THEORETICAL AND EXPERIMENTAL INVESTIGATIONS OF RESONANCE FLUORESCENCE LIDAR FOR MEASUREMENTS OF N_2^+ IN THE AURORAL ATMOSPHERE

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THEORETICAL AND EXPERIMENTAL INVESTIGATIONS OF RESONANCE FLUORESCENCE LIDAR FOR MEASUREMENTS OF N_2^+ IN THE AURORAL ATMOSPHERE

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Abstract

In this thesis, a series of experimental and theoretical studies of the resonance fluorescence lidar system at Poker Flat Research Range (located in Chatanika, Alaska) for use in obtaining measurements of aurorally produced molecular nitrogen ions (N_2^+) are presented. Obtaining measurements of N₂⁺ is made challenging by both the operational performance of the resonance lidar system and the high degree of geophysical variability inherent in the aurora. Analyses are conducted of measurements obtained by the operational sodium and iron resonance lidar systems to verify the lidar system performance. To increase the strength and quality of the lidar measurements, the telescope in the lidar receiver system was upgraded from a 0.6 m Newtonian telescope to a Cassegrain telescope with a 1.02 m diameter primary mirror. Lidar measurements from the system operating with this telescope are presented and compared to previous measurements to confirm an improvement to the overall operation. A spectroscopic analysis of the laser dye used in the previous development of the molecular nitrogen resonance lidar system is conducted to determine the cause of decreased lidar system performance at the operational wavelength relevant for studies of N_2^+ . A total of ten laser dyes are tested in the dye laser system. Based upon the performance of these dyes in the resonance lidar system, it is concluded that successful measurements of the strongest emission band in $N_{\,2}^{\,\scriptscriptstyle +}$ are unlikely due the transmittance of the diffraction grating at the relevant wavelength and low system efficiency in the dye laser. Therefore, the resonance lidar system is being developed to obtain measurements of the second strongest band of emissions in N₂⁺. To assess the capabilities of this system to obtain statistically significant measurements of aurorally produced N₂⁺, the expected resonance lidar signal is simulated by modifying an existing model. It is found that to obtain N_2^+ resonance lidar measurements of reasonable strength with the current operational system, the data would be obtained at high resolution and post integrated over selected temporal and spatial ranges.

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Dedication

То

Dr. David B. Friend

1954-2008

1 Introduction

Resonance fluorescence lidar is used to conduct studies of constituents of the middle atmosphere. In this chapter, the middle atmosphere and phenomena relevant to this study are introduced. The Rayleigh and resonance lidar systems used in this study are briefly discussed, while further details of the resonance lidar system are given throughout this work. The chapter concludes by discussing the previous work conducted and introducing the scope of this study.

1.1 The middle atmosphere

Earth's atmosphere is divided into a number of regions, called spheres, and each is separated by a boundary known as a pause [Figure 1.1]. The lowest layer of the atmosphere is the troposphere, where the air temperature decreases with height, being ~275 K at the surface. Containing approximately 80% of the mass of the atmosphere and 99% of the water vapor, this layer extends to an altitude of ~15 km where the temperature has decreased to ~225 K. The stratosphere extends from the top of the troposphere to an altitude of ~50 km. The temperature in the stratosphere increases with height due to absorption of ultraviolet (UV) solar radiation by ozone (O_3) . Above the stratopause is the mesosphere, extending to the mesopause at ~80 km and cooling with increasing altitude due to a decrease in solar heating relative to the stratosphere. Radiative cooling by carbon dioxide also contributes to the cooling of the mesosphere. The mesopause is the Earth's coldest climate [Chu and Papen, 2005] with temperatures less than ~175 K [Wayne, 2000]. The mesopause supports formation of noctilucent clouds (NLCs) [Collins et al., 2003]. The thermosphere, a layer of ionized molecules and charged particles which is also referred to as the ionosphere, extends from the mesopause into space ~1000 km above the surface of the Earth. Temperature increases with height in this region (up to \sim 1000 K) due to absorption of UV radiation by the trace amount of oxygen present there.

Below the thermosphere, the atmosphere is a well-mixed combination of gases, predominantly oxygen (O) and nitrogen (N), occupying ~21% and ~78% by volume,

respectively [Wallace and Hobbs, 2006]. The next two most abundant gases are argon (Ar) (less than 1%) and carbon dioxide (CO₂) (approximately 0.03%). The density of the atmosphere decreases with increasing altitude, and atmospheric mixing rapidly diminishes in the atmosphere, creating stratification of atmospheric constituents at altitudes above the troposphere. Molecular nitrogen and oxygen dominate at lower altitudes near 100 km, transitioning into atomic nitrogen and oxygen with increasing altitude [UCAR, 2008]. Hydrogen and helium dominate at the furthest reaches of the thermosphere.

Meteors entering the Earth's atmosphere begin to ablate at an altitude of ~90 km, depositing their atoms of matter, predominantly metals, into the upper atmosphere. These metals form layers, according to mass, above 80 km, and are composed of potassium, calcium, lithium, sodium, and iron. The concentration of metals in the atmosphere is variable due to seasonal changes in the atmospheric temperature profile, which affects air chemistry, as well as fluctuations in the frequency of meteorite impacts. Eventually, the metal atoms react with other atoms and molecules, forming molecular species, and become deposited on the Earth's surface. The existence of metal layers in the mesosphere is important to this study, as measurements of these stable atmospheric features aid in calibration of the lidar system.

The aurora borealis (known as aurora australis in the southern hemisphere), or northern lights, are among the most beautiful of atmospheric phenomena, occurring at altitudes from ~80 km to ~160 km. Auroral activity produces molecular nitrogen ions (N_2^+) in the middle atmosphere; It is the goal of this study to result in a resonance fluorescence lidar system which may obtain measurements of aurorally produced N_2^+ . The interactions between the Sun and the Earth which give rise to the aurora, the modification of the upper atmosphere due to auroral activity, and the production of N_2^+ are discussed in Chapter 2.



Figure 1.1 Simplified diagram of the thermal structure of the atmosphere and several upper atmospheric phenomena. Not to scale.

1.2 Resonance fluorescence lidar

Lidar (an acronym for light detection and ranging) is an active remote sensing technique. It is often referred to as laser radar because it is the optical equivalent of radar. A lidar system is composed of a laser, transmitter optics, a telescope, receiver optics, and various data acquisition devices and computerized controls [Figure 1.2]. The laser transmits pulses of light into the night sky with precise wavelengths, narrow spectral width, high energy, and small divergence [Chu and Papen, 2005]. In a Rayleigh lidar system, the photons transmitted by the laser are scattered elastically off of the particles in the atmosphere. This form of scattering is known as Rayleigh scattering and occurs when the scattering particles are much smaller than the wavelength of the incident light. Only a percentage of the total backscattered light is intercepted by the receiver system. The backscattered light is collimated before being filtered by the receiver optics and converted into an electrical signal by a photomultiplier tube (PMT). The data, comprised of raw photon counts, is recorded by a computer which synchronizes the transmitter and

receiver and determines the temporal and spatial resolution of the measurements from the known speed of light. The received lidar signal is measured as photon counts in height [Figure 1.3].

The lidar data in Figure 1.3 was obtained with the Rayleigh lidar system at Poker Flat Research Range, Chatanika, Alaska (PFRR, 65° N, 147° W). This system uses a neodymium yttrium aluminum garnet (Nd:YAG) solid state laser operating at a wavelength of 532 nm and was installed in November 1997 by researchers from the Geophysical Institute at the University of Alaska Fairbanks and the National Institute of Information and Communications Technology (NICT) in Japan. The lidar signal, due to Rayleigh scattering of the transmitted laser light, decreases with increasing altitude as the density of the atmosphere decreases. Electronic blanking suppresses the high signal received from the denser lower atmosphere because it would overload the detector. Exceptionally strong signals due to Mie scattering (scatter off of particles much larger than the wavelength of light) are often evident below ~25 km in the data. The signal above ~100 km is due to background light. The total lidar profile is the integrated signal from 22 data sets, or 352000 laser shots. One set is the integrated signal of 16 profiles; each profile is the sum of the return signals from 1000 laser shots. The Rayleigh lidar system is used to measure the density and temperature structure of the atmosphere at altitudes from 35 km to 80 km.

Resonance fluorescence lidar (often referred to as simply resonance lidar) differs from other lidar methods because it is species specific. The resonance lidar system probes a particular atmospheric constituent by transmitting laser light of the same frequency as a resonant line within the target atom or molecule. If the laser light is of the correct energy, it is absorbed by the atom or molecule, raising one of its electrons to a higher energy level. The lifetime of this excited state is short and the electron quickly returns to a lower energy level, giving up the energy it gained in the form of a photon with the same frequency as the first. This process is resonance fluorescence. The photons are emitted randomly, unlike Rayleigh scatter which is preferentially forward or backward. Resonance lidar techniques have the ability to measure concentrations of distinct atmospheric species precisely and to determine the populations of different vibrational and rotational energy states [Collins et al., 1997].



Figure 1.2 Simple schematic of lidar system.



Figure 1.3 Rayleigh lidar signal versus altitude profile.

An example of resonance lidar data is given in Figure 1.4. The altitude resolution is 75 m. The total resonance lidar signal profile has components due to both Rayleigh and resonance scattering. In this case (the number of profiles per set and the number of laser shots per profile are variable), each set is composed of eight profiles of 1000 shots each and four profiles were omitted when the data was obtained, for a total of 204000 shots. Electronic blanking suppresses the strong scatter signal below 15 km. The component of the signal due to Rayleigh scattering extends from 15 km to ~60 km. The signal due to resonance scattering, in this case from the mesospheric sodium layer, is observed from ~80 km to ~110 km. The background signal is visible above ~110 km.



Figure 1.4 Resonance lidar signal versus altitude profile.

A resonance fluorescence lidar system was installed at PFRR in April 1999. The laser transmitter is an excimer-pumped dye laser, consisting of an LPX220i® excimer laser and a Scanmate2® pulsed dye laser. The excimer laser is produced by passing a high voltage electric discharge (of 18 or 20 kV, selected by the user) through a gas mixture of xenon and chlorine. The atoms in the gas repulse each other in their ground

states, but attract each other when excited by the electrical discharge and form a xenon chloride molecule [Hecht, 1992]. The ground state does not exist for xenon chloride, so a population inversion, a condition for lasing, exists. A population inversion exists when there are more electrons in an excited state than in lower energy states. The excimer laser beam is used to excite dye molecules, suspended in a liquid solvent and pumped through a cuvette, in the Scanmate2[®] dye laser system. The excited dye molecules absorb photons from the pump beam, and fluoresce over a range of wavelengths. For a detailed analysis of dye lasers see Schäfer [1990]. The precise wavelength of the output laser light is selected by inputting the wavelength into the control computer which uses a stepping motor to vary the tilt of a Littrow mounted diffraction grating. The wavelength of the laser beam is tuned until its frequency corresponds to the frequency of the transition within the target species; when these two frequencies match, resonance fluorescence will occur. Operational details of the dye laser system are discussed throughout this work.

The resonance lidar system became operational during the summer of 1999. This system has since been used to conduct studies of the mesospheric sodium (at an operational wavelength, λ , of ~589 nm) and iron layers (at an operational wavelength, λ , ~372 nm) [Breese, 2001; Hou, 2002; Su, 2007]. A resonance lidar system for profiling aurorally excited molecular nitrogen ions (N⁺₂) was proposed by Garner and Dao [1995]. Collins et al. [1997] conducted an optimization analysis of such a lidar system. Obtaining measurements of an aurorally excited molecular species could yield significant data for use in fundamental upper atmospheric studies and space weather research. Atmospheric ions and their chemistry play a central role in determining the concentration and distribution of electrons in the upper atmosphere [Rees, 1989; Wayne, 2000] and diatomic nitrogen has great potential as a diagnostic tool for this region due to its rich spectrum and the diverse and complex nature of its lower excited electronic states [Morrill and Benesch, 1996]. Furthermore, diatomic nitrogen ions are well suited for lidar studies due to their short lifetimes and relatively large absorption cross sections, which allow for strong interactions between the ions and the transmitted laser light.

1.3 Previous work

Liguo Su worked with Dr. Richard L. Collins to develop a resonance lidar system for obtaining measurements of aurorally produced N₂⁺ and documented his work in his doctoral dissertation [2007]. Su based the nitrogen resonance system on the operational iron resonance system. The iron resonance system operates at wavelengths near 372 nm and the performance of this system has been verified [Breese, 2001; Hou, 2002]. The optimum vibrational band of N₂⁺ transitions for resonance lidar measurements occurs between the $X\,^2\Sigma_g^{_+}$ and $B\,^2\Sigma_u^{_+}$ electronic energy levels. The optimum rotational resonance line in the vibrational band, determined by Collins et al. [1997], occurs between the two lowest vibrational levels at a wavelength of ~390.303 nm for an atmospheric temperature of 500 K (the spectroscopy of N_2^+ and the determination of the optimum operational wavelengths for resonance lidar studies are discussed in Chapter 5). Su developed a computer simulation to model the expected measurements of the N₂⁺ resonance lidar system. Based upon the results of this model, Su concluded that the lidar system was capable of making statistically significant measurements of aurorally produced N₂⁺ at an operational wavelength of 390.303 nm. The dye known as QUI (3,5,3"",5""-tetra-t-butylp-quinquephenyl, C₄₆H₅₄) [Brackmann, 1997], used in the dye laser for iron studies, also fluoresces at the relevant wavelength for nitrogen ion studies, so it was thought that calibrating iron measurements could be taken, followed by nitrogen measurements, without having to change the type of dye. As the N_2^+ is more difficult to measure than the mesospheric iron layer due to the variability in auroral production of N_2^+ , obtaining resonance measurements of iron would allow for calibration of the laser and a basis of comparison in signal levels.

In the spring of 2007, Su conducted field tests of the prototype N_2^+ resonance lidar system. Su began by conducting a series of tests confirming the operation of the lidar system at both visible and ultraviolet wavelengths. Su used resonance lidar measurements of the mesospheric sodium layer at 589 nm to confirm and optimize the operation of the resonance lidar. These measurements were followed by measurements of the mesospheric iron layer at 372 nm, which were compared to lidar signals received from transmitted laser light with a wavelength of 390 nm. The results showed a significant and unexpected loss of system efficiency (a factor of 10) when the resonance lidar system was operated at 390 nm [Su, 2007] [Figure 1.5, courtesy of Su]. Su carefully tested the receiver optics and found that the optical elements were behaving as designed and did not seem to be the source of the signal loss. Su postulated that the QUI dye used in the laser system could support significant amplified spontaneous emission at 390 nm, where the dye has maximum gain [Figure 1.6], and this could lead to the decrease in signal. Su further recommended a careful realignment of the laser to yield higher laser pulse energy, which would facilitate more thorough insight into lidar system performance.



Figure 1.5 Rayleigh signal from 20-25 km for resonance lidar system operating with QUI dye. Figure courtesy of Su, 2007.



Figure 1.6 Energy output of QUI dye laser with wavelength.

1.4 The scope of this study

This thesis consists of seven chapters and presents the field tests and theoretical investigations conducted to further the development and understanding of an operational N_2^+ resonance lidar system at PFRR.

In Chapter 2, I discuss how interactions between the Sun and the Earth cause the aurora and lead to the production of molecular nitrogen ions in the middle atmosphere. The modification of the upper atmosphere caused by auroral activity is also discussed.

In Chapter 3, the studies of lidar system performance that have been conducted are presented. Following the work of Su [2007], the cavity optics in the excimer pumped dye laser were realigned. Resonance lidar measurements of the mesospheric sodium and iron layers are compared to similar measurements obtained prior to the realignment of laser cavity optics. I discuss the alignment of a 1.02 m Cassegrain telescope and its integration into the resonance lidar receiver system. Data obtained with the 1.02 m telescope is presented and the improved strength of the measurements is discussed. I

conduct a spectroscopic analysis of the laser tuning system and confirm that it is operating as designed. I also explore characteristics of the diffraction grating used in the dye laser system.

In Chapter 4, I examine the fluorescent characteristics of the dyes used in the dye laser in the resonance lidar system. A study of spontaneous emission within QUI and Rhodamine 6G (Benzoic Acid, 2-[6-(ethylamina)-3-(ethylimino)-2,7-dimethyl-3Hxanthen-9-yl]-ethylester, monohydrochloride) [Brackmann, 1997] laser dyes is discussed. These studies were conducted to explain the lack of lidar system performance at an operational wavelength of 390 nm observed by Su [2007]. A total of ten laser dyes were tested in the resonance lidar system for use in measurements of N_2^+ and the results of these tests are presented. The choice of one of these dyes as a likely candidate for successful resonance lidar studies at an operational wavelength of ~428 nm (causing resonance fluorescence in the second strongest band of transitions in N_2^+) is explained.

Chapter 5 presents the spectroscopy of N_2^+ and an analysis conducted to determine the optimum operational wavelengths for lidar measurements employing resonant fluorescence in two vibrational bands.

A resonance lidar system for profiling N_2^+ at an operational wavelength of ~428 nm is simulated in Chapter 6. The lidar model is described and the simulated resonance lidar signals are discussed. The model output is post integrated to demonstrate how signal processing methods would be used operationally to achieve statistically significant measurements. In Chapter 7, I present my conclusions and suggestions for future studies.

2 The Auroral Atmosphere

A simplified description of the interactions between the Sun and the Earth which result in the production of the aurora and molecular nitrogen ions in the upper atmosphere is given in this chapter. The optical emissions which occur during auroral activity are discussed, as well as how auroral activity modifies the upper atmosphere.

2.1 Interactions between the Sun and Earth

Viewed from the Earth's surface, aurora often resemble curtains of shimmering green, red, or purple light. Their bottommost limit of altitude above Earth's surface is over 10 times greater than the altitude at which jet aircraft fly [Akasofu, 2009]; Aurora is not considered a meteorological phenomenon. The cause of the aurora was not easily discovered, and the field of auroral physics still has many unanswered questions. The current understanding of the aurora was brought about by the effort of many scientists over the past century, though the earliest records of auroral sightings date to ~350 B.C. [Akasofu, 2009]. The history of auroral science is well discussed in Exploring the Secrets of the Aurora by Akasofu [2007]. It is now understood that the aurora is caused by energized particles from the solar wind impacting constituents of Earth's upper atmosphere.

The Sun is composed primarily of hydrogen and helium. At its core, temperatures and pressures are high enough to result in nuclear fusion, the process which powers the Sun. The visible surface of the Sun is called the photosphere. Above the photosphere is the corona, a low density, high temperature gas, thought of as the solar atmosphere. At a temperature of $\sim 1 \times 10^6$ K, coronal temperatures far surpass the ~ 6000 K of the photosphere. The heating mechanism operating within the Sun's corona is not well understood, but these high temperatures render the Sun's atmosphere unstable. It expands away from the Sun at supersonic speeds and is known as the solar wind. The solar wind is a gas composed primarily of protons and electrons, known as plasma. Typical charged particles in the solar wind have energies of ~ 1 keV [Carroll and Ostlie, 1996]. Plasma is generally electrically neutral, having approximately equal numbers of each charge carrier [Baumjohann and Treumann, 1997].

The most basic characteristics of plasmas are defined by three criteria [Baumjohann and Treumann, 1997]. The first of these criteria is that the free charges must be close enough together that they interact with many nearby charges, rather than just the nearest. In this way, the plasma has bulk fluid behavior. The length scale over which significant charge separation can occur is defined as the Debye length. Therefore, this criterion is valid when the number of charges in a sphere with a radius equal to the Debye length is much greater than unity. The second plasma criterion requires that the Debye length must be much smaller than the physical size of the plasma. When this requirement is met, bulk interactions will dominate over boundary interactions and the plasma behaves collectively. The third criterion is that if a parcel of plasma is displaced from its quasi-neutral state by an external force, the electrons will be accelerated in an attempt to restore neutrality. Their inertia causes them to oscillate about their equilibrium positions. For the plasma to remain plasma, the frequency of the electron oscillations (called the plasma frequency) must be greater than the frequency of collisions with neutrals in the plasma. If the collision frequency is greater than the plasma frequency, the electrons will equilibrate with the neutrals and the gas will no longer be plasma. The typical density of the solar wind is ~7 ions cm⁻³ [Carroll and Ostlie, 1996], allowing for a low frequency of collisions.

In addition to the qualities outlined so far, plasma is highly conductive, meaning it transmits electrical signals efficiently. Also, as the charged particles within the plasma move, they generate a current. This variable current induces a magnetic field, which subsequently influences the charge carriers and changes the current, which changes the magnetic field, and so forth. In short, moving plasma interacts with itself, and it is this characteristic of internal field generation that complicates plasma physics.

The plasma of the solar wind varies in speed and composition due to changes in the Sun's activity due to complex internal plasma interactions. An active Sun is characterized by eruptions of plasma from the surface of the Sun due to local variability in the Sun's magnetic field. Sunspots, solar flares, and coronal mass ejections are all associated with an active Sun, while the absence of these and other features define a quiet Sun. Solar activity waxes and wanes on an \sim 11 year cycle referred to as the solar cycle. Also varying on an \sim 11 year cycle is the orientation of the Sun's dipolar magnetic field, which switches orientation with each cycle.

The accepted reason for this 11 year variability in solar activity is due to the nonrigid body rotation of the Sun [Freedman and Kaufmann, 2002]. It is a sphere of dense plasma, which flows as a fluid. This allows differential rotation from pole to pole on the Sun's surface, meaning the plasma at the equator rotates faster than the plasma at the poles. As the plasma flows, magnetic fields flow with it in accordance with the Frozen Field Theorem, which states that in a field of high electrical conductivity, like plasma, the magnetic field and the material motion are coupled as if the magnetic field were frozen in the medium [UCAR, 2008]. Meaning, if two parcels of plasma are connected by a common magnetic field, they will continue to be connected by that field no matter the degree of deformation in the plasma. The theorem is valid when the electrical conductivity is high, the current density is not to large, and the spatial and temporal scales are not too small.

As the Sun rotates, a longitudinal magnetic field line frozen into the plasma will rotate faster at the equator than at the poles. After successive rotations, the field line will no longer be longitudinal, but stretched out at the equator in the direction of the Sun's rotation. The field winds around the Sun as it rotates. Where it becomes 'tangled' and turbulent, loops of plasma associated with concentrated magnetic fields may erupt out of the solar surface. The points where the loops connect to the solar surface are referred to as footprints. The Babcock magnetic dynamo model [Freedman and Kaufmann, 2002] suggests that sunspots should first appear near the poles and later form nearer to the equator, and this is what is observed. The model also agrees with many other observed qualities of sunspots.

Sunspots in the northern and southern hemisphere have opposite polarities, with the leading 'footprint' of the spot having the same magnetic orientation as the hemisphere it is in. As sunspots form closer to the equator, the opposing magnetic fields of the turbulent plasma begin to cancel each other out and eventually reverse the Sun's overall magnetic field, beginning the solar cycle anew.

The solar wind is divided into two components: the fast wind and the slow wind. The fast wind originates from coronal holes near the poles. A coronal hole is associated with locally open field lines, which do not close until far out in interstellar space. The magnetic field of the Sun is generally dipolar on a local scale [Carroll and Ostlie, 1996]. Particles cannot cross magnetic field lines, except due to collisions, and freely stream out along these lines independently of the solar activity cycle at speeds between 400 and 800 km s⁻¹ [Carroll and Ostlie, 1996; Freedman and Kaufmann, 2002]. The slow wind is due to active solar features, and is, therefore, highly variable. It is slowed by the closed field lines associated with the more equatorial latitudes where active solar features appear. The slow wind has speeds between 350 and 400 km s⁻¹ [Freedman and Kaufmann, 2002]. Coronal mass ejections are massive eruptions on the solar surface that cause an acceleration of the particles in the solar wind. A coronal mass ejection may lead to solar wind speeds of 400 to 2000 km s⁻¹ [Freedman and Kaufmann, 2002] in addition to changing the composition of the plasma in the solar wind.

The solar wind streams through interplanetary space, carrying the Sun's magnetic field along with it in accordance with the Frozen Field Theorem. When the solar wind encounters a magnetic field, like that of a planet, the charges in the plasma will be deflected. In this way, it is Earth's magnetic field, or magnetosphere, which protects it from direct bombardment by the particles of the solar wind. Earth's magnetic field is approximately dipolar and northward oriented. The axis of the dipole is tilted 11° with respect to the Earth's rotation axis. The solar wind flows around the magnetosphere at a distance of ~10 Earth radii (the Earth's radius is ~6378 km).

The solar wind's speed changes from supersonic to subsonic when it impacts the Earth's magnetic field. A common analogy for this situation is that of a rock in a stream. As the flowing fluid comes in contact with the obstacle, it slows down and piles up on the upstream side. The fluid slows, and then flows around the obstacle. The area of

compressed, slowed, fluid on the upstream side is called the shock wave. In the case of the flowing solar wind encountering the obstacle of the Earth's magnetic field, the resultant shock wave is called the bow shock.

To understand further why the bow shock is formed, beyond the 'rock in a stream' analogy, we examine how a shock wave is formed in Earth's atmosphere. These shock waves are called sonic booms and are associated with aircraft traveling faster than the speed of sound. The shockwave forms because the air molecules are being pushed away from the traveling aircraft faster than they can physically move. This causes the displaced air to become highly compressed, creating a high density wave or shock wave. It is easily understood that the creation of this shock wave is related to the mean free path of the air molecules between collisions, which is quite short in Earth's lower atmosphere. However, the mean free path of solar wind particles is about 10^{12} m (about the distance from the Earth to the Sun), meaning the solar wind particles are practically collisionless [Kallenrode, 2001]. The magnetospheric bow shock is the first proof of the existence of collision-less shock waves, which were theoretical until the discovery of the bow shock. The mechanism that allows for frequent interaction between the nearly collision-less particles of the solar wind is the frozen-in magnetic field that they share. The magnetic field acts as a coupling device, binding the particles together.

Due to the shock transition between the freely flowing solar wind and the Earth's magnetosphere, the magnetosphere is compressed on the dayside and streams out with the deflected solar wind on the night side to a distance of ~1000 Earth radii [UCAR, 2008]. The magnetopause is the outer boundary of the magnetosphere that defines the boundary where the outward directed magnetic pressure of the Earth's magnetosphere is counterbalanced by the solar wind. Most of the particles in the solar wind are deflected around the magnetopause and continue to flow through interplanetary space. Some particles become trapped in the Earth's magnetic field via the breaking and reconnection of solar and terrestrial magnetic fields.

When magnetic fields in two adjacent regions are anti-parallel, the Frozen Field Theorem may break down, meaning the magnetic field lines vanish locally. They lose their identity, 'break', 'reconnect', and then are rapidly repelled from each other by the magnetic tension force. The reason for this behavior is the existence of current sheets, which are a consequence of adjacent anti-parallel magnetic fields [UCAR, 2008]. Current sheets, which exist in these regions to maintain a force balance at the boundary, are narrow regions of high current density. Current sheets are formed between all such magnetic boundaries and multiple current sheets exist between the magnetic fields of the solar wind and Earth. The current density in the sheet between the magnetic fields of the solar wind and Earth can be sufficiently high enough to cause invalidation of the Frozen Field Theorem. When the solar wind field lines break and reconnect with the terrestrial field lines, particles from the solar wind gain access to the Earth's magnetosphere. Reconnection occurs at different areas in the magnetosphere depending on the orientation of the solar wind's magnetic field.

Solar wind particles that end up on a closed terrestrial magnetic field line with both ends in the Earth's atmosphere are in a magnetic mirror geometry which affects their motion. The equations that govern the interactions of charged particles in electric E and magnetic B fields are referred to as Maxwell's Equations (for a full treatment of Maxwell's Equations see Griffiths [1999]). From them, it is shown that the force exerted on a charged particle in an electric field is equal to the charge q times the electric field E. This force is called the Coulomb force:

$$\vec{F}_c = q\vec{E}$$
(2.1)

The force exerted on a moving charge in an external magnetic field is equal to the charge times the curl of the velocity vector \vec{v} and the magnetic field vector. This force is the Lorentz force:

$$F_{L} = q(v \times B) \tag{2.2}$$

Therefore, the equation of motion for a charged particle of mass m traveling through both external B and E fields is:

$$\vec{F}_{Net} = m \frac{d\vec{v}}{dt} = q(\vec{E} + \vec{v} \times \vec{B})$$
(2.3)

If the motion of a charged particle in the absence of an electric field (E=0) is considered, with a magnetic field with a component only in the z direction, it is found that, by the right hand rule, the resultant force is perpendicular to both the velocity and magnetic field vectors. The resultant motion is a circular trajectory, and the particle is said to have gyratory motion. Whenever the particle has a nonzero vertical velocity component, the particles trajectory will be a helix. The pitch angle of the helix α (the angle between the particle's velocity vector and the local magnetic field) depends on the ratio between the perpendicular and parallel velocity components. For a full discussion of charged particle motion see Baumjohann and Treumann [1997] and Kallenrode [2001].

If an electric field is present, a drift will be superimposed on the gyratory-helical motion. How this drift affects the particle's motion depends on the relationship between the vector components; Several types of drifts are observed. Also, a magnetic drift will likewise be superimposed if the magnetic field is not homogeneous.

A particle's magnetic moment μ (which quantifies its contribution to the net magnetic field of the system) and its kinetic energy are invariant over the typical scale of particle motions considered. A particle's magnetic moment is mathematically expressed as the ratio of its kinetic energy to the external magnetic field strength:

$$\mu = \frac{mv^2 \sin^2 \alpha}{2B}$$
(2.4)

Here the perpendicular vector components are considered. As a particle gyrates about a terrestrial magnetic field line, it eventually moves toward the regions of increased magnetic field strength at the poles (B_{max}). If the magnetic field strength B increases, either the mass, velocity, or pitch angle must also increase if the magnetic moment is to remain constant. The energy is a constant of motion, so only the particle's pitch angle may vary. If the particle's pitch angle exceeds 90°, the kinetic energy term cannot increase any further and the particle cannot enter a region of higher magnetic field strength. Particles which do not have enough energy to travel far enough along the terrestrial field lines to intersect the Earth's atmosphere are left to bounce back and forth between the magnetic mirror points (the points at which the pitch angle reaches its maximum value). These charged particles are 'stored' in the Earth's magnetic field and
form the Van Allen radiation belts. Only particles with pitch angles and energies in a limited range may escape the magnetic mirror and precipitate into the atmosphere. This range of angles is referred to as the loss cone.

The particles that manage to precipitate into the atmosphere near the poles, where the terrestrial field lines extend into the atmosphere, cause the aurora in an oval centered on the magnetic pole called the auroral oval. The oval is fixed with respect to the Sun, so the daily rotation of the Earth beneath it varies the distance between the oval and any point of geomagnetic latitude [Akasofu, 2009]. At 1 p.m. AK time (12 magnetic local time) the distance between Fairbanks, Alaska, and the auroral oval is the greatest. As the Earth rotates, Fairbanks advances toward the auroral oval until at 1 a.m. (AK time) it is overhead. The oval is ever present but brightens and expands with the injection of charged particles.

2.2 Modification of the upper atmosphere

The particles which precipitate into the atmosphere are primarily electrons. The electrons trapped in the Earth's magnetic field have energies of only a few eV or a few keV. They must be accelerated in order achieve the energy required to survive several hundred collisions in the upper atmosphere to reach auroral altitudes. The method of this acceleration is not well understood and is currently one of the major problems faced by auroral physics [Akasofu, 2009]. Accelerated particles which cause the aurora typically have energies of a few 100 eV to 10 keV [Prölss, 2004], but their energies may range as high as 100 keV [Kallenrode, 2001]. As they precipitate into the atmosphere, they collide with high altitude neutral or ionized particles. The lowest altitude at which aurora appear is determined by how energetic the incoming particles are and how many collisions they can withstand before they no longer have the energy required to excite atmospheric particles. Furthermore, most of an auroral particle's energy is deposited into the atmosphere at the lowest altitude it reaches. Most electrons with energies of 0.1 keV deposit all their energy above ~200 km, while those with energies of 1 keV penetrate to altitudes above 130 km [Prölss, 2004]. Particles with energies of 10 keV typically deposit their energies at altitudes of ~100 km [Prölss, 2004].

As auroral particles collide with atmospheric constituents, they impart some of their energy, which can cause ionization, dissociation or excitation. When ionization occurs, an electron is removed from an atom or molecule, resulting in a positively charged ion. Dissociation occurs when a molecule is split into smaller particles or atoms by the collision. Electrons or particles resulting from dissociation or ionization carry away excess energy and may cause a number of secondary energy distribution processes, including secondary ionization and dissociation, scattering, charge exchange, dissociation exchange, excitation exchange, dissociation recombination, radiative recombination, collisional quenching, and combinations of these [Prölss, 2004]. Positive and negative molecular ions may also interact to yield neutral particles that disperse energy as internal motion [Rees, 1989; Wayne, 2000]. This cascading effect of energy redistribution caused by the precipitation of auroral particles has important consequences for the dynamics and energetics of the upper atmosphere near the poles.

Most of the energy (50%) deposited in the upper atmosphere by precipitating auroral particles is expended as heat. Approximately 30% of the energy is converted to chemical potential energy, and less than 1% is converted into radiation [Prölss, 2004]. The rest is scattered back into the magnetosphere. If the ionization density produced by auroral electrons is 2x10¹¹ m⁻³ the heating rate is calculated to be 10⁻⁸ W m⁻³ [Prölss, 2004]. An immediate consequence of the intense heating caused by the precipitation of auroral particles is the expansion of atmospheric gases, which is generally upward due to the minimal back pressure in the high atmosphere. At high altitudes, the expanding gases drive horizontal winds that distribute the heat over a wide area. Vertical wind are generated as well which lead to a decrease in the abundance of lighter gases at lower altitudes and an increased abundance of heavier gases at high altitudes. This effect may alter air chemistry on a number of scales.

2.3 Auroral optical emissions

Optical emissions result when excitation is caused by a collision between an auroral particle and a constituent of the atmosphere. In the excitation process, an electron within an atom or molecule is raised from a lower energetic state to a higher one. Optical energy emissions are resultant because the excited states are unstable. A photon with an energy equivalent to the energy separation between the excited state and the lower state is emitted when the electron returns to a lower energy state from the unstable excited state. In equation form, the energy separation between the upper (E_2) and lower (E_1) energy levels is related to the wavelength λ of the emitted light by

$$E_2 - E_1 = hv = \frac{hc}{\lambda}$$
(2.5)

where h is Planck's constant $(6.63 \times 10^{-34} \text{ J s})$, v is the frequency of the emitted radiation, and c is the speed of light $(2.998 \times 10^8 \text{ m s}^{-1})$. The emitted auroral spectrum is not continuous because the atoms and molecules in the atmosphere emit light only at discrete wavelengths corresponding to the separation of their respective energy levels.

The features of the auroral spectrum are almost all due to emission lines or bands of neutral or ionized N₂, O, O₂, N, and NO. The most common auroral light is caused by the least energetic auroral particles, which create aurora that appears green [Figure 2.2]. This light has a wavelength of ~557.7 nm and corresponds to transitions within atomic oxygen (O). Attributing this auroral green line to oxygen was complicated by the understanding that oxygen exists only in its diatomic form in the atmosphere. This is true in the lower atmosphere, but at higher altitudes in the ionosphere absorption of solar UV radiation leads to chemical reactions which result in atomic oxygen, whose emission lines differ from the band emissions of O₂. The excited state of O has a relatively long lifetime of approximately 0.7 s [Akasofu, 2009], which gives green auroral light a lingering nature relative to other auroral optical emissions. At lower altitudes, collisions may take away the excitation energy in O during the long lifetime of its excited state, and green emissions will not be visible. In regions of high vacuum in the atmosphere (above ~240 km) low energy particles may excite O to emit in the red at wavelengths of 630 nm and 636.4 nm [Figure 2.3] [Prölss, 2004]. The transitions yielding red O emissions are particularly long lived, with lifetimes of ~110 s [Prölss, 2004].

The most energetic of auroral particles reach relatively low altitudes where they may excite molecular nitrogen (N_2) [Figure 2.4]. Molecular nitrogen band emissions occur in the red between 650 nm and 680 nm, and are viewed as a reddish lower border

on bright, active, auroral arcs [Jones, 1974]. Collisions with molecular nitrogen may also result in the ejection of an electron, resulting in ions (N_2^+). N_2^+ band emissions are ultraviolet to violet (390 nm to 470 nm), with the strongest band of emissions centered at 391.4 nm. Approximately 40% of the ~390 nm wavelength light from N_2^+ is produced by electrons with energy less than 85 eV [Feldman and Doering, 1975] and it is estimated that one of every 25 ionizations of a nitrogen molecule leads to the emission of a 391.4 nm photon [Prölss, 2004]. The N_2^+ excited state is short lived, with a lifetime of ~62 ns, [Collins et al., 1997], as are the N_2^+ molecules themselves, which are rapidly consumed in secondary chemical reactions with atomic and diatomic oxygen [Wayne, 2000]. The difference between the lifetimes of excited O and N_2^+ gives the appearance of lingering green light chasing short-lived magenta light.



Figure 2.1 Green aurora due to oxygen emission. Photo by Dirk Lummerzheim, Geophysical Institute. Not protected under copyright; used with permission.



Figure 2.2 High altitude red aurora due to oxygen emission. Photo by Poul Jensen. Not protected under copyright; used with permission.



Figure 2.3 Magenta lower border on aurora due to molecular nitrogen emission. Photos by Poul Jensen. Not protected under copyright; used with permission.

3 Lidar System Performance

In this chapter, the relevant governing equations for this lidar study are introduced. This presentation of the lidar method follows the recent review of Chu and Papen [2005], Collins et al. [1997], and the established practice of previous theses by members of the Lidar Research Laboratory at the University of Alaska Fairbanks [Breese, 2001; Hou 2002; Su, 2007]. The lidar method has been presented in a variety of styles since its inception and the reader is referred to The International Society for Optical Engineering (SPIE) Milestone Series of Selected Papers on Laser Applications in Remote Sensing [Grant et al., 1997] for discussions regarding the discovery and development of the lidar method.

This chapter also examines the performance of the resonance lidar system following realignment of the laser cavity optics and again after integration of a larger telescopic receiver. Finally, aspects of the laser tuning control system are investigated to assess their impact on lidar system performance.

3.1 The lidar equation

The lidar equation relates the received photon count from a scattering region to the number of emitted laser photons, the concentration of scatterers, the interaction between the laser radiation and the scatterer, and the lidar system efficiency [Chu and Papen, 2005]. It is assumed that scattering processes are independent and only single scattering occurs.

The expected total photon count, $N_{TOT}(z)$, from the altitude range z- $\Delta z/2$ to $z+\Delta z/2$ in a time interval Δt is expressed as

$$N_{TOT}(z) = N(z) + N_B + N_D$$
(3.1)

where N(z) is the signal from scatterers at altitude z, N_B is the signal due to background light (moonlight, starlight, auroral emissions, anthropogenic light emissions, etc.), and N_D is signal due to noise intrinsic to the detector system. Each of these terms is dependent on a number of factors. The signal due to resonance scattering, $N_S(z)$, is expressed as

$$N_{s}(z) = [\eta T^{2}] \cdot \left[\frac{E_{L}R_{L}\Delta t}{hc/\lambda_{L}} \right] \cdot [C_{eff}(v_{L}, T, v)\rho(z)\Delta z] \cdot \left[\frac{A_{R}}{4\pi z^{2}} \right]$$
(3.2)

where η is the efficiency of the receiver, T is the transmission of the atmosphere at the laser frequency λ_L (s⁻¹), E_L is the laser energy per pulse (J), R_L is the repetition rate of the laser (pulse per second, pps), $\rho(z)$ is the concentration of scatterers at altitude z (m⁻³), C_{eff} is the effective scattering cross section of scatterers at wavelength λ_L (m⁻²) and temperature T (K) at height z with velocity v (m s⁻¹), and A_R is the area of the telescope's primary mirror. The factor of 4π is due to the isotropic nature of resonance scatter. The Rayleigh lidar signal, $N_R(z)$, is expressed as

$$N_{R}(z) = [\eta T^{2}] \cdot \left[\frac{E_{L}R_{L}\Delta t}{hc/\lambda_{L}}\right] \cdot [\sigma_{\pi}^{R}(\lambda_{L})\rho(z_{R})\Delta z] \cdot \left[\frac{A_{R}}{z_{R}^{2}}\right]$$
(3.3)

where the Rayleigh backscatter cross section, σ_{π}^{R} , is given by

$$\sigma_{\pi}^{R} = 5.45 \cdot \left(\frac{550}{\lambda_{L}}\right)^{4} \cdot 10^{-32} \,\mathrm{m}^{2} \mathrm{sr}^{-1} \tag{3.4}$$

and $\rho(z_R)$ is the concentration of Rayleigh scatterers.

The background signal N_B is expressed as

$$N_{B} = \eta \cdot \left(H_{N} R_{L} \Delta t \pi \left(\frac{\Delta \theta_{R}}{2} \right)^{2} A_{R} \Delta \lambda \right) \cdot \left(\frac{1}{hc^{2} / \lambda_{R}} \right)$$
(3.5)

where H_N is the background sky radiance (W m⁻² µm⁻¹ sr⁻¹), $\Delta \theta_R$ is the field-of-view of the receiver (rad), $\Delta \lambda$ is the bandwidth of the detector (µm), and λ_R is wavelength of the received light (m). N_D is expressed as

$$N_{\rm D} = (C_{\rm N} R_{\rm L} \Delta t) \cdot \left(\frac{2\Delta z}{c}\right)$$
(3.6)

where C_N is the dark current count rate of the detector (s⁻¹).

The terms in equations 3.2 and 3.3 can be placed into four groups, separated here by brackets. The first (leftmost) term is an efficiency term for the lidar system and is always less than unity. The second term gives the number of photons being transmitted into the atmosphere by the laser. The third bracketed term gives the probability of scattering by the target species. The fourth term is the probability that a scattered photon is detected.

There are several ways to increase the expected lidar signal, beyond increasing the operating specifications of the laser system (such as E_L and R_L , which may be invariable in some cases). By increasing the light gathering power (related to A_R) of the telescope, more of the backscattered light can be intercepted and a greater signal can be measured. The expected lidar signal can also be increased by increasing Δt and Δz , which reduces the resolution of the measurements.

The density of resonance scatterers at an altitude z can be determined from the ratio of the resonance lidar signal at altitude z and the Rayleigh lidar signal at altitude z_R (typically, 30 km). The resultant expression for density is

$$\rho(z) = \frac{N_{s}(z) - \widetilde{N}_{B}}{N_{R}(z_{R}) - \widetilde{N}_{B}} \cdot \frac{\sigma_{\pi}^{R}(\lambda_{L})}{C_{eff}(\lambda_{L})} \cdot \frac{4\pi z^{2}}{z_{R}^{2}} \cdot \rho(z_{R})$$
(3.7)

where \tilde{N}_{B} is the average background signal from 120 to 125 km. The density at the Rayleigh altitude is calculated from the temperature and pressure profile measured by a radiosonde launched from Fairbanks International Airport, which is available for any given day from the University of Wyoming archives at http://weather.uwyo.edu/upperair. The terms for system efficiency and laser energy cancel out in this ratio and, therefore, do not have to be determined to find the density of scatterers. The value of the effective scattering cross section, C_{eff} , however, must be known.

In atmospheric physics, a cross section refers to the probability of a type of interaction occurring. The effective scattering cross section, C_{eff} , is an important quantity defined by the interaction of an atom, based on its absorption cross section, and the laser radiation's characteristic line shape function. The absorption cross section refers to the ability of an atom to absorb a photon of appropriate energy and is determined by atomic structure. C_{eff} is discussed further in the next section. By taking the ratio of the total Rayleigh lidar signal to the total resonance lidar signal and rearranging, an expression for C_{eff} is obtained:

$$C_{eff}(\lambda_{L}) = \frac{N(z) - \widetilde{N}_{B}}{N_{R}(z_{R}) - \widetilde{N}_{B}} \cdot \sigma_{\pi}^{R}(\lambda_{L}) \cdot \frac{4\pi z^{2}}{z_{R}^{2}} \cdot \frac{\rho(z_{R})}{\rho(z)}$$
(3.8)

To operationally obtain a value for C_{eff} , a series of lidar measurements are made at wavelengths spanning the range of resonant interaction with the target species. It is assumed the ratio of $\rho(z_R)$ to $\rho(z)$ remains constant over the time necessary to obtain these measurements. The ratio of the resonance signal to the Rayleigh signal is, therefore, directly proportional to C_{eff} . The procedure which is used to determine the value of C_{eff} and the linewidth of the laser is discussed further in the next section.

An expression for the efficiency of the resonance lidar system is also obtained by taking the ratio between the Rayleigh and resonance signals and rearranging:

$$\eta T^{2} = \frac{N_{s}(z) - \tilde{N}_{B}}{\left[\frac{E_{L}R_{L}\Delta t}{hc/\lambda_{L}}\right] \cdot \left[\rho(z)C_{eff}(\lambda_{L})\Delta z\right] \cdot \left[\frac{A_{R}}{4\pi z^{2}}\right]}$$
(3.9)

 C_{eff} is experimentally determinable (and, therefore, the density of scatterers and the efficiency of the system) for the sodium and iron resonance lidar systems, but would not be for the N_2^+ resonance lidar system because of the high temporal and spatial variability of the aurorally produced N_2^+ . It cannot be assumed that the density of scatterers remains constant over the measurement time. Measurements of C_{eff} and the linewidth of the laser for the sodium (Na) and iron (Fe) systems, however, provide good indicators of the overall system performance that may be assumed for N_2^+ studies. By determination of their values, the efficiency of the lidar system operating at both visible and UV wavelengths may be determined.

3.2 Calculation of effective scattering cross section

 C_{eff} is mathematically calculated by the convolution of the absorption cross section, σ_{abs} , and the laser lineshape function, g_L . The experimentally determined values of C_{eff} (proportional to the ratio of the total Rayleigh lidar signal to the total resonance lidar signal (equation 3.8), determined experimentally in the manner discussed in the previous section) are fitted to the calculated theoretical values to determine the value of

 C_{eff} and laser linewidth for the resonance lidar system. The calculation and determination of C_{eff} is discussed separately for the Na and Fe resonance lidar systems, as it is calculated differently based on the spectroscopy of each element.

3.2.1 Sodium

Sodium has 11 electrons in the configuration $1s^22s^22p^63s^1$. There is one electron in the unfilled outermost energy level, and it is this electron which interacts with light and yields radiative transitions. The spectrum of sodium emission is easily derived due to its relatively simple atomic structure. The spectral line used in sodium resonance lidar arises from the transition from the 3p excited state to the 3s ground state. This transition is known as Na D and the excited state is split into two fine states, ${}^{2}P_{1/2}$ and ${}^{2}P_{3/2}$, by the magnetic interaction between the electron's spin and its orbital angular momentum (spinorbital coupling) [Atkins, 1991]. This fine structure gives rise to the D₁ and D₂ emission lines as the electron returns to the ground state at approximate wavelengths of 589.7558 nm and 589.1583 nm [Chu and Papen, 2005], respectively. The D₁ and D₂ emission lines are responsible for the dominant yellow color seen in sodium emission. These lines are further split into six hyperfine states due to the nuclear spin of 3/2. An energy level diagram showing the fine and hyperfine structure of sodium is given in Figure 3.1 (courtesy of Su [2007]) (here, f denotes the total angular momentum of the atom).

Following Su [2007], the absorption cross section for sodium is approximated as

$$\sigma_{abs}(v) = \frac{\sigma_0}{\sqrt{2\pi} \cdot \sigma_D} \left[S_a \exp\left(-\frac{(v - v_a)^2}{2\sigma_D^2}\right) + S_b \exp\left(-\frac{(v - v_b)^2}{2\sigma_D^2}\right) \right]$$
(3.10)

where

$$\sigma_0 = \frac{e^2 f}{4\varepsilon_0 m_e c} = \frac{A_{ul} \lambda_0^2 g_u}{8\pi g_1}$$
(3.11)

with e being the elementary charge $(1.602177 \times 10^{-19} \text{ C})$, f is the oscillator strength, ε_0 is the vacuum permittivity $(8.85410 \times 10^{-12} \text{ J}^{-1} \text{ C}^2 \text{m}^{-1})$, m_e is electron mass $(9.10939 \times 10^{-31} \text{ kg})$, A_{ul} is the Einstein coefficient, λ_0 (v₀=c/ λ_0) is the D₂ center wavelength, and g_u and g_l are the degeneracies of the upper and lower states, respectively. An electron will be in a

degenerate state if it has the same quantum characteristics as any other electron in the system. The Einstein A coefficient is the probability (rate, s⁻¹) that an electron in an upper energy level E_2 will decay spontaneously to a lower energy level E_1 , giving up a photon with energy $hv=E_2-E_1$.



Figure 3.1 Energy level diagram for atomic sodium. Figure courtesy of Su [2007].

The oscillator strength f is written as,

$$f = \frac{A_{ul}\varepsilon_0 m_e c\lambda_0^2 g_u}{2\pi e^2 g_1}$$
(3.12)

The Doppler width of the spectral line shape, σ_D , is due to random thermal motions of the atoms in the atmosphere with respect to the observer and is given by

$$\sigma_{\rm D}^2 = \frac{kT}{m_{\rm e}\lambda_0^2} \tag{3.13}$$

where k is the Boltzmann constant $(1.38066 \times 10^{-23} \text{ J K}^{-1})$ and T is again temperature (K). The intensity profile of a homogeneously Doppler broadened beam is a Gaussian, which often obscures the Lorentzian profile due to natural line broadening. Natural broadening

is due to the gradual decrease in the amplitude of electron oscillation within an atom or molecule, which gives rise to a distribution of oscillation frequencies [Dëmtroder, 2003]. At visible and UV frequencies the Doppler width exceeds the natural broadening by two orders of magnitude [Dëmtroder, 2003]. S_a and S_b denote line strengths and are given by

$$S_a = \frac{A_1 + A_2 + A_3}{A_{ul}}$$
, $S_b = \frac{A_4 + A_5 + A_6}{A_{ul}}$ (3.14)

where the spontaneous emission coefficients are those of the six hyperfine transitions within Na. v, v_a , and v_b are the frequencies of the laser (v=c/ λ) and the center frequencies of the two sets of hyperfine lines, respectively.

Having an expression for the absorption cross section, C_{eff} is yielded by convolving it with the laser line shape function g_L :

$$C_{\text{eff}}(\nu_{\text{L}}) = g_{\text{L}}(\nu_{\text{L}},\nu) * \sigma_{\text{abs}}(\nu) \cdot \Phi = \int_{0}^{\infty} g_{\text{L}}(\nu_{\text{L}},\nu) \sigma_{\text{abs}}(\nu) d\nu$$
(3.15)

where Φ denotes the branching ratio of 100% for this transition [Su, 2007]. The laser line shape is a Gaussian in the form

$$g_{L}(v) = \frac{1}{\sqrt{2\pi} \cdot \sigma_{L}} \left[\exp\left(\frac{(v - v_{L})^{2}}{2\sigma_{L}^{2}}\right) \right]$$
(3.16)

with linewidth σ_L and center laser frequency $\nu_L.$ Combining equations 3.10 and 3.16, C_{eff} is

$$C_{\rm eff}(v_{\rm L}) = \frac{\sigma_0}{\sqrt{2\pi} \cdot \sigma_{\rm eff}} \left[S_{\rm a} \exp\left(-\frac{(v_{\rm L} - v_{\rm a})^2}{2\sigma_{\rm eff}^2}\right) + S_{\rm b} \exp\left(-\frac{(v_{\rm L} - v_{\rm b})^2}{2\sigma_{\rm eff}^2}\right) \right]$$
(3.17)

with effective width, σ_{eff} , of

$$\sigma_{\rm eff} = \sqrt{\sigma_{\rm L}^2 + \sigma_{\rm D}^2} \tag{3.18}$$

The linewidth of the laser is expressed as the full-width at half maximum (FWHM), Δv , and is related to the rms width, σ , as follows, [Su, 2007]

$$\Delta v = 2\sqrt{2\ln 2} \cdot \sigma \tag{3.19}$$

3.2.2 Iron

Iron has 26 electrons in the configuration $1s^22s^22p^63s^23p^63d^64s^2$. The greater number of valance electrons in iron results in a spectrum far more complex than that of sodium. It is so complex that it is often determined experimentally rather than by quantum mechanical calculations. The transition used for iron resonance lidar occurs between the iron ground state a^5D and the iron excited state z^5F_0 [Fuhr et al., 1988]. The wavelength corresponding to this transition is in the near UV range of wavelengths at ~372 nm. A partial energy level diagram is given in Figure 3.2 (figure courtesy of Su [2007], referencing Fuhr et al. [1988]). Iron has many strong UV transitions, but the light of these wavelengths is absorbed in the stratospheric ozone layer, and are therefore poor choices for lidar studies.

Following Su [2007], Ceff for iron is expressed simply as

$$C_{\rm eff}(v_{\rm L}) = \frac{\sigma_0}{\sqrt{2\pi} \cdot \sigma_{\rm eff}} \exp\left(-\frac{(v_{\rm L} - v_0)^2}{2\sigma_{\rm eff}^2}\right)$$
(3.20)

obtained from the convolution of the laser lineshape function and the iron absorption cross section σ_{abs} for the 372 nm transition,

$$\sigma_{abs}(v) = \frac{\sigma_0}{\sqrt{2\pi \cdot \sigma_D}} \left[exp\left(-\frac{(v - v_0)^2}{2\sigma_D^2} \right) \right]$$
(3.21)

The expression for C_{eff} of iron is simpler than the expression obtained for sodium because the 372 nm transition is a singlet rather than a doublet.

3.3 Determination of system efficiency following realignment of cavity optics

In November 2007, a laser technician and researchers at the Lidar Research Laboratory realigned the laser cavity optics in the excimer-pumped dye laser system. Prior to servicing, the excimer laser beam energy was 240 mJ (measured by an internal photodiode) with the high voltage discharge set at 18 kV. At 20 kV, the energy was measured to be 306 mJ. After replacement, cleaning, and aligning of excimer cavity optics, the energy was increased to 430 mJ at 20 kV. The energy of a Rhodamine 6G dye laser beam (used for sodium resonance lidar studies) produced by optical pumping from



the excimer beam was 27 mJ when operated at a wavelength of 581 nm, giving a lasing efficiency of $\sim 8\%$.

Figure 3.2 Energy level diagram for atomic iron. Figure courtesy of Su [2007].

To confirm the improved performance of the laser system, resonance lidar data was taken of the mesospheric sodium and iron layers and compared to data that preceded the realignment of cavity optics. Data for determination of the value of C_{eff} for sodium was obtained on the night of January 26, 2008. Consecutive measurements (sets 1/1-15/8) were taken at laser wavelengths from 589.0329 nm to 589.0419 nm. The total integrated signal profile for sets 16/1-21/8 is plotted in Figure 3.3. All times are Local Standard Times (LST is equivalent to Universal Time (UT) minus 9 hours).

The maximum total resonance signal occurs at an altitude of 86.1 km. The lidar signal measured at 589.038 nm and 589.0413 nm are compared in Figure 3.4 to highlight the variation in signal strength with wavelength. The maximum resonance signal decreases by a factor of 3.67 between these two measurements, separated by 0.33 pm.

The decrease in signal is due to decreased interaction between the sodium atoms and the emitted laser photons at that operational wavelength.

The experimentally determined value of C_{eff} as a function of laser wavelength for the night of January 26, 2008 appears in Figure 3.5, along with the expected values (equation 3.17). The value of the laser linewidth is 4.7 pm (FWHM) and the maximum value of the effective scattering cross section is 3.42×10^{-16} m². Signals less than 25% of the maximum value are not used in the determination of C_{eff} . These values are comparable to the measurements of Su [2007], which yielded a linewidth of 4.5 pm and a C_{eff} of 3.50×10^{-16} m². The effective scattering cross section is greater for a laser of narrower linewidth.

The sodium concentration profile determined from the data displayed in Figure 3.3 and the maximum C_{eff} in Figure 3.5 is plotted in Figure 3.6 (the density is determined by equation 3.7). The peak of the sodium layer on this night occurs at 86.1 km (corresponding to the height of the maximum total resonance signal). The raw data is smoothed with a 2 km running average and the average peak sodium density from 85-87 km is 6389 cm⁻³.

The determined values of the effective scattering cross section (C_{eff}) and the density of scatterers ($\rho(z)$) are combined with the operating specifications of the resonance lidar system and the measured lidar signal to yield a value for the efficiency of the sodium lidar system by means of equation 3.9. The values used in this calculation are tabulated in Table 3.1. A 2 km (Δz) altitude range (z_{range}) is considered around the peak density of the mesospheric sodium layer. The altitude at which the peak density occurs is z. The integrated resonance photon counts from this range is determined ($N_s(z_{range})$). The average background signal (\tilde{N}_B) has been scaled for the number of bins (data points) considered in the integrated background signal versus the integrated resonance signal.



Figure 3.3 Sodium resonance lidar signal profile on the night of January 26, 2008.



Figure 3.4 Sodium resonance signal profiles. (top) Measurement with laser wavelength at 589.0380 nm; (bottom) Measurement with laser wavelength at 589.0413 nm.



Figure 3.5 Measurement of sodium resonance lidar $C_{\rm eff}$ as a function of laser wavelength on January 26, 2008.



Figure 3.6 Sodium lidar concentration profile measured on January 26, 2008, smoothed over 2 km.

Date	26 January 2008
Integrated signal 85-87 km (N _s (z _{range}))	160629 photon counts
Average signal 120-125 km (\tilde{N}_B)	2740 photon counts
Energy per laser pulse (E _L)	19.6 mJ
Repetition rate of laser (R _L)	10 Hz
Time span over which data was obtained (Δt)	5040 s
Laser shots	44000
Average laser operational wavelength (λ_L)	589.0382 nm
Average peak sodium density 85-87 km ($\rho(z)$)	6388.26 cm ⁻³
Effective scattering cross section (Ceff)	$3.42 \times 10^{-16} \text{ m}^2$
Spatial range (Δz)	2 km
Area of telescope receiver (A_R)	0.28 m^2
Scattering height (z)	86.1 km
Integrated signal 20-25 km)	1631090
Integrated signal (30-35 km)	176763

Table 3.1 Values used to determine the value of the sodium resonance lidar system efficiency onJanuary 26, 2008.

The value of ηT^2 is determined to be 0.41%, which is lower than the 1% expected for this type of system. A similar analysis was conducted by Su [2007] for sodium data obtained on April 7, 2007. The data referenced by Su [2007] leads to a value of 0.17% for ηT^2 , a factor of 2.41 times lower than the efficiency of the system after the realignment of the laser optics and cavity.

Iron resonance data for determination of the value of C_{eff} was obtained on February 8, 2008. Consecutive measurements were taken at laser wavelengths from 372.0181 nm to 372.0201 nm (sets 9/1-16/4). The total integrated signal profile for sets 19/1-23/8 is plotted in Figure 3.7. The comparatively low resonance signal is due to a decrease in system performance and atmospheric transmission at UV wavelengths versus the visible wavelengths used in sodium resonance measurements. The effective scattering cross section of iron is also much smaller than the sodium cross section. The maximum total resonance signal occurs at an altitude of 84.6 km.

The value of the laser linewidth for the iron resonance lidar system is 1.9 pm and the maximum value of C_{eff} is 23.9x10⁻¹⁸ m² [Figure 3.8]. Signals less than 25% of the maximum value are not used in the determination of C_{eff} . Su [2007] measured a linewidth of 1.1 pm and a C_{eff} of 4x10⁻¹⁷ m² on March 14, 2005. The value of ηT^2 is not calculated for the iron resonance lidar system.

Iron resonance data for determination of the value of C_{eff} was also taken on March 1, 2008. Consecutive measurements were taken at laser wavelengths from 372.0234 nm to 372.0267 nm (sets 2/1-13/4). The total integrated signal profile for sets 14/1-25/8 is plotted in Figure 3.9. The maximum total resonance signal occurs at an altitude of 85.275 km. The value of the laser linewidth for the iron resonance lidar system is 1.7 pm and the maximum value of C_{eff} is 26.7x10⁻¹⁸ m² [Figure 3.10]. Signals less than 25% of the maximum value are not used in the determination of C_{eff} .



Figure 3.7 Iron resonance lidar signal profile on February 8, 2008.



Figure 3.8 Measurement of iron resonance lidar C_{eff} as a function of laser wavelength on February 8, 2008.



Figure 3.9 Iron resonance signal profile on March 1, 2008.



Figure 3.10 Measurement of iron resonance lidar C_{eff} as a function of laser wavelength on March 1, 2008.

Based on the calculated value of the efficiency term for the sodium resonance lidar system, I conclude that the resonance lidar system's performance has been increased for studies at visible wavelengths (sodium) by realigning the laser cavity optics. Signal levels at UV wavelengths appear to be comparable to pre-alignment performance documented by Su, with an increase in the width of the laser line.

3.4 Performance of 1.02 m telescope

In October of 2008, a Cassegrain telescope with a 1.02 m diameter primary mirror was integrated into the resonance lidar receiver system. Data prior to this date was obtained with a 0.6 m Newtonian telescope. By increasing the area of the telescope (in this case by a factor of 2.89), the measured lidar signal should increase by a similar factor given consistency in other lidar operational parameters (equation 3.2). The primary and secondary mirrors in the telescope were aligned and the focusing capabilities of the telescope examined in the fall of 2007.

To align the primary mirror so it is parallel to secondary mirror (confirmed to be horizontal), a HeNe (helium neon) laser beam was optically split into three beams with horizontal paths (normal to the secondary mirror face) directed at the primary mirror from above [Figure 3.11, upper left]. The paths were determined to be horizontal by reflecting them off of an un-perturbed liquid surface (in this case, olive oil) and then aligning the transmitted and reflected beams so that they overlapped [Figure 3.11, upper right]. The focused image of the three beams was then examined at the focal plane of the telescope [Figure 3.11, lower right]. The primary mirror was then shifted until proper focus was achieved [Figure 3.11, lower right], indicative of proper mirror alignment.

To verify the increase in the received resonance lidar signal due to the increase in the area of the receiving telescope, data obtained with the 1.02 m and 0.6 m telescope receivers is compared. Sodium resonance lidar obtained using the 1.02 m telescope on the night of February 17, 2009, is analyzed for means of comparison [Figure 3.12] (similar data was obtained on the nights of February 1, 2, 12, and 23, 2009; the February 17th data is chosen because measurements for determination of C_{eff} were obtained on that night). The sodium resonance lidar system was operating at this time in support of a rocket mission flown from PFRR. The excimer high voltage was 18 kV with beam energy of 348 mJ per pulse and the pulse rate was 10 Hz. The signal was blanked for 100 μs. The signal (photon counts per laser pulse) was relatively constant in the 20-25 km, 30-35 km, and 40-45 km altitude ranges as slight adjustments to the operational wavelength were made through the course of the night [Figure 3.13]. The signal is examined over these altitude ranges because the signal is due to Rayleigh scattering. The lower atmosphere is considered a relatively constant target, unlike the mesospheric metal layers, so any variations in signal should be due to system performance rather than variations in resonance scattering. Determination of the signals in these altitude ranges allows comparison of the resonance lidar system operating in different modes. The average signals from each height range on February 17, 2009, were compared with the data obtained on January 26, 2008, with the 0.6 m telescope [Table 3.2]. On January 26, 2008, the excimer energy was 363 mJ with the high voltage at 18 kV. The pulse rate and the blanking had the same settings as on February 17, 2009.



Figure 3.11 Aligning a 1.02 m telescope for use in the lidar receiver system.



Figure 3.12 Sodium resonance lidar signal profile on February 17, 2009.



Figure 3.13 Rayleigh signal from 20-25 km, 30-35 km, and 40-45 km for Na resonance lidar system on February 17, 2009.

Table 3.2 Average signal in three height ranges for sodium resonance lidar system operating with1.02 m (February 17, 2009) and 0.6 m receivers (January 26, 2008).

	Avg. signal w/	Avg. signal w/
Height range (km)	1.02 m telescope	0.6 m telescope
20-25	89.2	36.8
30-35	8.8	3.9

It is found that the average signal in the 20-25 km height range increases by a factor of 2.4 when the 1.02 m telescope is in the receiver system. The average signals in the 30-35 km range increases by factors of 2.3.

Iron resonance lidar data was obtained using the 1.02 m telescope on the night of October 28, 2008 [Figure 3.14]. The excimer high voltage was 20 kV with beam energy of 426 mJ per pulse and the pulse rate was 10 Hz. The blanking was set at 100 μ s. The signal in the three altitude ranges considered was nearly constant, as was the operational

wavelength [Figure 3.15]. The average signals from each height range were compared with the iron resonance data taken on February 8, 2008 with the 0.6 m telescope [Table 3.3]. There is a consistent increase by a factor of \sim 1.3 between the 20-25 km and 30-35 km signals in the two receiver systems.

For both sodium and iron resonance lidar data acquisition, the 1.02 m telescope results in an increase in signal. At visible wavelengths, the signal from 20-25 and 30-35 km has been more than doubled. However, it is not found that the signal has increased by a factor comparable to the 2.89 times increase in the area of the receiver. The lack of direct proportionality between the increase in lidar signal and the increase in the area of the receiving telescope's primary mirror is likely due to inconsistency in other lidar operational parameters between the two nights. For UV wavelengths, the signal has been increased by only 1.3 times in the 20-25 and 30-35 km height ranges.



Figure 3.14 Iron resonance lidar profile on October 28, 2008.



Figure 3.15 Rayleigh signal from 20-25 km, 30-35 km, and 40-45 km for Fe resonance lidar system on October 28, 2009.

	Avg. signal w/	Avg. signal w/
Height range (km)	1.02 m telescope	0.6 m telescope
20-25	20.5	16.5
30-35	2.16	1.68
40-45	0.26	0.24

Table 3.3 Average signal in three height ranges for iron resonance lidar system operating with1.02 m (October 28, 2008) and 0.6 m (February 8, 2008) receivers.

The efficiency of the sodium resonance lidar system operating with the 1.02 m telescope receiver is examined through the night of February 17, 2009, as this night resulted in an unusually long data set (totaling ~10 hours). The lidar system was operating that night in support of a rocket launch flown from Poker Flat Research Range (PFRR); a data set spanning the night was required for the research purposes of the launch team. The laser linewidth and C_{eff} determined for this night are plotted in Figure 3.16. The values for each term used to determine the system efficiency (equation 3.9) for

each set of ten profiles for sets 1-70 (eight profiles per set) are tabulated in Table 3.4, along with the determined value of ηT^2 for each period. E_L, R_L, C_{eff}, Δz , and A_R are considered constant through the night, being 11.2 mJ, 10 s⁻¹, 3.082x10⁻¹⁶ m², 2 km, and 0.817 m², respectively.



Figure 3.16 Measurement of sodium resonance lidar linewidth and C_{eff} on February 17, 2009.

Set range	1-10	11-20	21-30	31-40	41-50
N _s (z _{range})	169206	191851	208267	173065	220837
$z_{range}(km)$	87.6-89.6	86.25-88.25	86.5-88.5	88.3-90.3	83.8-85.8
\widetilde{N}_{B}	1500	1359	1246	1151	1159
$\Delta t(s)$	4080	4500	4080	4080	4320
$\lambda_L(nm)$	589.0455	589.0455	589.0458	589.0458	589.0445
$\rho(z_{range})(cm^{-3})$	3863.75	3986.98	4738.47	3997.29	4468.35
z (km)	88.6	87.25	87.5	89.3	84.8
$N_{20\text{-}25\ km}$	3520340	3698540	3449030	3582030	3655720
N _{30-35 km}	345659	363284	340675	352633	361291
ηT^2	0.63%	0.62%	0.61%	0.63%	0.62%

Table 3.4 Values of lidar system parameters used to calculate system efficiency on February 17,2009.

0.4	51 (0	(1.70
Set range	51-60	61-70
$N_s(z_{range})$	275793	246217
$z_{range}(km)$	84.15-86.15	85.2-87.2
\widetilde{N}_{B}	1141	1167
Δt (s)	4080	4740
$\lambda_L(nm)$	589.0445	589.0446
$\rho(z_{range})(cm^{-3})$	5451.57	5280.63
z (km)	85.15	86.20
N _{20-25 km}	3699970	3520620
N _{30-35 km}	365055	344654
ηT^2	0.67%	0.55%

The efficiency of the lidar system is approximately constant over the 8 hours and 18 minutes of data considered in this analysis, with an average system efficiency of

0.62%. This value of efficiency is 1.5 times the efficiency of the sodium resonance lidar system on January 26, 2008.

From the lidar equation (equation 3.2), the efficiency of the receiver is independent of the operational parameters of the laser and the characteristics of the sodium layer. The simultaneous Rayleigh lidar measurements on both nights show similar signal levels, and thus this increase in the efficiency represents an improvement in the receiver efficiency and not in the atmospheric transmission. The improvement reflects the optical quality of the two telescopes. The 0.6 m telescope is a Newtonian telescope built in-house (at the Lidar Research Laboratory) by NICT in 1997. The 1.02 m Cassegrain telescope is an astronomical quality telescope, whose optics were recoated in 2002 just before it was installed at the Lidar Research Laboratory in 2003. The Newtonian telescope is a dual channel system that supports simultaneous Rayleigh and resonance lidar measurements. The receiver separates these signals (at 532 nm and 589 nm, respectively) by employing dichroic beam-splitters [Su, 2007]. The Cassegrain telescope is configured as a single channel receiver and does not include the optical losses associated with the dichroic beamsplitters. In general, Cassegrain telescopes are fundamentally better aligned and are more stable than Newtonian telescopes and yield higher optical transmission [e.g., Schroeder, 2000]. Furthermore, as discussed earlier in this chapter, the Cassegrain was carefully aligned in 2007 at the beginning of these studies. Thus, we conclude that the improvement in the efficiency of the receiver is due to the higher optical quality of the Cassegrain telescope.

3.5 Tuning accuracy

To confirm the relative accuracy of the laser tuning system, the wavelength of transmitted laser light was compared to a known spectral reference. The spectral line produced by a Rhodamine 6G dye laser beam operating at 589 nm (used in Na resonance lidar) was compared to the spectrum of a sodium discharge lamp. Light from both sources was diffracted by a Mark V Half-Meter Ebert Scanning Spectrometer. The light was detected by a photodiode and recorded in time by a high resolution digital oscilloscope. The resolution of the data was 0.4 μ s (~12.7 pm). The unsmoothed data is



shown in Figure 3.17, where the three signal peaks are due to the sodium D_2 and D_1 emission and the laser light.

Figure 3.17 Superimposed spectra of Rhodamine 6G dye laser tuned to 589 nm and the sodium D_2 and D_1 emission lines.

The data was smoothed over 200 data points [Figure 3.18]. The laser peak occurs at 4.513 ms and the centroid in time of the laser peak is 4.643 ms. The centroid in time is determined for points with signal amplitudes greater than one fourth (25%) of the peak wavelength. The centroid is given by

$$t_{centroid} = \frac{\sum_{i=1}^{n} ((Signal) \cdot (Time))}{\sum_{i=1}^{n} (Signal)}$$
(3.22)

By using only the upper 75% of the peak, the centroid value is not skewed by fitting to values in the wings of the spectral shape. The D_2 line peaks at 4.117 ms, and the D_1 line peaks at 7.807 ms.



Figure 3.18 Same as Figure 3.17, but smoothed over 200 data points.

To match the spectral peaks in time to wavelengths, the spectrum of the Rhodamine 6G dye laser beam tuned to 589 nm was recorded with an Ocean Optics (OO) USB2000 Spectrometer with a resolution of 0.38 nm. The groove density of the diffraction grating used in the OO USB2000 is 600 mm⁻¹ (2048 pixels measuring wavelengths from 177.34 nm to 879.07 nm). The efficiency of the spectrometer grating is not constant with wavelength, being ~51% at 300 nm, ~70% at 400 nm, ~60% at 500 nm, and ~49% at 600 nm [Ocean Optics, 2009].

To confirm the performance of the spectrometer used in this study, a spectrum was taken for a sodium discharge lamp [Figure 3.19] and a mercury discharge lamp [Figure 3.20; The line at ~546 nm has saturated the detector at 4000 intensity units- this spectra is shown to highlight the spectral features of all the lines]. The USB2000 spectrometer does not resolve the D_2 and D_1 Na emission peaks separately when measuring the light from the sodium discharge lamp. The spectrum is averaged over 300 ms and the peak intensity occurs at a wavelength of 589.12 nm. The Na D_2 emission line has a reported vacuum wavelength of 589.1583 nm.

Seven of mercury's many spectral lines are captured by the OO USB2000 spectrometer, with two sets of closely spaced spectral lines resolved [Figure 3.20]. The measured wavelengths of these lines (centroids of non-saturated spectral lines) are compared to the reported wavelengths for these mercury lines [Sansonetti, 1996] in Table 3.5. In general, the measured wavelengths agree well with the reference data and the spectrometer measurements are assumed to yield relatively accurate wavelengths. Furthermore, the relative intensities of the two spectral lines measured at \sim 576 nm and \sim 579 nm agree well with the spectral reference data [Sansonetti, 1996].



Figure 3.19 Sodium lamp spectrum acquired with Ocean Optics USB2000 spectrometer.



Figure 3.20 Mercury lamp spectrum acquired with Ocean Optics USB2000 spectrometer.



Figure 3.21 Mercury emission doublets at 404.66 nm and 407.78 nm (right) and 576.96 nm and 579.07 nm (left).

Wavelength in air	Measured
(nm)	wavelength (nm)
365.0158	365.015
404.6565	404.52
407.7837	407.456
435.8335	435.597
546.0750	546.082
576.9610	576.883
579.0670	579.067

Table 3.5 Comparison of wavelengths in air (nm) for prominent spectral lines in mercury [Sansonetti,1996] and wavelengths measured with OO USB2000 Spectrometer.

The spectra obtained with the OO USB2000 spectrometer of the Rhodamine 6G dye laser beam tuned to 589 nm is shown in Figure 3.22. The OO USB2000 spectrometer data is much lower resolution than the data obtained from the Half-Meter Ebert Scanning Spectrometer (~30 times lower). The peak intensity of the spectrum occurs at 588.780 nm, but, due to the comparatively low resolution of the data, a centroid is again a more accurate representation of the actual spectral peak. The value of the centroid wavelength varies by almost a tenth of a nanometer depending upon the choice of how much of the peak to consider; we take $\lambda_{centroid}$ =588.843 nm, determined using only those data points of intensity greater than one third (33%) of the measured peak intensity. The calculation of $\lambda_{centroid}$ is similar to equation 3.22 with wavelength λ replacing time.

We match this centroid in wavelength to the centroid in time of the laser peak in the Half-Meter Ebert Scanning Spectrometer data. Based on this one matching point and the known separation of the D₂ and D₁ emission lines, we are able to assign wavelengths to all data points in time [Figure 3.23]. In this data, the D₂ emission line occurs at 588.7579 nm and the D₁ emission line occurs at 589.3549 nm, with $\Delta\lambda$ =0.5970 nm. The reported vacuum wavelengths of the D₂ and D₁ lines are 589.1583 nm and 589.7558 nm, respectively, with $\Delta\lambda$ =0.5975 nm [National Institute of Standards and Technology (NIST), 2008]. The vacuum wavelengths λ_{vac} become λ_{STP} when refracted at standard temperature (15°C) and pressure (1013.25 hPa). The two are related by

$$\lambda_{\rm STP} = \frac{\lambda_{\rm vac}}{n_{\rm air}} \tag{3.23}$$

[e. g. Halliday et al., 2001]. The index of refraction, n, of dry air is calculated from the expression

$$(n-1) \cdot 10^{-8} = 8342.54 + 2406147(130 - \sigma^2)^{-1} + 15998(38.9 - \sigma^2)^{-1}$$
(3.24)

where $\sigma = \lambda_{\text{vac.}}^{-1}$ and λ_{vac} has units of μ m [CRC Handbook of Chemical and Physical Reference Data, 2006]. Therefore, the D₂ and D₁ lines occur at wavelengths of 588.9951 nm and 589.5924 nm, respectively, with $\Delta\lambda$ =0.5973 nm, when refracted under standard conditions. There is a 0.237 nm (~1%) disagreement between these wavelengths and the wavelengths assigned to the sodium D doublet by this method of centroid matching.

Based on this analysis, we conclude that the Scanmate2® dye laser tuning is not absolutely accurate, but reasonably accurate given uncertainties in temperature, pressure, and limitations in measurement methods. The tuning system is operating as designed.



Figure 3.22 Spectrum of Rhodamine 6G dye laser operating at 589 nm.


Figure 3.23 Overlapping spectra of Rhodamine 6G dye laser tuned to 589 nm and the sodium D₂ and D₁ emission lines, centroid in time matched to centroid in wavelength.

3.6 Diffraction grating orders

As previously discussed, Collins et al. [1997] determined that, for N_2^+ studies, the resonance fluorescence lidar optimum operational wavelength would be ~390 nm. The excited dye molecules in the dye laser system fluoresce over a range of wavelengths; The precise wavelength of the output laser light is selected by a Littrow mounted diffraction grating. In this configuration, the incident light is normal to the groove face. The spectrum is scanned by rotating the grating, thereby reorienting the grating normal so the incident angle and diffraction angle change (even though they are equal for all wavelengths) [Pedrotti and Pedrotti, 1993]. The desired wavelength of operation is input by the laser operator into the laser control computer, which sets the pitch of the diffraction grating is 0.14 pm at 589 nm and 0.089 pm at 372 nm and 390 nm [Hou, 2002; Peshave, 2004]. The wavelength of the light varies for any wavelength due to temperature and pressure variations within the room where the laser is operated [Peshave, 2004]. As a

result, the laser must be stepped over several pm to find the operational wavelength that will cause fluorescence in the target atoms or molecules.

The order of the grating is determined by the pitch angle of the grating relative to the optical axis of the laser oscillator (discussed in section 4.1) [Su, 2007]. The diffraction grating transmits light of wavelengths 1000 nm to 300 nm over five gratings orders [Table 3.6]. 390 nm light is near the edge of the eighth order. Light of wavelengths greater than 396 nm is output in seventh order [Table 3.7]. The 390 nm light necessary for N_2^+ studies is therefore close to a change in grating order. The 372 nm light necessary for Fe studies and the 589 nm light for Na studies are found in the middle of orders eight and five, respectively. To determine what effect, if any, the placement on the grating has on the 390 nm laser beam, the variation of grating orders with wavelength was examined.

Wavelength, λ (nm)	Grating order
1000	3
900	3
800	3
700	4
600	5
500	6
400	7
300	8

 Table 3.6 Grating order corresponding to tuning wavelength.

Table 3.7 Wavelengths at which transitions between orders occurs.

Transition between	Wavelength at which		
orders:	transition occurs (nm)		
3 to 4	792		
4 to 5	634		
5 to 6	529		
6 to 7	452		
7 to 8	396		

The grating equation for a diffraction grating in a Littrow mount configuration is $m\lambda = 2a \sin \theta_{\rm b}$ (3.24)

[e.g. Pedrotti and Pedrotti, 1993] where m is the grating order, a is the groove spacing, and θ_b is the blaze angle of the grating which is equal to θ_i (the angle of the incident light) in the Littrow mount. Therefore, the angle of the refracted light in the mth order is θ_m =- θ_i . To calculate the wavelength λ_a light in one order, m_a , would have in another order, m_b , take the ratio of grating equations and solve for λ_b , obtaining

$$\lambda_{a} \cdot \frac{m_{a}}{m_{b}} = \lambda_{b} \tag{3.25}$$

Therefore, to output 390 nm light in order seven instead of eight, the laser would be tuned to a wavelength of 446 nm, and so forth. To confirm that the input tuning wavelength may be varied to output light in different grating orders without affecting the true wavelength of the light, Rhodamine 6G dye laser light was examined at 589 nm where the energy output of this dye is greatest. Spectra were obtained with the OO USB2000 Spectrometer at input tuning wavelengths of 589 nm (order 5), 736 nm (589 nm light in order 4), and 491 nm (589 nm light in order 6) [Figure 3.24]. All three spectral peaks occur at 589 nm, confirming that the light retains its true wavelength across orders.

To examine the energy output of the laser beam across grating orders, the QUI dye laser was tuned in 5 nm steps from 360 nm to 455 nm, across grating orders eight and seven. The energy of the output beam was measured at each step with a Molectron Energy Meter [Figure 3.25]. Energy is measured in mV and converted to mJ (16.4 J V⁻¹). The reported fluorescent range of QUI dye (used for Fe resonance lidar studies) is 368 nm to 402 nm [Brackmann, 1997]. The repetition of dye spectra across grating orders is also observed for other laser dyes. The energies measured at each wavelength in order 8 and their corresponding wavelengths in order 7 are comparable. An increase in output energy is observed at wavelengths immediately following a change in grating order. The reported characteristics of the QUI dye do not account for the increased energy measured at 405 nm. An increase in energy following a change in grating order is observed for all

dyes examined in the dye laser system (a total of ten) and is therefore not a feature unique to QUI [Appendix A], but a characteristic of the output of the grating.



Figure 3.24 Spectra of Rhodamine 6G dye laser beam at input wavelengths of 589 nm (order 5), 736 nm (order 4), and 491 nm (order 6).

The output laser light with wavelength for QUI dye was also measured with the USB2000 spectrometer. When the QUI dye laser is tuned to 360 nm and 365 nm, where the gain of the dye is low, the spectral peak is observed at wavelengths of 411.06 nm and 416.85 nm, respectively [Figure 3.26]. The dye laser grating transmits 360 nm light in the eighth order, according to the Scanmate2® tuning program, but 360 nm is 411 nm light in the seventh order (similarly, 365 nm light in the eighth order is 416 nm in the seventh, by equation 3.25). This behavior is an optical occurrence we only see when operating at wavelengths where the dye emission is weak and should not affect resonance lidar system performance.



Figure 3.25 QUI dye laser energy output with wavelength in grating orders seven and eight.

Clearly, if 390 nm light is on the edge of a grating order, it will be on the edge of any other grating order. Therefore, any effect the placement of the wavelength within an order has on the output light cannot be circumvented by operating in a different grating order. Furthermore, light of any given wavelength produced by any laser dye will occur at the same place within each grating order, so any effects due to the grating will be uniform for all dyes. The energy of the output light is also not increased by varying the grating order, so there is no discernible advantage in operating in other grating orders.



Figure 3.26 Spectra of QUI dye laser operating at 360 nm.

4 Analysis of Laser Dyes

In April 2008, following the careful realignment and verification of the performance of the resonance lidar system, a sky test similar to that of Su [2007] was conducted with a QUI dye laser beam. The transmitted laser light was tuned from 372 nm to 390 nm and a similar drop in signal at 390 nm was observed [Figure 4.1]. The signal in the 20-25 km and 30-35 km altitude ranges is again examined because this signal comes from Rayleigh scattering of the laser light by the atmosphere. The atmosphere is considered a constant target, so any variations in the received signal should be due to the lidar system performance. Such an analysis allows comparison between different operational modes of the resonance lidar system (Fe versus Na, etc.). The NICT Rayleigh lidar system was operated simultaneously, yielding data indicating that the sky conditions remained stable during the time of this test, confirming that a drop in atmospheric transmission (due to clouds overhead) was not the cause of the decrease in signal at 390 nm.



Figure 4.1 Rayleigh signal from 20-25 km and 30-35 km altitude ranges for the QUI dye laser on April 23, 2008.

Su [2007] had postulated that operating the dye laser at 390 nm, at the peak of the dye gain, could cause a significant amount of amplified spontaneous emission, decreasing the amount of coherent laser light. To investigate the ratio of stimulated emission to spontaneous emission in the QUI dye laser beam, measurements of the output laser light were taken with the OO USB2000 spectrometer. For comparison, spontaneous emission within Rhodamine 6G dye was also examined. A total of nine dyes, not including QUI, were tested in the dye laser system to assess their potential for obtaining N_2^+ resonance lidar measurements. One of the nine dyes (Exalite 428) was selected as a likely candidate for obtaining such measurements. Before these results and others are discussed, some details of dye lasers and laser dyes are given.

4.1 Dye lasers and laser dyes

An LPX220i® excimer laser is used as the pump source for a Scanmate2® pulsed dye laser in the resonance lidar system at PFRR. Dye lasers are attractive and unusually flexible tools because they are easily tunable over a wide range of frequencies and wavelengths [Schäfer, 1990; Hecht, 1992]. Dye lasers are capable of producing ultra-short pulses of smaller half-width than other lasers due to the broad spectral bandwidth produced by the pumped dye molecules [Hecht, 1992]. The concentration of dye molecules (and therefore their absorption and gain) is easily controlled. Cooling is also easily managed by simply circulating the liquid dye. The cost of the organic dyes is also negligibly small compared to that of solid state lasers, many of which are similarly tunable.

There are, however, several disadvantages in the use of dye lasers. Their versatility comes at a cost in complexity [Hecht, 1992]. The emission range of any dye is narrow compared with the entire spectrum and the dye must be changed several times to tune across the entire visible spectrum. Different laser dyes, fluorescing over different spectral regions, are used for sodium and iron resonance lidar studies. Use of one dye for both iron and N_2^+ studies would have been serendipitous. The dyes are suspended in a liquid solvent, many of which are carcinogenic in nature, and require a complex liquid

handling system. The dye molecules can experience thermal and photochemical degradation when excited by the pump source and must be replaced over time.

Laser dyes are complex organic compounds (hydrocarbons and their derivatives) with an extended system of conjugated bonds, i.e. alternating single and double bonds [Schäfer, 1990]. Due to this internal ring structure, dye molecules readily absorb and emit light over a range of wavelengths as interactions among electronic, vibrational, and rotational energy levels create a continuum of levels [Hecht, 1992]. Absorption of a photon from the excimer laser beam raises an electron in a dye molecule to a highly excited state. Within picoseconds, non-radiative processes drop it to the upper level of the laser transition (the lowest vibronic level [Drexhage, 1990]) where the electron undergoes stimulated emission. Stimulated emission occurs when a photon perturbs an electron in an upper energy state, causing it to drop to a lower energy state and emit a photon with the same energy as the first. The energy of the photons causing stimulated emission varies depending on the type of emitter. The emitted photon has the same characteristics of the first (frequency, polarization, phase, and direction). After the laser light is emitted, non-radiative processes remove electrons from the lower level of the laser transition.

Stimulated emission occurs when the excited electron within the dye molecule is in what is known as a singlet state; in a μ s or less, the electron will move to a long-lived triplet state and begin to absorb the lasing wavelength. This non-radiative process can effectively compete with the light emission and reduce the fluorescent efficiency of the dye [Drexhage, 1990]. Pump pulses must be short in duration to limit the amount of triplet state absorption, which limits the gain in the dye (and therefore the laser energy). Pump pulses in the 10 ns range or shorter, such as those of an excimer beam, are too short to produce triplet state absorption [Hecht, 1992]. The gain in the dye can also be limited by amplified spontaneous emission.

Amplified Spontaneous Emission (ASE) or super-luminescence, is spontaneous emission that has been amplified by stimulated emission. Spontaneous emission is when an electron moves from a higher level to a lower level, unbidden. ASE is produced in the dye when it is pumped to produce a population inversion. When the excited electrons spontaneously decay, they may cause incoherent stimulated emission along their direction of propagation. Only coherent laser radiation is useful in the dye laser system, and the amount of it that may occur in the dye is decreased by the occurrence of incoherent ASE. Spontaneous emission can occur at a wide range of wavelengths in the lower level of the lasing transition, with the highest probability of emission at central wavelengths [Hecht, 1992]. Amplification is strongest at the peak of the laser gain curve, where the difference between emission and absorption is greatest [Hecht, 1992].

Complex optics are required to produce the ultra-narrow linewidth pulses of a dye laser. Dye lasers pumped by pulsed lasers are often operated in an oscillator-amplifier configuration. The excimer pumped dye laser at PFRR employs this common configuration (see Hecht [1992] for a more detailed discussion). The oscillator stage in the Scanmate2[®] dye laser is briefly discussed to aid in future understanding.

The oscillator stage consists of a dye cell, beam expanding optics, a tuning grating, output mirrors, and the pump source (in this case, the excimer laser beam). A very simplified diagram of the oscillator, modeled after the diagrams of Hou [2002] and the Lambda Physik [1997] technical diagrams, is given in Figure 4.2. (Note that the actual optical system used in the Scanmate2[®] dye laser is far more complex and proprietary to the manufacturer.)



Figure 4.2 Simplified diagram of Scanmate2 laser oscillator.

A single cuvette, through which dye is pumped, acts as both the pre-amplifier and oscillator stages. The pump beam (the excimer laser beam) is split into two beams to provide two regions of excited dye in the cuvette. To limit the amount of ASE, the oscillator pump beam is slightly skewed relative to the oscillator axis. Spontaneous emission therefore occurs off-axis and should not be amplified by being reflected by the end mirror back through the gain medium. The ASE beam should miss the turning mirror and not be directed through the preamplifier. The oscillator beam is expanded by a prism to yield higher wavelength sensitivity and decrease the energy density, thereby preventing burning of optics, before it is incident on the Littrow mounted diffraction grating. The output wavelength of light is selected by rotating the diffraction grating. This grating serves as one of the lasing cavity end mirrors. A totally reflective mirror acts as the other end mirror. The desired light that is reflected by the diffraction grating must be amplified to yield the amount of coherent radiation desired. The laser beam exits the oscillator stage when it is reflected off of the prism and directed by the turning mirror to the preamplifier stage and on to the amplifier. The beam path of one of the pump beams is chosen to lie on the same path as the pre-amplified output beam. The optics in the

oscillator cavity result in three beams: the tunable laser light, ASE, and stray reflections off the faces of the cuvette.

4.2 Spontaneous emission in QUI

The dye laser was operated with QUI dye with a pulse repetition rate of 10 Hz and the high voltage in the excimer laser at 20 kV. The output laser beam was sent through a beam expander and an iris was used to block the stray light and the light due to spontaneous emission from entering the amplifier stage. The resulting output beam was scattered off of a white card and the OO USB2000 spectrometer was used to analyze the spectrum of this light.

Using the Scanmate2[®] tuning control, the laser was stepped in 1 nm increments from 370 nm to 374 nm and from 388 nm to 393 nm. These wavelength ranges cover the two emission peaks of QUI (refer to Figure 1.6). A spectrum was recorded at each wavelength. The wavelength where peak intensity was measured in the spectral data corresponds well with the input laser wavelength, further confirming the relatively precise tuning of the laser beam.

Two spectral peaks are observed in the resultant spectrum for tuning wavelengths of 370 nm to 374 nm, as is seen in Figures 4.3 and 4.4. A small peak is observed at the wavelength corresponding to the input laser wavelength and a larger peak is observed at the peak gain of the dye, near 390 nm. This secondary peak is due to spontaneous emission within the dye. There is a baseline background noise of ~175 intensity units.



Figure 4.3 Spectra of QUI dye laser beam tuned to 372 nm.



Figure 4.4 Relative intensities of spectral peaks observed in QUI dye.

An analysis of the ratio between the amount of light due to stimulated emission and that due to spontaneous emission in the QUI dye for operational wavelengths of 370 nm to 374 nm was conducted. The integrated intensity of the stimulated emission line, the spontaneous emission line, and the baseline background were determined (refer to marked ranges in Figure 4.3). The background light was scaled for the width (number of pixels, or data points) of the emission lines and subtracted from their integrated intensities. The stimulated emission peak was discernible from the background noise for measurements at 371, 372, 373, and 374 nm. Table 4.1 shows the relative optical signals of the stimulated emission peak, R₁, corresponding in wavelength to the input laser wavelength and the spontaneous emission peak, R₂, as well as the ratio between the two. The highest stimulated emission output between 371 nm and 374 nm is measured at 372 nm, where the laser operates for iron resonance lidar measurements. Spontaneous emission at 390 nm is 61 times stronger than the stimulated emission at this wavelength.

$\lambda_{\text{Laser}}(\mathbf{nm})$				R ₁ (arbitrary	R ₂ (arbitrary	R_2/R_1
	I_1	I_2	I ₃	units)	units)	
370	912	9386	2615	-22	6397	N/A
371	1169	10833	2722	3	7723	2574
372	1275	10519	2692	121	7442	61
373	1203	11069	2672	58	8015	138
374	919	10313	2653	90	7281	81

Table 4.1 Ratio of spontaneous emission R₂ to stimulated emission R₁ in QUI dye, where I₁, I₂, and I₃ are the intensities of the stimulated and spontaneous emission peaks and the background signal, respectively.

The amount of light attributable to spontaneous emission is indistinguishable from the stimulated emission at wavelengths from 388 nm to 393 nm, in the second energy output peak of QUI dye. At λ_{Laser} equal to 390 nm, the relative optical signal of the spectral peak is 13703, 113 times more intense than the stimulated emission peak at 372 nm (refer to Figure 4.4).

4.3 Spontaneous emission in Rhodamine 6G dye laser spectra

Rhodamine 6G is a common laser dye used for sodium resonance lidar studies. Its emission peak occurs in the visible range of wavelengths near 585 nm [Figure 4.5]. The dye laser was operated with Rhodamine 6G dye at 1 Hz, 18 kV. A beam expander and reflecting mirror were used to transmit the beam a distance of approximately 30 feet where it was intercepted by a white card. Spectra were captured at the site of the card. Transmitting the beam over this distance allowed a ~23 cm separation between the laser beam associated with stimulated emission and the stray light from the optics and the light due to spontaneous emission [Figure 4.6]. The bright orange spot on the right in Figure 4.6 is the light of the laser beam tuned to 610 nm. The green band of light at top left is stray light from the optics, and the bright yellow light at lower left is amplified spontaneous emission at the peak of the dye. The laser is operated at 610 nm in this analysis because the dye gain is weak at this wavelength, allowing more dye molecules to be available for spontaneous, rather than stimulated, emission processes.



Figure 4.5 Energy output of Rhodamine 6G dye laser as a function of wavelength.



Figure 4.6 Separation of Rhodamine 6G dye laser light from stray light and light due to spontaneous emission at λ =610.



Figure 4.7 Same as Figure 3.5 except λ =580 nm.

The optical distinction of differing colors between the types of output light occurs only at wavelengths where the emission of the dye is weak. At 580 nm, where the energy output of the dye is greater, the laser beam, the light due to spontaneous emission, and the stray light are the same color (alternatively, the same wavelength) [Figure 4.7].

In the first set of measurements, the stray light and the light due to spontaneous emission were blocked, isolating the light of the laser beam only. The laser was then tuned in 5 nm increments from 570 nm to 615 nm, across the emission peak of the Rhodamine 6G dye. A spectrum was recorded at each wavelength. The wavelength where peak intensity was measured corresponds well with the input laser wavelength [Figure 4.8].



Figure 4.8 Input laser wavelength versus wavelength corresponding to spectral peak in Rhodamine 6G dye laser beam.

Light due to spontaneous emission at the peak of the dye is only visible in the laser beam for spectra taken where the dye emission is weak, at 570 nm and 615 nm. The intensity of the spontaneous emission at 580 nm is comparable in these two spectra. Figure 4.9 displays the spectrum obtained at λ_{Laser} equal to 615 nm. The relative optical signals of the stimulated emission and spontaneous emission are similar, though the spontaneous emission line shape is broader. Only at wavelengths where the gain of the dye is weak is the spontaneous emission measurable for Rhodamine 6G dye.

In the second set of measurements, the light due to stimulated emission (the laser beam light) was blocked and only the spectrum of the spot attributed to spontaneous emission was recorded. The laser was tuned in 5 nm increments from 570 nm to 615 nm. At 570 nm the spectral peak is merged with the more intense and broader peak of the spontaneous emission at 580 nm. Both peaks are of greater intensity than in the first set



Figure 4.9 Spectra of Rhodamine 6G dye laser beam tuned to 615 nm, spontaneous emission blocked.

of measurements. At 575 nm, the spectrum looks almost identical to that obtained at the same wavelength in the first set of measurements, with no secondary peak visible at 580 nm. As the wavelength is stepped toward 615 nm, the spectral peak at the wavelength corresponding to the tuning wavelength becomes more intense than in the spectra taken in the first set of measurements. Emission at 580 nm also grows until at 610 nm, it is half as intense as the peak at 610 nm and approximately three times as broad. At 615 nm, the peak at 580 nm dwarfs the 615 nm emission [Figure 4.10].



Figure 4.10 Spectra of Rhodamine 6G dye laser beam tuned to 615 nm, stimulated emission blocked.

Following these two sets of measurements the cavity optics were adjusted to maximize the energy of the output laser beam. The laser was tuned to 605 nm, where the stimulated emission peak is still sufficiently intense to be discerned from the background noise and the spontaneous emission light at 580 nm is clearly visible in the spectrum. In this spectrum, the relative optical signal from the stimulated emission is five times the signal from the spontaneous emission. Spectra were then taken with the diffraction grating blocked, unblocked, and with the pre-amplifier blocked [Figure 4.11]. When the grating was unblocked the Molectron Energy Meter measured beam energy of 8 mJ, versus 4 mJ when the grating was blocked. The light from spontaneous emission, the lasing peak and the background were isolated and integrated for each spectrum. The background was subtracted, yielding the intensities of the spontaneous and stimulated emission light for the three cases. The intensities are shown in Figure 4.12.



Figure 4.11 Spectra of 605 nm Rhodamine 6G dye laser beam with the grating blocked (upper left), unblocked (upper right), and with the pre-amplifier output blocked (lower middle).

When the grating is unblocked, stimulated emission dominates. When the grating is blocked, the total output is attributable to spontaneous emission, which is expected. With the pre-amplifier blocked, we see a similar relationship as for the case of a blocked grating. The ratio of the light due to stimulated and spontaneous emission measured when the grating is unblocked to the spontaneous emission measured when the grating is blocked is 1.5 for this case, where the tuning wavelength is 605 nm.



Figure 4.12 Spontaneous (at λ =605 nm) and stimulated emission (at λ =580 nm) intensities for three cases of output for Rhodamine 6G dye laser.

4.4 Conclusions regarding spontaneous emission

The double peaked emission spectra of the QUI dye allows examination of the spontaneous emission at the peak of the dye gain near 390 nm for laser tuning wavelengths of 370-374 nm. Over this range of wavelengths, the resultant ratio of spontaneous to stimulated emission is calculable. The spontaneous emission is greater than the stimulated emission over this range, being over 2000 times more intense at 371 nm and 81 times more intense at 374 nm (the stimulated emission at 370 nm is not discernible from the background light). Based on these findings, it is likely that a significant portion of the optical signal measured at wavelengths from 388 nm to 393 nm is spontaneous emission. The amount is not quantifiable in this set of spectrometer data, however, because it is indiscernible from the stimulated emission, because they are occurring at similar wavelengths.

A further possible indication of the dominance of spontaneous emission in the second emission peak of the QUI dye is the shape of the spectral peak over this range of

wavelengths. Consider the line shape of Nd:YAG laser beam [Figure 4.13]. An asymmetrical peak is observed near 532 nm having a sharp rise on the left side and slightly more gradual decay and trailing rise on the right. This shape is a characteristic emission line shape for the gain mediums examined, where the trailing rise is attributed to collisions within the medium. The shapes of the spectral peak for the Rhodamine 6G dye laser operating at 590 nm and an Exalite 428 (this dye is discussed in section 4.6 and beyond) dye laser operating at 427 nm also display this characteristic shape [Figures 4.14 and 4.15].



Figure 4.13 Spectrum of NICT Rayleigh lidar Nd:YAG laser.



Figure 4.14 Spectrum of Rhodamine 6G dye laser operating at 590 nm.



Figure 4.15 Spectrum of Exalite 428 dye laser operating at 427 nm.



Figure 4.16 Spectrum of QUI dye laser operating at 390 nm.

The shape of the spectral peak of the QUI dye laser for tuning wavelengths from 388 nm to 393 nm does not have this characteristic shape [Figure 4.16]. The peak is more symmetrical, and does not have the rise on the right side. This shape is similar to the line shape of the spontaneous emission peaks seen in the QUI dye laser beam from 370 nm to 374 nm (refer to Figure 4.3) and the Rhodamine 6G dye laser beam at 570 nm and 615 nm (refer to Figure 4.10). The spectral line shape measured would seem to be indicative of a relatively high ratio of spontaneous emission occurring at the tuning wavelengths in the peak of the dye gain.

In the Rhodamine 6G dye, spontaneous emission is only observable at the peak of the dye gain for input laser wavelengths in the wings of this peak, at 570 nm and 615 nm. At these wavelengths, the gain of the dye is low, and more dye is available for spontaneous emission. Spontaneous emission in Rhodamine 6G does not affect the quality of sodium resonance lidar measurements, as the measurements are taken at 589 nm, where stimulated emission dominates in the dye.

4.5 Analysis of five dyes for 390 nm lidar studies

Prompted by the failure of the QUI dye laser in the lidar transmitter system to perform at 390 nm, an analysis of five other laser dyes fluorescing over a range including 390 nm was undertaken to analyze the effect of operating in a different wavelength range on the lidar system performance. Dyes were chosen based on their fluorescent range as well as their solubility in P-dioxane (the solvent used for QUI; having a common solvent simplifies dye handling procedures). The common names of the dyes and their reported lasing maximum wavelengths are listed in Table 4.2 [Exciton, 2006]. Dyes with reported lasing maximums near 390 nm, but not at this wavelength, were chosen so as to attempt to avoid to recurrence of spontaneous emission dominance at the peak of the dye. QUI is included in Table 4.2, and the following figure, for comparison. Collectively, this group of dyes is referred to as the 390 nm dyes.

The energy output with wavelength was measured for each of the dyes [Figure 4.17]. Note the increase in energy output for each dye as the grating order changes at 395 nm (refer to section 3.6). Excluding PBBO (2-[1,1', biphenyl]-4-yl-6-phenyl-benzoxazole) [Exciton, 2006], these dyes have peak energy higher or comparable to QUI. A trend of the peak energy output occurring at or very near to 390 nm was observed in three of the dyes. Of the 390 nm dyes, only QUI and BBQ (4',4'"-Bis(2-butyloctyloxy)-p-quaterphenyl) [Brackmann, 1997] spectra were examined. BBQ shows emission trends similar to Rhodamine 6G; spontaneous emission is visible in the spectra when the laser is operated at wavelengths in the wings of the BBQ emission [Figure 4.18], but the stimulated emission at wavelengths near the peak gain of the dye does not appear to be dominated by it [Figure 4.19].

Common name of dye	Reported lasing maximum (nm)
QUI	390
Exalite 392A	392
BBQ	387
TBS	398
PBBO	396
Exalite 389	388

Table 4.2 Dyes analyzed for 390 nm lidar studies.



Figure 4.17 Comparison of energy output with wavelength for 390 nm dyes.



Figure 4.18 Spectrum of BBQ dye laser operating at 380 nm.



Figure 4.19 Spectrum of BBQ dye laser operating at 390 nm.

On the nights of October 28, October 31, November 7, November 19, December 1, 2008, and January 8, 2009, sky tests were conducted for the 390 nm dyes. The NICT Rayleigh lidar system was operated simultaneously on each of these nights to provide a measure of sky transmission. When sky conditions warranted it (as was the case on the nights of November 19 and December 1, 2008) the resonance lidar signals were corrected for decreased atmospheric transmission due to sub-visual cirrus clouds.

Three bandpass filters were used in these studies: one with a center wavelength of 372 nm and a bandwidth of 1 nm (372 nm/1 nm), one with a center wavelength of 390 nm and a bandwidth of 10 nm (390 nm/10 nm), and one with a center wavelength of 400 nm and a bandwidth of 40 nm (400 nm/40 nm). When measurements were taken at wavelengths on the edges of the filter transmission function [Su, 2007], the signal was compensated for the value of transmission at this wavelength.

When the lidar system was operated with each of the five 390 nm dyes in the dye laser system, a decrease in lidar signal or a very low signal was measured when the laser was tuned to 390 nm [Appendix A]. Results for tests conducted with Exalite 389 are shown as an example of this recurrent behavior. The energy output of Exalite 389 peaks at 390 nm [Figure 4.20], but the lowest lidar signal per laser pulse from 20-25 km and 30-35 km occurs when the laser is tuned to this wavelength [Figure 4.21]. The signal has been compensated for both filter and atmospheric transmission. Measurements were taken with the 400 nm/40 nm bandpass filter. The normalized Rayleigh lidar signal used to correct for decreased atmospheric transmission is shown in Figure 4.22. Signals are normalized to the signal obtained for stable atmospheric transmission (in this case, the transmission was stable for sets 6 through 14). Resonance signals are matched to the Rayleigh normalized signals in time to determine the atmospheric transmission value for each measurement.

Due to the persistent decrease in signal at 390 nm, the success of N_2^+ resonance lidar measurements at wavelengths at or near 390 nm is unlikely with the current lidar system. While spectral evidence indicates a relatively high occurrence of spontaneous emission in the QUI dye, it is unlikely this dominance is a common characteristic of all

dyes examined, as the dyes are molecularly dissimilar. All dyes manufactured by Exciton [2006], fluorescing over a range including 390 nm and soluble in p-dioxane (the solvent used), where considered in this study. Due to the persistent lack of lidar system performance at 390 nm despite the wide variety of dyes tested, it is likely the recurrent decrease in signal is due to a characteristic of the dye laser itself, rather than the laser dye, at this wavelength. Su [2007] ruled out the possibility of the optical elements in the lidar receiver system being responsible for the decreased signal at 390 nm. Therefore, it is further likely the lack of performance is attributable to the transmitter system.

Key optical elements of the transmitter system were considered. The throughput of light through the beam expander and off of the 90° reflecting mirror which amplify and transmit the laser beam output from the dye laser into the sky was examined and found to be good. The optical system of the prism beam expander known as the Lambda Superpure® in the dye laser oscillator stage is proprietary to Coherent, Inc. (previously, Lambda Physik), and cannot be investigated. The placement of 390 nm light within a grating order, discussed in section 3.6, is a likely factor for the lack of system performance at this wavelength because the placement would be the same for every dye tested in the dye laser system. The conclusion that the diffraction grating fails to properly transmit light at 390 nm, so close to the edge of a grating order, agrees well with the consistent observation of a perceived increase in the energy output of laser dyes immediately following a change in grating order. The reported spectral properties of the laser dyes do not account for this observed increase and it is likely caused by an increase in the performance of the grating immediately following a change in grating order.

The tilt of the diffraction grating at tuning wavelengths of 589 nm, 372 nm, 390 nm, and 400 nm was examined by Su [2007]. Su found that at a tuning wavelength of 390 nm the tilt of the grating was at a relative maximum angle compared to the other tuning wavelengths examined.



Figure 4.20 Energy output of Exalite 389 dye laser with wavelength.



Figure 4.21 Compensated signal from 20-25 km and 30-35 km altitude ranges for Exalite 389 dye laser.



Figure 4.22 NICT Rayleigh lidar signals in each altitude range normalized to stable sky conditions for November 18, 2008.

4.6 Analysis of four dyes for 428 nm lidar studies

Based on the failure of the dye laser operating with the 390 nm dyes to provide a beam of sufficient performance to conduct resonance lidar studies of the 390 nm band transitions of N_2^+ , four dyes fluorescing in ranges including 428 nm where tested for the possibility of conducting measurements of the second strongest band of N_2^+ transitions (the spectroscopy of N_2^+ is discussed in Chapter 5). The common names of these dyes (referred to collectively as the 428 nm dyes) and their reported lasing maximums are listed in Table 4.3 [Exciton, 2006].

The energy output with wavelength was measured for each of the dyes [Figure 4.23]. The peak output of these dyes is between 2.5 (POPOP (2,2'-(1,4-phenylene)bis[5-phenyl-oxazole], $C_{24}H_{16}N_2O_2$ [Exciton, 2006]) and 4.7 (Exalite 417) times higher than the peak energy of QUI. Three of the four dyes peak near 420 nm, and have fallen to approximately a third of this peak energy at 428 nm. The fourth dye, Exalite 428, peaks strongly at 428 nm. Exalite 428 is the only dye whose spectrum was observed with the

OO USB2000 spectrometer. The spectral data shows that at wavelengths where the stimulated emission is weak (where spontaneous emission was observed in other dyes), the spontaneous emission at the peak of the dye is barely discernible from the background light [Figure 4.24]

Common name of dye	Reported lasing maximum (nm)
Exalite 417	416
Exalite 428	428
Bis-MSB	421
POPOP	421

Table 4.3 Dyes analyzed for 428 nm lidar studies.



Figure 4.23 Comparison of energy output with wavelength for 428 nm dyes.



Figure 4.24 Spectrum of Exalite 428 dye laser operating at 437 nm.

Sky tests of the Bis-MSB (1,4-bis[2-(2-methylphenyl)ethenyl]-benzene) [Exciton, 2006] and POPOP dye laser systems where conducted on the night of November 7, 2008. The NICT Rayleigh lidar system was operated simultaneously. The Exalite 417 dye laser was sky tested on the night of November 19, 2008, along with Exalite 428, which was also tested on December 1, 2008. All measurements for these dyes were conducted with the 400 nm/40 nm bandpass filter. The measurements taken on November 7, 2008, did not need to be compensated. The November 19 and December 1, 2008, measurements were compensated for both filter and atmospheric transmission.

The three dyes which peaked in energy at 420 nm (POPOP, Bis-MSB, and Exalite 417) do not show a marked drop in sky performance when the laser is tuned to the peak of their gain [Appendix A]. However, none of them have sufficient output at 428 nm to make them viable for N_2^+ studies. Exalite 428 also does not show a drop in signal when the laser is tuned to the peak of the dye, and since the dye peaks at 428 nm, Exalite 428 is a likely dye for use in obtaining N_2^+ resonance lidar measurements at 428 nm [Figure 4.25]. A signal of ~10.5 photon counts per shot is measured in the 20 to 25 km altitude

range. The Rayleigh lidar data used to compensate the signal plotted in Figure 4.25 is given in Figure 4.26.



Figure 4.25 Compensated signal from 20-25 km and 30-35 km altitude ranges for Exalite 428 dye laser on December 1, 2008.



Figure 4.26 NICT Rayleigh lidar signals in each altitude range normalized to stable sky conditions for December 1, 2008.

The transmission of the 400 nm/40 nm bandpass filter was determined and plotted by Su [2007]. 428 nm is in the wings of the filter's transmission; only approximately 10% of 428 nm wavelength light is transmitted. The decrease in filter transmission at a wavelength of 428 nm is corrected for by dividing the signal by the filter transmission at the wavelength of the measurement. To obtain more accurate measurements, requiring less correction, a bandpass filter with a center wavelength of 426.7 nm and a bandwidth of 3 nm (426.7 nm/3 nm) was introduced into the lidar receiver system in May of 2009.

Sky tests of the Exalite 428 dye laser system with the 426.7 nm/3 nm filter in the receiver system were conducted on the night of September 15, 2009. The excimer energy was 288 mJ, 75 mJ lower than on December 1, 2008. These measurements were taken with the 426.7 nm/3 nm bandpass filter and the 1.02 m diameter telescope in the receiver system. The measured Rayleigh scatter of the Exalite 428 laser beam in the 30-35 km height range is plotted with wavelength in Figure 4.27. The 20-25 km signal fell below the background signal for part of the night, as sky conditions deteriorated, and is not plotted. The blanking was decreased from 50 μ s to 20 μ s for these measurements. The normalized Rayleigh lidar signal used to correct for decreased atmospheric transmission is shown in Figure 4.28.

The signals measured on September 15, 2009, are lower than the signals measured on December 1, 2008 for the same height range, despite the increase in the area of the receiving telescope between the two nights and the use of the 426.7 nm/3 nm bandpass filter. The excimer laser energy was lower on September 15, 2009, however, and both signals required significant compensation for poor sky conditions. Further tests of the resonance lidar system operating with Exalite 428 dye should be conducted to verify the performance of this system for use in measurements of aurorally excited molecular nitrogen ions.

The successful performance of the 428 nm dyes in the dye laser further confirm the conclusions reached regarding the lack of system performance at 390 nm. The operating wavelength of 428 nm occurs in the middle of the wavelength range for grating order 7, and is therefore free from any effects due to operating at a wavelength near the edge of a grating order.



Figure 4.27 Compensated signal from 30-35 km altitude ranges for Exalite 428 dye laser on September 15, 2009.



Figure 4.28 NICT Rayleigh lidar signals in each altitude range normalized to stable sky conditions for September 15, 2009.
5 Spectroscopy of N₂⁺

In this chapter, the quantum mechanics of diatomic molecules is briefly reviewed, followed by a discussion of the determination, via spectroscopic calculation, of the optimum operational wavelengths for N₂⁺ resonance lidar. The fundamental principle of resonance fluorescence lidar is not wavelength matching, but frequency matching, between the strongest transition within the target species and the transmitted laser light. The atomic and molecular transitions within the target species for lidar measurements occur at characteristic frequencies, defined by the respective atomic characteristics. Operationally, the wavelength of the laser beam is tuned until its frequency matches the frequency of the strongest transition within the target species, thereby yielding the maximum resonance scattering signal. The following treatment of the spectroscopy of N_2^+ is concerned with determining the wavelength corresponding to the strongest transitions, rather than their frequency, because wavelength is the quantity controlled operationally when obtaining lidar measurements. Due to variations in the reported molecular constants of N_2^+ , there is a degree of uncertainty in the determined wavelengths. Variations in atmospheric temperature also allow for a range of possible optimum operational wavelengths. Both the (0, 0) (centered at ~390 nm) and the (1, 0) (centered at ~428 nm) vibrational transition bands are considered.

The following discussion of molecular spectroscopy follows the treatment of Herzberg [1950], Brown [1998], and Hollas [2004]. The interested reader is referred to these works for a detailed discussion of the molecular spectroscopy of diatomic molecules, as a full treatment of all the relevant quantum mechanics is beyond the scope of this work.

5.1 Introduction

The spectroscopy of a diatomic molecule such as N_2^+ is far more complicated than that of atomic Na or Fe. A molecule can rotate about an axis passing through the center of gravity and perpendicular to the internuclear axis. The atoms can also vibrate relative to each other along the internuclear axis [Herzberg, 1950]. These complex atomic interactions give rise to a host of possible transitions. The wavelength corresponding to any transition within the N_2^+ molecule is dependent on the energy contributions from the electronic, vibrational, and rotational states. Molecular spectra can rarely be calculated from quantum mechanics, but must be experimentally determined. Fortunately, the nitrogen molecule, as a primary constituent of the atmosphere, has been extensively studied [Scholl et al., 1998].

Collins et al. [1997] determined the optimum lidar system for profiling aurorally produced N_2^+ would operate at wavelengths corresponding to rotational-vibrational transitions between electronic states $X^2\Sigma_g^+$ and $B^2\Sigma_g^+$ (referred to as the first negative band of transitions). The two strongest vibrational transitions occur between the $\upsilon''=0$, $\upsilon'=0$ (0,0) state and the $\upsilon''=1$, $\upsilon'=0$ (1, 0) state, where υ'' and υ' denote the vibrational quantum numbers of the lower ($X^2\Sigma_g^+$) and upper ($B^2\Sigma_g^+$) states, respectively. The population of electrons available for transitions in a given rotational state depends on the temperature T, which varies between 200 and 1000 K in the thermosphere [Collins et al., 1997]. The optimum operational wavelength for N_2^+ resonance lidar studies is the wavelength corresponding to the strongest transition in the current population. The strongest transition is found by maximizing the product of the ratio of degeneracies between the upper and lower states, the Honl-London factor (diatomic rotational line strength), and the temperature dependent population.

Before discussing the determination of the optimum transitions and corresponding operational wavelengths for lidar studies of aurorally produced N_2^+ , the quantum mechanics of diatomic molecules is reviewed.

5.1.1 Review of quantum mechanics

An atom or molecule, quantum mechanically, is described by its wavefunction, Ψ , which is a function obtained by solving the Schrödinger equation. The Schrödinger equation is the fundamental equation for connecting wave behavior to energy [Krane,

1996] and, therefore, fundamental to spectroscopy, as spectroscopy is the study of the interaction between radiation and matter. The Schrödinger equation must satisfy certain symmetry principles and conservation laws. If the wavefunction of a system is known, all the observable properties of the system may be deduced by performing the appropriate mathematical operations [Atkins, 1991].

In its basic form the Schrödinger equation is expressed as,

$$H\Psi = E\Psi \tag{5.1}$$

where H is the Hamiltonian operator and E is the energy of the system. The Hamiltonian is derived by taking the complete classical expression for the energy of a system (the sum of the kinetic and potential energies), re-expressing it in terms of linear momenta and position coordinates, and transforming the resultant equation into operator form. In operator form, a position coordinate is represented simply by itself: $x \rightarrow \hat{x}$. The component of linear momentum is represented by a complex, partial derivative: $p_x \rightarrow \hat{p}_x = -i\hbar \frac{\partial}{\partial x}$, where $\hbar = \frac{h}{2\pi}$ (the quantity h/2 π occurs frequently in wave mechanics and is given the special symbol h-bar, \hbar [Krane, 1996]). If a single particle of mass m within a potential well is examined, the Hamiltonian

$$H = \frac{-\hbar^2}{2m} \nabla^2 + U(x, y, z)$$
(5.2)

is obtained. Ψ is an eigenfunction of H and E is its corresponding eigenvalue. H will have a set of eigenfunctions and corresponding eigenvalues that will satisfy the Schrödinger equation. The energy levels of a system are determined by defining the appropriate Hamiltonian of a system, and then determining the eigenfunctions and eigenvalues of this Hamiltonian.

Application of the Born-Oppenheimer approximation allows determination of the wavefunction for electrons in an atom. The approximation assumes that in an atom the electrons and nuclei are subject to similar forces, but the electrons move much faster than the nuclei under this same force because they are far less massive [Atkins, 1991]. Therefore, the nucleus (or nuclei in a molecule) is treated as motionless, which reduces the number of coordinates considered. The motion, or energy, of the electrons can then be

separated into electronic and nuclear parts. In molecules, the nuclear energy is further divided into vibrational and rotational energy. The separation of vibrational energy states is greater than the separation of rotational energy states because the characteristic frequency of vibration is greater [Brown, 1998]. The total energy of a state can be expressed as a simple sum of the contributions from the three types of motion: $E_{tot}=E_{el}+E_{vib}+E_{rot}$.

The eigenvalues, or allowed energy levels, of the rotational Hamiltonian for a diatomic molecule, are given by the relation

$$E_{rot} = \left(\frac{h^2}{8\pi^2 I}\right) J(J+1) = BJ(J+1) \quad (cm^{-1})$$
 (5.3)

where J is the rotational quantum number (J=0, 1, 2...) and I is the moment of inertia of the molecule. E_{rot}, the energy of a given rotational energy level, is not expressed here in joules (J) because what is measured in experimental spectroscopy is not energy, but frequency or wavenumber. The wavenumber \tilde{v} of a transition is the inverse of the wavelength ($\tilde{v} = \frac{1}{\lambda}$) and is expressed in units of cm⁻¹, by convention [Brown, 1998]. The quantity $\frac{h^2}{8\pi^2 I}$ in equation 5.3 is known as the rotational constant B and is also commonly

defined in cm⁻¹. B is determined spectroscopically and allows determination of internuclear distances [Hollas, 2004]. In a transition between rotational energy levels, the selection rule $\Delta J=0, \pm 1$ is obeyed. ΔJ is conventionally taken to refer to J'-J'', where J' is the rotational quantum number of the upper state and J'' that of the lower state [Hollas, 2004]. Rotational transition wavenumbers, or rotation term values for a spherical rotor, are given by $\tilde{v} = 2B(J+1)$ where J is used conventionally instead of J''. From equation 5.3 it is clear that the rotational energy level separation increases quadratically with J.

The rotational constant is slightly vibrationally dependent. For diatomic molecules in excited vibrational states, the vibrational dependence of B is given (to a close approximation) by

$$B_{\upsilon} = B_{e} - \alpha \left(\upsilon - \frac{1}{2} \right)$$
(5.4)

where B_e refers to the hypothetical equilibrium state of the molecule, α is the vibrationrotation interaction constant, and v is the vibrational quantum number. B_e is obtained by measuring B_v in at least two vibrational states. If there is an insufficient population of the v=1 level to obtain B_1 then rotational spectroscopy can give only B_0 [Hollas, 2004].

The vibrational energy levels of a diatomic molecule, treated in the harmonic oscillator approximation, are given by

$$E_{\upsilon} = hv \left(\upsilon + \frac{1}{2}\right) \tag{5.5}$$

where the vibrational quantum number v=0, 1, 2... Like the rotational energy levels, the vibrational energy levels are expressed as term values in cm⁻¹. The vibrational term T_v value is given by

$$\frac{E_{\upsilon}}{hc} = T_{\upsilon} = \omega \left(\upsilon + \frac{1}{2}\right)$$
(5.6)

where ω is the vibrational wavenumber. The vibrational selection rule is $\Delta v = 0, \pm 1$ and all vibrational energy levels are equally spaced. Vibrational transitions give rise to what are called 'bands' in the spectrum. The word 'line' describes transitions between the rotational levels associated with the two vibrational levels.

5.2 Determination and variations of N_2^+ transition wavelengths

The optimum lines for N_2^+ resonance lidar studies are determined from the reported rotational and vibrational term values. The base terms of the upper and lower vibrational states are calculated as well as the vibrationally dependent rotational constants for each state. The rotational energy contributions are combined with those from the electronic and vibrational states to yield the total energy of each state. The wavelength corresponding to each transition between energy levels is determined by use of equation 2.5.

In our analysis, rotational transitions with J''=0 to 50 were considered in the (0,0) and (1, 0) vibrational states of the first negative band of N_2^+ . Even at high temperatures, no states with J>40 are sufficiently populated to be of interest. A partial energy level diagram of

 N_2^+ is given in Figure 5.1 (the optimum rotational transitions for lidar studies at T=500 K are shown). As was previously stated, rotational transitions must follow the selection rule $\Delta J=0$ or ±1. However, $\Delta J=0$ is forbidden for electronic systems in which both states have zero angular momentum along the internuclear axis (such states are designated Σ) [Garner and Dao, 1995].



Figure 5.1 Partial energy level diagram for rotational transitions in molecular nitrogen ions.

As both of the electronic states under consideration are Σ states, $\Delta J=0$ is forbidden for the N_2^+ first negative band. Transitions with $\Delta J= -1$ are referred to as the P branch, and $\Delta J= +1$ are in the R branch. Therefore, for example, if J''=0 (in $X^2\Sigma_g^+$ state), J'=1 (in $B^2\Sigma_g^+$ state), $J''_P = 2$, $J''_Q=1$ (Q branch forbidden), and $J''_R=0$. The R transitions are chosen for lidar because this allows the received wavelength to correspond to the transmitted wavelength (refer to Figure 5.1). Based on the value of J'' and the known selection rule, the excitation wavelength to level J' and the subsequent J''_P and J''_Q transition wavelengths are calculated. The Honl-London factor for each rotational transition is

determined from selection rules depending on the quantum numbers of the lower state (see Brown [1998]). The Honl-London factor for each transition is combined with the temperature dependent population N (T=200 K, 500 K, and 1000 K are considered) of the J" state, given by the Boltzmann distribution

$$N(J) = \frac{hcB_{v}}{kT} \cdot (2J'' + 1)e^{\frac{-hcB_{v}}{kT}J''(J'' + 1)}$$
(5.7)

along with the ratio of degeneracies between the upper state and the lower state (g=2J+1), to determine the optimum transition for N_2^+ reasonance lidar studies.

The Honl-London factor increases for higher rotational states, while the degeneracy decreases [Figure 5.2; here the Honl-London factor is given as the ratio of the R state factor to the total factor for the P, Q, and R states]. Both values quickly plateau and then change little between high rotational states. Neither value is temperature dependent, so it is largely the temperature dependent population which determines which transitions are strongest [Figure 5.3]. A baseline uncertainty in the wavelengths corresponding to transitions appropriate for lidar studies exists due to variations in the reported N_2^+ molecular constants, which is discussed next.



Figure 5.2 Honl-London factors and degeneracy ratio for (0, 0) rotational transitions with relative strengths > 5% of maximum value.



Figure 5.3 Temperature dependent population of rotational states (left) is the primary factor determining the optimum wavelengths for lidar studies of molecular nitrogen ions (right).

5.2.1 Quantum uncertainty due to reported molecular constants

Differing transition wavelengths within the N_2^+ molecule (λ_{vac}) are calculated from vibrational and rotational constants reported by two sources: Laher and Gilmore [1991], and Scholl et al. [1998]. Laher and Gilmore [1991] combined the spectroscopic data on N_2^+ that was available at the time to derive the vibrational and rotational constants they reported. Scholl et al. [1998] were able to probe the hyperfine structure of N_2^+ for the first time by means of fast-ion-beam laser spectroscopy and derive, what they called, more refined values for the vibrational and rotational constants. The method of fast-ion-beam spectroscopy involves an arrangement where a laser beam travels collinearly with a focused ion beam in which the ions have been mass (velocity) selected by a Wien filter [Cameron et al., 1995]. The beam of ions is accelerated or decelerated by the application of an external electric field. Application of this voltage further narrows the velocity distribution of the ions. Narrowing the velocity distribution drastically reduces, to the point of elimination, the effects of Doppler broadening, allowing resolution of fine spectral features.

The molecular constants reported by each source for both states are shown in Table 5.1. Note that Scholl et al. [1998] provides a more limited set of constants than do Laher and Gilmore [1991]. Also, Scholl et al. [1998] do not report values of T_0 , the energy of an excited or ionic state relative to the v=0, J=0 level of the corresponding neutral ground state. Laher and Gilmore [1991] tabulated T_0 rather than the equilibrium term value, T_e , because it is directly measured and does not depend on the choice of molecular constants ω_e , etc. The relation between T_e and T_0 is [Laher and Gilmore, 1991]

$$T_{e} = T_{0} - \frac{\omega_{e}}{2} + \frac{\omega_{e}\chi_{e}}{4} - \frac{\omega_{e}y_{e}}{8} - \frac{\omega_{e}z_{e}}{16} - \frac{\omega_{e}a_{e}}{32}$$
(5.8)

The vibrational and rotational base terms were calculated with the following polynomials [Laher and Gilmore, 1991]:

$$T_{v} = T_{e} + \omega_{e} (\upsilon + \frac{1}{2}) - \omega_{e} \chi_{e} (\upsilon + \frac{1}{2})^{2} + \omega_{e} y_{e} (\upsilon + \frac{1}{2})^{3} + \omega_{e} z_{e} (\upsilon + \frac{1}{2})^{4} + \omega_{e} a_{e} (\upsilon + \frac{1}{2})^{5}$$
(5.9)

$$B_{v} = B_{e} - \alpha_{e} (\upsilon + \frac{1}{2}) + \gamma_{e} (\upsilon + \frac{1}{2})^{2} + \delta_{e} (\upsilon + \frac{1}{2})^{3} + \varepsilon_{e} (\upsilon + \frac{1}{2})^{4}$$
(5.10)

The relevant transition wavelengths were calculated using molecular constants from both sources, for atmospheric temperatures of 200 K, 500 K, and 1000 K. The strongest (0,0) transition in N_2^+ is between rotational levels nine and ten at 500 K and has a vacuum wavelength of 390.303 nm when using Laher and Gilmore's [1991] constants.

	Laher,	Scholl et al.		Laher,	Scholl et al.
$\mathbf{X}^{2} \mathbf{\Sigma}_{g}^{+}$	Gilmore [1991]	[1998]	$\mathbf{B}^{2} \mathbf{\Sigma}_{u}^{+}$	Gilmore [1991]	[1998]
T ₀	125667.5 [†]	*	T ₀	151233.5	
T _e		0	T _e		25461.064
ω _e	2207.37	2207.252	ω _e	2420.83	2421.14
ω _e χ _e	16.302	16.256	ω _e χ _e	23.851	24.07
ω _e y _e	-2.67e-3	0	ω _e y _e	-0.3587	-0.3
$\omega_e z_e$	-2.61e-3		ω _e z _e	-6.192e-2	
ω _e a _e	3.7e-5		weae		
Be	1.93177	1.9317315	Be	2.0845	2.0853257
α _e	1.900e-2	1.874e-2	α _e	2.132e-2	2.12e-2
γe	-1.91e-5		γe	-8.5e-4	
δ_{e}	-5.0e-6	-9.7e-5	δ _e		-5e-4
E _e	4.6e-8		Ee		

Table 5.1 Vibrational and rotational molecular constants for the $X^2\Sigma_{\,g}^{\,+}$ and $B^2\Sigma_{\,u}^{\,+}$ states.

[†] Values given in cm⁻¹.

*Fields left blank were not reported by source.

The wavelength calculated using the constants reported by Scholl et al. [1998] differs by 2.01 pm (390.301 nm). The differences between the wavelengths of the first 50 rotational transitions calculated from each source become more pronounced for weaker transitions,

increasing from a few pm near 390 nm, to 37 pm near 382 nm [Figure 5.4 (left); The 'vacuum wavelength' is taken to be the wavelength calculated from the Laher and Gilmore [1991] molecular constants].

The strongest (1, 0) transition is also between rotational levels nine and ten at 500 K. This transition has a vacuum wavelength of 426.474 nm from the Laher and Gilmore [1991] constants, and a wavelength of 426.472 nm from Scholl et al. [1998]. The (1, 0) band exhibits similar difference trends, with differences near 427 nm of a few pm and differences as great as 45 pm near 416 nm [Figure 5.4 (right)].



Figure 5.4 (left) Difference between (0,0) transition wavelengths and (right) differences between (1, 0) transition wavelengths calculated from Laher and Gilmore [1991] and Scholl et al. [1998] molecular constants.

The differences seen in the wavelengths of the strongest transitions due to use of constants from two sources are baseline uncertainties arising from the limitations and variations in determination of the molecular constants of N_2^+ . Therefore, there is a fundamental quantum uncertainty of approximately 2 pm in the theoretical optimum operational wavelengths. The linewidth of the laser spans this range of uncertainty, however. At ~589 nm, the linewidth of the laser is ~5 pm and ~2 pm at 372 nm.

5.2.2 The effect of atmospheric temperature

The variations in atmospheric temperature alter the Boltzmann distribution of population within the states of the N_2^+ molecule. The available population in a given state affects which rotational transition will be the strongest and, therefore, the optimum wavelength for obtaining measurements. The rotational transition between the ninth and tenth states is the strongest at 500 K, but at 200 K the transition between the fifth and sixth states is stronger. At 1000 K, the strongest transition is between states 13 and 14. The differences between the wavelengths derived from the Laher and Gilmore [1991] constants and those derived from the Scholl et al. [1998] values increase for higher rotational states, so the uncertainty in the wavelength of the strongest transition increases with increasing temperature. Temperature variations over the range considered can vary the optimum operational wavelength over several hundred picometers [Table 5.2].

Table 5.2 Optimum operational wavelengths in nm derived from Laher and Gilmore [1991].

Vib. Transition	T=200 K	T=500 K	T=1000 K
(0, 0)	390.695	390.303	389.839
(1, 0)	426.963	426.474	425.888

5.2.3 Effects of refraction in the atmosphere

If the laser is tuned to the wavelength corresponding to the frequency of the calculated strongest transition within the N_2^+ molecule, the light reaching the molecules in the atmosphere will have a slightly different wavelength due to the effects of refraction (equation 3.23 in section 3.5). The calculated shift in wavelength due to refraction is approximately 100 pm for all wavelengths of light being considered for N_2^+ resonance lidar studies. By tuning the laser wavelength at the ground to maximize the received lidar signal, any effect of atmospheric refraction is included in the operational wavelength; we do not have to correct for it.

5.3 Conclusions regarding lidar operation

Tables giving the wavelengths of all rotational transitions with a relative strength $\geq 5\%$ calculated from both the Laher and Gilmore [1991] and Scholl et al. [1998] molecular constants and all pertinent information for atmospheric temperatures of 200 K, 500 K, and 1000 K are given in Appendix B. Based on the variations in the wavelength of the strongest transition in the N⁺₂ molecule due to slightly differing molecular constants and variable atmospheric temperatures, we expect the operational wavelength λ_{op} to fall in the ranges of 389.836 nm $\leq \lambda_{op} \leq 390.694$ nm for the (0,0) transitions and 425.884 nm $\leq \lambda_{op} \leq 426.962$ nm for the (1, 0) transitions. Operationally, the laser would be stepped through these ranges until a resonance signal is measured. It would then be possible to tune to the line by fine tuning according to increasing signal, similar to the manner in which sodium and iron maximum resonance is tuned to. Tuning to the wavelength of maximum resonance would, however, be more difficult for N⁺₂ measurements due to the high temporal and spatial variability of the aurora.

The variation in operational wavelength for obtaining iron resonance and sodium resonance lidar data has been documented over the years of resonance lidar operation at PFRR [Figures 5.5 and 5.6]. For each night resonance lidar data was obtained, the most common wavelength at which measurements were taken was recorded (the mode). The dashed line in each figure indicates the value of λ_{STP} corresponding to the vacuum wavelength of the strongest transition for sodium and iron. Over the seven years of reviewed resonance lidar data, the optimum operational wavelength is generally longer than λ_{STP} and varies widely. Iron data was obtained at wavelengths ranging from 371.99 nm to 372.04 nm, with a standard deviation of 9 pm. Sodium data was taken at wavelengths from 589.004 nm to 589.054 nm, with a standard deviation of 15 pm. Neither the iron or sodium vacuum wavelengths fall within these ranges. All observations, except one, were taken at longer wavelengths than λ_{STP} .



Figure 5.5 Variations in operational wavelength for iron resonance lidar measurements over 7 years.



Figure 5.6 Variations in operational wavelength for sodium resonance lidar measurements over 7 years.

6 Simulation of N₂⁺ (1, 0) Band Resonance Lidar System

Su [2007] expanded the work of Collins et al. [1997] in simulating the expected N_2^+ (0, 0) band resonance lidar signal. For a complete discussion of the simulation see Su [2007]. Given the unlikelihood of successful resonance lidar measurements at ~390 nm due to the lack of performance in the dye laser system at this wavelength, the simulation was modified to model the expected signal from the (1, 0) band of transitions in N_2^+ at ~428 nm. The pre-existing simulation is described and the modifications made are discussed. The modeled resonance lidar measurements of aurorally produced N_2^+ at ~428 nm are discussed and compared to the results of Su [2007]. Finally, the model is altered to provide a more accurate representation of the current capabilities of the resonance lidar system. Post integration of the model output is demonstrated to yield statistically significant resonance lidar measurements of aurorally produced N_2^+ .

6.1 The lidar simulation

The lidar simulation determines the resonance, background, and total lidar signal in altitude and time for the night of February 6-7, 1994, from 18:25 LST to 6:55 LST. A simple diagram of how the total lidar signal is derived from the model inputs is given in Figure 6.1. This night was chosen from reviewing 2 years of Meridian Scanning Photometer (MSP) data taken at PFRR. The MSP obtains integrated line-of-sight measurements of the aurora at four wavelengths: 557.7 nm, 427.8 nm, 620.0 nm, and 486.1 nm. The MSP scans along the magnetic meridian from horizon to horizon every 16 seconds with a 1° instantaneous field-of-view [Morse and Romick, 1982]. The observed brightness is compensated for atmospheric extinction by assuming emission occurs at an altitude of 100 km. The auroral activity of the night of February 6-7, 1994, was chosen as being representative of moderate auroral activity. Furthermore, the aurora is highly variable through the night allowing the assessment of the lidar signal during periods of active and quiet aurora. A more active aurora produces higher concentrations of N_2^+ at lower altitudes, resulting in an increase in the resonance lidar signal, while less active aurora yields lower quality lidar measurements [Collins et al., 1997]. The aurora at 19:05 LST, 19:55 LST, and 00:55 LST as imaged through an all-sky camera at PFRR is displayed for the night of February 6-7, 1994, in Figure 6.2.



Figure 6.1 Simplified diagram of the inputs determining the total expected N₂⁺ resonance lidar signal.

The expected N_2^+ resonance lidar signal is determined from the relation

$$N_{s}(z) = [\eta T^{2}] \cdot \left[\frac{E_{L}R_{L}\Delta t}{hc/\lambda_{L}} \right] \cdot \left[(C_{eff}(\nu, T)\gamma)\rho_{N_{2}^{+}}(z)\Delta z \right] \cdot \left[\frac{A_{R}}{4\pi z^{2}} \right]$$
(6.1)

which is identical to equation 3.2, except for the inclusion of the branching ratio, γ , which is the product of the vibrational and rotational branching ratios (0.71 and 0.48, respectively) and has a value of 0.34 for this band of transitions within N₂⁺. The effective scattering cross section varies with temperature because it is proportional to the temperature dependent Doppler width of the absorption cross section [Collins et al., 1997, Su, 2007]. The atmospheric temperature profile in altitude is input into the lidar model from a single MSISE-90 (Mass Spectrometer-Incoherent Scatter Model) atmospheric profile from 00:00 LST on February 6-7, 1994 [Figure 6.3]. Within the lidar model, the atmospheric temperature profile is considered to be constant in time. The lidar model, therefore, does not account for the heating of the atmosphere due to dissipation of the energy input into the atmosphere by the precipitating electrons causing the aurora. The MSISE-90 data is for altitudes from 0 to 500 km with a spatial resolution of 2 km.





Figure 6.2 All-sky camera images from the night of February 6-7, 1994, taken at (clockwise from top left) 19:05 LST, 19:55 LST, and 00:55 LST.

The MSISE-90 density profile of the atmosphere is used to determine the expected Rayleigh scattering component of the expected lidar signal by means of equation 3.3. Other lidar system parameters are given in Table 6.1. In the N_2^+ (0, 0) band lidar simulation, only two terms differ from those in Table 6.1: the operational wavelength λ is 390.303 nm and the transmittance T is 0.67. The factors that change in the model between the two vibrational-rotational bands of N_2^+ considered for resonance fluorescence lidar studies are discussed further in the next section.



Figure 6.3 Temperature profile from MSISE-90 atmospheric model used in the lidar model. (MSISE-90: http://modelweb.gsfc.nasa.gov/models/miss.html).

Parameter	Symbol	Value
System efficiency	η	0.05
One-way atmospheric transmittance	Т	0.75
Laser energy per pulse	E_L	50 mJ
Laser pulse repetition rate	RL	20 pps
Laser wavelength	λ	426.474 nm
Laser linewidth	Δλ	1.0 pm
Lower rotational state	J_2	9
Upper rotational state	J_1	10
Honl-London factor	Н	0.48
Telescope diameter	$(A_R/\pi)^{1/2}$	1.02 m
Field-of-view	$\Delta \theta_R$	0.5 mrad
Filter bandwidth	BW	0.3 nm
Measurement temporal resolution	Δt	300 s
Measurement altitude resolution	Δz	6000 m

Table 6.1 N_2^+ (1, 0) band lidar simulation system parameters.

The density of aurorally produced N_2^+ ($\rho_{N_2^+}(z)$) is derived in time and altitude by an auroral chemistry model which simulates auroral precipitation and the resultant modification of the thermosphere [Lummerzheim, 1987; Lummerzheim et al., 1989]. The auroral chemistry model consists of an electron transport code, a chemistry and diffusion code, and an energy balance code. The simulation uses successive MSP optical measurements, averaged together and smoothed with a 5 minute running average, to determine the auroral conditions, total energy flux, and characteristic energy of the incident electrons for the night of February 6-7, 1994. The electron characteristics determined from the MSP measurements drive the auroral chemistry model. Other inputs into the auroral chemistry model include the concentration of neutral particles as a function of altitude, the energy spectrum of the precipitating particles, the incident solar EUV (extreme ultra-violet) flux, and the downward heat flux in the thermosphere [Su, 2007]. Unlike the lidar model, the auroral chemistry model resolves the fluctuations in the atmospheric temperature profile in time. The auroral chemistry model, therefore, accounts for the heating of the atmosphere due to dissipation of the energy input into the atmosphere by the precipitating electrons which are causing the aurora. The time dependent chemistry code is coupled with the electron transport code to determine the auroral emissions that eventually result from the electrons incident in the upper atmosphere. The model output includes excitation rates for all excited neutral and ionic states for which cross sections are known, electron and ion temperatures, and the concentration and composition of the ionospheric plasma through the night. The model calculation of N_2^+ density and the ion excitation rate for the observations of February 6-7, 1994, are plotted in Figures 6.3 and 6.4. The spatial resolution of the auroral model output is 2 km with a temporal resolution of 300 s. The layer of N_2^+ is visible between 100 and 300 km with the peak density (8.17x10³ cm⁻³) occurring at an altitude of 166 km at 19:05 LST. The ionization rate peaks at 100 km with a value of $3.62x10^5$ cm⁻³ s⁻¹ at

output is 2 km with a temporal resolution of 300 s. The layer of N_2^+ is visible between 100 and 300 km with the peak density (8.17x10³ cm⁻³) occurring at an altitude of 166 km at 19:05 LST. The ionization rate peaks at 100 km with a value of 3.62x10⁵ cm⁻³ s⁻¹ at 00:55 LST. The average N_2^+ density profile is plotted in Figure 6.5. The population of the lowest vibrational level (v''=0) is taken as 0.3 of the total population of N_2^+ ions [Morrill and Benesch, 1996]. The N_2^+ density peaks when the ionization rate is relatively high over the broadest range of altitudes at ~19:00 LST, ~1:00 LST, and ~5:00 LST. The density of N_2^+ through the night is highly variable, corresponding to the variability in the level of auroral activity. The variance in N_2^+ determines the strength of the expected resonance scatter through the night. Stronger resonance scatter is expected at the times of greatest ionization and the subsequent production of N_2^+ .

The background lidar signal is determined by use of the MSP measurements of photon flux in time and is due to emissions within the aurora occurring at a similar wavelength as the resonance wavelength at which the laser in the lidar transmitter system is operating. The photon flux density measured by the MSP is the optical auroral emissions integrated over all altitudes. The MSP signal also includes any other light sources emitting in the wavelength range of the MSP filter's sensitivity, such as starlight. The contribution to the total background photon counts from sources other than auroral emissions is considered to be insignificant. The measured photon flux density is only a fraction of the total flux density from the (1, 0) band due to the transmission function of the bandpass filter used. To determine what fraction of the entire band emission is measured by the MSP due to its bandpass filter, the entire vibrational-rotational line spectrum at each altitude is calculated from the spectral reference data for the N_2^+ (1, 0) transition, the ionization rate in time and altitude from the auroral chemistry model, and the MSISE-90 atmospheric temperature profile, which is constant for each altitude bin in time. The spectrum varies in altitude with the variance of temperature and varies in time due to the variation in the altitude of emission, which is due to the variance with altitude of the ionization rate. The derived spectrum is combined with the known properties of bandpass filter used in the MSP to determine what percentage of the entire spectrum is being measured at each time point. By determining what percentage of the entire spectrum is being transmitted at each time point, the MSP photon flux measurements are corrected for the effects of the filter, yielding the total optical emissions from the ~428 nm band in time. The expected background lidar signal in time is then found by applying the filtering characteristics of the bandpass filter used in the lidar receiver system to the derived total optical emissions from the ~428 nm band. The expected background photon counts from the (1, 0) band of N_2^+ emissions in time is plotted in Figure 6.6.



Figure 6.4 N_2^+ density (ions cm⁻³) as a function of local time and altitude in false color. The contour levels are 1, 10, 100, and 1000.



Figure 6.5 N_2^+ ionization rate (ions cm⁻³ s⁻¹) as a function of local time and altitude in false color. The contour levels are 1, 10, 100, and 1000.



Figure 6.6 Average N_2^+ density profile for February 6-7, 1994.



Figure 6.7 Background lidar signal for N₂⁺ (1, 0) band resonance lidar system determined from MSP measurements on the night of February 6-7, 1994.

Realistically, the background lidar signal would also vary in time with temperature, as the temperature profile is not constant through the night (as it is considered to be in the model) due to the heating caused by the precipitating electrons. To investigate the change in the expected background lidar signal, 25 K is added to the MSISE-90 temperature profile at all altitudes. 25 K is an exaggeration of the atmospheric heating expected for this level of auroral activity and physical auroral heating would not be evenly distributed at all altitudes over the time scales considered. The ratio of the background lidar signal derived from the MSISE-90 temperature profile for 00:00 LST on February 6-7, 1994, and the background lidar signal derived from a temperature profile to which 25 K has been added at all altitudes is plotted in time in Figure 6.8. The maximum value of this ratio is 1.012, while the minimum is 0.958, and the average ratio is 0.997. It is found that the background lidar signal varies the most with increasing temperature for the period where the ionization rate is the strongest. Therefore, the modeled increase in the background signal during periods of heightened auroral activity would be even greater if the model also included the temperature increase through time due to the heating caused by precipitating electrons. As these time periods also correspond to the times where the N_2^+ density is the greatest and the highest resonance scattering signals will be measured, the statistical significance of the peak resonance lidar measurements would be lower in reality than they are in the model.



Figure 6.8 The ratio of the background lidar signal and the background lidar signal when the temperature has been increased by 25 K at all altitudes.

6.2 Modification of simulation for N_2^+ (1, 0) band lidar measurements

Su presented model results for a lidar system operating at 390.303 nm, the wavelength corresponding to the strongest rotational transition between the lowest vibrational levels of the X-B band in N_2^+ at 500 K. Based on the persistently low signals obtained from the resonance lidar system operating at 390 nm, and the relatively good performance seen when operating at wavelengths near 428 nm, the simulation was modified to model the performance of an N_2^+ resonance lidar system operating at 426.474 nm, the wavelength corresponding to the strongest rotational transition in the (0, 1) band for an atmospheric temperature of 500 K (only wavelengths derived from the Laher and Gilmore [1991] molecular constants are considered in the lidar simulation). By examining the model output for a lidar system operating at ~428 nm, the capabilities of this system in obtaining statistically significant measurements may be evaluated. Such an analysis will provide a useful indicator as to the likelihood of successful resonance lidar measurements of N_2^+ in the (1, 0) band of transitions.

The expected lidar signal determined by the model changes when considering the (1, 0) band of transitions due to several factors. Firstly, the spontaneous emission coefficient is three times smaller for the (1, 0) band than for the (0, 0) band, meaning the (1, 0) band excited states are three times longer lived ($A_{00}=1.14 \times 10^7 \text{ s}^{-1}$, $A_{10}=3.71 \times 10^6 \text{ s}^{-1}$, $\tau_{00}=8.77 \times 10^{-8} \text{ s}$, $\tau_{10}=2.70 \times 10^{-7} \text{ s}$ [Gilmore et al., 1992; Collins et al., 1997]). As the effective absorption cross section is proportional to the spontaneous emission coefficient, the effective scattering cross section is smaller for this band, resulting in a decrease in the expected resonance lidar signal. The longer lifetime of the (0, 1) transitions would also limit the minimum resolution of the lidar measurements, as the distance the light travels in the lifetime of the excited state increases with longer lifetimes. The transmittance of the atmosphere also varies from 0.67 for ~390 nm light to 0.75 for ~428 nm light (these values where obtained from the MODTRAN® (MODerate resolution atmospheric TRANsmission) atmospheric model under subarctic conditions) [Berk et al., 1989].

The expected background lidar signal will also decrease, as the spectrum of emission from this band of N_2^+ is not as bright as the (0, 0) band emissions. Su [2007] used the derived value of brightness for the (1, 0) band to determine the value of the total brightness from the (0, 0) band by means of the relation

$$\mathbf{K}_{00} = \mathbf{K}_{10} \frac{\mathbf{A}_{00}}{\mathbf{A}_{10}} \tag{6.2}$$

where K_{00} is the true brightness of the (0, 0) band, and K_{10} is the true brightness of the (1, 0) band. The background signal derived from the MSP data does not need to be scaled for the (1, 0) band measurements as the MSP data is obtained for the band emissions of interest in deriving the background lidar signal.

6.3 Results of simulation of (1, 0) band $\,N_2^{\,*}\,$ resonance lidar system

The resonance lidar parameters used to obtain the simulated measurements of N_2^+ are tabulated in Table 6.1. The spatial resolution of the model output is 6 km and the temporal resolution is 300 s. The component of the lidar signal due only to resonance scatter from the (1, 0) band of N_2^+ is plotted in Figure 6.8. The resonance lidar signal

profile integrated over the observation period is plotted in Figure 6.9. The total Rayleigh scattering component of the lidar signal is shown in Figure 6.10. Combination of the resonance scattering and Rayleigh scattering signals with the background lidar signal yields the total lidar signal, which is plotted in both altitude and time in Figure 6.11. The total lidar signal is dominated by the background signal, appearing as vertical stripes, above ~100 km, and by the Rayleigh scatter below 80 km. The SNR (signal-to-noise ratio) of the lidar signal in altitude and time is plotted in Figure 6.12. The total resonance lidar signal profile, not including the background signal, integrated over the entire observation period (18:15 to 6:55 LST, 12 hours and 40 minutes) is plotted in Figure 6.13, along with the corresponding total SNR. The SNR is given by the relation

$$SNR = \frac{N_s}{\sqrt{N_{TOT}}}$$
(6.3)

where N_{TOT} and N_S represent the total lidar signal (equation 3.1) and the resonance lidar signal (equation 6.1). The SNR is used to determine the statistical significance of the lidar measurements.

The peak signal from the N_2^+ layer for the total lidar profile (the resonance signal plus the Rayleigh and background signals, integrated over the observation period) is 57298 photon counts with a background signal of 55088 photon counts (2210 resonance counts), occurring at an altitude of 126 km. The associated SNR is 9.23.

For comparison, the total resonance lidar signal profile, not including the background signal, integrated over the entire observation period for the N_2^+ (0, 0) band resonance lidar system operating at 390.303 nm [Su, 2007] is plotted in Figure 6.13, along with the corresponding SNR values. The peak signal for the total lidar profile is 199130 photon counts with a background signal of 187943 photon counts (11187 resonance counts). The associated SNR is 25.07. The total resonance counts from the N_2^+ (1, 0) band resonance lidar system operating at 426.474 nm is ~5 times smaller than the total resonance counts from the (0, 0) band system.



Figure 6.9 N_2^+ (1, 0) resonance lidar signal (photon counts) in time and altitude (false color). The contour levels are at 0 and 10.



Figure 6.10 $N_2^+(1, 0)$ total resonance lidar signal integrated over the observation period (left) and the corresponding SNR (right).



Figure 6.11 $N_2^+(1, 0)$ total Rayleigh scattering signal integrated over the observation period.



Figure 6.12 N_2^+ (1, 0) total lidar signal (photon counts) in time and altitude (false color). The contour levels are at 1, 10, 100, and 1000.



Figure 6.13 N_2^+ (1, 0) SNR in time and altitude (false color). The contour levels are at 1, 10, 100, and 1000.



Figure 6.14 N₂⁺ (0, 0) total resonance lidar signal integrated over the observation period (left) and the corresponding SNR (right).

The N_2^+ (1, 0) band resonance signal has a maximum value of 69 photon counts at an altitude of 114 km at 00:55 LST (corresponding to the time of peak ionization). The maximum SNR of 1.95 occurs at an altitude of 126 km at 19:05 LST (corresponding to the time of the peak N_2^+ density), while the minimum SNR of 0.302 occurs at an altitude of 162 km at 19:55 LST. The fact that the maximum and minimum SNRs are found within 1 hour of each other and at altitudes separated by 48 km highlights the temporal and spatial variability that may be expected in auroral lidar measurements [Su, 2007]. The maximum resonance signal and maximum and minimum SNRs of the N_2^+ (0, 0) band lidar system occur at the same times and altitudes but have values of 341 photon counts, 5.32, and 0.85, respectively. The total lidar signal (N_{TOT}), resonance lidar signal (Ns), background signal (N_B), and associated SNRs at the times of maximum SNR, minimum SNR, and maximum resonance signal (19:05 LST, 19:55 LST, and 00:55 LST, respectively) are tabulated for the N_2^+ (1, 0) band and (for comparison) the (0,0) band resonance lidar systems in Table 6.2. The total lidar signal (N_{TOT}) for the measurements at 426.474 nm is a consistent factor of ~3.5 less than the total lidar signal at 390.303 nm. The resonance signal is 5 times smaller for the $(1, 0) N_2^+$ resonance lidar system, and the SNRs are ~3 times less. The resonance signal profiles and corresponding SNRs for each of the three time periods are plotted in Figure 6.14 for the N_2^+ (1, 0) band resonance lidar system. For comparison, the correspondent figures for the N_2^+ (0, 0) band resonance lidar system are plotted in Figure 6.15.

The simulated resonance lidar system is therefore capable, under the lidar parameters defined, of obtaining statistically significant measurements of aurorally produced N_2^+ in both the (0, 0) and (1, 0) bands. However, the measurements of the (1, 0) band yield much lower resonance signals, which have lower associated SNRs, than the measurements of the (0, 0) band. This simulation has also assumed ideal lidar system parameters, such as energy per laser pulse (E_L) of 50 mJ. The peak output measured for a 390 nm laser dye at ~390 nm was ~17 mJ (Exalite 392A). The peak energy output of Exalite 428 dye (at 428 nm) is ~15 mJ. To determine the performance that can be

expected from the resonance lidar system operating under current system parameters, we next determine the expected resonance lidar signal from a system operating with an energy per laser pulse, E_L , of 15 mJ.

N_{2}^{+} (1, 0) lidar	Max. SNR	Min. SNR	Max N _S
LST (hour:minute)	19:05	19:55	00:55
Altitude (km)	126	162	114
N _{TOT}	793	134	2544
N_S	55	3.5	69
N_B	738	130.5	2475
SNR	1.95	0.30	1.36

Table 6.2 Comparison of (1, 0) and (0, 0) $N_2^{\scriptscriptstyle +}\,$ resonance lidar system counts.

N ₂ ⁺ (0,0) lidar	Max. SNR	Min. SNR	Max N _S
LST (hour:minute)	19:05	19:55	00:55
Altitude (km)	126	162	114
N _{TOT}	2774	452	8898
N_S	280	18	341
N_B	2494	434	8557
SNR	5.32	0.85	3.62



Figure 6.15 (1, 0) system resonance signal (left) and associated SNR (right) at 19:05, 19:55, and 00:55 LST.



Figure 6.16 (0,0) sytem resonance signal (left) and associated SNR (right) at 19:05, 19:55, and 00:55 LST.

6.4 Scaling model results to measured lidar system performance

To assess the N_2^+ resonance signal that could potentially be measured by the current resonance lidar system, the simulation of the $N_2^+(1, 0)$ band resonance lidar system was modified to include the current measured value of E_L for the Exalite 428 dye laser (~15 mJ). This energy per laser pulse is over 3 times lower than the 50 mJ previously used in the simulations, resulting in a ~3 times lower total resonance signal. When the simulation is run for these conditions (the resolution remains at 6 km and 300 s, only E_L is changed), the $N_2^{\scriptscriptstyle +}$ (1, 0) band resonance signal has a maximum value of 21 photon counts at an altitude of 114 km at 00:55 LST. The maximum SNR of 0.60 occurs at an altitude of 126 km at 19:05 LST, while the minimum SNR of 0.091 occurs at an altitude of 162 km at 19:55 LST. These results are tabulated in Table 6.3. The total resonance lidar signal profile, not including the background signal, integrated over the entire observation period (18:15-6:55, 12 hours and 40 minutes) is plotted in Figure 6.17, along with the corresponding SNR values. The peak signal from the $N_2^{\scriptscriptstyle +}$ layer for the total lidar profile (resonance signal plus background signal integrated over the observation period) is 55751 photon counts with a background signal of 55088 photon counts (663 resonance counts), occurring at an altitude of 126 km. The associated SNR is 2.81. The total lidar profile (including the background signal) is plotted in Figure 6.18: Note that the resonance signal above 100 km is barely discernible.

$\mathbf{N}_{2}^{\scriptscriptstyle +}$ (1, 0) lidar	Max. SNR	Min. SNR	Max N _s
LST (hour:minute)	19:05	19:55	00:55
Altitude (km)	126	162	114
N _{TOT}	755	132	2496
N_s	16	1	21
N_B	739	131	2475
SNR	0.60	0.091	0.41

Table 6.3 Modeled N_2^+ (1, 0) band resonance lidar measurements when E_L =15 mJ.



Figure 6.17 N_2^+ (1, 0) total resonance lidar signal integrated over the observation period (left) and the corresponding SNR (right) for E_L =15 mJ.



Figure 6.18 N_2^+ (1, 0) total lidar signal profile for E_L =15 mJ.

The model results appearing in Figure 6.17 and 6.18 are representative of the actual lidar measurements the current resonance lidar system could be capable of obtaining given moderate auroral activity, precise tuning to the optimum resonance line, and similar lidar system parameters as those listed in Table 6.1. These measurements are
not of acceptable statistical significance. To achieve statistically significant measurements, the resolution of the lidar signal data must be decreased. However, the spatial and temporal resolutions cannot be simply increased without losing the scientific results of the data. Increasing the spatial resolution results in a significant loss in the resonance signal contribution of the total signal profile, as the strong Rayleigh signal overpowers the weak resonance signal as they are averaged together [Figure 6.19]. Increasing the temporal resolution has a similar effect, as the auroral activity creating N_2^+ is highly temporally and spatially variable. By increasing the temporal resolution from 300 s to 900 s, the fine scale temporal variability of the aurora is no longer evident in the lidar measurement [Figure 6.20]. Due to the large number of parameters that may be varied, many possible model outcomes may be explored. The altitude resolution could be reduced and the temporal resolution increased to highlight temporal variations in the aurora, and vice versa. Operationally, as is possible in the model, the lidar measurements would be obtained at high resolution and post integrated in selected altitude and time ranges to yield statistically significant measurements.



Figure 6.19 N₂⁺ (1, 0) resonance lidar signal in time and altitude (false color). Spatial resolution increased to 30 km. The contour levels are at 1, 10, 100, and 1000.



Figure 6.20 N₂⁺ (1, 0) resonance lidar signal in time and altitude (false color). Temporal resolution increased to 900 s. The contour levels are at 1, 10, 100, and 1000.

As an example of post integration of the modeled lidar signal obtained at high resolution, a 30 km range, centered on the respective altitude where the maximum SNR, minimum SNR, and maximum resonance signal occur is chosen. For the (1, 0) band model results for an E_L of 15 mJ, the resonance signal (N_s) and total signal (N_{TOT}) are summed in these altitude ranges for each time considered and the associated SNR is determined. This method alters the resolution of the measurement, but only in the height range of interest, so as to decrease the amount of resonance signal corruption by the strong Rayleigh scattering signal. These results are tabulated in Table 6.4.

N_{2}^{+} (1, 0) lidar	Max. SNR	Min. SNR	Max N _s
LST (hour:minute)	19:05	19:55	00:55
Avg. altitude (km)	126	162	114
N _{TOT}	3768	659	12467
N_s	77	5	89
N_B	3691	654	12378
SNR	1.25	0.19	0.80

Table 6.4 Modeled N_2^+ (1, 0) band resonance lidar measurements obtained at 6 km, 300 s resolution, integrated over a 30 km height range centered on the altitude of peak signal for E_L =15 mJ.

The N_2^+ (1, 0) band resonance signal has a maximum value of 89 photon counts in the altitude range 102 to 126 km at 00:55 LST. The maximum SNR of 1.25 occurs in the altitude range of 114 to 138 km at 19:05 LST, while the minimum SNR of 0.19 occurs in the altitude range of 150 to 174 km at 19:55 LST. The peak signal from the N_2^+ layer for the total lidar profile in the 114 to 138 km range is 278590 photon counts with a background signal of 275438 photon counts (3152 resonance counts). The associated SNR has more than doubled from the 6 km resolution analysis with a value of 5.97.

The statistical significance of the N_2^+ (1, 0) band resonance lidar measurements for $E_L=15$ mJ may be further improved for the time period containing the maximum resonance signal by integrating over a specific time range within the night. The value of the maximum SNR in altitude through time is plotted in Figure 6.21, along with the altitude at which the maximum value occurs. The time period 00:55 to 2:00 LST is chosen to integrate over, as the average SNR is relatively high in this time range. Furthermore, the altitude at which the maximum SNR occurs over this time period is also relatively constant, indicating that integrating over this time period and height range will not drastically distort the signal in altitude. The lidar signals measured in this time range at the altitude of the maximum resonance signal are tabulated in Table 6.5.



Figure 6.21 Maximum SNR and altitude at which this value occurs through the night of February 6-7, 1994.

Table 6.5 N_2^+ (1, 0)	band resonance lidar measurements integrated over 00:55 to 2:00 LST.
	E _L =15 mJ.

${ m N}_2^{\scriptscriptstyle +}$ (1, 0) lidar signals at 114 km	00:55-2:00 LST	
N _{TOT}	14289	
Ns	159	
N_B	14130	
SNR	1.33	

Next, the lidar signal integrated over the selected time range is integrated over a 30 km altitude range centered on the altitude of peak resonance at 114 km. Doing so yields values for N_{TOT} , N_S , N_B of 71339, 689, and 70650 photon counts, respectively. The associated value of the SNR is 2.58. This value of the SNR is over 4 times greater than the maximum SNR occurring at 19:05 LST in the high resolution resonance lidar data for the (1, 0) band model with E_L =15 mJ. The SNR value for the modeled lidar signal

achieved by this method of post integration is statistically significant while maintaining the integrity of the resonance lidar measurements in altitude and time. Lidar measurements of aurorally excited N_2^+ experimentally obtained would be processed in a similar manner to achieve statistically significant results.

6.5 Comparison of modeled signal to signal achieved with Exalite 428 dye laser

To compare the modeled resonance lidar signal to the current signal measurements, the simulated Rayleigh lidar signal from the N_2^+ (1, 0) band resonance lidar system is compared to the measured Rayleigh signal from the Exalite 428 resonance lidar system on September 15, 2009 (refer to section 4.6). The Rayleigh signals in the 19.5 to 25.5 km altitude range are compared. The modeled signal from the 19.5 to 25.5 km altitude range is 5551 photon counts per shot from a laser beam tuned to 426.474 nm having an E_L of 50 mJ per pulse. The energy per pulse of the Exalite 428 laser beam tuned to a wavelength of 428 nm was measured to be 15.6 mJ on September 15, 2009. When the modeled lidar signal is scaled to this measured value of E_L, the Rayleigh scattering signal from the 19.5 to 25.5 km altitude range decreases to 2 photon counts per shot. The lidar signals measured on September 15, 2009, at wavelengths from 425.50 nm to 429.50 nm in the 19.5 to 25.5 km and 29.5 to 35.5 km height ranges are plotted in Figure 6.22 and tabulated in Table 6.6. (The NICT Rayleigh lidar signals from this night were similarly re-binned to allow the resonance lidar signal to be compensated for decreased atmospheric transmission.) When the Exalite 428 dye laser is tuned to 425.50 and 427.50 nm, ~5 photon counts per shot are measured in the 19.5 to 25.5 km altitude range. In general, however, signals below 1 photon count per laser pulse are recorded.

Based on the measured photon counts per laser pulse resulting from the Exalite 428 dye laser beam, the current resonance lidar system may be capable of making statistically significant measurements of aurorally produced N_2^+ . Such measurements would be obtained at high resolution, allowing for careful signal processing in the data analysis process (as was discussed in the previous section). Time ranges of stable auroral activity, where the SNRs remain relatively stable, would be ideal for experimenting with

signal processing methods. Temporal and spatial resolutions would be determined on a case-by-case basis.



Figure 6.22 Compensated signal from 19.5-25.5 km and 29.5-35.5 km altitude ranges for Exalite 428 dye laser on September 15, 2009.

	J	-
Wavelength (nm)	Compensated signal 19.5-25.5 km	Compensated signal 29.5-35.5 km
425.50	4.68	0.25
426.50	1.80	0.12
427.50	4.83	0.39
428.50	0.89	0.08
429.50	0.026	0.48

 Table 6.6 Compensated signals for September 15, 2009 lidar measurements in two altitude ranges obtained with Exalite 428 dye laser.

7 Conclusions and Further Work

Obtaining resonance fluorescence lidar measurements of aurorally produced N_2^+ is made challenging by both the operational performance of the resonance lidar system used at PFRR and the high degree of geophysical variability inherent in the aurora. In the course of the study presented in this dissertation, we have field tested and analyzed key components of the resonance lidar system, as well as theoretically investigating the use of this system in obtaining statistically significant measurements of N_2^+ .

The improved performance of the PFRR resonance lidar system following a realignment of the laser cavity optics has been verified by obtaining and analyzing sodium and iron resonance lidar measurements. A 1.02 m diameter telescope has successfully been integrated into the lidar receiver system and is yielding an increase in the strength and quality of the lidar system's measurements.

Ten laser dyes have been examined in the dye laser for use in resonance lidar measurements of N_2^+ . Six of these dyes resulted in a decrease in measured lidar signal when the dye laser was tuned to 390 nm, the wavelength necessary for lidar measurements of the (0, 0) band of N_2^+ , near the peak of their optical emission. Spectroscopic measurements of two of these dyes showed a significant amount of spontaneous emission at the peak of the dye gain when operating at wavelengths where the dye emission is relatively weak. Consultation with a researcher in the field of laser dye development could yield further insight into the observed dye performance. The spectroscopic evidence does not support the conclusion that all the 390 nm dyes fail at 390 nm due to amplified spontaneous emission. We conclude that the lack of lidar system performance at 390 nm is likely due to the proximity of this wavelength to a change in the diffraction grating order. Light with a 390 nm wavelength falls on the edge of a grating order and a marked increase in the measured energy of the laser beam is observed immediately following a transition from this order to the next. Based on the lack of performance for the lidar system operating at wavelengths near and at 390 nm,

measurements of N_2^+ at operational wavelengths near 428 nm (corresponding the (1, 0) band of transitions within N_2^+) are being pursued.

Of the four dyes tested which fluoresce over a range of wavelengths relevant for lidar measurements of the (1, 0) band (centered at ~428 nm) of N_2^+ , Exalite 428 was chosen for pursued study due to its relatively high level of sky performance. On December 1, 2008, Rayleigh scattering signals when the laser was tuned to 428 nm measured in the 20 to 25 km and 30 to 35 km altitude ranges were approximately half of the signal levels measured in the same altitude ranges for the iron resonance lidar system (operating with the 1.02 m diameter telescope in the receiver system). On September 15, 2009, however, much lower signals were measured for roughly the same lidar operational parameters. Further sky tests with the Exalite 428 dye laser beam are warranted to confirm the performance of this system and assess its capabilities regarding obtaining successful measurements of N_2^+ . Further sky tests of some of the other 428 nm laser dyes are required, given the seemingly unstable performance of Exalite 428 thus far.

Another tunable laser system may also be considered for conducting resonance fluorescence lidar measurements of aurorally produce N_2^+ . A solid state alexandrite laser may be an alternative to use of a dye laser. Alexandrite lasers are high powered and tunable, fluorescing in the range of 730 to 810 nm at room temperatures [Hecht, 1992]. Frequency doubling methods would be used to achieve the wavelengths necessary for N_2^+ resonance lidar studies. It is unlikely that the laser beam strength would be similarly attenuated for operational wavelengths near 390 nm in another laser system. A resonance lidar system for profiling atomic iron at the Leibniz-Institute of Atmospheric Physics in Kühlungsborn, Germany, uses a frequency-doubled alexandrite laser with a pulse energy of 85 mJ at a wavelength of 386 nm [Lautenbach and Höffner, 2004]. As this operational wavelength is near to 390 nm, it is likely that use of a similar system would make it possible to obtain measurements of the (0, 0) band of transitions within N_2^+ . It is desirous to obtain measurements of the (0, 0) band, rather than the (0, 1) band, as these transitions are of greater strength, yielding a higher resonance lidar signal.

The performance of an operational N₂⁺ resonance lidar system operating at 426.474 nm, causing resonance fluorescence in the (1, 0) band of transitions, and at 390.303 nm, causing resonance fluorescence in the (0, 0) band of transitions, has been simulated. For the same set of lidar operation parameters used by Su [2007], the simulation of the expected lidar signal from the (1, 0) band yields resonance signals ~ 5 times lower and SNRs ~3 times lower. The decrease in performance is due to the decreased strength of resonance scatter within the (1, 0) band. The SNR corresponding to the peak signal of the total lidar signal profile (integrated over the entire observation period) is ~26 for the (0, 0) band lidar system and ~9 for (1, 0) band lidar system. When the lidar parameters are changed to reflect the current operational laser energy per pulse (E_L) of the resonance lidar system operating with an Exalite 428 dye laser beam, the SNR of the total lidar profile decreases to \sim 3, with a maximum SNR of only 0.60 at the time of peak ionization (19:05 LST) corresponding to high levels of auroral activity. To operationally achieve lidar measurements of similar quality, we find that at least 2 photon counts per laser pulse from the 19.5 to 25.5 km altitude range are necessary. The resonance lidar system operating with Exalite 428 dye may be capable of yielding this signal level; Further sky tests of the Exalite 428 resonance lidar system are needed. However, the SNRs yielded from the system operating with a laser energy per pulse of 15 mJ are too low to be considered statistically significant. However, post integration methods of processing the modeled lidar data, obtained at high resolution, over selected time and altitude ranges have been shown to be capable of yielding statistically significant results. The temporal and spatial ranges over which the data is integrated must account for the high degree of variability in the auroral activity, which affects the density of N_2^+ and the background skylight conditions. Operationally, the resolution of auroral lidar measurements would also be adapted to signal levels in altitude and time to yield statistically significant data.

Obtaining measurements of an aurorally excited molecular species could yield significant data for use in fundamental upper atmospheric studies and space weather research. Space weather affects a wide variety of aspects of human life, from day-to-day activities to space exploration. Much of the motivation for atmospheric research on charged species has also derived from the need to understand and provide secure communications in the near-Earth space environment [Prölss, 2004]. While the fundamental ionospheric processes are understood, space weather studies demand realtime, high resolution, and high accuracy knowledge of the Earth's near space environment. Atmospheric ions are central in the chemical interplay between species in the upper atmosphere. Knowledge of the concentrations of ions within a particular energy state during an auroral event could improve models and the understanding of energetics in the upper atmosphere. The long term goal of these studies is to establish rangeresolved measurements of specific electronic-vibrational-rotational states with accuracy and precision allowing complete characterization of the chemical and physical processes in the upper atmosphere. Species specific measurements could be combined with incoherent scatter radar data to yield a more comprehensive measurement of the upper atmospheric disturbance created by precipitation of auroral particles. On the broadest scale, measurements of this caliber will enhance current space weather studies, by reducing current uncertainties in the physics, chemistry and energetics of the ionosphere.

References

Angelopoulos, V., 2008: Tail Reconnection Triggering Substorm Onset, Science, 321, 931-935.

Akasofu, S., 2007: Exploring the Secrets of the Aurora, 2nd Ed., Springer, 288 pp.

Akasofu, S., 2009: The Northern Lights: Secrets of the Aurora Borealis, Alaska Northwest Books, 192 pp.

Atkins, P. W., 1991: Quanta: A Handbook of Concepts, Oxford University Press, 440 pp.

Baumjohann, W. and R. A. Treumann, 1997: Basic Space Plasma Physics, Imperial College Press, 329 pp.

Berk, A., Bernstein, L. S., and D. C. Robertson, 1989: MODTRAN: a moderate resolution model for LOWTRAN 7, AFGL-TR-89-0122, U.S. Air Force Geophysics Laboratory, Hanscom Air Force Base, Massachusetts.

Brackmann, U., 1997: Lambdachrome® Laser Dyes, 2nd Revised Edition, Lambda Physik GmbH, 284 pp.

Breese, J., 2001: Development of Fe Boltzmann Temperature Lidar, M.S. Thesis, University of Alaska Fairbanks, 98 pp.

Brown, J. M., 1998: Molecular Spectroscopy, Oxford University Press, 89 pp.

Cameron, R., Scholl, T. J., Zhang, L., Holt, R. A., and S. D. Rosner, 1995: Fast-Ion-Beam Laser Spectrscopy of the $B^2\Sigma_u^+ - X^2\Sigma_g^+$ and $B^2\Sigma_u^+ - A^2\Pi$ Systems of SiO⁺: Experiment, Journal of Molecular Spectroscopy, 169, 352-363. Carroll, B., and D. Ostlie, 1996: An Introduction to Modern Astrophysics, Addison-Wesley Publishing Company, Inc., 1400 pp.

Chu, X. and G. Papen, 2005: Fujii, T. and T. Fukuchi, editors, Laser Remote Sensing, Taylor and Francis, 888 pp.

Collins, R. L., Lummerzheim, D., and R. W. Smith, 1997: Analysis of lidar systems for profiling aurorally excited molecular species, Applied Optics, 36, 24, 6024-6034.

Collins, R. L., Kelley, M. C., Nicolls, M. J., Ramos, C., Hou, T, Stern, T. E., Mizutani, K., and T. Itabe, 2003: Simultaneous Lidar Observations of a Noctilucent Cloud and an Internal Wave in the Polar Mesosphere, Journal of Geophysical Research, 108, D8, 8435.

CRC Handbook of Chemistry and Physics, 87th edition, 2006: David R. Lide, editor, CRC Press, variable pagination.

Dëmtroder, W., 2003: Laser Spectroscopy: Basic Concepts and Instrumentation, 3rd Edition, Springer, 987 pp.

Drexhage, K. H., 1990: Schäfer, F. P., editor, Topics in Applied Physics, Volume 1: Dye Lasers, 3rd Enlarged and Revised Edition, Springer-Verlag, 244 pp.

Exciton, 2006: Laser dye data sheets, http://www.exciton.com/wavelength_chart.html, first accessed website spring 2008.

Falthammar, C., 1999: Space Physics, Alfven Laboratory, 173 pp.

Feldman, P., and J. Doering, 1975: Auroral Electrons and Optical Emissions of Nitrogen, Journal of Geophysical Research, 80, 19, 2808-2812.

Freedman, R. A. and W. J. Kauffman, 2002: Universe, W.H. Freeman and Company, 800 pp.

Fuhr, J. R., Martin, G. A., and W. L. Weiss, 1988: Atomic Transition Probabilities Iron through Nickel, Journal of Physical and Chemical Reference Data, 17, Supplement 4, 493 pp.

Garner, R. C., and P. Dao, 1995: Molecular nitrogen fluorescence lidar for remote sensing of the auroral ionosphere, Journal of Geophysical Research, 100, 74, 131-140.

Gilmore, L., Laher, R., and P. Espy, 1992: Franck-Condon Factors, r-Centroids, Electronic Transition Moments, and Einstein Coefficients for Many Nitrogen and Oxygen Band Systems, Journal of Physical and Chemical Reference Data, 21, 5, 1005-1107.

Grant, W. B., Browell, E. V., Menzies, R. T., Sassen, K., and C. Y. She, editors, 1997: Selected Papers on Laser Applications in Remote Sensing, SPIE Milestone Series, MS 141, SPIE Optical Engineering Press, 662 pp.

Griffiths, D. J., 1999: Introduction to Electrodynamics, 3rd Edition, Prentice Hall, 576 pp.

Halliday, D., Resnick, R., and J. Walker, 2001: Fundamentals of Physics, 6th Edition, John Wiley and Sons, 1144 pp.

Hecht, J., 1992: The Laser Guidebook, 2nd Edition, McGraw-Hill, 498 pp.

Herzberg, G., 1950: Molecular Spectra and Molecular Structure I. Spectra of Diatomic Molecules, 2nd Edition, D. Van Nostrand Company, 658 pp.

Hollas, J. M., 2004: Modern Spectrsocopy, 4th Edition, John Wiley and Sons, 422 pp.

Hou, T., 2002: Development of High Spectral Resolution Iron Boltzmann Lidar, M.S. Thesis, University of Alaska Fairbanks, 114 pp.

Jones, A. V., 1974: Aurora, D. Reidel Publishing Company, 301 pp.

Kallenrode, M., 2001: Space Physics: An Introduction to Plasmas and Particles in the Heliosphere and Magnetospheres, 2nd Edition, Springer-Verlag, 428 pp.

Krane, K., 1996: Modern Physics, 2nd Edition, John Wiley and Sons, 581 pp.

Laher, R., and F. R. Gilmore, 1991: Improved Fits for the Vibrational and Rotational Constants of Many States of Nitrogen and Oxygen, Journal of Physical and Chemical Reference Data, 20, 4, 685-712.

Lambda Physik, 1997: Dye laser SCANMate Instruction Manual, Lambda Physik Technik, 262 pp.

Lautenbach, J., and J. Höffner, 1988: Scanning iron temperature lidar for mesopause temperature observation, Applied optics, 43, 23, 4559-4563.

Lummerzheim, D., 1987: Electron Transport and Optical Emissions in the Aurora, Ph.D. Thesis, University of Alaska Fairbanks, 128 pp.

Lummerzheim, D., Rees, M., and H. Andersen, 1989: Angular dependent transport of auroral electrons in the upper atmosphere, Planetary and Space Science, 37, 109.

Morrill, J., and W. Benesch, 1996: Auroral N_2 emissions and the effect of collisional processes on N_2 triplet state vibrational populations, Journal of Geophysical Research, 101, A1, 261-274.

Morse, T. H. and G. J. Romick, 1982: The Fluctuation and fading of auroral arcs preceding auroral substrom onsets, Geophysical Research Letters, 9, 1065-1068.

National Institute of Standards and Technology (NIST), 2008: Atomic Spectra Database version 3.1.2, http://physics.nist.gov/PhysRefData/ASD/index.html, first accessed website September 2008.

Ocean Optics, 2009: Grating efficiency curves, http://www.oceanoptics.com/Products/ bench_gratingcharts.asp#Gratings_1,_2,_3,_4,_14,_H1,_H2,_H3,_H4,_H14%3EH3, first accessed website September 2009.

Pedrotti, F., and L. Pedrotti, 1993: Introduction to Optics, 2nd Edition, Prentice Hall, 602 pp.

Peshave, M., 2004: Design and Implementation of a Microcontroller-Based Closed-Loop Tuning-Controller for a Tunable Dye Laser, M.S. Thesis, University of Alaska Fairbanks.

Petrukovich, A. A., 2008: The Elusive Onset of Geomagnetic Substorms, Science, 321, 920-921.

Prölss, G. W., 2004: Physics of the Earth's Space Environment: An Introduction, Springer, 513 pp.

Rees, M. H., 1989: Physics and chemistry of the upper atmosphere, Cambridge University Press, 289 pp.

Sansonetti, C., Salit, M., and J. Reader, 1996: Wavelengths of spectral lines in mercury pencil lamps, Applied Optics, 35, 74-77.

Scholl, T. J., Holt, R. A., and S. D. Rosner, 1998: Fine and Hyperfine Structure in ${}^{14}N_{2}^{+}$: The B² Σ_{μ}^{+} –X² Σ_{μ}^{+} (0,0) Band, Journal of Molecular Spectroscopy, 192, 424-434.

Schäfer, F. P., 1990: F. P. Schäfer, editor, Topics in Applied Physics, Volume 1: Dye Lasers, 3rd Enlarged and Revised Edition, Springer-Verlag.

Schroeder, D. J., 2000: Astronomical Optics, Second Edition, Academic Press, 478 pp. She, C. Y., and J. R. Yu, 1995: Doppler-free saturation fluorescence spectroscopy of Na atoms for Atmospheric Applications, Applied Optics, 34, 1063-1075.

Su, L., 2007: Development of Resonance Fluorescence Lidar for Studies of the Aurora, Ph.D. Dissertation, University of Alaska Fairbanks, 187 pp.

University Corporation for Atmospheric Research (UCAR) Office of Programs, 2008: Cooperative Program for Operational Meteorology Education and Training (COMET), Physics of the Aurora: Earth Systems, Online module, http://www.meted.ucar.edu/zearch/zearch_results.php?hq=site%3Ameted.ucar.edu&cx=0 12446052473863902991%3Ar8nkgnwzzsc&cof=FORID%3A11&q=%22aurora+borealis

%22#0, first accessed website September 2008.

Wallace, J. M. and P. V. Hobbs, 2006: Atmospheric Science, 2nd Edition, Academic Press, 483 pp.

Wayne, R. P., 2000: Chemistry of Atmospheres, Oxford University Press, 775 pp.



Figure A.1 Energy output of QUI dye laser with wavelength.



Figure A.2 Signal from 20-25 km and 30-35 km altitude ranges for QUI dye laser.



Figure A.3 Energy output of Exalite 392A dye laser with wavelength.



Figure A.4 Signal from 20-25 km and 30-35 km altitude ranges for QUI dye laser.



Figure A.5 Energy output of BBQ dye laser with wavelength.



Figure A.6 Compensated signal from 20-25 km and 30-35 km altitude ranges for BBQ dye laser.



Figure A.7 Energy output of TBS dye laser with wavelength.



Figure A.8 Signal from 20-25 km and 30-35 km altitude ranges for TBS dye laser.



Figure A.9 Energy output of PBBO dye laser with wavelength.



Figure A.10 Compensated signal from 20-25 km and 30-35 km altitude ranges for PBBO dye laser.



Figure A.11 Energy output of Exalite 389 dye laser with wavelength.



Figure A.12 Compensated signal from 20-25 km and 30-35 km altitude ranges for Exalite 389 dye laser.



Figure A.13 Energy output of Exalite 417 dye laser with wavelength.



Figure A.14 Compensated signal from 20-25 km and 30-35 km altitude ranges for Exalite 417 dye laser.



Figure A.15 Energy output of Exalite 428 dye laser with wavelength.



Figure A.16 Compensated signal from 20-25 km and 30-35 km altitude ranges for Exalite 428 dye laser.



Figure A.17 Energy output of Bis-MSB dye laser with wavelength.



Figure A.18 Compensated signal from 20-25 km and 30-35 km altitude ranges for Bis-MSB dye laser.



Figure A.19 Energy output of POPOP dye laser with wavelength.



Figure A.20 Compensated signal from 20-25 km and 30-35 km altitude ranges for POPOP dye laser.

Appendix B

All wavelengths are given in units of nm. LG Wvl. and SHR Wvl. denote wavelengths calculated from the molecular constants reported by Laher and Gilmore [1991] and Scholl et al. [1998], respectively. Wvl. STP is the wavelength refracted at standard temperature and pressure. The strength reported is the relative strength of the particular transition. Only transitions with a relative strength greater than 1 are reported.

1=200 K, v'' =	=0, 0'=0					
LG Wvl.	SHR Wvl.	Wvl. STP	Strength	Pop.	J"	J'
390.695	390.694	390.58	100	10.1	5	6
390.604	390.603	390.49	98	10.1	6	7
390.781	390.781	390.67	95	9.44	4	5
390.508	390.507	390.40	93	9.56	7	8
390.863	390.863	390.75	85	8.2	3	4
390.408	390.406	390.30	83	8.69	8	9
390.303	390.301	390.19	72	7.57	9	10
390.940	390.940	390.83	69	6.36	2	3
390.194	390.192	390.08	60	6.34	10	11
391.013	391.013	390.90	49	4.04	1	2
390.080	390.078	389.97	48	5.13	11	12
389.962	389.959	389.85	37	4	12	13
389.839	389.836	389.73	28	3.01	13	14
391.081	391.081	390.97	25	1.38	0	1
389.712	389.708	389.60	20	2.2	14	15
389.580	389.576	389.47	14	1.55	15	16
389.443	389.439	389.33	9	1.06	16	17
389.302	389.297	389.19	6	0.7	17	18
389.157	389.151	389.05	4	0.45	18	19
389.007	389.001	388.90	2	0.28	19	20
388.853	388.846	388.74	1	0.17	20	21

T-200 V y'' = 0 y y' = 0

ŕ	·					
LG Wvl.	SHR Wvl.	Wvl. STP	Strength	Pop.	J"	J'
390.301	390.303	390.19	100	6.39	9	10
390.406	390.408	390.30	99	6.31	8	9
390.192	390.194	390.08	98	6.32	10	11
390.507	390.508	390.40	96	6.09	7	8
390.078	390.080	389.97	95	6.13	11	12
390.603	390.604	390.49	91	5.7	6	7
389.959	389.962	389.85	90	5.83	12	13
389.836	389.839	389.73	84	5.46	13	14
390.694	390.695	390.58	83	5.15	5	6
389.708	389.712	389.60	77	5.02	14	15
390.781	390.781	390.67	73	4.46	4	5
389.576	389.580	389.46	69	4.55	15	16
389.439	389.443	389.33	62	4.05	16	17
390.863	390.863	390.75	61	3.62	3	4
389.297	389.302	389.19	54	3.56	17	18
390.940	390.940	390.83	47	2.68	2	3
389.151	389.157	389.04	47	3.09	18	19
389.001	389.007	388.89	40	2.64	19	20
388.846	388.853	388.74	33	2.22	20	21
391.013	391.013	390.90	32	1.64	1	2
388.687	388.694	388.58	28	1.85	21	22
388.523	388.531	388.41	23	1.52	22	23
388.354	388.363	388.24	18	1.23	23	24
391.081	391.081	390.97	16	0.55	0	1
388.182	388.191	388.07	14	0.98	24	25
388.004	388.015	387.89	11	0.77	25	26
387.823	387.834	387.71	9	0.6	26	27

T=500 K, $\upsilon'' = 0, \upsilon' = 0$

387.637	387.649	387.53 7	0.46	27	28
387.446	387.459	387.34 5	0.35	28	29
387.251	387.265	387.14 4	0.27	29	30
387.052	387.067	386.94 2	0.2	30	31
386.849	386.864	386.74 2	0.14	31	32
386.641	386.657	386.53 1	0.1	32	33
386.429	386.446	386.32 1	0.07	33	34

I 1000 IK, 0	0,0 0					
LG Wvl.	SHR Wvl.	Wvl. STP	Strength	Pop.	J"	J'
389.839	389.836	389.73	100	4.51	13	14
389.962	389.959	389.85	99	4.49	12	13
389.712	389.708	389.60	99	4.49	14	15
390.080	390.078	389.97	98	4.42	11	12
389.580	389.576	389.46	97	4.41	15	16
390.194	390.192	390.08	95	4.28	10	11
389.443	389.439	389.33	94	4.3	16	17
390.303	390.301	390.19	92	4.1	9	10
389.302	389.297	389.19	91	4.15	17	18
389.157	389.151	389.04	87	3.97	18	19
390.408	390.406	390.30	87	3.85	8	9
389.007	389.001	388.89	82	3.77	19	20
390.508	390.507	390.40	80	3.55	7	8
388.853	388.846	388.74	77	3.55	20	21
390.604	390.603	390.49	73	3.2	6	7
388.694	388.687	388.58	72	3.31	21	22
388.531	388.523	388.41	67	3.07	22	23
390.695	390.694	390.58	65	2.8	5	6
388.363	388.354	388.24	61	2.82	23	24
388.191	388.182	388.07	56	2.58	24	25
390.781	390.781	390.67	55	2.36	4	5
388.015	388.004	387.89	50	2.34	25	26
387.834	387.823	387.71	45	2.1	26	27
390.863	390.863	390.75	45	1.87	3	4
387.649	387.637	387.53	40	1.88	27	28
387.459	387.446	387.34	36	1.67	28	29
390.940	390.940	390.83	34	1.36	2	3

387.265	387.251	387.14 31	1.47	29	30
387.067	387.052	386.94 27	1.29	30	31
386.864	386.849	386.74 24	1.12	31	32
391.013	391.013	390.90 23	0.83	1	2
386.657	386.641	386.53 21	0.97	32	33
386.446	386.429	386.32 18	0.83	33	34
386.230	386.212	386.10 15	0.71	34	35
386.011	385.991	385.88 13	0.6	35	36
391.081	391.081	390.97 11	0.28	0	1
385.787	385.766	385.66 10	0.51	36	37
385.558	385.537	385.43 9	0.42	37	38
385.326	385.303	385.197	0.35	38	39
385.089	385.066	384.96 6	0.29	39	40
384.848	384.824	384.71 5	0.24	40	41
384.603	384.577	384.47 4	0.2	41	42
384.354	384.327	384.22 3	0.16	42	43
384.100	384.072	383.96 2	0.13	43	44
383.843	383.814	383.70 2	0.1	44	45
383.581	383.551	383.44 1	0.08	45	46
383.316	383.284	383.17 1	0.07	46	47
383.046	383.013	382.90 1	0.05	47	48

1-200	$\mathbf{K}, \mathbf{U}^r = \mathbf{I}, \mathbf{U}^r = \mathbf{U}$					
LG Wv	l. SHR Wvl.	Wvl. STP	Strength	Pop.	J''	J
426.963	426.962	426.84	100	9.99	5	6
426.850) 426.848	426.73	98	10	6	7
427.069	427.068	426.95	95	9.37	4	5
426.731	426.729	426.61	93	9.54	7	8
427.170) 427.169	427.05	85	8.13	3	4
426.606	6 426.604	426.48	84	8.69	8	9
426.474	426.472	426.35	73	7.59	9	10
427.264	427.264	427.14	69	6.31	2	3
426.337	426.334	426.21	61	6.38	10	11
426.194	426.190	426.07	49	5.17	11	12
427.352	427.352	427.23	48	4	1	2
426.044	426.040	425.92	38	4.04	12	13
425.888	3 425.884	425.76	29	3.06	13	14
427.434	427.434	427.31	25	1.37	0	1
425.727	425.722	425.60	21	2.24	14	15
425.559	425.554	425.43	15	1.59	15	16
425.385	5 425.379	425.26	10	1.09	16	17
425.205	5 425.199	425.08	6	0.73	17	18
425.019	425.012	424.89	4	0.47	18	19
424.828	3 424.820	424.70	2	0.29	19	20
424.630) 424.621	424.50	1	0.18	20	21

T=200 K, $\upsilon'' = 1, \upsilon' = 0$

1 200 I I , 0	1,0 0					
LG Wvl.	SHR Wvl.	Wvl. STP	Strength	Pop.	J''	J'
426.474	426.472	426.35	100	6.36	9	10
426.606	426.604	426.48	99	6.28	8	9
426.337	426.334	426.21	98	6.30	10	11
426.731	426.729	426.61	96	6.05	7	8
426.194	426.190	426.07	95	6.11	11	12
426.850	426.848	426.73	91	5.66	6	7
426.044	426.040	425.92	90	5.83	12	13
425.888	425.884	425.76	84	5.46	13	14
426.963	426.962	426.84	83	5.11	5	6
425.727	425.722	425.60	77	5.03	14	15
427.069	427.068	426.95	73	4.42	4	5
425.559	425.554	425.43	70	4.56	15	16
425.385	425.379	425.26	62	4.07	16	17
427.170	427.169	427.05	61	3.59	3	4
425.205	425.199	425.08	55	3.59	17	18
425.019	425.012	424.89	47	3.11	18	19
427.264	427.264	427.14	47	2.65	2	3
424.828	424.820	424.70	40	2.67	19	20
424.630	424.621	424.50	34	2.25	20	21
427.352	427.352	427.23	32	1.63	1	2
424.426	424.417	424.30	28	1.88	21	22
424.216	424.206	424.09	23	1.54	22	23
424.001	423.990	423.87	19	1.25	23	24
427.434	427.434	427.31	16	0.550	0	1
423.779	423.768	423.65	15	1.00	24	25
423.552	423.539	423.42	12	0.790	25	26
423.319	423.305	423.19	9	0.620	26	27

423.080	423.065	422.95	7	0.480	27	28
422.835	422.819	422.70	5	0.370	28	29
422.584	422.568	422.45	4	0.280	29	30
422.327	422.310	422.19	3	0.210	30	31
422.065	422.047	421.93	2	0.150	31	32
421.797	421.778	421.66	1	0.110	32	33
421.523	421.503	421.38	1	0.080	33	34

1 1000 14, 0	1,0 0					
LG Wvl.	SHR Wvl.	Wvl. STP	Strength	Pop.	J"	J'
425.888	425.884	425.76	100	4.49	13	14
426.044	426.040	425.92	99	4.47	12	13
425.727	425.722	425.60	99	4.47	14	15
426.194	426.190	426.07	98	4.39	11	12
425.559	425.554	425.43	97	4.40	15	16
426.337	426.334	426.21	95	4.25	10	11
425.385	425.379	425.26	94	4.29	16	17
426.474	426.472	426.35	91	4.07	9	10
425.205	425.199	425.08	91	4.15	17	18
425.019	425.012	424.89	87	3.97	18	19
426.606	426.604	426.48	86	3.82	8	9
424.828	424.820	424.70	83	3.77	19	20
426.731	426.729	426.61	80	3.52	7	8
424.630	424.621	424.50	78	3.55	20	21
426.850	426.848	426.73	73	3.17	6	7
424.426	424.417	424.30	72	3.32	21	22
424.216	424.206	424.09	67	3.08	22	23
426.963	426.962	426.84	64	2.77	5	6
424.001	423.990	423.87	62	2.84	23	24
423.779	423.768	423.65	56	2.59	24	25
427.069	427.068	426.95	55	2.33	4	5
423.552	423.539	423.42	51	2.36	25	26
423.319	423.305	423.19	46	2.12	26	27
427.170	427.169	427.05	45	1.85	3	4
423.080	423.065	422.95	41	1.90	27	28
422.835	422.819	422.70	36	1.69	28	29
427.264	427.264	427.14	34	1.35	2	3

T=1000 K, $\upsilon'' = 1, \upsilon' = 0$

422.584	422.568	422.45	32	1.49	29	30
422.327	422.310	422.19	28	1.31	30	31
422.065	422.047	421.93	24	1.14	31	32
427.352	427.352	427.23	23	0.820	1	2
421.797	421.778	421.66	21	0.990	32	33
421.523	421.503	421.38	18	0.850	33	34
421.244	421.222	421.10	15	0.730	34	35
420.959	420.936	420.82	13	0.620	35	36
427.434	427.434	427.31	11	0.270	0	1
420.668	420.644	420.53	11	0.520	36	37
420.372	420.347	420.23	9	0.440	37	38
420.070	420.043	419.92	7	0.360	38	39
419.762	419.734	419.62	6	0.300	39	40
419.449	419.420	419.30	5	0.250	40	41
419.131	419.100	418.98	4	0.200	41	42
418.807	418.775	418.66	3	0.170	42	43
418.477	418.444	418.33	2	0.130	43	44
418.142	418.107	417.99	2	0.110	44	45
417.802	417.765	417.65	1	0.0900	45	46
417.456	417.418	417.30	1	0.0700	46	47
417.105	417.065	416.95	1	0.0500	47	48