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Reconciling the centimeter- and millimeter-wavelength ammonia absorption spectra under jovian conditions: Extensive millimeter-wavelength measurements and a consistent model

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ABSTRACT

Over 1000 laboratory measurements of the 2–4 mm-wavelength opacity of ammonia have been made under simulated jovian atmospheric conditions using a high-precision laboratory system developed at Georgia Tech. These laboratory measurements of the opacity of ammonia were made of various gas mixtures of hydrogen (∼77.5–85.5%), helium (∼12.5–13.5%), and ammonia (1–10%) at pressures between 1 and 3 bars and temperatures between 200 and 300 K. Laboratory measurements were also made of the opacity of pure ammonia at pressures between 0.05 and 1 bar and temperatures between 200 and 300 K. Using these millimeter-wavelength measurements and close to 2000 cm-wavelength measurements made by Hanley et al. (2009), a new consistent model has been developed to accurately characterize the absorption spectra of ammonia in a hydrogen/helium atmosphere in the 1 mm to 30 cm wavelength range. This model can be used in the 1–30 cm wavelength range at pressures up to 20 bars and temperatures from 200 to 500 K and in the 1 mm to 1 cm wavelength range at pressures up to 3 bars and temperatures from 200 to 300 K. These measurements and the accompanying model will enable better interpretation of the centimeter- and millimeter-wavelength emission spectra of the jovian planets.

1. Introduction

Millimeter-wavelength astronomy is a powerful tool for studying the temperature structure, composition, and dynamics of jovian planetary atmospheres. Furthermore, at millimeter-wavelengths, the planets Uranus and Neptune with small apparent diameters and large flux densities are frequently used as primary calibrators of astronomical sources and telescope parameters (Ulich, 1981; Kramer et al., 2008). To date, ground-based millimeter-wavelength observations have yielded disk-averaged emission measurements of the jovian planets (Ulich, 1974; Griffin et al., 1986; Muhleman and Berge, 1991; Griffin and Orton, 1993; Kramer et al., 2008), interferometric mapping of Saturn (Dowling et al., 1987; van der Tak et al., 1999; Dunn et al., 2005), and interferometric observations of limb darkening of Jupiter (Valdes et al., 1982). Accurate interpretation and modeling of the emission spectra of the jovian planets depend on the knowledge of the atmospheric abundances of various constituents and the opacity of each constituent. The presence of any broad absorption lines in the spectrum of the planet that are not properly accounted for can lead to erroneous predictions of the flux densities. Ammonia is one such constituent known to contribute to strong absorption in the jovian planets and has broad absorption features in the centimeter- and millimeter-wavelength range. Hence, accurate knowledge of the opacity of gaseous ammonia directly impacts the interpretation of the emission spectra of the jovian atmospheres at those wavelengths. Furthermore, since ammonia is one of the predominant centimeter- and millimeter-wavelength absorbers in the jovian planets, its opacity should be known before the potential effects of other absorbing constituents on the emission spectra can be assessed.

The strong absorption of ammonia in the centimeter- and millimeter-wavelength range stems from the presence of a series of strong inversion transitions around 1.25 cm, several strong rotational transitions in the submillimeter region, and a strong ν_2 roto-vibrational transition at 2.15 mm. There has been tremendous interest in understanding the absorption properties of ammonia in the centimeter-wavelength region since they were first measured in the laboratory by Cleeton and Williams (1934). Recently, Hanley et al. (2009) made close to 2000 high-accuracy measurements of the centimeter-wavelength properties of ammonia under simulated jovian atmospheric conditions (pressures up to 12 bars and temperatures up to 450 K) and developed a new model to estimate

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the opacity of ammonia in the centimeter-wavelength range. The millimeter-wavelength absorption properties of pure ammonia were first investigated more than 50 years ago by Nethercot et al. (1952) who measured the absorption of one atmosphere of pure ammonia up to 260 GHz. A few 3.2 mm-wavelength measurements of ammonia gas properties under simulated jovian conditions were made by Joiner and Steffes (1991) and Mohammed and Steffes (2004). However, these millimeter-wavelength measurements had large uncertainties because of the coarse instrumentation used at that time and did not account properly for the adsorption of ammonia on the surface of the test instruments.

Several ammonia absorption models are currently used to estimate the opacity of gaseous ammonia under jovian conditions in the millimeter-wavelength region (Joiner and Steffes, 1991; Mohammed and Steffes, 2003, 2004). These models were derived based on a limited number of millimeter-wavelength measurements and do not accurately represent the absorptivity of ammonia because of limited wavelength ranges measured and the large uncertainties associated with those measurements. There were also difficulties in reconciling the centimeter- and millimeter-wavelength opacity of ammonia (see, e.g., Mohammed and Steffes, 2004). Hence, there has been a strong impetus to make a large number of highly accurate measurements of the millimeter-wavelength properties of ammonia under simulated jovian conditions and to develop a model to estimate the opacity of ammonia in a hydrogen/helium atmosphere over a wide range of pressures, temperatures, and mixing ratios, that is consistent in the centimeter- and millimeter-wavelength range.

Over 700 measurements of the opacity of ammonia in a hydrogen/helium atmosphere at pressures up to 3 bars and temperatures from 200 to 300 K and nearly 300 measurements of the opacity of pure ammonia at pressures up to 1 bar and temperatures from 200 to 300 K have now been made in the 2–4 mm-wavelength range. These millimeter-wavelength measurements and close to 2000 cm-wavelength measurements of the opacity of ammonia made by Hanley et al. (2009) have been used to develop a consistent ammonia opacity model for the reliable interpretation of the ground-based and spacecraft-based observations of the jovian planets in the 1 mm to 30 cm wavelength range. This model can be used in the 1–30 cm wavelength range at pressures up to 20 bars and temperatures from 200 to 500 K and in the 1 mm to 1 cm wavelength range at pressures up to 3 bars and temperatures from 200 to 300 K.

2. Measurement theory and system

The reduction in the quality factor (Q) of a resonant mode of a resonator in the presence of a lossy gas is used to measure the absorption of the gas (see, e.g., Bleaney and Penrose, 1947; Bleaney and Loubser, 1950). The Q is computed as the resonant frequency (f_0) divided by its half-power bandwidth (*HPBW*). When a lossy gas is introduced into the resonator, the resonances broaden due to the opacity of the gas and the center frequency shifts due to the refractivity of the gas mixture. The Q of the resonances of a Fabry–Perot resonator are monitored in the presence of the lossy gas mixture. Subsequently, a non-absorbing or low-loss gas such as argon or carbon dioxide is added to shift the center frequency of the resonances by the same amount as the lossy gas, and dielectrically matched measurements are made. The insertion loss (S) of the resonator is measured in dB at the center frequencies of each resonance and the transmissivities are obtained by $t = 10^{-S/10}$. The formula used for calculating absorptivity is given as (DeBoer and Steffes, 1994)

$$\alpha = 8.686\frac{\pi}{\lambda}\left(\frac{1-\sqrt{t_{loaded}}}{Q^m_{loaded}} - \frac{1-\sqrt{t_{matched}}}{Q^m_{matched}}\right)\ \text{dB/km}, \tag{1}$$

where Q^m_{loaded} and $Q^m_{matched}$ are the measured Qs of the resonances in the loaded and dielectrically matched conditions, respectively, and t_{loaded} and $t_{matched}$ are the transmissivities of the resonances in the loaded and matched conditions, respectively.

The high-sensitivity millimeter-wavelength system used for measuring the opacity of gaseous ammonia under simulated jovian conditions has been described by Devaraj and Steffes (2011). This system operates in the 2–4 mm-wavelength range and consists of a planetary atmospheric simulator, a millimeter-wavelength subsystem (W-band/F-band), and a data handling subsystem. The planetary atmospheric simulator controls and monitors the environment experienced by the measurement system, including the pressure and temperature conditions of the gas under test. The simulator consists of a glass pressure vessel capable of withstanding up to 3 bars of pressure, a temperature chamber capable of operating in the 200–300 K range, gas-handling subsystems, a vacuum pump, and pressure and temperature gauges. The millimeter-wavelength subsystem consists of a W-band system used for measurements in the 3–4 mm-wavelength range and an F-band system used for measurements in the 2–3 mm-wavelength range which is shown in Fig. 1. At the heart of the measurement system is a spherical mirror Fabry–Perot resonator (FPR) that operates in a near confocal configuration and is enclosed in the pressure vessel. The FPR has low losses which correspond to high Qs of between 45,000 and 190,000 at ambient room temperature. The effective path length (EPL) of the electromagnetic energy is given as (Valkenburg and Derr, 1966)

$$\text{EPL} = \frac{Q\lambda}{2\pi}. \tag{2}$$

The EPL for the FPR is between 20 and 120 m, depending on the wavelength and the quality factor of the particular resonance. A swept signal generator (HP 83650B) is used to generate signals which are fed to a times-six active multiplier chain/frequency tripler (Millitech AMC-10-RFH00/Pacific Millimeter Products D3WO) and coupled to the input port of the FPR. The signals from the output port of the FPR are fed to a spectrum analyzer (HP 8564E) via a W-band/F-Band harmonic mixer (QuinStar 922WHP-387/Pacific Millimeter Products DM) and a diplexer (MD1A/MD2A). The data acquisition system consists of a computer connected to the spectrum analyzer (HP 8564E), the swept signal generator (HP 83650B), and a CW signal generator (HP 83712B) via a general purpose interface bus (GPIB). The instruments are controlled via Matlab® and the Standard Commands for Programmable Instruments (SCPI). The software used is similar to that used by Hanley and Steffes (2007) with modifications to account for the suite of instruments used in this measurement system. An elaborate description of the measurement theory and system is given by Devaraj and Steffes (2011).

3. Measurement procedure and uncertainty

The measurement procedure followed in this investigation for characterizing the opacity of polar molecules such as ammonia broadened by non-polar molecules such as hydrogen and helium has been described by Devaraj and Steffes (2011). The measurement process begins by characterizing and selecting standard resonances (TEM_{q00}) with high Q, low asymmetry, and high signal to noise ratio at a particular measurement temperature. Any change in the measurement temperature requires a wait time of several days because of the long thermal time-constant and the necessity to identify the standard resonances at each measurement temperature. Hence, multiple measurements were performed at each temperature using different concentrations of ammonia gas mixture and different pressures. The standard resonances were used to measure the gas opacity for all the measurements performed