



The spectrum of Jupiter's Great Red Spot: The case for ammonium hydrosulfide (NH₄SH)



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ABSTRACT

Here we present new ultraviolet–visible spectra of irradiated ammonium hydrosulfide (NH₄SH), a reported jovian atmospheric cloud component, for a range of temperatures and radiation doses and make assignments to the spectral features. We show that the combination of radiolysis and thermal annealing of NH₄SH causes the originally featureless ultraviolet–visible reflectance spectrum to evolve into one that absorbs in the ultraviolet–visible region. Furthermore, we find that our laboratory spectra resemble HST spectra below 500 nm, suggesting that the more stable reaction products of NH₄SH radiolysis are likely an important component of the Great Red Spot.

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1. Introduction

Jupiter's Great Red Spot (GRS) is arguably one of the more recognizable features in the Solar System. The GRS was observed in the nineteenth century and perhaps earlier (Falorni, 1987), and since its discovery many candidate materials have been proposed to explain its color (West et al., 1986). However, no consensus has emerged concerning a unique chromophore, chromophores, or composition. One reason for the difficulty in understanding the GRS's color is that this feature's spectrum lacks distinct absorption bands, possessing only a strong slope below 600 nm and into the ultraviolet region, which results in the aforementioned red appearance (Simon et al., 2015a). We note that the colors associated with the GRS are not as pronounced as is indicated in Voyager and Pioneer images and that the GRS is actually more orange than red (Simon et al., 2015a).

The GRS is believed to originate in jovian tropospheric clouds made of NH₃, NH₄SH, and H₂O (Weidenschilling and Lewis, 1973; Wong et al., 2015). Although a simple explanation for the GRS's color might involve one of these three cloud components in solid form, none of the laboratory reflectance spectra of the corresponding ices is an adequate match at ultraviolet and visible wavelengths (Lebofsky and Fegley, 1976). However, because these same cloud components are exposed to cosmic rays (Whitten et al., 2008) and possibly solar UV photons, they will undergo chemical reactions.

Such chemical alterations could contribute to an explanation of jovian colors, but the pursuit of this possibility has been hindered by a lack of laboratory data. Only the photochemical alteration of the ultraviolet–visible (UV–vis) reflectance of NH₄SH has been reported in the refereed literature (Lebofsky and Fegley, 1976), and only at a single temperature far below that of the GRS, and the resulting spectrum does not match that of the GRS.

Given that there is only one published photochemical study of NH₄SH available (Lebofsky and Fegley, 1976) and no radiation–chemical studies, and that there are many variables that could alter the reflectance spectrum of this solid, we investigated whether ion-irradiated ammonium hydrosulfide (NH₄SH) could be an important contributor to the GRS's spectrum. As a radiation source we used ~1 MeV protons (p⁺), which serve as an analog to low-energy cosmic rays and high-energy cosmic rays that have been slowed down in Jupiter's atmosphere. Our proton-irradiation results also give insight into what changes other energy sources, such as vacuum-UV photons, electrical discharges, and high-energy electrons, might cause as the reaction products and spectral changes induced by various types of energetic processing are often similar (Baratta et al. 2002; Hudson & Moore 2001), although this has not been checked for NH₄SH until now. Our previous work on NH₄SH relied on infrared (IR) spectroscopy to identify radiation products, which were found to be predominantly polysulfur ions and radicals (Loeffler et al., 2015). We now report the first correlations between our infrared studies and the ultraviolet–visible region.

We point out that equilibrium thermodynamic models (Wong et al., 2015) predict that the jovian NH₄SH cloud component is

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stable at higher temperatures than those used in our experiments, the difference being due to a greater pressure in Jupiter's atmosphere than in our sample chamber. Thus, in this study we investigated whether heating our irradiated samples to the predicted NH_4SH cloud temperatures (~ 200 K; Wong et al. 2015) caused their UV–vis reflectance spectra and color to change significantly and to match GRS observations. Our new laboratory results are presented here, compared with HST spectra of the GRS, and discussed in light of 40 years of research into sulfur chemistry since the early work of Lebofsky and Fegley (1976).

2. Methods

2.1. Laboratory details

The experimental approach and preparation of NH_4SH are described in Loeffler et al. (2015), so only a few points require additional comments. Samples approximately $17\ \mu\text{m}$ thick were irradiated with $0.924\ \text{MeV p}^+$ to a fluence of $5 \times 10^{13}\ \text{ions cm}^{-2}$ (or an average absorbed dose of $2.83\ \text{MGy}$), corresponding to 150 years on Jupiter based on the cosmic-ray energy flux of $9 \times 10^{-3}\ \text{ergs cm}^{-2}\ \text{s}^{-1}$ (Sagan and Thompson, 1984). The film thickness was chosen to be smaller than the range of the impinging ions (Ziegler, 2010). After irradiation, samples were warmed at $0.4\ \text{K min}^{-1}$ to the desired temperature. Before and after various irradiation and warming steps, spectra of NH_4SH were recorded with an Avantes ULS2048XL fiber-optic spectrometer from 250 to 1000 nm at a resolution of 1.5 nm. To make our spectra as comparable as possible to those derived from HST observations, we measured the diffuse reflectance of our ices on a nearly Lambertian aluminum substrate, created by sandblasting the substrate's surface. The incident light was aimed normal (perpendicular) to the sample's surface and the reflected light was collected at an angle of 9° from normal. Again, this small phase angle was chosen to make our results more directly comparable to those from ground-based and HST observations, which are limited in jovian phase angle coverage. The laboratory reflectance spectra shown here were obtained by dividing the intensity (I) of the light collected from the sample by a reference spectrum of the blank substrate, after subtracting a dark-current spectrum from each. This procedure gave the ice's reflectance as $R = (I_{\text{sample}} - I_{\text{dark}}) / (I_{\text{reference}} - I_{\text{dark}})$. In addition to measuring spectral reflectance, we also photographed our samples with a Canon Eos Rebel T3i camera.

To assess the stability of our equipment, we performed a variety of blank experiments that tested to what degree factors such as source fluctuation, detector drift, and background contamination caused changes to the UV–vis reflectance of our samples. Over the timescale of our experiments, we found that all such reflectance variations due only to our equipment were under 1% over the wavelengths used (250–1000 nm). In addition to the errors determined from these blank experiments, which are errors analogous to the error bars given for the HST data (see below), the various steps in the synthesis, irradiation, and annealing of our NH_4SH ices raised the possibility of sample-to-sample variations. Thus, to assess the reproducibility of our laboratory samples and their spectra, multiple experiments were repeated under identical conditions. Comparisons of the results showed that reflectance values in the initial NH_4SH spectrum in Fig. 1 were reproducible to within 5%, while those of the spectra of irradiated and of irradiated and heated NH_4SH were reproduced to better than 10%.

2.2. HST details

The HST data used in this study were taken from WFPC2 and WFC3 over 20 years (1995–2015). The acquired HST images were

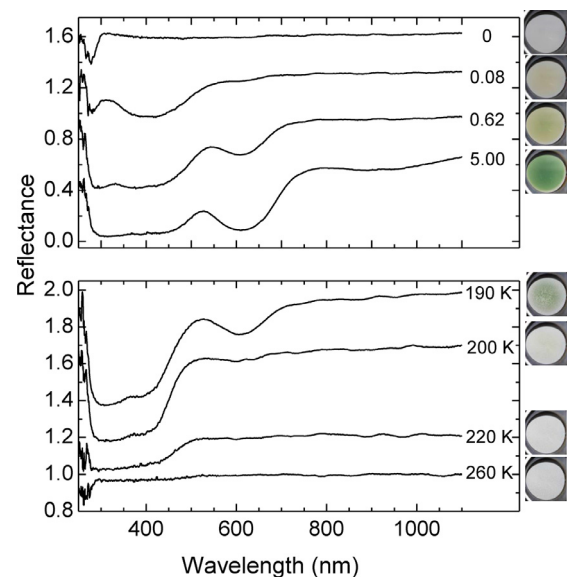


Fig. 1. Ultraviolet–visible reflectance spectra and photographs of initially-crystalline NH_4SH before and after irradiation at 120 K with 0.9 MeV protons, followed by post-irradiation heating. The proton fluence (top panel; units of $10^{13}\ \text{p}^+\ \text{cm}^{-2}$) and sample temperature during heating (bottom) are given to the right of the spectra. For clarity, spectra after doses labeled 0, 0.08, and 0.62 have been vertically offset by 0.9, 0.6, and 0.25, while spectra taken at temperatures of 190 K, 200 K, and 220 K have been vertically offset by 1.2, 0.8, and 0.25.

converted into absolute calibrated I/F units (reflectance), as described previously (Simon-Miller & Gierasch 2010), so that the spectral characteristics of the GRS could be studied. The data shown here were acquired in pixels covering the central core of the GRS, which is typically the region with the most intense color (Simon et al. 2015a).

The photometric stability of WFPC2 and WFC3 over time has been monitored using standard stars (Gonzaga et al. 2006, Kalirai et al. 2009, Kalirai et al. 2010). For WFPC2, the UV filters ($<336\ \text{nm}$) showed the greatest long-term change with the reflectance of Jupiter varying less than 2–8%, while the longer wavelength filters were stable to within $\sim 2\%$ (see Simon-Miller & Gierasch 2010). The WFC3 photometric performance has been even more stable than WFPC2 2015 with variation of less than 1% (Kalirai et al. 2009, 2010). Possible variations due to the solar flux across the HST pass bands has previously been calculated using data from Colina et al. (1996) and found to be constant within 0.1% over time (Krivova et al. 2009).

3. Results

Fig. 1 shows photographs and the corresponding UV–vis spectra of an initially-crystalline NH_4SH sample before and after proton irradiations at 120 K and during post-irradiation warming. The only absorption in the spectrum of the unirradiated sample is a drop in reflectance below 300 nm, likely from SH^- (Ellis and Golding, 1959; Guenther et al., 2001). On irradiation of the NH_4SH ice, a broad feature appeared near 400 nm, followed by one near 610 nm, and then a much weaker feature near 900 nm. The photographs in the figure show that the irradiated sample had a greenish appearance, a color that was rapidly lost on warming. By about 200 K, the strong green color and the peak near 610 nm were gone, but a spectral slope remained at 300–500 nm that was absent from the initial spectrum. Heating to higher temperatures caused this slope to decrease, yet a weak slope was still observed at 260 K. By room temperature, no slope or absorption remained in the spectrum, indicating that any residual material present did not absorb

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