A near-IR absorption band at 2.2 μm linked to ammonia-containing ice has been detected on icy bodies throughout the solar system and appears in the extensive volume of data for Pluto and Charon returned by New Horizons. This band is an important clue for understanding the abundance of ammonia and ammoniated compounds on the surface of outer solar system bodies and requires new laboratory data for its full analysis. To satisfy this data need, the complex refractive index of amorphous ammonia ice was calculated from experimental infrared transmission spectra with ice deposition and measurements conducted at 40 K, a characteristic surface temperature for outer solar system bodies. The measured imaginary part of the complex refractive index and associated band strength calculations are generally larger than prior published values for amorphous ammonia ice at 30 K. The complex refractive index for amorphous ammonia at 40 K computed in the mid-infrared region (2.5–22.73 μm) will also be valuable for interpreting observations of both solar system and astrophysical sources anticipated with the Near InfraRed Spectrograph and Mid-Infrared Instrument on the James Webb Space Telescope.

**Supporting material:** data behind figures

**1. Introduction**

A near-IR absorption feature generally occurring at 2.2 μm has been detected in the reflectance spectra of icy bodies throughout the outer solar system: Pluto (Cruikshank et al. 2019; Dalle Ore et al. 2019) and Charon (Brown & Calvin 2000); Nix and Hydra (Cook et al. 2018); Orcus (Barucci et al. 2008); the Uranian moons Miranda, Ariel, Umbriel, Titania, and Oberon (Bauer et al. 2002; Cartwright et al. 2018, 2020); and possibly Enceladus (Emery et al. 2005; Verbiscer et al. 2006) and Quaoar (Jewitt & Luu 2004; Barucci et al. 2015). One possibility is that this feature represents the 2.2 μm band of ammonia, with an associated ammonia feature at ~2.0 μm being obscured by typically strong water ice absorption features. This observed 2.2 μm feature is therefore a tantalizing clue suggesting that ammonia in some form may be present on the surfaces of these bodies. Analyzing the 2.2 μm feature in detail is challenging since ammonia hydrates and several ammoniated minerals and salts also have a prominent absorption band at or near 2.2 μm, thus complicating any certainty in the detection of ammonia ice itself (e.g., Berg et al. 2016; Cook et al. 2018; Fastelli et al. 2020). Beyond a general identification of ammonia, ammonia hydrates, and other ammoniated compounds as the origin of this feature, a detailed analysis of the 2.2 μm feature in the observational data using radiative transfer modeling has been severely hampered by a general lack of relevant experimental data (Cruikshank et al. 2019). For example, Dalle Ore et al. (2019) only incorporated nonammoniated surface components into radiative transfer models of Pluto reflectance spectra obtained from New Horizons.

The complex-valued refractive indices (or just “refractive indices” hereafter) of solid media are key input parameters for the reflectance modeling of outer solar system bodies and are needed to improve the radiative transfer modeling of the 2.2 μm feature for ammonia-containing ices. Integrated band strengths can also be determined using the refractive index and are important for deducing abundances of surface ices from absorption features in the observational data (Richey & Gerakines 2012). Refractive indices of ammonia and ammonia-containing compounds at surface temperatures relevant to outer solar system bodies are therefore an important step toward better constraining the presence of these materials in the outer solar system.

The 2.2 μm feature is the only diagnostic spectral feature for outer solar system ammonia currently available for analysis because of limitations of wavelength coverage and interference by other strongly absorbing species such as H2O and CH4, and it will be one target for the James Web Space Telescope (JWST) Near InfraRed Spectrograph (NIRSpec; e.g., Keszthelyi et al. 2016). There is also a need for mid-IR refractive indices of ammonia and ammonia-containing compounds. In some astrophysical sources and in solar system sources such as Iapetus or Dione, a 2.97 μm band tentatively assigned to ammonia might become more easily discernible with the high spectral resolution and signal precision attainable with JWST, even in cases where the contribution of H2O is strong (e.g., Clark et al. 2008, 2012; Rocha et al. 2020). In addition, observations to be made by JWST’s Mid-Infrared Instrument (MIRI) will provide mid-IR spectra of astrophysical environments with molecular ices containing ammonia (e.g., Milam et al. 2016). The ammonia “umbrella” mode at 9.3 μm (1070 cm−1) in particular is considered to be an important diagnostic spectral feature of ammonia since it is unobscured by water ice bands (Kerkhof et al. 1999) and can be diagnostic of the crystalline versus amorphous structure of ammonia ice.
Detection of the 9.3 μm band in outer solar system surface ices would be an important clue for investigating the structure of surface ammonia, and observations including this band would require mid-infrared refractive indices of all relevant species for their full analysis.

Experimental refractive indices measured for ices deposited at 40 K, a characteristic surface temperature in the outer solar system, and the approximate surface temperatures of the two KBOs most well studied by the New Horizons mission, Pluto and Charon (Earle et al. 2017; Holler et al. 2017), are also needed to ensure that radiative transfer models are accurately interpreting the surface compositions of these bodies. However, previous efforts to measure near-infrared or mid-infrared refractive indices of ammonia ices have focused on ice temperatures at 80 K or higher (see Howett et al. 2007 and references therein) or 15–30 K temperatures more appropriate for ice mantles on dust grains in the interstellar medium (e.g., Roux et al. 1979; Wood & Roux 1982; Martonchik et al. 1984; Mukai & Kratschmer 1986; Zanchet et al. 2013; Rocha & Pilling 2014).

To address these data needs, this work describes new refractive index and integrated band strength measurements for pure ammonia ice that was vacuum deposited at 40 K. Section 2 describes the experimental apparatus used to record transmission spectra in the near-IR and mid-IR spectra ranges. Section 3 describes a methodology for analysis of transmission spectra prior to a refractive index determination, the application of this methodology to the experimental measurements from Section 2, and an algorithm for computation of refractive indices from laboratory transmission spectra. Section 4 presents the calculated near-IR and mid-IR refractive indices and integrated band strengths. Finally, Section 5 summarizes and briefly discusses these results.

2. Experimental Measurements

The experimental measurements were carried out using the Matrix Isolation/Optical Constants of Ices (MIOCI) facility at NASA Ames Research Center with a schematic diagram shown in Figure 1. It consists of a diffusion-pumped vacuum chamber and a Sumitomo RDK-408D cryostat with a sample holder containing a cesium iodide (CsI) substrate window mounted at its tip. The system’s base pressure is 6.0 × 10⁻⁸ Torr, with a decrease to 2.5 × 10⁻⁸ Torr with the cryostat running. The temperature of the sample holder is controlled by an attached cartridge heater and measured by an uncalibrated Lakeshore model DT-670B-CU silicon diode. The accuracy of the diode temperature reading during experiments is ±0.5 K, with temperature stability maintained with a Lakeshore model 331 temperature controller.

When measuring transmission spectra, the cryostat is positioned for normal-incidence transmission of the infrared beam from a Digilab model FTS-4000 Fourier transform infrared (FTIR) spectrometer. The infrared beam from the spectrometer passes through Axiot system conduits connecting the spectrometer to the vacuum system and enters and leaves the vacuum system through KBr windows mounted onto two small side ports of the vacuum chamber. The spectrometer and conduits enclosing the infrared beam path are purged with compressed air provided by a dryer/carbon dioxide scrubber.

Experiments used Matheson anhydrous ammonia (99.99% purity) stored in a glass bulb (internal volume 2.2 ± 0.1 L) at a pressure of ~50 mbar and isolated from the vacuum chamber with a metering valve. The ammonia bulbs were prepared using a separate, diffusion-pumped vacuum system dedicated to gas handling with a base pressure of ~10⁻⁵ Torr or better.

In a typical experiment, the substrate is first cooled to its ultimate temperature of 5 K before the cartridge heater is used to maintain a steady substrate temperature of 40 K. A background transmission spectrum of the ice-free substrate at this working temperature is measured. The substrate is then rotated 90° about a vertical axis to its deposition position, and a flow of ammonia gas (with the metering valve slightly adjusted as necessary to maintain a constant vacuum system pressure during deposition) is introduced into the vacuum chamber. The nozzle used for ammonia deposition is 0.125 inches diameter, stainless steel tubing with the tubing end positioned approximately 3 inches from the CsI substrate and aimed at the substrate at a ~45° angle from below. After ice deposition is completed, a transmission spectrum of the ice-coated substrate is measured, and the ratio of ice-coated substrate spectrum to associated background spectrum determines the transmittance measurement.

Transmission spectra for refractive index determinations in the wavenumber range 7000–4000 cm⁻¹ were measured using the tungsten-halogen light source, a KBr beamsplitter, and a mercury cadmium telluride (MCT) detector with a resolution setting of 1.0 cm⁻¹. The standard ceramic mid-IR light source and a resolution setting of 0.5 cm⁻¹ were used for measuring transmission spectra for refractive index determinations in the mid-infrared wavenumber range 4000–440 cm⁻¹. A relatively thin ice deposit was deposited for the mid-IR measurement to avoid saturating the strong mid-IR bands, and a much thicker ice deposit was used for the near-IR measurement to better investigate weak near-IR bands (Mastrapa et al. 2008, 2009).

Experimental data in the mid-infrared region had several corrections applied. To remove gas-phase water vapor bands or absorption from water frost onto the MCT detector, the experimental spectrum was divided by an estimated transmittance calculated from a rescaled “water vapor only” or “frost
only” optical depth spectrum. The small regions around 2350 cm$^{-1}$ or 668 cm$^{-1}$ containing small CO$_2$ bands were replaced with piecewise-linear representations of the local spectral baseline.

Finally, each of the measured spectra exhibited interference fringes due to the ice layer forming on the substrate surface that is directly exposed to the flow of gas into the vacuum system (denoted the “front” ice layer) and a back ice layer forming from molecular effusion of background gas in the vacuum chamber. Infrared light from the FTIR light sources is initially incident upon this front ice layer when transmittance measurements are performed.

From the ice thickness estimates described below, the average deposition rates of ammonia ice onto the substrate are estimated to be $(1-3) \times 10^{-2}$ μm minute$^{-1}$ for the front ice layer and $(2-5) \times 10^{-3}$ μm minute$^{-1}$ for the back ice layer. With the assumption that the vacuum chamber base pressure of $2.5 \times 10^{-6}$ Torr is entirely water vapor, water deposition would add $3 \times 10^{-4}$ μm minute$^{-1}$, or roughly 1–2 orders of magnitude smaller change in deposited ice thickness per unit time than that produced by ammonia deposition from the gas flows into the system.

3. Analysis of Measured Transmission Spectra

3.1. Transmittance Calculations

The first step for determining refractive indices from the transmission spectra is to determine an appropriate transmission model for generating simulated transmission spectra, which are necessary for determining the imaginary part of the refractive index at each step of the iterative refractive index algorithm described below. A close match between the simulated spectral baseline and the spectral baseline in an experimental measurement is necessary for generating realistic refractive index values.

Prior refractive index algorithms typically required the spectral baseline to be subtracted from the spectrum before determination of the refractive index can begin (Gerakines & Hudson 2020) or were limited to using optically thick spectra (Mastrapa et al. 2008). In our method, we used a parameterized transmission model that does not require the spectral baseline to be subtracted from the spectra before analysis. This model can be used for both optically thin spectra and optically thick transmission spectra (by setting the parameter $f_c$ described below to 0 for modeling optically thick spectra). One limitation of this method is that the transmission model used must be sufficiently complex to accurately represent the spectral baseline in the spectra. More sophisticated transmission models than the one described below may be required for analyzing transmission spectra of thin films that are more complex than single-component, vacuum-deposited ice layers.

The refractive indices of the two ice layers deposited on the substrate might depend on the deposition method. Loeffler et al. (2016) measured the refractive indices of CO$_2$ ices prepared using several different deposition methods, including direct deposition and background deposition, and found a 4% variation in refractive indices measured at 670 nm. Dohnálek et al. (2003) measured the refractive index of water ice at 623.8 nm as a function of deposition angle and found a decrease of 7% with background deposition compared to direct deposition. In this work’s experiments, the back ice layer is approximately 15%–20% of the thickness of the front ice layer, so the computed refractive index should be dominated by the front ice layer values. For simplicity, the adopted transmission model assumes that the front and back ice layers have the same composition.

To match the amplitude of ammonia ice interference fringes observed in the near-IR spectrum, the Fernández-Torre et al. (2005) method of using a linear combination of the equations for fully coherent and fully incoherent propagation was adopted for light propagation through the front ice layer. A decrease in the height of observed interference fringes could be due to thickness inhomogeneities or refractive index inhomogeneities in the deposited ice layers (Swanepoel 1984), so conclusions about the ice composition based on the value of this parameter were not possible. It was not necessary to introduce an equivalent parameter for the back ice layer.

The transmission model adopted here assumes planar, parallel interfaces for the substrate and deposited ices and uses standard normal-incidence equations for the computations (see Heavens 1995; Katsidis & Siapkas 2002; Fernández-Torre et al. 2005). The total simulated transmittance $T_i[n(\vec{r}) + ik(\vec{r})]$ is

\[ T_i[n(\vec{r}) + ik(\vec{r})] = f_c \frac{T_{coh}[n(\vec{r}) + ik(\vec{r})]}{T_{substrate}} + (1 - f_c) \frac{T_{inc}[n(\vec{r}) + ik(\vec{r})]}{T_{substrate}}, \]

where $f_c$ is a coherence parameter for the front ice layer, $T_{coh}[n(\vec{r}) + ik(\vec{r})]$ is the transmittance of the ice-coated substrate calculated assuming coherent light propagation through front and back ice layers, $T_{inc}[n(\vec{r}) + ik(\vec{r})]$ is the transmittance of the ice-coated substrate assuming incoherent light propagation through the front ice layer and coherent light propagation through the back ice layer, and $T_{substrate}$ is the calculated transmittance of the bare substrate. Expressions for $T_{coh}$ and $T_{inc}$ are provided in the Appendix. Their dependence on the ice thicknesses and the refractive index is contained within complex phases $i\delta_f$ and $i\delta_b$:

\[ i\delta_f = 2\pi i n(n(\vec{r}) + ik(\vec{r}))d_f - \Delta_f \]  \hspace{1cm} (2a)
\[ i\delta_b = 2\pi i n(n(\vec{r}) + ik(\vec{r}))d_b - \Delta_b. \]  \hspace{1cm} (2b)

Parameters $\Delta_f$ and $\Delta_b$ are analogous to scattering losses in the ice layers and were introduced to produce a better match between the transmission model and a measured transmission spectrum as described below.

We assume fully incoherent light propagation through the bare, nonabsorbing substrate:

\[ T_{substrate} = \frac{T_{es} T_{se}}{1 - R_{es} R_{se}}, \]

where $R_{es} = R_{se} = |r_{es}|^2$, $T_{es} = T_{se} = 1 - |r_{es}|^2$, and $r_{es}$ is the Fresnel reflection coefficient for light incident upon the substrate from the embedding medium (vacuum).

The refractive index $n_e$ of the cesium iodide substrate is necessary in order to calculate Fresnel coefficients at the substrate surfaces. For the near-IR measurement, we used refractive index data cataloged by Li (1976) to calculate an average value of $n_e = 1.77$ for the refractive index of cesium iodide at 40 K in the 7000–4000 cm$^{-1}$ range. For the mid-IR wavenumber measurement, the calculated average value of $n_e = 1.76$ for the 4000–440 cm$^{-1}$ range produced calculated ammonia ice interference fringes that were too small in amplitude to match observed interference fringes in the spectra.
The Planetary Science Journal, 2:240 (11pp), 2021 December

Roser et al.

Table 1
Estimated Thicknesses of Deposited Ices in Microns and Final Parameters \( f_c \), \( \Delta f \), and \( \Delta \delta \) Assigned to the Mid-IR and Near-IR Transmission Spectra

| Spectrum   | Front Thickness  | Back Thickness | \( f_c \) | \( \Delta f \) | \( \Delta \delta \) |
|------------|------------------|----------------|--------|-----------|-----------|
| Mid-IR     | 0.68±5%          | 0.14±17%       | 1.0    | 0.002 8   | 0.000 58  |
|            | 0.031 1 at 3 999.873 cm\(^{-1}\) | 0.004 9 at 3 999.873 cm\(^{-1}\) |          |
|            | 0.032 0 at 4 653.258 cm\(^{-1}\) | 0.005 0 at 4 653.258 cm\(^{-1}\) |          |
|            | 0.046 2 at 5 200.076 cm\(^{-1}\) | 0.007 2 at 5 200.076 cm\(^{-1}\) |          |
|            | 0.051 0 at 5 599.822 cm\(^{-1}\) | 0.008 0 at 5 599.822 cm\(^{-1}\) |          |
| Near-IR    | 10.16±3%         | 1.6±4%         | 0.2 at 4 000.355 cm\(^{-1}\) | 0.047 5 at 5 738.214 cm\(^{-1}\) | 0.007 78 at 6 350.130 cm\(^{-1}\) |
|            | 0.02 at 6 350.130 cm\(^{-1}\) | 0.01 at 6 999.657 cm\(^{-1}\) | 0.007 78 at 6 350.130 cm\(^{-1}\) |
|            | 0.053 6 at 6 999.657 cm\(^{-1}\) | 0.008 44 at 6 999.657 cm\(^{-1}\) |          |

Note. \( f_c \), \( \Delta f \), and \( \Delta \delta \) are defined for the near-IR spectrum as piecewise-linear functions with linear segments defined by the given end points.

3.2. Selection of Parameters in the Transmission Model

Distinct values for the front and back ice thicknesses \( d_f \) and \( d_b \) and parameters \( f_c \), \( \Delta f \), and \( \Delta \delta \), were determined for both the near-IR and mid-IR experimental spectra. Thicknesses and parameter values were kept fixed during the iterative Kramers–Kronig algorithm described below, so that \( T_d \) for each spectrum is purely a function of \( n(\varphi) + ik(\varphi) \) in those calculations. An initial assumption of \( n(\varphi) + ik(\varphi) = n_0 \), where \( n_0 \) is the refractive index in the visible range, throughout the wavelength range of a spectrum was adopted during this analysis. Satorre et al. (2013) measured \( n_0 = 1.45±2.5\% \) for amorphous ammonia ice deposited at 40 K. The criterion for selecting parameter values was fidelity to the spectra in regions where \( k \approx 0 \). The final parameters assigned to the mid-IR spectrum and near-IR spectrum are listed in Table 1.

The thicknesses \( d_f \) and \( d_b \) of the deposited layers were determined first by selecting values that produced a match between the wavenumber positions of extrema of the interference fringes observed in the experimental spectra and the wavenumber positions of extrema of the interference fringes observed in the simulated transmission spectra. The base error in this thickness determination is the error in \( n_0 \), which is 2.5%. For a given \( n_0 \), an estimate of the additional error in determining ice layer thicknesses by matching extrema positions is 0.02 \( \mu m \), which corresponds to a total percentage error ranging from 17% for an ice layer with 0.14 \( \mu m \) thickness to a total percentage error of 3% for an ice layer with 10.16 \( \mu m \) thickness.

Once assignments of the ice layer thicknesses in the model were made, \( f_c \) was determined by matching the amplitude of interference fringes in the simulated transmission spectra to the amplitude of interference fringes observed in the experimental spectra. Note that optically thick spectra can be modeled by choosing \( f_c = 0 \), but some other method of determining the thickness of the ice layers would be needed when there are no observable interference fringes. Finally, the parameters \( \Delta f \) and \( \Delta \delta \) were selected to produce agreement between the spectral baseline and the experimental spectrum in regions where \( k \approx 0 \). A further constraint that

\[
\frac{\Delta f}{\Delta \delta} = \frac{d_f}{d_b}
\]

was adopted to determine unique values of \( \Delta f \) and \( \Delta \delta \).

3.3. Analysis of Pure Ammonia Spectra

Once the transmission model and the procedure for selecting the model parameters had been determined, a detailed analysis of the experimental spectra was performed. The pure ammonia spectrum measured in the mid-IR is shown in Figure 2. The left panel of Figure 2 shows a simulated transmission spectrum in which only a single surface of the cesium iodide substrate is coated with ammonia ice with thickness 0.68 \( \mu m \), \( f_c = 1.0 \), and \( \Delta f = \Delta \delta = 0 \). This ice thickness replicates the maximum in the interference fringes at \( \sim 2800 \) cm\(^{-1}\), but there is disagreement between model and measurement in the range above \( \sim 3300 \) cm\(^{-1}\).

The middle panel of Figure 2 shows a simulated transmission spectrum that takes into account a back ice layer with thickness 0.14 \( \mu m \), \( f_c = 1.0 \), and \( \Delta f = \Delta \delta = 0 \). This corrects the slope of the transmission curve but causes this model to slightly overestimate the transmittance at all wavenumbers. Finally, the right panel of Figure 2 shows a model that uses the back ice layer with thickness 0.14 \( \mu m \), \( f_c = 1.0 \), and \( \Delta f = 0.0028 \) and \( \Delta \delta = 0.00058 \) to improve the agreement between measured and simulated spectra.

The pure ammonia spectrum measured in the near-IR region 7000–4000 cm\(^{-1}\) is shown in Figure 3. The interference fringes in this spectrum are more numerous than in the mid-IR measurement, indicating that a much thicker ice layer has been deposited. The top left panel of Figure 3 shows a model transmission spectrum of a single ice layer with a thickness of 10.16 \( \mu m \), \( f_c = 1.0 \), and \( \Delta f = \Delta \delta = 0 \). The major variation in the transmittance level is attributed to interference fringes from a back ice layer deposited onto the cesium iodide substrate. The simulated spectrum shown in the bottom left panel of Figure 3 has a back ice layer with thickness 1.6 \( \mu m \) added with \( f_c = 1.0 \) and \( \Delta f = \Delta \delta = 0 \) to better match the magnitude of this variation in the transmittance.

For further agreement between the transmission model and the measured near-infrared spectrum, it was necessary to introduce wavenumber dependence to the parameters \( f_c \), \( \Delta f \), and \( \Delta \delta \) as shown in Figure 4 and specified in Table 1. We used piecewise-linear functions for convenience in specifying these functions and incorporating them into the optimization code described below. As shown in the top right panel of Figure 3, the effect of using the chosen function \( f_c(\varphi) \) with \( \Delta f(\varphi) = \Delta \delta(\varphi) = 0 \) in the transmission model is to reduce the amplitude of the interference fringes to match the observed amplitude. In our choice of \( f_c \), we have implicitly assumed that there is no decrease in \( f_c \) in the vicinity of the absorption bands at \( \sim 4500 \) and \( \sim 5000 \) cm\(^{-1}\). We discuss the potential error in determinations of \( k(\varphi) \) due to
3.4. Iterative Algorithm for Computing the Refractive Index

The analysis of each transmission spectrum described above determined parameter values for a simulated transmission spectrum $T_i(n(\tilde{v}) + ik(\tilde{v}))$ as a function of the current trial values of $n(\tilde{v})$ and $k(\tilde{v})$. An iterative algorithm generated the complex refractive index $n(\tilde{v}) + ik(\tilde{v})$ from an experimental transmission spectrum $T_i(\tilde{v})$ and its associated simulated spectrum function $T_i[n(\tilde{v}) + ik(\tilde{v})]$. This algorithm starts with $n(\tilde{v}) + ik(\tilde{v}) = n_0$ when entering into the first iteration and uses the function $F(\tilde{v}) = (T_i(\tilde{v}) - T_i[n(\tilde{v}) + ik(\tilde{v})])^2$ to guide optimization steps in this process. Each iteration progresses through the following steps:

1. Computation of $k(\tilde{v})$ values by Nelder–Mead optimization of $F(\tilde{v})$ at each value of $\tilde{v}$ in the experimental spectrum. For each fixed value of $\tilde{v}$, $n(\tilde{v})$ is kept fixed and only the value of $k(\tilde{v})$ is allowed to be changed to optimize $F(\tilde{v})$.

2. Calculation of refined $n(\tilde{v})$ values from the newly determined $k(\tilde{v})$ values using the Kramers–Kronig relation (Bergren et al. 1978) and Maclaurin’s formula for calculating the integral (Ohta & Ishida 1988):

$$n(\tilde{v}) = n_0 + \frac{2}{\pi} P \int_0^{\infty} \frac{\tilde{v}' k(\tilde{v}')}{{\tilde{v}'}^2 - \tilde{v}^2} d\tilde{v}',$$

where $n_0$ is the refractive index in the visible range and $P$ represents the Cauchy principal value of the given integral. The Cauchy principal value in Equation (5) overcomes the problem of the singularity when $\tilde{v}' = \tilde{v}$ (Rocha & Pilling 2014; R. Bell, private communication).

The ice thicknesses, the $f_i$ parameter, and the $\Delta_f$ and $\Delta_b$ parameters are kept fixed throughout this iterative process. Overall convergence of the refractive index was tracked using the post-iteration value of $\sum_i F(\tilde{v}_i)$, with a decrease by less than 1% relative to the previous iteration indicating convergence and termination of iterations. Convergence during analysis of the experimental data reported here was generally reached in 10–30 iterations using this convergence criterion.

3.5. Strategy for Using the Kramers–Kronig Relation

Since the Kramers–Kronig relation entails evaluating an integral over all wavenumber values, approximations to this integral representing wavenumber ranges inaccessible to these experiments were required. Calculations using the Kramers–Kronig relation used $n_0 = 1.45 \pm 2.5\%$, the refractive index of pure amorphous ammonia at 632.8 nm measured by Satorre et al. (2013).

For the near-IR range, the Kramers–Kronig calculations used $n_0$ to represent the approximately constant value of the integral over wavenumber values above 7000 cm$^{-1}$ (Bartatta & Palumbo 1998; Bergren et al. 1978). A separate determination of the refractive index in the mid-IR wavenumber range 4000–440 cm$^{-1}$ (2.5–22.73 $\mu$m) described below was used to compute the mid-IR contribution to the near-IR refractive index. The effect of the mid-IR correction on the near-IR refractive index calculations is to produce a gradual change in the baseline value of $n$ by $\sim 0.01$ across the near-IR range. Errors in $k$ measured in the mid-IR are unlikely to meaningfully affect this baseline; it would require doubling all $k$ values in the mid-IR to increase the magnitude of this baseline change in the
The integral over wavenumber values below 440 cm$^{-1}$ is expected to have a negligible contribution to $n(n)$ in the near-IR region and was not included in the near-IR calculations (Biermann et al. 2000). For the determination of the refractive index from the mid-IR transmission measurement, $n_0$ was also used to represent the approximately constant value of the integral over wavenumber values above 4000 cm$^{-1}$, and the contribution of weak bands in the near-IR and far-IR was neglected. An absorption band at $\sim$440 cm$^{-1}$ was partially cut off by the low-wavenumber limit of the mid-IR experimental measurements. As an alternative to attempting to design an ad hoc correction based on a guess about the missing part of the band profile, the Kramers–Kronig calculations did not include the contribution from the part of this band below 440 cm$^{-1}$. If it is assumed that this band has mirror symmetry with a maximum at 440 cm$^{-1}$, a rough estimate from the Kramers–Kronig relation is that this would change $n(n)$ measured at 1080 cm$^{-1}$ by less than 0.01 and would have an even smaller contribution to $n(n)$ at higher wavenumber values. The calculated refractive index at wavenumber values in close proximity to the 440 cm$^{-1}$ feature will see the largest errors from neglecting the far-IR portion of this band.

An error estimate for $n(n)$ can be determined from a Lambert–Beer expression for the absorbance $\alpha$ that neglects reflectance effects (Fernández-Torre et al. 2005):

$$\alpha = \frac{4\pi d_f}{\log_e 10} k(n)[d_f + d_b].$$

Assuming that $\alpha$ is fixed, a lower bound for the error in $k(n)$ is therefore the error estimate of the total thickness, which is 7% for the mid-IR measurement and 3% for the near-IR measurement. From the Kramers–Kronig relation, a lower bound for the error in $n(n)$ is the error estimate of the total thickness plus the additional 2.5% error in $n_0$. In the near-IR measurement, incorrect specification of the front ice layer’s interference fringes in regions with $k > 0$ could also be a source of error. A shift in transmittance equal to the maximum-to-minimum difference in the transmittance spectrum is assumed to be equal to the corresponding change in the refractive index. This assumption is valid because the Kramers–Kronig relations neglect the contribution of weak bands in the near-IR and far-IR regions. An alternative to this approach is to use the Kramers–Kronig relations to calculate the refractive index at wavenumber values in close proximity to the 440 cm$^{-1}$ feature. The calculated refractive index at these wavenumber values will see the largest errors from neglecting the far-IR portion of this band.

**Figure 3.** Comparisons of the near-IR measurement for pure ammonia deposited at 40 K (black) to transmittance models (red) with a front ice layer only (top left), with both ice layers (bottom left), with both ice layers and partially incoherent light propagation (top right), and with both ice layers, partially incoherent light propagation, and scattering effects (bottom right). The near-IR transmission spectrum is available as Data behind the Figure (See Figure 2).

**Figure 4.** Wavenumber-dependent parameters $f_f(n), \Delta_f(n)$, and $\Delta_b(n)$ for the near-IR ammonia transmission measurement.
minimum amplitude of the front ice layer’s interference fringes is produced by an estimated change in $k$ of 0.00075 $\text{cm}^{-1}$, or $1.5 \times 10^{-4}$ from 6350–5000 $\text{cm}^{-1}$ and proportionally lower above 6350 $\text{cm}^{-1}$ (see Table 1). This corresponds to an additional 3% error at points where $k$ peaks at $\sim 0.005$.

4. Computational Results

4.1. Refractive Index

The refractive index calculated for amorphous ammonia ice deposited at 40 K is shown in Figures 5 and 6. Band assignments in Figure 6 follow Zanchet et al. (2013) for bands above 440 $\text{cm}^{-1}$. The broad feature at 440 $\text{cm}^{-1}$ is an intermixture of two discrete bands in crystalline ammonia that correspond to collective motions of the H atoms (Giuliano et al. 2014). The structure at $\sim 800$ $\text{cm}^{-1}$ in Figure 6 is due to residual water frost in the MCT detector that was not fully removed by the data processing. A small feature at $\sim 5260$ $\text{cm}^{-1}$ in Figure 6 might be the feature that Zheng & Kaiser (2007) observed at 5235 $\text{cm}^{-1}$ in amorphous ammonia.

No previously published measurements of the infrared refractive index of amorphous ammonia deposited at 40 K were found. The most similar data available are refractive index measurements for amorphous ammonia ice deposited at 30 K (Zanchet et al. 2013), which are also shown in Figures 5 and 6. The major difference between the two measurements in Figures 5 and 6 is that $k(n)$ is 8%–50% higher for the peaks at 1077, 3378, 4470, and 4992 $\text{cm}^{-1}$ at 40 K than at 30 K. $n(n)$ has correspondingly larger swings in value from its baseline value at 40 K than at 30 K. An increase in $k$ values with deposition at 40 K compared to deposition at 30 K is consistent with a finding that $k$ values for amorphous ammonia increased with increasing deposition temperature (Zanchet et al. 2013).

Figures 7 and 8 compare values of $k(n)$ computed by this work for the $\nu_2$ band and overlapping $\nu_3$ and $\nu_1$ bands of amorphous ammonia to values computed by Zanchet et al.
and to values from other publications that reported results of experiments conducted at temperatures below 50 K, the threshold temperature for depositing amorphous ammonia ice (Holt et al. 2004). Thompson et al. (1974) was omitted since these calculated $n$ values deviate greatly from all other measurements, which calls these results into question. There is a noticeable resemblance between this work’s $k$ values at 40 K and $k$ values of the 20 K (Roux et al. 1979) and 14 K (Rocha & Pilling 2014) measurements. The few data points of the 23 K measurement (Mukai & Kratschmer 1986) are ambiguous. This work’s spectra also share a general resemblance with another published graph of the ammonia refractive index measured at 10 K (Trotta & Schmitt 1996) and refractive index values using a combination of 15 K (mid-IR) and 35 K (far-IR) measurements (Trotta 1996; Trotta & Schmitt 1996; Schmitt et al. 2018).

In almost all cases, the $k$ values at the peaks of the $\nu_2$ band and $\nu_3$ band of these similar spectra are substantially higher than values in the 15 and 30 K data sets in Zanchet et al. (2013). There are two well-known experimental effects that could introduce differences in refractive index determinations:

1. As discussed above, it has been recognized that differences in the ice deposition conditions such as the geometry of a nozzle source relative to the sample substrate and direct deposition onto the substrate versus background deposition can introduce variations in the measured refractive indices. Variations in transmission spectra could also depend on the deposition rate (e.g., Jenniskens et al. 1995).

2. Differences between observed results might also be due to the presence of minute variations in water contamination of the deposited ices. Zheng & Kaiser (2007) suggested that this could be the origin of a supposed “metastable” phase of ammonia ice that is often observed in experiments.
Table 2

Peak Position (or Peak Position of a Complex of Overlapping Bands), Integration Ranges in cm$^{-1}$, and Integrated Band Strengths $A$ (in Units of $10^{-18}$ cm molecule$^{-1}$) for Pure Ammonia Deposited at 40 K Calculated from Integrated Optical Depths

| Band(s) | Peak (μm) | Peak (cm$^{-1}$) | Integration Range (cm$^{-1}$) | $A$ (This Work) | $A$ (Zanchet et al. 2013) |
|---------|-----------|-----------------|-----------------------------|----------------|-------------------------|
| ~440 cm$^{-1}$ feature | ... | ... | 440–930 | 6.6 | ... |
| $\nu_2$ | 9.29 | 1077 | 940–1285 | 19 ± 2 | 14 |
| $\nu_4$ | 6.15 | 1627 | 1285–1740 | 5.9 ± 0.7 | ... |
| $\nu_4$, $\nu_4$ | 2.96 | 3378 | 2915–3610 | 28 ± 3 | 18 |
| $\nu_1 + \nu_2$, $\nu_2 + \nu_3$ | 2.24 | 4470 | 4120–4595 | 1.14 ± 0.09 | 0.8 |
| $\nu_3 + \nu_4$ | 2.00 | 4992 | 4750–5095 | 1.03 ± 0.08 | 0.75 |
| ~5260 cm$^{-1}$ feature | 1.90 | 5260 | 5215–5420 | 0.021 ± 0.002 | ... |
| $\nu_2 + \nu_4 + \nu_4$ | 1.64 | 6108 | 6030–6150 | 0.0082 ± 0.0007 | 0.013 |
| 2$\nu_1$ | 1.54 | 6510 | 6400–6795 | 0.13 ± 0.01 | 0.079 |

Note. Errors cited for this work’s $A$ values are 12% for mid-IR features and 8% for near-IR features.

4.2. Integrated Band Strengths

Once the refractive index was determined, integrated band strengths for the principal ammonia bands were calculated using the formula

$$A = \frac{m}{\rho N_A} \int 4\pi \bar{k}(\bar{\nu}) \, d\bar{\nu}. \quad (7)$$

Here $m = 17.031$ g mol$^{-1}$ is the molar mass of ammonia, $N_A$ is Avogadro’s number, and $\rho = 0.80 \pm 5\%$ g cm$^{-3}$ is the density of ammonia at 40 K from Satorre et al. (2013). Error estimates for the integrated band strengths depend on errors in the computed $k$ values and error in the ammonia ice density. Integrated band strengths with errors based on error estimates for $k$ and the fractional error of 5% in the density value are listed in Table 2 for ammonia ice deposited at 40 K, along with analogous integrated band strength values from Zanchet et al. (2013) (with estimated 1% uncertainties) for ammonia ice deposited at 30 K.

Only an integrated band strength of the observed portion of the feature at ~440 cm$^{-1}$ that is only partially accessible in the mid-IR measurement can be determined. Giuliano et al. (2014) calculated a full band strength of $(8.1 \pm 1.5) \times 10^{18}$ cm molecule$^{-1}$ for the complete band in amorphous ammonia deposited at 10 K and $(1.2 \pm 0.2) \times 10^{-17}$ cm molecule$^{-1}$ for crystalline ammonia after warm-up to 100 K.

For most bands in Table 2, the measured band strengths with deposition at 40 K are 40%–60% larger than corresponding values with deposition at 30 K published in Zanchet et al. (2013). Peak positions at 40 K are similar to the 30 K values, although the $\nu_2 + \nu_3$ band at ~4470 cm$^{-1}$ in this work’s measurement possesses a very slight double peak. Zanchet et al. (2013) noted that a sharp side peak on this band appeared in 30 K experiments when ice thicknesses larger than 1 μm were deposited, and this was attributed to a morphological effect, perhaps connected to an observation that film thicknesses greater than ~1 μm exhibited “some film fracture or scattering phenomenon.” This work only observed a sharp side peak on this band to a very small degree for a film thickness of ~10 μm.

Zanchet et al. (2013) also recomputed $\nu_2$ and $\nu_3$, $\nu_1$ band strengths originally determined by d’Hendecourt & Allamandola (1986) for amorphous ammonia deposited at 10 K. These band strengths were determined to be $2.2 \times 10^{-17}$ cm molecule$^{-1}$ and $2.9 \times 10^{-17}$ cm molecule$^{-1}$, respectively, when the correct amorphous ammonia density value was used. These band strength values are very similar to this work’s computed values for these bands and suggest that the result in d’Hendecourt & Allamandola (1986) was impacted by the same systematic effect as observed in the refractive index comparisons above.

5. Conclusion

The 2.2 μm band seen throughout the outer solar system is the best available diagnostic of solid ammonia and ammoniated compounds and a tantalizing clue that surface ammonia is present on these bodies. To date, a detailed analysis of this band has been severely hampered by a lack of refractive indices of ammonia and ammoniated compounds at relevant outer solar system surface temperatures. As a first step toward providing these data for ammoniated materials for analysis of the 2.2 μm band, the refractive index for amorphous ammonia ice at 40 K was measured throughout the infrared region 1.43–22.73 μm (7000–440 cm$^{-1}$) and used to compute integrated band strengths for the experimentally observed absorption bands. The measured imaginary part of the refractive index and associated band strength calculations are generally larger than values in Zanchet et al. (2013) for amorphous ammonia ice at 30 K but resemble previously published measurements conducted for amorphous ammonia ice deposited at temperatures of 10–23 K. This might be due to subtle variations in the deposited ices due to differences in the deposition conditions or contamination from background gases in the vacuum systems.

The search for ammonia and other chemical components of outer solar system bodies will also be expanded as new observational resources—very large ground-based telescopes, future spacecraft probes, and the imminently launching JWST mission—investigate the small objects that hold important clues to the origin and evolution of our planetary system. In particular, future JWST observations of the 9.3 μm ammonia band will open up another diagnostic feature for investigating ammonia ice. The optical constants for ammonia at 40 K computed in the mid-infrared region 2.5–22.73 μm (4000–400 cm$^{-1}$) in this work will therefore contribute to interpreting ammonia ice features discovered within the large volume of mid-infrared data that will be returned by JWST.

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propagation defined above. These terms are
\[
T_{f, \text{inc}}, T_{b, \text{inc}} = \frac{T_{e, \text{inc}} T_{t, \text{inc}} \exp(-2 \Im \delta_f)}{1 - R_{r, \text{inc}} R_{e, \text{inc}} \exp(-4 \Im \delta_f)} \times \frac{T_{a, \text{inc}} T_{b, \text{inc}} \exp(2 \Im \delta_b)}{1 - R_{r, \text{inc}} R_{e, \text{inc}} \exp(-4 \Im \delta_b)} \quad (A4a)
\]
\[
R_{f, \text{inc}} = R_{f, \text{inc}} + \frac{T_{f, \text{inc}} T_{t, \text{inc}} R_{a, \text{inc}} \exp(-4 \Im \delta_f)}{1 - R_{r, \text{inc}} R_{e, \text{inc}} \exp(-4 \Im \delta_f)} \quad (A4b)
\]
\[
R_{b, \text{inc}} = R_{b, \text{inc}} \quad (A4c)
\]
Here \( R_{re} = |r_{re}|^2 \) and \( T_{ei} = 1 - R_{ei} \), with similar expressions for other interfaces between media. Finally, calculations in Section 3 showing the effect of using only a front ice layer used \( R_{b, \text{inc}} = R_{f, \text{inc}} = |r_{ei}|^2 \) and \( T_{b, \text{inc}} = 1 - |r_{ei}|^2 \) for the uncoated back surface of the substrate.

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**Transmittance Equations for the Ice-coated Substrate**

Transmittance equations for light propagation through media with planar, parallel interfaces are well known in the scientific literature (see Heavens 1995; Katsidis & Siapkas 2002; Fernández-Torre et al. 2005). Here we presume that light is initially incident upon the vacuum–ice interface of the front ice layer at normal incidence, that both ice layers are the same material, and that light propagation through the millimeters-thick substrate material is fully incoherent. The complex phases \( \delta_f \) and \( \delta_b \) at normal incidence are given by Equations (2(a)) and (2(b)) above.

With coherent propagation through the ice layers deposited on the substrate, the transmittance \( T \) is given by

\[
T_{coh} = \frac{T_{f, coh} T_{b, coh}}{1 - R_{f, coh} R_{b, coh}}, \quad (A1)
\]

where \( T_{f, coh} \) and \( R_{f, coh} \) are transmission and reflection factors for coherent propagation of light from vacuum to substrate through the front ice layer and \( T_{b, coh} \) and \( R_{b, coh} \) are transmission and reflection factors for coherent propagation of light from substrate to vacuum through the back ice layer. These terms are

\[
T_{f, coh} = \frac{T_{f, inc} T_{t, inc} \exp(i2\delta_f)}{1 + r_{r, inc} r_{e, inc} \exp(2i\delta_f)} \quad (A2a)
\]

\[
R_{f, coh} = -\frac{r_{r, inc} + r_{e, inc} \exp(2i\delta_f)}{1 + r_{r, inc} r_{e, inc} \exp(2i\delta_f)} \quad (A2b)
\]

\[
R_{b, coh} = -\frac{r_{r, inc} + r_{e, inc} \exp(2i\delta_b)}{1 + r_{r, inc} r_{e, inc} \exp(2i\delta_b)} \quad (A2c)
\]

Here \( r \) and \( t \) are Fresnel coefficients, with the left subscripted symbol indicating the medium on the incident side of the interface and the right subscripted symbol indicating the medium on the opposite side of the interface. Subscript “e” denotes the embedding medium (vacuum), subscript “i” denotes the ice medium (amorphous ammonia ice), and subscript “s” denotes the substrate material (cesium iodide).

For incoherent propagation at normal incidence through the ice-coated substrate, the transmittance \( T \) is given by

\[
T_{inc} = \frac{T_{f, inc} T_{b, inc}}{1 - R_{f, inc} R_{b, inc}}, \quad (A3)
\]

where \( T_{f, inc} \), \( R_{f, inc} \), \( T_{b, inc} \), and \( R_{b, inc} \) are incoherent propagation versions of the transmission and reflection factors for coherent propagation through the ice-coated substrate and through the uncoated back surface of the substrate.
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