Infrared Optical Constants of Low Temperature H_2 Ices for Astrophysics Research

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Abstract

This is a proposal for laboratory measurements of the infrared (λ=1 - 20 μm) optical constants of hydrogen (H_2) ices at temperatures between 2 and 5 K for astrophysics research. The optical constants – complex index of refraction – of materials are widely used in astrophysics as input parameters required for simulations of light scattering, calculations of radiation transfer, and to determine the chemical composition of matter. Since the 1960s the database of optical constants for materials involved in astronomical calculations has grown enormously and now includes a wide variety of ices (H_2O, CO, CO_2, NH_3, CH_4). However, the optical constants of H_2 ices in the visible or infrared regions are not available in the literature. We propose to measure the complex index of refraction of pure H_2 ices by extracting them from high-resolution infrared transmission spectra of H_2 ices grown on an optical substrate attached to the cold finger of a liquid helium cryostat. This new capability to extract optical constants from IR spectroscopic data can then be applied to other interstellar and planetary ices to take my research program into qualitatively new directions related to NASA missions. The optical constants will be reported in refereed journal articles and made available to online databases for astronomical research.

I. Description of Proposed Research

Modern observational astrophysics continues to utilize a wider range of the electromagnetic spectrum to take advantage of the properties that these different spectral regions offer. Infrared (IR) observations allow the study of molecules in the gas and/or condensed phases which can be used as fundamental tools to investigate the physical and chemical composition of matter in space. Observations in the mid-IR (λ=2.5-25 μm) led to the identification of various interstellar ices (H_2O, CO, CO_2, CH_4) which form around dust grains present in the interstellar medium.[1] In addition, molecular ices have been identified on planetary bodies such as Pluto, Charon, Triton, and Europa.[1] A major component of the research into interstellar and planetary ices is laboratory based measurements that allow the optical properties of these materials to be well characterized under conditions that simulate the space environment.

This proposal describes experimental characterization of the IR (λ=1 - 20 μm) optical constants of H_2 ices at temperatures from 1.6 to 4.3 K using high-resolution Fourier transform infrared (FTIR) spectroscopy. The complex index of refraction is defined by,

$$\tilde{n}(\tilde{\nu}) = n(\tilde{\nu}) + ik(\tilde{\nu}) \qquad (1)$$

where $n(\tilde{\nu})$ and $k(\tilde{\nu})$ are the wavenumber (IR frequencies are commonly expressed as wavenumbers $\tilde{\nu}=1/\lambda$ in units of cm^{-1}) dependent refractive and absorption indices or optical constants, respectively.[2] These two wavenumber dependent optical constants define the full optical behavior of the material under investigation. Knowledge of these optical constants is thus crucial to understanding how the material interacts with light (scattering, absorption, and emission), which is clearly important for modeling the behavior of H_2 ices in astrophysical contexts. Careful review of the literature reveals that the optical constants of H_2 ices have not been reported despite the IR spectrum of H_2 ices being well known.[3-5]

Hydrogen is the most abundant chemical species in the known universe[6] however H_2 ices have not received much attention from the astrophysics community presumably because H_2 ices are only stable at extremely low temperatures. The melting point[7] of H_2 is only 13.8 K and thus the general consensus is there are no regions in the universe where H_2 ices can form and be stable. However, without knowledge of the optical constants of H_2 there is no way to accurately predict or test whether H_2 ices can form under certain conditions. For example, H_2 ices have been postulated to exist in low temperature clouds in the Galactic halo but the authors had to estimate the values of the optical constants.[8] The assumed values of the optical constants of H_2 ices represent "*one of the main areas of uncertainty*" in the model.[8] Therefore we propose to make these difficult measurements in order to provide the opportunity to better model the radiative properties of H_2 ices.

The proposed research is a significant departure from my current research interests. My research program at UW currently utilizes H_2 ices as materials to study ultra-low temperature chemistry. Thus, while we have built-up the expertise on how to grow H_2 ices and how to characterize them using FTIR spectroscopy, what would be new is how to extract the optical constants from the IR data.[9] This is not a trivial extension since our proposed method of measuring the optical constants relies on quantitative intensity measurements. However, the procedures we would follow have been worked out in other research groups and thus we would use this knowledge to develop this new capability in our laboratory.[10, 11] We have been in contact with Prof. Margaret A. Tolbert's group at UC Boulder and Prof. Emeritus John E. Bertie at the University of Alberta on how to best extract the IR optical constants of H_2 ices from the FTIR transmission spectra. Given our background, the H_2 ices are a natural first system to develop the experimental methodology and analysis skills to extract optical constants from IR transmission data. In addition, since we are one of only a handful of laboratories in the world that can grow optical quality H_2 ices, it makes sense to measure the optical constants for this important fundamental system.[4, 5] Due to the special properties of H_2 ices, specifically pronounced quantum mechanical effects,[4, 5] we may find significant surprises in the extracted optical constants when compared to other more common molecular ices (e.g., H_2O and CO_2). These measurements will be of interest to the condensed phase physics community but we will emphasize the astrophysical significance of these results and look to make the research more interdisciplinary.

The optical constants will be extracted from the absorption spectra of H_2 ices in the mid- and near-IR regions. The research laboratory is equipped with a liquid helium cryostat that is used to prepare H_2 ices under high vacuum conditions at temperatures between 1.6 and 4.3 K that are well-suited (highly transparent) for IR transmission studies. The IR absorption spectra can be recorded at high-resolution (0.006 cm^{-1}) using an FTIR spectrometer equipped with a variety of IR sources, beamsplitters, and IR detectors. This allows the IR spectrum to be recorded from

600 to 10,000 cm^{-1} with no gaps and since the IR absorption path is purged with dry nitrogen, the measurements will be free from atmospheric absorptions. The IR optical constants are extracted by measuring the transmission spectra of the H_2 ices of varying thickness (0.1 to 3 mm) and then using an iterative Kramers-Kronig technique both the real and imaginary indices of refraction can be determined from the data.[10, 11] By varying the thickness of the sample the real and imaginary components of the index of refraction can be better quantified.[10, 11] In addition, our experimental apparatus has the capability to enrich the H_2 ices in the para nuclear spin state of hydrogen. There are two nuclear spin states of H_2, the so-called para and ortho nuclear spin states and the para nuclear spin state is the lower energy form of hydrogen.[4, 5] This will allow the optical constants to be studied as a function of the ortho and para content of the H_2 ice. The H_2 ices can also be doped with varying concentrations of other chemical species (e.g., He) to measure the optical constants of binary mixtures of helium and hydrogen.

The general research plan is (i) to grow H_2 ices using our existing apparatus, (ii) characterize the samples using FTIR spectroscopy under a variety of experimental conditions (growth conditions, annealing histories, and temperatures), (iii) use the acquired IR spectra to deduce the corresponding real and imaginary indices of refraction, and (iv) use this data in collaboration with astrophysicists to make predictions about the stability of H_2 ices under a range of astronomical contexts.

II. Relationship to Space Grant and NASA Goals

The proposed research is most inline with the Science Mission Directorate of NASA within the Astrophysics Division. If our reported H_2 optical constants lead to the discovery of H_2 ices in space this would have large implications on the structure of the universe. Once the methodology for extracting optical constants from FTIR data is well established, we could then apply this method to other interstellar and planetary ices. The James Webb Space Telescope (JWST) scheduled to launch in 2013 will attempt to study the processes of star formation in the interstellar medium of external galaxies.[1] This new capability for extracting fundamental near-IR data for interstellar ices would allow us to compete for grants aimed at supporting the JWST mission. The proposed research would also allow one of my best graduate students, Sharon C. Kettwich, to gain new experience in interdisciplinary astrophysics research.

III. Products

One of the products of this research would be to publish a paper reporting the IR optical constants of H_2 ices. We would work in collaboration with Dr. Mark Walker, Director of Oxford & Manly Astrophysics in Australia to interpret our results in an astrophysical context and would plan to submit the paper to *The Astrophysical Journal*. Dr. Walker is one of the authors of reference 8 and he contacted us to see if we could measure the infrared optical constants of H_2 ices. We would also publish the H_2 optical constants in online databases.[12] The goal would also be to use this grant to develop these new analysis capabilities and collect preliminary data for a proposal to NASA.

IV. Timeline

We have all the necessary equipment to perform the proposed research and thus we could get started right away and in the first three months develop the methodology to extract optical constants from the IR data. This would consist of first using programs we have gotten from Prof. Emeritus John E. Bertie to analyze existing data for pure H_2 ices. Then we would design experiments specifically to measure quantitative absorption spectra of H_2 ices of varying thickness. The next 6 months would focus on studying the effect of dopant species (e.g., He and ortho-H_2) and crystal morphology on the measured optical constants. We would also adapt this new methodology to explore new chemical systems such as CH_4 ices which can be studied over a larger temperature range. During this time we would continue the analysis of the H_2 ices and try to submit a paper reporting the IR optical constants of pure H_2 ices by the end of 9 months. We would also at this time publish the results in online databases. The remainder of the grant period would be spent acquiring additional preliminary data for a proposal to NASA. These preliminary data will steer the focus of the proposal between two different scenarios: the H_2 ices are high risk but potentially large impact research studies while detailed measurements on CH_4 ices are more likely to be of interest to NASA.

V. References

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Budget Summary

3. Labor	1. Request	Proposed Cost Share Non-Federal	Federal
Faculty/Applicant Labor – Salary	$8402.00	$0.00	$0.00
Benefits	$3361.00	$0.00	$0.00
4. Other costs:			
Supplies	$1000.00	$1500.00	$0.00
5. TOTALS	$12763.00	$1500.00	$0.00

Budget Justification

Salaries: Funds are requested for one-month of summary salary for the PI ($8402).

Benefits: Fringe benefits for the PI are calculated at 40% of the salary.

Supplies: The requested funds will cover the cost of cryogens, liquid helium ($10/L) and liquid nitrogen ($0.5/L). We estimate that the total supply budget should allow for roughly 10 experiments to be conducted over the one-year grant period. The definition of an experiment is approximately 12-14 hours of acquisition time using the cryostat to measure infrared spectra of H_2 ices.

Cost share: Other expenses that are required for the proposed research that will be covered from other funding sources include the cost of chemicals, use of equipment, and cryogen costs. The major supply cost is cryogen costs, liquid helium and nitrogen, and $1500.00 of cost share will be available through Chemistry Departmental funds.