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H_2 IN INTERSTELLAR AND EXTRAGALACTIC ICES: INFRARED CHARACTERISTICS, ULTRAVIOLET PRODUCTION, AND IMPLICATIONS

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ABSTRACT

H_2 is the most abundant molecule in the universe. We demonstrate that this molecule may be an important component of interstellar and possibly intergalactic ices, both because it can be formed in situ, within the ices, and because gas phase H_2 can freeze out onto dust grains in some astrophysical environments. The condensation-sublimation and infrared spectral properties of ices containing H_2 are presented. We show that solid H_2 in H_2O-rich ices can be detected by an infrared absorption band at 4137 cm^{-1} (2.417 μm). The surface binding energy of H_2 to H_2O ice was measured to be $\Delta H_s/k = 555 \pm 35$ K. Surface binding energies can be used to calculate the residence times of H_2 on grain surfaces as a function of temperature. Some of the implications of these results are considered.

Subject headings: infrared: interstellar: lines — ISM: molecules — molecular processes

1. INTRODUCTION

Because of its great abundance, H_2 is of fundamental astrophysical importance. For example, it is *the* central species in all interstellar chemistry models, and it dominates the mass of clouds, thereby playing the key role in determining cloud dynamics. In spite of this, H_2 generally escapes direct detection. It can only be detected in slightly reddened clouds, shocked portions of clouds, or regions exposed to intense radiation (see, e.g., Shull & Beckwith 1982; Brand 1993; and references therein). It is generally assumed that H_2 forms by H atom recombination on cold grains in dense molecular clouds (cf. Hollenbach & Salpeter 1970, 1971; Flower & Pineau des Forets 1990).

In this *Letter* we show that solid H_2 can be detected by infrared spectroscopy. To evaluate the volatility of H_2 we have also determined the trapping efficiencies and surface binding energy of H_2 onto H_2O:CH_3OH interstellar ice analogues. We further demonstrate that significant quantities of H_2 can be produced by the UV photolysis of interstellar ices in a manner similar to that proposed for cosmic-ray exposure (Pironello & Averna 1988). The possibility that appreciable amounts of solid state H_2 may be present in many astronomical environments should be considered since this would have profound effects on our understanding of the physical, chemical, and dynamical evolution of dense molecular clouds, galaxies, and perhaps galactic systems. The presence of H_2 in interstellar ices was considered as early as 1949 (van de Hulst 1949). This *Letter* is a full discussion of preliminary results presented earlier (Sandford & Allamandola 1992).

2. EXPERIMENTAL TECHNIQUES AND ANALYSIS

For a detailed description of the experimental equipment and sample preparation and analysis techniques used here, see Sandford & Allamandola (1990, 1993) and references therein. Ice samples were made by condensing premixed gas samples (all with purity > 99.5%) onto a temperature-controlled CsI window at pressures of $\sim 3 \times 10^{-8}$ mbar. Typical deposition rates were $\sim 1.5 \times 10^{-5}$ moles hr^{-1} (~ 5 μm hr^{-1}), and substrate temperatures ranged from 10 to 70 K. The infrared spectra of the ices were measured from 7000 to 400 cm^{-1} at a resolution of 0.9 cm^{-1} and a frequency accuracy of ± 0.2 cm^{-1}. The ices were then processed either by warming or vacuum-UV radiation.

The number of molecules in each sample was determined by measuring the absorption band areas of the characteristic vibrational bands of each molecule and using the equation

$$N_i = \frac{\int \tau(\nu) d\nu}{A_i}, \quad (1)$$

where N_i is the column density of component i in molecules cm^{-2}, $\tau(\nu)$ is the optical depth of an absorption band as a function of frequency, and A_i is the integrated absorbance of the band in cm $molecule^{-1}$. A list of appropriate A-values for the molecules of interest here can be found in Hudgins et al. (1993), Sandford & Allamandola (1993), and references therein.

The measurement of the surface binding energy of H_2 onto various ices was made using the standard assumption that the sticking efficiency of a deposited molecule is identically equal to 1.0, and escape from the surface (reevaporation) takes place at a rate given by

$$R_s = \nu_0 \exp(-\Delta H_s/kT), \quad (2)$$

where ν_0 is the lattice vibrational frequency of the molecule within its surface matrix site, ΔH_s is the binding energy of the molecule on the ice surface, k is the Boltzmann constant, and T is the ice temperature in kelvins. Solving for the binding energy yields

$$\Delta H_s/k = T_0 \ln(\nu_0 t), \quad (3)$$

where t is the residence time of a molecule on the surface of the grain. In the slow deposition regime, if the time t is set equal to the time between the deposition of successive molecular monolayers of ice, then the binding energy can be found by determining the temperature T_0 at which the trapping efficiency drops to zero, that is, the temperature at which the guest molecules leave the ice at the same rate they arrive during deposition. (Throughout this paper we will use the standard notation of expressing surface binding energies in terms of $[\Delta H_s/k]$, that is, in kelvins.)

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3. RESULTS

Since H_2 is a homonuclear, diatomic molecule, its vibrations and rotations do not produce a change in dipole moment, making it infrared inactive and radio quiet. Thus, in the gas phase, most H_2 is not detectable by the usual astronomical techniques (see the thorough review by Shull & Beckwith 1982). However, when H_2 is frozen in a solid, some of its transitions are sufficiently perturbed to become weakly infrared active. These transitions have been studied using matrix isolation techniques in Ne, Ar, Kr, Xe, N_2, O_2, and CO (cf. De Remigis & Welsh 1970; Smith, Warren, & Guillory 1976; Warren, Smith, & Guillory 1980), and it has been found that the band strengths, positions, and widths produced by solid H_2 are *strongly* dependent on matrix composition and H_2 concentration. None of these studies considered pure H_2 or H_2 in H_2O-dominated matrices, although Warren et al. (1980) did examine Ar:H_2 samples containing small amounts of H_2O. They found that the $Q_{R1}(1)$ and $Q_1(1)$ bands near 4245 and 4130 cm^{-1}, respectively, increased linearly with H_2O concentration while other bands did not.

3.1. *The Infrared Spectral Properties of* H_2 *in Cosmic Ice Analogues*

Figure 1 shows the 4250–4050 cm^{-1} (2.35–2.47 μm) spectra of several astrophysical ice analogues. The top spectrum is of an H_2O:CH_3OH:NH_3: CO = 100:50:1:1 ice deposited at 10 K. The composition of this ice is thought to be representative of many dense molecular clouds (cf. Allamandola & Sandford 1988; Allamandola et al. 1992). The sole feature in this spectrum is due to an overtone/combination band of CH_3OH (Sandford & Allamandola 1993). The middle spectrum is of an H_2O:CH_3OH:NH_3:CO:H_2 = 100:50:1:1:10 ice deposited at 10 K. The presence of the H_2 produces new bands at 4137 cm^{-1} (2.417 μm; Fig. 1) and 3690 cm^{-1} (2.710 μm; not shown). The FWHM of these two bands are approximately 10 and 15 cm^{-1}, respectively.

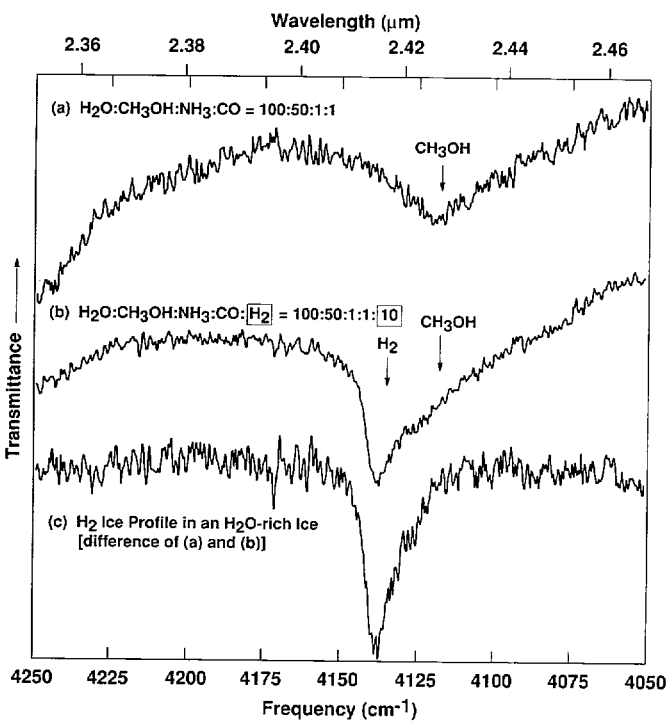

FIG. 1.—The 4250–4050 cm^{-1} (2.35–2.47 μm) spectra of several astrophysical ice analogues. (*a*) A H_2O:CH_3OH:NH_3:CO = 100:50:1:1 ice deposited at 10 K, (*b*) H_2O:CH_3OH:NH_3:CO:H_2 = 100:50:1:1:10 ice deposited at 10 K, and (*c*) the difference between (*a*) and (*b*). The bottom spectrum is the profile of the $Q_1(1)$ pure vibrational transition of H_2 in an H_2O-rich ice.

We attribute the 4137 cm^{-1} band to the $Q_1(1)$ pure vibrational transition of H_2, the same transition noted to increase with H_2O concentration by Warren et al. (1980). We do not, however, detect a counterpart of the $Q_{R1}(1)$ transition, implying that H_2 does not rotate freely in an H_2O-rich ice. *Thus, an absorption band at* 4137 cm^{-1} *is characteristic of* H_2 *in* H_2O-*rich ices.* This weak band provides an excellent diagnostic of H_2-containing ices in space.

The second band induced by the presence of H_2 falls at 3690 cm^{-1}, a position not reported for H_2 in other matrices. This feature is due to the stretching vibrations of dangling OH groups of H_2O molecules that are prevented from H-bonding with neighboring H_2O molecules due to the presence of H_2 (e.g., Hagen, Tielens, & Greenberg 1983; Rowland, Fisher, & Devlin 1991). Since the 3690 cm^{-1} band can also be produced by many other species in H_2O ices, its presence in the spectrum is not diagnostic of H_2.

The integrated absorbance value (A) of the 4137 cm^{-1} band due to H_2 in H_2O-rich ices was determined by measuring the spectrum of an H_2O:CH_3OH:H_2 = 10:5:1 gas deposited at 10 K. The A-value for the H_2 feature was then derived by scaling its area by the A-value of the 1026 cm^{-1} CH_3OH band (1.8×10^{-17} cm $molecule^{-1}$; d'Hendecourt & Allamandola 1986) and its associated band area. This process was repeated for five independent experiments yielding $A_{4137} \geq (9.4 \pm 0.9) \times 10^{-20}$ cm $molecule^{-1}$. (The greater than sign is included because this procedure *assumes* that all the components of the gas mixture stuck to the sample window with 100% efficiency. If some of the H_2 did not stick then this value represents a *lower* limit.)

3.2. *The Production of* H_2 *by Photolysis*

Figure 2 shows how the 4250–4050 cm^{-1} spectrum of an interstellar ice analogue evolves with UV photolysis and warm up. The top spectrum is of an H_2O:CH_3OH:NH_3:CO = 100:50:10:10 ice deposited at 10 K. The next spectrum is of an ice deposited for 70 minutes at 10 K from the same gas with simultaneous UV photolysis. Photolysis produced a strong H_2 feature at 4143 cm^{-1} (2.414 μm; FWHM $\approx$ 15 cm^{-1}) and weakened the methanol feature near 4120 cm^{-1}. The strength of the H_2 band did not increase appreciably after the first 70 minutes although irradiation was continued. Using $A = 9 \times 10^{-20}$ cm $molecule^{-1}$ for the H_2 band, the abundance of H_2 relative to H_2O saturated at a value of $\sim$35% (since the H_2 A-value is a lower limit, this is an upper limit to the H_2 abundance).

Comparing the total UV dose over the first 70 minutes with the amount of H_2 produced, we estimate a photolytic H_2 production efficiency on the order of 10% (one H_2 produced per 10 incident photons). Thus, for an unattenuated diffuse interstellar radiation field between 6 and 13.6 eV (10^8 cm^{-2} s^{-1}; Habing 1968), an H_2O:CH_3OH-rich interstellar ice grain would become saturated in H_2 in only 300 yr. Since ices only exist in the denser medium where the interstellar radiation field is attenuated, the actual production rate will fall as e^{-2A_v} going into a cloud. Therefore, up to $A_v \approx 5$, interstellar ice grains should become saturated with H_2 on time scales less than 10^7

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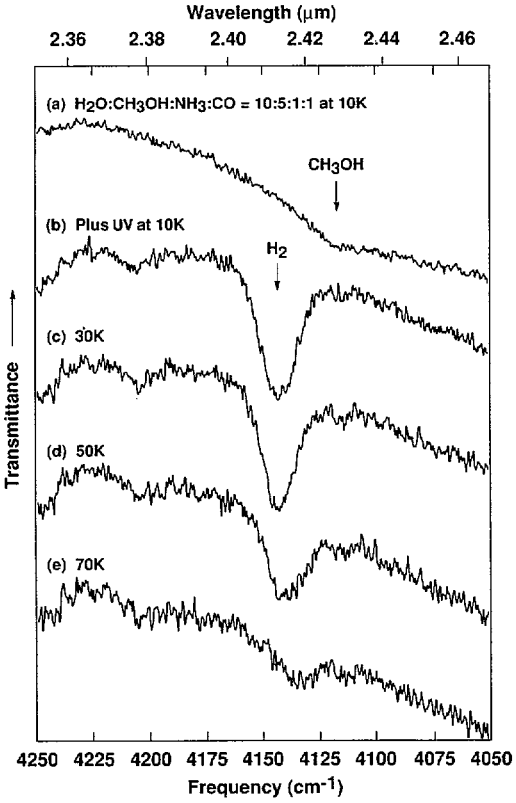

FIG. 2.—Production of H_2 by UV photolysis of an astrophysical ice analogue. (*a*) A $H_2O:CH_3OH:NH_3:CO = 100:50:10:10$ ice deposited at 10 K without photolysis. (*b*) 10 K ice deposited from the same gas mixture with simultaneous UV photolysis followed by additional irradiation. The prominent band at 4143 cm^{-1} is due to photolytically produced H_2. The other spectra are of the ice after warming to temperatures of (*c*) 30 K, (*d*) 50 K, and (*e*) 70 K.

yr. At $A_v > 5$ the ambient UV flux due to shocks driven by T Tauri winds (Norman & Silk 1980) and cosmic-ray interactions with hydrogen (Prasad & Tarafdar 1983) is expected to be on the order of 10^{-4} to 10^{-5} that of the diffuse medium, that is, comparable to the attenuated diffuse radiation field at $A_v \approx 5$. Thus, *independent of their position within dense clouds, it should be possible for interstellar ices to become saturated in* H_2 *on time scales that are less than or comparable to the lifetimes of typical dense molecular clouds.* As a result, it should be possible to detect the weak 4137 cm^{-1} solid state H_2 absorption feature in the spectra of some interstellar dense molecular clouds.

The UV production of H_2 will ultimately be limited by the H content of the ices, and the H_2 production rate will fall rapidly as the ices become depleted in H-bearing precursor molecules. Since the H content of interstellar ices is only about 10^{-3} that of the total hydrogen in dense clouds, the UV production of H_2 is not expected to be a major contributor to overall H_2 abundances. It is, however, the most likely way in which H_2 will become incoporated into normal interstellar ices (see also § 3.3).

The lower spectra in Figure 2 show how H_2 is lost from the interstellar ice analogue as it is warmed after photolysis. Note that little H_2 is lost until the temperature exceeds 30 K and that some still remains in the ice even at 70 K! *The retention of solid* H_2 *at such high temperatures occurs because it is produced within the volume of the ice rather than at its surface.* Once produced, it is trapped by the surrounding, less-volatile matrix. The position of the band is slightly temperature dependent. From 10 to 30 K it falls at 4143 cm^{-1}, but it moves to lower frequency with H_2 loss, ending up near 4135 cm^{-1} at 70 K.

3.3. *The Surface Binding Energy of* H_2 *on* $H_2O:CH_3OH$ *Ices*

Figures 1 and 2 show that gas phase H_2 can stick to H_2O-rich ices at 10 K. To quantify the depletion of gas phase H_2 onto grains, the surface binding energy of H_2 onto an $H_2O:CH_3OH = 2:1$ ice was determined using the technique discussed in § 2. Measurements were made at 10, 14, 16, 18, 20, and 22 K, and a well-determined value of $T_0 = 22$ K was found for the 0% trapping efficiency of H_2 when an $H_2O:CH_3OH:H_2 = 10:5:1$ gas mixture was deposited at $\sim 1.5 \times 10^{-5}$ moles hr^{-1} (~ 5 μm hr^{-1}).

Assuming an average surface site size for H_2 of 10 $Å^2$ (that of H_2O), we find that the time between the deposition of successive monolayers on the ice using our flow rate was $t = 0.013$ s. To the best of our knowledge the ν_0 value for H_2 in H_2O-rich ices has not appeared in the literature. One can estimate the value of ν_0, however, by mass scaling the observed lattice vibrational frequencies reported for H_2O, CO, and CO_2. This yields an expected value for ν_0 of about 7.5×10^{12} s^{-1} for H_2. Warren et al. (1980) reported an unidentified band near 256 cm^{-1} in their studies of H_2 in Ar matrices. If this band is due to the lattice mode of H_2 in the Ar matrix, it corresponds to $\nu_0 = 7.68 \times 10^{12}$ s^{-1}, a value very near that derived by mass scaling. We therefore adopt $\nu_0 = 7.5 \times 10^{12}$ s^{-1} for the lattice vibrational frequency of H_2. Using equation (4) and the values given above for T_0, ν_0, and t, *we derive an* H_2-$H_2O:CH_3OH$ *surface binding energy of* $\Delta H_s/k = 555 \pm 35$ K. This value is excellent agreement with the theoretically calculated value of $\Delta H_s/k = 550$ K for H_2 on H_2O from Hollenbach & Salpeter (1970) and the ~ 540 K value of Yuberov & Busol (1967).

3.4. *The Surface Binding Energy of* H_2 *to Pure* H_2 *Ices*

Temperature- and pressure-dependent surface binding energies for H_2 on pure H_2 are available in the literature. For astrophysical environments where pure H_2 might conceivably condense, values between $\Delta H_s/k \approx 90$ and 100 K are indicated (cf. Brown & Ziegler 1980; Souers 1986). As discussed in the next section, the much lower H_2-H_2 binding energy has significant implications for the condensation of pure H_2 ices or large quantities of H_2 onto H_2O-rich interstellar ices.

4. ASTROPHYSICAL IMPLICATIONS

These results demonstrate that H_2 can stick to the surfaces of H_2O-rich interstellar ices for grain temperatures below 20 K, temperatures which are typical of dense molecular clouds. However, once the hydrogen coverage on an H_2O-rich grain has increased to 100% (one monolayer), the lower H_2-H_2 surface binding energy will control subsequent H_2 condensation. Thus, the trapping efficiency of H_2 onto an H_2O-rich ice will decrease as its coverage increases, an effect observed in the laboratory (Lee 1972). As a result, at the colder temperatures common in dense clouds, the grains may accumulate a monolayer of H_2 which subsequently "fends off" the condensation of additional H_2. The critical temperature at which the loss of H_2 from the surface of an H_2O-rich grain balances

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its arrival rate satisfies the equation

$$N_s f \nu_0 \exp(-\Delta H_s/kT) = \tfrac{1}{4} n_{H_2} v_{th} \eta , \quad (4)$$

where N_s is the number of surface sites on the grain per unit area, f is the fraction of the sites covered by H_2, n_{H_2} is the density of gas phase H_2, v_{th} is the thermal velocity of the gas phase H_2, and η is the H_2 sticking probability. Setting $f = 1$, taking $\eta = 1$, and solving for the critical temperature yields

$$T_{crit} \approx \frac{(\Delta H_s/k)}{\ln[(m/k)^{1/2}(4N_s \nu_0/n_{H_2})]} . \quad (5)$$

Inserting appropriate values for the constants and setting $\Delta H_s/k = 555$ K, $\nu_0 = 7.5 \times 10^{12}$ s^{-1}, $N_s = 10^{15}$ cm^{-2} (appropriate for a 10 Å^2 site size), and $n_{H_2} = 10^4$ cm^{-3} yields a value of $T_{crit} \approx 12$ K. For a value of $\Delta H_s/k = 100$ K, the critical temperature drops to about 2.1 K. Thus, grains in dense molecular clouds will be partially covered by a monolayer of molecular H_2 and the coverage should be nearly complete in the colder, denser cores within clouds. However, few, if any, clouds are expected to attain the temperatures at which additional layers of H_2 will condense. Thus, *interstellar ices should not contain large contributions from the condensation of gas phase* H_2.

However, as discussed in § 3.2, significant amounts of H_2 may still be incorporated into the ice through in situ production, and once it resides in the *bulk* of the ice, H_2 can remain in the solid state to significantly higher temperatures. Our laboratory results strongly suggest that some H_2 should be in the solid state in many astrophysical environments.

4.1. *Frozen* H_2 *in Dense Molecular Clouds*

The presence of H_2 in ice grains could have significant effects on the physical properties of the grains as well as on the chemistry associated with both the gas and solid phases. At grain temperatures below 70 K, UV- or cosmic-ray produced H_2 frozen in ices may well represent a significant component of the ices. Thus, a rigorous search for the 4137 cm^{-1} absorption band due to soid H_2 band trapped in H_2O-rich ices is very important.

It should be possible to detect the 4137 cm^{-1} band of solid H_2 in some interstellar clouds. For example, if H_2 is present at the 30% level of the H_2O ice in W33A ($N_{H_2O} \approx 10^{19}$ molecules cm^{-2}), a band with an optical depth of $\sim$0.03 would be expected in its spectrum at 4137 cm^{-1}. In order to obtain a 3 σ detection of this feature with current instrumentation would require integrations on the order of an hour at a large telescope.

4.2. *Detection of Pure Solid State* H_2 *in Intergalactic and Extragalactic Space: Some Speculations*

We have shown that H_2-containing ices may be fairly common in dense molecular clouds. It is interesting to consider the possibility that pure H_2 ices might exist at the edges of or between galaxies and be responsible for some or all of the "missing mass." This possibility has received some attention in the past (cf. Lee, Gowland, & Reddish 1971; Hegyi & Olive 1986). The general consensus of the theoretical calculations is that such pure H_2 ices should not be stable against sublimation, although it is difficult to know whether the conditions assumed for these environments are appropriate or not. Regardless of the theoretical considerations, solid H_2 in these environments may have escaped detection because, in its pure solid form, it is not amenable to straightforward measurement. However, given that the direct detection of H_2-bearing ices in extragalactic environments could potentially be of fundamental importance for our understanding of the universe, it is clearly worth pursuing. In this regard, we note that pure H_2 ices produce a complex family of infrared absorption bands that span the K-band atmospheric window between 5000 and 4150 cm^{-1}. Included are bands at 4770 cm^{-1} [$S_R(1)$], 4738 cm^{-1} [$Q_1(0) + S_0(1)$], 4732 cm^{-1} [$Q_1(1) + S_0(1)$], 4703.3 cm^{-1} [$S_1(1)$], 4547 cm^{-1} [$S_R(0)$], 4503.5 cm^{-1} [$Q_1(0) + S_0(0)$], 4497 cm^{-1} [$Q_1(1) + S_0(0)$], 4484.5 cm^{-1} [$S_1(0)$], 4223 cm^{-1} [Q_R], 4153.0 cm^{-1} [$Q_1(0)$], and 4146.5 cm^{-1} [$Q_1(1)$] (cf. Gush et al. 1960; Souers 1986). Combined, these features have an intrinsic absorption strength of $A \approx 4 \times 10^{-20}$ cm molecule^{-1}. Thus, following the arguments in § 4.1, moderately long integrations with large telescopes should be able to place meaningful upper limits on the abundance of pure H_2 ices in space. We also note that the profiles and relative strengths of the characteristic solid H_2 absorptions are sensitive to the structure of the ice and its ortho- to para-hydrogen ratio (Gush et al. 1960). Thus, if they are detected, the absorption features of solid H_2 will provide information not only on its abundance but also on its history and local environment.

A telescopic search for the infrared absorption features of H_2 in H_2O-rich ices in dense molecular clouds and H_2 ices in extragalactic space is presently under way.

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