Signatures of Quantum-Tunneling Diffusion of Hydrogen Atoms on Water Ice at 10 K

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Reported here is the first observation of the tunneling surface diffusion of a hydrogen (H) atom on water ice. Photostimulated desorption and resonance-enhanced multiphoton ionization methods were used to determine the diffusion rates at 10 K on amorphous solid water and polycrystalline ice. H-atom diffusion on polycrystalline ice was 2 orders of magnitude faster than that of deuterium atoms, indicating the occurrence of tunneling diffusion. Whether diffusion is by tunneling or thermal hopping also depends on the diffusion length of the atoms and the morphology of the surface. Our findings contribute to a better understanding of elementary physicochemical processes of hydrogen on cosmic ice dust.

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The diffusion of hydrogen (H) atoms, the most abundant element in the Universe, has attracted the interest of researchers in various fields such as material and vacuum sciences, geoscience, and astrochemistry. Given that H and deuterium (D) atoms exhibit a prominent wave nature at low temperatures, their diffusion by quantum tunneling on solids has received significant attention. In particular, the tunneling diffusion of H atoms on water ice has been often investigated theoretically because of its relevance to astronomy and astrochemistry [1–3]. In interstellar molecular clouds, where the temperatures are as low as 10 K, H_2 molecules are formed on the surface of cosmic ice dust by the barrierless recombination reaction following H-atom diffusion: $H + H \rightarrow H_2$. Therefore, the diffusion process is of key importance for our understanding of H_2 formation in space. Theoretical models often assume tunneling diffusion of H atoms in addition to classical thermal hopping [4–11], but have lacked supporting experimental evidence.

Many experimental attempts to observe tunneling diffusion of H atoms on single crystalline metal surfaces have been successful; they have mostly employed field emission microscopy and scanning tunneling microscopy [12–14]. Evidence of tunneling diffusion is typically provided by the observation of the diffusion rate being temperature independent. Therefore, diffusion rates need to be monitored over a wide range of temperatures that includes the transition from classical thermal hopping (Arrhenius-type temperature dependence) to tunneling diffusion (temperature independent). Such transition temperatures for metals have been determined to span the range of 60–140 K [12–15]. Unlike on metallic surfaces, H atoms readily desorb from water ice at low temperatures below 20 K [16]. Therefore, the temperature window in which diffusion occurs is too narrow to allow measurement of its surface-temperature dependence. Consequently, it is not easy to monitor the migration of H atoms *in situ* on water ice, nor the transition from classical thermal hopping to tunneling diffusion.

Water ice is stable in two morphological structures at around 10 K in a vacuum; i.e., as amorphous solid water (ASW) and polycrystalline ice (PCI). The diffusion of H and D atoms on ASW has been studied based on temperature-programmed desorption (TPD) experiments: the measured TPD spectra of HD or D_2 reflect the diffusive recombination of atomic H and D [17–22]. However, there have been no reports of diffusion on PCI. The PCI surface consists of small pieces of a single crystal, with many steps and grain boundaries [23]. The diffusion mechanism should depend on the migration distance, that is, tunneling diffusion may occur for a short distance within each single crystal, whereas it should be highly suppressed for long-distance diffusion beyond the steps and boundaries. In addition, TPD experiments are not appropriate to clarify the diffusion process on ASW and PCI, because the shapes of the TPD spectra pertaining to ASW and PCI are characterized by diffusion and desorption from various potential sites, unlike for single crystalline surfaces, which requires analysis of multiparameter fits [16,24]. An alternative approach to TPD is desirable for studying tunneling diffusion on amorphous and polycrystalline surfaces.

We recently developed a novel experimental design to study the surface diffusion of H and D atoms: the system combines photostimulated desorption (PSD) and resonance-enhanced multiphoton ionization (REMPI) [25,26]. We measured the attenuation rates of the H- or D-atom signals owing to diffusion-limited recombination, *after* atomic deposition on ASW at 8 K. Only a weak isotope dependence of the diffusion rate was found, and the results can be explained by classical thermal hopping [26]. However, we could not measure the attenuation of H or D atoms on the PCI surface, because the diffusion was too rapid to be monitored *after* the deposition had stopped. This fast diffusion rate suggests that shallow potential sites dominate the PCI surface, where tunneling diffusion may be noticeable. The motivation of the present study is to clarify whether tunneling diffusion is observable on ASW and

PCI. The present experiment explores the isotope effects on surface diffusion of atoms *during* atomic deposition for a wide range of fluxes F (10^{12}–10^{15} atoms $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$) to evaluate the possibility of quantum tunneling. By changing the flux, we control effectively the distance between atoms on the surface and thus the encounter rate. Here we report, for the first time, experimental evidence of quantum-tunneling diffusion on PCI. Surface diffusion on ASW was predominantly found to be by thermal hopping, although tunneling diffusion may also partly contribute to the observed recombination rate.

The basis of the present experiment is as follows. When H atoms are *continuously* deposited onto a surface, the surface number density of H atoms [H] in steady state should obey

$$p_s F = k_{\mathrm{H+H}}[\mathrm{H}]^2 + k_{\mathrm{des}}[\mathrm{H}], \tag{1}$$

where F, p_s, $k_{\mathrm{H+H}}$, and k_{des} are the atomic flux, the atomic sticking coefficient, and the rate constants of $\mathrm{H}+\mathrm{H}$ recombination and of monoatomic desorption, respectively. At sufficiently low temperatures, $k_{\mathrm{des}}[\mathrm{H}]$ can be negligible [27]. We have previously shown that monoatomic desorption from ASW can be neglected at 8 K because the rate of diffusive recombination of H atoms absorbed on ASW at 8 K is much larger than the monoatomic desorption rate during and after atomic deposition; i.e., $k_{\mathrm{H+H}}[\mathrm{H}]^2 \gg k_{\mathrm{des}}[\mathrm{H}]$ [26]. In Eq. (1), the term due to recombination by the Eley-Rideal (ER) process where an incoming atom from the gas phase directly hits an adsorbed atom on the surface is not included because of the reason discussed later. A number of molecular dynamics (MD) calculations have reported that the sticking probability of H atoms with low incident energy (100 K) is near unity at 10 K on both ASW and PCI [27–29]. Another MD calculation also showed that the difference in sticking coefficients between H and D atoms is very small when the kinetic temperature of H and D atoms impinging is around 100 K [30]. Therefore, it is reasonable to assume that $p_{s,\mathrm{D}}/p_{s,\mathrm{H}} \approx 1$. When the fluxes of H and D atoms are almost identical in separate experiments, the ratio of the surface number density of H and D atoms $[\mathrm{D}]/[\mathrm{H}]$ can be expressed simply as

$$\frac{[\mathrm{D}]}{[\mathrm{H}]} \approx \sqrt{\frac{k_{\mathrm{H+H}}}{k_{\mathrm{D+D}}}}. \tag{2}$$

Given that recombination is a radical-radical barrierless reaction, the recombination rates are limited by the diffusion of the adsorbed atoms. Therefore, we can derive the ratio of the diffusion rates from that of the surface number densities of the H and D atoms in steady state *during* the continuous deposition of atoms.

The experimental apparatus was described elsewhere [25,26] (also, see Supplemental Material [31]). Briefly, the ASW and PCI samples were produced by $\mathrm{H_2O}$ vapor deposition through a capillary plate onto a mirror-polished aluminum (Al) substrate at 15 and 145 K, respectively. The column densities of ASW and PCI were estimated to be approximately 2×10^{16} molecules cm^{-2} using the absorption coefficient reported in the literature [32]. For reference, the coverage of unity corresponds to about 1.0×10^{15} molecules cm^{-2} on the surface of hexagonal ice, considering the relevant lattice parameters [33]. The sample was recooled and maintained at 10 K during atomic deposition. The H (D) atoms were produced through dissociation of $\mathrm{H_2}$ ($\mathrm{D_2}$) gas in microwave-induced plasma in a water-cooled Pyrex tube in a differentially pumped atomic source chamber. The atoms were subsequently cooled to ~100 K by passing them through the cold Al pipe, and *continuously* deposited onto the sample surfaces. The flux of atoms was controlled to fall in the range of 4.9×10^{12}–7.9×10^{14} atoms $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ by inserting apertures with diameters of 0.5, 1, 2, and 4 mm before the Al-pipe entrance and also removing a PTFE pipe attached between the Pyrex tube and the Al pipe. To estimate the atomic fluxes, we first estimated the flux of the $\mathrm{H_2}$ molecular gas from the atomic source without microwave discharge by a cold cathode gauge and also simultaneously monitored the signals of $\mathrm{H_2}$ ($v = 0$; $J = 1$) just above the aluminum substrate at room temperature by the REMPI technique. The base pressure in the sample chamber started at 10^{-8} Pa and increased to 10^{-4} Pa during atomic deposition. Subsequently, the decrease of $\mathrm{H_2}$ in the REMPI signals during H-atom production was measured when the microwave was switched on for the same gas-flow rate, thus yielding the H-atom flux. In this measurement, recombinative desorption from the aluminum substrate was negligible. Using the REMPI method, we also confirmed that the fluxes of H and D atoms were identical by directly measuring the gaseous atoms originating from the atomic source.

The H or D atoms adsorbed on the surface were photodesorbed by weak PSD laser radiation at a wavelength of 532 nm with a beam diameter of approximately 1.5 mm (0.3–0.4 mJ pulse^{-1} at 10 Hz). The atoms desorbed into vacuum were selectively ionized by a 2 + 1 REMPI laser at 1 mm above the ice surface, and further analyzed by a time-of-flight method. The REMPI employed the two-photon $2s(^2\mathrm{S}_{1/2}) \leftarrow 1s(^2\mathrm{S}_{1/2})$ transition and also the weaker $3s(^2\mathrm{S}_{1/2})$ or $3d(^2\mathrm{D}_{3/2,5/2}) \leftarrow 1s(^2\mathrm{S}_{1/2})$ transition to avoid signal saturation for the lower and higher fluxes, respectively [34,35]. The PSD-REMPI signals were recorded at three different REMPI wavelengths on a resonance peak as a function of delay time between the PSD and REMPI lasers to detect all the atoms with various kinetic energies. We previously confirmed that the H- and D-signal intensities are proportional to the number densities of the H and D atoms present on the ice surface and that the PSD laser does not cause any undesired heating or energetic processes [25].

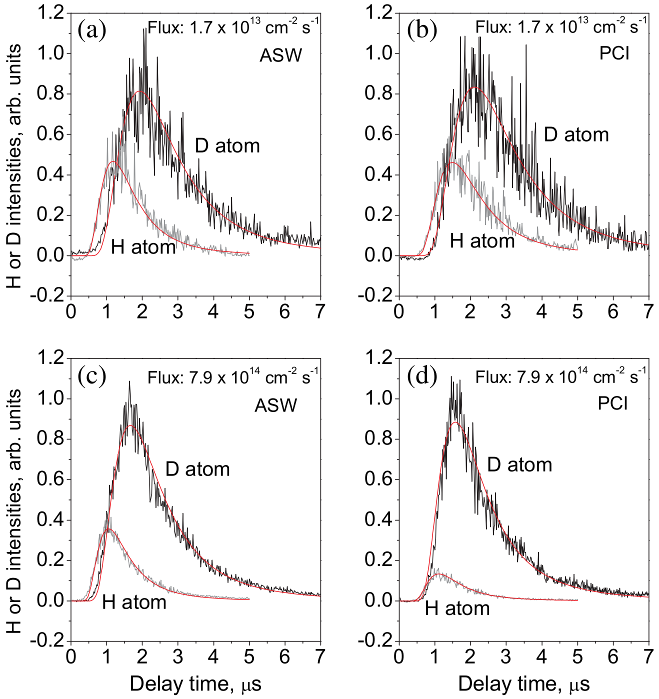

FIG. 1 (color online). Delay-time spectra of photodesorbed H (gray line) and D (black line) atoms from ices at 10 K: (a) ASW, (b) PCI for the atomic flux of 1.7×10^{13} atoms s^{-1}, (c) ASW, (d) PCI for the atomic flux of 7.9×10^{14} atoms s^{-1}. The red lines are fits to the data derived assuming Maxwell-Boltzmann distributions with translational temperatures in range 25–50 K for the spectra of the H and D atoms.

Figure 1 shows the typical delay-time spectra of PSD-REMPI signals obtained from the ASW and PCI surfaces during continuous H or D atomic deposition. Each data point is the sum of signals obtained at three different REMPI wavelengths. The spectra were reproduced by a single Maxwell-Boltzmann distribution with a translational temperature T_{trans} of 25–50 K which corresponds to 0.4–0.8 kJ mol^{-1}, assuming a translational energy of $2k_BT_{\mathrm{trans}}$ [36–39]. These low energies indicate that the PSD laser did not induce undesired heating of the surface [26]. We confirmed that the signal intensities increased linearly as the power of the 532 nm photodesorption laser increased to 0.6 mJ pulse^{-1} (see Supplemental Material [31]). The absorption coefficient of water ice is known to be negligibly small for photons at 532 nm [40], and the PSD-REMPI signal intensity decreased as the ice thickness increased, and almost disappeared for 316 L deposition (1 L = 10^{-6} torr × 1 s) [26]. Hence, photodesorption may be induced from the Al surface; propagation of phonons from the substrate is a possible source. In Fig. 1, the PSD-REMPI signal intensities of the D atoms are approximately twice stronger than those of the H atoms for a low atomic flux of 1.7×10^{13} atoms cm^{-2} s^{-1} for both ASW and PCI. Remarkably, the D/H ratio increases with increasing atomic flux, particularly on PCI. Figure 2 shows the ratios of the D/H signal intensities obtained by summing the delay-time spectra as a function of the flux of H (D) atoms. The error bars pertaining to the x axis in Fig. 2 are dominated by the statistical errors of the H_2 intensities in the REMPI signals used for estimating the atomic fluxes. The D/H ratio strongly depends on the atomic fluxes, especially on the PCI surface, and it exhibits a strong dependence on structure (amorphous versus polycrystalline). From Eq. (2), this result indicates that the relative diffusion efficiency of D and H atoms followed by recombination is strongly affected by the atomic fluxes, as well as by the ice-surface structure, and that the isotope effect becomes significant on the PCI surface at higher atomic fluxes.

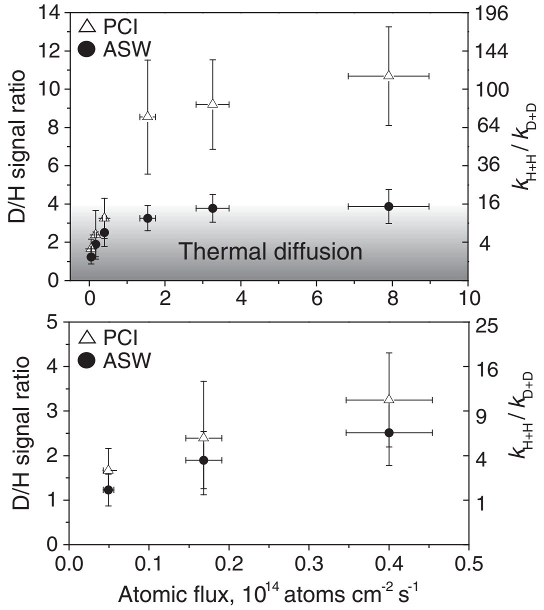

FIG. 2. Upper: D/H signal intensity ratio of the delay-time spectra as a function of the flux of H (D) atoms, obtained from the PCI (open triangles) and ASW (filled circles) surfaces. The right Y axis is the ratio of surface diffusion rates calculated from Eq. (2). The shaded area represents the thermal-diffusion dominated region derived from Eq. (4). Lower: enlarged view in the low flux region.

We first consider the [D]/[H] ratio expected when surface diffusion of H (D) atoms is limited to thermal hopping. Given that the recombination reaction is a radical-radical barrierless reaction, its rate $k_{\mathrm{H+H}}$ for H atoms is dominated by H-atom diffusion and can be expressed as

$$k_{\mathrm{H+H}} = s\nu_{\mathrm{H}} \exp(-E_{\mathrm{diff,H}}/k_B T), \tag{3}$$

where s is the unit area of the surface site, ν is the frequency factor, and E_{diff} is the diffusion activation energy of the atoms [41]. Here, ν is generally proportional to the inverse of the square root of the mass when the mass of diffusing particle is significantly smaller than that of surface molecule [42,43]. Therefore by adopting Eq. (3), Eq. (2) can be rewritten as

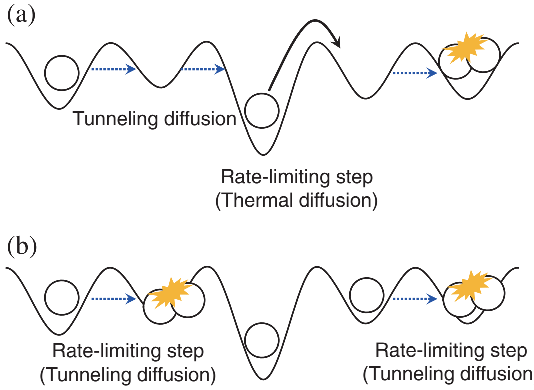

FIG. 3 (color online). Schematic illustration of the diffusion of atoms on a surface for (a) low atomic coverage and (b) high atomic coverage. For lower fluxes, the average distance between adatoms is significant [case (a)] and, therefore, each atom is required to migrate a long distance across the steps and boundaries to encounter its reaction partner. For higher fluxes, adatoms locate nearby and thus can encounter through the tunneling diffusion.

$$\frac{[\mathrm{D}]}{[\mathrm{H}]} \approx \sqrt{\frac{k_{\mathrm{H+H}}}{k_{\mathrm{D+D}}}} \approx [\sqrt{2}\exp(\Delta E_{\mathrm{diff,D\text{-}H}}/k_B T)]^{1/2}, \qquad (4)$$

where $\Delta E_{\mathrm{diff,D\text{-}H}} = E_{\mathrm{diff,D}} - E_{\mathrm{diff,H}}$. In practice, $\Delta E_{\mathrm{diff,D\text{-}H}}$ is the zero-point energy difference for thermal hopping between H and D atoms and causes a semiclassical kinetic isotope effect (KIE) for thermal hopping. We previously reported a small KIE in relation to the surface diffusion of H and D atoms on ASW at 8 K, and activation energies of 22 and 23 meV for H- and D-atom diffusion, respectively [26]. Substituting the difference of the activation energies $\Delta E_{\mathrm{diff,D\text{-}H}} = 1$ meV into Eq. (4) gives $[\mathrm{D}]/[\mathrm{H}] \approx 2.12$, and thus $k_{\mathrm{H+H}}/k_{\mathrm{D+D}} \sim 4.5$ at 10 K. As shown in Figs. 1 and 2, the D/H ratios for both PCI and ASW in low atomic flux conditions ($<5 \times 10^{13}$ cm^{-2} s^{-1}) are consistent with this value, indicating that the observed diffusion is limited by thermal hopping. In contrast, at higher atomic fluxes the D/H ratio—particularly for PCI—increases to ~10. According to Eq. (2), a $[\mathrm{D}]/[\mathrm{H}]$ of ~10 corresponds to $k_{\mathrm{H+H}}/k_{\mathrm{D+D}} \approx 100$. This large KIE cannot be explained by thermal hopping, and is clear evidence of quantum-tunneling diffusion of atoms on the PCI surface. The D/H ratios for ASW remain around 4 (i.e., $k_{\mathrm{H+H}}/k_{\mathrm{D+D}} \approx 16$) for high atomic fluxes of 1.5×10^{14}–7.9×10^{14} cm^{-2} s^{-1}. These values are much smaller than those for PCI. Although the increase of the D/H values implies the coexistence of quantum tunneling and thermal diffusion, thermal hopping may still be the dominant mechanism controlling the surface diffusion of H and D atoms on the ASW surface because of poor energy-level matching in irregular potentials [1–3]. The observed flux dependence of the PCI diffusion mechanism can be explained as follows (see also Fig. 3). For lower fluxes, the average distance between adatoms is significant and, therefore, each atom is required to move a long distance across the steps and boundaries to encounter a reaction partner. This limits diffusion by thermal hopping at irregular potential sites. On the other hand, at higher fluxes, the reaction partners would always be located nearby within the single crystals, and thus recombination can occur, as a result of tunneling diffusion. The present results are consistent with the theoretical prediction of Smoluchowski, who proposed that diffusion by quantum tunneling is suppressed on ASW because of the nonperiodic potential [1–3], while on PCI surfaces H atoms would spread instantaneously to their boundaries through quantum tunneling and become localized there [3].

We confirmed that the ER process and the “hot-atom” mechanism (i.e., the transient migration of incident atoms before fully thermalized on the surface) did not affect the recombination rate in the present experiment. When the H atoms were deposited with the different incident kinetic energies corresponding to 60 and 300 K controlled by varying the temperature of the Al pipe in the atomic gas line [44], the recombination rates were independent from the kinetic energy (see Supplemental Material [31]). Since both the ER and hot atom processes should depend on the kinetic energy, we conclude that these processes are negligible in the present experiment. MD calculations showed that the average distance relevant to hot-atom migration is approximately 10 and 25 Å for incident kinetic temperatures of 60 and 300 K, respectively [27]. This suggests that the H atoms on the PCI surface are individually distributed across an area in excess of $\pi(25\ \text{Å})^2 = 2 \times 10^{-13}$ cm^2 in the present study. In other words, the H atoms diffuse over more than $(2 \times 10^{-13}) \times (1 \times 10^{15}) = 200$ sites to recombine following tunneling diffusion. We also examined the effect of coexisting molecules on the diffusion. Although the dissociation fraction in our atomic source is as high as 70%–80%, [25,26] undissociated $\mathrm{H_2}$ ($\mathrm{D_2}$) from the source and residual $\mathrm{H_2}$ gas still remained in the chamber. We codeposited atoms on ASW and PCI with additional $\mathrm{H_2}$ or $\mathrm{D_2}$ gas from another gas line. The resulting ratios of the H and D intensities were almost independent of the gas pressure (see Supplemental Material [31]), indicating that the presence of molecules barely affects the diffusion mechanism of atoms on the ice surfaces.

In conclusion, the surface number densities of H and D atoms on ice surfaces were monitored in the steady state, where the deposition of incoming atoms balances their loss through diffusive recombination at 10 K. For the PCI surface, the $[\mathrm{D}]/[\mathrm{H}]$ number density ratios increase significantly up to ~10 with increasing flux, indicating that the surface diffusion of H atoms is 100 times faster than that of D atoms, which cannot be explained by classical thermal hopping. This is the first experimental evidence of quantum-tunneling diffusion on the PCI surface. For the ASW surface, tunneling diffusion may partly contribute to the recombination rate, but the diffusion is predominantly thermal hopping given the smaller KIE than that observed on PCI.

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