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Signatures of quantum-tunneling diffusion of hydrogen atoms on water ice at 10 K

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(1) Experimental apparatus and procedure

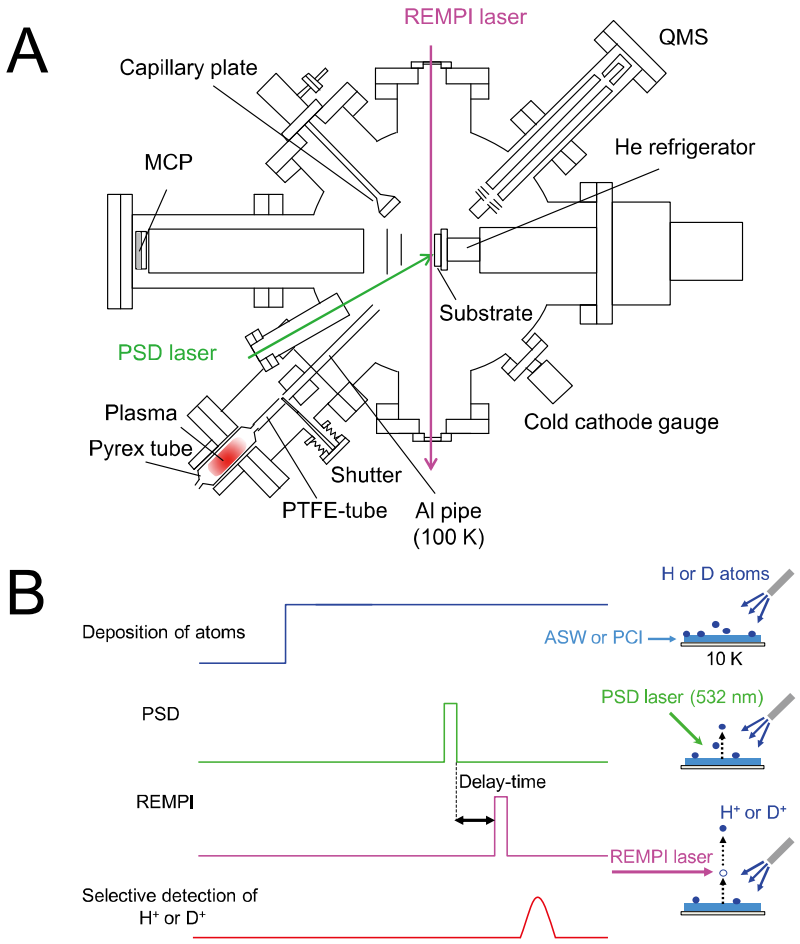

Fig. S1. (A) The experimental apparatus. MCP: microchannel plate, PTFE: polytetrafluoroethylene, QMS: quadrupole mass spectrometer, PSD: photo-stimulated desorption. REMPI: resonance-enhanced multiphoton ionization. (B) Schematic illustration of the timing chart for the present experiment. The PSD and REMPI are performed under continuous atomic deposition onto amorphous solid water (ASW) or polycrystalline ice (PCI).

(2) Energy dependence of the 532 nm PSD laser on the PSD-REMPI signal intensity of H and D atoms from ASW and PCI.

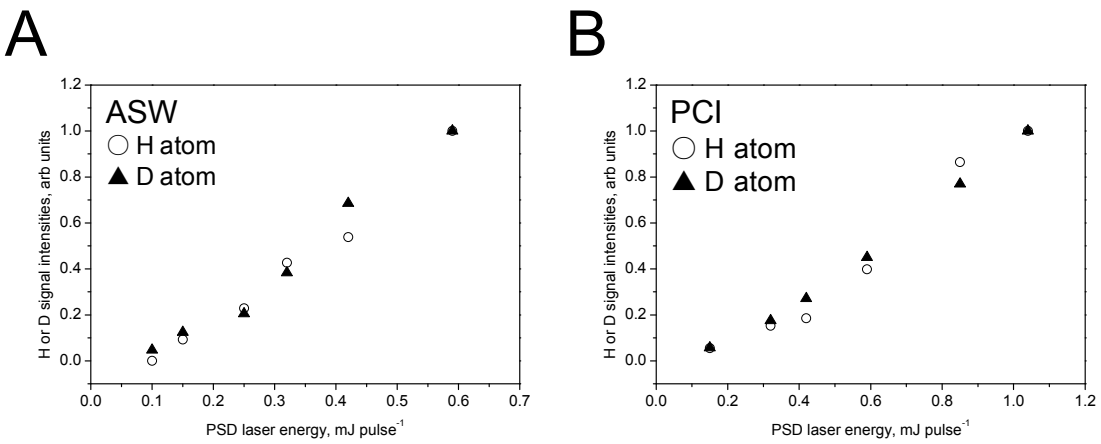

FIG. S2. PSD-REMPI signal intensity of H and D atoms from (A) ASW and (B) PCI as a function of the 532 nm PSD laser energy. The signal intensities of H and D atoms are normalized by those at 0.59 mJ pulse^{-1} for ASW and 1.04 mJ pulse^{-1} for PCI.

(3) Incident energy dependence of H atoms on the surface number density of adsorbed atoms on the PCI surface at 10 K.

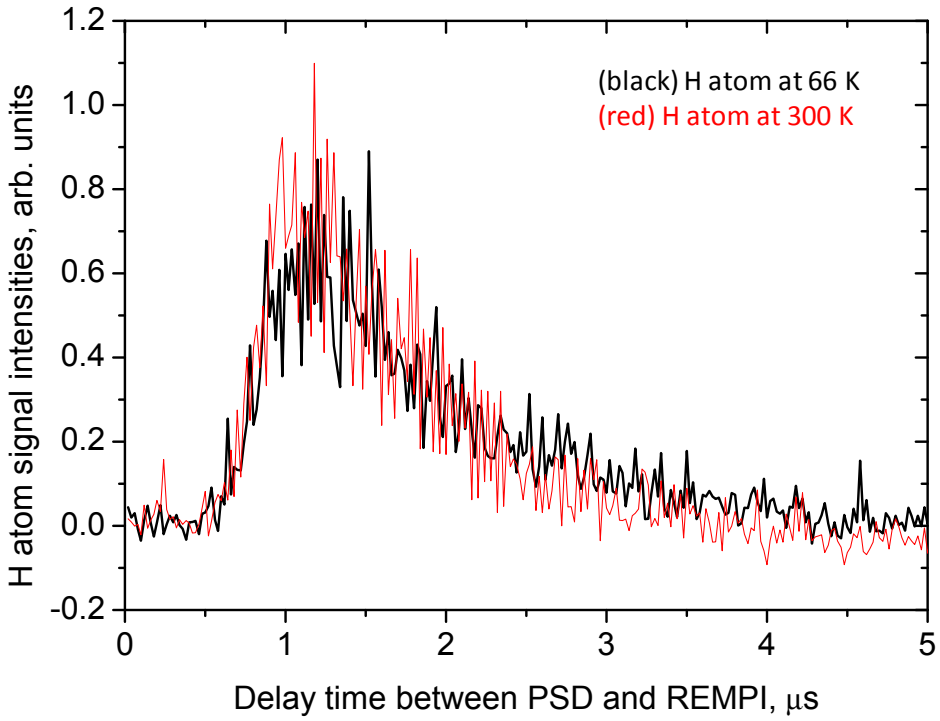

FIG.S3. Delay-time spectra of the PSD–REMPI signal of H atoms from the PCI surface at 10 K, for Al-pipe temperatures (used for atomic cooling) of 66 K (black) and 300 K (red). The H-atom flux is approximately 1.5×10^{14} cm^{-2} s^{-1}.

(4) Additional H_2 and D_2 co-deposition experiments

Figure S4 shows the PSD–REMPI signal intensities of H and D atoms from the ASW and PCI surfaces as a function of the pressure of additional H_2 or D_2. These additional molecules were introduced into the chamber by background vapor deposition up to 3×10^{-4} Pa, which corresponds to fluxes of approximately 3×10^{15} cm^{-2} s^{-1}. The PSD–REMPI signals for both H and D atoms gradually decrease as the deposition of H_2 and D_2 increases, respectively. These results show that atomic adsorption was inhibited by competitive long-term adsorption of molecules on the surface at 10 K. However, the ratio of the signals of the H and D atoms was similar for each pressure, indicating that the presence of molecules barely affects the diffusion mechanism of atoms on the ice surfaces. Figure S4 also indicates that the coverage of H_2 or D_2 molecules is less than unity during H_2 or D_2 co-deposition, because the amount of adsorbed molecules increased with increasing fluxes (deposition pressure). This demonstrates that thermal desorption of the molecules should occur at 10 K, also indirectly suggesting the occurrence of monoatomic desorption of H and D atom from the ASW and PCI surfaces at 10 K. However, we have previously shown that the diffusive recombination rate, $k_{H+H}[H]^2$, is much faster than the monoatomic desorption rate, $k_{des}[H]$ at around 10 K. [1] Thus, monoatomic desorption did not strongly contribute to the observed [H] or [D].

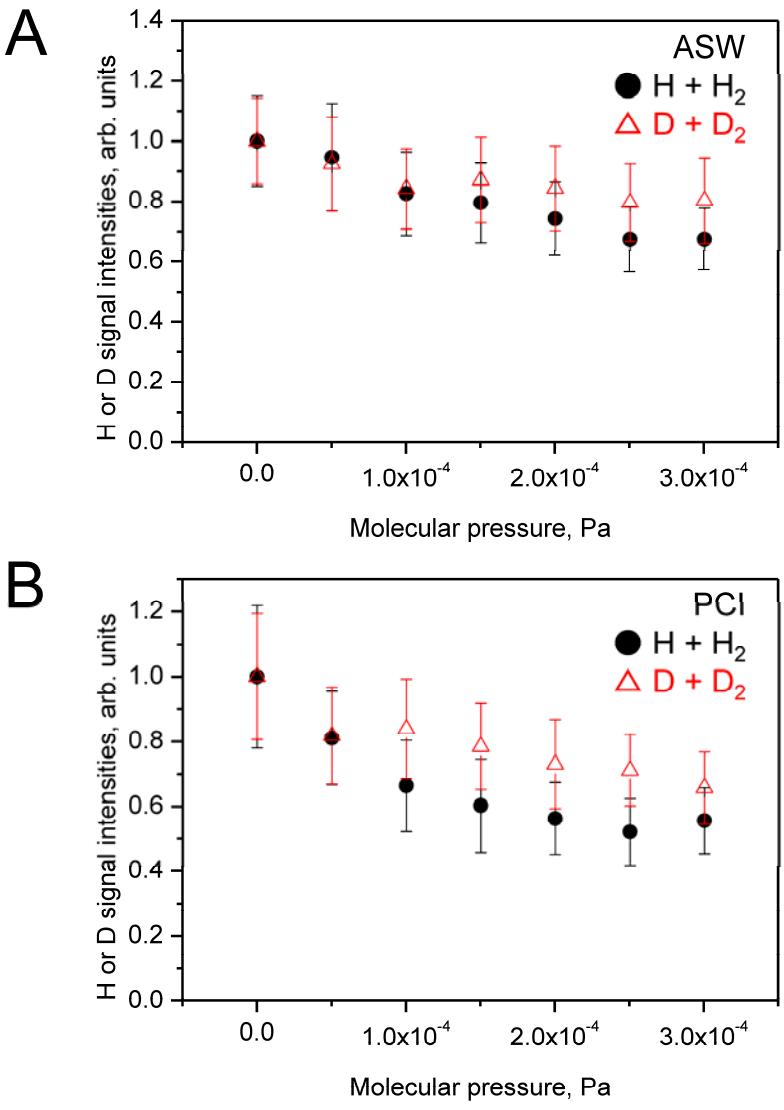

FIG. S4. PSD–REMPI signal intensity of H and D atoms from (A) ASW and (B) PCI surfaces as a function of the pressure of additional H_2 or D_2 co-deposition by background vapor deposition.

References

[1] T. Hama, K. Kuwahata, N. Watanabe, A. Kouchi, Y. Kimura, T. Chigai, and V. Pirronello, Astrophys. J. **757**, 185 (2012).