Supporting Information

Intact water adsorption on Co(0001) at 100 K: transition from ordered bilayer to amorphous ice structures

Ping Yi[1, #], Yalong Jiang[1, #], Yitian Cao[1], Fangfang Liu[2], Yun Zhu[1], Jiayi Xu[1], Zechao Yang[1], Chuanqi Huang[2], Wenshao Yang[2], Hongying Mao[1, *], Jian-Qiang Zhong[1, *]

[1] School of Physics, Hangzhou Normal University, Hangzhou, Zhejiang 311121, China

[2] Hangzhou Institute of Advanced Studies, Zhejiang Normal University, Hangzhou, Zhejiang 311231, China

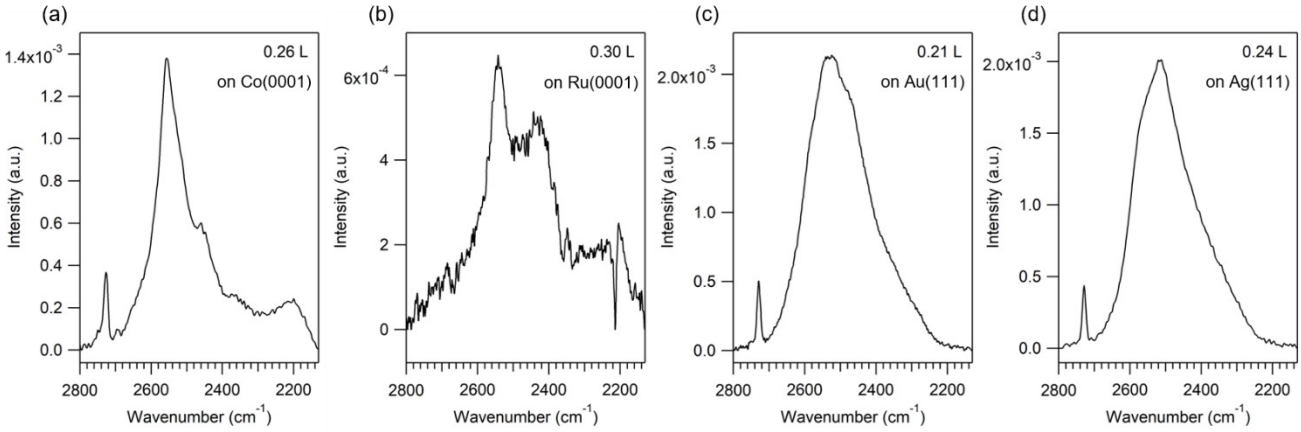

Figure S1. IRRAS spectra of the OD stretching mode for D_2O adsorbed on different metal surfaces at 100 K. (a) Co(0001), (b) Ru(0001), (c) Au(111), and (d) Ag(111). The respective coverages of D_2O are indicated in the figures.

The above spectra were acquired using similar experimental procedures. Notably, D_2O on Co(0001) forms two structures at approximately one adsorbed layer coverage: D-up and D-down geometries, as discussed in the paper. Conversely, for D_2O/Ru(0001), the overall intensity of the OD stretching mode is suppressed, and no free OD stretching is observed at a coverage of 0.3 L, indicating intact adsorption with almost flat-lying orientations, consistent with previous studies. In contrast, D_2O/Au(111) and D_2O/Ag(111) exhibit broad peaks with the highest intensity of the OD stretching mode. Furthermore, the intensity of the free OD stretching is nearly double that on Co(0001). However, no peak is observed around 2200 cm^{-1}, suggesting an upright orientation of D_2O on Au(111) and Ag(111).

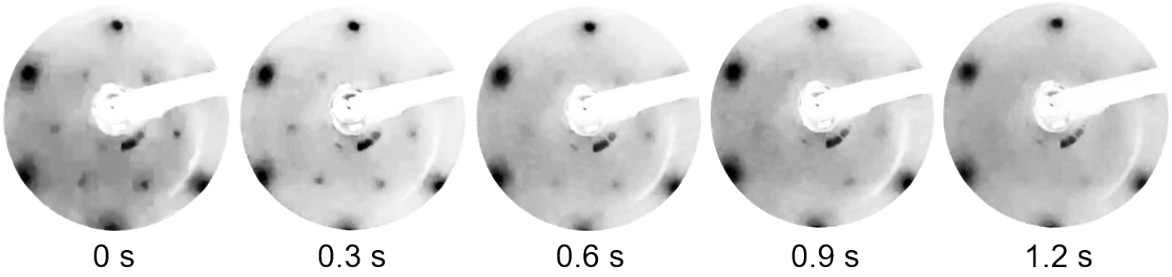

Figure S2. The effect of low-energy electron beams on the LEED patterns for 0.30 L D_2O adsorbed on Co(0001) at 100 K. E = 80 eV.

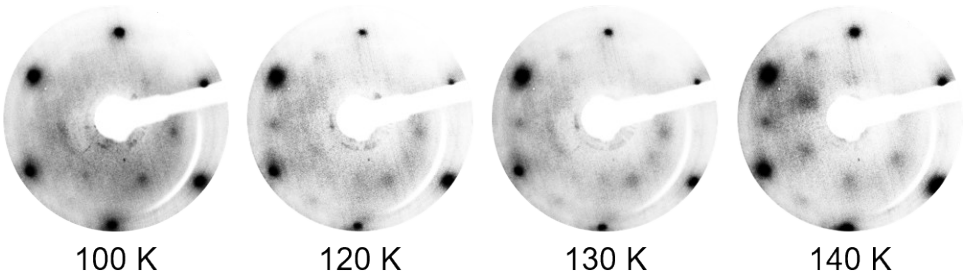

Figure S3. The effect of annealing temperatures on the LEED patterns for 027 L D_2O adsorbed on Co(0001) at 100 K. E = 84 eV.

It has been observed that the water layer is susceptible to damage from the low-energy electron beam (Figure S2). Therefore, all LEED patterns in our studies were acquired within one second on fresh spots or freshly prepared samples to minimize electron beam-induced damage. Additionally, the temperature of the Co(0001) substrate can potentially lead to the reconstruction or dissociation of water molecules. As shown in Figure S3, the $(\sqrt{3}\times\sqrt{3})R30°$ spots transformed into p(2×2) spots during gradual annealing of the sample from 100 K. These p(2×2) spots were observable around 130 – 150 K. consistent with the findings of Xu et al. (J. Phys. Chem. C, 114, 17023-17029, 2010).

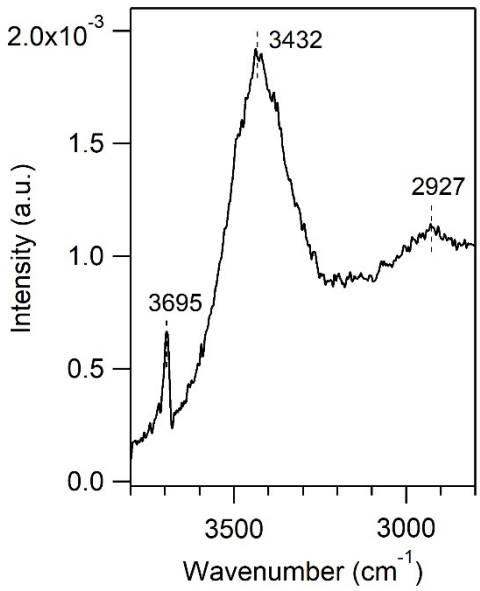

Figure S4. IRRAS spectra of the OH stretching mode for 0.3 L H_2O adsorbed on Co(0001) at 100 K.

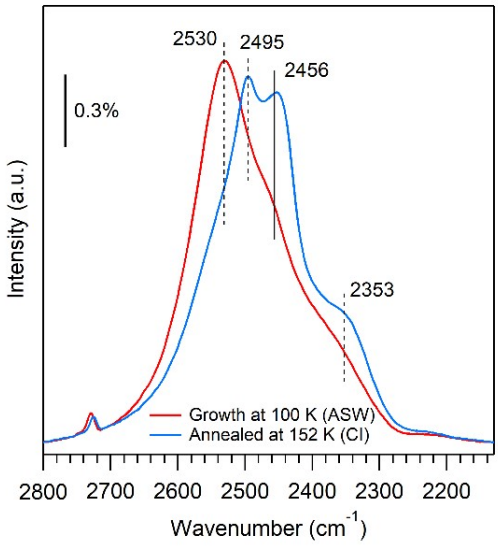

Figure S5. IRRAS spectra of amorphous solid water (ASW) and crystalline ice (CI) films on Co(0001). The ASW film is approximately 6.6 BL thick, while the CI film was obtained by annealing the ASW in UHV at 152 K.

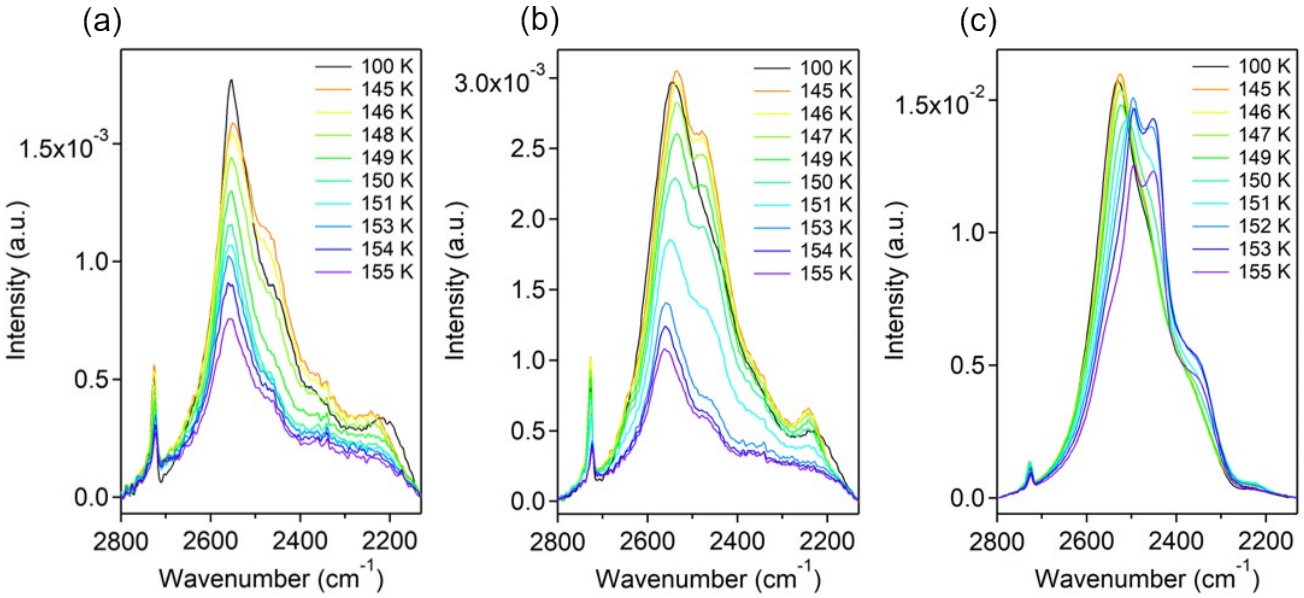

Figure S6. Temperature-dependent IRRAS spectra of the OD stretching mode for D_2O/Co(0001) adsorbed at 100 K with coverages of (a) 0.32 L, (b) 0.53 L, and (c) 1.78 L.