



Title	Surface Temperature Dependence of Hydrogen Ortho-Para Conversion on Amorphous Solid Water
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Supplemental Material for Surface Temperature Dependence of Hydrogen Ortho-Para Conversion on Amorphous Solid Water

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(1) Experimental procedure

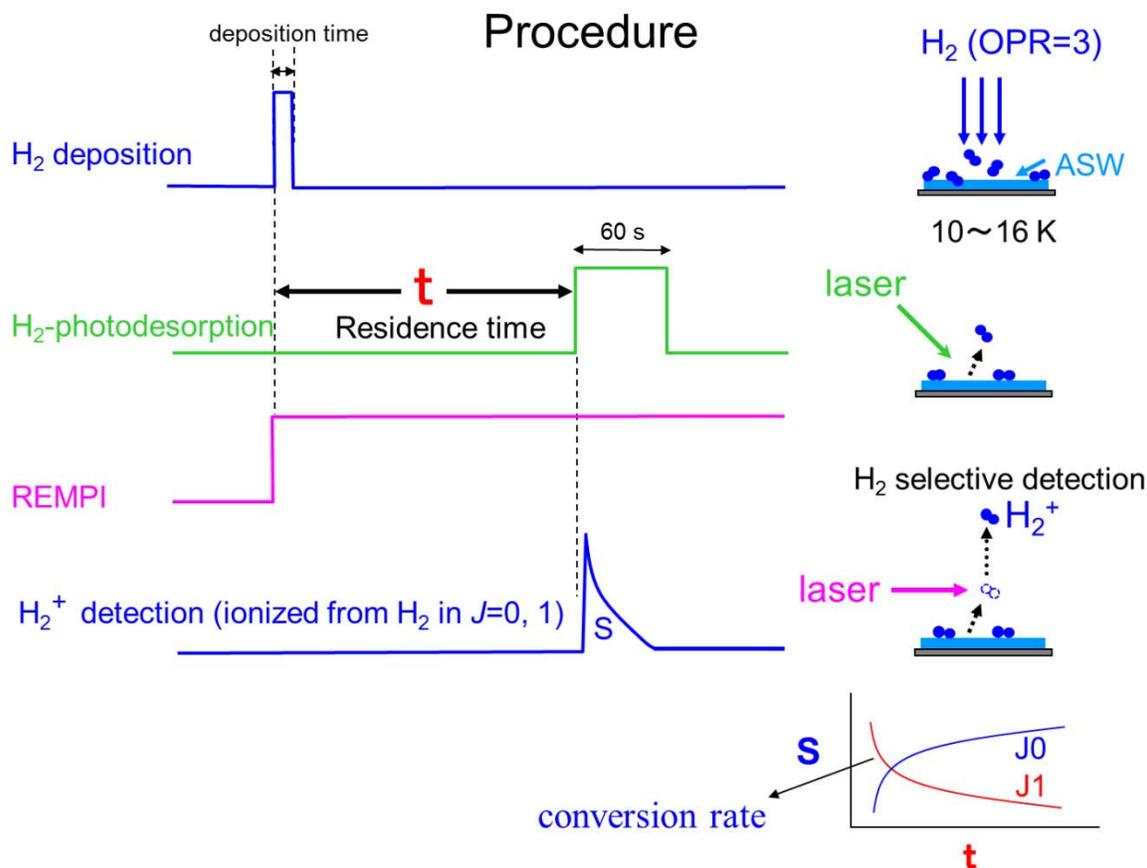


Fig. S1. Experimental procedure. A series of H₂ molecular pulses are first deposited onto ASW produced by vapor deposition of H₂O (>18 MΩ cm resistance) carefully degassed by repeating freeze-pump-thaw cycles (upper blue line). The REMPI laser injection at the resonant wavelength is started after the H₂ deposition (pink line). After the period of time (residence time) following the H₂ deposition, the PSD laser at a repetition rate of 10 Hz photodesorbs H₂ on ASW for 60 s (green line). The photodesorbed H₂ is state-selectively ionized by REMPI and detected (lower blue line, corresponding to H₂ intensity in Fig 1). It should be noted that although the REMPI laser is always running at a repetition rate of 10 Hz with the power of 200-250 μJ/pulse but no signal is detected before the PSD laser injection (during residence time). The H₂ molecular pulse is produced by expansion of pure H₂ gas (300 kPa) through a pulsed valve (100 μm-diameter hole) and subsequent a series of skimmer and orifice at room temperature. The diameter of the H₂ beam spot at the sample surface was about 3 mmφ. The kinetic energy of H₂ beam was not measured. However, based on the ideal gas law in which internal energies of molecule are converted to the kinetic energy, an upper limit of H₂ incident kinetic energy is 90 meV under our experimental condition. As a reference, E_i of ~73 meV was reported under similar expansion condition for pure H₂ beam (Farias & Miranda, Prog. Surf. Sci. **86**, 222 (2011); Groot et al. JCP, **127**, 244701 (2007)). In any case, since the ortho-

to-para ratio of H₂ photodesorbed after 10 s from the deposition was almost 3, the kinetic energy of H₂ beam does not affect the conversion process.

(2) Para-to-ortho conversion for D_2 molecule on ASW at 16 K

At 16 K, the D_2 molecules were deposited on ASW and the para-to-ortho conversion was measured.

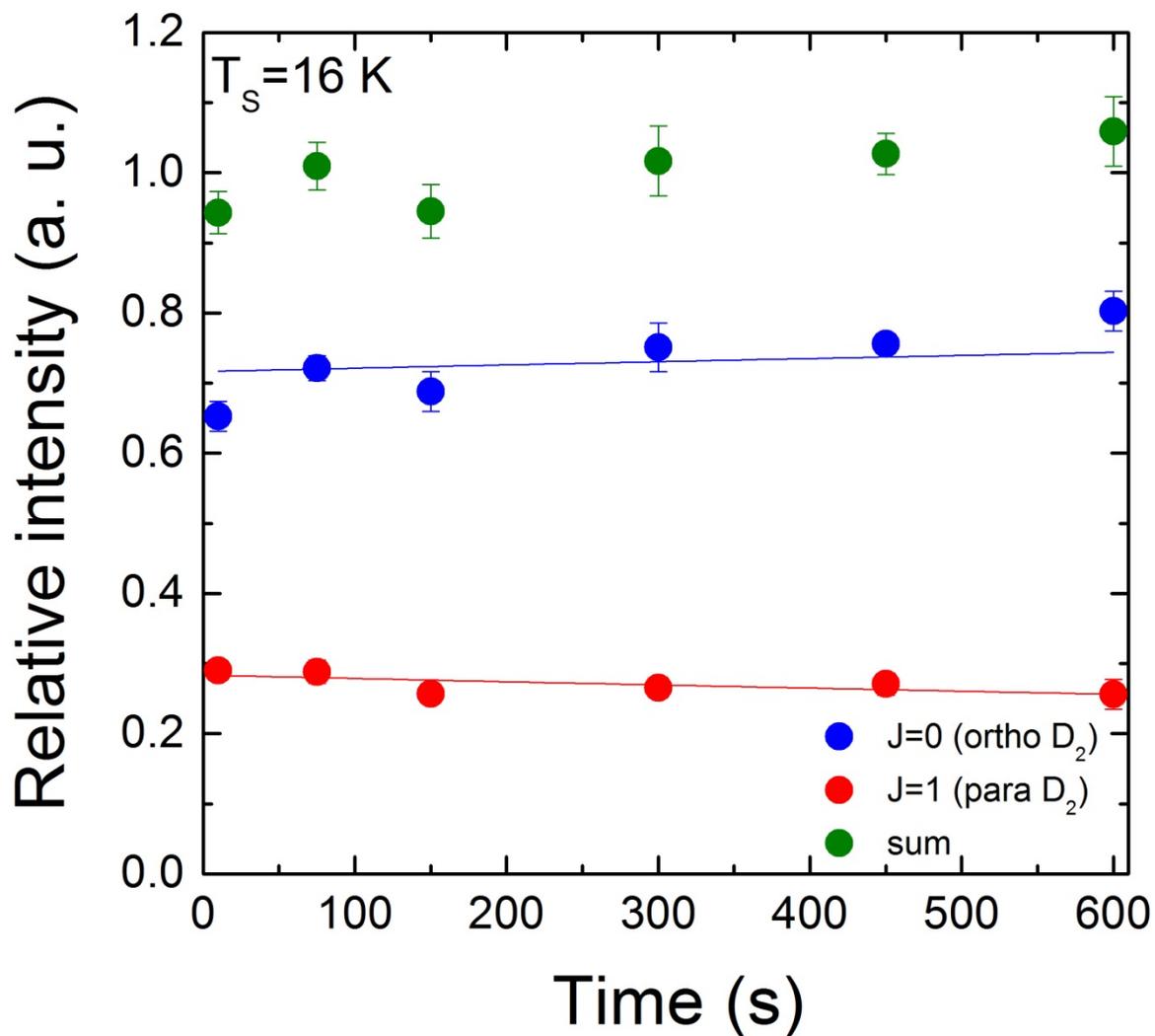


Fig. S2. The time evolution of photo-desorbed D_2 intensities at $T_s = 16$ K. The origin of the x axis corresponds to the opening of the pulsed valve of the D_2 molecular beam source. The error bars represent the confidence reliabilities.

(3) Comparison of conversion rates at 10 K between samples for H₂-depositions at 10 K and 12 K

The OP conversion was measured for H₂ at 10 K which was initially deposited at 12 K. In this experiment, the temperature of ASW was lowered to 10 K within ~15 s immediately after H₂-deposition at 12 K. As shown in Fig. S3, the OP conversion rate obtained in the measurement is almost equivalent to that shown in Fig. 2 (a), where the surface temperature was held at 10 K for entire experiment including H₂ beam dosing.

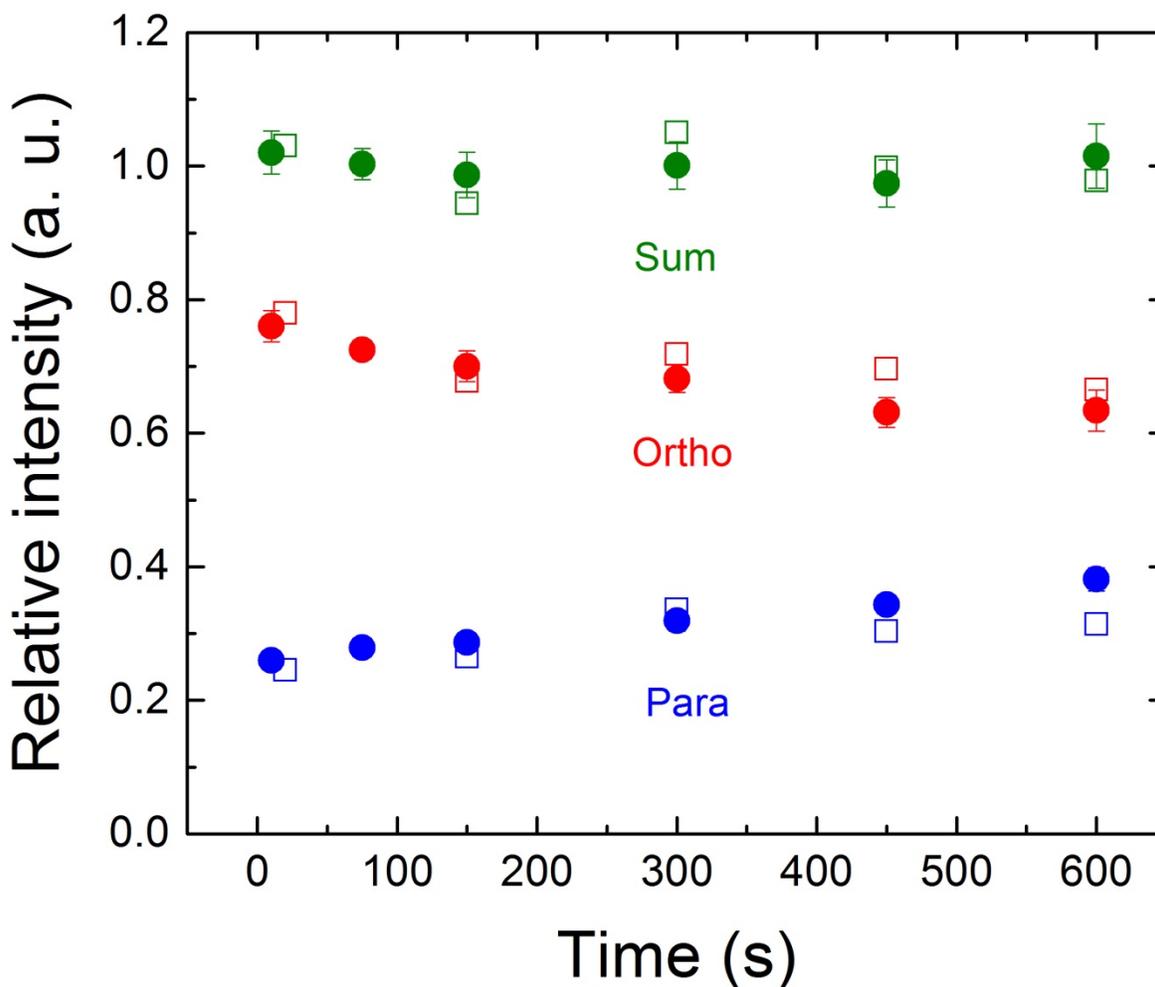


Fig. S3. The time evolution of photodesorbed H₂ intensities for J=0 (blue) and J=1 (red) and their sum (green). Open squares and solid circles represent the results for H₂-deposition at 12 K and that for H₂ deposition at 10 K taken from Fig. 2 (a). The scale in x-axis is defined by H₂ beam dose and includes the time of cooling down to 10 K from 12 K.