NUCLEAR-SPIN TEMPERATURE OF WATER MOLECULES THERMALLY DESORBED FROM ICE: A LABORATORY STUDY

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Introduction: The nuclear-spin temperatures (T_{spin}) are derived from the ortho-to-para ratio (OPR) of molecules. In the case of H_2O, it contains two protons with spin of 1/2; thus, its total spin state can be either 0 (singlet, para) or 1 (triplet, ortho). The OPR of H_2O is equal to 3 in statistical equilibrium, which is achieved at temperatures above ~50 K. In cometary coma, T_{spin} of H_2O molecules has been derived in the range from 21 to greater than 50 K, typically ~30 K [1]. These values have been implicated as the temperature of cold grains at molecular condensation or formation in a molecular cloud, or in the solar nebula [2]. However, the real meaning of the observed T_{spin} remains a topic of continuing debate. The present study experimentally measured the T_{spin} of H_2O thermally desorbed from pure amorphous solid water (ASW) deposited at 8 K by a resonance-enhanced multiphoton ionization (REMPI) method. We also produced ASW at 8 K by photolysis of a CH_4/O_2 mixture (photoproduced ASW), considering that T_{spin} of H_2O molecules may relate to formation processes on cold dust grains.

Experimental: The experiment was performed with the RASCAL apparatus at Institute of Low Temperature Science, which consists of a vacuum sample chamber, a Fourertransform infrared (FTIR) spectrometer, a UV excimer lamp, and a laser system. An aluminum sample substrate connected to the cold head of an He refrigerator was installed at the center of the sample chamber. The thickness of vapor-deposited pure ASW was approximately 500 monolayers. The present study also employed photolysis of CH_4 and O_2 mixed ices using the UV excimer lamp to yield H_2O molecules at the low temperature surface. The prepared ice samples were heated to ~150 K to sublime H_2O from the solid samples at a heating rate of typically 4 K min^{-1}. The desorbed H_2O molecules were analyzed rovibrationally by the 2+1 REMPI method in the laser wavelength range of 248.1–248.6 nm. The focal point of the REMPI probe laser was about 1 mm from the substrate surface. H_2O^+ ions formed by REMPI were detected using a time-of-flight mass analyzer.

Results: The present experimental procedures and results are summarized in Fig. 1. Thermally desorbed H_2O molecules from all ice samples prepared at 8K showed T_{spin} almost at the statistical high-temperature limit (≥ 30 K). The value of T_{spin} was almost identical even after leaving the deposited ASW for 9 days at 8 K. Figure 2 shows the REMPI spectrum of desorbed H_2O for the ASW produced from the photolysis of CH_4 and O_2 solid mixture at 8 K. The spectrum was best reproduced by the simulation with $T_{rot} = T_{spin} = 150$ K, where T_{rot} represents rotational temperature. These results suggest that the T_{spin} of gaseous H_2O molecules thermally desorbed from ice does not necessarily reflect the surface temperature at which H_2O molecules condensed or formed. We discuss the possibility of nuclear-spin conversion of H_2O in water ice [3].

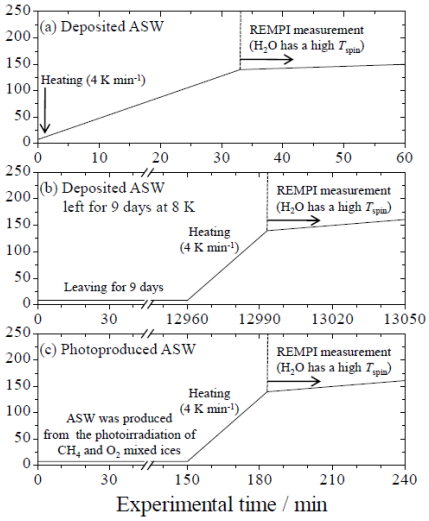

Figure 1. Typical temperature profiles of (a) deposited ASW, (b) deposited ASW left for 9 days at 8 K, and (c) photoproduced ASW. The ice samples were prepared at 8 K. Thermally desorbed H_2O molecules from all ice samples show T_{spin} almost at the statistical high-temperature limit (≥30 K).

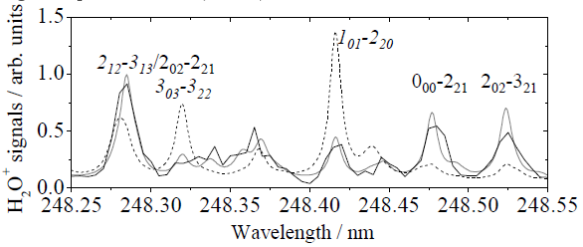

Figure 2. (The black line) 2+1 REMPI spectrum of thermally desorbed H_2O from photoproduced ASW. (The gray line) The best-fitting simulations with $T_{rot} = T_{spin} = 150$ K. (The dotted line) Reference simulations with T_{spin} =8 K. T_{rot} is fixed at 150 K. Indications ($J'_{Ka',Kc'}$-$J_{Ka,Kc}$) are rotational assignments in H_2O. H_2O molecules with (K_a + K_c = odd) are ortho, while those with (K_a + K_c = even) are para species (*italic transitions*).

References: [1] Bonev, B. P. et al. (2007) *ApJ, 661,* L97-L100. [2] Shinnaka, Y. et al. (2010) *PASJ, 62,* 263-271. [3] Hama, T. et al., (2011) *ApJ, 738,* L15 (5pp).