Temperature dependence of ortho-to-para conversion of H_2 on amorphous solid water at around 10 K

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The ortho-to-para abundance ratio (OPR) of H_2 is crucial for chemical evolution and deuterium fractionation of molecules in molecular clouds, because H_2 in the ortho-ground state (J=1) is more energetic (and thus reactive) than that in the para-ground state (J=0) by approximately 14.6 meV corresponding to 170 K, which is significantly higher than typical temperatures of molecular clouds. Since the radiative transformation of molecular nuclear spins is forbidden in the gas phase, the OPRs of hydrogen molecule (H_2) observed toward various astronomical objects have been often considered as tracers of chemical history of the molecule.

In the gas phase, spin-exchange reaction with either ionic or neutral hydrogens can alter the OPR of H_2. In contrast, little is known about how the nuclear spins behave on cosmic ice dust. It was often assumed without the experimental evidences that the OPR of H_2 formed on the cosmic dust surface is statistical value of 3. Recently, our group has tackled this issue experimentally and found that the OPR of nascent H_2 is 3 at the formation by H-H recombination on cosmic ice dust [1]. In the preset study, using experimental techniques of molecular beam, photostimulated-desorption, and resonance-enhanced multiphoton ionization, we measured the OPRs of H_2 photodesorbed from amorphous solid water (ASW) at temperatures around 10 K, which is an ice dust analogue. We demonstrated that the OPR of H2 easily varies on ASW and that the rate of spin conversion from ortho to para steeply increases from 2.4×10^{-4} to 1.6×10^{-3} s^{-1} within the very narrow temperature window of 9.5 to12 K and reach a plateau at temperatures above 12 K [2]. This temperature dependence cannot be explained solely by state-mixing models ever proposed [3]. The temperature dependence of the conversion rate was very well represented by the power law of T^7 where T is the surface temperature. This indicates that the conversion rate at lower temperatures is dominated by the two-phonon energy dissipation process (Raman process) for the excess energy (~14.6 meV) arising from the conversion, and at higher temperatures the rate may be limited by a spin-flip transition rate.

From the present and our previous results, astrochemical history of interstellar H_2-OPR is depicted as follows. When H_2 molecule is produced by H-H recombination on cosmic ice dust, the OPR of nascent H_2 is 3 [1]. If the H_2 is immediately desorbed at the formation, the OPR of H_2 released should be near 3. However, if H_2 is trapped even for a short period on the ice dust surface before desorption, the OPR at desorption strongly depends on the surface temperature and duration of trapped.

References

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