RESONANT TUNNELING OF HYDROGEN IN PD

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Hydrogen (H) exhibits quantum natures at low temperature. Quantum tunneling has been observed for H on metal surfaces and muon in metals. At intermediate temperature, two types of transitions were experimentally observed in different materials: abrupt and gradual transitions. However, the role of nuclear quantum effects at the crossover region from thermal hopping to quantum tunneling is still not clear. From a theoretical viewpoint, vibrational excitation of H has to be taken into account to elucidate the H hopping mechanism at the intermediate temperature. In this study, we successfully measured the H hopping rate in Palladium (Pd) at the intermediate temperature region and calculated the quantum state of H in three-dimensional potential of Pd. It is revealed that the hydrogen hopping is governed by resonant tunneling via excited vibrational states around the crossover from thermal to quantum regimes.

The sample was 50-nm-thick polycrystalline Pd films deposited by magnetron sputtering. Resistance change called the 50 K anomaly was observed after hydrogenation followed by rapid cooling, which is known to be related to H ordering. The H hopping rate was derived from the time variation of resistance. Figure 1(a) shows the temperature dependence of hydrogen hopping rates of $PdH(D)_x$ with various x's. Enhancement of hopping was observed around 60 K only for PdH_x with small x. To elucidate the origin of its enhancement, we calculated the quantum state of H(D) by solving the Schrödinger equation for H(D) in three-dimensional potential of Pd. As shown in Fig. 1(b), the quantum state extended between the second excited vibrational state at the octahedral (O) site and the ground state at the tetrahedral (T) site was observed for H beneath the barrier top, indicating that H can diffuse via quantum tunneling between the two states. This corresponds to resonance tunneling. Concentration dependence of the site energy showed that the T site becomes unstable as the H concentration increases, which results in the energy level mismatch between the two states. We revealed that the vibrational excitation and the energy level matching between the discrete vibrational states determine the H(D) hopping via the quantum tunneling around the crossover region between thermal and quantum regimes.

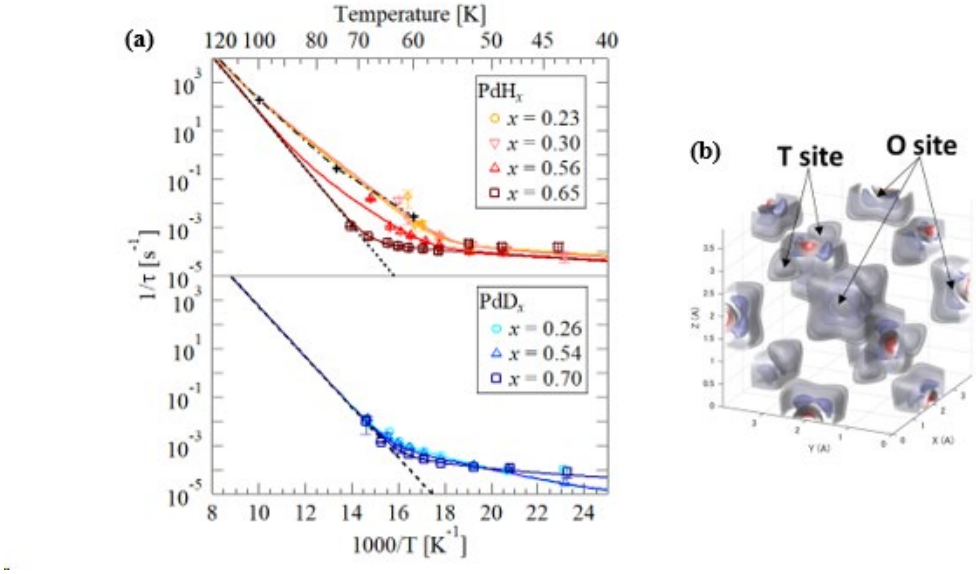

Figure 1 (a) Hopping rates of H and D in a 50-nm-thick Pd film. (b) Calculated quantum state of H in Pd.