

Nuclear spin conversion of hydrogen molecules trapped and polarized in solid carbon dioxide

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According to the total nuclear spin I , a hydrogen molecule (H_2) is classified into either the ortho ($I=1$) or para ($I=0$) isomer. Since the total wave function of H_2 must be antisymmetric under the proton permutation, the ortho (para) species takes only odd (even) values of the rotational quantum number J . This coupling allows us to monitor the nuclear spin conversion (NSC) with use of rovibrational spectroscopy. The conversion from ortho to para is strictly forbidden in the isolated state but has been found to proceed in condensed systems [1]. In particular, NSC of H_2 interacting with a diamagnetic insulator has been intensively investigated recently, and its mechanism is still open to discussion [2,3]. In the present study, we detected weak infrared absorption by H_2 trapped and polarized in solid carbon dioxide (CO_2) and measured the NSC rate of H_2 to discuss the NSC mechanism [4].

Gases of H_2 and CO_2 were mixed in a gas handling system at the molar ratio of $CO_2/H_2=100$. The gas mixture was introduced into an ultrahigh-vacuum chamber and was condensed on a gold plate, which was cooled down to 5.4 K. Spectra were measured with a Fourier transform infrared spectrometer and an external HgCdTe detector in the reflection configuration.

Figure 1 shows the time evolution of the infrared absorption band of H_2 trapped in solid CO_2 . We defined $t = 0$ s as the moment when the condensation was completed. The absorption band had two components; P_1 at 4149 cm^{-1} grew with time whereas P_2 at 4138 cm^{-1} decayed. By using Gaussian fitting, we determined the integrated intensities of P_1 and P_2 as functions of time, which are displayed in Fig. 2. We found that both the components exhibited the monoexponential time-dependence and obtained the relaxation rates as follows: $k_1 = (9.6 \pm 0.4) \times 10^{-4}\text{ s}^{-1}$ for P_1 and $k_2 = (9.2 \pm 0.4) \times 10^{-4}\text{ s}^{-1}$ for P_2 . Since these rates coincided within the experimental error, we ascribed the time evolution observed in Fig. 1 to the rotational relaxation of H_2 from $J = 1$ to $J = 0$ accompanied by NSC from ortho to para; in other words, we assigned P_1 to para H_2 and P_2 to ortho H_2 . The inset of Fig. 2 is a schematic diagram of the vibrational transitions corresponding to P_1 and P_2 .

In a previous study, the electric-field-induced conversion model was proposed to describe NSC of H_2 adsorbed on amorphous ice [2]. However, the small vibrational shift of P_1 from the gas-phase value of 4161 cm^{-1} eliminates the possibility that this model is applicable to H_2 inside solid CO_2 . Instead, we found that the measured NSC-rate agrees with the rate of $1 \times 10^{-3}\text{ s}^{-1}$ estimated from the three-step conversion model [3], where the Coulomb interaction causes the electron

exchange between H_2 and solid CO_2 in the first step, the hyperfine interaction brings about the simultaneous singlet-triplet transitions of the electronic and nuclear states of H_2 in the second, and the spin-orbit interaction finally realizes the electronic singlet-triplet transition inside the solid. The way of non-destructively measuring NSC of H_2 , established in the present study, can be applied to other systems and will help to reveal the more detailed mechanism of NSC in diamagnetic insulators.

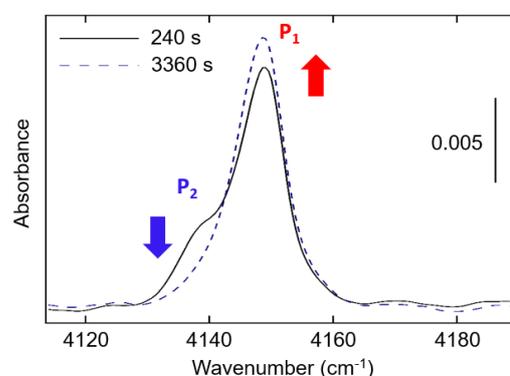


Fig.1 Time development of the infrared absorption band of H_2 trapped and polarized in solid CO_2 at 5.4 K.

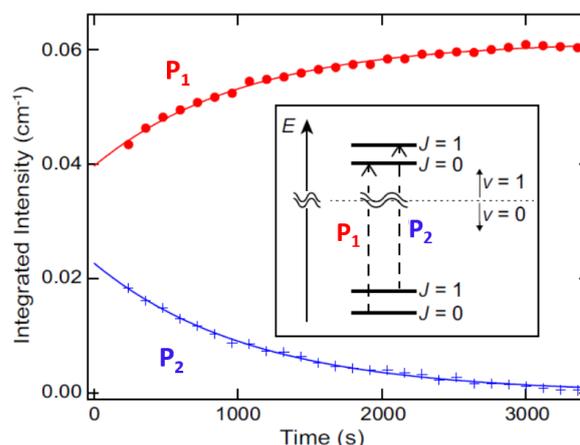


Fig.2 Time-dependent integrated intensities of the absorption components of para H_2 (P_1 , circles) and ortho H_2 (P_2 , crosses). The results of monoexponential fitting are represented by solid lines. The inset schematically shows the infrared transitions of P_1 and P_2 . The quantum number of the stretching vibration is represented by v .

References

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