CO および CO_2 凝縮層中に捕捉された H_2 の赤外分光

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Infrared spectroscopy of H_2 trapped in condensed films of CO and CO_2

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Field and co-workers have recently suggested that dipolar molecules such as CO and N_2O, which are vapor-deposited at a low temperature, spontaneously align and exhibit large electric fields ($> 10^8$ V/m).[1,2] They employed two kinds of experimental methods: direct measurement of the surface electric potential of a substrate[1] and analysis of phonon-induced splitting in the infrared absorption band due to the internal vibration of condensed dipolar molecules.[2] We focused on the fact that measurement of the induced dipole moment of a homonuclear diatomic molecule is also useful for the evaluation of the electric field strength. In fact, Dai et al. detected induced infrared absorption of H_2 on an annealed NaCl film and calculated its infrared intensity (IRI), from which the electric field intensity on the film surface is derived.[3] In this work, we obtained the electric field strength in solid CO, as well as that in solid CO_2, CH_4 and Ar, evaluating IRI of contained H_2.

The experimental apparatus consists of a vacuum chamber equipped with a liquid helium cryostat, a Fourier transform infrared spectrometer, and a HgCdTe detector. The base pressure of the vacuum chamber was 1×10^{-8} Pa. Gas mixtures of X (X = CO, CO_2, CH_4, Ar) and H_2 were prepared at the ratio of X / H_2 = 10 and were condensed on a gold substrate at 5 K. Infrared absorption spectra were measured in the reflection geometry with the incident angle of 80 degrees.

In the spectrum of H_2-containing solid CO at 5 K, we detected a strong and broad infrared absorption band of ^{12}CO along with a sharp peak of ^{13}CO. Since the integrated absorbance (IA) of ^{13}CO was much easier to be analyzed than that of ^{12}CO, the column density of ^{12}CO was determined from IA, IRI, and the natural abundance (1.1%) of ^{13}CO. Using the mixing ratio of CO / H_2 = 10, we also obtained the column density of H_2. Here, it was

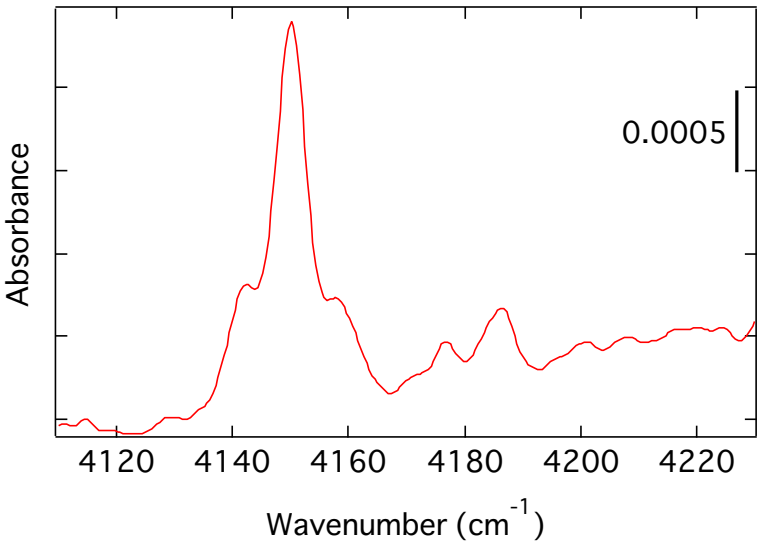

Fig.1 : Infrared spectrum of H_2 in solid CO vapor-deposited at 5 K. (CO / H_2 = 10)

assumed that the trapping coefficient of H_2 is unity. Figure 1 shows the spectrum in the range of the stretching vibration of H_2. The induced infrared absorption band of H_2 extends from 4130 to 4190 cm^{-1}. This indicates that the field in solid CO induces the electric dipole moment in H_2. The minute structure of the absorption band reflects the existence of different binding sites of H_2 in solid CO. IRI of H_2 was determined from IA of this band and column density of H_2 obtained above. Using the relation between the electric field strength, E_{CO}, and IRI of H_2,[3] we obtained $E_{CO} = 5.5 \times 10^9$ V/m.

In the case of solid CH_4 and Ar, infrared absorption by H_2 was not detected. This result is reasonable because of the following facts: 1) neither electric dipole nor quadrupole moment exists in CH_4; 2) Ar has no electric multipole moment. On the other hand, infrared absorption by H_2 in solid CO_2 was clearly detected. This is shown in Fig. 2. Using IA of $^{13}CO_2$, the electric field strength was derived in the same way as solid CO: $E_{CO2} = 1.2 \times 10^{10}$ V/m. The result that a solid of CO_2 without the dipole moment has larger electric field than that of CO is rather surprising. However, the trapping coefficient of H_2 as well as the film structure should be properly taken into account. In fact, the cryotrapping effects of CO, CO_2, CH_4 and Ar on H_2 were discussed by Wallen.[4] Additionally, the calculated electric field intensities were found to become smaller when the mixing ratio was set to be X / H_2 = 1 (X = CO, CO_2). In this presentation, we will discuss the electric field strength in solid CO and CO_2, considering the structures of the condensed layers and their trapping coefficients.

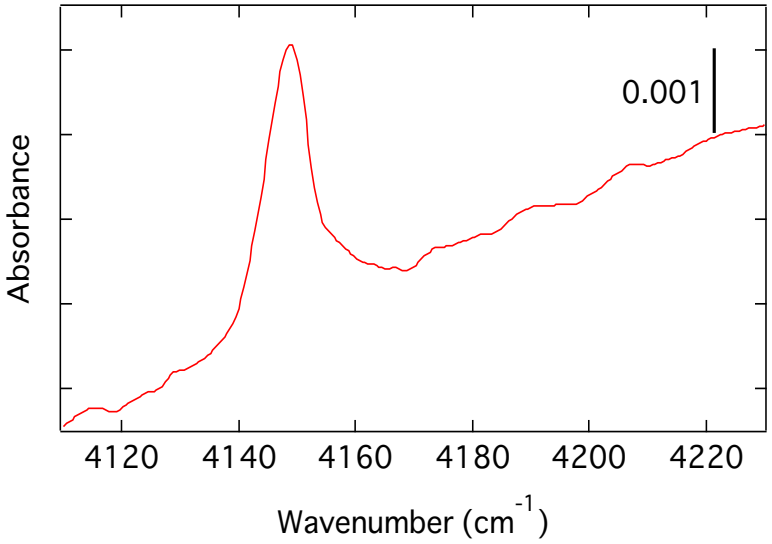

Fig. 2 : Infrared spectrum of H_2 in solid CO_2 vapor-deposited at 5 K. (CO_2 / H_2 = 10)

[1] R. Balog et al., Phys. Rev. Lett. 102 (2009) 073003.
[2] J. Lasne et al., Phys. Chem. Chem. Phys. 17 (2015) 20971.
[3] D. J. Dai and G. E. Ewing, J. Chem. Phys. 98 (1993) 5050.
[4] E. Wallen, J. Vac. Sci. Technol. A 14 (1996) 2916.