ROVIBRATIONAL MOTION OF METHANE IN PARAHYDROGEN CRYSTAL, AND ANOMALOUS TEMPERATURE DEPENDENCE OF THE SPECTRAL LINEWIDTH

HIROYUKI KATSUKI, HIROMICHI HOSHINA, and TAKAMASA MOMOSE, *Division of Chemistry, Graduate School of Science, Kyoto University, Kyoto 606-8502, JAPAN.*

During the past several years we have been utilizing parahydrogen crystal as a new matrix for matrix isolation spectroscopy.[a] Most of the observed spectral lines are much sharper than those observed in the conventional matrices, which makes parahydrogen crystal an excellent host medium for the spectroscopic study of rovibrational states of guest molecules. This paper presents a high-resolution infrared spectroscopic study of rovibrational states of methane and its deuterated species isolated in parahydrogen crystals.[b] The observed triply degenerate stretching (ν_3) and the triply degenerate bending (ν_4) modes of methane are fully analyzed in terms of a model in which the methane molecule occupies a substitutional site of the crystal. The crystal field potential as well as the rotational constant, Coriolis coupling constant, and the band origins of methane in the crystal is determined by the least-squares fitting of the observed transition frequencies. The rotational constant of methane in the hydrogen crystal is found to be about 90 % of the gas phase value. The reduction of the constant is caused by the dispersive interaction between methane and hydrogen molecules. It should also be added that a remarkable dependence of the linewidth upon temperatures in the range of 4 to 8 K is observed, which suggests an intrinsic nature of rovibration-lattice interaction in the quantum crystal.

[a] T. Momose and T. Shida, *Bull. Chem. Soc. Jpn.* **71**, 1 (1998).

[b] T. Momose, M. Miki, T. Wakabayashi, T. Shida, M. -C. Chan, S. S. Lee and T. Oka, *J. Chem. Phys.* **107**, 7707 (1997); H. Hoshina, T. Wakabayashi, T. Momose, and T. Shida, *J. Chem. Phys.* **110**, in press (1999).