P154-032 **Room: 201B** **Time: May 17 9:39-9:52**

Phase Changes of Solid Methane under High Pressure and their Implications in Interior of Icy Planets

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High pressure experiments of solid methane were performed in the pressure range of 0.5 to 86 GPa at room temperature and high temperature using diamond-anvil cell. In-situ X-ray diffractmetry and Raman spectroscopy, above 25 GPa, two phase changes were revealed at 38 and 60 GPa and at room temperature . The X-ray diffraction patterns of the high-pressure phases, called HP2 and HP3 in this study, were indexed as simple cubic lattice, and these phases were inferred to possess different orientation of methane molecules in some common fundamental structure.

A metastable phase was found between phase A and B which were previously reported. This phase was characterized by a single peak of n1 mode of Raman spectrum different from three-split peaks of phase B. Although the X-ray diffraction pattern was indexed as simple cubic lattice, the relative intensities of the peaks were totally different from those of phase B. Thus, this phase, called pre-B phase in this study, was thought to be different from phase A and B in the basic structure and the intramolecular vibration mode of methane. A series of A/pre-B/B phase changes were observed as follows. At first, phase changed rapidly to the phase pre-B at approximately 10GPa. The phase pre-B changed sluggishly to phase B, accompanied with preferred orientation of methane molecules. Phase changes from this pre-B phase without passing phase B were also investigated. The phase changes from the pre-B phase were observed at 25, 45 and 67 GPa, and their XRD patterns were indexed as simple cubic lattice which are same as that of pre-B phase. In the Raman spectrum, splitting of n1 mode which is characteristic to phase B were not observed.

High temperature experiments were made under various conditions by using CO2 and YAG laser heating. X-ray diffractmetry and Raman spectroscopy were performed after the samples were cooled. Under a condition of 66GPa and 700K, any change was not observed. Under those of 53GPa/~2500K, and 81GPa/~2500K, some melting-like texture suggesting to melting was observed after cooling. The XRD pattern indicated amorphization of the sample, and Raman spectrum showed any other peak of hydrocarbon but methane were not observed. These results indicated existence of only methane molecules and absence of ethane, propane and diamond. On the basis of the experimental results, interiors of icy planets, Uranus and Neptune were considered. It is likely to exist methane molecules in shallow or middle region of Middle Ice Layer. And their methane-rich atmosphere might be kept by supplying methane from their melting locally in Ice Layers.