Abstract Submitted
for the SHOCK13 Meeting of
The American Physical Society

Phase changes induced by guest ordering of filled ice Ih structure of methane hydrate under high pressure and low temperature HISAKO HIRAI, TAKEHIKO TANAKA, Geodynamics Research Center, Ehime University, TAKAHIRO MATSUOKA, KYOKUGEN, Osaka University, YASUO OHISHI, Japan Synchrotron Radiation Research Institute, TAKEHIKO YAGI, SHINGO KAGAWA, Geodynamics Research Center, Ehime University, MICHIKA OHTAKE, YOSHITAKA YAMAMOTO, The National Institute of Advanced Industrial Science and Technology — Orientational ordering of guest methane molecules in a filled ice Ih structure of methane hydrate (MH-FIIhS) was observed above 20GPa by our previous Raman study, whereas change in a fundamental structure was not detected by XRD. In this study, in-situ XRD studies were performed in the temperature range from 300 to70 K at the pressures up to 57 GPa. The results revealed that the lattice parameters changed continuously with increasing pressure, however, clear changes in axes ratios were shown. At about 20 GPa the slopes of axis ratios, b/c and c/a, changed abruptly at room temperature. The Raman spectroscopy showed split of CH vibration mode of the methane molecules, which indicates the orientational ordering of the guest methane molecules, at the almost same pressure. These results demonstrated that the changes in axis ratio were caused by orientational ordering of the guest molecules. Similar changes in the axis ratios and split of CH vibration mode were observed at low temperature regions. The regions of the guest-ordered phase and the guest-rotated phase were roughly estimated from the experimental results.

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Date submitted: 26 Feb 2013 Electronic form version 1.4