TETRAHYDROFURAN CLATHRATE HYDRATE FORMATION STUDIED BY X-RAY RAMAN SCATTERING

Heiko Conrad$^1$, Felix Lehmkühler$^1$, Christian Sternemann$^1$, Arto Sakko$^2$, Omid Feroughi$^1$, Laura Simonelli$^3$, Simo Huotari$^2$, Dietmar Paschek$^4$, Keijo Hämäläinen$^2$, and Metin Tolan$^1$

$^1$ Technische Universität Dortmund, Fakultät Physik/DELTA, Maria-Goeppert-Mayer-Str. 2, 44221 Dortmund, Germany
$^2$ Department of Physics, P.O.B. 64, FIN-00014 University of Helsinki, Finland
$^3$ ESRF, BP 220, 38043 Grenoble Cedex 9, France
$^4$ Rensselaer Polytechnic Institute, 110 8th Street, Troy, 12180 NY, USA
email: felix.lehmkuehler@tu-dortmund.de

Clathrate hydrates are ice-like inclusion compounds where guest molecules are embedded in water nano-cages. A possible future application of hydrates is the storage of gases, in particular H$_2$ in THF hydrate for fuel cells [1]. Therefore, a detailed knowledge of the hydrate formation process is important which is still not fully understood on a microscopic level. Competing formation models are reported in literature, which predict hydrate precursors in a supercooled state before the hydrate nucleation starts or a stochastic formation without any hydrate pre-structures, see Fig. 1.

The formation of clathrate hydrates in the system water-Tetrahydrofuran (THF) was studied by means of non-resonant x-ray Raman scattering (XRS). XRS is an energy loss spectroscopy which allows to measure light element x-ray absorption using hard x-rays. In the dipole limit XRS directly corresponds to x-ray absorption spectroscopy (XAS). XRS spectra are sensitive to possible hydrate precursors in a supercooled state and thus yield unique information about the hydrate formation process. X-ray Raman spectra of the oxygen K-edge were measured of the hydrate, of a supercooled liquid THF/water mixture, and of a mixture at a temperature above the region of hydrate stability. The measurements were compared with a DFT calculation method [3] employing local structures obtained from molecular dynamics simulation snapshots.