ESRF	**Experiment title:** Probing the guest dynamics and configurational energies of THF hydrate by x-ray Compton scattering	**Experiment number**: HE-3157
Beamline: ID15	**Date of experiment**: from: 04.11.2009 to: 11.11.2009	**Date of report**: 02.12.2012
Shifts: 18	**Local contact(s)**: Dr. D. Pontoni	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

F. Lehmkühler*, C. Sternemann*, S. Tiemeyer*, A. Nyrow,* I. Steinke,* Ch.J. Sahle*, and M. Tolan
Fakultät Physik/DELTA, Technische Universität Dortmund, D-44221 Dortmund, Germany

S. Galambosi,* A. Sakko, M. Hakala, and K. Hämäläinen*
Department of Physics, POB 64, FIN-00014 University of Helsinki, Finland

Report:

Parts of this experiment have been published in:

F. Lehmkühler, A. Sakko, I. Steinke, C. Sternemann, M. Hakala, Ch.J. Sahle, T. Buslaps, L. Simonelli, S. Galambosi, M. Paulus, T. Pylkkänen, M. Tolan, and K. Hämäläinen, Temperature induced structural changes of tetrahydrofuran clathrate and of the liquid water/tetrahydrofuran mixture, Journal of Physical Chemistry C **115**, 21009 (2011).

Abstract: We present two complementary inelastic X-ray scattering studies on the structure of tetrahydrofuran (THF) clathrate hydrate and the supercooled stoichiometric liquid mixture of water and THF. Compton scattering experiments of the liquid mixture show that formation of hydrate precursors is unlikely. By comparing experimental spectra of THF hydrate and water/THF mixtures at temperatures above 250 K with density functional theory calculations, structural changes that manifest in OH bond length changes are observed. X-ray Raman scattering measurements of the oxygen K-edge in the same temperature range corroborate these results. The experimental results of THF hydrate at temperatures between 20 and 244 K can be modeled best by assuming thermal expansion only. Therefore, dependency on the system's temperature different structural behavior of THF hydrate is reported.

F. Lehmkühler, A. Sakko, C. Sternemann, M. Hakala, K. Nygard, C.J. Sahle, S. Galambosi, I. Steinke, S. Tiemeyer, A. Nyrow, T Buslaps, D. Pontoni, M. Tolan, and K. Hämäläinen, Anomalous energetics in tetrahydrofuran clathrate hydrate revealed by x-ray Compton scattering, Journal of Physical Chemistry Letters **1**, 2832 (2010).

Abstract: Changes in the ground-state electron momentum density of tetra-hydrofuran clathrate hydrate are studied in a temperature range between 93 and 275 K by means of X-ray Compton scattering. At

temperatures above 253 K, large rather unexpected differences from the Compton profiles of ice are observed. Configurational enthalpies are extracted and exhibit a rapid rise above 253 K, whereas a constant configurational heat capacity of 0.23 +/- 0.07 J g(-1) K(-1) is found below 253 K. Density functional theory calculations suggest that this abomalous behavior originates from the structural change of the hydrate, however, no indication was found for the formation of hydrogen bonds between tetrahydro-furan and water molecules.