

CPP 7: Liquids

Time: Monday 18:00–19:30

Location: ZEU 160

CPP 7.1 Mon 18:00 ZEU 160

Doppler-broadening (DB) measurement of ionic liquids using a monoenergetic positron beam. — ●PHILIP PIKART¹, CHRISTOPH HUGENSCHMIDT², and KLAUS SCHRECKENBACH² — ¹Physik-Department E21, Technische Universität München, James-Frank-Strasse, 85748 Garching, Germany — ²ZWE FRM II, Technische Universität München, Lichtenbergstrasse 1, 85747 Garching, Germany

In this project, room-temperature ionic liquids are studied by positron annihilation spectroscopy which is a standard method for defect characterization of solid state samples. These "organic salts" contain essentially only weakly coordinated ions, resulting in a remarkably low melting point. One of their key properties is the low vapour pressure of less than 10^{-6} mbar at liquid state, which enables measurements of the bare substances in high-vacuum and therewith the application of positron beam studies. 1-Butyl-3-methyl-imidazolium-hexafluorophosphat is chosen for this study, that has well known thermal properties like glass-transition- and triple-point temperature.

The (C)DB-spectrometer and the high intense monoenergetic positron beam of the NEPOMUC beam facility located at the FRM II in Garching are used. The measurements demonstrate the diffusion effects of positrons in a liquid as well as phase transitions, induced by variation of sample temperature between 80K and 300K during the measurement.

CPP 7.2 Mon 18:15 ZEU 160

Cation Dynamics in Ionic Liquids as seen by QuasiElastic Neutron Scattering — ●JAN PETER EMBS^{1,2} and ROLF HEMPELMANN² — ¹Laboratory for Neutron Scattering, ETH Zurich & Paul Scherrer Institut, CH - 5232 Villigen-PSI — ²Physical Chemistry, Saarland University, D-66041 Saarbrücken

Quasi-elastic Neutron Scattering (QENS) is a powerful tool to investigate the dynamics of species containing protons on a picosecond (ps) time and Angstrom length scale. With this method one can investigate crystalline as well as liquid samples. We use QENS to probe the cation dynamics in pyridinium and imidazolium based ionic liquids containing anions that are not visible due to their negligible scattering cross sections. In this contribution we will present data obtained using different time-of-flight (FOCUS@SINQ, TOFTOF@FRM-II) as well as backscattering spectrometers MARS@SINQ, IN10@ILL). Different spectrometers are needed in order to obtain experimental results on different time and spatial scales and thus deepen the insight into the dynamical processes occurring in ionic liquids. Our results show that even in the liquid phase the corresponding cations perform localized motions (translational and rotational jump processes inside confining regions (cages) built up by surrounding ions) together with slower processes that can be attributed to rearrangements of the ions forming the confining cages. Furthermore using QENS we also can get insight into inelastic processes and thus completing the inelastic region that can be probed by far-infrared spectroscopy.

CPP 7.3 Mon 18:30 ZEU 160

Charge transport and glassy dynamics in ionic liquids — ●JOSHUA SANGORO, CIPRIAN IACOB, ANATOLI SERGHEI, SERGEJ NAUMOV, JÖRG KÄRGER, and FRIEDERICH KREMER — Institute of Experimental Physics I, University of Leipzig, Linnéstr. 5, 04103, Leipzig, Germany

Charge transport and glassy dynamics of a variety of glass-forming ionic liquids (ILs) are investigated in a wide frequency and temperature range by means of Broadband Dielectric Spectroscopy(BDS), Pulsed Field Gradient Nuclear Magnetic Resonance (PFG NMR), AC Calorimetry, Differential Scanning Calorimetry and Rheology. While the absolute values of dc conductivity and viscosity vary over more than 11 decades with temperature and upon systematic structural variation of the ILs, quantitative agreement is found between the characteristic frequency of charge transport and the structural α -relaxation. This is traced back to dynamic glass transition assisted hopping as the underlying mechanism of charge transport. A novel approach is applied to extract diffusion coefficients from BDS spectra in quantitative agreement with PFG NMR values but in a much broader range. References: [1] J. R. Sangoro, A. Serghei, S. Naumov, P. Galvosas, J. Kärger, C. Wespe, F. Bordusa, and F. Kremer, Phys. Rev. E 77, 051202 (2008);

J. R. Sangoro et al, J. Chem. Phys. 128, 214509 (2008); J.R. Sangoro, C. Iacob, A. Serghei, C. Friedrich, F. Kremer, Phys. Chem. Chem. Phys., In press.

CPP 7.4 Mon 18:45 ZEU 160

Tetrahydrofuran clathrate hydrate formation studied by non-resonant inelastic x-ray scattering — ●FELIX LEHMKÜHLER¹, CHRISTIAN STERNEMANN¹, HEIKO CONRAD¹, ARTO SAKKO², MIKKO HAKALA², KIM NYGÅRD³, DIETMAR PASCHEK⁴, OMID FEROUGH¹, THOMAS BUSLAPS⁵, LAURA SIMONELLI⁵, METIN TOLAN¹, and KEIJO HÄMÄLÄINEN² — ¹TU Dortmund, Fakultät Physik/DELTA, Dortmund, Germany — ²Division of Materials Physics, Department of Physics, FIN-00014 University of Helsinki, Finland — ³Paul Scherrer Institut, Villigen PSI, Switzerland — ⁴Rensselaer Polytechnic Institute, Troy, 12180 NY, USA — ⁵ESRF, 38043 Grenoble, France

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, especially H₂ in THF hydrate for fuel cells. In general, the hydrate formation on a microscopic level is still not understood. 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.

The bulk formation in the system water-THF was investigated by measuring x-ray Compton profiles and non-resonant x-ray Raman spectra of the hydrate, a mixture at temperatures above the region of hydrate stability, and the supercooled liquid mixture in order to detect hydrate precursors. The measurements were compared with DFT calculations employing local structures obtained from molecular dynamics simulation snapshots.

CPP 7.5 Mon 19:00 ZEU 160

Chelation in a water/triamine mixture: investigation of structure formation in space and time — ●ULRICH MÜLLER¹, MARTINE PHILIPP¹, JÖRG BALLER¹, ROLAND SANCTUARY¹, PIERRE SECK¹, MICHAEL VEITH², and JAN KRISTIAN KRÜGER¹ — ¹Université du Luxembourg, FSTC, Campus Limpertsberg, 162a avenue de la Faiencerie, L-1511 Luxembourg — ²Universität des Saarlandes, Anorganische Chemie, Fachbereich 8.11, Gebäude C 4.1, Postfach 151150, D-66041 Saarbrücken

Chelation is the binding or complexation of a bi- or multidentate ligand. Usually this term refers to complexes of metal ions and the chelating agent but can also refer to the complexation of water by polyamines. We will present first results of the chelation of water by diethylene triamine (DETA). DETA is tridentate ligand and exhibits a good miscibility with water. Our study comprises the investigation of the DETA concentration dependence of the mass density, of the refractive index and the longitudinal elastic modulus as well as time and space dependence of the chelation process at a water/DETA interface as measured by time resolving Brillouin microscopy. The results show that the longitudinal elastic modulus and the mass density possess a maximum over concentration whereas the refractive index does not. This strong deviation from the Lorenz-Lorentz behaviour indicates a significant influence of chelation and the accompanying change in structure on the electronic polarizability. Furthermore we are able to show a complex structure formation process at the water-DETA interface leading to an astonishingly asymmetric interphase.

CPP 7.6 Mon 19:15 ZEU 160

Molecular motions in liquid medium-chain *n*-alkanes — ●TOBIAS UNRUH¹, CHRISTOPH SMUDA¹, SEBASTIAN BUSCH¹, and GERD GEMMECKER² — ¹Technische Universität München, Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), D-85747 Garching — ²Technische Universität München, Bayerisches NMR-Zentrum, Chemie Department

Different diffusive motions in liquid medium-chain *n*-alkanes could be disentangled by resolution resolved quasielastic time-of-flight neutron spectroscopy (TOF-QENS). It was found that the dominating motion causes a Q^2 -dependence of the QENS signal for any observation time in the picosecond time range. For the longest times the Q^2 -dependence describes the long range center-of-mass diffusion of the alkane molecules as confirmed by PFG-NMR measurements. By com-

parison with molecular dynamics simulations, the identified motions for short times are attributed to displacements of hydrogen atoms reflecting local molecular trajectories. Despite the rod-like shape of the molecules the center-of-mass diffusion was found to be essentially

isotropic. A coherent picture of the diffusional process ranging from the fast tumbling of CH₂ groups to the slow long range molecular diffusion will be presented.