

DY 20: Glasses III (joint session DF/DY)

Time: Wednesday 14:30–16:15

Location: EB 407

DY 20.1 Wed 14:30 EB 407

Impedance- and IR-spectroscopy on sputtered borate glasses — ●GERD-HENDRIK GREIWE and GUIDO SCHMITZ — Institut für Materialphysik, WWU Münster, Wilhelm-Klemm-Str. 10, D-48149 Münster

Ion-conducting oxide glasses are considered as components of thin film batteries. In our study, glass films of the compositions $x \text{Li}_2\text{O} \cdot (1-x) \text{B}_2\text{O}_3$ with $x = 0.15, 0.20, 0.25, 0.30,$ and 0.35 are prepared by ion beam sputtering in a thickness range between 100 and 1000 nm. TEM cross section investigations show a homogeneous, amorphous structure of the films, while the correspondence of their chemical composition with the glass targets is proved by EELS analysis. The specific dc-conductivity of the glass films is determined by temperature-dependent impedance spectroscopy and found to be up to three orders of magnitude higher compared to the conductivity of the corresponding bulk glasses prepared from the melt. This conductivity increase is explained by a modification of the network structure of the thin glass films. The concentration of the Non-Bridging Oxygen atoms of the network is assumed to be increased by the sputter process. This increase is expected to be the main reason for the observed conductivity enhancement. IR-spectroscopy is used to determine the content of the Non-Bridging Oxygens atoms of the network, to correlate structural and electrical properties of the thin film glasses.

DY 20.2 Wed 14:45 EB 407

Decoupling of Atomic Diffusion in Glass-Forming Mixtures — ●THOMAS VOIGTMANN and JÜRGEN HORBACH — Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt (DLR), 51170 Köln

The diffusion in dense binary soft-sphere mixtures with disparate sizes is studied. While the system approaches a glass transition, only the transport of large particles ceases, while small particles are still able to diffuse through the almost frozen background formed by the large ones, leading to an order-of-magnitude decoupling in the respective transport coefficients. This mechanism qualitatively describes the phenomenology of ion-conducting melts such as sodium silicate mixtures. Upon further increasing the density, the small-particle mean-squared displacement shows an increasing regime of anomalous power-law-like diffusion, which is interpreted as the precursor of a second localization transition following the glass transition in this system.

DY 20.3 Wed 15:00 EB 407

In-situ study of dynamics in hydrous silicate melts with quasielastic neutron scattering — ●FAN YANG¹, ANDREAS MEYER², TOBIAS UNRUH³, and JOSEF KAPLONSKI¹ — ¹Physik-Department E13, TU München, 85748 Garching, Germany — ²Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt, 51147 Köln, Germany — ³Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), TU München, 85748 Garching, Germany

Dissolved water in silicate melts and glasses is of great interest in geoscience as well as in technical applications. We investigate water dynamics in silicate melts and the correlation between macroscopic properties like viscosity and the microscopic structure. With the intrinsic q resolution of quasielastic neutron scattering diffusion can be studied in great detail. Diffusion of water in sodium trisilicate melt was studied in-situ at different temperatures under a pressure of 200 MPa. The temperature dependence of the H diffusion coefficient obeys an Arrhenius law with an activation energy of about 0.36 eV.

DY 20.4 Wed 15:15 EB 407

Conductivity of Alkali Glasses in Quantum Statistics — ●JOACHIM SOHNS and MICHAEL SCHULZ — University of Ulm

Our aim is to find an analytically solvable model of the conductivity of mixed alkali glasses. Our starting point is the schrodinger equation of the multy particle wave function of the system. As a consequence of the structural and dynamical disorder the system is irreversible in time. The dynamics of the system is given by the matrix greens functions of the keldysh technique. The linear response of the system to an external electrical field may be calculated within the kubo formalism. In addition to the usual paramagnetic and diamagnetic current we find another contribution to the current which is caused by the finite life times of the states. The dependence of the conductivity on the con-

centration of alkali ions, on the temperature and on the frequency of the external electrical field are calculated. The mixed alkali effect is reproduced by our model.

DY 20.5 Wed 15:30 EB 407

Barium diffusion in mixed cation glasses — ●MICHAEL GROFMEIER, FRANK NATRUP, and HARTMUT BRACHT — Institute of Materials Physics, University of Münster, Germany

Diffusion of barium in mixed cation glasses of the composition $x\text{Na}_2\text{O} \cdot (3-x)\text{BaO} \cdot 4\text{SiO}_2$ with $x = 0.0, 0.1, 0.3$ and 1.0 and $0.4\text{K}_2\text{O} \cdot 2.6\text{BaO} \cdot 4\text{SiO}_2$ was investigated by means of the radiotracer diffusion technique below the respective glass transition temperatures. In accord with our previous results of calcium diffusion in soda-lime silicate glasses, the mobility of alkaline-earth ions increases with the alkali content in all analyzed glass systems with no decrease in the diffusion activation enthalpy, but a raise in the pre-exponential factor. A distinct dependency of the activation enthalpy of alkaline-earth ions on the type and content of the alkali ions in the glass is observed. The results provide evidence for elastic and electrostatic contributions to cation diffusion in glasses and support the formation of dissimilar cation pairs, that were derived from nuclear magnetic resonance investigations of soda-lime silicate glasses and glasses containing sodium and barium. Finally, a striking correlation between the pre-exponential factor of alkaline-earth ion diffusion in soda-lime and potassium barium glasses is found whose origin remains unsolved.

DY 20.6 Wed 15:45 EB 407

Ion Dynamics in Room Temperature Ionic Liquids — MONIKA MUTKE, ●RADHA DILIP BANHATTI, and KLAUS FUNKE — Institut für Physikalische Chemie und SFB 458, Universität Münster, Corrensstr. 30, D-48149 Münster

Room temperature ionic liquids (RTIL) are molten salts consisting of a bulky organic cation such as 1-butyl-3-methyl-imidazolium (BMIM) and anions such as BF_4^- and PF_6^- . Above T_G , RTILs exhibit non-Arrhenius type DC conductivities and are classified as of intermediate fragility. Earlier, the broadband conductivity spectra of a fragile ionic melt [1] and of a polymer electrolyte [2] were modelled providing a link between the short-time and the long-time dynamics of the ions, via the dispersive features of the spectra. We could thus obtain the activation energy of the elementary displacive step, E^* . In this contribution, we present and analyse the conductivity spectra of BMIMBF_4 from 1 MHz up to about 6 GHz in the temperature range 193 K - 353 K. We show that BMIMBF_4 exhibits spectral features similar to those of the polymer electrolyte, indicating the importance of structure mediated ion-ion interactions. Moreover, both for BMIMPF_6 and BMIMBF_4 , E^* is found to be about 0.18 eV, which is similar to the conformational reorientation energy of the cation [3].

[1] P. Singh, R.D. Banhatti and K. Funke, Phys. Chem. Glasses **46**, 241 (2005).

[2] S.J. Pas, R.D. Banhatti and K. Funke, Solid State Ionics **177**, 3135 (2006).

[3] A. Rivera and E. Rössler, Phys. Rev. B **73**, 212201 (2006).

DY 20.7 Wed 16:00 EB 407

Crystal precursor nucleation: A connection between crystallization and vitrification. — ●HANS JOACHIM SCHÖPE¹, GARY BRYANT², and WILLIAM VAN MEGEN² — ¹Johannes Gutenberg-Universität Mainz, Institut für Physik, Staudinger Weg 7, 55099 Mainz, Deutschland — ²Department of Applied Physics, Royal Melbourne Institute of Technology, GPO Box 2476V, Melbourne 3001, Australia

A complete understanding of the solidification process (crystallization, vitrification) is one of the long-standing problems in condensed matter physics. The use of colloidal model systems provides an ideal controlled experimental system to reduce this lack of knowledge. We investigated the solidification scenario in suspensions of colloidal hard spheres for three polydispersities between 4.8% and 5.8%, from near freezing to near the glass transition. We identify four stages in the crystallization process: (i) an induction stage where large numbers of precursor structures are observed; (ii) a conversion stage as precursors are converted to close packed structures; (iii) a second nucleation stage; and (iv) a ripening stage. Near the glass transition the crystallization process is

entirely frustrated, and the sample is locked into a compressed crystal precursor structure. Interestingly neither polydispersity nor volume fraction significantly influence the precursor stage, suggesting that the crystal precursors are present in all solidifying samples. We speculate

that these precursors are related to the dynamical heterogeneities observed in a number of dynamical studies linking the two processes of crystallization and vitrification. JCP 127, 084505 (2007)