CPP 30: Glasses and Glass Transition I (jointly with DY, DF)

Time: Wednesday 10:15–13:00 Location: KÖN Farb

CPP 30.1 Wed 10:15 KÖN Farb

Boson-peak in glasses and random-matrix statistics — •Walter Schirmacher — Institut für Physik, Univ. Mainz — Physik-Department E13, TU München

The enhancement of the vibrational density of states (DOS) with respect to the Debye expectation ("boson peak"), which is universally observed in glasses, is considered by means of of symmetry arguments. Low-frequency wave-like excitations in a disordered solids probe a homogeneous and isotropic material. Due to these symmetries the vibrational wave-like states are highly degenerate (Debye regime). This degeneracy is lifted at higher frequencies. In this regime the discretized equation of motions are governed by a sparse random matrix. The eigenvalues of this matrix are non-degenerate and show the distance statistics of the Gaussian orthogonal ensemble ("level repulsion"). It is conjectured that the integrated density of levels in this regime increases linearly. The cross-over from the Debye regime to the random-matrix regime leads in three dimensions to an enhancement of the DOS. In two dimensions this is not the case. Model calculations using a field-theoretical approach [1] and inspection of simulation data [2] confirm this reasoning.

[1] W. Schirmacher, Europhys. Lett. Europhysics Letters, 73, 892 (2006); [2] see e.g. S. K. Sarkar, G. S. Matharoo, A. Pandey, Phys. Rev. Lett. 92,215503 (2004)

CPP 30.2 Wed 10:30 KÖN Farb

Identification of facilitation effects in supercooled liquids — •Andreas Heuer and Christian Rehwald — Institute of Physical Chemistry, University of Muenster

In a first step we analyze the information content of the finite-size effects of a glass-forming system. Interestingly, the diffusion constant shows a very weak and the structural relaxation time a very strong finite-size effect [1]. This result reflects the dynamic coupling of different regions in real space via a facilitation mechanism. We can formulate a minimum model of the glass transition which can reproduce in detail the observed features of these finite-size effets. It can be regarded as a generalization of the kinetically constrained models [2].

In a second step we search for a direct evidence of this facilitation mechanism in computer simulations. Using the setup of a highly non-equilibrium configuration these causal relations between successive relaxation events can indeed be found and characterized.

[1] C. Rehwald, O. Rubner, A. Heuer, Phys. Rev. Lett. 105, 117801 (2010).

[2] Y. J. Jung, J. P. Garrahan, and D. Chandler, Phys. Rev. E 69, 061205 (2004).

CPP 30.3 Wed 10:45 KÖN Farb

Dynamics of shear transformation zones during mechanical cycling of glassy CuTi - a molecular dynamics study — •Lennart Fricke und S. G. Mayr — Leibniz-Institut fuer Oberflaechenmodifizierung, Translationszentrum fuer regenerative Medizin und Fakultaet fuer Physik und Geowissenschaften der Universitaet Leipzig, 04318 Leipzig

Plastic deformation of bulk metallic glasses at low temperatures occurs in highly localized regions, called shear-transformation-zones (STZ) [1] - as corroborated recently in detailed experimental and simulational studies. After activation, these STZs should possess a memory of their configuration prior to transformation due to confinement by the surrounding elastic matrix, i.e. the Eshelby back-stress. While this picture surely is intuitive, it is particularly interesting whether it applies in a strict or only statistical sense and up to what strain levels. With this background we study shear behavior during mechanical cycling of CuTi with a maximum of 1% to 10% shear strains in large–scale MD simulations using realistic embedded atom method (EAM) potentials. Evaluating suitable quantities, including non-affine displacements and atomic-level Basinski–Duesbery–Taylor (BDT) stresses, we address the reversibility of STZs and the underlying physics on the atomic scale.

[1] A. S. Argon, Acta Metall. 27, 47 1979

This project is funded by the German DFG - PAK 63

CPP 30.4 Wed 11:00 KÖN Farb

Describing experimentally obtained stress overshoots in sheared colloidal dispersions with schematic MCT — •Christian Peter Amann[1], Matthias Fuchs[1], Miriam Siebenbürger[2], and Matthias Ballauff[2] — [1]Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany — [2]Helmholtz Zentrum für Materialien und Energie, 14109 Berlin, Germany

Sheared viscoelastic media show a stress overshoot between elastic and plastic deformation regime, i.e. a maximum in the shear stress vs. strain plot after switching on a constant shear rate. We found a way to model such overshoots with the $F_{12}^{(\dot\gamma)}$ model, a schematic model of a microscopic mode-coupling theory (MCT) approach to describe glass forming liquids. The enhancement of the schematic model is tested by fitting experimental strain-stress curves from sheared colloidal dispersions of thermosensitive core-shell particles. Flow curves and linear stress response moduli of the same experimental setup could be fitted well with the $F_{12}^{(\dot\gamma)}$ model [1]. Furthermore this model has been able to describe nonlinear stress response to oscillatory shear rates [2]. The implementation of stress overshoots in this schematic model was motivated and guided by the recently identified mechanism within microscopic MCT framework causing such overshoots to occur [3]. This mechanism is also identified to be highly connected to a super-diffusive motion regime of the colloids [3].

[1] M. Siebenbürger et al., *J. Rheol.* **53**, 707–726 (2009)
[2] J.M. Brader et al., arXiv:1010.2587v1 (2010)
[3] J. Zausch et al., *J. Phys.: Condens. Matter* **20**, 404210 (2008)

CPP 30.5 Wed 11:15 KÖN Farb

Evaluation of MD force fields for ion transport in glassy materials — •Christian Trott[1], Martin Körner[1], Michael Schuch[2], and Philipp Maass[2] — [1]Theoretical Physics II, Technische Universität Ilmenau, 98684 Ilmenau, Germany. — [2]Fachbereich Physik, Universität Osnabrück, 49076 Osnabrück, Germany

Ion conducting glasses are an interesting class of materials with a wide range of possible applications, including batteries, supercapacitors, and smart windows. Considerable efforts have been undertaken to understand their properties with the help of experimental and theoretical investigations such as molecular dynamics (MD) simulations. Most of the previous MD studies of ion conducting glasses have been limited to structural analysis and the existing investigations of ion transport were almost all performed at comparatively high temperatures just below the "computer glass transition" temperature. Recent advances in hardware and MD software, namely the development of the GPU-MD code $\text{LAMMPS}_{\text{CUDA}}$, allow for a much more comprehensive investigation of long term dynamics. We assess the power of a number of interaction models for investigating long range ion transport in glasses. Specifically we determine diffusion constants and selected activation energies in several ion conducting glass systems for which force fields have been suggested in the literature.

CPP 30.6 Wed 11:30 KÖN Farb

Lorentz-like power-law decay of velocity anti-correlations in a supercooled liquid — •Felix Höfling[1] and Peter Colberg[2,3] — [1]Max-Planck-Institut für Metallforschung, Stuttgart, and Institut für Theoretische und Angewandte Physik, Universität Stuttgart — [2]Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Canada — [3]Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt (DLR), Köln

Harnessing the compute power of recent graphics processors, we have measured the velocity-autocorrelation function (VACF) in the supercooled Kob-Andersen mixture for large systems of 50,000 particles, providing data with an excellent signal-to-noise ratio. The emergence of glassy dynamics upon supercooling is systematically accompanied by a power-law decay of the VACF at intermediate times with exponent 5/2 and negative prefactor, similarly as observed recently for the hard sphere liquid (Williams *et al.*, PRL 2006). Such anti-correlations are reminiscent of the well-known, universal long-time tail in the Lorentz model with the same exponent and sign. The role of dynamic heterogeneities for the power-law decay is addressed by considering correlation functions that are restricted to the most mobile or immobile particles. We find that the Lorentz-like decay is absent in the VACF of the most mobile particles and conclude that the power law is *not* a manifestation of dynamic heterogeneities. For the most immobile particles, however, the power-law decay is well pronounced and we propose that the relevant mechanism is given by repeated encounters

with the quasi-arrested, microscopic particle cages.

CPP 30.7 Wed 11:45 KÖN Farb

Universal jamming phase diagram in the hard-sphere limit and comparison of the dynamics of soft and hard spheres — •Michael Schmiedeberg[1], Thomas K. Haxton[2], Sidney R. Nagel[3], and Andrea J. Liu[4] — [1]Institut für Theoretische Physik 2: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany — [2]Theory of Nanostructured Materials, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA — [3]The James Franck Institute, The University of Chicago, Chicago, IL 60637, USA — [4]Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, PA 19104, USA

We present a new formulation of the jamming phase diagram for a class of glass-forming fuids consisting of spheres interacting via finite-ranged repulsions. Our phase diagram is universal at low pressure, i.e. observables such as the relaxation time are insensitive to details of the interaction potential and collapse onto the values for hard spheres.

Furthermore, we show that the dynamics of soft spheres can be described in terms of the dynamics of hard spheres. By introducing an effective hard sphere diameter that is determined from the soft-sphere pair potential via the Andersen-Weeks-Chandler approximation, the relaxation times of soft spheres can be mapped onto the curve known for hard-sphere liquids. These results indicate that the dynamics of soft spheres depend on an effective free volume in a universal way.

CPP 30.8 Wed 12:00 KÖN Farb

Investigation of the dephasing of tunneling systems in glasses using two-pulse polarisation echo experiments — •Masoomeh Bazrafshan, Paul Fassl, Martin Schwarze, Angela Halfar, Annina Luck, Andreas Fleischmann, and Christian Enss — Kirchhoff Institut für Physik, Universität Heidelberg

Low temperature properties of glasses are governed by atomic tunneling systems. Many aspects are well described within the phenomenological standard tunneling model. Tunneling systems couple to their local strain fields which gives rise to a strain-mediated coupling among them. These interactions cause time-dependent variations in the energy splittings of tunneling systems when their neighbors undergo thermal transitions. This is the basis of the spectral diffusion model, which describes the dephasing of tunneling systems at very low temperatures. Experimentally, this dephasing of tunneling systems can be studied by two-pulse polarisation echo experiments. We have performed such echo decay measurements with an improved setup allowing us to observe echoes at very long delay times where the echo has decayed five orders of magnitude from its original amplitude. We have analysed the time and temperature dependent results in the framework of spectral diffusion model, finding very good qualitative agreement for the echo decay, but clear shortcoming in terms of the temperature dependence.

CPP 30.9 Wed 12:15 KÖN Farb

Structural investigations on Eu-doped fluorobromozirconate glass ceramics. — •Marie-Christin Wiegand[1], Bernd Ahrens[2], Bastian Henke[2,3], and Stefan Schweizer[2,3] — [1]Department of Physics, University of Paderborn, Warburger Str. 100, 33100 Paderborn, Germany — [2]Fraunhofer Center for Silicon Photovoltaics, Walter-Hülse-Str. 1, 06120 Halle (Saale), Germany — [3]Centre for Innovation Competence SiLi-nano®, Martin Luther University of Halle-Wittenberg, Karl-Freiherr-von-Fritsch-Str. 3, 06120 Halle (Saale), Germany

Modified Eu-doped fluorozirconate glasses are regarded as promising materials for photovoltaic and medical applications. In these glasses, a substantial fraction of the fluorine ions was replaced by bromine ions resulting in the formation of $BaBr_2$ nanocrystals upon subsequent thermal treatment of the as-made glass. Interestingly, the metastable hexagonal phase of $BaBr_2$ is always formed first before further annealing leads to the formation of orthorhombic phase $BaBr_2$, i.e., a phase transition from hexagonal to orthorhombic phase $BaBr_2$ occurs upon annealing. During the annealing a part of the doped Eu^{2+} is incorporated into the $BaBr_2$ nanocrystals enabling fluorescent transitions of Eu^{2+} in hexagonal and orthorhombic $BaBr_2$, respectively, upon ultraviolet excitation. The nanocrystal size and the structural phase depend on the addition of InF_3 and YF_3 and on the Br/(F+Br)-ratio, which was investigated by differential scanning calorimetry and x-ray diffraction. In addition, photoluminescence experiments were performed to monitor the phase transition by optical means.

CPP 30.10 Wed 12:30 KÖN Farb

Optical and structural properties of fluorozirconate-based glass ceramics doped with divalent and trivalent europium. — •Christian Passlick[1], Bastian Henke[1,2], Jacqueline Anne Johnson[3], and Stefan Schweizer[1,2] — [1]Centre for Innovation Competence SiLi-nano®, Martin Luther University of Halle-Wittenberg, Karl-Freiherr-von-Fritsch-Str. 3, 06120 Halle (Saale) — [2]Fraunhofer Center for Silicon Photovoltaics, Walter-Hülse-Str. 1, 06120 Halle (Saale) — [3]Department of Materials Science and Engineering, University of Tennessee Space Institute, Tullahoma, TN 37388, USA

Eu-doped fluorozirconate-based glass ceramics can be used for x-ray detection in medical diagnostics as well as for down-converting top layers in photovoltaics. A modified ZBLAN composition consisting of a mixture of Zr, Ba, La, Al, and Na fluorides was additionally doped with chlorine ions to initiate the formation of $BaCl_2$ nanocrystals upon thermal treatment of the as-poured glasses. During annealing some of the Eu^{2+} ions are incorporated into the nanocrystals enabling a strong blue fluorescence upon ultraviolet excitation or x-ray irradiation. In this work, focus is put on the amount of divalent and trivalent Eu fluoride and chloride additives since it is known that the expensive Eu^{2+} can be produced by melting the cheaper Eu^{3+} raw material. Influences of the different Eu oxidation states on the $BaCl_2$ crystallization and the optical response of the glass ceramics are presented.

CPP 30.11 Wed 12:45 KÖN Farb

Multi-phonon relaxation in Eu-doped fluorozirconate-based glasses and glass ceramics — •Charlotte Pfau[1], Christian Bohley[1], Manuela Miclea[1], Paul-Tiberiu Miclea[2,3], and Stefan Schweizer[2,1] — [1]Centre for Innovation Competence SiLi-nano®, Martin Luther University of Halle-Wittenberg, Karl-Freiherr-von-Fritsch-Str. 3, 06120 Halle (Saale) — [2]Fraunhofer Center for Silicon Photovoltaics, Walter-Hülse-Str. 1, 06120 Halle — [3]Institute of Physics, Martin-Luther-University of Halle-Wittenberg, Heinrich-Damerow-Str. 4, 06120 Halle

Eu-doped fluorozirconate(FZ)-based glasses are of interest for various fluorescence applications such as photon down-conversion layers for high efficiency solar cells or ionizing radiation imaging plates. Multi-phonon relaxation (MPR) is one of the major quenching processes of the rare-earth (RE)-related fluorescence therein. The MPR is significantly reduced in hosts providing low phonon frequencies such as FZ-based glasses and glass ceramics; the latter contain barium halide nanocrystals with even lower phonon frequencies. However, the MPR rate depends not only on the phonon frequency, but also on the electron-phonon coupling between the rare-earth ion and the host lattice. The local vibrational environment of the RE ion is investigated by phonon sideband spectroscopy. To analyze the vibrational spectra and their influence on the fluorescence properties, a series of Eu-doped FZ-based glasses and glass ceramics has been studied by Raman, phonon sideband, and fluorescence spectroscopy. The MPR rate is determined for the levels involved in the fluorescence process.