CPP 14: Poster: Glasses I (jointly with DY)

Time: Tuesday 18:15–20:15 Location: Poster A

CPP 14.1 Tue 18:15 Poster A

Elastic Properties of Amorphous Solids — •Christian L. Klix, Florian Ebert, Georg Maret, and Peter Keim — University of Konstanz

In this contribution, we present experimental results on the elastic properties of a two-dimensional colloidal glass former. Given that glasses are solids, one expects a mechanical behavior similar to that of crystals, i.e., glasses exhibit a finite shear modulus μ. Using positional data from video microscopy, we study the displacement field and connect it to the dynamical matrix $D(q)$ via the equipartition theorem [1]. The resulting dispersion relation of the system hints at structural change upon decreasing the temperature in the glassy state. Next, this data is used to derive the Lamé coefficients and the corresponding moduli from thermally excited modes in the long wavelength limit [1] using continuum elasticity theory. We consider finite size and time effects and find the expected frequency dependence of the shear modulus μ. By cooling the system, the significant increase of μ allows us to determine the glass transition temperature T_g precisely. Following the method described in [2], we compute the short wavelength excitations in our system and analyse the density of states as well as the structure of normal modes in a two-dimensional colloidal system.

[1] H. H. Von Grünberg, P. Keim, and G. Maret, Phys. Rev. Lett. 93, 255703 (2004)

[2] A. Gosh, V. K. Chikkadi, P. Schall, J. Kurchan, and D. Bonn, Phys. Rev. Lett. 104, 248305 (2010)

CPP 14.2 Tue 18:15 Poster A

Segmental and chain dynamics of polyisoprene confined in thin layers — •Emmanuel Urandu Mapesa, Martin Tress, and Friedrich Kremer — Institute for Experimental Physics I, University of Leipzig, 04103 Leipzig, Germany

Broadband Dielectric Spectroscopy (BDS) - in combination with a nanostructured electrode arrangement - is used to study thin layers of cis-1,4-polyisoprene (cis-PI). cis-PI belongs to a special type of polymers because a part of its molecular dipole moment attached to each monomer unit is aligned along the main chain. This fraction adds up to one dipole moment of the whole polymer chain corresponding to the end-to-end vector of the molecule. Consequently, the investigation of two distinct relaxation modes taking place at two different length scales is possible: the segmental motion which involves structures of about one nanometer in size, and the normal mode which represents the dynamics of the whole macromolecule. In the current study, where highly insulating silica nanostructures are used as spacers, the need to evaporate a counter-electrode is circumvented. The spin-cast samples are checked before and after dielectric measurement so as to exclude the possibility of dewetting affecting the measured dynamics. Down to 7 nm, for three molecular weights (44.5, 53 and 75 kg/mol), it is observed that: (i) the segmental mode (and hence the dynamic glass transition) is unaffected by 1-D confinement, and (ii) the normal mode becomes faster with decreasing layer thickness in dependence on molecular weight.

CPP 14.3 Tue 18:15 Poster A

Holographic Studies of New Azobenzene-Containing Low-Molecular-Weight Phase-Change Materials — •Christoph Meichner[1], Lothar Kador[1], Christian Probst[2], and Hans-Werner Schmidt[2] — [1]University of Bayreuth, Institute of Physics and Bayreuther Institut für Makromolekülforschung (BIMF), 95440 Bayreuth, Germany — [2]University of Bayreuth, Macromolecular Chemistry I and Bayreuther Institut für Makromolekülforschung (BIMF), 95440 Bayreuth, Germany

Using a plane-wave holographic setup, we investigated new azobenzene-containing low-molecular-weight materials which reveal a light-induced phase change from glassy to liquid-crystalline. Different parameters such as refractive-index modulation, long-term stability, writing speed, and sensitivity have been measured for different mixing ratios between the photo-active material and a similar but inactive compound. The experiments demonstrated that the inactive compound showed partial reorientation effects, which were driven by the *trans-cis-trans* isomerization cycles of the azobenzene moieties. In addition, an improved setup for the parallel inscription and reconstruction of two-dimensional data pages was built to test the feasibility of fast and high-density data storage. It is based on an LCD spatial light modulator and high-quality camera objectives.

CPP 14.4 Tue 18:15 Poster A

Heat capacity spectroscopy of the dynamic glass transition in chosen room temperature ionic liquids — •Evgeni Shoifet[1], Heiko Huth[1], Sergey Verevkin[2], and Christoph Schick[1] — [1]University of Rostock, Institute of Physics, 18051 Rostock, Germany — [2]University of Rostock, Institute of Chemistry, 18051 Rostock, Germany

Many of the ionic liquids are good glass formers. Nevertheless, only a few studies of the glass transition in ionic liquids are available so far. Particularly the frequency dependence of the dynamic glass transition (alpha-relaxation) is not known for most ionic liquids. The standard technique for such studies - dielectric spectroscopy - is not easily applicable to ionic liquids because of the high electrical conductivity. The aim of this study is to use different calorimetric techniques to obtain complex heat capacity and to investigate the dynamic glass transition of room temperature ionic liquids (RTILs) in a wide frequency range. This can give an insight in cooperative motions of ions and ion clusters in RTILs. The techniques that were used are DSC, TMDSC, and AC-chip calorimeter covering a total range of 0.001-10000 Hz. The trace of the dynamic glass transition in the activation diagram for RTILs was fitted to the Vogel-Fulcher-Tammann-Hesse function and the parameters compared for different RTILs.

CPP 14.5 Tue 18:15 Poster A

Differential AC chip calorimeter for in situ investigation of vapor-deposited thin films — •Mathias Ahrenberg[1], Evgeni Shoifet[1], Katie Whitaker[2], Heiko Huth[1], Mark Ediger[2], and Christoph Schick[1] — [1]Universtät Rostock, Institut für Physik, Universitätsplatz 3, 18055 Rostock — [2]Department of Chemistry, University of Wisconsin - Madison, Madison, Wisconsin 53706, USA

Physical vapor deposition (PVD) can be used to produce thin films with interesting material properties including extraordinarily stable organic glasses. We describe an AC chip calorimeter for in-situ heat capacity measurements of as-deposited nanometer thin films of organic glass formers. The calorimetric system is based on a differential AC chip calorimeter which is placed in the vacuum chamber for physical vapor deposition. The sample is directly deposited onto one calorimetric chip sensor while the other sensor is protected against deposition. The device and the temperature calibration procedure are described. The latter makes use of the phase transitions of cyclopentane and the frequency dependence of the dynamic glass transition of toluene and ethylbenzene. Sample thickness determination is based on a finite element modeling (FEM) of the sensor sample arrangement. In the modeling, a layer of toluene was added to the sample sensor and its thickness was varied in an iterative way until the model fit the experimental data.

CPP 14.6 Tue 18:15 Poster A

High-frequency laser modulated AC-chip calorimetry investigations of glass transition of polymers — •Yeong Zen Chua, Evgeni Shoifet, Gunnar Schulz, and Christoph Schick — Universität Rostock, Institut für Physik, Wismarsche Straße 43-45, 18051 Rostock, Germany

Combination of different techniques for heat capacity spectroscopy and temperature-modulated calorimetry allows the determination of complex heat capacity in a wide frequency range. We study the dynamic glass transition and its corresponding temperature-dependent cooperativity of polymers. An AC-chip calorimeter gives the possibility to decrease the size of the measured sample, hence increases the sensitivity and frequency range. However, the investigation of the dynamic glass transition is still limited to the frequencies up to a few kHz, due to capacitive coupling between heater driving voltage and thermopile voltage at high frequencies. A new AC-chip calorimeter with laser heating, which decouple the temperature measurement and heating, is capable of measuring the glass transition temperature of thin film samples in nanogram range and in the frequency range up to 500 kHz. The dynamic glass transition of polystyrene and poly(methyl methacrylate) was studied from 10^{-5} Hz to 500 kHz.

CPP 14.7 Tue 18:15 Poster A

Molecular dynamics of glass forming liquids confined in two dimensional constraint of uni-directional nanopores — •Wycliffe Kipnusu, Ciprian Iacob, Joshua Sangoro, and Friedrich Kremer — Institute of Experimental Physics I, University of Leipzig, Linnestr. 5, 04103 Leipzig, Germany.

Broadband Dielectric Spectroscopy (BDS), Pulse Field Gradient-Nuclear Magnetic Resonance (PFG-NMR) spectroscopy, and Fourier Transform Infrared (FT-IR) spectroscopy are combined to study the molecular dynamics of low molecular weight glass forming liquids (e.g. alkylcitrates) in the bulk state and confined in unidirectional nanopores (diameter: 4, 8, and 10.4 nm, length: 50 microns) prepared by electrochemical etching of highly doped p-type $< 100 >$ silicon and subsequent oxidation. The dynamics of the structural (alpha)relaxation and the diffusion are enhenced by more than 2 orders of magnitude in 4 nm pores at lower temperatures in comparison to the bulk. This is attributed to changes in molecular packing due to geometrical constraint. Apart from a beta- relaxation, a broad process (slower than the alpha- process) assigned to relaxation of interfacial layers is observed for the confined molecules. Silanization of the pores removes this process but does not alter the structural alpha- relaxation.

References:

(1). M. Arndt, etal. Physical review. E, 1996, 54, 5377. (2). F. Kremer Broadband Dielectric Spectroscopy, Springer, Berlin, 2003. (3). R. Prisk, and P. E. Sokol, J. Chem. Phys., 2011, 134, 114506

CPP 14.8 Tue 18:15 Poster A

Shear moduli of two dimensional binary glasses — •Rabea Seyboldt[1], David Hajnal[2], Fabian Weysser[1], and Matthias Fuchs[1] — [1]FB Physik, Universität Konstanz, 78457 Konstanz, Germany — [2]Institut für Physik, Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany

We studied the shear moduli of two-component glasses in two dimensions within mode coupling theory. Varying the concentration, strong mixing effects can be observed along the glass transition lines for two interaction potentials. Hard disks with size ratios between 0.3 and 0.9, and point particles interacting with (magnetic) dipoles of strength ratio between 0.1 and 0.6 are considered. Equilibrium structure factors (partially obtained from Monte Carlo simulations) and glass form factors show that a softening of the elastic shear constant of glass upon adding another component arises from a dilution effect of the majority component. For very disparate mixtures, an anomalous elastic strengthening results from what we interpret as clustering of the smaller particles in the voids between the larger ones. This among other things might indicate a close-by glass-glass transition. Simulation data on hard disk mixtures show that the theory underestimates the moduli by around 50%, but otherwise captures the qualitative trends (within the rather large simulational error bars).

CPP 14.9 Tue 18:15 Poster A

Excess heat capacity and fictive temperature of polystyrene in a wide range of cooling and heating rates — •Gunnar Schulz[1], Timur V. Tropin[2], Jürn W. Schmelzer[1], and Christoph Schick[1] — [1]University of Rostock, Institut of Physics, 18055 Rostock, Germany — [2]Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, 141980 Dubna, Russia

The physical characteristics of polystyrene allow us straightforward and reproducible measurements of heat capacity, C_p, glass transition temperature, T_g, and other properties. The possibility to reuse one sample for numerous cooling and heating cycles permits the investigation of the influence of the cooling and heating rates on C_p and T_g.

In our research, we conduct the cooling phases with various (constant) cooling rates, but the respectively following heating phases with only one heating rate. The comparison of the heat capacities observed after different cooling rates results in an excess C_p, which we also calculate by means of an appropriate model.

Our measurements furthermore yield the fictive temperature in the cooling rate range from 10^{-4} Ks^{-1} to 10^{4} Ks^{-1}. We compare these results with the dynamic T_g observed by means of temperature-modulated differential scanning calorimetry (TMDSC). The dependence of the fictive T_g on the cooling rate and the dependence of T_g on the modulation frequency turn out to be closely related.

CPP 14.10 Tue 18:15 Poster A

Dielectric relaxation of alcohols in silica nanochannels — •Daniel Rau, Carsten Biehl, Rene Berwanger, and Rolf Pelster — Experimentalphysik, Universität des Saarlandes

We studied the molecular dynamics and glass transition of n-butanol and 2-ethyl-1-hexanol confined in silica nanochannels using broadband dielectric spectroscopy. For this purpose we have varied the pore radii in the range from 3 to 7 nm[1]. In contrast to the bulk material, confined n-butanol exhibits a glass transition upon slow cooling. Interestingly, in larger pores partial recristallization occurs upon heating. This behaviour reflects the different temperature dependence of nucleation and crystal growth.

[1] Berwanger et al., Eur. Phys. J. Special Topics 189, 239-249 (2010)

CPP 14.11 Tue 18:15 Poster A

Probing a statical, critical length scale in supercooled liquids — •Ingo Füreder, Patrick Ilg, and Hans Christian Öttinger — ETH Zürich, Switzerland

Nonequilibrium molecular simulations of glassy systems pose several challenges due to their long relaxation times and the large time scale gap between micro- and macroscopic dynamics. Guided by general principles of nonequilibrium thermodynamics we develop a constitutive model which should enable us to describe the behaviour of supercooled liquids in a quantitative manner on a macroscopic scale. The building blocks of the model contain a naturally arising length scale in glassy systems being a signature of growing amorphous order near the glass transition. This length is associated with the hindrance of affine deformations of an inherent structure configuration and could be extracted by investigating the system's non-affine response to static shear.

CPP 14.12 Tue 18:15 Poster A

Molecular signature of the dynamic glass transition as probed by spectroscopies sensing inter- and intra-molecular interactions — •Wilhelm Kossack[1], Periklis Papadopoulos[2], and Friedrich Kremer[1] — [1]Fakultät für Physik und Geowissenschaften, Linnéstraße 5, 04103 Leipzig, Germany — [2]Max-Planck-Institut für Polymerforschung, Ackermannweg 10, 55128 Mainz, Germany

The dynamic glass transition reflects a continuous slowing down of relaxations between structural substates of a glass forming system. Conventially its temperature dependence is described by the empirical Vogel-Fulcher-Tammann dependence. In the present study Fourier Transform InfraRed (FTIR) spectroscopy is employed to study the inter-molecular interactions between different molecular moieties of glass forming systems by measuring the temperature dependence of the spectral positions of its characteristic vibrations. In combination with Broadband Dielectric Spectroscopy (BDS) and DC- and AC- calorimetry this delivers a novel detailed insight into the molecular signature of the dynamic glass transition being specific for each glass forming system.

CPP 14.13 Tue 18:15 Poster A

Space-time phase transition in an atomistic model of glass formers — •Thomas Speck — Institut für Theoretische Physik II, Heinrich-Heine-Universität Düsseldorf, Germany

The transformation of a liquid into a solid crystal is a first-order phase transition that occurs through nucleation. All liquids can, therefore, be supercooled to a certain degree; and some turn into highly viscous, amorphous solids. One of the striking features is the emergence of dynamic heterogeneity, i.e., while overall particle motion becomes more and more restricted small regions of activity persist. Here we study ensembles of trajectories for a popular atomistic glass former, the Kob-Andersen binary Lennard-Jones mixture. We characterize mobility in a single trajectory through the number of particles that undergo a long-lived displacement. These excited particles record the underlying excitations that lead to dynamic heterogeneity. Using umbrella sampling we determine the probability distribution of this order parameter and show that it is non-concave. This can be interpreted as a phase transition in space-time between an active, liquid-like phase and an inactive, jammed phase in which the system remembers its initial conditions over the observation time. Structural and dynamical properties of these two distinct dynamic phases are analyzed and consequences for the glass transition are discussed.

CPP 14.14 Tue 18:15 Poster A

Differential AC chip calorimeter for in situ investigation of vapor-deposited thin films — •Mathias Ahrenberg[1], Evgeni Shoifet[1], Katie Whitaker[2], Heiko Huth[1], Mark Ediger[2], and Christoph Schick[1] — [1]Universtät Rostock, Institut für Physik, Universitätsplatz 3, 18055 Rostock — [2]Department of Chemistry, University of Wisconsin - Madison, Madison, Wisconsin 53706, USA

Physical vapor deposition (PVD) can be used to produce thin films with interesting material properties including extraordinarily stable organic glasses. We describe an AC chip calorimeter for in-situ heat capacity measurements of as-deposited nanometer thin films of organic glass formers. The calorimetric system is based on a differential AC chip calorimeter which is placed in the vacuum chamber for physical vapor deposition. The sample is directly deposited onto one calorimetric chip sensor while the other sensor is protected against deposition. The device and the temperature calibration procedure are described. The latter makes use of the phase transitions of cyclopentane and the frequency dependence of the dynamic glass transition of toluene and ethylbenzene. Sample thickness determination is based on a finite element modeling (FEM) of the sensor sample arrangement. In the modeling, a layer of toluene was added to the sample sensor and its thickness was varied in an iterative way until the model fit the experimental data.

CPP 14.15 Tue 18:15 Poster A

Phase diagram of confined hard sphere glasses — •Suvendu Mandal[1], Dierk Raabe[1], and Fathollah Varnik[1,2] — [1]Max-Planck Institut fur Eisenforschung, Max-Planck Str. 1, 40237 Dusseldorf, Germany — [2]Interdisciplinary Centre for Advanced Materials Simulation (ICAMS), Ruhr-Universitat Bochum, Stiepeler Strasse 129, 44801 Bochum, Germany

Spatial confinement is known to induce a dramatic change in the viscosity and relaxation time of liquids near the glass transition point (see, e.g., the topical review in [1] and references therein). While this issue has been the subject of a large number of studies for polymers and molecular glasses, only few investigations focus on colloidal hard sphere systems [2]. Quite recently, computer simulation studies of a monodisperse hard sphere (HS) system revealed an oscillatory behavior of the diffusion coefficient as the wall-to-wall distance, H, was varied between 2 and 4 particle diameters [3]. Suppressing crystallization via polydispersity, we extend these studies to considerably higher volume fractions, thereby addressing, for the first time in simulations, the effect of confinement on the mode coupling critical volume fraction. Our results are in good agreement with recent theoretical predictions [4]. A thorough test of other aspects of theoretical predictions is also provided [5].

[1] J. Baschnagel, F. Varnik, J. Phys.: Condens. Matter 17, R851-R953 (2005). [2] C. R. Nugent et al. PRL 99, 025702 (2007). [3] J. Mittal et al. PRL 100, 145901 (2008). [4] S. Lang et al. PRL 105, 125701 (2010). [5] S. Mandal et al. in preparation.

CPP 14.16 Tue 18:15 Poster A

Aging and elementary relaxation processes in amorphous solids — •Nima Hamidi Siboni[1,3], Fathollah Varnik[2], and Dierk Raabe[3] — [1]Aachen Institute for Computational Engineering Science, Aachen, Germany — [2]The Interdisciplinary Centre for Advanced Materials Simulation, Bochum, Germany — [3]Max-Planck Institut für Eisenforschung, Düsseldorf, Germany

Aging affects both dynamical [1] and mechanical (e.g., yield stress [2]) properties of amorphous solids. We study here aging effects on the distribution of elementary relaxation processes for the case of a binary Lennard-Jones glass. Both the distribution of first relaxation events, $\xi(t_1)$, and the waiting time between two successive events, $\phi(\tau)$ is modified upon aging. This is at first sight not surprising since these two distributions are known to satisfy the relation $\xi(t_1) = \int_{t_1}^{\infty} \phi(t')t'dt'/\langle\tau\rangle_\phi$, where $\langle\tau\rangle_\phi = \int_0^{\infty} \phi(t')t'dt'$ is the mean waiting time [3]. Recent simulation studies of this issue, however, report that only $\xi(t_1)$ changes upon aging, while $\phi(\tau)$ seems to remain stationary [4]. This unexpected behavior has been attributed to a possible power-law tail of $\phi(\tau)$ and the resulting divergence of $\langle\tau\rangle$. We thoroughly study this issue and propose possible explanations of this apparent discrepancy [5].

[1] W. Kob, J.-L. Barrat, Phys. Rev. Lett. 78, 4581 (1997).

[2] F. Varnik, L. Bocquet, J.-L. Barrat, J. Chem. Phys. 120, 2788 (2004).

[3] O. Rubner, A. Heuer 78, 011504 (2008).

[4] M. Warren, J. Rottler 88, 58005 (2009).

[5] N. Hamidi Siboni, D. Raabe, F. Varnik (in preparation).

CPP 14.17 Tue 18:15 Poster A

THz dynamics in glasses studied by means of Inelastic Neutron Scattering — •Nicolo' Violini — Jülich Center of Neutron Scattering, Forschungszentrum Jülich, D-52425 Jülich, Germany — Dipartimento di Fisica, Universitá di Perugia, Via A. Pascoli, I-06123, Perugia, Italy

The THz dynamics of two different glasses has been studied by means of Inelastic Neutron Scattering: the germanium diselenide (v-GeSe2) [1], an amorphous glass belonging to the family of continuous random network forming glasses and the glucose (C6H12O6). In both cases two well-defined peaks are resolved in the dynamic structure factor in a broad Q region, below and above the position of the first sharp diffraction peak in the static structure factor. Their common characteristics and possible differences will be discussed during the talk. The presence of two pseudo-branches in glasses seems to be quite general, but their nature is not completely understood, mainly due to the lack of theoretical models able to provide a complete description of this intriguing features[3].

[1] L. Orsingher, G. Baldi, A. Fontana, L. E. Bove, T. Unruh, A. Orecchini, C. Petrillo, N. Violini and F. Sacchetti Phys. Rev. B 82, 115201 (2010).

[2] N. Violini and F. Sacchetti, paper in preparation (unpublished).

[3] N. Violini, PhD Thesis (2011).