CPP 17: Glasses II (joint session DY/DF/CPP)

Time: Tuesday 9:30–12:30 Location: H46

CPP 17.1 Tue 9:30 H46

Towards reliable structural information of multicomponent glass systems — •Christoph Scherer[1,2], Friederike Schmid[1], and Martin Letz[2] — [1]Johannes Gutenberg-Universität, Mainz, Deutschland — [2]Schott AG, Mainz, Deutschland

Glasses have a huge range of applications, however, they are still theoretically not well understood. Also experimental access to the structure of glasses is limited. This motivates the study of glass systems by means of computer simulations.

In this work, a set of glass structures is generated on the computer by equilibrating a system of a few hundred atoms at high temperature, well above the glass transition temperature, with a classical molecular dynamics simulation (MD). Afterwards the system is cooled down to 0 K and structurally relaxed to the next (local) minimum by means of a quantum mechanical density functional (DFT) calculation. The glass properties before and after the structural relaxation are compared to experimental results. Especially, the phonon density of states is of interest, as it provides access to thermodynamical quantities.

This sets the basis for the next steps: The force fields for the MD simulation are generated by means of a structural fitting procedure. Here, the force field parameters are fitted in a way that the structure, namely the radial distribution function, of a short MD run at high temperature matches as closely as possible that one of a short DFT run at the same temperature. The dependence of the fitting accuracy of the classical force field on the final glass structure and glass properties is examined.

CPP 17.2 Tue 9:45 H46

On the behavior of supercooled liquid water in Confinements formed by frozen water molecules: a molecular dynamics simulation study — •Felix Klameth and Michael Vogel — Institut für Festkörperphysik, TU Darmstadt, 64289 Darmstadt

Molecular dynamics simulations are performed to study the influence of an amorphous ice confinement on liquid water. Investigating water in confinement is believed to reveal information unaccessible for bulk water due to crystallization. Therefore, there are numerous studies on confined water, which claim, e.g., existence of a second critical point associated with a liquid-liquid phase transition in the supercooled regime [1]. However, transfer of information from confined water to bulk water is not straightforward because introducing walls changes the static properties of water due to specific interactions at the interfaces. To avoid this drawback, we use a neutral confinement comprised of immobilized water molecules. We compare static and dynamical properties found in pores with different radii to that of bulk water. The static characteristics, like the tetragonal order parameter, are not changed even near the pore wall. On the contrary, the dynamics inside the pore are dramatically influenced. We find a tremendous increase of the structural relaxation time of liquid water when approaching the pore wall. Thus, we observe a strong change of the local dynamics, which is neither accompanied by a variation of the local structure nor caused by specific wall-liquid interactions. Possible origins for this effect are discussed. [1] P. Kumar et al, PRL (2006), 97, 177802

CPP 17.3 Tue 10:00 H46

Deuteron-NMR investigation on the dynamics of supercooled, confined water — •Matthias Sattig and Michael Vogel — TU Darmstadt, Institut für Festkörperphysik

The dynamical behaviour of water in the regime of the supercooled liquid is a topic of large interest. In particular, the existence of a fragile-to-strong transition (FST) at T=225K related to the transition between two distinct phases of liquid water is controversially discussed [1]. Due to crystallization the temperature range proposed for the FST is hardly accessible in bulk water. Therefore, we confine heavy water to narrow pores in the mesoporous sillicate MCM-41. This suppresses the freezing of a substantial fraction of water, enabling direct investigation of the interesting temperatures. Deuteron-NMR methods are utilised to determain the rotational correlation times τ of water on time scales from ns up to s. The spin-lattice-relaxation time T_1 exhibits a typical minimum at about T=230K. Above this minimum the correlation times follow a Vogel-Fulcher-Tammann law. Below the minimum, two relaxation processes could be observed. The low-temperature processes show a different temperature dependence, where the curves τ(T) of all processes intersect at about T=230K. A comparison with literature data [2] from neutron scattering and dielectric spectroscopy gives rise to the idea that the observed crossover is due to this intersection of processes rather than to a FST. To test this idea studies on water confined to MCM-41 with different pore sizes and fillings are in progress.

[1] Mishima; Nature, Vol. 396, 329(1998) [2] Hedström; EPJST 141, 53(2007)

CPP 17.4 Tue 10:15 H46

Modelling the relaxation of glass-forming systems at low temperatures: a potential energy approach — •Andreas Heuer and Christian Rehwald — Institute for Physical Chemistry, Corrensstr. 28/30, D-48149 Münster

Based on finite-size effects of a model glass-forming system we have introduced a model which allows one to express the dynamics of a macroscopic glass-former in terms of coupled subunits of temperature-independent size and temperature-dependent coupling constant [1]. The results are obtained from computer simulations on a binary mixture Lennard-Jones model, interpreted in terms of the underlying potential energy landscape. The model is denoted coupled landscape model (CLM).

After a short review of the CLM we present key predictions of this approach for temperatures far below the range accessible by computer simulations. In particular we present results for the violation of the Stokes-Einstein relation (connecting diffusivity and structural relaxation) and the validity of the time-temperature superposition. Finally, the CLM is compared with other models presently discussed for the explanation of the glass-transition phenomena.

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

[2] C. Rehwald, A. Heuer, Phys. Rev. E 86, 051504 (2012)

CPP 17.5 Tue 10:30 H46

Microrheology on supercooled liquids in terms of a Potential Energy Landscape approach — •Carsten Friedrich Erich Schroer and Andreas Heuer — Westfälische Wilhelms-Universität Münster, Münster, Germany

We perform MD simulations of a binary Lennard-Jones mixture where an external force is applied on a single tracer particle. The dynamics of the tracer particle includes several interesting features like non-linear mobilities and anomalous diffusion parallel to the force direction. Our main focus relies in the investigation of the underlying Potential Energy Landscape (PEL), especially in the energetic minima the system explores during its time evolution. Equally to equilibrium systems a coarse graining of these minima to mesoscopic regions allows the description of the system dynamics in terms of a continuous time random walk (CTRW). Extending the concept of the CTRW towards stationary non-equilibrium systems turns out to be an efficient tool for the understanding of non-equilibrium dynamics. First, the approach contains a decomposition between linear and non-linear effects, thus enables a detailed study of the transition between these dynamical regimes. Second, it allows the quantitative understanding of the anomalous diffusion of the tracer particle. Third, for the first time a connection between the non-Gaussian parameter α_2 in equilibrium and superdiffusivity in non-equilibrium can be established. With the help of the underlying PEL, important information can be gained about the dynamics, e.g. about the onset of non-linear effects. The non-linear regime can be discussed in terms of a rejuvenation scenario.

15 min. break.

CPP 17.6 Tue 11:00 H46

Simulation of Aging in SiO2: Single Particle Jump Analysis — •Katharina Vollmayr-Lee[1], Robin Bjorkquist[2], and Landon Chambers[3] — [1]Bucknell University, USA — [2]Cornell University, USA — [3]Texas A&M, USA

Using molecular dynamics computer simulations, we study the aging dynamics of amorphous SiO2. Starting from fully equilibrated configurations at high temperatures the system is quenched to temperatures which are below Tc. We then observe the resulting microscopic dynamics as a function of the waiting time tw, the time elapsed since

the temperature quench. We use single particle trajectories to identify "jumps" when the particle's average position changes over a short time interval significantly compared to its fluctuations. We find that the only tw-dependent microscopic quantity is the number of jumping particles per unit time. Similar to previous studies for fragile glass formers, we show here for the strong glass former SiO2 that neither the distribution of jump lengths nor the distribution of times spent in the cage are tw-dependent. We therefore find a surprising similarity of the jump dynamics of fragile and strong glass formers.

CPP 17.7 Tue 11:15 H46

Excess free energy of supercooled liquids at disordered walls — •Ronald Benjamin and Jürgen Horbach — Institut für Theoretische Physik II - Soft Matter, Heinrich-Heine-Universität Düsseldorf

We perform NVT molecular dynamics simulations of a supercooled liquid confined between identical walls of two types. In the first case flat structureless walls, represented by an external field are considered. In the second case we consider disordered walls consisting of the same supercooled liquid frozen into an amorphous configuration. Using a thermodynamic integration scheme [R.Benjamin and J. Horbach, J. Chem. Phys. **137**, 044707 (2012)] we are able to obtain the excess free energy of the supercooled liquid with respect to both kinds of walls. While a positive excess free energy (of the order of $10k_BT/\sigma^2$) is obtained with respect to a flat structureless wall, the excess free energy between the supercooled liquid and the frozen disordered walls turns out to be negative ($\approx -0.5k_BT/\sigma^2$) even though the potential energy of the supercooled liquid in presence of the disordered walls is the same as that of the bulk. This shows the purely entropic contribution to the excess free energy of the supercooled liquid in presence of the disordered walls. The existence of a negative excess free energy also shows that the thermodynamic properties of such a confined supercooled liquid is not identical to that of the bulk.

CPP 17.8 Tue 11:30 H46

Multiple reentrant glass transitions of soft spheres at high densities — •Michael Schmiedeberg — Institut für Theoretische Physik 2: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, 40204 Düsseldorf, Germany

We study the dynamics of soft spheres by using Molecular Dynamics simulations. The relaxation time varies non-monotonically as a function of density at constant temperature (cf. [1,2]). We determine and study the jamming phase diagrams that indeed show multiple reentrant glass transitions if temperature and density are used as control parameters. However, if we switch to a new formulation of the jamming phase diagrams [3], where temperature over pressure and pressure are employed as control parameters, no non-monotonic behavior can be observed.

[1] L. Berthier, A.J. Moreno, and G. Szamel, Phys. Rev. E 82, 060501(R) (2010).
[2] M. Pica Ciamarra and P. Sollich, arXiv:1209.3334.
[3] T.K. Haxton, M. Schmiedeberg, and A.J. Liu, Phys. Rev. E 83, 031503 (2011).

CPP 17.9 Tue 11:45 H46

Exact Nonlinear Response in the driven lattice Lorentz gas — •Sebastian Leitmann and Thomas Franosch — Institut für Theoretische Physik, Universität Erlangen-Nürnberg, Staudtstraße 7, 91058, Erlangen, Germany

We determine the nonlinear time-dependent response of a tracer on a lattice with randomly distributed hard obstacles as a force is switched on. The calculation is exact to first order in the obstacle density and holds for arbitrarily large forces. In particular, we show that the nonlinear mobility in the stationary state becomes non-analytic in the driving force. Furthermore we demonstrate that the stationary velocity is approached exponentially fast for any finite values of the force, in striking contrast to the power-law relaxation predicted within linear response. We discuss the range of validity of our analytic results by comparison to Monte Carlo simulations.

CPP 17.10 Tue 12:00 H46

From beta-relaxation to alpha-decay: Atomistic picture from molecular dynamics simulations for glass-forming Ni0.5Zr0.5 melt — •Helmar Teichler — Inst. Materialphysik, Univ Göttingen

In glass-forming melts the decay of structural fluctuation shows the well known transition from beta-relaxation (von-Schweidler law with exponent b) to alpha-decay (KWW law with exponent beta). Here we present results from molecular dynamics simulations for a metallic glass forming Ni0.5Zr0.5 model aimed at giving an understanding of this transition on the atomistic scale. At the considered temperature below mode coupling Tc, the dynamics of the system can be interpreted by residence of the particles in their neighbour cages and escape from the cages as rare processes. Our analysis yields that the fraction of residing particles is characterized by a hierarchical law in time, with von-Schweidler b explicitly related to the exponent of this law. In the alpha-decay regime the stretching exponent reflects, in addition, floating of the cages due to strain effects of escaped particles. Accordingly, the change from beta-relaxation to alpha-decay indicates the transition from low to large fraction of escaped particles.

CPP 17.11 Tue 12:15 H46

Interaction between tunnelling systems in glasses — •Gudrun Fickenscher[1], Christian Schötz[1], Paul Fassl[1], Alexander Archer[1], Alexander Burin[2], Manfred von Schickfus[1], Andreas Fleischmann[1], and Christian Enss[1] — [1]Kirchhoff-Institut für Physik, Universität Heidelberg — [2]Tulane University, New Orleans, USA

The low-temperature properties of glasses are governed by tunnelling systems as described in the well-established standard tunnelling model. Interactions between tunnelling systems and phonons lead to energy relaxation. In addition, the interaction between individual tunnelling systems, as predicted by spectral diffusion theory, causes phase decoherence phenomena. To study these interaction processes we have measured the decay of different types of polarization echoes in the standard glass BK7 with respect to the delay time at temperatures between 7.5mK and 70mK. The decay of 2- and 3-pulse echoes is strongly influenced by spectral diffusion. In the case of 3-pulse echoes we expect, in addition, a significant contribution to the decay by energy relaxation processes. On comparing the measured data to numerical calculations we find that the decay of the echo amplitude is slower than predicted by the standard theory at all temperatures. This leads us to the assumption, that there exists a small subspace of tunnelling systems which interact very little with phonons due to a very small coupling constant. Including this subspace in the calculations we can accurately fit the data for all echo types and temperatures with one consistent set of parameters.