CPP 51: Glasses (joint session CPP/DY, organized by DY)

Time: Thursday 9:30–13:00   Location: H48

Invited Talk   CPP 51.1   Thu 9:30   H48
A new look at atomic tunneling systems in glasses containing isotopes with nuclear quadrupol moments — •Andreas Reiser — Kirchhoff Institut für Physik, Universität Heidelberg, INF 227, 69120 Heidelberg, Germany

The physical properties of disordered materials at low temperatures are governed by atomic or molecular tunneling systems. The standard tunneling model explains many properties such as thermal conductivity and heat capacity well. In dynamic experiments for example dielectric susceptibility measurements or polarization echo experiments clear deviations from the standard tunneling model are observed. In particular, the strong influence of nuclear moments inherent in tunneling atoms was proven experimentally recently. We will discuss the mechanism that leads to the interplay of nuclear moments and atomic tunneling systems and will show that at ultra low temperatures a new type of relaxation process enabled by nuclear moments is present. Contrary to previous assumptions this process governs the energy relaxation of the tunneling systems in the limit of lowest temperatures rather than the one phonon process.

CPP 51.2   Thu 10:00   H48
Theoretical investigation of mechanism and statistical properties of a model two-dimension silica glass. — •Projesh Kumar Roy[1] and Andreas Heuer[2] — [1]Graduate school of chemistry, Institute of physical chemistry , Correnstrasse 28/30, D-48149, Muenster — [2]Institute of physical chemistry , Correnstrasse 28/30, D-48149, Muenster

The discovery of 2D silica glass [1] [2] has offered a realistic description of random network theory of silica glass structure; debated for over 80 years[3]. This extremely thin material consists of two layers of silica and exhibits 2D properties in all phases. In the glass phase, the silica network shows a log-normal ring distribution with a typical microstructure or triplet distribution. For simulations two different models based on a Soft - core Yukawa potential[4] and a Stillinger Weber type multi-body potential, have been employed with a binary mixture of silicon and oxygen type particles. After energy minimization, the defect free structures were identified and their ring statistics were compared with the experiment. Going beyond the experiment, the observations can be related to the underlying local energies. Furthermore, two major mechanisms turn out to be responsible for creating various random networks. On this basis a new Monte-Carlo algorithm is proposed for generating random networks of defect free 2D silica.

[1] Heyde M., Shaikhutdinov S., Freund H.-J., Chem. Phys. Lett. 550, 1 (2012). [2] Huang P. Y. et al, Nano Lett. 12, 1081 (2012). [3] Zachariasen W.H., J. Am. Chem. Soc. 54, 3841 (1932). [4] Mendez-Maldonado G.A., et al., J. Chem. Phys. 137, 054711 (2012).

CPP 51.3   Thu 10:15   H48
Relevance of Nuclear Quadrupoles to the Low Temperature Dielectric Properties of Glasses — •Annina Luck, Benedikt Frey, Wiebke Scholz, Guido Homann, Anne Zeissner, Nils Hauff, Philipp Wesp, Andreas Reiser, Andreas Fleischmann, and Christian Enss — Kirchhoff-Institut für Physik, Universität Heidelberg, 69120 Heidelberg

The standard tunneling model based on two level systems describes many properties of solids at low temperatures. Over the years the measurements of dielectric and acoustic properties have, however, shown significant deviations from predictions made by this model. Despite enormous efforts these deviations are yet unexplained.

One aspect which is missing in the prediction of low frequency dielectric properties by the tunneling model is the relevance of nuclear spins, which has been proven to be important in dielectric polarization echo experiments at 1 GHz. Despite of this, modelling the consequences of this effect on the low frequency dielectric and acoustic susceptibility has proven difficult.

In order to experimentally investigate the role of nuclear quadrupoles in the dielectric susceptibility we have investigated multiple glass samples containing elements with different quadrupole moments in a wide frequency range spanning from Hz to GHz.

We have found clear indications for a novel nuclear spin driven two level system relaxation process at very low temperatures.

CPP 51.4   Thu 10:30   H48
Glass transition and stable glass formation of carbon tetrachloride — •Yeong Zen Chua[1], Michael Tylinski[2], Mark D. Ediger[2], and Christoph Schick[1] — [1]Institute of Physics, University of Rostock, Rostock 18051 Germany — [2]Department of Chemistry, University of Wisconsin-Madison, Madison, Wisconsin 53706 USA

There have been attempts to find correlations between the ability to form stable glasses and general characteristic of the materials. It was argued that asymmetric molecules, which allow anisotropic packing on the surface during deposition, are a prerequisite for stable glass formation. This leads to the question whether symmetric molecules can form stable glasses. Carbon tetrachloride (CCl_4) is an ideal molecule to verify this hypothesis, since the molecule is pseudo spherical with no orientation. On the other side, the formation of stable glasses is thought to be mediated by a highly mobile surface layer. So it might be expected that CCl_4, as a small and simple organic molecule, has enhanced mobile surface layer and thus increases its ability to form stable glasses. These conflicting factors lead to our investigation of CCl_4 glasses produced by physical vapor deposition with *insitu* AC chip nanocalorimetry. Kinetically stable glasses have been observed to form at substrate temperature around $0.8\,T_g$, consistent with previous work on stable glass formers. The isothermal transformation of the as-deposited glasses into supercooled liquid state gave further evidence to support the stable glass formation, thus disproving molecular asymmetry as a prerequisite. The glass transition temperature is determined as $T_g = (78\pm2)$ K, which is different from previously reported values.

CPP 51.5   Thu 10:45   H48
Influence of MCM-41 on the dynamical and phase behaviors of aqueous mixtures — •Matthias Sattig and Michael Vogel — Institut für Festkörperphysik, TU Darmstadt, Hochschulstraße 6, 64289 Darmstadt, Germany

The dynamics of aqueous mixtures in bulk and confinement is a topic of great interest. Two model systems, which exhibit rather different behavior upon varring water concentration are propylene glycol- and propylene glycol monomethyl ether-mixtures. This was attributed to the different ability of both alcohols to form hydrogen bonds in neat systems and in aqueous mixtures. The formation of H-bond networks can be altered by geometrical confinement, whose surface interacts with the guest molecules and provides spatially restriction. Here we provide temperature dependent deuteron-NMR data of heavy water in mixture with PG, PGME or di-PGME confined in mesoporous MCM-41 in a broad dynamic range. We examin the rotational autocorrelation using stimulated echo experiments and spin-lattice-relaxation. The latter exhibits a typical minimum in its relaxation time T_1, below which several relaxation processes could be identified in the different mixtures, supporting the idea of a phase separation in confinement, that was proposed in QENS [1] and dielectric [2] investigations. We compare our results with the dynamics of water [3] and glycerol confined in the same material to discuss similarities and differences relating to the influence of confinement on the dynamics of neat and mixed systems.
[1] Swenson et al., JCP, 141, 214501, 2014 [2] Elamin et al., PCCP, 17, 12978, 2015 [3] Sattig et al., JCPL, 5, 174 - 178 , 2014

CPP 51.6   Thu 11:00   H48
Spatio-temporal Correlations of Glass-forming Systems in Terms of Coupled Energy Landscapes — •Carsten Schroer[1,2] and Andreas Heuer[1,2] — [1]Westfälische Wilhelms-Universität Münster, Institut für physikalische Chemie, Corrensstraße 28/30, 48149 Münster, Germany — [2]NRW Graduate School of Chemistry, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany

The concept of the potential energy landscape is an effective tool for the physical description of supercooled liquids in the vicinity of the glass-transition. In this framework, typically small elementary systems are studied for which a strong correlation between its thermodynamic state and its dynamics can be found[1]. The transfer of these insights to macroscopic systems can be achieved by regarding the larger system as a superposition of elementary systems. The observed finite-size effects of the structural relaxation time contain important information about the coupling between these subsystems[2].

In this talk we discuss the impact of this coupling mechanism on the occurrence of spatio-temporal correlations, measured by four-point cor-

relation functions. The results obtained by our model are compared with numerical data obtained by molecular dynamics simulations. We demonstrate that this model, based on the energy landscape of the small system plus the coupling, is sufficient to describe complex behavior of glass-formers.

[1] A. Heuer, *J. Phys.: Condens. Matter* **20**, 37 (2008).
[2] C. Rehwald and A. Heuer, *Phys. Rev. E* **86**, 051504 (2012).

CPP 51.7   Thu 11:15   H48

Colloidal monodisperse hard ellipsoids — •PATRICK PFLEIDERER[1], STEFAN SCHÜTTER[2], NICOLAI SÄNGER[2], MATTHIAS KLEIN[2], and ANDREAS ZUMBUSCH[2] — [1]Department of Physics, University of Konstanz, 78457 Konstanz, Germany — [2]Department of Chemistry, University of Konstanz, 78457 Konstanz, Germany

The hard sphere system has been generating hundreds of publications per year for the past decades. The focus of attention has recently been shifted towards anisotropic particles, as rotational degrees of freedom have been identified as key for progress in statistical mechanics models and engineering alike. One prominent puzzle is the violation of Stokes-Einstein(-Debye) relations near the glass transition.

The next obvious simple system besides spheres is the hard ellipsoid system. It lends itself to theoretical descriptions and simulations. While an experimental technique to prepare colloidal poly(methyl methacrylate) (PMMA) ellipsoids exists since the 1990s, little progress has been made towards an establishment anywhere near the 'fruit fly' hard spheres. This is due to the difficulties in endowing several desired properties simultaneously to the system: mass-density and refractive-index matching, hard-core interactions, long-term stability etc. We present the first 3D hard ellipsoid system which meets all these goals, fluorescently labeled with a unique core-shell structure [1], polydispersity as low as 3%, and study the interplay of rotational and translational degrees of freedom approaching the colloidal glass transition.

[1] M.K. Klein, N. Saenger, S. Schuetter, P. Pfleiderer, and A. Zumbusch, Langmuir 30, 12457 (2014).

15 min. break

CPP 51.8   Thu 11:45   H48

Electron bombardment induced cation transport in an ion conducting glass — •ANNELI HEIN, JAN WIEMER, and KARL-MICHAEL WEITZEL — Fachbereich Chemie, Philipps-Universität Marburg

The bombardment induced ion transport (BIIT) technique has been developed for measuring ionic conductivities and activation energies for ion hopping. The original version is based on attaching a cation beam to the sample (cation-BIIT) inducing the transport of such cations towards a grounded backside electrode. In the current work the BIIT technique is extended to electron bombardment (e- - BIIT). Here a low energy electron beam has been attached to a mixed sodium and potassium conducting borosilicate glass mounted on a single grounded backside electrode. Attachment of the electrons to the surface induces cation transport towards this surface. In the first part of the experiment current-voltage curves have been measured. The specific conductivities as well as the activation energy for ion hopping derived agree with cation-BIIT data. In the second part of the experiment we have performed a long term electron bombardment of the glass sample at -25 eV. Ex situ - after the bombardment - the sample has been analyzed by time-of-flight secondary ion mass spectrometry (ToF-SIMS). The depth profiles obtained from the ToF-SIMS analysis clearly demonstrate that sodium has been neutralized at the front side of the glass sample due to the recombination of electrons with sodium ions. Reduction of the less noble potassium ions appears to be suppressed. The electrochemical implications of this observation will be discussed.

CPP 51.9   Thu 12:00   H48

Residual stresses in glass forming systems — •GAURAV PRAKASH SHRIVASTAV[1], PINAKI CHAUDHURI[2], and JÜRGEN HORBACH[1] — [1]Heinrich-Heine-Universität Düsseldorf, Düsseldorf, Germany — [2]The institute of Mathematical Sciences, Chennai, India

When a sheared glass is allowed to relax, stress does not decay to zero but tends towards a finite value. How the resulting residual stress is spatially distributed in the system is only poorly understood, especially in the case of a sheared system with shear bands. Also, the effect of different shear rates, with which the initial glass is deformed, and the effect of the amount of strain present in the initial deformed glass on the residual stress distribution are open issues.

In the present work, we study, using molecular dynamics simulations, relaxation of stresses in a sheared model glass former. We first deform a binary glass forming Lennard-Jones mixture by shearing it with a constant strain rate. Then, we switch of the shear in different regimes of the stress-strain curve and allow the system to relax. We find that the amount of residual stress in the system depends on the initial strain before the shear cessation. By looking at the spatially resolved mean square displacement of particles, we find that the residual stress remains localized in regions where the shear band has been present before. We also find that the deformed glass has higher Poisson's ratio than the undeformed glass, consistent with previous observations.

References
[1] I Binkowski, G. P. Shrivastav, J. Horbach, S. V. Divinski, G. Wilde arXiv:1506.03031 (2015).

CPP 51.10   Thu 12:15   H48

Nonlinear Microrheology of Supercooled Liquids in Terms of an Effective Temperature — •CARSTEN SCHROER[1,2] and ANDREAS HEUER[1,2] — [1]Westfälische Wilhelms-Universität Münster, Institut für physikalische Chemie, Corrensstraße 28/30, 48149 Münster, Germany — [2]NRW Graduate School of Chemistry, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany

We perform computer simulations of a fragile model glass-former in which a single particle is driven by an external force through the liquid. Thereby, we track the path the system takes through its underlying Potential Energy Landscape (PEL) and aim for understanding how this is altered by the external field[1,2] and how the altering is related to the nonlinear responses of dynamic quantities. In this talk, we show that, for strong forces, the thermodynamic state and the local kinetics of the system is altered by the external force in a similar way as an increase of the (bath) temperatures. This behavior gives rise to the definition of an *effective temperature* that turns out to be characterized by a single force and temperature-independent parameter only, even for bath temperature below the computer glass transition. Combining these observations enables us to derive scaling relations for the nonlinear mobility and the diffusive properties of the tracer particle that are tested with our numerical data [3].

[1] C. F. E. Schroer, A. Heuer, *J. Chem. Phys.* **138**, 12A518 (2013).
[2] C. F. E. Schroer, A. Heuer, *Phys. Rev. Lett.* **110**, 067801 (2013).
[3] C. F. E. Schroer, A. Heuer, *J. Chem. Phys.* (accepted) (2015).

CPP 51.11   Thu 12:30   H48

Nonlinear Microrheology of Supercooled Liquids in Terms of an Effective Temperature — •CARSTEN SCHROER[1,2] and ANDREAS HEUER[1,2] — [1]Westfälische Wilhelms-Universität Münster, Institut für physikalische Chemie, Corrensstraße 28/30, 48149 Münster, Germany — [2]NRW Graduate School of Chemistry, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany

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[1] C. F. E. Schroer, A. Heuer, *J. Chem. Phys.* **138**, 12A518 (2013).
[2] C. F. E. Schroer, A. Heuer, *Phys. Rev. Lett.* **110**, 067801 (2013).
[3] C. F. E. Schroer, A. Heuer, *J. Chem. Phys.* (accepted) (2015).

CPP 51.12   Thu 12:45   H48

Potential energy landscape analysis of sheared glass-forming systems — •MARKUS BLANK-BURIAN and ANDREAS HEUER — Institut für Physikalische Chemie, WWU Münster, Deutschland

We performed molecular dynamics simulations of small binary Lennard-Jones mixtures ($65 \leq N \leq 1040$) under constant shear rates and at very low temperature ($T = 0.01$) as well as above T_g. We also performed shear reversal simulations.

In previous work on unsheared systems it was shown, that most of

the physical properties of macroscopic systems are already encoded in small systems. We can show that finite size effects have only limited influence on major observables of the sheared system at $N = 130$.

For the analysis of the sheared system, we perform energy minimization using the strain as an additional variable. We then use this information to identify inherent structures (IS) and metabasins (MB) from the trajectories. The IS have zero strain and are therefore comparable to the unsheared system.

From the resulting statistical data we gain a microscopic understanding of macrorheological phenomena like the initial stress overshoot as well as shear thinning in the plastic flow regime from a potential energy landscape perspective.