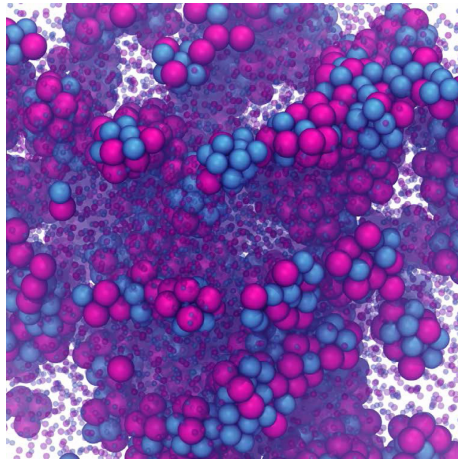# The role of local structure in dynamical arrest

July 21, 2015 - July 23, 2015
Johannes Gutenberg University Mainz, Germany

C. Patrick Royall
University of Bristol, United Kingdom

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1 General Information

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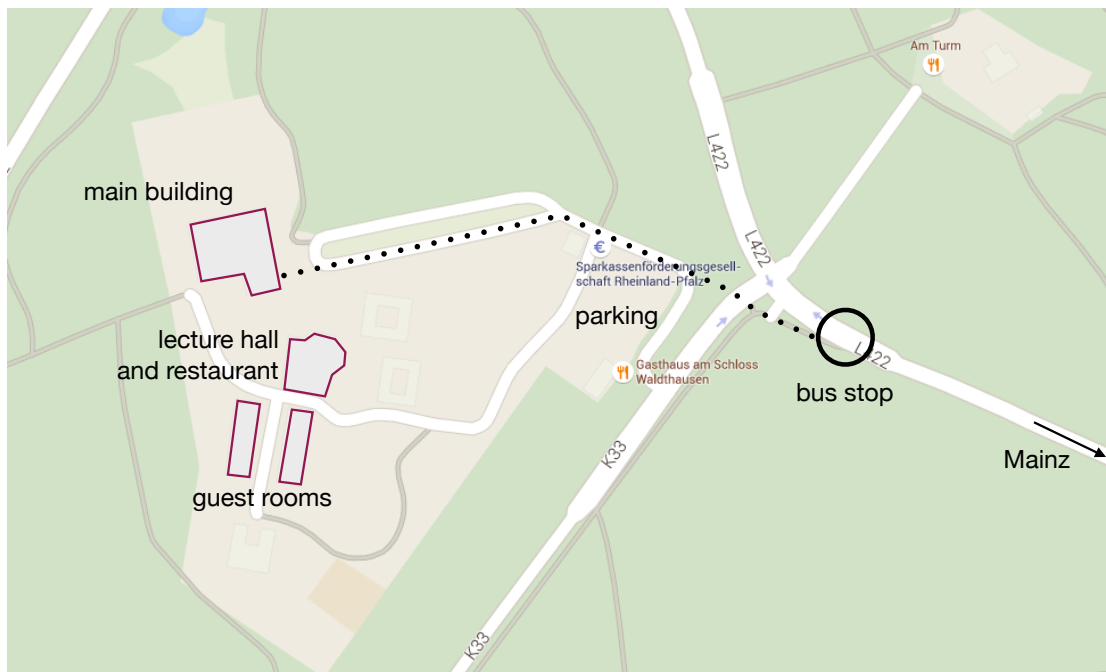
Opening hours:
- "Weinstube" (bar located in the basement of the main building)
  open from 7:30 pm to 12:00 midnight (closed on Sundays)
- Restaurant:
  Breakfast from 7:00 to 9:00 am
  Lunch from 12:00 to 1:45 pm
  Dinner from 6:00 to 7:30 pm

2 Program

	Tuesday	Wednesday	Thursday
8:45	*opening*		
9:00 / 9:20	Harrowell	Tarjus	Charbonneau
9:40 / 10:00	Chen	Ediger	Pedersen
10:20	*break*	*break*	*break*
10:40 / 11:00	Kelton	Cammarota	Angell
11:20 / 11:40	Wilson	Garrahan	Del Gado
12:00	Janssen	Godfrey	Turci
12:20	*lunch*	*lunch*	*lunch*
14:20 / 14:40	Coslovich	Salmon	Jack
			Tanaka
15:00	Zhang	Pastore	
15:20	*break*	*break*	*discussion*
15:40	Shokef	Bhattacharyya	
16:00	Liu		*closing*
16:20 / 16:40	Wuttig	Palberg	
17:00	*posters*	*break*	
17:20 / 17:40		Samwer	
18:00		*discussion*	
18:20 / 18:40 / 19:00	*dinner*	*barbeque*	
	posters		

Day 1 - Tuesday July 21, 2015

Registration
- 8:00 to 8:45 - Registration

Welcome
- 8:45 to 9:00 - Welcome

Session 1
- 9:00 to 9:40 - **Peter Harrowell**
  The role of the geometry and multiplicity of favoured local structures in determining the stability of supercooled liquids

- 9:40 to 10:20 - **Mingwei Chen**
  Structure of metallic glasses: from short-range order, medium range order to long range disorder

Break

- 10:20 to 10:40 - Coffee Break

Session 2
- 10:40 to 11:20 - **Ken Kelton**
  Correlation between kinetic and structural fragility in metallic liquids

- 11:20 to 12:00 - **Mark Wilson**
  Structure and dynamics in network-forming materials

- 12:00 to 12:20 - **Liesbeth Janssen**
  Quantitative relaxation dynamics of supercooled liquids from first principles

Lunch

- 12:20 to 14:20 - Lunch

Session 3
- 14:20 to 15:00 - **Daniele Coslovich**
  Non-universal role of local structure around the dynamic crossover

- 15:00 to 15:20 - **Zexin Zhang**
  Glassy dynamics in two-dimensional melting of colloidal crystals

Break

- 15:20 to 15:40 - Coffee Break

Session 4
- 15:40 to 16:00 - **Yair Shokef**
  The relation between structure and relaxation in kinetically-constrained models

- 16:00 to 16:20 - **Amelia Liu**
  Bond-orientational order in metallic glasses and colloids measured by limited-volume diffraction

- 16:20 to 17:00 - **Matthias Wuttig**
  The role of structure and bonding for aging and crystallization in phase change materials

Posters
- 17:00 to 18:20 - Poster Session

Dinner

- 18:20 to 19:30 - Dinner

Day 2 - Wednesday July 22, 2015

Session 5
- 9:00 to 9:40 - **Gilles Tarjus**
  Frustration, structure and dynamical slowing down in glass-forming liquids

- 9:40 to 10:20 - **Mark Ediger**
  Control of stability and molecular orientation in organic glasses

Break

- 10:20 to 10:40 - Coffee Break

Session 6
- 10:40 to 11:20 - **Chiara Cammarota**
  Random pinning glass transition: features, origin, consequences

- 11:20 to 12:00 - **Juan Garrahan**
  Metastability, transitions in overlap and activity, and dynamic facilitation

- 12:00 to 12:20 - **Michael Godfrey**
  Dynamical heterogeneity in a system of hard disks in a channel

Lunch

- 12:20 to 14:20 - Lunch

Session 7
- 14:20 to 15:00 - **Phil Salmon**
  Identification and characterisation of the different structural length scales in glass-forming materials

- 15:00 to 15:20 - **Raffaele Pastore**
  Cage–jump motion reveals universal dynamics and non-universal structural features in glass forming liquids

Break

- 15:20 to 15:40 - Coffee Break

Session 8
- 15:40 to 16:20 - **Sarika Bhattacharyya**
  Role of crystal structures and their frustration in the stability of a supercooled liquid

- 16:20 to 17:00 - **Thomas Palberg**
  Limits of homogeneous crystallization in suspensions of charged colloidal spheres approaching monodisperse size distributions

Break

- 17:00 to 17:20 - Coffee Break

Session 9
- 17:20 to 18:00 - **Konrad Samwer**
  Strain (-rate) induced fragility transition in bulk metallic glasses

- 18:00 to 18:40 - Discussion

Dinner
- 19:00 to 22:00 - Social Dinner

Day 3 - Thursday July 23, 2015

Session 10
- 9:00 to 9:40 - **Patrick Charbonneau**
  Cavity point-to-set correlations: recent advances

- 9:40 to 10:20 - **Ulf Pedersen**
  Statistics of density fluctuations in supercooled viscous liquids

Break

- 10:20 to 10:40 - Coffee Break

Session 11
- 10:40 to 11:20 - **Austen Angell**
  Tuning of second critical temperature in the s-w model gives insight into the ultrastable glass challenge, and supercooled water conundrum.

- 11:20 to 12:00 - **Emanuela Del Gado**
  Length scales, soft modes and non-affine rearrangements in the inherent structures of supercooled liquids

- 12:00 to 12:20 - **Francesco Turci**
  Evidence for a second order phase transition to an ideal glass from trajectory sampling

Lunch

- 12:20 to 14:20 - Lunch

Session 12
- 14:20 to 14:40 - **Rob Jack**
  Overlap fluctuations in glassy systems

- 14:40 to 15:20 - **Hajime Tanaka**
  Role of structural ordering in slow dynamics and crystallization of a supercooled liquid

Closing
- 15:20 to 16:00 - Discussion

- 16:00 to 16:10 - Summary & Conclusions

3 Abstracts

The role of the geometry and multiplicity of favoured local structures in determining the stability of supercooled liquids

Peter Harrowell[1] and Pierre Ronceray[2]

[1] School of Chemistry, University of Sydney, Sydney, Australia
[2] Laboratoire de Physique Théorique et Modèles Statistiques, Université Paris-Sud, Orsay, France

We investigate the connection between the geometry of Favoured Local Structures (FLS) in liquids and the associated liquid and solid properties. We introduce a lattice spin model – the FLS model on a face centered cubic lattice – where this geometry can be arbitrarily chosen among a discrete set of 115 possible FLS [1]. We find crystalline groundstates for all choices of a single FLS. Sampling all possible FLS's, we identify the following trends: (i) low symmetry FLS's produce larger crystal unit cells but not necessarily higher energy groundstates, (ii) chiral FLS's exhibit peculiarly poor packing properties, (iii) accumulation of FLS's in supercooled liquids is linked to large crystal unit cells, and (iv) low symmetry FLS's tend to find metastable crystal structures on cooling. Apart from the depression of the freezing point due to the high energy of their crystals, we find no compelling evidence to suggest that low symmetry FLS's exert any particular influence in stabilizing the supercooled liquid. In contrast, when we extend our model to include multiple degenerate FLS's we find that the stability of the supercooled liquid is systematically enhanced as we increase the multiplicity of the FLS's.

[1] P. Ronceray and P. Harrowell, Soft Matt. 11, 3322 (2015)

Structure of metallic glasses: from short-range order, medium range order to long range disorder

Mingwei Chen
Tohoku University, Japan

Atomic structure of metallic glasses is a long-standing issue in materials science and solid-state physic. Extensive theoretical and experimental investigations have suggested that the disordered glasses possess pronounced short-range order (SRO) arising from densely-packed arrangements of nearest-neighbor atoms. However, it remains unclear how SRO evolves into long-range disorder with retained dense atomic packing. Here we show experimental atomic configurations of the best metallic glass former, Pd42.5Cu30Ni7.5P20, using scanning Angstrom-beam electron diffraction (ABED) which provides a series of adjacent SROs through point-by-point acquisitions of ABED. Atomic configurations of the metallic glass can be experimentally reconstructed by the assembly of the interconnected SRO. The experimental atomic structure reveals the hidden ordering beyond SRO and provides a new insight into the inherent correlation between SRO and long-range disorder as well as the structural origins of dynamic arrest of supercooled metallic liquids.

Correlation between kinetic and structural fragility in metallic liquids

Ken Kelton
Department of Physics & Institute of Materials Science and Engineering, Washington University, St. Louis Missouri 63130

Fragility is a concept that is often correlated with glass formability. It is generally defined in terms of dynamical aspects of the liquid, usually based on the rate of increase of the viscosity as a function of the inverse temperature scaled to the glass transition temperature, T_g (i.e. T_g/T). Our experimental studies of the temperature dependence of the liquid structure factor, $S(q)$, for a range of metallic alloy liquids demonstrate a link between structural and kinetic fragility [1]. A new scaling temperature, T_A, is also identified, which, based on the results of molecular dynamics simulations, corresponds to the onset of cooperative flow in the liquid [2]. Our recent experimental and MD results show that the rate of short-range and medium-range structural

ordering in the liquid accelerates below the temperature of this dynamical crossover, further demonstrating the link between structure and dynamics. Further, for all metallic glass-forming liquids studied $T_g \cong T_A/2$, suggesting a deep connection between the onset of cooperative dynamics and the glass transition [3], which is supported by our recent MD results [4]. Finally, by scaling the inverse temperature by T_A, and the magnitude of the viscosity by the extrapolated viscosity at infinite temperature, η_o, a universal curve from T_A down to T_g is obtained for all of the metallic liquids studied. On average, η_o is equal to the product of the number density and Planck's constant [5]. These points will be discussed and suggestions made for further studies. Research was partially supported by the National Science Foundation (DMR-12-06707) and NASA (NNX10AU19G).

[1] N. A. Mauro, M. E. Blodgett, M. L. Johnson, A. J. Vogt and K. F. Kelton, Nature Comm., 5, 4616 (2014)
[2] T. Iwashita, D. M. Nicholson and T. Egami, Phys. Rev. Lett. 110, 205504 (2013)
[3] R. Soklaski, V. Tran, Z. Nussinov, K. F. Kelton, L. Yang, arXiv:1502.01739
[4] M. Blodgett, T. Egami, Z. Nussinov and K. F. Kelton, arXiv:1407.7558
[5] Z. Nussinov, F. S. Nogueira, M. Blodgett and K. F. Kelton (arXiv:1409.1915)

Structure and dynamics in network-forming materials

Mark Wilson
Department of Chemistry
University of Oxford

The structure and dynamics of network-forming materials of MX2 stoichiometry are investigated by molecular dynamics. Two approaches are considered. In the first, ``good'' models for key target systems (SiO2 and ZnCl2) are developed and applied. In the second, a single parameter (the anion polarizability) is varied to control the network topology. At ambient pressure the systems have common tetrahedral coordination polyhedra (short-range order - SRO) but differ in the way in which these tetrahedra inter-link, which in turn control ordering on longer length-scales. The concept of amorphous structure being described as a network(constructed from relatively simple repeating units) goes back to the two-dimensional representation proposed by Zachariasen in 1932. In three-dimensions systems such as C, Si or Ge, or more complex systems like SiO2, GeO2 or GeSe2 can usefully considered as network materials. However, diffraction experiments which are most commonly used to probe the structures of these systems, highlight structure primarily at a pair-wise level often displaying ordering on multiple length-scales. Potentially more complex structure (bond angles, ring structure...) must be inferred from the experimental data often augmented with parallel simulation studies. Recent experimental developments have allowed for the generation of two- (or near two-) dimensional amorphous structures for C and SiO2. Electron microscopy allows the atomistic structure of these systems to be determined unequivocally and, as a result, complex structural correlations, such as those expressed in terms of the ring structure, are easily obtained. However, these experiments are in their infancy and the effect of the formation conditions on the underlying structure is unclear. As a result, a detailed understanding of the underlying interactions is required if materials are to be grown in a controlled fashion. In this talk key target systems will be modelled in both three- and two-dimensions. The evolution of the structure and dynamics both in which the local coordination polyhedra change, and in which the spatial relationships between these units change, will be investigated. The role of the different underlying length-scales in determining the bulk relaxation to glass will be discussed.

Quantitative relaxation dynamics of supercooled liquids from first principles

Liesbeth Janssen and David R. Reichman
Columbia University, New York, USA

Understanding the liquid-to-glass transition remains one of the deepest unsolved problems in condensed matter science. Here we present a new theoretical framework, referred to as generalized mode-coupling theory (GMCT), which can predict the microscopic dynamics of glass-forming systems with near-quantitative accuracy on a strictly first-principles basis. The theory is based on the well-established standard mode-coupling theory (MCT) of the glass

transition, but rigorously incorporates higher-order dynamic density correlations neglected in standard MCT. We demonstrate that, using only the static structure factor of a system as input, GMCT can already accurately describe the microscopic dynamics over an unprecedentedly large time and density domain [1]. Moreover, we show that GMCT is capable of predicting novel types of glass transitions, including type-A, type-B, and avoided transitions, as well as different types of relaxation-time scaling behaviors [2]. This suggests that GMCT may constitute the first microscopic, first-principles theory that can account for different fragilities in glass-forming materials. Overall, this framework paves the way towards a systematically correctable and ultimately exact theory of glassy dynamics, in which the importance of higher-order static and dynamic correlations in glassy systems can be rigorously and systematically explored.

[1] L.M.C. Janssen and D.R. Reichman, under review in Nature Phys.
[2] L.M.C. Janssen, P. Mayer, and D.R. Reichman, Phys. Rev. E 90, 052306 (2014)

Non-universal role of local structure around the dynamic crossover

Daniele Coslovich[1], G. Hocky[2], A. Ikeda[3], and D.R. Reichman[4]
[1] Université de Montpellier, France
[2] University of Chicago, USA
[3] Kyoto University, Japan
[4] Columbia University, USA

The dynamics of glass-forming liquids is heterogeneous and displays growing spatial correlations upon cooling. Whether such behavior arises from heterogeneities in the local structure or more complex forms of amorphous order is a highly debated question. To clarify this issue, we study several model liquids within a coherent simulation framework based on the iso-configurational ensemble. We find that the correlation between the preferred local structure and the propensity of motion is pronounced in systems that deviate markedly from the mean-field picture of glassy dynamics and weak in models that adhere to it to a good extent [1]. We conclude that the degree to which structural correlations affect the dynamics around the dynamic crossover is, in general, system-dependent.

[1] G.M. Hocky, D. Coslovich, A. Ikeda, D.R. Reichman, Phys. Rev. Lett. 113, 157801 (2014)

Glassy dynamics in two-dimensional melting of colloidal crystals

Zexin Zhang, H. Wang, and H. Xi
Center for Soft Condensed Matter Physics and Interdisciplinary Research, Soochow University, Suzhou, China

We study glassy dynamics in two-dimensional (2D) melting by colloidal experiments and computer simulations. The 2D melting proceeds from a solid phase into a intermediate hexatic phase then into a liquid phase, which is consistent with the KTHNY theory [1]. Intriguingly, we find, in the hexatic phase, hallmarks of glassy systems such as slow dynamics, strong dynamical heterogeneity and string-like structures of particles moving cooperatively. We identify the structural origin for such glassy behavior, and reveal a direct relation between local ordering and the dynamics [2-5]. Thus, our studies shed new light on the nature of 2D melting, and provide a novel perspective on studying glass and dynamical arrest in general [6].

[1] D. R. Nelson, Defects and Geometry in Condensed Matter Physics, Cambridge Univ. Press (2002)
[2] D.N. Perera, P. Harrowell, J. Chem. Phys. 111, 5441 (1999)
[3] T. Kawasaki, T. Araki, H. Tanaka, Phys. Rev. Lett.99, 215701 (2007)
[4] C.P. Royall, S.R.Williams, T. Ohtsuka, H. Tanaka, Nat. Mater. 7, 556 (2008)
[5] B. Charbonneau, P. Charbonneau, G. Tarjus, J. Chem. Phys. 138, 12A515 (2013)
[6] H. Wang, H. Xi, Z. Zhang, submitted for publication (2015)

The relation between structure and relaxation in kinetically-constrained models

Yair Shokef, Nimrod Segall, and Eial Teomy
Tel Aviv University, Tel Aviv, Israel

In a liquid all the particles are mobile, while in a glass only some of them are mobile at any given time. Although overall the structure is amorphous in both cases, the difference is that in glasses there are local structures that inhibit the movement of particles inside them. We investigate the size of these structures by considering the minimum number of particles that need to move before a specific particle can move. In kinetically constrained models this structural property, the mean culling time, is easy to find by iteratively culling mobile particles from a snapshot of the system. We use the Kob-Andersen, Fredrickson-Andersen, and the spiral kinetically-constrained models, which are either lattice gases in which a particle may hop to a nearby site if its local environment satisfies some constraint, or Ising-like models in which a spin, representing regions of high and low mobility, can flip if its environment satisfies some constraint. Due to these constraints and the lattice nature of the models, there is a clear unambiguous definition for which particles are mobile in a given configuration, and various analytical tools may be used to calculate their fractions. We compare these structural properties to the dynamics in these models by measuring the persistence time, which is the average time it takes a particle to move for the first time. Again, due to the lattice nature of these models, this relaxation time is unambiguously defined, and various analytical tools may be used to calculate it. We find an algebraic relation between the mean culling time and the persistence time, with a model-dependent exponent.

The role of structure and bonding for aging and crystallization in phase change materials

Matthias Wuttig
RWTH Aachen University of Technology, Germany

Phase change media are among the most promising materials in information technology. They are already employed in rewriteable optical data storage, where the pronounced difference of optical properties between the amorphous and crystalline state is employed. This unconventional class of materials is also the basis of a storage concept to replace flash memory. In this application, the material is rapidly and reversibly switched between the amorphous and crystalline state. Hence, it is essential to understand the structure, bonding and the resulting material properties of both the amorphous and crystalline state, as well as the transformation kinetics between the different states. This talk will discuss the unique material properties, which characterize phase change materials as well as their transformation kinetics. In particular, it will be shown that the amorphous and the crystalline state of phase change materials differs in the bonding mechanism, which is utilized [1,2]. While resonance bonding is utilized in crystalline phase change materials, ordinary covalent bonding is found in the amorphous state. Interesting enough, the transformation from the amorphous to the crystalline state can be remarkably fast [3]. Phase change materials are bad glass formers and fragile liquids [4]. The origin of this fragile behaviour will be discussed. Furthermore, we will discuss the unconventional aging in these materials [5].

[1] K. Shportko et al., Nature Materials 7, 653 (2008)
[2] D. Lencer et al., Nature Materials 7, 972 (2008)
[3] G. Bruns et al., Appl. Phys. Letters 95, 043108 (2009)
[4] M. Salinga et al., Nature Communications 4, 2371 (2013)
[5] J.-Y. Raty et al., Nature Communications, in press

Bond-orientational order in metallic glasses and colloids measured by limited-volume diffraction

Amelia Liu[1], Rico Tabor[1], Laure Bourgeois[1], Timothy Petersen[1], Martin de Jonge[2], and Stephen Mudie[2]
[1] Monash University, Clayton, Australia
[2] Australian Synchrotron, Clayton, Australia

The existence of a structural motif or motifs that frustrate periodic order and give rise to good glass-formability has not been experimentally confirmed despite long-standing hypotheses bolstered by simulation. This lack of evidence reflects a current grand challenge in the

experimental determination of disordered structures. It is well known that the 2-body correlation function measured using standard broad-beam diffraction is not sufficient to discriminate between competing structural models in disordered materials [1]. By limiting the coherence length of the incident radiation to a size comparable to that of the short-medium range structural units, fluctuations in the diffracted intensity may be detected [2, 3]. These fluctuations relate to higher-order correlation functions (3- and 4-body) in the material, and are usually averaged out if the diffracted volume samples too many short-range configurations. These higher-order functions are extremely sensitive probes of atomic-range order in the material. We examine angular correlations in electron nano- and x-ray micro- diffraction patterns of metallic glasses and glassy colloids, respectively [2,4]. We develop the dynamical diffraction theory of disordered structures to enable us to interpret these angular symmetries in the diffraction plane in terms of the symmetries of the short-range polyhedral clusters in the structure itself. We present statistical measurements of these symmetries. This novel technique shows distinct promise as a tool to understand the short-range order in glassy materials.

[1] J. M. Ziman, 'Models of Disorder: The Theoretical Physics of Homogeneously Disordered Systems', Cambridge University Press, (1979)
[2] A. C. Y. Liu et al., Phys. Rev. Lett. 110, 205505 (2013)
[3] M. M. J. Treacy, J. M. Gibson, L. Fan, D. J. Paterson, I. McNulty, Rep. Prog. Phys. 68, 2899 (2005)
[4] P. Wochner et al., Proc. Natl. Acad. Sci. 106, 11511 (2009)

Frustration, structure and dynamical slowing down in glass-forming liquids

Gilles Tarjus
LPTMC, CNRS/UPMC, Paris, France

The slowing down of relaxation leading to glass formation may be induced by several mechanisms, which is of course unfortunate for devising a universal theoretical description of the phenomenon. Even considering only those explanations that involve the putative growth of some static characteristic length scale as one approaches the glass transition, two main mechanisms may be at play: on the one hand, the frustrated extension of some local order characterizing a given liquid and, on the other hand, the development of generic point-to-set spatial correlations as predicted from the mean-field scenario of glassy systems. In this talk, I will consider some ways to disentangle these two possibilities and argue that their relative importance is connected to the degree of frustration found in a specific glass-former.

Control of stability and molecular orientation in organic glasses

Mark Ediger
University of Wisconsin-Madison

The large number of local packing arrangements in glasses underlies their important properties, including macroscopic homogeneity (e.g., the clarity of window glass) and the ability to be tuned by composition changes. Their nonequilibrium nature means that many different glasses of the same composition can be prepared and some of these glasses may have quite exotic and useful properties. A problem with glasses, also associated with their large number of packing arrangements, is that they may not be kinetically stable with respect to lower energy glasses and crystalline states. We have used physical vapor deposition and the mobility of glassy surfaces to prepare what are likely the most kinetically stable glasses on the planet, in comparison to liquid-cooled glasses of the same composition. Our materials have the properties expected for "million-year-old" glasses, including high density, low enthalpy, and high mechanical moduli. In fact, these glasses are so well-packed that thin films melt via constant velocity fronts similar to crystals. We have discovered deposition conditions that combine high stability with substantial molecular orientation. We have used computer simulations to understand why some deposition conditions orient molecules vertically and others orient them horizontally. These developments present major opportunities to expand our understanding of amorphous packing and to design new classes of anisotropic solids for applications such as organic electronics.

Random pinning glass transition: features, origin, consequences

Chiara Cammarota[1] and Giulio Biroli[2]
[1] La Sapienza/University of Rome, Rome, Italy
[2] IPhT/CEA Saclay, Gif-sur-Yvette, France

Pinning a fraction of particles from an equilibrium configuration in supercooled liquids is a structural-agnostic way to bias the system in favour of a specific, self-induced amorphous order. This procedure has been recently proposed [1,2] as a way to induce a new kind of glass transition, the Random Pinning Glass Transition (RPGT). The RPGT has been predicted to share some features of standard thermodynamic glass transitions and usual first order ones [3]. Thanks to its special nature, the approach and the study of the RPGT appears to be a fairly reachable task compared to the daunting problem of inspecting standard glass transitions [4,5]. Moreover, it provides a new and very promising way to probe the ideal glass critical point and its features to ascertain or disprove the validity of the theories of the glass transition[6,7]. Finally it gives rise to peculiar equilibrium and out-of-equilibrium phenomenology to be tested by numerical simulations and experiments [8,9]. Besides a review of the RPGT physics, I will discuss the origin of the specific features of the RPGT suggesting some directions for numerical and experimental checks and possible extensions of this procedure to test the role of structure or the role of dynamical constraints.

[1] C.Cammarota and G.Biroli, PNAS 109, 8850 (2012)
[2] C.Cammarota and G.Biroli, J. Chem. Phys. 138, 12A547 (2013)
[3] C.Cammarota, EPL 101, 56001 (2013)
[4] W.Kob and L.Berthier PRL 110, 245702 (2013)
[5] M.Ozawa, W.Kob, A.Ikeda and K.Miyazaki arXiv:1412.4911 to appear in Proc. Nat. Acad. Science (2015)
[6] C.J.Fullerton and R.Jack, PRL 112, 255701 (2014)
[7] S.Gokhale, K.H.Nagamanasa, R.Ganapathy and A.K.Sood, Nature Comm. 5, 4685 (2014)
[8] C.Cammarota and G.Biroli, EPL 98, 16011 (2012)
[9] S.Nandi, G.Biroli, J.P.Bouchaud, K.Miyazaki and D.R.Reichman, PRL 113, 245701 (2014)

Metastability, transitions in overlap and activity, and dynamic facilitation

Juan Garrahan
University of Nottingham

The cooperative and fluctuating dynamics of systems close to their glass transition can be traced back to the existence of configurations with atypically large relaxation times, the phenomenon we call metastability. These rare long-lived states can be found and explored by dynamical large deviation techniques, an approach which reveals a novel non-equilibrium transition between ergodic and non-ergodic phases, the relaxing "liquid" and the arrested "glass". The transition is dynamical because rare long-lived states are suppressed exponentially with system size in the statics, but this suppression is overcome when the time dimension is included: the transition occurs in ensembles of trajectories of the dynamics rather than in ensemble of configurations. This is in essentially the dynamical facilitation (DF) view of glasses. Still, metastable states can play a role in the thermodynamics. One can drive certain thermodynamic transitions by constraining the system, for example by coupling two copies of a system: under certain conditions the coupled system can undergo a transition from a phase where the copies are independent to one where they are highly overlapped. The limiting behaviour where the coupling is removed is an indicator of whether an ideal thermodynamic glass transition exists (as would be predicted by for example by the random first-order transition theory) or not. I will discuss how all these ideas relate to each other from the point of view of DF, and what the dynamical large-deviation "in silico" approach can tell us about glasses prepared by standard means.

Dynamical heterogeneity in a system of hard disks in a channel

Michael Godfrey and Michael Moore
University of Manchester, Manchester, United Kingdom

Disks confined to a narrow channel have features in common with the glassy behaviour of hard spheres in three dimensions. Caging of the disks sets in at a characteristic packing fraction, leading to a crossover from fragile- to strong-fluid behaviour. We have used event-driven molecular dynamics simulations to extract a time-dependent dynamical length scale from four-point correlation functions. This length scale increases with time and, on the equilibration time scale, is proportional to the static length scale for structural ordering in the system. We can identify the structural features responsible for this dynamical behaviour from the structure factor, which we have determined exactly by a transfer-matrix approach.

Identification and characterisation of the different structural length scales in glass-forming materials

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The identification and characterisation of the different structural length scales in glass-forming materials will be approached, largely from an experimental perspective. The emphasis will be on binary network glass-forming materials with the MX2 stoichiometry, where two length-scales manifest themselves at distances larger than the nearest-neighbour distance. One of these is associated with an intermediate range, and the other is associated with an extended range that can persist to nanometer distances. The network properties are influenced by a competition between the topological ordering on these length scales, which can be manipulated by changing the chemical identity of the atomic constituents or by varying state parameters such as the temperature and pressure [1,2,3]. In the case example of ZnCl2, the structure has been investigated from the boiling point to the glass [4, 5]. Here, the glass comprises a network of corner-sharing ZnCl4 tetrahedra in which the chloride ion packing fraction is close to that expected for a random close packing of hard spheres. The fragility of the melt is associated with large structural variability, e.g., there is an increase with temperature in the fraction of edge-sharing tetrahedral motifs, where the latter are common structural motifs in fragile MX2 glass-formers [6]. The correlation lengths associated with the intermediate and extended ranges are inversely proportional to temperature. The structural transformations on vitrification of the very fragile glass-forming system CaAl2O4 will also be considered [7]. Here, structural changes on multiple length scales occur on glass formation. Pressure can profoundly affect the structure and dynamics of network glass-forming materials. Here, the focus will be on prototypical oxides such as SiO2, GeO2 and B2O3 [8,9] where an analysis of the available information shows that the coordination number of network-forming structural motifs, which plays a key role in defining the topological ordering, can be rationalized in terms of the oxygen-packing fraction over an extensive pressure and temperature range [10]. The result is a structural map for predicting the likely regimes of topological change for a range of oxide materials. This information can be used to forecast when changes may occur to the transport properties and compressibility of, e.g., fluids in planetary interiors, and is a prerequisite for the preparation of new materials following the principles of rational design.

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Cage–jump motion reveals universal dynamics and non-universal structural features in glass forming liquids

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Glass-forming materials are characterized by an intermittent motion at the microscopic scale, since particles diffuse through rare jumps out of the cages formed by their neighbours. A puzzling issue concerns the relationship of this cage-jump motion with the macroscopic dynamics and with the material structure. In this talk, I'll show that cage-jumps allows to predict the macroscopic dynamics and structure-dynamics relations, in both simulations of different glass forming systems [1-3] and experiments of hard-sphere colloids [4]. First, I demonstrate that cage-jumps are the elementary relaxation events allowing for a Continuous Time Random Walk description of the single particle dynamics. This leads to a short time prediction of the diffusion constant [1,4] and to a measure of the time dependent distribution of single particle diffusivities $P(d,t)$ [2,3]. Approaching the deeply supercooled regime, $P(d,t)$ acquires a transient bimodal shape, before achieving the expected asymptotic Gaussian shape, with a variance to mean ratio which increases on cooling. The approach to the asymptotic distribution occurs on a timescale larger than the relaxation time, with the dynamic correlation length of diffusivities achieving the maximum on the same timescale. This is the first direct observation of the dynamic coexistence of two phases with different diffusion coefficients and provides a scenario complementary to that inspired by dynamic heterogeneities. In the second part of the talk, I show how the bimodal shape of $P(d,t)$ allows to identify two populations of mobile an immobile particles, without introducing any arbitrary threshold, so as to facilitate the study of structure-dynamics relationships. In particular, in 2d harmonic systems the dynamic phase coexistence is accompanied by a striking structural signatures: the distribution of the hexatic order parameter is also bimodal, the more the lower the temperature, and the overlap between the most ordered and the immobile particles increases on cooling [3]. However, the same structural counterpart is absent in other models, such as the standard Kob-Andersen LJ mixture, whereby more sophisticated order parameter might be relevant.

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Role of crystal structures and their frustration in the stability of a supercooled liquid

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We have studied different binary mixtures and have shown that although all of them have a global crystalline minima some undergoes crystallization and some are stable in their supercooled liquid state [1]. Our study suggest that when a global structure is a mixed crystal where a single species contributes to both the crystal form and where the two crystal forms have large difference in some order parameter related to that species then this induces frustration between the locally preferred structure and the global structure. This frustration is the origin of stability of the supercooled liquid state. We have further shown that the well known Kob-Anderson model which is known to be good glass former sits at the bottom of two V - shaped phase diagram - one against the size of the intermolecular distance of the two species and temperature and other being the composition of the larger species and temperature. These kinds of V shaped phase diagrams are known to induce stability against crystallization [2]. Next we have performed a calculation of the free energy barrier for crystallization for the different systems which are stable in the supercooled liquid domain. We show that the comparative study of the free energy barriers and the different crystal structure compatibility in the respective global crystalline minima provides us the origin and the stability of these systems [3].

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Limits of homogeneous crystallization in suspensions of charged colloidal spheres approaching monodisperse size distributions

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We have studied the homogeneous nucleation rate densities of charged colloidal spheres by optical experiments. Interpreting our results in terms of classical nucleation theory, we find that the extrapolated equilibrium reduced interfacial free energy increases with decreasing polydispersity. This suggested that there may be a state of an undercooled melt of monodisperse charged spheres, which will crystallize only very hesitantly. Inspired by this thought we investigated a sample of polydispersity index 0.001 (standard deviation divided by mean radius). This highly charged sample was prepared at volume fractions between 0.001 and 0.01 under thoroughly deionized conditions. It displays a liquid-like static structure factor with a peak value exceeding 4, as well as a finite shear rigidity of 0.04Pa. Preliminary measurements of the intermediate scattering function indicate a two step decay. We thus far are tempted to conclude, that this system forms an amorphous solid at very low volume fraction, rather than the expected polycrystalline bcc solid. Crystallization of such samples appears to be present only on the time scale of many hours to days. Our observations raise some interesting questions including: What is the nature of the observed amorphous state? Is it a Wigner-glass? How does this state nucleate crystals? Is the sample crystallization correlated to the observed alpha-relaxation in the amorphous state? For the time being there appear many more questions than answers and the only conclusive statement that can be made is that this observation is not explicable in terms of the kinetic glass transition observed for hard spheres, the formation of attractive glasses nor the frustration of crystalization observed for charged spheres at large volume fractions.

Strain (-rate) induced fragility transition in bulk metallic glasses

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Since the famous work of Austen Angell the fragility parameter is worldwide used to classify glass forming systems into "fragile" and "strong". More recently it is also used to describe very successfully glass forming ability in fast and slow quenched bulk metallic glasses. Here we discuss the fragility parameter on the basis of the interatomic potentials and demonstrate a fragility transition from strong to fragile by a variation of the local strain(-rate) using MD-simulations. The results will be discussed in terms of a universal Lindemann criterion for liquefying of a glass by thermal heating or/and mechanical excitation.

Cavity point-to-set correlations: recent advances

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The glassy slowdown is accompanied by the growth of the cavity point-to-set (PTS) correlation length, which is expected to diverge together with the relaxation time. Yet efficiently measuring and interpreting PTS correlations remains an open computational and theoretical challenge. In order to obtain PTS correlations, we have developed a numerical approach based on parallel tempering that couples cavities immersed in high-temperature and less-frustrated environments. This scheme is particularly efficient for small cavities, which are otherwise difficult to sample properly. We have also developed a model-free approach to extracting the PTS length from the cavity-size dependence of the PTS susceptibility, and a modified cavity PTS correlation for assessing the contribution of orientational correlations. These advances allow us to cleanly assess the role of PTS structural correlations on the dynamical slowdown.

Statistics of density fluctuations in supercooled viscous liquids

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It is well known that small length scale density fluctuations in normal homogeneous liquids obey Gaussian statistics over many orders of magnitude [1, 2]. This fact underlies successful theories of normal liquids [3]. Here, we examine the statistics of density fluctuations outside the realm of normal liquids, by going into the supercooled regime. We use enhanced sampling methods [4] to compute probability distributions of density fields, and we consider a range of length scales and amplitudes. We investigate of both a non-associated liquid, a networked liquid and two diatomic mixtures. Gaussian statistics of the collective density field persists to a remarkable extent. Deviations of from Gaussian statistics are found in small sub-volumes due to the formation of sub-critical crystallites.

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Tuning of second critical temperature in the s-w model gives insight into the ultrastable glass challenge, and supercooled water conundrum.

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Following Vasisht et al's identification of the second critical point (Tc2,Pc2) for liquid silicon in the Stillinger-Weber (S-W) model for silicon [1], we study the variation of Tc2,Pc2 with tetrahedral repulsion parameter in an extension of the earlier "potential tuning" study of this system [2]. We use the simple isochoric cooling approach (and "isochore crossing" diagnostic [3]) to identify the location of the second critical point as a function of the "tuning" or "tetrahedrality", parameter Î», and identify two phenomena of high interest content. The first is that the second critical point pressure Pc2, becomes less negative as Î» decreases from the silicon value (meaning the drive to high tetrahedrality is decreased) and reaches zero pressure at the same value of Î» as was found to mark the onset of glassforming ability in an earlier study of this tunable system [2]. The second is that, as Î» increases above the silicon value, and the Tc,2 (already at -0.6 GPa for silicon [1]), approaches the temperature of the liquid-gas spinodal, the behavior of the temperature of maximum density TMD (minimum pressure TMP) for an isochore, switches from the behavior seen in most current water pair potential models (locus of TMDs has a maximum [4]), to the behavior seen in empirical engineering multiparameter equations of state (EoS [5]) (and also by two parameter Speedy expansion EoS) for water. According to these latter, the locus of TMDs has no maximum, and the EoS has no second critical point, but rather has a line of liquid-liquid transitions, that promote low diffusivity (due to the rapid isochoric entropy loss) and consequent structural arrest on the computation time scale (evidenced by a flattening of the isochore to give crystal-like behavior). Some discussion of the likelihood of the empiricists being correct, based on observations made on water studied at large negative pressure [6], will be given.

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Length scales, soft modes and non-affine rearrangements in the inherent structures of supercooled liquids

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Spatial distributions of soft modes and non-affine rearrangements in the inherent structures of supercooled liquids provide useful insight into possible structural changes underlying the liquid to solid transition. The non-affine displacement (NAD) field resulting from a shear deformation applied to the inherent structures of a liquid changes qualitatively between the high temperature liquid and supercooled state. In addition to being very sensitive to temperature and strongly correlated with the inherent structure energy, the NAD field detects spatial correlations that increase in range upon lowering the temperature towards the supercooled regime, with features strongly reminiscent of critical phenomena. A direct connection with the dynamics of the supercooled liquid can be established: upon lowering temperature, the regions where structural relaxation tend to occur are statistically correlated to regions undergoing large NAD in the inherent structures. The origin of this correlation is, interestingly, in the sensitivity of the spatial distribution of the soft normal modes to the hierarchical structure of the potential energy landscape and hence to the spatial distribution of the dynamical heterogeneity. Nevertheless, when considering only the very softest modes, those are only weakly correlated with the dynamical heterogeneities in the supercooled liquid, while displaying higher statistical overlap with the NAD field, a feature reminiscent of the behavior of nonaffine deformations in amorphous solids, where the very softest modes identify the loci of plastic instabilities.

Evidence for a second order phase transition to an ideal glass from trajectory sampling

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Dynamical heterogeneities in a model glassformer are linked to the presence of locally favoured structures (LFS). With sampling dynamical observables in trajectory space, it is possible to highlight a non-equilibrium first order phase transition between LFS-rich and LFS-poor trajectories [1]. This can be projected into configuration space and interpeted as the coexistence of two amorphous phases in the presence of an external chemical potential bias. We present strong numerical evidence suggesting that, in the limit of zero bias, the LFS-poor and the LFS-rich phases become undistinguishable when approaching from above the Vogel-Fulcher-Tammann temperature, at which the relaxation times are customarily expected to diverge and lead to the Kauzmann scenario of the so-called "ideal glass". We additionally show that structural signs of the LFS-rich phase can be detected at higher temperatures, hidden in the inherent state potential energy landscape.

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Overlap fluctuations in glassy systems

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In mean-field theories of the glass transition [1], one considers the overlap between two copies of a system, which acts as an order parameter for the transition. These theories predict several kinds of phase transition that occur when the overlap is biased, by a field eps. Analysis of fluctuations of the overlap can reveal complex many-body correlations within the glass state. I will present measurements of overlap fluctuations in two glassy systems: a Lennard-Jones binary mixture and a three-dimensional spin plaquette model. In the Lennard-Jones model, we demonstrate overlap fluctuations consistent with the existence of such phase transitions [2], and we discuss their interpretation in terms of multiple metastable states, and the interfaces between them. I will also discuss the extent to which these phase transitions occur in the plaquette model [3].

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Role of structural ordering in slow dynamics and crystallization of a supercooled liquid

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Glass transition takes place if crystallization is avoided upon cooling or increasing density. However, the physical factors controlling the glass-forming ability and the nature of glass transition remain elusive. For hard disks and spheres, we systematically control the size polydispersity, which can be regarded as the strength of frustration effects on crystallization. We revealed that crystal-like bond orientational order grows in both size and lifetime when approaching the glass transition point [1-3]. Our study suggests an intriguing scenario that the strength of frustration controls both the glass-forming ability and the fragility of liquid. When the strength of frustration on crystallization is so strong that glassy order may no longer has a link to the symmetry of the crystal, but we speculate that it may still be associated with low local free-energy configurations. We show a direct correlation between structural order and slow mobility, indicating a link between static growing length and the diverging relaxation time [1-4]. Furthermore, we revealed that such structural order also plays a crucial role in crystal nucleation [1,5-7]: Crystallization is a process of the enhancement of spatial coherence of crystal-like bond orientational order and not driven by translational order at least in the nucleation stage. We also show that this scenario is not limited to liquids interacting with spherically symmetric interactions, but local ordering also plays an important role in vitrification and crystallization of water-type liquids [8]. These results clearly indicate that liquid-state theories at the two-body level may not be enough for the description of these phenomena and it is crucial to take into account many-body correlations.

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4 Posters

Connecting structure to free energy: affine-to-non-affine transition in flowing glasses

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The structure of glasses plays a crucial role in their relaxation and response to applied shear. Nevertheless, the complex interplay of structure, affine and non-affine displacements remains a crucial challenge in our understanding of the flow of glasses. Structural modifications during flow are central to the flow behaviour, but are difficult to interpret and resolve experimentally.In this talk, we connect the structure of the colloidal glass directly to its free energy: in hard-sphere systems, the structure provides a unique route to the free energy of the system. We explore this relation to follow the structure and free energy of the glass upon application of shear. We observe that the nonequilibrium free energy decreases due to complex interplay of non-affine displacements and dissipation. We measure this free energy directly in strained colloidal glasses, and use mean-field theory to relate it to affine and nonaffine displacements. Nonaffine displacements grow with applied shear due to shear-induced loss of structural connectivity. Our direct observations and mean-field model allow for the first time to disentangle the complex contributions of affine and nonaffine displacements and dissipation in the transient deformation of glasses.

Structural aspects of nearly jammed sheared suspensions

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Structural information of nearly jammed -- and sheared -- suspensions is extracted from the free volume distribution. The free volume of a particle is defined as the geometrically available space to the centre of the particle keeping its' neighbours fixed. At densities not far below close packing, the free-volume distribution has a strong delta peak at zero free volume and a power law tail for 5-10% particles (rattlers).Interestingly, we find that shearing, on average, destroys free volume by comparing with configurations of the same density but decompressed from jammed-packings. In fact, by shearing the structure approaches that of jammed-packings also by comparing the radial distribution function. In this context we further explore the role of rattlers in preserving the structure of sheared configurations due to weak thermalization.

Multiple reentrant glass transitions in confined hard-sphere glasses

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Glass forming liquids exhibit a rich phenomenology under confinement. This is often related to the effects arising from wall-fluid interactions. Here, we focus on the strong confinement limit (wall-to-wall separation of the order of a few particle diameters). As expected, the confined liquids microscopically relax differently than the bulk liquids. We report on a recently developed generalization of the mode-coupling theory to account for the slab geometry [1,2]. The theory considered symmetry-adapted modes and use generalized static structure factors as input. Hence the prediction of the non-equilibrium state diagram is direct manifestation of the structure

of the fluid in confinement. We find a multiple reentrantrant scenario driven by theinterplay of local packing and confinement [2]. In the presentation we discuss the general features of the theory and emphasizes the changes with respect to the standard mode-coupling theory of the glass transition. Motivated by this idea, we performed event driven molecular dynamics (EDMD) simulations of model polydisperse hard sphere liquids in a slit geometry. This strong confinement induces structural changes due to competing length scales, for example, a non-monotonic variation of the first diffraction peak with the wall separation [2]. For the distances investigated, the first diffraction peak is maximal for non-commensurate wall distance. As expected, this must have a strong impact on the dynamics of a liquid. Indeed, a non-monotonic dependence of the self diffusion coefficient D on plate separation is observed. By increasing the packing fraction, the self diffusion coefficient varies by a factor of 1000 upon variation of H at packing fraction 0.49. This dramatic amplification is one of our principle results [2]. We find that the self diffusivity exhibits power-law behaviour for a fixed wall distance, which is asymptotically predicted by the mode-coupling theory (MCT) [1,2]. The state diagram obtained from the simulation has been compared to the MCT calculation. The most prominent feature is the variation of the glass-transition line with a period comparable to the hard sphere average diameter. As a consequence, along such paths, first a transition from a confined liquid to a non-ergodic glass state occurs, followed by a melting to a fluid state upon further shrinking the dimension. In the second part of our work, we consider the experimentally more accessible confined geometry, i.e., a wedge-shaped channel. By transferring the slit geometry results using thermodynamic relations, we are able to predict that the above reentrant effect also persists here. This effect may find interesting applications in nano-technology, for example, fluids confined in a nano-scale roughness. To provide direct evidence, we performed a number of EDMD simulations of a polydisperse hard-sphere system in a wedge-shaped geometry with a very small tilt angle. These simulations clearly demonstrate that the self diffusion coefficient in a wedge exhibits variation as a function of the distance from the corner of the wedge. This strongly suggests that a liquid-glass phase coexistence may occur at a sufficiently high external pressure.

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Active microrheology in a colloidal glass: mode-coupling theory and molecular dynamics simulations

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Active microrheology, viz. driving a probe particle through a complex material, allows to resolve the spatio-temporal evolution of its nonlinear response and of the underlying structural processes. We use molecular dynamics simulations and mode-coupling theory (MCT) to study the dynamics of a tracer particle embedded in a glass-forming colloidal dispersion under the action of a constant force. On the MCT side, this work extends previous approaches [1, 2] to the regime of strong applied forces. We focus on the structure of the probe particle displacements, described by the probe van Hove correlation function in force direction. The long time limit of the van Hove function of a localized probe in a glassy host exhibits a Gaussian central part and a long exponential tail in force direction. The tail becomes more pronounced for larger forces. Two length scales emerge from this picture: the cage size (width of the Gaussian part) and the correlation length of the exponential tail. We relate this structure to the coexistence of two types of trajectories of probe particles: those localized inside the cage, contributing to the Gaussian part, and those probe particles that perform jumps to other cages, generating the tail [3]. In the delocalized regime, simulation and MCT results for the probe displacements indicate strong anisotropy in the probe dynamics. In the direction perpendicular to the force, the motion of the probe is diffusive, with diffusion enhanced by increasing the applied force. Parallel to the force,

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Residual stresses after cessation of shear flow

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We present results from Brownian-dynamics computer simulation for the nonequilibrium transient dynamics in a colloidal glass former after the cessation of shear flow. In the glass, persistent residual stresses are found that depend on the flow history. The partial decay of stresses from the steady state to this residual stress is governed by the previous shear rate. Using a glassy hard-disk system, we also link this macroscale dynamics to microscopic particle motion, monitoring the transient mean-squared displacement and radial distribution function measured during the relaxation from the steady state. A flow-induced second plateau is found in the mean-squared displacement at long times. For sufficiently small previous shear rates we observe a rich structural relaxation after the interruption of the flow.

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Shear modulus of glass-forming systems

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The static shear modulus G can be thought of as an order parameter distinguishing the liquid (G = 0) from the glass (solid, G > 0). There has been recent interest in determining the temperature (T) dependence of G when approaching the glass transition temperature (Tg) from below. Here we present simulation results for G(T) for binary glass-forming liquids in 2 and 3 dimensions. We compare various approaches, employed in the literature, to calculate G(T), such as the (equilibrium) stress-fluctuation formalism and non-equilibrium methods determining the response of the glass to an externally imposed stress or strain. Our results indicate that G(T) vanishes continuously as Tg is approached from below, and we can understand this behavior from the interplay of affine and non-affine particle displacements [1]. This is, however, in contrast to other works, suggesting that G jumps discontinuously at Tg. In some of these studies, G is identified with the plateau found at intermediate times for the shear-stress auto-correlation function [C(t)] in the canonical ensemble. It could be that this difference is related to the ensemble (constant strain versus constant stress) in which C(t) is determined. Some support for this speculation comes simulations for a network system with permanent crosslinks [2].

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Structural origins for 'avalanche' crystallisation in hard sphere glasses

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The crystallisation of hard spheres is one of the most important, fundamental phenomena in condensed matter physics. Yet, the dynamics of the process at high volume fraction (> 60%), where the structural rearrangements are significantly slowed down, is not yet fully understood. Recent molecular dynamics simulations by Sanz et al. [1] have revealed that a supercooled

hard-sphere liquid at high volume fraction devitrifies simultaneously with periods of sudden, large displacements (`avalanches') in a sub-population of particles, separated by long periods of metastability. Curiously, particles that undergo `avalanches' do not become part of the crystal nuclei themselves. It was claimed that they `mediate' the process and were predominantly stochastic in origin, with limited correlation with the local structural environment. However, this is significantly at odds with other glassy systems, where a structural signature is clearly correlated with dynamics [2]. Thus, through Brownian Dynamics simulation, we sought structural origins and changes associated with the avalanche phenomenon.

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Heterogeneous glassy dynamics near a random critical point

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The study of metastable states and the search for growing structural correlations are challenging tasks in glass science, which can be attacked using suitably coupled replicated systems in real glass models [1]. Given two configurations (x,y), it is useful to consider a system where the replica y is frozen and only the replica x can evolve under the influence of the other replica. The total system is described by the following Hamiltonian: $H_\varepsilon(x,y) = H(x) - \varepsilon Q(x,y)$, where ε is a coupling field and the overlap Q(x,y) between the two replicas is defined as $Q(x,y) = \Sigma_{i,k=1,N} w(x_i - y_k)$, where w(x) is a smooth function which evolves from one when the inter particle distance is small, to zero when it gets large. Therefore the overlap is of order one when the two copies are in a similar state and close to zero otherwise. In this context, some interesting properties of this system have recently been revealed. The most important one is the existence of a first-order liquid-to-glass phase transition at equilibrium [2,3], ending at a second order critical point. After defining an effective potential function as $V(Q) = -\frac{T}{N} \ln P(Q)$, where T is the temperature and P(Q) the probability distribution of the overlap, it has been shown that by increasing ε at constant T, the system presents two different phases. For small ε, replica x is very different from replica y, the overlap Q is very small. For larger ε, replica x is close to replica y and the overlap is large. These two situations are separated by a sharp discontinuity in the (ε,T) plane. These properties are directly reflected in the evolution of the effective potential V(Q). In this contribution [4] we study a 80:20 binary mixture of Lennard-Jones particles using the Kob-Andersen potential and implementing a Monte-Carlo dynamics of the Hamiltonian H_ε with a system size of N = 1000 particles. We carefully analyze the phase diagram in the plane (ε,T) both in the high-Q and in the low-Q phases, in order to characterize the dynamical behaviour and its glassy features in equilibrium conditions in the vicinity of the phase transition. We study in particular the dynamical slowing down related to the approach of the random critical point. We observe a massive change in the dynamical properties of the system when the phase transition is approached. This implies a dramatic slowing down of the mean-squared displacement and of the relaxation times of density correlation functions. We also focus our analysis on the vicinity of the random critical point, where we find a sudden growth of dynamic heterogeneity. Moreover, we analyse the behaviour of the temporal fluctuations of the microscopic order parameter, the global overlap Q, near the random critical point.

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Cluster-glass transition of ultrasoft-potential fluids

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Ultrasoft-potential fluids are model systems whose interaction is characterized by a finite replusion [1]. They are known to exhibit exotic and rich thermodynamic behavior such as cluster-crystal formation and reentrant phase boundaries. Surprisingly little is known about their

dynamical properties. (Notable exceptions are works by Coslovich et al. [2] and Ikeda and Miyazaki [3]). Here we study the slow dynamics of a fluid with a representative ultrasoft potential, a generalized Hertzian potential, v(r) = epsilon (1-r/sigma)^alpha for r.

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Molecular dynamics simulations of silicate and borate glasses and melts: structure, diffusion dynamics and vibrational properties

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Computer simulations of the model glass formers SiO2 and B2O3 are presented, using the techniques of classical MD simulations and quantum mechanical DFT calculations. The latter limits the system size to about 100 - 200 atoms. SiO2 and B2O3 are the two most important network formers for industrial applications of oxide glasses. Glass samples are generated by means of a quench from the melt with classical MD simulations and a subsequent structural relaxation with DFT forces. In addition, (a) full ab initio quench(es) are carried out with a significantly faster cooling rate. In principle, the structural properties are in good agreement to experimental results from neutron and X-ray scattering, in all cases. Regarding the model glass former B2O3, a new classical interaction potential is parametrized, based on the liquid trajectory of an ab initio MD run at 2300 K. In this course, the structural fitting routine of [1] is used, extended to include 3-body angular interactions which leads to a significantly improved agreement of the liquid properties of the classical MD and ab initio MD simulations. However, the generated glass structures, in all cases, show a significantly lower fraction of 3-membered boroxol rings as predicted by experimental results (f=60%-80%) and compared to an alternative model structure (f=75%) [2]. The largest value of f=15% is observed in the full ab initio quenches from 2300 K. In case of SiO2, the glass structures after the quantum mechanical relaxation are the basis for calculations of the linear thermal expansion coefficient, employing the quasi-harmonic approximation. This shows a change of sign and a temperature range of negative thermal expansion at low temperatures, in good agreement with experimental results. To my knowledge this has not been observed so far.

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Effects of dynamic heterogeneity and density scaling of molecular dynamics on the relationship among thermodynamic coefficients at the glass transition

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One of the strong evidences that the glass transition cannot be treated as a pure second order thermodynamic transition is the difference between the prediction of well-known Ehrenfest's equations, which determine the pressure coefficient of the glass transition temperature dTg/dp, and the value of dTg/dp obtained from experimental measurements. Furthermore the Progogine-Defay ratio, which is a combination of Ehrenfest's equations are not equal to unity for majority of glass formers, against expectations. A very hopeful alternative for estimating the pressure coefficient of the glass transition temperature and improving the Prigogine-Defay ratio are our new equations, which describe dTg/dp. The first one [1] is developed in the thermodynamic scaling regime, i.e., if structural relaxation times can be plotted on one master

curve (independently of thermodynamic conditions) as a function of TV^{γ} (γ is a material constant interpreted as m/3 where m is the exponent of the repulsive part of effective intermolecular potential). The second one employs the concept of the dynamic heterogeneity and their approximations according to which the degree of the dynamic heterogeneity triggered by enthalpy fluctuations is equal to a good approximation to the sum of the degrees of the dynamic heterogeneity triggered by temperature and volume fluctuations [2]. We show that the values of dTg/dp from our new equations are consistent with those estimated from experimental Tg(p) and the Prigogine-Defay ratio which results from our new equations is equal to unity to a very good approximation. It is worth noting that our relations, which link dTg/dp with the exponent γ, reflect the relative role of thermal activation (γ → 0) and free volume (γ → ∞) in molecular dynamics near the glass transition, therefore it takes part in the discussion on a single- or two-order-parameter model of glass-forming liquids [4,5] and shows when single-order-parameter models can be used.

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Adam-gibbs model in the density scaling regime and its implications for the configurational entropy scaling.

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To solve a long-standing problem of condensed matter physics with determining a proper description of the thermodynamic evolution of the time scale of molecular dynamics near the glass transition, we extend the well-known Adam-Gibbs model to describe the temperature-volume dependence of structural relaxation times, $\tau_\alpha(T,V)$. We employ the thermodynamic scaling idea reflected in the density scaling power law, $\tau_\alpha = f(T^{-1}V^{-\gamma})$, recently acknowledged as a valid unifying concept in the glass transition physics, to discriminate between physically relevant and irrelevant attempts at formulating the temperature-volume representations of the Adam-Gibbs model. As a consequence, we determine a straightforward relation between the structural relaxation time τ_α and the configurational entropy S_c, giving evidence that also $S_c(T,V) = g(T^{-1}V^{-\gamma})$ with the exponent γ that enables to scale $\tau_\alpha(T,V)$. This important finding has meaningful implications for the linkage between thermodynamics and molecular dynamics near the glass transition, because it implies that τ_α can be scaled with S_c.

The structure of the phase space for polydisperse hard spheres

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We computer-generated monodisperse and polydisperse frictionless hard-sphere packings of 10,000 particles with log-normal particle diameter distributions in a wide range of volume packing densities φ (for monodisperse packings φ = 0.46-0.72). We equilibrated these packings and searched for their inherent structures. We analysed the dependence of inherent structure densities φ_{IS} vs. the initial packing densities φ. This analysis demonstrates that for monodisperse packings $\varphi_{IS} \approx 0.64$ corresponds to the J-point density [1] and $\varphi_{IS} \approx 0.65$ corresponds to the ideal glass density [2]. The ideal glass transition density for such packings is estimated from this analysis at φ ≈ 0.585, whereas the phase space demonstrates a change in its structure from "liquid-like" to "glassy" at φ ≈ 0.52. We measure the corresponding quantities for polydisperse packings as well.

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Dynamics and structure in a toy model

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We investigate the slow dynamics and the associated length scales in a model system at equilibrium. The latter consists in dilute 2-levels systems formed by a particle in a double-well potential, which are linearly coupled to a lattice of harmonic oscillators. At low temperature, the relaxation function is exponential with a characteristic time following an Arrhenius temperature dependence. On the other hand, dynamical heterogeneities, characterized by multi-points space-time correlation functions, develop as temperature decreases. This model allows us to clarify the difference between cooperativity and dynamical heterogeneity in the relaxation slowdown.

Active microrheology below the delocalization transition

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By active microrheology local viscoelastic properties of complex fluids can be probed. For this purpose, a strong external force is applied to a colloidal probe immersed in the test sample. From the resulting motion one can infer information about the local viscoelastic properties. For soft solids there is a delocalization transition when the force on the probe particle is large enough to pull it free [1]. We study the long time limit of the spatial probability distribution of a tracer particle in a glassy host as seen by active microrheology in constant force mode. The model system consists of hard spheres performing Brownian motion with one of them being actively pulled by a constant external force. Mode-coupling theory is used to obtain the spatial probability distribution, refining the previous model [2] by decomposing the mobility-tensor kernel as suggested by [3]. This procedure allows us to approach larger forces, while maintaining physically meaningful results. Already below the critical force there show up highly nonlinear effects, for example in mean and mean square displacements. One reason is the emergence of exponentially decaying tails of the probability distribution in both directions, perpendicular and parallel to the applied force. This feature can also be found in molecular dynamic computer simulations. We interpret these tails as average of hopping and caging events of the tracer particle.

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Hard spheres dynamics in high dimension

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Glass is made cooling a liquid down to some transition temperature where a dynamical arrest of particles motion occurs. This is due to a dramatic viscosity increase, related to a similar increase of a relaxation time of the correlations of the system. The hallmark of this dynamics is the emergence of a plateau on mean square displacements of the particles. When the dynamical transition temperature TD is reached, the relaxation time diverges and the system becomes stuck in an amorphous state. Unfortunately, in spite of considerable experimental, numerical and theoretical efforts, a theory of this transition from first principles is still lacking. The mode coupling theory (MCT) is one such example, but relies on an approximation, which cannot be refined systematically. Recently, breakthroughs about the thermodynamics of hard

spheres glasses in infinite spatial dimensions have been obtained [1] and applied to the jamming transition, deep in the glass phase, through replica techniques, as was done for spin glasses 30 years ago. The aim of the present work is to build the dynamical aspect of the problem, needed to understand purely dynamical phenomena such as the increase of viscosity. The computation of the exact MCT-like kernel would tell if MCT is the correct dynamical theory in high dimension. Several dynamical quantities can then be computed exactly. A solution in infinite dimension would be a basis to understand 3D glasses. Besides, qualitative but also quantitative agreement with 3D glasses is expected for several key quantities, as a weak dependence upon dimension is seen in simulations [2].

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Defect dynamics in the network glass SiO$_2$

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We study the dynamics of the strong glass former SiO$_2$ via molecular dynamics simulations below the glass transition temperature.T$_o$ focus on microscopic processes, we average single particle trajectories over time windows of about 100 particle oscillations. The structure on this coarse-grained time scale is very well defined in terms of coordination numbers, allowing us to identify ill-coordinated atoms, called defects in the following. The most numerous defects are O-O neighbors, whose lifetimes are comparable to the equilibration time at low temperature. On the other hand SiO and OSi defects are very rare and short lived. The lifetime of defects is found to be strongly temperature dependent, consistent with activated processes. Single-particle jumps give rise to local structural rearrangements. We show that in SiO$_2$ these structural rearrangements are coupled to the creation or annihilation of defects, giving rise to very strong correlations of jumping atoms and defects [1].

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Rounding of the localization transition in model crowded media

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Transport in dense environment can be significantly suppressed and even become anomalous. Here we investigate obstructed transport in two dimensions by computer simulation for increasingly complex frozen host structures to test if the predictions of the localization transition in the Lorentz model apply. We introduce hard-core correlations in the cherry-pit model, and employ a soft potential model for realistic interactions of a frozen liquid. We observe anomalous transport of an ideal gas of tracer particles in these structures up to several decades in time. This and the suppression of diffusion is attributed to an underlying percolation transition. The rounding of the transition observed in the soft potential model can be understood as an average over the tracers -- each with its own critical density.

Glasses of binary colloidal mixtures: structure and dynamics under shear

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We investigate the glass state of binary colloidal hard sphere mixtures with large size asymmetry (size ratio 1:5). Increasing the fraction of small spheres, a transition in the caging is

observed; the large particles, initially caged by the large spheres, become localised in a cage of small spheres [1]. During this transition, the dynamics accelerate and a reduction of the yield strain, partly as a result of the shift of random close packing, is observed [2]. Similarly, the one-component glass melts at a free volume, i.e., distance to random close packing, while the binary mixtures at the same free volume are still in a weak but solid-like state with a small finite yield strain. We performed step rate experiments and compared the rheological results to the particle dynamics [3]. The transition in caging affects the shear-induced cage constriction. It closely follows changes in the magnitude of the stress overshoot. We found that yielding is dominated by the short-time in-cage dynamics. In contrast, the storage of stress is controlled by the dynamics that is observed on the time scale of the long-time structural relaxation time of the caging species.

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Evolution of metastable glassy states in mean field hard-spheres: a replica approach

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We consider the adiabatic evolution of glassy states under external perturbations. Although the formalism we useis very general, we focus here on mean-field hard spheres where an exact analysis is possible. We consider perturbationsof the boundary, i.e. compression or (volume preserving) shear-strain, and we compute the response of glassy states to suchperturbations. We also study the stability of the glass basins towards breaking into sub-basins, corresponding to a Gardner transition, and we present results for the regime beyond the Gardner Point.

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Geometrical effects in the crystallization of supercooled quantum binary liquid mixtures

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The study of crystallization in supercooled liquids is important not only for understanding the fundamental mechanisms of crystal growth, but also to investigate the nature of the glass transition, which poses a serious challenge to both theory and experiments. Indeed, it is increasingly believed that the rates of crystal growth may represent useful probes of the glass-

forming ability of super-cooled liquids. The simplest systems that can be used as test beds for our current microscopic under-standing of the behavior of supercooled liquids are binary mixtures. Liquid binary mixtures are generally harder to crystallize compared to pure fluids, showing a very rich, though not fully understood behavior in dependence of particle size ratio and composition. For example, understanding why for some compositions certain binary metallic mixtures invariably form crystal phases that compete with glass formation in terms of stability is an open and fundamental question for understanding both crystallization and its interplay with glass-forming ability. While the effects of mixing have been widely investigated by classical molecular dynamics simulations of particles interacting through a simple Lennard-Jones (LJ) potential, comparable experimental studies have so far remained out of reach. Here we present experimental data on the crystallization kinetics of supercooled binary mixtures of parahydrogen, orthodeuterium, and neon obtained by employing the liquid microjet technique [1]. The mixtures are essentially nearly isotopic as the interparticle interactions can be approximated by the same LJ potential. We show that starting with a pure substance (either parahydrogen or orthodeuterium) the crystallization slows down considerably by increasing the amount of the impurity species, the effect being at strongest for the parahydrogen-neon mixture. The observed surprising behavior has its origin in the quantum effects that play a major role in the unique binary systems: Quantum delocalization due to zero-point motion tends to increase the effective radius of the molecules, yet the strength of this effect differs for the three species due to their different masses, resulting in an effective particle size ratio different from unity [2]. Our experimental results thus support a strong correlation between dense packing and slower crystallization kinetics, offering in particular a suggestive trend: the magnitude of the observed reduction in the crystal growth rate upon mixing appears to be directly linked to the particle size ratio for the investigated mixtures.

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Topological cluster classification of colloid in liquid-glass transition regime.

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We conducted experiments to understand of the hard sphere colloidal liquid-glass transition with an emphasis on experimental structure observations. As hard sphere colloids we used polydisperse of fluorescently dyed sterically stabilized dispersions of polymethylmeththacrylate (PMMA) particles in index and density matched solvent mixtures [1]. The volume fraction of colloids in liquid-glass transition regime was treated as the control parameter. Real space coordinates of particles were obtained by tracking particle from video of confocal microscopy. Then the structure was analysed in terms of clusters using the topological cluster classification (TCC) [2-3]. This is followed by detailed analysis of structure in trajectories space which reveals the information into the mechanism for liquid-glass transition [4-6].

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Crystallization of sheared nearly hard-spheres

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Understanding crystallization pathways and kinetics is a long-standing challenge in condensed matter. Additionally, the effect of impurities or external fields is also matter of debate and misunderstanding. In this study, we look at the effect of a simple linear shear flow on the crystallization of nearly hard-spheres. By varying the density from low to high supersaturation,

we were able to follow the crossover from shear suppressed crystallization to an enhanced kinetics. We attribute this crossover to the vanishing nucleation barrier from an activated to a diffusive limited process.

Relative resolution: a hybrid formalism for fluid mixtures

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For over a decade, several frameworks have been formulated for connecting Fine-Grained (FG) and Coarse-Grained (CG) models, yet no strategy can guarantee optimality across state space for both structural and thermal behavior [1-2]. Here, we overcome this problem by combining molecular resolution via relative separation: Molecules interact by a FG potential if close to each other and by a CG potential if far from each other. By imposing energy conservation, we present an analytical expression for relating the two models. While connecting with other theories for liquids [3-4], we importantly retrieve in molecular simulations structural correlations and thermal properties of various fluids (e.g., a tetrachloromethane-thiophene mixture with a metastable cavity). This hybrid approach may be of much relevance in the study of glassy phenomena, which require correct and efficient modeling throughout the phase trajectory.

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