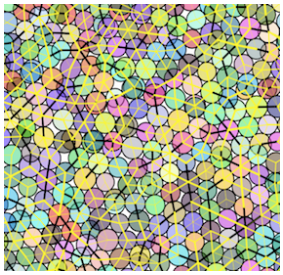

ID de Contribution: **26** Type: **Non spécifié**

Structure and dynamics of highly disordered systems

lundi 13 juin 2022 14:00 (1 heure)

1. Dynamic Aperiodic Matter (DAM)
   1.1 Glassy behavior of systems with strong disorder
   The properties of an isolated lattice defect depend strongly on the nature of the mother lattice. But as the defect density increases the interaction among them becomes more important than the interaction with lattice, and they start to assume their own identity and show different, emergent, properties. Often, they show behaviors similar to those of glasses. For instance, magnetic impurities show spin-glass behavior, and certain crystals with high twin density show strain-glass behavior. Ferroelectric solids with strong chemical disorder show relaxor ferroelectric behavior, very similar to that of spin-glass. Soft-matter, including biological matter, often shows viscous liquid/glass behavior. Such glassy behaviors are ubiquitous, but not fully understood because of complexity, in spite of long history of research. This represents a challenging broad field of research.
   1.2 Dynamic aperiodic matter; liquid, glass, and human society
   Liquids, gels, colloids, and polymers are called "soft-matter", but this naming is misleading. Softness is relative, and really does not catch the special nature of these materials which are outside the scope of the usual condensed-matter physics focused on crystals. A better name is "dynamic aperiodic matter (DAM)". DAM includes not only the conventional soft-matter, but also large objects such as human society. They are not random assembly of particles as in gas, but are condensed matter, held together by a cohesive force. The structure and dynamics of the system are highly cooperative with strong interparticle correlation. They key of understanding DAM is to determine and elucidate these dynamic correlations.
   1.3 Fluctuation-dissipation theorem
   The correlation functions that characterize the cooperative dynamics are directly related to the properties of the system through the fluctuation-dissipation theorem for linear response. The derivation of this theorem, an important milestone in statistical physics, is presented, and examples on diffusivity and viscosity are discussed.
2. Real-space characterization of DAM by scattering
   2.1 Pair-distribution function
   In characterizing the structure and dynamics of crystals through x-ray or neutron scattering, the reciprocal space is a natural arena because of the periodicity of the lattice. But it is not the most convenient place to describe the deviations, such as defects. Deviations from periodicity result in diffuse scattering, which requires extensive modeling to interpret. A more direct approach is to Fourier-transform the structure function, S(Q), including diffuse scattering (total scattering), to real space to obtain the pair-distribution function (PDF), g(r). The PDF tells you the distribution of atomic distances for both periodic and aperiodic structures. In crystals with chemical disorder the local structure is often quite different from the average structure. If a low-symmetry phase changes into a high-symmetry phase at high temperatures, the high-symmetry phase is usually a dynamic mosaic of low-symmetry local structure. Because the properties are usually determined by the local structure rather than the average structure the accurate knowledge of the local structure is important in elucidating the properties of the solid. Some technical details of the PDF method and examples of its application are discussed.
   2.2 Van Hove function
   The atomic structure of liquid is customarily described by the PDF. However, the term, "structure of liquid"is an oxymoron, because liquid is inherently dynamic and there is no elastic scattering from liquid. The PDF is the same-time correlation function, a thermal average of snapshots. This can be expanded to the two-time correlation function, the Van Hove function (VHF), G(r, t). The VHF is obtained

through the double-Fourier-transformation of the dynamic structure factor, S(Q, ω), where E = ⊠ω is the energy transfer in inelastic scattering. The VHF describes how the atomic correlation evolves with time. The decay of the VHF with time typically shows two steps. The first decay is due to vibrational motions (phonons), whereas the second decay describes the topological excitations due to the changes in the local topology of atomic connectivity. Technical details of the VHF measurement by inelastic x-ray/neutron scattering and examples of its application are discussed.

3. Understanding strong correlation

   3.1 Potential energy landscape

   In the absence of periodicity, it is difficult to describe the structure and dynamics of DAM in a meaningful way. A powerful approach is the potential energy landscape (PEL) concept. The PEL is the energy surface of a N-particle system in 3N dimensions, made of numerous valleys and hills. The PEL is so vast that it is difficult even to visualize it; it is usually hand-written. However, the relevant and accessible portion of the PEL is not so large, because the information transfer is limited in space and time by dynamic disorder. We discuss the local PEL of high-temperature liquid, and the nature of the saddle-point of the PEL where local configurational melting disrupts memory transfer.

   3.2 Nature of the glass transition

   The viscosity, η, of a typical liquid, such as water, is of the order of 10-2 poise (= 10-3 Pa.s). As temperature is reduced, if crystallization can be avoided by supercooling, viscosity increases rapidly by many orders of magnitude. The glass transition is defined by viscosity reaching 1013 poise, when the Maxwell relaxation time, τM = η/G∞, where G∞ is the high-frequency shear modulus, reaches of the order of a minute, so that the system behaves like a solid in the experimental time-scale. Why viscosity increases by as much as 15 orders of magnitude over a relatively small temperature range has been a major question in condensed-matter theory. We discuss various theories and speculations on the nature of the glass transition. A leading idea assumes there is a real phase transition, but the transition is frustrated and the system slows down into practical freezing before the critical temperature is reached.

   3.3 Medium-range order in liquid and glass

   The PDF of liquid and glass shows many peaks extending to an nm-scale. The first peak describes the short-range order (SRO) in the nearest neighbor atoms, whereas the peaks beyond the first peak depicts the medium-range order (MRO). However, they are different in nature. The SRO describes the atom-atom, or point-to-point correlation, while the MRO describes the correlation between the central atom and a group of atoms, point-to-set correlation. The MRO is produced by density waves induced by the interatomic potential. Currently many theories attempt to link the SRO, the structure of the near neighbors, directly to properties. However, atomic dynamics that controls viscosity and diffusivity in supercooled liquid is characterized by dynamic cooperativity. Evidence suggests that the MRO, which describes the cooperative dynamics, is the key quantity in atomic transport and the glass transition.

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