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Perspective

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Physics and chemistry perspectives on three unsolved problems in glass science

Jeppe C. Dyre[1] & Mark D. Ediger[2]

Abstract

The study of glasses and glass formation is a rich and technologically important area of research. This Perspective reviews recent progress in the field from the different points of view of chemistry and physics. At the heart of glass science are the super-Arrhenius temperature dependence of molecular relaxation processes and the nonexponential nature of relaxation functions. We discuss three questions motivated by these features and describe the progress of the past decade in answering them. The paper reviews experiments and simulations that probe growing and possibly diverging length scales associated with supercooling, the possibility of a thermodynamic transition to an ideal glass state underlying the glass transition, and new indications for universal relaxation functions describing molecular motion in deeply supercooled liquids. For each of these still largely unsolved problems, we illuminate the interplay between the universality often favoured by physicists and the specificity typical of the chemist approach.

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[1]Glass and Time, IMFUFA, Department of Science and Environment, Roskilde University, Roskilde, Denmark. [2]Department of Chemistry, University of Wisconsin-Madison, Madison, WI, USA. e-mail: dyre@ruc.dk; ediger@chem.wisc.edu

Introduction

Glass science can be reasonably placed somewhere in between physics and chemistry, each of which has its own way of thinking about what constitutes an important scientific problem. Although there is a continuous spectrum of approaches bridging the paradigms of purist physicists and chemists, as exemplified by the existence of major interdisciplinary journals such as *Journal of Chemical Physics* and *Journal of Physical Chemistry*, here we juxtapose the typical physicist and chemist approaches to glass science.

In reviewing important recent developments as the main purpose of the paper, we reflect on how physics and chemistry view three deep problems that have occupied scientists for years. A recurrent theme is that physics generally emphasizes universality whereas chemistry is more attuned to specificity. In this regard, theoretical physics and experimental chemistry differ most, with many experimental physicists and theoretical chemists placing themselves somewhere in between.

In this Perspective, we first give a brief overview of relevant features of the supercooled liquid that precedes the glass phase as one approaches the glass transition via cooling. Although almost all applications involve glasses and not the liquid phase[1], progress in understanding the glass-forming liquid impacts the development of glassy materials and their use. We then discuss progress on three questions:

- Do supercooled liquids have diverging length scales?
- Do supercooled liquids approach a state of zero configurational entropy at a finite temperature? If so, does the average relaxation time diverge at this temperature?
- Do supercooled liquids have an underlying universal linear response?

The physics and chemistry approaches to these questions differ substantially, as discussed below. Progress over the past 10–15 years from both sides attests to the creativity and innovation in the field of glasses.

Supercooled liquids

To set the stage, we remind the reader that below the melting temperature T_m, the equilibrium state of a chemically pure substance is a crystal. Although T_m is well defined thermodynamically, the barrier to forming a crystallite is very large just below T_m; in fact, it diverges as $T \to T_m$. As a consequence, even a very slowly cooled liquid experiences some degree of supercooling before it begins to crystallize. Liquids such as water and molten monatomic metals cannot be cooled far below T_m unless done extremely rapidly[2], but other liquids, such as most molten oxides and organic liquids, are difficult to crystallize and readily supercooled. At the glass transition temperature T_g, the structure of the supercooled liquid freezes because there is insufficient time to equilibrate[3–29]. There are several ways to form glasses, but cooling is most common and our primary focus.

'Glass' is so common in everyday experience — and so different from liquids such as ambient water — that one might not immediately see any connection between the two. However, glass formation from a supercooled liquid is a universal phenomenon, whether the substance in question is bonded by van der Waals, covalent, metallic, ionic or hydrogen bonds. Glasses may also form from polymeric liquids, which, for simplicity, we also refer to as supercooled even when there is no known crystalline state.

Whereas the supercooled liquid phase is fully characterized by temperature and pressure — although it is thermodynamically metastable by having a Gibbs free energy larger than that of the crystalline state — the structure of a glass depends on how it was prepared. As a result, there are an infinite number of different glasses that can be made of a given substance, a fact that from a general perspective makes the scientific study of glasses more challenging than the study of supercooled liquids. In this Perspective, we focus on the metastable liquid phase above T_g, not the glass, but note that the past 10–15 years has brought tremendous progress also in the understanding, for example, of mechanical properties of glasses and other non-crystalline solids, soft matter, how glass formation relates to the jamming of granular media, and glassy behaviour of active matter and biological systems[30–43].

The average relaxation time τ of a liquid increases an astonishing 12–15 orders of magnitude upon cooling. Rarely does τ follow the Arrhenius expression $\tau(T) = \tau_0 \exp(\Delta E/k_B T)$, in which τ_0 is a microscopic time of order a picosecond or smaller, ΔE the (free) energy of activation, and k_B the Boltzmann constant. Although this expression by itself would imply a huge increase of τ upon cooling, for almost all supercooled liquids, one observes that ΔE increases with decreasing temperature. Explaining this 'super-Arrhenius' behaviour is a long-standing problem of glass science, and this challenge has motivated the first two of the three questions discussed in this Perspective.

The intriguing super-Arrhenius behaviour is one reason that glass science, a century-old field of study[44], keeps attracting attention from the physics and chemistry communities alike. Another stubborn problem is to explain the ubiquitous nonexponential time dependence of linear and nonlinear relaxation functions, which is addressed in our third question. If all glass-forming liquids had an Arrhenius $\tau(T)$ and all relaxation functions were exponential in time, glass science would only attract limited attention from the physics and chemistry basic-science communities.

An often-invoked explanation of the dramatic slowing down of liquids upon supercooling is that relaxation becomes increasingly cooperative, that is, that the constituents of a liquid need to move in a more and more coordinated manner as temperature is lowered. A crude analogy of such cooperative motion would be the difficulty in getting off a crowded subway — first, everyone near the door needs to reorganize or step out to create the needed pathway.

Length scales

Our first question is the following: Do supercooled liquids have diverging length scales? We define 'length scale' as follows. If some recognizable pattern extends over the length l, the system has a length scale that is at least l. An example is the electric potential of a charge in an electrolyte or a plasma, which decays with distance r as $\propto \exp(-r/l)/r$ (Yukawa or screened Coulomb potential). Another example is the spin–spin correlation length that diverges as the ferromagnetic phase transition is approached from the high-temperature side, such as in the 2D Ising model in which like-spin islands grow in size when temperature is lowered towards the transition temperature (Fig. 1a).

A large number of different correlation lengths can be identified for a glass-forming liquid. These may be classified into static and dynamic lengths[14,45]. Knowing the molecular interactions, a static correlation length can be extracted, in principle, from a single snapshot of all molecular positions[46,47]. Static correlation reflects some sort of order — of density, molecular orientation, elastic properties and so on[37,48–51] — and such lengths can be defined both in terms of pair and many-body correlations[50]. Conversely, a dynamic correlation length can only be identified by monitoring the molecules of a liquid over some time interval[52,53]. Almost all static and dynamic correlation

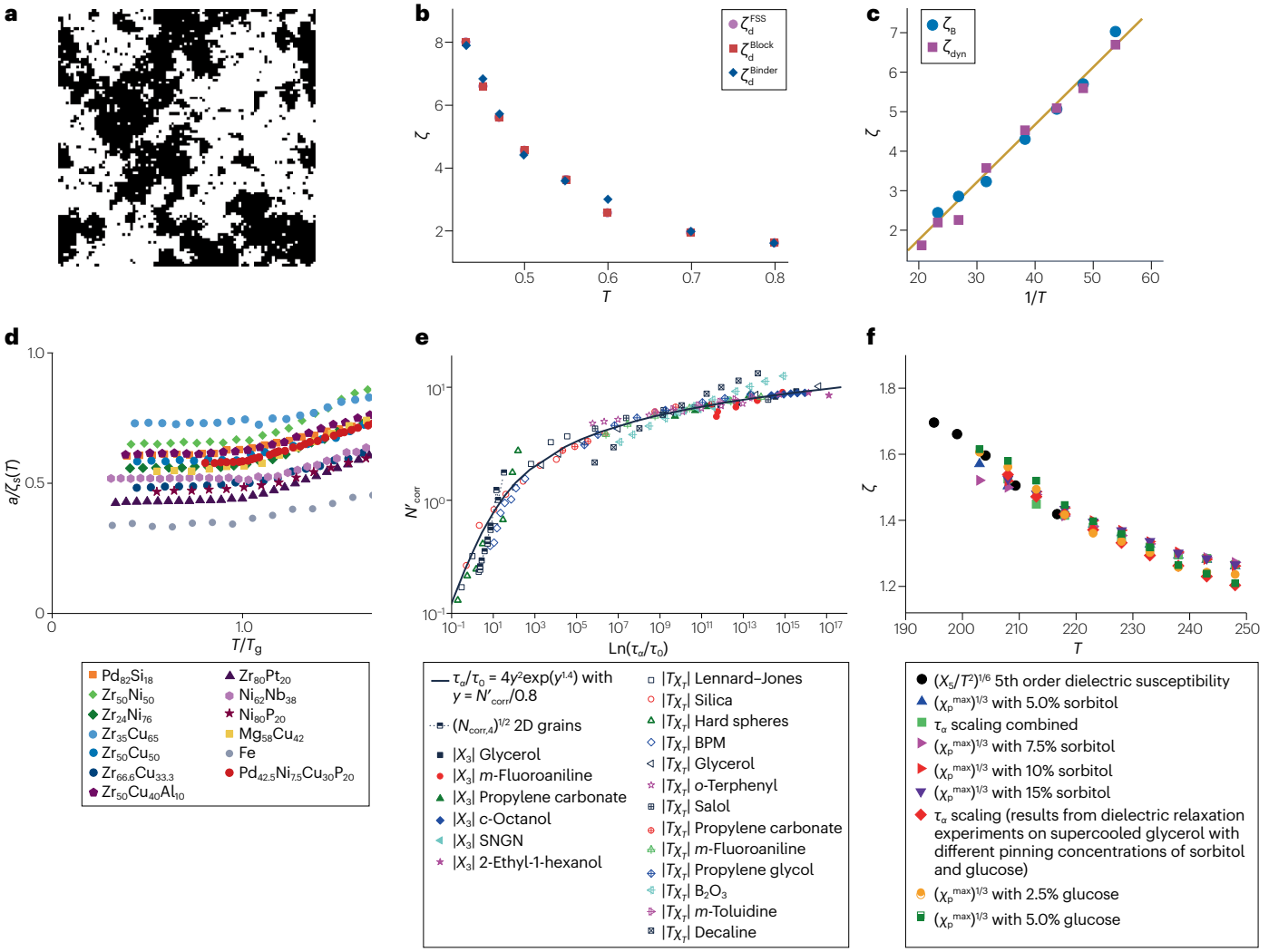

Fig. 1 | Correlation lengths derived from simulations and experiments, generally increasing as temperature is lowered. a–**c**, Simulation results. **a**, Snapshot of the 2D Ising model slightly above the phase transition, showing a large correlation length. **b**, Different dynamical correlation lengths ζ of the Kob–Andersen binary Lennard–Jones system as functions of temperature T. **c**, Static correlation length ζ_B and dynamic correlation length ζ_{dyn} as functions of inverse temperature of a 2D binary mixture of particles interacting through nonadditive potentials that scale with distance r as r^{-12}. **d**–**f**, Experimental results. **d**, Inverse correlation length a/ζ_s of metallic glasses as functions of temperature relative to the glass transition temperature T_g (derived from the static structure factor). **e**, Number of correlated molecules N'_{corr} as a function of the logarithm of the relaxation time τ_α derived from linear and nonlinear dielectric responses. **f**, Correlation lengths ζ of glycerol modified by sorbitol or glucose pinning agents as functions of temperature. Panel **b** is adapted with permission from ref. 230, ACS. Panel **c** is adapted from ref. 231, Springer Nature Limited. Panel **d** is adapted from ref. 232, CC BY 4.0. Panel **e** is adapted from ref. 29, CC BY 4.0. Panel **f** is adapted from ref. 233, CC BY 4.0.

lengths of glass-forming liquids increase upon cooling, although a few exceptions have been reported[54,55].

If the length scales are indeed diverging, an obvious follow-up question is whether the divergence is important for understanding supercooled liquids and the glass transition. Is the growing length scale the cause or merely a consequence of the slowing down[56,57]? We shall not discuss this interesting question in this Perspective, but focus on actual length-scale data of experiments and simulations. Before summarizing developments illuminating the question, however, we first highlight the generic chemist and physicist inclinations towards the existence of diverging length scales.

Physics and chemistry perspectives

The chemist generally says 'no' to large length scales. Chemical reaction rates are typically controlled by characteristics of the first shell of neighbours in a liquid, such as polarity and efficiency of local packing. For instance, the concept of 'biological water' indicates that water molecules directly adjacent to a protein can have distinct properties, whereas molecules further away than two monolayers are typically described as bulk water. There are many examples of very slow chemical reactions in molecules of modest size, in which case there is no large length scale, only a large activation barrier to be overcome.

By contrast, many physicists are open to the idea of a diverging length scale, hoping for a universal explanation of the glass transition[17,21,29,58]. A drive for universality has characterized physics ever since Isaac Newton, and the search for it received further emphasis from one of the important physics breakthroughs of the second half of the twentieth century: the understanding of critical phenomena (second-order phase transitions) as characterized by scaling and universality[59–61]. For instance, in the approach to ferromagnetic ordering upon cooling (Fig. 1a), the spin correlation length increases beyond bounds; in this case, the physics is universal because details of the spin interactions become irrelevant close to the transition temperature. If there is a universal theory of glass formation, such a theory must likewise be independent of the precise molecular interactions, and this is the case if there is a controlling length scale much larger than microscopic lengths[62].

The physics–chemistry difference relates also to the question at what point the liquid becomes a solid on cooling[63]. The view of the pragmatic chemist is that this is when the liquid looks solid in experiment, but many physicists regard that as an arbitrary definition, arguing instead that a solid is characterized by a diverging relaxation time corresponding to an energy barrier for flow that diverges in the thermodynamic limit[64]. Thus, the possibility of a growing length is not only the result of an aesthetic whim but can also be grounded in the substantive question of how solidity is defined.

A further difference to the focus of the chemist on the interactions between neighbouring molecules is the role of elasticity, a discipline that is absent in the standard chemistry curriculum. The fact that a glass-forming liquid approaching T_g acquires an extremely large viscosity means that long-ranged elastic interactions may become important[65–68], interactions that are not present in less viscous liquids.

Progress on the question

The concept of a length that diverges upon cooling is old[10,14,52], but the past 10–15 years has generated a much more complete picture. Figure 1 shows examples of different correlation lengths. Figure 1b shows dynamic correlation lengths of a binary Lennard–Jones system as functions of temperature. Figure 1c compares a static and a dynamic correlation length for a 2D system, and Fig. 1d shows experimental data for the inverse correlation length both above and below T_g for metallic glasses, in which case the correlation length is derived from the first peak of the structure factor. Figure 1e plots the number of 'correlated molecules' derived from linear and nonlinear dielectric measurements, a number that increases by a factor of 100 when the relaxation time increases by 18 decades. Finally, Fig. 1f shows correlation lengths derived from dielectric measurements on glycerol with different pinning agents.

All these observations could be interpreted as the start of a diverging length scale. It is not a very impressive start, though, typically reaching only 5–8 molecular diameters at the lowest temperatures probed. In comparison, the Ising model spin correlation length diverges as $(T-T_c)^{-\nu}$ wherein $\nu = 1$ in 2D and $\nu = 0.63$ in 3D, and in simulations of this model, it is straightforward to generate data with correlation lengths that are orders of magnitude larger than the lattice spacing (Fig. 1a).

In contrast to the work described above, some investigations from the past decade have suggested that length scales of 100 nm or more can be important near the glass transition. One very useful experimental approach is to measure how a nearby interface perturbs dynamics and related properties[69–72], for instance, by using the emission properties of a probe molecule to determine how the 'local T_g' changes as a function of distance away from an interface between two glass-forming polymers[73]. Results for one of the systems investigated, with an interface between polystyrene and poly(*n*-butyl)methacrylate, are shown in Fig. 2a. The composition profile of the interface is sharp (dashed line) whereas the local-T_g profile is extremely broad. Thus, more than 200 nm away from the interface, the local T_g on the polystyrene side of the interface has not yet attained the value of bulk polystyrene. Acoustic measurements show that the modulus of such a bilayer sample also changes over more than 100 nm near the interface[74], a finding that supports the local-T_g measurements as there is a strong correlation between mechanical moduli and relaxation time[75,76]. Results similar to those shown in Fig. 2a have been obtained for several different pairs of polymeric glass formers[77], with the observation that the length scale over which the high-T_g polymer is perturbed is larger when in contact with a low-modulus polymer[78].

Building on these results and extending them beyond polymeric glass formers, it has been demonstrated that the density of a vapour-deposited molecular glass depends on the modulus of the underlying substrate[79] (Fig. 2b). In qualitative agreement with ref. 73, a low-modulus substrate can apparently increase mobility at the surface of the deposited glass, even when that surface is more than 100 nm away from the substrate. A related work shows that the translational diffusion of a probe molecule near T_g can be slowed by a factor of 5 by interfaces that are 50 nm distant[80].

However, the existence of very large (>100 nm) length scales near glassy interfaces is controversial. Many additional experimental approaches have been used to examine interfaces; some approaches yield results supportive of large length scales[81,82] whereas others might not be[54,83–85]. Computer simulations have addressed both physical ageing and the influence of interfaces on the dynamics[72,86–88]. These generally do not find large length scales, although this may be a result of the limited time and length scales accessible in simulations. Most experiments near interfaces involve the nonequilibrium glassy state whereas most simulations do not, and this difference may be a key factor[89]. Although the length scale of interfacial perturbation (Fig. 2) need not match the bulk correlation length (Fig. 1), the large difference between these length scales is not easily reconciled. Explaining this discrepancy is a key challenge for the future.

We finally note that simulations have advanced a percolation approach for explaining the mechanical properties of glasses in a reasoning that emphasizes the importance of the large-scale connectivity of the domains of mobility and of immobility[58,90]. This relates to the solidity length l_s (ref. 91), below which a glass-forming liquid behaves more like a 'solid that flows'[68] than as the less viscous liquids of conventional liquid-state theory[92,93]. We note that there is no agreement, however, on the exponent x relating l_s to the average relaxation time τ of the liquid in the scaling rule $l_s \propto \tau^x$, with proposals being made of $x = 1/2$ (ref. 94), $x = 1/3$ (ref. 95) or $x = 1/4$ (ref. 91). In all cases, however, the solidity length is at least 1 μm at the calorimetric glass transition. Note also that l_s diverges at a finite temperature only if τ does. This brings us to the next question that relates to a possibly diverging relaxation time.

The ideal glass

Classical questions in glass science are as follows: Why is the relaxation time super-Arrhenius and what controls its temperature dependence[28,96–98]? What is the role of entropy[99,100] in explaining the super-Arrhenius behaviour? Are the dynamics dominated by collective effects or by the local geometry[56,57,75,97,99–102]? We address these questions indirectly by enquiring into their connection to the fascinating concept of an ideal glass.

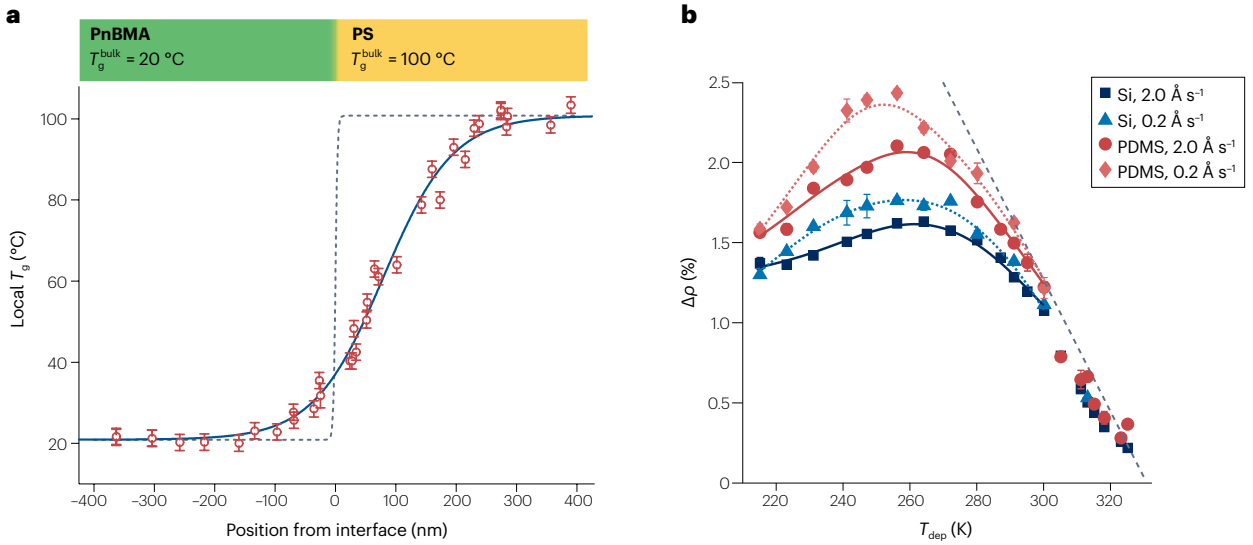

Fig. 2 | Experimental data indicating the potential role of a large length scale near the glass transition. a, Local glass transition temperature T_g near the interface between two polymer glass formers — poly(*n*-butyl)methacrylate) (PnBMA) and polystyrene (PS) — with the interface (composition profile indicated by the dashed line) affecting the local T_g over a range of ≈200 nm. **b**, The density of vapour-deposited glasses of the organic semiconductor TPD (*N*,*N*′-bis(3-methylphenyl)-*N*,*N*′-diphenylbenzidine) as a function of substrate temperature T_{dep}. Deposition onto soft substrates (red symbols) produces higher-density glasses than deposition onto hard substrates (blue symbols), for films of 200-nm thickness. PDMS, polydimethylsiloxane. Panel **a** is adapted with permission from ref. 73, AIP. Panel **b** is adapted from ref. 79, Springer Nature Limited.

The difference in the level of molecular organization between the crystalline and liquid states is quantified by the entropy of fusion $S_{fus} \equiv S_{liq} - S_{cryst} > 0$. Because entropy reflects the number of microscopic states, the liquid has more of these than the crystal, which makes sense in view of the disorder of the liquid. However, the supercooled liquid usually also has a notably larger heat capacity than the crystal, which implies that S_{fus} decreases substantially as the liquid is cooled below T_m. Thus, the number of molecular packings decreases upon cooling, which means that the liquid becomes increasingly organized as it is supercooled. For many liquids, S_{fus} extrapolates to zero well before $T = 0$ (refs. 3,44,103), at the so-called Kauzmann temperature T_K (Fig. 3a).

One can write the entropy of a glass-forming liquid as a sum of a configurational contribution, S_{conf}, reflecting the different ways the molecules may pack, and a vibrational contribution, S_{vib} (refs. 104–106). A perfect crystal has zero configurational entropy and typically a similar S_{vib} as the liquid. Thus, by extrapolation, $S_{conf} \cong S_{fus}$ becomes negative below T_K. This is not possible, however, because S_{conf} is proportional to the logarithm of the number of (appropriately defined) amorphous states[106]. Although the supercooled liquid could have a lower vibrational entropy than the crystal[107], this can hardly save the situation — something must happen around T_K to avoid the entropy crisis. No supercooled liquid state with $S_{conf} = 0$ has been reached experimentally because the liquid always falls out of equilibrium before reaching T_K, forming a glass.

A possible solution to the entropy crisis is that there is a second-order phase transition at T_K to an ideal glass defined as a state of zero (sub-extensive) configurational entropy. If this occurs, are there consequences for the dynamics above the transition? In the 1950s and 1960s, it was proposed that owing to the 'dearth of configurations' upon approaching T_K, the relaxation time of the supercooled liquid diverges as $T \to T_K$ (refs. 99,108). The compelling idea is that it is very difficult for a system to relax if there are only few states to transit into. Note that this proposal implies that the liquid must fall out of equilibrium before reaching T_K, no matter how slowly it is cooled. This thinking has dominated large parts of glass science for half a century, in particular in terms of the random first-order transition (RFOT) theory[100,109–113]. Our second question is whether this picture is correct.

Physics and chemistry perspectives

To many physicists, the concept of a phase transition underlying the glass transition provides an attractive scenario, in particular, because this typically implies universality as for critical phenomena. However, the concept of a unique amorphous state, an ideal glass, may be difficult to accept because such a state would a priori be expected to have a simple mathematical description[107].

The chemist trusts thermodynamics and statistical mechanics and does not dispute that liquids become more organized as they are increasingly supercooled. However, what happens by extrapolation is not an overwhelming concern unless the existence of an ideal glass influences properties wherein the supercooled liquid is accessible to experiment. The chemist might ask: If an ideal glass state exists, what unique properties can be achieved by approaching it? As for the idea of a relaxation time approaching infinity at a finite temperature, most chemists are suspicious. As an example, some organic molecules show substantial self-diffusion in the single-crystalline state[114]; therefore, an extremely low S_{conf} does not necessarily imply an extremely low mobility. Thus, at least the single-particle dynamics does not need to come to a halt in an ideal glass.

Progress on the question

We begin by highlighting an important theoretical breakthrough of the past 10–15 years: The glass-transition problem has been rigorously solved for systems of point particles in infinite dimensions. This ground-breaking achievement conforms to the RFOT scenario[100,109–113]. For more discussions on it, the reader is referred to refs. 110,111. As a

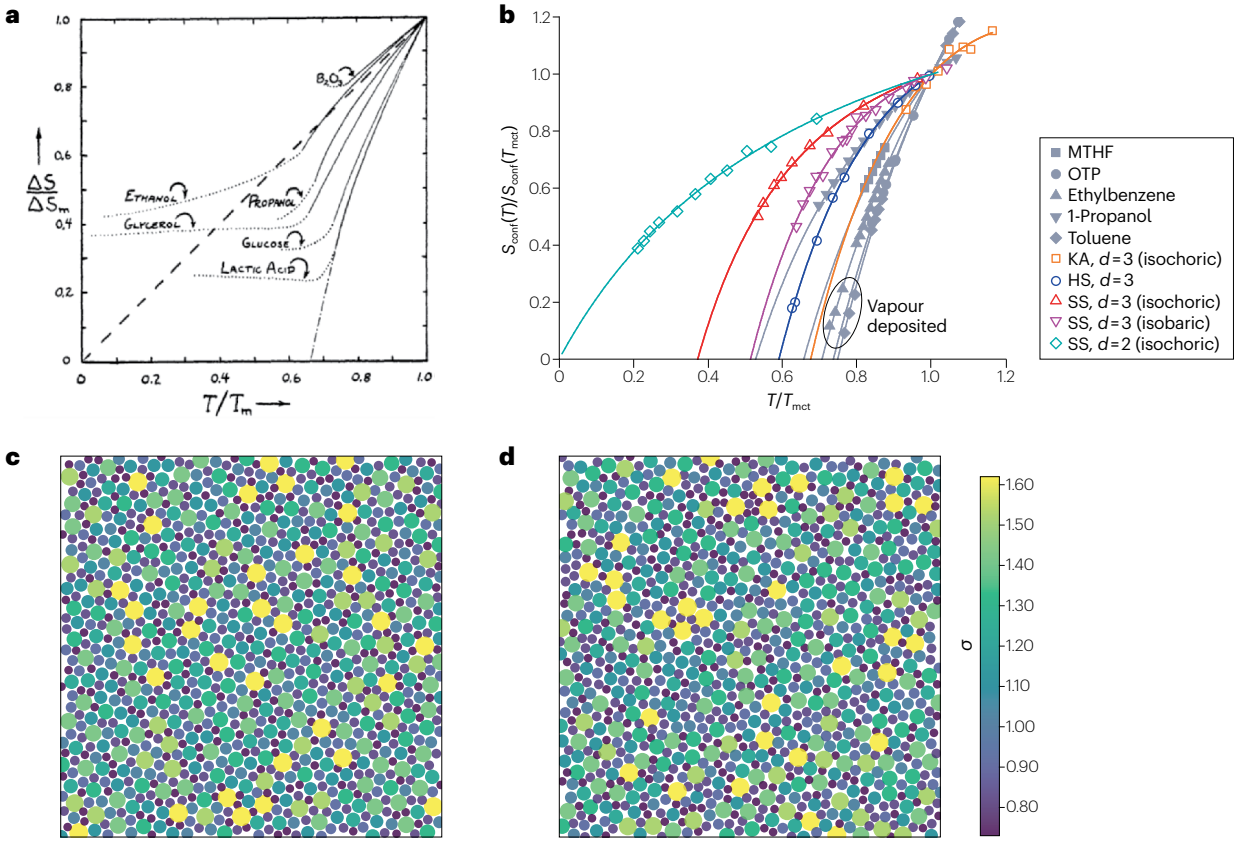

Fig. 3 | Probing the existence of an ideal glass state. a, A figure from Kauzmann's 1948 review showing that the entropy of fusion of the supercooled liquid (full curves) extrapolates to zero at a finite temperature; the dotted curves mark entropy differences between liquid and crystal calculated by integrating the measured specific heat below the glass transition. **b**, Compilation of numerical and experimental data for a normalized configurational entropy S_{conf}. The grey points show data for S_{conf} of several organic glass formers produced by cooling (upper points) and vapour deposition (lower points). The blue points show the configurational entropy of a swap-equilibrated polydisperse hard-sphere system. Swap brings one very close to the Kauzmann temperature at which $S_{conf} = 0$. **c**,**d**, Equilibrium configurations differing by a factor of 10^{12} in relaxation time, coloured according to the particle size σ. The difference in structure is subtle — panel **c** is the slow configuration. ΔS_m, entropy of fusion; HS, hard-sphere potential; KA, Kob–Anderson binary Lennard–Jones system; MTHF, 2-methyltetrahydrofuran; OTP, *ortho*-terphenyl; SS, soft-sphere potential (inverse power-law potential) model; T_m, melting temperature; T_{mct}, mode-coupling transition temperature. Panel **a** is reproduced with permission from ref. 3, ACS. Panel **b** is adapted with permission from ref. 106, AIP. Panel **c** and **d** images are courtesy of Anshul Parmar.

consequence, the two questions posed in this section can be answered affirmatively in infinite dimensions, at least for point-particle systems: the ideal glass does exist in infinite dimensions and the relaxation time does diverge as the ideal glass is approached upon cooling. We focus below on experimental and numerical data illuminating whether an ideal glass state also exists in 2D and 3D, in which the physics can be quite different.

In the past 20 years, it has become possible to produce the so-called ultrastable glasses by vapour deposition, with properties corresponding to systems aged for thousands of years[115–118] with utility, for example, for organic electronics[119]. This breakthrough, which takes advantage of the dynamics at the free interface being enhanced owing to fewer constraints on the molecular motion[120–123], has produced samples with properties much closer to T_K than those previously produced. Data for S_{conf} of several organic glass formers produced by cooling and vapour deposition do not show signs of a change of the temperature dependence of $S_{conf}(T)$ close to T_K (ref. 124) (Fig. 3b). These results do not mean that there must be a phase transition and that the ideal glass phase exists, but if not, S_{conf} needs to do something dramatic within a very narrow temperature range to avoid going negative.

In an equally important development, the past decade has witnessed a revolution in computer simulations by the introduction of the so-called swap dynamics[125–127]. By swapping non-identical particles every now and then during a simulation, it is possible to produce systems equilibrated at low temperatures, resulting in glasses corresponding to those produced in the laboratory at cooling rates of Kelvin per minute[128]. The configurational entropy of a swap-equilibrated polydisperse hard-sphere system indicates that swap brings one very close to T_K (blue symbols in Fig. 3b). A further illustration of the power of swap dynamics is provided in Fig. 3c,d, which shows two configurations of a 2D polydisperse system differing by 12 orders of magnitude in average relaxation time. This plot, incidentally, demonstrates that slowing down is associated with few obvious structural signatures

(try to guess which configuration is slower if you have not yet read the figure caption).

The entropy crisis is avoided if the temperature extrapolation breaks down and S_{conf} changes its temperature dependence dramatically[4,129,130]. Few experimental data support this possibility, but that it can happen has recently been demonstrated for a 2D model[131]. Here, results from simulations of small-system ternary mixtures of Lennard–Jones particles — obtained by combining swap dynamics with parallel tempering and population-annealing Monte Carlo algorithms — show that the Kauzmann temperature moves to zero as the system size is increased, whereas for very small system size, the entropy crisis is avoided because S_{conf} changes its temperature dependence before going negative. As mentioned, the entropy crisis would likewise be avoided if there is a phase transition at T_K to an 'ideal glass' defined as the amorphous state with the lowest position on the potential energy landscape[14,111,132,133]. Because the S_{conf} of the ideal glass is zero, the ideal glass state is basically unique, that is, it involves at most a subextensive number of configurations. Because of this, the approach to T_K has been associated with a diverging 'amorphous' correlation length[99,108,109,111].

In summary, experimental vapour deposition and computational advances have generated systems both in the laboratory and in simulations that are equilibrated much closer to T_K than previously thought possible, that is, with a very low configurational entropy. For small simulation systems in 2D, it is now possible to equilibrate at T_K and even near to $T = 0$ (ref. 131). In the next few years, a key challenge will be to perform analogous studies of larger systems and in 3D, which would enable important connections to real liquids and materials.

While awaiting further data on the existence of the ideal glass, one may ask: what is the structure of the proposed unique equilibrium state at T_K? No general mathematical description of it has been proposed, even for point-particle model glasses, which is a serious worry for the physicist. However, there has been a great deal of activity in identifying amorphous order in the form of structures that dominate as liquids are cooled, the so-called locally favoured structures[134–138]. The oldest and most famous locally preferred structure is the icosahedron often observed in the hard-sphere system[139–141]. This is the optimal packing of 12 identical spheres around one sphere, an arrangement that cannot be continued into a crystal (in contrast to the 2D case, compare compressing many identical coins on a table resulting in spontaneous crystallization into a hexagonal pattern). Figure 4a gives examples of locally favoured structures of different glass-forming binary liquids[135]. Another example is the so-called Frank–Kasper bond of six smaller spheres around two touching larger spheres, a structure that becomes increasingly common upon cooling a binary Lennard–Jones system (Fig. 4b).

If an ideal glass exists, it probably involves one or more of the locally favoured structures that dominate upon cooling. However, not all systems have a small number of locally favoured structures, as illustrated in Fig. 4c for simulations of the standard CuZr metallic glass. It is not clear how all these local structures could merge into a unique arrangement at T_K. Nevertheless, it is an obvious idea that long-range orientational order becomes important as glass-forming liquids are cooled[138,142]. It has also been shown that locally favoured structures lead to less frequent rearrangements[143]. A critical discussion of the utility of structural descriptors in general is given in ref. 49.

An alternate approach to learning about the structure of the ideal glass is to drive simulations non-thermally towards particular motifs to examine the properties of the resulting amorphous structures[141,144]. In recent years, efforts along these lines have optimized triangular packing or a steric order parameter in 2D[142,144]; in 3D, density fluctuations[145] or the local virial stresses[141] have been minimized to produce new packing arrangements. All of these efforts yield amorphous packing with very low energy and very high kinetic and mechanical stability, and thus produce structures that are candidates for the ideal glass. Figure 4d shows an ideal 2D structure produced by optimizing triangular packing. The algorithms achieve their impressive results by allowing the particle size distribution to change during structural optimization. Unfortunately, for laboratory glasses there is no analogue of particle size optimization, so it is possible that these motif-driven algorithms generate amorphous structures with characteristics that cannot be achieved in the hypothetical ideal glass state of a real system. Understanding the connection between structures produced in equilibrium — using swap dynamics, for example — and those produced by driving specific motifs is an important area for future work.

Although many physicists are content with studying the structure of point-particle glass formers — either by simulations or by experiments on metallic glasses or colloidal systems — chemists are far more interested in molecular systems. Locally favoured structures and a possible ideal glass state may look quite different for molecular systems, and this is a key area for future work. Recent efforts have provided a step in this direction by applying machine-learned potentials or swap dynamics to simulations of molecular systems[146–148].

Does the relaxation time of an equilibrium supercooled liquid diverge at the temperature of the ideal glass transition, as predicted by the Adam–Gibbs and RFOT approaches[99,109,113,149–155]? (The mode-coupling theory also predicts a finite-temperature divergence, but not at a temperature connected with the configurational entropy[156,157]). Answering this question inevitably requires extrapolation from the region in which equilibrium can be attained. That extrapolation is often done using the Vogel–Fulcher–Tammann (VFT) equation $\tau(T) \propto \exp(A/(T - T_0))$ from which is inferred a temperature of divergence T_0, but the VFT expression consistently overestimates how much the relaxation time increases upon cooling[158]. Indeed, a study involving 42 supercooled liquids has shown that dielectric data are fitted well by non-diverging functional forms[159].

How can one reconcile the evidence for an impending entropy crisis as the temperature is lowered with a lack of evidence for a corresponding relaxation-time divergence? Simulations on small 2D systems have indicated a transition from super-Arrhenius to Arrhenius dynamics — opposite to a divergence — when the ideal glass is approached[160], as predicted by trap models[161–163]. In this scenario, transitions occur below T_K via thermally accessible excited states, analogous to self-diffusion in crystals. A related possibility is that the dynamics are governed by local barriers with no cooperative effects[56,67,75,76,102,164–167], in which case the configurational entropy is irrelevant and proximity to T_K has no impact on relaxation times. A key challenge for future work will be to clarify whether or not, and in which sense, thermodynamics relates to dynamics.

Universal features of atomic and molecular motion

The dynamic properties of a glass-forming liquid are dictated by how the molecules of a liquid move about each other. Any universality of the latter must, therefore, be reflected in linear-response quantities such as dielectric relaxation data[168], dynamic shear and bulk moduli[21], and frequency-dependent specific heat, among others. Three classical experimental indications of possible universality of the dynamics are as follows:

1. The β process is an extra relaxation process at frequencies higher than those of the main (α) process, which is seen not only in polymers but also in liquids of rigid molecules[169]. Although many liquids do not exhibit a separate β process but merely a high-frequency tail, the consensus is that this tail derives from a β process that is partly merged with the α process[170,171] and only becomes clearly visible in the glass. Thus, the existence of a β relaxation channel may be universal for glass-forming liquids[172].
2. In glass-forming liquids, translational diffusion and other measures of mobility are decoupled. In comparison with either the viscosity or the α relaxation process, self-diffusion becomes 10–100 times 'too fast' as T_g is approached[173,174]. This observation is possibly universal. It is generally understood as an effect of dynamic heterogeneity, the observation that the molecular dynamics always varies notably in space, at any given time, with some regions being slow and others fast[52,175–179].
3. In the frequency domain, complex linear-response functions $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$ are often fitted by the Laplace transform of the stretched-exponential time-autocorrelation function $\exp(-(t/\tau)^{\beta})$, with $0 < \beta < 1$ (refs. 172,180,181). The significance of this function lies in its two limits: $\chi''(\omega) \propto \omega$ when $\omega\tau \ll 1$ and $\chi''(\omega) \propto \omega^{-\beta}$ when $\omega\tau \gg 1$. Thus, a stretched-exponential loss peak is asymmetric; it has the same low-frequency dependence as that of a simple exponential time-autocorrelation function, $\exp(-t/\tau)$, that leads to the Debye response function $\chi(\omega) \propto 1/(1 + i\omega\tau)$. An example of the universal $\chi''(\omega) \propto \omega$ low-frequency behaviour is given in Fig. 5a, which shows dynamic shear-modulus data of eleven glass formers.

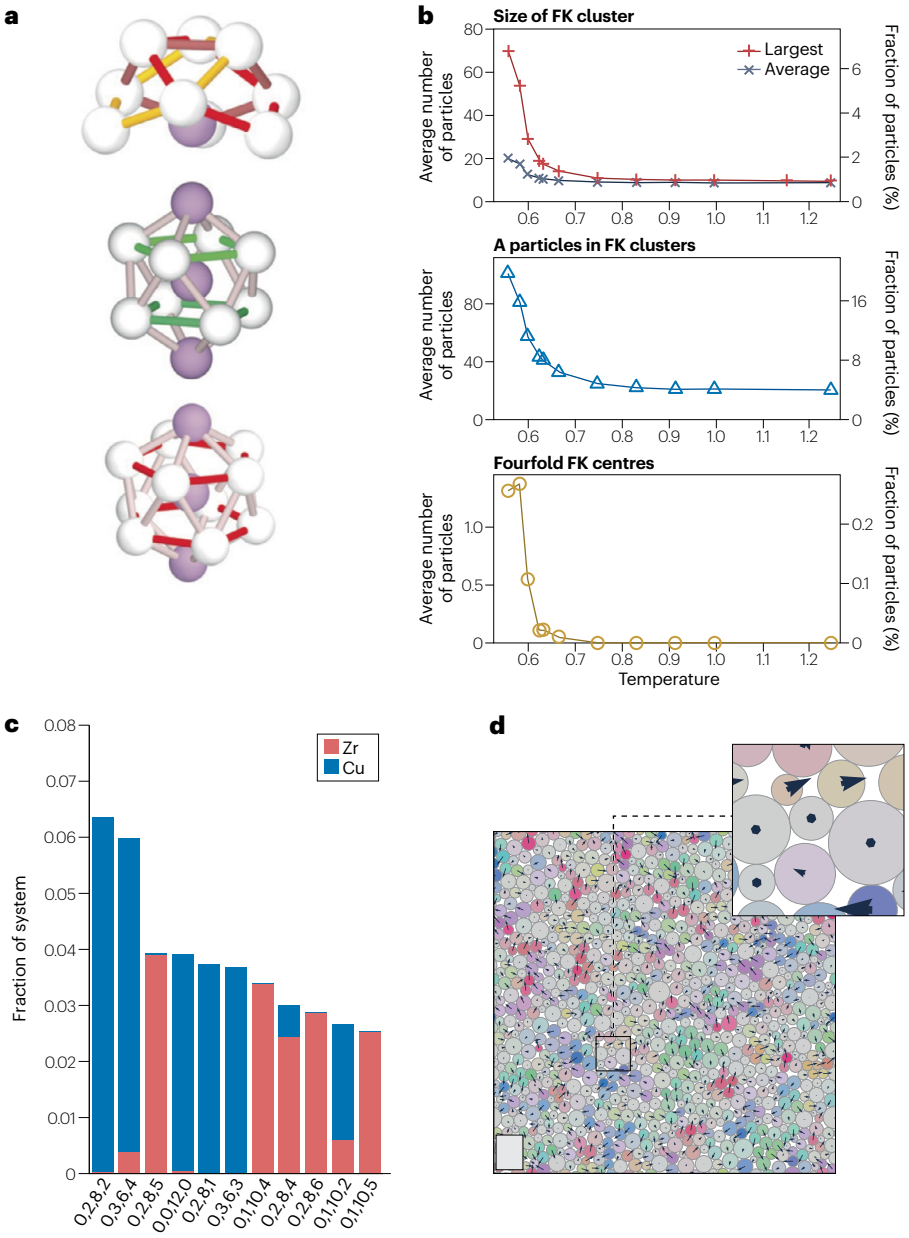

Fig. 4 | Locally preferred structures. a, Examples of popular structural motifs in glass-forming systems. The top motif is found in hard-sphere systems and colloidal gels, the middle motif (bicapped square antiprism) in the Kob–Andersen binary Lennard–Jones (LJ) liquid, and the bottom motif (icosahedron) in the Wahnström binary LJ liquid. **b**, Temperature dependence of the number of Frank–Kasper (FK) bonds in the Kob–Andersen binary LJ liquid. **c**, Fraction of atoms in a CuZr model in environments classified by the Voronoi descriptors (n_3, n_4, n_5, n_6) in which n_i is the number of facets with i edges. A multitude of structures is observed. **d**, Possible ideal glass packing of disks in 2D prepared by adjusting particle diameters. Panel **a** is adapted with permission from ref. 135, Elsevier. Panel **b** is adapted with permission from ref. 234, APS. Panel **c** is adapted from ref. 49, CC BY 4.0. Panel **d** image is courtesy of Viola Bolton-Lum.

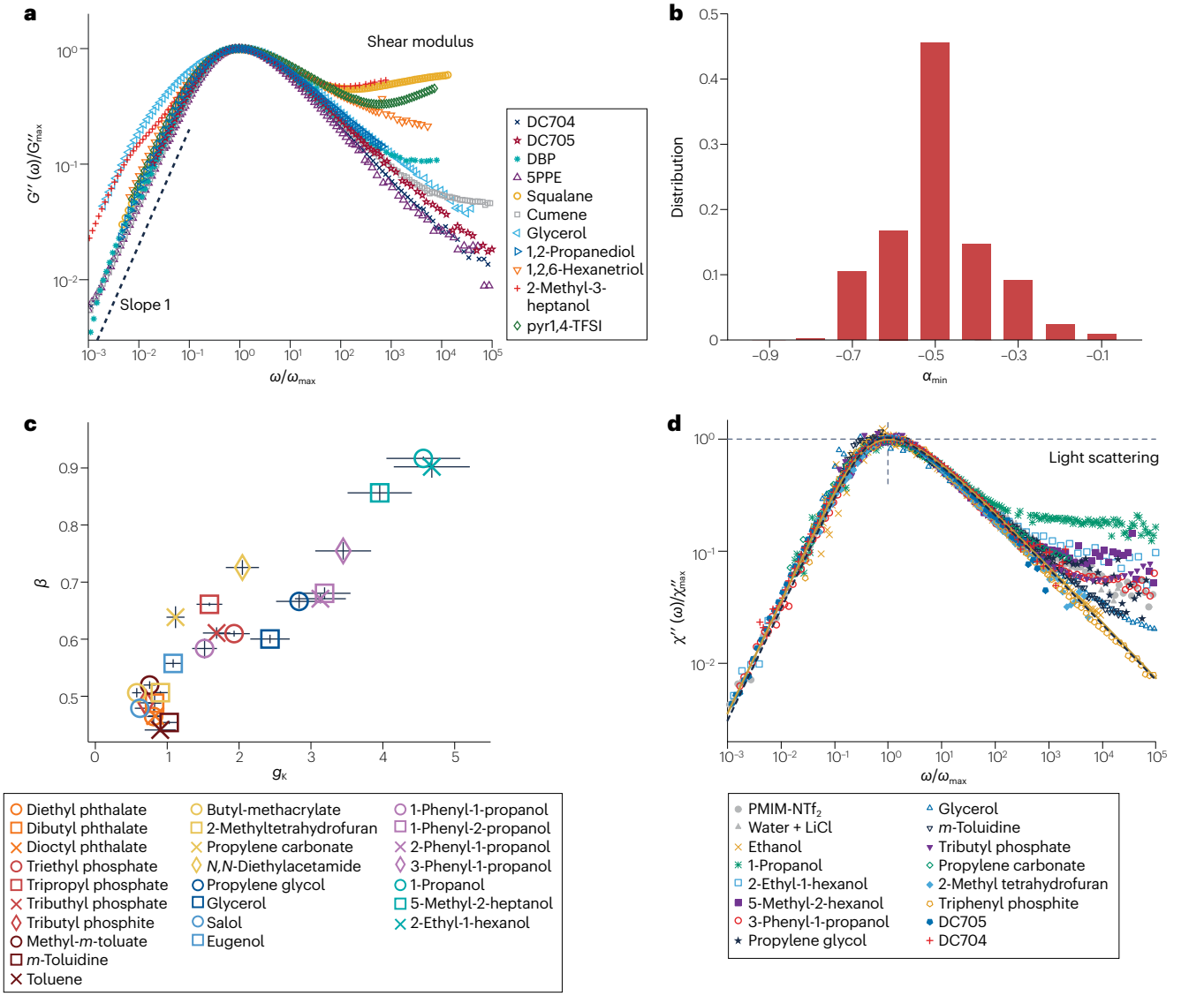

Fig. 5 | Linear-response data of glass-forming liquids. a, Log–log plot of the imaginary part of the dynamic shear modulus G'' of several molecular liquids, normalized to its maximum G''_{max}, plotted as a function of (angular) frequency ω normalized to the loss-peak frequency ω_{max}. All data have a low-frequency loss proportional to ω. **b**, Distribution of log–log plot minimal slopes above the loss peak, α_{min}, of 342 dielectric spectra. **c**, The β parameter of stretched exponential fits to dielectric data of 25 molecular liquids, plotted versus the Kirkwood correlation factor g_K. Whenever dipoles are uncorrelated ($g_K \cong 1$), β is close to 0.5. **d**, Dynamic light scattering data. The data conform to equation (1) as indicated by the yellow and black dashed lines of slope −1/2 above the loss-peak frequency; deviations at high frequencies are presumably owing to β processes. Panel **a** is adapted from ref. 186, CC BY-NC-ND 4.0. Panel **b** is adapted from ref. 201, CC BY 4.0. Panel **c** is adapted with permission from ref. 204, APS. Panel **d** is adapted with permission from ref. 205, ACS.

Our final question is as follows: Do supercooled liquids have an underlying universal linear response?

Physics and chemistry perspectives

The chemist emphasizes the variety among glass-forming systems: atomic systems (metallic alloys), molecular systems and covalently bonded systems (silica, organic polymers). The chemist sees weak, non-directional van der Waals attractions in some systems, stronger and more directional interactions, such as hydrogen bonding, in others, and very strong, highly directional covalent bonds in still others. Even in the subset of organic glass formers with only van der Waals interactions, there is a wide variety of molecular shapes (disks, rods, flexible chains). The chemist expects all this variety in interactions and molecular shapes to have a strong impact on molecular motion in supercooled liquids, and would therefore be astounded if generic features span all these systems.

As for the previous two questions, many physicists would be delighted if some form of dynamic universality describes very different glass-forming liquids. The physicist is encouraged in this regard by exact results for the dynamics of point-particle systems in infinite dimensions[182].

Progress on the question

Because a liquid is disordered, one expects a continuous distribution of energy barriers for molecular rearrangements. Apparently contradicting this, the general low-frequency behaviour of linear-response losses, $\chi''(\omega) \propto \omega$ – as shown in Fig. 5a for the dynamic shear modulus and in Fig. 5d for dynamic light scattering data – indicates a sharp cut-off in the barrier distribution at large barriers. This can be understood in terms of facilitation, a phenomenon that has recently come into renewed focus[88,171,183–186]: regions of low mobility can acquire mobility from neighbouring regions of higher mobility, in effect making barriers above a certain threshold irrelevant[68]. The ability of facilitation to relax low-energy amorphous states on distance scales as large as microns has been dramatically visualized[187,188].

In many early papers on glass-forming liquids – some from more than half a century ago – it was noted that linear-response losses at high frequencies often to a good approximation decay as

$$\chi''(\omega) \propto \omega^{-1/2} \quad (\omega\tau \gg 1). \tag{1}$$

This corresponds to $\beta = 1/2$ if a stretched exponential is used to fit the time-autocorrelation function (although that is not always a good fit). Over the years, several models have been proposed to explain equation (1), many of which were based on constituents or defects obeying the diffusion equation[189–200]; these older models are briefly summarized in ref. 199.

With the advent of modern frequency analysers, dielectric relaxation, with its high precision and very wide frequency range, became the gold standard for monitoring molecular motion in glass-forming liquids[168]. In 2009, an analysis of 347 dielectric spectra at different temperatures of 53 organic liquids (excluding monoalcohols) investigated how well equation (1) applies above the loss peak by identifying the effective decay exponent as the minimum slope α_{min}, that is, the slope at the inflexion point above the loss peak in a log–log plot of $\chi''(\omega)$ (ref. 201). The resulting α_{min} distribution is shown in Fig. 5b. Although there is a considerable spread at variance with equation (1), almost half of the spectra obey $-0.55 < \alpha_{min} < -0.45$. The conclusion at the time was that equation (1) is likely generic for the main (α) process. This was supported by the observations that the better time–temperature superposition applies, the closer α_{min} is to −0.5 (refs. 201,202), and that, furthermore, the α_{min} distribution narrows around −0.5 as temperature is lowered[201].

The past decade has brought much progress in elucidating equation (1) in relation to dielectric relaxation. In 2016, a correlation between the stretching exponent β and the dielectric strength of a liquid was demonstrated[203]. This was later confirmed by demonstrating that the high-frequency power-law exponent $\beta \equiv -\alpha_{min}$ obeys $\beta \to 0.5$ for $g_K \to 1$ (ref. 204) (Fig. 5d) in which g_K is the Kirkwood correlation factor, a quantity that is unity if the dipole vectors of different molecules are statistically uncorrelated, as expected, for example, for systems of small dielectric strength.

Further clarification of the situation came by comparing dielectric and dynamic light scattering (DLS) data[205]. Whereas the former show a sizeable spread of high-frequency exponents α_{min} (Fig. 5b), equation (1) is clearly visible in the DLS data on quite different glass-forming liquids (Fig. 5d). The deviations at high frequencies for some liquids are probably owing to β processes. Dynamic shear-modulus data present a similar picture (Fig. 5a).

Arguing that the cross-correlation terms in the time-autocorrelation function are insignificant for DLS but important for the dielectric relaxation[206,207], the new understanding is that orientational self-correlations give rise to equation (1) whereas cross-correlations are responsible for deviations from equation (1). The cross-correlations lead to a low-frequency Debye process, just as previously established for monoalcohols in which the slow Debye process completely dominates the dielectric response[204,208]. The cross-correlations are also visible on the low-frequency side of shear-modulus data of monoalcohols, explaining the lack of overlap on the low-frequency side of Fig. 5a.

These new results make a strong case for an underlying generic linear response in supercooled molecular liquids. There is evidence that this applies also for covalently bonded systems[209]. Important questions remain, however, including the temperature range over which the generic behaviour is observed[210]. It is important to note that even if there is an underlying universal linear response, knowing the generic shape is not enough to determine everything of relevance. For instance, the decoupling of translational motion from the α process increases progressively as temperature is lowered, so the degree of decoupling – which, as mentioned, reflects dynamic heterogeneity[52,175,176,178,211] – cannot be causally related to any generic linear response. Conversely, it is also worth noticing that the possible existence of an underlying universal linear response has consequences for glassy states because the standard Narayanaswamy material-time glass-ageing theory[212–215] is based on a linear-response convolution kernel of the equilibrium liquid[216].

An obvious way to test for equation (1) would be by computer simulations. Unfortunately, computers are not yet fast enough to cover the full dynamics of realistic molecular glass-forming liquids approaching the calorimetric glass transition[217,218]. Long simulations of the dielectric relaxation of *ortho*-terphenyl were fitted to a stretched exponential, resulting in an exponent β that decreased with decreasing temperature to approach 0.5, although with no sign of stabilizing at this value[219]. Simulations of polydisperse point particles have managed to go to an impressive 30 ms, with high-frequency linear-response losses $\propto \omega^{-\beta}$ in which $\beta \cong 0.45$ in 2D and $\beta \cong 0.38$ in 3D[126]. It is not clear how to reconcile these results with the generic experimental findings, and this is an important area for future work.

An alternative suggestion of a generic linear response of glass-forming liquids, which is not rigorously compatible with equation (1), focuses on the dynamic mechanical properties[220]. For a range of different liquids, the dynamic 'fluidity' (inverse complex frequency-dependent shear viscosity) is well described by the prediction of the random barrier model (RBM) that has no shape parameters. This finding is surprising because the RBM is highly idealized by modelling the hopping of non-interacting particles in a random landscape with identical energy minima[221,222]. The result has been confirmed by extensive computer simulations of a binary Lennard–Jones model[223]. Future simulations reaching longer times and exploring additional observables will hopefully shed light on this; it will also be important to test experimentally for RBM-type universality with additional liquids.

Relaxation phenomena can be described by replacing time with the distance travelled in configuration space[90,224,225]. Different measures have been proposed to quantify this distance: for example, the inherent structure minimal displacement, in terms of which temperature, pressure, ageing and other effects are unified via a power-law relation with the mechanical damping factor[224,225]. Yet another perspective is that a number of 'simple' behaviours constitute the essential features of many supercooled liquids[21]. Such behaviours include time–temperature superposition, density (thermodynamic) scaling and isochronal superposition[217,226] (the property that any two state points in the thermodynamic phase diagram with the same relaxation time have the same relaxation function). The latter behaviour, along with density scaling, has been identified both

in experiments and simulations, and it can be understood with isomorph theory: the properties of systems with isomorphs depend on just a single state variable, a fact that makes the 2D thermodynamic phase diagram effectively 1D[227,228]. Reference [21] suggests that addressing the origin of simple behaviours of supercooled liquids, rather than focusing on universality, may provide a fruitful path of reasoning.

The past decade has seen a rapid advance in the power of computer simulations, enabled by new equilibration algorithms and faster processors. In addition, machine learning is increasingly being used to rationalize glassy dynamics[43,137] and, in particular, to develop realistic force fields that describe laboratory systems of interest[147,229]. These developments will lead to closer connections between simulations and experiments, and we anticipate that such connections will be particularly important for evaluating whether — and to what extent — the linear response of supercooled liquids is universal.

Outlook

Glass-forming liquids are extremely diverse. Compared to the classical case of universality in critical phenomena, glass science is far more complex and confusing. It provides a perfect illustration of the latent 'physics-versus-chemistry' enigma of honouring a huge diversity while, at the same time, keeping an open mind towards potential universality[21,29].

Physics and chemistry are both indispensable for revealing how nature works. Having in mind the contrasting emphasis of the two disciplines, one is reminded about the statement of Niels Bohr that while the opposite of a simple truth is plainly wrong, the opposite of a deep truth is another deep truth. In our opinion, when it comes to glass science, universality and specificity both reveal deep truths.

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