

Multiple Time Scales in Amorphous Materials

Tina Hecksher

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Abstract Amorphous materials display dynamic properties on many time scales from picoseconds to millions of years and – depending on the time scale of observation – may appear solid-like or liquid-like or something in between. Measuring this enormous span in time scales is a monumental challenge for experimentalists, and often several techniques are involved in the attempt. Here, a pedestrian introduction to the topic is given and a personal view of the current status of experimental achievements, focusing on the possible generic relaxation shape and a model for relaxation time that connects short-time elastic properties to the long-time viscous flow.

Keywords Viscoelasticity • Glasses • Supercooled liquids • Relaxation • Spectroscopy

Mathematics Subject Classification (MSC2020) Primary 74-02, 93A15, 93C70 • Secondary 74-05, 78A02, 82D30

1 Introduction

Materials that we use to craft tools, furniture, and art, to build houses, instruments, and computers, are all made of densely packed molecules or atoms. The physics of studying materials and material properties is appropriately named *condensed matter* physics. Many materials – for instance,

T. Hecksher
Glass & Time, IMFUFA, Department of Science and Environment,
Roskilde University, Roskilde, Denmark
e-mail: tih@ruc.dk

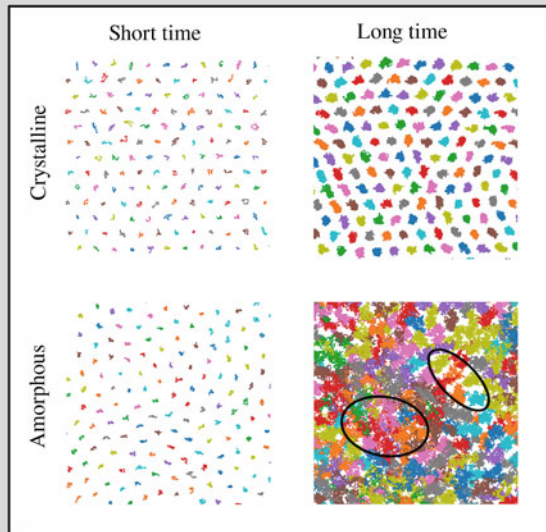
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most metals – are crystalline in their solid form, but a large group of materials are *amorphous*. Amorphous condensed matter embraces all materials that lack the long-ranged microscopic order and periodicity characteristic of crystals (see Box 1). Examples of such materials include most plastics and rubbers, glass in all forms, and some metallic alloys as well as materials with structure on intermediate length scales such as gels, emulsions, foams, colloidal suspensions, and granular materials. Amorphous materials are often easy to mold, and their properties may be tuned to a given purpose, e.g., precision optics [Vu et al. 2020], space application [Strock 1992; Rawal 2001], ultra-sharp surgical knives and medical implants [Meagher et al. 2016; Li and Zheng 2016], and environmentally friendly plastic materials for toys [Andersen et al. 2019]. In the industry, there is often a good engineering understanding of how to manipulate and mix compounds to obtain the desired properties of the end product. But a fundamental understanding of how the macroscopic properties are linked to the molecular interactions is still lacking.

Box 1 Microscopic Structure and Dynamics

Amorphous and crystalline materials differ in both their microscopic structure and dynamics. The crystal displays long-range order, while the amorphous material is disordered on the microscopic scale. Since tools to study real-time motion of molecules have not yet been developed, we turn to computer simulations to illustrate these differences.



(continued)

Box 1 (continued)

The figure shows trajectories of individual particles from a computer simulation of a two-dimensional model system [Booss-Bavnbek et al. 2023, Sec. 3]. The model emulates the interaction between molecules, and the simulation solves the equations of motion for a large system of particles and maps the position of all particles as a function time. The left-hand side of the figure shows trajectories of the particle movements in a short period of time, while the right-hand side shows the trajectories for a long period of time. Top panel shows a perfect crystal where all particles line up in a honeycomb pattern and the particles stay located except for vibrations around their equilibrium positions. The only difference between short and long times is the amount of vibrational motion observed. The bottom panel shows an amorphous state. At short times, it looks very much like the crystal: the dynamics consists of vibrations of each particle. Even the structure resembles the crystal in the sense that the average distance between particles is roughly the same. However, there is no long-range order – the system is disordered. At long times, the difference between the amorphous and crystalline materials is striking: Some particles stay more or less in the same position for the duration of the simulation, but other particles start to diffuse, and thus the trajectories begin to cross each other and the colors get smeared out (for an example of both stationary and non-stationary particles, see the two marked areas in the bottom right).

The crystal in this simulation is a so-called single crystal. Single crystals are perfect crystals with no defects and are extremely rare in nature (and in production). Most crystals are polycrystalline, which means that they consist of domains that have perfect order, but the different domains do not line up.

Glasses are a subset of amorphous materials. Hard candy is an everyday example of a glass; in fact, most sugars are good glass formers. A less familiar example is amorphous drugs, which can be superior to their crystalline counterparts, because they are easier to dissolve and thus easier for the body to assimilate [Babu and Nangia 2011]. The most common way to produce a glass is by cooling a liquid fast through its melting point to prevent it from crystallizing. Upon continued cooling, the liquid becomes more and more viscous until it solidifies (vitrifies) into a glass [Tammann 1925; Kauzmann 1948]. All liquids can be supercooled, i.e., cooled below their melting point without crystallizing, and most can even be deeply supercooled and eventually form a glass. Water is a prominent counterexample to this statement. Making a glassy water is notoriously difficult and cannot be done by simple cooling. Commonly used alternative routes to the glassy state

include elevated pressure and physical vapor deposition where a glassy layer is built by condensing vaporized molecules on a substrate [Angell 1995].

The microscopic structure of a glass is disordered like the liquid, but the molecules are “frozen” in their positions (see Box 1). Thus for all practical purposes, the glass is a solid and has many properties in common with crystals, e.g., low thermal expansion coefficient and high mechanical moduli. The transition from supercooled liquid to a glass – the *glass transition* – is understood as a purely kinetic phenomenon. When the characteristic time for structural relaxation exceeds the inverse relative cooling rate, the molecules no longer have time to adjust to the new temperature and they simply “freeze in.” This implies that different cooling rates result in different glass transition temperatures and glasses with different properties (see Box 2). The glass transition is thus a kinetic phenomenon and not a genuine phase transition like for instance the liquid–gas phase transition. However, since the glassy state can be obtained nearly in any substance, some consider it the fourth state of ordinary matter, in addition to the three classical thermodynamic states: gas, liquid, and crystal [Dyre 2006].

Box 2 Equilibrium and Non-equilibrium Systems

In condensed matter physics, the concept of thermodynamic equilibrium is central. A system *in equilibrium* does not change its (average) internal energy state with time. Its state is completely determined by two thermodynamic variables, for instance, temperature and pressure. A system *out of equilibrium* (non-equilibrium system) has an internal energy that is higher than the corresponding equilibrium state at that thermodynamic state point. This can either be because the system is *driven* (there is a constant flux of energy through the system) or because it has not had time to reach the equilibrium state. The non-equilibrium state requires at least one additional parameter to describe. If left undisturbed, any non-equilibrium system will gradually approach an equilibrium state.

Glasses are non-equilibrium systems, and thus their properties evolve over time. Moreover, the properties of a glass are also dependent on *how* it was made. Two glasses made from the same substance monitored at the same temperature and pressure can have different densities, refractive indices, mechanical moduli, etc. How fast these properties change over time depends dramatically on how far from equilibrium the system is.

Almost no systems found in nature are in equilibrium. They are constantly subjected to perturbations, to flux of matter and energy to and from other systems, and to chemical reactions. Nevertheless, the concept of equilibrium as something always sought is central for understanding non-equilibrium systems and their time development.

2 Viscoelasticity and Spectroscopic Methods

Supercooled liquids at temperatures above their glass transition display *viscoelastic* behavior. This means that their properties are dynamic, i.e., they depend on the time scale of our observation: on the short time scales, the material is solid-like (elastic), and on long time scales, the material is liquid-like (viscous). A familiar example is the children's toy "Silly Putty," which can be shaped into a ball that will bounce when thrown to the floor. But if left undisturbed for some time, it will flow out in a puddle [Cross 2012]. Another example is the famous Pitch Drop Experiment. This longest running experiment in the world was initiated in 1927 at the University of Queensland [Johnston 2013]. A heated sample of pitch was placed in a sealed funnel and allowed to settle for three years. Then the seal was cut at the neck of the funnel and large droplet slowly formed. Once around every ten years a droplet falls. Clearly, the pitch is flowing, albeit very slowly with a viscosity (*flow resistance*) of around 10^{11} (100 billion) times that of water. However, if the pitch sample were hit hard by a hammer, it would shatter like a solid. A third example is the flow of the mantle of the Earth that is responsible for the tectonic movements [Conrad et al. 2013]. The mantle consists of semi-crystalline rock at elevated temperature and pressure, which flows extremely slowly in convective patterns from the core to the crust. The estimated viscosity of the Earth's mantle is 10^{24} times that of water [Cathles 1975, Part IV]. Still, seismic waves after an earthquake travel through the material as if it were an elastic solid. Common for these examples is that the material flows when subjected to a force (for instance, gravity) for a long time. In the first case, the time scale for the flow is hours, in the second the time scale is years, and in the third example it is millions of years.

The time dependence of material properties can of course be studied as a function of time after some perturbation, but it is often more convenient to do such studies as a function of frequency. This means applying a sinusoidally varying perturbation at a given frequency (where frequency means cycles per second) and then scanning all relevant frequencies. Frequency-domain spectroscopy gives information that is equivalent to doing an experiment in the time domain. High frequencies correspond to short times and thus probe the elastic part of the response and low frequencies to long times and thus probe the viscous part of the response. In between, there is a crossover from elastic to viscous behavior. The location of this transition gives the characteristic time scale for molecular rearrangement and is often called *structural relaxation* or the alpha relaxation. The structural relaxation is extremely temperature-dependent: at high temperatures where the liquid is easy flowing

the time scale is picoseconds (10^{-12} s). When temperature is lowered toward the glass transition, the structural relaxation time increases 16 orders of magnitude to approach 100s of seconds. Measuring material properties over all relevant temperatures is thus a tremendous technical challenge.

Note that the characteristic time scale for the structural relaxation is an *intrinsic* time scale, while the time scales for flow mentioned in the previous section are *macroscopic* time scales that are generally much longer. Macroscopic flow – where the substance moves visibly – requires many molecular rearrangements and depends on geometrical parameters: flow through a small pipe takes longer than flow through a large pipe even if the intrinsic time scale of the flowing material is the same. A substance near its glass transition will have an intrinsic time scale of ~ 100 s (or roughly a minute), but if we tried to pour this substance out of a glass, it would take ~ 30 years [Dyre 2006]. We always refer to the intrinsic time scale, when we talk about the *relaxation time* of a liquid.

Viscoelasticity is described in terms of mechanical perturbation (e.g., a force) and mechanical response (e.g., a displacement or flow). However, any perturbation and response of such materials will exhibit this characteristic transition between a solid-like and a liquid-like response. A standard method for probing the dynamic properties of materials is dielectric spectroscopy. Here the electrical response of the material is probed by varying an electrical field and measuring the frequency-dependent polarization of the sample, i.e., how much the molecular dipoles are able to align with the imposed field at a given frequency [Kremer and Schönhals 2002]. The method is convenient because it is a relatively simple and precise way to probe the time dependence of material properties and an extremely broad range of frequencies is available. Broadband dielectric spectroscopy routinely covers 9–10 orders of magnitude and with special equipment a staggering 18 orders of magnitude from μHz (corresponding to one cycle taking almost 12 days to complete) to THz (with 1,000,000,000,000 cycles per second) [Lunkenheimer et al. 2000].

From the above, one could get the impression that there is only one relevant time scale to map in viscoelastic materials. In reality, most amorphous materials are much more complex than that. The schematic graphs in Fig. 1 show how the dielectric spectrum typically evolves with temperature. As described above, the structural alpha relaxation becomes slower when temperature is decreased. In Fig. 1, this corresponds to the dominant peak in the spectrum (yellow area) moving *down* in frequency. As it moves down in frequency, we begin to see other processes separating out, what is usually referred to as time-scale separation. Already at quite high temperatures, the boson peak (pink area), which is vibrational in nature, separates from the structural alpha relaxation, Fig. 1a. The boson peak is virtually temperature-

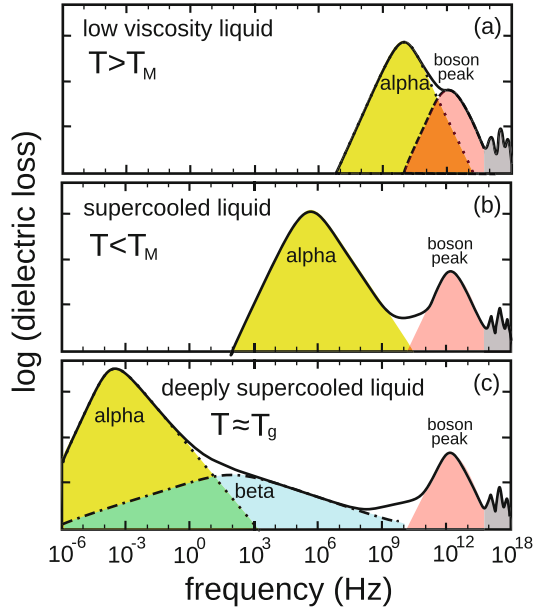


Figure 1 Complex materials have dynamics on all time scales. The schematic presentation of isothermal dielectric spectra of a supercooled liquid shows how the structural alpha relaxation process (yellow peak) moves down in frequency when temperature is lowered. With decreasing temperature, more processes separate out: at high temperature, the boson peak (pink peak) separates from the structural relaxation. At even lower temperatures, usually one or more distinct peaks show up between the structural relaxation and the boson peak, here illustrated by a blue beta process. Adapted after Lunkenheimer et al. [2017] with permission from Springer Nature

independent and thus stays in the same position in the spectrum as temperature is lowered. As the glass transition is approached, one or more smaller processes usually appear between the structural alpha and the boson peak. These processes are termed secondary processes: often denoted beta, gamma, etc. in order of appearance. In Fig. 1c, this is represented by the blue peak appearing at intermediate frequencies, overlapping with the alpha at the low-frequency flank (green area). Each process has its own temperature dependence, and thus to characterize the dynamic properties of viscous materials, it is necessary to map out the entire frequency range as a function of temperature.

Secondary relaxations are diverse in origin. In substances consisting of large molecules, they can originate from internal degrees of freedom in the molecule, e.g., the rotation or bending of a part of the molecule. But secondary relaxations also appear in substances with a simple and rigid

molecular structure [Johari and Goldstein 1970]. In those cases, the origin has to involve movement of the entire molecule, but there is no general consensus to the specific mechanism and in what way it separates from the alpha relaxation. Notably, there are also materials that display no secondary processes [Niss and Hecksher 2018].

Other spectroscopic techniques show similar patterns. One interesting complementary technique covering a similarly large range of frequencies is dynamic light scattering [Berne and Pecora 1976]. Dynamic light scattering also probes rotation of molecules and thus carries some of the same information as dielectric spectroscopy, but not always resulting in identical spectra. Dynamic light scattering is less common than dielectric spectroscopy, because it is a much more demanding technique and generally has a lower resolution.

Finally, mechanical spectroscopy – actually probing the flow properties of materials by “squeezing” or “twisting” the sample – is frequently used to characterize materials. Mechanical spectroscopy has the clear advantage that it probes directly the properties we are interested in, but it is technically much more difficult, especially to obtain high-frequency ($\gg 100$ Hz) response. However, combining a number of techniques, it is possible to cover almost 14 orders of magnitude from mHz to 100s of GHz [Hecksher et al. 2017] (see Fig. 2).

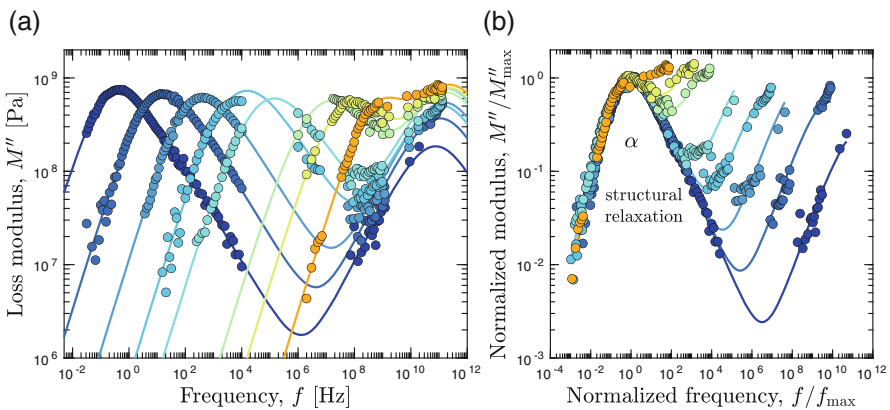


Figure 2 Broadband mechanical spectra over temperatures from the melting point to just above the glass transition. **(a)** shows the mechanical data as a function of frequency for different temperatures. **(b)** shows the spectra scaled to the alpha peak position. This collapses all spectra onto one master curve except for the contribution from the high-frequency microscopic peak. Data from Hecksher et al. [2017]

3 What Do We Learn?

The two main unsolved puzzles in glass science connect to the time and temperature evolution of material properties [Debenedetti and Stillinger 2001]. This is often summarized as the two non's: non-Arrhenius temperature dependence of the structural relaxation time and non-exponential relaxation shape [Dyre 2006]. The non's imply a deviation from the simplest models for the time and temperature dependence of the structural relaxation, that one could come up with, namely Arrhenius temperature dependence of the relaxation time and exponential time dependence of material properties. Instead, most systems have varying degrees of super-Arrhenius behavior and *stretched exponential* relaxation. The stretched exponential relaxation corresponds to an asymmetric/skewed alpha peak in Fig. 1.

Physicists usually look for universal patterns and simple models to describe them. So what patterns do these broadband spectra of viscous liquids and other amorphous materials reveal? Over the many years of glass research, numerous correlations between different quantities have been proposed, hence also correlations between the two non's [Böhmer et al. 1993; Niss et al. 2007]. The paragraphs below give an opinion of the emerging patterns. This opinion is not shared by all colleagues in the field.

3.1 Spectral Shapes

One interesting observation is that in some materials, the spectral shape of the structural relaxation is temperature-independent. This is called time-temperature superposition (TTS). In Fig. 1, TTS of the structural relaxation corresponds to the alpha peak keeping the same shape, when temperature is changed; temperature only shifts peak on the frequency axis as if the same movie was played in slow motion (when temperature is lowered) or in high speed (if the temperature is increased).

Figure 2a shows mechanical broadband spectra of a silicone oil at several temperatures from above the melting point of the liquid and down to the glass transition temperature [Hecksher et al. 2017]. Each color is an isothermal spectrum; reddish colors are the high temperatures with fast dynamics, and blueish colors are the low temperatures with slow dynamics. The spectra are pieced together by seven different techniques, each covering a certain frequency range, and thus there are gaps in the spectrum. Full lines are fits to connect the different parts of the spectrum. In these spectra, the structural alpha relaxation peak is clearly resolved, and only the onset of the vibrational

peak is within the frequency window of the applied techniques. Interestingly, there is no sign of an intermediate beta relaxation in this substance. As in the schematic in Fig. 1, the structural alpha relaxation peak moves down in frequency as the temperature is lowered, while the vibrational peak roughly stays in the THz region. In Fig. 2b, the spectra are collapsed by scaling to the structural alpha relaxation peak frequency and intensity. This shows that the spectral shape is unchanged for the structural peak, except toward the high frequencies where the vibrational peak interferes. As temperature is lowered, the separation between the two peaks increases and more of the alpha peak is revealed.

TTS is found in some, but not all systems [Niss and Hecksher 2018]. It may apply to different processes separately, i.e., the alpha and beta processes each can have a temperature-independent shape, but because of the different temperature dependencies of the processes, TTS does not apply to the full spectrum. In the alpha process, TTS seems to be correlated to a particular spectral shape [Olsen et al. 2001; Nielsen et al. 2009]. This is a quite remarkable property that suggests some kind of simplicity is at play.

In light-scattering spectra, TTS is found frequently [Schmidtke et al. 2013; Blochowicz et al. 2003]. In fact, recent results suggest that there is a generic alpha shape across many different systems [Pabst et al. 2021]. Figure 3 shows a superposition of a single spectrum for a range of different systems. It is

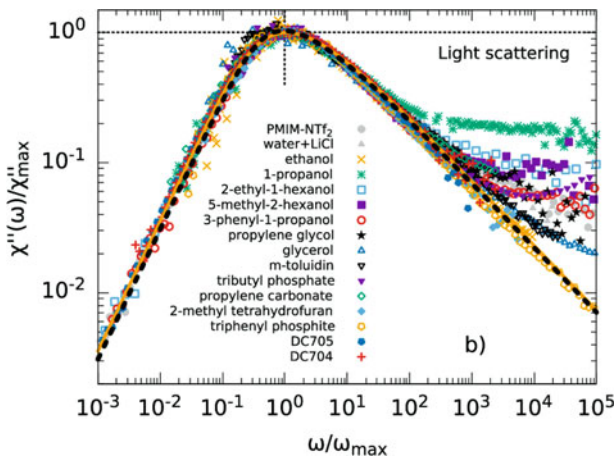


Figure 3 Dynamic light-scattering spectra for a range of different systems. Strikingly, all spectra collapse when they are all scaled to their peak positions. Except for varying deviations on the high-frequency side of the alpha peak arising from different beta contributions, there seems to be a generic alpha relaxation shape independent on the type molecular interaction is dominant in the system. Reprinted with permission from Pabst et al. [2021]. Copyright 2021 American Chemical Society

clear that the shape of the peak is strikingly similar, except toward the high frequencies where secondary relaxations begin to interfere. A similar generic shape across different substances has been observed for mechanical spectra [Gainaru 2019].

The emerging picture seems to be that the structural relaxation, although not as simple as the simplest models would suggest, could have a temperature-independent generic or universal shape. This would be an important input to the theorists in the field.

3.2 Temperature Dependence of the Relaxation Time

The second big puzzle of glass science is the non-Arrhenius temperature dependence of the structural relaxation time. As is the case with any open question in science, there are a number of competing models on the market. Some suggest that entropy controls the structural time scale [Gibbs and DiMarzio 1958; Adam and Gibbs 1965], and others that it is the free volume available to the rearranging molecules [Cohen and Turnbull 1959] (more examples can be found in Dyre 2006 and Dyre et al. 2009).

Here we will focus on one model (the shoving model [Dyre et al. 1996]) having the beauty that: (1) it is easy to understand and (2) it is testable: it involves quantities that can be measured directly. The basis of most models is that flow in a (viscous) liquid consists of a series of flow events, i.e., local rearrangement of molecules [Kauzmann 1948; Goldstein 1969]. These flow events in themselves are fast, and the reason for the structural relaxation slowing down with temperature is that flow events become increasingly rare: at low temperatures, molecules are locked in position by their neighbors most of the time and only occasionally move.

The structural relaxation time (τ) is then the characteristic time that molecules have to wait to rearrange. The argument of the shoving model is that because of the strong repulsion between molecules, moving past each other at constant volume is very energetically costly. Thus the flow events happen primarily when a thermal fluctuation expands the locally available volume – much like if you and your friend want to switch places at a crowded concert, it is often easier to do so when there is a movement in the rest of the crowd that suddenly opens up a space for your swap than to try squeeze past each other.

The energy associated with a rearrangement is thus the energy needed to create the extra space, i.e., the energy needed to “shove aside” the surroundings. This energy is much larger than the energy needed to separate the

rearranging molecules, since the attraction between molecules is weak [Dyre et al. 1996]. Because the flow event is fast, the surrounding material behaves like an elastic solid during a flow event, and the showing model thus argues that the energy associated with the flow is determined by the short-time elastic properties of the liquid.

Mathematically, the model is written as follows:

$$\tau(T) = \tau_0 \exp\left\{\frac{G_\infty(T)V_c}{k_B T}\right\}, \quad (1)$$

where τ_0 is the microscopic vibration time (roughly temperature-independent), $G_\infty(T)$ is the temperature-dependent short-time elastic modulus, V_c is a microscopic volume (assumed roughly temperature-independent), k_B is Boltzmann constant, and T is temperature.

Thus by measuring the structural relaxation time, τ , and the short-time elastic modulus, G_∞ , as functions of temperature, this model can be tested even without knowing V_c .

It is a daunting proposition that the long-time flow properties – extending over seconds to millions of years – are in fact controlled by the short-time elastic response, i.e., the elastic properties on the picosecond (10^{-12} s) time scale. Nevertheless, several experimental tests of this relation supports the model [Hecksher and Dyre 2015].

4 Concluding Remarks and Outlook

Amorphous materials are everywhere around us: from technologically applicable materials to daily-life non-crystalline materials, such as various foods and amorphous drugs. Even many cold-tolerant living organisms use vitrification as part of the survival strategy to avoid freezing that is detrimental to living cells [Debenedetti 1996, Ch. 1.2]. For the most part, this chapter has focused on model systems, i.e., well-behaved glass-forming systems studied above the glass transition. This is because these are the simplest systems to study – and yet fundamental puzzles remain unsolved regarding what governs their dynamical properties and how that connects microscopic interactions between the constituent molecules. This is basic research, but with applications in the immediate vicinity as stressed in the introduction. From a physicists' point of view – with the goal to understand the fundamental mechanisms of nature – it makes sense to start with the simplest and go up in complexity from there.

A consensus regarding what governs the structural relaxation time of a viscous liquid has not been reached in the field. The model presented here connecting the short- and long-time properties is both promising and fascinating. It is a model for the *equilibrium* properties (see Box 2), but it also has actually been shown in one case to consistently describe the dynamics out of equilibrium [Hecksher et al. 2019]. In its current form, the model might be too simplistic, but it seems to capture the essence of the problem and is thus a good starting point for a more complete description. The question about the relaxation shape is less mature. It is mostly discussed among experimentalists and has not reached the attention of many theoreticians in the field. The notion of a generic, yet non-trivial, relaxation shape for a range of very different substances with different molecular interactions is appealing, because it indicates that there is a universal model for the dynamics in highly viscous liquids that only relates to fact that they are disordered and not to their molecular composition or interactions.

In the future, prevailing theories should be tested experimentally through more measurements and the development of more precise techniques. But an important contribution might also come from computer simulations. Currently, computer simulations of more or less realistic models of molecules are able to simulate to the millisecond time scale. Measurements can reach far longer time scales with far more particles. But computers are getting exponentially faster and may in the future even overtake experiments regarding time scales. The beauty of a computer simulation is that all the microscopic information is available and interactions can easily be tweaked to test connections between microscopic and macroscopic properties.

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