

Unified percolation scenario for the α and β processes in simple glass formers

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Given the vast differences in interaction details, describing the dynamics of structurally disordered materials in a unified theoretical framework presents a fundamental challenge to condensed-matter physics and materials science. Here we numerically investigate a double-percolation scenario for the two most important relaxation processes of supercooled liquids and glasses, the so-called α and β relaxations. For several simple glass formers, we find that when monitoring the dynamic shear modulus as temperature is lowered from the liquid state, percolation of immobile particles takes place at the temperature locating the α process. Mirroring this, upon continued cooling into the glass state, the mobile-particle percolation transition pinpoints a β process whenever the latter is well separated from the main (α) process. For two-dimensional systems under the same conditions, percolation of mobile and immobile particles occurs nearly simultaneously, and no β relaxation can be identified. Our findings suggest that a general description of glassy dynamics should be based on a percolation perspective.

A liquid close to the glass transition relaxes extremely slowly towards equilibrium when subjected to an external disturbance^{1–10}. Depending on the temperature, the main so-called α relaxation time, τ_α , can be seconds, hours or even months, with no other limit than the patience of the experimentalist^{2–5,9–11}. In the Maxwell model of viscoelasticity^{5,12}, a glass-forming liquid behaves like a solid on time scales shorter than τ_α and flows on longer time scales. Interestingly, most liquids exhibit additional, faster relaxations. The most prominent one is the Johari–Goldstein β process, which is observed in virtually all glass-forming organic liquids, polymers, metallic glasses and so on^{13–18}.

It was known by the 1960s that polymers exhibit relaxation processes above the α relaxation frequency $1/\tau_\alpha$, which were attributed to side-chain motion¹³. It was a surprise, however, when Johari and Goldstein reported in 1970 that fast processes also occur in glasses of small rigid molecules¹⁹. To explain this, they proposed the existence of ‘islands of mobility’^{19,20}; this constituted an early example of the dynamic heterogeneities subsequently identified as a universal feature of glass-forming liquids^{6,21–23}. Nowadays, the term ‘ β relaxation’ is used for the first relaxation process at frequencies higher than those of the main (α) process, independent of its origin. A further relaxation

process worth mentioning is the β' (or γ) process, which has been observed in both experiments and molecular dynamics simulations. It originates from high-frequency particle diffusion in glass-forming liquids and is considered an extension of the dynamics observed in high-temperature liquids^{24,25}.

Research in recent decades has demonstrated that β relaxation plays a crucial role in the mechanical and thermal properties of amorphous materials^{26–30}. Ngai has suggested that the β process is a precursor of the main α relaxation: before the onset of β relaxation, one finds a regime in which molecules are confined to cages defined by the anharmonic intermolecular potential^{31–33}. A related proposal was discussed in 1999 by Kudlik et al., who suggested that the β process in molecular liquids is a local, spatially restricted reorientation process preceding the α relaxation³⁴, an idea that is reminiscent of the fundamental prediction of mode-coupling theory³⁵. Experiments have confirmed the caged-molecule picture of the β process by detecting small-angle jumps³⁶, but interestingly, large-angle jumps are sometimes also involved in β relaxation³⁷.

Although most papers on the β process report results for the glass phase, β relaxation is present also in the equilibrium (metastable)

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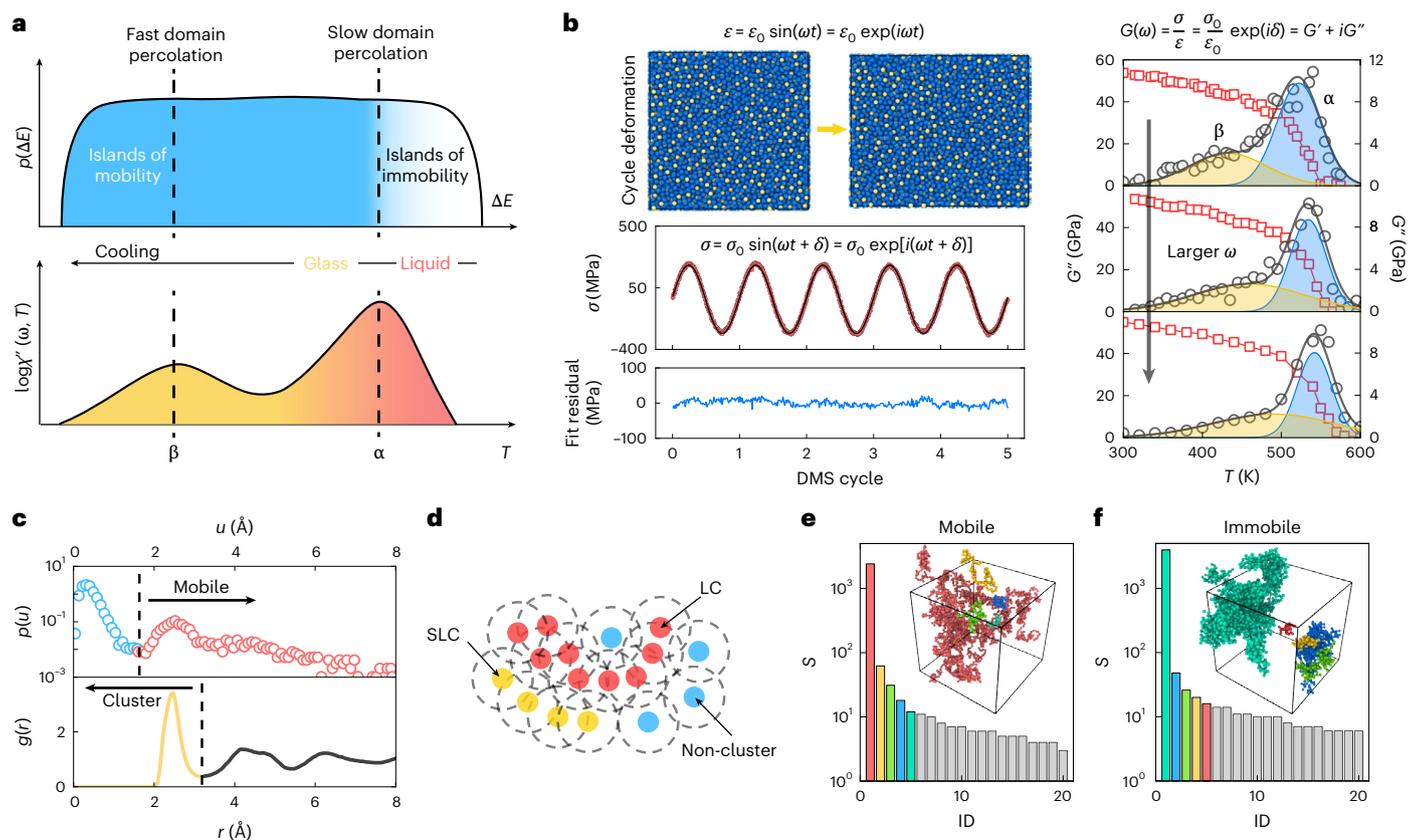


Fig. 1 | Main concepts. **a**, Generic double-percolation scenario for a glass-forming system's linear-response properties when cooled through the glass transition. The loss $\chi''(\omega)$ is probed at a fixed frequency during the cooling (lower panel), corresponding to activation energies that decrease with decreasing temperature (upper panel). Assuming that the activation-energy distribution is much wider than $k_B T$, at a certain temperature the high-activation-energy 'islands of immobility' corresponding to times longer than the probe frequency will percolate. This locates the α process. At a lower temperature well within the glass phase, the low-activation-energy 'islands of mobility' stop percolating, which pinpoints the β relaxation^{48,63,64}. **b**, Molecular dynamics simulations mimicking experimental DMS for a $\text{Ni}_{30}\text{P}_{20}$ mixture. At selected times

during the cooling, the sample is deformed to determine the dynamic shear modulus $G(\omega)$. **c**, Definition of particles that are mobile on the time scale $\Delta t = 2\pi/\omega$. The upper panel shows the distribution of all-particle displacements, the van Hove function $p(u, \Delta t)$. Particles with displacement larger than the minimum (dashed line) are designated as 'mobile' and the remaining particles as 'immobile'. The lower panel shows the all-particle radial distribution function $g(r)$. Whenever two mobile/immobile particles are closer than the first minimum of $g(r)$, they are defined to belong to the same cluster⁹¹. **d**, Example of the LC (red) and SLC (yellow), concepts that are defined for the mobile and the immobile particles separately. **e, f**, Examples of mobile (e) and immobile (f) particle clusters (insets) and their size (S) histograms.

liquid phase above the glass transition temperature T_g . In most cases, however, the β process is here partly merged with the α process and observed only as a wing of the latter^{38–41}. Lengthy annealing of a glass to approach the liquid phase may in some cases separate the β relaxation wing from the α peak and establish it as an independent, well-defined process³⁹, but in other cases annealing annihilates the β process by decreasing its magnitude to below the resolution limit¹⁴.

Because a glass-forming liquid is disordered, it must be expected that the energy barriers of flow events vary in space. The existence of a wide barrier distribution was recently documented in simulations by Pica Ciamarra and co-workers⁴². Assuming the barriers vary randomly in space amounts to replacing complexity with randomness, which is an old and venerated strategy of simplification⁴³. Once spatial randomness is introduced into the modelling, the phenomenon of percolation comes to mind by connecting randomness and geometry^{44–47}: if finite-size domains in space are marked randomly one after the other, at some point the marked domains will percolate throughout the sample. The value of the percolation threshold depends on the spatial dimension and the model in question. In one dimension, the percolation threshold is unity. On a two-dimensional (2D) cubic lattice, the link-percolation threshold is 50% by self-duality^{45–47}. The percolation threshold decreases with increasing spatial dimension D , and for $D \rightarrow \infty$

it approaches zero. The threshold is 0.25 for link percolation on a three-dimensional (3D) simple cubic lattice and 0.31 for site percolation on the same lattice^{45–47}.

Recently, a double-percolation picture was proposed, linking the α and β processes in the liquid phase above T_g to the percolation of immobile and mobile regions, respectively⁴⁸. Associating a given energy barrier, ΔE , with a relaxation time proportional to $\exp(\Delta E/k_B T)$ where T is the temperature and k_B is the Boltzmann constant, the von Neumann type 'minimax' idea is that τ_α is controlled by the lowest energy barriers on the percolation cluster formed by the most immobile regions: that is, those of largest barriers. On time scales longer than τ_α , this cluster breaks up and flow becomes possible. As a consequence, the solid-like structure maintaining the energy barriers throughout the sample by keeping surrounding molecules in place disappears on time scales longer than τ_α (refs. 49–53). This means that the largest energy barriers are never overcome; hence their light colour in Fig. 1a illustrating double percolation. Borrowing a term from nuclear magnetic resonance^{54,55}, this phenomenon was long ago referred to as 'exchange', whereas nowadays the term 'facilitation' is preferred^{56–62}. Considering next the opposite limit of very short times, only the lowest-barrier regions are relevant, and these are spatially separated. Extended fast motion becomes possible when these regions percolate

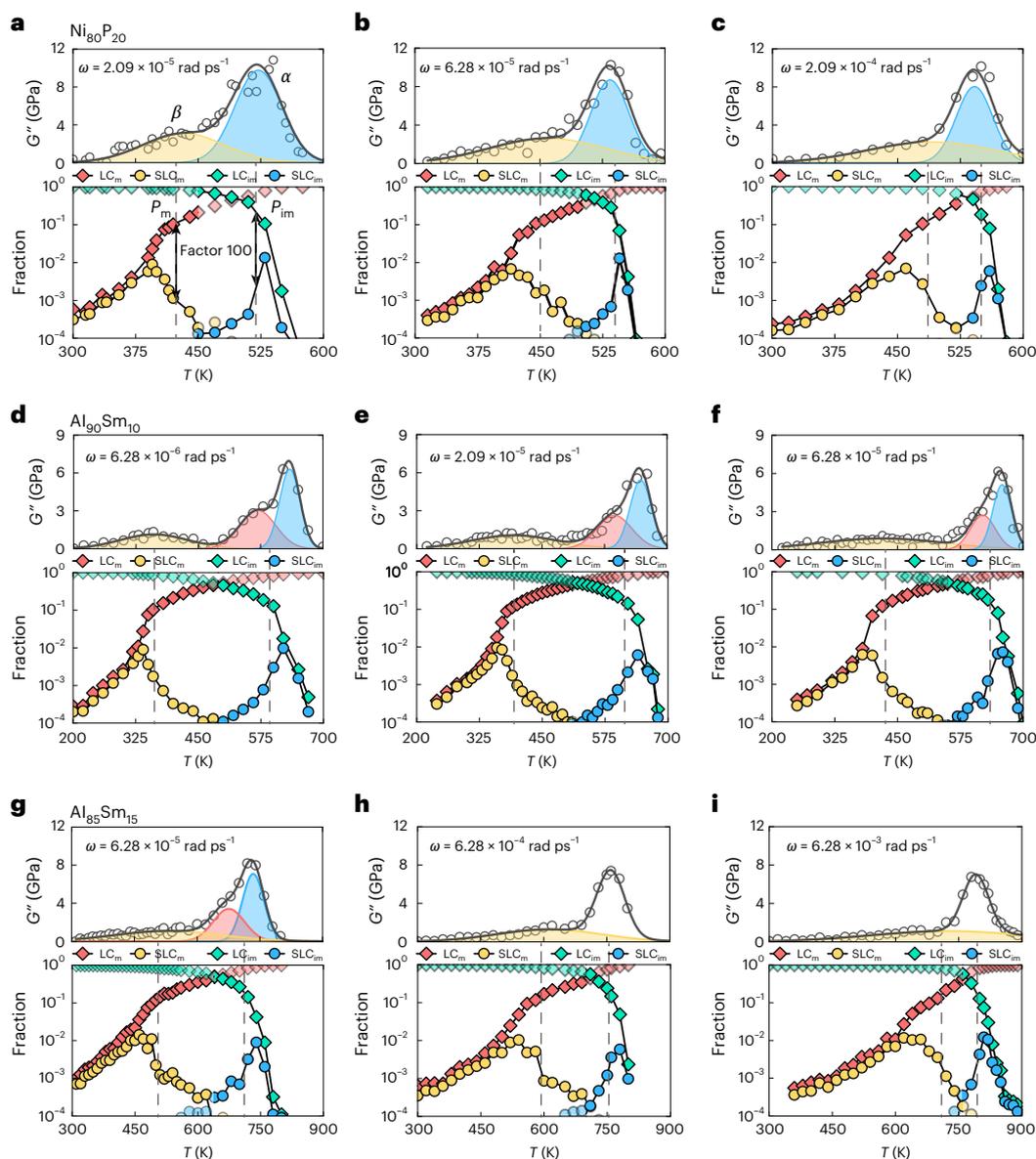


Fig. 2 | Double-percolation studies. **a–i**, Results at three frequencies for $\text{Ni}_{80}\text{P}_{20}$ (**a–c**), $\text{Al}_{90}\text{Sm}_{10}$ (**d–f**) and $\text{Al}_{85}\text{Sm}_{15}$ (**g–i**). For each panel, the upper graph displays the shear-mechanical loss modulus $G''(\omega)$ at a fixed frequency monitored when cooling at 0.1 K ns^{-1} . The loss modulus is fitted to a sum of Gaussian peaks, corresponding to α (blue), α_2 reflecting the α process asymmetry⁴⁸ (pink, not needed in all spectra) and β (yellow) processes. The lower panels show the fraction of particles belonging to the LC (pink/green) and SLC (yellow/blue) of

mobile (left) and immobile (right) particles, respectively. The dashed lines mark the mobile- and immobile-particle percolation temperatures defined from the criterion that the LC is 100 times larger than the SLC. **h** and **i** do not show separate α and α_2 processes because of the uncertainty of their relative positions (Supplementary Figs. 5–7 give more details on the α_2 process and the fitting procedure).

on longer time scales. This gives rise to a new relaxation channel that we propose is the β process^{48,63,64}.

The idea that percolation is important for understanding relaxations in viscous liquids and glasses has a long history^{24,48,50,51,53,63–75}. A double-percolation picture was proposed in 1996 by Novikov et al. in the context of percolation of liquid-like and solid-like domains defined by the largest and smallest vibrational mean-square displacement, respectively⁷⁶, but no relation to the α and β processes was proposed at the time.

Figure 1 illustrates our main idea and how it is tested in simulations. With current computers, it is not possible to test numerically the double-percolation scenario in the equilibrium-liquid phase in the way this scenario was discussed in ref. 48; it would require extremely long low-temperature simulations to separate the α and β processes,

which as mentioned is a challenge even in experiments lasting a long time. As an alternative, we proceed as follows. Figure 1a presents the double-percolation scenario for a linear-response loss at the fixed (angular) frequency ω , $\chi''(\omega)$, monitored as a function of temperature when the system is cooled from the equilibrium-liquid state into the glass phase. This procedure allows one to monitor the activation-energy distribution in much the same way as a constant-temperature equilibrium-liquid frequency scan. The largest activation energies are probed at high temperatures, with gradually smaller ones becoming relevant as the system is cooled. Two ‘percolation temperatures’ are conjectured to pinpoint the α and β processes, respectively. At any given time, all particles are marked as either mobile or immobile. The immobile particles percolate below the upper percolation temperature, and the mobile particles percolate above

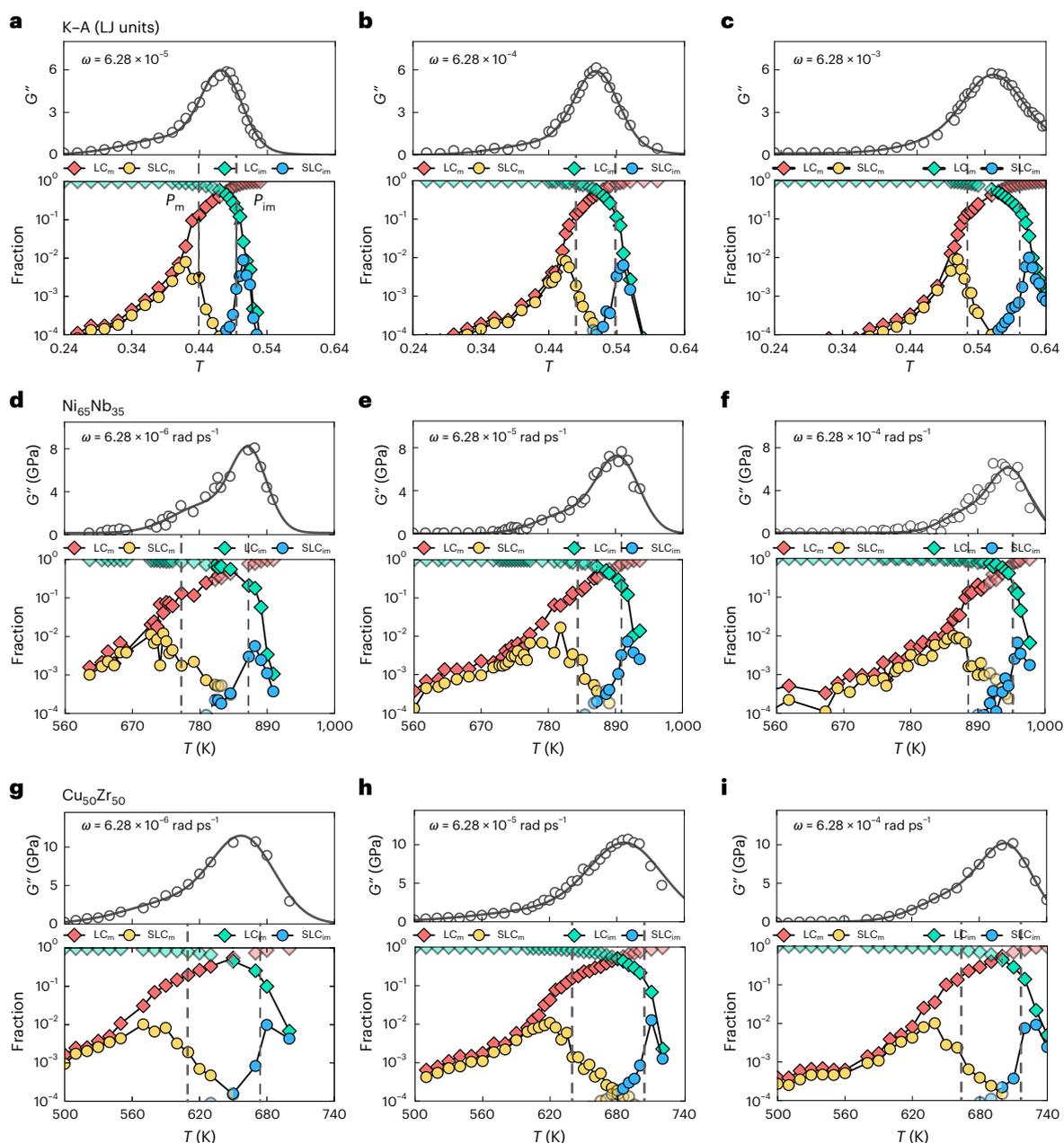


Fig. 3 | Double-percolation studies of systems with close percolation temperatures. **a–i**, Results at three frequencies for 65:35 Kob–Andersen (K–A) (**a–c**), $\text{Ni}_{65}\text{Nb}_{35}$ (**d–f**) and $\text{Cu}_{50}\text{Zr}_{50}$ (**g–i**) (similar data for the ternary system $\text{La}_{50}\text{Ni}_{35}\text{Al}_{15}$ are given in Supplementary Fig. 9). The upper panels display $G'(\omega)$ for samples cooled at the rates 2×10^{-7} (K–A, Lennard–Jones (LJ) units)

and 0.1 K ns^{-1} ($\text{Ni}_{65}\text{Nb}_{35}$ and $\text{Cu}_{50}\text{Zr}_{50}$). As in Fig. 2, all α processes are found at the immobile-particle percolation temperature, but in contrast to Fig. 2, these three samples have no well-defined β process at the mobile-particle percolation threshold.

the lower percolation temperature. Thus both species percolate between these two temperatures. Because this is not possible in two dimensions, we predict that separate α and β relaxations should not be observed here. Note that the system is assumed to have an activation-energy distribution much wider than $k_B T$, an assumption that may not always be realistic.

Our simulations follow the strategy of ref. 64, which mimicked experimental dynamic mechanical spectroscopy (DMS) by occasionally, during a slow cooling through the glass transition, subjecting the sample to a periodic deformation to probe the dynamic shear modulus. The numerical study of α and β processes in realistic molecular models is extremely challenging because it requires both specially designed models and lengthy computations^{41,71,77,78}. For this reason, the

present paper tests the double-percolation scenario with simulations of point-particle models.

We find that whenever the α and β processes are well separated, they correspond to the percolation transition of immobile and mobile particles, respectively. This confirms the double-percolation scenario. There are also cases where the two processes are not well separated, however; in particular, this is always the situation in two dimensions. Most of the models studied are binary metallic glass formers. Our investigation also includes data on a binary Lennard–Jones system, the metalloid NiP and a ternary metallic glass, but relaxations of ionic or covalent glasses involving mobile cations remain to be explored from the double-percolation perspective, as do relaxations in molecular models.

Results

In DMS, the glass transition is seen as a maximum in a plot of the temperature dependence of the fixed-frequency mechanical loss monitored during cooling, and the β process manifests itself as a smaller peak below T_g . Figure 1b illustrates our simulation procedure. At selected times during the cooling, a binary 32,000-particle sample is subjected to a small periodic elongation while the two transverse dimensions simultaneously are decreased to keep the volume fixed. This results in a periodic shear stress on a constant-volume sample, the magnitude and phase of which determine the complex frequency-dependent shear modulus $G(\omega)$ (more details are given in the Methods section and the Supplementary Information). The right side of Fig. 1b shows an example of $G(\omega) = G'(\omega) + iG''(\omega)$ at three frequencies, plotted as a function of temperature where squares and circles give the real and imaginary parts of $G(\omega)$. The β process is most clearly visible at the lowest frequency (upper panel).

There is considerable freedom in how to define mobile and immobile particles. In order not to introduce arbitrary parameters, we adopted the simple approach of treating all particles on an equal footing by proceeding as follows (Fig. 1c). For a given time interval, Δt , mobile particles are defined via the all-particle van Hove function $p(u, \Delta t)$, which in all cases simulated has a well-defined first minimum or transition from peak to a long tail (Supplementary Figs. 3 and 4). Particles with a displacement larger than this length are designated as ‘mobile’ and all other particles as ‘immobile’. This depends on Δt , of course, which is put equal to $2\pi/\omega$ of the mechanical deformation. Note that in this approach, any particle is either mobile or immobile. This is different from what is usually done; in ref. 69, for instance, fewer than one-fifth of the particles were classified as either mobile or immobile.

Having defined mobile and immobile particles at any given time, two particles of same class are designated to belong to the same cluster if their distance is smaller than the minimum of the all-particle radial distribution function, $g(r)$; compare the lower panel of Fig. 1c. In this way, at any time during the cooling, one identifies the largest cluster (LC) and the second-largest cluster (SLC) of mobile and immobile particles, respectively (Fig. 1d–f). In summary, the probe frequency defines the time scale used to classify the particles as either mobile or immobile, and each of these two classes is subsequently divided into clusters.

Figure 2a–c shows our results for $\text{Ni}_{80}\text{P}_{20}$ cooled through the glass transition and monitored at the three frequencies in Fig. 1b. Figure 2a gives results for the lowest frequency where the α and β processes are best separated, and Fig. 2b,c give results for higher frequencies. The upper panels show the shear-mechanical loss modulus as a function of temperature during the cooling. The lower panels show the fraction of particles belonging to the LC (orange and green diamonds) and SLC (yellow and blue circles) of mobile and immobile particles, respectively (left and right). The two dashed vertical lines mark the mobile- and immobile-particle percolation temperatures, $T_{p,m}$ and $T_{p,im}$, defined by the criterion that the LC is 100 times larger than the SLC.

Focusing first on the immobile particles, for all three frequencies we find that during cooling, the LC and SLC fractions are virtually identical down to the temperature $T_{p,im}$ close to that of the α peak, below which LC completely dominates. Upon continued cooling into the glass phase, an almost-mirror behaviour is found for the β peak: here, the LC and SLC fractions are different down to a temperature, $T_{p,m}$, close to that of the β peak, at which all the mobile-particle clusters become small (~1%). Thus the α peak is found where the immobile particles percolate, and the β peak is found where the mobile particles percolate.

To investigate the generality of these findings, we carried out simulations of five other metallic glasses, $\text{Al}_{90}\text{Sm}_{10}$, $\text{Al}_{85}\text{Sm}_{15}$, $\text{Ni}_{65}\text{Nb}_{35}$, $\text{Cu}_{50}\text{Zr}_{50}$ and $\text{La}_{50}\text{Ni}_{35}\text{Al}_{15}$, as well as of a Kob–Andersen-type binary Lennard–Jones mixture⁷⁹ (Figs. 2 and 3 and Supplementary Fig. 9). The results can be summarized as follows: (1) the α process is in all cases characterized by immobile-particle percolation; (2) whenever

Table 1 | Data for all simulations in 3D, showing that the β process manifestation is predicted by the ratio between the mobile- and immobile-particle percolation temperatures

	ω (radps ⁻¹)	$T_{p,m}$ (K)	$T_{p,im}$ (K)	$T_{p,m}/T_{p,im}$	β manifestation
$\text{Ni}_{80}\text{P}_{20}$ (EAM)	2.09×10^{-5}	425	520	0.82	Shoulder
	6.28×10^{-5}	450	540	0.83	Shoulder
	2.09×10^{-4}	485	550	0.88	Wing
	6.28×10^{-4}	505	562	0.90	Wing
	2.09×10^{-3}	538	578	0.93	Wing
	6.28×10^{-3}	558	600	0.93	Wing
$\text{Al}_{90}\text{Sm}_{10}$ (EAM)	6.28×10^{-6}	365	595	0.61	Peak
	2.09×10^{-5}	395	615	0.64	Peak
	6.28×10^{-5}	422	633	0.67	Peak
	2.09×10^{-4}	460	660	0.70	Peak
	6.28×10^{-4}	495	665	0.74	Peak
	6.28×10^{-3}	585	708	0.83	Shoulder
$\text{Al}_{85}\text{Sm}_{15}$ (EAM)	6.28×10^{-5}	505	710	0.71	Peak
	6.28×10^{-4}	590	755	0.79	Shoulder
	6.28×10^{-3}	710	795	0.89	Wing
$\text{Ni}_{65}\text{Nb}_{35}$ (EAM)	6.28×10^{-6}	750	860	0.87	Wing
	6.28×10^{-5}	827	900	0.92	Wing
	6.28×10^{-4}	875	948	0.92	Wing
$\text{Cu}_{50}\text{Zr}_{50}$ (EAM)	6.28×10^{-3}	950	1,010	0.94	Wing
	6.28×10^{-6}	610	673	0.91	Wing
	6.28×10^{-5}	640	703	0.91	Wing
	2.09×10^{-4}	662	717	0.92	Wing
	6.28×10^{-4}	680	735	0.93	Wing
	2.09×10^{-3}	700	755	0.93	Wing
$\text{La}_{50}\text{Ni}_{35}\text{Al}_{15}$ (DNN)	6.28×10^{-3}	720	780	0.92	Wing
	6.28×10^{-5}	527	590	0.89	Wing
	6.28×10^{-4}	570	620	0.92	Wing
K–A (LJ units)	6.28×10^{-3}	610	672	0.91	Wing
	6.28×10^{-6}	0.40	0.46	0.87	Wing
	6.28×10^{-5}	0.44	0.50	0.88	Wing
	6.28×10^{-4}	0.48	0.54	0.89	Wing
	6.28×10^{-3}	0.52	0.60	0.87	Wing

The β process classification into ‘shoulder’, ‘wing’ or ‘peak’ is detailed in Supplementary Fig. 8. DNN, deep neural network.

there is a well-defined β process in the form of a peak or a shoulder, it is characterized by mobile-particle percolation (Fig. 2); (3) for the Kob–Andersen, $\text{Ni}_{65}\text{Nb}_{35}$, $\text{Cu}_{50}\text{Zr}_{50}$ and $\text{La}_{50}\text{Ni}_{35}\text{Al}_{15}$ systems, the β process is not well separated from the α process and is merely visible as a wing of the latter (Fig. 3 and Supplementary Fig. 9).

What is the difference between the systems with a clearly visible β relaxation (Fig. 2) and those with only a wing (Fig. 3)? Whenever the mobile- and immobile-particle percolation temperatures are close, one cannot expect to find well-separated α and β relaxations, and the β process will be at most a wing of the α process. The percolation temperatures are reported in Table 1; recall that these depend on the frequency/time scale in question. The table reveals a threshold of $T_{p,m}/T_{p,im} \cong 0.85$ below which the mobile- and immobile-particle percolation transitions are well enough separated for a β relaxation to be identifiable, which is not the case above the 0.85 threshold. A genuine β peak is seen for the systems with the lowest percolation-temperature ratios.

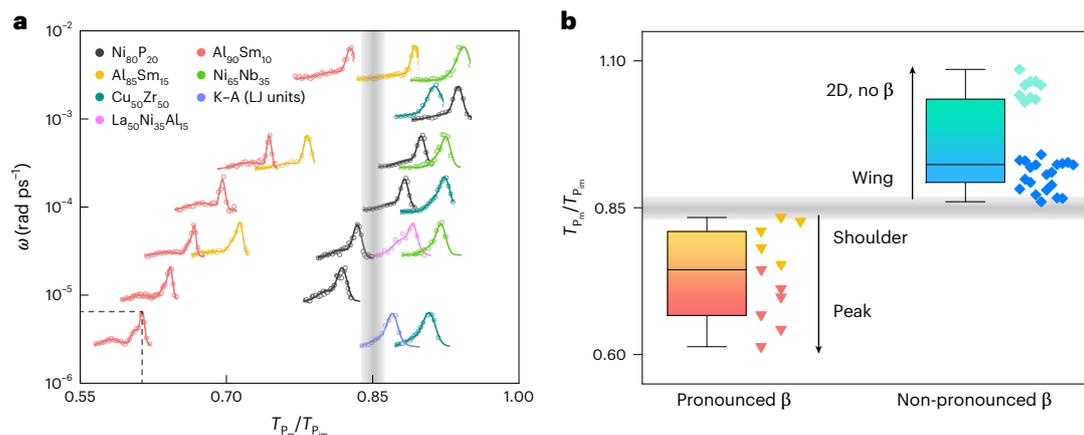


Fig. 4 | The ratio between mobile and immobile percolation temperatures determines the β process manifestation. a, Selected loss spectra of 3D mixtures at different frequencies. When the percolation-temperature ratio is below roughly 0.85 (marked in grey), the β process is a ‘shoulder’ or a ‘peak’, whereas the β process is absent or at most a ‘wing’ for percolation-temperature ratios larger than 0.85. The placement of each loss spectrum is determined by its maximum,

as indicated by the two dashed lines. **b**, Summary of all data for the percolation-temperature ratio and its correlation with the β process manifestation. The data behind the box plots are shown nearby. Each box includes the middle 50% of the data: that is, the edges of each box mark the first and third quartiles. The lines inside the boxes are the medians, and the whiskers mark the smallest and largest data values.

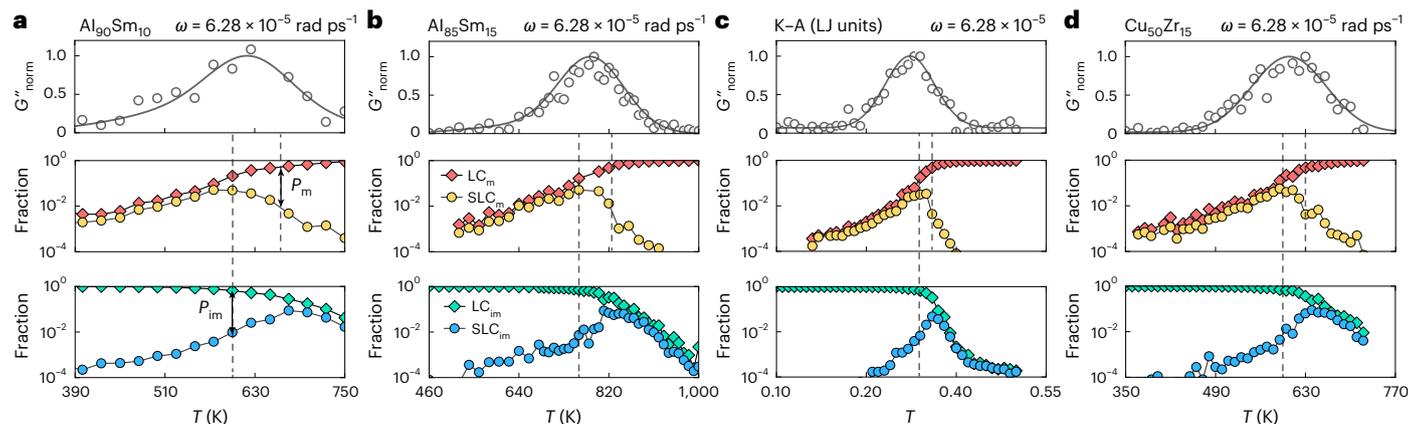


Fig. 5 | Double-percolation studies of 2D models. a, Data for $\text{Al}_{90}\text{Sm}_{10}$. **b**, Data for $\text{Al}_{85}\text{Sm}_{15}$. **c**, Data for the Kob–Andersen mixture. **d**, Data for $\text{Cu}_{50}\text{Zr}_{15}$. (Supplementary Fig. 20 gives data for the 2D $\text{La}_{50}\text{Ni}_{35}\text{Al}_{15}$ mixture.) Unlike in 3D, the two percolation temperatures are close; compare Fig. 4b. This is consistent

with the finding that there are no separate α and β processes (upper panels), reflecting the fact that in two dimensions, mobile- and immobile-particle percolation cannot occur at the same time.

Figure 4a illustrates our results by showing representative loss spectra in a plot where the x coordinate is the ratio of the two percolation temperatures and the y coordinate is the probe frequency. There is a pronounced β process at low ratios of the percolation temperatures. When the two percolation temperatures are close ($T_{P_m}/T_{P_{im}} > 0.85$), on the other hand, the β process is at most manifested as a wing. Figure 4b summarizes our findings by reporting the percolation-temperature ratios for all simulations.

We also simulated systems in 2D where one cannot at the same time have percolation of both the mobile and the immobile particles: if one type of particles percolates, their percolation cluster will necessarily sever any infinite cluster of the opposite type of particles (think of the paths and walls of a labyrinth: if the walls percolate, the paths do not, and vice versa). Thus, according to the double-percolation scenario, no separate α and β processes should exist in two dimensions, and the ratio of the two percolation temperatures should be close to unity. This is tested for five systems in Fig. 5 and Supplementary Figs. 16–20 (the 2D versions of the remaining two systems simulated in 3D, $\text{Ni}_{80}\text{P}_{20}$ and $\text{Ni}_{65}\text{Nb}_{35}$, could not be included in this analysis because they crystallized

upon cooling). In no cases do we find a β process, and the ratio of the two percolation temperatures is always significantly above 0.85 (actually, it is slightly above unity; compare Fig. 4b).

Because the two percolation temperatures are defined by reference to the specific time scale $\Delta t = 2\pi/\omega$, repeating the simulations for different frequencies allows one to identify the two time scales’ temperature dependencies. Results are shown for four systems in Fig. 6 for frequencies covering more than 3 decades. The figure also plots the α and β loss-maximum temperatures at the corresponding frequencies. Within the numerical uncertainty, the latter coincide with the immobile- and mobile-particle percolation temperatures, respectively, confirming the connection between double percolation and mechanical response. The lower panels of Fig. 6 show that the same picture is seen if one uses an alternative percolation-temperature definition based on the observed percolation thresholds, 10% and 25% of the mobile and immobile particles, respectively (compare Supplementary Figs. 22–24; the different percolation thresholds reflect the interesting fact that the geometries of the immobile- and mobile-percolation clusters differ).

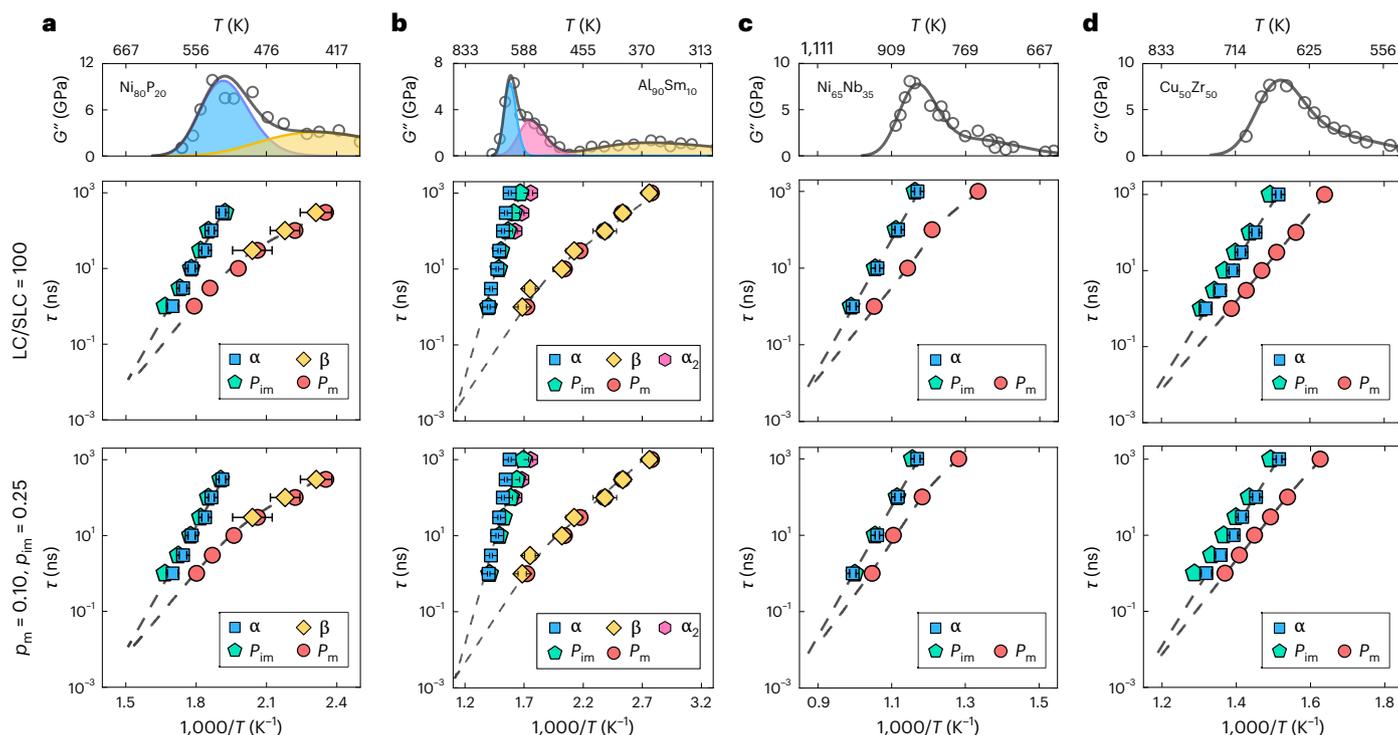


Fig. 6 | Temperature dependence of the α and β average relaxation times and of the immobile- and mobile-particle percolation times. a, Data for $\text{Ni}_{80}\text{P}_{20}$, b, Data for $\text{Al}_{90}\text{Sm}_{10}$, c, Data for $\text{Ni}_{65}\text{Nb}_{35}$, d, Data for $\text{Cu}_{50}\text{Zr}_{50}$. The upper panels show examples of the loss moduli at a fixed frequency. For $\text{Ni}_{65}\text{Nb}_{35}$ and $\text{Cu}_{50}\text{Zr}_{50}$, the α and β processes are not well separated; for $\text{Al}_{90}\text{Sm}_{10}$, there is an additional α_2 process reflecting the asymmetry of the α process (pink hexagon in the lower panel). The middle panels plot the percolation times, defined as the inverse angular frequencies for which the temperature in question is the percolation temperature, as well as the relaxation times based on the LC/SLC = 100 criterion used above. The α and α_2 processes (blue square) give the temperature

dependence of the average relaxation time (inverse loss-peak frequency), which is the same as that of immobile-particle percolation (green pentagon). Likewise, the β process (yellow diamond) has the same temperature dependence of its average relaxation time as that of mobile-particle percolation (orange circle). The dashed lines are guides to the eye illustrating that the two processes merge at high temperatures. The lower panels demonstrate the same picture if one instead uses the percolation thresholds derived from a detailed analysis of the percolation-cluster geometry (see Supplementary Fig. 22): $P_m = 0.10$ and $P_{im} = 0.25$, in which P is the fraction of mobile/immobile particles. The error bars show the uncertainty of the fitting of peak positions for each process.

Discussion and outlook

Percolation is important in many contexts involving disordered solids⁸⁰ by determining, for example, thermodynamics, fragility and stability of chalcogenide glasses^{81,82}, ac conduction at extreme disorder⁸³, spatial heterogeneity of soft modes⁸⁴, vibrational anomalies⁸⁵, yielding⁸⁶ and so on. This Article has investigated numerically a scenario according to which percolation also controls the two main relaxation processes of glass-forming liquids. The starting point is the assumption of extreme dynamic heterogeneity in the form of a wide barrier distribution for flow events (Fig. 1a). From this, one arrives at a picture characterized by percolation of the mobile and of the immobile particles. The former percolation transition is linked to the β process and the latter to the main (α) relaxation. Note that the proposed scenario does not take into account local facilitation: that is, the recently discussed mechanisms according to which one flow event makes nearby flow events more likely by lowering their barriers—for example, by long-ranged elastic interactions^{41,48,61,87–89}.

To summarize our findings, extensive computer simulations of systems in three and two dimensions establish the following. Two temperatures can be identified marking the percolation of mobile and immobile particles, respectively, defined by reference to the particle displacements on a specific time scale. One percolation temperature marks the immobile-particle percolation threshold, which is found above the glass transition temperature; the second percolation temperature marks mobile-particle percolation taking place in the glass. Whenever the two percolation temperatures are well separated, they pinpoint the α and β shear-modulus loss-peak

temperatures for the frequency corresponding to the time scale in question. A ratio of approximately 0.85 of the two percolation temperatures separates two cases; whenever the ratio is below 0.85, the α and β processes are well separated, and whenever this ratio is above 0.85, the processes are partly or fully merged. In two dimensions, where one cannot have percolation of both mobile and immobile particles at the same time, there are no separate α and β processes—here the ratio of the two percolation temperatures is close to unity (in fact, slightly above).

Although we have found no exceptions to this connection between percolation and the mechanical linear-response properties, it should be emphasized that one cannot yet conclude that a general, causal relation exists between the percolation of mobile and immobile particles and the α and β processes. More work is needed before generality of the double-percolation picture can be concluded. It would be interesting to relate double percolation to well-known features of the α and β relaxations, in the hope that the scenario may help provide answers to questions like these: Why is the β relaxation Arrhenius but the α usually is not? How do the α and β relaxations merge at high temperatures? Why is the β process usually symmetric in $\log(\text{frequency})$? Our conclusions are based on numerical studies of metallic glasses⁹⁰, confirmed by data on the Kob–Andersen model and the metalloid NiP. To investigate whether double percolation always controls the α and β processes of glass formers, for future work it will be important to simulate other inorganic/non-metallic systems, as well as more complex systems like molecular and polymeric glass formers.

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Methods

Molecular dynamics simulations and DMS

Twelve different mixtures were simulated using LAMMPS⁹². The 3D mixtures are Ni₈₀P₂₀ (32,000 particles), Al₉₀Sm₁₀ (32,000/10,976/2,916 particles), Al₈₅Sm₁₅ (9,088 particles), Ni₆₅Nb₃₅ (9,088 particles), Cu₅₀Zr₅₀ (32,000 particles), La₅₀Ni₃₅Al₁₅ (8,788 particles) and 65:35 Kob–Andersen (70,304 particles). The 2D mixtures are Al₉₀Sm₁₀ (7,200 particles), Al₈₅Sm₁₅ (7,200 particles), Cu₅₀Zr₅₀ (7,200 particles), La₅₀Ni₃₅Al₁₅ (7,200 particles) and 65:35 Kob–Andersen (7,200 particles). It is demonstrated by example in Supplementary Fig. 21 that these samples are large enough to represent the genuine bulk response⁹³, with the correct percolation temperatures. With the exception of the Kob–Andersen and La₅₀Ni₃₅Al₁₅ mixtures, which use Lennard–Jones and deep neural network potentials⁹⁴, respectively, all mixtures employ embedded-atom-method (EAM) potentials⁹⁵.

Simulations were initiated using the melt–quench method that involves the following two steps: (1) annealing the mixture at a high temperature above the melting point until its energy stabilizes, thereby producing a high-temperature equilibrium liquid; (2) cooling this liquid to below room temperature (the final temperature of the Kob–Andersen mixtures is below 0.2). The time step of the annealing is 1 fs, and it is 2 fs for cooling (0.001 and 0.002 for Kob–Andersen mixtures, respectively). The simulations employed periodic boundary conditions and used a Nose–Hoover thermostat, and most were performed at constant pressure and temperature; the 2D mixtures and the 3D Kob–Andersen mixture were simulated at constant volume and temperature, however.

The simulations were designed to numerically mimic the protocol of real DMS experiments. Thus a sinusoidal volume-preserving strain $\epsilon(t) = \epsilon_0 \sin(\omega t)$ is applied at selected times, with strain amplitude ϵ along the x or x – y direction of the simulation box (leading to the same results). The resulting shear stress σ is fitted by $\sigma(t) = \sigma_0 \sin(\omega t + \delta)$. The storage and loss moduli are calculated from $G' = \sigma_0/\epsilon_0 \cos(\delta)$ and $G'' = \sigma_0/\epsilon_0 \sin(\delta)$, respectively. The deformation amplitude must be small enough to be within the linear-response regime. To ensure this, we used a deformation of 1.4%; the two transverse dimensions were simultaneously decreased by 0.7% to maintain the volume. To strike a balance between system stability and simulation duration, a time step of 10 fs is utilized when probing the shear-mechanical properties. Supplementary Table 1 summarizes the details of the molecular dynamics simulations.

Cluster and percolation analysis

Our percolation analysis involves all particles by dividing them into two classes, mobile and immobile. The division is controlled by the magnitude of the displacement, u , of each particle over one molecular dynamics DMS cycle. To identify a suitable critical displacement to distinguish between mobile and immobile particles, u_c , the van Hove function $p(u) \equiv [P(u + \Delta u) - P(u)]/\Delta u$ is used, in which $P(u)$ is the cumulative distribution giving the probability of finding the value $X \leq u$, normalized according to $\int_0^\infty p(u) du = P(\infty) = 1$. The threshold displacement u_c is defined from the first minimum (or transition to long tail) of $p(u)$; by definition, all mobile particles have a displacement greater than u_c and all immobile particles a displacement smaller than u_c . A cluster consists of particles ‘close’ to neighbours of the same class (mobile or immobile), defined by reference to the all-particle radial distribution function $g(r)$: when the distance between two particles is below that of the first minimum of $g(r)$, r_c , the particles by definition belong to the same cluster. The values of u_c and r_c depend on the frequency, thermodynamic state point and system in question, but for all systems these

values are almost identical at different temperatures and frequencies (compare Supplementary Fig. 4). Supplementary Table 2 reports u_c and r_c for all mixtures.

Data availability

All of the data used to generate Figs. 1–6 are available via Zenodo at <https://doi.org/10.5281/zenodo.13925660> (ref. 96).

Code availability

The simulation package LAMMPS (<https://www.lammps.org>) was used for all molecular dynamics simulations. The code and scripts of the analysis are available via Zenodo at <https://doi.org/10.5281/zenodo.13925660> (ref. 96).

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Author contributions

H.-B.Y. and J.C.D. devised the project. L.G. and H.-B.Y. performed the simulations with input from T.B.S. and J.C.D. All authors wrote and revised the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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