

REVIEW ARTICLE | JULY 21 2026

Computational methods toward ultrastable glasses

Special Collection: [Manipulating the structure and stability of glasses](#)

Fabio Leoni  ; Misaki Ozawa  ; John Russo  ; Taiki Yanagishima  ; Andrea Ninarello  



J. Chem. Phys. 165, 031001 (2026)

<https://doi.org/10.1063/5.0341332>



Articles You May Be Interested In

Surface dynamics of glasses

Appl. Phys. Rev. (March 2022)

Understanding and engineering spontaneous orientation polarization in organic light-emitting devices

Chem. Phys. Rev. (May 2023)

3D-printed lithium-metal batteries: Multiscale architectures, hybrid technologies, and monolithic integration for next-generation energy storage

Appl. Phys. Rev. (October 2025)



The Journal of Chemical Physics

Special Topics Open for Submissions

[Learn More](#)

Computational methods toward ultrastable glasses

Cite as: J. Chem. Phys. 165, 031001 (2026); doi: 10.1063/5.0341332

Submitted: 30 April 2026 • Accepted: 29 June 2026 •

Published Online: 21 July 2026



View Online



Export Citation



CrossMark

Fabio Leoni,¹ Misaki Ozawa,² John Russo,¹ Taiki Yanagishima,^{3,4} and Andrea Ninarello^{1,5,a)}

AFFILIATIONS

¹ Department of Physics, Sapienza University of Rome, Piazzale Aldo Moro 2, 00185 Roma, Italy

² Univ. Grenoble Alpes, CNRS, LIPhy, 38000 Grenoble, France

³ Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan

⁴ Department of Physics, Tokyo Metropolitan University, Tokyo 192-0397, Japan

⁵ CNR Institute of Complex Systems, Uos Sapienza, Piazzale Aldo Moro 2, 00185 Roma, Italy

Note: This paper is part of the Special Topic on Manipulating The Structure and Stability of Glasses.

a) Author to whom correspondence should be addressed: andrea.ninarello@cnr.it

ABSTRACT

Ultrastable glasses, amorphous solids with exceptionally low-energy states and enhanced kinetic, thermodynamic, and mechanical stability, have long been a subject of intense experimental interest. Over the past decade, their computational realization has emerged as a major goal in condensed matter physics, as numerical methods can exploit unphysical moves to access deeply supercooled and nonequilibrium glassy states far beyond the reach of conventional cooling protocols, thereby providing key insights into the nature of the glass transition and amorphous states and enabling the design of mechanically robust glassy materials. In this review, we outline the key steps underlying the most effective algorithms developed across the field. For each approach, we discuss its efficiency, limitations, and physical interpretation. We finally present a comparative analysis of the stability achieved across these methods, with the aim of equipping both newcomers and experts with an intuitive and comprehensive understanding of the field's current state and the opportunities it presents.

© 2026 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/5.0341332>

I. INTRODUCTION

Glasses have accompanied human technology since antiquity and currently underpin applications ranging from optics to electronics. Despite their ubiquity, they remain conceptually intriguing because rigidity emerges without long-range order and the resulting materials are inherently out of equilibrium.^{1,2} A particularly active frontier is the exploration of ultrastable glasses (UG), amorphous states that lie much deeper in the energy landscape than those obtained by conventional cooling.^{3,4} Studying UG is fundamental from both theoretical and technological perspectives, as it sheds light on the nature of the glass transition and amorphous states while enabling the design of mechanically robust glassy materials for engineering applications.³ Understanding the computational routes by which such states can be generated and manipulated is the guiding question of this review.

A necessary starting point is to clarify what is meant by *stability* in a system that is intrinsically out of equilibrium. Unlike thermodynamically equilibrium materials, glasses require an operational notion that captures how long they persist, how deep they lie in the landscape, and how they respond to perturbations. Accordingly, stability is commonly characterized through three complementary perspectives, i.e., kinetic, thermodynamic, and mechanical, summarized schematically in Fig. 1.

The glass transition observed during the fast cooling of a liquid is phenomenologically characterized by a dramatic increase in viscosity, reaching values of order $\eta \approx 10^{12}$ Pa s, beyond which the material behaves as a rigid, disordered solid on experimental timescales.⁵ This kinetic arrest implies that the stability of conventional glasses is inherently limited by the time available for equilibration during preparation: as the glass transition is approached, relaxation times grow so rapidly that the system becomes trapped

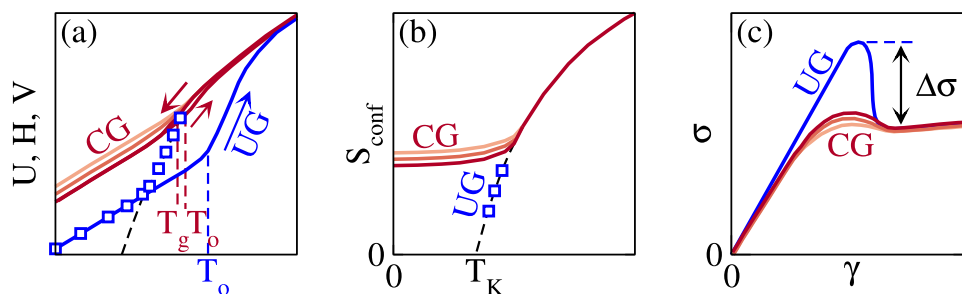


FIG. 1. (a) Kinetic (T_o) signatures of stability as represented by internal energy U , enthalpy H , or volume V vs temperature during heating ramps (right-up arrows), compared with cooling for a conventional glass (left-down arrow). T_g and T_o denote the glass transition temperature and the onset temperature, respectively. (b) Thermodynamic signature of stability as represented by the configurational entropy S_{conf} vs temperature T for a conventional glass (CG, dark red for low cooling rates and light red for high cooling rates) and an ultrastable glass (UG, blue squares). T_K denotes the Kauzmann temperature. (c) Mechanical signature of stability as represented by the stress-strain curve (σ vs γ). The stress overshoot $\Delta\sigma$ is shown for a conventional glass (CG) and an ultrastable glass (UG).

in out-of-equilibrium configurations before it can fully explore its energy landscape.⁶

The kinetic stability of a glass is naturally assessed through its response to heating protocols.^{7,8} A more stable glass resides deeper in the energy landscape and, thus, requires more time and thermal energy to relax. In practice, kinetic stability is quantified by the onset temperature T_o at which, during heating, physical quantities such as energy, enthalpy, or volume recover their equilibrium values, with higher T_o indicating greater stability, as shown in Fig. 1(a). T_o also slightly increases with an increasing heating rate. Originally introduced for conventionally prepared glasses, where slower cooling generically yields more stable states, this characterization remains equally valid for ultrastable glasses obtained through any preparation route.

From a thermodynamic perspective, stability reflects how deep a configuration lies in the potential or free-energy landscape.^{9–11} More stable glasses correspond to states trapped in deeper minima, characterized by lower energies and a reduced number of accessible configurations. In this picture, supported by theoretical frameworks that attribute a thermodynamic origin to glass formation,^{12–15} the configurational entropy S_{conf} decreases as the system explores progressively deeper regions of the landscape, as illustrated in Fig. 1(b). Extrapolating this trend suggests the Kauzmann temperature T_K ,¹⁶ where the configurational entropy would supposedly vanish, defining the hypothetical ideal glass. Thus, the lowest value of S_{conf} achieved can serve as a measure of thermodynamic stability.

Finally, stability can be probed through mechanical response. Under applied strain, glasses initially deform elastically before yielding through irreversible rearrangements. More stable glasses typically exhibit larger elastic moduli and higher stress overshoots $\Delta\sigma$, reflecting an increased resistance to plastic flow.¹⁷ This behavior is illustrated in Fig. 1(c), where the ultrastable glass shows a pronounced peak of σ compared to a conventional glass.

Taken together, these kinetic, thermodynamic, and mechanical viewpoints provide a unified picture: increasing stability corresponds to preparing amorphous states that persist longer before transforming, reside deeper in the energy landscape, and resist deformation more strongly.

From an experimental perspective, a major breakthrough that overcame the limitations imposed by extremely long relaxation times in conventional preparation methods came in 2007, when Swallen *et al.* demonstrated that physical vapor deposition (PVD) can produce glasses whose stability corresponds to thousands or even millions of years of conventional aging.⁷ In PVD, molecules are deposited onto a substrate held near an optimal temperature, where enhanced surface mobility allows them to equilibrate efficiently before being buried by subsequent layers. This layer-by-layer growth enables the formation of exceptionally well-packed amorphous states. PVD glasses became the first example of ultrastable glasses, i.e., states that display markedly lower energies, higher onset temperatures, reduced configurational entropy, and increased mechanical rigidity compared to conventionally cooled glasses. They also exhibit improved resistance to devitrification and reduced gas permeability, properties that are technologically relevant, for example, in organic electronic devices such as organic light emitting diodes (OLEDs).^{3,4}

At the same time, PVD glasses strongly motivated the development of computational strategies capable of producing ultrastable glasses *in silico*, allowing a microscopic understanding of the mechanisms underlying their formation. Indeed, molecular simulations provide atomistic access to the structure and dynamics of glass-forming systems, but they operate on length and time scales that are vastly shorter than those of laboratory experiments.

Even state-of-the-art molecular dynamics simulations reach equilibrium relaxation times of at most $\tau_\alpha \sim 10^{-6}$ – 10^{-3} s.^{2,18} Using the Maxwell relation $\eta \simeq G_\infty \tau_\alpha$, with a typical high-frequency shear modulus $G_\infty \sim 1$ – 10 GPa,⁴ these timescales translate into effective viscosities in the range $\eta \sim 10^3$ – 10^7 Pa s, between those of peanut butter and tar pitch at room temperature.¹⁹ This falls dramatically short of experimental glasses, whose relaxation times near T_g are of order 10^2 s, corresponding to the aforementioned viscosities of $\sim 10^{12}$ Pa s. The resulting mismatch between simulated and experimental timescales spans five to eight orders of magnitude, implying that conventional simulations probe dynamical regimes far less viscous and, hence, far less stable than those accessible in the laboratory. A further constraint is that simulations must carefully avoid crystallization, which is particularly delicate in simple or

weakly frustrated models, precisely those most commonly employed for their analytical tractability and conceptual transparency.

Overcoming these intrinsic limitations requires a change of perspective. Unlike experiments, simulations are not constrained to follow physical dynamics and can instead employ non-physical sampling strategies that accelerate the exploration of configuration space while preserving statistical consistency. Over the past two decades, this idea has led to a broad family of approaches. Early numerical efforts to reconcile experiments with simulations were strongly influenced by methods developed in the spin-glass community,²⁰ leading to the adoption of parallel tempering²¹ and mean-field-inspired approaches such as random pinning^{22–24} and, more recently, random bonding.²⁵ More than a decade ago, event-chain algorithms²⁶ pushed the boundaries of advanced Monte Carlo algorithms. In parallel, experimental advances in vapor deposition inspired computational analogs²⁷ as well as nonequilibrium protocols such as cyclic shear.^{28,29} A turning point came in 2017 with the optimization of swap Monte Carlo for polydisperse systems,³⁰ enabling the equilibration of supercooled liquids beyond experimentally accessible regimes and yielding glasses of unprecedented stability. This development underscored the importance of enhanced phase-space exploration, effectively introducing polydispersity as an additional control dimension. Subsequent developments along this line have introduced grand-canonical dynamics,³¹ in which particle attributes such as size are allowed to evolve dynamically to facilitate equilibration, and further algorithmic refinements.³² A complementary route, based on trajectory sampling,³³ targets rare dynamical fluctuations in configuration space, thereby granting access to exceptionally deep glassy states. Finally, more recently, machine-learning-based strategies, ranging from adaptive Monte Carlo schemes to generative models, have been explored to evaluate their potential for generating ultrastable configurations.³⁴

Some of these techniques now generate amorphous states whose stability rivals or even exceeds that of conventionally prepared experimental glasses and, in some cases, approaches that of ultrastable PVD glasses. For this reason, they are often described as producing ultrastable glasses *in silico*. Beyond their algorithmic significance, these methods provide a powerful route to probe deeply metastable regions of the landscape, enabling direct tests of theoretical ideas on configurational entropy, relaxation mechanisms, and mechanical response. This review focuses on these computational strategies, their physical principles, and the extent to which they allow simulations to close the stability gap with experiments. Each technique is presented according to a unified framework: we first provide a concise description of the method and its underlying principles (description), followed by a detailed account of its implementation (algorithm). We typically discuss its advantages, limitations, and performance relative to alternative approaches (considerations) and conclude with an overview of its applications, highlighting its adoption and evolution within the community (applications).

We organize this review into five broad categories. The first covers physical and quasi-physical preparation protocols, such as vapor deposition (Sec. II) and cyclic shear (Sec. III), which are the closest in spirit to experimental routes. The second category comprises equilibrium-sampling accelerators, including swap Monte Carlo (Sec. IV), cluster moves (Sec. V), and parallel tempering (Sec. VI), which employ unphysical yet statistically valid

moves to efficiently sample the Boltzmann distribution. The third covers phase-space and Hamiltonian modification methods, such as random pinning (Sec. VII) and random bonding (Sec. VIII), which stabilize glassy states by constraining or modifying microscopic degrees of freedom. The fourth includes target-oriented and nonequilibrium methods, ranging from structural and packing optimization (Sec. IX) to trajectory-ensemble approaches (Sec. X). Finally, the fifth category covers machine-learning-based methods (Sec. XI), which combine and extend ideas from all previous categories. However, this categorization should not be viewed as rigid, as several algorithms admit variants that may fall into different classes, for instance, equilibrium and nonequilibrium formulations, or schemes that enforce only global balance. A dedicated section (Sec. XII) provides a systematic comparison of the stability achieved by the different methods, summarizing results from the literature in a unified framework, complementing the primary metrics discussed above (and illustrated in Fig. 1). We conclude by discussing conceptual connections between algorithms and by outlining future directions toward even more efficient routes to ultrastable glasses.

II. VAPOR DEPOSITION

A. Description

Inspired by an established experimental technique,⁷ physical vapor deposition (PVD) involves depositing particles from a vapor onto a cold substrate, where controlled tuning of the deposition rate and substrate temperature enables the formation of ultrastable films.

B. Algorithm

Begin by preparing the substrate: (1) take a bulk configuration with a box size larger than spatial correlations (typically larger than $\sim 10\sigma$, with σ being the particle's size). Therefore, quench the bulk configuration below its glass-transition temperature (T_g) in the NPT ensemble at zero pressure in preparation for its exposure to vacuum. (2) Extract an xy -slab whose thickness in the z (deposition) direction

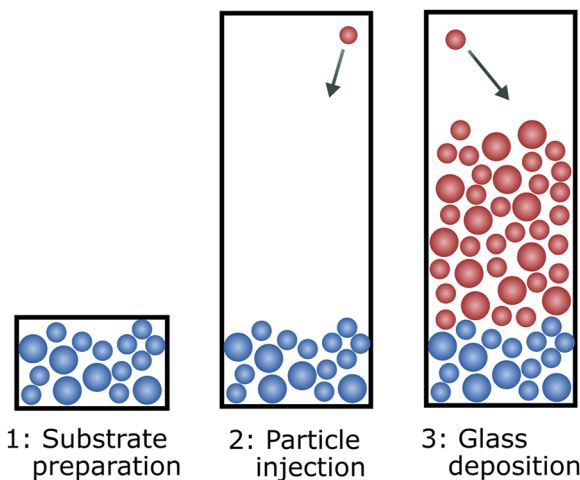


FIG. 2. Schematic representation of PVD. Blue and red particles represent the substrate and deposited particles, respectively.

exceeds the potential cutoff (see panel 1 in Fig. 2). (3) Embed this slab in an elongated simulation box exposed to vacuum, with open boundaries along z and periodic boundaries along x and y (see panel 2 in Fig. 2). (4) Next, initialize the deposition loop: inject with rate γ one or a small group of particles at a time from the top with velocities sampled from a high-temperature Maxwell distribution (considering also rotational degrees of freedom for non-spherical molecules) (see panel 3 in Fig. 2). The deposition rate γ , defined as the thickness of the deposited layer divided by the elapsed time, is an explicit control parameter of the algorithm together with the temperature of the substrate. Newly inserted particles are propagated in the NVE ensemble, while the pre-existing substrate is thermostatted in NVT in the range $0.8T_g - 0.9T_g$. Deposited particles are usually evolved in the NVE ensemble, since they thermalize to the substrate temperature between arrivals. Continue iterating insertion–integration cycles until the target film thickness is reached.

C. Considerations

The mechanism behind vapor deposition lies in the enhanced surface mobility of glass-forming materials.^{35–48} At the free surface, particles experience drastically faster diffusion than in the bulk, remaining mobile for several bulk relaxation times,^{7,49} allowing them to explore configuration space and relax into low-energy states before being buried by subsequent depositions. The dynamic acceleration extends a few molecular layers into the film, its penetration length being significantly larger than that of the structural inhomogeneities induced by the interface, so that the resulting ultrastable glass is formed in the subsurface region.⁴⁸ Consistently, the degree of ultrastability correlates with the ratio between surface, τ_{surf} , and bulk, τ_{α} , relaxation times.⁵⁰ Indeed, the most stable glass that can be prepared at a given substrate temperature is thought to be the equilibrium supercooled liquid, and its kinetic stability is characterized by τ_{α} . Accordingly, the maximum kinetic stability achievable for a specific material is constrained by the value of $\tau_{\text{surf}}/\tau_{\alpha}$, with lower ratios favoring enhanced stability. This ratio has been found to positively correlate with liquid fragility in several studies,^{4,50,51} suggesting that strong liquids are less effective at forming ultrastable glasses. Despite this trend, theoretical calculations³⁵ indicate that liquids spanning the full range of fragilities are, in principle, capable of producing highly stable glasses. As a result, fragility by itself does not constitute a universal predictor of glass stability.⁵⁰ Other favorable factors include low deposition rates⁷ (allowing equilibration at the surface), optimal substrate temperatures near $0.85T_g$,⁷ and molecular shape influencing the anisotropy,^{52–60} or orientational ordering, which can further enhance packing efficiency and kinetic stability. More recently, the elasticity of the substrate has been experimentally investigated as an additional parameter to enhance stability.⁶¹

A first important aspect of the substrate is its structure, which can influence the formation of the deposited layer depending on the material used. In experiments employing organic molecules such as indomethacin, the substrate is typically a crystalline solid, most commonly a silicon wafer, which provides a smooth and thermally stable surface for controlled deposition.³ Similar to experiments, organic molecules can be deposited on a crystal surface in simulations to form an ultrastable glass.⁶² However, when considering atoms, symmetrical or simple molecules, crystalline substrates can favor heterogeneous nucleation over a wide range of conditions.⁶³

In contrast, disordered substrates restrict nucleation to a narrower temperature range above T_g and tend to remain homogeneous, at least for simple liquids.⁴⁷

The typical control parameters optimized in simulations are the substrate temperature and the deposition rate. The optimal substrate temperature is usually located between 0.8 and $0.9T_g$, similar to what is found experimentally. The deposition rates attainable in simulations (of the order of $\gamma \sim 10^7$ nm/s) are orders of magnitude larger than in experiments ($\gamma \sim 0.1 - 1$ nm/s for organic glasses⁴), and this is the major reason why simulations can only attain moderate ultrastability: while the gain in stability for experimental vapor deposited glasses over annealed glasses is of ~ 8 orders of magnitude, for simulations it reduces to ~ 3 orders of magnitude.^{64,65}

Finite system sizes further restrict the bulk region of the film, which is not affected by surface effects, and limit the exploration of film-thickness-dependent properties such as those observed in experiments for deposited layers as thick as hundreds of nm,⁶⁶ while typical simulated layers reach a thickness of ~ 10 nm.

At temperatures close to T_g , diffusion from the substrate may occur; in such cases, harmonic springs should be applied to confine the substrate molecules.

The properties of the deposited species (atoms or molecules) play a key role in determining the characteristics of the resulting layer, including its structural complexity. For spherically symmetric interactions, the deposited structure is, on average, isotropic.⁴⁸ In contrast, asymmetric molecules forming organic glasses such as TPD tend to produce anisotropic molecular packing.^{3,4} Another structural feature influenced by the nature of the deposited species is the development of porosity: porous structures have been observed, for instance, in films formed by silica molecules⁶⁷ and by water.⁶⁸

Despite the limited stability range achievable in simulations, vapor deposition is the main algorithmic route to glass formation with a direct experimental counterpart, providing a unique bridge between computational and laboratory studies of ultrastable glasses.

D. Applications

The concept of producing ultrastable glasses by physical vapor deposition (PVD) originated from experiments by Ediger and co-workers, who demonstrated that vapor-deposited organic films could reach equilibrium states equivalent to those of glasses aged for millennia.³⁹ This finding immediately raised fundamental questions about how far out-of-equilibrium systems could approach equilibrium by tuning kinetic pathways during formation.

Shortly thereafter, computer simulations began to reproduce and rationalize these observations. De Pablo and collaborators implemented atomistic and coarse-grained deposition protocols, showing that controlled deposition on cold substrates indeed leads to glasses of enhanced stability.^{27,39,41,57,60,69–72} Since then, many works have explored the connections between vapor deposition and theoretical ideas of glass equilibration via surface mobility and tried to extend the range of systems capable of forming ultrastable glasses, including low-fragility liquids⁵⁰ and tetrahedral, network-forming materials.⁵¹ Other works have explored the structural properties of deposited glasses and their connection to bulk quantities such as viscosity and relaxation dynamics. A related but much less explored route is the formation of ultrastable glasses by precipitation from

solution,⁷³ where a glass-forming solute grows at a solvent interface and the enhanced interfacial mobility enables the formation of highly stable amorphous structures.

III. CYCLIC SHEAR

A. Description

Under cyclic, or oscillatory, shear deformation, the energy of a glass can either decrease or increase depending on key parameters such as strain amplitude.^{28,74} Similar to the compaction of granular materials under cyclic shear,⁷⁵ tapping,⁷⁶ or compression–decompression cycles,⁷⁷ the deformation amplitude must be carefully chosen to enhance stability by balancing overaging and rejuvenation effects.⁷⁸

B. Algorithm

To prepare a glass with enhanced stability using cyclic shear (schematically illustrated in Fig. 3), one typically proceeds as follows: (1) initialize a periodic simulation box containing N particles and quench the system to obtain a glass; (2) impose shear deformation in a strain-controlled setting with appropriate boundary conditions, such as Lees–Edwards periodic boundary conditions;^{79–81} and (3) apply an oscillatory strain of amplitude $\gamma_{\max}$ slowly in a quasi-static manner.

The energy is typically monitored stroboscopically each time the strain γ returns to zero. It decreases as the number of cycles increases when $\gamma_{\max}$ is below the yielding value. The final configuration at $\gamma = 0$ is then used to assess the stability of the glass.

C. Considerations

Deformation, such as shear, is widely used to probe the mechanical properties of materials, including steady-state flow,⁸² oscillatory response,⁸³ shear-band formation,^{29,84} failure of amorphous solids,^{85,86} shear-melting and resolidification,⁸⁷ and the yielding transition.^{17,74,88–92}

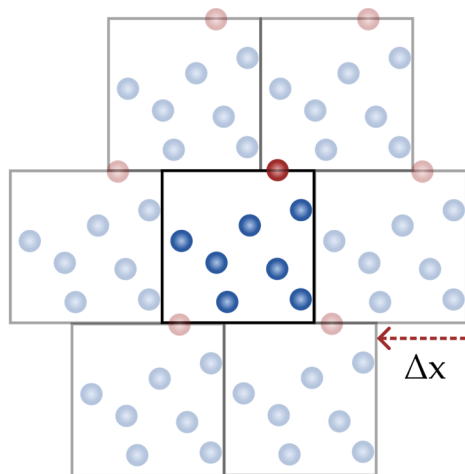


FIG. 3. Schematic illustration of cyclic shear applied to a glass-forming system under Lees–Edwards periodic boundary conditions.

When the applied shear strain is sufficiently large, the glass yields and enters a flowing state. Interestingly, however, when cyclic shear is applied with an amplitude below the yielding point, it can anneal the system and enhance its stability.^{28,74,93,94} The underlying reason is that shear-induced plastic rearrangements can progressively drive the system toward lower-energy states under periodic deformation. As the number of cycles increases, the system approaches a steady state. The final energy level depends on the strain amplitude $\gamma_{\max}$: the energy decreases with increasing $\gamma_{\max}$ up to the yielding point, above which the system yields and undergoes rejuvenation. Therefore, the strain amplitude must be carefully tuned to maximize the annealing effect. However, for a given shear protocol, cyclic annealing cannot reduce the energy below a certain threshold. To reach deeper energy levels, alternative annealing methods, such as thermal annealing, must be employed.^{95,96}

D. Applications

Most studies of cyclic shear have been performed either using athermal quasi-static shear (AQS) simulations or finite-strain-rate simulations at very low shear rates,^{28,74,95,96} with only a few exceptions considering finite-temperature, finite-strain-rate protocols such as SLOD.²⁹ In AQS simulations, small strain increments are applied, each followed by energy minimization,⁹⁷ and these two steps are repeated iteratively. This protocol isolates purely mechanical responses, such as plastic events, avalanches, and yielding, from thermal effects, thereby revealing how strain drives transitions between energy minima in a complex energy landscape.

Building on these insights, cyclic or oscillatory shear was first studied in the context of memory encoding, whereby the system retains information about the training amplitude in the energy landscape.²⁸ It was later exploited as a means of annealing glasses by tuning the strain amplitude.^{29,74} Recent work⁹⁸ showed that multidirectional oscillatory shear reaches lower steady-state energies than unidirectional shear, suggesting that carefully designed shear cycles can anneal glasses and enhance their stability rather than degrade it.

A shear protocol applied to soft glass suspensions⁹⁹ found a connection between stability and microscopic mechanical features such as narrowing and symmetrization of local stress distributions, a trend also observed in polydisperse glassformers,¹⁰⁰ reinforcing the link between stability and stress homogenization.

Hyperuniform states, which may be associated with enhanced glass stability,^{101,102} have been observed in cyclically driven glasses below yielding, whereas above yielding, the system exhibits enhanced heterogeneity and fluctuations.¹⁰³

Recent works have explored the connection between shear deformation and active forces in relation to glass stability. In Ref. 104, internal activity in amorphous solids is shown to play a role similar to external shear in controlling glass stability. Moderate activity anneals the system into deeper energy minima, thereby enhancing stiffness, whereas excessive activity fluidizes the system and erases memory of the preparation history. This activity-induced tuning also drives a ductile-to-brittle transition, mirroring shear-driven glasses: low activity leads to homogeneous flow, whereas high activity promotes shear-band-mediated failure. Priya and Karmakar¹⁰⁵ emphasized the critical role of activity in shaping the mechanical memory of ultrastable glasses, particularly through its regulation of shear-band formation and evolution.

Cyclic shear is also relevant to experiments, particularly in soft-matter rheology¹⁰⁶ and fatigue failure in metallic glasses.¹⁰⁷

IV. SWAP MONTE CARLO

A. Description

The Swap Monte Carlo (SMC) algorithm extends standard Monte Carlo dynamics by supplementing local particle displacement moves with random diameter-swap moves, both of which accepted or rejected according to the Metropolis criterion to preserve detailed balance. SMC has demonstrated high efficiency in simulations of simple glass-forming mixtures of multicomponent and polydisperse particles.

B. Algorithm

SMC enables the generation of equilibrium simulations of supercooled liquids, which can subsequently be driven out of equilibrium to form glasses through rapid cooling or compression. During SMC simulations, at each Monte Carlo step, in addition to standard particle displacement moves, a swap move is attempted with probability p . A swap move proceeds as follows: (1) Two particles are randomly selected, with diameters σ_i and σ_j . (2) The particle diameters are exchanged, and the resulting energy difference between the new and old configurations is computed: $\Delta U = U_{\text{new}} - U_{\text{old}}$. (3) The move is accepted or rejected according to the Metropolis criterion,

$$p_{\text{acc}}(i \leftrightarrow j) = \min \{1, \exp(-\beta \Delta U)\}. \quad (1)$$

Those three steps are illustrated schematically in Fig. 4.

C. Considerations

To ensure that crystallization does not occur, throughout the simulation, simple observables such as the potential energy must be monitored. Structural relaxation times τ_α can then be extracted from time-dependent correlation functions, such as the self-intermediate scattering function or overlap function, as a function of temperature or density. These relaxation times are used both to verify equilibration and to quantify the dynamical speedup achieved relative to standard MC simulations.

An equivalent implementation of the SMC algorithm can be obtained by exchanging particle positions rather than diameters. However, this approach prevents the tracking of

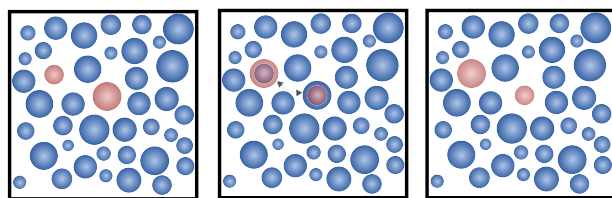


FIG. 4. Schematic representation of the Swap Monte Carlo (SMC) algorithm. In addition to standard particle displacement moves, pairs of particles are randomly selected and their diameters are exchanged with a given probability. The proposed swap is accepted or rejected according to the Metropolis criterion based on the resulting energy change.

single-particle dynamics, since particles undergo arbitrarily large effective displacements during swap moves.

The SMC algorithm constitutes one of the most significant advances in the numerical study of glass-forming systems and supercooled liquids. Using Metropolis-accepted displacement and swap moves, SMC preserves detailed balance while enabling non-local exploration of configuration space. This simple yet effective approach establishes SMC as the most efficient known method for equilibrating polydisperse mixtures across a broad range of dimensions, owing to its independence from geometric constraints. It yields an enormous dynamical speedup, with the most effective systems exhibiting estimated gains of at least ten orders of magnitude, although accurate quantification remains challenging, as it requires extrapolating standard dynamical timescales into thermally inaccessible regimes, and the true speedups could in fact be considerably larger.

The algorithm's efficiency can be improved by adjusting the swap fraction p to reduce relaxation times and limiting swaps to particles with diameter differences smaller than an optimized value. Using highly polydisperse or carefully designed discrete (e.g., ternary) particle size distributions is essential to suppress crystallization and achieve a large dynamical gain. For continuous polydisperse systems, the optimal polydispersity for hard spheres and repulsive particles is $\sim 23\%$ of polydispersity,³⁰ while discrete mixtures can achieve comparable performance through optimized composition and size ratios.^{108–110} Moreover, as a Monte Carlo-based method, its stochastic and inherently serial nature makes large-scale parallelization nontrivial.¹¹¹

D. Applications

The method has its origins in early lattice MC studies of binary alloys, where swap-like exchanges were used to compute order parameters and phase transitions.¹¹² It was later generalized to off-lattice systems such as Lennard-Jones microclusters¹¹³ and hard-sphere mixtures.¹¹⁴ The first applications to glass-forming liquids employed binary mixtures, demonstrating the potential of swap dynamics to reach deeply supercooled states.¹¹⁵ Its algorithmic efficiency has also made it a valuable tool for mapping phase diagrams^{108,116} and testing theoretical frameworks such as the random first order transition theory.¹¹⁷ However, these early binary models were later shown to crystallize easily.¹¹⁸ A 2017 reassessment of SMC³⁰ identified optimal parameters (particle size distribution, softness, and non-additivity) that suppress crystallization and yield extremely high equilibration speedups, redefining glass simulation standards.

One of the key achievements of SMC is its contribution to understanding the entropy crisis in glass formation, as it enabled the observation of a pronounced decrease in configurational entropy over an unprecedented cooling range⁶⁵ and, in two dimensions, hinted at a possible zero-temperature transition characterized by vanishing entropy and a growing static length scale.¹¹⁹ SMC also provides insights into the evolution of the rugged energy landscape upon deep supercooling.¹²⁰

SMC has been instrumental in elucidating the vibrational properties of ultrastable glasses, revealing that quasi-localized modes (soft excitations distinct from phonons) govern low-frequency vibrations, thermal anomalies, and tunneling phenomena.^{121–124} These soft modes generally follow $D(\omega) \sim \omega^4$, with deviations linked

to stability and system size.^{124,125} Sound attenuation serves as a key probe of these excitations, with more stable glasses exhibiting reduced damping and delayed quartic scaling.^{124,126} Furthermore, tunneling two-level systems, long associated with glass anomalies, are increasingly linked to soft quasi-localized modes, although their connection remains subtle and nonlinear.^{122,123,127}

SMC has been crucial as well for exploring thermodynamic transitions predicted by high-dimensional mean-field glass theory. In three-dimensional hard-sphere glasses, simulations have revealed signatures of a Gardner-like transition,^{128,129} at which the glass free-energy basin fragments into a hierarchically organized, marginally stable landscape. However, softer particle models show no clear evidence,^{127,130} and in two dimensions, the phenomenon appears only as a strong crossover.¹³¹ Moreover, simulations across dimensions $d = 3$ –10 reveal finite-dimensional remnants of mean-field Ising-like spinodal criticality, strongly softened in low dimensions and gradually approaching mean-field behavior as d increases.¹³²

The rheological behavior of glasses, including their yielding and failure, has been extensively studied through athermal quasi-static simulations (see Sec. III for further details), with SMC enabling access to ultrastable configurations. These simulations revealed a brittle, discontinuous yielding transition with macroscopic failure reminiscent of experimental observations.¹³³ Further work suggested that the crossover from brittle to ductile behavior may correspond to a critical point,¹³⁴ although this interpretation remains debated.¹³⁵ Recent studies have also examined cyclic deformation⁹⁶ and plastic events¹³⁶ to deepen understanding of this transition.

The role of the SMC algorithm in the glass transition debate has generated significant theoretical interest, as it has been used as a probe to distinguish between competing theories of glass formation. One line of interpretation holds that SMC's efficiency undermines thermodynamic, cooperative theories such as RFOT, since altering purely local dynamical rules dramatically changes the relaxation time.¹³⁷ However, several works push back on this view: within RFOT, the speedup can be understood as a postponement of the onset of glassy dynamics through “crumbling metastability,” leaving the underlying free-energy landscape intact.¹³⁸ At the mean-field level, replica liquid theory confirms that SMC shifts the dynamical transition point relative to standard Monte Carlo, suggesting a modification rather than a refutation of thermodynamic glass theory.¹³⁹ More explicitly, SMC dynamics can be governed by an effective potential that stabilizes configurations at lower energies, shifting the glass transition to lower temperatures, with the magnitude of the effect tied to polydispersity.³¹ A mode-coupling-inspired framework further shows that size swaps open an additional relaxation channel for density fluctuations, moving the dynamic glass transition to higher volume fractions in hard-sphere mixtures.¹⁴⁰ Simulations have further confirmed this shift in the mode-coupling temperature induced by SMC.¹⁴¹ A unifying perspective comes from the notion of time-reparameterization softness:¹⁴² local constraints and global landscape complexity are not mutually exclusive but complementary, as mean dynamics is invariant under time reparameterization, while the landscape governs the structure of correlations, and SMC-like algorithms exploit precisely this softness. Taken together, these works suggest that the sensitivity of the glass transition to dynamical rules reflects not the irrelevance of thermodynamic complexity, but a subtle interplay between local kinetics and the free-energy landscape.

V. EVENT-CHAIN CLUSTER MOVES

A. Description

One of the central advantages of Monte Carlo simulations over molecular dynamics is the possibility of employing non-physical collective or *cluster moves* that dramatically accelerate relaxation. A wide variety of cluster moves have been developed for fluids and complex systems. In what follows, we focus on the family of *lifted* Markov chain algorithms,^{143,144} which have recently shown great promise for tackling exceptionally hard sampling problems, including glass-forming systems.³² For simplicity, we limit our discussion to the case of hard interactions, but generalizations to arbitrary potentials are available.^{145–147}

B. Algorithm

Lifted Monte Carlo algorithms proceed through an irreversible Markov chain in an extended configuration space. We illustrate the construction for two classes of collective moves: translational updates, realized through Event-Chain Monte Carlo (ECMC),¹⁴⁴ and compositional updates, realized through the Collective Swap (cSwap) algorithm.³² The two schemes can also be combined in hybrid implementations.

- (1) Begin by defining an extended configuration space by introducing additional degrees of freedom (the *lifting variables*) that govern how the dynamics unfolds (first column in Fig. 5). In ECMC, these variables consist of the index i of the active particle and a propagation direction $\mathbf{d}$ (the colored particle and the arrow in the top panel in Fig. 5), while in cSwap, the lifted variable is an index i identifying a particle within an array where all diameters are sorted in increasing size (the colored particle in the bottom panel in Fig. 5).

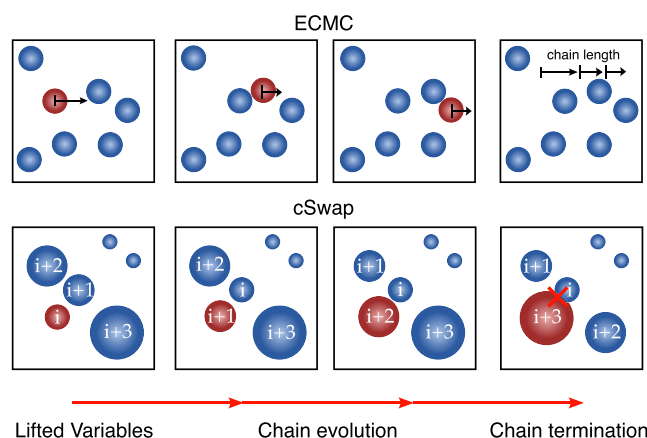


FIG. 5. Schematic representation of lifted Monte Carlo dynamics for Event-Chain Monte Carlo (ECMC, top row) and Collective Swap (cSwap, bottom row). In ECMC, the lifted variables consist of the active particle index i and the propagation direction $\mathbf{d}$. The active particle moves deterministically along $\mathbf{d}$ until a collision occurs, at which point the lifting rule transfers motion to the collided particle, maintaining the same direction. In cSwap, the lifted variable corresponds to the index i in the ordered diameter array. The active particle i sequentially exchanges its diameter with those of neighboring particles ($i + 1$, $i + 2$, ...) until an overlap event terminates the chain.

- (2) Evolve the variables deterministically according to the chosen dynamics (second column in Fig. 5). In ECMC, the active particle i moves along $\mathbf{d}$ with constant velocity until an event, such as a collision, occurs. When periodic boundary conditions are employed, the propagation direction is typically chosen from the set $\{+\mathbf{e}_x, +\mathbf{e}_y, +\mathbf{e}_z\}$, cycling through the Cartesian axes to ensure isotropic sampling. In cSwap, the evolution takes place in composition space rather than real space: the active particle i successively exchanges its diameter with its right-hand neighbor in the sorted array, and each accepted swap immediately updates the configuration before the next attempt.
- (3) Define a lifting rule specifying how activity is transferred when deterministic propagation is interrupted (third column in Fig. 5). In ECMC, upon a collision, the moving particle stops and the collision partner becomes active, inheriting the same propagation direction. In cSwap, when a swap attempt leads to an overlap, the chain terminates, and the activity label shifts to a neighboring particle, either to the left or to the right in the sorted array.
- (4) Terminate each chain after a prescribed cumulative displacement (in ECMC) or after the first rejected exchange (in cSwap), forming a collective update that satisfies global balance with respect to the Boltzmann distribution (last column in Fig. 5).
- (5) Ensure ergodicity by periodically resampling the lifting variables. In ECMC, select a new active particle and propagation direction after each chain, while in cSwap, uniformly reassign the activity index i along the ordered array with a small probability $p_r \sim 1/N$. Together, these steps define a complete lifted Monte Carlo cycle capable of generating irreversible yet statistically exact trajectories through configuration space.

C. Considerations

The dynamics of the lifted variables intentionally violate detailed balance, yet obey the more general condition of global balance, ensuring that the steady-state distribution remains Boltzmann.^{144,146,148–150} Although the resulting trajectories are irreversible and, therefore, unphysical, they often yield substantially faster equilibration than standard, reversible Monte Carlo schemes.

A second major advantage of the lifted approaches described above is that they are *rejection-free*: all proposed micro-moves are accepted by construction so that the acceptance rate does not decay as the system becomes dense or strongly correlated. In Event-Chain Monte Carlo (ECMC), this feature leads to dramatic gains in sampling efficiency. For instance, ECMC achieves speedup factors on the order of 10^3 with respect to standard Metropolis Monte Carlo for hard-sphere systems and remains one of the most efficient algorithms available for dense fluids.¹⁵¹ In two-dimensional polydisperse disks, the ratio of relaxation times between ECMC and standard Monte Carlo decreases from roughly 22 to 10 as the packing fraction increases, demonstrating that the advantage persists even near the glassy regime.³²

A similar acceleration is observed in the compositional space explored by the Collective Swap (cSwap) algorithm. Compared to standard swap Monte Carlo, cSwap yields a dramatic reduction in equilibration times, with speedup factors that *increase* with volume

fraction, reaching values of about 40 at the highest packing fractions studied.³² This inverted trend, opposite to that of ECMC, suggests that lifted dynamics in composition space become particularly efficient as steric constraints intensify.

Several extensions have further broadened the applicability of lift algorithms. The introduction of the *factorized Metropolis filter* allows ECMC to be generalized beyond hard interactions, making it applicable to continuous,¹⁴⁵ long-range potentials^{152,153} and to parallel programming.¹⁵⁴

D. Applications

The development of non-local Monte Carlo methods traces back to the pioneering work of Swendsen and Wang,¹⁵⁵ who introduced a cluster algorithm capable of suppressing critical slowing down in spin systems near continuous phase transitions. This demonstrated that non-physical, collective updates, if properly constructed to respect detailed balance, could drastically accelerate equilibration in strongly correlated regimes.^{156–159}

The concept of irreversibility in Monte Carlo dynamics emerged later with the introduction of algorithms such as Event-Chain Monte Carlo (ECMC).¹⁴⁴ they replaced stochastic trial moves with deterministic, rejection-free propagation in an extended configuration space that satisfies global rather than detailed balance. Originally developed for hard-sphere and hard-disk systems, ECMC contributed to solving a long-standing problem of the nature of the liquid-to-hexatic transition in two dimensions.²⁶

The application of lifted algorithms to the study of ultrastable glasses is a more recent development.^{32,147} The Collective Swap (cSwap) algorithm extends the idea of event-driven, rejection-free dynamics to composition space. Together, these advances mark the convergence of ideas from critical phenomena, nonequilibrium statistical mechanics, and glass physics into a unified framework of lifted Monte Carlo algorithms for the efficient sampling of complex free-energy landscapes.

VI. PARALLEL TEMPERING

A. Description

Parallel tempering is an advanced Monte Carlo sampling method that runs multiple simulations in parallel at different temperatures, enabling the system to overcome kinetic barriers and more efficiently explore the complex energy landscape of supercooled liquids.

B. Algorithm

The algorithm is schematically shown in Fig. 6.

- (1) Initialize the system: Begin by preparing a disordered initial configuration of the system. Launch a dynamical simulation, which can be either Monte Carlo or molecular dynamics, at a chosen reference temperature T_0 . At this stage, the system explores the phase space at a high enough temperature to avoid being trapped in local energy minima, ensuring a broad sampling of configurations.
- (2) Create temperature replicas: Duplicate the current configuration and initiate a second simulation at a lower temperature, $T_1 < T_0$. Each replica now evolves independently according to its assigned temperature.

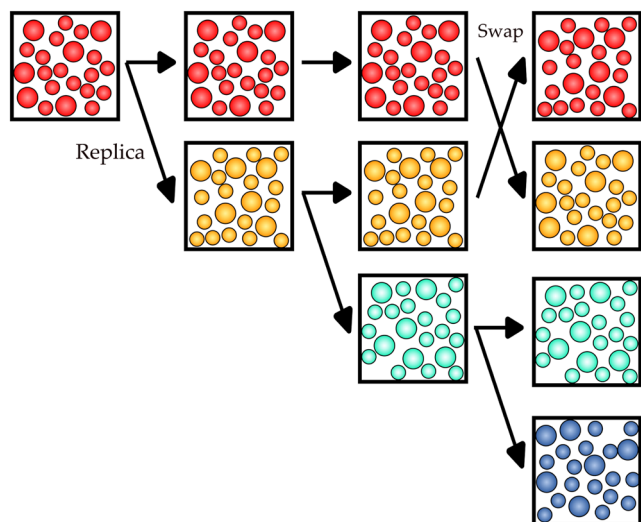


FIG. 6. Schematic representation of the parallel tempering algorithm. From left to right: initialize the system, generate replicas at different temperatures, construct a temperature ladder, and perform configuration exchanges between replicas to enhance sampling efficiency.

- (3) Build a temperature ladder: Continue this process iteratively, creating a series of n replicas down to the target temperature T_{n-1} . This set of simulations at different temperatures forms a *temperature ladder*, which is crucial for the parallel tempering method: higher-temperature replicas facilitate barrier crossing, while lower-temperature replicas provide detailed sampling of low-energy configurations.
- (4) Perform configuration exchanges: At regular intervals of N_{swap} simulation steps, attempt to exchange configurations between replicas at adjacent temperatures. The swaps are governed by a Metropolis-like acceptance criterion,

$$p_{\text{acc}}(j \leftrightarrow j \pm 1) = \exp[-(\beta_j - \beta_{j \pm 1}) \Delta U], \quad (2)$$

where $\Delta U = E_{\text{pot}}(x_j) - E_{\text{pot}}(x_{j \pm 1})$ is the difference in potential energy between the two configurations being swapped, and $\beta_j = (k_B T_j)^{-1}$ is the inverse temperature of replica j . This exchange mechanism allows lower-temperature replicas to escape local minima by swapping with higher-temperature replicas, enhancing sampling efficiency.

- (5) Iterate and equilibrate: Repeat the simulation and swap procedure for sufficiently long times so that all replicas can equilibrate and explore their respective configuration spaces thoroughly. The combination of independent evolution at different temperatures and periodic exchanges ensures that the overall ensemble represents the correct Boltzmann distribution at each temperature.

C. Consideration

Parallel tempering is a powerful sampling method for supercooled liquids, allowing equilibration at very low temperatures and

thereby enabling the production of stable glassy states. Its key advantage is the ability to cross large energy barriers by exchanging configurations between replicas at different temperatures. In simple terms, a configuration trapped in a narrow basin at low temperature can be swapped to a higher temperature, where it can freely move across the landscape into another basin and then return to low temperature, now trapped in a different and previously inaccessible region. This leads to improved equilibration and more accurate estimation of thermodynamic and structural properties in deeply supercooled regimes. However, parallel tempering also has limitations: it requires the simultaneous simulation of multiple replicas, increasing computational cost, and the efficiency of exchanges depends sensitively on the choice of temperature ladder, with poorly chosen spacing leading to low swap acceptance rates. To optimize parallel tempering, one can carefully tune the temperature ladder to maintain moderate acceptance rates (typically 20%–50%), balance the number of replicas against computational resources, or employ adaptive schemes that adjust temperatures dynamically during the simulation.

Another computational method, called population annealing, can be viewed as a version of parallel tempering in which configuration exchanges are achieved entirely via probabilistic reweighting instead of running separate simulations at each temperature. In this method, a large ensemble of configurations is initialized at a high temperature $\beta_1 = (k_B T_1)^{-1}$ and gradually annealed to lower temperatures in small steps. At each step, configurations are reweighted according to $W_i = \exp[-(\beta_j - \beta_{j+1}) E_{\text{pot}}(x_i)]$ and resampled proportionally to W_i . This enables efficient exploration of low-temperature states without the explicit need to run computationally demanding simultaneous simulations at multiple temperatures, combining the benefits of enhanced sampling and barrier crossing.

D. Applications

Parallel tempering was first introduced in the spin-glass community,^{20,160} where it also became known as the replica Monte Carlo method²⁰ or replica exchange.¹⁶¹ Its earliest applications to structural glass formers involved simulations of small binary soft-sphere systems with $N = 36$ particles,¹⁶² later extended to significantly larger systems of up to $N = 1000$ particles.²¹ These pioneering studies reported only a limited speedup, typically on the order of one to two decades. The method became particularly attractive with the increasing availability of parallel computing resources, as the replicas at different temperatures can be simulated largely independently and efficiently distributed across multiple processors. In the context of supercooled liquids, parallel tempering was initially employed mainly for equilibration^{21,163} and, subsequently, used in investigations of random pinning¹⁶⁴ and in measurements of the point-to-set correlation length.^{165,166} More recently, it has served as a benchmark for assessing modern enhanced-sampling strategies, including swap Monte Carlo and normalizing-flow-based methods.¹⁶⁷

VII. RANDOM PINNING

A. Description

Random pinning stabilizes a system by artificially freezing a subset of particles, rather than by lowering the temperature. More specifically, a fraction of particles is randomly selected from an equilibrium supercooled liquid configuration, and their positions

are permanently pinned. The stability of the remaining mobile (unpinned) particles is then examined. As the concentration of pinned particles increases, the dynamics of the mobile particles slow down and the system becomes progressively more stable.

B. Algorithm

The random pinning method starts from an equilibrium bulk configuration of a glass-forming liquid at low temperature, irrespective of the specific interaction potential type. From this configuration, a fraction of particles are randomly selected, and their positions are permanently frozen (or pinned),^{22,24,164,168} as schematically illustrated in Fig. 7. Alternatively, one can significantly increase the mass of the selected particles.¹⁶⁹ We then assess the stability, for example, by examining the enhancement of relaxation times of the remaining mobile (unpinned) particles in equilibrium,¹⁷⁰ monitoring hysteresis in heating-cooling cycles,²³ and probing the mechanical response.¹⁷¹ As one can imagine, due to the presence of pinned particles, the dynamics of the unpinned particles become progressively more glassy as the fraction of pinned particles increases. In this sense, random pinning provides an artificial way to solidify the system by introducing the concentration of pinned particles, c_{pin} , as an additional control parameter.

We note that the enhancement of stability induced by random pinning becomes more pronounced when the initial bulk configuration is taken at a lower temperature.^{24,164}

It is also important to emphasize that if the pinned particles are selected from a dense, thermally equilibrated configuration, the remaining mobile particles remain in thermal equilibrium immediately after pinning,^{172,173} although with enhanced stability. In contrast, if the pinned particles are chosen, e.g., in a purely random (Poisson) manner, thermal equilibrium is no longer guaranteed, and such an approach offers no algorithmic advantage.

C. Considerations

The enhanced stability induced by random pinning can be interpreted from the perspective of the potential energy landscape.

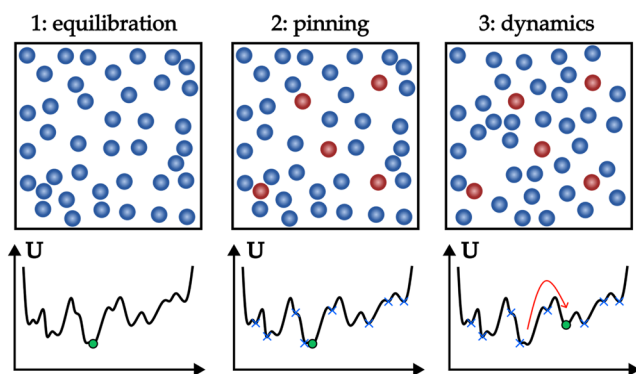


FIG. 7. Starting from an equilibrium liquid configuration (top left), a fraction of particles is randomly pinned (shown in red, top middle), and the dynamics of the remaining mobile particles are, subsequently, examined (top right). Bottom: Corresponding schematic illustration of how random pinning modifies the potential-energy landscape. The accessible configuration space becomes restricted (indicated by blue crosses) upon the introduction of pinned particles, effectively increasing the energy barriers separating metastable states.

Consider an initial bulk equilibrium configuration located at a local minimum of the energy landscape, corresponding to a relatively high-energy basin, as schematically illustrated in Fig. 7. Introducing pinned particles effectively restricts the accessible phase space of the system. Increasing the fraction of pinned particles progressively restricts the number of accessible metastable states available to the remaining mobile particles, thereby reducing the configurational entropy. As a result, the effective glass transition temperature shifts to higher temperatures (or lower densities), allowing the system to remain equilibrated deeper in the glassy regime. In this constrained energy landscape, configurations correspond to relatively deeper minima, which manifests as enhanced thermodynamic and kinetic stability and a pronounced slowdown of structural relaxation.

D. Applications

The random pinning protocol was first introduced by Kang Kim to study glassy dynamics by using the concentration of pinned particles as an additional control parameter, alongside temperature.¹⁶⁸ This protocol has subsequently been generalized to other geometries, such as walls and cavities, in order to study the influence of pinned particles, including the extraction of associated correlation length scales.^{117,165,172} It was shown that thermal equilibrium for the unpinned particles is maintained when the pinned particles are selected from a dense, equilibrated bulk configuration.^{172,173} Cammarota and Biroli theoretically demonstrated that an equilibrium ideal glass (Kauzmann) transition can be achieved through random pinning,²² a prediction that was later validated by molecular dynamics simulations.^{24,164} Thus, random pinning provides a unique approach to accessing an equilibrium glass state (where the configurational entropy vanishes) in computer simulations, without the need to explicitly solve the difficult equilibration or optimization problem.

Since then, various aspects of randomly pinned glass formers have been investigated, including their thermodynamics and dynamics in the supercooled liquid regime,^{170,174–178} non-equilibrium heating-cooling cycles,²³ vibrational properties,^{179,180} and mechanical responses.^{171,181}

It is also worth mentioning that random pinning can be realized experimentally in colloidal glasses, where the positions of selected particles can be frozen using optical tweezers.^{182,183} For molecular systems, a similar idea has been explored by considering mixtures of two substances with different masses to mimic the pinning situation.^{184,185}

VIII. RANDOM BONDING

A. Description

Starting from a low-temperature configuration of a monomer (particle) system, pairs of particles are randomly selected and permanently bonded. The stability of the resulting molecular glass former is then investigated.

B. Algorithm

First, a low-temperature thermal equilibrium configuration of a particulate system is prepared (e.g., via standard molecular dynamics annealing). Starting from such a “monomer” configuration, one monomer is randomly selected, and a second monomer is chosen

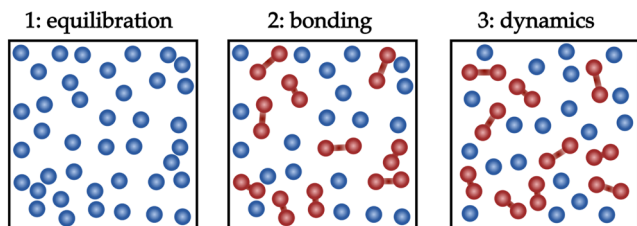


FIG. 8. Starting from an equilibrium liquid configuration (left), a fraction of particle pairs is permanently bonded (middle), forming a stable molecular glass former. The resulting bonded system exhibits enhanced stability, characterized by significantly slower dynamics (right).

from its neighbors, typically defined as non-bonded particles located within the first coordination shell in the radial distribution function. A permanent bond is then introduced between this pair of monomers,²⁵ forming a “dimer.” For bonding, one may employ either rigid-body constraints, such as those implemented in the RATTLE algorithm,¹⁸⁶ or harmonic potentials with sufficiently large spring constants.

This random pairing-and-bonding procedure is repeated until a desired fraction of dimers is obtained, which defines a control parameter analogous to the pinning concentration in random pinning protocols. As schematically shown in Fig. 8, this process yields a densely packed dimer configuration characterized by the dimer concentration c_{bond} .

It is important to emphasize that the resulting stability is enhanced when the initial (monomer) configuration is prepared at a lower temperature and when c_{bond} is larger, akin to the behavior observed in random pinning.^{22,24,164} We also note that, although previous work has focused on randomly bonded dimers, the method can naturally be extended to trimers, polymers, or more complex molecular shapes.

C. Considerations

Random bonding is conceptually inspired by the random pinning method. The physical essence behind the success of random pinning lies in the introduction of constraints in phase space, which effectively narrows the accessible configurations (see Fig. 7). In the case of random bonding, the frozen degree of freedom is not the particle position itself but rather the interparticle distance between selected pairs, leading to the idea of bonding. In contrast to random pinning, random bonding does not break the translational invariance of the system, an important property of bulk materials. Consequently, this approach allows one to study mechanical responses such as brittle yielding and the emergence of sharp shear bands,²⁵ phenomena that are typically absent in the yielding of randomly pinned glasses.¹⁷¹

As with random pinning, it can be shown that if bonding pairs are chosen completely at random from all possible monomer pairs in the simulation box, the resulting molecular glass former remains mathematically guaranteed to be in thermal equilibrium immediately after bonding.¹⁸⁷ However, if bonding pairs are chosen among neighboring monomers, which is physically more reasonable, this selection introduces a bias that formally violates the equilibrium guarantee. Nevertheless, numerical studies have demonstrated that

the resulting configurations remain nearly equilibrated, showing only negligible deviations from equilibrium.¹⁸⁷ Having said this, equilibrium guarantees are not a relevant issue when studying ultrastable glasses in non-equilibrium states.

D. Applications

The random bonding protocol was introduced in Ref. 25, where its stability was assessed through heating–cooling cycles, mechanical responses under shear deformation, and liquid-state dynamics. In particular, Ref. 25 generated deeply annealed, nearly equilibrium configurations corresponding to relaxation times that are $\sim 10^7$ times longer than those accessible by standard molecular dynamics simulations. Moreover, non-equilibrium mechanical deformation tests revealed a pronounced stress overshoot, characterized by a stress-overshoot index $(\sigma_{\text{max}} - \sigma_{\text{min}})/\sigma_{\text{min}}$ of ~ 1.4 , where σ_{max} denotes the peak stress and σ_{min} denotes the steady-state stress after the overshoot. Following this study, the issue of strict equilibration has been examined more thoroughly, both theoretically and numerically.¹⁸⁷ In addition, random bonding has been used to investigate how constraining the degrees of freedom modifies the fraction of unstable modes and their connection to glassy dynamics.¹⁸⁸

It was also suggested in Ref. 25 that random bonding could be realized experimentally with existing techniques, for example, by suddenly inducing attractive interactions between patchy colloids via changes in salt concentration or by applying ultraviolet irradiation in colloidal systems with DNA linkers.

IX. STRUCTURAL OPTIMIZATION

A. Description

The unprecedentedly deep equilibration enabled by the swap algorithm is largely due to the availability of unphysical moves, i.e., the switching of particle sizes between particles. In a generalization of this approach, one can consider a situation where such variables were not only allowed to be swapped but also freely evolved. There are two ways in which this might be applied. In one, local particle sizes evolve in equilibrium with the chemical potential associated with a particular particle population,¹⁸⁹ i.e., the definition of an equation of motion for particle size (grand canonical dynamics). The other is where unphysical moves might artificially optimize the system to have specific physical features that might be associated with ultrastability, e.g., homogeneity of virial stress.¹⁰⁰ This second, non-equilibrium, approach is described below.

B. Algorithm

Starting from an initial, quenched glassy state, a local particle parameter is adjusted to achieve a target characteristic. A specific example might be in a Lennard-Jones system, when the target is a system with homogeneous local virial stress over space, as illustrated in Fig. 9. At each step, particles are individually resized by some value to reduce the variation in the virial stress over the particle population. The modified glassy state is then re-quenched to remove any force imbalances generated by the modifications. On comparing the new state to the original glass, the process may be repeated if the standard deviation in virial stress contributions is reduced. If not, the step size is likely too large and to be scaled down. Convergence is arbitrarily defined but may be reached when the standard deviation

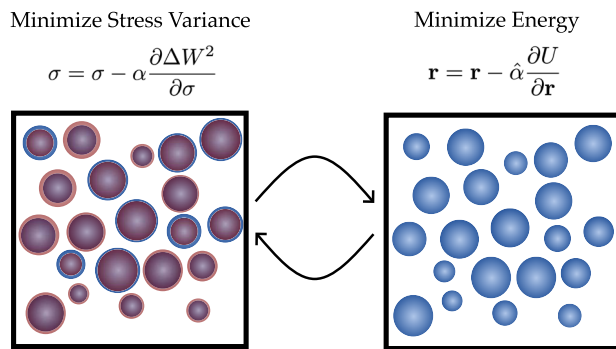


FIG. 9. Structural optimization procedure combining stress homogenization and energy minimization. Starting from a glassy configuration, particle sizes σ are first updated to reduce spatial variations in the local virial stress. The system is then re-equilibrated by minimizing the potential energy through updates of particle positions. These two steps are iterated until convergence, yielding configurations with reduced stress heterogeneity.

reaches some set value or further reduction of particle size modifications does not yield a reduction in the standard deviation of the virial stress.

Regarding the selection of the step size, there are many parallels with standard optimization techniques, such as steepest descent or conjugate gradient. The step size may be scaled by the susceptibility of the target parameter, much like step sizes are made smaller in an energy optimization when an energy minimum is approached. For example, instead of resizing individual particles by a set, small amount (e.g., $0.01\sigma_i$, where σ_i is the local Lennard-Jones size) to correct for locally low/high virial stress, one might scale the step size by considering how far the local virial stress is from the population average and scale it by the gradient of the local virial stress as a function of particle size. In practice, this is further scaled by some factor (usually < 0.1) to ensure that modifications are not too large, such that unstable oscillations about a minimum are seen.

C. Considerations

The most significant limitation of these techniques is the change in the specific particle population of the initial glassy state. While it may be shown that the modified particle population quenched in a conventional manner gives rise to qualitatively different, rejuvenated glasses,^{100,101} it should be noted that properties that are specific to certain particle populations may be modified.

Another consideration is the nature of the steps themselves. While the steps in grand-canonical dynamics¹⁸⁹ may be defined based on equilibrium with a set particle population, allowing for a form of detailed balance, optimization based algorithms are not constrained. Thus, the path taken during structural optimization as described here is strictly out of equilibrium, although it may remain close to it in some specific cases. This renders any analogies between optimization based methods and swap configurations non-trivial.

D. Applications

Early work by Brito, Wyart, and Lerner framed the swap algorithm as a special instance of grand-canonical dynamics,^{31,189} where particle sizes may also be adjusted during equilibration within the

constraint of a chemical potential defined particle size population with which a glassy state is in equilibrium. They noticed that relaxation of the strict particle population in this manner led to the generation of ultrastable glasses in significantly less time than what is required with slow-quenching protocols.¹⁸⁹

In parallel with this work, two independent strands of research led to the development of algorithms that leveraged modifications to particle size that were unconstrained by equilibrium with a particle reservoir. One was the study of glassy states with features of hyperuniform packings.¹⁹⁰ It was argued that amorphous hyperuniform packings might define a special instance of a well-equilibrated glass, a “perfect” glass.¹⁹¹ To this end, an algorithm designed to create large, sparse hyperuniform packings¹⁹² was applied to create dense soft sphere packings.¹⁹³ Unlike for sparse hard-spheres, modifications to soft spheres led to mechanically unstable states, necessitating mechanical relaxations and iterative adjustments until convergence was reached. The glasses generated in this work featured spatially correlated soft modes that broadly spanned the system size.

The correlation between such states and stability against glass aging was established through another body of work, where localized density inhomogeneities were correlated with the incidence of collective avalanche-like relaxation events in a deeply quenched glass.¹⁹⁴ In an effort to create glasses that were more stable to thermally activated relaxation, an algorithm inspired by Ref. 193 was applied to modify quenched repulsive glasses to create glasses with a sharper distribution of local densities. The result was states that did not feature any of the avalanche relaxation events seen in the original glasses¹⁰¹ and exhibited qualitatively different devitrification behavior when driven to crystallize through templating.¹⁹⁵

While thermodynamic quantities such as bond-orientational order remained largely unchanged, it was found that the mechanical environment of individual particles was significantly modified to “homogenize” the response of the glass to perturbations. To achieve this more directly, an algorithm was proposed targeting not the local density but the local virial stress. Importantly, homogenization of the local virial stress may be achieved in a much wider range of particulate systems. Applying the algorithm to binary glasses revealed a significant drop in energy and enhanced kinetic stability, which mirrored what is seen in physical vapor deposition glasses.^{48,100}

Target variables for this approach are not limited to density and virial stress. For example, optimization of local packing efficiency has given rise to ultrastable glasses with exceptionally low energy, as reviewed.¹⁹⁶ Tong and Tanaka optimized a local packing parameter based on bond-angles,¹⁹⁷ creating amorphous states that have many of the salient properties of a Debye solid.¹⁰² Such properties were mirrored by density uniform packings with stronger convergence criteria¹⁹⁸ than in the original work.¹⁰¹ In work on a two-dimensional glass former, Bolton-Lum *et al.*¹⁹⁹ modified local particle sizes to create strictly triangulated packings with a convergence in relaxation time with temperature following a Volger–Fulcher–Tammann (VFT) form approaching the ideal glass transition. These recent works exemplify efforts to explore the potential of target-oriented, non-equilibrium pathways to create ultrastable glasses.

Finally, the role of polydispersity remains less well understood than in swap-based approaches. Recent work has indeed shown that enhanced hyperuniformity and local ordering do not necessarily

imply high thermodynamic stability,²⁰⁰ concluding instead that the relaxation of particle-size degrees of freedom is the key ingredient for achieving ultrastability.

X. TRAJECTORY SAMPLING

A. Description

Differently from schemes that alter the dynamics, trajectory sampling leaves the equations of motion unchanged while still giving access to long lived glassy states that are otherwise difficult to sample. Here, we focus on the s -ensemble, where a Markov chain is performed in the space of trajectories and where each path is assigned a statistical weight (the s variable) that penalizes dynamical activity.²⁰¹ In this way, the method selects trajectories that are dynamically inactive and deeply metastable, even though such trajectories are rarely observed in equilibrium.

B. Algorithm

The preparation of a biased trajectory in the s -ensemble proceeds as follows. (1) One starts from an initial trajectory of fixed duration t_{obs} , generated by the unbiased dynamics (left panel in Fig. 10). This initial trajectory (denoted as X) acts as the starting point for the Markov chain in trajectory space. (2) A time slice t_s is chosen at random along the trajectory, and the configuration at that time is used as the shooting point. (3) The momenta at the shooting point are perturbed by a small random change, producing a slightly modified state from which new dynamics can be launched (middle panel in Fig. 10). (4) From this state, the equations of motion are integrated forward and backward (by inverting momenta) to reconstruct a full trajectory of duration t_{obs} . This yields a trial trajectory X' . (5) The dynamical observable associated with the s -ensemble is

then evaluated along the trial trajectory. This observable is called the *activity* and measures the total amount of particle motion along the trajectory, for example, through the sum of particle displacements between successive time intervals. (6) The trial trajectory is accepted with a probability that depends on the change in activity, using a Metropolis rule,

$$p_{\text{acc}} = \min \left\{ 1, e^{-s (C[X'] - C[X])} \right\},$$

where $C[X]$ is the activity of trajectory X . (7) If the trial trajectory is accepted, it replaces the previous trajectory as the next state of the Markov chain; otherwise, the previous trajectory is retained. Repeating this procedure generates a sequence of trajectories sampled from the biased ensemble.

C. Considerations

In the s -ensemble, the biased distribution of trajectories is defined by the weight $\exp[-sC]$, where C is the activity. The field s acts as a conjugate variable to the activity, in close analogy with a Legendre transform: increasing s suppresses particle motion and favors the sampling of slow, inactive trajectories. In this way, the method continuously tunes the frequency of rare dynamical fluctuations that are essentially inaccessible in equilibrium sampling.

A central outcome is the identification of two distinct dynamical regimes.^{201–203} For small values of s , the sampled trajectories remain liquid like and active, while for sufficiently large s , the ensemble becomes dominated by trajectories with strongly suppressed mobility. These regimes can be represented in proposed dynamical phase diagrams, where active and inactive trajectories occupy separate regions of trajectory space and are separated by sharp crossovers that become more pronounced at larger observation times. Such diagrams provide a useful framework for interpreting how rare dynamical fluctuations organize in space and time.

In addition to the standard shooting move, many other trajectory moves have been proposed within the transition path sampling framework.²⁰⁴ These include shifting moves, which translate the whole trajectory forward or backward in time; partial path moves, which regenerate only a segment of the trajectory; and replica exchange in trajectory space, which facilitates sampling across different biasing fields or observation times. Such extensions can improve ergodicity and efficiency, especially when the dynamical landscape contains multiple competing inactive pathways or when the observation time is large.

A practical limitation of trajectory sampling is the restricted system sizes that can be treated. Each step of the Markov chain requires the generation and storage of an entire trajectory of duration t_{obs} , so the computational cost grows quickly with both system size and observation time. As a result, existing studies focus on relatively small systems, of the order of a few hundred particles.

D. Applications

The conceptual foundation of trajectory based sampling originates from the development of transition path sampling, which introduced the idea that rare dynamical events can be studied by constructing a Markov chain in the space of trajectories.²⁰⁵ This framework established shooting and shifting moves as practical tools for generating new dynamical paths without imposing a predefined

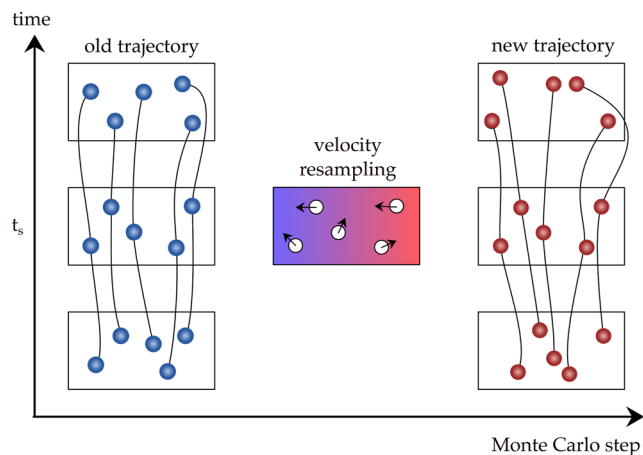


FIG. 10. Schematic representation of trajectory sampling in the s -ensemble. Each column represents a full dynamical trajectory of duration t_{obs} , with particle positions shown at a sequence of discrete physical times. A shooting move selects a configuration at an intermediate time slice (center panel), perturbs the particle momenta, and regenerates the forward and backward parts of the trajectory to produce a new trial path. Repeating this procedure constructs a Markov chain in trajectory space, from which inactive and long lived trajectories are sampled with a weight that depends on their dynamical activity.

reaction coordinate and demonstrated that entire trajectories can be treated as statistical objects.

Work on kinetically constrained models, analyzed within dynamical large deviation theory, showed that rare fluctuations of activity can be accessed by introducing a field that biases trajectories according to their dynamical activity.^{206,207} This led to the formulation of the s -ensemble, in which the trajectory distribution displays distinct active and inactive regimes interpreted as a form of phase coexistence in space-time.²⁰² The same methodology was subsequently extended from simplified lattice models to atomistic glass formers, where analogous transitions between liquid like and inactive trajectories were identified.^{201,203}

A further development was the introduction of the μ -ensemble, which biases trajectories according to their structural content, for example, the time spent in locally favored structures.^{33,208–210} This approach established a direct link between dynamical large deviations and specific structural motifs associated with slow relaxation. For example, in Ref. 33, the authors were able to equilibrate a Kob–Andersen mixture down to $T \approx 1.2T_K$, where T_K is the (estimated) Kauzmann temperature.

XI. MACHINE LEARNING APPROACHES

The recent surge of machine learning (ML) techniques has revolutionized many areas of science, including computational physics and related fields. Consequently, there is growing anticipation that ML approaches will eventually tackle complex simulation problems in glass physics and even generate ultrastable glasses, much as ML has surpassed conventional or human-designed methods in numerous other domains. As of the time of writing this review, however, no ML-based methods are yet available that can generate ultrastable glasses in a way that significantly outperforms existing standard techniques. Nevertheless, we present a few pioneering works that may inspire future developments toward this ambitious goal.

Since ML-based approaches for glass generation are still in their infancy and no ready-to-use preparation protocols are currently available, this section departs from the structure used previously. Instead, we divide the discussion into three paragraphs, each presenting the essential information on the method and its main outcomes to date.

A. Inverse design of ultrastability

A novel strategy for generating mechanically stable glass configurations was proposed in Ref. 211, which formulates the problem as an inverse-design task enabled by machine learning. In particular, Wang and Zhang²¹¹ developed a Monte Carlo-like update algorithm that iteratively modifies particle configurations so as to minimize the propensity for plastic activity under deformation, thereby steering the system toward increasingly mechanically stable glassy states.

Measuring plasticity for every Monte Carlo trial configuration would be computationally prohibitive (typically quantified through the non-affine displacement measure $D_{\min}^2$ ²¹² under applied deformations). To overcome this bottleneck, the authors first trained a *graph neural network* (GNN) to predict local plastic propensity directly from a static configuration, without explicitly performing deformation simulations. The training data for this surrogate model

were generated from independent molecular dynamics simulations in which plastic events were computed athermally under a set of controlled deformations. Once trained, the GNN serves as a fast and accurate surrogate for evaluating plasticity: it provides instantaneous predictions of local $D_{\min}^2$ -like quantities, effectively replacing the need for repeated MD simulations. This surrogate model is then embedded within a Monte Carlo loop, where the GNN-predicted plasticity plays the role of an “effective Hamiltonian” that defines the acceptance probability of configuration updates. In practice, the algorithm seeks configurations with progressively lower effective Hamiltonian (predicted plasticity) during the Monte Carlo

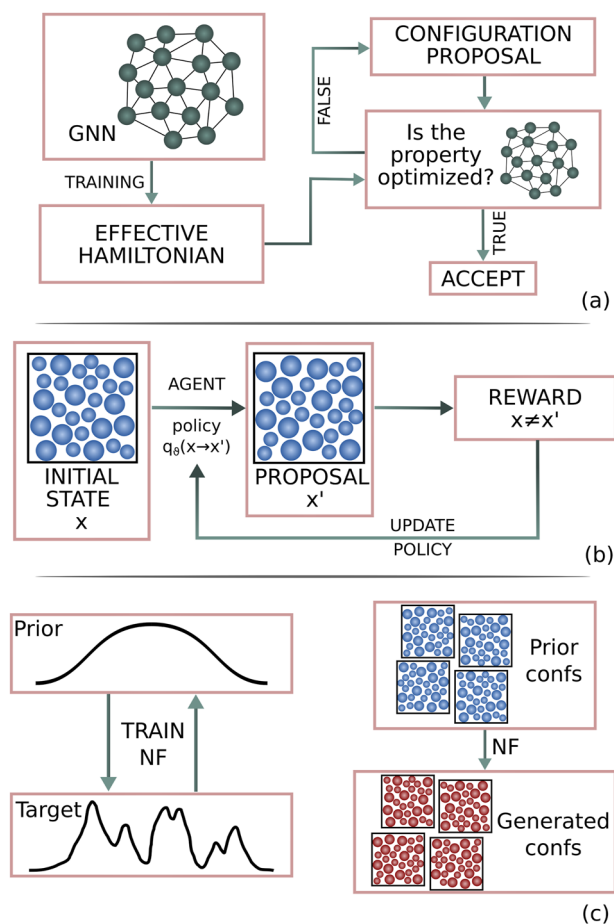


FIG. 11. Three schematic examples of ML algorithms used to study supercooled liquids and glasses. (a) A graph neural network is first trained to learn the mapping between configurations and one or more target properties, which define an effective Hamiltonian to be minimized. The trained GNN is then employed as a fast surrogate model that evaluates this effective Hamiltonian for any given configuration and is, subsequently, used within a Monte Carlo-like update scheme. (b) A reinforcement-learning-inspired strategy in which, starting from an initial state x , an agent generates a proposed configuration x' according to a policy $q_\theta(x \rightarrow x')$. A reward is then computed, and the policy is updated accordingly. This procedure is repeated recursively. (c) A normalizing flow is trained to map a simple prior distribution onto a desired target distribution. After training, it generates configurations in the target distribution by applying this learned transformation to samples drawn from the prior.

sampling. The structural updates themselves are performed using swap Monte Carlo, allowing the algorithm to explore configuration space far more efficiently than conventional particle displacements alone. Through this MC-like optimization process, the method progressively identifies configurations with reduced plastic propensity, yielding glass states that are more stable against mechanical deformation.

An overview of the algorithm is shown in Fig. 11(a). This method has been successfully applied to a binary $\text{Cu}_{64}\text{Zr}_{36}$ metallic glass model,²¹¹ enabling targeted optimization of plastic propensity from MD deformation data. Notably, it reveals non-trivial glassy states, such as configurations that are geometrically stable yet energetically metastable—states that conventional thermal protocols cannot easily access. The study also demonstrates a degree of transferability across different alloy compositions.

Despite its promise, the approach has limitations. Its performance depends critically on the accuracy and generalization ability of the GNN, which may bias the sampling toward restricted regions of the configuration landscape, especially when separated by high energy barriers. Moreover, generating sufficiently diverse and labeled training data requires substantial computational resources.

Possible future developments include optimizing multiple target properties simultaneously, online retraining of the GNN as new configurations are discovered to improve predictive accuracy, incorporating reinforcement-learning-based or more advanced search strategies, extending the inverse-design protocol to other material families via transfer learning, and improving scalability or integrating the method with experimental feedback loops.

B. Machine learning assisted MC

One promising application of ML techniques for generating stable glass configurations is ML-assisted Monte Carlo sampling, where the proposal moves in the MC algorithm are optimized using a learning strategy. Within the general formulation of the Monte Carlo framework,²¹³ the acceptance probability p_{acc} for updating a configuration x to a new configuration x' is given by

$$p_{\text{acc}} = \min \left\{ 1, \frac{q(x' \rightarrow x) \rho(x')}{q(x \rightarrow x') \rho(x)} \right\}, \quad (3)$$

where $\rho(x)$ is the target Boltzmann distribution and $q(x \rightarrow x')$ is the proposal distribution for generating candidate configurations x' from x . Equation (3) is a generalization of the standard Metropolis algorithm, which corresponds to the case of symmetric proposals, $q(x' \rightarrow x) = q(x \rightarrow x')$. Asymmetric updates introduced through a general proposal distribution $q(x \rightarrow x')$ allow significantly greater flexibility in sampling configurations. Such flexibility has long been exploited through human-designed, system-specific insights to improve sampling efficiency.^{213,214} ML-assisted Monte Carlo aims to take this idea further by constructing or learning an optimal proposal distribution $q_{\theta}(x \rightarrow x')$, parameterized by a set of trainable parameters θ , with the goal of accelerating sampling.²¹⁵

While this approach has been used in various domains to tackle diverse problems, a recent and promising work implemented it specifically for investigating supercooled liquids.³⁴ In this paper, by Galliano, Rende, and Coslovich,³⁴ the authors employed a policy-guided Monte Carlo scheme inspired by reinforcement learning (RL).²¹⁶ In this approach, the proposal distribution $q_{\theta}(x \rightarrow x')$

(referred to as the *policy* in RL terminology) is learned so as to maximize the magnitude of decorrelation (the *reward*) induced by the Monte Carlo update, such as particle displacements. The proposal distribution $q_{\theta}(x \rightarrow x')$ is parameterized in a physically motivated manner, combining displacement and swap moves, while also incorporating structural information such as local forces and local energies. A schematic illustration of the algorithm is provided in Fig. 11(b). The hope is that ML can discover more flexible and effective proposal distributions compared to traditional (or human-designed) displacement or swap moves.

The authors reported an efficiency gain of roughly two orders of magnitude for soft-sphere models, whereas no significant improvement was observed for the Kob–Andersen mixture, indicating that the performance of the method is model-dependent. Although the performance is still limited and far from enabling the preparation of ultrastable glasses, the proposed framework is nevertheless promising. In particular, the design and parameterization of $q_{\theta}(x \rightarrow x')$ offer substantial room for improvement and further development, especially by incorporating collective moves using deep learning.²¹⁷

C. Sampling with generative models

Generative models are making a huge impact across virtually all domains of science, engineering, and society. In essence, they learn or approximate an underlying probability distribution $\rho(x)$, which is generally unknown, from data by constructing a parameterized model $\rho_{\theta}(x)$ such that $\rho_{\theta}(x) \approx \rho(x)$. Using generative models to accelerate Monte Carlo sampling has recently gained significant attention.^{218–220} The core idea is that once the model $\rho_{\theta}(x)$ is learned, it can efficiently generate new configurations distributed according to $\rho_{\theta}(x)$, typically from Gaussian noise (or a high-temperature configuration). Leveraging this property, one can construct the proposal distribution in Eq. (3) as

$$q(x \rightarrow x') = \rho_{\theta}(x'). \quad (4)$$

In other words, a new configuration x' can be proposed directly from the generative model, without referring to the current configuration x at all. Moreover, in the ideal limit where the generative model perfectly reproduces the target distribution, $\rho_{\theta}(x) = \rho(x)$, the Metropolis–Hastings acceptance probability becomes unity. In practice, the closer $\rho_{\theta}(x)$ is to $\rho(x)$, the higher the acceptance rate. Thus, at each Monte Carlo step, the simulation can propose nearly decorrelated configurations that are already very close to equilibrium.

In Ref. 167, Jung, Biroli, and Berthier employed a specific class of generative models, namely, *normalizing flows* (NFs), for a ternary Lennard–Jones mixture. As schematically shown in Fig. 11(c), normalizing flows construct an explicit mapping between a simple *prior* distribution $\rho_p(x)$, which is easy to sample (corresponding to high-temperature configurations), and an approximation $\rho_{\theta}(x)$ of a more complex target distribution $\rho(x)$ of interest. Importantly, the target low-temperature distribution $\rho(x)$ does not need to be known or sampled in advance. Instead, the flow is trained iteratively using configurations drawn from an easier-to-sample reference distribution together with a reweighting scheme that incorporates the Boltzmann weight of the target temperature.

Once trained, the normalizing flow enables efficient sampling of configurations approximately distributed according to $\rho_{\theta}(x)$ by

transforming samples drawn from the prior distribution $\rho_p(x)$. These generated configurations can then be used as proposal moves according to Eq. (4), combined with a Metropolis–Hastings correction to ensure unbiased sampling of the target Boltzmann distribution.

Normalizing flows show strong potential for studying supercooled liquids, providing substantial speedups over conventional molecular dynamics and achieving performance competitive with advanced sampling techniques such as parallel tempering and population annealing, while still remaining inferior to swap Monte Carlo. Their main current limitation lies in the accessible system size.¹⁶⁷ Extending these approaches to larger system sizes is, therefore, an obvious and important next step.

A complementary line of work has developed global annealing procedures in which a generative model is trained sequentially at decreasing temperatures to learn the low-temperature distribution and is coupled to local MC steps, thereby bridging generative methods and ML-assisted Monte Carlo sampling. This approach, however, has not yet been applied to structural glass formers.²²¹

XII. STABILITY ACROSS METHODS

In Sec. I, we outlined several approaches commonly used to characterize glass stability and presented a few representative quantitative indicators. We now systematically compare the computational strategies discussed thus far for producing ultrastable glasses by reviewing the stability metrics and quantitative results reported in the literature. Because *ultrastability* does not admit a unique definition and different algorithms emphasize distinct aspects of stability, direct comparisons are not straightforward. To address this issue, we organize the reported measures within a unified framework based on kinetic, thermodynamic, and mechanical properties of glasses. In each case, stability is quantified through relative measures, typically by comparing properties of ultrastable glasses to those of conventional glasses or the equilibrium liquid branch. For clarity, representative quantitative indicators are summarized in

Table I, which enables a direct, although necessarily approximate, comparison between methods. While not exhaustive, this overview includes the most widely used and informative measures, providing a practical reference for assessing the relative performance of different algorithms in generating ultrastable glasses.

Kinetic stability is associated with the dynamical response and mobility of the glass system, particularly during reheating and melting processes, i.e., under non-equilibrium protocols. A widely used measure of kinetic stability is the *stability ratio* S , defined phenomenologically and commonly employed in experiments. It is expressed as the ratio between the isothermal melting or transformation time t_m , i.e., the time required for a glass to transform into the supercooled liquid at a temperature $T > T_g$ (typically $T/T_g \approx 1.25$), and the structural relaxation time τ_α measured at the same temperature, $S = t_m/\tau_\alpha$. Values of S exceeding 10^3 – 10^4 are generally considered characteristic of experimental ultrastable glasses. Values observed in simulations of ultrastable glasses prepared via physical vapor deposition (PVD) and random pinning are around $S \approx 10^3$, as reported in Refs. 23, 41, and 51. Swap Monte Carlo simulations have produced even higher ratios, reaching $S \approx 10^4$ up to even 10^5 in small systems,²²⁵ whereas trajectory-based sampling methods typically yield more modest values,²⁰⁸ $S \approx 10$.

A common phenomenological measure of kinetic stability compares the onset temperature, T_o , of a glass, as previously introduced, to the glass transition temperature, T_g , both determined at the same heating and cooling rates. For conventionally prepared glasses, this ratio depends on the applied cooling and heating rates, which affect both T_g and T_o , although their values remain very close. For ultrastable organic glasses prepared via vapor deposition, typical experimental values are $T_o/T_g \approx 1.05$.^{3,229} In simulations, where rates are many orders of magnitude higher than in experiments, much larger ratios have been reported: $T_o/T_g \approx 1.19$ for PVD glasses,⁵¹ $T_o/T_g \approx 1.39$ for virial-homogenized configurations,¹⁰⁰ and $T_o/T_g \approx 1.75$ for random-bonding protocols.²⁵ Indeed, in addition to increasing with stability, T_o also increases with heating rate, preventing direct comparison of the ratio T_o/T_g across different conditions.

TABLE I. Comparison of stability in different glassy systems obtained with different methods. The symbols are defined as follows: S is the kinetic stability ratio. T_o , T_g , T_f , and T_K are, respectively, the onset, the glass transition, the fictive, and the Kauzmann temperatures. $\mathcal{R}_s$ is the configurational entropy ratio as defined in the main text. τ_α is the relaxation time at the lowest accessible temperature for different methods. $\Delta\sigma/\sigma$ quantifies the stress overshoot at yielding. Some values were not explicitly reported in the original works and have, therefore, been inferred from the available data.

Algorithm	Kinetic stability	Thermodynamic stability	Mechanical stability
Conventional	$S \approx 7 \times 10^{1222}$	$\mathcal{R}_s \approx 0.69^{223}$	$\Delta\sigma/\sigma \approx 0.5^{135}$
PVD	$T_o/T_g \approx 1.19,^{51} S \approx 3 \times 10^{251}$	...	...
Cyclic shear	...	...	$\Delta\sigma/\sigma \approx 0.9^{224}$
Swap	$S \approx 1.25 \times 10^{4225}$	$T_f/T_g \approx 1.18,^{226} \mathcal{R}_s \approx 0.2^{227}$ $\tau_\alpha(\text{MC})/\tau_\alpha(\text{Swap}) \approx 10^{1030}$	$\Delta\sigma/\sigma \approx 1.7^{133}$
Structural optimization	$T_o/T_g \approx 1.39^{100}$	...	$\Delta\sigma/\sigma \approx 3.0^{189}$
Cluster moves	...	$\tau_\alpha(\text{Swap})/\tau_\alpha(\text{Chain}) \approx 10^{232}$	...
Random pinning	$S \approx 4.5 \times 10^{223}$	$\mathcal{R}_s \approx 0 - 0.2^{24,179}$	$\Delta\sigma/\sigma \approx 1.2^{171}$
Random bonding	$T_o/T_g \approx 1.75^{25}$	...	$\Delta\sigma/\sigma \approx 1.4^{25}$
Parallel tempering	...	$\tau_\alpha(\text{MC})/\tau_\alpha(\text{PT}) \approx 10^{228}$	...
Trajectory sampling	$S \approx 10^{1208}$	$\mathcal{R}_s \approx 0.43,^{33} T_K/T_{eq} \approx 0.81^{33}$	...

An alternative approach to probe kinetic stability involves analyzing the melting dynamics using the Avrami exponent^{230–233} n_A , which characterizes the rate of phase transformation over time. It should be noted, however, that the exponent depends not only on the melting rate but also on factors such as the dimensionality of growing liquid clusters. While an Avrami exponent exceeding the standard value $d + 1$ (with d being the spatial dimension) may indicate enhanced stability relative to conventional glasses, as found for a vapor deposited ice model with $d = 3$ ($n_A \approx 4.4$ ²³³) or for soft repulsive spheres in $d = 2$ dimensions ($n_A \approx 4.5$ ²³¹), its value alone is insufficient to unambiguously determine the underlying transformation mechanism. Therefore, its interpretation must be supported by additional structural or dynamical evidence,²³³ and for this reason, it is not included in Table I.

Thermodynamic stability is associated with fundamental quantities such as energy, enthalpy, and entropy, as well as relaxation times in *thermal equilibrium*. Several approaches have been proposed to quantify how deeply a glassy state lies within the (free) energy landscape. However, comparing energetic or temperature scales across different glass-forming systems is challenging due to their widely varying microscopic properties. In this context, entropy provides a more reliable basis for comparison, as it is directly related to the total number of accessible states at a given temperature.

In glassy systems, the configurational component of the entropy is of particular relevance, as discussed in Sec. I. Moving in this direction, we consider the configurational entropy at the lowest accessible temperature (or highest density) and compare it to its value at the mode-coupling transition temperature, T_{MCT} , when this quantity is available for the different methods, defining an associated stability ratio as $\mathcal{R}_s = S_c(T)/S_c(T_{MCT})$. According to this criterion, recent simulations of conventional glass formers have been able to reach ratios of $\mathcal{R}_s \approx 0.69$.²²³ Trajectory-sampling methods allow access to significantly deeper states, with estimated ratios around 0.43,³³ while swap Monte Carlo simulations achieve even lower values, down to $\mathcal{R}_s \approx 0.2$.²²⁷ Random pinning allows one to access glassy states with very small values of the configurational entropy, $\mathcal{R}_s \approx 0 - 0.2$, although the precise definition of the entropy in such constrained systems remains subtle.^{24,179} A very recent study combining parallel tempering, swap Monte Carlo, and population annealing in a carefully chosen order achieves $\mathcal{R}_s \approx 0$ for relatively small system sizes.²³⁴

Thermodynamic stability can also be assessed indirectly through relaxation times in thermal equilibrium. Although relaxation times are dynamical quantities, in the context of algorithms that employ unphysical moves, they provide insight into how efficiently a given protocol can access low-energy states. For example, in swap Monte Carlo simulations, the ratio of structural relaxation times between standard and swap dynamics at the respective lowest accessible temperatures has been conservatively estimated to be as large as $\tau_\alpha(\text{MC})/\tau_\alpha(\text{Swap}) \approx 10^{10}$ in three-dimensions.³⁰ Similarly, for irreversible chain-based collective-swap algorithms, a ratio of $\tau_\alpha(\text{Swap})/\tau_\alpha(\text{Chain}) \approx 10^2$ has been reported at the highest accessible densities,³² indicating a further enhancement in sampling efficiency beyond standard swap Monte Carlo. By contrast, parallel tempering yields more modest improvements, with reported ratios of $\tau_\alpha(\text{MC})/\tau_\alpha(\text{PT}) \approx 10^2$.²²⁸

In experiments, a way to assess thermodynamic stability is through the *fictive temperature* T_f , defined as the temperature at which the glass structure would be in thermal equilibrium with the liquid.³ In simulations, however, the limited accessible time scales compared to experiments make the determination of T_f less robust, and it is, therefore, infrequently evaluated. An exception is provided by swap Monte Carlo simulations, where T_f has been computed, showing that swap-generated configurations lie close to the experimental glass transition of the numerical model with $T_f/T_g \approx 1.18$.²²⁶

Alternative definitions of thermodynamic stability have also been employed in simulations. For example, in trajectory sampling of the Kob–Andersen mixture, thermodynamic stability can be evaluated using the ratio between (estimated) T_K , the Kauzmann temperature, and T_{eq} , the lowest temperature at which the inherent-structure energy remains well described by a quadratic approximation ($T_{eq} \approx 0.37$).³³

Mechanical stability, which complements other measures of glass stability, refers to the material's response to mechanical loading and can be quantified using rheological observables.²³⁵ Under quasi-static deformation, glasses prepared with increasing stability exhibit a qualitative change in their stress-strain curves. In particular, the transition from conventional to ultrastable glasses is marked by a sharp, discontinuous change in the mechanical response.^{25,133,171,198} The stress overshoot at yielding, $\Delta\sigma$, can therefore serve as an effective order parameter distinguishing these regimes. In Table I, we report $\Delta\sigma/\sigma = (\sigma_{\max} - \sigma_{\min})/\sigma_{\min}$, where $\sigma_{\max}$ and $\sigma_{\min}$ are the values of the maximum stress and its value just after the overshoot, respectively. The reported values range from the moderate value of 0.5¹³⁵ for conventional glasses to as high as 3.0 for glasses prepared using structural optimization.¹⁸⁹

Several alternative notions of stability have been proposed, often based on assumed correlations between specific structural or geometrical features and enhanced glass stability.^{236,237} As these approaches do not provide direct quantitative measurements of stability, we do not include them in Table I. Nevertheless, they remain relevant and will be collectively referred to here as structural stability.

The population of locally favored structures (LFSs), i.e., geometric motifs that often locally minimize the energy, provides an efficient probe of structural stability. Several studies have shown that LFS populations correlate strongly with thermodynamic control parameters such as temperature or packing fraction²³⁷ and with dynamics.^{238–242} This correlation has been observed across a wide range of systems, including colloidal glasses^{48,243,244} and metallic glasses.^{27,245} Icosahedra are found to be the most frequently observed LFS in many Lennard-Jones-like systems.^{48,237,246–248} A larger concentration of icosahedra in UG with respect to CG has been found in glasses obtained with different methods, such as vapor deposition,⁴⁸ structural optimization,¹⁰⁰ or swap MC.²⁴⁹

More recently, machine-learning-based strategies have been developed to detect structurally relevant local environments in amorphous configurations. In both unsupervised²⁵⁰ and supervised^{251,252} approaches, high-dimensional order parameters are employed to identify local structural signatures associated with thermodynamic quantities such as energy^{251,252} or with dynamic properties such as dynamical heterogeneities.^{250,253}

An alternative measure of glass stability is the centrosymmetry parameter, which quantifies the degree of local inversion symmetry in the arrangement of neighbors around each particle.^{254–256} High centrosymmetry suppresses non-affine rearrangements that would otherwise reduce the shear modulus. In a perfectly centrosymmetric crystal, forces from opposite neighbors cancel under affine deformation, keeping particles in mechanical equilibrium. In disordered structures, this cancellation is absent, generating residual forces that induce non-affine displacements. Non-affinity is typically quantified by the parameter μ_{sc} , which measures how closely a glass mimics the elastic response of a crystal under shear or compression. Smaller values indicate a more ordered, crystal-like response,^{102,257} and thus a more stable glass.

Structural stability can also be explored through vibrational properties. In computer-generated glasses, the vibrational density of states, $D(\omega)$, universally follows ω^4 scaling at low frequencies,^{258–261} with a prefactor that decreases as stability increases.¹²¹ Experiments on vapor-deposited glasses similarly show that low-frequency modes are progressively suppressed in more stable samples.^{7,262,263} In the limit of extremely stable two-dimensional amorphous systems obtained through steric optimization, a crossover toward crystal-like Debye scaling $D(\omega) \sim \omega^{d-1}$ has recently been reported.¹⁰² The same study also identified broader signatures of emerging crystal-like order, including affine displacement and hyperuniformity, the latter manifested as suppressed long-wavelength density fluctuations with a small- q spectrum $\chi(q) \sim q^\alpha$, approaching the crystalline limit $\alpha = d + 1$ in d dimensions.^{102,198}

XIII. PERSPECTIVES

To help navigate this broad landscape, the algorithms discussed in this review can also be viewed through the lens of three encompassing paradigms, distinct from the five categories used to structure the presentation of individual methods, offering a complementary perspective on their underlying approach.

The first paradigm exploits the selective freezing or freeing of degrees of freedom. Swap Monte Carlo exemplifies this logic: by temporarily liberating particle sizes as additional degrees of freedom, it enables rapid traversal of configuration space. Random pinning operates on the opposite principle, freezing a subset of particles to restrict phase space and lower configurational entropy. Physical vapor deposition fits naturally within this framework as well, since the enhanced mobility at the free surface provides faster degrees of freedom that allow local equilibration before particles are incorporated into the bulk. Structural optimization methods and random bonding follow analogous strategies, selectively activating or suppressing degrees of freedom either temporarily or permanently. Finally, cyclic shear can also be interpreted in these terms, with the strain amplitude acting as an effective control parameter that opens additional relaxation pathways in the energy landscape and grants access to deeper regions of that landscape. Following a recent work,¹⁴² all these different preparation protocols, with the exception of the structural optimization, may access deeper states not by altering the underlying free-energy landscape but by accelerating the exploration of configuration space, effectively reparameterizing the flow of time along a common relaxation trajectory.

The second paradigm relies on biasing the target distribution and applying appropriate reweighting in order to effectively

flatten free-energy or potential-energy barriers. This category naturally encompasses importance sampling methods, parallel tempering, population annealing, and trajectory-sampling techniques. The common principle is to enhance the sampling of configurations that would otherwise be exponentially suppressed in the equilibrium ensemble, while restoring statistical consistency with the Boltzmann distribution through reweighting.

The third paradigm encompasses sampling methods that generate configurations through non-equilibrium dynamics with finite entropy production, while still ensuring that the stationary distribution follows the Boltzmann measure. Examples include lifted and irreversible Monte Carlo schemes, where detailed balance is relaxed while global balance is maintained, enabling faster equilibration without affecting the equilibrium distribution.

Machine-learning-based approaches do not fit neatly into any single paradigm but can, in principle, intersect with all three: ML models may implicitly learn to free or constrain effective degrees of freedom, construct biased proposal distributions for importance sampling, or generate non-equilibrium dynamics that enhance exploration while preserving the correct equilibrium measure. More broadly, machine learning and artificial intelligence have shown a remarkable ability to navigate high-dimensional spaces, as illustrated by their success in complex games such as Go,²⁶⁴ a capability that tends to emerge when the rules of the problem are well defined and performance is unambiguously measurable. Sampling a target distribution can be framed in precisely these terms: the objective is to decorrelate configurations as rapidly as possible while preserving the equilibrium measure. Although current ML-based approaches do not yet outperform state-of-the-art sampling algorithms, the preparation of ultrastable glasses is a well-posed computational problem, and the ability of ML to learn efficient exploration strategies in rugged, high-dimensional landscapes makes it a promising direction for future developments.

The quest for ultrastability remains, however, ongoing. From a theoretical perspective, it is intimately connected to the unresolved question of whether an ideal glass transition exists. At the same time, a pressing challenge is the extension of these sophisticated algorithms, often developed for simplified model systems, to more realistic glass formers featuring rotational degrees of freedom and more complex interaction potentials. Whether further progress will arise from entirely new algorithmic paradigms, from refinements of existing methods, or from hybrid combinations of current strategies remains an open question.

A compelling recent example of combining different algorithms is provided by Ref. 234, which introduces a protocol merging parallel tempering, swap Monte Carlo, and population annealing to achieve full equilibration of a two-dimensional Lennard-Jones ternary mixture down to zero temperature for systems up to $N = 77$ particles and provides new insight into the nature of the Kauzmann transition in two dimensions. The protocol first uses swap Monte Carlo combined with parallel tempering to reach temperatures far below those accessible with standard approaches. At these low temperatures and for small system sizes, the potential-energy distribution develops a rare low-energy tail approaching the ground-state energy. Population annealing then amplifies these rare configurations through replication and reweighting, promoting them to typical equilibrium states. This work highlights how the careful combination and ordering of advanced sampling techniques

can be crucial for achieving full equilibration, suggesting promising directions for future developments, including larger system sizes, three-dimensional models, and molecular liquids.

Taken together, the pursuit of ultrastable glasses will continue to drive methodological innovation, with advances that are likely to resonate well beyond the glass community, benefiting neighboring fields in statistical physics, soft matter, and computer science.

ACKNOWLEDGMENTS

F.L. and J.R. acknowledge the support from ICSC—Centro Nazionale di Ricerca in High Performance Computing, Big Data, and Quantum Computing, funded by the European Union—NextGenerationEU. M.O. acknowledges the support from MIAI@Grenoble Alpes and the Agence Nationale de la Recherche under France 2030 (Grant No. ANR-23-IACL-0006. T.Y. acknowledges the support from JSPS KAKENHI (Grant No. JP24K00597).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Fabio Leoni: Conceptualization (equal); Data curation (lead); Methodology (equal); Project administration (equal); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (equal). **Misaki Ozawa:** Conceptualization (equal); Data curation (equal); Funding acquisition (equal); Methodology (equal); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (equal). **John Russo:** Conceptualization (equal); Data curation (equal); Funding acquisition (equal); Methodology (equal); Project administration (equal); Visualization (lead); Writing – original draft (equal); Writing – review & editing (equal). **Taiki Yanagishima:** Conceptualization (equal); Funding acquisition (equal); Methodology (equal); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (equal). **Andrea Ninarello:** Conceptualization (equal); Data curation (equal); Methodology (equal); Project administration (lead); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this article, as no new data were created in this study.

REFERENCES

- 1 M. D. Ediger, C. A. Angell, and S. R. Nagel, “Supercooled liquids and glasses,” *J. Phys. Chem.* **100**, 13200–13212 (1996).
- 2 L. Berthier and G. Biroli, “Theoretical perspective on the glass transition and amorphous materials,” *Rev. Mod. Phys.* **83**, 587–645 (2011).
- 3 M. D. Ediger, “Perspective: Highly stable vapor-deposited glasses,” *J. Chem. Phys.* **147**, 210901 (2017).
- 4 C. Rodriguez-Tinoco, M. Gonzalez-Silveira, M. A. Ramos, and J. Rodriguez-Viejo, “Ultrastable glasses: New perspectives for an old problem,” *Riv. Nuovo Cimento* **45**, 325–406 (2022).

- 5 C. A. Angell, “Formation of glasses from liquids and biopolymers,” *Science* **267**, 1924–1935 (1995).
- 6 C. A. Angell, K. L. Ngai, G. B. McKenna, P. F. McMillan, and S. W. Martin, “Relaxation in glass-forming liquids and amorphous solids,” *J. Appl. Phys.* **88**, 3113–3157 (2000).
- 7 S. F. Swallen, K. L. Kearns, M. K. Mapes, Y. S. Kim, R. J. McMahon, M. D. Ediger, T. Wu, L. Yu, and S. Satija, “Organic glasses with exceptional thermodynamic and kinetic stability,” *Science* **315**, 353–356 (2007).
- 8 K. L. Kearns, T. Still, G. Fytas, and M. D. Ediger, “High-modulus organic glasses prepared by physical vapor deposition,” *Adv. Mater.* **22**, 39 (2010).
- 9 P. G. Debenedetti and F. H. Stillinger, “Supercooled liquids and the glass transition,” *Nature* **410**, 259–267 (2001).
- 10 F. Sciortino, “Potential energy landscape description of supercooled liquids and glasses,” *J. Stat. Mech.: Theory Exp.* **2005**, P05015.
- 11 P. K. Gupta and W. Kob, “Basis glass states: New insights from the potential energy landscape,” *J. Non-Cryst. Solids: X* **3**, 100031 (2019).
- 12 T. R. Kirkpatrick and P. G. Wolynes, “Connections between some kinetic and equilibrium theories of the glass transition,” *Phys. Rev. A* **35**, 3072 (1987).
- 13 M. Mézard and G. Parisi, “A first-principle computation of the thermodynamics of glasses,” *J. Chem. Phys.* **111**, 1076–1095 (1999).
- 14 V. Lubchenko and P. G. Wolynes, “Theory of structural glasses and supercooled liquids,” *Annu. Rev. Phys. Chem.* **58**, 235–266 (2007).
- 15 G. Biroli and J.-P. Bouchaud, “The random first-order transition theory of glasses: A critical assessment,” in *Structural Glasses and Supercooled Liquids: Theory, Experiment, and Applications* (John Wiley & Sons, Inc., 2012), pp. 31–113.
- 16 W. Kauzmann, “The nature of the glassy state and the behavior of liquids at low temperatures,” *Chem. Rev.* **43**, 219–256 (1948).
- 17 L. Berthier, G. Biroli, L. Manning, and F. Zamponi, “Yielding and plasticity in amorphous solids,” *Nat. Rev. Phys.* **7**, 313–330 (2025).
- 18 L. Berthier and D. R. Reichman, “Modern computational studies of the glass transition,” *Nat. Rev. Phys.* **5**, 102–116 (2023).
- 19 T. Koop, J. Bookhold, M. Shiraiwa, and U. Pöschl, “Glass transition and phase state of organic compounds: Dependency on molecular properties and implications for secondary organic aerosols in the atmosphere,” *Phys. Chem. Chem. Phys.* **13**, 19238–19255 (2011).
- 20 R. H. Swendsen and J.-S. Wang, “Replica Monte Carlo simulation of spin glasses,” *Phys. Rev. Lett.* **57**, 2607–2609 (1986).
- 21 R. Yamamoto and W. Kob, “Replica-exchange molecular dynamics simulation for supercooled liquids,” *Phys. Rev. E* **61**, 5473 (2000).
- 22 C. Cammarota and G. Biroli, “Ideal glass transitions by random pinning,” *Proc. Natl. Acad. Sci. U. S. A.* **109**, 8850–8855 (2012).
- 23 G. M. Hocky, L. Berthier, and D. R. Reichman, “Equilibrium ultrastable glasses produced by random pinning,” *J. Chem. Phys.* **141**, 224503 (2014).
- 24 M. Ozawa, W. Kob, A. Ikeda, and K. Miyazaki, “Equilibrium phase diagram of a randomly pinned glass-former,” *Proc. Natl. Acad. Sci. U. S. A.* **112**, 6914–6919 (2015).
- 25 M. Ozawa, Y. Iwashita, W. Kob, and F. Zamponi, “Creating bulk ultrastable glasses by random particle bonding,” *Nat. Commun.* **14**, 113 (2023).
- 26 E. P. Bernard and W. Krauth, “Two-step melting in two dimensions: First-order liquid-hexatic transition,” *Phys. Rev. Lett.* **107**, 155704 (2011).
- 27 S. Singh, M. D. Ediger, and J. J. De Pablo, “Ultrastable glasses from *in silico* vapour deposition,” *Nat. Mater.* **12**, 139–144 (2013).
- 28 D. Fiocco, G. Foffi, and S. Sastry, “Oscillatory athermal quasistatic deformation of a model glass,” *Phys. Rev. E* **88**, 020301 (2013).
- 29 A. D. S. Parmar, S. Kumar, and S. Sastry, “Strain localization above the yielding point in cyclically deformed glasses,” *Phys. Rev. X* **9**, 021018 (2019).
- 30 A. Ninarello, L. Berthier, and D. Coslovich, “Models and algorithms for the next generation of glass transition studies,” *Phys. Rev. X* **7**, 021039 (2017).
- 31 C. Brito, E. Lerner, and M. Wyart, “Theory for swap acceleration near the glass and jamming transitions for continuously polydisperse particles,” *Phys. Rev. X* **8**, 031050 (2018).
- 32 F. Ghimenti, L. Berthier, and F. van Wijland, “Irreversible Monte Carlo algorithms for hard disk glasses: From event-chain to collective swaps,” *Phys. Rev. Lett.* **133**, 028202 (2024).

- ³³F. Turci, C. P. Royall, and T. Speck, "Nonequilibrium phase transition in an atomistic glassformer: The connection to thermodynamics," *Phys. Rev. X* **7**, 031028 (2017).
- ³⁴L. Galliano, R. Rende, and D. Coslovich, "Policy-guided Monte Carlo on general state spaces: Application to glass-forming mixtures," *J. Chem. Phys.* **161**, 064503 (2024).
- ³⁵J. D. Stevenson and P. G. Wolyne, "On the surface of glasses," *J. Chem. Phys.* **129**, 234514 (2008).
- ³⁶L. Zhu, C. W. Brian, S. F. Swallen, P. T. Straus, M. D. Ediger, and L. Yu, "Surface self-diffusion of an organic glass," *Phys. Rev. Lett.* **106**, 256103 (2011).
- ³⁷C. R. Daley, Z. Fakhraai, M. D. Ediger, and J. A. Forrest, "Comparing surface and bulk flow of a molecular glass former," *Soft Matter* **8**, 2206–2212 (2012).
- ³⁸C. W. Brian and L. Yu, "Surface self-diffusion of organic glasses," *J. Phys. Chem. A* **117**, 13303–13309 (2013).
- ³⁹I. Lyubimov, M. D. Ediger, and J. J. de Pablo, "Model vapor-deposited glasses: Growth front and composition effects," *J. Chem. Phys.* **139**, 144505 (2013).
- ⁴⁰Y. Zhang, E. C. Glor, M. Li, T. Liu, K. Wahid, W. Zhang, R. A. Riggelman, and Z. Fakhraai, "Long-range correlated dynamics in ultra-thin molecular glass films," *J. Chem. Phys.* **145**, 114502 (2016).
- ⁴¹D. R. Reid, I. Lyubimov, M. D. Ediger, and J. J. de Pablo, "Age and structure of a model vapour-deposited glass," *Nat. Commun.* **7**, 13062 (2016).
- ⁴²Y. Zhang and Z. Fakhraai, "Invariant fast diffusion on the surfaces of ultrastable and aged molecular glasses," *Phys. Rev. Lett.* **118**, 066101 (2017).
- ⁴³L. Berthier, P. Charbonneau, E. Fleener, and F. Zamponi, "Origin of ultrastability in vapor-deposited glasses," *Phys. Rev. Lett.* **119**, 188002 (2017).
- ⁴⁴S. Samanta, G. Huang, G. Gao, Y. Zhang, A. Zhang, S. Wolf, C. N. Woods, Y. Jin, P. J. Walsh, and Z. Fakhraai, "Exploring the importance of surface diffusion in stability of vapor-deposited organic glasses," *J. Phys. Chem. B* **123**, 4108–4117 (2019).
- ⁴⁵A. R. Moore, G. Huang, S. Wolf, P. J. Walsh, Z. Fakhraai, and R. A. Riggelman, "Effects of microstructure formation on the stability of vapor-deposited glasses," *Proc. Natl. Acad. Sci. U. S. A.* **116**, 5937–5942 (2019).
- ⁴⁶P. Harrowell, "Orientationally ordered glasses via controlled deposition," *Proc. Natl. Acad. Sci. U. S. A.* **116**, 21341–21342 (2019).
- ⁴⁷F. Leoni, H. Tanaka, and J. Russo, "Crystal nucleation in a vapor deposited lennard-jones mixture," *J. Mol. Liq.* **391**, 123178 (2023).
- ⁴⁸F. Leoni, F. Martelli, C. P. Royall, and J. Russo, "Structural signatures of ultrastability in a deposited glassformer," *Phys. Rev. Lett.* **130**, 198201 (2023).
- ⁴⁹G. Sun, S. Saw, I. Douglass, and P. Harrowell, "Structural origin of enhanced dynamics at the surface of a glassy alloy," *Phys. Rev. Lett.* **119**, 245501 (2017).
- ⁵⁰A. Sepúlveda, M. Tylnski, A. Guiseppi-Elie, R. Richert, and M. D. Ediger, "Role of fragility in the formation of highly stable organic glasses," *Phys. Rev. Lett.* **113**, 045901 (2014).
- ⁵¹F. Leoni, F. Martelli, and J. Russo, "Correlating ultrastability with fragility and surface mobility in vapor deposited tetrahedral glasses," *J. Phys. Chem. Lett.* **15**, 8444–8450 (2024).
- ⁵²H.-W. Lin, C.-L. Lin, H.-H. Chang, Y.-T. Lin, C.-C. Wu, Y.-M. Chen, R.-T. Chen, Y.-Y. Chien, and K.-T. Wong, "Anisotropic optical properties and molecular orientation in vacuum-deposited ter (9, 9-diarylfuorene) s thin films using spectroscopic ellipsometry," *J. Appl. Phys.* **95**, 881–886 (2004).
- ⁵³H.-W. Lin, C.-L. Lin, C.-C. Wu, T.-C. Chao, and K.-T. Wong, "Influences of molecular orientation on stimulated emission characteristics of oligofluorene films," *Org. Electron.* **8**, 189–197 (2007).
- ⁵⁴D. Yokoyama, A. Sakaguchi, M. Suzuki, and C. Adachi, "Horizontal molecular orientation in vacuum-deposited organic amorphous films of hole and electron transport materials," *Appl. Phys. Lett.* **93**, 173302 (2008).
- ⁵⁵D. Yokoyama, "Molecular orientation in small-molecule organic light-emitting diodes," *J. Mater. Chem.* **21**, 19187–19202 (2011).
- ⁵⁶J. Y. Kim, D. Yokoyama, and C. Adachi, "Horizontal orientation of disk-like hole transport molecules and their application for organic light-emitting diodes requiring a lower driving voltage," *J. Phys. Chem. C* **116**, 8699–8706 (2012).
- ⁵⁷S. S. Dalal, D. M. Walters, I. Lyubimov, J. J. de Pablo, and M. D. Ediger, "Tunable molecular orientation and elevated thermal stability of vapor-deposited organic semiconductors," *Proc. Natl. Acad. Sci. U. S. A.* **112**, 4227–4232 (2015).
- ⁵⁸J. Jiang, D. M. Walters, D. Zhou, and M. D. Ediger, "Substrate temperature controls molecular orientation in two-component vapor-deposited glasses," *Soft Matter* **12**, 3265–3270 (2016).
- ⁵⁹L. W. Antony, N. E. Jackson, I. Lyubimov, V. Vishwanath, M. D. Ediger, and J. J. de Pablo, "Influence of vapor deposition on structural and charge transport properties of ethylbenzene films," *ACS Cent. Sci.* **3**, 415–424 (2017).
- ⁶⁰D. M. Walters, L. Antony, J. J. de Pablo, and M. D. Ediger, "Influence of molecular shape on the thermal stability and molecular orientation of vapor-deposited organic semiconductors," *J. Phys. Chem. Lett.* **8**, 3380–3386 (2017).
- ⁶¹P. Luo, S. E. Wolf, S. Govind, R. B. Stephens, D. H. Kim, C. Y. Chen, T. Nguyen, P. Wasiq, M. Zhernikov, B. McClimon, and Z. Fakhraai, "High-density stable glasses formed on soft substrates," *Nat. Mater.* **23**, 688–694 (2024).
- ⁶²A. A. Sokolov and M. I. Yagofarov, "All-atom molecular dynamics simulations of vapor deposition as a tool to study thermodynamic stability and structure of organic glasses," *J. Chem. Phys.* **164**, 174506 (2026).
- ⁶³L. Lupi, N. Kastelowitz, and V. Molinero, "Vapor deposition of water on graphitic surfaces: Formation of amorphous ice, bilayer ice, ice I, and liquid water," *J. Chem. Phys.* **141**, 18C508 (2014).
- ⁶⁴W. Zhang, J. F. Douglas, and F. W. Starr, "Dynamical heterogeneity in a vapor-deposited polymer glass," *J. Chem. Phys.* **146**, 203310 (2017).
- ⁶⁵L. Berthier, P. Charbonneau, D. Coslovich, A. Ninarello, M. Ozawa, and S. Yaida, "Configurational entropy measurements in extremely supercooled liquids that break the glass ceiling," *Proc. Natl. Acad. Sci. U. S. A.* **114**, 11356–11361 (2017).
- ⁶⁶Y. Jin, A. Zhang, S. E. Wolf, S. Govind, A. R. Moore, M. Zhernikov, G. Freychet, A. Arabi Shamsabadi, and Z. Fakhraai, "Glasses denser than the supercooled liquid," *Proc. Natl. Acad. Sci. U. S. A.* **118**, e2100738118 (2021).
- ⁶⁷N. S. Shcheblanov, C. Léonard, Q.-D. To, M. E. Povarnitsyn, and A. Lemaitre, "Molecular dynamics study of the formation of porous films by room-temperature physical vapor deposition of silica," *J. Phys. Chem. C* **128**, 17606–17618 (2024).
- ⁶⁸Z. Amato, T. F. Headen, P. Ghesquière, and H. J. Fraser, "A molecular dynamics study of the effect of annealing temperature on the structure of ASW," *Phys. Chem. Chem. Phys.* **27**, 14864–14883 (2025).
- ⁶⁹S. Singh and J. J. de Pablo, "A molecular view of vapor deposited glasses," *J. Chem. Phys.* **134**, 194903 (2011).
- ⁷⁰P.-H. Lin, I. Lyubimov, L. Yu, M. D. Ediger, and J. J. de Pablo, "Molecular modeling of vapor-deposited polymer glasses," *J. Chem. Phys.* **140**, 204504 (2014).
- ⁷¹I. Lyubimov, L. Antony, D. M. Walters, D. Rodney, M. D. Ediger, and J. J. de Pablo, "Orientational anisotropy in simulated vapor-deposited molecular glasses," *J. Chem. Phys.* **143**, 094502 (2015).
- ⁷²B. Seoane, D. R. Reid, J. J. de Pablo, and F. Zamponi, "Low-temperature anomalies of a vapor deposited glass," *Phys. Rev. Mater.* **2**, 015602 (2018).
- ⁷³I. Douglass and P. Harrowell, "Formation of ultrastable glasses via precipitation: A modeling study," *Phys. Rev. Lett.* **122**, 088003 (2019).
- ⁷⁴P. Leishangthem, A. D. S. Parmar, and S. Sastry, "The yielding transition in amorphous solids under oscillatory shear deformation," *Nat. Commun.* **8**, 14653 (2017).
- ⁷⁵O. Pouliquen, M. Belzons, and M. Nicolas, "Fluctuating particle motion during shear induced granular compaction," *Phys. Rev. Lett.* **91**, 014301 (2003).
- ⁷⁶J. B. Knight, C. G. Fandrich, C. N. Lau, H. M. Jaeger, and S. R. Nagel, "Density relaxation in a vibrated granular material," *Phys. Rev. E* **51**, 3957–3963 (1995).
- ⁷⁷N. Kumar and S. Luding, "Memory of jamming-multiscale models for soft and granular matter," *Granular Matter* **18**, 58 (2016).
- ⁷⁸D. J. Lacks and M. J. Osborne, "Energy landscape picture of overaging and rejuvenation in a sheared glass," *Phys. Rev. Lett.* **93**, 255501 (2004).
- ⁷⁹A. W. Lees and S. F. Edwards, "The computer study of transport processes under extreme conditions," *J. Phys. C: Solid State Phys.* **5**, 1921 (1972).
- ⁸⁰F. Weik, R. Weeber, K. Szuttor, K. Breitsprecher, J. de Graaf, M. Kuron, J. Landsgeßel, H. Menke, D. Sean, and C. Holm, "ESPResSo 4.0—An extensible software package for simulating soft matter systems," *Eur. Phys. J.: Spec. Top.* **227**, 1789–1816 (2019).
- ⁸¹S. Bindgen, F. Weik, R. Weeber, E. Koos, and P. de Buyl, "Lees–Edwards boundary conditions for translation invariant shear flow: Implementation and transport properties," *Phys. Fluids* **33**, 083615 (2021).

- ⁸²D. Bonn, M. M. Denn, L. Berthier, T. Divoux, and S. Manneville, "Yield stress materials in soft condensed matter," *Rev. Mod. Phys.* **89**, 035005 (2017).
- ⁸³E. D. Knowlton, D. J. Pine, and L. Cipelletti, "A microscopic view of the yielding transition in concentrated emulsions," *Soft Matter* **10**, 6931–6940 (2014).
- ⁸⁴M. Golkia, G. P. Shrivastav, P. Chaudhuri, and J. Horbach, "Flow heterogeneities in supercooled liquids and glasses under shear," *Phys. Rev. E* **102**, 023002 (2020).
- ⁸⁵C. A. Schuh, T. C. Hufnagel, and U. Ramamurty, "Mechanical behavior of amorphous alloys," *Acta Mater.* **55**, 4067–4109 (2007).
- ⁸⁶I. Procaccia, C. Rainone, and M. Singh, "Mechanical failure in amorphous solids: Scale-free spinodal criticality," *Phys. Rev. E* **96**, 032907 (2017).
- ⁸⁷L. Pérez-Ocampo, A. Zaccone, and M. Laurati, "A well defined glass state obtained by oscillatory shear," *J. Rheol.* **62**, 197–207 (2018).
- ⁸⁸T. Kawasaki and L. Berthier, "Macroscopic yielding in jammed solids is accompanied by a nonequilibrium first-order transition in particle trajectories," *Phys. Rev. E* **94**, 022615 (2016).
- ⁸⁹I. Regev, J. Weber, C. Reichhardt, K. A. Dahmen, and T. Lookman, "Reversibility and criticality in amorphous solids," *Nat. Commun.* **6**, 8805 (2015).
- ⁹⁰J. Lin, E. Lerner, A. Rosso, and M. Wyart, "Scaling description of the yielding transition in soft amorphous solids at zero temperature," *Proc. Natl. Acad. Sci. U. S. A.* **111**, 14382–14387 (2014).
- ⁹¹C. Liu, E. E. Ferrero, F. Puosi, J.-L. Barrat, and K. Martens, "Driving rate dependence of avalanche statistics and shapes at the yielding transition," *Phys. Rev. Lett.* **116**, 065501 (2016).
- ⁹²P. Das, H. A. Vinutha, and S. Sastry, "Unified phase diagram of reversible-irreversible, jamming, and yielding transitions in cyclically sheared soft-sphere packings," *Proc. Natl. Acad. Sci. U. S. A.* **117**, 10203–10209 (2020).
- ⁹³N. V. Priezjev, "Molecular dynamics simulations of the mechanical annealing process in metallic glasses: Effects of strain amplitude and temperature," *J. Non-Cryst. Solids* **479**, 42–48 (2018).
- ⁹⁴N. V. Priezjev, "Accelerated relaxation in disordered solids under cyclic loading with alternating shear orientation," *J. Non-Cryst. Solids* **525**, 119683 (2019).
- ⁹⁵H. Bhaumik, G. Foffi, and S. Sastry, "The role of annealing in determining the yielding behavior of glasses under cyclic shear deformation," *Proc. Natl. Acad. Sci. U. S. A.* **118**, e2100227118 (2021).
- ⁹⁶W.-T. Yeh, M. Ozawa, K. Miyazaki, T. Kawasaki, and L. Berthier, "Glass stability changes the nature of yielding under oscillatory shear," *Phys. Rev. Lett.* **124**, 225502 (2020).
- ⁹⁷C. E. Maloney and A. Lemaitre, "Amorphous systems in athermal, quasistatic shear," *Phys. Rev. E* **74**, 016118 (2006).
- ⁹⁸V. V. Krishnan, K. Ramola, and S. Karmakar, "Annealing effects of multidirectional oscillatory shear in model glass formers," *Phys. Rev. Appl.* **19**, 024004 (2023).
- ⁹⁹M. Bantawa, P. Edera, M. Cloitre, and R. T. Bonnecaze, "Stress distributions in soft particle glasses: Insights from a thermodynamic model," *J. Rheol.* **69**, 611–620 (2025).
- ¹⁰⁰F. Leoni, J. Russo, F. Sciortino, and T. Yanagishima, "Generating ultrastable glasses by homogenizing the local virial stress," *Phys. Rev. Lett.* **134**, 128201 (2025).
- ¹⁰¹T. Yanagishima, J. Russo, R. P. A. Dullens, and H. Tanaka, "Towards glasses with permanent stability," *Phys. Rev. Lett.* **127**, 215501 (2021).
- ¹⁰²X. Fan, D. Xu, J. Zhang, H. Hu, P. Tan, N. Xu, H. Tanaka, and H. Tong, "Ideal non-crystals as a distinct form of ordered states without symmetry breaking," *Nat. Mater.* **25**, 1020–1027 (2026).
- ¹⁰³S. Mitra, A. D. S. Parmar, P. Leishangthem, S. Sastry, and G. Foffi, "Hyperuniformity in cyclically driven glasses," *J. Stat. Mech.: Theory Exp.* **2021**, 033203.
- ¹⁰⁴R. Sharma and S. Karmakar, "Activity-induced annealing leads to a ductile-to-brittle transition in amorphous solids," *Nat. Phys.* **21**, 253–261 (2025).
- ¹⁰⁵R. Priya and S. Karmakar, "Inverse Bauschinger effect in active ultrastable glasses," *Phys. Rev. E* **112**, 035414 (2025).
- ¹⁰⁶D. V. Denisov, M. T. Dang, B. Struth, A. Zaccone, G. H. Wegdam, and P. Schall, "Sharp symmetry-change marks the mechanical failure transition of glasses," *Sci. Rep.* **5**, 14359 (2015).
- ¹⁰⁷H. Jia, G. Wang, S. Chen, Y. Gao, W. Li, and P. K. Liaw, "Fatigue and fracture behavior of bulk metallic glasses and their composites," *Prog. Mater. Sci.* **98**, 168–248 (2018).
- ¹⁰⁸R. Gutiérrez, S. Karmakar, Y. G. Pollack, and I. Procaccia, "The static length-scale characterizing the glass transition at lower temperatures," *Europhys. Lett.* **111**, 56009 (2015).
- ¹⁰⁹A. D. Parmar, M. Ozawa, and L. Berthier, "Ultrastable metallic glasses *in silico*," *Phys. Rev. Lett.* **125**, 085505 (2020).
- ¹¹⁰G. Jung, G. Biroli, and L. Berthier, "Predicting dynamic heterogeneity in glass-forming liquids by physics-inspired machine learning," *Phys. Rev. Lett.* **130**, 238202 (2023).
- ¹¹¹L. Berthier, E. Flenner, C. J. Fullerton, C. Scalliet, and M. Singh, "Efficient swap algorithms for molecular dynamics simulations of equilibrium supercooled liquids," *J. Stat. Mech.: Theory Exp.* **2019**, 064004.
- ¹¹²L. D. Fosdick, "Calculation of order parameters in a binary alloy by the Monte Carlo method," *Phys. Rev.* **116**, 565 (1959).
- ¹¹³N.-H. Tsai, F. F. Abraham, and G. M. Pound, "The structure and thermodynamics of binary microclusters: A Monte Carlo simulation," *Surf. Sci.* **77**, 465–492 (1978).
- ¹¹⁴D. Gazzillo and G. Pastore, "Equation of state for symmetric non-additive hard-sphere fluids: An approximate analytic expression and new Monte Carlo results," *Chem. Phys. Lett.* **159**, 388–392 (1989).
- ¹¹⁵T. S. Grigera and G. Parisi, "Fast Monte Carlo algorithm for supercooled soft spheres," *Phys. Rev. E* **63**, 045102 (2001).
- ¹¹⁶L. A. Fernández, V. Martín-Mayor, and P. Verrocchio, "Phase diagram of a polydisperse soft-spheres model for liquids and colloids," *Phys. Rev. Lett.* **98**, 085702 (2007).
- ¹¹⁷G. Biroli, J.-P. Bouchaud, A. Cavagna, T. S. Grigera, and P. Verrocchio, "Thermodynamic signature of growing amorphous order in glass-forming liquids," *Nat. Phys.* **4**, 771–775 (2008).
- ¹¹⁸Y. Brumer and D. R. Reichman, "Numerical investigation of the entropy crisis in model glass formers," *J. Phys. Chem. B* **108**, 6832–6837 (2004).
- ¹¹⁹L. Berthier, P. Charbonneau, A. Ninarello, M. Ozawa, and S. Yaida, "Zero-temperature glass transition in two dimensions," *Nat. Commun.* **10**, 1508 (2019).
- ¹²⁰Y. Nishikawa, M. Ozawa, A. Ikeda, P. Chaudhuri, and L. Berthier, "Relaxation dynamics in the energy landscape of glass-forming liquids," *Phys. Rev. X* **12**, 021001 (2022).
- ¹²¹L. Wang, A. Ninarello, P. Guan, L. Berthier, G. Szamel, and E. Flenner, "Low-frequency vibrational modes of stable glasses," *Nat. Commun.* **10**, 26 (2019).
- ¹²²D. Khomenko, C. Scalliet, L. Berthier, D. R. Reichman, and F. Zamponi, "Depletion of two-level systems in ultrastable computer-generated glasses," *Phys. Rev. Lett.* **124**, 225901 (2020).
- ¹²³D. Khomenko, D. R. Reichman, and F. Zamponi, "Relationship between two-level systems and quasilocalized normal modes in glasses," *Phys. Rev. Mater.* **5**, 055602 (2021).
- ¹²⁴L. Wang, L. Fu, and Y. Nie, "Density of states below the first sound mode in 3D glasses," *J. Chem. Phys.* **157**, 074502 (2022).
- ¹²⁵W. Schirmacher, M. Paoluzzi, F. C. Mocanu, D. Khomenko, G. Szamel, F. Zamponi, and G. Ruocco, "The nature of non-phononic excitations in disordered systems," *Nat. Commun.* **15**, 3107 (2024).
- ¹²⁶G. Szamel and E. Flenner, "Microscopic analysis of sound attenuation in low-temperature amorphous solids reveals quantitative importance of non-affine effects," *J. Chem. Phys.* **156**, 144502 (2022).
- ¹²⁷C. Scalliet, L. Berthier, and F. Zamponi, "Nature of excitations and defects in structural glasses," *Nat. Commun.* **10**, 5102 (2019).
- ¹²⁸B. Seoane and F. Zamponi, "Spin-glass-like aging in colloidal and granular glasses," *Soft Matter* **14**, 5222–5234 (2018).
- ¹²⁹Y. Jin, P. Urbani, F. Zamponi, and H. Yoshino, "A stability-reversibility map unifies elasticity, plasticity, yielding, and jamming in hard sphere glasses," *Sci. Adv.* **4**, eaat6387 (2018).
- ¹³⁰C. Scalliet, L. Berthier, and F. Zamponi, "Absence of marginal stability in a structural glass," *Phys. Rev. Lett.* **119**, 205501 (2017).
- ¹³¹Q. Liao and L. Berthier, "Hierarchical landscape of hard disk glasses," *Phys. Rev. X* **9**, 011049 (2019).

- ¹³²L. Berthier, P. Charbonneau, and J. Kundu, "Finite dimensional vestige of spinodal criticality above the dynamical glass transition," *Phys. Rev. Lett.* **125**, 108001 (2020).
- ¹³³M. Ozawa, L. Berthier, G. Biroli, A. Rosso, and G. Tarjus, "Random critical point separates brittle and ductile yielding transitions in amorphous materials," *Proc. Natl. Acad. Sci. U. S. A.* **115**, 6656–6661 (2018).
- ¹³⁴M. Ozawa, L. Berthier, G. Biroli, and G. Tarjus, "Role of fluctuations in the yielding transition of two-dimensional glasses," *Phys. Rev. Res.* **2**, 023203 (2020).
- ¹³⁵D. Richard, C. Rainone, and E. Lerner, "Finite-size study of the athermal quasistatic yielding transition in structural glasses," *J. Chem. Phys.* **155**, 056101 (2021).
- ¹³⁶M. Ozawa, L. Berthier, G. Biroli, and G. Tarjus, "Rare events and disorder control the brittle yielding of well-annealed amorphous solids," *Phys. Rev. Res.* **4**, 023227 (2022).
- ¹³⁷M. Wyart and M. E. Cates, "Does a growing static length scale control the glass transition?," *Phys. Rev. Lett.* **119**, 195501 (2017).
- ¹³⁸L. Berthier, G. Biroli, J.-P. Bouchaud, and G. Tarjus, "Can the glass transition be explained without a growing static length scale?," *J. Chem. Phys.* **150**, 094501 (2019).
- ¹³⁹H. Ikeda, F. Zamponi, and A. Ikeda, "Mean field theory of the swap Monte Carlo algorithm," *J. Chem. Phys.* **147**, 234506 (2017).
- ¹⁴⁰G. Szamel, "Theory for the single-particle dynamics in glassy mixtures with particle size swaps," *J. Stat. Mech.: Theory Exp.* **2019**, 104016.
- ¹⁴¹N. Küchler and J. Horbach, "Understanding the swap Monte Carlo algorithm in a size-polydisperse model glassformer," *Phys. Rev. E* **108**, 024127 (2023).
- ¹⁴²F. Ghimenti, L. Berthier, J. Kurchan, and F. van Wijland, "Clever algorithms for glasses work by time reparameterization," *Proc. Natl. Acad. Sci. U. S. A.* **123**, e2520818123 (2026).
- ¹⁴³F. Chen, L. Lovász, and I. Pak, "Lifting Markov chains to speed up mixing," in *Proceedings of the Annual ACM Symposium on Theory of Computing* (Association for Computing Machinery, 1999), pp. 275–281.
- ¹⁴⁴E. P. Bernard, W. Krauth, and D. B. Wilson, "Event-chain Monte Carlo algorithms for hard-sphere systems," *Phys. Rev. E* **80**, 056704 (2009).
- ¹⁴⁵M. Michel, S. C. Kapfer, and W. Krauth, "Generalized event-chain Monte Carlo: Constructing rejection-free global-balance algorithms from infinitesimal steps," *J. Chem. Phys.* **140**, 054116 (2014).
- ¹⁴⁶W. Krauth, "Event-chain Monte Carlo: Foundations, applications, and prospects," *Front. Phys.* **9**, 663457 (2021).
- ¹⁴⁷Y. Nishikawa, F. Ghimenti, L. Berthier, and F. van Wijland, "Irreversible swap algorithms for soft sphere glasses," *Phys. Rev. E* **111**, 045416 (2025).
- ¹⁴⁸S. C. Kapfer and W. Krauth, "Sampling from a polytope and hard-disk Monte Carlo," *J. Phys.: Conf. Ser.* **454**, 012031 (2013).
- ¹⁴⁹A. C. Maggs and W. Krauth, "Large-scale dynamics of event-chain Monte Carlo," *Phys. Rev. E* **105**, 015309 (2022).
- ¹⁵⁰A. Monemvassitis, A. Guillin, and M. Michel, "PDMP characterisation of event-chain Monte Carlo algorithms for particle systems," *J. Stat. Phys.* **190**, 66 (2023).
- ¹⁵¹B. Li, Y. Nishikawa, P. Höllmer, L. Carillo, A. Maggs, and W. Krauth, "Hard-disk pressure computations—A historic perspective," *J. Chem. Phys.* **157**, 234111 (2022).
- ¹⁵²S. C. Kapfer and W. Krauth, "Cell-veto Monte Carlo algorithm for long-range systems," *Phys. Rev. E* **94**, 031302 (2016).
- ¹⁵³M. F. Faulkner, L. Qin, A. C. Maggs, and W. Krauth, "All-atom computations with irreversible Markov chains," *J. Chem. Phys.* **149**, 064113 (2018).
- ¹⁵⁴B. Li, S. Todo, A. C. Maggs, and W. Krauth, "Multithreaded event-chain Monte Carlo with local times," *Comput. Phys. Commun.* **261**, 107702 (2021).
- ¹⁵⁵R. H. Swendsen and J.-S. Wang, "Nonuniversal critical dynamics in Monte Carlo simulations," *Phys. Rev. Lett.* **58**, 86 (1987).
- ¹⁵⁶C. Dress and W. Krauth, "Cluster algorithm for hard spheres and related systems," *J. Phys. A: Math. Gen.* **28**, L597 (1995).
- ¹⁵⁷A. Buhot and W. Krauth, "Numerical solution of hard-core mixtures," *Phys. Rev. Lett.* **80**, 3787 (1998).
- ¹⁵⁸L. Santen and W. Krauth, "Absence of thermodynamic phase transition in a model glass former," *Nature* **405**, 550–551 (2000).
- ¹⁵⁹J. Liu and E. Luijten, "Rejection-free geometric cluster algorithm for complex fluids," *Phys. Rev. Lett.* **92**, 035504 (2004).
- ¹⁶⁰E. Marinari, G. Parisi, and J. Ruiz-Lorenzo, "Numerical simulations of spin glass systems," in *Spin Glasses and Random Fields* (World Scientific, 1998), pp. 59–98.
- ¹⁶¹Y. Sugita and Y. Okamoto, "Replica-exchange molecular dynamics method for protein folding," *Chem. Phys. Lett.* **314**, 141–151 (1999).
- ¹⁶²B. Coluzzi and G. Parisi, "On the approach to the equilibrium and the equilibrium properties of a glass-forming model," *J. Phys. A: Math. Gen.* **31**, 4349 (1998).
- ¹⁶³C. De Michele and F. Sciortino, "Equilibration times in numerical simulation of structural glasses: Comparing parallel tempering and conventional molecular dynamics," *Phys. Rev. E* **65**, 051202 (2002).
- ¹⁶⁴W. Kob and L. Berthier, "Probing a liquid to glass transition in equilibrium," *Phys. Rev. Lett.* **110**, 245702 (2013).
- ¹⁶⁵S. Yaida, L. Berthier, P. Charbonneau, and G. Tarjus, "Point-to-set lengths, local structure, and glassiness," *Phys. Rev. E* **94**, 032605 (2016).
- ¹⁶⁶L. Berthier, P. Charbonneau, and S. Yaida, "Efficient measurement of point-to-set correlations and overlap fluctuations in glass-forming liquids," *J. Chem. Phys.* **144**, 024501 (2016).
- ¹⁶⁷G. Jung, G. Biroli, and L. Berthier, "Normalizing flows as an enhanced sampling method for atomistic supercooled liquids," *Mach. Learn.: Sci. Technol.* **5**, 035053 (2024).
- ¹⁶⁸K. Kim, "Effects of pinned particles on the structural relaxation of supercooled liquids," *Europhys. Lett.* **61**, 790 (2003).
- ¹⁶⁹E. Anwar, P. Patel, M. Sharma, and S. Maitra Bhattacharyya, "Exploring the soft pinning effect in the dynamics and the structure-dynamics correlation in multicomponent supercooled liquids," *J. Chem. Phys.* **161**, 154501 (2024).
- ¹⁷⁰S. Chakrabarty, S. Karmakar, and C. Dasgupta, "Dynamics of glass forming liquids with randomly pinned particles," *Sci. Rep.* **5**, 12577 (2015).
- ¹⁷¹B. P. Bhowmik, P. Chaudhuri, and S. Karmakar, "Effect of pinning on the yielding transition of amorphous solids," *Phys. Rev. Lett.* **123**, 185501 (2019).
- ¹⁷²P. Scheidler, W. Kob, and K. Binder, "The relaxation dynamics of a supercooled liquid confined by rough walls," *J. Phys. Chem. B* **108**, 6673–6686 (2004).
- ¹⁷³V. Krakoviack, "Statistical mechanics of homogeneous partly pinned fluid systems," *Phys. Rev. E* **82**, 061501 (2010).
- ¹⁷⁴P. Charbonneau and G. Tarjus, "Decorrelation of the static and dynamic length scales in hard-sphere glass formers," *Phys. Rev. E* **87**, 042305 (2013).
- ¹⁷⁵R. L. Jack and C. J. Fullerton, "Dynamical correlations in a glass former with randomly pinned particles," *Phys. Rev. E* **88**, 042304 (2013).
- ¹⁷⁶C. J. Fullerton and R. L. Jack, "Investigating amorphous order in stable glasses by random pinning," *Phys. Rev. Lett.* **112**, 255701 (2014).
- ¹⁷⁷J. Russo and H. Tanaka, "Assessing the role of static length scales behind glassy dynamics in polydisperse hard disks," *Proc. Natl. Acad. Sci. U. S. A.* **112**, 6920–6924 (2015).
- ¹⁷⁸U. K. Nandi, P. Patel, M. Moid, M. K. Nandi, S. Sengupta, S. Karmakar, P. K. Maiti, C. Dasgupta, and S. Maitra Bhattacharyya, "Thermodynamics and its correlation with dynamics in a mean-field model and pinned systems: A comparative study using two different methods of entropy calculation," *J. Chem. Phys.* **156**, 014503 (2022).
- ¹⁷⁹M. Ozawa, A. Ikeda, K. Miyazaki, and W. Kob, "Ideal glass states are not purely vibrational: Insight from randomly pinned glasses," *Phys. Rev. Lett.* **121**, 205501 (2018).
- ¹⁸⁰K. Shiraishi, Y. Hara, and H. Mizuno, "Low-frequency vibrational states in ideal glasses with random pinning," *Phys. Rev. E* **106**, 054611 (2022).
- ¹⁸¹A. Mutneja, B. P. Bhowmik, and S. Karmakar, "Finite-disorder critical point in the brittle-to-ductile transition of amorphous solids in the presence of particle pinning," *Phys. Rev. Lett.* **135**, 198201 (2025).
- ¹⁸²S. Gokhale, K. Hima Nagamanasa, R. Ganapathy, and A. K. Sood, "Growing dynamical facilitation on approaching the random pinning colloidal glass transition," *Nat. Commun.* **5**, 4685 (2014).
- ¹⁸³I. Williams, F. Turci, J. E. Hallett, P. Crowther, C. Cammarota, G. Biroli, and C. P. Royall, "Experimental determination of configurational entropy in a two-dimensional liquid under random pinning," *J. Phys.: Condens. Matter* **30**, 094003 (2018).

- ¹⁸⁴G. Kikumoto, N. Torii, K. Fukao, C. P. Royall, H. Yao, Y. Saruyama, and S. Tatsumi, "Towards the ideal glass transition by pinning in a dimer-polymer mixture," *arXiv:2003.06089* (2020).
- ¹⁸⁵R. Das, B. P. Bhowmik, A. B. Puthirath, T. N. Narayanan, and S. Karmakar, "Soft pinning: Experimental validation of static correlations in supercooled molecular glass-forming liquids," *PNAS Nexus* **2**, pgad277 (2023).
- ¹⁸⁶H. C. Andersen, "Rattle: A 'velocity' version of the shake algorithm for molecular dynamics calculations," *J. Comput. Phys.* **52**, 24–34 (1983).
- ¹⁸⁷M. Ozawa, J.-L. Barrat, W. Kob, and F. Zamponi, "Creating equilibrium glassy states via random particle bonding," *J. Stat. Mech.: Theory Exp.* **2024**, 013303.
- ¹⁸⁸G. Sun and P. Harrowell, "Influence of bond and pinning constraints on configurational restraint in a supercooled liquid," *Phys. Rev. E* **112**, 035407 (2025).
- ¹⁸⁹G. Kapteijns, W. Ji, C. Brito, M. Wyart, and E. Lerner, "Fast generation of ultra-stable computer glasses by minimization of an augmented potential energy," *Phys. Rev. E* **99**, 012106 (2019).
- ¹⁹⁰S. Torquato, "Hyperuniform states of matter," *Phys. Rep.* **745**, 1–95 (2018).
- ¹⁹¹G. Zhang, F. H. Stillinger, and S. Torquato, "The perfect glass paradigm: Disordered hyperuniform glasses down to absolute zero," *Sci. Rep.* **6**, 36963 (2016).
- ¹⁹²J. Kim and S. Torquato, "Methodology to construct large realizations of perfectly hyperuniform disordered packings," *Phys. Rev. E* **99**, 052141 (2019).
- ¹⁹³J. R. Dale, J. D. Sartor, R. C. Dennis, and E. I. Corwin, "Hyperuniform jammed sphere packings have anomalous material properties," *Phys. Rev. E* **106**, 024903 (2022).
- ¹⁹⁴T. Yanagishima, J. Russo, and H. Tanaka, "Common mechanism of thermodynamic and mechanical origin for ageing and crystallization of glasses," *Nat. Commun.* **8**, 15954 (2017).
- ¹⁹⁵T. Yanagishima, J. Russo, R. P. A. Dullens, and H. Tanaka, "From ultra-fast growth to avalanche growth in devitrifying glasses," *J. Chem. Phys.* **159**, 064510 (2023).
- ¹⁹⁶L. Berthier and M. Ediger, "Designing disordered materials beyond equilibrium: Glasses," *Nat. Mater.* **25**, 899–900 (2026).
- ¹⁹⁷H. Tong and H. Tanaka, "Structural order as a genuine control parameter of dynamics in simple glass formers," *Nat. Commun.* **10**, 5596 (2019).
- ¹⁹⁸Y. Wang, Z. Qian, H. Tong, and H. Tanaka, "Hyperuniform disordered solids with crystal-like stability," *Nat. Commun.* **16**, 1398 (2025).
- ¹⁹⁹V. M. Bolton-Lum, R. C. Dennis, P. K. Morse, and E. I. Corwin, "Ideal glass and ideal disk packing in two dimensions," *Phys. Rev. Lett.* **136**, 058201 (2026).
- ²⁰⁰L. Galliano and L. Berthier, "Identifying the relevant parameters in design strategies for stable glasses," *arXiv:2605.12127* (2026).
- ²⁰¹D. Chandler and J. P. Garrahan, "Dynamics on the way to forming glass: Bubbles in space-time," *Annu. Rev. Phys. Chem.* **61**, 191–217 (2010).
- ²⁰²M. Merolle, J. P. Garrahan, and D. Chandler, "Space-time thermodynamics of the glass transition," *Proc. Natl. Acad. Sci. U. S. A.* **102**, 10837–10840 (2005).
- ²⁰³L. O. Hedges, R. L. Jack, J. P. Garrahan, and D. Chandler, "Dynamic order-disorder in atomistic models of structural glass formers," *Science* **323**, 1309–1313 (2009).
- ²⁰⁴P. G. Bolhuis and D. W. H. Swenson, "Transition path sampling as Markov chain Monte Carlo of trajectories: Recent algorithms, software, applications, and future outlook," *Adv. Theory Simul.* **4**, 2000237 (2021).
- ²⁰⁵P. G. Bolhuis, D. Chandler, C. Dellago, and P. L. Geissler, "Transition path sampling: Throwing ropes over rough mountain passes, in the dark," *Annu. Rev. Phys. Chem.* **53**, 291–318 (2002).
- ²⁰⁶F. Ritort and P. Sollich, "Glassy dynamics of kinetically constrained models," *Adv. Phys.* **52**, 219–342 (2003).
- ²⁰⁷R. L. Jack and J. P. Garrahan, "Metastable states and space-time phase transitions in a spin-glass model," *Phys. Rev. E* **81**, 011111 (2010).
- ²⁰⁸T. Speck, A. Malins, and C. P. Royall, "First-order phase transition in a model glass former: Coupling of local structure and dynamics," *Phys. Rev. Lett.* **109**, 195703 (2012).
- ²⁰⁹F. Turci, T. Speck, and C. P. Royall, "Structural-dynamical transition in the Wahnström mixture," *Eur. Phys. J. E* **41**, 54 (2018).
- ²¹⁰C. P. Royall, F. Turci, and T. Speck, "Dynamical phase transitions and their relation to structural and thermodynamic aspects of glass physics," *J. Chem. Phys.* **153**, 090901 (2020).
- ²¹¹Q. Wang and L. Zhang, "Inverse design of glass structure with deep graph neural networks," *Nat. Commun.* **12**, 5359 (2021).
- ²¹²M. L. Falk and J. S. Langer, "Dynamics of viscoplastic deformation in amorphous solids," *Phys. Rev. E* **57**, 7192 (1998).
- ²¹³M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids* (Oxford University Press, 2017).
- ²¹⁴D. Frenkel and B. Smit, *Understanding Molecular Simulation: From Algorithms to Applications* (Elsevier, 2023).
- ²¹⁵J. Liu, Y. Qi, Z. Y. Meng, and L. Fu, "Self-learning Monte Carlo method," *Phys. Rev. B* **95**, 041101 (2017).
- ²¹⁶T. A. Bojesen, "Policy-guided Monte Carlo: Reinforcement-learning Markov chain dynamics," *Phys. Rev. E* **98**, 063303 (2018).
- ²¹⁷V. Bihani, S. Manchanda, S. Sastry, S. Ranu, and N. A. Krishnan, "StriderNet: A graph reinforcement learning approach to optimize atomic structures on rough energy landscapes," in *International Conference on Machine Learning* (PMLR, 2023), pp. 2431–2451.
- ²¹⁸F. Noé, S. Olsson, J. Köhler, and H. Wu, "Boltzmann generators: Sampling equilibrium states of many-body systems with deep learning," *Science* **365**, eaaw1147 (2019).
- ²¹⁹M. Gabrié, G. M. Rotskoff, and E. Vanden-Eijnden, "Adaptive Monte Carlo augmented with normalizing flows," *Proc. Natl. Acad. Sci. U. S. A.* **119**, e2109420119 (2022).
- ²²⁰T. Marchand, M. Ozawa, G. Biroli, and S. Mallat, "Multiscale data-driven energy estimation and generation," *Phys. Rev. X* **13**, 041038 (2023).
- ²²¹L. M. Del Bono, F. Ricci-Tersenghi, and F. Zamponi, "Performance of machine-learning-assisted Monte Carlo in sampling from simple statistical physics models," *Phys. Rev. E* **112**, 045307 (2025).
- ²²²H. Staley, E. Flenner, and G. Szamel, "Cooling-rate dependence of kinetic and mechanical stabilities of simulated glasses," *J. Chem. Phys.* **142**, 244508 (2015).
- ²²³P. Das and S. Sastry, "Crossover in dynamics in the Kob-Andersen binary mixture glass-forming liquid," *J. Non-Cryst. Solids: X* **14**, 100098 (2022).
- ²²⁴N. V. Priezjev, "Structural relaxation and delayed yielding in cyclically sheared Cu–Zr metallic glasses," *Metals* **14**, 984 (2024).
- ²²⁵C. J. Fullerton and L. Berthier, "Density controls the kinetic stability of ultrastable glasses," *Europhys. Lett.* **119**, 36003 (2017).
- ²²⁶A. D. Parmar, B. Guiselin, and L. Berthier, "Stable glassy configurations of the Kob-Andersen model using swap Monte Carlo," *J. Chem. Phys.* **153**, 134505 (2020).
- ²²⁷L. Berthier, M. Ozawa, and C. Scalliet, "Configurational entropy of glass-forming liquids," *J. Chem. Phys.* **150**, 160902 (2019).
- ²²⁸E. Flenner and G. Szamel, "Hybrid Monte Carlo simulation of a glass-forming binary mixture," *Phys. Rev. E* **73**, 061505 (2006).
- ²²⁹M. Tylinski, Y. Z. Chua, M. S. Beasley, C. Schick, and M. D. Ediger, "Vapor-deposited alcohol glasses reveal a wide range of kinetic stability," *J. Chem. Phys.* **145**, 174506 (2016).
- ²³⁰M. Fanfoni and M. Tomellini, "The Johnson–Mehl–Avrami–Kohnogorov model: A brief review," *Nuovo Cimento Soc. Ital. Fis., D* **20**, 1171–1182 (1998).
- ²³¹C. Herrero, C. Scalliet, M. D. Ediger, and L. Berthier, "Two-step devitrification of ultrastable glasses," *Proc. Natl. Acad. Sci. U. S. A.* **120**, e2220824120 (2023).
- ²³²R. N. Chacko, F. P. Landes, G. Biroli, O. Dauchot, A. J. Liu, and D. R. Reichman, "Dynamical facilitation governs the equilibration dynamics of glasses," *Phys. Rev. X* **14**, 031012 (2024).
- ²³³F. Leoni, F. Martelli, and J. Russo, "Devitrification and melting in vapor deposited ice," *J. Chem. Phys.* **163**, 104501 (2025).
- ²³⁴G. Jung, M. Ozawa, G. Biroli, and L. Berthier, "Numerical investigation of the equilibrium kauzmann transition in a two-dimensional atomistic glass," *arXiv:2507.03590* (2025).
- ²³⁵A. Nicolas, E. E. Ferrero, K. Martens, and J.-L. Barrat, "Deformation and flow of amorphous solids: Insights from elastoplastic models," *Rev. Mod. Phys.* **90**, 045006 (2018).
- ²³⁶T. Kawasaki and H. Tanaka, "Structural evolution in the aging process of supercooled colloidal liquids," *Phys. Rev. E* **89**, 062315 (2014).
- ²³⁷T. Jenkinson, P. Crowther, F. Turci, and C. P. Royall, "Weak temperature dependence of ageing of structural properties in atomistic model glassformers," *J. Chem. Phys.* **147**, 054501 (2017).

- ²³⁸G. M. Hocky, D. Coslovich, A. Ikeda, and D. R. Reichman, "Correlation of local order with particle mobility in supercooled liquids is highly system dependent," *Phys. Rev. Lett.* **113**, 157801 (2014).
- ²³⁹R. L. Jack, A. J. Dunleavy, and C. P. Royall, "Information-theoretic measurements of coupling between structure and dynamics in glass formers," *Phys. Rev. Lett.* **113**, 095703 (2014).
- ²⁴⁰J. E. Hallett, F. Turci, and C. P. Royall, "Local structure in deeply supercooled liquids exhibits growing lengthscales and dynamical correlations," *Nat. Commun.* **9**, 3272 (2018).
- ²⁴¹S. Ishino, Y.-C. Hu, and H. Tanaka, "Microscopic structural origin of slow dynamics in glass-forming liquids," *Nat. Mater.* **24**, 268–277 (2025).
- ²⁴²H. Tanaka, "Structural origin of dynamic heterogeneity in supercooled liquids," *J. Phys. Chem. B* **129**, 789–813 (2025).
- ²⁴³C. Patrick Royall, S. R. Williams, T. Ohtsuka, and H. Tanaka, "Direct observation of a local structural mechanism for dynamic arrest," *Nat. Mater.* **7**, 556–561 (2008).
- ²⁴⁴M. Leocmach and H. Tanaka, "Roles of icosahedral and crystal-like order in the hard spheres glass transition," *Nat. Commun.* **3**, 974 (2012).
- ²⁴⁵C. P. Royall and S. R. Williams, "The role of local structure in dynamical arrest," *Phys. Rep.* **560**, 1–75 (2015).
- ²⁴⁶F. C. Frank, "Supercooling of liquids," *Proc. R. Soc. London, Ser. A* **215**, 43–46 (1952).
- ²⁴⁷P. J. Steinhardt, D. R. Nelson, and M. Ronchetti, "Bond-orientational order in liquids and glasses," *Phys. Rev. B* **28**, 784–805 (1983).
- ²⁴⁸H. Jónsson and H. C. Andersen, "Icosahedral ordering in the Lennard–Jones liquid and glass," *Phys. Rev. Lett.* **60**, 2295 (1988).
- ²⁴⁹M. Ozawa, L. Berthier, and D. Coslovich, "Exploring the jamming transition over a wide range of critical densities," *SciPost Phys.* **3**, 027 (2017).
- ²⁵⁰E. Boattini, S. Marín-Aguilar, S. Mitra, G. Foffi, F. Smallenburg, and L. Filion, "Autonomously revealing hidden local structures in supercooled liquids," *Nat. Commun.* **11**, 5479 (2020).
- ²⁵¹F. Martelli, F. Leoni, F. Sciortino, and J. Russo, "Connection between liquid and non-crystalline solid phases in water," *J. Chem. Phys.* **153**, 104503 (2020).
- ²⁵²F. Leoni and J. Russo, "Nonclassical nucleation pathways in stacking-disordered crystals," *Phys. Rev. X* **11**, 031006 (2021).
- ²⁵³G. Jung, R. M. Alkemade, V. Bapst, D. Coslovich, L. Filion, F. P. Landes, A. J. Liu, F. S. Pezzicoli, H. Shiba, G. Volpe *et al.*, "Roadmap on machine learning glassy dynamics," *Nat. Rev. Phys.* **7**, 91–104 (2025).
- ²⁵⁴R. Milkus and A. Zacccone, "Local inversion-symmetry breaking controls the boson peak in glasses and crystals," *Phys. Rev. B* **93**, 094204 (2016).
- ²⁵⁵A. C. Y. Liu, E. D. Bøjesen, R. F. Tabor, S. T. Mudie, A. Zacccone, P. Harrowell, and T. C. Petersen, "Local symmetry predictors of mechanical stability in glasses," *Sci. Adv.* **8**, eabn0681 (2022).
- ²⁵⁶A. Zacccone, "Ultrastable 2D glasses and packings explained by local centrosymmetry," *arXiv:2602.03770* (2026).
- ²⁵⁷H. Tong, P. Tan, and N. Xu, "From crystals to disordered crystals: A hidden order-disorder transition," *Sci. Rep.* **5**, 15378 (2015).
- ²⁵⁸U. Buchenau, Y. M. Galperin, V. L. Gurevich, and H. R. Schober, "Anharmonic potentials and vibrational localization in glasses," *Phys. Rev. B* **43**, 5039–5045 (1991).
- ²⁵⁹V. L. Gurevich, D. A. Parshin, and H. R. Schober, "Anharmonicity, vibrational instability, and the boson peak in glasses," *Phys. Rev. B* **67**, 094203 (2003).
- ²⁶⁰D. Richard, K. González-López, G. Kapteijns, R. Pater, T. Vaknin, E. Bouchbinder, and E. Lerner, "Universality of the nonphononic vibrational spectrum across different classes of computer glasses," *Phys. Rev. Lett.* **125**, 085502 (2020).
- ²⁶¹S. Bonfanti, R. Guerra, C. Mondal, I. Procaccia, and S. Zapperi, "Universal low-frequency vibrational modes in silica glasses," *Phys. Rev. Lett.* **125**, 085501 (2020).
- ²⁶²T. Pérez-Castañeda, C. Rodríguez-Tinoco, J. Rodríguez-Viejo, and M. A. Ramos, "Suppression of tunneling two-level systems in ultrastable glasses of indomethacin," *Proc. Natl. Acad. Sci. U. S. A.* **111**, 11275–11280 (2014).
- ²⁶³H. B. Yu, M. Tylinski, A. Guiseppi-Elie, M. D. Ediger, and R. Richert, "Suppression of β relaxation in vapor-deposited ultrastable glasses," *Phys. Rev. Lett.* **115**, 185501 (2015).
- ²⁶⁴D. Silver, T. Hubert, J. Schrittwieser, I. Antonoglou, M. Lai, A. Guez, M. Lancot, L. Sifre, D. Kumaran, T. Graepel *et al.*, "A general reinforcement learning algorithm that masters chess, shogi, and Go through self-play," *Science* **362**, 1140–1144 (2018).