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Hidden Instabilities in ML Molecular Dynamics of Solid-State Electrolytes: Failure Modes under Finite Temperature

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Abstract

Machine learning molecular dynamics has become a cornerstone for exploring solid-state electrolytes in next-generation batteries. Researchers rely on these potentials to predict ionic conductivity, lithium diffusion, and phase behavior at speeds far beyond traditional density functional theory while maintaining near-DFT accuracy. Yet a critical gap remains: most ML potentials are trained exclusively on static, zero-kelvin structures and energies. When deployed in finite-temperature simulations between 300 K and 1000 K—the actual operating regime of solid-state electrolytes—hidden instabilities emerge that standard benchmarks never detect. This failure-mode analysis identifies five specific instabilities that compromise the reliability of ML-driven molecular dynamics for solid-state electrolytes. Energy drift in long NVT or NVE runs violates conservation laws and produces artificial heating or cooling. Unphysical lithium diffusion pathways appear because transition states and saddle-point configurations are absent from training data, leading to either barrierless motion or spurious trapping. Force discontinuities arise from non-smooth descriptor cutoffs and become amplified by thermal motion. Phonon softening is mispredicted, distorting the vibrational precursors to superionic transitions. Finally, the superionic transition itself is either shifted by more than 100 K, entirely absent, or incorrectly sharp or gradual. These instabilities are invisible in conventional zero-kelvin tests such as energy mean-absolute error or force root-mean-square error on equilibrium structures. They only surface during extended nanosecond-scale simulations at operating temperatures. The present work systematically dissects why each failure mode occurs, provides clear detection signatures observable in any ML molecular-dynamics workflow, and outlines practical mitigation strategies grounded in the literature. By treating finite-temperature stability as a core validation requirement rather than an afterthought, the field can move from promising prototypes to trustworthy tools for solid-state electrolyte design. The analysis draws on recent advances in machine-learning interatomic potentials while highlighting the urgent need for temperature-aware training and testing protocols.

Keywords Machine learning molecular dynamics, Solid-state electrolytes, Finite-temperature instabilities, Energy drift, Ionic conductivity prediction, Interatomic potential transferability

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Introduction

Solid-state electrolytes are essential for safer, higher-energy-density batteries that avoid the flammable liquids used in conventional lithium-ion cells [1, 2]. Researchers have turned to machine learning molecular dynamics because it promises density-functional-theory-level accuracy at a fraction of the computational cost, enabling nanosecond-scale simulations of lithium diffusion and ionic conductivity that were previously inaccessible [3–5]. Early demonstrations on

materials such as Li₆PS₅Cl have shown that ML potentials can reproduce diffusion pathways and conductivity trends with impressive fidelity when tested under carefully controlled conditions [3].

Nevertheless, a hidden problem undermines many of these promising results. Most ML potentials are trained on static, zero-kelvin configurations obtained from density functional theory calculations [6]. When the same potentials are placed into molecular-dynamics runs at realistic operating temperatures of 300–1000 K, instabilities appear that were never anticipated by standard benchmarks [7, 8]. Energy conservation breaks down over time, lithium ions follow pathways that do not exist in reality, forces exhibit abrupt jumps, phonon spectra fail to evolve correctly with temperature, and superionic phase transitions are either missed or misplaced [9–11].

These failure modes are not minor numerical artifacts. They directly corrupt the predicted ionic conductivity—the very property that determines whether a candidate electrolyte will succeed or fail in a battery [11, 12]. Because the instabilities remain invisible in short zero-kelvin tests or brief picosecond runs, many published studies inadvertently report misleading performance metrics [13, 14]. The community therefore needs a systematic failure-mode analysis that moves beyond accuracy tables and focuses on long-term dynamical stability at finite temperature.

This paper provides exactly that analysis. It first explains why solid-state electrolytes are uniquely challenging for current ML potentials. It then dissects five distinct failure modes, each with a precise definition, mechanistic explanation, observable detection signature, and practical consequences for electrolyte design. The work builds directly on recent studies of ML-accelerated modeling of thiophosphate and sulfide electrolytes [15, 16], while extending insights from broader investigations of temperature transferability in interatomic potentials [17–19]. By the end of this sectioned analysis, readers will possess both a diagnostic toolkit and a set of mitigation principles that can be applied immediately to any new ML potential intended for solid-state electrolyte research.

The urgency is clear. As the field accelerates toward high-throughput screening of thousands of candidate compositions, undetected finite-temperature instabilities risk steering materials discovery toward false positives or discarding genuinely promising candidates [1, 13, 20]. Only by confronting these hidden instabilities head-on can machine learning molecular dynamics fulfill its promise for solid-state battery innovation.

What Makes Solid-State Electrolytes Challenging for ML Potentials

Solid-state electrolytes present a combination of physical and data-related difficulties that expose limitations in today's machine learning interatomic potentials. Five interlocking factors turn what appears to be a straightforward regression task into a demanding test of generalization [6].

The first factor is anharmonicity. At operating temperatures of 300–1000 K, atomic vibrations in solid-state electrolytes deviate strongly from the simple harmonic picture assumed in zero-kelvin training data [8]. Lattice vibrations become large enough that atoms explore regions of the potential-energy surface far from equilibrium minima. Most ML potentials, trained on near-equilibrium configurations, cannot accurately extrapolate into these anharmonic regimes [7].

The second factor is lithium mobility. Lithium ions must hop through a relatively rigid anion framework via narrow channels and saddle-point configurations. Accurate prediction of the energy barriers—typically between 0.1 and 0.5 eV—is essential [12]. Yet the training sets used for most potentials contain few or no explicit transition-state geometries, leaving the models to interpolate across regions where small errors produce qualitatively wrong diffusion behavior [15, 16].

The third factor concerns long time scales. Realistic lithium diffusion in solid-state electrolytes occurs on nanosecond or longer timescales [9, 10]. Molecular-dynamics simulations therefore require millions of time steps. Even tiny per-step force inaccuracies accumulate, producing macroscopic trajectory drift that invalidates conductivity predictions. Studies of

thiophosphate electrolytes have already shown that seemingly accurate short-run simulations can diverge dramatically once extended [15].

The fourth factor is the superionic transition. Many promising solid-state electrolytes undergo a first-order or continuous transition into a disordered, highly conductive phase. Conductivity can jump by several orders of magnitude within a narrow temperature window [10, 11]. Capturing this sharp change requires the potential to reproduce both the low-temperature ordered structure and the high-temperature disordered state with equal fidelity—an extremely difficult requirement when training data are drawn almost exclusively from zero-kelvin minima.

The fifth and perhaps most fundamental factor is training-data bias. Virtually all current ML potentials for solid-state electrolytes are fitted to static density-functional-theory snapshots of equilibrium crystal structures or small thermal ensembles at low temperature [6]. Deployment, however, occurs at finite temperature with continuous lithium diffusion and frequent exploration of non-equilibrium geometries. This creates a severe distribution shift that standard validation metrics—energy and force errors on held-out zero-kelvin structures—completely miss [8, 17]. Recent reviews of machine-learning force fields have emphasized the same distribution-shift problem across materials classes, yet the consequences are especially acute for solid-state electrolytes where ionic transport is the central property of interest [6].

Taken together, these five factors explain why ML potentials that perform well on conventional benchmarks can still fail catastrophically once placed into production molecular-dynamics runs at operating temperature [13, 14].

Figure 1 maps the manuscript's core argument as a hierarchical failure pathway, showing how zero-kelvin training bias propagates through finite-temperature deployment challenges into five distinct dynamical instabilities that ultimately distort conductivity prediction.

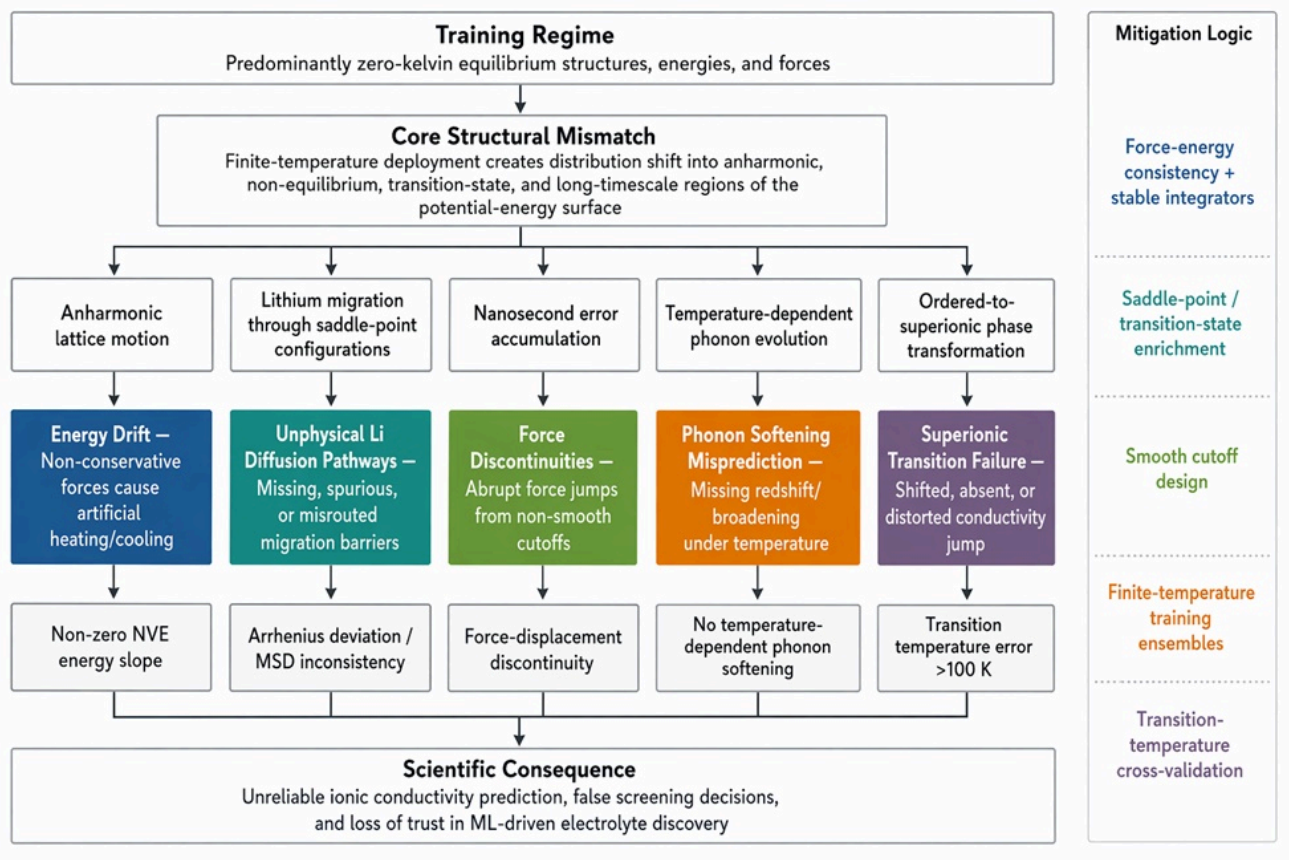


Figure 1. Hierarchical failure pathway linking zero-kelvin training bias to finite-temperature instability in ML molecular dynamics of solid-state electrolytes

The remainder of this analysis examines the concrete failure modes that result.

Energy Drift in NVT/NPT Simulations

Energy drift is the most fundamental and easily quantifiable instability in ML molecular dynamics of solid-state electrolytes. It is defined as a systematic, linear increase or decrease in the total conserved energy of the system during NVE (microcanonical) or properly thermostatted NVT (canonical) simulations. In an ideal conservative force field the total energy should fluctuate around a constant mean; any persistent upward or downward trend signals that the predicted forces are not exact derivatives of the predicted energy [21].

The root cause lies in imperfect force-energy consistency. Even when an ML model is trained with a loss term that penalizes force errors, the learned potential surface is never perfectly differentiable everywhere. Tiny inconsistencies accumulate with each integration step. At finite temperature the effect is amplified because thermal fluctuations repeatedly push atoms into regions where the local curvature of the potential is poorly constrained [18, 22]. Over hundreds of picoseconds to nanoseconds the cumulative error produces a measurable drift rate, often exceeding 1×10^{-5} eV per picosecond per atom.

For solid-state electrolytes the consequences are immediate and severe. Artificial energy drift translates into uncontrolled heating or cooling of the simulation cell. Because ionic conductivity is exponentially sensitive to temperature, even a few kelvin of spurious drift can change the predicted lithium diffusion coefficient by tens of percent [10, 12]. Conductivity values extracted from such runs therefore become unreliable for materials screening. In extreme cases the simulation cell can drift into unphysical temperature regimes that trigger premature phase changes or structural collapse.

Standard zero-kelvin benchmarks miss this failure entirely. A static force root-mean-square error calculation on a handful of equilibrium structures cannot reveal whether forces remain conservative over millions of dynamical steps [6]. Short test runs of 10 ps or less rarely show statistically significant drift; the instability only becomes apparent in the long trajectories required for converged conductivity statistics. Recent work on machine-learning potentials for high-temperature dynamics has begun to document similar drift phenomena in metallic systems, underscoring that the problem is not unique to electrolytes but is especially damaging when the scientific goal is quantitative transport prediction [21].

Detection is straightforward: monitor the total energy time series in an NVE run at the target operating temperature and fit a linear slope. A non-zero slope above the threshold of 1×10^{-5} eV/ps/atom flags the failure mode. Once identified, the drift must be addressed before any conductivity or diffusion results can be trusted.

Unphysical Li Diffusion Pathways

Unphysical lithium diffusion occurs when an ML potential predicts hopping mechanisms, barriers, or site occupancies that contradict both higher-level density-functional-theory calculations and experimental observation. Four subtypes are commonly observed. Type A is missing barriers, where lithium appears to move almost freely through the lattice because the model has created artificially low-energy corridors. Type B is spurious barriers that trap lithium ions in sites that should be transit points. Type C involves entirely wrong pathways that route lithium through interstitial positions forbidden by the anion framework. Type D concerns collective motion errors, in which correlated lithium hops—known to dominate transport in many thiophosphates—are replaced by independent single-ion jumps [9, 15].

The underlying mechanism is training-data incompleteness. Most datasets contain only equilibrium crystal structures and perhaps small thermal displacements around those minima [6]. Transition states and the high-energy configurations

visited during actual diffusion are statistically rare and therefore underrepresented or absent. The ML model is forced to extrapolate, often producing smooth but incorrect interpolations between known minima. Finite-temperature molecular dynamics then samples precisely those extrapolated regions, exposing the flaws [17].

Detection signatures are clear in post-processing. Lithium diffusivity extracted from mean-square-displacement curves can be more than ten times higher or lower than reference density-functional-theory values at the same temperature [10, 12]. Arrhenius plots of log diffusivity versus inverse temperature become non-linear or exhibit slopes that deviate markedly from experiment. Trajectory visualization reveals lithium ions occupying interstitial sites never seen in reference calculations. In collective-motion failures, the Haven ratio (ratio of tracer to conductivity diffusion) deviates from values established by nuclear magnetic resonance or impedance spectroscopy.

The practical impact on solid-state electrolyte research is profound. Overestimated diffusivity produces false-positive candidates that appear superionic in simulation but fail in real devices. Underestimated diffusivity leads to premature rejection of viable materials. Both outcomes waste experimental resources and slow the discovery pipeline. Earlier studies of ionic conductivity prediction with machine-learning potentials have already encountered similar discrepancies, yet the community has not yet adopted routine diffusion-pathway validation as standard practice [11, 12].

Force Discontinuities, Phonon Softening, Superionic Transition Failure

ML potentials can exhibit force discontinuities when atomic configurations cross descriptor cutoff boundaries or when non-smooth switching functions are activated, leading to abrupt changes in predicted forces [23]. Under finite-temperature conditions, thermal fluctuations repeatedly drive atoms across these regions, effectively injecting non-physical impulses into the dynamical evolution. This undermines energy conservation and can induce artificial energy transfer between degrees of freedom, occasionally destabilizing trajectories. In practice, such non-smoothness is most clearly revealed by inspecting force components under infinitesimal atomic displacements, where discontinuities emerge as sharp, unphysical jumps.

A related limitation arises in the treatment of lattice dynamics, where phonon softening is often misrepresented. In real solid-state electrolytes, increasing temperature enhances anharmonicity, progressively reducing phonon frequencies and signaling lattice softening that can precede superionic transitions [10]. However, models trained predominantly on zero-Kelvin configurations frequently yield phonon densities of states that remain effectively invariant with temperature [8]. This absence of thermally driven spectral evolution leads to systematic errors in vibrational entropy and distorts the free-energy landscape governing disorder.

These inaccuracies propagate directly into the prediction of superionic transitions, where the conductivity increase may be misplaced, suppressed entirely, or reproduced with unrealistic sharpness or excessive smearing [9-11]. Since this transition defines functional performance in many electrolyte systems, such deviations fundamentally compromise the reliability of materials screening pipelines. In practice, the issue becomes evident when ionic conductivity is evaluated across temperature trajectories and the predicted transition temperature or magnitude fails to align with experimental or density-functional-theory benchmarks.

These behaviors are not independent but emerge from a shared deficiency in how finite-temperature anharmonic configurations are represented during training [17, 19]. Force-level discontinuities destabilize dynamical sampling, while incorrect phonon softening reshapes the inferred thermodynamic landscape, jointly biasing the emergence of the superionic phase. Although recent efforts in temperature transferability and multi-fidelity learning partially address related challenges, robust validation frameworks specifically tailored to solid-state electrolytes remain limited [13, 14].

Table 1 consolidates the five failure modes into a single analytical matrix that links physical origin, benchmark blind spot, observable simulation signature, quantitative failure threshold, and downstream scientific consequence.

Table 1. Analytical failure-mode matrix for finite-temperature instability in ML molecular dynamics of solid-state electrolytes

Failure mode	Immediate physical origin	Why zero-kelvin benchmarking misses it	Primary observable signature in production MD	Quantitative failure criterion	Scientific consequence
Energy drift	Imperfect force-energy consistency; local non-conservative behavior amplified during long integration	Static energy/force error on equilibrium structures does not test conservation over millions of steps	Monotonic slope in total energy during long NVE/NVT runs	Drift rate > 1×10^{-5} eV/ps/atom	Artificial heating/cooling and distorted ionic conductivity
Unphysical Li diffusion pathways	Missing saddle points, transition states, and correlated hopping configurations in training data	Held-out equilibrium snapshots do not probe migration topology or barrier fidelity	Nonphysical Li occupancy, wrong pathways, anomalous Arrhenius behavior	Barrier error > 0.1 eV or diffusivity deviation > order-of-magnitude vs reference	False positives or false negatives in electrolyte screening
Force discontinuities	Non-smooth descriptors, hard cutoffs, or switching artifacts	Conventional metrics average errors and obscure localized discontinuities	Abrupt force jumps under infinitesimal displacement; simulation instability	Detectable discontinuity in force-displacement scan	Spurious impulse transfer, poor stability, possible trajectory collapse
Phonon softening misprediction	Inability to represent anharmonic temperature dependence of lattice dynamics	Zero-kelvin validation contains no test of vibrational evolution with temperature	No systematic redshift or broadening in phonon DOS across temperature ladder	Absence of expected temperature-dependent softening	Misrepresentation of vibrational entropy and transition precursors
Superionic transition failure	Poor free-energy balance between ordered and disordered states; missing finite-temperature configurations	Static benchmarks do not test conductivity discontinuities or phase crossover behavior	Shifted, absent, smeared, or unrealistically sharp conductivity jump	Transition temperature error > 100 K	Invalid prediction of usable operating window and material ranking

Detection Principles

Detection of hidden instabilities in ML molecular dynamics for solid-state electrolytes necessitates moving beyond static zero-Kelvin benchmarks toward diagnostics that interrogate long-timescale behavior under operating conditions, where a set of complementary validation strategies grounded in established computational workflows becomes essential [9, 10, 21]. A central requirement is the long-timescale NVE stability assessment, in which nanosecond-scale trajectories at relevant temperatures (typically 300–600 K) are used to evaluate total energy conservation through linear drift fitting; deviations exceeding 1×10^{-5} eV per picosecond per atom indicate a breakdown of dynamical consistency [21]. In parallel, transport fidelity is assessed through lithium self-diffusion analysis derived from mean-square displacement, where Arrhenius relationships between diffusivity and inverse temperature expose both slope distortions and non-linearity as signatures of unphysical migration mechanisms [10, 12]. Complementing these dynamical checks, migration energetics are probed via nudged elastic band calculations with the ML potential, where discrepancies greater than 0.1 eV relative to density-functional-theory barriers reveal inadequate representation of saddle-point regions and latent training-data sparsity [15, 17].

Beyond transport and barrier consistency, lattice dynamical behavior provides a more sensitive probe of anharmonic fidelity, as physically accurate potentials must reproduce temperature-dependent phonon softening and spectral broadening extracted from equilibrated trajectories across increasing thermal conditions; the absence of such systematic evolution indicates a failure to capture essential anharmonic effects [8, 10]. This deficiency propagates directly into phase-transition physics, where ionic conductivity evaluated across a temperature ladder should exhibit a well-defined superionic transition; deviations manifested as shifted, absent, or excessively smeared conductivity jumps by more than 100 K undermine predictive reliability for electrolyte screening [9, 11]. Taken together, these diagnostics expose how seemingly stable short-time behavior can mask deeper thermodynamic inconsistencies, underscoring that robust validation of ML-driven molecular dynamics for solid-state electrolytes must explicitly interrogate long-time stability, transport, vibrational spectra, and emergent phase behavior within a unified evaluation framework [13, 14].

Mitigation Principles

Once instabilities are detected, targeted mitigation strategies restore reliability without sacrificing the speed advantage of machine learning potentials. Twelve principles, grouped by failure mode, provide actionable fixes drawn from recent advances in interatomic-potential development [17, 19, 21].

Table 2 translates the manuscript's detection and mitigation sections into an operational validation-to-mitigation framework, clarifying which test should govern model acceptance and which corrective action is most appropriate when a given test fails.

Table 2. Validation-to-mitigation alignment framework for making ML molecular dynamics of solid-state electrolytes decision-ready

Validation principle	Failure mode(s) primarily tested	What the test reveals conceptually	Minimum practical implementation	Decision rule	Most directly aligned mitigation principle(s)
Long MD energy-conservation test	Energy drift; secondarily force discontinuities	Whether the learned potential behaves as a dynamically conservative force field rather than	NVE run ≥ 1 ns at target temperature; fit energy slope	Fail if slope $> 1 \times 10^{-5}$ eV/ps/atom	Energy-conserving integrators; force-energy consistency constraints;

		only a good static regressor			equivariant architectures
Diffusion validation across temperatures	Unphysical Li diffusion pathways; superionic transition failure	Whether transport topology and activation behavior remain physically credible under deployment conditions	MSD-derived diffusivity at multiple temperatures; Arrhenius comparison	Fail if slope/intercept deviate strongly or curve becomes nonphysical	Transition-state enrichment; active learning on diffusion paths; periodic barrier validation
Migration-barrier benchmarking	Unphysical Li diffusion pathways	Whether the model reproduces the topology of the transport landscape rather than only endpoint stability	NEB barriers between known Li sites compared with DFT	Fail if barrier error > 0.1 eV	Saddle-point inclusion; active learning; barrier-based stopping criteria
Phonon temperature-dependence test	Phonon softening misprediction; superionic transition failure	Whether the model captures anharmonic lattice evolution that precedes fast-ion disordering	Phonon DOS from equilibrated trajectories at 0 K / 300 K / 600 K or equivalent	Fail if no systematic softening/broadening appears	Finite-temperature training ensembles; temperature-aware model inputs
Transition-temperature ladder	Superionic transition failure; integrates all upstream errors	Whether conductivity emergence occurs at the correct thermodynamic point and with credible sharpness	Conductivity calculation across dense temperature ladder around expected transition	Fail if transition is absent, shape is implausible, or error > 100 K	Multi-fidelity refinement; experimental cross-validation
Integrated decision gate	All five failure modes	Whether the potential is fit for screening use, mechanistic interpretation, or neither	Pass all five validation layers before reporting conductivity claims	Use only if all critical tests pass	Treat finite-temperature stability as a design constraint, not post-hoc verification

For energy drift, Principle 1 recommends energy-conserving integrators such as the reversible reference system propagator algorithm, which reduces numerical accumulation of inconsistencies during long runs. Principle 2 requires explicit force-energy consistency constraints during training so that predicted forces remain exact derivatives of the predicted energy surface [21]. Principle 3 favors equivariant network architectures that inherently respect physical symmetries and improve long-term conservation [17, 19, 24].

For unphysical lithium diffusion, Principle 4 calls for deliberate inclusion of transition-state and saddle-point configurations in the training set, either generated by density-functional-theory nudged-elastic-band calculations or extracted from short ab-initio molecular-dynamics runs. Principle 5 advocates active learning loops that iteratively sample high-uncertainty lithium diffusion pathways and add them to the training data [15]. Principle 6 embeds periodic validation of migration barriers against density-functional-theory during model refinement, halting training when barrier errors exceed 0.1 eV.

For force discontinuities, Principle 7 replaces hard cutoffs with smooth cosine-taper functions that eliminate abrupt changes in descriptor values. Principle 8 increases the cutoff radius while ensuring smooth decay, preventing thermal motion from repeatedly crossing artificial boundaries [23].

For phonon softening misprediction, Principle 9 shifts training data from purely zero-kelvin snapshots to finite-temperature density-functional-theory ensembles, allowing the model to learn anharmonic effects directly [8]. Principle 10 incorporates temperature as an explicit input feature during model construction, enabling the network to condition its predictions on the thermodynamic state [18].

For superionic transition failure, Principle 11 employs multi-fidelity learning: low-fidelity potentials screen large numbers of candidates quickly while high-fidelity models refine the transition temperature for promising compositions [13]. Principle 12 mandates experimental cross-validation of the predicted transition temperature before any ML-derived conductivity data are used in materials discovery campaigns [11].

Applying these principles in combination transforms ML potentials from fragile prototypes into robust tools for solid-state electrolyte research. The key is to treat finite-temperature stability as a design constraint rather than a post-hoc check [14].

Relation to Other Failure Modes

The five instabilities identified here are not isolated; they interconnect with broader challenges documented in the machine-learning interatomic-potential literature. Energy drift, for example, shares mechanistic roots with long-term stability problems observed in high-temperature molecular-dynamics simulations of metals and alloys [21, 25]. Earlier proposals for error-correcting potentials address similar cumulative inaccuracies, yet the present analysis pinpoints why solid-state electrolytes are particularly vulnerable because of their nanosecond-scale lithium transport requirements [21, 26].

Missing long-range electrostatic interactions exacerbate both energy drift and force discontinuities in ionic materials such as thiophosphates [15, 27]. Many solid-state electrolytes rely on long-range Coulomb forces that short-range ML descriptors often under-represent, leading to the same instabilities that appear when temperature drives atoms across cutoff boundaries. Recent foundation models that incorporate explicit polarizable long-range terms demonstrate reduced drift, confirming the link [27].

Temperature extrapolation lies at the core of all five failure modes. Most potentials are trained on zero-kelvin or low-temperature data and then deployed at 300–1000 K, creating the distribution shift that triggers unphysical diffusion, phonon misprediction, and superionic-transition errors [8, 17]. This explains why instabilities remain hidden in conventional benchmarks yet surface immediately in production runs [7].

Active learning offers a unifying mitigation pathway. By iteratively sampling configurations that expose high uncertainty—especially transition states and high-temperature disordered structures—active learning directly addresses the training-data bias responsible for unphysical pathways and transition failures [13, 15]. When combined with the detection principles outlined earlier, active learning closes the loop between diagnosis and model improvement.

Overall, the hidden instabilities in solid-state electrolytes are special cases of general limitations in ML force fields, amplified by the unique demands of ionic transport at finite temperature [6, 28]. Recognizing these relations allows the community to borrow solutions developed for other material classes while tailoring them to electrolyte-specific needs [14].

Implications for Solid-State Electrolyte Simulation

The failure-mode analysis carries direct consequences for three stakeholder groups: practitioners, model developers, and benchmark designers.

For practitioners running ML molecular dynamics on candidate solid-state electrolytes, the central message is caution. Never trust predicted ionic conductivity or lithium diffusivity without first applying the full suite of finite-temperature detection tests. A brief zero-kelvin energy and force check is insufficient; long nanosecond-scale runs at operating temperature must become routine before any publication or device-design decision [9, 10]. When drift or unphysical diffusion is detected, results should be discarded or flagged until mitigation strategies are applied.

For model developers, the implications are design-oriented. Finite-temperature configurations, transition states, and explicit temperature dependence must enter the training pipeline from the outset rather than as afterthoughts [8, 17, 19]. Reporting only zero-kelvin accuracy metrics is no longer acceptable; every new potential intended for solid-state electrolytes should include energy-drift rates, Arrhenius-plot fidelity, and superionic-transition temperatures in its publication. Equivariant architectures and smooth descriptors should be prioritized because they address multiple failure modes simultaneously [17].

For benchmark designers, the analysis highlights the need for electrolyte-specific suites that go beyond static error tables. Future benchmarks must incorporate long MD stability tests, temperature-dependent phonon calculations, and conductivity-versus-temperature curves derived from nanosecond trajectories [13, 14]. Such suites would prevent the publication of models that look excellent on paper but collapse under realistic operating conditions.

Collectively these implications shift the culture of ML-driven materials simulation from speed-first accuracy to stability-first reliability. Only when finite-temperature validation becomes standard will machine learning molecular dynamics deliver on its promise to accelerate solid-state electrolyte discovery and deployment [6, 11, 20].

Conclusion

Machine learning molecular dynamics offers unprecedented speed for exploring solid-state electrolytes, yet hidden instabilities at finite temperature undermine its reliability. This failure-mode analysis has identified five interconnected problems: energy drift in long simulations, unphysical lithium diffusion pathways, force discontinuities, phonon softening misprediction, and superionic transition failure. Each arises because most potentials are trained on static zero-kelvin data and then deployed in regimes where anharmonicity, long time scales, and distribution shift expose their weaknesses.

The instabilities are undetectable by conventional benchmarks yet produce quantitatively wrong ionic conductivity predictions—the property that ultimately decides electrolyte performance in batteries. Detection is straightforward using long MD runs, Arrhenius validation, barrier calculations, phonon temperature scans, and transition-temperature ladders. Mitigation is equally practical: energy-conserving integrators, transition-state inclusion via active learning, smooth cutoffs, finite-temperature training data, and multi-fidelity validation.

The field must now treat finite-temperature stability as a non-negotiable requirement rather than an optional extra. Practitioners should run the proposed detection tests before trusting any conductivity result. Developers must embed temperature-aware constraints in model architecture and training. Benchmark creators should build electrolyte-specific suites that reward dynamical robustness.

By confronting these hidden instabilities head-on, machine learning molecular dynamics can evolve from a promising prototype into a trustworthy engine for solid-state battery innovation. The five failure modes and their corresponding detection and mitigation principles provide a clear roadmap. Implementing them will ensure that the next generation of solid-state electrolytes is discovered and optimized with confidence rather than hidden artifacts.

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