

Loop-level surrogate modeling of dopant-distribution effects in Ba(Zr, Ti)O₃

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Barium titanate-based perovskites are important candidates for lead-free dielectric and electromechanical technologies. In Zr-substituted BaTiO₃, functional behavior is usually discussed in terms of the average Zr concentration, while the influence of dopant spatial distribution beyond average concentration is less understood and difficult to explore systematically. Here we present an accelerated materials-design workflow that links controlled dopant distributions to full field-driven response curves. We generate a broad set of Zr distributions spanning a continuum of nanoscale arrangements, with layers, rods, dots, and lamellae serving as representative end-member motifs, and encode each configuration using a compact, parametrized descriptor model. Effective-Hamiltonian molecular dynamics is used to compute polarization–electric-field and strain–field hysteresis loops and we train a conditional autoencoder surrogate to predict complete loops directly from the distribution parameters. This surrogate enables rapid screening and dense, property-selective design maps at scales that are not feasible with direct simulations alone and it supports targeted follow-up simulations in regions of interest. Using the predicted loop database, we screen the distribution space for multiple functional targets, including energy-storage performance, electromechanical response, and switching behavior, and identify the corresponding dopant distribution motif families. The resulting design maps show that dopant distribution is an independent tuning parameter that can strongly affect hysteresis behavior and loop-derived figures of merit: layerlike motifs, vertical lamellae, and nanoplatelike inclusions emerge in different performance regimes. More generally, predicting full response curves enables screening of other loop-derived targets and multiobjective design in substituted ferroelectrics.

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I. INTRODUCTION

Ferroelectric materials [1,2] show a broad range of functional responses, including dielectric [3], electromechanical [2], and electrocaloric effects [4,5], and therefore remain important for many technologies. A key advantage of perovskite ferroelectrics is their chemical and structural tunability: relatively small changes in composition, local order, or defects can strongly modify phase stability and field-driven switching. Among lead-free perovskites, BaTiO₃ (BT) remains a benchmark system because its phase sequence, switching behavior, and chemical tunability are particularly well established [6–8]. BT exhibits the well-known sequence of structural phases [9] (cubic paraelectric → tetragonal → orthorhombic → rhombohedral) and its rich phase behavior already illustrates that functional response is governed not only by average composition but also by how local environments and correlations develop across length scales.

Chemical substitution on the A or B site is widely used to tune perovskite ferroelectrics [8,10–14]. Most studies parametrize substitution primarily through the average

dopant fraction, which shifts phase boundaries, modifies susceptibilities, and can induce relaxorlike behavior [10]. At fixed composition, however, the same dopant content can be realized through very different real-space arrangements—random, clustered, compositionally modulated, layered, or lamellar—and these arrangements need not yield the same field-driven response. Layering, compositional modulation, and atomically sharp substitution profiles are already established design motifs in ferroelectric heterostructures and superlattices. However, a systematic and quantitative understanding of how concentration and distribution jointly shape field-driven response, and how distribution effects compare at a given dopant fraction, is still limited.

In this work we focus on Zr-substituted BaTiO₃, Ba(Zr_xTi_{1-x})O₃ (BZT) [12–15]. Substitution of Ti by Zr on the B site modifies phase stability and functional behavior: increasing Zr content changes the BT phase sequence [16,17], suppresses ferroelectric order, and eventually drives a crossover toward relaxorlike response [13,16–18]. More broadly, prior work on complex perovskites has shown that the emergence of ferroelectric versus relaxorlike behavior is closely tied to local structural arrangement [19] and simulation studies have emphasized that dielectric response is highly sensitive to local heterogeneity and its dynamics [20]. Recent effective-Hamiltonian work on BZT has further shown that Zr substitution can stabilize nontrivial topological polarization textures, underscoring that chemically controlled local structure can generate qualitatively distinct polarization states beyond trends set by average composition alone [21]. Most

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studies of BZT nonetheless treat the average Zr concentration as the main control variable. Here we shift the emphasis and ask how much additional control resides in the nanoscale spatial arrangement of Zr at fixed composition.

Answering this question requires two ingredients. First, the distribution space must be described in a controlled way so that different motifs can be compared quantitatively. Second, the resulting structure space must be explored at a scale that is large enough to reveal reliable trends and to support design decisions. This is difficult with direct atomistic sampling alone: the number of distinct dopant arrangements grows rapidly with system size and motif complexity and many figures of merit are determined by full polarization–electric-field (P–E) and strain–field (S–E) response curves rather than by a small set of scalar parameters. Moreover, meaningful comparisons often require separating concentration-driven changes from distribution-driven changes, for example by analyzing distributions at similar Zr concentration ($zcon$). For this reason, a loop-level description is particularly useful: once full response curves are available, energy-storage measures, electromechanical indicators, and switching characteristics can all be extracted consistently and multiobjective screening becomes possible when several targets must be balanced.

We therefore develop a simulation-driven surrogate workflow that makes dopant-distribution engineering computationally tractable. We first generate controlled Zr configurations in BT supercells, spanning layers, rods, dots, lamellae, and intermediate motifs, and describe them using a compact parametrized descriptor model inspired by geological pattern characterization. This provides a low-dimensional but physically interpretable representation of the distribution space, enabling systematic sampling and direct comparison across motif classes. We then compute the field-driven response of these configurations using first-principles-parametrized effective-Hamiltonian molecular dynamics (EH–MD), obtaining finite-temperature P–E and S–E hysteresis loops. To move beyond the directly simulated data set, we train a conditional autoencoder surrogate [22–24] that predicts complete hysteresis responses from the distribution descriptors. The gain in screening efficiency is substantial: while generating on the order of 10^5 loops with EH–MD would require roughly 2.7 million core hours, the trained surrogate predicts the same number in minutes on a conventional computer. This makes it possible to construct dense design maps, screen for property-specific targets at scale, and select promising candidates for targeted EH–MD follow-up.

Machine learning has also been used to analyze ferroelectric switching and hysteretic responses in several related settings, including deep recurrent autoencoders for piezoresponse spectroscopy and encoder-decoder models linking domain structure to switching behavior [25,26]. More recently, Lahbabi *et al.* [27] combined phase-field simulations and variational autoencoders to connect hysteresis-loop features to microstructural parameters in $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$. Relative to that literature, the present work targets a different design variable—dopant distribution in BZT—and conditions the surrogate directly on explicit real-space distribution descriptors to predict coupled loop-level responses.

Using BZT as a test case, we show that dopant distribution acts as an additional design variable that can strongly

reshape hysteresis behavior and the resulting loop-derived figures of merit. Based on surrogate-generated design maps, we screen the descriptor space for energy-storage performance, electromechanical response, and switching characteristics and identify the motif families associated with different response regimes. Although layered compositional modulation and related superlattice concepts are already well established in ferroelectrics [28–32], the present study is not limited to that motif class. Its main contribution is a quantitative loop-level workflow that connects compact descriptors of dopant arrangement to full field-driven responses, thereby enabling systematic screening, interpretation, and targeted optimization across concentration and distribution variables.

II. METHODOLOGY

In this section, we introduce the simulation and modeling framework used to generate and analyze Zr-substituted BaTiO_3 supercells. We first describe the effective-Hamiltonian molecular-dynamics approach used to compute P–E and S–E responses. We then introduce the parametrized model used to generate controlled spatial distributions of Zr within the supercells, followed by a description of the data set construction and the extraction of key functional quantities. Together, these components form the basis for the machine-learning framework developed in the subsequent section.

A. Effective Hamiltonian simulations

The atomistic response in this work is computed using molecular dynamics based on first-principles-derived effective Hamiltonians [17,33–37], a framework that has been used extensively to describe finite-temperature behavior in ferroelectric perovskites and their chemically complex variants [38–41]. In this approach, the potential-energy surface is projected onto a reduced set of physically relevant lattice degrees of freedom [33,34], most importantly local polar distortions and their couplings to strain, which allows large-scale finite-temperature simulations [18] at tractable cost while retaining the key ingredients that govern ferroelectric switching and phase behavior [17,42].

For pure BT we employ the effective-Hamiltonian parametrization of Mayer *et al.* [17], which incorporates anharmonic couplings to additional optical modes [17,42] and reproduces the experimental transition sequence with improved accuracy [17]. For $\text{Ba}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ we use the corresponding BZT extension introduced by the same authors [12], which explicitly accounts for Zr substitution on the B site and reproduces the main features of the experimental composition-temperature phase behavior. All simulations were performed with a modified version of *feram* [11,36,37].

Because a central objective of this study is the evaluation of dynamic hysteresis behavior, we briefly summarize the key simulation parameters. All simulations were carried out at 300 K with a time step of 2 fs. Prior to field cycling, systems were equilibrated for 200 ps. Polarization vs electric-field (P–E) and strain vs electric-field (S–E) loops were obtained by applying an oscillating electric field along the z direction (pseudocubic [001] direction) with a maximum amplitude of 1500 kV/cm and a frequency of 1 GHz. Two full cycles were

simulated: the first to eliminate transient effects and the second for data collection. The field-cycling protocol is chosen as a computationally consistent probe of relative hysteresis behavior across configurations rather than as a direct representation of experimental operating conditions. Temperature control was maintained using a velocity-scaling thermostat. All simulations employed a $40 \times 40 \times 40$ ($16 \text{ nm} \times 16 \text{ nm} \times 16 \text{ nm}$) supercell (described in detail in the following section) with fully periodic three-dimensional boundary conditions.

B. Parametrized Zr distribution model

As outlined in the Introduction, a central objective of this work is to examine how different spatial arrangements of Zr ions within a BT matrix influence dielectric and electromechanical behavior. All simulations are based on $40 \times 40 \times 40$ supercells with periodic boundary conditions, providing 64 000 B-site lattice positions available for $\text{Ti} \rightarrow \text{Zr}$ substitution. Even at fixed Zr concentration, the number of possible substitution patterns is combinatorially large, making an explicit enumeration infeasible. A controlled parametrization of spatial distributions is therefore essential to enable systematic exploration and subsequent machine-learning analysis.

To address this challenge, we introduce a constructive parametrization of an idealized but diverse motif space for Zr distributions, inspired by geological facies modeling [43,44]. Accordingly, the present model focuses on controlled ordered motifs and their intermediates. Supercells are generated through a discrete layer-based procedure governed by a small set of tunable descriptors. Within this construction scheme, each three-dimensional Zr distribution is generated from five structural descriptors: $zcon$ (Zr concentration), $intv$ (number of active intervals), hff (horizontal fill factor), hfr (horizontal form ratio), and vsr (vertical superposition ratio).

The overall Zr concentration ($zcon$) is the fundamental parameter specifying the fraction of substituted B sites and therefore constraining all other descriptors. Although it could range from 0 to 1, this study restricts $zcon$ to 0–0.3, where structural distribution effects remain pronounced; at higher concentrations the response increasingly approaches flattened P–E behavior with reduced configurational sensitivity.

The number of active vertical intervals ($intv$) determines how Zr-containing layers are distributed along the supercell z direction. An interval represents either a single active layer or a contiguous stack of active layers and values between 1 and 40 are permitted. This parameter therefore controls the degree of vertical segmentation and the emergence of layered motifs.

Within each interval, the density of substitution is governed by the horizontal fill factor (hff), which specifies the fraction of substituted sites per active layer. Larger hff values yield denser layers and reduce the number of layers required to satisfy the global concentration constraint. Consequently, hff mediates the trade-off between layer thickness and spatial extent of substitution.

The in-plane geometry of substituted regions is controlled by the horizontal form ratio (hfr), which sets the aspect ratio of a continuous Zr-rich patch within each active layer. Values range from squarelike domains to maximally elongated rectangular regions, enabling interpolation between isotropic and anisotropic substitution patterns.

Finally, the vertical superposition ratio (vsr) introduces lateral translation between successive intervals. This parameter determines relative in-plane offsets between layers and thereby enables shifted stacking motifs under periodic boundary conditions, producing structures ranging from aligned nanolayers to staggered lamellae.

To examine deviations from these sharply defined motifs, we additionally introduce an optional blurring parameter (rnd). The parameter progressively redistributes Zr substitutions away from their original motif positions while preserving the total Zr concentration. Thus $rnd = 0$ represents the unmodified parent structure, whereas increasing rnd produces progressively more diffuse or disordered variants. The blurring parameter is not used as an input to the surrogate model, but is applied separately in selected EH–MD calculations discussed in Sec. V C.

Representative distributions generated by this parametrization are shown in Fig. 1. Panels (a)–(f) illustrate layered configurations obtained through controlled variation of individual parameters, including interval count, layer thickness, lateral extent, and interlayer shifts. Additional examples demonstrate lamellar, rodlike, and dotlike motifs, highlighting that these recognizable structures represent limiting cases within a continuous design space rather than isolated constructions.

Overall, the five primary descriptors ($zcon$, $intv$, hff , hfr , and vsr) provide a compact and interpretable representation of the motif space suitable for systematic sampling and surrogate modeling. The optional blurring operation provides a density-preserving baseline for assessing the sensitivity of the identified motif-property relationships to substitutional disorder, but is not intended to reproduce a specific synthesis or interdiffusion mechanism. Additional algorithmic details are provided in the Supplemental Material [45].

C. Key functional quantities

We now briefly introduce the key functional quantities that will be discussed throughout the following sections. Several descriptors can be extracted directly from the P–E loops, including the maximum polarization $P_{\max}$, the remanent polarization P_{rem} , and the coercive field E_c . These quantities are illustrated in Fig. 2 for clarity.

We are further interested in the energy-storage characteristics of the material, in particular the recoverable energy density W_{rec} and the energy loss W_{loss} , which are standard figures of merit for dielectric capacitors [46]. The recoverable energy density is computed according to Eq. (1) and corresponds to the green-shaded area in Fig. 2. Here, E_{dis} denotes the discharge field and P the polarization. The losses are calculated using Eq. (2), which requires both the charging curve E_{ch} and the discharging curve E_{dis} ; the enclosed area between these curves, as illustrated in Fig. 2, represents W_{loss} :

$$W_{\text{rec}} = \int_{P_r}^{P_{\max}} E_{\text{dis}}(P) dP, \quad (1)$$

$$W_{\text{loss}} = \int_0^{P_{\max}} E_{\text{ch}}(P) dP - \int_{P_r}^{P_{\max}} E_{\text{dis}}(P) dP. \quad (2)$$

From these equations, it becomes apparent that high energy losses are inherently associated with high remanent

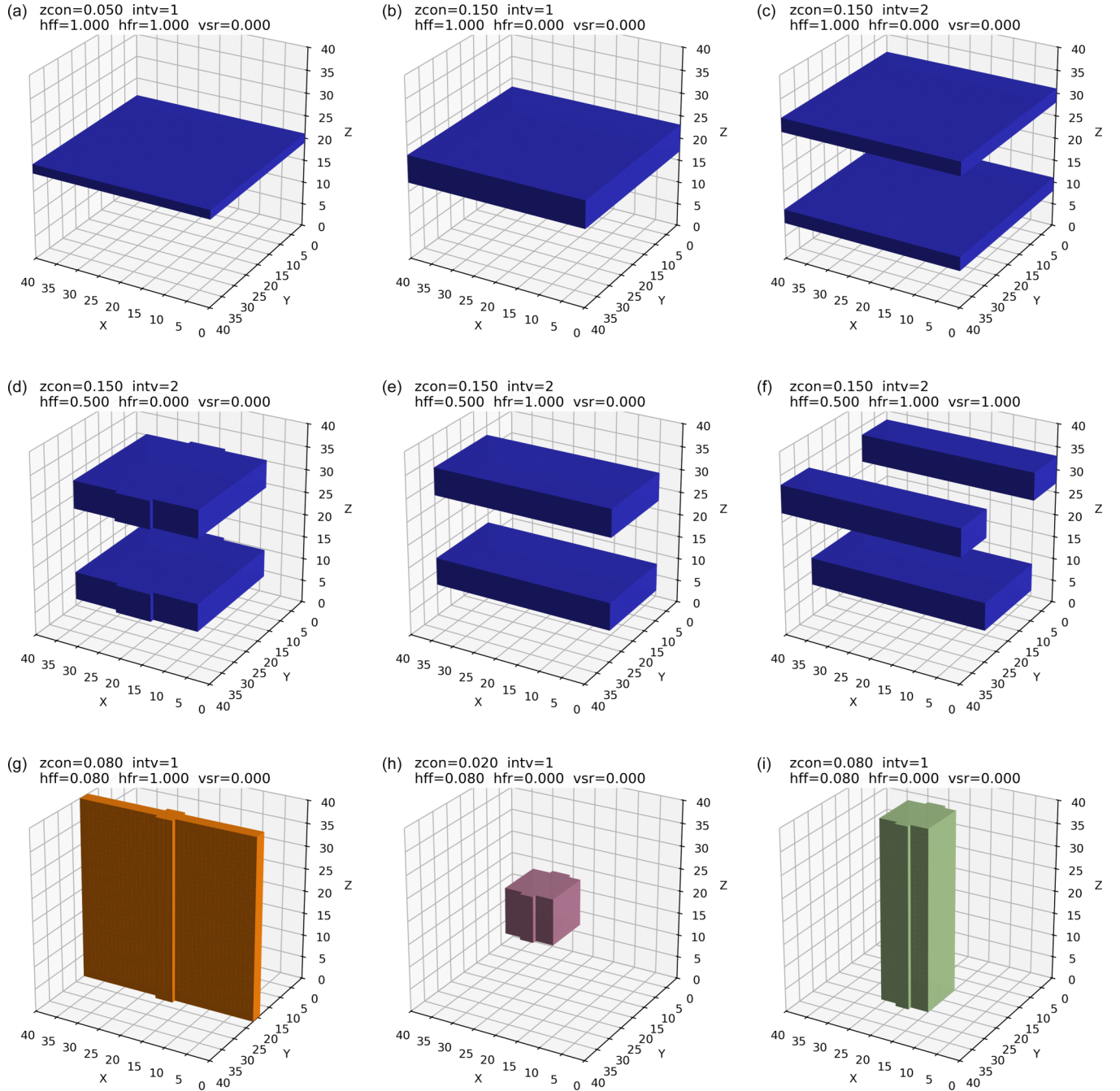


FIG. 1. Examples of supercell configurations generated by the parameter-based modeling approach. Solid regions indicate cells where Ti is substituted by Zr. Panels (a)–(f) show a sequence of configurations obtained by varying only a single parameter between realizations. Panels (g)–(i) illustrate representative structures such as a nanolamella, a nanodot, and a nanorod, all produced using the same parametrized scheme.

polarization and large coercive fields. Consequently, when the remanent polarization and coercive field are large, the recoverable energy density W_{rec} is reduced, as a larger portion of the stored energy is dissipated.

In addition to dielectric properties, we evaluate the electromechanical performance via the bipolar strain loops ($\eta_3 - E$ curves), as shown in Fig. 2(b). While the absolute maximum strain η_{max} describes the peak deformation at maximum field, practical actuator applications under cyclic loading are governed by the reversible electromechanical response. We therefore introduce the usable dynamic strain range, defined as

$\Delta\eta_3 = \eta_{\text{max}} - \eta_{\text{rem}}$, where η_{rem} represents the remanent strain remaining at zero applied field after discharging. This subtraction effectively decouples the functional cyclic response from the static baseline strain shift induced by lattice expansion via Zr substitution.

It is important to note that EH–MD simulations of this class are known to overestimate the electric-field scale by roughly one order of magnitude [15]. The field-dependent quantities reported here should therefore be interpreted primarily in comparative rather than absolute terms. In the present study, the main emphasis is on relative trends, rankings, and

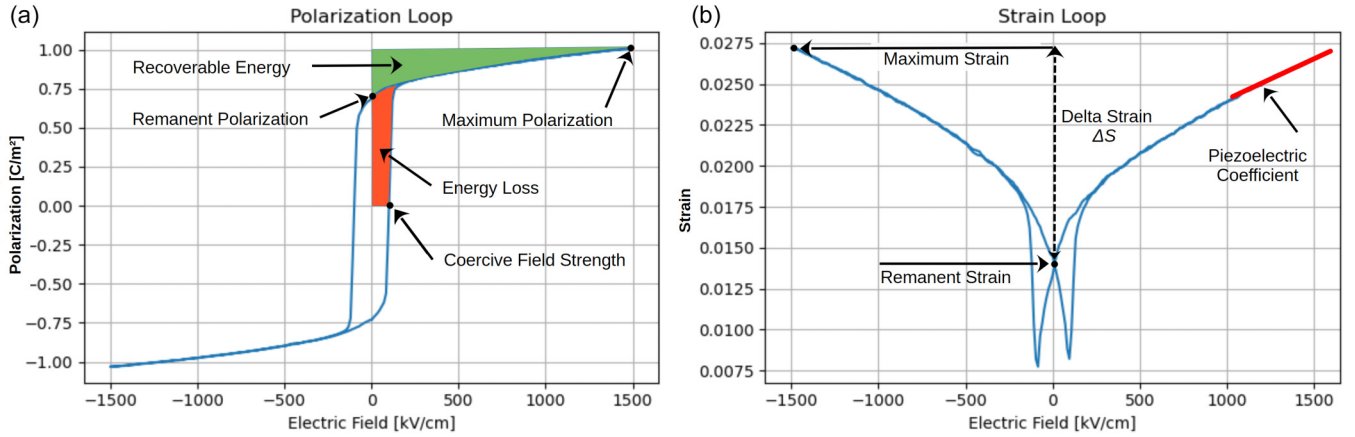


FIG. 2. Schematic illustration of the key functional quantities extracted from the simulated hysteresis loops. (a) Polarization loop ($P - E$) defining the maximum polarization $P_{\max}$, remanent polarization P_{rem} , and coercive field strength E_c . The shaded areas represent the recoverable energy density (W_{rec} , green) and the energy loss (W_{loss} , red). (b) Corresponding bipolar strain loop ($\eta_3 - E$) detailing the maximum absolute strain, the remanent strain at zero field, and the resulting usable dynamic strain range ($\Delta\eta_3$ or Delta Strain ΔS). The red solid line indicates the high-field linear fit used to evaluate the effective large-signal longitudinal piezoelectric coefficient (d_{33}).

motif-dependent differences across configurations evaluated under a common simulation protocol. Quantitatively accurate comparison to experiment would require explicit calibration of the field scale.

III. COMPUTATIONAL EXPERIMENTS AND DATA SET

Having defined the EH framework and the parametrized Zr-distribution model in Sec. II, we now describe how these tools were combined to generate the data set that underpins all subsequent analysis and machine-learning development. This section details how the structural parameter space was sampled, how EH-MD simulations were performed to obtain P-E and S-E responses, and how the resulting hysteresis loops were distilled into key functional quantities. Together, these computational experiments provide the quantitative foundation for training and validating the conditional autoencoder introduced in the following section.

Supercell sampling and MD simulations

Using the parametrized distribution model introduced in Sec. II B, we generated a data set of 2680 Zr-substituted BT supercells and evaluated their electromechanical response by EH-MD under the common protocol described in Sec. II A. Data set construction proceeded in three stages. First, 100 configurations were selected from the boundaries of the descriptor space to probe the range of accessible responses. Second, 1000 additional configurations were sampled from the initial discrete grid of 7000 parameter combinations. Third, the data set was expanded to 2680 configurations by generating off-grid samples that interpolate between the discrete descriptor values, thereby increasing coverage of the interior of the parameter space and reducing the bias associated with a purely grid-based design. Selected parent configurations were additionally evaluated at several blurring ratios using direct EH-MD simulations. These calculations were not used for surrogate training and are discussed separately in Sec. V C to

assess the influence of partial dopant disorder on the identified motif-property relationships.

Figure 3 provides an overview of how different Zr distributions affect these hysteresis behaviors. Figures 3(a) and 3(b) show loops for a low Zr concentration of 1%. The color coding indicates the number of intervals ($intv$). Even at 1% Zr, noticeable variations appear, primarily in the coercive field. The maximum polarization remains nearly constant, which can be explained by the fact that Zr-substituted unit cells embedded in BT behave nearly nonpolar, preferring centrosymmetric positions [12,13]. Increasing Zr content therefore reduces the maximum polarization, although at 1% this effect is still minimal. The strain-field loops also show small but distinct differences near the coercive field.

Figures 3(c) and 3(d) illustrate results for 5% Zr. At this concentration, the impact of Zr arrangement becomes significantly more pronounced. Some configurations exhibit well-defined ferroelectric loops, while others show much flatter, weakly hysteretic responses reminiscent of relaxor-like behavior under the present simulation protocol. The maximum polarization remains relatively similar across configurations, as 5% Zr still leads to only moderate suppression. The strain loops again display a wide range of responses.

Figures 3(e) and 3(f) present examples for 20% Zr, where the variability becomes even more substantial. Differences appear not only in loop shape but also in maximum polarization, coercive fields, and the overall electromechanical response. The electromechanical response exhibits a markedly different behavior, which can be attributed to two primary factors. First, Zr substitution in BT induces a lattice expansion even in the absence of an electric field, leading to an increased baseline strain. Second, Zr can simultaneously suppress the ferroelectric and piezoelectric response. As a result, the strain under applied field may show only minimal variation or, conversely, a substantial change, depending on whether the Zr-induced change allows or hinders further field-driven response.

Across the sampled configurations, the spread in coercive field, maximum polarization, and field-induced strain response increases strongly with Zr concentration,

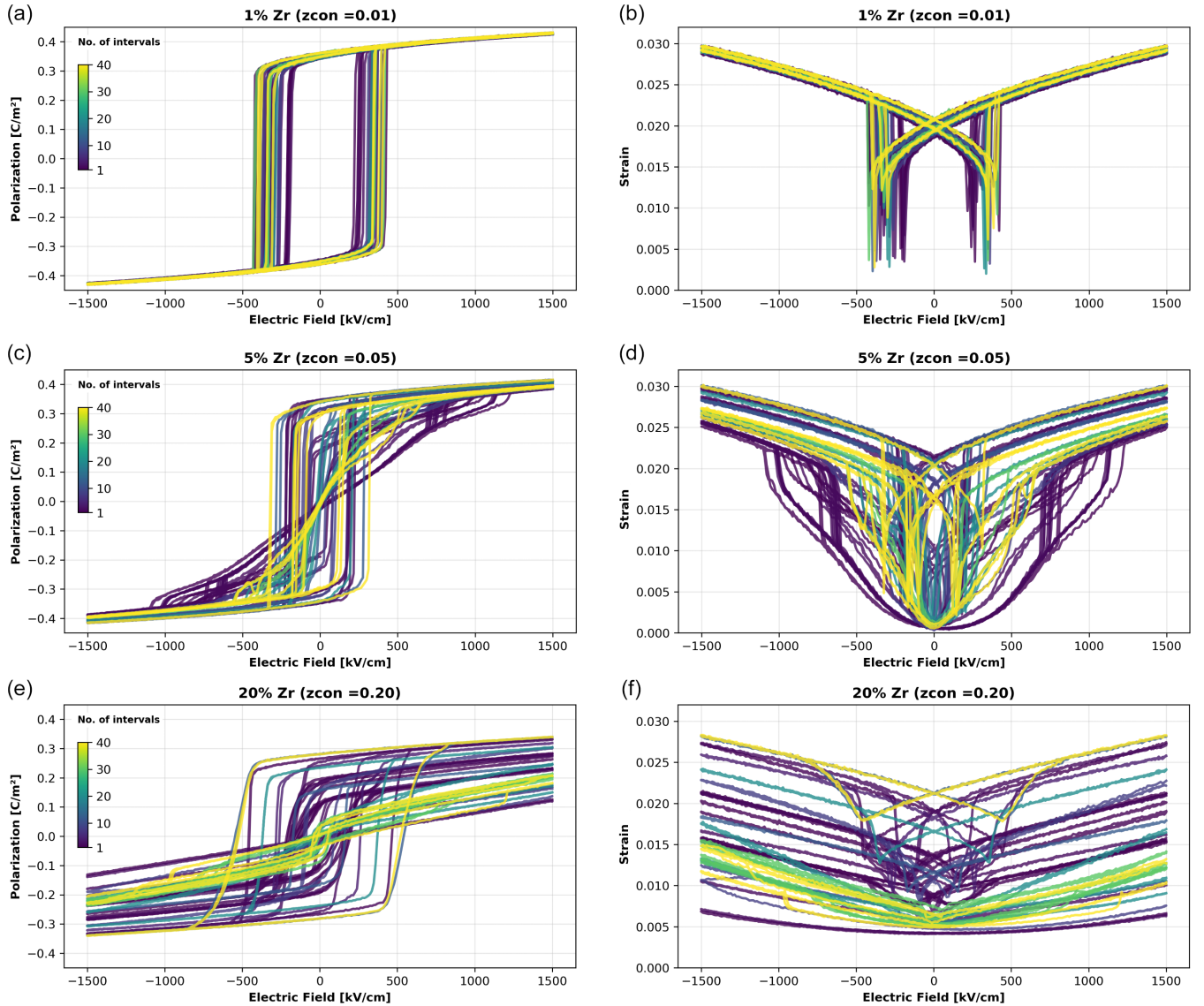


FIG. 3. Polarization and strain-field loops for supercells with varying Zr distributions. Panels (a), (b): 1% Zr, showing minor variations mainly in coercive field; maximum polarization remains nearly constant. Panels (c), (d): 5% Zr, with more pronounced differences—some loops are ferroelectric, others flatter, approaching relaxorlike behavior. Panels (e), (f): 20% Zr, exhibiting substantial variability in loop shape, maximum polarization, coercive fields, and overall electromechanical response. The parametrized distribution approach captures a wide range of behavior across Zr concentrations.

confirming that distribution effects become more pronounced as the accessible motif space broadens.

Overall, these results demonstrate that even small concentrations of Zr can produce significant variations in dielectric and electromechanical behavior when the Zr ions are arranged differently. At higher concentrations, these effects become increasingly strong due to the greater structural degrees of freedom available. The final data set therefore spans both descriptor-space extremes and interpolating interior configurations, providing a more suitable basis for surrogate training than a purely discrete grid.

IV. MACHINE LEARNING (ML) MODEL

As outlined in the Introduction, our goal is to develop a methodology that supports systematic exploration

of dopant distributions in substituted systems for efficient materials design. While EH-MD is far more efficient than direct density-functional theory (DFT) and enables large-scale finite-temperature simulations, screening a large design space of dopant distributions with EH-MD alone still becomes computationally expensive. The parametrized generation of Zr distributions together with EH-MD thus provides an ideal foundation for applying machine-learning (ML) methods [22,23] to accelerate exploration and to identify promising regions of the design space. With suitable ML architectures, full response predictions can be obtained at a small fraction of the computational cost of MD, enabling high-throughput studies and rapid screening. All machine learning models were implemented using the PyTorch deep learning framework [47].

A. General aspects

Each Zr distribution is defined by five structural descriptors: Zr concentration ($zcon$), number of intervals ($intv$), horizontal fill factor (hff), horizontal form ratio (hfr), and vertical superposition ratio (vsr). Within the adopted construction scheme, these parameters determine the spatial arrangement of Zr within the $40 \times 40 \times 40$ supercell. For every configuration, EH–MD simulations produce two response sequences: the polarization–electric field (P–E) loop and the strain–electric field (S–E) loop. From these sequences, functional descriptors such as recoverable energy density and piezoelectric response are derived. The data set therefore consists of five structural inputs, two response sequences per configuration, and associated scalar descriptors.

From a materials-design perspective, the primary targets are scalar performance descriptors rather than the response sequences themselves. A direct regression from structural descriptors to these quantities is therefore a natural baseline and in the present descriptor space it already captures much of the underlying structure–property relationship (see Supplemental Material [45]). However, direct scalar prediction does not provide access to the full hysteresis response and treats the derived quantities as independent outputs. We therefore adopt a loop-level surrogate strategy in which the model reconstructs the full P–E and S–E response sequences from the structural descriptors and derives the functional quantities only afterwards. This preserves the physical coupling embedded in the hysteresis behavior, enforces internal consistency between derived metrics, and allows additional descriptors to be evaluated without retraining. The resulting model is therefore not only predictive, but also more flexible as a framework for exploring the structural design space.

B. Sequence representation and autoencoder design

To model electromechanical response, the P–E and S–E loops are treated as primary prediction targets. Autoencoders provide an effective framework for representing structured high-dimensional signals by encoding them into a compact latent representation and reconstructing them with minimal information loss [48,49]. In this context, the latent representation captures essential switching behavior, curvature, and coupling characteristics of the response sequences.

The encoder processes the sequences using one-dimensional convolutional layers followed by dense transformations. Convolutional operations capture local correlations along the field axis and efficiently represent switching features and loop curvature [50]. The decoder mirrors this structure through dense expansion and transposed convolutions to recover the full sequences.

P–E and S–E loops are modeled jointly within a shared architecture. Since both responses originate from the same atomistic simulations, their coupled treatment encourages the learned representation to capture shared electromechanical structure rather than independent signal features. This formulation ensures that structural information directly influences sequence generation, enabling stable prediction for previously unsimulated configurations within the sampled descriptor space while maintaining a compact and interpretable representation. Additional details on architectural exploration,

conditioning strategies, and training and optimization procedures are provided in the Supplemental Material [45].

C. Conditional autoencoder design

To enable prediction of response sequences from structural descriptors, the autoencoder is extended to a conditional formulation in which reconstruction is guided by the distribution model parameters ($zcon$, $intv$, hff , hfr , vsr) [51]. These descriptors are processed through a dedicated encoder branch and embedded into the latent representation during decoding, allowing the network to learn a unified structure–response mapping.

Unlike probabilistic generative formulations based on stochastic latent variables, such as variational autoencoders and their conditional extensions [52], the present deterministic conditional autoencoder targets reconstruction of configuration-specific physical responses, aligning directly with the objective of predicting unique hysteresis behavior.

The final architecture integrates sequence and conditional pathways within a single optimization framework as illustrated in Fig. 4. One encoder compresses the response sequences, while the second processes structural descriptors. Their latent representations are aligned through an auxiliary loss penalizing discrepancies between the two latent embeddings, encouraging consistency without overloading either pathway.

During decoding, conditional features are concatenated with latent sequence features and propagated through a shared decoder that produces both P–E and S–E outputs. Shared decoder weights enforce coherent reconstruction of the coupled responses and reduce the risk of divergence between predicted sequences. At inference time, only the conditional pathway is used: structural descriptors are mapped to the latent representation and decoded into predicted P–E and S–E loops.

This integrated formulation ensures that structural information directly influences sequence generation, enabling stable prediction for previously unsimulated configurations within the sampled descriptor space while maintaining a compact and interpretable latent representation. Further architectural details, ablation experiments, and alternative design variants explored during development are documented in the Supplemental Material [45].

D. Training and performance evaluation

Training of the conditional autoencoder was carried out iteratively as the data set expanded. An initial set of approximately 1000 simulated P–E and S–E loops was used to establish the modeling workflow and refine the architecture. As additional configurations were generated through extended sampling of the structural parameter space, the data set was progressively enlarged and the model retrained. The final network was trained on 2680 loop pairs, ensuring that the learned representation captured the diversity of the explored configuration space. Given the moderate data set size relative to network capacity, model complexity was deliberately constrained through latent compression and weight sharing. These architectural choices act as implicit regularization

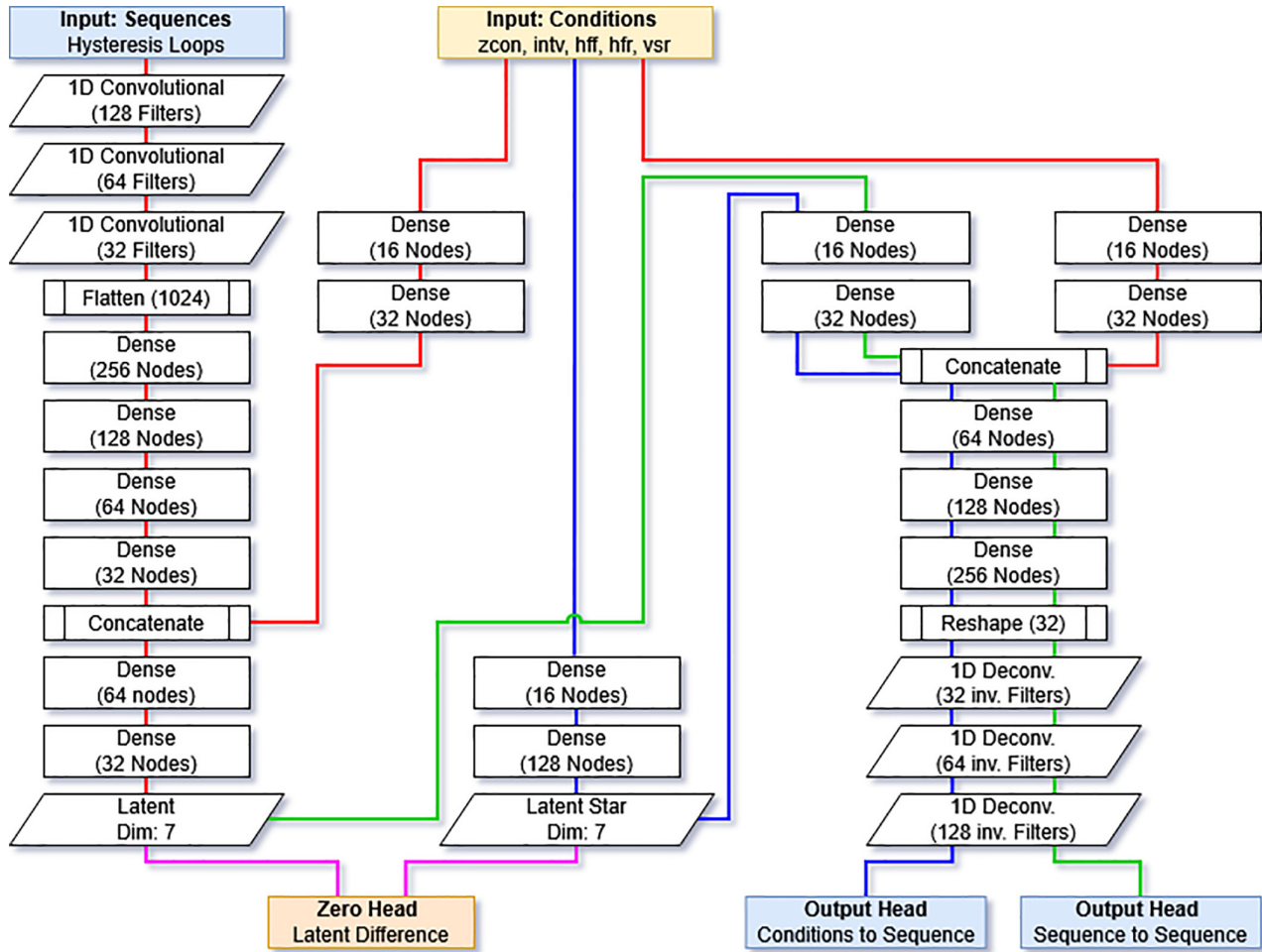


FIG. 4. Schematic of the conditional autoencoder used to simultaneously predict P-E and S-E loops from structural parameters. The model has two inputs: the sequences themselves and the conditional data, corresponding to the supercell distribution parameters. The architecture features three output heads: two for reconstructing the sequences (sharing decoder weights) and one latent difference output head, a fixed-zero output that enforces alignment between the main latent space (*Latent*) and the auxiliary latent space (*Latent Star*).

mechanisms, limiting overfitting while preserving sufficient expressive power for accurate loop reconstruction.

Training employed a composite loss function that supervises all output branches simultaneously. The dominant contribution enforces accurate reconstruction of the P-E and S-E sequences, while auxiliary terms promote consistent encoding of the structural parameters and alignment of latent representations. Parameters were optimized using the Adam optimizer [53]. Unless stated otherwise, model performance was evaluated using random training/validation/test partitions of the available data set.

Overall convergence behavior and prediction accuracy indicate stable training dynamics and reliable reconstruction across the explored parameter space. Representative loop reconstructions (Fig. 5) illustrate the model's ability to reproduce strongly ferroelectric, weakly hysteretic, and nearly linear responses across distinct regimes of loop morphology, supporting its use as a surrogate for large-scale screening of structural configurations. Further details on architectural evolution, loss-function variants, and extended performance evaluation are provided in the Supplemental Material [45].

E. Key functional quantities prediction

Having established a stable and well-trained conditional autoencoder capable of accurately reproducing P-E and S-E loops, we now assess its ability to predict the derived functional quantities that ultimately define material performance. Because these descriptors—including recoverable energy density, remanent polarization, and piezoelectric response—are extracted from the reconstructed sequences rather than learned directly, their accuracy provides a stringent validation of the surrogate model. The predictive performance is summarized in Fig. 6.

Overall, the agreement between predicted and reference values is strong. Coefficients of determination exceed 0.948 for all quantities except the piezoelectric coefficient, indicating that the model reliably captures both dielectric and electromechanical response characteristics across the explored parameter space.

Quantities derived from global loop features—namely total energy density (*d*), recoverable energy density (*e*), and energy loss (*f*)—are reproduced with particularly high fidelity. Because these metrics depend on integrated loop areas and discharge pathways, they are inherently smooth functionals

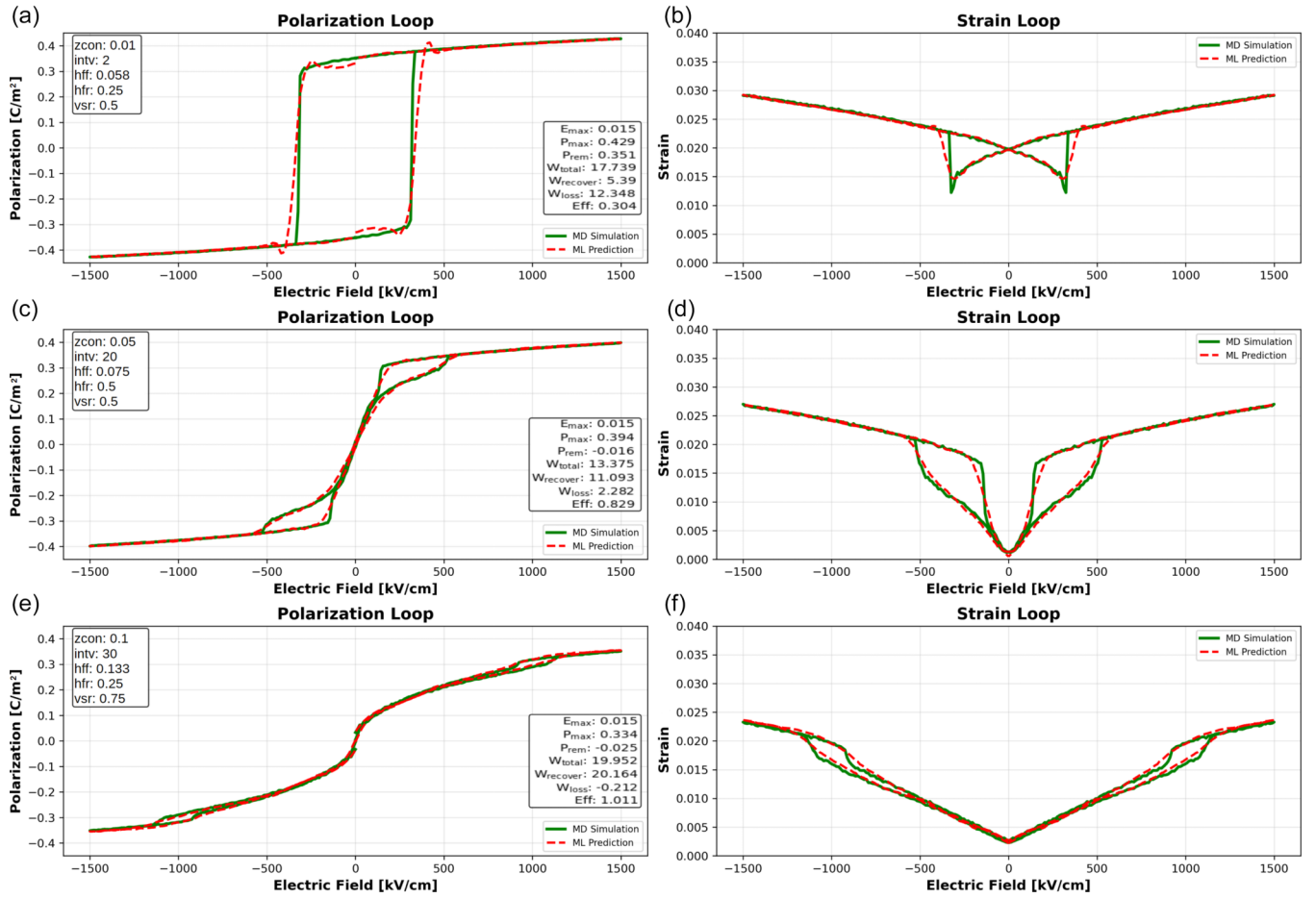


FIG. 5. Comparison of EH-MD reference loops and cAE reconstructions for representative response regimes within the sampled descriptor space. Panels (a), (b) show a strongly ferroelectric configuration with sharp switching and pronounced hysteresis in both the P-E and S-E responses. Panels (c), (d) illustrate a flatter, weakly hysteretic double-loop-like response with a paraelectriclike central region and subsidiary branch structure. Panels (e), (f) show a nearly linear, very weakly hysteretic response. Across these examples, the cAE reproduces the main loop morphology, including switching location, loop opening, curvature, and strain evolution.

of the sequences and therefore less sensitive to local reconstruction errors. Their tight correlation confirms the model's accurate reproduction of overall loop geometry.

Descriptors determined by localized loop features exhibit slightly larger scatter. Remanent polarization (b), for example, is evaluated near steep and sometimes tilted loop segments; small deviations in reconstruction therefore translate into amplified variation in the extracted value. This effect remains moderate and does not alter the observed trends.

The piezomechanical response quantities showcase the strength of the model across different strain-related features. The maximum strain $S_{\max}$ (g) and the recoverable strain ΔS (h) both demonstrate excellent predictability. The largest dispersion appears for the piezoelectric coefficient d_{33} (i). This quantity is obtained from a numerical derivative evaluated only at the high-field tail of the S-E loop, making it highly sensitive to pointwise deviations and statistical noise. Despite this intrinsic sensitivity, the dominant trends remain well preserved.

Taken together, these results indicate that the surrogate is sufficiently accurate for ranking, screening, and motif-level interpretation across the sampled descriptor space, even

though the most localized derivative-based quantities remain more sensitive to reconstruction error. To benchmark the loop-level surrogate against a simpler alternative, we additionally trained a standard multilayer perceptron (MLP) to predict the principal loop-derived scalar quantities directly from the five structural descriptors ($zcon$, $intv$, hff , hfr , vsr). The MLP performs well across all targets, indicating that the adopted descriptor set already captures much of the relevant structure-property mapping within the sampled design space. However, the cAE provides the best overall performance for most quantities, with particularly clear improvements for remanent polarization and the energy-density-related metrics, while the MLP remains competitive and can match or slightly exceed the cAE for some localized targets on individual splits. A detailed comparison is provided in the Supplemental Material [45]. This baseline comparison shows that the principal advantage of the cAE is not only predictive accuracy, but also representational flexibility: unlike the direct scalar-regression baseline, it reconstructs complete P-E and S-E loops and therefore allows additional loop-derived figures of merit to be evaluated without retraining.

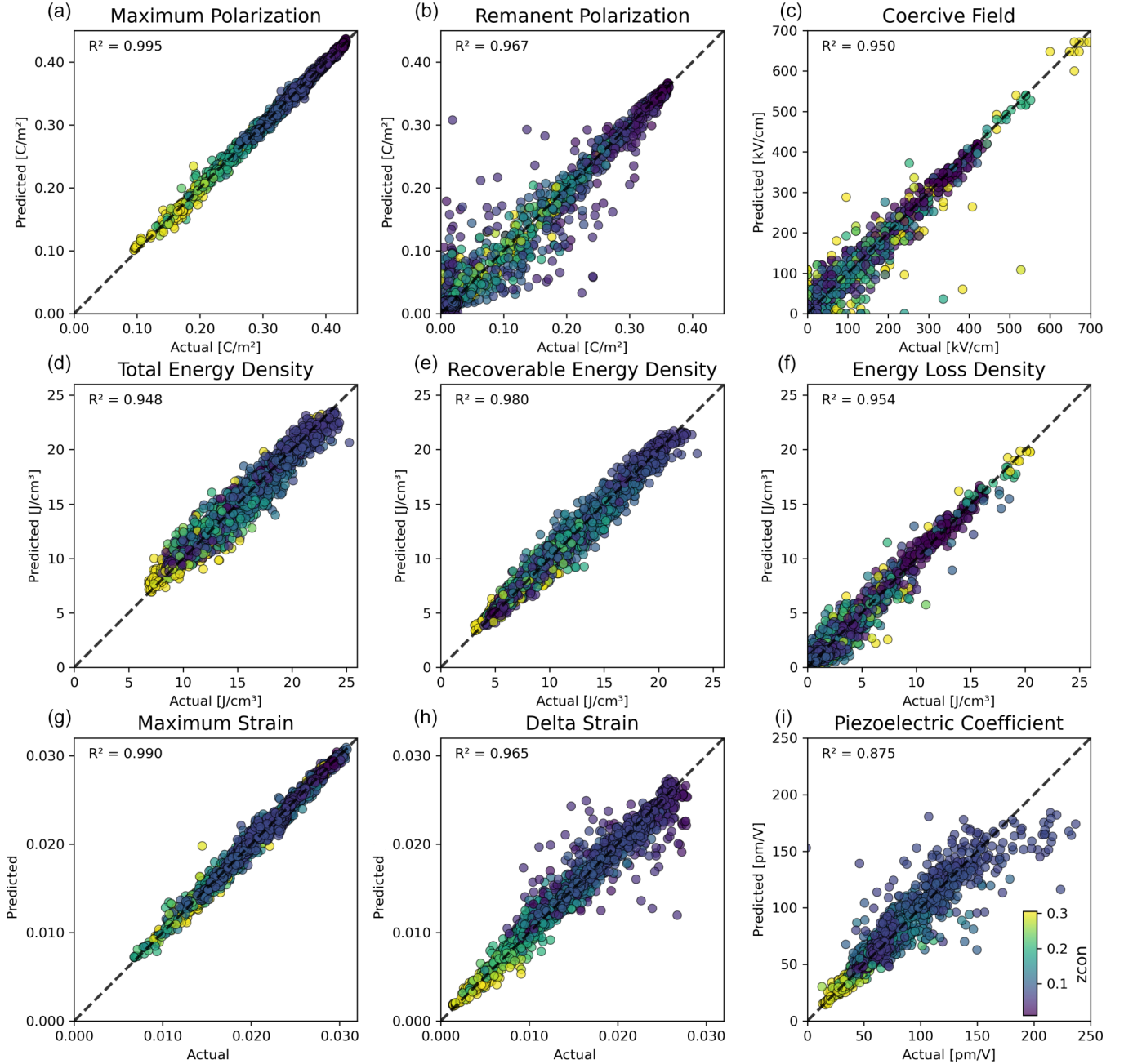


FIG. 6. Predicted versus reference values of loop-derived functional quantities for the conditional autoencoder surrogate. Panels (a)–(f) correspond to the electrical properties, including (a) maximum polarization $P_{\max}$, (b) remanent polarization P_{rem} , (c) coercive field E_c , (d) total energy density, (e) recoverable energy density W_{rec} , and (f) energy loss density W_{loss} . Panels (g)–(i) present the piezomechanical properties: (g) maximum strain $S_{\max}$, (h) recoverable strain ΔS , and (i) the high-field effective slope denoted here as the piezoelectric coefficient d_{33} . Most quantities are reproduced with high fidelity, with $R^2 > 0.948$ for all metrics except d_{33} . The larger scatter in P_{rem} and especially d_{33} reflects their stronger sensitivity to localized loop features, whereas integrated quantities such as W_{rec} and W_{loss} are less affected by small reconstruction errors. Colors indicate the Zr concentration ($zcon$).

V. DESCRIPTOR-BASED DESIGN MAPS FOR ZR DISTRIBUTIONS

Now that a surrogate model is available that can predict complete P–E and S–E loops efficiently, we use it to explore the response landscape and to construct property-selective design maps that link Zr-distribution descriptors to loop-derived figures of merit. In the following, we consider three case studies that illustrate how the database can be

screened for different functional targets. First, we analyze energy-storage performance and identify Zr distributions that combine high recoverable energy density with low hysteresis loss. Second, we examine electromechanical behavior using two contrasting objectives: configurations combining a high high-field effective slope d_{33} with large recoverable strain ΔS and mechanically quiet configurations with reduced electromechanical activity. Third, we analyze switching behavior by identifying distributions that yield easily switchable

systems and, conversely, hard-switching systems. In the present manuscript, direct EH–MD follow-up for the energy-storage target is presented as the primary validation case in the main text, while additional EH–MD validation is included in the Supplemental Material [45].

The results in this section should be interpreted mainly in terms of relative trends and rankings. As discussed above, effective-Hamiltonian simulations typically require applied electric fields that are about one order of magnitude larger than experimental values [15] and absolute energy densities are therefore expected to be higher than in experiment.

A. Surrogate-generated loop database and screening protocol

In this section we establish quantitative links between functional properties and the structural parameters of the parametrized Zr-distribution model. Each configuration is described by five distribution parameters (see Sec. II B). To identify correlations and to locate regions with favorable properties, we use the trained cAE to generate a dense prediction data set. Specifically, we sample the descriptor space within the predefined parameter bounds (Sec. II B) and evaluate the cAE for 50 000 parameter sets. This yields a database of predicted P–E and S–E loops together with their associated distribution parameters. We then compute loop-derived key quantities from each predicted loop and add them to the database, which allows systematic screening by applying percentile-based filters to the derived metrics. The percentile thresholds used below are heuristic screening choices introduced to highlight the most informative regions for each case study.

1. Energy-storage screening

We first use the database to identify configurations with favorable energy-storage properties. For this purpose, we apply a filter that selects configurations with high recoverable energy density ($W_{\text{rec}} \geq 90\text{th percentile}$) and low hysteresis loss ($W_{\text{loss}} \leq 5\text{th percentile}$). The resulting subsets are highlighted in Fig. 7. The first row of Fig. 7 relates W_{rec} to selected distribution parameters; a complete set of correlation plots is provided in the Supplemental Material [45]. In Figs. 7(a)–7(c), the red points correspond to the energy-storage filter described above.

Because mechanical actuation can be undesirable in capacitor operation, we further identify a subset of mechanically quiet candidates. Starting from the red subset, we apply a second filter requiring low recoverable strain ($\Delta S \leq 30\text{th percentile}$) and low high-field piezoelectric coefficient ($d_{33} \leq 30\text{th percentile}$). These doubly filtered configurations are highlighted in blue in Figs. 7(a)–7(c). This second filter is relevant for capacitor applications where repeated field-induced strain could promote mechanical degradation.

Two main high-performance regions are visible for the energy-storage filter. Both occur in a similar Zr concentration range (z_{con}), but they differ in other descriptors. In particular, Fig. 7(a) indicates two red clusters with comparable z_{con} but different horizontal fill factor (h_{ff}). The same separation becomes clear in Fig. 7(b) (similar z_{con} but different interval counts) and Fig. 7(c) (one cluster shows a relatively narrow range in v_{sr} , whereas the other spans a broader range).

These two regions correspond to two distinct motif families. The first family is characterized by large h_{ff} , a small number of intervals, and a broader spread in v_{sr} . In real-space terms, these configurations correspond to quasicontinuous horizontal Zr-rich nanolayers that periodically interrupt BT-rich regions, i.e., a superlatticelike modulation along the field direction. An example is shown in Fig. 8(a), with the corresponding P–E loop presented in Fig. 8(g). Such layerlike architectures are associated with high recoverable energy and low loss, plausibly because the Zr-rich layers disrupt long-range ferroelectric correlations, which narrows the P–E loop and reduces hysteresis, while the BT-rich regions retain a high maximum polarization that supports a large energy density. This general mechanism is consistent with prior theoretical and experimental work on compositional modulation and superlattices [28–32]. The present analysis identifies the corresponding region in descriptor space and quantifies the associated trends. Notably, the blue subset largely overlaps with this first red cluster, indicating that these high-energy candidates can also be mechanically quiet and therefore attractive for capacitor operation.

The second energy-storage cluster shows lower h_{ff} , a larger number of intervals, and a more concentrated v_{sr} . These configurations are different from extended nanolayers. A representative example is shown in Fig. 8(b), with the corresponding P–E loop presented in Fig. 8(g). In real space, these motifs resemble distributed rectangular nanoplatelike Zr-rich regions embedded within BT. They are likewise consistent with a disruption of long-range ferroelectric correlations and can lead to narrower loops, while the surrounding BT-rich volume maintains a high P_{max} . In this cluster, however, the mechanically quiet subset is less prominent, suggesting that the same energy-storage performance is achieved with somewhat stronger electromechanical activity.

2. Electromechanical response screening

We next examine electromechanical response using the same screening approach, not primarily to identify actuator materials, but to understand how field-induced strain activity varies across the descriptor space and how it overlaps with other functional targets, especially energy-storage behavior. To this end, we apply two complementary filters. The first selects configurations with strong electromechanical activity, defined here by high high-field effective slope d_{33} and high recoverable strain ($d_{33} \geq 85\text{th percentile}$ and $\Delta S \geq 85\text{th percentile}$). The second selects mechanically quiet configurations with very low strain response ($d_{33} \leq 2\text{nd percentile}$ and $\Delta S \leq 2\text{nd percentile}$). These subsets are shown in Figs. 7(d)–7(f). The complete correlation structure and EH–MD validation are provided in the Supplemental Material [45]. As described in Sec. II, d_{33} is extracted from the high-field region of the S–E response and therefore represents a large-signal effective slope, whereas ΔS denotes the recoverable strain, defined as the difference between the maximum and remanent strain.

For the high-electromechanical-activity subset, Figs. 7(d)–7(f) indicate two distinguishable regions. One occurs at somewhat higher z_{con} and is concentrated at low h_{ff} , while the other spans a broader range of h_{ff} . For the low- h_{ff} region (moderate z_{con} combined with large h_{fr} and small v_{sr}), we

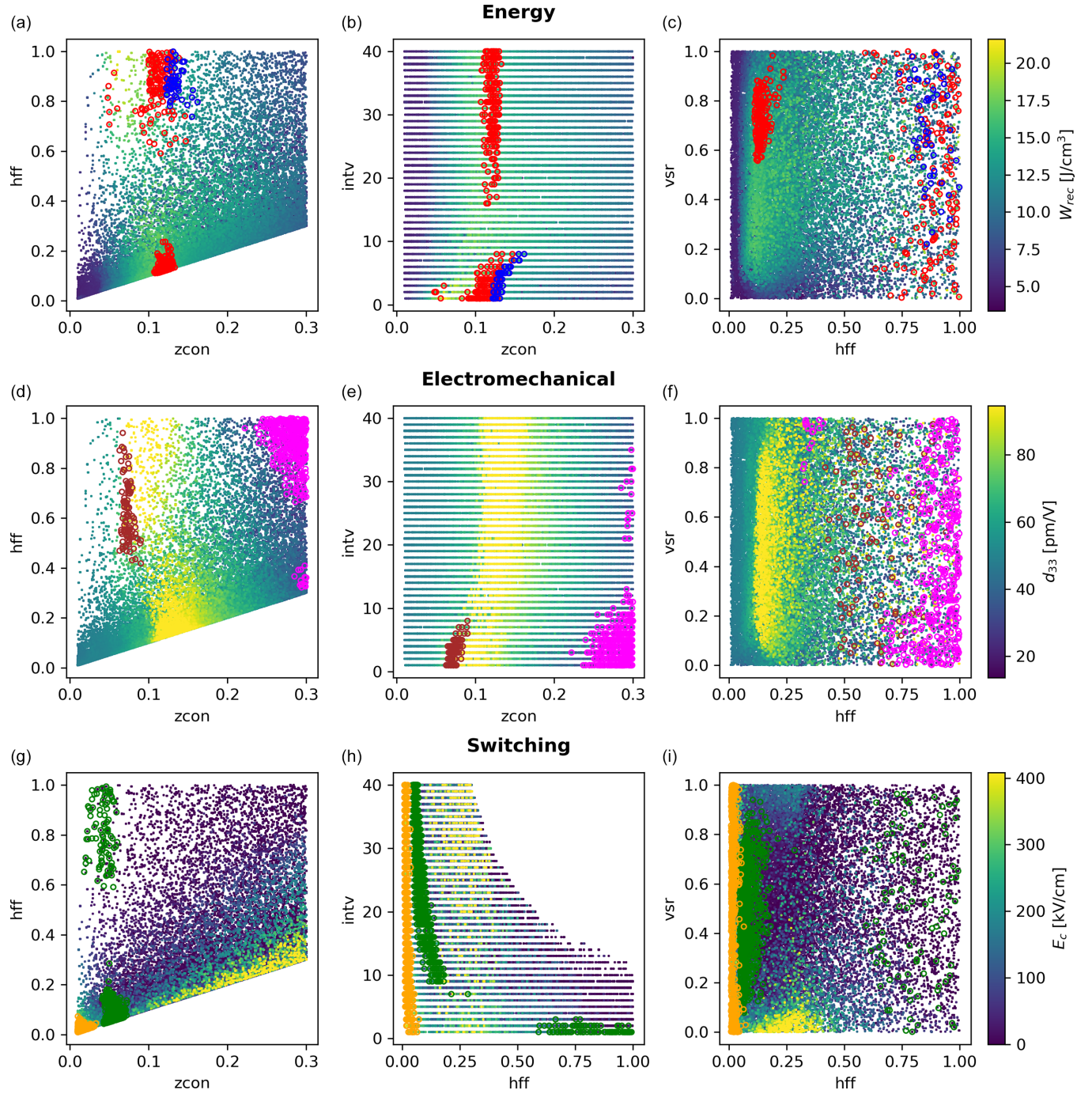


FIG. 7. Property-selective design maps derived from 50 000 cAE-predicted hysteresis loops. Each point corresponds to one predicted configuration and is plotted against selected distribution descriptors. In panels (a)–(c), red points highlight configurations with high recoverable energy density ($W_{rec} \geq 90$ th percentile) and low hysteresis loss ($W_{loss} \leq 5$ th percentile), while blue points indicate a subset of these configurations that are additionally mechanically quiet ($\Delta S \leq 30$ th percentile and $d_{33} \leq 30$ th percentile). Panels (d)–(f) show electromechanical response screening using two complementary filters: high electromechanical activity (dark red points, $d_{33} \geq 85$ th percentile and $\Delta S \geq 85$ th percentile) and mechanically quiet response (magenta points, $d_{33} \leq 2$ nd percentile and $\Delta S \leq 2$ nd percentile). Panels (g)–(i) show switching-related screening: easy switching (green points, low E_c and P_r with high P_{max}) and hard switching (orange points, high E_c and P_r). A complete set of correlation plots for all descriptor pairs is provided in the Supplemental Material [45].

observe vertically oriented lamellalike Zr-rich features that interrupt the BT host, as illustrated in Fig. 8(e). These configurations combine a high high-field effective slope d_{33} with a moderate recoverable strain ΔS . As shown by the corresponding S–E response in Fig. 8(h), the lamellalike structures

exhibit an approximately linear strain evolution over a broad field range, which may be advantageous for proportional and predictable actuation. The second region is associated with nanoplatelike motifs [Fig. 8(d)], which typically exhibit a larger ΔS but a somewhat smaller d_{33} . Their strain response is

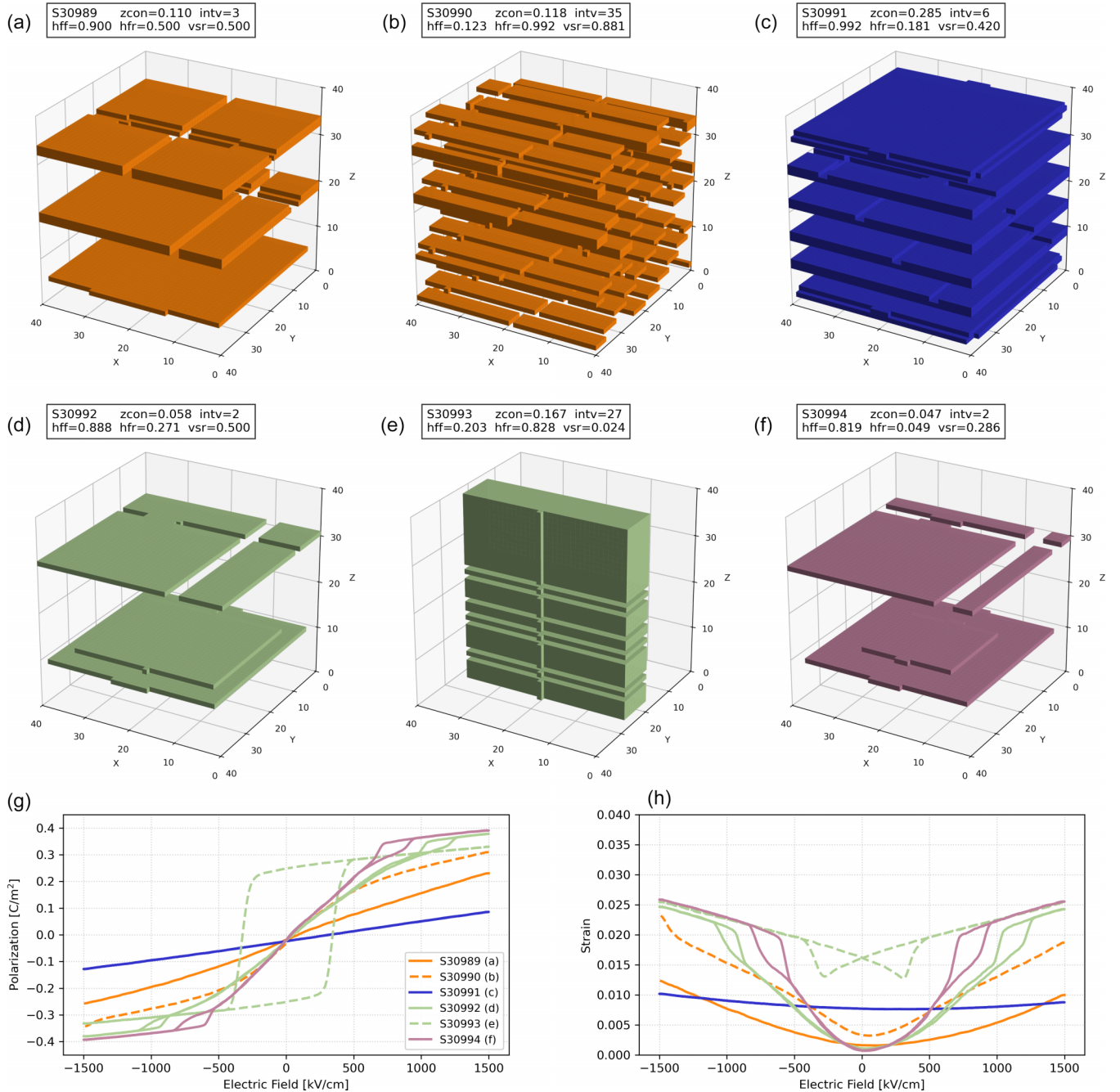


FIG. 8. Representative three-dimensional Zr-distribution motifs corresponding to the screened regions in Fig. 7, along with their predicted functional responses. Panels (a) and (b) illustrate two high energy-storage motif families selected by the W_{rec}/W_{loss} filter: (a) quasicontinuous Zr-rich nanolayers periodically interrupting BT-rich regions (superlattice-like modulation along z) and (b) distributed Zr-rich nanoplattelike inclusions embedded in the BT host. Panels (c)–(e) show representative electromechanical motifs: (c) a mechanically quiet layered configuration with small layer spacing, (d) a nanoplattelike configuration associated with large recoverable strain ΔS and a somewhat lower d_{33} , and (e) a vertical lamellalike motif combining moderate ΔS with a high high-field effective slope d_{33} . Panel (f) shows a representative configuration from the easy-switching subset, illustrating that layerlike motifs can also produce slim loops with reduced coercive field and remanent polarization while maintaining high P_{max} . All structures are shown for the same supercell size. Zr-rich regions are highlighted for clarity. Panels (g) and (h) display the corresponding cAE-predicted polarization ($P - E$) and strain ($S - E$) hysteresis loops, respectively, color coded to match the individual structural configurations (a)–(f). The loop profiles directly illustrate how tailored spatial distribution motifs dictate macroscopic ferroelectric switching, energy-storage efficiency, and electromechanical strain curves.

more strongly nonlinear, allowing a large recoverable strain to accumulate over the applied-field range. Thus the two motif families offer complementary electromechanical characteristics: lamellalike structures favor a nearly linear response with

a large effective slope, whereas nanoplattelike structures favor a larger recoverable strain.

For the mechanically quiet subset, the filters identify configurations with simultaneously small recoverable ΔS and

small high-field d_{33} . In the surrogate screening, these configurations fall predominantly into a layerlike motif family [Fig. 8(c)], but with markedly smaller spacing between adjacent Zr-rich layers than in the high-electromechanical-activity layer case shown in Fig. 8(d). Correspondingly, the associated S–E response is nearly flat over the entire applied-field range, as shown in Fig. 8(h). This highlights that “layerlike” is not sufficient as a descriptor on its own: layer spacing and thickness strongly affect whether layering promotes large strain response or suppresses it. This result is particularly relevant for the energy-storage screening, because it shows that mechanically quiet behavior can be obtained within the same motif class by tuning the geometric details of the layering, rather than requiring an entirely different type of distribution.

3. Switching behavior screening

Finally, we examine switching behavior by screening for configurations with easy versus hard switching. For easy switching, we select configurations with small coercive fields ($E_c \leq 15$ th percentile), small remanent polarizations ($P_r \leq 15$ th percentile), and simultaneously large maximum polarizations ($P_{\max} \geq 80$ th percentile). This combined filter targets loops that are slim and easy to switch while still maintaining a high polarization amplitude, which is of interest for energy storage. For hard switching, we select configurations with large coercive fields and remanent polarizations ($E_c \geq 95$ th percentile and $P_r \geq 95$ th percentile). The results are summarized in Figs. 7(g)–7(i).

For the hard-switching filter, highlighted as orange points, the selected configurations lie, as expected, close to the BT-like regime. Pure BT exhibits a comparatively hard ferroelectric loop and within the combined hard-switching criteria it appears as the most difficult-to-switch limit. If one considered only E_c in isolation, other distributions could also yield elevated coercive fields, but the combined requirement of simultaneously large E_c and large P_r selects BT-like behavior most strongly.

For the easy-switching filter, marked as green points, the selected configurations overlap substantially with the layerlike motifs identified in the energy-storage screening. In particular, quasicontinuous Zr-rich nanolayers yield slim loops with reduced coercive field and remanent polarization while maintaining high $P_{\max}$, consistent with the requirement for easy switching and high energy density. A representative example is shown in Fig. 8(f), with the corresponding P–E and S–E loops presented in Figs. 8(g) and 8(h), respectively. Differences within this family are mainly quantitative, such as layer spacing, layer thickness, and the presence of small interruptions or secondary features, which can tune loop shape without changing the dominant motif class.

Overall, this section shows that cAE-based loop prediction combined with loop-derived key quantities enables systematic screening for target behavior and direct identification of the corresponding Zr-distribution motifs. Since the results presented here are based on surrogate predictions, we next validate the predicted trends and selected candidates by follow-up EH–MD simulations for representative configurations.

B. Surrogate-guided validation and targeted sampling

To translate the surrogate-derived design rules into validated microstructures, we combined rapid screening with targeted EH–MD simulations. Starting from the design maps, we carried out a denser screening in the region that combines high recoverable energy density with low hysteresis loss. The purpose of this step is to focus on configurations where the P–E loop becomes narrow while the polarization amplitude remains high, which is required for strong capacitor performance.

The main structural signatures of this region were identified from the previous analysis of the top- W_{rec} subset (Fig. 7). High recoverable energy is associated with a narrow concentration window, quasicontinuous layering (large horizontal fill factor), and only a small number of Zr-rich intervals. Based on these signatures, we selected 300 candidate configurations and validated them by direct EH–MD simulations. For this selection, we constrained the three most influential parameters ($zcon$, $intv$, and hff), while the remaining two parameters were sampled randomly. As a result, a small number of candidates fall outside the optimal region and show higher losses. These outliers are rare and mainly reflect unfavorable combinations of the unconstrained parameters; constraining all five parameters would further reduce this effect.

Figure 9 summarizes the validation results. The figure shows the energy-storage map, i.e., W_{loss} versus W_{rec} and $P_{\max}$ versus W_{rec} , using EH–MD data only and color coded by Zr concentration ($zcon$). The newly simulated configurations are highlighted in red. The red points populate the high- W_{rec} and low-loss region much more densely than the initial sampling, confirming that the surrogate-guided selection drives the search toward improved energy-storage behavior. The $P_{\max} - W_{\text{rec}}$ projection further shows that most targeted candidates lie in the regime where high W_{rec} coincides with high $P_{\max}$, consistent with the mechanism identified above: high recoverable energy requires both a large polarization amplitude and a narrow hysteresis loop.

With this region now more densely sampled and partially validated by EH–MD, the task reduces to selecting a representative configuration that combines high W_{rec} with low loss. In our parametrized space, the best-performing candidate corresponds to a layered architecture consisting of BT nanolayers (thickness ~ 14 nm) periodically interrupted by quasicontinuous Zr-rich layers (thickness ~ 2 nm). These Zr-rich sheets are BaZrO₃-like nanolayers, such that the resulting structure is a simple superlattice along the z direction, which is also the direction of the applied electric field. Although superlattices of ferroelectric and paraelectric materials are not a new concept [28], this result illustrates the main value of the present workflow: rather than assuming a layered architecture *a priori*, the approach identifies the relevant distribution features from a broader descriptor space, narrows the search to high-performing regions, and supports targeted atomistic validation.

C. Influence of substitutional disorder on the identified motif–property relationships

The primary design space considered above consists of controlled nanoscale Zr-distribution motifs with well-defined

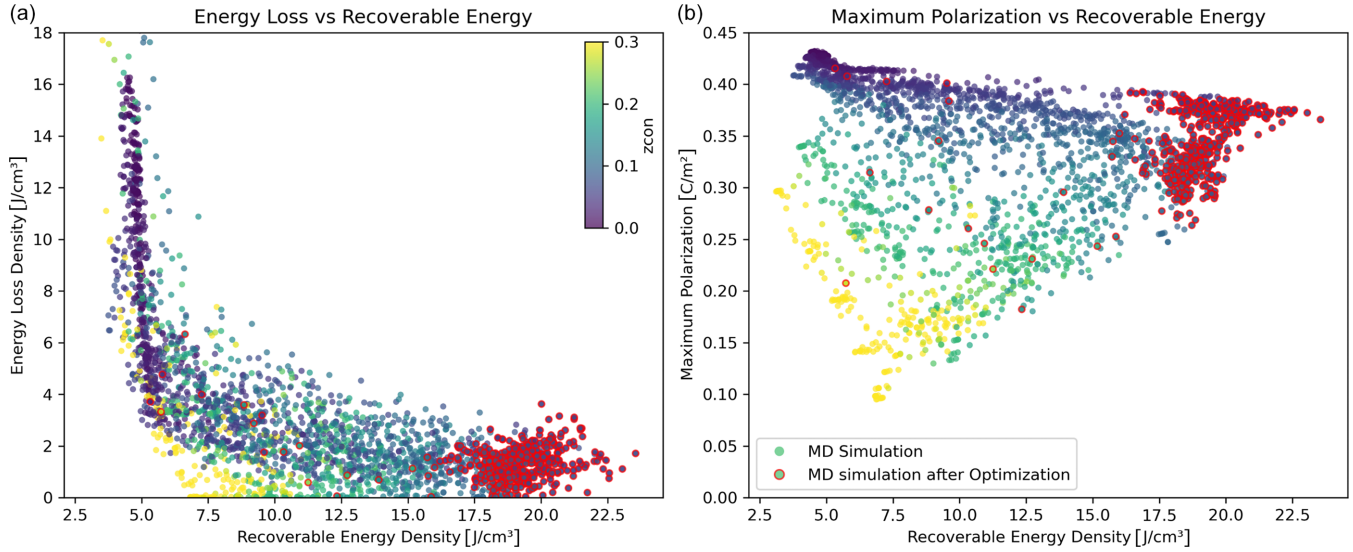


FIG. 9. Updated EH-MD simulation results (red points) targeted to the ML-identified high-performance region. (a) Energy-loss density W_{loss} versus recoverable energy density W_{rec} . (b) Maximum polarization P_{max} versus W_{rec} . The additional simulations densely populate the high- W_{rec} , low-loss regime, confirming that moderate Zr concentrations combined with quasicontinuous layered motifs and a small number of intervals yield improved energy-storage performance.

interfaces and spatial correlations. Experimentally realized substituted materials, however, may exhibit intermixing, interface roughness, and substitutional disorder. We therefore performed additional direct EH-MD simulations for selected parent structures to examine how progressive randomization of the Zr distribution affects the identified functional regimes.

The degree of disorder is controlled by the blurring parameter rnd . Starting from a parent configuration, an increasing fraction of the Zr ions defining the original motif is redistributed among randomly selected B sites while preserving the overall Zr concentration. Accordingly, $rnd = 0$ represents the sharply defined parent motif, whereas $rnd = 1$ corresponds to a fully randomized solid solution at the same composition. Figure 10 illustrates the resulting evolution for three representative motif families identified in Fig. 8: a high-energy-storage layered motif, a mechanically quiet densely layered motif, and a vertical lamellalike motif characterized by a high d_{33} and moderate ΔS . These examples illustrate three distinct motif-dependent responses to progressive blurring.

The influence of disorder is particularly pronounced for the high-energy-storage layered configuration shown in Fig. 10(a). In the unblurred structure, quasicontinuous Zr-rich layers interrupt the BT matrix perpendicular to the applied field and produce a slim P-E loop with favorable energy-storage characteristics. As rnd increases, the sharp compositional modulation is progressively weakened and the corresponding loops evolve toward a more open ferroelectric response, as shown in Fig. 10(d). Both the maximum and remanent polarization increase, while the curvature of the discharge branch becomes less favorable for recoverable energy storage. Consequently, W_{rec} decreases with increasing blurring, as shown in Fig. 10(g). The sharply layered configuration therefore substantially outperforms its randomized counterpart at the same Zr concentration. This confirms that the enhanced energy-storage response does not arise from

composition alone, but depends critically on the spatial organization and continuity of the Zr-rich layers.

A similarly clear effect is observed for the mechanically quiet layered configuration shown in Fig. 10(b). The closely spaced Zr-rich layers strongly suppress the field-induced strain response, resulting in an almost flat S-E loop for $rnd = 0$. Progressive blurring weakens this dense layered modulation and leads to an increasingly pronounced strain response, as shown in Fig. 10(e). Both the recoverable strain ΔS and the high-field effective slope d_{33} increase with rnd [Fig. 10(h)]. This behavior is consistent with the loss of the continuous compositional interruptions that constrain the electromechanical response of the BT-rich regions. The mechanically quiet state is therefore promoted by the deliberately structured layering and is gradually lost as the Zr distribution approaches the random limit.

The vertical lamellalike configuration exhibits a different sensitivity to disorder. As shown in Fig. 10(c), this motif combines a moderate recoverable strain with a comparatively high d_{33} and an approximately linear S-E response over a broad field range. Blurring produces only moderate changes in the loop shape and in the derived electromechanical quantities [Figs. 10(f) and 10(i)] and the response of the fully randomized configuration remains relatively similar to that of the ideal lamella. Unlike the horizontal layers, the vertical Zr-rich feature does not form a continuous compositional barrier perpendicular to the applied field. Connected BT-rich regions therefore remain available to accommodate the field-induced strain in both the lamellar and randomized configurations. The electromechanical response of this motif is consequently less dependent on an atomically sharp interface and more robust against substitutional disorder.

Overall, the blurring analysis demonstrates that substitutional disorder has a substantial but strongly motif-dependent influence on the functional response. Disorder progressively erodes the extreme properties associated with sharply defined

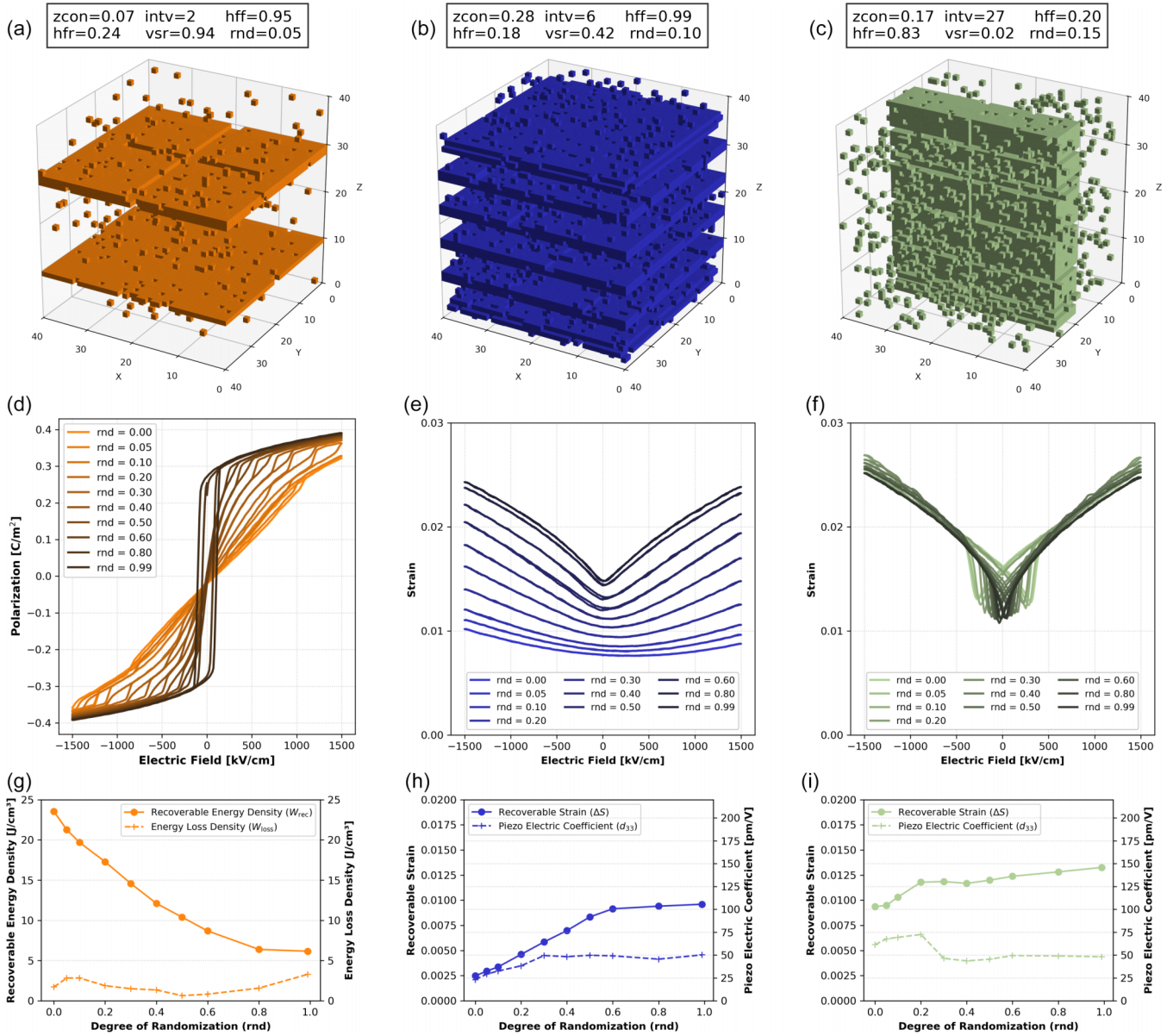


FIG. 10. Impact of progressive spatial randomization on the functional response of three representative Zr-distribution motif families corresponding to those identified in Fig. 8: a high-energy-storage layered motif (a), a mechanically quiet densely layered motif (c), and a vertical lamellalike motif (e). Panels (a)–(c) show representative three-dimensional realizations of these motifs at blurring ratios $rnd = 0.05, 0.10, 0.15$, respectively, while preserving the overall Zr concentration. Panels (d)–(f) show the evolution of the corresponding response curves over the full investigated range, $rnd = 0.00-0.99$: (d) $P-E$ loops for the high-energy-storage layered motif and (e), (f) $S-E$ loops for the mechanically quiet layered and vertical lamellalike motifs, respectively. Panels (g)–(i) summarize the associated loop-derived quantities as functions of rnd : (g) recoverable energy density W_{rec} and energy-loss density W_{loss} for the high-energy-storage motif and (h), (i) recoverable strain ΔS and high-field effective slope d_{33} for the mechanically quiet layered and vertical lamellalike motifs, respectively. Increasing rnd progressively weakens the parent structural motif and approaches the disordered solid-solution limit.

horizontal layers: it lowers the recoverable energy density of the optimized energy-storage configuration and increases the strain activity of the mechanically quiet structure. By contrast, the high- d_{33} , nearly linear response of the vertical lamella is comparatively insensitive to blurring. These results support the central conclusions of the controlled-motif screening. The spatial distribution of Zr remains a decisive design variable, while disorder primarily modifies the magnitude and sharpness of the targeted response rather than overturning the identified motif–property relationships. This two-stage

strategy enables the identification of interpretable structural design rules followed by an independent assessment of their sensitivity to substitutional disorder.

VI. SUMMARY AND CONCLUSIONS

We have presented a loop-level workflow for dopant-distribution engineering in substituted ferroelectrics by combining a controlled descriptor model for nanoscale dopant arrangements, effective-Hamiltonian molecular dy-

namics (EH–MD), and a machine-learning surrogate based on a conditional autoencoder. Using $\text{Ba}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (BZT) as a test case, we showed that dopant distribution acts as an additional design variable within the explored descriptor space: at fixed composition, different spatial arrangements of Zr can produce markedly different polarization–electric-field and strain–electric-field hysteresis responses and therefore different loop-derived figures of merit.

A central outcome of the present work is methodological. Rather than predicting a small set of scalar properties directly, the surrogate model reconstructs complete P–E and S–E loops from compact descriptors of dopant arrangement. This preserves the internal coupling between dielectric and electromechanical response and makes it possible to evaluate multiple derived quantities from a common predicted response representation. Once trained on EH–MD data, the surrogate enables rapid generation of dense response databases at a computational cost far below that of direct simulation, thereby making large-scale screening of dopant-distribution space tractable. Comparison with a direct scalar-regression MLP baseline further shows that the descriptor space is already highly informative, while the cAE remains advantageous because it provides the best overall performance across most targets and, more importantly, predicts complete response curves rather than only a predefined set of scalar outputs.

Within this framework, we constructed property-selective design maps for three representative targets: energy-storage performance, electromechanical response, and switching behavior. Across these cases, the screening results show that distinct motif families occupy different response regimes, demonstrating that nanoscale dopant arrangement can systematically reshape hysteresis behavior beyond trends set by average composition alone. For the energy-storage target, targeted EH–MD follow-up confirmed that moderate Zr concentrations combined with quasicontinuous Zr-rich layering yield slim hysteresis loops with high recoverable energy density, illustrating how surrogate-guided exploration can be translated into validated candidate structures. At the same time, the broader analysis shows that layered motifs are only one part of the accessible design space. Lamellar and nanoplate-like arrangements emerge in other functional regimes. Furthermore, systematic randomization testing showed that substitutional disorder modifies the functional response in a strongly motif-dependent manner. Progressive blurring reduces the enhanced recoverable energy density of the optimized layered configuration and weakens the suppression of electromechanical activity in the mechanically quiet layer motif, whereas the response of the vertical lamella remains comparatively less sensitive. The disorder analysis therefore supports the identified motif–property relationships while showing that the magnitude of the targeted response depends on the preservation of the underlying spatial correlations.

The present results should be interpreted within the scope of the adopted model hierarchy. First, the descriptor space is intentionally restricted to controlled motif families and their intermediates, rather than arbitrary disordered substitution patterns. Second, the surrogate is trained and validated primarily for interpolation within the sampled configuration space, so its predictions should be understood in that domain. Third, the EH–MD framework is known to overestimate the electric-field scale quantitatively, meaning that the field-dependent quantities reported here are most meaningful in comparative rather than absolute terms. Accordingly, the main value of the present study lies in identifying trends, rankings, and motif–response relationships under a common simulation protocol, rather than in making direct quantitative predictions for experiment. These limitations also define the most important directions for future work. On the structural side, while our density-preserving randomization model provided a fundamental baseline for spatial disorder, future extensions can incorporate more sophisticated, synthesis-aware blurring mechanisms—such as thermodynamic phase segregation, diffuse Gaussian interdiffusion fronts, or strain-mediated defect clustering—to directly reflect processing-induced interfacial morphologies. On the surrogate-modeling side, more stringent validation strategies, uncertainty-aware prediction, and active-learning or Bayesian-optimization loops could further strengthen the connection between screening and targeted EH–MD sampling. Finally, the same general framework should be transferable to other substituted ferroic and functional oxide systems in which compact structural descriptors can be linked to field-driven response curves. In this sense, the present study provides not only a case study for BZT, but also a general route for combining physics-based simulation and surrogate modeling to accelerate exploration of complex composition–structure–property landscapes.

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DATA AVAILABILITY

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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