

Emergent ferroelectric order in strained KTaO_3 by anharmonic and machine learned force fields methods

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Ferroelectric materials are a class of dielectrics that exhibit spontaneous polarization which can be reversed under an external electric field. The emergence of ferroelectric order in incipient ferroelectrics is a topic of considerable interest from both fundamental and applied perspectives. Despite evidence from first-principles calculations that strain triggers ferroelectricity in KTaO_3 , conventional methods cannot reliably characterize the soft-mode dynamics that govern its ferroelectric behavior. In this study, we investigate the impact of in-plane uniaxial and biaxial strain, ranging from 0 to 1%, on pristine KTaO_3 to explore its potential for ferroelectricity induction via inversion symmetry breaking. By integrating density-functional theory calculations with the stochastic self-consistent harmonic approximation assisted by on-the-fly machine learned force fields, we obtain accurate structural information and dynamical properties under varying strain conditions while incorporating higher-order anharmonic effects. Employing the Berry-phase method, we obtained the ferroelectric polarization of the strained structures over the entire temperature range up to 300 K. Our findings provide valuable insights into the role of strain in stabilizing ferroelectricity in KTaO_3 , offering guidance for future experimental and theoretical studies on strain-engineered ferroelectric materials.

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I. INTRODUCTION

Potassium tantalate (KTaO_3) is a prototypical quantum paraelectric, in which strong quantum fluctuations suppress the onset of ferroelectric order even at the lowest temperatures [1,2]. It exhibits a temperature-dependent transverse optical soft phonon mode whose frequency decreases upon cooling but never vanishes [3,4], leading to a pronounced deviation of the inverse dielectric constant from the classical Curie-Weiss law [5,6]. This soft-mode saturation and associated dielectric anomaly place KTaO_3 in close proximity to the ferroelectric quantum critical point (QCP) [6,7] in Fig. 1. Such proximity implies that small external perturbations—such as strain [8,9], hydrostatic pressure [10–12], or isotopic substitution [13–15]—can drive the system across the QCP and induce long-range ferroelectricity, a hallmark of so-called incipient ferroelectrics.

Over the past decades, numerous experiments have confirmed that the dielectric properties of KTaO_3 are highly tunable [8,16–18], yet the microscopic mechanisms governing the interplay between external perturbations and quantum

fluctuations remain incompletely understood [19–26]. From a theoretical standpoint, modeling quantum paraelectric from first principles is challenging (Fig. 1). Conventional density-functional theory (DFT), formulated at zero temperature, cannot capture quantum or thermal fluctuations and thus fails to describe the suppression of ferroelectricity in materials like KTaO_3 [27]. Path-integral Monte Carlo simulations with effective Hamiltonian [28–30] and low-dimensional lattice nuclear Schrödinger equation [31,32] approaches have provided insights, but they often neglect mode coupling and are restricted to simplified models. In a related direction, theoretical model work on double-well-type nonlinear electron-phonon interactions has further highlighted the relevance of such energy landscapes to quantum paraelectric [33]. More recently, the stochastic self-consistent harmonic approximation (SSCHA) [34–37] has emerged as a powerful framework to treat anharmonic phonons nonperturbatively while fully including nuclear quantum and thermal effects, offering a route to qualitatively describe quantum criticality in incipient ferroelectrics [38–42]. However, the requirement of large supercells for accurate phonon research makes the computational cost prohibitively high. Machine learning algorithms have been demonstrated to effectively accelerate such simulations while maintaining accuracy, thus providing a promising strategy to overcome this technical limitation [43–45].

Despite these advances, a key open question remains: how do quantum fluctuations compete with strain-induced ferroelectric order in KTaO_3 ? Understanding this competition is essential to determine whether long-range ferroelectricity can be sustained in strained KTaO_3 and how its temperature

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dependence evolves near the quantum critical regime. To address this question, we employ the SSCHA method [34–37] combined with on-the-fly machine learned force fields (MLFF) scheme [46–48] to investigate the competition between ferroelectric order and quantum fluctuation in strained KTaO_3 with first-principles accuracy.

II. METHODOLOGY

The computational work in this study was conducted by combining the Vienna *Ab initio* Simulation Package (VASP) [49,50] with the SSCHA method [34–37]. DFT calculations were carried out at the meta-generalized-gradient approximation level. Specifically, the SCAN functional [51] was employed alongside projector augmented wave potentials [52]. A plane-wave energy cutoff of 800 eV was used to ensure the accuracy and convergence of the results. The Berry-phase theory was conducted to compute the electrical polarization [53–56]. The pristine KTaO_3 is modeled using the experimental lattice of 3.9842 Å [38,57,58], and strain is applied on this basis. To prevent the emergence of additional soft ferroelectric modes, only the atomic positions are relaxed, while all lattice constants are kept fixed.

To fully capture quantum and anharmonic effects, we employ SSCHA, a variational method that determines the free energy by optimizing an auxiliary harmonic density matrix. This approach naturally incorporates both thermal and quantum fluctuations beyond the harmonic approximation. Within the SSCHA framework, stochastic ensembles of ionic configurations are generated according to Gaussian distribution centered around the equilibrium positions. These ensembles are used to evaluate the free energy and its gradients, which are iteratively minimized using force-constant gradient descent scheme. The required stochastic averages are computed via Monte Carlo sampling in a suitably chosen $4 \times 4 \times 4$ supercell for KTaO_3 . To make these calculations computationally feasible, we integrate an on-the-fly machine learned force field (MLFF) [46–48].

The MLFF, trained on the fly from DFT molecular dynamics with Bayesian error estimation, accelerates the stochastic sampling in SSCHA by several orders of magnitude while retaining full fidelity to the *ab initio* potential-energy surface. Based on previous study [38], we had already developed a machine learned force field adapted to pristine KTaO_3 , which performed well and significantly accelerated SSCHA calculations. Therefore, we retrain the model using dataset from a $4 \times 4 \times 4$ strained supercell and a $2 \times 2 \times 2$ Gamma-centered k -point mesh. MD simulations were performed at increasing temperatures up to 300 K, sampling 10 000 steps at each temperature with a time step of 2 fs. The final structure at each temperature served as the starting configuration for the next higher temperature. A Langevin thermostat was employed [59]. For the descriptors representing local atomic environments in the MLFF, the cutoff radius for the two- and three-body atomic density distributions was set to 8 Å, and the Gaussian broadening to 0.3 Å. Equal weights were assigned to both descriptors. This updated MLFF was optimized to accurately describe both uniaxially and biaxially strained KTaO_3 structures.

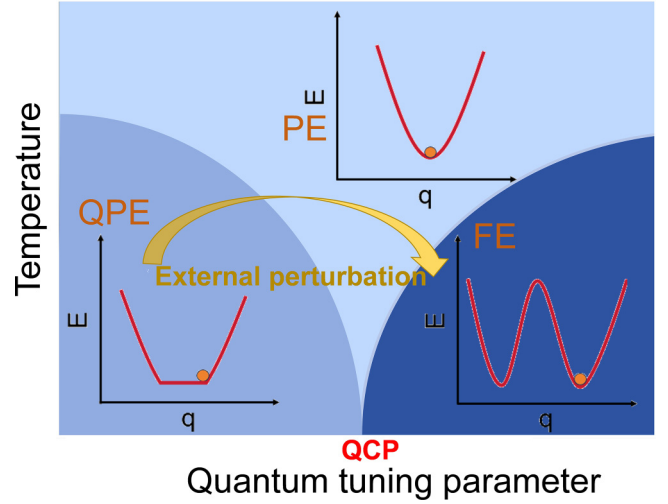


FIG. 1. Qualitative phase diagram of the crossover among the ferroelectric (FE), paraelectric (PE), and quantum paraelectric (QPE) phases as the function of quantum tuning parameter and temperature.

Overall, our workflow combines a VASP-SSCHA interface with the on-the-fly MLFF to compute energies and forces for ensembles generated within the SSCHA framework. The structure of this workflow is illustrated schematically in Fig. 2. This integrated framework enables us to compute the temperature-dependent ferroelectric soft-mode frequency $\omega_{\text{FE}}(T)$, anharmonic phonon spectra, and free-energy landscape across a range of strain conditions, thereby mapping the critical strain and temperature thresholds for stabilizing the FE phase. Details of the computational methodology are provided in Supplemental Material [60].

III. RESULTS

A. Structural properties and symmetry breaking in strained KTaO_3

We apply in-plane uniaxial and biaxial strain ranging from 0 to 1% on pristine KTaO_3 to investigate its structural evolution and dielectric response over the temperature range of 0–300 K. The SSCHA + MLFF workflow, introduced in Methodology (Sec. II), enables us to obtain accurate structural information under different strain conditions while incorporating higher-order anharmonic effects.

Regarding the structural details, as shown in Fig. 3, both uniaxial and biaxial strain induce symmetry transformation and significant lattice distortions. The application of strain leads to structural transformation from cubic to tetragonal (uniaxial strain) or orthorhombic (biaxial strain) phase. Our calculations indicate that at low temperatures, KTaO_3 under uniaxial and biaxial strain adopts the space group $P4mm$ and $Amm2$, respectively. In contrast, pristine KTaO_3 belongs to the $P4/mmm$ space group. Both $P4mm$ and $Amm2$ exhibit lower symmetry than $P4/mmm$, with broken inversion symmetry, enabling ferroelectricity. And, the $Amm2$ phase possesses even lower symmetry than the $P4mm$ phase, as the tetragonal fourfold rotational symmetry is broken.

The O_1 –Ta bond and O_2 –Ta bond extend along the a and b directions, respectively, and their changes can represent

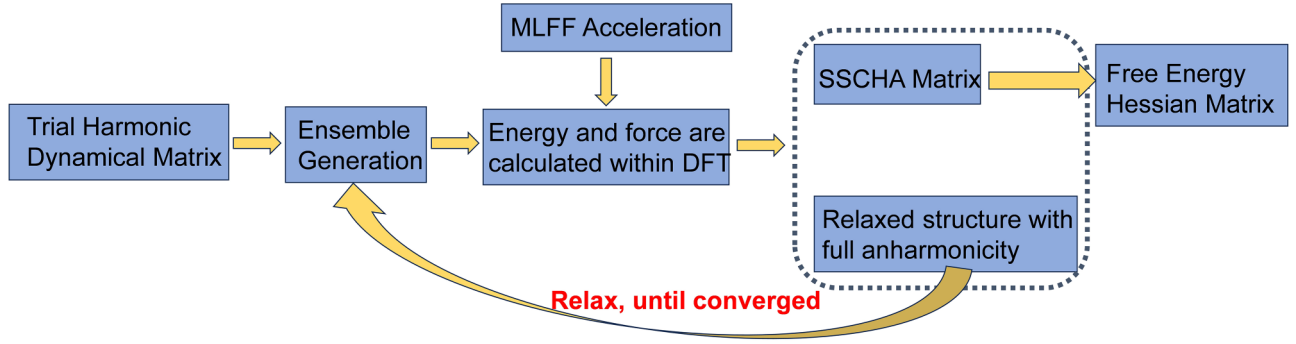


FIG. 2. Sketch of the SSCHA + MLFF workflow.

structural distortions along these two directions. Therefore, to further illustrate the impact of strain and temperature on structural distortions in strained KTaO_3 , we fix the potassium atomic positions and analyze the variation of O_1 –Ta bond length and O_2 –Ta bond length (see Fig. 4 and Fig. S1 [60]). Here, the relative change in bond length is defined as $\Delta l/l_0 = (l - l_0)/l_0$, where l and l_0 correspond to the bond lengths in the strained- and reference cubic structures, respectively. These bond-length variations serve as a key indicator of strain- and temperature-induced structural distortions. It is evident that for a given strain, the variation in the O_1 –Ta bond-length decreases with increasing temperature. This implies that the system exhibits reduced distortion at higher temperatures, progressively transitioning from a ferroelectric state to a paraelectric state. Under uniaxial strain, the space group evolves from $P4mm$ to $P4/mmm$, and under biaxial strain, from $Amm2$ to $P4/mmm$.

This transition is characterized by distinct Curie temperatures, marking the temperature at which the system loses its spontaneous polarization and restores higher symmetry. The system undergoes a symmetry transformation from a polar to a nonpolar space group. Furthermore, the lowest-temperature condition yields the most significant bond-length variation across all strain levels, highlighting the strong temperature dependence of the strain-induced lattice distortions.

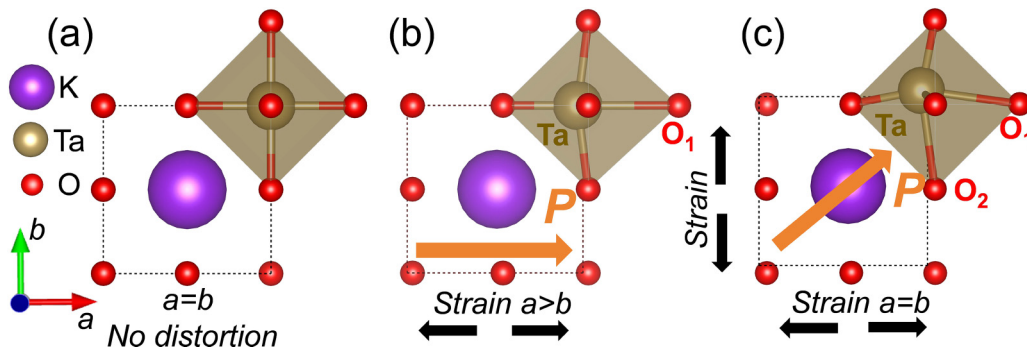
At a fixed temperature, the bond-length variation increases as the strain magnitude increases, indicating a direct correlation between strain and structural distortion. Notably, the largest applied strain consistently exhibits the most pronounced bond-length variation across the entire temperature range. The most pronounced distortion is observed under

1% strain at 0 K, emphasizing the strong interplay between strain and temperature in governing the structural properties of KTaO_3 .

Compared to uniaxial strained KTaO_3 , the only difference under biaxial strain is that structural distortions emerge along both the a and b directions. The distortion magnitudes along these two directions are essentially identical, with only negligible differences arising from numerical uncertainty. Furthermore, the overall distortion amplitudes under uniaxial and biaxial strain are comparable. The results underscore the critical role of both external strain and thermal effects in modulating the lattice distortions. Since the bond-length variation is directly correlated with ferroelectric distortion, it provides valuable insight into the emergence of ferroelectric polarization, which will be discussed in detail in subsequent analysis.

B. Lattice dynamics, anharmonic effects, and ferroelectric instability

For the dynamical properties, we obtained the phonon spectra of both uniaxial strained KTaO_3 and biaxial strained KTaO_3 under 1% strain and 0 K, as shown in Fig. 5. The calculated data show that, similar to pristine KTaO_3 [38], including anharmonic terms can stabilize the spurious imaginary ferroelectric soft mode predicted by harmonic approximation. Moreover, the other phonon branches remain unaffected by the anharmonic effects. This highlights the crucial role of anharmonicity, which significantly impacts the renormalized frequency of soft-mode phonons. Ultimately, this interplay determines whether a material can cross the QCP into a

FIG. 3. Geometric structure of (a) pristine KTaO_3 , (b) uniaxial strained KTaO_3 , and (c) biaxial strained KTaO_3 .

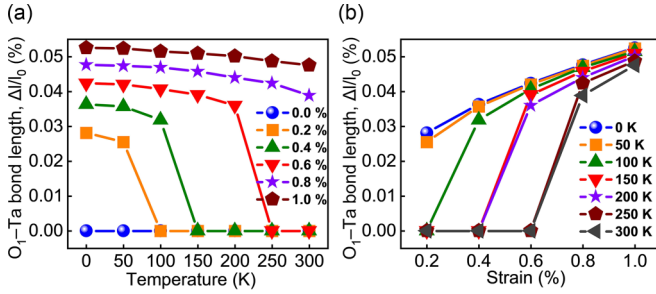


FIG. 4. Relative change in the O₁-Ta bond length [$\Delta l/l_0$ (%)] in KTaO₃ shown (a) as a function of temperature and (b) under uniaxial strain.

ferroelectric phase. Neglecting anharmonicity can lead to severe misjudgments in predicting polarization dynamics and phase-transition temperatures.

For the quantum paraelectric phase KTaO₃, the energy barrier for ferroelectricity is small compared to quantum fluctuations preventing the system from transitioning into ferroelectric state at any time. According to previous studies, the energy barrier of pristine KTaO₃ is approximately 0.306 meV per atom [38], while that of BaTiO₃ is around 4 meV per atom [39]. This comparison indicates that the energy barrier in KTaO₃ is significantly lower, allowing quantum fluctuations

to readily overcome it and thereby stabilizing the quantum paraelectric phase. These results are obtained from DFT calculations using the Strongly Constrained and Appropriately Normed (SCAN) functional [51], which we also employ for this study.

In the case of strained KTaO₃, it is crucial to understand the relationship between the ferroelectric energy barrier and quantum fluctuations. We analyze the one-dimensional potential-energy surface for KTaO₃ under 1% uniaxial and 1% biaxial strain at 0 K using the same SCAN functional and computational parameters. The energy barrier for 1% uniaxial and 1% biaxial strained KTaO₃ in Figs. S2(a) and S2(b) [60] are found to be 2.820 and 4.406 meV per atom, respectively, within DFT calculations, which are comparable to that in BaTiO₃ [39]. The results suggest that under strain, quantum fluctuations are no longer sufficient to overcome the energy barrier, leading to the stabilization of the ferroelectric phase. Additional to the DFT calculation, we also calculate the potential-energy surface by MLFF, which is shown by red line in Fig. S2 [60]. In comparison, the MLFF method predicts the energy barrier of 2.758 meV per atom (uniaxial strain) and 4.206 meV per atom (biaxial strain), demonstrating excellent agreement with the DFT result.

C. Temperature-dependent ferroelectricity and soft-mode evolution

As previously discussed, strained KTaO₃ exhibits significant and systematic structural distortions across various temperatures. To further investigate these distortions, we compute the ferroelectric polarization using the Berry-phase method [53–56]. The evolution of ferroelectric phase diagrams under different strains and temperatures, presented in Fig. 6(a) and Fig. S3 [60], is consistent with the bond-length trends observed in Fig. 4 and Fig. S1 [60]. The exact polarization values for each ferroelectric structure are displayed in Fig. S4 and Fig. S5 [60], with color intensity increasing proportionally to the magnitude. In the uniaxial case, the largest ferroelectric polarization occurs at the highest strain, reaching a value of 23.3 $\mu\text{C}/\text{cm}^2$. Under biaxial strain, the polarizations along both in-plane directions exhibit comparable magnitudes, resulting in a net polarization of 29.3 $\mu\text{C}/\text{cm}^2$ along the diagonal direction. Notably, the maximum ferroelectric polarization in both uniaxially and biaxially strained KTaO₃ is comparable to the polarization observed in BaTiO₃ [54,61–63], emphasizing the significant enhancement of ferroelectric properties due to strain. In the ferroelectric phase of strained KTaO₃, the soft-mode frequency remains positive across the entire low-temperature range up to the Curie temperature. As ferroelectric properties are closely linked to soft phonon modes, and given the approximately linear relationship between ferroelectric polarization and strain, it is reasonable to expect that the soft modes will also exhibit a linear dependence. To characterize this behavior, we exact the phonon spectrum of KTaO₃ under both 1% uniaxial and biaxial strain, examining the evolution of soft modes with temperature. Figure 6(b) and Fig. S6 [60] illustrate the evolution of the soft mode at Gamma point in phonon dispersion for KTaO₃ under both 1% uniaxial and biaxial strain at different temperatures. From the figures, it can be observed that in both cases, the

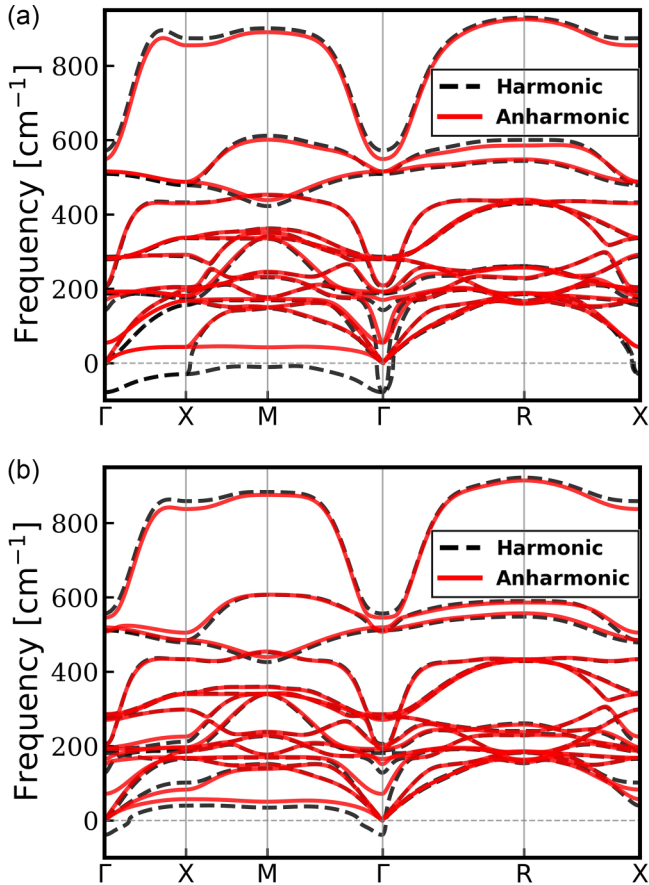


FIG. 5. Phonon spectrum of (a) 1% uniaxial strained KTaO₃ and (b) 1% biaxial strained KTaO₃ at 0 K without (black dashed line) and with (red line) anharmonic effects.

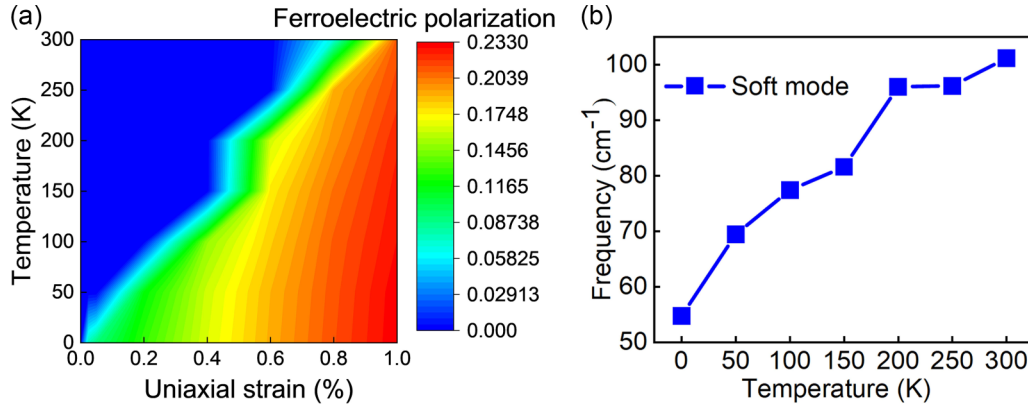


FIG. 6. (a) Phase diagram of KTaO_3 under uniaxial strain and at different temperatures, and (b) Variation of soft mode at Gamma point in ferroelectric phase for 1% uniaxial strained KTaO_3 across the temperature range of 0–300 K.

soft-mode frequency shifts to higher values as the temperature increases. This behavior aligns well with the linear trend observed in the ferroelectric polarization. The results demonstrate that anharmonic effects not only eliminate the imaginary frequencies associated with the ferroelectric soft mode but also exhibit the temperature-dependent evolution.

IV. CONCLUSION

In this study, we systematically investigated the effect of in-plane uniaxial and biaxial strain, ranging from 0 to 1%, on pristine KTaO_3 to evaluate its potential for ferroelectricity induction via inversion symmetry breaking over the temperature range of 0–300 K. By employing the SSCHA + MLFF workflow and Berry-phase method, we obtained accurate structural information and polarization values under different strain conditions while accounting for higher-order anharmonic effects.

Our calculations reveal that under uniaxial strain and biaxial strain in the range of 0.2 to 1%, the system undergoes a ferroelectric to paraelectric phase transition as temperature increases. Specifically, the space group evolves from $P4mm$ to $P4/mmm$ in the uniaxial strain case and from $Amm2$ to $P4/mmm$ under biaxial strain. Furthermore, our results demonstrate that similar to pristine KTaO_3 , the inclusion of anharmonic effects stabilizes the spurious imaginary ferroelectric soft mode predicted by the harmonic approximation.

Additionally, we found that at 0 K, the energy barrier for the system under 1% uniaxial strain is 3.1 meV per atom, which is comparable to the energy barrier in BaTiO_3

(4 meV per atom), suggesting that under strain, quantum fluctuations are no longer sufficient to overcome the energy barrier, thereby stabilizing the ferroelectric phase in strained KTaO_3 . Using the Berry-phase method, we constructed the ferroelectric phase diagram for both uniaxial and biaxial strained cases. Our calculations indicate that the maximum spontaneous polarization occurs at 1% strain and 0 K, reaching approximately $23.3 \mu\text{C}/\text{cm}^2$ along a direction for the uniaxial case and $29.3 \mu\text{C}/\text{cm}^2$ along the in-plane diagonal direction for the biaxial case.

By considering both temperature effects and anharmonicity, our study offers theoretical predictions that closely reflect realistic experimental conditions. These findings offer valuable guidance for future experimental and theoretical research on strain-engineered ferroelectricity in KTaO_3 .

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DATA AVAILABILITY

The data that support the findings of this study are available in Ref. [64].

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