A large modulation of electron-phonon coupling and an emergent superconducting dome in doped strong ferroelectrics

Jiaji Ma¹, Ruihan Yang¹ & Hanghui Chen¹,²✉

We use first-principles methods to study doped strong ferroelectrics (taking BaTiO₃ as a prototype). Here, we find a strong coupling between itinerant electrons and soft polar phonons in doped BaTiO₃, contrary to Anderson/Blount’s weakly coupled electron mechanism for “ferroelectric-like metals”. As a consequence, across a polar-to-centrosymmetric phase transition in doped BaTiO₃, the total electron-phonon coupling is increased to about 0.6 around the critical concentration, which is sufficient to induce phonon-mediated superconductivity of about 2 K. Lowering the crystal symmetry of doped BaTiO₃ by imposing epitaxial strain can further increase the superconducting temperature via a sizable coupling between itinerant electrons and acoustic phonons. Our work demonstrates a viable approach to modulating electron-phonon coupling and inducing phonon-mediated superconductivity in doped strong ferroelectrics and potentially in polar metals. Our results also show that the weakly coupled electron mechanism for “ferroelectric-like metals” is not necessarily present in doped strong ferroelectrics.
Electron-phonon coupling plays an important role in a variety of physical phenomena in solids. In metals and doped semiconductors, low-energy electronic excitations are strongly modified by the coupling of itinerant electrons to lattice vibrations, which influences their transport and thermodynamic properties. Furthermore, electron-phonon coupling provides an attractive electron-electron interaction, which leads to conventional (i.e., phonon-mediated) superconductivity in many metals. Recent studies on hydrogen-rich materials show that when their electron-phonon coupling is strong enough, the transition temperature of conventional superconductors can reach as high as 260 K at 180–200 GPa. One general way to increase the electron-phonon coupling of solids is to find a particular phonon to which itinerant electrons are strongly coupled and whose softening (i.e., the phonon frequency approaches zero) across a structural phase transition may consequently increase the total electron-phonon coupling. However, identifying a strong coupling between a soft phonon and itinerant electrons in real materials is no easy task, which relies on material details. On the other hand, the superconductivity in doped SrTiO$_3$ has drawn great interests from both theorists and experimentalists. One beautiful experiment is Sr$_{1-x}$Ca$_x$TiO$_3$, in which Ca doping leads to a weak ferroelectric distortion in SrTiO$_3$ and oxygen vacancies provide itinerant electrons. Increasing the carrier concentration in Sr$_{1-x}$Ca$_x$TiO$_3$ induces a polar-to-centrosymmetric phase transition and a superconducting “dome” emerges around the critical concentration. The nature of the superconductivity in doped SrTiO$_3$ is highly debated, because the superconductivity in doped SrTiO$_3$ persists to very low carrier density, which seriously challenges the standard phonon pairing mechanism. It is not clear why superconductivity in doped SrTiO$_3$ vanishes above a critical concentration in spite of an increasing density of states at the Fermi level. Attention has been paid to recent proposals on soft polar phonons, but the coupling details and strength are controversial. Furthermore, according to Anderson and Blount’s original proposal that inversion symmetry breaking by collective polar displacements in metals relies on the weak coupling between itinerant electrons and soft phonons responsible for inversion symmetry breaking, it is not obvious that across the polar-to-centrosymmetric phase transition the soft polar phonons can be coupled to itinerant electrons in Sr$_{1-x}$Ca$_x$TiO$_3$ or more generally in doped ferroelectrics and polar metals.

Motivated by the above experiments and theories, we use first-principle methods with no adjustable parameters to demonstrate a large modulation of electron-phonon coupling in doped strong ferroelectrics by utilizing soft polar phonons. We study BaTiO$_3$ as a prototype, because previous studies found that in doped SrTiO$_3$ vanishes above a critical concentration in spite of an increasing density of states at the Fermi level. Attention has been paid to recent proposals on soft polar phonons, but the coupling details and strength are controversial. Furthermore, according to Anderson and Blount’s original proposal that inversion symmetry breaking by collective polar displacements in metals relies on the weak coupling between itinerant electrons and soft phonons responsible for inversion symmetry breaking, it is not obvious that across the polar-to-centrosymmetric phase transition the soft polar phonons can be coupled to itinerant electrons in Sr$_{1-x}$Ca$_x$TiO$_3$ or more generally in doped ferroelectrics and polar metals.

Motivated by the above experiments and theories, we use first-principle methods with no adjustable parameters to demonstrate a large modulation of electron-phonon coupling in doped strong ferroelectrics by utilizing soft polar phonons. We study BaTiO$_3$ as a prototype, because (1) previous studies found that in doped SrTiO$_3$, increasing the carrier density gradually reduces its polar distortions and induces a continuous polar-to-centrosymmetric phase transition; and (2) the critical concentration for the phase transition is about $10^{21}$ cm$^{-3}$, which is high enough so that the electron-phonon coupling can be directly calculated within the Migdal’s approximation (in contrast, in doped SrTiO$_3$, superconductivity emerges at a much lower carrier concentration $10^{17}$–$10^{20}$ cm$^{-3}$ so that its Debye frequency is comparable to or even higher than the Fermi energy $\hbar \omega_D \sim 1$–$10^{28}$, which invalidates the Migdal’s approximation and Eliashberg equation). The key result from our calculation is that, contrary to Anderson/Blount’s argument for “ferroelectric-like metals”, we find that the phonon bands associated with the soft polar optical phonons are strongly coupled to itinerant electrons across the polar-to-centrosymmetric phase transition in doped BaTiO$_3$. As a consequence, the total electron-phonon coupling of doped BaTiO$_3$ can be substantially modulated via carrier density and in particular is increased to about 0.6 around the critical concentration. Eliashberg equation calculations find that such an electron-phonon coupling is sufficiently large to induce phonon-mediated superconductivity of about 2K. In addition, we find that close to the critical concentration, lowering the crystal symmetry of doped BaTiO$_3$ by imposing epitaxial strain further increases the superconducting temperature via a sizable coupling between itinerant electrons and acoustic phonon bands.

While ferroelectricity and superconductivity have little in common, our work demonstrates an experimentally viable approach to modulating electron-phonon coupling and inducing phonon-mediated superconductivity in doped strong ferroelectrics and potentially in polar metals. Our results show that the weakly coupled electron mechanism in “ferroelectric-like metals” is not necessarily present in doped strong ferroelectrics and as a consequence, the soft polar phonons can be utilized to induce phonon-mediated superconductivity across a structural phase transition.

Results

Structural phase transition induced by electron doping. In this study, electron doping in BaTiO$_3$ is achieved by adding extra electrons to the system with the same amount of uniform positive charges in the background. For benchmarking, our calculation of the undoped tetragonal BaTiO$_3$ gives the lattice constant $a = 3.930$ Å and $c/a = 1.012$, polarization $P = 0.26$ C/m$^2$, and Ti-O and Ba-O relative displacements of 0.105 Å and 0.083 Å, respectively, consistent with the previous calculations. We note that upon electron doping, BaTiO$_3$ becomes metallic and its polarization is ill-defined. Therefore, we focus on analyzing ionic polar displacements and $c/a$ ratio to identify the critical concentration.

We test four different crystal structures of BaTiO$_3$ with electron doping: the rhombohedral structure (space group $R3m$ with Ti displaced along (111) direction), the orthorhombic structure (space group $Ammm$ with Ti displaced along (011) direction), the tetragonal structure (space group $P4mm$ with Ti displaced along (001) direction) and the cubic structure (space group $Pm3m$ with Ti at the center of oxygen octahedron). Figure 1a shows that as electron doping concentration $n$ increases from 0 to 0.15e/f.u., BaTiO$_3$ transitions from the rhombohedral structure to the tetragonal structure, and finally to the cubic structure. The critical concentration is such that the crystal structure of doped BaTiO$_3$ continuously changes from tetragonal to cubic (see Supplementary Note 6). While the structural transition from tetragonal to cubic is first-order and thus does not show phonon softening (see Supplementary Note 7). Furthermore the low electron concentration in the rhombohedral structure invalidates Migdal’s theorem and electron-phonon coupling cannot be calculated within Migdal’s approximation (see Supplementary Note 5).

Figure 1b shows $c/a$ ratio and Ti-O cation displacements $\delta$ as a function of the concentration $n$ in the range of 0.06–0.14e/f.u. It is evident that the critical concentration $n_c$ of doped BaTiO$_3$ is $0.10e$/f.u. (about $1.6 \times 10^{21}$ cm$^{-3}$), at which the polar displacement $\delta$ is just completely suppressed and the $c/a$ ratio is reduced to unity. This result is consistent with the previous theoretical studies. Experimentally, in metallic oxygen-deficient BaTiO$_3$, the low-symmetry polar structure can be retained up to an electron concentration of $1.9 \times 10^{21}$ cm$^{-3}$ (close to the theoretical result)44,46. However, weak localization and/or phase separation may exist in oxygen-deficient BaTiO$_3$, depending on sample quality.

Electronic structure and phonon properties. Figure 2a shows the electronic structure of doped BaTiO$_3$ in the tetragonal...
structure at a representative concentration ($n = 0.09 \text{e/f.u.}$, close to the critical value). Undoped BaTiO$_3$ is a wide gap insulator. Electron doping moves the Fermi level slightly above the conduction band edge of the three Ti t$_{2g}$ orbitals and thus a Fermi surface is formed. We use three Wannier functions to reproduce the Ti t$_{2g}$ bands, upon which electron-phonon coupling is calculated. Figure 2b shows the phonon spectrum of doped BaTiO$_3$ in the tetragonal structure at 0.09e/f.u. concentration. We are particularly interested in the zone-center (Γ-point) polar optical phonons, which are highlighted by the green dots in Fig. 2b. The vibrational modes of those polar phonons are explicitly shown in Fig. 2c. In the tetragonal structure of BaTiO$_3$, the two polar phonons with the ion displacements along x and y directions ($\omega_x$ and $\omega_y$) are degenerate, while the third polar phonon with the ion displacements along z direction ($\omega_z$) has higher frequency. Figure 2d shows that electron doping softens the zone-center polar phonons of BaTiO$_3$ in the tetragonal structure until it reaches the critical concentration where the three polar phonon frequencies become zero. With further electron doping, the polar phonon frequencies of BaTiO$_3$ increase in the cubic structure (see Supplementary Note 13 for a discussion about doping’s effect on polar phonon behavior).

Electron-phonon coupling and phonon-mediated superconductivity. The continuous polar-to-centrosymmetric phase transition in doped BaTiO$_3$ is similar to the one in “ferroelectric-like metals” proposed by Anderson and Blount. They first argued, later recast by Puggioni and Rondinelli, that inversion symmetry breaking by collective polar displacements in a metal relies on a weak coupling between itinerant electrons and soft phonons responsible for removing inversion symmetry. According to this argument, one would expect that across the polar-to-centrosymmetric phase transition, the soft polar phonons are not strongly coupled to itinerant electrons in doped BaTiO$_3$. In order to quantify the strength of electron-phonon coupling and make quantitative comparison, we introduce the mode-resolved electron-phonon coupling $\lambda(q)$, and around-zone-center branch-resolved electron-phonon coupling $\lambda_c$:

$$\lambda(q) = \frac{1}{nN_p} \int_{|q|<q_c} d\mathbf{q}' \lambda_{\mathbf{q}'}, \quad \lambda_c = \sum_{|q|<q_c} d\mathbf{q}$$

where Im$\Pi_{\mathbf{q}}$ is the imaginary part of electron-phonon self-energy, $q_c$ is a small phonon momentum. The reason we define $\lambda_c$ within $|q| < q_c$ is because: (1) exactly at the zone-center Γ point, the acoustic phonon frequency is zero and thus the contribution from the acoustic mode is ill-defined at Γ point; (2) when $q_c$ is sufficiently small, there are no phonon band crossings within $|q| < q_c$ and hence each branch $\nu$ can be assigned to a well-defined phonon mode (for a general $\mathbf{q}$ point, it is not trivial to distinguish which phonon band corresponds to polar modes and which to other optical modes). We choose $q_c = 0.05\sqrt{\alpha}$ where $\alpha$ is the lattice constant (the qualitative conclusions do not depend on the choice of $q_c$, as long as no phonon band crossings occur within $|q| < q_c$).

Figure 3a, b show the imaginary part of electron-phonon self-energy Im$\Pi_{\mathbf{q}}$, for each phonon mode $\mathbf{q}$ of doped BaTiO$_3$ along a high-symmetry path (panel a corresponds to 0.09e/f.u. doping in a tetragonal structure and panel b corresponds to 0.11e/f.u. doping in a cubic structure). Since within the double delta approximation Im$\Sigma_{\mathbf{q}}$ is positive definite (see Supplementary Note 2), the point size in panels a and b is chosen to be proportional to the value of Im$\Pi_{\mathbf{q}}$. Our calculations find that, contrary to Anderson/Blount’s weak coupled electron mechanism, the phonon bands associated with the zone-center polar phonons have the strongest coupling to itinerant electrons, while the couplings of other phonon bands are weaker. Specifically, in the case of 0.09e/f.u. doping:

$$\lambda_{\text{acoustic}} = \sum_{\nu=1-3} \lambda_{\nu} = 3.83$$
$$\lambda_{\text{polar}} = \sum_{\nu=4-6} \lambda_{\nu} = 10.92$$
$$\lambda_{\text{others}} = \sum_{\nu=7-15} \lambda_{\nu} = 0.53$$

and in the case of 0.11e/f.u. doping:

$$\lambda_{\text{acoustic}} = \sum_{\nu=1-3} \lambda_{\nu} = 0.27$$
$$\lambda_{\text{polar}} = \sum_{\nu=4-6} \lambda_{\nu} = 5.58$$
$$\lambda_{\text{others}} = \sum_{\nu=7-15} \lambda_{\nu} = 0.11$$

In both cases, $\lambda_{\text{polar}}$ is larger than $\lambda_{\text{acoustic}}$ and $\lambda_{\text{others}}$. An intuitive picture for the strong coupling is that in doped BaTiO$_3$, the soft polar phonons involve the cation displacements of Ti and O atoms, and in the meantime itinerant electrons derive from Ti-d states which hybridize with O-p states (see Supplementary
The total electron-phonon coupling \( \lambda \) is obtained by taking the upper bound \( \omega \) to \( \infty \) in Eq. (5). The green shades are \( a^2F(\omega) \) and the dashed lines are the corresponding cumulative electron-phonon coupling. The total electron-phonon coupling \( \lambda \) of doped BaTiO\(_3\) in the tetragonal structure at 0.09/e/f.u. concentration is 0.61, while that in the cubic structure at 0.11/e/f.u. concentration is 0.50. Both \( \lambda \) are sufficiently large to induce phonon-mediated superconductivity with measurable transition temperature. Figure 3d shows the total electron-phonon coupling \( \lambda \) of doped BaTiO\(_3\) for a range of electron concentrations (exactly at the critical concentration, we find some numerical instabilities and divergence in the electron-phonon calculations, rendering the result unreliable). An increase of \( \lambda \) around the critical concentration is evident, consistent with the strong coupling between the soft polar phonons and itinerant electrons in doped BaTiO\(_3\).

Based on the electron-phonon spectrum \( a^2F(\omega) \), we use a three-orbital Eliashberg equation (see Supplementary Note 3) to calculate the superconducting gap \( \Delta(T) \) and estimate the superconducting transition temperature \( T_c \) as a function of electron concentration. Because the three Ti \( t_{2g} \) orbitals become identical at the critical concentration, when solving the three-orbital Eliashberg equation, we set Morel-Anderson pseudopotential \( \mu_i^* \) to be 0.1 for each orbital pair (i.e., \( i, j = 1, 2, 3 \))\(^{47}\). Figure 3e shows the superconducting gap \( \Delta(T) \) of doped BaTiO\(_3\) as a function of temperature \( T \) at two representative concentrations (0.09/e/f.u. in the tetragonal structure and 0.11/e/f.u. in the cubic structure). Since both concentrations are close to the critical value, the three Ti \( t_{2g} \) orbitals are almost degenerate in doped BaTiO\(_3\). For clarification, we show the superconducting gap of one orbital for each concentration. From the Eliashberg equation, we find that at 0.09/e/f.u. concentration, \( \Delta(T = 0) = 0.27 \) meV and \( T_c = 1.75 \) K; and at
Thus $\Delta(T=0)/k_B T_c = 1.79$ at 0.09 e/f.u. concentration and 1.68 at 0.11 e/f.u. concentration, both close to the BCS prediction of 1.77. Figure 3f shows the estimated superconducting transition temperature $T_c$ of doped BaTiO$_3$ for a range of electron concentrations. $T_c$ notably exhibits a dome-like feature as a function of electron concentration. The origin of the superconducting “dome” is that the electron-phonon coupling of doped BaTiO$_3$ is increased by the softened polar phonons around the critical concentration. When the electron concentration is away from the critical value, the polar phonons are “hardened” (i.e., phonon frequency increases) and the electron-phonon coupling of doped BaTiO$_3$ decreases. We note that the estimated $T_c$ strongly depends on $\mu^*_g$. Therefore in the inset of Fig. 3f, we study doped BaTiO$_3$ at a representative concentration (0.09 e/f.u.) and calculate its superconducting transition temperature $T_c$ as a function of $\mu^*_g$. As $\mu^*_g$ changes from 0 to 0.3, the estimated $T_c$ decreases from 9.3 K to 0.4 K, the lowest of which (0.4 K) is still measurable in experiment. We make two comments here: (1) The superconducting transition temperature is only an estimation due to the uncertainty of Morel-Anderson pseudopotential $\mu^*_g$ and other technical details. But the picture of an increased electron-phonon coupling around the structural phase transition in doped BaTiO$_3$ is robust. (2) Experimentally in Sr$_{1-x}$Ca$_x$TiO$_3$, the optimal doping for superconductivity is larger than the “ferroelectric” critical concentration, while in our calculations of doped BaTiO$_3$, the two critical concentrations (one for optimal superconducting $T_c$ and the other for suppressing polar displacements) just coincide due to polar phonon softening and an increased electron-phonon coupling. Comparison of these two materials implies that the microscopic mechanism for superconductivity in doped SrTiO$_3$ is probably not purely phonon-mediated.

Fig. 3 Electron-phonon properties and phonon-mediated superconductivity in doped BaTiO$_3$. a The imaginary part of the electron-phonon self-energy $\Im\Pi_{q\nu}$ for each phonon mode of doped BaTiO$_3$ at 0.09 e/f.u. concentration (tetragonal structure $T$). The point size is proportional to $\Im\Pi_{q\nu}$. The largest point corresponds to $\Im\Pi_{q\nu} = 4.6$ meV. The green dots highlight the zone-center polar optical phonons. b The imaginary part of the electron-phonon self-energy $\Im\Pi_{q\nu}$ for each phonon mode of doped BaTiO$_3$ at 0.11 e/f.u. concentration (cubic structure $C$). The point size is proportional to $\Im\Pi_{q\nu}$. The largest point corresponds to $\Im\Pi_{q\nu} = 3.2$ meV. The green dots highlight the zone-center polar optical phonons. c Electron-phonon spectral function $\alpha^2 F(\omega)$ and accumulative electron-phonon coupling $\lambda(\omega)$ of doped BaTiO$_3$ at 0.09 e/f.u. and 0.11 e/f.u. concentration. The total electron-phonon coupling $\lambda$ is 0.61 for the former and 0.50 for the latter. d Total electron-phonon coupling $\lambda$ of doped BaTiO$_3$ as a function of electron concentration $n$. e Superconducting gap $\Delta$ of doped BaTiO$_3$ as a function of temperature $T$ at 0.09 e/f.u. concentration (red) and at 0.11 e/f.u. concentration (blue), calculated by the three-orbital Eliashberg equation. The Morel-Anderson pseudopotential $\mu^*_g = 0.1$ is used for each orbital pair. f Superconducting transition temperature $T_c$ of doped BaTiO$_3$ calculated by the Eliashberg equation as a function of electron concentration $n$. The inset shows $T_c$ of BaTiO$_3$ in the tetragonal structure at 0.09 e/f.u. concentration as a function of Morel-Anderson pseudopotential $\mu^*_g$. 

For 0.11 e/f.u. concentration, $\Delta(T=0) = 0.11$ meV and $T_c = 0.76$ K. Thus $\Delta(T=0)/(k_B T_c) = 1.79$ at 0.09 e/f.u. concentration and 1.68 at 0.11 e/f.u. concentration, both close to the BCS prediction of 1.77. Figure 3f shows the estimated superconducting transition temperature $T_c$ of doped BaTiO$_3$ for a range of electron concentrations. $T_c$ notably exhibits a dome-like feature as a function of electron concentration. The origin of the superconducting “dome” is that the electron-phonon coupling of doped BaTiO$_3$ is increased by the softened polar phonons around the critical concentration. When the electron concentration is away from the critical value, the polar phonons are “hardened” (i.e., phonon frequency increases) and the electron-phonon coupling of doped BaTiO$_3$ decreases. We note that the estimated $T_c$ strongly depends on $\mu^*_g$. Therefore in the inset of Fig. 3f, we study doped BaTiO$_3$ at a representative concentration (0.09 e/f.u.) and calculate its superconducting transition temperature $T_c$ as a function of $\mu^*_g$. As $\mu^*_g$ changes from 0 to 0.3, the estimated $T_c$ decreases from 9.3 K to 0.4 K, the lowest of which (0.4 K) is still measurable in experiment. We make two comments here: (1) The superconducting transition temperature is only an estimation due to the uncertainty of Morel-Anderson pseudopotential $\mu^*_g$ and other technical details. But the picture of an increased electron-phonon coupling around the structural phase transition in doped BaTiO$_3$ is robust. (2) Experimentally in Sr$_{1-x}$Ca$_x$TiO$_3$, the optimal doping for superconductivity is larger than the “ferroelectric” critical concentration, while in our calculations of doped BaTiO$_3$, the two critical concentrations (one for optimal superconducting $T_c$ and the other for suppressing polar displacements) just coincide due to polar phonon softening and an increased electron-phonon coupling. Comparison of these two materials implies that the microscopic mechanism for superconductivity in doped SrTiO$_3$ is probably not purely phonon-mediated.
Crystal symmetry and acoustic phonons. We note that in Figure 3a, b, in addition to the large Im\(\Pi_{q\nu}\) in the polar optical phonon bands, there is also sizable Im\(\Pi_{q\nu}\) in the acoustic phonon bands (from \(\Gamma\) to \(X\)) in the tetragonal structure at 0.09/e/f.u. concentration. Since the mode-resolved electron-phonon coupling \(\lambda_{q\nu} \propto \text{Im}\Pi_{q\nu}/\omega_{q\nu}^2\), the small frequency of acoustic phonons can lead to a substantial \(\lambda_{q\nu}\), given a sizable Im\(\Pi_{q\nu}\). However, in the cubic structure at 0.11/e/f.u. concentration, Im\(\Pi_{q\nu}\) in the acoustic phonon bands almost vanishes from \(\Gamma\) to \(X\). To exclude that the concentration difference may have an effect, we perform a numerical experiment: we start from the cubic structure doped BaTiO\(_3\) at 0.11/e/f.u. concentration, and then we impose a slight (001) compressive bi-axial 0.8% strain by fixing the two in-plane lattice constants \((a\text{ and }b)\) to a smaller value. This compressive strain makes the crystal structure of doped BaTiO\(_3\) tetragonal and polar (space group \(P4mm\)). Figure 4a shows the optimized crystal structures of the two doped BaTiO\(_3\). For doped BaTiO\(_3\) at 0.11/e/f.u., without strain, the ground state structure is cubic and the optimized lattice constant \(a\) is 3.972 Å; under a 0.8% bi-axial (001) compressive strain, the ground state structure becomes tetragonal with the in-plane lattice constants \(a\) and \(b\) being fixed at 3.940 Å and the optimized long lattice constant \(c\) being 4.019 Å. We find that the total electron-phonon coupling \(\lambda\) increases from 0.50 in the \(Pm\bar{3}m\) structure to 0.57 in the \(P4mm\) structure. Figure 4b, c compare the imaginary part of the electron-phonon self-energy \(\text{Im}\Pi_{q\nu}\) and the mode-resolved electron-phonon coupling \(\lambda_{q\nu}\) along the \(\Gamma\rightarrow X\) path for the two doped BaTiO\(_3\). Similar to Figure 3a and b, we find that there is a notable difference in Im\(\Pi_{q\nu}\) from the acoustic phonon bands. The difference in Im\(\Pi_{q\nu}\) is further “amplified” by the low phonon frequencies \(\omega_{q\nu}\) which results in

\[
\text{without epitaxial strain } \lambda_{\text{acoustic}} = 0.27
\]

\[
\text{under 0.8% (001) compressive strain } \lambda_{\text{acoustic}} = 4.45
\]

At the same time, we find that for polar modes,

\[
\text{without epitaxial strain } \lambda_{\text{polar}} = 5.58
\]

\[
\text{under 0.8% (001) compressive strain } \lambda_{\text{polar}} = 3.21
\]

This shows that under 0.8% (001) compressive strain, \(\lambda_{\text{polar}}\) remains substantial (albeit reduced by about 40%), but \(\lambda_{\text{acoustic}}\) is increased by one order of magnitude, which altogether leads to an enhancement of the total electron-phonon coupling \(\lambda\). Note that in the numerical experiment, the two doped BaTiO\(_3\) have exactly the same electron concentration, indicating that the additional increase in Im\(\Pi_{q\nu}\) of the acoustic phonons arises solely from the crystal structure difference. A possible explanation, which is based on our calculations, is that in the cubic structure, some electron-phonon vertices \(g^{ij}_{q\nu}(k, q)\) are exactly equal to zero because some atoms are frozen in the acoustic phonons, while in the low-symmetry structure, those \(g^{ij}_{q\nu}(k, q)\) become non-zero. Because Im\(\Pi_{q\nu}\) \(\propto |g^{ij}_{q\nu}(k, q)|^2\), this leads to an increase in Im\(\Pi_{q\nu}\). In addition, the frequencies of acoustic phonon modes \(\omega_{q\nu}\) are very small and \(\lambda_{q\nu} \propto \text{Im}\Pi_{q\nu}/\omega_{q\nu}^2\), therefore even a slight increase in Im\(\Pi_{q\nu}\) results in a substantial enhancement in \(\lambda_{q\nu}\).
doped BaTiO₃, when the electron concentration is close to the critical value, a small (001) compressive strain that lowers the crystal symmetry may also enhance its superconducting transition temperature due to the increased electron-phonon coupling, similar to doped SrTiO₃.¹⁸,₂²

Discussion
Finally we discuss possible experimental verification. Chemical doping¹⁴,⁴⁵,⁵⁰–⁵⁴ and epitaxial strain⁵⁵,⁵⁶ have been applied to ferroelectric materials such as BaTiO₃. La-doped BaTiO₃ has been experimentally synthesized. High-temperature transport measurements show that Ba₁₋ₓLaₓTiO₃ exhibits polar metallic behavior but ultra-low-temperature transport measurements are yet to be performed⁵⁰–⁵⁴. We note that La doping in BaTiO₃ may result in some chemical disorder. While the randomness of La distribution in LaBa₁₋ₓTiO₃ may affect the transport properties in the normal state, Anderson’s theorem asserts that superconductivity in a conventional superconductor is robust with respect to non-magnetic disorder in the host material⁵⁷. As a consequence, the superconducting transition temperature Tc of a conventional superconductor barely depends on the randomness of defects. In our case, the superconductivity in doped BaTiO₃ is phonon-mediated (i.e., conventional) and La is a non-magnetic dopant. Therefore Anderson’s theorem applies and we expect that even if chemical disorder may arise in actual experiments, it does not affect the superconducting properties of doped BaTiO₃.

In addition, we perform supercell calculations which include real La dopants. We find that even in the presence of real La atoms, the conduction electrons on Ti atoms are almost uniformly distributed in LaBa₁₋ₓTiO₃ (see Supplementary Note 8 and Supplementary Note 9 for details). Since our simulation does not consider dopants explicitly, a more desirable doping method is to use electrostatic carrier doping³⁸–⁶⁰, which does not involve chemical dopants and has been successfully used to induce superconductivity in KTaO₃.⁶¹ We clarify two points concerning the electrostatic doping method. (1) The electrostatic gating by ionic liquid can achieve a two-dimensional carrier density as high as 8 × 10¹⁴ cm⁻². The induced electrons are usually confined in a narrow region that is a few nanometers from the surface/interface, which leads to an effective three-dimensional carrier density of about 1 × 10¹¹ – 5 × 10¹³ cm⁻³. In our current study, the critical concentration of doped BaTiO₃ is about 1.6 × 10¹⁵ cm⁻³, which is feasible by this approach. (2) While the electrostatic doping method induces the carriers in the surface/interface area, we show that our results on bulk doped BaTiO₃ can still be used as a guidance to search for superconductivity in the surface area of BaTiO₃.

In summary, we use first-principles calculations to demonstrate a large modulation of electron-phonon coupling and an emergent superconducting “dome” in n-doped BaTiO₃. Contrary to Anderson/Blount’s weak electron coupling mechanism for “ferroelectric-like metals”³²–³⁴, our calculations find that the soft polar phonons are strongly coupled to itinerant electrons across the polar-to-centrosymmetric phase transition in doped BaTiO₃ and as a consequence, the total electron-phonon coupling increases around the critical concentration. In addition, we find that lowering the crystal symmetry of doped BaTiO₃ by imposing epitaxial strain can also increase the electron-phonon coupling via a sizable coupling between acoustic phonons and itinerant electrons. Our work provides an experimentally viable method to modulating electron-phonon coupling and inducing phonon-mediated superconductivity in doped strong ferroelectrics. Our results indicate that the weak electron coupling mechanism for “ferroelectric-like metals”³²–³⁴ is not necessarily present in doped strong ferroelectrics. We hope that our predictions will stimulate experiments on doped ferroelectrics and search for the phonon-mediated superconductivity that is predicted in our calculations.

Methods
We perform first-principles calculations by using density functional theory⁴⁴–⁴⁷ as implemented in the Quantum ESPRESSO package⁶⁸. We use norm-conserving pseudo-potentials⁶⁹ with local density approximation as the exchange-correlation functional. For electronic structure calculations, we use an energy cutoff of 100 Ry. We optimize both cell parameters and internal coordinates in atomic relaxation. We find that the optimized crystal structures are in good agreement with experiments (see Supplementary Note 1). The detailed structural information is reported in Supplementary Note 7. In the strain calculations, the in-plane lattice constants are fixed while the out-of-plane lattice constant and internal coordinates are fully optimized. The electron Brillouin zone integration is performed with a Gaussian smearing of 0.005 Ry over a Γ-centered k mesh of 12 × 12 × 12. The threshold of total energy convergence is 10⁻⁷ Ry; self-consistency convergence is 10⁻¹² Ry; force convergence is 10⁻⁶ Ry/Bohr and pressure convergence for variable cell is 0.5 kbar. For phonon calculations, we use density functional perturbation theory⁶⁸ as implemented in the Quantum ESPRESSO package (see Supplementary Note 11 for the validation of this method on a prototypical oxide SrTiO₃). The phonon Brillouin zone integration is performed over a q mesh of 6 × 6 × 6. For the calculations of electron-phonon coupling and superconducting gap (see Supplementary Note 2), we use maximally localized Wannier functions and Migdal-Eliashberg theory, as implemented in the Wannier90 and EPW code⁷¹. The Fermi surface of electron-doped BaTiO₃ is composed of three Ti t₂g orbitals. We use three maximally localized Wannier functions to reproduce the Fermi surface. The electron-phonon matrix elements gₚ,k(q) are first calculated on a coarse 12 × 12 × 12 k-grid in the electron Brillouin zone and a coarse 6 × 6 × 6 q-grid in the phonon Brillouin zone, and then are interpolated onto fine grids via maximally localized Wannier functions. The fine electron and phonon grids are both 50 × 50 × 50. We check the convergence on the electron k-mesh, phonon q-mesh and Wannier interpolation and no significant difference is found by using a denser mesh. Details can be found in Supplementary Note 4. We solve a three-orbital Eliashberg equation to estimate the superconducting transition temperature Tc (see Supplementary Note 3). The only Eliashberg equation we use is the three-orbital Eliashberg equation when electron doping concentration is high enough so that Λγ/Tc < 0.1 and Migdal’s theorem is valid⁵⁹ (Λ is electron-phonon coupling, Tc is Debye temperature and Tγ is Fermi temperature). Validation test of Migdal’s theorem is shown in Supplementary Note 5.

We solve a three-orbital Eliashberg equation to estimate the superconducting transition temperature Tc. This method is compared to McMillan Equation. Details of Eliashberg Equation and McMillan Equation can be found in Supplementary Note 3.

Reporting summary. Further information on research design is available in the Nature Research Reporting Summary linked to this article.

Data availability
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Code availability
The electronic structure calculations were performed using the open-source code Quantum Espresso⁶⁸. Quantum Espresso package is freely distributed on academic use under the Massachusetts Institute of Technology (MIT) license.

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Author contributions
H.C. conceived and supervised the project. J.M. and H.C. performed the calculations. R.Y. contributed to the data analysis. H.C. and J.M. wrote the paper and all the authors commented on the paper.

Competing interests
The authors declare no competing interests.

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Correspondence and requests for materials should be addressed to H.C.

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