

Electric Field Control of Phonon Lifetimes and Thermal Conductivity in Relaxor-Based Ferroelectric

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The demand for high energy efficiency drives intense interest in thermal-management technology. Phonons are a major contributor to heat transfer in solids and controlling them through external stimuli is a key challenge for thermal management due to their weak interactions with applied fields. We report significant changes in phonon transport with the application of an electric field over a broad temperature range in a relaxor-based ferroelectric using neutron-scattering and -transport measurements. Phonon lifetimes increase along the applied poling field and this results in a tripling of the thermal conductivity in that direction. We also observe a suppression of nanoscale antiferroelectric fluctuations along the poling direction and argue that this increases the phonon lifetimes. Our results highlight a promising yet underexplored route to realistic solid-state heat switching from electric-field-modified nanostructures altering the phonon lifetimes in disordered functional materials.

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I. INTRODUCTION

The pursuit of energy efficiency drives research on harnessing and controlling heat flow. Precise control of heat flow is fundamental to the efficiency of all thermal cycles, a fact first codified in Carnot's theoretical upper limit for the efficiency of a heat engine [1] derived in 1824 and applied today in a wide variety of modern energy technologies. Tailoring heat flow in a solid-state material has a wide range of thermal-management applications including thermal control of caloric energy conversion [2], thermal regulators [3], magnetic refrigeration [4], and thermoelectric conversion systems [5].

Significant research over the past decade has been aimed at discovering the mechanisms needed to modify the thermal conductivity (κ) of materials. These efforts have

targeted structural phase transitions [6–8], mass transfer by ion insertion or removal [9,10], charge density distribution [11], or heat carriers [12,13]. Heat capacity and thermal conductivity are key elements in thermal-management devices. Perovskite-based ferroelectric (FE) materials have served as model systems [14] and have been significantly explored both theoretically and experimentally due to their rapid, repeatable, and well-integrated triggering mechanisms [15–19]. Dynamic control of thermal conductivity in FE materials subjected to an electric field can be achieved by inducing an antiferroelectric (AFE) to FE phase transition [8,19–21] or by reconfiguring an appropriate domain-wall structure to scatter phonons [8,15,19,21,22]. However, relaxor-based ferroelectrics (FEs) provide a unique opportunity to regulate phonon scattering at length scales comparable to phonon mean free paths through the presence of polar nanoregions (PNRs). Unlike solid-state phase transitions, which typically occur in a narrow temperature window, PNR-mediated control remains effective across a broad temperature range. Moreover, in contrast to domain engineering, PNRs act uniformly throughout the material, offering a more consistent mechanism for tuning phonon transport.

Phonons can be manipulated by external stimuli such as electric [15] or magnetic [12,29,30] fields, strain [31,32], pressure, [33] or light [6]. According to the phonon gas

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model, the phonon contribution to thermal conductivity is the weighted sum of contributions from the different modes, [34]

$$\kappa = \frac{1}{3} \sum_{\mathbf{q}s} C_{\mathbf{q}s} v_{\mathbf{q}s}^2 \tau_{\mathbf{q}s}, \quad (1)$$

where $C_{\mathbf{q}s}$ is the phonon specific heat capacity, $v_{\mathbf{q}s}$ is the phonon group velocity, and $\tau_{\mathbf{q}s}$ is the phonon lifetime. The phonon thermal transport can be tailored by impacting the phonon group velocities or lifetimes. In a recent experiment, an enhancement of thermal conductivity under an applied electric field in $\text{PbZr}_{(1-x)}\text{Ti}_x\text{O}_3$ (PZT) has been attributed to changes in phonon velocities [17]. Achieving electric-field-driven enhanced thermal conductivity by modifying phonon lifetimes has not been considered experimentally. The relaxor-based ferroelectric system $(1-x)[\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3]-x\text{PbTiO}_3$ (PMN- x PT) has been extensively studied for its outstanding piezoelectric response, high dielectric constant, and minimal strain–electric field hysteresis as a function of composition [35–37], temperature, and electric field [38,39], but its thermal-transport properties under electric fields remain largely unexplored. The only previous study is a measurement of the dynamic electric field dependence of $\kappa(E)$ in single-crystal PMN-33%PT [40], which, being just beyond the morphotropic phase boundary, exhibits less prominent PNR diffuse scattering. Domain-wall engineering of poled (ac or dc) PMN- x PT single crystals [22,41–45] could significantly enhance thermal conductivity, but achieving a domain size comparable to the short phonon mean free paths in bulk materials is challenging.

In previous work, the changes in thermal conductivity of the pristine samples with electric field are explained using a model based solely on the change in the longitudinal acoustic phonon velocity. However, after thermal cycling and electric field alterations, the effect increased tenfold and reversed sign [40]. While the sign change could be explained by changes in the piezoelectric coefficient, particularly d_{33} , the major increase remains unexplained and is tentatively attributed to changes in phonon lifetimes. In this work, we demonstrate increased phonon lifetimes directly using neutron scattering and shed light on controlling thermal conductivity in PMN-30%PT under electric field application. We report that the thermal conductivity of the [100]-poled PMN-30%PT is enhanced approximately threefold along the poling direction compared to the perpendicular direction over a wide range of temperatures. Neutron-scattering measurements show that this enhancement results primarily from changes in phonon lifetimes with poling. The phonon linewidths, which are inversely proportional to the lifetimes, are broad and diffuse for modes with atomic displacements ($\mathbf{Q}$) perpendicular to electric field poling and relatively sharp for modes with

displacements along the poling axis. We also observe suppressed antiferroelectric fluctuations at zone boundaries from atomic displacements along the poling direction, which aligns with the preference for ferroelectric over antiferroelectric displacements under an electric field. We argue that this suppression of the antiferroelectric fluctuations changes the phonon lifetimes with poling. Our results highlight the potential for using electric field manipulation of the nanostructure to actively control the thermal conductivity in solid-state devices.

II. RESULTS

A. Thermal conductivity of [100]-poled PMN-30%PT

To explore the impact of field poling on thermal conductivity, we poled a PMN-30%PT crystal along the [100] direction (see details in Sec. V). This orientation was chosen because it induces a 4R “engineered” domain structure that generates the largest piezoelectric response [46] and comes with changes in the phonons and local structure [47]. In Fig. 1, we show the giant effect in the κ values measured along two different directions, parallel and perpendicular to the poling directions, of the poled sample. We find that κ is about 3 times larger along the poling direction than in the perpendicular direction. Also, κ along the perpendicular direction closely matches that of the unpoled PMN- x PT near the morphotropic phase boundary ($x = 27, 32$) [23].

B. Time-of-flight inelastic neutron scattering on poled PMN-30%PT

To understand the giant poling effect on κ in PMN-30%PT, we used inelastic neutron scattering to study the heat-carrying phonons. The electric field poling has a large effect on the phonon linewidths, as shown in Fig. 2. The neutron-scattering maps in Figs. 2(a), 2(h) show that the phonon-dispersion curves are broad for momentum transfer, $\mathbf{Q}$, perpendicular to the poling field both through the zone center and along the zone boundary, respectively. The pseudocubic equivalent maps shown in Figs. 2(b), 2(i) reveal that these modes are relatively sharp for $\mathbf{Q}$ parallel to the poling field. This sharpening of the modes is clearly observed in constant-energy slices at 12 meV, where the TO mode appears to have an oval and a stretched oval shape in the $(0,K,L)$ and $(H,0,L)$ planes, respectively [Figs. 2(f), 2(g)]. Moreover, the modes at zone-boundary M points in Fig. 2(f) appear diffuse, whereas these same modes appear comparatively sharp in Fig. 2(g). The hint of broadening and sharpening of modes from PNRs [48] with electric field poling is also observed in Figs. 2(f), 2(g) at $K = -2$ and $H = 2$ r.l.u. (reciprocal lattice units), respectively. These results show that phonons with atomic displacements parallel to the poling field, as determined by $\mathbf{Q}$, are generally sharper.

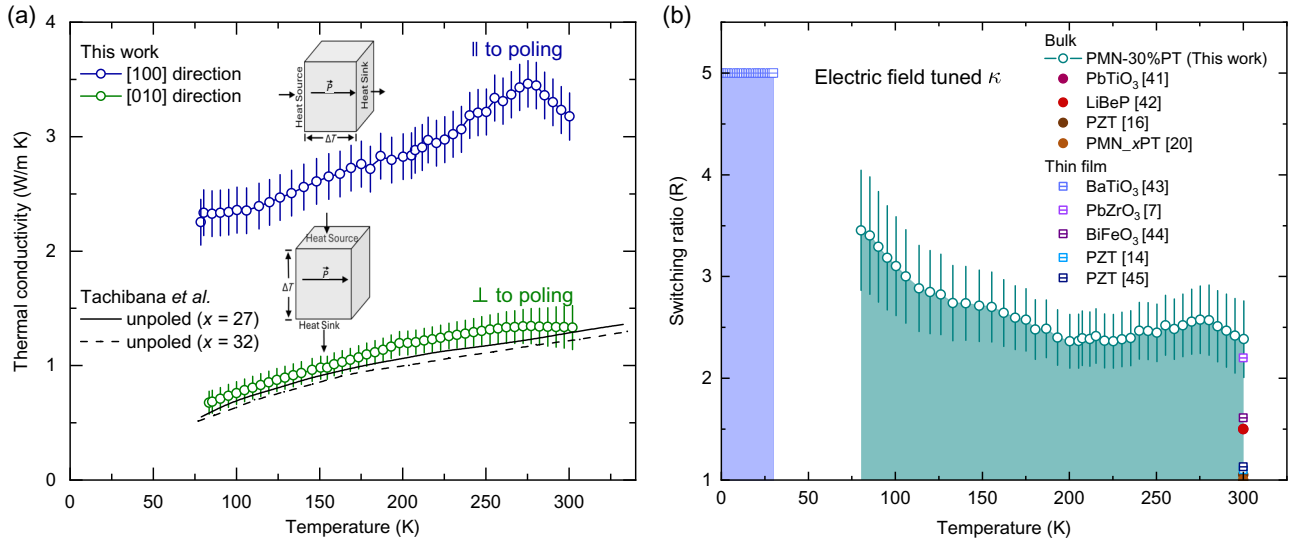


FIG. 1. Thermal-conductivity (κ) measurements in poled PMN-30%PT. (a) The thermal conductivity measured along directions parallel to poling (blue) and perpendicular to poling (green). The solid and dashed lines are the κ values in unpoled PMN- x PT for $x = 27$ and $x = 32$, respectively, measured by Tachibana *et al.* [23]. The insets in (a) are schematic diagrams of the thermal-conductivity measurement. (b) The thermal-switching ratio (R), defined as the ratio of κ values after and before applying the electric field, on bulk and thin films. The R values in this work (open green circles over the temperature range from 80 K to 300 K) represent the ratio of κ along the electric-field-poling direction to that perpendicular to it. This definition is consistent with the previous case, as the κ values perpendicular to the electric field closely match those of the unpoled sample (as shown in (a)). The R values in (b) are adopted from Refs. [8,15,17,22,24–28].

The relative changes in phonon linewidths are depicted in the multi-peak-fitted Lorentzian profiles [Figs. 2(c)–2(e) and 2(j)] to one-dimensional cuts [dashed white rectangular boxes in Figs. 2(a), 2(b), 2(h), and 2(i)] taken along L at constant energies. The reciprocal of the full width at half maximum (FWHM) of the Lorentzian function gives a measure of the phonon mean free path (MFP). The MFP of the TO phonon at $E = 10$ meV (Fig. 2(d)) is 2.104 ± 0.06 nm for atomic displacements perpendicular to the poling and is 2.66 ± 0.065 nm for atomic displacements along the poling direction. The MFP of the TA phonon branch [Fig. 2(j)] at $L \approx 0.14$ r.l.u. is 1.219 ± 0.273 nm for atomic displacements perpendicular to the poling and 2.912 ± 0.135 nm for atomic displacements along the poling direction. A similar mode-specific effect of poling is shown in Fig. S1 of the Supplemental Material [49] for other zone boundaries.

C. Effect of poling and temperature on phonon dispersion in PMN-30%PT

Along with the broadening and sharpening of phonon linewidths with poling, we also observe an unusual condensation of soft optic phonon modes at zone-boundary point $\mathbf{Q}_M$ (0.5, -2.5 , 0), which has a scattering vector perpendicular to the poling axis [Fig. 3(a)]. This feature is weaker at $\mathbf{Q}_M$ (2.5, -0.5 , 0) along the poling axis [Fig. 3(b)]. These modes are quantitatively depicted by an

arrow in the one-dimensional cuts in Fig. 3(c) (for similar poling effects at equivalent (0.5, -1.5 , 0) and (1.5, -0.5 , 0) points, see Fig. S2 of the Supplemental Material [49]). This field effect is similar to the temperature dependence of inelastic scattering at zone-boundary point $\mathbf{Q}_M$ (0.5, -2.5 , 0) observed in an unpoled PMN-30%PT crystal [Figs. 3(d)–3(f)]. Phonon softening at this position corresponds to antiferroelectric type atomic displacements.

The scattering shows that antiferroelectric fluctuations are suppressed in the poling direction. This makes sense, since ferroelectric displacements are favored over antiferroelectric displacements under an applied electric field. The reduction in antiferroelectric fluctuations likely reduces the phonon scattering rate for modes displacing atoms along the poling direction. The scattering phase space becomes more restricted with the flattening of the inelastic column at the M point along the poling direction, analogous to a similar temperature-driven effect discussed in Ref. [50]. This helps to explain the sharpening of the phonon linewidths for scattering vectors in the poling direction.

Lattice heat conduction along a direction is determined by phonons propagating in that direction, not by the atomic displacement direction. In Fig. 4, we illustrate the schematic diagrams for two different phonon-propagation directions, parallel to and perpendicular to the poling direction. Poling ($\vec{P}$) is along the [100] direction (black arrow). In Fig. 4(a), the phonon-propagation vector (long blue

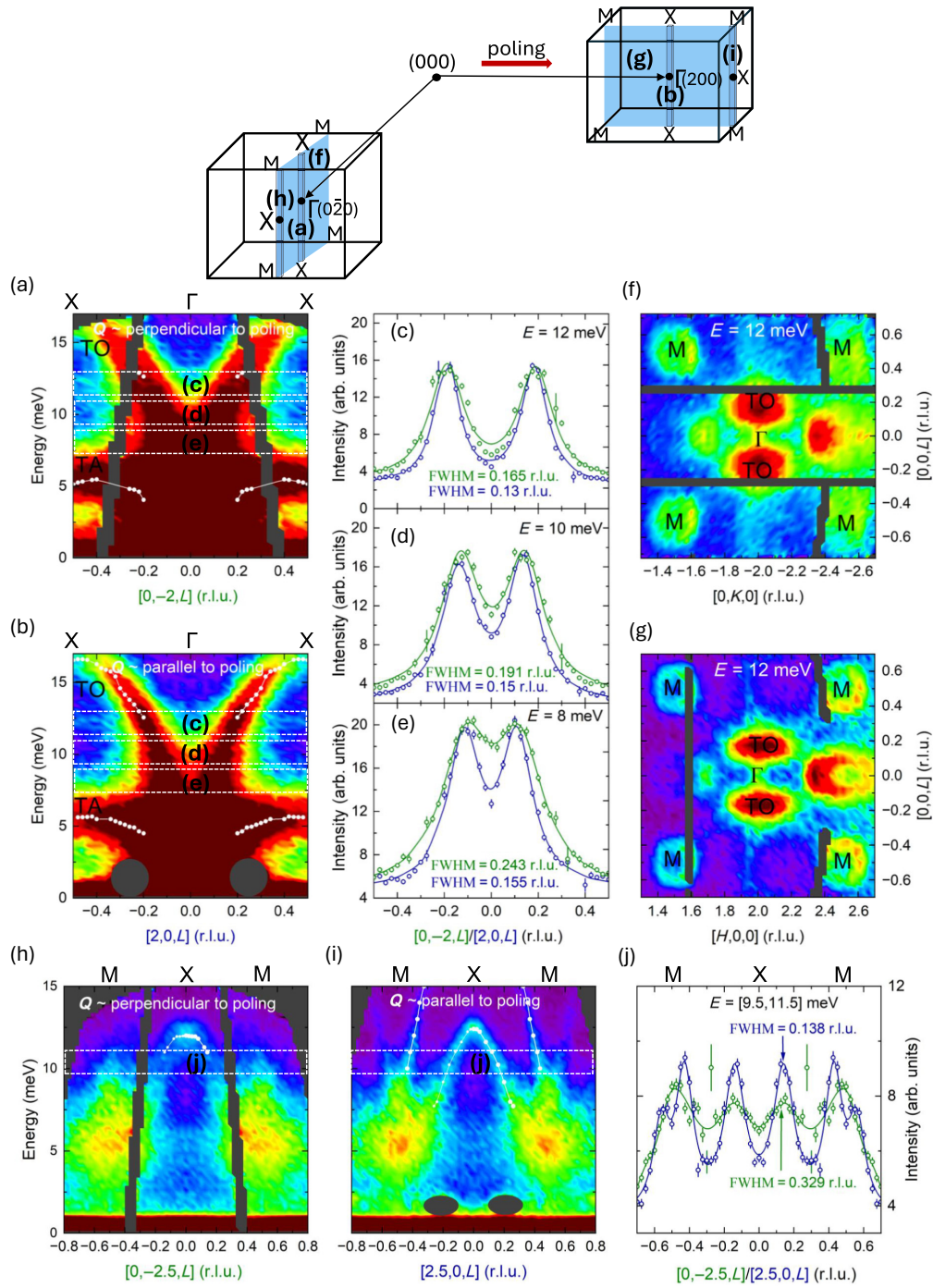


FIG. 2. The poling effect on phonon dispersion in PMN-30%PT. (a),(b) The transverse phonon dispersion measured along (a) $\mathbf{Q} = [0, -2, L]$ and (b) $\mathbf{Q} = [2, 0, L]$, respectively, in [100]-poled PMN-30%PT. (c)–(e) Fits to one-dimensional cuts shown as dashed white rectangular boxes in (a) and (b) at different energies [(c) 12, (d) 10, and (e) 8 meV, respectively] with integration, $\Delta E = \pm 1$ meV, in E and $\Delta q = \pm 0.05$ r.l.u. (reciprocal lattice units) in the orthogonal direction to the L axis along both $\mathbf{Q} = [0, -2, L]$ (green) and $\mathbf{Q} = [2, 0, L]$ (blue). (f),(g) Constant-energy slices at 12 meV taken in the (f) (0KL) and (g) (H0L) planes, respectively. (h),(i) The phonon dispersion measured along zone boundaries (h) $\mathbf{Q} = [0, -2.5, L]$ and (i) $\mathbf{Q} = [2.5, 0, L]$, respectively. (j) Fits to the corresponding cuts through the data in (h) and (i) with intensities integrated in E from 9.5 to 11.5 meV and $\Delta q = \pm 0.05$ r.l.u. in the direction orthogonal to the L axis, shown as dashed white rectangular boxes in (h) and (i). In (a), (f), and (h), the scattering vectors are perpendicular to the poling axis, whereas in (b), (g), and (i), by contrast, they are along the poling axis. Phonons are broad for atomic displacements perpendicular to the poling direction [(a),(f),(h)] and relatively sharp for displacements along the poling axis [(b),(g),(i)]. The data points overlaid on the images in (a), (b), (h), and (i) indicate the peak positions from fits to the TO and TA modes, and the white lines running through these points are guides to the eye.

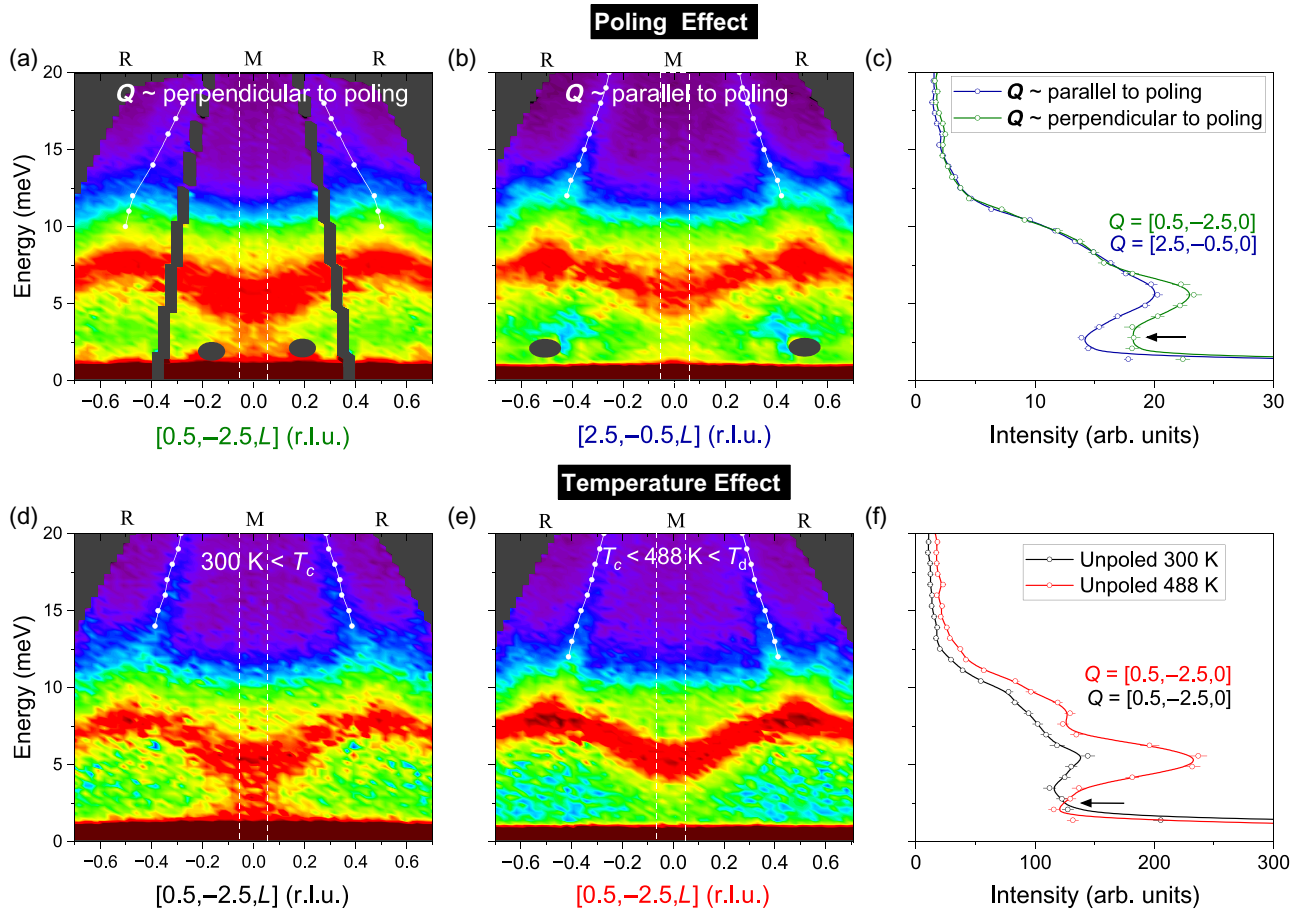


FIG. 3. The phonon dispersion in PMN-30%PT, comparing (a)–(c) poling and (d)–(f) temperature effects along the zone edge. (a) The phonon dispersion measured along $\mathbf{Q} = [0.5, -2.5, L]$ perpendicular to the poling direction. (b) The phonon dispersion measured along $\mathbf{Q} = [2.5, -0.5, L]$ parallel to the poling direction. (c) The inelastic scattering intensity at $L = 0$ in (a) and (b) with the integration of $\Delta q = \pm 0.05$ r.l.u. along all H , K , and L (dashed white rectangular boxes), showing marked antiferroelectric fluctuations extending down to the elastic line for a scattering vector perpendicular to the poling direction. All the phonon modes have relatively broad linewidths compared to those modes along the poling direction. (d),(e) The temperature dependence of similar phonon dispersion in unpoled PMN-30%PT along $\mathbf{Q} = [0.5, -2.5, L]$ measured (d) below the critical temperature at 300 K and (e) in between the critical temperature and the Burns temperature at 488 K ($T_c \approx 400$ K and $T_d \approx 688$ K), respectively. (f) The inelastic scattering intensity integrated over a range of -0.05 to 0.05 r.l.u. in L (dashed white rectangular boxes), showing antiferroelectric fluctuation softening to the elastic line below T_c . The data points overlaid on the images in (a), (b), (d), and (e) indicate the peak positions from fits, and the white lines running through these points are guides to the eye.

arrow) is along the poling direction, while in the bottom panel (long green arrow) it is perpendicular to the poling direction ([001]). Each phonon propagation is associated with $2N$ transverse (T) (small colored arrows) and N longitudinal (L) (small colored lines) polarizations, i.e., atom displacement directions, where $3N$ is the total number of phonon branches. For phonons propagating along the poling direction, both transverse modes have atomic displacements perpendicular to the poling direction and the longitudinal mode has atomic displacements along the poling direction. For phonons propagating perpendicular to the poling direction, one transverse mode has atomic displacements along the poling direction, while the other transverse mode and the longitudinal mode have atomic displacements perpendicular to the poling direction.

From the inelastic scattering measurements in the [100]-poled PMN-30%PT, we find that the transverse phonon modes propagating along the poling direction exhibit two opposing effects (yellow traffic signal), shorter lifetimes competing with higher group velocities [see Fig. S3(a) of the Supplemental Material [49]]. The longitudinal branch in this direction exhibits longer lifetimes along the zone boundary [see Figs. S4(a), S4(c), and S4(g) of the Supplemental Material [49]] that increase (green signal) the κ values. On the other hand, for the phonons propagating perpendicular to the poling direction, one transverse branch exhibits two opposing effects (yellow signal), higher lifetimes with lower velocities [see Fig. S3(b) of the Supplemental Material [49]], while the other transverse branch [Fig. 2(a)] and the longitudinal branch [see Figs. S4(b),

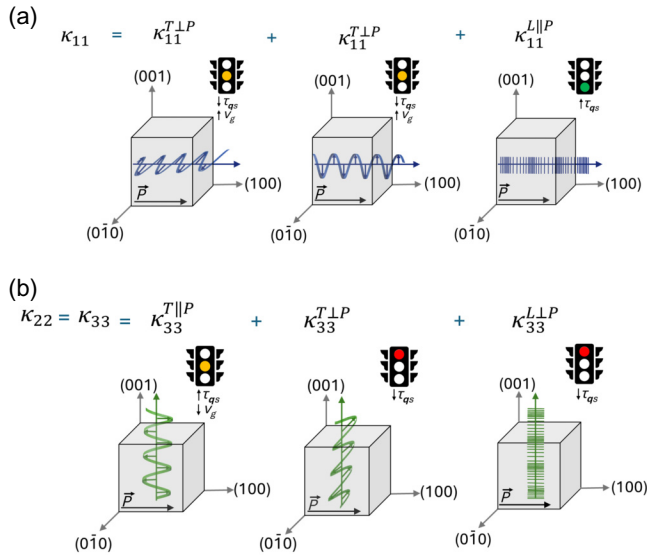


FIG. 4. An illustration of two different phonon-propagation directions in the [100]-poled system. (a) Schematic diagrams representing (a) the phonon propagation along the electric field (P along the [100] direction), whereas (b) shows the phonon propagation perpendicular to the electric field (along the [001] direction) with their three different atomic displacements—two transverse (T) (small colored arrows) and one longitudinal (L) (colored lines). The traffic light symbols are used as an analogy for the factors contributing to κ . The green, red, and yellow symbolize the increase, decrease, and two canceling effects, respectively, with regard to κ .

S4(c), and S4(g) of the Supplemental Material [49] exhibits decreased lifetimes along the zone boundary that reduce (red signal) the κ values in this direction. Overall, with regard to κ , we observed one increasing factor along the poling direction and two reducing factors perpendicular to the poling direction. This difference is reflected in our κ values (Fig. 1) measured along these two different directions. Hence, the changes in the phonon dispersion and linewidths provide a basis for understanding the electric-field-induced thermal-conductivity switching effect.

III. DISCUSSION

The large change in thermal conductivity with poling in Fig. 1 can be understood by examining how the electric field modifies the structure that affects heat-carrying phonons. PMN-30%PT sits on the morphotropic phase boundary with maximum polar nanostructure. The structure remains rhombohedral (R) after poling [51] with original $8R$ domain polarization along [111] equivalent directions in the unpoled state reduced to $4R$ domains aligned toward the [100] poling axis. The local atomic displacement of Pb atoms aligns along the [100] field-poling direction, which appears as a weak mode of approximately 11 meV in the phonon spectrum [47]. Furthermore, the

diffuse background perpendicular to the [100] poling direction decreases monotonically, which corresponds to the decrease in the local displacement of atoms separated by half of the lattice unit [47]. While poling along the [111] direction redistributes the [110]-type PNR diffuse scattering in PZN [52], these changes do not occur with the [100] poling used here [47].

Our neutron-scattering results reveal that phonon modes with atomic displacements along the poling axis have significantly sharper linewidths than modes with displacements perpendicular to it [see Figs. 2(a), 2(b), 2(h), 2(i), 3(a), and 3(b), and also Figs. S1(a), S1(b), S2(a), S2(b), S3(a), S3(b), S4(a), S4(b), S5(a), S5(b), S6(a), S6(b), and S7(b) of the Supplemental Material [49]]. However, even the sharper modes indicate phonon mean free paths of just a few nanometers, indicating that structural modifications at the nanoscale are affecting the phonons. On the other hand, the mechanisms behind poling effects on phonons are influenced by the varying length scales of different domains present in PMN- x PT [53]. Strong competition between long-range ferroelectric domains, short-range correlated antiferroelectric microdomains, and PNRs in PMN- x PT near the morphotropic phase boundary (MPB) [48] result in its very low strain-electric field hysteresis, high dielectric constants, and record-breaking piezoelectric coefficients. The TO modes observed at the zone center Γ point [Figs. 2(a), 2(b)] are linked to the ferroelectric domains, while the TO phonon located at the M point is characterized by atomic displacements that, if frozen, would form an antiferroelectric pattern of displacements. The unpoled PMN- x PT crystal exhibits strong composition-dependent antiferroelectric scattering at the M point, likely due to short-range antiferrodistortive displacements of lead atoms [36,54,55]. The soft vertical column of inelastic scattering at the high-symmetry M point along the zone edge Q_M (0.5, -2.5, 0) [Fig. 3(a)] is extended in energy up to approximately 6 meV but is confined in momentum space perpendicular to the poling direction [Fig. 3(a)]. A similar temperature-dependent soft column of inelastic scattering has also been observed in the parent PMN compound at Q_M (1.5, 1.5, 1) [56]. Along the poling direction, by contrast, this phonon is stabilized or hardened, with stabilization reflected in pushing the column intensity to higher energies (approximately 6 meV). This soft phonon column signals an instability driving antiferroelectric distortions, though without necessarily forming the static feature. Such antiferroelectric distortions should provide a strong source of scattering for phonons. Since an electric field favors ferroelectric distortions over antiferroelectric distortions, this destabilizes static antiferroelectric distortions (which means stabilizing the soft phonon columns) along the poling direction.

This suggests that competition between ferroelectric and antiferroelectric distortions persists in the direction perpendicular to the poling axis, while mainly ferroelectric

domains remain along the poling axis after the antiferroelectric distortions are suppressed. This suppression likely reduces phonon scattering rates for modes with atomic displacements along the poling axis, thereby increasing their lifetimes.

We considered other potential scattering factors that might contribute to the observed changes in κ values in PMN- x PT such as domain sizes or structural defects. However, none of these factors aligned with the findings from our inelastic-neutron-scattering experiment. While it is true that poling can influence domain morphology that could impact κ , if phonon scattering from the domain walls were the dominant factor governing κ , we would expect κ to increase uniformly in both directions due to the overall reduction of the domain-wall density with poling in PMN- x PT, as reported in Ref. [57]. Instead, we observe an increase only in the poling-field direction, showing a directionality that strongly aligns with nanoscale phonon scattering effects, as revealed by our inelastic-neutron-scattering data. Moreover, the size of the domain walls (approximately micrometers) is orders of magnitude larger than the mean free paths of the phonons (approximately nanometers). This disparity in length scales indicates that domain walls cannot contribute significantly to the phonon lifetimes that we observe. The other possible factor contributing to κ that warrants inspection here is structural defects—i.e., features with a polar nature that may change with the electric field poling if this defect is impacting κ . In the previous related work [47], we have reported the alignment of the Pb atom along the applied field direction in [100]-poled PMN-30%PT, which appears as a weak local dynamic mode around approximately 11 meV. While it is plausible that this local mode has a little impact on thermal conductivity, the observed modifications in the diffuse-neutron-scattering column—particularly at the antiferroelectric points—are far more pronounced and span a significantly larger region of Q - E space. These diffuse intense columns, revealed through our neutron-scattering experiment, serve as a substantial source of phonon scattering within the crystal lattice. Hence, we believe that this plays a critical role in driving the observed changes in phonon linewidths that alone is sufficient to account for the pronounced thermal-switching behavior observed in PMN- x PT.

To assess the impact of phonon changes on thermal conductivity, we have to weigh the controlling factors. In general, the total lattice thermal conductivity is the sum of the contributions from the phonon channel and the diffuson channel [58], with their relative impact depending on the dispersion curves and the phonon scattering rates. The diffuson channel emerges from the mode mixing of broad phonon branches. In PMN- x PT, the diffuson contribution to κ cannot be ignored due to its complex structural dynamics, which likely facilitate the coupling of different phonon branches at the Γ point in Figs. 2(a) and 2(b),

at the M points in Figs. 2(h) and 2(i), or at the R points in Figs. 3(a), 3(b), 3(d), and 3(e). This mode coupling exhibits directional dependence when the material is poled [59], likely resulting in a softening of the $[H, -2, 0]$ TO and $[2 + H, -2 + H, 0]$ TA modes [47]. The phonon lifetimes, which are crucial in determining the phonon channel, are discussed below.

The relative contributions to κ due to this change in phonon linewidths are illustrated in Fig. S4 of the Supplemental Material [49]. For comparison, two equivalent LA modes along $[H, 0, 0]$ and $[0, -K, 0]$ [see Figs. S4(e)–S4(h) of the Supplemental Material [49]] and TA modes along $[2.5, 0, L]$ and $[0, -2.5, L]$ [see Figs. S4(i)–S4(l) of the Supplemental Material [49]], corresponding to the directions parallel and perpendicular to the poling axis, are considered, which would be identical in the unpoled state. The change in LA modes upon poling is insignificant near the zone center Γ point along both directions [the left panel in Figs. S4(a) and S4(b) of the Supplemental Material [49]]. The LA mode along the poling axis appears to sharpen on moving toward the zone-boundary X point [see Fig. S4(c) of the Supplemental Material [49]]. Also, the TA modes become significantly sharper (higher τ_{qs}) along the poling axis compared to the direction perpendicular to the poling axis, on moving from the X point to the M point [see Figs. S4(a), S4(b) (right panel), and S4(d) of the Supplemental Material [49]]. The combined effect of $v_{qs}^2 \tau_{qs}$ of this TA mode has an enormous contribution to κ along the poling axis [the area under the blue curve in Fig. S4(l) of the Supplemental Material [49]] when compared to the mode perpendicular to the poling axis [the area under the green curve in Fig. S4(l) of the Supplemental Material [49]] (see also additional details in the Supplemental Material [49]). Likewise, the steeper and sharper TO mode at the M point along the poling axis [Fig. 2(i)] likely enhances κ along that direction, whereas the pseudocubic equivalent hardly resolvable TO mode perpendicular to the poling axis [see Fig. S2(h) of the Supplemental Material [49]] has an insignificant impact on κ . Hence, the changes are significant even though they are concentrated near the zone boundaries.

Relaxor-ferroelectric-based thermal switching provides new opportunities in regulating thermal transport within solid-state devices. Strong phonon scattering at the nanoscale is an intrinsic feature of relaxor ferroelectrics and our comprehensive neutron-scattering measurements reveal that the electric-field-driven changes to the thermal conductivity are largely controlled by changes in the phonon lifetimes, resulting in a large increase in thermal conductivity (approximately 3 times higher when poled) across a broad temperature range in a bulk material. This is a promising yet underexplored mechanism for precise manipulation of heat flow in solid-state devices that does not require special geometry, interfaces, or domain engineering. Future optimization to reduce the coercive

field needed for poling will enable unprecedented device-level control of heat-flows.

IV. CONCLUSIONS

We observe a giant electric-field-induced enhancement of thermal conductivity in a relaxor-based ferroelectric that reaches 300% of the unpoled material. This enhancement arises primarily from the suppression of phonon scattering along the poling-field direction, attributed to electric-field-induced modifications in the nanostructure, particularly the suppression of antiferroelectric fluctuations. These findings offer a promising application of relaxor ferroelectrics in developing thermal-switching devices, enabling improved thermal management under an applied electric field.

V. MATERIALS AND METHODS

A. Crystal growth

Bulk PMN-PT single crystals were grown using a modified Bridgman growth method. The crystals were aligned along the major crystallographic orientations $[001]_c$, $[011]_c$, and $[111]_c$ using Laue x-ray and x-ray diffractometry techniques. Discs 0.5 cm thick and 5 cm in diameter were sliced out from the bulk crystal using an inner diameter (ID) saw. The composition of the crystals was initially estimated based on their positions within the single-crystal ingot. This was later confirmed by measuring the transition temperatures of the individual cut pieces using high-temperature x-ray diffraction and comparing the results to known phase diagrams.

B. Electric field poling of the crystal

For poling, the two sliced pieces (0.5 cm thick and 5 cm diameter) were cleaned and coated first with an approximately 50-nm-thick layer of chromium and then later with an approximately 200-nm-thick layer of gold, using a sputtering process. The coated part was then poled using a polarization bath. The crystal slice was submerged in a bath of Fluorinert oil (silicone oil with high dielectric strength), and a dc electric field was applied. For the rhombohedral PMN-30%PT crystal, poling was carried out by applying a dc electric field of 4 kV/cm at room temperature. The piezoelectric charge coefficient (d_{33}) was measured using a d_{33} meter to confirm whether the crystal slices had been successfully poled into a piezoelectric state. After poling, polarization changes, as a function of the applied electric field, were observed on batch qualification samples using a hysteresis-loop device based on the Sawyer-Tower circuit. This method uses an integrator to measure the charge per unit area formed on the electrode surface of a dielectric when exposed to an electric field. The voltage applied follows a biased (unipolar) triangular waveform (from 0 to peak) for a few cycles (approximately

three). The area of the hysteresis loop, between the field-ascending and field-descending segments, is proportional to the energy lost as heat during each charge-discharge cycle and is characteristic of ferroelectric materials.

C. Thermal conductivity parallel and perpendicular to poling

Thermal conductivity was measured along directions parallel and perpendicular to $[100]$ poling axis using the static heater-sink method on two distinct samples. Both approximately 0.5-mm-thick samples were mounted identically. Type-T thermocouples (copper-constantan) were affixed using GE varnish, which is thermally conductive but electrically insulating. A strain gauge attached with GE varnish to the samples served as the heater. The samples were bonded to a lithium fluoride (LiF) slab, which served as an intermediate heat sink due to its high thermal conductivity. The use of this additional heat sink was necessary because the samples were too long to fit inside the cryostat while allowing space for the radiation shield; the LiF mount resolved this issue. LiF was then mounted on an alumina base for mechanical support and to enable placement inside the cryostat. Figure S10 of the Supplemental Material [49] shows a mounting configuration. The samples were then individually placed in a vacuum environment inside a Lakeshore nitrogen-cooled cryostat. During each experiment, the temperature was allowed to stabilize for an hour before recording the thermal conductivity. The uncertainty, represented by error bars in Fig. 1(a), arises from radiative heat losses and geometrical uncertainties; noise in the thermocouple measurement does not contribute significantly. The most important source of error is geometrical error, typically the distribution of the heat-flux lines in the sample and sample dimensions. The width and thickness of the sample can be measured quite accurately. The geometrical uncertainty is dominated by the uncertainty in the distance between the thermocouples. Its relative value is equal to the ratio of the thermocouple contact size to the spacing between them and is of the order of 10%.

The error due to radiative losses was quantified using a standard iron bar with known thermal conductivity, yielding thermal conductance values of 0.10 mW K^{-1} at 200 K and 0.44 mW K^{-1} at 300 K. For the PMN-30%PT samples, these losses correspond to $< 10\%$ at 200 K and $< 40\%$ at 300 K. Although these losses were subtracted from the measured conductance, the correction carries an uncertainty due to differences in emissivity between the PMN-30%PT samples and the iron standard, estimated to contribute a 30% error to the correction. As a result, the radiative-loss contribution to the absolute uncertainty in thermal conductivity is estimated to be 0.3% (parallel) and 3% (perpendicular) at 200 K, and 2% (parallel) and 12% (perpendicular) at 300 K.

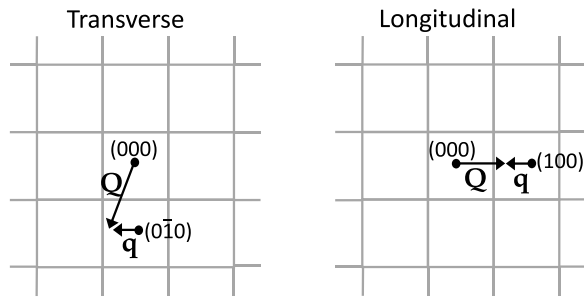


FIG. 5. An illustration of the scattering vector.

D. Time-of-flight inelastic neutron scattering

Time-of-flight measurements of [100]-poled PMN-30%PT and unpoled PMN-30%PT were performed using the ARCS spectrometer at the Spallation Neutron Source, Oak Ridge National Laboratory [60]. Measurements were performed with the crystal oriented in the (HK0) plane, with an incident neutron energy of 25 meV. Comprehensive four-dimensional Q - E data volumes were collected by rotating the angle between the [100] axis and the incident beam in 0.5° steps and combining and visualization of the data using the HORACE software [61]. For the unpoled PMN-30%PT crystal, data were collected at 300 and 488 K at each angle from 0° to 60° . For the poled PMN-30%PT crystal, data were collected at 300 K at each angle from -70° to 55° to obtain a complete data set that includes Q values in directions both perpendicular and parallel to the [100] poling direction in the (HK0) plane.

In an inelastic-neutron-scattering experiment, the change in the momentum of the incident neutron is referred to as the scattering vector ($\mathbf{Q}$). The scattering vector selects for phonon modes that displace atoms in the direction of momentum transfer. This appears as the mode polarization factor in the scattering function, $[\mathbf{Q} \cdot \mathbf{e}_j]$, where $\mathbf{e}_j$ is the polarization of the j th phonon [62]. Combining this polarization factor with the extended zone scheme enables neutron scattering to distinguish between transverse and longitudinal modes. Figure 5 illustrates how to orient the $\mathbf{Q}$ vector so as to measure the transverse (left) and longitudinal (right) modes at the same point in the zone, where $\mathbf{q}$, the phonon wave vector, gives the direction and wavelength of the phonon. This allows us to distinguish between transverse and longitudinal branches in the scattering-intensity map in Figs. 2 and 3.

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M.E.M. designed and led the INS experiments and J.P.H. designed and led the thermal-conductivity experiments. P.U. analyzed the INS data, with assistance from R.P.H. and M.E.M. Author D.R. measured and analyzed the thermal-conductivity data, with guidance from J.P.H. Author R.S. grew and field poled the crystals. D.L.A. and M.E.M. performed the INS measurements. All authors contributed to writing and editing the manuscript.

All authors declare no financial or nonfinancial competing interests.

DATA AVAILABILITY

The data that support the findings of this paper are openly available [63].

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