High-temperature thermal conductivity of ferroelectric and antiferroelectric perovskites

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We report thermal conductivity (κ) above 300 K for perovskite ferroelectrics BaTiO3 (Tc ≈ 402 K) and PbTiO3 (763 K), as well as antiferroelectrics PbZrO3 (503 K) and PbHfO3 (476 and 433 K). BaTiO3 and PbTiO3 show similar κ in the paraelectric phase. In contrast, smaller and glasslike κ is found above Tc for PbZrO3 and PbHfO3, signifying the presence of large anharmonic distortions in the antiferroelectric phase. Low-temperature heat capacity on PbZrO3 shows a lack of glasslike thermal behavior in the antiferroelectric phase. © 2022 The Author(s). Published on behalf of The Japan Society of Applied Physics by IOP Publishing Ltd

The perovskite oxides BaTiO3, PbTiO3, PbZrO3, and PbHfO3 are some of the most important compounds in the study of ferroelectricity and antiferroelectricity.1 At high temperatures, these compounds have the same paraelectric cubic structure (space group Pm 3 m), where the transition metal ions are located at the center of the oxygen octahedron, and the Ba/Pb ions occupy the 12-fold coordinated sites between the octahedra. Different sequences of first-order structural transitions are observed on cooling: (1) BaTiO3 shows a series of three ferroelectric (FE) transitions, from the cubic to tetragonal (P4mm) at Tc ≈ 402 K, tetragonal to orthorhombic (Ammn) at 278 K, and finally to rhombohedral (R3m) at 183 K. Each of these transitions is marked by uniform displacement of Ti ions, which is driven by the softening and condensation of the zone-center transverse optic (TO) mode. However, the soft mode is overdamped and the Ti ions show dynamic displacements along [111] in the cubic phase, leading to much debate on whether the order–disorder mechanism plays important roles in BaTiO3.2,3) (2) PbTiO3 displays a single FE transition at Tc ≈ 763 K to the tetragonal (P4mm) phase, where the displacement arises mostly from Pb ions. The FE soft mode in this case is underdamped (well-defined), such that PbTiO3 is often regarded as showing a “textbook” example of displacive FE transition. (3) PbZrO3 undergoes an antiferroelectric (AFE) transition4,5) at Tc ≈ 503 K to an orthorhombic (Pbam) phase, which contains eight formula units per cell. The AFE structure is described by the anti-parallel shifts of Zr ions along the cubic [110] and the antiferrodistortive (AFD) tilting of oxygen octahedra around [110]; these displacements correspond to lattice modes at the Σ and R points, respectively, of the Brillouin zone. In addition, the dielectric susceptibility shows Curie-Weiss behavior in the cubic phase (as in BaTiO3 and PbTiO3), implying the softening of zone-center TO mode. Recent studies6–9) are divided on which mode takes the role of the primary driving force for the AFE transition. The importance of order–disorder character has also been discussed.9) (4) PbHfO3 shows two AFE transitions at Tc ≈ 476 and 433 K, the former to an incommensurate (IC) phase10) and the latter to an orthorhombic (Pbam) phase that is isostructural with PbZrO3. It has been suggested that PbHfO3 holds a key to understanding PbZrO3, as a similar intermediate IC phase is found in PbZrO3 under high pressure.11) Because the FE and AFE instabilities interact with the anharmonic scattering of heat-carrying acoustic phonons, thermal conductivity (κ) measurements should provide important insights into these compounds. Indeed, the high-quality κ data for perovskite SrTiO3, obtained from both experiments12,13) and first-principles calculations,14) contributed much to the recent discussion on its phonon dynamics. In contrast, κ data15–18) for BaTiO3, PbTiO3, PbZrO3, and PbHfO3 are still mostly lacking, though κ of thin films has been examined in recent studies.19–21) To remedy this situation, we provide bulk high-temperature (T ≥ 300 K) κ data for the four perovskites. The results reveal a strong difference between the FE and AFE compounds: only the latter exhibit distinct glasslike behavior in their cubic phase. We also present low-T heat capacity (Cp) on PbZrO3 single crystals, which shows a lack of glasslike thermal behavior in the AFE phase.

Polycrystalline samples of BaTiO3, PbTiO3, PbZrO3, and PbHfO3 were prepared by solid-state reactions. High purity (≥ 99.9%) powders of BaCO3, PbO, TiO2, ZrO2, and HfO2, with particle sizes of ~1–20 μm, were dried and weighed to either the stoichiometric ratio (for BaTiO3) or 1% excess PbO. The powders were then mechanically ground for 30 min and calcined in air for 3 h at 1373 K (BaTiO3) or 1123 K (the others). After the calcined products were ground again, they were divided into two portions. To the first portion was added 2 wt% polyvinyl alcohol (PVA) binder, which was then pelletized under an isostatic pressure of 200 MPa and heated at 723 K for 3 h to remove the binder. For PbTiO3, PbZrO3, and PbHfO3, these pellets were buried in the second portion in a tightly covered Pt crucible, which was in turn sealed in an Al2O3 crucible using alumina cement. This procedure minimized the loss of PbO during the 3 h sintering at 1523 K (PbTiO3) or 1573 K (PbZrO3 and PbHfO3). For BaTiO3, the pellets were sintered directly in the air at 1623 K for 4 h. Powder X-ray diffraction confirmed that the products were single-phase.
The $\kappa$ data were obtained using the relation $\kappa = D\rho C_p$, where $D$ is thermal diffusivity and $\rho$ is density. The $D$ values were determined in a nitrogen atmosphere on heating direction by the flash technique, using either Netzsch LFA 467 for 300–773 K or LFA 467HT for 300–993 K. For each measurement, a $10 \times 10 \text{mm}^2$ square plate of $\sim 1.5 \text{mm}$ thickness was coated on both sides with a thin layer of graphite. The $C_p$ values were taken from the literature, as described below. The $\rho$ was determined by the Archimedes method, and its $T$ dependence was calculated using thermal expansion data.\(^{22-25}\) As the $\rho$ of sintered pellets were 88%–95% of the theoretical values, the raw $\kappa$ values ($\kappa_{\text{raw}}$) were converted to the $\kappa$ of a fully dense solid through the relation $\kappa_{\text{raw}}/\kappa = 1 - 4\phi/3$, where $\phi$ is the porosity of the specimen.\(^{26}\) The overall uncertainty in $\kappa$ is estimated to be less than $\pm 10\%$ for BaTiO$_3$, PbZrO$_3$, and PbHfO$_3$, and slightly larger for PbTiO$_3$ (see below).

Single crystals of PbZrO$_3$ with $< 2$ mm in size were grown by the flux method using excess PbO as the flux. The powder X-ray diffraction pattern was indistinguishable from the polycrystalline data, and differential scanning calorimetry showed a sharp peak with the onset $T = 506.5 \text{K}$ on heating and 505.5 K on cooling. A collection of three single crystals was used to measure $C_p$ between 0.5 and 2 K using the relaxation method of the Physical Properties Measurement System by Quantum Design.

As the accuracy of our $\kappa$ is constrained by that of $C_p$, we first examine the $C_p$ values used in this study. The inset of Fig. 1 shows published $C_p$ for PbTiO$_3$, PbZrO$_3$, and PbHfO$_3$, in units of $J \text{K}^{-1} \text{mol}^{-1}$.\(^{27}\) The high quality of the data can be ascertained by the observations that (1) they join smoothly with the lower-$T$ data obtained from other measurements,\(^{27}\) and (2) a clean, sharp peak is observed for each of the first-order transitions. Also, in the cubic phase of each compound, $C_p$ saturates at a value slightly below the Dulong–Petit limit of $125 J \text{K}^{-1} \text{mol}^{-1}$, and this continues at least up to 1050 K for PbTiO$_3$.\(^{28}\) In other perovskites, including BaTiO$_3$, SrTiO$_3$,\(^{31}\) and BaZrO$_3$,\(^{32}\) high-$T$ $C_p$ exceeds the Dulong–Petit limit due to anharmonic lattice expansion. We associate the unusual behavior of Pb-based perovskites with the easily polarizable Pb ions, although further studies are certainly needed to confirm this idea. In any case, the data provide reliable $C_p$ for the present purpose, except in the vicinity of $T_c$ where even small sample dependence leads to a large difference in $C_p$. For this reason, we exclude $\kappa$ data in such $T$ regions.

The $C_p$ for BaTiO$_3$ is shown in the main panel of Fig. 1. The data up to 420 K were obtained by Moriya using an adiabatic calorimeter,\(^{33}\) which has an accuracy of 0.2% above 300 K. (The sharp peak at $T_c$ climbs up to $210.35 J \text{K}^{-1} \text{mol}^{-1}$ at 401.72 K.) The figure also shows Coughlin and Orr’s $C_p$\(^{29,30}\) which lacks the sharp peak but exhibits larger values just above $T_c$. Accordingly, we represent the data by the $C_p$ of SrTiO$_3$ (Ref. 31) multiplied by 1.03, which joins smoothly with Moriya’s data. It is noted that more recent\(^{34}\) $C_p$ on BaTiO$_3$ appears to be too large by $\sim 5\%$.

The main panel of Fig. 2 shows the $\kappa$ of BaTiO$_3$, for which the present result is labeled as polycrystalline. Also shown is $\kappa$ for a single crystal, obtained from Hofmeister’s $D$ data\(^{35}\) using the same $C_p$. (As it shows a lower $T_c$, $C_p$ in the cubic phase was extrapolated.) These results exhibit several important features. First, polycrystalline values are lower than those of single crystal by $\sim 20\%$, which is similar to the case\(^{13}\) of SrTiO$_3$ shown in the inset. Such reduction in $\kappa$ should result from the scattering of thermal phonons at grain boundaries. Second, the single crystal shows strongly anisotropic $\kappa$ in the tetragonal phase, and their weighted average is closely matched in its slope by the polycrystalline data. The polycrystalline $\kappa$ of 2.8 W m$^{-1}$ K$^{-1}$ at 300 K agrees well with the published room temperature values.\(^{36}\) Third, $\kappa$ in the cubic phase is less than half the values in SrTiO$_3$, and it decreases almost linearly with $T$, unlike the more typical $T^{-1}$
drop in SrTiO$_3$. From the kinetic expression, $\kappa = (1/3)Cv_l$, where $C$ is the heat capacity per unit volume, $v$ is the averaged phonon group velocity, and $l$ is the mean free path of the phonons. As comparable $v$ for BaTiO$_3$ and SrTiO$_3$ can be deduced from the acoustic dispersion curves, the smaller $\kappa$ in BaTiO$_3$ is attributed to a shorter $l$, arising from stronger phonon scattering by anharmonic phonon-phonon interactions.

Possible sources of additional phonon scattering in cubic BaTiO$_3$ are (i) the $\langle 111 \rangle$ disorder of Ti ions, which is most dynamic in character, and (ii) the FE soft mode. (For SrTiO$_3$, the former is practically absent and the latter occurs at much lower $T$.) To further evaluate these contributions, we next compare the $\kappa$ of BaTiO$_3$ and PbTiO$_3$, shown in Fig. 3; for PbTiO$_3$, the $\langle 111 \rangle$ Ti disorder is negligible and the FE soft mode is reported to interact strongly with the heat-carrying phonons. Moreover, PbTiO$_3$ shows much flatter acoustic dispersion curves than BaTiO$_3$, leading to a lower $v$. Vibration energies at zone boundaries are less than half the values in BaTiO$_3$. Considering these differences, the observation of similar cubic $\kappa$ for BaTiO$_3$ and PbTiO$_3$ ($\sim$2–2.5 W m$^{-1}$ K$^{-1}$) implies shorter $l$ in BaTiO$_3$, which in turn can be attributed to the $\langle 111 \rangle$ Ti disorder. Recently, this disorder in BaTiO$_3$ was incorporated into the calculation of phonon dispersion curves. It would be of great interest to extend such works to obtain $\kappa$ from the first-principles.

Figure 3 also reveals stronger anomaly across $T_c$ for PbTiO$_3$, reflecting its larger polarization and tetragonal strain ($c/a = 1.06$ in PbTiO$_3$ and 1.01 in BaTiO$_3$ at room temperature). Also, when the measurements were repeated after the sample was cooled from the cubic phase, only PbTiO$_3$ showed significant difference (4% larger) from the first measurements; similar difference (3% smaller) occurred on another PbTiO$_3$ sample heated only to 773 K. This feature suggests that $\kappa$ in the FE phase depends on how the large tetragonal strain is accommodated in the sample, which varies each time it experiences the transition. Perhaps related to this, the present $\kappa$ shows a stronger slope in the FE phase than the previous polycrystalline data, and $\kappa = 3$ W m$^{-1}$ K$^{-1}$ at 300 K is much lower than 5 W m$^{-1}$ K$^{-1}$ for a multidomain single crystal [see Fig. 4(a)]. Thus, measurements on a single domain crystal are probably needed to assess the intrinsic $\kappa$ in the FE phase, and to make a close comparison with the results of first-principles $\kappa$ calculations.

The $\kappa$ of AFE compounds PbZrO$_3$ and PbHfO$_3$ are also shown in Fig. 3. Their small values compared to BaTiO$_3$ and PbTiO$_3$ are obvious, and the smaller $\kappa$ for PbHfO$_3$ is ascribed to the heavier mass of Hf compared to Zr. Furthermore, both the magnitude and $T$ dependence in the cubic phase resemble those of glasses, as evidenced through comparison with the $\kappa$ of amorphous silica ($\alpha$-SiO$_2$). Such glasslike $\kappa$ occurs when $l$ is limited to the order of interatomic distance, where heat is carried by a random walk of energy between localized oscillators. Empirically, this condition is satisfied in amorphous solids and a range of chemically disordered crystals. On the other hand, PbZrO$_3$ and PbHfO$_3$ do not possess chemical disorder, so that, the glasslike $\kappa$ should be related to large anharmonic distortions, which are presumably stronger than the Ti disorder in BaTiO$_3$. The evidence for such distortions can be found in structural studies, where

![Fig. 3](image-url) (Color online) Thermal conductivity of polycrystalline BaTiO$_3$, PbTiO$_3$, PbZrO$_3$, and PbHfO$_3$. The $\kappa$ of amorphous silica ($\alpha$-SiO$_2$) is shown as a dashed line.

![Fig. 4](image-url) (Color online) (a) Thermal conductivity of single crystal PbTiO$_3$, single crystal PbMg$_{1/3}$Nb$_{2/3}$O$_3$, and $\alpha$-SiO$_2$, and polycrystalline PbZrO$_3$. For PbZrO$_3$, both the present result and Lawless’s data are shown. (b) Low-temperature heat capacity of the same perovskites, plotted as $C_p/T$ versus $T^2$. For PbZrO$_3$, Lawless’s and present single crystal data are shown. The solid lines are fits to $C_p = \gamma T + \beta T^3$. © 2022 The Author(s). Published on behalf of The Japan Society of Applied Physics by IOP Publishing Ltd.
large displacements of Pb ions and oxygen octahedral tilting are reported for the cubic phase. Moreover, the dynamic nature of such distortions is seen through the relaxation polar mode, and these results have been interpreted as signatures of order–disorder mechanism for the AFE transition. Interestingly, the present results provide additional support for the order–disorder mechanism, as $\kappa$ changes to crystalline-like $d\kappa/dT < 0$ below $T_c$.

We view the $d\kappa/dT < 0$ behavior in the AFE phase as an important result, since previous data on PbZrO$_3$ showed either a constant $\kappa$ or a weakly positive $d\kappa/dT$ below $T_c$; only the present result strongly questions Lawless’s conclusion that PbZrO$_3$ exhibits a glasslike thermal behavior in the AFE state. To discuss this problem in sufficient detail, we now focus on the low-$T$ thermal properties.

Figure 4(a) shows the $\kappa$ of PbTiO$_3$, PbMg$_{1/3}$Nb$_{2/3}$O$_3$ (PMN), a-SiO$_2$, and PbZrO$_3$, plotted in the double logarithmic scale. Here, both the present result and Lawless’s data are plotted for polycrystalline PbZrO$_3$, while the data for PbTiO$_3$ and PMN come from single crystals. As an example of normal ferroelectrics, PbTiO$_3$ exhibits the usual crystalline $\kappa$ behavior, with a peak at 65 K. In contrast, a FE relaxor PMN shows the prototypical glassylike behavior, with a signature plateau at ~10 K. The glassylike behavior in PMN can be attributed to the presence of randomly oriented polar nanoregions. As for the AFE compound PbZrO$_3$, Lawless suggested that a glassylike plateau should occur above 40 K, the upper limit of the data. However, the glassylike plateau should not occur at such a high-$T$. Moreover, the present data suggest the presence of a broad peak above 40 K.

To make a more definitive statement on PbZrO$_3$, we have measured the low-$T$ $C_p$ on single crystals. The result plotted as $C_p/T$ versus $T^2$, is shown with the other perovskites in Fig. 4(b). Here, the linear lines are the fit to $C_p = \gamma T + \beta T^2$, where the intercept $\gamma$ arises from the glassylike two-level tunneling systems and the slope $\beta$ is due to phonons. For PbTiO$_3$, $\gamma = 0$ and $\beta$ agrees with the value obtained from elastic constants, which are the expected behavior of a crystalline insulator. In contrast, PMN has a glassylike $\gamma$ and $\beta$ is much larger than the elastic value, indicating the presence of glassylike nonacoustic contributions. As for PbZrO$_3$, an even larger $\gamma$ (=0.8 mJ K$^{-1}$ mol$^{-1}$) is seen in Lawless’s data, and it was used as evidence for glasslike behavior. However, this result is not reproduced by our data on single crystals, which show a lack of $\gamma$ but similar $\beta$. In particular, our $C_p$ above ~1.1 K closely follows that of PbTiO$_3$, which is consistent with similar elastic Debye temperatures (~396 K for PbZrO$_3$ and ~360–370 K for PbTiO$_3$). There is an additional weak upturn below 1.1 K in our data, and this can be attributed to extrinsic contributions from lattice defects and/or impurities. Thus, these observations lead us to suggest that PbZrO$_3$ exhibits crystalline thermal behavior in the AFE phase. It should be noted that the accuracy of Lawless’s $C_p$ for other compounds has already been questioned.

In summary, we have examined the high-$T$ $\kappa$ for BaTiO$_3$, PbTiO$_3$, PbZrO$_3$, and PbHfO$_3$. In contrast, $\kappa$ in the cubic phase is found between the FE and AFE perovskites, where only the latter shows glasslike behavior. The AFE transition is accompanied by a change to the crystalline thermal behavior, which is also seen in the low-$T$ $C_p$ of PbZrO$_3$. With the recent advances in first-principles $\kappa$ calculations, the present results are expected to serve as an important database for anharmonic lattice dynamical studies of these technologically important materials.

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