

Superconductivity-localization interplay and fluctuation magnetoresistance in epitaxial $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ thin films

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$\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ is a superconductor, with transition temperature $T_c = 11$ K, whose parent compound BaBiO_3 possesses a charge ordering phase and perovskite crystal structure reminiscent of the cuprates. The lack of magnetism simplifies the $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ phase diagram, making this system an ideal platform for contrasting high- T_c systems with isotropic superconductors. Here we use high-quality epitaxial thin films and magnetotransport to demonstrate superconducting fluctuations that extend well beyond T_c . For the thickest films (thickness above ~ 100 nm) this region extends to ~ 27 K, well above the bulk T_c and remarkably close to the higher T_c of $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ($T_c = 31$ K). We drive the system through a superconductor-insulator transition by decreasing thickness and find the observed T_c correlates strongly with disorder. This material manifests strong fluctuations across a wide range of thicknesses, temperatures, and disorder presenting new opportunities for understanding the precursor of superconductivity near the 2D-3D dimensionality crossover.

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In contrast to the layered cuprate superconductors, $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ (BPBO, $T_c = 11$ K) and $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ (BKBO, $T_c = 31$ K) are isotropic and nonmagnetic; however, there are still interesting similarities [1,2]. The bismuthates are complex oxides with oxygen octahedra similar to the cuprates, and the parent insulating BaBiO_3 (BBO) possesses a competing phase, a charge density wave (CDW), which is suppressed for superconducting compositions. The study of the simpler, conventional bismuthate may lead to a deeper understanding of the role of CDW physics in the more complicated cuprates. The cuprate phase diagram is characterized by numerous electronic and magnetic phases and the properties are strongly influenced by disorder [3]. In thin conventional superconductors, disorder can lead to a pseudogap reminiscent of the high- T_c cuprates, suggesting a possible connection between the layered cuprate structure and dimensionally confined conventional superconductors [4]. In superconducting BPBO single crystals, Luna *et al.* [5] found a reduction in the density of states consistent with a disorder-driven metal-insulator transition and predicted a disorder-free T_c of 17 K in the strong-coupling limit and 52 K in the weak-coupling limit for $x = 0.25$.

Here we demonstrate an extended region of positive magnetoresistance in epitaxial thin films of $\text{BaPb}_{0.75}\text{Bi}_{0.25}\text{O}_3$ that is well described by superconducting fluctuations. This fluctuation regime persists for the thickest films that are well within the 3D regime, consistent with the high disorder found in our films. Restricting film thickness causes a superconductor-to-insulator transition (SIT) that correlates with disorder. Although our results are consistent with the disorder levels found in bulk

single crystals [5], we find that the critical thickness for superconductivity depends on extrinsic factors related to the poor lattice matching of BPBO with common perovskite substrates.

The high-quality epitaxial growth of BBO-based materials presents additional challenges since $\text{B}(\text{K},\text{Pb})\text{BO}$ exhibits one of the largest lattice parameters ($a_{pc} = 4.26\text{--}4.36$ Å) among the ABO_3 perovskites. There is little understanding of how the large lattice mismatch, $>10\%$ on typical commercial perovskite substrates such as SrTiO_3 (STO), impacts the structural and electronic properties of these materials. Reports in the literature demonstrate epitaxial growth on STO [6–8], MgO ($a = 4.21$ Å) [9–11], or by using buffer layers [12]. We use recently developed LaLuO_3 (LLO, $a_{pc} = 4.187$ Å) single crystals [13] as a substrate to grow BPBO ($x = 0.25, a_{pc} = 4.29$ Å) and BBO films, demonstrating that reduction of the lattice mismatch from $\sim 10.1\%$ (STO) to $\sim 2.7\%$ (LLO) improves crystallinity, surface roughness, and superconducting transitions.

Epitaxial films of optimally doped $x = 0.25$ BPBO were grown using 90° off-axis rf-magnetron sputtering on (001) STO and (110) LLO substrates (see Supplemental Material [29] for detailed methods). Thick films grown on both substrates exhibit a room temperature resistivity of 0.8 m Ω cm, the lowest reported for BPBO with $x = 0.25$, and a slightly negative $d\rho/dT$ typical for optimally doped BPBO, as seen in Fig. 1 [7,14,15]. The thick films exhibit sharp transitions with transition widths of 0.2 K (90%–10% of normal state) and T_c (50% of normal state) of 10.9 K, comparable to bulk single crystal [16] and polycrystalline ceramics [14], and the highest reported for thin films [7]. We also find no evidence of inhomogeneous transitions as seen in bulk single

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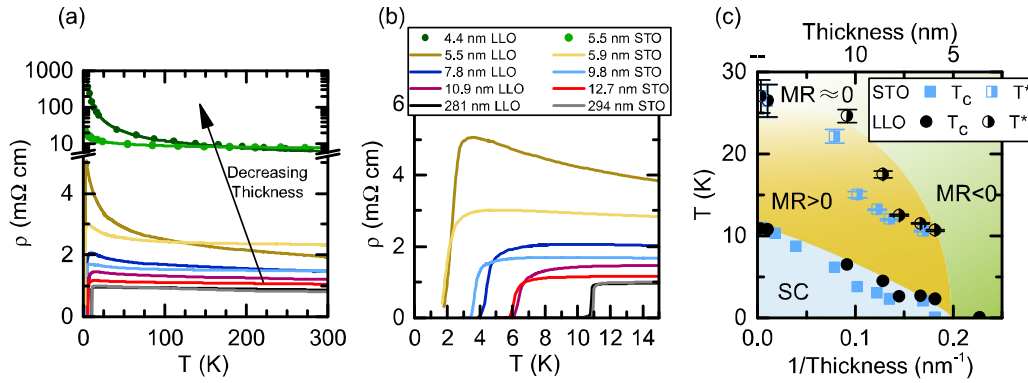


FIG. 1. Resistivity measurements from 300 K to 2 K. (a) Representative resistivity measurements for films on LLO and STO. For 4.4 nm on LLO and 5.5 nm on STO, the dots are experimental data and the solid line is a fit to a variable range hopping model. (b) Transition region for superconducting samples from (a). (c) Phase diagram of the BPBO system vs thickness for films on LLO (black circles) and STO (blue squares). The shaded regions are guides to the eye.

crystals [16]. The quality of BPBO films is further demonstrated in Figs. 2(a)–2(c) by the narrow ω -rocking curves, presence of Kiessig fringes, and out-of-plane lattice constant of 4.276 Å, in good agreement with the bulk value.

Decreasing the film thickness leads to higher resistivity, more negative $d\rho/dT$, depressed T_c , and broadened transitions as shown in Figs. 1(a) and 1(b). The thinnest films on both LLO and STO show insulating behavior well fitted by a 2D variable range hopping model ($T_0 = 1070$ K and 7.5 K, respectively) over the entire temperature range [solid lines in Fig. 1(a)]. The LLO films exhibit higher T_c when compared with films grown on STO. Figure 1(c) shows the extracted T_c (50% of normal state), showing a smaller critical thickness for films on LLO ($d_c \sim 5$ nm) than those on STO ($d_c \sim 5.5$ nm).

Previous reports of BPBO polycrystalline films found significant suppression of T_c already apparent at 200 nm, a much larger thickness than the onset of T_c reduction in our epitaxial films [17]. Grain boundaries in polycrystalline BPBO form

Josephson junctions [18,19], leading to reentrant behavior [20]. Our epitaxial films do not show reentrant behavior, and the high crystalline quality evident in the rocking curves makes the existence of a granular structure unlikely.

Disorder can be parametrized using the Mott-Ioffe-Regel parameter, k_{Fl} , determined from the normal state resistivity ρ and the Hall coefficient R_H by using the free electron formula $k_{Fl} = (3\pi^2)^{2/3} \hbar R_H^{1/3} / (\rho e^{5/3})$, which describes the limit of scattering in a system before localization occurs. In our system we find strong correlation between k_{Fl} and T_c , as shown in Fig. 3, with films on LLO showing higher k_{Fl} for a given thickness. The disorder-induced superconductor-insulator transition, controlled by a variety of parameters including thickness, has been extensively studied in conventional and high-temperature systems, with recent efforts focusing on the quantum nature of the SIT; however, no complete theoretical understanding exists for the variety of phenomena experimentally observed [20,21].

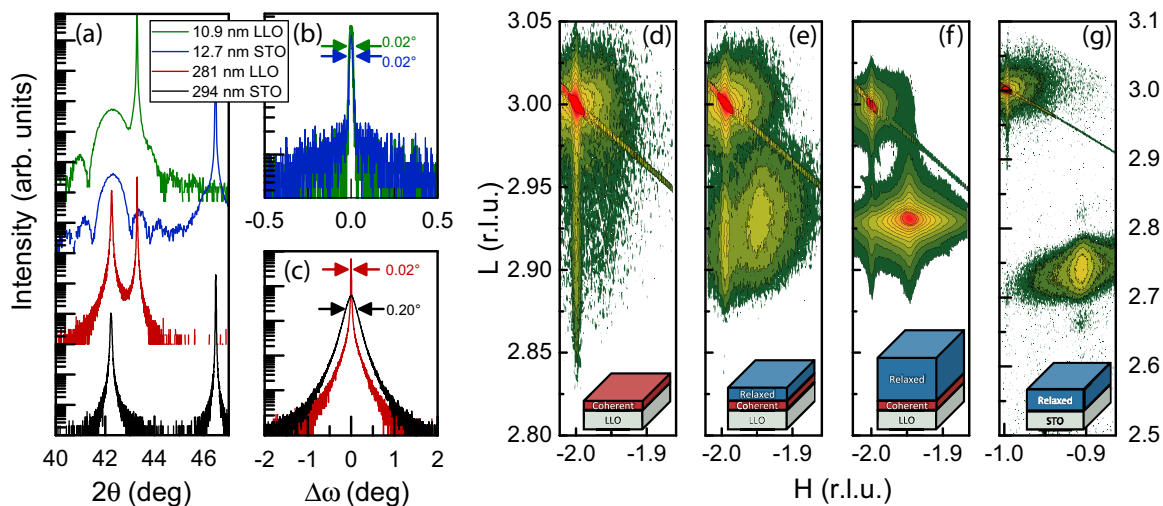


FIG. 2. X-ray diffraction characterization. (a) Representative out-of-plane 2θ - ω scans around the 002_{pc} peak for thin (~ 11 nm) and thick (~ 290 nm) BPBO films on STO and LLO. The corresponding rocking curves around the 002_{pc} film peaks for (b) thin and (c) thick films with the full width at half maximums indicated. Reciprocal space maps around the 103_{pc} reflection for (d) a fully coherent 4.4 nm thick film on LLO, as well bilayer relaxed films on LLO (e) 7.8 nm thick and (f) 281 nm thick and (g) fully relaxed 8.2 nm on STO. The inset cartoons show the film structure for each space map.

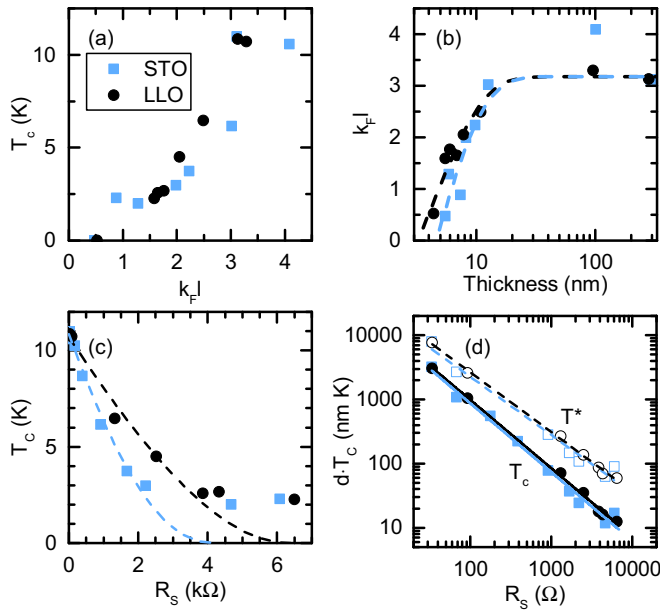


FIG. 3. Disorder and superconductivity. (a) Transition temperature vs the Mott-Ioffe-Regel parameter, $k_F l$ (extracted from room temperature measurements), and (b) $k_F l$ vs thickness (the dashed lines are guides for the eye). (c) T_c vs the sheet resistance at 20 K. The dashed lines are fits to Finkel'stein's model for a homogeneously disordered superconductor. (d) Power-law scaling dependence of thickness times transition temperature ($d \times T_c$) versus R_s based on the phenomenological formula $d \times T_c = AR_s^{-B}$, where A and B are fitting parameters [31].

For 2D homogeneously disordered films, Finkel'stein developed a model for T_c suppression from Coulomb interactions assuming no change to the bulk electron gas properties with changes to R_s [22]. The data for both sets of films initially fitted well Finkel'stein's model, Fig. 3(c), indicating an increase

in the scattering time for films on LLO compared to STO; however, the model breaks down for films close to the critical thickness. Epitaxial films can experience thickness-dependent strain relaxation, resulting in changes to material structure and properties with decreasing thickness. We investigate the strain state of our films using reciprocal space maps, shown in Figs. 2(d)–2(g). On STO, BPBO grows relaxed, as reported by other groups [23,24]. On LLO, we obtain coherent growth for films up to ~ 4.5 nm, with an abrupt relaxation at ~ 4.5 nm that forms a layered structure [see inset schema Figs. 2(d)–2(f)] that remains present for the thickest films studied. Similar relaxation behavior has been reported for other oxide epitaxial systems [25–28] and occurs in undoped BaBiO₃ films as shown in supplemental Fig. 1 of Ref. [29]. The relaxed BPBO phase on both substrates has lattice constants of $a_{pc} = 4.29 \text{ \AA}$ and $c_{pc} = 4.28 \text{ \AA}$, in good agreement with bulk values from powder diffraction [30]. Films on both substrates show variations in the out-of-plane lattice parameter at low thicknesses, see supplemental Fig. 2 in Ref. [29], indicating assumptions are likely violated for the Finkel'stein model. Both sets of films show a power-law relationship between $d \times T_c$ and R_s , shown in Fig. 3(d), an empirical observation found in many thin superconducting systems that is not well understood theoretically [31].

The x-ray diffraction in Fig. 2 reveals rocking curves with higher diffuse backgrounds and reciprocal space maps with broader in-plane components for all films grown on STO. The mosaic spread seen in the broadening of rocking curves and reciprocal space maps is indicative of a high dislocation density [32,33]. Improving the film-substrate lattice match by switching to LLO reduces the diffuse background, consistent with a reduction in dislocations and mosaicity. Additionally, surface and interface scattering become more important in thinner films and atomic force microscopy consistently reveals smoother surfaces for films grown on LLO, as shown in Fig. 4. The smaller critical thickness for superconductivity for films

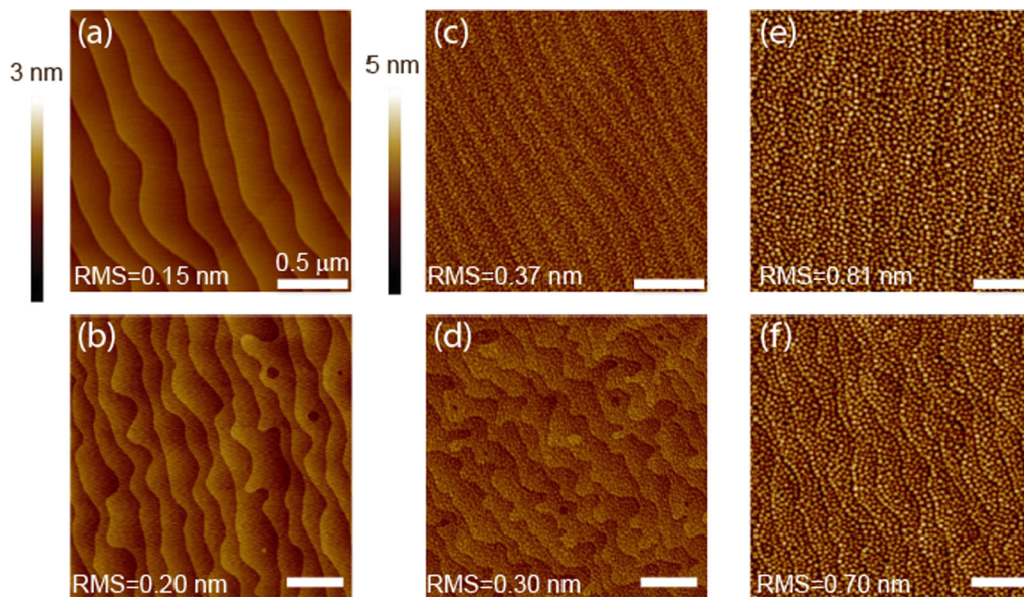


FIG. 4. Atomic force microscopy images. Surface topography images for (a) treated STO and (b) LLO, (c) 12.7 nm BPBO on STO, (d) 10.9 nm on LLO, (e) 101 nm on STO, and (f) 96.5 nm on LLO.

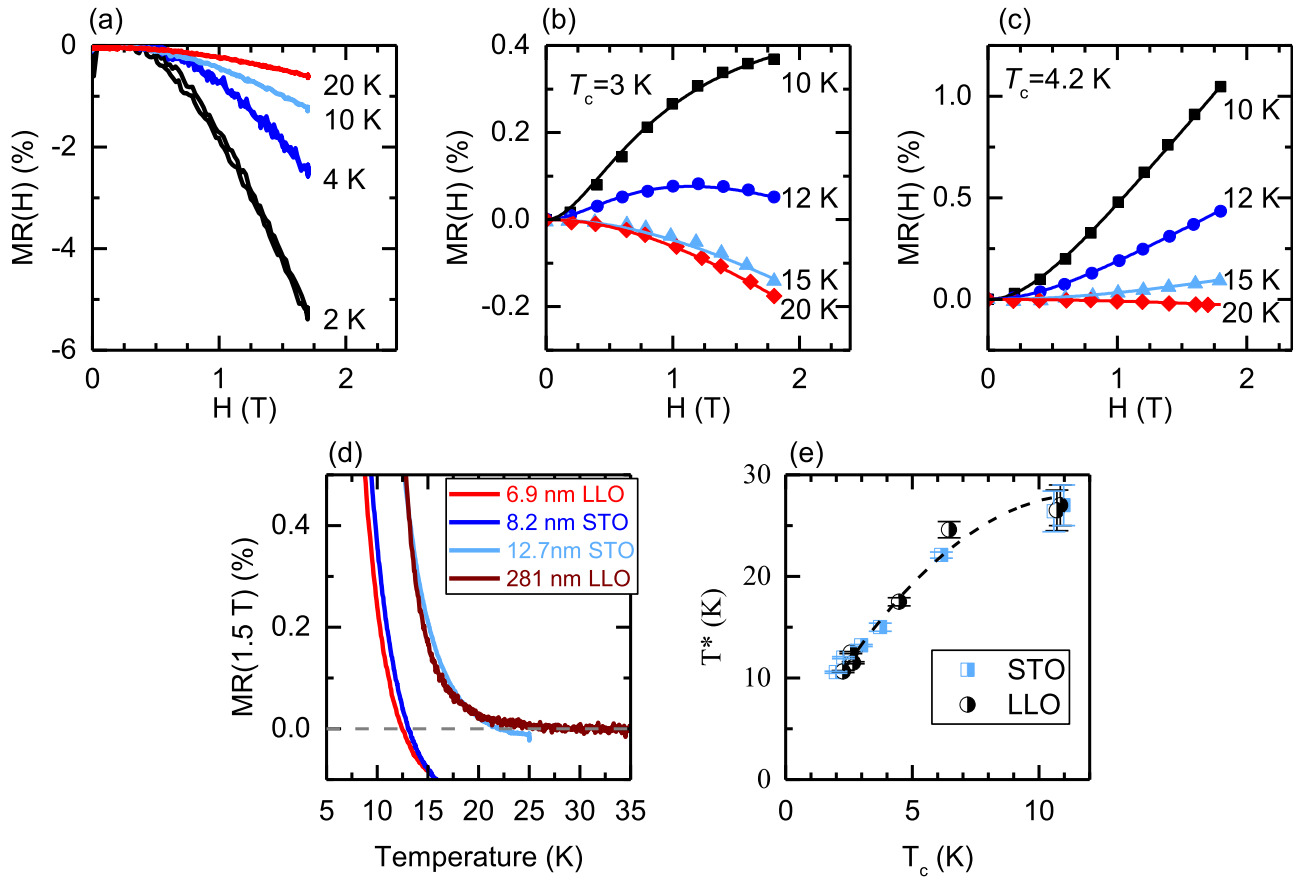


FIG. 5. Magnetoresistance measurements above T_c . Magnetoresistance at fixed temperatures for (a) insulating 5.5 nm on STO, (b) superconducting 8.2 nm BPBO on STO, and (c) superconducting 7.8 nm BPBO on LLO. In (b) and (c) the markers are experimental data points with the solid lines fits to superconducting fluctuations and weak localization in a 2D disordered system. (d) Representative magnetoresistance vs temperature. (e) The MR inflection temperature T^* vs T_c (the dashed line is a guide for the eye).

on LLO is consistent with the reduced disorder in the higher quality, smoother films [34–36].

Thin disordered superconductors routinely show evidence of superconductivity above the measured T_c [20,37]. We investigated the insulating-to-superconducting transition via magnetotransport, revealing positive magnetoresistance (MR) in all superconducting films well above T_c . Nonsuperconducting films (with thickness less than a critical thickness $d_c \sim 5\text{--}6$ nm) show only negative magnetoresistance that increases in magnitude as the temperature is lowered, as shown in Fig. 5(a) for an insulating film on STO. Magnetoresistance in the insulating phase of disordered superconductors can show a variety of responses. For instance, positive magnetoresistance reported in insulating samples is evidence of the existence of localized superconductivity persisting beyond the superconductor-insulator transition [38,39]. However, the observation of positive magnetoresistance in insulating samples is not universal even within the same material system, suggesting that localization can occur through different mechanisms of disorder resulting in distinct insulator states [40–43].

In contrast to the insulating samples, superconducting films show an extended region of positive magnetoresistance above T_c that extends to a temperature T^* that is highly correlated with T_c , as shown in Fig. 5(e), suggesting a strong link between the positive MR and superconductivity [44–47]. In

superconductors, a positive magnetoresistance above T_c is associated with the presence of superconducting fluctuations and can arise from several different mechanisms [37]. For thin ~ 8 nm films, we fit our experimental MR data to models for fluctuations and weak localization in 2D disordered systems, see the Supplemental Material [29], and find excellent agreement in the region far above the transition and at low fields, as shown in Figs. 5(b) and 5(c). We note that while we used the readily available expressions for magnetoresistance of a 2D film [37,48,49] and obtained excellent agreement with experiment, far from T_c we expect a shortened coherence length and that our films are likely in the 3D regime [50]. The theory of fluctuations for 3D systems is incomplete with no expressions for the field dependence for all terms available, in part due to difficulty in preparing metallic systems with high enough disorder for measurable fluctuations [51]. The BPBO system exhibits measurable fluctuations across a wide range of thicknesses and at temperatures easily accessed by common He cryostats, allowing comparison with theory and study of the crossover regime from 2D to 3D. Although we are beyond the strict limits of 2D fluctuation theory, the correlation between T_c and T^* [Fig. 5(e)] and the good fits of experimental data strongly suggest the positive MR originates from superconducting fluctuations. Weak antilocalization (WAL) can also lead to positive MR in systems with strong spin-orbit

coupling. Separation of the various terms that contribute to MR is difficult, but we point out the strong correlation between T_c and T^* [Fig. 5(e)], the power-law scaling of T^* in a manner similar to T_c [Fig. 3(d)], and the smooth divergence of MR as T_c is approached [Fig. 5(d)]. Further demonstration of the presence of fluctuations is beyond the scope of the present study but could be searched for in magnetic susceptibility or other above- T_c phenomena [37].

We extracted T^* for each film by fixing field and sweeping temperature, as shown in Fig. 5(d), resulting in the phase diagram shown in Fig. 1(c). The existence of fluctuations far above the observed T_c is expected for films approaching the critical thickness for superconductivity [40]; however, surprisingly, the thickest films (100–300 nm) show positive magnetoresistance extending to ~ 27 K [Fig. 5(d)], significantly higher than the bulk transition $T_{c0} = 11$ K. Clean, bulk superconductors are expected to have only a very narrow temperature regime where fluctuations manifest [37]. The superconducting coherence length of BPBO is ~ 8 nm and isotropic, placing these thick films firmly in the 3D regime [14,52]. Measurements on single crystals of BPBO reveal scaling consistent with a 2D material [15] and transmission electron microscopy measurements show some evidence of striped polymorph ordering on the scale of the coherence length [53], suggesting nanostructuring of the material could contribute to this phenomenon. We, however, have no evidence of such stripes in our films, and our measurements suggest a high level of disorder in BPBO. The Mott-Ioffe-Regel parameter, $k_F l$, extracted from room temperature measurements for the thick films is ~ 3 – 4 , consistent with bulk single crystal measurements [16,52], and in the regime of a disordered material. This finding agrees with tunneling measurements on bulk BPBO crystals that show changes to the tunneling density of states arising from disorder [5].

The role of disorder and superconducting fluctuations has interested the community, in part due to the layered structure of the high- T_c cuprates and nearby insulating phases that promote the presence of fluctuations. Similarly, thin conventional superconductors exhibit strong fluctuations, proximity to a superconductor-insulator transition, and evidence for a pseudogap in the density of states, suggesting a connection between thin conventional superconductors and the higher- T_c layered materials [4]. Disordered NbN films exhibit magnetoresistance inflection at temperatures close to the where the energy gap

from scanning tunneling spectroscopy vanishes [39]. There is one report of a pseudogap in $\text{Ba}_{0.67}\text{K}_{0.33}\text{BiO}_3$ [54] and a pseudogap has been suggested for BPBO [55]; however, experimentally the normal state properties are poorly explored in BBO superconductors.

Although the physical origin of disorder in BPBO is as of yet undetermined, there are several possibilities. Optical and terahertz spectroscopy measurements hint at local CDW fluctuations that could compete with superconductivity prior to the onset of a semiconducting band gap from a long-range CDW [56,57]. In BPBO there is also evidence that many samples consist of two structural polymorphs, tetragonal and orthorhombic, and that superconductivity is correlated with the tetragonal volume fraction [30,58]. Our measured T^* of ~ 27 K in thick films is very close to the higher T_c of $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$, and in reasonable agreement with the prediction for disorder-free BPBO from Luna *et al.* [5]. The similar temperature scales of our T^* , the disorder-free T_c prediction, and the onset of CDW fluctuations strongly suggests a connection of these phenomenon. Models of T_c suppression, such as the Finkel'stein model, do not consider effects such as a competing CDW phase. The surprisingly large temperature range with measurable fluctuation effects is consistent with BPBO possessing a significantly higher T_c that is obscured by disorder.

Our work demonstrates a smaller critical thickness for films grown on LLO; however the lattice mismatch is still large compared to other perovskite heterostructures, preventing growth of films thick enough to study epitaxial strain engineering [59,60]. Further improvements in crystalline and interface qualities are important for BBO heterostructures that could use thin layers to control the CDW [23] or interface superconducting BPBO with the predicted topological insulating phase of BBO [61]. Our results show how extrinsic contributions related to material limitations play an important role in the ultrathin limit, as well as reveal above- T_c phenomena in BPBO.

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