Incoherent Cooper Pairing and Pseudogap Behavior in Single-Layer $FeSe/SrTiO_3$

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In many unconventional superconductors, the presence of a pseudogap—a suppression in the electronic density of states extending above the critical temperature—has been a long-standing mystery. Here, we employ combined *in situ* electrical transport and angle-resolved photoemission spectroscopy measurements to reveal an unprecedentedly large pseudogap regime in single-layer $FeSe/SrTiO_3$, an interfacial superconductor where incoherent Cooper pairs are initially formed above $T_\Delta \approx 60$ K but where a zero-resistance state is achieved only below $T_0 < 30$ K. We show that this behavior is accompanied by distinct transport signatures of two-dimensional phase fluctuating superconductivity, suggesting a mixed vortex state hosting incoherent Cooper pairs which persist well above the maximum clean limit T_c of approximately 40 K. Our work establishes the critical role of reduced dimensionality in driving the complex interplay between Cooper pairing and phase coherence in two-dimensional high-T_c superconductors, providing a paradigm for understanding and engineering higher-T_c interfacial superconductors.

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I. INTRODUCTION

Single-layer FeSe grown on $SrTiO_3$ ($FeSe/SrTiO_3$) has attracted interest due to its characteristics as an atomically thin, interfacially enhanced high-T_c superconductor. $FeSe/SrTiO_3$ exhibits a spectroscopic gap-opening temperature (T_Δ) between 60 and 70 K [1–4], nearly one order of magnitude higher than that of bulk FeSe (8 K) [5] and in excess of related electron-doped FeSe-based bulk compounds (approximately 40 K) [6,7]. The combination of its high T_c, relative simplicity, and inherently two-dimensional (2D) nature positions $FeSe/SrTiO_3$ as an ideal platform for exploring the importance of superconducting fluctuations and the possibility of interfacial enhancement in high-T_c materials.

Nevertheless, significant challenges impede the systematic study of $FeSe/SrTiO_3$, as its air sensitivity, variability in the postgrowth annealing process, and potential impact of capping layers make meaningful comparisons across different techniques and studies, both *in situ* and *ex situ*, difficult [8,9]. Consequently, there remains a widely observed but heretofore unexplained discrepancy between the gap-opening temperature T_Δ observed by angle-resolved photoemission spectroscopy (ARPES) ($T_\Delta \approx 60$ K) and the temperature at which a zero-resistance state has been measured by electrical transport, T_0 ($T_0 < 30$ K) [2,10–13]. A potential resolution to this puzzle is the existence of Cooper pair fluctuations above T_c, which are known to play an important role in two-dimensional superconductors as well as underdoped cuprates but have not been widely investigated for $FeSe/SrTiO_3$.

To reveal the intrinsic nature of superconductivity and the pseudogap in $FeSe/SrTiO_3$, we employ, for the first time, a combination of ARPES and *in situ* resistivity measurements to simultaneously probe both the spectroscopic and electrical transport properties of pristine single-layer $FeSe/SrTiO_3$ samples in ultrahigh vacuum. Through a systematic investigation of a large number of such samples, we reveal the presence of intrinsic superconducting

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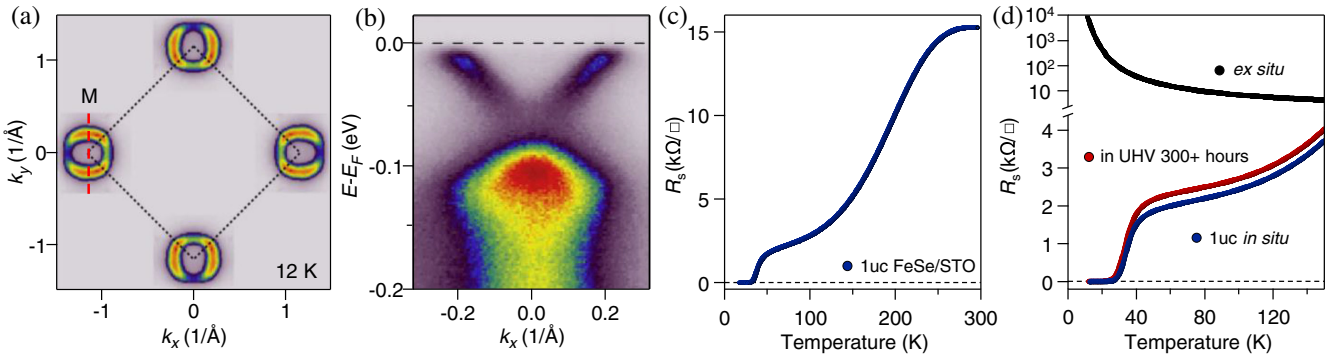

FIG. 1. Combined ARPES and *in situ* electrical resistivity measurement of single-layer $FeSe/SrTiO_3$. Measurements in (a)–(c) are conducted on the same sample. (a) Fermi surface intensity map for an as-grown 1 uc $FeSe/SrTiO_3$ sample held at 12 K, integrated over ± 5 meV of E_F. The black dashed line indicates the boundary of the 2-Fe Brillouin zone. The electron pockets comprise 5.5% of the Brillouin zone area. (b) Photoemission intensity at M [dashed red line in (a)] taken at 12 K. (c) Temperature-dependent sheet resistance for 1 uc $FeSe/SrTiO_3$. (d) Stability of the superconducting state for a separate sample after growth (blue line), long-term UHV storage (red line), and after momentary exposure to an inert gas atmosphere (black line).

fluctuations over an unprecedentedly broad temperature range, as characterized by the window between the onset of spectroscopic gap T_Δ and the onset of zero resistance T_0. This result establishes the essential role that reduced dimensionality plays in the superconductivity of $FeSe/SrTiO_3$ and resolves the long-standing confusion surrounding the critical temperature of $FeSe/SrTiO_3$.

II. RESULTS

In Fig. 1, we show combined *in situ* resistivity and ARPES measurements conducted on the same sample of single-layer $FeSe/SrTiO_3$. The Fermi surface [Fig. 1(a)] is comprised of electron pockets centered at the M point consistent with an electron doping of $0.11e^-$ per unit cell, in good agreement with earlier reports [1,3,14], and exhibits the expected spectroscopic signatures of superconductivity (a well-defined gap and band backbending). Because of photoemission matrix elements in our measurement geometry, only one band is observed in the high-statistics cut shown in Fig. 1(b), despite the expectation of two nearly degenerate elliptical pockets at M [15]. In Fig. 1(c), we show the sheet resistance $R_s(T)$, which exhibits a humplike feature at 280 K, characteristic of heavily electron-doped bulk FeSe-derived compounds [16], and a broad superconducting transition which onsets at $T_{\text{onset}} = 44 \pm 3$ K, eventually falling below 0.1% of $R_{70\text{ K}}$ at $T_0 = 29 \pm 0.2$ K. When measured *in situ*, $FeSe/SrTiO_3$ samples exhibit residual resistivity ratios (RRRs, defined as $R_{300\text{ K}}/R_{70\text{ K}}$) of approximately 10, in contrast to RRRs of approximately 1–2 for capped single-layer films reported in the literature [10]. While samples remain robust for hundreds of hours and over numerous cooling and warming cycles when maintained under ultrahigh vacuum [red curve, Fig. 1(d)], pristine films deteriorate instantaneously upon exposure to atmosphere [black curve, Fig. 1(d)].

To explore this behavior more systematically, we perform detailed temperature-dependent measurements of the energy gap $\Delta(T)$ using ARPES. In Fig. 2, we show a quantitative comparison between $\Delta(T)$ and $R_s(T)$ measurements on the same sample shown in Fig. 1. In Fig. 2(a), we plot over 100 energy distribution curves (EDCs) symmetrized about E_F from 12 to 94 K, measured at k_F of the electron pocket, where false color represents the intensity of the EDCs. In Fig. 2(b), we plot select EDCs extracted from the temperature series in Fig. 2(a). Figure 2(c) tracks Δ as a function of the temperature, defined as half the separation between quasiparticle peaks of the symmetrized EDCs from Figs. 2(a) and 2(b), as well as the evolution of the spectral gap depth δ_{SW}, defined as the difference between the coherence peak amplitude normalized to unity and the corresponding spectral weight at E_F. In Fig. 2(d), we show $R_s(T)$, as well as its derivative dR_s/dT. As the superconducting transition is broad, we define three characteristic temperatures to describe the shape of the transition: T_0, where the resistance reaches 0.1% of $R_s(70\text{ K})$; T_{onset}, the intersection between the extrapolated normal-state sheet resistance and a linear fit to the superconducting transition region; and T^*, where $R_s(T)$ exhibits an inflection point at the onset of the broad resistive rollover (as determined by a local minimum in dR_s/dT). For the sample shown in Fig. 2, $T_0 = 29 \pm 0.2$ K, while $T_{\text{onset}} = 44 \pm 3$ K, and $T^* = 72 \pm 4$ K. Deep within the superconducting state ($T < T_0$), a clear superconducting gap ($\Delta = 12.8 \pm 1$ meV) and sharp Bogoliubov quasiparticle peaks are observed in the ARPES spectra. In the broad transition region where $T_0 < T < T_{\text{onset}}$, the strength of the quasiparticle peak is gradually suppressed as the temperature increases, accompanied by a rapid filling of spectral weight within the gap [Figs. 2(a) and 2(c)], despite the energy separation between the peaks remaining largely constant. Upon

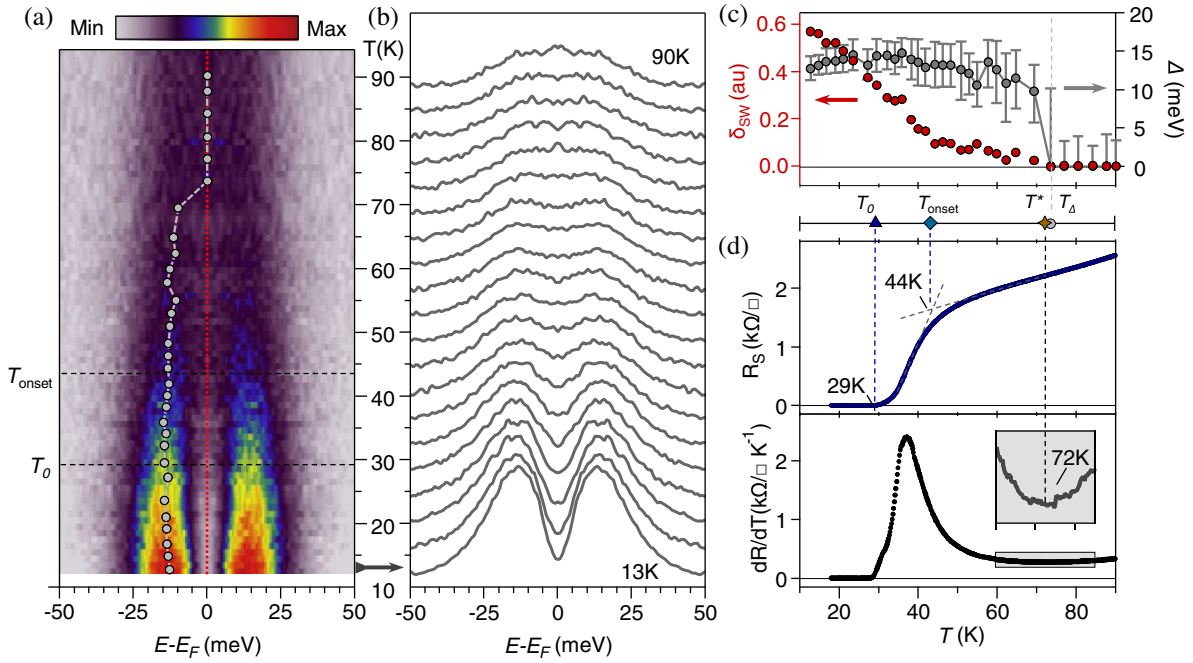

FIG. 2. Comparison of temperature-dependent gap closing behavior and corresponding resistive superconducting transition. (a) Symmetrized EDCs taken at k_F from over 100 individual spectra collected between 12 and 94 K. The color scale indicates the EDC intensity, with increasing temperature along the vertical axis. The gray circles track the quasiparticle peak position as a function of the temperature. (b) Selection of symmetrized EDCs from the data in (a). The temperature (vertical) axis is matched to that of (a). (c) Extracted energy gap Δ (right axis) as a function of the temperature from the data in (a). Δ is defined as half of the separation between the EDC peak positions indicated in (a). The red symbols (left axis) track the suppression spectral weight δ_{SW} at E_F [dotted line in (a)] relative to 90 K. (d) Sheet resistance (blue line, top) and dR/dT (black line, bottom) as a function of the temperature measured *in situ* for the same sample presented in (a) and (b).

increasing the temperature further ($T_{\rm onset} < T < T^*$), the energy gap continues to fill in at a more gradual rate, until eventually Δ is no longer discernible above $T_\Delta = 73 \pm 5$ K, a temperature that corresponds closely to T^*. We confirm that alternative methods for fitting the symmetrized EDCs to a model spectral function for Bogulibov quasiparticles yields comparable results for T_Δ (Supplemental Material, Sec. III [17]).

This behavior is in stark contrast to what is observed in bulk conventional superconductors, where the resistivity drops abruptly to zero at the same temperature at which the superconducting gap opens (i.e., $T_0 \approx T_{\rm onset} \equiv T_\Delta$). The most notable exception to this behavior is underdoped cuprates, where the pseudogap at the d-wave antinode measured by numerous techniques including ARPES also opens at significantly higher temperatures than the bulk T_c [18]. In contrast, in bulk Fe-based superconductors, it is widely shown that $T_{\rm onset}$ and T_Δ match closely [19,20], including in electron-doped bulk FeSe-based compounds such as $A_x{\rm Fe}_2{\rm Se}_2$ ($A = $ K, Cs) and $({\rm Li}_{1-x}{\rm Fe}_x){\rm OHFeSe}$ [6,7]. Thus, the observed discrepancy in ${\rm FeSe/SrTiO_3}$ cannot be solely attributed to the unconventional nature of Fe-based superconductivity. Furthermore, by using spatially resolved ARPES measurements with a 100-μm-diameter beam spot, we observe that Δ is largely uniform across the entire sample, ruling out percolation or spatial variations as the reason for the discrepancy between T_0 and $T_{\rm onset}$ and T_Δ (Appendix B).

On the other hand, such behavior is expected in 2D superconductors which can exhibit a broad Berezinskii-Kosterlitz-Thouless (BKT) transition [21], where vortex-antivortex fluctuations prevent long-range phase coherence at temperatures well above where a zero-resistance state is finally achieved ($T_{\rm BKT}$). BKT transitions have been extensively studied in disordered 2D superconductors as well as more recently in atomically thin crystalline superconductors or interfaces such as ${\rm LaAlO_3/SrTiO_3}$ [22] and twisted bilayer graphene [23]. Probes such as ARPES or tunneling spectroscopy detect the initial formation of pairs but are not sensitive to their phase coherence, so a spectroscopic gap can be found to open at temperatures well above a broad resistive transition ($T_\Delta > T_{\rm BKT}$). Recently, combined tunneling and transport measurements of disordered ultrathin films of the BCS superconductors TiN [24] and NbN [25] have verified such a picture.

To quantitatively investigate the possibility of BKT phase fluctuations in ${\rm FeSe/SrTiO_3}$, we show $V(I)$ characteristics from the ${\rm FeSe/SrTiO_3}$ films in Fig. 3 on a log-log scale, measured from 24 to 37 K. The slopes of the curves in Fig. 3(a) indicate the power-law exponent α at low

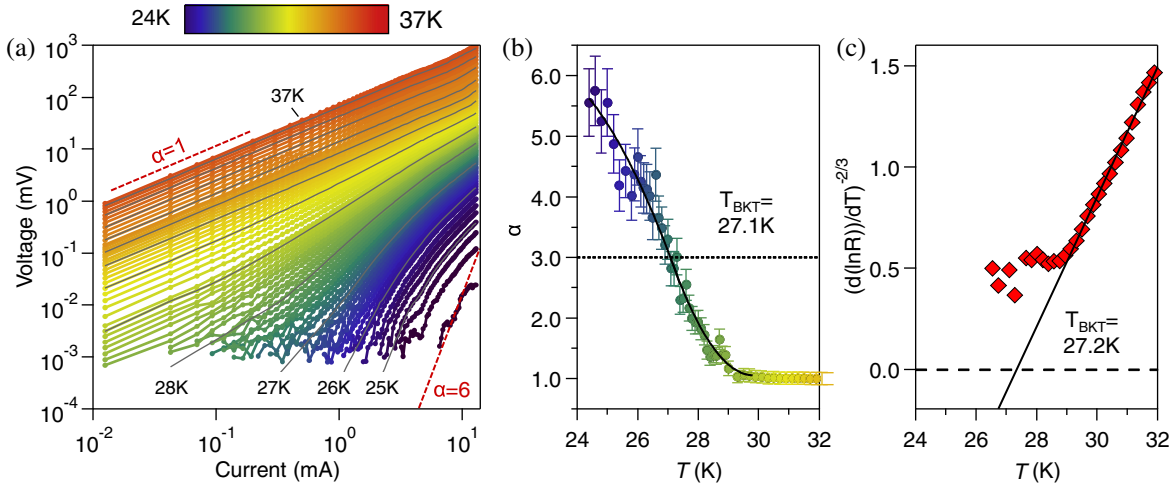

FIG. 3. *In situ* $V(I)$ characteristics and $R(T)$ analysis. (a) Current-voltage characteristics for 1-uc-thick $FeSe/SrTiO_3$, shown on log-log scale. Color indicates the temperature for a given $V-I$ curve, where the gray lines indicate temperature spacings of 1 K. Red dashed lines indicate power-law behavior for $V \propto I^{\alpha}$. (b) Extracted temperature dependence of α from power-law fits to the data in (a). (c) $[d(\ln R)/dT]^{2/3}$ plotted versus T. The solid black line indicates behavior consistent with $T_{\mathrm{BKT}} = 27.2$ K.

voltages for $V(I) \propto I^{\alpha}$ [Fig. 3(b)]. As expected for a BKT-like transition, the values of α are highly temperature dependent, crossing $\alpha = 3$ at $T_{\mathrm{BKT}} = 27.1 \pm 0.5$ K. A plot of $[d(\ln R)/dT]^{2/3}$ [Fig. 3(c)] matching the Halperin-Nelson form of $R_s(T)$ [26] yields a value of $T_{\mathrm{BKT}} = 27.2 \pm 0.5$ K, in agreement with T_{BKT} extracted from the critical exponent analysis. Signatures of a BKT transition are also reported in *ex situ* measurements of capped $FeSe/SrTiO_3$ thin films, albeit with lower values of T_{onset} and T_{BKT} [11].

Since T_0 in 2D superconductors can be strongly influenced by disorder, we systematically investigate a large number of samples with varying degrees of disorder, using the extrapolated residual sheet resistivity R_0 as a metric, and controlled primarily through the postgrowth annealing process [27]. A comparison with ARPES data shows close correspondence between R_0 and increased quasiparticle broadening, consistent with sample-to-sample variation in the disorder strength (Appendix C). At the limit where films become insulating, distinct quasiparticle peaks vanish entirely, and the spectral weight at E_F is strongly suppressed. In Fig. 4(a), we show $R_s(T)$ for a selection of single-layer $FeSe/SrTiO_3$ films, which clearly demonstrates the obvious dependence of T_0 and T_{onset} on R_0. Figure 4(c) summarizes all samples measured in this study, with values of T_0, T_{onset}, T_{Δ}, and T^* extracted from additional samples following the conventions in Fig. 2 (Supplemental Material, Sec. IV [17]). As shown, T_0 decreases linearly with increasing R_0, approaching 40 K in the clean limit. The crossover from a superconducting to insulating regime occurs around $R_0 \approx 7.2$ kΩ, close to the quantum of resistance for pairs, $R_Q = h/(2e)^2$, as would be expected for a 2D superconductor limited by phase fluctuations [28]. The importance of disorder on 2D phase fluctuations naturally explains the wide variation in T_0 and T_{onset} values [2,10–13] previously reported in the literature from capped films [Fig. 4(b)]. The highest values of $T_{\mathrm{onset}} \approx 45$ K reported here on pristine films are slightly higher than the maximum T_{onset} observed in capped films from the literature (approximately 40 K) and are inconsistent with the singular report of $T_c > 100$ K by Ge *et al.* [29].

In contrast to T_0, both T_{Δ} and T^* show relatively little dependence on disorder [Fig. 4(c)], with the values of T_{Δ} reported here generally consistent with the values extracted from the literature using the same analysis method for our own data [Fig. 4(b), gray symbols] [1,3,4,14,30–32]. The close correspondence of T_{Δ} and T^* strongly suggests that the beginning of the resistive transition at T^* is directly related to the appearance of Cooper pairs below T_{Δ}. This incoherent Cooper pairing persists within a high-temperature pseudogap regime ($T_{\mathrm{onset}} < T < T_{\Delta}$) well above the temperature range where 2D BKT-like phase fluctuations are clearly observed ($T < 40$ K).

III. DISCUSSION AND CONCLUSIONS

Taken together, these measurements present, for the first time, a self-consistent picture for the previously mysterious superconducting behavior of $FeSe/SrTiO_3$. At low temperatures ($T < T_0$), the influence of phase fluctuations is minimal, resulting in sharp Bogoliubov quasiparticle peaks and a zero-resistance state. As the temperature is increased, the zero-resistance state is destroyed by a BKT-like vortex-unbinding transition, at a temperature dependent on the level of disorder, while spectral weight begins to fill within the gap. Since T_0 should asymptote to T_c in the clean limit for a 2D superconductor [33], the trend in T_0 demonstrated in Fig. 4(c) suggests that maximum intrinsic T_c of $FeSe/SrTiO_3$ is approximately 40 K, when accounting for disorder and phase fluctuations [Fig. 4(c)], comparable

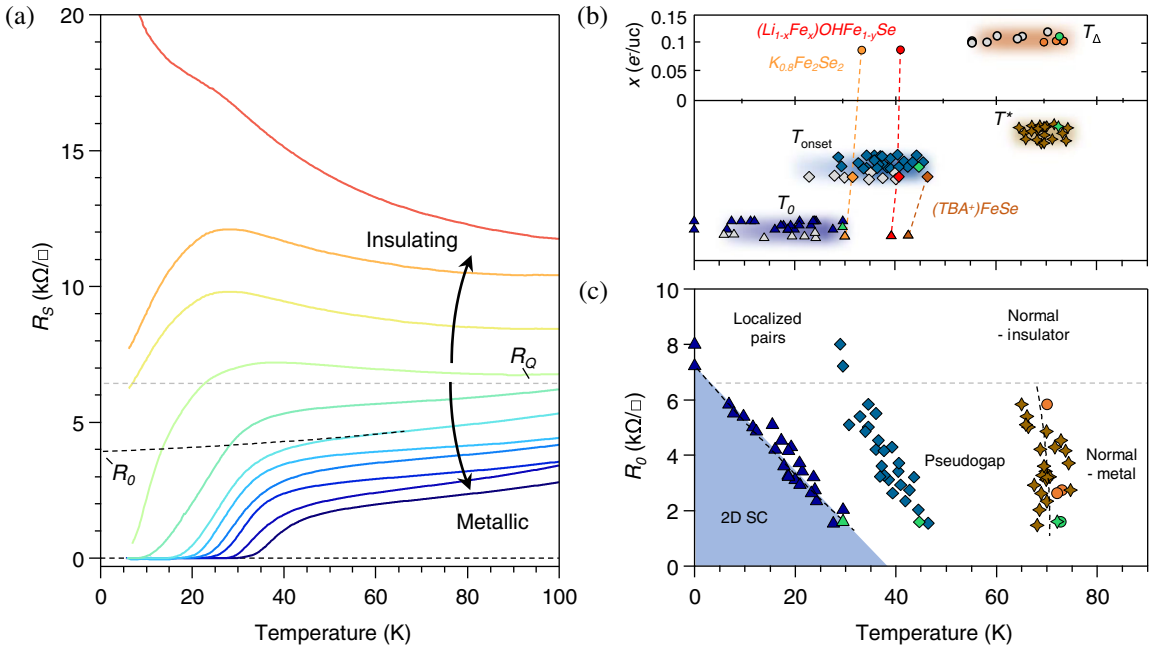

FIG. 4. Disorder-driven superconductor-insulator transition in single-layer FeSe/$SrTiO_3$. (a) R_s for a selection of single-layer FeSe/$SrTiO_3$ films. The residual resistance R_0 for each curve is determined by an extrapolation of the normal state data above T^* (black dashed line). (b) Extracted characteristic temperatures of single-layer FeSe/$SrTiO_3$. Marked symbols indicate T_0 (triangles), T_{onset} (diamonds), T^* (stars), and T_Δ (circles) versus x (Luttinger volume) as measured *in situ* by this study (solid colored symbols) and from the literature (gray filled symbols). T_0, T_{onset}, and T^* values are offset along the Y axis for clarity. Literature data for T_0 and T_{onset} are taken from capped single-layer FeSe/$SrTiO_3$ films. Bright green symbols indicate values for the sample presented in Figs. 1–3. ARPES gap values and doping levels from the literature are reported from Refs. [1,3,4,14,30–32]. *Ex situ* transport data from Refs. [2,10–13]. T_0, T_{onset}, and T_Δ values are also plotted for bulk compounds $K_{0.8}Fe_2Se_2$ (orange, solid line), $(Li_{1-x}Fe_x)$OHFeSe (red, solid line), and (TBA+)FeSe (brown, solid line). (c) R_0 versus T_0, T_{onset}, T^*, and T_Δ for films measured *in situ* for this work. Extended data for T_Δ and T^* are provided in Supplemental Material, Sec. IV [17].

to typical values of T_c for bulk electron-doped FeSe-based compounds [Fig. 4(b)] such as $(Li_{1-x}Fe_x)$OHFeSe [7] but well short of the 60–70 K T_c previously interpreted from spectroscopic results alone.

Finally, we speculate on the high-temperature pseudogap regime for FeSe/$SrTiO_3$ ($T_{onset} < T < T_\Delta \approx 60$–70 K), when compared to bulk electron-doped FeSe-based materials which do not exhibit a pseudogap and show $T_\Delta \approx T_c \approx 40$ K. One possibility is that Gaussian fluctuations above T_c account for the behavior observed in the high-temperature pseudogap regime. However, this scenario is contradicted by the observed behavior of $\Delta(T)$, which shows no evidence of closing near 40 K, as well as by the shape of $R_s(T)$, which is poorly reproduced by the Aslamazov-Larkin framework (Supplemental Material, Sec. II [17]). Instead, this behavior suggests that the microscopic, mean-field pairing temperature of FeSe/$SrTiO_3$ is intrinsically higher than that of bulk FeSe-based compounds, even if the ultimate maximum T_c set by the onset of phase coherence (approximately 40 K) for FeSe/$SrTiO_3$ is comparable to those of bulk compounds. Much speculation focuses on the possible influence of interfacial electron-phonon coupling from the $SrTiO_3$ substrate in enhancing the T_c [3,4]. Alternatively, recent work on the highly two-dimensional bulk compound (TBA^+)FeSe, where the distance between FeSe layers is expanded to 15.5 Å by intercalation of ion tetrabutyl ammonium organic molecules [compared to 5.5 Å for bulk FeSe or 9.32 Å for $(Li_{1-x}Fe_x)$OHFeSe], also reports evidence of incoherent preformed pairing up to 60 K, comparable to FeSe/$SrTiO_3$, but in the absence of any substrate [34]. That similar pseudogap features are also observed in the more two-dimensional (TBA^+)FeSe suggests that the increased two-dimensional nature of the electronic or crystal structure could potentially be the origin of the enhanced mean-field pairing temperature T_Δ in FeSe/$SrTiO_3$. While it is empirically known that a two-dimensional electronic structure appears to be a key ingredient for unconventional high-temperature superconductivity (e.g., cuprates, Fe-based superconductors, and nickelates), most Fe-based superconductors exhibit some degree of three-dimensionality in their electronic structure, as evidenced by k_z dispersion in ARPES [35], as well as their resistivity anisotropy ρ_c/ρ_{ab} being in the range of 2–3 for the 11 and 111 families or up to 10^2 for the 122 compounds [36–38]. This behavior is in contrast to their more two-dimensional, higher T_c cuprate analogues such as $Bi_2Sr_2CaCu_2O_{8+\delta}$, $YBa_2Cu_3O_{7-\delta}$, or $La_{2-x}Sr_xCuO_4$,

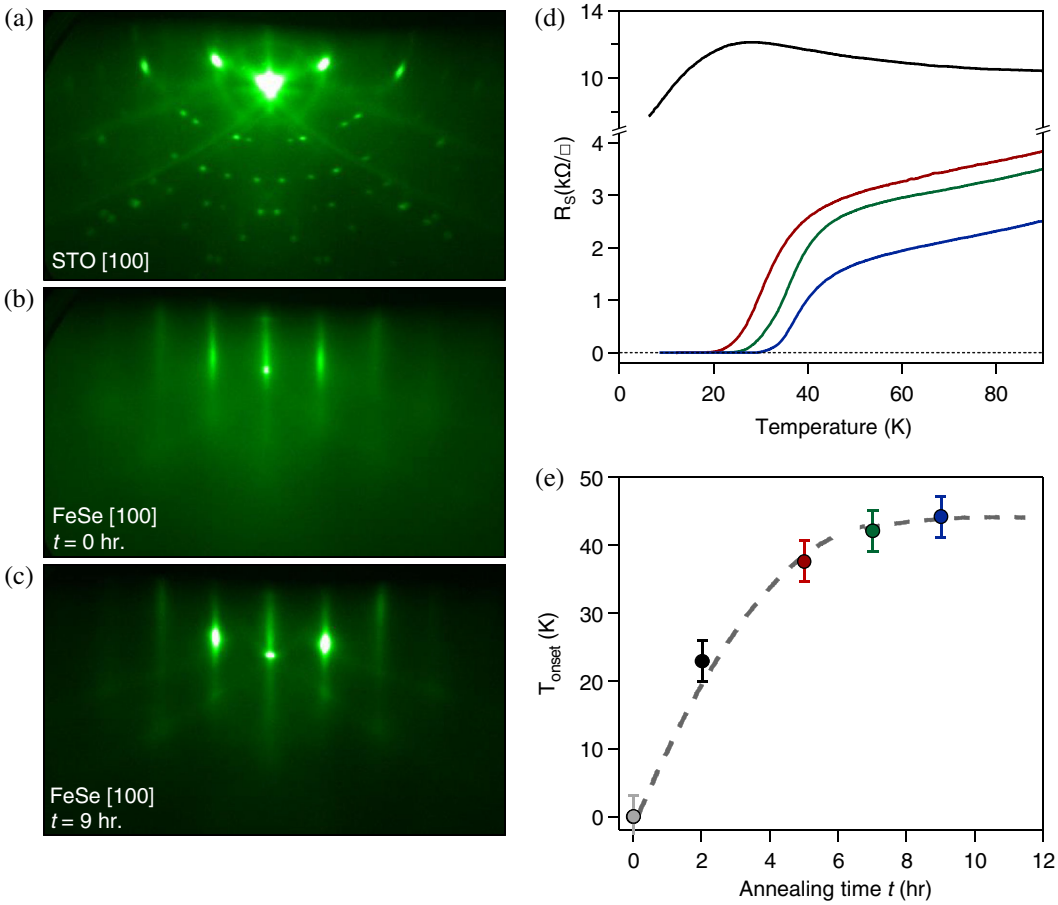

FIG. 5. RHEED images along the crystallographic [100] direction for (a) $SrTiO_3$ substrate prior to film deposition, (b) single-layer FeSe film immediately after growth, and (c) single-layer FeSe upon reaching maximum T_c following 9 h accumulated annealing time at 450 °C. (d) *In situ* resistivity data as a function of accumulated annealing time t. (e) T_{onset} as a function of cumulative annealing time t.

where ρ_c/ρ_{ab} is much larger, in the range of 10^3–10^6 [39]. By pushing Fe-based superconductors closer to the idealized two-dimensional limit as in (TBA+)FeSe ($\rho_c/\rho_{ab} \approx 10^5$) or in the ultimate case of single-layer $FeSe/SrTiO_3$, it is possible that the strength of the microscopic pairing is increased but at the cost of 2D phase fluctuations and enhanced sensitivity to disorder which limit T_0.

IV. METHODS

Single-layer $FeSe/SrTiO_3$ films are synthesized on $SrTiO_3$ (001) substrates using a chalcogenide molecular beam epitaxy system as reported previously [3]. Se (99.999% purity) and Fe (99.995% purity) are coevaporated at a nominal flux ratio of 5∶1 and at a nominal growth rate of 1.8–2 unit cells (uc) per minute, with source fluxes calibrated by a quartz crystal monitor and film crystallinity monitored in real time using reflection high-energy electron diffraction [RHEED, Figs. 5(a)–5(c)]. To enable reliable resistivity measurements of the FeSe monolayer, we utilize undoped insulating $SrTiO_3$ for all films presented in this work. After growth films are progressively annealed until optimal superconducting properties are achieved [Figs. 5(d) and 5(e)], followed by deposition of 20-nm-thick Au electrodes at the sample corners using a shadow mask to provide reliable four-point electrical contact (Fig. S3 [17]).

In situ resistivity measurements are performed using a custom-built UHV four-point transport probe with a base temperature of 5.2 K and a base pressure of 7×10^{-11} Torr. Contact is applied directly to the film using a set of Au-plated spring-loaded probes in a van der Pauw geometry, with a nominal instrumental contact spacing of 7 mm. Resistance measurements are taken using a Keithley 6221/2182A current source–voltmeter combination in delta mode (Fig. S2 [17]) with a typical applied current of $1 - 10\ \mu$A.

ARPES measurements are taken with a VG Scienta R4000 electron analyzer equipped with a VUV5000 helium discharge lamp using He-I photons at 21.2 eV. The base pressure in the ARPES system is 5×10^{-11} Torr. The energy resolution is nominally set at 12 meV for mapping and 9 meV for gap measurements. To avoid sample charging during ARPES measurement, the film is grounded

using a retractable contact pin built onto the sample manipulator [Fig. S3(a) [17]]. For gap measurements, the Fermi level is referenced to the measured Fermi edge of the Au electrodes [Fig. S3(c) [17]].

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APPENDIX A: FILM SYNTHESIS AND OPTIMIZATION OF SUPERCONDUCTING PROPERTIES

An additional postgrowth annealing step is known to be critical for producing superconductivity in ultrathin $FeSe/SrTiO_3$ films. This postgrowth annealing serves several purposes. First, it removes excess Se from the film present due to the adsorption-control growth regime, improving the stoichiometry [40,41]. Superconductivity in both FeSe films and crystals is known to be highly sensitive to nonstoichiometry [42]. Additionally, the annealing reduces disorder in the form of Fe-vacancy defects [27], increasing the electron mean free path and reducing the sheet resistance, macroscopically.

In Fig. 5, we present RHEED and *in situ* transport characteristics for a representative single-layer $FeSe/SrTiO_3$ film both before growth and afterward as it is progressively annealed to achieve optimal superconducting properties. Prior to film growth, undoped $SrTiO_3$ substrates (10 mm × 10 mm, Shinkosha) are annealed at 600 °C for 3 h and then cooled to 420 °C for deposition. $SrTiO_3$ substrates prepared in this fashion typically exhibit a clear $\sqrt{13} \times \sqrt{13}$ surface reconstruction, indicating the presence of a TiO_2 double-layer structure at the substrate surface [12,43]. Upon initial growth, the film exhibits weak RHEED spots and strongly insulating low-temperature transport behavior. With progressive annealing at 450 °C postgrowth, the film eventually becomes metallic and superconducting, reaching an optimal T_c (in this case) after 9 h. The RHEED pattern for

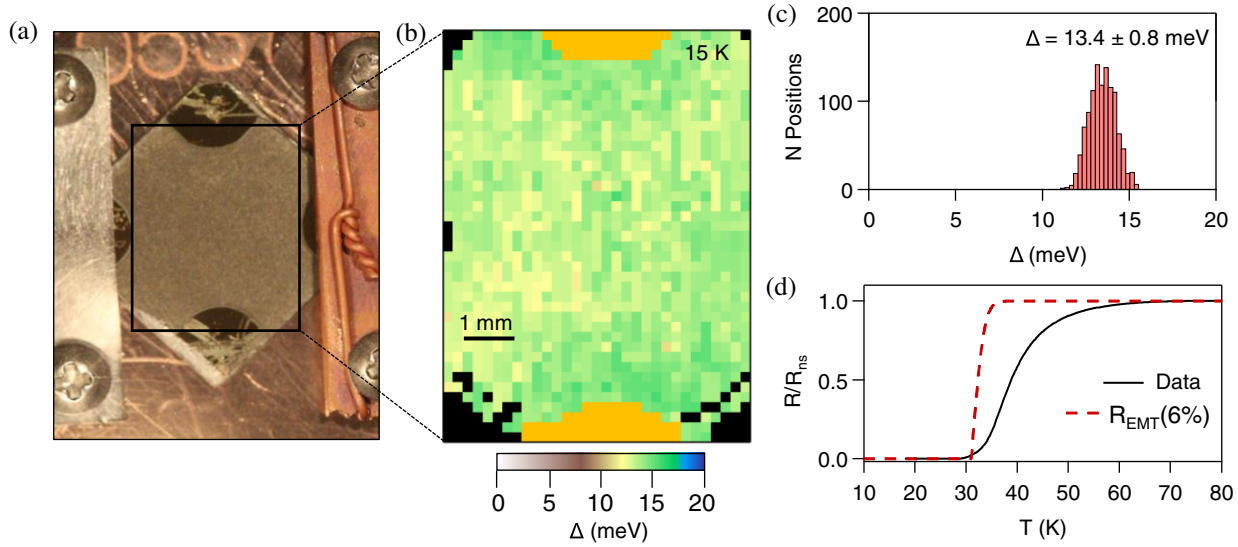

FIG. 6. Spatially resolved ARPES measurements of the superconducting gap. (a) Camera image of the single-layer $FeSe/SrTiO_3$ sample as mounted for beam line ARPES measurements. (b) Spatially resolved gap distribution measured across the sample surface of the region indicated in (a). The region probed is 8 mm × 6 mm. The gold-colored regions indicate areas with Au electrodes, and the black indicate regions where no FeSe signal is observed. (c) Histogram showing the statistical distribution of Δ_e from the data in (b). The local gap values form a Gaussian-like distribution with a mean of 13.4 meV and a standard deviation of 0.8 meV. (d) $R(T)$ data normalized to the extrapolated normal state resistance (black line) compared to expected behavior for a percolative superconducting transition (dashed red line) assuming a local T_c distribution matching the data in (c).

optimally annealed films shows sharp, well-defined spots and distinct Kikuchi lines, indicating an atomically flat surface with improved crystallinity. This behavior is consistently observed across all films prepared for this study, although the optimal annealing time is found to vary somewhat across films, falling within the range of 5–12 h typically.

APPENDIX B: EVALUATION OF INHOMOGENEITY EFFECTS

Because our measurements probe a macroscopic average of the film, the discrepancy in transport and ARPES results as well as the broad resistive transitions could conceivably be explained by gross sample inhomogeneity. To rule this possibility out, we perform spatially resolved measurements of the same single-layer $FeSe/SrTiO_3$ sample presented in Figs. 1–3, transported under vacuum to beam line 7.0.2 (MAESTRO) of the Advanced Light Source at Lawrence Berkeley National Laboratory. For beam line ARPES measurements, we set the photon energy and polarization to 24 eV and p polarization, respectively, and fix the beam diameter to a 100-μm spot for spatially resolved measurements. Figure 6(b) shows the spatially resolved distribution of Δ across an 8 mm × 6 mm region of the film, as measured at 15 K. All regions of the film show single-layer FeSe band structure, except for the

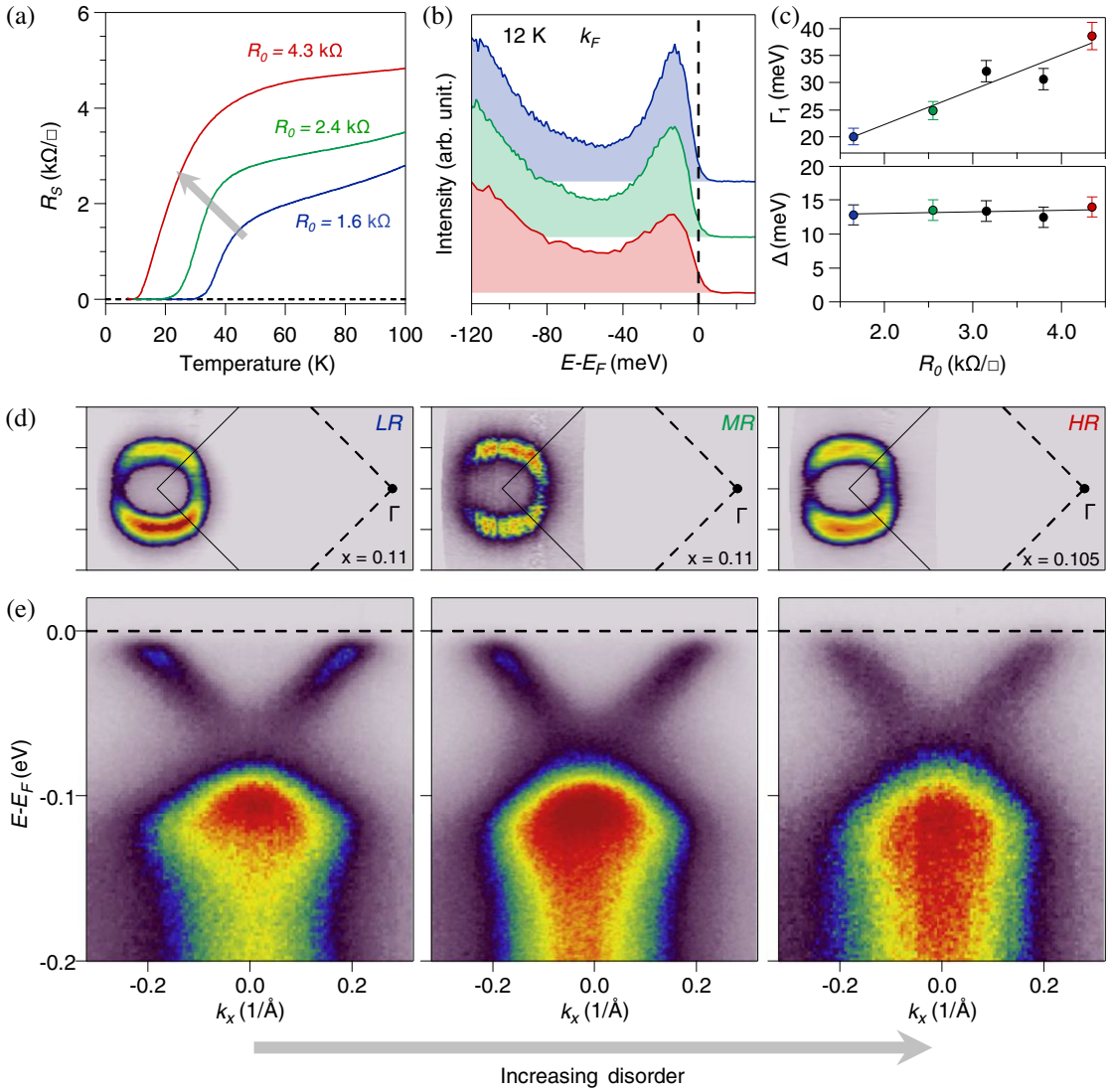

FIG. 7. Comparison of the resistive transition and ARPES band structure for films at different annealing conditions. (a) *In situ* $R_s(T)$ measurements show that the onset of zero resistance is suppressed to lower temperatures in more resistive films. (b) Comparison of raw EDCs at k_F for the same films measured in (a). (c) Comparison of quasiparticle half-width Γ_1 (top) and Δ (bottom) against residual resistance R_0. Black markers indicate data for samples also measured by ARPES and *in situ* resistivity at low temperature but not presented in (a),(b). (d) Fermi surface maps and (e) high statistics cuts at M, showing minimal variation in the doping content x based on Luttinger volume.

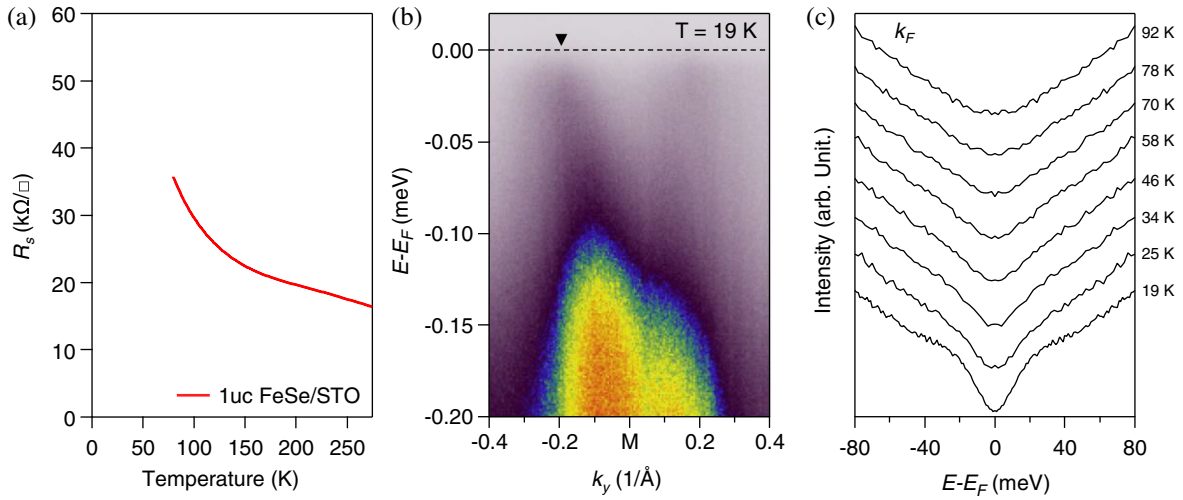

FIG. 8. ARPES behavior of insulating single-layer $FeSe/SrTiO_3$. (a) *In situ* $R_s(T)$ data for an insulating single-layer $FeSe/SrTiO_3$ film. (b) ARPES spectra taken at M at $T = 19$ K. (c) Temperature-dependent symmetrized EDCs at k_F as indicated by the arrow in (b).

corners which host Au electrodes. In particular, we observe no multilayer band structure or void regions. Therefore, we conclude that the film is indeed a macroscopically homogeneous monolayer. The measurable variation in superconducting gap at 15 K follows a normal distribution, with $\Delta = 13.4$ meV and $\sigma_\Delta = 0.82$ meV [Fig. 6(c)]. We observe no FeSe regions over which a superconducting gap is not present.

To show that this level of inhomogeneity cannot account for our broad resistive transition, in Fig. 6(d) we simulate the expected behavior of $R(T)$, assuming a percolative network with a T_c distribution matching the ARPES gap data from Fig. 6(a), based on the predictions of effective medium theory [44]. The simulated transition (red dashed curve) is far too narrow to account for the broadness of the transition that we observe by *in situ* measurement (solid black curve). Therefore, spatial inhomogeneities cannot explain the discrepancy between the temperature dependence of the superconducting gap by ARPES and our electrical resistivity measurements.

APPENDIX C: INFLUENCE OF DISORDER AND DOPING VARIATION ON ARPES AND TRANSPORT BEHAVIOR

The wide variation in T_0 observed across samples raises natural questions about what drives the suppression of superconductivity in monolayer $FeSe/SrTiO_3$ films. One possibility is that natural variation in the interfacial charge transfer from the $SrTiO_3$ interface leads to variation in the electron doping x across samples. To rule out this scenario, in Fig. 7 we compare ARPES and transport behavior across a series of films with substantially different values of T_0. Figure 7(a) shows low-temperature $R_s(T)$ behavior for three separate samples labeled as LR (low-resistance), MR (medium-resistance), and HR (high-resistance), with T_0's that span the observed range for superconducting films presented in Fig. 4(c). LR is identical to the sample presented in Figs. 1–3 of the main text. The residual resistance R_0 in each case is determined by extrapolating the high-temperature $R(T)$ behavior to 0 K and is found to be 1.6, 2.4, and 4.3 kΩ for samples LR, MR, and HR, respectively. Figure 7(b) shows corresponding EDCs at k_F extracted from the ARPES spectra on the same samples [Fig. 7(e)], and Fig. 7(c) tracks the extracted scattering rate Γ_1 (top) and low-temperature gap magnitude (bottom) versus R_0 for all films for which ARPES and *in situ* transport data are available. Despite the substantial variation in T_0, both the gap magnitude [Fig. 7(c)] and doping level (as deduced from the Luttinger volume [Fig. 7(d)]) are highly consistent across films, ruling out irregular charge transfer as the cause of the variation in the resistive behavior.

Figure 8 shows combined *in situ* resistivity and ARPES data for an even more disordered single-layer $FeSe/SrTiO_3$ film, which is "insulating" (negative dR/dT) at low temperatures [Fig. 8(a)]. The main difference between this sample and the superconducting samples is the lack of distinct quasiparticle peaks, coincident with a significant suppression of the weight near E_F [Fig. 8(c)].

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