

Disorder-Induced Suppression of Superconductivity in Infinite-Layer Nickelates

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The pairing symmetry of superconducting infinite-layer nickelates is a fundamental yet experimentally challenging question. We employ high-energy electron irradiation to induce disorder in superconducting $\text{Nd}_{0.825}\text{Sr}_{0.175}\text{NiO}_2$ thin films, examine the impact of pair-breaking defects on superconductivity, and elucidate the nature of the superconducting gap. Our measurements reveal a complete suppression of superconductivity with increasing disorder, suggesting an unconventional, sign-changing order parameter.

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Superconductivity in hole-doped infinite-layer rare-earth nickelates was realized two decades after their theoretical prediction [1] through a combination of crystalline thin-film synthesis with topotactic chemical reduction to stabilize the requisite nickel $3d^9$ valence [2], providing an experimental platform to explore a close electronic and structural analog to high- T_c cuprates [3,4]. Despite the similarities between the phase diagrams of the Ni- and Cu-based systems, several key distinctions between their electronic landscapes and hybridization have been identified [5–7].

One fundamental and outstanding question is the symmetry of the superconducting order parameter in infinite-layer nickelates. Experimentally, this has remained challenging to address, hindered largely by the thin-film geometry and sample synthesis. An early single-particle tunneling experiment, for example, found both s -wave and d -wave-like gap structures across a single film, suggesting limitations from sample surface quality [8]. Temperature-dependent measurements of superfluid density by mutual inductance [9] and tunnel-diode-oscillator [10] approaches

have so far led to differing interpretations, particularly complicated by the low-temperature behavior in the Nd-based nickelates. Recent optical measurements by terahertz spectroscopy exhibit responses which could be attributed to d -wave pairing [11]. Other conventional experimental probes of pairing symmetry such as thermal conductivity and specific heat are generally not compatible with a very thin (< 10 nm) film geometry [12].

Alternatively, the dependence of superconductivity on local nonmagnetic pair-breaking disorder can be an effective way to distinguish between nodal and fully gapped systems following the formulation of Abrikosov and Gor'kov [13–15]. In some materials, nonmagnetic chemical impurities can be introduced during synthesis as a way to tune disorder [16–19]. Given the myriad synthetic challenges already posed by the infinite-layer nickelates, however, a more practical approach in this system is postsynthetic introduction of pair-breaking disorder into the highest-quality superconducting films.

Here, we harness high-energy electron irradiation to controllably induce pointlike disorder in optimally hole-doped superconducting $\text{Nd}_{0.825}\text{Sr}_{0.175}\text{NiO}_2$ (NSNO) thin films. Primary electrons with megavolt-range energies undergo nearly elastic collisions with atoms in the sample, displacing single atoms from their crystalline positions to create vacancy-interstitial pairs [20,21]. Importantly, however, the transferred energy is not sufficient for the displaced atom to create additional defects (cascade processes), such that the resulting disorder is considered to be comprised of

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single pointlike rather than extended defects [22,23] (for further discussion, see Supplemental Note A [24]). With this ability to systematically tune pointlike disorder, high-energy electron irradiation has been used as an effective platform to investigate the pairing symmetries of a wide range of superconducting compounds, such as cuprates [21,27–29], ruthenates [30], organic superconductors [31], iron pnictides [32], kagome systems [33], doped SrTiO_3 [34], and topological superconductors [35].

Our measurements show that increasing disorder systematically and reproducibly suppresses superconductivity in infinite-layer nickelates, pointing to a sign-changing superconducting order parameter. Electrical resistivity also increases with disorder in all samples, but we reveal quantitative variation in the evolution of parameters such as residual resistivity and the normal state linear-in-temperature slope, which we attribute to additional sources of scattering which cannot be fully captured by simplified considerations. These results point to unconventional nickelate superconductivity and also provide a uniquely systematic study of disorder signatures in these materials, instructive for both physical insights as well as ongoing material optimization efforts.

Thin films of infinite-layer $\text{Nd}_{0.825}\text{Sr}_{0.175}\text{NiO}_2$ were obtained by first synthesizing perovskite precursor $\text{Nd}_{0.825}\text{Sr}_{0.175}\text{NiO}_3$ thin films on $(\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{TaAlO}_6)_{0.7}$ (LSAT) (001)-oriented substrates (lattice constant = 3.868 Å) by pulsed-laser deposition using a KrF excimer laser ($\lambda = 248$ nm), followed by a topotactic reduction using CaH_2 . All films used here are ~ 5 nm thick and capped with a few unit cells of epitaxial SrTiO_3 which act as a supporting and protective layer during the apical oxygen removal process. Further details of the synthesis are discussed in [2,36–38]. Prior to electron irradiation, sample substrates were backpolished with diamond grit lapping paper to reduce the overall thickness to < 300 μm , which reduces heating effects and attenuation of the high-energy electron beam during irradiation.

Electron irradiation was performed using 2.5 MeV electrons at the SIRIUS Pelletron accelerator at Laboratoire des Solides Irradiés, École Polytechnique (Palaiseau, France). Electrons with this energy have a large penetration depth on the order of millimeters, ensuring a homogeneous and complete transmission of the beam through the entire thin-film sample [22]. During irradiation, the samples are held at ~ 22 K by a flow of liquid hydrogen within the CRYO-1 irradiation cell to immobilize irradiation-induced defects upon formation and remove the heat produced by relativistic electrons upon collision with the atoms [Fig. 1(a)]. The irradiation dose is monitored by measuring the current from transmitted electrons with a Faraday cage placed behind the sample stage.

Total irradiation doses were accumulated over multiple runs, between which samples were periodically removed to perform electronic transport measurements. It is expected

that some fraction of the defects induced by irradiation are lost (e.g., recombined through annealing) when the sample is warmed to room temperature for external measurements [22,23,30]. In our study, this annealing makes quantitative correlation between defect density and electron dose challenging, but the relative trends between irradiation-induced disorder and various properties measured by electronic transport are robust. We confirm by x-ray diffraction measurements and atomic-resolution scanning transmission electron microscopy that the overall structure and crystallinity of the infinite-layer phase are preserved following electron irradiation, finding no evidence of additional structural changes (Supplemental Notes D and E [24]).

Figures 1(b)–1(d) show the effect of accumulated electron irradiation on two superconducting NSNO thin films S1 and S2, namely, the decrease of superconducting transition temperature (T_c) and increase in resistivity (ρ) with increasing dose. Progressive measurements of temperature-dependent electronic transport of S1 and S2 are shown in Figs. 1(c) and 1(d), both illustrating a

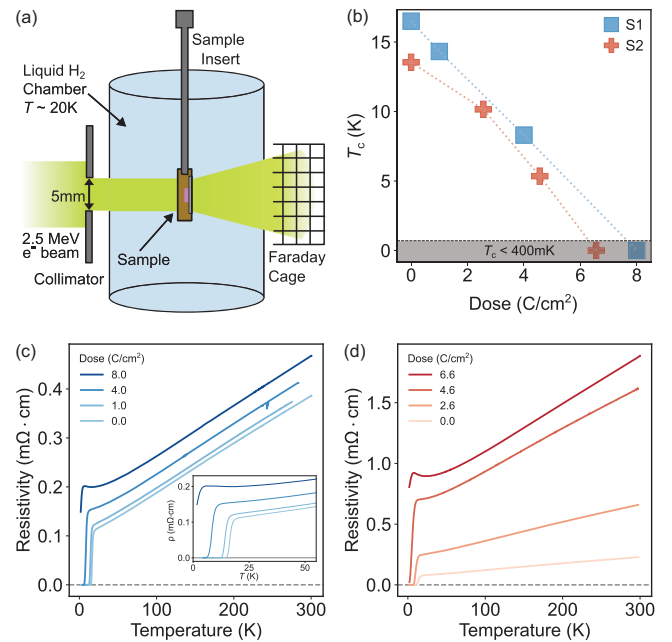


FIG. 1. (a) Schematic of the high-energy electron irradiation experiment: the sample (purple) is mounted on an insert, placed in a flowing liquid hydrogen chamber (blue), and rastered with a 2.5 MeV electron beam (green). A Faraday cage measures the transmitted current to monitor the accumulated dose. (b) Evolution of T_c with accumulated 2.5 MeV electron irradiation dose for multiple NSNO thin films. (c) Resistivity as a function of temperature for NSNO thin film S1 with electron irradiation at 2.5 MeV following progressive total doses of 0 (pristine), 1.0, 4.0, and 8.0 C/cm^2 . Inset: the superconducting transition is suppressed with increasing dose. (d) Resistivity as a function of temperature for a second NSNO thin film S2 for 2.5 MeV electron irradiation with progressive total doses of 0 (pristine), 2.6, 4.6, and 6.6 C/cm^2 .

characteristic evolution with decreasing T_c from the as-synthesized or pristine film (0 C/cm^2) across total accumulated doses up to 8 C/cm^2 . Here, we define T_c as the temperature where the resistivity falls to 50% within the downturn; details of extracting T_c and other parameters are discussed in Supplemental Fig. S2 [24].

The sharpness of the superconducting transition shows negligible changes between measurements, confirming both the pointlike nature of the induced defects and the homogeneity of their distribution across an entire thin-film sample (Supplemental Fig. S3 [24]) [22]. Similar T_c suppression is observed across several irradiated samples (Supplemental Figs. S4 and S5 [24]).

As the disorder increases with additional irradiation, low-temperature resistive upturns appear for both S1 and S2. Similar upturns were observed in electron irradiation studies of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO), where they have been discussed in the context of combined single impurity scattering and localization effects [28,39]. Low-temperature resistive upturns are also observed in as-synthesized under- and overdoped NSNO films of similar quality [37]. We note, however, that the resistive upturn we observe here is dependent on increasing disorder (similar to the pristine overdoped and distinct from pristine underdoped NSNO films) but not affected by applied magnetic fields (Supplemental Fig. S6 [24], similar to pristine underdoped and distinct from pristine overdoped films). Further investigations with more densely sampled irradiation doses may elucidate the microscopic origin of the upturn in the specific case here.

Full suppression of the superconducting state (i.e., $T_c < 400 \text{ mK}$) is achieved in both samples with the total accumulated doses of 8.0 and 6.6 C/cm^2 for S1 and S2, respectively [Fig. 1(b)]. The systematic and consistent suppression of T_c at a rate of $\sim 2 \text{ K per C/cm}^2$ irradiation across all samples [Fig. 2(a)] demonstrates this method as a reliable and controllable way of tuning pointlike disorder in these films.

Also accompanying the decrease in superconducting transition temperature is a systematic increase in resistivity

with accumulated dose, albeit at notably different rates between films [Supplemental Fig. S5(b) [24]]. The residual resistivity (resistivity just before the superconducting downturn) ρ_0 is commonly used as a convenient parameter to quantify the degree of intrinsic disorder in a system [17,19,40]. In many real materials, however, other extrinsic factors such as extended defects or epitaxial strain can also contribute to additional scattering. Here, we define the residual resistivity of pristine (preirradiated) films ρ_0^{pristine} as a baseline for all the existing disorder and/or defects present in as-synthesized samples and use the change in residual resistivity at a given dose $\Delta\rho_0 = \rho_0 - \rho_0^{\text{pristine}}$ as a proxy for the controllably induced disorder [23,28,30,32]. Unlike ΔT_c , $\Delta\rho_0$ per dose is highly sample dependent [Fig. 2(b)], suggesting additional scattering contributions, for example, within extended defects, which are also affected by irradiation-induced disorder.

This is further hinted by the evolution of the normal state slope with increasing disorder. The linear-in-temperature resistivity in optimally doped nickelates is phenomenologically similar to superconducting cuprates and other materials also dubbed “strange metals” [37,41–46]. Strange metallic resistivity has a general form $\rho(T) \sim \rho(T=0) + AT$ [47], where the slope $A = \partial\rho/\partial T$ captures (in principle) the scaling $\rho \propto k_B T$ within the frameworks of linear scattering rates [48–50] and the Drude approximation. It was previously shown that optimally doped nickelates exhibit an intriguingly similar normal state slope to hole-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) [37,51–53].

Here, we find the normal state slope $\partial\rho/\partial T$ in sample S1 remains consistent with increasing disorder, reminiscent of substitutional disorder studies in LSCO [17], both of which follow Matthiessen’s rule that the total scattering rate is a sum of all forms of scattering assuming all scattering processes are independent. Similar evolution has also been observed upon electron irradiation in iron pnictide superconductors, ascribed to increased impurity (elastic) scattering [54]. Notably, however, in sample S2 we observe a substantial change in $\partial\rho/\partial T$ with increasing electron dose, in an apparent violation of Matthiessen’s rule.

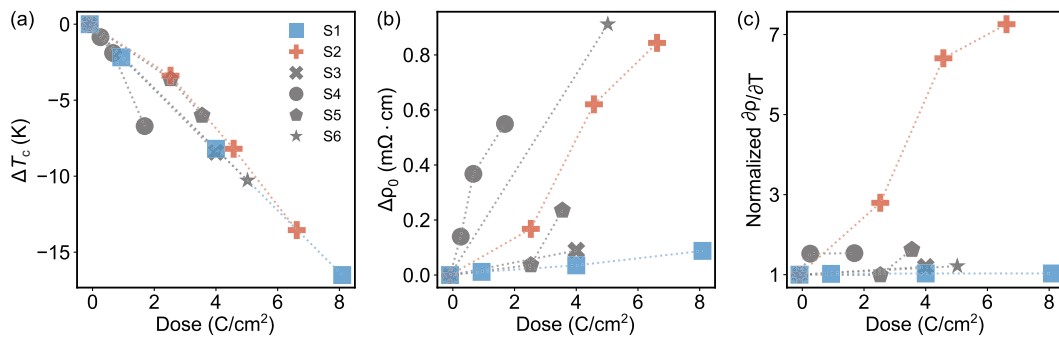


FIG. 2. Evolution of transport properties for several NSNO thin films with increasing dose, including (a) change in superconducting transition T_c , (b) change in residual resistivity with respect to the pristine values ($\Delta\rho_0$ as defined in the main text), and (c) normalized T -linear normal state slope $\partial\rho/\partial T$.

Broadly, we find that samples which exhibit larger changes in normal state slope $\partial\rho/\partial T$ (i.e., violate Matthiessen's rule) also tend to exhibit larger changes in ρ_0 and $\Delta\rho_0$ with increasing disorder [Figs. 2(b) and 2(c); Supplemental Fig. S5 [24]], suggesting that these effects may be linked. We therefore posit that this apparent violation of Matthiessen's rule may be related to the presence of other sources of scattering, such as sparse extended defects or other types of native disorder in the as-synthesized films (e.g., solid solution cation disorder and surface or interface scattering in the thin-film geometry), which cannot be fully accounted for by our measurements at this stage. We emphasize, however, that all films presented in this study are among the highest-quality superconducting nickelates produced to date, as evidenced by their relatively low pristine resistivity and T -linear slopes $\partial\rho/\partial T$ down to lowest temperatures prior to irradiation [37] and confirmed by atomic-scale structural characterization (Supplemental Fig. S7 [24]).

These results provide clear caution against the over-interpretation of macroscopic electronic transport quantities, e.g., ρ_0 , in the context of microscopic physics in these—and likely other—material systems.

Samples S1 and S2 thus represent the spread of disorder-dependent evolution observed in NSNO films studied here. The normal state behavior, in particular, exhibits a wide sample-to-sample variation, limiting our ability to meaningfully and quantitatively interpret these measurements at this stage. The effect on T_c , on the other hand, is remarkably consistent between all measured films [Fig. 2(a)], which we further parametrize to investigate the superconducting gap symmetry.

The sensitivity of unconventional superconductivity to nonmagnetic disorder can be described through a generalization of the Abrikosov-Gork'kov (AG) pair-breaking theory for magnetic impurities in a fully gapped superconductor [13,14,55,56]. While the order parameter for conventional fully gapped s -wave superconductivity is largely insensitive to nonmagnetic disorder [33,34,57,58], an unconventional sign-changing order parameter should be more susceptible to pair breaking such that increased disorder suppresses superconductivity [19,27,28]. More explicitly, within the AG formulation, the superconducting transition temperature T_c follows:

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \Psi\left(\frac{1}{2} + \frac{\alpha T_{c0}}{2\pi T_c}\right) - \Psi\left(\frac{1}{2}\right), \quad (1)$$

where Ψ is the digamma function, the pair-breaking parameter is represented by $\alpha = \hbar/2\tau k_B T_{c0}$, τ is the mean free time due to elastic scattering by impurities, and T_{c0} is T_c in the low-disorder limit ($\alpha \rightarrow 0$). In a simple theory, the scattering rate manifests in resistivity as $1/\tau = (\omega_{pl}^2/4\pi)\rho_0$, where $\hbar\omega_{pl}$ is the characteristic plasma frequency of in-plane charge carriers. For historical reasons, we note the

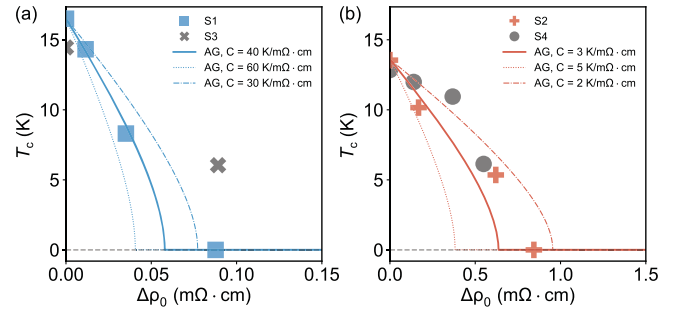


FIG. 3. Superconducting transition temperature T_c as a function of change in residual resistivity $\Delta\rho_0$ for (a) S1 and S3 and (b) S2 and S4 measured after progressive irradiation doses. The solid lines in blue (red) are generated by fitting the generalized Abrikosov-Gork'kov pair-breaking function to data points from S1 (S2) with a scaling parameters $C = 40$ (3) K/m $\Omega \cdot \text{cm}$, as described in Eq. (2). Additional AG functions (dotted and dashed lines) corresponding to other C values are plotted for comparison. The points for S3 and S4 are not included in the fits.

use of Gaussian centimetre-gram-second (CGS) system of units, i.e., the vacuum permittivity $\epsilon_0 = 1/4\pi$, in this formulation. When parameters such as τ or ω_{pl} are known for a given material, this relation can—in principle—provide quantitative insight to disorder and superconductivity [30].

Here, we fit the results of our irradiation experiments to a more generalized version of Eq. (1):

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \Psi\left(\frac{1}{2} + C \frac{\Delta\rho_0}{T_c}\right) - \Psi\left(\frac{1}{2}\right), \quad (2)$$

where we use a scaling parameter $C = \omega_{pl}^2(\epsilon_0\hbar)/(4\pi k_B)$ to fit the experimental transport data measured in SI units [59]. For each sample, we take T_{c0} as the pristine T_c measured prior to irradiation, 16.5 and 13.6 K for S1 and S2, respectively, but note that with the sparsity of data points here the quality of the fit is roughly independent of T_{c0} values within this range.

As shown in Fig. 3, the disorder dependence of both samples S1 and S2 can be fit to this generalized AG model with appropriately chosen C values, here 40 and 3 K/m $\Omega \cdot \text{cm}$, respectively. Additional fits are included for each sample to illustrate the relatively low sensitivity on the precise value of C [60]. Samples S3 and S4 are pieces from the same original perovskite film growths as S1 and S2, respectively (i.e., “sister samples”) which are not included in the AG fits but here included for comparison to emphasize that both characteristic magnitudes of $\Delta\rho_0$ are reproducible across multiple samples.

In principle, the critical resistivity at which superconductivity is fully suppressed can be related to other quantitative parameters such as characteristic length scales or critical disorder limits [19,28,62,63]. As in the normal state transport, however, the huge differences in our C

scaling factors or critical resistivities for individual samples [note the order-of-magnitude difference in the x axes in Figs. 3(a) and 3(b)] suggest that, even by accounting for different ρ_0^{pristine} through $\Delta\rho_0$, we cannot fully capture varying degrees of non-point-like disorder across different films [64].

In addition to the AG model considered above, within the density of our sampling along the disorder parameter, our results also appear generally consistent with other proposed models describing the impact of disorder in superconductors, such as that of phase fluctuations proposed by Emery and Kivelson [62,63] or spatial variance in the order parameter described by Franz *et al.* [68], both of which predict systematic suppression of T_c with disorder. It is possible that future experiments with more sampling points guided by the universal dose-dependent suppression rate of T_c reported here could be used to more clearly distinguish between such scenarios.

Most generally, the evolution of superconductivity with high-energy electron irradiation allows us to narrow the possible pairing symmetries in superconducting infinite-layer nickelates. Our results are inconsistent with the expected progression for a fully gapped order parameter, in which the superconducting transition temperature would be largely unaffected by irradiation-induced nonmagnetic disorder [69]. In the case of an anisotropic s -wave order parameter, we would expect an initial disorder-driven decrease in T_c to eventually plateau, never reaching a complete suppression [33]. Here, the clear and complete suppression of superconductivity with pair-breaking disorder in NSNO thin films indicates a momentum-space sign change in the phase of the superconducting order parameter which averages to zero with sufficient scattering (disorder) [15], pointing to unconventional superconductivity in infinite-layer nickelates.

In future experiments, it may be instructive to consider the wider family of square-planar nickelate superconductors and investigate whether the universality we find in the NSNO films here extends to the Pr- and La-based $(\text{Pr}/\text{La})_{1-x}(\text{Sr}/\text{Ca}/\text{Eu})_x\text{NiO}_2$ compounds [70–73] as well as the multilayered $T'\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$ nickelates [74]. The impact of disorder at different hole dopings may also provide some insights to proximal phases on the under- and overdoped sides of the superconducting dome [17,37,75–77].

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Data availability—The data that support the findings of this Letter are openly available [78].

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- [59] More explicitly, we focus on the second term of the first digamma function argument in Eq. (1) and start by substituting $\alpha = \hbar/2\tau k_B T_{c0}$ to rewrite as
- $$\frac{\alpha T_{c0}}{2\pi T_c} = \frac{\hbar}{4\pi k_B T_c \tau}.$$
- Further substituting the Drude expression for scattering time $1/\tau = (\omega_{pl}^2/4\pi)\rho_0$ into the same term, we get
- $$\frac{\rho_0^G}{T_c} \frac{\hbar \omega_{pl}^2}{4\pi k_B 4\pi},$$
- where ρ_0^G denotes the resistivity in Gaussian CGS units (dimensions of time). For comparison to our experimental measurements, we convert into SI units by
- $$\rho_0^G = \rho_0^I 4\pi \epsilon_0,$$
- where ρ_0^I is the resistivity in SI units (dimensions of Ohm meters). Combining the previous two expressions and taking the assumption $\rho_0 \rightarrow \Delta\rho_0$ as discussed in the text, we obtain a simplified digamma argument term $C\Delta\rho_0^I/T_c$, where
- $$C = \frac{\epsilon_0 \hbar}{4\pi k_B} \omega_{pl}^2$$
- in SI units, with dimensions of Kelvin per resistivity. All resistivities reported within the text are ρ^I .
- [60] From optical measurements of $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films on SrTiO_3 , Cervasio *et al.* [61] reported a room-temperature plasma frequency of $\sim 5500 \text{ cm}^{-1}$ ($\hbar\omega_{pl} \sim 0.68 \text{ eV}$), which would correspond to $C \sim 57 \text{ K/m}\Omega \cdot \text{cm}$ in our formulation. Encouragingly, this falls within the same order of magnitude as our fits to S1, though we note several challenges for more precise comparison, which include accounting for sample-to-sample variation with the reported transport data, temperature dependence, and low sensitivity of the fit dependence on C .
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use the 2D expression $\lambda \cong (1/\rho)(\hbar d/(e^2 k_F))$, where d is the interlayer spacing and k_F the Fermi wave vector, taking $k_F \sim 0.45 \text{ \AA}^{-1}$ from recent photoemission experiments [65,66], considering only the large quasi-two-dimensional Fermi surface and neglecting the electron pockets near the zone corners. The resulting estimate of $\lambda^{\text{crit}} \cong \xi \cong 33 \text{ \AA}$ is, therefore, only a rough one, but yields a value for ξ in reasonable agreement with estimates from other measurements [2,67].

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