

# Tunneling studies in a homogeneously disordered $s$ -wave superconductor: NbN

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We report the evolution of superconducting properties as a function of disorder in epitaxial NbN thin films grown on (100) MgO substrates. The effective disorder in these films encompasses a large range, with  $k_F l \sim 1.38$ – $8.77$ . Tunneling measurements reveal that for films with large disorder the superconducting transition temperature is not associated with a vanishing of the superconducting energy gap but with a large broadening of the superconducting density of states. Our results provide strong evidence of the loss of superconductivity via phase fluctuations in a homogeneously disordered  $s$ -wave superconductor.

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## I. INTRODUCTION

The effect of disorder on superconductivity<sup>1–4</sup> which has been a topic of considerable interest for several decades has been enjoying a serious revival,<sup>5–9</sup> which is primarily motivated around the phenomenon of the loss of bulk superconductivity not coincident with the loss of pairing. While this is natural in granular superconductors<sup>10</sup> where bulk superconductivity can be destroyed through phase fluctuations between grains, while the pairing amplitude inside individual grains remains finite, the same is not expected in a homogeneously disordered system. In fact, it was shown by Anderson<sup>1</sup> in the late 1950s that nonmagnetic disorder in a  $s$ -wave superconductor should not significantly affect the superconducting transition temperature ( $T_c$ ) as long as the system remains a metal. However, it has been suggested in recent years<sup>5–7</sup> that in the presence of strong disorder [where the mean-free path ( $l$ ) is of the order of the inverse Fermi wave vector ( $1/k_F$ )], the superconductor can spontaneously break up into domains, giving rise to a scenario similar to granular systems. Superconductivity can get destroyed due to phase fluctuations between these domains, giving rise to novel metallic and insulating phases<sup>8,9</sup> with finite pairing amplitude but no global superconductivity.

Most of the theoretical investigations<sup>5,6,9</sup> and experimental studies<sup>3,4,7,8,11</sup> on disordered superconductors have concentrated in the region close to the superconductor-insulator (SI) transition where quantum fluctuations are believed to drive the system into an insulating state with a finite gap in the electronic spectrum. In this paper, we investigate the evolution of the superconducting energy gap ( $\Delta$ ) with temperature in three-dimensional (3D) homogeneously disordered epitaxial NbN films, which are far from the SI transition. The effective disorder in these films can be tuned<sup>12</sup> from moderately clean to strongly disordered limit without destroying the crystallographic structure of NbN and the epitaxial nature of these films, making them an ideal test bed for investigating the effect of homogeneous disorder in an  $s$ -wave superconductor. All films reported in the present work have thicknesses of  $>50$  nm which is much larger than the coherence

length,  $\xi_0 \sim 5$  nm. The effective disorder in these films, characterized by the Ioffe-Regel parameter ( $k_F l$ ) varies in the range of  $k_F l \sim 1.38$ – $8.77$ . Within this range of  $k_F l$  the  $T_c$  decreases from 16.8 to 7.7 K. However, all our films have carrier densities typical of a good metal and are therefore far from the Anderson superconductor-insulator phase boundary. The central result of this paper is that even far from the SI transition, as  $k_F l$  approaches unity, the bulk  $T_c$  of an  $s$ -wave superconductor is not associated with vanishing of the superconducting energy gap ( $\Delta$ ) but rather a large broadening of the quasiparticle density of states (DOS).

## II. EXPERIMENTAL DETAILS

Epitaxial NbN films were synthesized on (100)-oriented single crystalline MgO substrates by reactive dc magnetron sputtering by sputtering an Nb target in Ar/N<sub>2</sub> gas mixture. The disorder in these films can be controlled by controlling the deposition parameters during growth. Details of the synthesis and characterization of NbN thin films have been reported in Ref. 12. Planar tunnel junctions were fabricated by first depositing a 300- $\mu$ m-wide NbN strip and oxidizing its surface at 250 °C in oxygen atmosphere for 2 h. Two tunnel junctions were fabricated on each device by subsequently evaporating 300  $\mu$ m cross strips of Ag. Under optimal growth conditions,<sup>13</sup> this process resulted in highly reproducible tunnel junctions, with high bias ( $V \gg \Delta/e$ ) tunnel junction resistance varying between 2–10  $\Omega$ . Devices fabricated by oxidizing for longer times had higher junction resistance but poorer tunneling characteristics in the superconducting state, possibly due to formation of defect states in the tunnel barrier. The device was configured in such a way that we could perform both  $I$ - $V$  measurements on the tunnel junction as well as resistance versus temperature ( $R$ - $T$ ) measurements of the NbN film on which the tunnel junction was fabricated. The conductance [ $G(V) = \frac{dI}{dV}|_V$ ] versus voltage ( $V$ ) characteristics of the tunnel junction were obtained by measuring the current versus voltage ( $I$ - $V$ ) characteristics at various temperatures and numerically differentiating the  $I$ - $V$  data. In all these measurements the NbN electrode was connected to the

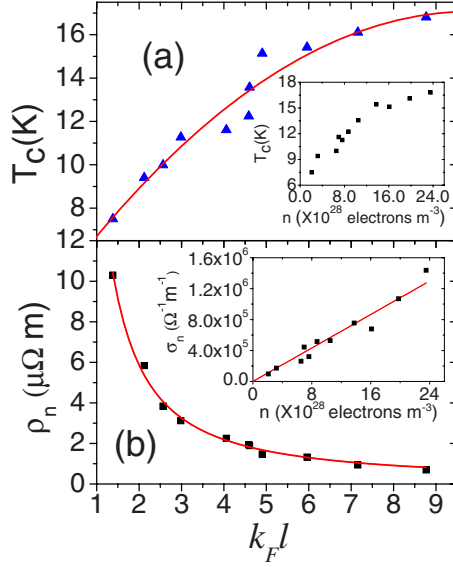


FIG. 1. (Color online) (a) Variation in  $T_c$  with  $k_F l$  in NbN films with different levels of disorder; the solid line is a polynomial fit to the data. The inset shows the variation in  $T_c$  with  $n$ . (b) Variation in  $\rho_n$  with  $k_F l$  for the same films; the solid line is a polynomial fit in powers of  $(1/k_F l)$ . The inset shows the variation in  $\sigma_n (=1/\rho_n)$  with  $n$  and the linear fit to  $\sigma_n \propto n$ .

virtual ground whereas the voltage was applied on the silver electrode. The  $T_c$  and resistivity [ $\rho(T)$ ] of the film on which the tunnel junction was fabricated were independently measured using a standard four-probe technique. Hall effect was measured on samples patterned in a Hall bar geometry. Runs of several samples showed that for identical growth conditions the  $T_c$  of the films are within 10% of each other.

### III. RESULTS AND DISCUSSION

Figure 1(a) shows the variation in  $T_c$  with  $k_F l$  for several NbN films with different levels of disorder. The values of  $k_F l$  are determined from the  $n$  extracted from Hall effect and the normal state resistivity ( $\rho_n$ ) (both measured at 18 K) using free-electron relations.<sup>14</sup> Figure 1(b) shows the variation in  $\rho_n$  versus  $k_F l$  for the same films. The inset of Figs. 1(a) and 1(b) show the variation in  $T_c$  and normal state conductivity  $\sigma_n (=1/\rho_n)$  with  $n$ . As a first approximation, both the variations in  $\rho_n$  and  $T_c$  are related to the variation<sup>12</sup> in  $n$ . While the disorder tuning in these films is likely to be a result of Nb deficiencies in highly disordered films, the large change in  $n$  cannot be accounted for by chemical considerations alone.<sup>12</sup> This points toward an unusual localization of the electronic carriers with increase in disorder which will be dealt with in a separate paper. The Thomas-Fermi screening length estimated from  $n$  indicates that the disorder arising from vacancies is screened over atomic length scales which is much smaller than the coherence length<sup>12</sup> ( $\xi \sim 5$  nm) of the superconductor. These epitaxial films are therefore homogeneous, with no physical source of extrinsic granularity. The  $k_F l$  values of the NbN films on which tunnel junctions are fabricated are estimated from the measured  $\rho_n$  and  $T_c$  using the polynomial fit to the variations in  $T_c$  and  $\rho_n$  with  $k_F l$  and

$1/k_F l$ , respectively [solid lines in Fig. 1(a) and 1(b)].

Figures 2(a)–2(c) show the  $G(V)$  vs  $V$  spectra at different temperatures down to 2.2 K for three films with different levels of disorder corresponding to (a)  $T_c = 14.9$  K, where  $k_F l \sim 6$ ; (b)  $T_c = 9.5$  K, where  $k_F l \sim 2.3$ ; and (c)  $T_c = 7.7$  K, where  $k_F l \sim 1.4$ , respectively.<sup>15</sup> An asymmetry between positive and negative biases as well as a slope at high bias, similar to the tunneling spectra of high  $T_c$  cuprates,<sup>16,17</sup> are observed for all the spectra. This asymmetry is larger for samples with smaller  $k_F l$ . Before theoretically fitting the spectra, the spectra were symmetrized by subtracting a linear background passing through the origin. The conductance spectra were fitted with the tunneling equation<sup>18</sup>

$$G(V) = \frac{dI}{dV} \Big|_V = \frac{d}{dV} \left\{ \frac{1}{R_N} \int_{-\infty}^{\infty} N_s(E) N_n(E - eV) \{f(E) - f(E - eV)\} dE \right\},$$

where  $N_s(E)$  and  $N_n(E) = N_n$  are normalized densities of states of the superconducting and normal metal electrodes, respectively,  $f(E)$  is the Fermi-Dirac distribution function, and  $R_N$  is the resistance of the tunnel junction for  $V \gg \Delta/e$ . For  $N_s(E)$  we used the DOS given by Bardeen-Cooper-Schrieffer (BCS) theory<sup>18</sup> with an additional broadening parameter<sup>19</sup>  $\Gamma$  so that  $N_s(E) = \text{Re}\{(E - i\Gamma)/[(E - i\Gamma)^2 - \Delta^2]^{1/2}\}$ .  $\Gamma (= \hbar/\tau)$  is formally introduced in this expression to take into account the effect of finite lifetime ( $\tau$ ) of the superconducting quasiparticles. However, while fitting a spectrum,  $\Gamma$  phenomenologically incorporates all nonthermal sources of broadening in the BCS DOS. The symmetrized spectra along with the corresponding fits are shown in Figs. 2(d)–2(f). We observe an increase in  $\Gamma_0/\Delta_0$  [corresponding to their respective values at 2.2 K shown in Figs. 3(a)–3(c)] from 0.002 in the most ordered film to 0.054 in the most disordered film, similar to earlier observation in granular Al samples<sup>3</sup> and disordered<sup>7</sup> TiN.

The central result of this paper, namely, the temperature variation in  $\Delta$  and  $\Gamma$  obtained from the best fit values of the spectra, is shown in Figs. 3(a)–3(c). In the same graph we show the  $\rho(T)$  versus  $T$  of the NbN films on which the tunnel junctions are fabricated. For the film with least disorder ( $k_F l \sim 6$ ),  $\Delta$  and  $\Gamma$  follow the temperature variation expected for a strong-coupling superconductor.<sup>20</sup> For this film  $T_c$  is associated with a vanishing of the  $\Delta$ . For the films with large disorder on the other hand,  $\Delta$  does not go to zero as  $T \rightarrow T_c$  but remains finite even for  $T \approx T_c$ . This is most clearly seen for the sample with  $T_c = 7.7$  K,  $k_F l \sim 1.4$  [Fig. 3(c)], where  $\Delta$  decreases to only 60% of its low-temperature value at 7.3 K. On the other hand  $\Gamma$  increases rapidly as the temperature approaches  $T_c$ . We can see from the trend in variation in  $\Delta$  and  $\Gamma$  that  $T_c$  phenomenologically corresponds to the point where  $\Delta \approx \Gamma$ .

In conventional mean-field theories of  $s$ -wave superconductors, such as BCS theory<sup>18</sup> or its strong-coupling counterparts,<sup>21</sup> the superconducting  $T_c$  is defined as the temperature where  $\Delta$  goes to zero. However, the zero resistance state in a superconductor is characterized not only by finite pairing amplitude ( $\propto \Delta$ ) but also by global phase coherence.

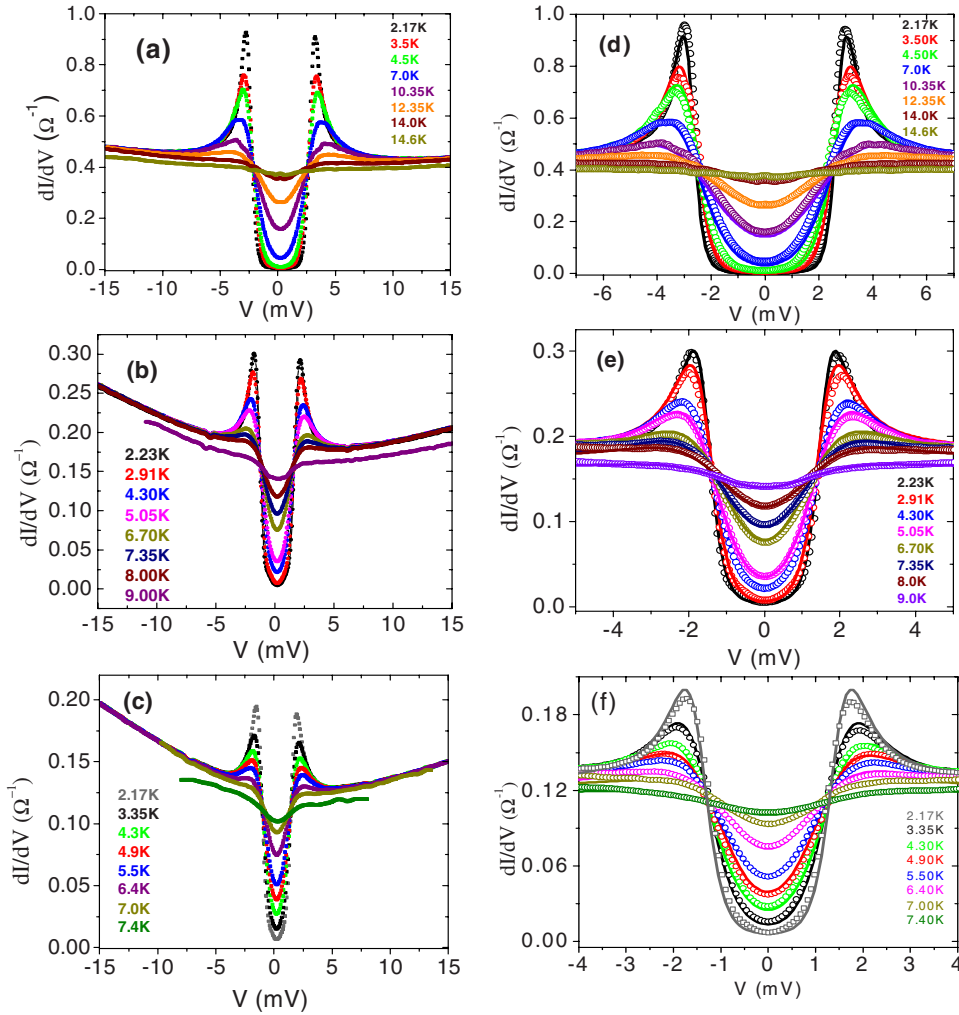


FIG. 2. (Color online) (a)–(c)  $G(V)$ - $V$  spectra at different temperatures of NbN/oxide/Ag tunnel junction on three NbN films with (a)  $T_c = 14.9$  K, where  $k_F l \sim 6$ ; (b)  $T_c = 9.5$  K, where  $k_F l \sim 2.3$ ; and (c)  $T_c = 7.7$  K, where  $k_F l \sim 1.4$ . Some temperatures have been removed for clarity. (d)–(f) The same set of spectra corresponding to (a)–(c) after removing the asymmetric background (points) along with the corresponding theoretical fits (lines). The fits are restricted to bias values equal to  $2.5\Delta_0/e$ .

Therefore, in principle bulk superconductivity can also be lost due to the loss of superfluid stiffness whereby phase fluctuations between different regions of the superconductor destroy the macroscopic zero resistance state even if the amplitude of the order parameter remains finite. This mechanism has been explored<sup>10</sup> in the context of Josephson junction (JJ) networks and granular superconducting films where the superconductor can be envisaged as a disordered network of Josephson junctions. Recent numerical simulations<sup>5</sup> on disordered two-dimensional (2D) superconductors as well as scanning tunneling spectroscopy experiments<sup>7</sup> on homogeneously disordered TiN films close to SI transition reveal that the situation in a strongly disordered  $s$ -wave superconductor is similar to a disordered JJ network. Under strong disorder the superconductor electronically segregates into superconducting domains separated by insulating regions. In this scenario, the bulk  $T_c$  corresponds to the temperature where the global phase coherence<sup>5</sup> is lost between these islands.<sup>22</sup> In such a situation the amplitude of the order parameter (and hence  $\Delta$ ) will not go to zero at  $T \approx T_c$  but BCS DOS will broaden due to thermal phase fluctuations. While we cannot make a detailed comparison with existing numerical simulations<sup>5</sup> which have been performed at  $T=0$  in 2D systems close to the SI transition, the nonvanishing of  $\Delta$  and the large increase in  $\Gamma$  as  $T \rightarrow T_c$  in strongly disordered

( $k_F l \sim 1$ ) epitaxial NbN films are consistent with this scenario.

Extrapolating the  $\Delta(T)$  versus  $T$  data for the NbN film with strong disorder, it seems natural to assume that the gap in the DOS will persist even in the normal state. This naturally points to the formation of a state with a finite value of energy gap but no global phase coherence for  $T > T_c$ . In such a situation, the bulk  $T_c$  is smaller than the mean-field transition temperature ( $T^*$ ) expected in the absence of phase fluctuations. While we cannot obtain spectroscopic information for  $T > T_c$  due to low resistance of our tunnel junctions compared to the sample resistance, an indirect evidence of this comes from the measurement of  $2\Delta_0/k_B T_c$  which is a measure of the electron-phonon coupling strength<sup>18</sup> within mean-field theories of superconductivity. Figure 4 shows the variation in  $2\Delta_0/k_B T_c$  as a function of  $T_c$  for six NbN films with different levels of disorder. For the least disordered film ( $T_c \sim 14.9$  K),  $2\Delta_0/k_B T_c = 4.36$  as expected for a strong-coupling superconductor. Since in our films  $n$  decreases with increase in disorder, for films with lower  $T_c$  the electron-phonon coupling strength is expected to decrease due to the decrease in DOS at Fermi level.<sup>12</sup> This trend is observed for films with  $T_c > 9.5$  K. However for films with  $T_c < 9.5$  K,  $2\Delta_0/k_B T_c$  shows an anomalous increase reaching a value of 4.43 for the film with  $T_c \sim 7.7$  K. This signals a breakdown

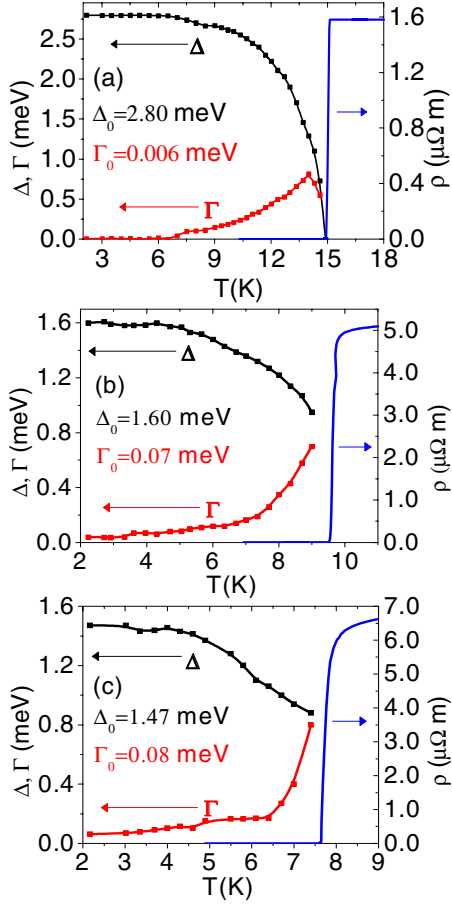


FIG. 3. (Color online) Temperature variation of  $\Delta$  and  $\Gamma$  and  $\rho$  for three NbN films with different levels of disorder: (a)  $T_c = 14.9$  K, where  $k_{Fl} \sim 6$ ; (b)  $T_c = 9.5$  K; where  $k_{Fl} \sim 2.3$ ; and (c)  $T_c = 7.7$  K, where  $k_{Fl} \sim 1.4$ . The values of  $\Delta$  and  $\Gamma$  obtained from the best fits at 2.2 K ( $\Delta_0$  and  $\Gamma_0$ ) are shown in the same figures.

of the mean-field scenario where the  $T_c$  obtained from the onset of zero resistance is smaller than the mean-field  $T_c^*$ . Between  $T_c$  and  $T_c^*$  one would expect a state with a gap in the quasiparticle spectrum but no overall phase coherence. Such a state should in principle be detectable in scanning tunneling spectroscopy measurements at temperatures higher than  $T_c$  where the tunneling resistance varies between few 100 M $\Omega$  to a few G $\Omega$  and the contribution of the normal state resistance of the sample is insignificant.

At this point it is worthwhile to explore whether other scenarios proposed in disordered superconductors could explain our data. It was suggested by Anderson *et al.*<sup>23</sup> that close to  $k_{Fl} \sim 1$  the loss of effective screening increases the electron-electron repulsion, leading to a decrease in  $T_c$ . Within McMillan theory of strong-coupling superconductors this effect leads to an increase in the effective Coulomb pseudopotential ( $\mu^*$ ). While the  $k_{Fl}$  of our films are in the range where this effect could manifest itself,<sup>24</sup> this mechanism gives a positive correction to the effective  $\mu^*$  which does not explain the increase in  $2\Delta_0/k_B T_c$  or our observation

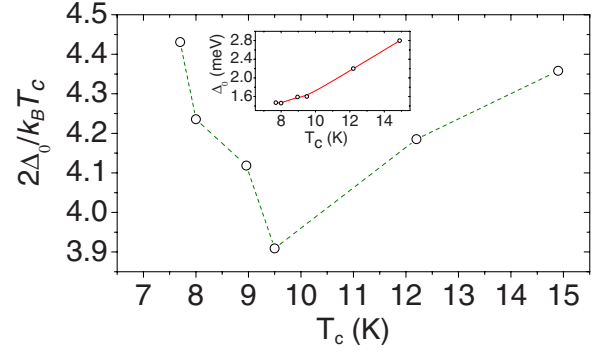


FIG. 4. (Color online) Variation in  $2\Delta_0/k_B T_c$  as a function of  $T_c$  for NbN films. The inset shows the variation in  $\Delta_0$  with  $T_c$ .

of finite  $\Delta$  at  $T \approx T_c$ . In another model proposed by Bergmann,<sup>25</sup> it was suggested that disorder causes an effective increase in the electron-phonon coupling strength due to phonon emission caused by inelastic-scattering processes at impurity sites. It was qualitatively argued<sup>26</sup> that in strong-coupling superconductors, this effect is compensated by “strong-coupling” effects, which in the presence of strong disorder could cause a decrease in  $\Delta$  and  $T_c$ . While this effect could in principle account for the increase in  $2\Delta_0/k_B T_c$ , it is not consistent with the nonvanishing of  $\Delta$  at  $T \approx T_c$ .

#### IV. CONCLUSION

In conclusion, we show that in 3D strongly disordered NbN epitaxial thin films, the superconducting energy gap does not vanish as  $T \rightarrow T_c$  as expected for a conventional  $s$ -wave superconductor. Furthermore,  $2\Delta_0/k_B T_c$  shows an anomalous increase with increasing disorder in this regime. Our results indicate that the DOS is likely to remain gapped even in the normal state, resulting in a pseudogap state at temperatures  $T > T_c$ . These results are in qualitative agreement with recent theoretical predictions on strongly disordered  $s$ -wave superconductors close to the SI phase boundary. However, the unique feature of our results is that even though our films are far from the SI phase boundary, we see that superconducting  $T_c$  is strongly influenced by phase fluctuations when  $k_{Fl}$  approaches 1. These results bear a strong similarity with the high  $T_c$  superconducting cuprates where a finite energy gap above  $T_c$  is well established<sup>16</sup> from tunneling measurements. It would therefore be instructive to obtain spectral information on the state at  $T > T_c$ , as well as its possible spatial variation using techniques such as scanning tunneling spectroscopy. These measurements are currently underway and will be reported in a future paper.

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- <sup>14</sup>For details regarding the calculation of  $k_{Fl}$ , see Ref. 12.
- <sup>15</sup>Since the absolute value of  $T_c$  is determined with greater accuracy than  $\rho_n$ , the  $k_{Fl}$  values mentioned are estimated from  $T_c$  using the polynomial fit in Fig. 1(a). However, we verified that the  $k_{Fl}$  values estimated from  $\rho_n$  are within 20% of the value determined from  $T_c$ .
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