

**Temperature dependence of resistivity and Hall coefficient in strongly disordered NbN thin films**

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We report the temperature dependence of resistivity ( $\rho$ ) and Hall coefficient ( $R_H$ ) in the normal state of homogeneously disordered epitaxial NbN thin films with  $k_F l \sim 1.68-10.12$ . The superconducting transition temperature ( $T_c$ ) of these films varies from 2.7 to 16.8 K. While our least disordered film displays usual metallic behavior, for all the films with  $k_F l \leq 8.13$ , both  $\frac{d\rho}{dT}$  and  $\frac{dR_H}{dT}$  are negative up to 285 K. We observe that  $R_H(T)$  varies linearly with  $\rho(T)$  for all the films and  $[\frac{R_H(T)-R_H(285\text{ K})}{R_H(285\text{ K})}] = \gamma[\frac{\rho(T)-\rho(285\text{ K})}{\rho(285\text{ K})}]$ , where  $\gamma = 0.68 \pm 0.11$ . Measurements performed on a 2-nm-thick Be film show similar behavior with  $\gamma = 0.69$ . This behavior is inconsistent with existing theories of localization and  $e-e$  interactions in a disordered metal.

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**I. INTRODUCTION**

The evolution of electronic properties with disorder remains one of the most challenging problems of modern condensed-matter physics.<sup>1</sup> In recent years, experimental investigations in this field have received renewed interest motivated by several novel phenomena observed in strongly disordered superconductors, such as magnetic field-driven superconductor-insulator transitions in disordered InO<sub>x</sub> and TiN films<sup>2,3</sup> close to the superconductor-insulator transition, unusual temperature dependence of normal-state resistivity<sup>4</sup> in underdoped High  $T_c$  cuprates and the formation of a pseudogapped normal state<sup>5,6</sup> in strongly disordered conventional superconductors. For weak disorder in a normal metal, scattering from the disorder potential results in an increase in the temperature-independent part of the electrical resistivity whereas the temperature-dependent part remains largely unaltered. However, it was shown by Anderson<sup>7</sup> that in the presence of strong disorder, all the electronic states in a three-dimensional (3D) metal can get localized giving rise to an insulating state. However, experimentally determining the exact disorder level where the Anderson metal-insulator transition happens in a 3D disordered system is often complicated due to the presence of strong electron-electron ( $e-e$ ) interactions close to the metal-insulator transition.  $e-e$  interactions, whose effect is enhanced by the diffusive motion of the electrons, can give rise to nontrivial temperature-dependent corrections to the resistivity ( $\rho$ ) and can even change the sign<sup>8,9</sup> of  $\frac{d\rho}{dT}$  while in the metallic phase. It is therefore important to investigate the evolution of the transport properties as a function of disorder strength to understand the role of  $e-e$  interactions in the presence of strong disorder.

Theoretically, in addition to  $\rho$ , the Hall coefficient ( $R_H$ ) is another quantity that is expected to get modified due to  $e-e$  interactions.<sup>10</sup> In the weak scattering regime the relative correction to  $\rho$  and  $R_H$  is predicted to be related by<sup>11</sup>  $\frac{\Delta R_H}{R_H} = 2 \frac{\Delta \rho}{\rho}$ . In this paper we investigate the temperature dependence of  $\rho$  and  $R_H$  in 3D disordered epitaxial NbN thin films with<sup>12</sup>  $k_F l \sim 1.68-10.12$ . Except for the most disordered film which

undergoes a broad superconducting transition with  $T_c \sim 2.7$  K, all the other films have a sharp superconducting transition ( $\Delta T_c < 0.3$  K) with the  $T_c$  ranging between 8.13 and 16.8 K. The thickness of these films ( $t > 50$  nm) is much larger than the dirty limit coherence length,  $\xi_0 \sim 5$  nm and the electronic mean-free path ( $l \sim 1.5-5$  Å). With increasing disorder the transport properties of these films evolve from conventional metallic behavior (e.g.,  $\frac{d\rho}{dT} > 0$ ,  $R_H$  independent of  $T$ ) for our most ordered films to an unusual metallic state where  $\frac{d\rho}{dT}, \frac{dR_H}{dT} < 0$ . The main result of this paper is that for all the films with  $\frac{d\rho}{dT}, \frac{dR_H}{dT} < 0$ ,  $R_H(T)$  varies linearly with  $\rho(T)$  over a large range of temperature. In addition, we observe a universal relation,  $[\frac{R_H(T)-R_H(285\text{ K})}{R_H(285\text{ K})}] = \gamma[\frac{\rho(T)-\rho(285\text{ K})}{\rho(285\text{ K})}]$  with  $\gamma \approx 0.68$ , in sharp contrast to the prediction in the weak scattering regime. For comparison, similar measurements done on ultrathin Be films show a very similar behavior with  $\gamma \approx 0.69$ .

**II. EXPERIMENTAL DETAILS**

Epitaxial NbN films (similar to the ones used in Refs. 6 and 13) were grown through reactive magnetron sputtering on (100)-oriented single-crystalline MgO substrate, by sputtering a Nb target in Ar/N<sub>2</sub> gas mixture. The effective disorder was controlled by changing the sputtering power or the Ar/N<sub>2</sub> ratio which effectively changed the Nb/N<sub>2</sub> ratio in the plasma. X-ray diffraction studies<sup>14</sup> and transmission electron microscopy reveal that all our films are epitaxial with no extrinsic source of physical granularity. The 2-nm-thick Be film was deposited on a polished glass substrate using  $e$ -beam deposition in an initial vacuum of  $\sim 0.1$   $\mu$  Torr. The Be film also showed no salient morphological features when investigated with scanning force microscopy. Further details of sample preparation and characterization have been reported in Refs. 13–16. Resistance, magnetoresistance, and Hall measurements were performed in a home built cryostat up to a maximum field of 12T.  $\rho$  and  $R_H$  at different temperatures were measured using a standard four-probe ac technique on films patterned in a Hall-bar geometry using a

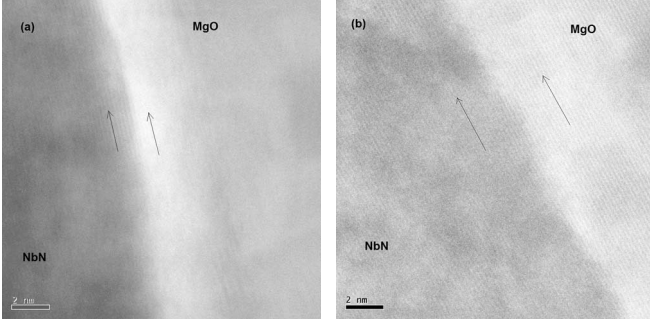


FIG. 1. (a) Transmission electron micrograph of disordered NbN film with a  $k_F l \sim 3.3$  and (b) transmission electron micrograph of an NbN film with  $k_F l \sim 9$ . The direction of lattice planes in the film and the substrate is shown by arrows.

shadow mask.  $R_H$  was calculated from Hall voltage deduced from reversed field sweeps from +12 to -12 T after subtracting the resistive contribution. The thickness of the films was determined within an accuracy of 15% using a stylus profilometer by measuring on different parts of the patterned sample.

III. RESULTS

Figure 1 shows the cross sectional transmission electron microscopy (TEM) images of two films with the highest and lowest level of disorder. The epitaxial nature of the films is evident from the NbN lattice planes which match with the lattice planes of the MgO substrate. This is also seen from x-ray diffraction studies.<sup>14</sup> This implies that the increase in disorder does not destroy the epitaxial nature of our films. Therefore disorder in our films does not result from physical

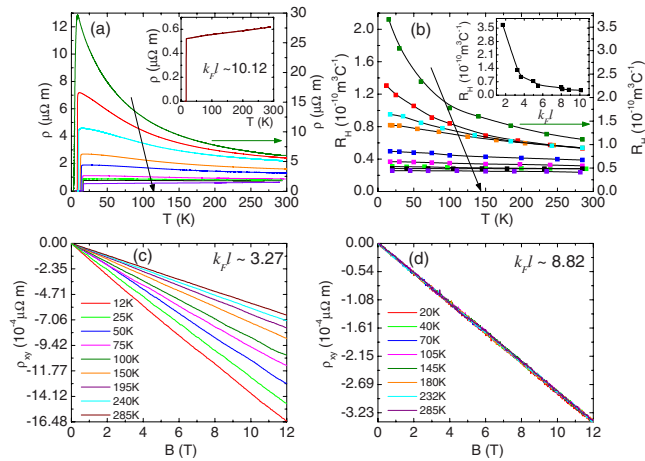


FIG. 2. (Color online) (a)  $\rho$  vs  $T$  for the films of different  $k_F l$  and (inset) an expanded view of the  $\rho(T)$  for the most ordered sample with  $k_F l \sim 10.12$ ; (b)  $R_H$  vs  $T$  for all the films and (inset)  $R_H$  at 18 K as a function of  $k_F l$ ; the black slanting arrows are in the direction of increasing  $k_F l$  values: 1.68, 3.27, 3.65, 4.98, 5.50, 8.01, 8.13, 8.82, and 10.12; in both (a) and (b) the scale for the film with  $k_F l$  1.68 is on the right-hand axis; [(c) and (d)]: Hall resistivity vs magnetic field at different temperatures for the samples with  $k_F l \sim 3.27$  and 8.82, respectively.

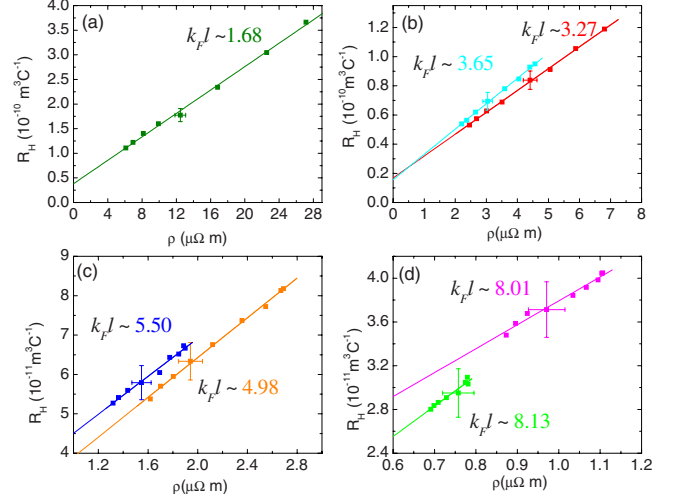


FIG. 3. (Color online)  $R_H(T)$  vs  $\rho(T)$  for the samples with (a)  $k_F l \sim 1.68$ , (b)  $k_F l \sim 3.27$  and 3.65, (c)  $k_F l \sim 4.98$  and 5.50, and (d)  $k_F l \sim 8.01$  and 8.13. The error bars indicate the errors in the absolute values of  $\rho$  and  $R_H$ .

granularity but rather from atomic-scale disorder such as vacancies in the crystalline lattice. Figure 2(a) shows  $\rho(T)$  as a function of temperature for films with different  $k_F l$  up to 300 K. An expanded view of  $\rho-T$  for the film with lowest disorder (showing conventional metallic behavior), is shown in the inset. All the films with  $k_F l \leq 8.13$  (Appendix) show a negative  $d\rho/dT$  extending up to room temperature. The temperature dependence of  $R_H(T)$  in the same temperature range extracted from the slope of  $\rho_{xy}-B$  curves [Figs. 2(c) and 2(d)], is shown in Fig. 2(b). The central result of this paper, namely, the linear variation in  $R_H$  with  $\rho$  obtained using the temperature variation in  $\rho(T)$  and  $R_H(T)$ , is shown in Figs. 3(a)–3(d). Within error bars of our measurement,  $R_H$  follows a relation  $R_H(T) = R_{H0} + A\rho(T)$  for all the disordered films.

We now turn our attention to the precise temperature dependence of  $\rho$  for the disordered samples. For the more disordered samples,  $\sigma$  varies as  $T^{1/2}$  over a large temperature range which gradually crosses over to a linear  $T$  behavior for temperatures below 100 K. This trend is shown in Figs. 4(a) and 4(b). It is interesting to note that for the films in the range  $k_F l \sim 3.27-5.50$ , the slopes of the linear parts of the  $\sigma-T$  and  $\sigma-T^{1/2}$  curves are independent of  $k_F l$  within error bars. All our films show an upward deviation from the linear  $T$  behavior at temperatures below  $\sim 40$  K. While the onset of superconductivity and the very high upper critical field preclude the possibility of investigating the normal-state conductivity down to very low temperatures, the trend in variation in  $\sigma-T$  suggests that  $\sigma \neq 0$  for  $T \rightarrow 0$ . Similar behavior in the resistivity has been reported earlier<sup>17</sup> in 3D disordered  $\text{In}_2\text{O}_{3-x}$  films. Since our measurements are done up to moderately high temperatures it is important to assess the role of electron-phonon ( $e$ -ph) scattering in  $\rho(T)$  at high temperatures. To estimate this contribution, in Fig. 4(c) we show an expanded view of  $\rho-T$  of the sample with  $k_F l \sim 8.82$ . The  $\rho-T$  curve has a shallow minimum around 240 K where  $d\rho/dT$  changes from negative to positive. The positive slope at high temperature which can be attributed to the  $e$ -ph scattering,

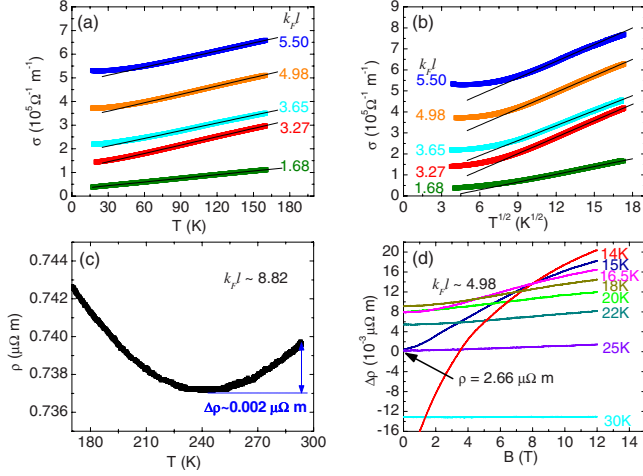


FIG. 4. (Color online) (a)  $\sigma$  vs  $T$  for the disordered films showing the linear behavior at lower temperatures. (b)  $\sigma$  vs  $T^{1/2}$  along with linear fits. (c) An expanded  $\rho(T)$  for the film with  $k_F l \sim 8.82$ . The upturn in resistivity above 240 K shows the relative magnitude of the contribution from electron-phonon scattering. (d) MR for the film with  $k_F l \sim 4.98$  at different temperatures.

results in a resistivity increase in  $\sim 0.002 \mu\Omega \text{ m}$  in the interval 240–300 K. Therefore the contribution of  $e$ -ph scattering to the overall  $\rho(T)$  is less than 1% for this sample. For more disordered samples this contribution is therefore negligible compared to impurity scattering. It is also worthwhile to explore whether the upward deviation of  $\sigma(T)$  at low temperatures from the linear  $T$  behavior in the highly disordered films is due to proximity to the superconducting transition. Since superconducting fluctuations<sup>18</sup> can be suppressed by the application of magnetic field we measured the magnetoresistance (MR) at  $T > T_c$  [Fig. 4(c)] and found that the MR is negligible for  $T > 25$  K. It is therefore likely that the deviation from  $\sigma \propto T$  behavior for  $T < 40$  K is an intrinsic property of the normal state and not associated with onset of superconductivity.

#### IV. DISCUSSION

We now compare our data with existing predictions for disordered metals. A power-law dependence of  $\sigma$  on  $T$  in a disordered metal has been predicted<sup>10</sup> to happen both from weak localization as well as  $e$ - $e$  interactions. However, for a 3D disordered system without  $e$ - $e$  interactions, scaling theory<sup>19</sup> predicts that  $R_H$  will retain its metallic character down to the metal-insulator transition and will be temperature independent.  $e$ - $e$  interactions on the other hand can change the values of both  $\rho$  and  $R_H$ . In the leading order in  $(1/k_F l)$ ,  $e$ - $e$  correlation in 3D predicts a conductivity of the form<sup>8,10</sup>

$$\sigma = \sigma_0 + \frac{e^2}{\hbar} \frac{1}{4\pi^2} \frac{1.3}{\sqrt{2}} \left( \frac{4}{3} - \frac{3}{2} \tilde{F}_\sigma \right) \sqrt{\frac{T}{D}}, \quad (1)$$

where  $D = \frac{v_F^2 \tau}{d}$  is the diffusivity,  $d$  is the dimensionality, and  $\tilde{F}_\sigma$  is a Fermi-liquid parameter such that  $(\frac{4}{3} - \frac{3}{2} \tilde{F}_\sigma)$  is of the

order of unity. Equation (1) is therefore strictly valid in the weak scattering regime where terms of higher order in  $(1/k_F l)$  do not contribute significantly. This is similar to the observed temperature dependence of our disordered films over a large range of temperature. There are, however, two difficulties in reconciling with this scenario. First, the overall change in the conductance is much larger than what is predicted from this effect. For example, calculation of the prefactor of  $T^{1/2}$  in Eq. (1) using the experimental value<sup>20</sup> of  $v_F = 1.758 \times 10^6 \text{ ms}^{-1}$  and  $\tau = 1.227 \times 10^{-16} \text{ s}$  for the sample with  $k_F l \sim 3.27$  shows that it is two orders of magnitude smaller than the corresponding experimental values of the slope obtained from the  $\sigma$  vs  $T^{1/2}$  curve in the temperature range where it is linear. More importantly, it has been shown that for  $e$ - $e$  interactions the corrections in  $(\sigma_{xy})$  in the conductivity diagrams exactly cancel each other such that  $\delta\sigma_{xy} = 0$ . Since  $R_H \cong \frac{\sigma_{xy}}{\sigma_{xx}}$ , it directly follows that

$$\frac{\Delta R_H}{R_H} = \frac{\delta\sigma_{xy}}{\sigma_{xy}} - 2 \frac{\delta\sigma_{xx}}{\sigma_{xx}} = 2 \frac{\Delta\rho}{\rho}, \quad (2)$$

namely, that the relative correction in  $R_H$  is double<sup>21</sup> that of  $\rho$ . In Figs. 5(a) and 5(b) we plot  $\frac{\Delta R_H}{R_H} [= \frac{R_H(T) - R_H(285 \text{ K})}{R_H(285 \text{ K})}]$  vs  $\frac{\Delta\rho}{\rho} [= \frac{\rho(T) - \rho(285 \text{ K})}{\rho(285 \text{ K})}]$  for all NbN samples with  $k_F l \leq 8.13$ . All the curves are linear within error bars of our measurements and the slope,  $\gamma = d(\frac{\Delta R_H}{R_H})/d(\frac{\Delta\rho}{\rho})$  has a universal value  $0.68 \pm 0.11$ . This is in clear contradiction with Eq. (2). While Eq. (2) is strictly valid for the limit  $H \rightarrow 0$ , taking this limit would not alter our results since for our samples  $\rho_{xy}$  is linear over the entire magnetic field range [Figs. 2(c) and 2(d)]. The inset of Fig. 5(a) shows that within error bars,  $\gamma$  is independent of  $k_F l$ .

We now consider possible scenarios where  $\gamma$  can deviate from 2. Since for localization effects  $\gamma = 0$  and for  $e$ - $e$  interaction  $\gamma = 2$ ,  $\gamma$  can, in principle, take any intermediate value in the presence of both localization and  $e$ - $e$  interactions. This scenario, however, can be ruled out for two reasons. First, since for samples with larger  $k_F l$ ,  $e$ - $e$  interactions will be more predominant than localization effects, a systematic deviation of  $\gamma$  towards 2 should have been observed with increasing  $k_F l$ . Secondly, in 3D the temperature dependence of  $\sigma$  due to localization is different from that due to  $e$ - $e$  interactions. Since  $R_H$  is affected only by  $e$ - $e$  interactions, the linear relation between  $\sigma(T)$  and  $R_H$  is not expected over a large temperature range. The second possibility is that our films are not in the weak scattering regime ( $k_F l \gg 1$ ) for which Eqs. (1) and (2) are applicable. However, since the level of disorder in the films shown in Fig. 5(a) spans a large range of  $k_F l \sim 1.68$ –8.13, one would have expected  $\gamma$  to be asymptotic to the theoretical value of 2 with increasing  $k_F l$ . Such a systematic change was actually observed in a two-dimensional (2D) electron gas<sup>22</sup> in a Si inversion layer where a gradual increase in  $\gamma$  towards the theoretical value of 2 was observed as the sheet resistance  $R_{sq} \rightarrow 0$ . No such systematic variation is observed in our data. To check if the observed behavior is specific to NbN, similar measurements were also performed on a 2-nm-thick Be film with sheet resistance,  $R_{sq} \approx 3.61 \text{ k}\Omega$ . While for this thickness the film is not super-

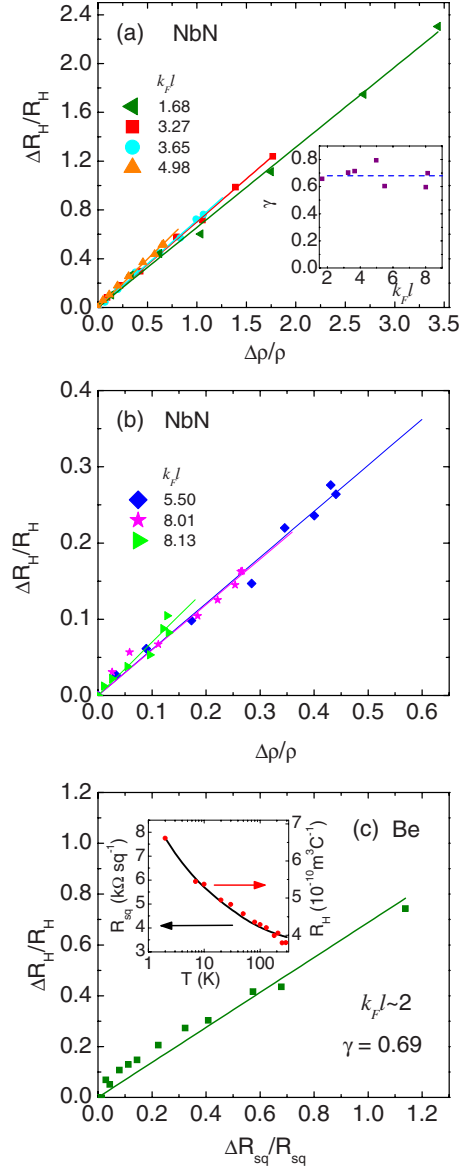


FIG. 5. (Color online) [(a) and (b)]  $\frac{\Delta R_H}{R_H} [= \frac{R_H(T) - R_H(285 \text{ K})}{R_H(285 \text{ K})}]$  vs  $\frac{\Delta \rho}{\rho} [= \frac{\rho(T) - \rho(285 \text{ K})}{\rho(285 \text{ K})}]$  (shown as points) for the different NbN samples. The linear fits (shown as lines) have an average value of  $0.68 \pm 0.11$ ; the inset in (a) shows  $\gamma [= d(\frac{\Delta R_H}{R_H})/d(\frac{\Delta \rho}{\rho})]$  as a function of  $k_F l$  with the average value shown as a dashed line. (c)  $\frac{\Delta R_H}{R_H} [= \frac{R_H(T) - R_H(290 \text{ K})}{R_H(290 \text{ K})}]$  vs  $\frac{\Delta R_{sq}}{R_{sq}} [= \frac{R_{sq}(T) - R_{sq}(290 \text{ K})}{R_{sq}(290 \text{ K})}]$  for the ultrathin Be film. The  $\gamma$  value is 0.69. The inset shows the temperature variation  $R_H(T)$  and  $R_{sq}(T)$  for the ultrathin Be film.

conducting the thickness is much smaller than the coherence length ( $\xi \sim 20$  nm) of its superconducting thick counterpart. This film is thus in the 2D limit with  $k_F l \sim 2$ . The  $R_{sq}(T)$  and  $R_H(T)$  measured in this film is summarized in Fig. 5(c). While this film displays a logarithmic temperature dependence of  $R_{sq}$  typical of a disordered 2D system, the slope,  $\gamma = d(\frac{\Delta R_H}{R_H})/d(\frac{\Delta R_{sq}}{R_{sq}}) \approx 0.69$ , is strikingly similar to the value observed in disordered NbN. It would be interesting to investigate whether the value of  $\gamma$  in the 2D Be films remains robust as a function of disorder ( $R_{sq}$ ), similar to NbN.

In this context we would like to note that the theoretical value of  $\gamma \approx 2$  has so far been observed in 2D electron gas in Si inversion layers<sup>23</sup> in the limit of large sheet resistance and at intermediate magnetic fields ( $\sim 0.1$ – $0.5$  T). However,  $\gamma$  decreases from 2 for both very low fields as well as higher field values. The former is attributed to the dominance of localization at very low fields whereas the latter presumably arises due to breakdown of the low-field limit where Eq. (2) is valid. The first effect is also observed in 3D disordered  $\text{In}_2\text{O}_3$  films<sup>24</sup> where  $R_H$  measured at very low magnetic field was reported to be temperature independent. While the very low limit of  $R_H$  is below our experimental resolution, we do not observe any nonlinearity in  $\rho_{xy}$  vs  $B$  at fields above  $\sim 0.2$  T. On the other hand, Hall-effect measurements on uncompensated Si:As samples<sup>25</sup> in the metallic regime showed that  $\gamma$  ranges from 0.4–0.6 for different samples. In this context we would like to note that when the transport is not in the diffusive regime (defined as  $k_B T \tau / \hbar \ll 1$ ), a decrease in  $\gamma$  from its value of 2 with increase in the quantity  $k_B T \tau / \hbar$  has been theoretically predicted by Zala *et al.*<sup>26</sup> However, our samples are at the opposite extreme with  $k_B T \tau / \hbar$  at 300 K ranging between  $4.5 \times 10^{-3}$ – $8.46 \times 10^{-3}$ . We therefore believe that further theoretical considerations may be needed to establish the value of  $\gamma$  in a disordered metallic system.

Since our measurements can be done only down to the superconducting transition temperature of the NbN thin films, we could also consider the possibility that our more disordered samples are actually in the insulating regime, where the temperature dependence of  $\sigma$  and  $R_H$  are expected to show similar behavior.<sup>27</sup> In that case it is expected that  $\sigma(T)$  will go to zero at a temperature below our minimum temperature of measurement. While such a possibility cannot be ruled out it is clear from the data that this would require a sharp downturn in the conductivity for  $T < 12$  K even for the most disordered sample. This would signify the onset of a new energy scale which is smaller than our lowest temperature of measurement ( $\sim 1$  meV). Therefore, even if such an energy scale exists it cannot manifest itself over the large temperature range of our observations.

## V. CONCLUSIONS

In summary, we report the unusual temperature dependence of  $\rho$  and  $R_H$  in the normal state of homogeneously disordered epitaxial NbN thin films with the disorder level varying from moderately clean to very dirty limit. All the samples are in the metallic regime as seen by a finite  $\sigma$  as  $T \rightarrow 0$ . The films with  $k_F l < 8.13$  display a large negative  $d\rho/dT$  extending up to room temperature. A quantitative analysis shows that the overall change in conductivity with temperature is much larger than what is expected from weak localization or  $e$ - $e$  interactions. We find a remarkably linear relation between  $R_H(T)$  and  $\rho(T)$  extending from low temperature up to 285 K in all the films with  $\frac{d\rho}{dT} < 0$ . While a linear relation between  $\rho(T)$  and  $R_H(T)$  is in agreement with the corrections in  $\rho(T)$  and  $R_H(T)$  arising from  $e$ - $e$  interactions, the relative change in the two quantities follow the relation  $\frac{\Delta R_H}{R_H(285 \text{ K})} = (0.68 \pm 0.11) \frac{\Delta \rho}{\rho(285 \text{ K})}$  independent of  $k_F l$ .

Measurements on ultrathin Be films suggest that this behavior is not specific to NbN but is a more general property of disordered metals. We believe that our results represent a fundamental contradiction with the weak scattering prediction of  $e$ - $e$  interactions in disordered metallic systems.

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### APPENDIX

In our earlier publications (Refs. 6 and 13) dealing with the superconducting state of disordered NbN, we have reported the  $k_{Fl}$  measured from the value of  $\rho$  and  $R_H$  at 18 K. In this paper we report the  $k_{Fl}$  values determined from the

TABLE I. Look-up table for  $k_{Fl}$  values calculated at 18 and 285 K.

$k_{Fl}$ (18 K)	$k_{Fl}$ (285 K)
0.56	1.68
1.54	3.27
2.14	3.65
3.33	4.98
4.08	5.5
6.36	8.01
7.47	8.13
8.38	8.82
12.07	10.12

corresponding quantities at 285 K. This is because at higher temperature there are no corrections in  $\rho$  and  $R_H$  due to localization<sup>11</sup> or  $e$ - $e$  interaction<sup>28</sup> up to the leading order in  $1/k_{Fl}$ . In Table I we provide a look-up table that gives the  $k_{Fl}$  values calculated at 18 and 285 K for the same samples in order to have a correspondence with Refs. 6 and 13.

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