Anomalous magnetoresistance in ultrathin Nb films

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(Recieved 15 August 1983)

The normal-state conductivity of thin Nb films was studied as a function of temperature and magnetic field. At liquid-He temperatures, the behavior of the zero-field conductivity is in agreement with recent electronic transport theories in the presence of disorder. The magnetoresistance, however, shows some anomalous features that cannot be simply reconciled with known mechanisms. The suppression of superconductivity with increased sheet resistance is discussed in light of these findings.

It has been recognized for quite some time that impurity scattering may have a detrimental effect on superconductivity. In three dimensions, this effect is usually small (and sometimes masked by enhancement phenomena), even when the energy uncertainty associated with the scattering process $\hbar/\tau$ is larger than the condensation energy $k_B T_c$. A theoretical explanation for this apparent "insensitivity" was given by Anderson[1] who had shown that only scattering that breaks time-reversal symmetry should be effective in reducing the BCS condensation energy. Intuitively, one expects these arguments to break down once the scattering is so intense as to cause the particle to be localized within a distance ξ_L when the latter is much smaller than the superconducting coherence length ξ_S. On the other hand, in the limit $\xi_L \gg \xi_S$ the Anderson theorem should hold true. Contrary to this expectation, experiments on thin films often show a pronounced decrease of the transition temperature T_c, with the inverse of the elastic scattering time τ, even in materials that do not show such an effect in the bulk form (e.g., lead). In fact, ΔT_c was shown to correlate with the sheet resistance $R_\Box$,[2-4] which perhaps is indicative of the restrictive geometry involved. At first glance, these phenomena are at variance with the Anderson theorem and clearly deserve a physical explanation. Various attempts to account for such experimental observations incorporating, in fact, the restricted dimensionality have been made in the past but with only limited success.[5] The recent progress[6-8] in the understanding of metallic transport properties in the presence of disorder has led to a new approach to this problem. Imry and Strongin[9] have suggested that some of the results on thin films of superconducting materials may be qualitatively understood within the framework of modern theories of localization and Coulomb interactions. In particular, they argued that in two dimensions (2D) superconductivity will vanish at the point where the transport properties change over from "weak" to "strong" localization, provided that the system can be treated as uniform (small grain limit). Although the physical mechanism responsible for the weakening of

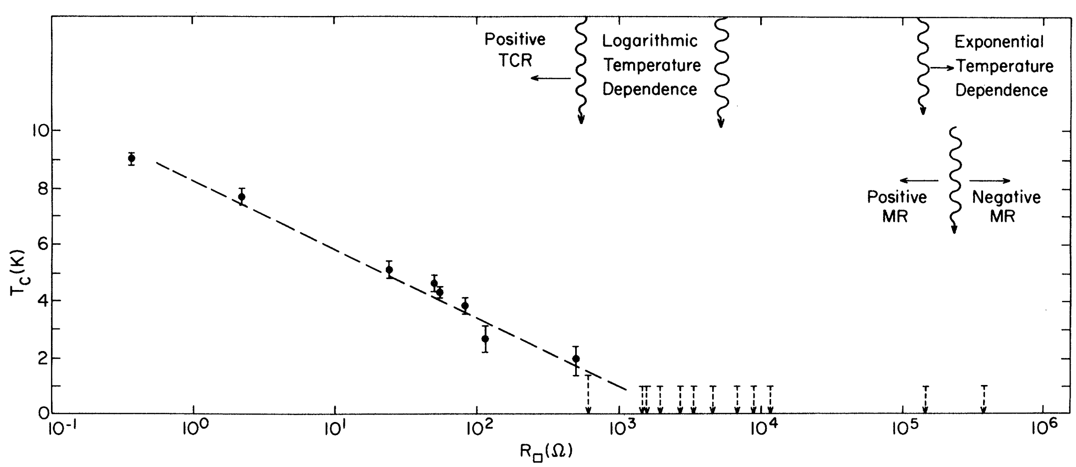

FIG. 1. Superconducting transition temperature T_c of the Nb films is plotted as a function of the sheet resistance at 4.2 K (the values of $R_\Box$ where electrical resistance measurements were made and no T_c were found are indicated by arrows). Also included are the ranges in terms of $R_\Box$ where qualitative change in the transport properties were observed.

superconductivity remains rather obscure, it is understood that both incipient localization and Coulomb-repulsion effects should correlate with the disappearance of the superconducting state.

In this work we have tried to look for evidence for the presence of these effects in the normal-transport properties of thin Nb films. Special emphasis was given to the region where T_c was essentially gone and the normal-state properties could have been measured on a wide range of temperatures. It will be shown that some features, such as the point where T_c vanishes and the temperature dependence of the resistivity, are in qualitative agreement with those theoretical ideas. On the other hand, the magnetoresistance (MR) results show features that seem to be inexplicable by the above models. Furthermore, suppression of T_c is observed and, in fact, is very prominent even when $R_\square$ is quite small and the transport properties in the normal state do not show any clear "anomalous" features.

The Nb films described below were prepared by dc magnetron sputtering at room temperature (in ultrahigh-purity argon $\sim 10^{-3}$ Torr; background pressure $\sim 10^{-7}$ Torr). With the use of a rotatory substrate holder, up to six samples with different thicknesses could be prepared at each run. The substrates, flat- (melted-) glass slides, were masked to obtain the desired geometry (0.6×2.5 cm) for the electrical measurements. In this manner, films with $R_\square$ from 0.4 to 5×10^3 Ω (at 4.2 K) were prepared. Some of these films were remeasured after exposure to air for several days. The film thickness (from 6000 to ~ 10 Å) was measured by interferometry and mechanical stylus (Dektak) or inferred from the sputtering rate for the thinnest films. A few samples were prepared for transmission electron microscopy, depositing the Nb films on Cu grids coated with carbon. These samples showed a uniform and homogeneous microstructure with grain size which was typically 10–20 Å. The electrical resistance and the superconducting transition were measured with a standard four-probe dc technique using the vapor pressure of He and calibrated Ge and Pt resistors for thermometry. The magnetoresistance measurements were made using a conventional split-coil magnet (7 kOe) that was able to vary the field axis continuously through $\sim 200°$ and a Nb-Ti superconducting magnet (40 kOe), both with the field perpendicular to the sample plane.

The transition temperature T_c of the thin Nb films studied is plotted in Fig. 1 as a function of the sheet resistance $R_\square$. Also included in this figure are the range in terms of $R_\square$, where qualitative changes in the transport properties are observed. Note that ΔT_c correlates with $\ln R_\square$ over a considerable range ($R_\square \approx 1-1000$ Ω). A similar dependence has been observed before in thin Pb and Bi layers deposited in cryogenic temperatures[3] and a theoretical model[10] indeed predicts that $\Delta T_c/T_c = A \ln R_\square$. The magnitude of the effect is, however, considerably larger than theoretically anticipated: We obtain from Fig. 1 that A is $\sim 3\times 10^{-4}$ whereas Kulik[10] predicts 0.3×10^{-4}.

The transition temperature vanishes by extrapolation at $R_\square \sim 5$ kΩ, at which point several anomalous effects have been established: The temperature coefficient of the resistance is negative, conductivity is non-Ohmic (electric field dependence is nonlinear), and there is a pronounced temperature-dependent magnetoresistance. The anomalous temperature dependence of the resistivity ρ is logarithmic, i.e., $\Delta\rho \propto \ln T$ (see, e.g., Fig. 2) for a certain range in $R_\square$ as indicated in Fig. 1. This behavior is expected in 2D systems by both the localization and interaction theories.[6,7] The constant of proportionality α' (in $\Delta R_\square / R_\square^2 = e^2/2\pi^2 n\alpha' \ln T$) was found to be $\alpha' = 1.1\pm 0.1$ in the above range and showed no systematic dependence on resistivity. [Note that in the last expression α' stands for αP or $(2F-2)$ for the localization and interaction pictures, respectively.] As $R_\square$ increases above 5 kΩ, the $\ln T$ dependence of the resistivity is replaced by a faster variation that eventually takes the form $\rho \propto \exp(T_0/T)^{1/2}$, an evolution that may be suggestive of the relevance of the localization picture. Further evidence for a qualitative change of behavior at $R_\square \sim 10^4$ Ω comes from studying

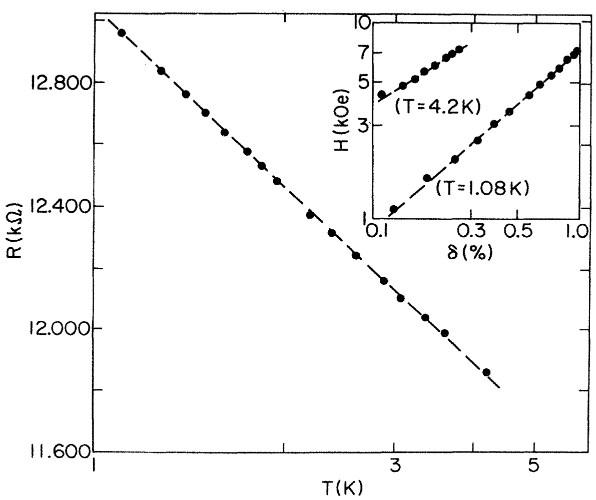

FIG. 2. Resistance vs temperature for a Nb film with $R_\square = 4.56$ kΩ (at 4.2 K). Inset shows positive MR for the same Nb film at 4.2 and 1.08 K (field perpendicular to the film's plane).

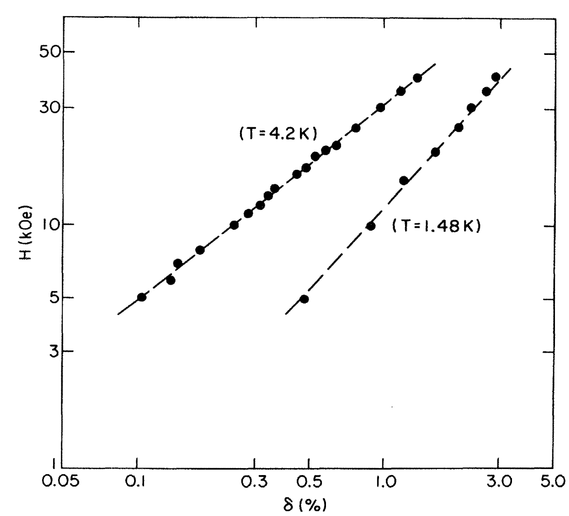

FIG. 3. Positive MR as a function of field for a Nb film with $R_\square = 11.8$ kΩ (at 4.2 K) measured at 4.2 and 1.48 K (field perpendicular to film surface).

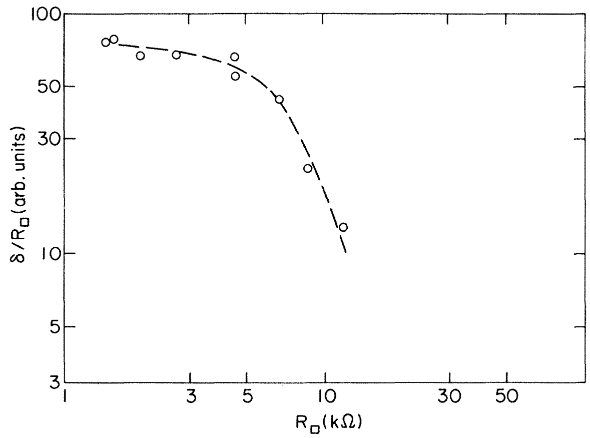

FIG. 4. Magnitude of the MR at 4.2 K and 7 kOe vs sheet resistance.

the magnetic field dependence of the conductivity which we will describe next.

Typical MR results are shown in the inset of Fig. 2 and in Fig. 3 for two samples that are characteristic of the anomalous regime. These results show the following systematic behavior.

(1) The fractional change of the resistance with field, $\Delta R/R \equiv \delta$, scales as H^{η} over a considerable range of fields and, usually, shows no sign of saturation or crossover to a slower variation up to very high fields (Fig. 3).

(2) The exponent of this power-law dependence, η, is temperature dependent and has the typical values of 1.5 ± 0.1 and 1.0 ± 0.1 at 4.2 and 1.1 K, respectively (Fig. 3).

(3) There is a definite amount of anisotropy in the results; $\Delta R/R$ for a perpendicular field is larger by about 20% than that for a parallel one.

(4) The magnitude of the effect depends on $R_\square$ as shown in Fig. 4. Note the sharp drop at $R_\square \approx 10$ kΩ, reflecting presumably a qualitative change of behavior. Indeed, with a further increase in $R_\square$, the MR becomes negative (Fig. 1).

These observations raise several questions. In particular, the magnetic field dependence is confusing.[11] All of the presently known theories[8,12–14] for the anomalous MR in 2D predict that δ should scale like H^2 for small fields and $\delta \propto \ln H$ for sufficiently large fields.

It is remarkable that similar MR results were recently reported for thin Pt films.[15] The fact that the MR is positive does not help, in this case, to identify the underlying mechanism. Both electron correlations[8,14] and localization effects[8,12] can yield such a result (for the latter mechanism one must assume the presence of strong spin-orbit scattering which, indeed, is very likely in this material). Furthermore, the anisotropy observed is neither weak nor sufficiently prominent to unambiguously support either possibility.

We have tried to fit the data to the expression[8]

$$\frac{\Delta R}{R} = g(T,H)\frac{e^2}{2\pi^2\hbar} Y\left[\frac{2DeH}{\pi c T}\right], \qquad (1)$$

where

$$Y(X) = \begin{cases} \ln X, & X \gg 1 \\ \sim 0.3X^2, & X \ll 1 . \end{cases}$$

Here, $g(T,H)$ is an effective coupling constant that has the following dependence for high fields:[8]

$$\frac{1}{g} = C - \ln\frac{DeH}{c\eta}, \qquad (2)$$

with η as a cutoff parameter, that is, in the present case, probably of the order of the Debye frequency ω_D. This procedure did not result in a satisfactory agreement. In particular, the observed functional dependence of δ on H could not have been reproduced. It appears that a special $g(H)$ is needed to account for the data if, indeed, Eq. (1) is the proper expression to be used in this case.

It may be possible to draw qualitative conclusions from these data on empirical grounds. For example, the fact that the magnitude of the anomalous MR rapidly diminishes at $R_\square \sim 10$ kΩ (see Fig. 4) might be related to the theoretically expected crossover from weak to strong localization behavior. It is also noteworthy that Fig. 4 depicts a dependence which is quite reminiscent of the way superconductivity is expected to be suppressed.[9] However, except for the observation that T_c seems to vanish at the transition to insulating behavior, there is no conclusive evidence that localization effects play an important role in suppressing superconductivity in our films. Note that T_c is already markedly depressed at $R_\square \sim 100$ Ω, at which point the localization length ξ_L is of the order of (Ref. 5) $\sim 10^4$ Å and is thus considerably larger than $\xi_S \sim 50$ Å. The other length that may be relevant to this problem, the inelastic mean free path l_{in}, is probably much smaller than ξ_L at the temperatures we are concerned with. Judging from the temperature where 2D behavior (i.e., logarithmic temperature dependence of the resistance) sets in, we approximately estimate l_{in} to be 50 Å at 15–20 K for a film with $R_\square \sim 1$ kΩ.

Thus, unless l_{in} has an unexpected dependence on R_0, this length should also be larger than ξ_S for all of our films with $R_\square < 1$ kΩ. These observations make it somewhat implausible to associate the mechanism for the suppression of superconductivity with localization effects in this range of resistances. The presumed role played by the restricted dimensionality in reducing the condensation energy must apparently be sought in other directions. For example, the question of electron screening in 2D systems might be of particular relevance here since it affects superconductivity in an obvious way. It is also one of the metallic properties that could be quite sensitive to the restricted dimensionality involved. Other possibilities, such as modifications of the density of states and the electron-phonon coupling strength, due to disorder, should also be considered.

To summarize, the transition temperature into the superconducting state of ultrathin Nb films is logarithmically depressed with the sheet resistance. The point where T_c vanishes apparently coincides with the "weak-to-strong localization" crossover point. In the vicinity of this point, several anomalous transport properties that may be associated with 2D localization and interaction effects are observed. The origin of the anomalous magnetoresistance observable in this range, as well as the mechanism for T_c suppression in the low-$R_\square$ regime remain enigmatic and clearly deserve further studies.

One of us (Z.O.) gratefully acknowledges instructive discussions with D. U. Gubser and S. A. Wolf. Work performed at Brookhaven National Laboratory was supported by the Division of Materials Sciences, U. S. Department of Energy, under Contract No. DE-AC02-76CH00016. The work of S. M. was supported in part by Fundação de Amparo à Pesqusa do Estado de São Paulo.

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