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Superconductivity enhancement in screened ultrathin metal films

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Abstract. We have studied superconductivity in thin quench-condensed bismuth films deposited on a 3 nm oxide layer covering a thick aluminium film. It was found that these Bi films have superconducting transition temperatures significantly larger than those of similar films deposited directly on glass. The enhancement of superconductivity is attributed to partial screening of the Coulomb interaction between conduction electrons in the bismuth films. The proximity of the thick metal film, even without any electrical contact to the thin film, reduces the interaction quantum correction to conductivity, the effect responsible for suppression of superconductivity in high-resistance films. However, the enhancement is less than predicted by the Finkelstein theory. It was also found that screened films showed an increased resistivity, a result attributed to reduced electron–electron scattering in the weak-localization quantum correction to conductivity.

1. Introduction

In our pilot experiments [1] it was shown that deposition of a thick gold film on top of thin anodized tantalum films resulted in an increase of the critical temperature of superconductivity of the thin film. Image charges in the bulk metallic conductor provided screening of the Coulomb interaction in the thin film, thereby reducing the repulsive component of the electron–electron interaction V and restoring $N(0)$, the density of states at the Fermi level. Both act to increase T_c as can be seen from the BCS result, $T_c \propto \exp[-1/N(0)V]$.

In those experiments, the largest observed T_c-enhancement was of the order of 0.02 K for 100 Ω films (resistance 'per square'). One could not expect larger values of T_c-enhancement because the films were prepared by ion-beam sputtering onto substrates held at room temperature. Such films start to conduct rather late, at mean thicknesses of the order of 5 nm or more and are expected to be of non-uniform thickness. To see greater superconductivity enhancement, much thinner conductive films are needed. Very thin, uniform (disordered) films may be prepared by quench condensation onto substrates held at liquid helium temperature. It is known that such films can become conductive at near-monolayer thicknesses. Bismuth was chosen as the superconducting material for our experimental structures. It is relatively easy to evaporate and has a convenient critical temperature, which is why it has been studied in a number of experimental works on quench-condensed thin films [2–6] which provide plenty of primary data with which to compare our results. Quench-condensed Bi is a good metal with a bulk superconducting transition temperature of about 6 K.

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2. Experimental details

Films were prepared in a small stainless steel cell, completely immersed in liquid helium. The cell contains a sample stage with a thermometer and a heater which allows the temperature of the samples to be raised above that of the surrounding helium. There are two miniature evaporation sources, one for Bi and one for Ge. A very thin layer of Ge, about one monolayer, was usually evaporated before the Bi as there is evidence [2] that this reduces the thickness at which bismuth films start to conduct. The sources required less than 1 W of power during evaporations and this caused the substrate temperature to rise by less than 0.5 K. However, most of the films used in these studies were prepared at a substrate temperature just above 6 K to ensure that they did not become superconducting during evaporation, allowing their changes of resistance to be monitored. The cell also contains an electromagnetically operated shutter to allow pre-evaporations from the sources before deposition of experimental films, and a quartz crystal, driven by specially developed helium-temperature electronics, to monitor film thicknesses during deposition. Film geometries were defined by a mask and electrical measurements were made via fine-wire connections passing through the mask to contact pads previously deposited at room temperature. Film resistances were measured by electrometers in the range $R = 10^{12}$–10^{4} Ω and by lock-in amplifiers at $R < 10^{6}$ Ω. To facilitate extraction of the effects of screening, two films were evaporated simultaneously, one screened and the other without screening (or with a different screening configuration).

As a degassing procedure after final assembly, the cell was held at about 100 °C under a vacuum of about 10^{-4} Pa for an hour or so. A needle valve on the chamber was then closed and the chamber completely submerged into the liquid helium bath. The residual pressure was estimated to be less than 10^{-9} Pa.

In these experiments, the bulk metal can only screen electron–electron interactions which involve spatial scales greater than the thickness of the insulating layer separating the bulk metal from the experimental film. Information about the spatial scale of the electron–electron interaction is therefore obtained by comparing results with different insulator thicknesses. We used two substrate configurations. The first consisted of a Ta disc which was anodized to different oxide thicknesses in the areas of the two samples. Here, the Ta itself provides the screening electrode. The second used a glass substrate on which an Al film of thickness about 100 nm was deposited at room temperature over the area where one of the pair of experimental films was to be deposited. The Al was then allowed to oxidize naturally for a couple of days. This produced the thinnest usable insulator layers, about 3 nm thick [7] which provided insulation exceeding 10^{12} Ω at helium temperature.

3. Experimental results and discussion

The maximum measurable resistivity in our experiments was about 10 TΩ. For Bi films deposited on a submonolayer of Ge, this corresponded to a mean Bi thickness of about 1.5 nm. This is a typical thickness for the onset of measurable conductivity with Bi films deposited on an insulator substrate [1, 4]. It is known that this value can be significantly reduced by coating the substrate with a thicker underlayer of Ge [5, 6]. We have found that fast evaporation of Bi on an underlayer of thickness of 0.6–0.8 nm provided us with conductive films at mean thicknesses as low as 0.5 nm but fast evaporation makes it more difficult to reach a desired resistivity. An added complication of the Ge is that there is uncertainty about its role. Thin Au films, quench condensed on a Ge underlayer, show superconductivity at low Au thicknesses indicating formation of superconducting

Au–Ge before the superconductivity is suppressed by increasing Au content [8, 9]. This implies that interdiffusion occurs during deposition despite the low substrate temperature. Presumably the kinetic energy of the incident species provides the necessary activation for the interdiffusion. Clearly, the Ge underlayer cannot be considered as inert with regard to the conductivity and superconductivity of the film deposited on it [5]. The fear of such complications was an added reason for avoiding the use of thick Ge underlayers.

All of the films discussed below had the onset of superconductivity at thicknesses 1.8–2.0 nm and resistivity of order 15 kΩ. Films with resistivity greater than 20 kΩ showed no superconductivity and an activated dependence of conductivity on temperature. Results with very-high-resistance films are described elsewhere [10], but it is worth mentioning here that the $R(T)$ dependencies as well as I–V characteristics of screened and unscreened films differed significantly, demonstrating suppression of the Coulomb gap by the presence of the screening layer.

The first sets of experiments used anodized tantalum substrates with oxide layers of different thickness, 10 nm for the thinner and 100 nm for the thicker oxide. These thicknesses were chosen because a characteristic distance between electrons participating in the electron–electron interaction effect, namely the thermal length $\sqrt{Dh/\pi kT}$, lies between these lengths. (D is the electron diffusion coefficient and the other symbols have their usual meanings.) However, no noticeable change of the critical temperature or of the resistances of the films were detected in these first experiments implying that contributions to electron–electron interactions at length scales between or greater than these oxide thicknesses were unimportant. We had to turn to much thinner oxide layers and used the glass/aluminium structures described earlier.

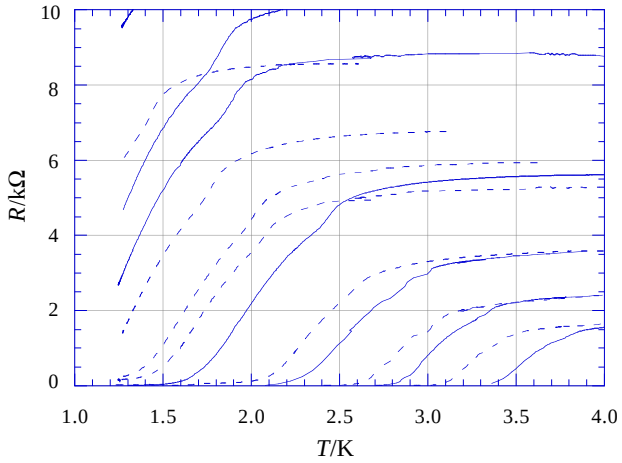

Figure 1. Superconducting transitions in the high-resistance region. Solid lines refer to films deposited on an oxidized Al film (screened films); broken lines refer to films deposited directly on glass (unscreened films).

An overview of superconducting transitions in the high-resistance region of superconductivity is shown in figure 1. It is apparent that screened films (solid lines) have noticeably higher critical temperatures than unscreened films of the same sheet resistance deposited directly on glass (broken lines). (The sample electrode configuration used in these measurements provided screening across most but not all of the 'screened' films and this produced the double transitions visible in the plot.) Thus, the presence of bulk metal at a

distance of a few nanometres from the experimental films does increase T_c without any direct electrical or tunnelling contact between them. The results suggest that the characteristic distances responsible for suppression of superconductivity are mostly smaller than the 3–10 nm thicknesses of our insulating layers, which is smaller than the typical distance between electrons believed to participate in the quantum interaction effect $\sqrt{D\hbar/\pi kT} \approx 50$ nm. It is worth remembering that no difference of T_cs was found between the films deposited on 10 nm and 100 nm tantalum oxide.

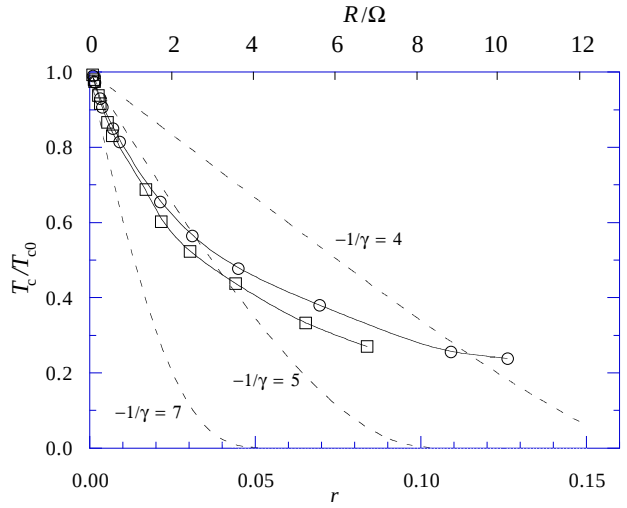

Figure 2. The dependence of critical temperature on sheet resistivity R for screened (circles) and unscreened (squares) films. The broken lines represent predictions of the Finkelstein formula [11] for suppression of superconductivity by interaction effects in two-dimensional disordered systems. The lower horizontal axis is the reduced resistivity, $r = Re^2/2\pi^2\hbar$.

The dependence of critical temperature on sheet resistance for screened (circles) and unscreened (squares) films is shown in figure 2. The theoretically predicted lowering of T_c from the bulk value T_{c0} caused by Coulomb interaction in two-dimensional systems is given by the Finkelstein formula [11]

$$\frac{T_c}{T_{c0}} = \exp\left(-\frac{1}{\gamma}\right)\left[\left(1+\frac{(r/2)^{1/2}}{\gamma - r/4}\right)\Big/\left(1-\frac{(r/2)^{1/2}}{\gamma - r/4}\right)\right]^{1/\sqrt{2r}}$$

where $r = Re^2/2\pi^2\hbar$ is the reduced film sheet resistivity (i.e. r is measured in units of $2\pi^2\hbar/e^2 \approx 81$ kΩ) and $\gamma = 1/\ln(kT_{c0}\tau/2\pi\hbar)$ characterizes the ratio of the bulk critical temperature and the elastic scattering frequency τ^{-1}. For films with relatively low resistivity (of the order of a hundred ohms) this formula can be approximated as

$$\frac{T_{c0} - T_c}{T_c} = \frac{Re^2}{12\pi^2\hbar}\left(\ln\frac{2\pi\hbar}{kT_{c0}\tau}\right)^3.$$

The theory has no free fitting parameters and predictions depend on only one material parameter, τ^{-1}, the elastic scattering rate of the electrons.

The broken lines in figure 2 are plots of the Finkelstein predictions for different values of γ. Obviously none gives a satisfactory fit with the data. The curve for $\gamma = -1/7$ is matched to the initial gradient, while the other values are chosen to straddle the data. It is not just the shapes of the 'fits' that present problems; the parameters are also quantitatively

unsatisfactory. The value $\gamma = -1/7$ gives $\tau \approx 10^{-14}$ s which, with a typical Fermi velocity, gives an electron mean free path of about 10 nm, which is about five times the film thickness. This seems too large since, regardless of scattering in the bulk of the film, there must be diffuse scattering from the surfaces. The other theoretical curves give even larger values of the elastic scattering length.

It is clear that the results are not consistent with the Finkelstein theory. We must ask why this might be the case. The theory applies to a homogeneous two-dimensional disordered system of uniform thickness. Recalling how the onset of conductivity depends on deposition conditions, it seems most unlikely that our films are uniform and homogeneous. We suggest that we are seeing percolation of superconductivity in films which are discontinuous ('island films') at lower thicknesses. The fact that superconductivity (and resistance) still depend on screening is entirely consistent with interpretation of the data in terms of a percolative transition since Coulomb energies will still be important on the metal/superconducting side of the transition. This is discussed further below.

That we first form discontinuous films is also consistent with the way that establishment of connectivity depends on evaporation rate. Similar behaviour is seen in thin films deposited at room temperature: it is well known that slow deposition (at normal temperatures) promotes growth of large islands before connectivity is established while rapid deposition, by giving less time for atomic rearrangement, produces more uniform films which become connected at lower mean thicknesses. It appears that similar processes occur in our films despite the low substrate temperatures during deposition: quench condensation does not totally suppress diffusion of incident species. The dependence on evaporation rate in our experiments cannot be explained in terms of slow surface diffusion because that will be totally frozen out at such low temperatures. However, incident atoms will diffuse for a short time after impact while they are still hot. The loss of their kinetic energy is likely to take several inverse Debye frequencies during which time they may diffuse several interatomic distances. Given the small spatial scale of our structures, such distances are important. Then what is the connection with deposition rate? We suggest that slow evaporation rates allow more time for release of adsorbed species from surfaces close to the sources and heated by them. The increased diffusion may then come from increased contamination of the Ge surface resulting in reduced binding for the incident Bi atoms. This explanation is consistent with the facts that film properties are totally stable once evaporation is stopped, and they are also stable if the sources are hot, provided that their temperature is not high enough for evaporation of more Bi. The difference has to be associated with the fate of incident Bi atoms after reaching the substrate.

As regards the resistances of the film, one interesting point should be noted. The screened films always have resistances which are slightly *larger* than those of the corresponding unscreened films. The difference of inverse resistance is of the order of the typical logarithmic correction, $e^2/2\pi^2\hbar$. The same was seen in the tantalum films in our pilot experiments. One might expect the opposite: if screening simply suppresses the electron–electron interaction part of the quantum corrections, the resistances should decrease. This assumes that screening does not affect the localization part of quantum corrections, which is treated in terms of interference of non-interacting electrons in the disordered system. However, electron–electron interaction does have an indirect influence on localization, and the effect is not necessarily small. The total value of localization correction to conductivity is determined by the upper limit of spatial integration of contributions to the correction, whose limit is the phase-breaking length. At low temperatures, this length is determined by inelastic electron–electron scattering. In the three-dimensional case, this scattering frequency is proportional to the ratio of the square of the temperature to the

Fermi energy, $\hbar\tau_i^{-1} = (kT)^2/\epsilon_F$. But, in the case of a two-dimensional system like a film, this scattering mechanism is greatly enhanced [12] and the dependence becomes linear with temperature: $2\pi\hbar\tau_i^{-1} = kT(e^2R/2\pi^2\hbar kT)\ln(\pi\hbar/e^2R)$. Even larger scattering rates, of the order of temperature $\hbar\tau_i^{-1} \approx kT$, have been found experimentally for ultrathin Cs and Rb films [13]. This means that reduction of Coulomb interaction can cause the localization part of quantum corrections to become much larger and that it will change with temperature as T^2 and not just as T. This results in larger resistance and larger logarithmic slope of the $R(T)$ curves. The total resistance behaviour is, of course, a superposition of both localization and interaction quantum corrections and depends on the interplay of diffusion and Cooper scattering channels, which in turn depends on the ratios of inelastic, spin, spin–orbit and thermal lengths. Screened Bi films in these experiments had a logarithmic slope approximately 50% larger, while the Ta films of our pilot experiments had a slope which was not only smaller but also of reversed sign when screened, consistent with the presence of weak *anti*localization [1].

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