

Superconducting properties of highly disordered bismuth films

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(Submitted 12 June 1987)

Pis'ma Zh. Eksp. Teor. Fiz. **46**, No. 2, 84–87 (25 July 1987)

Deposition of bismuth films through a layer of superfluid helium has resulted in the synthesis of films with a resistivity on the order of $3 \times 10^{-3} \Omega \cdot \text{cm}$. The superconducting properties of these films, including the critical temperature, are quite different from those of ordinary bismuth films synthesized by cold deposition.

The film deposition technique¹⁾ used in the present experiments is similar to that which we used previously in a study of cadmium films,¹ but an improved design of the apparatus made it possible to lower the deposition temperature to 0.46 K. Furthermore, the glass substrates were replaced by substrates of polished sapphire, and a film heater deposited on the opposite side of the substrate was used to heat the substrate, after the cooling of the apparatus, to a temperature of about 400 °C in high vacuum. Only after these steps was the helium admitted. This procedure made it possible to clean the substrate just before the deposition.

The bismuth is evaporated at a temperature of 10^3 K. The velocity of the atoms moving toward the surface of the liquid exceeds the sound velocity in the liquid helium. The mean free path of these atoms in the liquid should be short, and the atoms should be stopped quickly through the emission of quasiparticles. A stopping of this sort can occur until the atom is slowed to the critical velocity for superfluid motion, $v_c = 60$ m/s. Bismuth atoms with velocities $v < v_c$ in superfluid helium can interact only with normal excitations, but at such low temperatures the concentration of rotons is low, while the interaction with phonons is inconsequential because of their small momentum and small scattering cross section. It can therefore be assumed that the

situation which prevails in our experiments is one in which the bismuth atoms lose most of their energy in a thin layer of helium near the surface and then move as free particles through the superfluid liquid.

Although ordinary bismuth films synthesized by cold deposition have been studied thoroughly, we decided to repeat the experiments in order to test the procedure. Deposition was carried from the same evaporator, in the same apparatus, as for the deposition through the liquid helium, but in these particular experiments the helium was not admitted. In order to establish thermal contact between the substrate and the helium bath, we admitted liquid helium underneath the substrate, mounted with a hermetic seal. The temperature of this liquid helium was about 0.4 K during the deposition. A conductivity arises in vacuum films of this sort at a thickness²⁾ $d = 10 \text{ \AA}$. The temperature dependence of the resistance of comparatively thin and high-resistance films is of a semiconductor nature, and no superconducting transitions occurs in these films. For example, a film with $d = 17 \text{ \AA}$ had a resistance $R_{\square} = 23 \text{ k}\Omega$ at $T = 2.2 \text{ K}$ and $50 \text{ k}\Omega$ at $T = 0.4 \text{ K}$. An increase in the thickness to 17.5 \AA gave rise to a superconducting transition, at a temperature which increased with increasing film thickness (Fig. 1). The observed behavior of the critical temperature as a function of d and R_{\square} agrees well with data obtained in other studies.²⁻⁴

Completely different results were found in the case of deposition through liquid helium. In the first place, a conductivity on the order of 10^{-11} S arose in these films

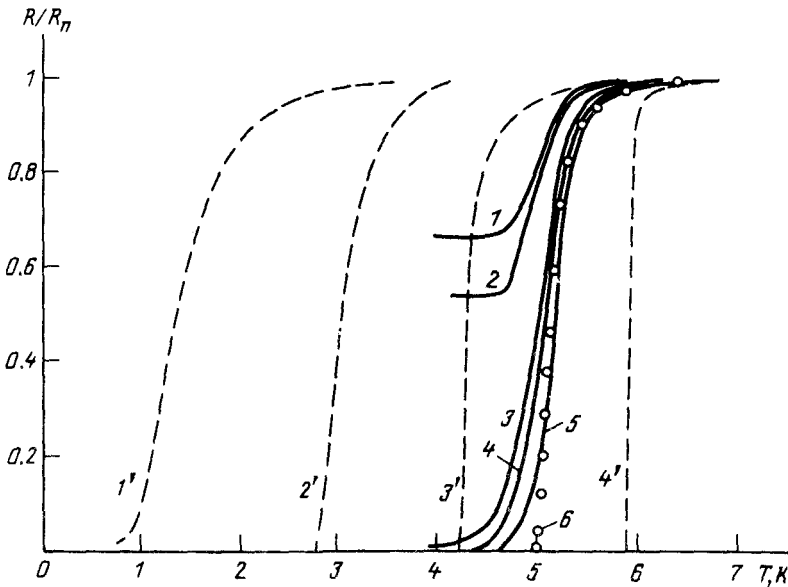


FIG. 1. Superconducting transitions of the bismuth films. Dashed lines: films synthesized by vacuum deposition. Solid lines: films deposited through helium. 1'— $d = 17.5 \text{ \AA}$, $R_n = 13 \text{ k}\Omega/\square$; 2'— $d = 23 \text{ \AA}$, $R_n = 2.4 \text{ k}\Omega/\square$; 3'— $d = 33 \text{ \AA}$, $R_n = 850 \text{ k}\Omega/\square$; 4'— $d = 113 \text{ \AA}$, $R_n = 62 \text{ k}\Omega/\square$. 1— $d = 123 \text{ \AA}$, $R_n = 18.6 \text{ m}\Omega/\square$; 2— $d = 124 \text{ \AA}$, $R_n = 7.4 \text{ m}\Omega/\square$; 3— $d = 131 \text{ \AA}$, $R_n = 350 \text{ k}\Omega/\square$; 4— $d = 136 \text{ \AA}$, $R_n = 60 \text{ k}\Omega/\square$; 5— $d = 183 \text{ \AA}$, $R_n = 2.5 \text{ k}\Omega/\square$; 6— $d = 233 \text{ \AA}$, $R_n = 1.1 \text{ k}\Omega/\square$.

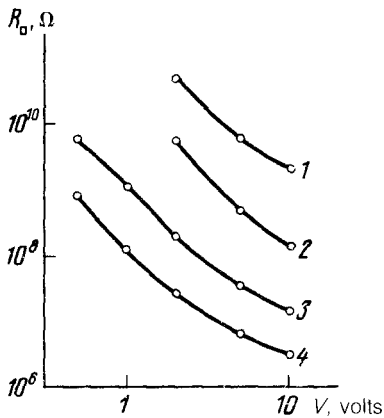


FIG. 2. Voltage dependence of the resistance of the films at $T = 1.7$ K. 1— $d = 120$ Å; 2— $d = 121.1$ Å; 3— $d = 122.4$ Å; 4— $d = 124$ Å.

only at $d \geq 120$ Å (we studied square films with dimensions 1.5×1.5 mm²). The resistance of the high-resistance films depends strongly on the applied voltage and falls off rapidly with increasing thickness (Fig. 2). Films with a resistance $R \sim 2 \times 10^5$ Ω were superconducting with a critical temperature $T_c = 5$ K and a resistance $R_s = 0$ below T_c .

We deposited a total of four series of films of various thicknesses. Film 1 was deposited initially to $d = 127$ Å; its resistance at $T = 0.46$ K was 300 kΩ. Raising the temperature only slightly, to 0.6 K, resulted in an irreversible 15% decrease in the resistance of this film. Films 2 and 3 were deposited to $d = 123$ Å; this thickness is

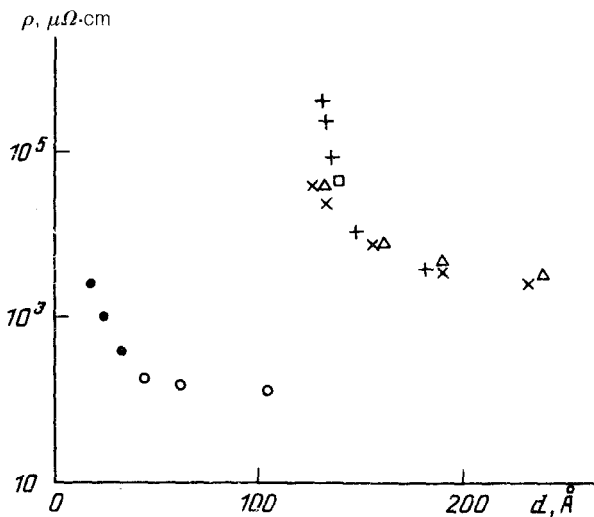


FIG. 3. Thickness dependence of the resistivity of the films at $T > T_c$. ●, ○—Films synthesized by vacuum deposition; □—film 1; ×—film 2; △—film 3; +—film 4.

slightly greater than the thickness at which the conductivity appears. These films were heated to $T = 6$ K, and then their thicknesses were increased by successive deposition steps. After each of these further deposition steps the films were heated, and the superconducting transition was recorded (Fig. 1). The superconducting transitions of the various films with identical values of R_n were the same. Film 4 was heated to 6 K after the deposition of the 60-Å layer; we observed no significant conductivity here. We then carried out some further deposition steps, each of about 10–15 Å, and heated the film after each step. A significant conductivity appeared in this film only at $d = 120$ Å.

The films with a resistance $R > 10^8 \Omega$ have a purely semiconductor $R(T)$ dependence. Reducing R to $(2-4) \times 10^7 \Omega$ causes the $R(T)$ dependence to become nonmonotonic; this is evidence of superconducting properties. If a comparatively high potential difference V is applied to such films, the $R(T)$ dependence essentially disappears at $T < T_c$ and $T > T_c$, and the superconducting transition takes the form of a characteristic step, with a significant and temperature-independent R_s (curves 1 and 2 in Fig. 1). The film with $d = 130$ Å has $R_n = 350$ k Ω and $R_s = 1$ k Ω ; a 1-Å increase in the thickness of this film reduces R_n to 200 k Ω and causes R_s to vanish.

The appearance of a conductivity in films with $d > 100$ Å during the deposition is very probably due to a change in the structure of the film as its thickness is increased. Each new atom which arrives at the surface of the film should cause some displacement of its nearest neighbors. Their displacements in turn cause displacements of more remote atoms; and so forth. These structural changes should be particularly noticeable in highly disordered and fairly thick films,³⁾ in which the position of each atom corresponds to a local minimum of the energy, set by the relative arrangement of the others. In films with a self-consistent arrangement of atoms of this sort we would naturally expect a significant annealing out at low temperatures.

The existence of a superconductivity with $R_s = 0$ in films with a resistance $R_{\square} \gtrsim 10^5 \Omega$ contradicts essentially all of the existing theoretical models of two-dimensional structures,⁵⁻⁷ which predict the disappearance of the superconducting transition at $R_{\square} \sim 10^4 \Omega$ as a result of localization effects. Nevertheless, a superconductivity in high-resistance films has also been observed previously in cold-deposited films of mercury⁸ and also in cadmium films deposited through liquid helium.¹ In all these cases, T_c has been essentially independent of the thickness.

All of the films which exhibit superconducting transitions at $R_{\square} \sim 10^5 \Omega$ are distinguished by their significant thickness and anomalously high resistivity ρ (Fig. 3). It turns out that the values of ρ of the films deposited through the helium are approximately 30 times the values of ρ of ordinary cold-deposited films. This result is evidence that these films are highly disordered. In these films we are apparently dealing with a situation in which the mean free path of the electrons is comparable to the distance between atoms. If this is the case, then these films cannot be regarded as two-dimensional, since the time scale for the diffusion of electrons from one film surface to the other may exceed the time scale of inelastic scattering. Films with $R_{\square} \sim 10^5 \Omega$, which should have been localized if they were two-dimensional, may constitute unlocalized three-dimensional entities.

We wish to thank Yu. V. Sharvin for many useful comments and E. G. Astrakharchik for a discussion of these questions.

¹A detailed description of the experimental procedure will be published separately.

²Actually, we measured the number of atoms per unit surface area of the substrate and converted this value into a thickness by making use of the density of bulk crystalline bismuth.

³Effects of this type should be substantially smaller in thin films because of the stabilizing effect of the fixed substrate atoms on the film structure.

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Translated by Dave Parsons