Electric phenomena in quench-condensed mercury film in the activation-conduction region

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The electric-field and temperature dependence of the resistance of high-resistance mercury films are investigated. It is shown that at a resistance \( R > 5 \times 10^{12} \Omega \) the presence of superconductivity in film increases the resistance, and both the critical superconducting temperature \( T_c \) and the critical field \( H_c \) are constant in the resistance range \( R = 10^{-6} - 10^{10} \Omega \).

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Quench-condensed films of various metals have by now been investigated in sufficient detail (see, e.g., Refs. 1 and 2). The structures and properties of such films have much in common. It is known that when a molecular metal beam of sufficiently low temperature is condensed on a helium-temperature surface the film produced is continuous and has an amorphous structure with maximum disorder (metallic glass). The critical superconducting temperature \( T_c \) of such a film differs generally speaking from \( T_c \) of the bulk material. When heated to a certain temperature, which is different for different metals, the film changes from amorphous to crystalline. This transition is accompanied by a change of \( T_c \) as well by a severalfold decrease of the resistance (but by an increase of the resistance of bismuth).

In this respect the quench-condensed mercury films investigated by Tsymbalenko and Shal'nikov3 are no exception. A freshly deposited amorphous film with \( T_c = 4.07 \) K recrystallizes at a temperature close to 70 K into a phase having the critical temperature of \( \alpha \)-mercury (4.16 K), and the resistance of thick films (thickness \( d > 120 \) Å) increases by approximately 10 times. The behavior of thinner films, however, have singularities that distinguish them greatly from films of other metals.

The principal and most interesting singularities of thin mercury films is that they are dielectric with resistance \( R > 10^{12} \Omega \) down to a thickness of \( \sim 20 \) atomic layers \( d < 60 \) Å, Fig. 1). Films in the thickness range \( d = 60-70 \) Å are "semiconducting" in the sense that their resistance increases with decreasing temperature:

\[
R(T) = R \exp \left( \frac{\alpha}{kT} \right)
\]

where \( \alpha \) is a certain characteristic energy that depends on the film thickness. Such films have essentially nonlinear current-voltage characteristics. Also unusual is the result of annealing thin films. The recrystallization takes place at the same temperature \( \sim 70 \) K as in thick films, and into the same \( \alpha \)-mercury phase with \( T_c = 4.16 \) K, but the change of resistance is in this case much larger, by \( 10^9 \) times. The annealed films become metallic, the growth of the resistance with decreasing temperature gives way to a transition into the superconducting state, and the nonlinear current-voltage characteristics become ohmic. Another interesting fact is that the critical superconducting temperature of thin films is practically independent of thickness. In the resistance interval \( R = 10^{6} - 10^{12} \) \( \Omega \) [\( d = 70-200 \) Å] we have \( T_c = 4.05 - 4.07 \) K for freshly deposited films and \( T_c = 4.16 \) for annealed ones.

These unusual results calls for a thorough check on the procedure. Since the film pressure was calculated from the mercury vapor pressure at a known evaporator temperature and from the geometric dimensions of the device, under the assumption that the accommodation coefficient of the mercury atoms on glass is \( \alpha = 1 \) at liquid-helium temperature [i.e., that all the atoms adhere to the substrate], we investigated the temperature dependence of the accommodation coefficient.\(^4\) It was established that \( \alpha = 1 \) already at \( T = 80 \) K.

We studied also Cs and Rb films produced in similar devices by the same procedure.\(^5\) It was found that the films of these metals begin to conduct already at a thickness \( \sim 0.7 \) of an atomic layer. The transition to metallic conduction was observed at a resistance \( R = 10^6 - 10^{12} \) \( \Omega \), corresponding to a thickness of approximately one atomic layer. Additional investigations were also made of mercury films on substrates of different geometries and different materials (polished glass, fused glass, single-crystal germanium, single-crystal silicon). The condensation on the substrate was at various temperatures in the interval 4.2 - 1.8 K and at different rates \((0.001-1 \text{ A/sec})\). Some of the films were produced after several successive depositions at the same or different evaporation rates. Different device variants were also used with different degree of cleaning the device itself and of evaporating the mercury. By now, more than forty devices were constructed with which approximately 300 film-condensation...
experiments were performed. The films obtained in all the experiments had identical characteristics. Thus, the anomalously large thickness at which conduction sets in, observed in Ref. 3, is not a consequence of contamination or peculiarity of the procedure, but a property of the mercury film as a metal.

Mercury has a minimum state density on the Fermi surface and the largest resistivity of all metals. In addition, the resistance of mercury increases with decreasing density much more strongly than in other metals, and the density of the material in the amorphous film (frozen vapor) is substantially lower than the density of the crystalline and liquid material.13

These peculiarities of the electric properties of mercury cause the resistivity \( \rho \) of the material in cold-deposited mercury films to be much higher than in films of other metals at the same electron mean free paths \( l \). The values of \( \sigma = \rho^{-1} \) and \( I \) can be estimated from the dependence of the film resistance on the thickness at large thicknesses, when

\[
\sigma / \rho \sim (1/e^2)/(l/d) (1-p),
\]

where \( \sigma \) is the conductivity of the film and \( p \) is the specularly coefficient of the electrons scattering from the surface. Comparison of the ratios \( (2) \) under the assumption that the specularity coefficient \( p \) is the same for Hg, Cs, and Rb films produced11 on identical substrates and under similar conditions shows that \( \sigma^2 = 1/a \) (a is the interatomic distance), which characterizes the effective density of the states at the Fermi level, is 16 times smaller for mercury than, say, for rubidium. The mean free path in these films is equal to several dozen interatomic distances. Since according to present-day theoretical premises it is precisely the resistance which is a good parameter that determines the properties of the electronic states, particularly the transition to exponential localization, it can be assumed that the large thickness at which a transition into metallic conduction occurs in mercury films is due to the high resistivity of the material in the amorphous mercury films condensed at helium temperature. It must noted right away that in a film with large thickness corresponding to the metal-dielectric transition the electrons cannot be localized at distances smaller than the film thickness even in the region of exponential localization, for this would mean three-dimensional localization that requires much smaller mean free paths.1 This means that such a film can be superconducting, since the electrons are localized, at any rate, not more strongly than in metal drops measuring ~ 20\( \mu \). It is known from experiment12 that in drops of this size \( T \), still does not decrease noticeably.

The situation is similar in the one-dimensional case, in which exponential localization is possible.6 Diffuse scattering of the electrons by the surface can lead here, too, to localization, with high values of the characteristic energy, under conditions when superconductivity can still exist. Imagine a wire of diameter \( d = 10\mu \). An electron has in it a mean free path \( l = d/l = d/10\mu \) and is localized in a length \( L \sim 300\mu \). The wire resistance at sufficiently low temperatures increases in accordance with \( (1) \), at an activation energy \( \varepsilon = \varepsilon_0 d^2/4 L \), on the order of several degrees (\( \varepsilon_0 \) is the Fermi energy). Nonetheless, as is known from experiments, in drops of size \( d = 10\mu \) superconductivity can still exist. The character of the change of the conductivity at low temperatures \( \sigma \rightarrow 0 \) or \( \sigma \rightarrow \infty \) as \( T \rightarrow 0 \) of such a system is determined by the competition between localization and superconductivity. It is possible that some phenomena described below in the article are connected precisely with the coexistence of exponential two-dimensional localization and conductivity in the mercury film.

A possible argument against homogeneity of mercury films with \( d \sim 20\mu \) is that such a film can be of the island type and consist of drops with characteristic dimensions of the order of the film thickness. But such a situation presupposes high mobility of the atoms on the substrate surface, or at least an ability of the mercury atom to move over a considerable length, several dozen interatomic distances, after striking the substrate. This possibility cannot be rejected a priori, but it must be noted that no one observed such phenomena at helium temperatures in any metal. In addition, this model cannot explain the behavior of annealed films. It is difficult to imagine that drops of mercury, a metal that wets glass poorly, would start to flow over the surface and increase the conductivity of the film. The assumption that the film has an island structure is contradicted also by the fact that the decrease of the resistance upon annealing coincides with the transition into the \( \alpha \) phase of mercury. Ordering of an amorphous material requires much smaller displacements of atoms through smaller potential barriers than for motion of an atom over a substrate, and therefore also lower temperatures. In fact, drop formation in mercury films is observed at much higher temperatures (near the melting point of bulk mercury) and this process is accompanied by an irreversible increase of the resistance.

**EXPERIMENTAL PROCEDURE**

The basic device chosen is described in Ref. 3. It comprises a glass ampoule in the upper part of which is placed the helium-cooled substrate, and in the lower a mercury drop that serves as the evaporator (Fig. 2). Such a construction permits multiple deposition of a film in a single setup, by recondensing the mercury from the substrate back to the...
The evaporator temperature was measured with a copper-constantan thermocouple and was maintained constant within \( \pm 0.05 \) \(^\circ\)C in the course of condensation by an automatic control system. The temperature measurement accuracy was monitored in each experiment against the melting (solidification) point of the mercury. The total error in the determination of the film thickness did not exceed 5%.

The scatter of the thickness dependence of the resistance in the different experiments did not exceed several percent in thickness. The temperature rise of the produced film relative to the liquid-helium bath did not exceed 5 \( \times \) \( 10^{-7} \) K for glass substrates and 10\(^{-6}\) K for substrates of Ge and Si single crystals 250 \( \mu\)m thick. Figure 2 shows the geometry of some of the employed substrates. All the resistance values cited hereafter in the article and in the figures have been recalculated to quadratic substrate geometry (into \( \Omega \).

Particular attention was paid to the absence of contamination and of residual gases in the evaporated mercury and in the setup itself. The mercury was initially outgassed by multiple distillation in a separate device, and then distilled into ampoules that could be sealed. Typical preconditioning of the ampoule consisted of heating it for 4 hours at 400 \(^\circ\)C in a vacuum of \( 10^{-3}\) Torr. After the end of the preconditioning, the ampoule with the mercury, the latter in a separate stub, was broken with a magnetic striker and the necessary amount of mercury distilled into the device. The residual pressure in the sealed device did not exceed \( 10^{-5} \) Torr and remained unchanged during the entire time of its use. The assembled device was inserted in a helium cryostat through a sluice in the upper part of the cryostat. Upon immersion in the liquid helium, first to be cooled were the side walls of the device, whereas the substrate temperature, monitored with an Allen-Bradley resistance thermometer, and the evaporator temperature, did not exceed 70 K. Thus, all the residual gases were condensed on the side walls of the device. Calculation shows that their content did not exceed one monatomic layer.

The resistance of the high-resistance films, \( R > 10^{4} \) \( \Omega \), was measured at constant voltage (by determining the current) using an F118 nanovoltmeter. For convenience in reducing the results, the signal was displayed in digital form with an F30 voltmeter at the output of the F118 meter. To measure the current voltage characteristics we used a voltmeter divider made up of resistors rated \( R = 1 \) k\( \Omega \) \( \pm \) 2%, which made it possible to apply to the film a voltage 1 AS \( K \times 10^{-3} \) V, where \( k = 1, 2, \ldots, 10 \) and \( n = 1, 2, 3 \). The time of setting of the voltage at the 99% level after switching over was \( \sim 5 \) sec, and the current-measurement procedure lasted \( \sim 20 \) sec. The resistance of low-resistance films with \( R < 10^{4} \) \( \Omega \) was measured with an R309 potentiometer accurate to 0.01%. A magnetic field up to 60 kOe, perpendicular to the substrate plane was produced by a superconducting solenoid.

RESULTS OF EXPERIMENT

The use of a magnetic field made it possible to separate the phenomena due to the presence of superconductivity in the film. It should be noted right away that in all films the change of the resistance in a magnetic field up to 60 kOe at 4.2 K (i.e., above \( T_{c} \)), did not exceed several percent. This is incomparably less than the resistance changes observed at \( T < T_{c} \) and due to the suppression of the superconductivity by the magnetic field. It can therefore be assumed in first-order approximation that the presence of a magnetic field \( H > 10 \) kOe does not change the magnitude and character of the exponential localization, but destroys the superconductivity completely. Figure 3 shows typical temperature dependences of the critical field \( H_{c}(T) \) for a thick \( (R = 192 \) \( \Omega \) and thin \( (R = 1350 \) \( \Omega \) ) film. The thick film with \( d = 480 \) \( \AA \) has \( H_{c}(0) \approx 2.3 \) kOe, much less than the \( H_{c}(0) \approx 27 \) kOe for the thin film. This value \( H_{c}(0) \approx 27 \) kOe remains practically unchanged, together with the constant \( T_{c} = 4.07 \) K, in a wide range of resistances \( R = 10^{2} \) to \( 10^{6} \) \( \Omega \), but the results of transition into the superconducting state for films with \( R > 10^{5} \) \( \Omega \) are the direct opposite of those for \( R < 5 \times 10^{5} \) \( \Omega \).

Figure 4 shows typical curves of destruction of superconductivity by a magnetic field. It can be seen that the presence of superconductivity in the high-resistance film leads to a substantial increase of the resistance, whereas films with \( R < 5 \times 10^{5} \) \( \Omega \) have zero resistance at \( T < T_{c} \) and \( H < H_{c} \). In the magnetic-field interval \( H = 30 \) to 60 kOe the resistance is independent of the magnetic field accurate to several percent.

More complete information on the state of electrons in high-resistance films can be extracted from the current-voltage characteristics. Actually, the deviations from Ohm's law, even when weak electric fields \( E \) are used, are so large that the film resistance can be determined by extrapolating, as \( E \to 0 \), the relation \( R(E) = E/I(E) \), where \( I(E) \) is the current through a film one centimeter wide. We present below
FIG. 5. Resistance of film 68.2Å thick vs the electric field at various temperatures (from the upper curves to the lower): 1.27; 1.61; 1.93; 2.24; 2.58; 2.94; 3.33; 3.78; 4.19K. a-H = 0, b, c-H = 50 kOe. On the left in Fig. 5a is shown the measurement error for small values of E. The measurement error at E > 1 V/cm does not exceed the size of the experimental point.

The results of investigations of R(E) in the resistance region 10^{10}−10^{12} Ω, at temperatures T = 4.2−1.2 K, and electric field intensities E = 9−0.009 V/cm, in a magnetic field H = 50 kOe and in a zero magnetic field.

Figures 5 and 6 show typical experimental data on the R(E) dependences in high-resistance films as functions of the temperature (Fig. 5) and of the film thickness (Fig. 6). Figure 5 shows, for a film thickness d = 68.2 Å, plots of R(E) obtained at constant temperatures ranging from T = 4.2 K (lower curves) to T = 1.27 K (upper curves). Figure 5a demonstrates the resistance increase due to the appearance of superconductivity in the film, compared with the film resistance at the same temperature but in the presence of a magnetic field (Fig. 5b). Figure 5c shows the same data as in Fig. 5, but in coordinates log R and E^{1/2}.

Since the error in the determination of the resistance is connected mainly with the current-measurement error, it increases with decreasing electric field intensity E and with increasing resistance. Subject to the maximum error in these figures are the data in the upper left corner, as is manifest by the large scatter of the experimental points.

Deviations from Ohm's law in semiconductors and other systems with activation dependence of the resistance on the temperature are usually described by the Frenkel-Poole law

$$R(E) = R_0 \exp \left(\frac{-C}{E}\right)$$

where C is a certain constant and the exponent k = 1 for systems with an R(T) dependence in the form (1) and k = 1/4 for systems in which the Coulomb interaction between the electrons is substantial. As can be seen from Fig. 5b, in the absence of superconductivity (at H > H_c or T > T_c) the experimentally observed R(E) dependences are not described by the law (3) with k = 1, which should result in straight-line plots of log R vs E. Nor does a relation in the form (3) hold at k = 1/4. It has turned out, however, that within the limits of experimental error the relation (3) is well satisfied at the empirical value k = 1/4 in the entire temperature interval for films with resistance R > 10^6 Ω (Figs. 5c and 6).

An approximation of R(E) as E → 0 by Eq. (3) with k = 1/4 yielded the temperature dependences of the resistance. Typical results are shown in Fig. 7. They agree sufficiently accurately with (1), where R_0 increases with decreasing film thickness. Figure 7 shows also the temperature dependence of the coefficient C (Eq. (3)), which characterizes the degree of deviation from Ohm's law. The result can be represented in the form C(T) = C_0/kT. Thus, the dependence of the film resistance on E and T in the absence of superconductivity is described by the empirical formula

$$R(E, T) = R_0 \exp \left(\frac{k}{E} - \frac{C_0}{kT}\right).$$

The degree C_0 of the deviation of the resistance from Ohm's law increases with decreasing film resistance (Fig. 6), i.e., as the metal-dielectric transition is approached. Near the trans-
sition, a relation in the form (3) with \( k = \frac{1}{2} \) is observed only in weak electric fields. In fields \( E \gtrsim 10^{6} \) \( \text{V/cm} \), the saturation sets in and the field dependence of the resistance is weaker. Detailed measurements were not made in stronger fields, but it can be noted that the degree of deviation from Ohm’s law is independent of temperature in this region.

According to currently prevailing notions, the transition from activation to metallic conduction in two-dimensional systems should take place at \( R \sim 10^{-9} \) \( \Omega \). In mercury films, the presence of superconductivity increases noticeably the threshold value of the resistance. Thus, a threshold value \( R (E) \sim 2 \times 10^{5} \) was determined from the dependence of the activation energy \( E \) in Eq. (4). The activation energy in the weak fields were measured there in an electric field \( E = 10 \) \( \text{V/cm} \), so that the observed value of \( E \) was substantially lowered. Measurements of \( R (E, T) \) in the temperature interval \( T = 4.2-1.2 \) \( K \) in weak fields have shown\(^{10}\) that in such a superconductor-dielectric transition the lowest activation energy, \( E \sim 9 \) \( K \), was observed directly prior to the transition into superconductivity with \( R = 0 \) at \( T < T_{c} \), i.e., in films with \( R > 10^{6} \) \( \Omega \). Films with resistances \( 10^{7}-10^{8} \) \( \Omega \) had essentially nonlinear current-voltage characteristics in weak fields at \( T > T_{c} \), thus indicating an exponential localization, but became superconducting with zero resistance at \( T < T_{c} \).

Measurements performed in a magnetic field \( H = 50 \) \( \text{kOe} \) (Fig. 8) show that in mercury films, as well as in films of other metals, the transition to metallic conduction takes place in the absence of superconductivity at \( R \sim 10^{-9} \) \( \Omega \).

The values of the activation energy \( E \), marked by the black circles in Fig. 8, were obtained by reducing the \( R (E, T) \) relations in accord with Eq. (4) as \( E \rightarrow 0 \). Unfortunately, the instability of the films near the transition (at \( R < 10^{7} \) \( \Omega \)) made impossible detailed measurements in the entire range of electric fields \( E \) at different temperatures, and we had to restrict ourselves to measurement of \( R (T) \) at the minimum voltages, corresponding to a current \( \sim 10^{-4} \) A through the film (light circles in Fig. 8). These measurements definitely underestimate the values of \( E \), but give a qualitatively correct picture of the transition to the metallic conduction.

The instability of the films in the resistance interval \( R \sim 10^{-7}-10^{-1} \) \( \Omega \) is due to the very strong dependence of the resistance on the thickness. A decrease of the resistance by one order of magnitude takes place when the thickness is changed by only 0.4 \( K \) (by 0.6%). Such films begin to change their resistance irreversibly even when heated to \( 8-10 \) \( K \) and can break down when the film voltage is reversed, with their resistance decreased irreversibly by several dozen percent. The high sensitivity of the films in this resistance region to the metal structure has enabled us to observe also so weak an effect as the mobility of the atoms in the film at low temperatures. The resistance of freshly condensed films decreased in our experiments by an approximate factor 1.5 in one day. Such a resistance change, of course, does not mean possibility of a phase transition into a crystalline structure at high temperature, but offer evidence of a certain ordering of the amorphous phase, as manifest by the annealing of the most unstable defects.

Comparison of the \( R (E) \) plots in a magnetic field \( H = 50 \) \( \text{kOe} \) (Fig. 8b) and \( H = 0 \) (Fig. 8a) shows that the presence of superconductivity in a film not only increases substantially the resistance and the degree of deviation from Ohm’s law, but also alters the character of this deviation. The \( R (E) \) dependencies at low temperatures and at \( H = 0 \) are described within the limits of errors, in the entire resistance range \( R \sim 10^{6}-10^{7} \) \( \Omega \), by the Frenkel-Poole law (3) with \( k = 1 \).

When the superconductor-dielectric transition is approached \( (R \sim 5 \times 10^{7} \) \( \Omega \)) this dependence is observed only in weak fields \( (E \lesssim 0.1 \) \( \text{V/cm} \)). In the saturation region \( E \sim 1-10 \) \( \text{V/cm} \), the film resistance becomes independent of temperature. The films closest to the transition behave in fields \( E \sim 1-10 \) \( \text{V/cm} \), when the temperature is lowered, as superconductors with a residual resistance at \( T < T_{c} \), although in weak fields one observes a substantial increase of the resistance as \( T \rightarrow 0 \), well described by the activation formula (1) with \( E \sim 9 \) \( K \).

The dependence of the film resistance on \( E \) and \( T \) at \( H = 0 \) and \( T < T_{c} \) can be written in the form

\[ R(E, T) = R_{1}(E, T) \exp \left( \frac{\Delta_{eff}}{kT} \right) \]

The characteristic length \( L \) increases on approaching to the superconductor-dielectric transition \( (R \sim 5 \times 10^{7} \) \( \Omega \)) near the transition, a fast growth of \( L \) is observed when the temperature is lowered. \(^{10}\) The value of \( R_{1}(E, T \rightarrow \infty) \) does not depend on the thickness and is equal to \( R_{1}(T \sim 10^{7} \) \( \Omega \)), in contrast, to the analogous parameter \( R_{2} \) for nonsuperconducting films.

**CONCLUSION**

The question of the interaction between localization and superconductivity was the subject of a number of experimental\(^{11-14}\) and theoretical\(^{15,16,17}\) studies. Experimental investigations of island films\(^{11,12}\) revealed a decrease of the critical temperature with increasing film resistance, this was theoretically explained\(^{18}\) as suppression of the superconductivity by localization effects. A similar conclusion that the critical temperature is lowered when the metallic-conduction threshold is approached was obtained in Ref. 12, where the suppression of Josephson tunneling between the islands was considered.

In quench-condensed films of pure metals, except mercury,\(^{1}\) a threshold resistance \( R \sim 3 \times 10^{7} \) \( \Omega \) was reached at thicknesses \( \sim a \), when the superconductivity is already suppressed to a considerable degree by the size effect. A transition from activation to metallic conduction was observed...
in Ref. 13 for quench-condensed lead films at a thickness $d \approx 0.15 \mu m$. For high-resistance films with $R = 10^{5}-10^{9} \Omega$, the $R(T)$ curves had near $T_c$, a break with an increase of the resistance at $T > T_c$, the critical temperature remaining constant in the entire range of thicknesses. Similar phenomena were observed on island films with large island dimensions. These results, just as the data for mercury films, show that exponential localization does not influence the value of $T_c$, although it does change the behavior of the conductivity as $T \to 0$. The large thickness given in Ref. 13 for the start of the conduction of lead films and differing greatly from the data by others was interpreted in Ref. 13 as an indication that an island film was involved. The experimental procedure of Ref. 13, however, is more readily evidence that the localization and superconductivity can apparently be observed in high-resistance films of metal deposited together with a dielectric, e.g., with an inert gas. An investigation of such "dirty" systems, however, indicates that the object of the investigation is not the substance but samples whose properties are poorly reproducible and depend on random factors. From among the pure metals, according to present-day experimental data, only mercury becomes superconducting at a sufficiently large initial thickness.

In conclusion, we list the features that distinguish cold-deposited mercury films from films of other pure metals: 1. Large thickness at which superconductivity sets in, $d \approx 60 \mu m$. 2. Low value of $\alpha V^2 / s$ in thick layers. 3. Abrupt decrease of resistance with change of thickness at $d \approx 0.70 \mu m$. 4. Existence of superconductivity with constant $T_c$ and $H_c^*$. 5. Nonconcurrence of the metal dielectric transition ($R \approx 10^{5}-10^{7} \Omega$) and the superconductor-dielectric transition ($R \approx 5 \times 10^{5} \Omega$). 6. Large characteristic lengths $L \sim 10^{-10} \mu m$, which determine the deviations from Ohm's law.

These phenomena can be qualitatively explained by assuming a two-dimensional character of the localization in mercury films.

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