Giant negative magnetoresistance of semi-insulating amorphous indium oxide films in strong magnetic fields

V. F. Gantmakher and M. V. Golubkov

Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow Region, Russia

J. G. S. Lok and A. K. Geim

High Field Magnet Laboratory, Katholieke Universiteit Nijmegen, 6500 GL Nijmegen, The Netherlands
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We have studied magnetoresistance in thin films of amorphous indium oxide with various degrees of oxidation, i.e., with various densities of states in the velocity of the Fermi level. A large negative magnetoresistance is observed when the films are in the insulating state. The negative field derivative $\partial R/\partial H$ of the resistance persists up to 20 T, the highest magnetic field in our experiment. The magnetoresistance is described in terms of a field-dependent gap at the Fermi level. Such a gap can arise due either to tunneling in a system of fluctuation-induced superconducting clusters or to the Cooper interaction between electrons localized in shallow minima of the random potential. In the latter case, if the depth of the minima is smaller than the Debye energy, the electrons are virtually delocalized by short-wavelength phonons.
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1. INTRODUCTION

There are several mechanisms for magnetoresistance which can take place under conditions of hopping conductivity. Negative magnetoresistance (NMr—we use the lower case "r" for the abbreviation to avoid confusion with nuclear magnetic resonance) often occurs in weak magnetic fields. In the variable-range hopping regime, the initial and final sites of a hopping electron are far apart, so that the virtual scattering from sites situated in between gives rise to an interference contribution to the probability of a hopping event.[1,2] The nature of this contribution is similar to that of weak localization in metals. Magnetic fields of the order of

$$H_{\text{int}}=\frac{\Phi_0}{r^{3/2}\xi^{1/2}} \tag{1}$$

(Φ_0 is the flux quantum, r is the hopping distance, and ξ is the localization length) destroy this contribution, leading to NMr. H_{int} is usually of the order of 1 T (Refs. 3 and 4).

Another type of NMr is discussed in Ref. 5 for 2D-materials which are either granular or near the metal–insulator transition, for which the resistance is determined by tunneling between metallic clusters. A typical value of such a field H_{tun} is also of the order of 1 T.

Negative magnetoresistance also follows from the general scaling approach and the renormalization group analysis,[6] which show that the localization length ξ increases in the presence of a magnetic field and that the field can cause delocalization.

These NMr effects should be superseded by strong positive magnetoresistance (PMr) if the magnetic field is high enough. The high field shrinks the wave functions of hopping electrons and reduces their overlap. The magnetoresistance of weakly doped GaAs is a typical example (see for instance, Ref. 7).

If there are both singly and doubly occupied sites contributing to the hopping transport and if two electrons occupying the same site can form a singlet state, Zeeman splitting may prove to be the main factor in magnetoresistance. A magnetic field suppresses hops between sites with different occupation numbers and induces strong PMr (Refs. 8 and 9).

Furthermore, experimental evidence suggests that in the hopping regime a large NMr occurs in high magnetic fields as well. Such NMr has been observed in amorphous films of Ge:Cu (Ref. 10) and Ge:Cr (Refs. 11 and 12), and in granular aluminum.[13] Recently, we published preliminary results[14] on high field NMr in amorphous indium oxide (a-In_2O_x). All these observations[10–14] have many features in common from the experimental point of view, viz.: a) NMr increases rapidly with decreasing temperature in the range $T<1$ K; b) there is no indication of saturation in high fields; and c) NMr depends strongly on the distance from the metal-insulator transition. The variety of materials where such NMr exists[10–14] motivates one to look for a common origin.

To this end, a possible field dependence of the density of states at the Fermi level was discussed in Refs. 12 and 14 following the idea of Efros and Shklovskiĭ about the Coulomb gap.[15] The Coulomb interaction changes the distribution of carriers over a set of possible sites and, due to the electric field created by the randomly distributed carriers, affects the energies of these sites. This leads to a minimum in the density of states at the Fermi level.

The Coulomb interaction and, accordingly, the Coulomb gap do not change with magnetic field. However, apart from electrostatic repulsion, there are other mechanisms of long-range interaction between electrons, which may well be field-dependent. Generally, the field can turn one of them off and thereby induce a redistribution of electrons over hopping centers. Then the density of states $g(\epsilon)$ in the vicinity of the

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Fermi energy ϵ_F may depend on H. A positive derivative $(\partial g/\partial H)_{\epsilon_F}>0$ would mean $\partial R/\partial H<0$, i.e., NMR.

The authors of Ref. 12, when discussing Ge:Cr, suggested that spin-spin interaction might be the reason for a field-dependent part of the total interaction between electrons. In respect to a-In_2O_x films, we have proposed[14] for such a role the Cooper interaction, which leads to superconductivity in our films in a metallic state. As this paper contains a detailed report on the experiments[14] with a-In_2O_x, we continue the discussion about the possible role of the Cooper interaction.

This paper is organized as follows.

In the next section we describe techniques employed to change the oxygen content in the a-In_2O_x films and summarize our understanding of the processes involved. The temperature dependence of the resistance of the as-grown film and its evolution with decreasing oxygen content give a classic example of the insulator–superconductor transition.

Section 3 contains our major new results. In particular, we have found that NMr of an a-In_2O_x film in the insulating state does not vanish even at 20 T. The observed temperature dependence of its resistance in high fields confirms unambiguously that the state is indeed insulating.

The first of four subsections of Sec. 4 contains analysis of our experimental data. We intend to prove that the Copper interaction plays the key role in the observed behavior. We have two reasons for this, both based on experimental observations.

First, while films gradually transform from metals to insulators, two processes go in parallel and seem to be interconnected. The PMr in low and moderate fields, which certainly originates from the superconducting response of the metallic state, decays, and the NMr in high fields arises and increases. Secondly, we appeal to the analogy between the transport properties of our films and those of macroscopically inhomogeneous materials. When grains in a granular material become superconducting, a gap Δ arises in their energy spectrum. Then the decrease of the one-particle tunnel current gives rise to an exponential factor in the resistance of such "superconducting insulators" (Refs. 16–19):

$$R(T)\propto\exp(\Delta/T). \tag{2}$$

The destruction of the superconductivity by the magnetic field eliminates the exponential factor (2) in the resistance and thus leads to NMr.

In the three remaining parts of Sec. 4, we consider different possibilities of how the superconductivity can affect the transport properties of our material. In subsection 4.2, referring to the similar behavior of granular materials and to theoretical results by Spivak and Zhou,[20] we examine the feasibility of superconducting clusters being formed by fluctuations against a random background. As an alternative to this, a homogeneous model is proposed next. It is formulated first in terms of the effect of the $e-e$ interaction on the density of states at the Fermi level (subsection 4.3). The same model is reformulated in subsection 4.4 in terms of pairing correlations and localized bosons. This last subsection also contains a comparison of our interpretation with one by Paalanen, Hebard, and Ruel[21] who have performed similar experiments on the same material.

In the concluding Sec. 5, the main results and hypotheses of this paper are summarized.

2. AMORPHOUS In_2O_x AS A VERSATILE AND CHANGEABLE MATERIAL

Indium oxide has proved to be a very useful material for investigation of the transport properties near the metal–insulator transition. Depending on the preparation method, films of indium oxide can be obtained in microcrytstalline, granular, and amorphous forms, all with different oxygen content. The oxygen deficiency compared to the fully stoichiometric insulating compound In_2O_3 gives rise to electrical conductivity in the films. Bellinghaus *et al.*[22] estimated the doping efficiency of oxygen vacancies to be 0.1. By changing the oxygen content, one can cover the range from a metallic, superconducting material to an insulating material which exhibits activational conductivity. Alternatively, transport measurements of the carrier density allow one to estimate the oxygen content.

Due to this variety of forms of indium oxide, many different physical phenomena have been studied in this material. Among those which should be mentioned first are metal–insulator transitions,[23,24] the Bose-insulator phase and superconductor–insulator transitions[21,25] observed in amorphous films, and the "superconducting insulator"[26] in granular indium oxide.

In the present work, we have employed a method due to Ovadyahu[23,24,27] to lower the resistance of an a-In_2O_x film by thermal annealing, and studied the magnetoresistance in various phases of a-In_2O_x by varying the property of a sample in a set of successive heat treatments. Our films were made at the Racah Institute of Physics in Jerusalem by electron-beam evaporation of high-purity In_2O_3 onto a glass substrate in an oxygen atmosphere. This method[23] enables one to prepare films with different oxygen deficiency by changing the oxygen pressure during evaporation. Our films have a value of x close to 3 and a thickness of 200–250 Å. The samples were in the form of a strip 0.5 mm wide with a distance of $\approx$1 mm between adjacent potential probes. We used pure indium for electrical contacts to the films.

Low-temperature data for an as-grown a-In_2O_x film are plotted in Fig. 1. The film exhibits the Arrhenius behavior in the temperature dependence of the resistance (2) with $\Delta\approx15$ K (see the inset). The minimum temperature in this figure was limited by the steep increase in the resistance with decreasing temperature. Similar limitations exist also for magnetoresistance measurements.

In a separate experiment we measured the anisotropy of the NMr. We compared the resistivity at the same fixed T and H but for three different directions of the magnetic field: *(1)*–perpendicular to the film, *(2)*–along the film perpendicular to the current, and *(3)*–along the current. The anisotropy was found to be rather small e.g., for $T=2.1$ K and $H=7.6$ T

$$\frac{\Delta R}{R(0)}\equiv\frac{R(0)-R(H)}{R(0)}\approx24\%,$$

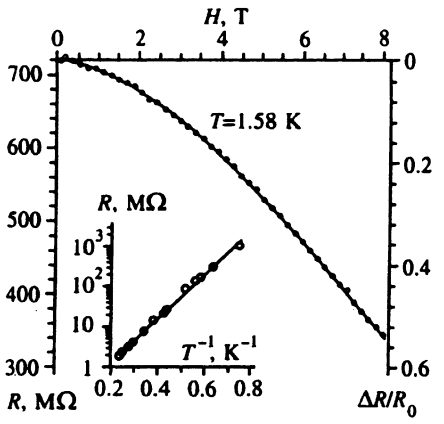

FIG. 1. Magnetoresistance of an as-grown a-In_2O_x film with dimensions 1 mm×0.5 mm×200 Å at a temperature of 1.58 K. Inset: resistance versus temperature for this film at zero magnetic field.

$$\frac{\delta R}{R(0)} \equiv \frac{R_{(2,3)}(H) - R_{(1)}(H)}{R(0)} \approx 0.5\%. \tag{3}$$

Therefore, we conclude that the main part of the NMr is isotropic and does not depend on the relative orientation of the film, field, and current. All of the data presented below correspond to the case of the magnetic field perpendicular to the film.

The amorphous rather than crystalline structure of the a-In_2O_x films makes them unstable, and the sample resistance changes in time. We observed three types of such changes.

A. After half a year in air at room temperature, the resistance had decreased by a factor of two even though the films remained amorphous. This corresponded to resistance changes by almost two orders of magnitude at 4.2 K. This slow decrease in oxygen content was irreversible. At least, it was impossible to resaturate the films with oxygen without causing recrystallization.

Two other types of observed changes can be described as reversible or quasireversible and are characteristic of films which have undergone the aging process. Their initial state has a considerably lower resistance than that shown in Fig. 1.

B. The exposure of the films to a temperature t in the interval

$$20\,^\circ\text{C} \leqslant t \leqslant 110\,^\circ\text{C} \tag{4}$$

in various gas atmospheres enables us to change the resistance R reversibly over some limited range. The lower limit on the temperature in (4) is imposed by the decrease in the relaxation rate, while the upper limit is due to recrystallization.

Exposure at some temperature t_1 in the interval (4) to air or oxygen atmosphere (we did not notice any difference) leads to a quasi-equilibrium state with resistance R_1. The higher t_1 the lower R_1. If the film is cooled to a lower temperature $t_2 < t_1$ after exposure, then its resistance remains the same or even decreases slightly in an atmosphere without oxygen or in vacuum, but it can start rising in an oxygen-bearing atmosphere. This is also the case for room-temperature storage: the reduced value R_1 can be maintained constant by placing the film in vacuum or in helium. However, if exposed to air, the sample relaxes to the initial value R with a time constant of several hundreds of hours.

Using the above procedure, we could reversibly transform the sample from the insulating state with NMr to the metallic state with superconductivity.[14] X-ray analysis showed that our films remained amorphous during all these transformations. Thus, the superconductivity in our films was not conditioned by the appearance of indium metal clusters.[24]

C. According to Ref. 28, a-In_2O_x films can be oxidized by ozone. We have tried two schemes for such oxidation. In the first scheme, the sample was placed inside a vacuum chamber with an oxygen pressure of 0.1 Torr and with an oxygen plasma produced by a gas discharge. The sample was shielded from direct exposure to the radiation produced by the discharge by means of a special shutter. In the second scheme, the sample was placed under normal pressure inside a tube through which air was pumped. The volume near the entrance of the tube was illuminated with ultraviolet light so that the air flowing along the sample contained an ionized component. In both schemes, the resistance of the sample at room temperature could rise as much as 50% after several hours of exposure. However, the resistance returned to the initial value a few seconds after the source of the ionization was turned off. Nevertheless, it was possible to preserve this high-resistance state: it could be frozen by cooling the sample before turning off the ionization source.

In order to concentrate below on the low-temperature transport properties of a-In_2O_x, we shall summarize here our understanding of the observed structure changes. Amorphous In_2O_x contains a framework formed by large indium atoms with mean distance r_{In} between them. In the aging process **A** the decrease of the oxygen content of the film is accompanied by "subsidence" of the indium framework, i.e., by decrease of r_{In}. The process **B** comprises oxygen exchange between the film and its surroundings and oxygen diffusion inside the film at a constant value of r_{In}. This assumes that there is a quasi-equilibrium value for x in a-In_2O_x films which depends on t and r_{In}. The thickness of the film is probably an important parameter also. The short relaxation time in process **C** indicates that the decrease in the resistance is not caused by diffusion of oxygen out of the bulk of the film. In this case we are probably dealing with oxygen adsorbed on the surface of the film which leaves the surface when the environment changes. We believe that the superconducting state results from changes in the oxygen shell of the indium atoms (e.g., a decrease in the number of indium valence electrons trapped by oxygen atoms in covalent bonds) and is not related to changes in the amorphous indium framework (i.e., the mean distance r_{In} remains constant).

A state with a given R can be reached through different sequences of heat treatment. On the other hand, as concerns the measured transport properties of such a state, it made no difference found whether this R value was set up after a lengthy diffusion process or after the adsorption of an oxygen layer. This universal behavior of the final state is illustrated in Fig. 2. States b–e were obtained from a film in state a by means of procedure **B**. We label this film No. 1. State

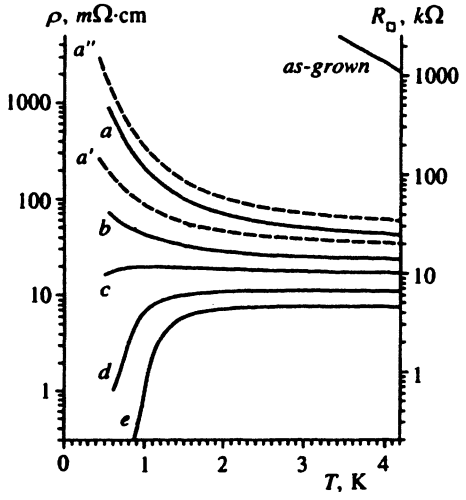

FIG. 2. Evolution of the temperature dependence of the resistance of a-In_2O_x films in zero magnetic field after different treatments. Film No. 1: states a (initial)–e, film No. 2: states a' (initial) and a''. A part of the dependence $R(T)$ for the as-grown film from the inset of Fig. 1 is also plotted.

a'' was obtained from state a' of another film (No. 2) by procedure C. Initial states a and a' of both films correspond to a half-year exposure of the as-grown film to air. This behavior suggests that it is not the disorder that is of prime importance for moving between the states in Fig. 2 but the density of unfixed electrons, which is controlled by broken chemical bonds or the electric field produced by an adsorbate at the surface of the films.

It is difficult to decide beforehand whether our films should be treated as 2- or 3-dimensional because we do not know what spatial scale is important in our case. For this reason, we give two scales in Fig. 2—resistivity ρ and sheet resistance $R_□$. Below we shall always use the resistance of the sample, which is approximately twice the sheet resistance and can be converted into resistivity by using a factor 10^{-6}:

$$R_□ \approx R/2, \quad \rho[\mathrm{m\Omega \cdot cm}] \approx R[\mathrm{k\Omega}].$$

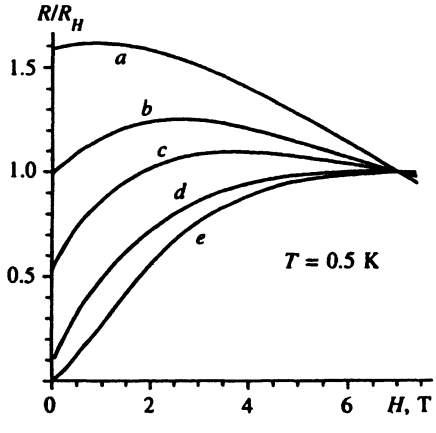

FIG. 3. Field dependence of $R(H)$ normalized to $R(7\ \mathrm{T})$ for various states of film No. 1.

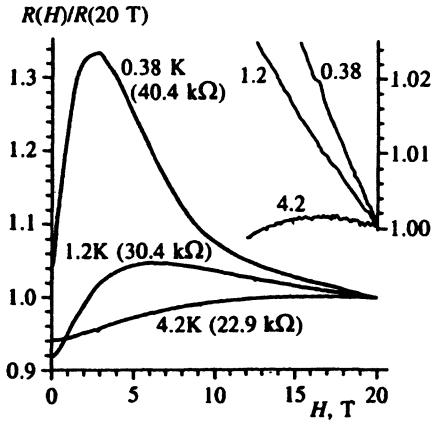

FIG. 4. Field dependence $R(H)$ normalized to $R(20\ \mathrm{T})$ at three different temperatures. Inset: high-field part of the curves at ten times the vertical scale. Film No. 3, state b.

3. EFFECT OF THE MAGNETIC FIELD

Figure 3 gives a general survey of the influence of the magnetic field on the resistance of the a-In_2O_x films. For the superconducting states e and d this influence is quite natural: magnetic fields up to 5–6 T gradually suppress superconductivity, and the magnetoresistance saturates in higher fields. However, this decay-of-superconductivity-like effect which leads to PMr in fields below 5–6 T can be also seen for states c–a, which do not exhibit any sign of superconductivity in the temperature dependence of the resistance (Fig. 2). As a measure of this superconducting response, it is convenient to use the reduced difference

$$D_{sc}=(R_{max}-R(0))/R_{max}, \qquad (5)$$

where R_{max} is the resistance at the maximum of the function $R(H)$. This difference D_{sc} equals 1 when the superconducting transition is complete and the resistance falls to zero.

A very intriguing feature in Fig. 3 is the NMr which appears at high magnetic fields. The farther the film moves into the insulating regime, the larger the derivative $\delta = -\partial(\ln R)/\partial H$. This tendency persists up to the most insulating of states. The δ values for the a'' state at 0.4 K and for the as-grown state at 1.6 K were approximately the same, but we note that δ for the as-grown film doubles when the temperature is lowered from 2.1 K to 1.6 K [cf. Fig. 1 and the values in Eq. (3)].

The qualitative conclusion about the correlation between δ and D_{sc} was reached after examining nine different films: as the states span the interval between the insulator and the superconductor, the difference D_{sc} from (5) changes from zero to unity, while the derivative δ declines to zero. However, any quantitative analysis can only be made by comparing different states of the same film.

One of the films (No. 3) was studied in magnetic fields up to 20 T in the b-like state. (We continue using the notation a–e for film No. 1 from Figs. 2 and 3 to give a general idea of what type of behavior we are dealing with: a- and b-like states are insulating, c-like is the one near the transition, and d- and e-like states are superconducting.) Figure 4 shows the magnetoresistance of this state at three different

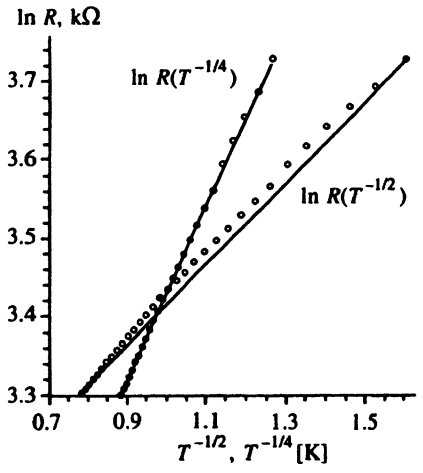

FIG. 5. Temperature dependence of $R(T)$ in a fixed magnetic field of $H=15$ T. While $\ln R$ is a straight line when plotted versus $T^{-1/4}$ in the range $T=0.4$–1.2 K, it is certainly not when plotted versus $T^{-1/2}$—here the straight line is only meant as a guide. The film and its state are the same as in Fig. 4.

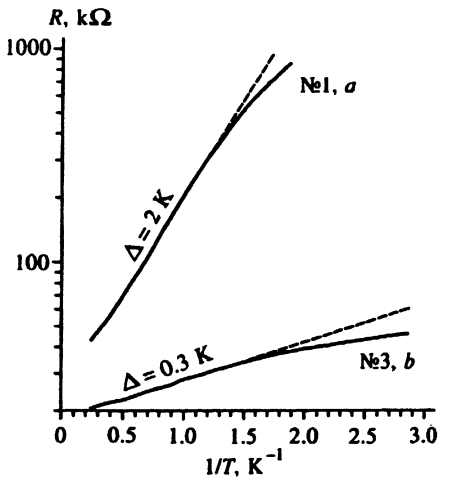

FIG. 6. Temperature dependences of the resistance of a-In_2O_x films in zero magnetic field, plotted as $\ln R$ versus $1/T$. The curves are labeled by the number of the film and by a letter representing the type of state (see the text).

temperatures. The data for different temperatures are normalized to the resistance at $H=20$ T. These values are shown in parentheses in Fig. 4. The high-field part of the curves is shown in the inset at ten times the vertical scale.

Two important facts can be deduced from Fig. 4. First, NMr extends to the highest field in the experiment. Secondly, the field at which $R(H)$ reaches its maximum, i.e., the field at which the NMr exceeds the low-field PMr, increases with increasing temperature. Even at 4.2 K, NMr still exists in fields above 17 T.

To determine what kind of material we get in strong magnetic fields, we have measured the temperature dependence $R(T)$ at a fixed field of 15 T. The result is shown in Fig. 5, where $\ln R$ is plotted against $T^{-1/2}$ and $T^{-1/4}$. The fact that the latter plot is a straight line indicates that after applying the magnetic field we are dealing with an insulator which obeys Mott's law

$$R=R_0\exp(T_0/T)^{1/4}, \tag{6}$$

with a characteristic Mott temperature $T_0=1.6$ K. At a temperature of $T=0.38$ K, we estimate the hopping distance to be $r \approx \xi(T_0/T)^{1/4} \approx 1.5\xi$, and the hopping energy $\epsilon_h \approx T_0^{1/4}T^{3/4} \approx 0.54$ K.

4. DISCUSSION

4.1. Analysis of the experimental data

The activational dependence (2) of the resistance indicates either that there is a gap Δ in the density of states at the Fermi level, or, that the states at the Fermi level are localized and the distance to the mobility edge is Δ. It is known from the paper of Shahar and Ovadyahu[24] that the resistance of a-In_2O_x films follows the dependence (2). It was found in Ref. 24 that the value of Δ decreased from 7 to 2 K when the state of the films approached the metal–insulator transition from the insulating side. Our as-grown state is far deeper in the insulating regime and, accordingly, we obtain $\Delta=15$ K (see inset in Fig. 1). Thus our data are in agreement with the data in Ref. 24.

However, both the measurements in Ref. 24 and our measurements with the as-grown film were performed above 1 K. When we extended the measurements to lower temperatures we found deviations from (2)—see Fig. 6. To explain these deviations, we recall the PMr in the left-hand parts of the magnetoresistance curves in Fig. 3. The evolution of the curves starting from the metallic states shows that PMr is due to destruction of the superconductivity (or its remnants), and the effect survives even in the insulating a- and b-type states. This means that in zero field the hopping resistance at low temperatures is partly compensated by the superconductivity—for example, by the emergence of superconducting clusters.

This interpretation makes the temperature dependence at zero field not very informative because of the superposition of two different phenomena. The situation in high magnetic fields is much simpler, since the usual superconducting response is completely suppressed by the field. As a result, a solid confirmation is obtained from Fig. 5 that the high-resistance states of the a-In_2O_x films represent an insulating material with variable-range hopping.

Let us start in our further reasoning from the high field limit. An increase in the resistance with decreasing field may be due either to a decrease in the transfer integral (probability of an elementary hopping event) or the density of states at the Fermi level. The first suggestion is unlikely, since the effect of a strong magnetic field consists mainly in shrinking the wave functions and thus reducing the transfer integral. This leaves us with changes in the density of states. The simplest way to model such changes is to introduce a field-dependent gap $\Delta(T,H)$ at the Fermi level[11,12,14]

$$R(T,H)=\exp[\Delta(T,H)/T]R_\infty$$
$$=R_0\exp[\Delta(T,H)/T]\exp(T_0/T)^{1/4}, \tag{7}$$

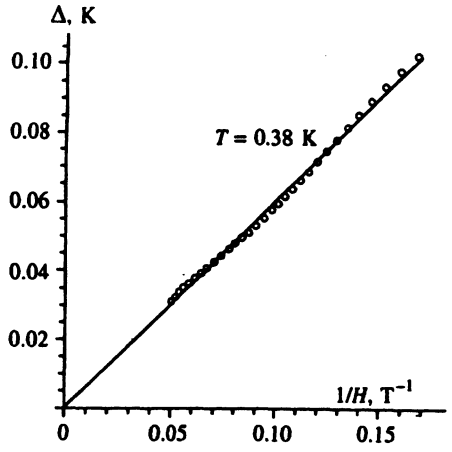

FIG. 7. Representation of the function $R(H)$ measured at $T=0.38$ K in the range 6 T$<H<$20 T in the form ln R versus $1/H$. The film and its state are the same as in Fig. 4.

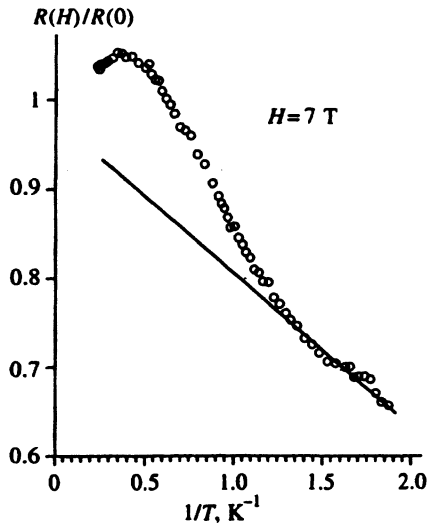

FIG. 8. Temperature dependence of the ratio $R(0)/R(7\text{ T})$ of the resistance of film No. 1 in state a.

where the resistance of the sample in the infinite field R_∞ is expressed using formula (6). We applied this expression to the data of Fig. 4 at $T=0.38$ K in the field interval 6–20 T, i.e., to the right of the maximum of the function $R(H)$. The inferred dependence $\Delta(H)$ is approximated in Fig. 7 by the function

$$\Delta(H)|_{T=0.38\text{ K}}=0.6/H, \tag{8}$$

where Δ and T are in K and H is in T. Our model expression (7) and the extrapolation of formula (8) to the high-field limit imply that a further increase in the field above 20 T can result only in an 8% decrease in the resistance at 0.38 K. Supposing that Δ vanishes in the high-field limit, we obtain Δ
(7 T)$\approx$0.1 K.

To find the temperature dependence of the first exponential factor in equation (7), we use data for the state a of film No. 1 from Figs. 2 and 3 and plot $(R(7\text{T})/R(0)$ versus $(1/T)$ in Fig. 8. It follows from the figure that below 0.8 K the gap Δ does not depend on temperature and the difference $\Delta(0)-\Delta(7\text{ T})$ is of the order of 0.25 K. Together with the estimate for $\Delta(7\text{T})$ obtained from the date in Fig. 7, this leads to the following value of Δ in zero field below 0.8 K:

$$\Delta(0,0)\approx 0.35\text{ K}. \tag{9}$$

The best way to check the extent to which the analytical form $\Delta(H)\propto(1/H)$ describes the whole set of data is to repeat the same procedure at another temperature. However, one can see from Fig. 4 that at 1.2 K the maximum of $R(H)$ is shifted above 6 T and becomes very broad. This makes the high-field interval of "pure" NMr too short for further analysis of the validity of the expression $R(T,H)$. The required verification can be obtained only at temperatures sufficiently lower than 0.4 K or, perhaps with samples further inside the insulating region.

4.2. Superconducting clusters created by fluctuations

The chosen probe function (7) reflects the idea that the density of states at the Fermi level is controlled by a field-independent $e-e$ interaction.[12,14] We can only guess the nature of such an interaction. Two indications have already been mentioned in the Introduction—a gradual transformation from the usual suppression of superconductivity to NMr and the analogy with the behavior of granular superconductors.

The transformation can be seen in Fig. 3. As one moves further into the insulating region, the superconducting response expressed by D_{sc} from Eq. (5) decreases from unity to zero, and there is an increase in the derivative δ which is a quantitative measures of the NMr.

Kowal and Ovadyahu[27] have previously noticed that a-In_2O_x films behave very similarly to granular insulators containing superconducting grains. The "superconducting insulators"[16–19] which we mentioned in the Introduction also have an exponential rise in resistance at low temperature and NMr that increases with decreasing temperature.[18] Sometimes the experimental plots even look very similar.[19]

If we agree that these indications imply that superconductivity might be a basis for the behavior of the a-In_2O_x films, we are left with two possibilities. We discuss the first of them in this subsection.

Despite an essentially amorphous underlying structure with a very short correlation length, the films can behave as if they were granular, with comparatively large superconducting clusters due to fluctuations.[27] This model finds support in the recent paper by Spivak and Zhou.[20] They demonstrated that in a disordered metal, fluctuations can lead to superconducting droplets embedded in a normal metal, and connected by Josephson links even in magnetic fields $H>H_{c2}$. The superconducting droplets in a-In_2O_x films, if they exist, are embedded in an insulator. Then the initial PMr at low fields can be attributed to breaking of the Josephson coupling between droplets and division of the superconducting clusters into smaller parts. In a network of superconducting clusters, some fraction β of the contacts between them are in a Josephson state when the magnetic field is absent,

but they all become normal in a moderate field. A similar process has been observed in inhomogeneous materials,[19] and a similar description has been used.[29] In the fully insulating state there is no Josephson coupling at all, $\beta=0$ even in zero field, and PMr is absent (Fig. 1).

In such a scheme, the exponential factor (2) appears in the resistance of the contact between neighboring superconducting clusters. When the field destroys the superconductivity in the clusters, this factor disappears. The critical field H_{c2} which destroys the superconductivity depends on the size of a grain a. Hence NMr arises over some extended range of the magnetic fields, which depends on the distribution of cluster sizes.

However, this approach has some difficulties in our case. If a grain is coupled to the environment only by single-particle tunneling, it must be treated as isolated. In this case, we can estimate the size of the grains:

$$H_{c2}/H_{cm}\approx\lambda/a.$$

For typical values of the thermodynamic critical field H_{cm} and penetration depth λ in metals, we find that 20 T destroys superconductivty in grains of size $a\approx\lambda/100\approx 10$ Å. On the other hand, a grain can remain superconducting only if the size quantatization of the grain spectrum $\delta\epsilon \approx (g_F a^3)^{-1}$ is smaller than Δ, i.e., until

$$a>a_0=(g_F\Delta)^{-1/3}, \qquad g_F\equiv g(\epsilon_F). \tag{10}$$

The lower limit a_0 imposed on a by (10) is greater than 10 Å: $a_0\approx 30$–40 Å. For "superconducting insulators" with the opposite relation between a and a_0, we need to find another model. For such a model, described below, we borrow ideas not only from superconductivity but mainly from variable-range hopping and Coulomb gap phenomena.

4.3. Homogeneous insulator: Cooper contribution to the Coulomb gap

Let us imagine a semiconductor with a density of donors N_d and a density of acceptors $N_{ac}<N_d$. At low temperatures all the acceptors are charged, and the remaining N_d-N_{ac} electrons are distributed over the N_d donors. For a moderate compensation

$$0\ll N_{ac}/N_d\ll 1, \tag{11}$$

there are many possibilities for such a distribution. Each one corresponds to a different configuration of the random electric field and hence to different energies of each particular donor site. According to Efros and Shklovskiĭ,[15] due to the Coulomb interaction the energetically favorable distribution of electrons over donors results in a density of states described by

$$g(\epsilon)\propto|\epsilon-\epsilon_F|^{d-1} \tag{12}$$

(d is the dimensionality), which is equal to zero at the Fermi level, i.e., $g_F=0$. This is the so-called Coulomb gap. It reduces significantly the variable range-hopping conductivity when the temperature is of the order of or below the width of the gap.

Consider now the indirect $e-e$ interaction via short-wavelength virtual phonons, which is responsible for superconductivity in metals. It is important in a Fermi gas when the electrons are delocalized. In our material the situation seems to be not far from this. Indeed, the change in conductivity of a-In_2O_x films from room temperature to liquid nitrogen temperature is rather small. Hence, the random sites where the carriers are localized at low temperature are expected to be rather shallow, with binding energy below the Debye energy typical of short-wavelength phonons. This means that such localized carriers interact via virtual phonons in delocalized fashion.

The whole scheme is rather simple. The Cooper interaction redistributes electrons over accessible potential minima just as the Coulomb interaction does. The nearly delocalized status of electrons makes this interaction sensitive to the field. Hence the incorporation of the Cooper interaction into the model can qualitatively explain the influence of the magnetic field—the key feature in our experiment.

4.4. Homogeneous insulator: localized bosons

In the preceding subsection, we proposed a picture of localized carriers redistributed over minima of the random potential through a virtual delocalization. This should lead to pair correlations in the carrier distribution and a gap in the density of states close to the Fermi level. It may also be helpful to discuss the same physics in other terms, using a model of localized bosons.[30]

Pair correlations in insulators have recently generated much theoretical and experimental interest. We mention the model of the superconductor–insulator transition in a system of dirty bosons,[30] the coherent propagation of interacting particles in a random potential,[31] and the observation of Coulomb correlations in resonant tunneling and in hopping.[32] Unfortunately, there are no quantitative predictions that would allow us to compare our results. Hence, we restrict our discussion to the simplest analogy with "superconducting insulators."

In regions with a high local density of pairs, their coherent propagation in a Josephson-like process is probable. This decreases the total resistivity of the material. In regions with a low local density of pairs, correlated tunneling is impossible, hopping is incoherent, and the gap at the Fermi level increases the resistivity. As usual, a magnetic field destroys the Josephson coupling first. At higher fields, Cooper pairs are destroyed due to Zeeman splitting. Thus we conclude that PMr in low fields and NMr in high fields can originate from the two components of the response to the destruction of pair correlations. As in inhomogeneous superconductors,[19] PMr results from the decay of the Josephson component, and NMr results from the increased probability of the incoherent one-particle hopping (i.e., from the elimination of the factor (2)).

We conclude this section by comparing our interpretation with those of Ref. 21. Paalanen, Hebard and Ruel[21] have measured field dependences of the resistance R and Hall resistance R_{xy} of an a-In_2O_x film in a d-like state at different temperatures. They have determined two fields H_{xx}^c and H_{xy}^c which separated three phases: vortex-glass (supercon-

ductor), Bose-insulator, and Fermi-insulator. The highest field, H_{xy}^c, corresponded to the vicinity of the maximum of $R(H)$ (see the curve at 0.38 K in Fig. 4 as an example). So, according to Ref. 21 NMr belongs completely to the Fermi-insulator state. The above description implies that pair correlations (dirty bosons[30]) are not destroyed by the field H_{xy}^c, but survive up to our highest fields, and that the Fermi-insulator phase is still not reached in a field of 20 T, in contrast to the arguments of Ref. 21. However, the difference is one of interpretation only, not of the experimental data.

5. CONCLUSION

We conclude by summarizing the main results and assumptions of this paper.

1. In accordance with many other experimental observations,[23–25] we assume that a-In_2O_x films can be maintained in the condition of homogeneous disorder over the whole range of states from fully insulating to superconducting. The main parameter altered by the reversible heat treatment of the a-In_2O_x films is carrier density, rather than disorder.

2. Fully insulating states exhibit only NMr in high fields, while fully superconducting states exhibit only PMr in low fields (suppression of superconductivity). Intermediate states have both features, and the smaller the positive $\partial R/\partial H$ in low fields, the larger the negative derivative in high fields. In the current experiment, we have dealt mainly with these intermediate states.

3. Films with a negative low-temperature derivative $\partial R/\partial T$ (i.e., films in the insulating state) exhibit temperature-activated conductivity, which can be described as $R_0\exp(T_0/T)^n$. At zero field, Arrhenius activation with $n=1$ is observed above 1 K, while below 1 K it is distorted by the superconducting response. In high fields, we observed Mott activation with $n=1/4$.

4. NMr, i.e., the negative derivative $\partial R/\partial H<0$ of a-In_2O_x films in high magnetic fields, can be described by a field-dependent gap in the density of localized states at the Fermi level. This gap is found to be of the order of 0.3–0.4 K in zero field. In high fields it decays as $1/H$.

5. The assumption that the gap results from Cooper interactions is based on two experimental observations: a) the correlation between NMr (in high fields) and PMr (in low fields)—the latter transforms into a superconducting response in metallic films, and b) the similarity between the behavior of a-In_2O_x films and inhomogeneous high-resistance superconductors.

6. Superconducting interactions can be invoked to explain NMr in our homogeneous insulator in two schemes: a) the films become effectively granular, with superconducting clusters due to fluctuations, and b) a gap in the density of states appears due to the Cooper attraction between electrons localized at shallow sites, with binding energies below the energy of virtual phonons. Both models require further theoretical and experimental analysis.

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Note added in proof (14 March 1996). While writing this article, the authors did not know that the relation (7) had already been derived theoretically by Ioselevich [A. S. Ioselevich, Phys. Rev. Lett. **71**, 1067 (1993)] for an insulator with a hard gap. This further justifies using this relation in our analysis.

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