The Coulomb gap in the phenomena of nonlinear screening and non-ohmic hopping conduction

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(Submitted 31 January 1992; revised 16 March 1993)
Zh. Eksp. Teor. Fiz. **104**, 2473–2482 (July 1993)

We study hopping conduction over Coulomb-gap states in strong electric fields for a-Si⟨Mn⟩ samples. All previously predicted nonlinear transport effects are discovered in experiments. We are able to observe the entire spectrum of field and temperature dependences of the conductivity σ, using a single sample. We observe a decrease in σ as the field gets stronger. In a narrow range of temperatures and field strengths the effect is so strong that negative resistance emerges.

1. INTRODUCTION

In disordered semiconductors the charge transfer at low temperatures takes place owing to electron hopping, and the hopping range grows as the temperature decreases (variable range hopping, or VRH). Study of the temperature dependence of the hopping conductivity σ in weak electric fields, in which Ohm's law is valid, makes it possible to gather information about the spectrum of localized states of a material.

In studying hopping conduction over the states of the impurity band of Mn and Fe created in amorphous silicon (a-Si) by ion implantation we discovered an anomalously wide and deep Coulomb gap[1] formed, according to the existing theoretical ideas,[2] in the electron spectrum because of the Coulomb interaction of electrons localized at different centers. Within such a gap the density of states $g(E)$ vanishes according to a power law when the energy tends to the Fermi energy:

$$g(E)=\alpha \frac{\varepsilon^3}{e^6}(E-E_f)^2, \tag{1}$$

where α is a numerical factor, e is the electron charge, and ε the dielectric constant of the medium. This paper is devoted to non-ohmic hopping conductivity in a-Si⟨Mn⟩ in intermediate and strong electric fields. The distinctive feature of the object, the presence of a large Coulomb gap, made it possible to observe in experiments practically all the predicted[3–8] nonlinear effects of hopping transport. Another remarkable feature of the system under investigation is that it was possible to observe the entire spectrum of field and temperature dependences of σ in studies of the current-voltage characteristics of one and the same sample. The main results of our work are the following:

1. The exponential increase in conductivity with field strength was observed only in fairly strong electric fields, when the local field was nonuniform due to the nonlinear screening by electrons localized within the Coulomb gap and followed the current conductor.

2. In the intermediate range of field strengths and temperatures (up to the temperature of thermal clogging of the Coulomb gap) and field varied little owing to the potential of the electrons redistributed on the critical subgrid and remained practically uniform, as in an insulator. In this case the experiment showed a drop in hopping conductivity as the field got stronger and the phenomenon of negative incremental resistance manifested itself.

3. Raising the temperature to the value at which the Coulomb gap clogged led the system to a state with superlinear current-voltage characteristics.

2. THE EXPERIMENT AND THE RESULTS

The layers of amorphous silicon were prepared by electron-beam evaporation of crystalline silicon in a vacuum onto quartz substrates with subsequent implantation of manganese ions. The leads to the samples were made by planar-geometry gold or aluminum sputtering. The electrode separation d amounted to 25–50 μm. To exclude the possibility of the samples getting too hot in strong electric fields, a pulsed voltage with a period $\tau=1$–$10\ \mu$s was applied to them. Measurements of the current in samples with different electrode separations showed, first, that the impedance grows with d and, second, that the non-ohmic threshold voltage U_c is proportional to d. These properties prove that the observed nonlinear effects are due to bulk processes in a-Si rather than to junction properties.

The preparation and properties of a-Si⟨Mn⟩ sample on the ohmic section of the current-voltage characteristics are described in greater detail in Refs. 1 and 10, where it is shown that at low temperatures the conductivity of this material follows the Shklovskiĭ–Éfros law

$$\sigma(T)\propto \exp\left\{-\left(-\frac{T_0}{T}\right)^{1/2}\right\}, \qquad T_0=\frac{\beta e^2}{k\varepsilon a}, \tag{2}$$

where k is the Boltzmann constant, a the range of localization of electrons at impurities, and β a numerical coefficient. Formula (2) corresponds to hopping charge transport over the Coulomb-gap states.[2]

To exclude the possibility of current oscillations at a constant voltage in conditions where the sample exhibits negative incremental resistance, we used a measurement setup in which the current was measured by the decrease of the voltage on the load resistance r_{load}. Here, if r_x is the absolute value of the sample resistance, the load resistance was chosen such that $r_{\text{load}}<r_x$. In this case the current-

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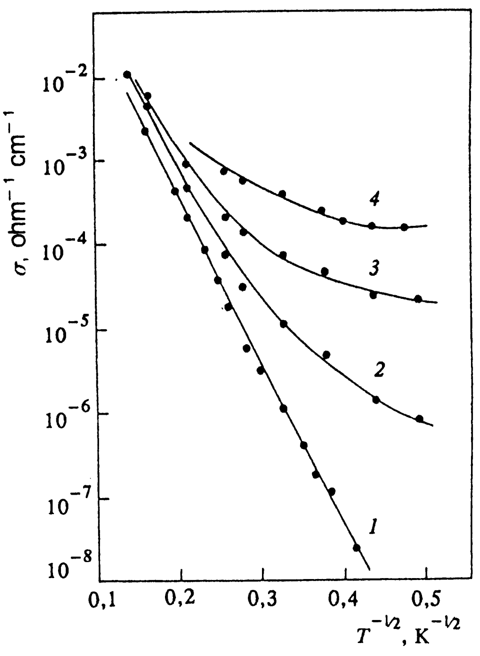

FIG. 1. Temperature curves for the conductivity of an a-Si⟨Mn⟩ sample with $N=7$ at. % at different electric field strengths F: curve *1* corresponds to 20 V cm^{-1}, curve *2* to 5×10^3 V cm^{-1}, curve *3* to 10^4 V cm^{-1}, and curve *4* to 1.5×10^4 V cm^{-1}.

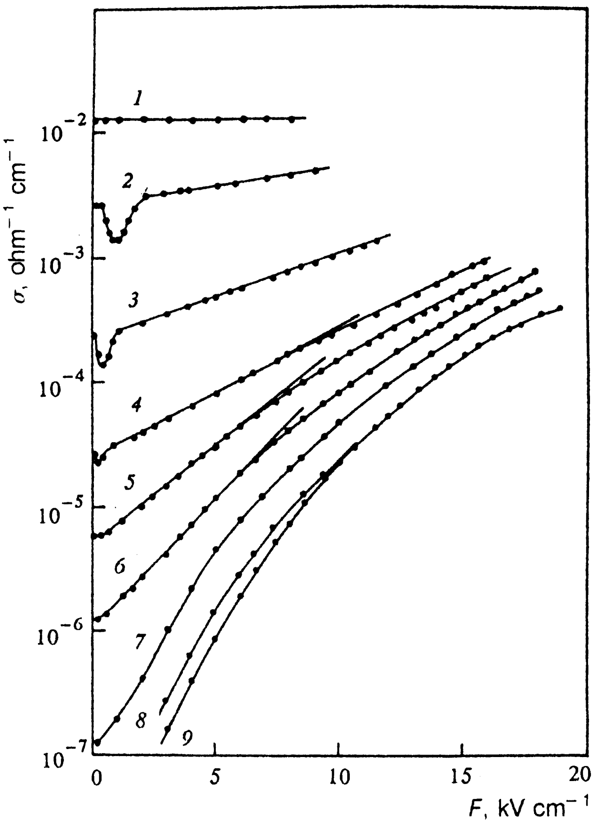

FIG. 2. The field dependence of the conductivity of an a-Si⟨Mn⟩ sample with $N=7$ at. % at different temperatures T: curve *1* corresponds to 50 K, curve *2* to 38 K, curve *3* to 22 K, curve *4* to 14.5 K, curve *5* to 12.5 K, curve *6* to 9.2 K, curve *7* to 6.9 K, curve *8* to 5.2 K, and curve *9* to 4.2 K.

voltage curves for the load and the sample, when considered as functions of the sample voltage, have only one point of intersection. If in conditions of negative incremental resistance no domain instability develops in the sample, such a circuit excludes current oscillations.

In the studied a-Si structures with Mn densities of $N=7$ and 8 at. % the temperature T_c of the onset of the Shklovskiĭ–Éfros law (2) is 47 and 41 K, respectively. Measurements of the temperature dependence of σ for $T<T_c$ have shown that as the electric field strength F increases, the temperature dependence of conductivity weakens considerably and departs from the law (2). The respective curves for a-Si⟨Mn⟩ with $N=7$ at. % are depicted in Fig. 1.

The measured σ as a function of F at different temperatures are illustrated by Fig. 2 ($N=7$ at. %). Ohm's law breaks down for $F>F_c=(1\text{–}4)\times10^2$ V cm^{-1}. The situation just above the non-ohmic threshold is remarkable: depending on the temperature, the conductivity in the field either exponentially grows ($T\leqslant12.5$ K; by Ref. 9 this is called "positive non-ohmic behavior") or decreases (14.5 K$\leqslant T\leqslant$50 K; "negative non-ohmic behavior"), which corresponds to superlinear current-voltage characteristics or to sublinear characteristics. The decrease in hopping conductivity accompanying an increase in field strength manifests itself only in a limited temperature range; this phenomenon disappears in fairly strong electric fields.

At $T\approx38$ K the drop of conductivity of the system in the field is so large that the current-voltage characteristic acquires a section with negative differential resistance (Fig. 3). As measurements in constant electric fields have shown, a characteristic feature of the observed negative

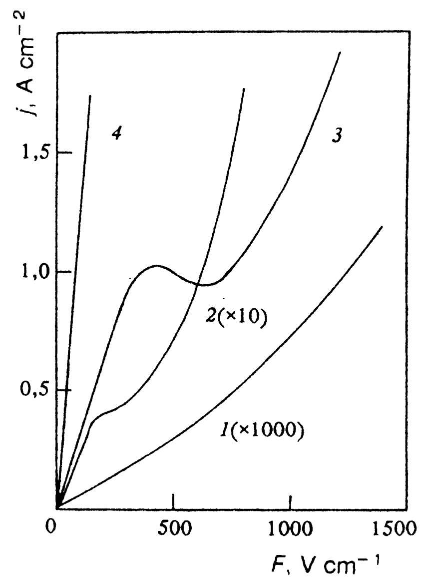

FIG. 3. Current-voltage characteristics of an a-Si⟨Mn⟩ sample with $N=7$ at. % at different temperatures T: curve *1* corresponds to 12.5 K, curve *2* to 22 K, curve *3* to 40 K, and curve *4* to 50 K.

differential resistance is the absence of any domain instability.

It was found that the high-temperature limit at which sublinear features of the current-voltage characteristics disappeared ($T_s \approx 50$ K at $N=7$ at. % Mn and $T_s \approx 40$ K at $N=8$ at. % Mn) coincides, with high accuracy, with the temperature of thermal clogging of the Coulomb gap determined in Ref. 1 ($T_b \approx 45$ K at $N=7$ at. % Mn and $T_b \approx 35$ K at $N=8$ at. % Mn). This is not accidental. The presence of a Coulomb gap in the spectrum of states explains the observed qualitatively different field dependences of hopping conductivity.

3. GENERAL INFORMATION ABOUT NON-OHMIC HOPPING CONDUCTION AND DISCUSSION OF THE EXPERIMENTAL RESULTS

The theory of non-ohmic hopping transport in the VRH region developed in Refs. 3–5 predicts markedly different temperature dependences for σ. In extremely high fields (Ref. 3), $eFa/kT \gg 1$, we have

$$\sigma(F) \propto \exp\left\{ -\left(\frac{F_0}{F} \right)^{1/2} \right\}. \tag{3}$$

In the theory of VRH conduction the average hopping radius R is equal to $\frac{1}{4}a\xi_c(T)$, where $\xi_c \gg 1$ is the percolation threshold. For the case of hopping conduction over Coulomb-gap states we have $\xi_c = \sqrt{T_0/T}$. In exceptionally high fields, the energy acquired in the hopping range, $W=eFR(T)$, becomes higher than the energy spread $\Delta E \sim kT\xi_c$ of the localized states over which hopping conduction takes place. Hence, the conduction is not of activation origin.

In weaker fields, $\Delta E > eFR(T) > kT$, we have

$$\sigma(F) \propto \exp\left\{ \frac{eFl(T)}{kT} \right\}. \tag{4}$$

Here $l(T)$ is a parameter with the dimension of length and is called the non-ohmic length, $l(T) = C_1 R(T)$, where $C_1 < 1$ (Ref. 4) or $C_1 = (14 \pm 2) \times 10^{-3} \xi_c$ (Ref. 6).

The main idea leading to this result is that the activation energy ΔE needed for hopping between localized states is decreased by the field by a quantity of the order of W.

Shklovskiĭ[5] shows that the non-ohmic behavior manifests itself even at lower fields. The explanation is as follows. In the ohmic region, hopping conduction over states localized at random points is equivalent to a grid of randomly connected resistors with resistance distributed according to an exponential law:

$$r_{ij} \propto \exp \xi_{ij}, \qquad \xi_{ij} \gg 1.$$

Hence, the conductivity of the system is determined by resistances with values ξ_{ij} close to the percolation threshold ξ_c (Ref. 2), which carry practically all the applied voltage. In the non-ohmic regime the role of $l(T)$ in (4) is then taken by the distance between the pivotal resistances,

$$L_c = R\xi_c^{\nu} \gg R(T), \tag{5}$$

where $\nu = 0.88$ is the critical exponent of percolation theory.[2] According to Ref. 5, if $eFL_c(T)/kT \gg 1$, we have

$$\sigma(F) \propto \exp\left(\frac{C_2 eFL_c}{kT} \right)^{1/(1+\nu)}. \tag{6}$$

Equations (4) and (6) contain constants C_1 and C_2 that are poorly defined in the theory.

The result (6) has been obtained in a concrete model of an infinite hopping-conduction cluster in the form of a twisting single-conductor grid with suspended "dead ends" (Ref. 11). Each conductor in the grid, which we call a macrolink, contains one pivotal resistance and ξ_c exponentially smaller resistances, so that the conductor is much longer than L_c. The geometric distance between the pivotal resistances, L_c, has the meaning of the characteristic cell size of such a grid, that is, the scale starting with which the system may be assumed homogeneous.

In the same model, an effect was predicted and experimentally observed[9] in which σ decreased as the field strength grew and even negative differential resistance manifested itself for the regime of hopping conduction over the nearest neighbors in the range of hopping-conductivity saturation. The sublinear nature of the current-voltage characteristics was linked to the capture of electrons by the dead ends and, as a result, to the decrease in their mobility caused by the exponentially long delay in the dead ends. Bottger and Bryskin[7] and Nguyen Van Lien and Shklovskiĭ[8] expressed the idea that the presence on the lines of current flow of sections directed against the force with which the field acts on an electron, or "bottlenecks," can also lead to sublinear current-voltage characteristics. Such sections of the critical subgrid (dead ends and bottlenecks) carry the generic name of traps induced by the electric field.

It is assumed that with VRH conductivity there is no negative differential resistance caused by capture at dead ends because in this case the number of electrons leaving a trap is exactly balanced by the number of electrons supplied by the Fermi level. However, bottlenecks must be here, and Aleshin and Shlimak[12] do mention observing sublinear current-voltage characteristics in the VRH regime.

Note that all the arguments concerning field-induced traps[7–9] implicitly assume that the field is uniform over the size of the trap. At the very least it is assumed that over such a distance there is no time for the field to change its direction. Yet the question of uniformity of the field is not discussed in the papers cited above.

It is clear, however, that the potential difference applied to the sample not only generates a hopping current but redistributes the electrons over the impurity states. The carriers will build up in the dead ends, and regions depleted of electrons will appear at entrances to bottlenecks. The emerging space-charge density $\rho(\mathbf{R})$ leads to potential fluctuations inside the sample. As a result the local fields $F_{\text{loc}}(\mathbf{R})$ differ from the average field F in both magnitude and direction. Thus, in the event of a current in the system there emerges another parameter of the dimension of length characterizing the size of regions in which the elec-

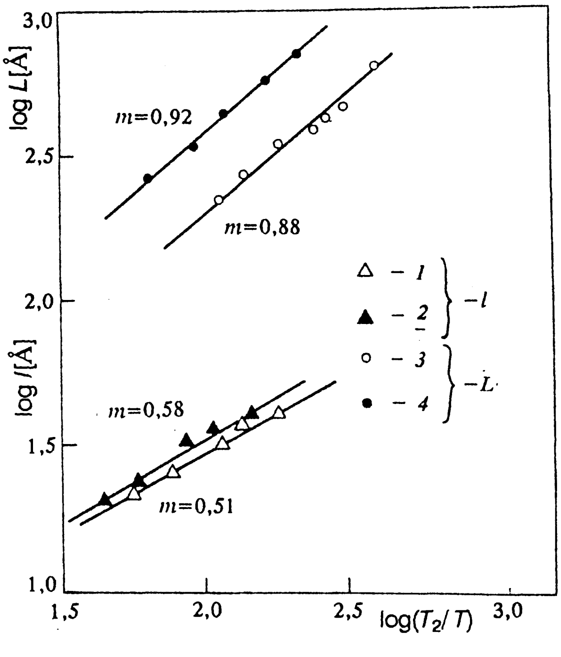

FIG. 4. Non-ohmic length l and $L \sim L_c$ as functions of the square of the percolation threshold, $\xi_c^2 = T_0/T$, in a-Si⟨Mn⟩ samples with the following impurity concentrations: Δ and ○, $N=7$ at. %; and ▲ and ●, $N=8$ at. %.

TABLE I. Numerical parameters of non-ohmic hopping conduction in a-Si⟨Mn⟩.

Impurity concentration, at. %	T, K	$\xi_c = (T_2/T)^{1/2}$	C_1	C_2	C_3
7	4.2	20.0		2.2	
	5.2	17.9		2.0	
	6.1	16.5		2.1	
	6.9	15.6		2.1	
	9.2	13.5	2.2	2.3	
	12.5	11.6	2.3	2.4	
	14.5	10.6	2.2	2.2	5.7
	22.0	8.7	2.1		5.8
	40.0	6.6	2.4		6.4
8	4.7	14.6	2.2	3.0	
	6.2	12.8	2.1	3.0	
	8.5	11.0	2.2	3.2	
	10.7	10.0	2.3	3.0	
	15.5	8.0	2.0	3.4	
	21.0	6.6	2.0		5.6

tric field may be assumed uniform, the screening radius R_s. From the very definition of field-induced traps[7,8] it is clear that a trap can be no larger than R_s. If $R_s \geqslant L_c$, the field is uniform over the entire system, and the greatest delay is in traps whose length along the field is the greatest (of the order of or greater than L_c). Since such traps are exponentially rare, the average delay time in the motion along a macrolink is determined by the competition between two exponents. One of these determines the probability of a trap appearing, the other the time of delay in the trap.[9]

If the screening radius is so small that the field follows the current conductor, no traps appear and the quasimetallic situation realizes itself. Then Shklovskiĭ's results,[5] which predict an increase in hopping conductivity in a field to follow Eq. (4) or (6), are valid. Thus, a negative incremental resistance exists only if

$$R_s \geqslant L_c. \tag{7}$$

In practice, maintaining a uniform field in the VRH conduction regime is fairly difficult. Indeed, if the energy dependence of the density of states near the Fermi level $g(F_f)$ is weak and the linear screening theory holds true, then

$$R_s = \sqrt{\frac{\varepsilon}{4\pi e^2 g(E_f)}}.$$

Using, for instance, the known data on Si⟨P⟩ (Ref. 13), $g(E_f) = 5\times 10^{18}$ eV^{-1} cm^{-3} and $\varepsilon = 12$, we get $R_s = 150$ Å. The correlation radius can be found from Eq. (5). Typical values of the percolation threshold ξ_c at low temperatures amount to roughly 10-15. Taking $a=20$ Å and $\nu=0.9$, we get $L_c = 1000$ Å. Thus, not only is condition (7) invalid but $R_s \approx (2\text{–}3)\times R$, as one can easily see. In this case the dead ends and bottlenecks cannot be electron traps and the current-voltage characteristics must be superlinear, a situation observed in most experiments. Uniformity in the field can be achieved only in the special case where the compensation ratio K does not exceed 10^{-4} (Ref. 9). In a semiconductor with a Coulomb gap the low concentration of screening carriers is achieved by nullifying the density of states at the Fermi level.

Another feature of a semiconductor with a Coulomb gap is the nonlinear nature of screening. The screening of a potential φ is achieved by electrons with an energy $E_f = \pm e\varphi$. According to Eq. (1), the farther we are from the Fermi level, the greater the density of states within the Coulomb gap, so that screening a large potential fluctuation requires a high electron number density [with $\rho(\varphi)$ growing faster than φ], that is, the effective screening radius decreases as the screening potential increases. Hence, in a semiconductor with a Coulomb gap, as the voltage grows, one can expect a transition from the quasi-insulator hopping-conduction regime (the case of a uniform field) to the quasimetallic regime, a situation that manifests itself in experiments by the change from negative non-ohmic behavior to positive. We believe that this explains the disappearance of negative differential resistance for $F > 1000$ V cm^{-1} in Fig. 3 and the minima in hopping conductivity in Fig. 2. To our mind, the absence of negative differential resistance at low temperatures is due to the increase in L_c as T lowers so much that conditions (7) becomes invalid [see Eq. (5)]. At high temperatures ($T > T_b$) the Coulomb gap in the spectrum of states blurs, which means that the screening radius decreases, condition (7) breaks down, and negative differential resistance vanishes.

Let us return to Fig. 1. We see that for $T \leqslant 12.5$ K beyond the non-ohmic threshold and for $14.5 \leqslant T < 50$ K following the decrease in conductivity, there is observed an exponential increase in σ with field strength. Here the law (4) holds true for $T \geqslant 9.2$ K in intermediate fields. From the slope of the straight lines in Fig. 2 one can find the

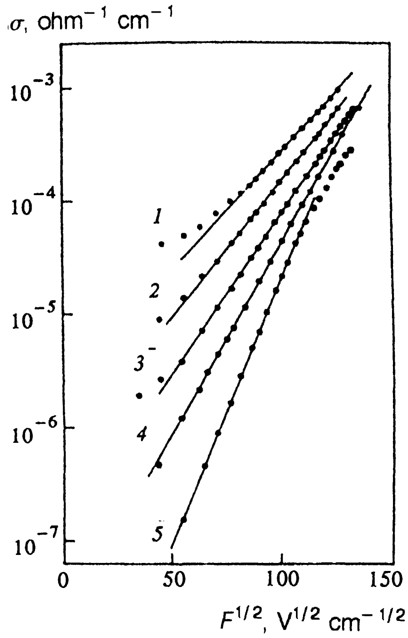

FIG. 5. Conductivity of an a-Si⟨Mn⟩ sample with $N=7$ at. % as a function of the electric field strength in log σ vs $F^{1/2}$ coordinates at different temperatures T: curve *1* corresponds to 14.5 K, curve *2* to 12.5 K, curve *3* to 9.2 K, curve *4* to 6.9 K, and curve *5* to 4.2 K.

non-ohmic length l. The dependence of l on the parameter $T_0/T=\xi_c^2$ for a-Si⟨Mn⟩ samples with impurity concentrations 7 and 8 at. % is depicted in Fig. 4. We found that

$$l \propto \left(\frac{T_0}{T}\right)^m, \tag{8}$$

where $m=0.51$ and 0.58 for $N=7$ and 8 at. %, respectively. On the other hand, the typical length of a hop over the critical subgrid is determined by the formula $R=a(T_0/T)^{1/2}/4$. On the basis of Eq. (4) we determined the size of parameter C_1, which links the non-ohmic length and the hopping length. We found that $C_1=2.0$–2.4 (see Table I) and is independent of ξ_c. Note that computer simulation predicts a linear increase in C_1 with the percolation threshold growing.[6]

As the electric field strength grows, the field dependence of conductivity weakens. Detailed analysis shows that in this case the link between σ and F is best described by the exponential function (6) (Fig. 5). For samples with Mn concentrations $N=7$ and 8 at. % an analysis of the ln σ vs $F^{1/2}$ dependence at different temperatures yielded a value $L=C_2L_c$ proportional to the correlation length of the conducting cluster. The results of such a procedure are depicted in Fig. 4.

If the spectrum of states of a semiconductor contains a Coulomb gap, L_c must grow, as the temperature lowers, like $(T_0/T)^{(1+\nu)/2}$. Setting the critical index at 0.9, we get $\frac{1}{2}(1+\nu)=0.95$. It was found in experiments that $L\propto(T_0/T)^m$, where $m=0.88$ and 0.92 for $N=7$ and 8 at. %, respectively, which supports the validity of the Coulomb-gap model.

Determining the constant C_2 from the experimental data is of practical interest because calculating it presents serious difficulties. From the formula $C_2=L/L_c=4L/a\xi_c^{1+\nu}$ we find that for different temperatures and impurity concentrations the value of C_2 lies between 2 and 3 (see Table I).

A detailed study of the sublinear sections of the experimental current-voltage characteristics has shown that the conductivity decrease in the field is described by the following formula:

$$\sigma(F)\propto \exp\left\{-\left(\frac{C_3eFL_c}{kT}\right)^{1/2}\right\} \tag{9}$$

with the factor C_3 in the 5.6 to 6.4 range.

We are grateful to V. A. Dravin for implanting Mn^+ ions in a-Si samples and to M. V. Éntin for numerous discussions of effects related to the fluctuations of φ in the event of a hopping-conductivity current.

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