Long-lived photoconductivity of AlGaAs: distribution of electrons between free and bound metastable states

I. E. Itskevich and V. D. Kulakovskiĭ

Institute of Solid-State Physics, Academy of Sciences of the USSR, Chernogolovka, Moscow Province
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A study was made of the behavior of a nonequilibrium electron system under conditions of long-lived photoconductivity of hydrostatically compressed ($P = 0$–12 kbar) $Al_x Ga_{1-x} As$ ($x \approx 0.29$) crystals doped with Si. Donor X states were found to be metastable (like free Γ states) relative to the L_5^* level (DX level). The distribution of photoexcited electrons between the Γ and X states was in thermodynamic equilibrium. Under pressures in excess of 9 kbar a qualitatively different metastable effect was observed: photoexcitation at 4.2 K transferred electrons from a deep L_5^* level to a higher X_5^* donor level without appearance of the long-lived photoconductivity, but in the range 20–50 K a metastable thermally activated conductivity was observed.

INTRODUCTION

The phenomenon of long-lived photoconductivity of $Al_x Ga_{1-x} As$ has been attracting attention for over a decade.[1] It is manifested as follows: when a sample is illuminated at low temperatures, its conductivity rises and this higher conductivity is retained for a considerable period (amounting to hours or days). The long-lived photoconductivity has been observed in $Al_x Ga_{1-x} As$ with aluminum concentrations in the range $x_{Al} = 0.20$–0.45 in GaAs under pressure and in n-GaAs/$Al_x Ga_{1-x} As$ heterojunctions, sometimes seriously degrading their properties.[2–8]

In the first investigations of the long-lived photoconductivity it has been suggested that a localized state from which photoexcitation takes place is a complex structure defect called a DX center (for example, a donor and vacancy). The long electron-trapping times of the DX center are explained by assuming that such trapping is accompanied by a change in the spatial configuration of the defect.[9,10] An alternative model is based entirely on the energy band structure. It is assumed in this model that photoexcitation takes place from a deep donor state associated with a side minimum of the conduction band. The long-lived photoconductivity is attributed to long tunneling times of electrons in $\mathbf{k}$ space.[11]

The published experimental results do not fit either of these models. For example, the first model does not explain why the number of the DX centers should be half the number of the donor impurities and why it is affected by hydrostatic compression which does not alter the structure of defects. The second model gives thermal activation energies much smaller than those found experimentally and, moreover, difficulties are encountered in explaining the long electron tunneling times, Currently the term DX center (or DX level) is used in the literature to denote a deep localized state from which the long-lived photoconductivity is excited irrespective of the nature of this center.

It seems to us that the best model is the one combining the ideas of changes in the spatial configuration of a defect as a result of capture of an electron with the characteristics of the energy band structure of the relevant semiconductor.[12] It is assumed that donor impurities in $Al_x Ga_{1-x} As$ are simple donors. Their special feature is the existence of two stable positions of the ionic core. All electron states, both free and localized at a donor, correspond to a stable position of the ionic core at a lattice site. Moreover, in the case of a donor level L_5^* which is not totally symmetric there is a second stable position of the ionic core, slightly displaced from a site (Jahn–Teller effect), which forms a DX level. If the first states are filled with electrons under thermodynamic equilibrium conditions, the second state is separated from them by an energy barrier. Calculations confirm the possibility of existence of such a state when an impurity atom is displaced along the $\langle 111 \rangle$ axis.[13,14]

The long-lived photoconductivity can appear if the following two conditions are satisfied: 1) the impurity concentration exceeds the Mott value for the $1s$ states of the Γ electrons but it is less than the Mott value for the L_5^* level with a shifted position of the ionic core (DX level); 2) the L_5^* level is the ground state and the Γ valley is located between this level and all the other electron states. This (or a similar situation) is indeed realized in mixed n-type $Al_x Ga_{1-x} As$ semiconductors doped with Si ($n_{Si} \gtrsim 10^{17}$ cm^{-3}) and with aluminum concentrations in the range $x = 0.20$–0.45, which exhibit long-lived photoconductivity below liquid nitrogen temperatures (when kT is much less than the height of the postulated potential barrier).

The Morgan theory[12] distinguishes sharply the DX level from the system of all other electron states separated from the former by a potential barrier and therefore metastable. Photoexcited electrons should be distributed uniformly between the free and localized states. The conductivity is one of the manifestations of the metastability associated with the Γ minimum. In principle, the model admits metastable effects associated also with other electron levels when the usual long-lived photoconductivity may be absent. These effects can be observed only if the "equilibrium" localized states have energies close to the Γ minimum or below it. In the case of $Al_x Ga_{1-x} As$ with $x > 0.40$ or with lower values of x this is the situation realized under pressure.

Very few experimental investigations have been made of these aspects of long-lived photoconductivity. In the range $x > 0.45$ this photoconductivity practically disappears in the case of $Al_x Ga_{1-x} As$ (Refs. 2 and 4). The ground state in the range $x > 0.45$ is a shallow donor with a binding energy ≈ 50 meV (Ref. 15). To the best of our knowledge, there have been no investigations of metastable effects (in the absence of the long-lived conductivity) in this range of x_{Al}. Our investigations of the long-lived photoconductivity

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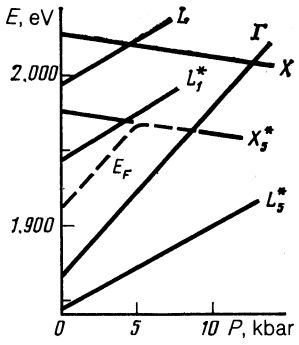

FIG. 1. Shifts of the Γ, L, and X valleys and of the donor L_5^* and X_5^* levels in $Al_{0.29}Ga_{0.71}As$ under pressure P. The expressions describing the energy positions E of the valleys are given in the text. The binding energies are assumed to be as follows: 50 meV for the X electrons and 50 and 150 meV for the L electrons (DX level). Here, E_F is the Fermi level of electrons in the Γ valley after illumination.

in n-GaAs/$Al_xGa_{1-x}As$ heterostructures under pressure have revealed a reduction in this photoconductivity attributed solely to changes in the energy band structure.[16] Clearly, this result also fits qualitatively the Morgan model.[12]

We carried out a further study of the nature of the long-lived photoconductivity by investigating the influence of the energy band structure on the properties of metastable electrons in $Al_xGa_{1-x}As$. We measured the Hall density of free electrons under the long-lived photoconductivity conditions applying hydrostatic pressures and varying temperatures from 4.2 K to ≈40 K. The pressures made it possible to modify the band structure, altering both the relative positions of "equilibrium" states as well as the position of the DX level relative to these states. The shifts of the levels under pressure in $Al_xGa_{1-x}As$ ($x \approx 0.29$) are shown schematically in Fig. 1. In the range of pressures from 0 to 10 kbar the X_5^* level crosses first the Fermi level of the Γ electrons and then the bottom of the Γ valley. Measurements carried out at different temperatures make it possible to study the filling of the Γ and X states with electrons. The present paper reports an investigation of the distribution of electrons between free and localized (bound) states under the long-lived photoconductivity conditions.

EXPERIMENTAL METHOD

Our measurements were carried out using a sample of n-type $Al_xGa_{1-x}As$ grown by the molecular beam epitaxy method on an insulating GaAs substrate carrying a buffer layer. The thickness of the $Al_xGa_{1-x}As$ film was 520 nm. A circuit needed for the Hall effect measurements was formed on the surface of a sample by lithography. The aluminum concentration $x = 0.29 \pm 0.01$ was deduced from the low-temperature luminescence spectrum. The concentration of the dopant (Si donor) was $\approx 8\times10^{17}$ cm^{-3}.

The sample was placed inside a self-contained low-temperature chamber where a fixed pressure was established; this chamber was filled with an oil–kerosene mixture.[17] The pressure inside the chamber was deduced from the temperature of the superconducting transition in an Sn manometer. Illumination was provided by a GaAs light-emitting diode placed inside the chamber next to the surface of a sample. The energy of the radiation emitted by this diode was always less than the band gap of $Al_{0.29}Ga_{0.71}As$. When temperature dependences were determined, the chamber was placed inside a vacuum container within a superconducting solenoid and the temperature was deduced using a calibrated thermistor.

The Hall emf u_{xy} was measured under ac conditions (at a frequency of ≈20 Hz) using a selective nanovoltmeter. The Hall electron density n was deduced from the linear slope of the dependence of u_{xy} on a magnetic field in the range $H \leqslant 4$ T; small changes in n were found from the deviation of u_{xy} from the initial values in a constant field of $H = 3.06$ T. These measurements were always repeated for opposite directions of H in order to eliminate the contribution of the component u_{xx}. The working values of the Hall resistance R_{xy} were tens of ohms, whereas the transverse magnetoresistance R_{xx} amounted to few or tens of kilohms, and measurements could be made right up to $R_{xx} \sim 1$ MΩ. We assumed that the conductivity was not observed when R_{xx} exceeded 10 MΩ ($\rho \approx 50$ Ω·cm).

EXPERIMENTAL RESULTS AND DISCUSSION

1. At zero (atmospheric) pressure the dark conductivity was retained by n-type $Al_{0.29}Ga_{0.71}As$ ($n_{Si} \approx 8\times10^{17}$ cm^{-3}) during cooling right down to helium temperatures (the Hall density of electrons at 4.2 K was $\approx 6\times10^{16}$ cm^{-3}). In the case of compressed samples ($P = 3.5$–12 kbar) the dark conductivity disappeared at 4.2 K.

At pressures up to 9 kbar it was found that illumination of a sample ($\lambda \approx 0.8$ μm) at 4.2 K created a conductivity. The conductivity increased during illumination, reaching finally a steady value. When illumination was interrupted, the conductivity first fell slightly (by no more than 2%) and then remained constant (to within 1%) for several days. Significant relaxation (representing an irreversible reduction) of the conductivity was observed at temperatures above 60 K. The maximum steady conductivity σ^* and the corresponding density n^* at 4.2 K depended on pressure; at a fixed pressure they were readily reproducible.

Figure 2 shows the pressure dependences of the maximum steady Hall density of electrons n^* and of the corresponding Fermi energy E_F^*. The value of E_F was found from the relationship[18]:

$$E_F = \frac{2\pi^2\hbar^2}{m^*}\left(\frac{3n}{8\pi}\right)^{1/3}\left[1 + \frac{\alpha}{E_g}\frac{2\pi^2\hbar^2}{m^*}\left(\frac{3n}{8\pi}\right)^{1/3}\right],$$

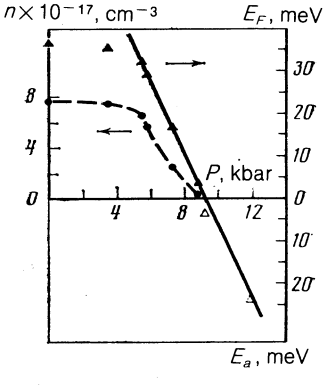

FIG. 2. Dependences, on the applied pressure P, of the maximum Hall density of free electrons after illumination n (●), of the corresponding Fermi energy E_F (▲), and of the activation energy of conduction electrons at high pressures E_a (△).

where m^* is the effective mass of the Γ electrons; α is the nonparabolicity coefficient; E_g is the band gap of $Al_{0.29}Ga_{0.71}As$; the pressure dependences of E_g and m^* were allowed for.

[The following data were used in the calculations. The energy positions of the Γ, X, and L valleys in GaAs at 4.2 K relative to the maximum of the valence band were assumed to be $E_\Gamma = 1.519$ eV, $E_L = 1.815$ eV, and $E_X = 1.981$ eV (Ref. 18). The shift of the valleys with x_{Al} was $\Delta E_\Gamma(x) = 1.247x$, $\Delta E_L(x) = 0.642x$, $\Delta E_x(x) = 0.125x + 0.143x^2$ eV (Ref. 19). The effective mass of the Γ electrons was[19]

$$m^* = m_0 \Big/ \left[1 + 7.51\left(\frac{2}{E_\Gamma} + \frac{1}{E_\Gamma + 0.341}\right)\right],$$

where m_0 is the mass of a free electron. It was also assumed that $\Delta E_\Gamma(P) = 12.6P$ (Ref. 18). The nonparabolicity coefficient was assumed to be $\alpha = -0.824$ (Ref. 18).]

It is clear from Fig. 2 that at pressures below 5 kbar the maximum electron density did not change greatly, but at higher pressures there was a considerable reduction in n^*. The electron Fermi energy decreased in this range of pressures linearly at a rate $dE_F^*/dP = (8.7 \pm 0.5)$ meV/kbar.

We shall analyze the behavior of $E_F^*(P)$ by considering changes in the energy band structure of $Al_{0.29}Ga_{0.71}As$ under pressure (Fig. 1). According to the published data, the Γ and X valleys converge under pressure at a rate $dE_{\Gamma-X}/dP = 12$–13 meV/kbar, whereas the Γ and L valleys converge at a rate $dE_{\Gamma-L}/dP = 6$ meV/kbar (Refs. 19 and 20). Therefore, it is logical to attribute the reduction in E_F^* to the capture of electrons by states in the X valley. The lowest of these is the donor level X_5^*, in resonance with the Γ valley. Since the density of states at the X_5^* level is much higher than in the Γ valley, it follows that after filling of the Γ states located below the energy of this level the capture of photoelectrons occurs mainly at this level.

The lower value of dE_F^*/dP compared with $dE_{\Gamma-X}/dP$ can be explained by broadening of the donor level due to inhomogeneity of the distribution of Al in the sample. The fall of E_F^* from its maximum value to zero occurs in the range of pressures from 5 to 9 kbar. At $P = 5$ kbar the level E_F^* is at the lower edge of the broadened donor level, whereas at $P = 9$ kbar it is at the upper edge. Hence, we can estimate the width of the donor level, which is ≈ 12–15 meV. It should be pointed out that the scatter of Al concentrations ($x_{Al} = 0.29 \pm 0.01$) deduced from optical measurements yields a similar estimate of the width of the level amounting to ≈ 12 meV.

The data presented in Fig. 2 can be used to estimate the binding energies E_{dX} and E_{dL} for the X and L electrons at an Si donor. An estimate of E_{dX} can be obtained by extrapolation of the linear part of the dependence $E_F(P)$ (in the range 5–9 kbar) to zero pressure, which gives $E_F(0) = 80$–100 meV and yields $E_{dX} = 50$–70 meV. The values of E_{dX} given in the literature lie within the range 40–50 meV (Refs. 2, 15, 21). Similarly, using the Fermi energy of the dark electrons at zero pressure $E_F \approx 7$–8 meV and bearing in mind that cooling to typical relaxation temperatures (≈ 80 K) causes freezing of electrons in the Γ valley, we can estimate the binding energy of electrons at the DX level, which is $E_{dL} \approx 150$ meV. According to the published data, this energy is $E_{dL} \approx 120$–160 meV (Refs. 4, 13, 19, and 21).

2. The reduction in the maximum density of the Γ electrons under pressure will be attributed to the capture of electrons by the X_5^* donor level, which is in agreement with a number of models. In the range of pressures 5–9 kbar the L_5^* level is the ground state, whereas the X_5^* level is located higher, in resonance with the Γ valley (Fig. 1). The principal question is what happens to electrons captured in the X state. According to the ideas based only on energy band structure, electrons may relax back via the same state to the level L_5^* from which they have been excited optically.[2,11] However, according to Morgan,[12] such relaxation is impossible and electrons fill the state X_5^* under thermodynamic equilibrium with the Γ valley. Both these states X_5^* and Γ are metastable relative to the L_5^* level, because overcoming of the potential barrier associated with the lattice deformation is the requirement for the capture of an electron by the L_5^* level from the Γ valley or from the X states.

It is possible to determine experimentally whether metastable filling of the X_5^* level takes place: this is done by recording the temperature dependences of the density of free electrons below typical relaxation temperatures. If electrons return to the DX level, then all the changes in the electron density as a result of a slight increase in temperature T are irreversible. However, if electrons fill the X state metastably then variation of T results in a reversible redistribution of electrons between the states and the Γ valley and the process increases in intensity the closer the X_5^* level is to the Fermi level of free electrons.

A sample was illuminated at a fixed pressure until the different densities of free electrons were established (in the range from zero to the maximum value) and then the change in the Hall resistance R_{xy} was determined in a static magnetic field $H = 3.06$ T as a function of T. The working range of temperatures was within 40 K, i.e., relaxation was avoided: all the changes in the Hall resistance were reversible. The dependences $\Delta R_{xy}(T)$ were recorded for two opposite directions of the magnetic field, smoothed out, and averaged, and then the change in the Hall density $\Delta n(T)$ was calculated. The Hall factor was assumed to be unity.

The dependences $\Delta n(T)$ obtained for different carrier densities and pressures are shown in Fig. 3. (The values of the density n, Fermi energy E_F, and pressure P corresponding to different curves are listed in Table I.) Clearly, some of the curves reproduce one another quite well, whereas others

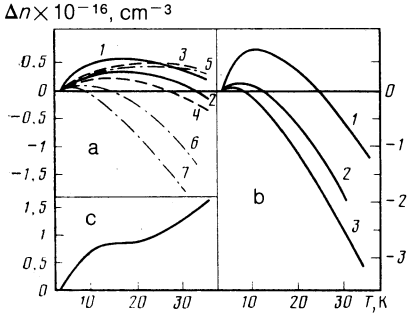

FIG. 3. Dependences of the Hall density of free electrons Δn on the temperature T of a sample obtained for different carrier densities (at 4.2 K) and pressures. The experimental dependences were smoothed out and averaged for two opposite directions of the magnetic field. The values of n, E_F, and P used for different curves are listed in Table I.

TABLE I.

Figure	Curve	P, kbar	$n\times10^{-17}$, cm^{-3}	E_F, meV
3, a	1	0	1.25	11.1
	2	0	7.65	37.0
	3	3.5	1.5	12.4
	4	3.5	7.45	35.5
	5	5.5	1.75	13.6
	6	5.5	5.3	28.0
	7	5.5	6.6	32.5
3. b	1	7.3	1.1	9.9
	2	7.3	2.0	14.6
	3	7.3	2.45	16.8
3, c		8.8	0.24	3.5

differ quite significantly. We can conclude that for all the dependences of the first group the energy gap between the Fermi level of the Γ electrons E_F and the X_5^* donor level exceeded 20 meV. A qualitative change in the dependences occurred when E_F approached the donor level or crossed it. It should be stressed once again that all the temperature dependences of the Hall density of carriers were accurately reversible.

The actual nature of the dependences $\Delta n(T)$ for the first group could not be identified with certainty. It could be due to the effect of temperature on the Hall factor. The ratio $\Delta n/n$ for these dependences was less than 3–4% and their nature was in qualitative agreement with the results of calculations of the Hall factor of the Γ electrons in GaAs (Ref. 22). In this case the dependences did not reflect the real changes in the density of free electrons. Deviations of the dependences $\Delta n(T)$ from this initial form (for E_F close to X_5^*) could be readily explained assuming that electrons excited optically from the DX level had a thermodynamic-equilibrium distribution between the Γ valley and the donor states X_5^*. When E_F approached a smeared donor level from below, the density of the states localized above E_F was considerably higher than below it: firstly, we were moving toward the maximum of the energy distribution of these states and, secondly, each unoccupied impurity state was degenerate. The density of the free states in the Γ valley changed only slightly and, therefore, heating caused the transfer of free electrons to the impurity level so that the density of free electrons fell. This effect was manifested more strongly as E_F approached the donor level. This occurred on increase in the carrier density (due to illumination) and on increase in the applied pressure, as demonstrated by the curves obtained at 5.5 and 7.3 kbar. At 7.3 kbar the maximum illumination established a situation in which the donor level was approximately half-filled and the difference between the densities of the localized states above and below the Fermi level was largest (after allowing for the degeneracy of the vacant states). In this situation the transfer of electrons from free states to the localized ones was manifested most strongly: heating from 4.2 to 35 K changed the carrier density by almost 14% and the change was fully reversible.

When the majority of the X_5^* donor states was filled after illumination, the density of the localized states above the Fermi level was lower than below it. Therefore, heating transferred electrons from the donor level to the Γ valley and the density of free electrons increased, as we indeed observed under a pressure of 8.8 kbar.

The results obtained thus demonstrated that under the long-lived photoconductivity conditions both the Γ valley and the X_5^* donor level were filled metastably with electrons excited from the DX level and the distribution between the Γ valley and X_5^* was in thermodynamic equilibrium.

3. A qualitatively new effect was observed at pressures above 9 kbar. When a sample was cooled, the dark conductivity disappeared already at $T\sim250$ K. Illumination at 4.2 K did not produce conductivity. This was usually regarded as the absence of the long-lived photoconductivity. However, when the sample illuminated at 4.2 K was heated to $T\approx15$–20 K, a conductivity σ did appear. At $T\approx40$–50 K the conductivity and the density of free electrons increased considerably; in this range of temperatures the changes were fully reversible. However, heating above 50–60 K caused the conductivity σ to disappear rapidly and irreversibly, and observation of the conductivity at 20–40 K required a second illumination.

The dependence of the Hall density of free electrons on the reciprocal temperature of a sample subjected to maximum illumination and to pressures of 9.2 and 11.8 kbar is plotted on a logarithmic scale in Fig. 4. The dependence is clearly quite linear. The corresponding activation energies E_a are plotted in Fig. 2. Clearly, these energies together with the maximum values of the Fermi energy fit well the same linear pressure dependence.

This qualitatively new form of the long-lived photoconductivity can be explained completely by the Morgan hypothesis.[12] The X_5^* donor level, which limits the maximum electron density n^*, drops at pressures above 9 kbar below the bottom of the Γ minimum and is the lowest of the "equilibrium" states. Therefore, illumination causes electrons excited from the L_5^* level to fill the X_5^* level, i.e., electrons are transferred from one localized state to another: there is no conductivity. Heating to 20–40 K activates electrons from the X_5^* level to the Γ valley: we observe conductivity. The linear nature of the dependences in Fig. 3 confirms that the Γ valley becomes filled with electrons in thermodynamic equilibrium with the X_5^* level. Further heating causes thermally activated relaxation of electrons to the level L_5^* from the Γ minimum and from the X donor states.

These results confirm directly that in the case of

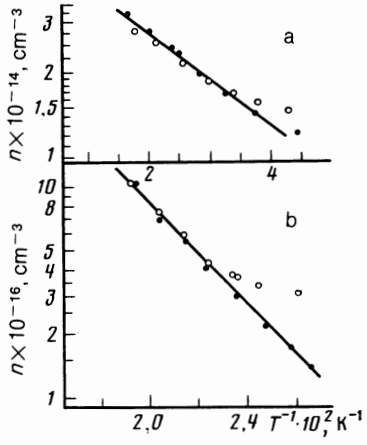

FIG. 4. Dependences of the density of free electrons n (on a logarithmic scale) on the reciprocal of the temperature T^{-1} of a sample subjected to pressures of 9.2 kbar (a) and 11.8 kbar (b). The black dots (●) denote the values obtained during heating, whereas the open circles (○) were recorded during cooling.

$Al_xGa_{1-x}As$ ($x \approx 0.3$) the L_5^* donor level (DX level) is separated by an energy barrier from both free and localized states and the distribution of electrons between these states corresponds to thermodynamic equilibrium. Illumination of a sample at liquid helium temperatures metastably fills the lowest of the "equilibrium" states. In this case there is no fundamental difference between free and localized states, but in the latter case the long-lived photoconductivity may appear in a qualitatively new form or not at all. For example, in the case of $Al_xGa_{1-x}As$ ($x \approx 0.3$) at $P \approx 13$–14 kbar we might expect the energy of activation of an electron from a metastable donor level to the nearest vacant state to be comparable with the energy barrier for the relaxation of electrons to the DX level. In this case illumination should result in metastable filling of the level X_5^*, but it is generally impossible to observe the metastable conductivity. Different methods have to be employed in order to detect metastable filling of the donor level under these conditions.

CONCLUSIONS

Our results demonstrate that both free and localized states in $Al_xGa_{1-x}As$ ($x \approx 0.3$) are separated from the DX level by an energy barrier. The distribution of electrons between these two types of states corresponds to thermodynamic equilibrium under the long-lived photoconductivity conditions. Hence, it follows that $Al_xGa_{1-x}As$ may in principle exhibit a wider range of metastable phenomena in addition to the long-lived photoconductivity, which is one of the manifestations of the metastability. The results reported above demonstrate that in the case of $Al_{0.29}Ga_{0.71}As$ under a pressure of $P \gtrsim 5$ kbar the lowest metastable donor state corresponds to the X valley. Under pressures in excess of 9 kbar a new metastable effect appears: photoexcitation at 4.2 K transfers electrons from one donor state to another (no long-lived photoconductivity is present); in a narrow range of temperatures (20–50 K) we may observe metastable thermally activated conductivity.

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