Electron Concentration Dependence of the Coulomb Gap in AlGaAs:Si

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(Received 22 July 1997)

The Efros-Shklovskii (ES) $T_{1/2}$ and Mott $T_{1/4}$ parameters and the width of the soft Coulomb gap in $Al_{0.3}Ga_{0.7}As$:Si have been determined for electron concentration n ranging from 1.2×10^{16} to 14×10^{16} cm^{-3}. The gap width presents a maximum at an n value corresponding to a compensation ratio K between 0.94 and 0.97. The $T_{1/2}$ and $T_{1/4}$ parameters decrease exponentially with n and provide the simple relation $e^2 g_0 \xi^2/\kappa \propto n$, between the localization length ξ and the dielectric constant κ. This is compatible with scaling theory. Our data also suggest that the validity limits for the Mott and ES regimes should be estimated, respectively, by $2.5(T_{1/2})^2/T_{1/4}$ and $(T_{1/2})^2/T_{1/4}$. [S0031-9007(98)05315-0]

PACS numbers: 71.30.+h, 71.20.–b, 71.55.Jv, 72.20.–i

A doped semiconductor is an insulator when its doping concentration is lower than a critical value n_c. When the majority dopants (supposed here to be donors) are not compensated, n_c is determined approximately from Mott's criterion $a_0 n_{\mathrm{Mott}}^{1/3} = q$, where a_0 is the dopant's Bohr radius and $q \approx 0.26$ [1,2]. For highly compensated samples n_c is larger than n_{Mott} and can be found from Mott's criterion with $0.36 < q < 0.40$ [3]. In the insulator regime and at low enough temperatures, electrical conduction is achieved by variable range hopping, with a conductivity $\sigma \approx \exp[(-T_p/T)^p]$ [1,4]. The most widely accepted values for p are $1/4$ and $1/2$, proposed by Mott and by Efros and Shklovskii (ES), respectively. However, other values for p have been reported [5–8]. The exponent p is related to the behavior of the density of states $g(E)$ close to the Fermi level E_F. Mott's model assumes a constant value g_0 for the density of states, whereas Efros and Shklovskii found that the Coulombic interaction between the carriers forces $g(E)$ to go smoothly to zero, with $(E - E_F)^2$ in 3D systems. When $g(E)$ behaves the same as $|E - E_F|^m$ it is said to have a soft gap at E_F, and it gives $p = (m + 1)/(m + 4)$ [9,10]. The Mott ($T_{1/4}$) and ES ($T_{1/2}$) parameters are given by

$$k_B T_{1/4} = \beta/\xi^3 g_0, \quad k_B T_{1/2} = 2.8 e^2/\kappa\xi\,, \qquad (1)$$

where ξ is the localization radius of the electrons, κ is the dielectric constant, k_B is the Boltzmann constant, e is the electron charge, and β is a constant whose value is not precisely known [1], but 18 and 21 are the most used values [2,4,11]. The mean hopping energies in Mott ($T_{1/4}$) and in ES ($T_{1/2}$) regimes are given, respectively, by $\Delta_{\mathrm{Mott}} = k_B T (T_{1/4}/T)^{1/4}/4$ and $\Delta_{\mathrm{ES}} = k_B T (T_{1/2}/T)^{1/2}/2$, and the half-width Δ_{CG} of the soft Coulomb gap can be estimated by [4]

$$\Delta_{\mathrm{CG}} = e^3 \sqrt{g_0/\kappa^3} \approx k_B T_{1/2} \sqrt{T_{1/2}/T_{1/4}}\,. \qquad (2)$$

Several experimental results have been interpreted on the grounds of the above-described models [4,10,11]. However, some interpretations have been questioned because the Efros-Shklovskii regime was ascribed to experimental results obtained at temperatures where kT is higher than the Coulomb gap obtained from the same results [11]. This regime is expected to appear at temperatures lower than some characteristic limit T^{**}, such that the thermal effects are not able to shadow the soft gap. At higher temperatures, one expects a crossover to the Mott regime that would become fully applicable at temperatures higher than another limit T^*. These limits have been estimated [11] by the criteria $\Delta_{\mathrm{ES}} = \Delta_{\mathrm{CG}}$ and $\Delta_{\mathrm{Mott}} = 2\Delta_{\mathrm{CG}}$, which turn into $T^{**} = 4(T_{1/2})^2/T_{1/4} = 4\Delta_{\mathrm{CG}}\sqrt{T_{1/2}/T_{1/4}}$ and $T^* = 4T^{**}$. The temperature range $T^{**} < T < T^*$ defines the crossover regime, where neither Mott's nor ES's rule is expected to apply. Aharony *et al.* [12] have proposed a universal formula to fit the resistivity in this crossover range. Another challenging point has been stressed by Pollak and co-workers [13] who argue that the gap in the density of states, rather than soft, could be a hard gap, meaning that $g(E)$ goes to zero at E_F more sharply than $|E - E_F|^m$. This would be caused by multielectron effects. Recent studies, however, predict that these effects are consistent with the $T_{1/2}$ behavior, with a smaller value for the $T_{1/2}$ parameter [14], or with the Aharony formula [15]. The hard gap hypothesis would lead to a conductivity with a simple thermally activated behavior or even with a power law dependence on temperature.

Experimental works have shown the presence of the Coulomb gap in three- [16], two- [17], and one-dimensional [18] systems. The presence of a soft gap has also been detected by the resistivity behavior at temperatures corresponding to the crossover from the Mott to the ES regime [5,12,19–21] and by tunneling spectroscopy [22]. The presence of a hard gap has also been reported at temperatures lower than those of the variable range hopping, which is created by magnetic correlations due to exchange [23,24] or simply by electrical interactions [25].

The dependence of the Coulomb gap width Δ_{CG} on the electron concentration and on the compensation ratio

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K, as far as we know, has not yet been experimentally established. One can predict from Eq. (2) that Δ_{CG} will decrease to zero at $K = 1$, where the density of states g_0 is also zero. This would also be true for $K = 0$, if the two Hubbard subbands are split.

We have obtained the dependence of the width of the Coulomb gap Δ_{CG} on the electron concentration n and on the compensation ratio K, in a wide range of these parameters, from the n dependence of the hopping conductivity. A simple relation between $T_{1/2}$ and $T_{1/4}$ was found, and the validity limits for Mott and ES regimes were analyzed. We used the persistent photoconductivity (PPC) properties of the DX centers in $Al_{0.3}Ga_{0.7}As$:Si to change the carrier concentration, instead of using different samples. This method has ingeniously been employed by Katsumoto *et al.* [8] in order to determine the exponent of the dependence of the conductivity on $(n - n_c)$, close to the metal-insulator transition. The DX center, according to Chadi and Chang [26], is a deep level state of the dopant Si atom with an extra electron. Therefore it corresponds to a state of the second Hubbard subband which is lowered, by lattice deformation, to a deep position in the crystal energy gap [26,27]. The shallow donor state is metastable with a lifetime as long as months or years at temperatures below 100 K. The formation of a DX center requires one dopant atom (with its own electron) and one more electron taken from another atom. Then the first atom will no longer be a shallow donor, and it works as a compensating center for the second one, which will be an ionized donor with an empty state in the shallow impurity band. So, the creation of DX centers leads to a decrease in the shallow donor concentration (N_d) and, simultaneously, to an increase in its ionized part. Therefore, the DX center plays two roles in controlling the electron concentration: It changes the donor concentration and also behaves as a compensating defect. Hence, disregarding any unintentional acceptor dopants, one has $N_d = (N_{Si} + n)/2$, $N_{DX} = (N_{Si} - n)/2$, $K = N_d/N_{DX}$, where N_{Si} is the silicon concentration and N_{DX} is the DX center concentration. A DX center can be converted into a shallow donor by shining light on the sample and this can be used to vary n, N_d, and K.

Our sample consisted of a 3.8 mμ thick $Al_{0.3}Ga_{0.7}As$ layer grown by molecular-beam epitaxy, at 620 °C, on the top of GaAs. It was processed by photolithographic techniques in a Hall bridge pattern with a 300 μm width central channel and 800 μm distance between successive arms. The sample was slowly cooled down to 0.32 K in order to get a high concentration of DX centers. The temperature was varied from 0.32 to 60 K. The electron concentration was varied by incidence of successive doses of light-emitting diode radiation with an energy quantum smaller than the forbidden band gap of the sample. From the light doses, always applied at the same temperature, the electron concentrations were obtained [28]. Figure 1 shows the temperature dependence of the resistivity for several consecutive light doses corresponding to n values between 1.2×10^{16} and 16.6×10^{16} cm^{-3} and to compensation ratios from 0.6 to 0.97, respectively. The donor concentration, however, varies only between 34×10^{16} and 42.4×10^{16} cm^{-3}. This gives an almost constant mean separation between donors, $r_d \approx (N_d)^{-1/3}$, which, by chance, is approximately equal to twice the Bohr radius $a_0 \approx 70$ Å. At the highest n value, 16.6×10^{16} cm^{-3}, the sample resistivity shows a diffusive behavior [1], $\rho^{-1} \sim \sigma_0 + \sigma_1 T^m$. At the next lower value, 13.6×10^{16} cm^{-3}, it already shows hopping conductivity, following perfectly the $T_{1/4}$ rule up to 10 K (see inset in Fig. 1). Therefore, the critical concentration n_c lies between these values of n and is much higher than n_{Mott}, which for this material is approximately 5×10^{16} cm^{-3}. These limits for n_c give a value for $a_0 n_c^{1/3}$ between 0.38 and 0.40, confirming the compensating character of the DX center [3] and so its negative charge. This conclusion reinforces the assumption of the Chadi-Chang model for the DX center, which, some years ago, was a big challenge [27]. All of the other data seem to present characteristics of both $T_{1/4}$ and $T_{1/2}$ regimes and, apparently, have no signature of a hard gap. Figure 1 also shows

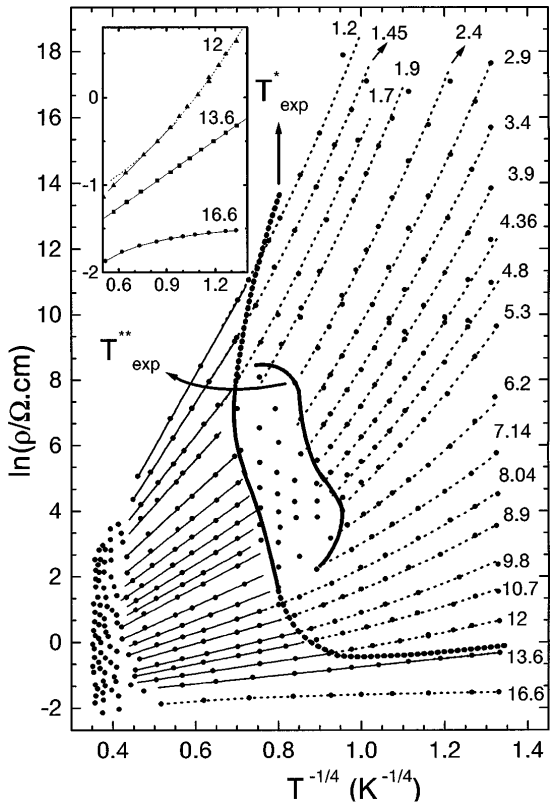

FIG. 1. Log of resistivity versus $-1/4$ for several carrier concentrations, whose values are indicated next to each curve in units of 10^{16} cm^{-3}. The lines named T^*_{exp} (solid for group A and dotted for group B) and T^{**}_{exp} define the limits of validity for Mott and ES regimes, respectively. The inset shows the data corresponding to the three highest values of n (16.6, 13.6, and 12) fitted with the formula for metallic, Mott, and Mott plus ES regimes, respectively. Its axes labels are the same as in the main figure.

the fitting curves of the data to Mott's regime (straight solid lines at higher temperatures) and to ES's regime (dotted parabolic lines at lower temperatures). The obtained $T_{1/4}$ and $T_{1/2}$ values are plotted in Fig. 2 for different electron concentrations. The validity limits for these regimes are shown by the bold lines, named T^{*}_{exp} and T^{**}_{exp}, drawn at the ends of their respective fitting curves. The estimated T^{*}_{exp} and T^{**}_{exp} limit values are shown in Fig. 3 for each n. From these limits, one can see that the data shown in Fig. 1 can be divided into two distinct groups. Group A, with $T^{*}_{\mathrm{exp}} > T^{**}_{\mathrm{exp}}$, is characterized by the existence of a crossover between $T_{1/4}$ and $T_{1/2}$ regimes, in a temperature range where the data do not follow either of them. This occurs for n ranging from 1.7×10^{16} to 7.1×10^{16} cm^{-3} (the region where the T^{*}_{exp} and T^{**}_{exp} lines in Fig. 1 are solid). For this group, one can doubtless rely on the experimental values of the validity limits for the Mott (T^{*}_{exp}) and ES (T^{**}_{exp}) regimes. Group B is formed by data with n within the ranges 1.2×10^{16} to 1.9×10^{16} cm^{-3} and 8.0×10^{16} to 12×10^{16} cm^{-3}. They apparently have coincident values for T^{*}_{exp} and T^{**}_{exp}. Good fits are obtained, with the $T_{1/2}$ formula, up to T^{*}_{exp} or even at temperatures above this limit. The T^{*}_{exp} line in Fig. 1 is dotted in the regions of group B. We have tried to use the universal formula proposed by Aharony *et al.* [12]. However, it does not fit the data in group A at all and, although at low temperatures it fits well the data in group B, at high temperatures it is worse than Mott's law. Additionally, Aharony's equation predicts $T_{1/4}$ values which are much lower than those obtained from Mott's law, and they do not agree with the values obtained for the data in group A. So a more appropriate model for this kind of crossover is still lacking and we cannot be very confident on the parameters obtained for the data in group B. However the fits of the data in group A are reliable and we will focus our analysis on them. The $T_{1/2}$ and $T_{1/4}$ parameters follow the relation

$$T_{1/2} \approx (0.524 r_d)^3 n T_{1/4} = 0.144 (n/N_d) T_{1/4}\,, \qquad (3)$$

as can be seen in the inset of Fig. 2. From Eqs. (1) and (3), one has

$$e^2 g_0 \xi^2/\kappa \approx n/N_d = 1 - K\,. \qquad (4)$$

This means that the number of states at the Fermi level (not considering the Coulomb gap), in a volume ξ^3 and in an energy interval equal to the electron-electron interaction at a distance ξ, is equal to n/N_d. This number is closely related to the characteristic overlap between neighbor states [1]. Equation (4) represents a striking result because, if it is also valid near the metal-insulator transition, where scaling arguments can be applied, then ξ^2 and κ will have the same scale behavior, as expected [1]. Therefore, Eq. (3) seems to be more appropriate than the $T_{1/2} \sim (T_{1/4})^{2/3}$ relation found by Zhang *et al.* [19]. The dependence of $T_{1/2}$ upon n can be described by the equation

$$T_{1/2} = (550\ \mathrm{K}) \exp(-n/2 \times 10^{16}\ \mathrm{cm}^{-3})\,, \qquad (5)$$

which fits well the data of both groups A and B. In the limit of very low concentration, one expects that the localization length is the Bohr radius for a shallow

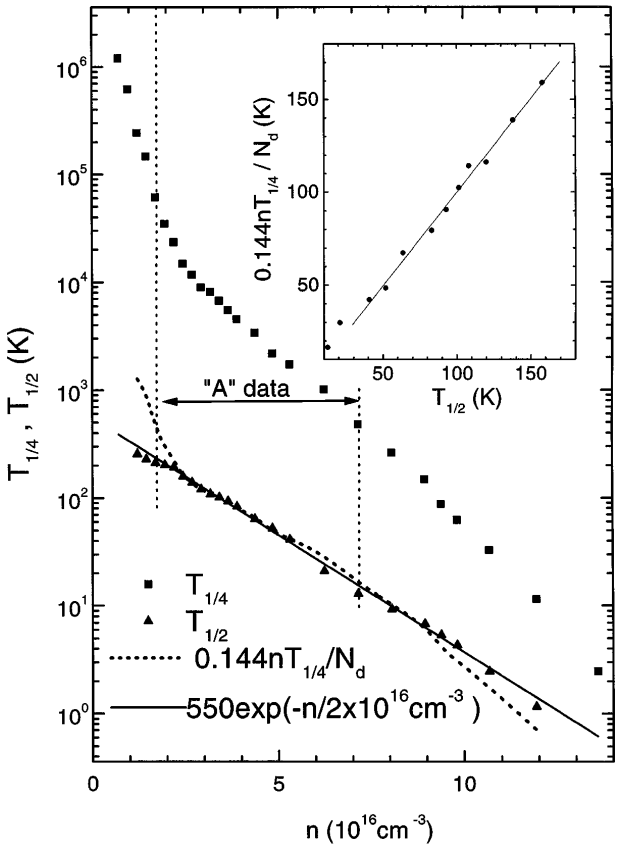

FIG. 2. Plot of $T_{1/4}$ and $T_{1/2}$ versus n. The solid lines is an exponential fit to the $T_{1/2}$ data. The region between the two vertical dotted lines corresponds to data in group A. The inset shows a fit which describes the relation between $T_{1/4}$ and $T_{1/2}$.

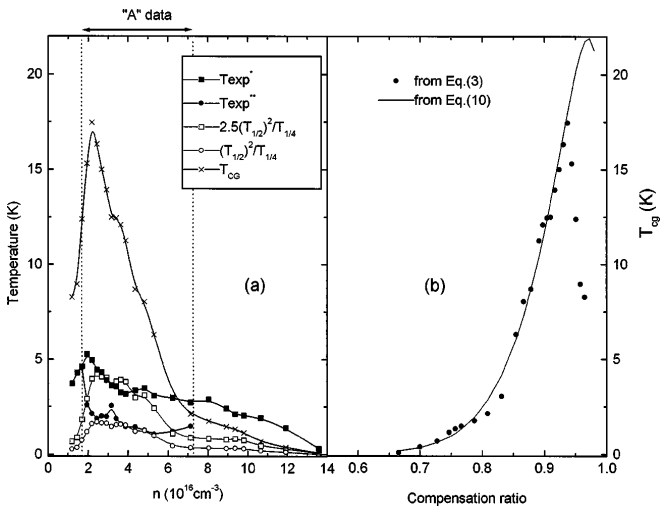

FIG. 3. (a) The dependence on n of the Coulumb gap, of the experimental crossover limits, and of $2.5(T_{1/2})^2/T_{1/4}$ and $(T_{1/2})^2/T_{1/4}$. (b) The Coulomb gap dependence on the compensation ratio.

impurity (70 Å) and the dielectric constant is close to the value 12.2, characteristic of the pure material (without free electron screening). These values and Eq. (1) give a low n limit for $T_{1/2}$ which agrees very well with Eq. (5). This suggests that the parameters obtained for the $T_{1/2}$ regime may be reliable even for the B group of data, mainly at low n values.

The Coulomb gap was estimated by Eq. (2), and in Fig. 3 $T_{\mathrm{CG}} \approx \Delta_{\mathrm{CG}}/k_B$ is plotted versus n and versus the compensation ratio K. These plots show a very sharp maximum at $n \approx 2 \times 10^{16}$ cm^{-3} ($K \approx 0.94$) due to the rapid increase of $T_{1/4}$ at low n values, not followed by $T_{1/2}$. As these parameters may have not been well estimated at low n values, we also used Eqs. (2), (3), and (5) to find the following n dependence of the Coulomb gap:

$$T_{\mathrm{CG}} = (209\ \mathrm{K})\,(n/N_d)^{1/2} \exp(-n/2 \times 10^{16}\ \mathrm{cm}^{-3})\,. \tag{6}$$

This relation predicts a maximum for the gap width at $n \approx 1 \times 10^{16}$ cm^{-3} (or $K \approx 0.97$). Therefore, our experiments indicate a maximum in T_{CG} at a compensation ratio K_0 between 0.94 and 0.97. As the gap width increases for K above 0.6 (the lowest K value), it should present a maximum at $K < 1$, because at $K = 1$ the gap has to disappear. Then the existence of the maximum is compatible with the behavior of the whole data. The dependence of the gap width on K was studied by Pollak and Ortuño [13] who predicted a maximum at $K = 0.5$. This result considers a fixed ratio between the characteristic Coulombic and disorder energies and so is not expected to apply to our results. At $K = 0.5$ our sample is already conducting. In order to estimate the correct behavior of the gap width, one has to be able to predict the dependence of that energy ratio on the electron concentration.

In Fig. 3 we show also the experimental estimates for the validity limits of the Mott and ES regimes ($T^{*}_{\mathrm{exp}}, T^{**}_{\mathrm{exp}}$). They indicate that, as far as group A is concerned, the lower limit for the Mott regime and the upper limit for the ES regime are much lower than T_{CG}. The figure shows that these limits compare well with $2.5(T_{1/2})^2/T_{1/4}$ and $(T_{1/2})^2/T_{1/4}$, respectively. These formulas correspond to $\Delta_{\mathrm{Mott}} \approx \Delta_{\mathrm{CG}}/2$ and $\Delta_{\mathrm{ES}} \approx \Delta_{\mathrm{CG}}/2$. Therefore, our data suggest that these could be better criteria for estimating T^{*} and T^{**} than those usually adopted.

We thank F. L. Veas for the discussion on the experimental procedures, and FAPEMIG, CNPq, and FINEP for financial support.

[1] N. F. Mott, *Metal-Insulator Transitions* (Taylor & Francis, London, 1990), 2nd ed.

[2] N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials* (Oxford University Press, New York, 1971).

[3] H. Fritzsche, Philos. Mag. B **42**, 835 (1980); N. F. Mott and J. H. Davies, Philos. Mag. B **42**, 845 (1980).

[4] B. I. Shklovskii and A. L. Efros, *Electronic Properties of Doped Semiconductors* (Springer-Verlag, Berlin, 1984).

[5] R. Rosenbaum, Phys. Rev. B **44**, 3599 (1991).

[6] F. Tremblay, M. Pepper, D. Ritchie, D. C. Peacock, J. E. F. Frost, and G. A. C. Jones, Phys. Rev. B **39**, 8059 (1989).

[7] W. N. Shafarman, D. W. Koon, and T. G. Castner, Phys. Rev. B **40**, 1216 (1989).

[8] S. Katsumoto, F. Komori, N. Sano, and S. Kobayashi, J. Phys. Soc. Jpn. **56**, 2259 (1987); S. Katsumoto, Inst. Phys. Conf. Ser. **108**, 17 (1991).

[9] F. D. Allen and C. J. Adkins, Philos. Mag. **26**, 1027 (1972).

[10] E. M. Hamilton, Philos. Mag. **26**, 1043 (1972).

[11] T. G. Castner, in *Hopping Transport in Solids*, edited by M. Pollak and B. Shklovskii (North-Holland, Amsterdam, 1991), pp. 1–47.

[12] A. Aharony, Y. Zhang, and M. P. Sarachik, Phys. Rev. Lett. **68**, 3900 (1992).

[13] M. Pollak and M. Ortuño, *Electron-Electron Interactions in Disordered Systems* (Elsevier, New York, 1985), pp. 287–408.

[14] A. Pérez-Garrido, M. Ortuño, E. Cuevas, and J. Ruiz, Phys. Rev. B **55**, R8630 (1997).

[15] Y. Meir, Phys. Rev. Lett. **77**, 5265 (1996).

[16] W. L. McMillan and J. Mochel, Phys. Rev. Lett. **46**, 556 (1981).

[17] M. J. Burns, W. C. McGinnis, R. W. Simon, G. Deutscher, and P. M. Chaikin, Phys. Rev. Lett. **47**, 1620 (1981).

[18] A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. Lett. **56**, 532 (1986).

[19] Y. Zhang, P. Dai, M. Levy, and M. P. Sarachik, Phys. Rev. Lett. **64**, 2687 (1990).

[20] I. Shlimak, M. Kaveh, M. Yosefin, M. Lea, and P. Fozooni, Phys. Rev. Lett. **68**, 3076 (1992).

[21] N. V. Angrinskaya, J. Cryst. Growth **138**, 493 (1994).

[22] J. G. Massey and M. Lee, Phys. Rev. Lett. **75**, 4266 (1995).

[23] I. Terry, T. Penney, and S. von Molnár, Phys. Rev. Lett. **69**, 1800 (1992).

[24] P. Dai, Y. Zhang, and M. P. Sarachik, Phys. Rev. Lett. **69**, 1804 (1992).

[25] J. Kim and H. J. Lee, Phys. Rev. Lett. **70**, 2798 (1993).

[26] D. J. Chadi and K. J. Chang, Phys. Rev. Lett. **61**, 873 (1988); Phys. Rev. B **39**, 10 063 (1989).

[27] P. Mooney, J. Appl. Phys. **67**, R1 (1990).

[28] The Hall concentration n, obtained in the metallic regime, was fitted to the formula $n = (N_{\mathrm{Si}} - n_0)(1 - e^{-aD_n}) + n_0$, where D_n is the total light dose used to achieve an electron concentration n. The obtained parameters, $N_{\mathrm{Si}} = 68 \times 10^{16}$ cm^{-3} and $n_0 = 0.6 \times 10^{16}$ cm^{-3} were used to estimate n, in the insulator regime, from the D_n values. This is justified by supposing that the DX center cross section for the photons does not change significantly with the electron concentration.