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Anomalous Hall-effect results in low-temperature molecular-beam-epitaxial GaAs: Hopping in a dense *EL2*-like band

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Molecular-beam-epitaxial GaAs grown at very low temperatures ($\sim 200^\circ\text{C}$) exhibits anomalous Hall-effect properties. Here we show conclusively that the room-temperature conduction is due to activated (nearest-neighbor) hopping in a deep defect band of concentration $3 \times 10^{19} \text{ cm}^{-3}$, and energy $E_c - 0.75 \text{ eV}$, along with conduction due to free carriers thermally excited from this band. At low measurement temperatures, variable-range hopping [$\sigma \propto \exp(-T_0/T)^{1/4}$] prevails. The conduction-band mobility can be well explained by neutral-deep-donor scattering in parallel with lattice scattering.

Molecular-beam-epitaxial (MBE) GaAs is typically grown at substrate temperatures of about 600°C . However, recently it has been shown that material grown at 200°C , when used as a "buffer" between the semi-insulating (SI) GaAs substrate and the active layer, greatly minimizes several common device problems, such as backgating, sidegating, and light sensitivity.¹ Because of the new-found technological importance of the material, it has been studied in some detail, and several facts have emerged: (1) surface morphology and crystallinity are excellent;^{1,2} (2) stoichiometry is heavily shifted toward the As-rich side, deviating from equal Ga and As proportions by as much as 1%;² (3) a large quantity of As_{Ga} exists,² perhaps accommodating most of the nonstoichiometry; and (4) the material is optically and electrically "dead," i.e., it exhibits very weak photoluminescence (PL) and, under certain conditions, very high resistivity.¹ The weak PL is obviously due to a high concentration of nonradiative-recombination centers, but the high resistivity, and, indeed, the general conduction mechanism, has eluded explanation. In this paper, we show conclusively that the anomalous Hall properties result from two-band mixing due to bound carriers hopping between deep, *EL2*-like defect centers along with free carriers excited to the conduction band from these deep centers. A remarkable feature is that both the wave-function size and the electron-scattering power of the deep, neutral donors may be simply scaled from the expressions normally used for shallow levels, in order to produce a good data fit. Infrared-absorption spectra verify the calculated defect concentrations as a function of annealing temperature.

The MBE layers were grown in a Varian 360 system under normal, As-stabilized conditions, at a growth rate of $0.8 \mu\text{m/h}$. To minimize the possible electrical shunting effects of the SI GaAs substrate, some of the layers

were grown as thick as $20 \mu\text{m}$; however, it turned out that the substrate conduction was not important in these experiments, as was shown explicitly in some cases by differential Hall measurements. Square $6\text{-mm} \times 6\text{-mm}$ samples were cut for Hall-van der Pauw measurements, and In contacts were soldered onto the corners. No contact alloying was necessary to produce Ohmic conduction. Before contacting, some of the samples were annealed, under a GaAs proximity wafer, at temperatures from 250 to 600°C , in a flowing inert gas. Also, larger samples were cut and annealed for infrared-absorption measurements, at 10 K , in a Cary model 2300 spectrophotometer.

The resistivity and Hall-effect results are illustrated in Fig. 1 for five of the annealing temperatures. As is seen, the unannealed material is quite conductive ($\rho \sim 14 \Omega \text{ cm}$ at 300 K) and not very temperature dependent, whereas the *measured* carrier concentration ($n \equiv 1/eR$) is anomalous in that it is strongly temperature dependent and *decreasing* with increasing temperature. Note that the Hall coefficient for the 200°C material was too small to be measured below 350 K , yielding an upper limit for the mobility of $\mu \equiv R/\rho < 1 \text{ cm}^2/\text{Vs}$. This situation strongly suggests hopping conduction;³ however, the observation of a rapidly increasing mobility between 350 and 400 K indicates a conduction-band component, also. Thus, we invoke a two-band formalism⁴ to explain the results (band 1, conduction band; band 2, defect band):

$$\rho^{-1} = \sigma = \sigma_1 + \sigma_2 = en_1\mu_1 + \sigma_2, \quad (1)$$

$$n = \frac{1}{eR} = \frac{1}{e} \frac{(\sigma_1 + \sigma_2)^2}{R_1\sigma_1^2 + R_2\sigma_2^2} = \frac{1}{e} \frac{(\sigma_1 + \sigma_2)^2}{R_1\sigma_1^2}, \quad (2)$$

where $R_1 = 1/en_1$ and it is assumed that $R_2 = 0$, since R_2 is too small to measure in regions where hopping conduction (σ_2) dominates. In fact, although a Hall effect for

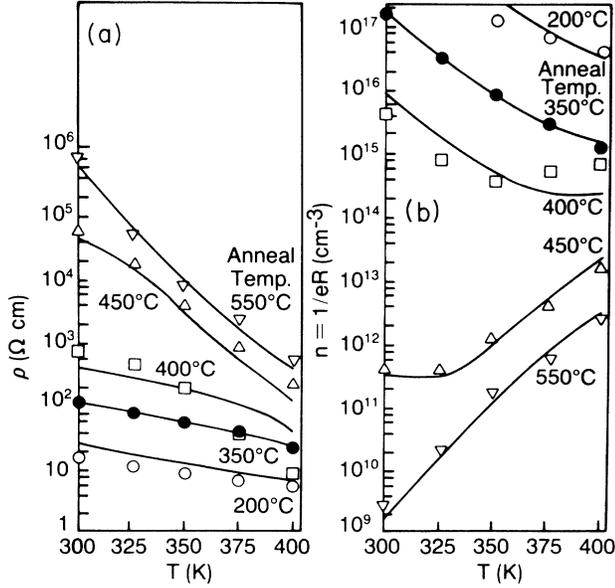


FIG. 1. The resistivity ρ and apparent carrier concentration n as a function of measurement temperature, for various annealing temperatures. The solid lines are theoretical fits with N_D as the only fitting parameter.

hopping conduction is theoretically possible,⁵ it has evidently never been observed in a semiconductor.⁶

For thermal excitation from a deep donor, at temperatures well below the exhaustion region, the conduction-band (CB) electron concentration n_1 can be written⁴

$$n_1 = \left[\frac{N_D}{N_A} - 1 \right] N'_c T^{3/2} \frac{g_0}{g_1} e^{\alpha/k} e^{-E_{D0}/kT} \\ = \left[\frac{N_D}{N_A} - 1 \right] C_1 T^{3/2} e^{-E_{D0}/kT}, \quad (3)$$

where $N'_c T^{3/2}$ is the effective CB density of states, g_0/g_1 is the degeneracy factor, and α is a linear temperature coefficient given by $E_D = E_{D0} - \alpha T$. In preliminary fits to the data represented in Fig. 1, it became clear that a deep donor near $E_c - 0.75$ eV was required to produce a good fit. This is not surprising, since it is already known that the arsenic antisite As_{Ga} is abundant in low- T MBE GaAs,² and As_{Ga} is a major component (if not the only component) of $EL2$, which has $E_{D0} = 0.75$ eV. From $EL2$ capture and emission data, Blakemore⁷ has given a result which can be shown to be equivalent to letting $E_{D0} = 0.748$ eV and $C_1 = 1.85 \times 10^{15} \text{ cm}^{-3} \text{ K}^{-3/2}$. Note that if $g_0/g_1 = \frac{1}{2}$ for $EL2$, then this value of C_1 requires $\alpha \approx 3.3 \times 10^{-4}$ eV/K, which then leads to E_D (296 K) = 0.65 eV, exactly the 296-K energy arrived at in the experimental studies of Duncan *et al.*⁸ Thus, we feel confident in using Blakemore's values of C_1 and E_{D0} and eliminating them as fitting parameters in our model.

We next model the conduction-band mobility μ_1 . Again, from preliminary fits, it becomes clear that μ_1 is quite low in the as-grown material, a few hundred $\text{cm}^2/\text{V s}$, and seems to vary inversely with the concentra-

tion of N_D centers, most of which are neutral (since, as will be shown, n_1 and N_A are small). In fact, mobilities calculated from the well-known Erginsoy formula, scaled from the usual shallow-donor value of 0.006 eV to our deep-donor value of 0.75 eV, are surprisingly accurate ($\mu_n \propto E_D$). McGill and Baron⁹ have noted that such scaling seems to be justified over a certain range, but nobody before, to our knowledge, has ever observed neutral scattering from a center as deep as 0.75 eV. Thus, combining neutral-center scattering and lattice scattering by Matthiessen's rule,⁴ we have

$$\mu_1^{-1} = \left[\frac{N_D}{K_1} + \frac{1}{8500[(300 \text{ K})/T]^{3/2}} \right]^{-1}, \quad (4)$$

where the second term in the large parentheses (the lattice-scattering term) yields scattering rates which are sufficiently accurate at the measurement temperatures of our experiments. Again, since $\mu_n \propto E_D$, a simple scaling of Erginsoy's formula to $E_D = 0.75$ eV gives $K_1 = 1.02 \times 10^{22} \text{ cm}^{-1}/\text{V s}$. Note that we need not insist that Erginsoy's theory is precisely correct for such deep centers, but only that $\mu_{\text{defect}} \propto N_D^{-1}$, with K_1 being the appropriate proportionality constant; our data cannot be fitted with K_1 much different than the above value.

Finally, we model the hopping conduction by a theory applicable when $N_A \ll N_D$, in which case the "carriers" are the "holes" (of concentration $N_A + n_1$) in the deep-donor band.^{3,10} An excitation energy ϵ_3 is needed to transfer a hole from its lowest-energy state (on a donor near an ionized acceptor) to a nearby neutral donor, after which the hole can hop through the lattice between donors, which are mostly isolated and close to the same energy. However, this nearest-neighbor hopping probability decreases rapidly with decreasing N_D , because of the necessity for wave-function overlap. In this model, the conductivity σ_2 is given by^{3,10}

$$\sigma_2 = C_2 e^{-\gamma R/a} e^{-\epsilon_3/kT} = C_2 e^{-\gamma/a N_D^{1/3}} e^{-\epsilon_3/kT}, \quad (5)$$

where C_2 and γ are constants, R is the average donor separation ($4\pi R^3/3 = 1/N_D$), a is the extent of the donor wave function [$a = \hbar/(2m^* E_{D0})^{1/2}$ in a hydrogenic-like model], and ϵ_3 is the difference between the isolated-donor energy and the Fermi energy ($\epsilon_3 = K_3 e^2 N_D^{1/3}/4\pi\epsilon$, where ϵ is the dielectric constant and K_3 is a constant of order unity). From the value $E_{D0} = 0.75$ eV we calculate $a = 8.67$ Å, and also Shklovskii¹⁰ has suggested that $\gamma \approx 1.8$. The constant C_2 depends, of course, on the details of the electron-phonon interaction which promotes the hopping, and will depend to some extent on N_D and N_A (probably $C_2 \propto N_A^{1/2}$),³ but not nearly as strongly as the term involving γ . Also, ϵ_3 should depend on N_D , as indicated above, but the temperature-dependent Hall-effect data, from 300 to 400 K, can be satisfactorily fitted by assuming a constant ϵ_3 . Thus, in this study, ϵ_3 is simply a fitting parameter, although it does come out to be of the expected order of magnitude.

Thus, to minimize the number of fitting parameters, we have taken E_{D0} , C_1 , K_1 , and γ from the literature. We have then fitted N_D , N_A , C_2 , and ϵ_3 to the ρ and n data

for the sample annealed at 350 °C, a temperature at which both defect-band and conduction-band effects are appreciable. The fits, shown in Fig. 1, are excellent, with the following fitting parameters: $N_D = 1.66 \times 10^{19} \text{ cm}^{-3}$, $N_A = 7.0 \times 10^{14} \text{ cm}^{-3}$, $C_2 = 3.7 \times 10^3 \text{ } \Omega^{-1} \text{ cm}^{-1}$, and $\epsilon_3 = 0.123 \text{ eV}$. This value of N_A is very close to what is routinely produced (probably due to carbon) in our MBE system. From the formula $\epsilon_3 = K_3 e^2 N_D^{1/3} / 4\pi\epsilon$, a value of $K_3 = 3.5$ is obtained, well above Mott's suggested value³ of 0.73 and Shklovskii's suggested value¹⁰ of 0.99. However, this calculation assumes that the *bulk* value of the relative dielectric constant holds, i.e., $\epsilon_{\text{rel}} = 12.91$, which is doubtful in this case because of the close spacings between the acceptors and their nearest donors. If we instead assume that the value $K_3 = 0.99$ is correct, then the experimental value of ϵ_3 is fitted with $\epsilon_{\text{rel}} = 3.6$, which is a change in the expected direction. However, this question will have to be studied further.

Finally, all the rest of the data at different annealing temperatures, from 200 to 600 °C, were fitted by fixing E_{D0} , C_1 , K_1 , γ , N_A , C_2 , and ϵ_3 , and varying only N_D . As can be seen in Fig. 1, all of the fits (including those not shown) are very good except for the fit of the 400 °C data. The mediocre fit of the 400 °C data is not surprising, because the conduction mechanism is rapidly changing from predominantly hopping conduction to free-carrier conduction at an annealing temperature of 400 °C, and the sample is probably somewhat inhomogeneous. However, the goodness of the ρ and n fits for all of the other data, over 8 orders of magnitude for n , leaves no doubt as to the basic correctness of the conduction model.

The fitted values of N_D are plotted as a function of annealing temperature in Fig. 2. As an independent check of N_D , infrared-absorption studies were also carried out. The absorption spectra, to be discussed in detail elsewhere, were very much like those due to *EL2*, so that approximate concentrations of the deep center could be estimated from the known photon-absorption cross section of *EL2*.¹¹ These data are also plotted in Fig. 2, and the agreement is remarkable, considering that the experiments are entirely different and involve no common parameters.

The conduction model can be summarized as follows: MBE GaAs grown at 200 °C, under the stated conditions, contains about 3×10^{19} defects per cm^3 of energy $E_c - 0.75 \text{ eV}$ (extrapolated to $T = 0 \text{ K}$). Conduction takes place by nearest-neighbor hopping at measurement temperatures above 300 K; at lower temperatures, however, variable-range hopping¹⁰ becomes important, as shown by the $\exp[-(T_0/T)^{1/4}]$ dependence below 175 K in Fig. 3. Upon annealing at 400–500 °C, the relevant defect concentration decreases to about $3 \times 10^{18} \text{ cm}^{-3}$, destroying the hopping conduction, mainly because of the strong decrease in the wave-function overlap factor, $\exp(-\gamma/aN_D^{1/3})$, in Eq. (5); the conduction-band contribution is then dominant and a Hall effect is easily measured at all temperatures. Note that even though the 300-K resistivity is very high, $\sim 8 \times 10^5 \text{ } \Omega \text{ cm}$, for the 550 °C-annealed material, still it is about 2 orders of magnitude lower than that in typical bulk, semi-insulating

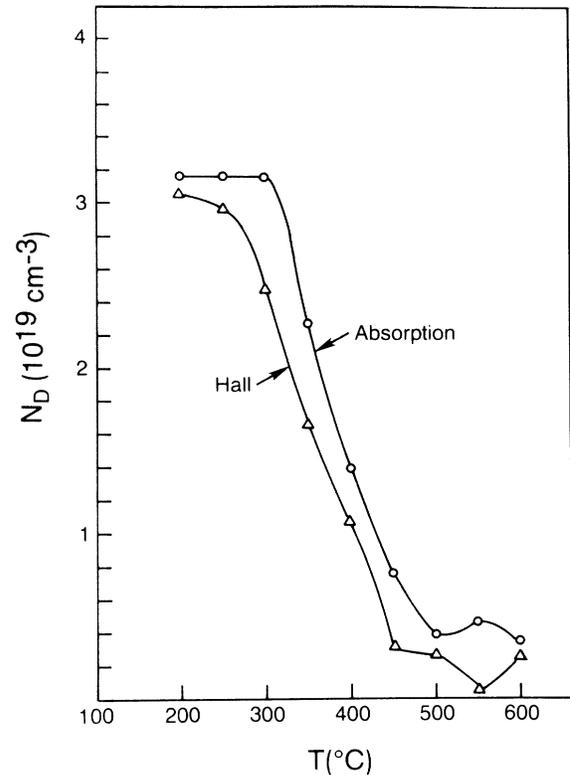


FIG. 2. The deep-donor concentration N_D , calculated separately from Hall-effect and infrared-absorption data, as a function of annealing temperature.

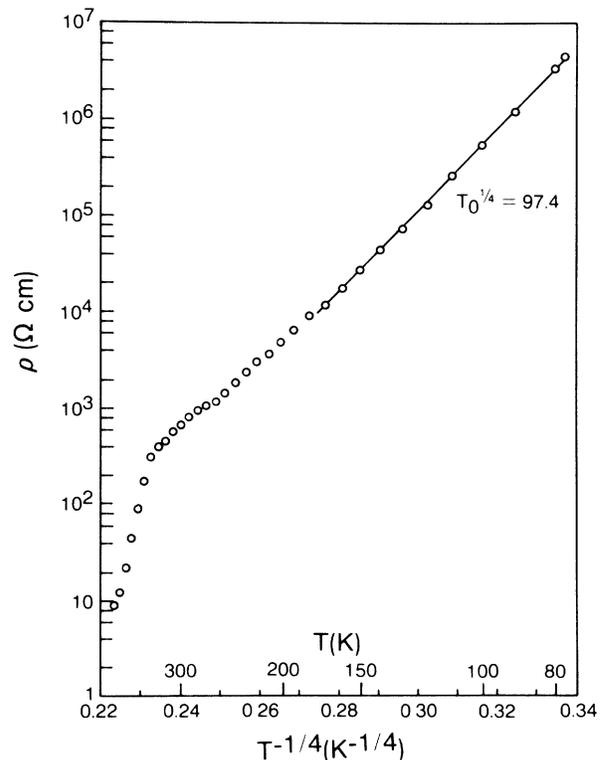


FIG. 3. The resistivity as a function of $T^{-1/4}$ for the sample annealed at 400 °C.

GaAs, because the latter has a much lower deep-center concentration, typically $[EL2] = 1 \times 10^{16} \text{ cm}^{-3}$.

To identify the dense defect responsible for the electrical properties, we note that an Auger analysis shows our material to be about 2% As rich, which should produce over 10^{20} cm^{-3} As_{Ga} , As_i , and/or V_{Ga} defects. High concentrations of V_{Ga} are doubtful, because it would be an acceptor, and we have measured an N_A of only about $1 \times 10^{15} \text{ cm}^{-3}$, i.e., the expected carbon concentration. On the other hand, high concentrations ($\sim 5 \times 10^{18} \text{ cm}^{-3}$) of As_{Ga} have already been identified in similar as-grown material,² and also in neutron-irradiated material.^{12,13} In the latter, a variable-range-hopping coefficient $T_0^{1/4} \simeq 94$ has been measured,¹² which is close to our value of 97. One might question whether our defects are not simply *EL2*, since the parameters in Eq. (3), which fitted the data well, were based on known *EL2* parameters. However, in conjunction with the transmission measurements, we have also carried out photon-quenching experiments, considered to be the definitive test of *EL2*, and found that only about 10% of the defects

are quenchable. Thus, the bulk of our electrically active defects are associated with As_{Ga} , but not the variation or complex of As_{Ga} which constitutes *EL2*. Note that the thermal excitation properties of As_{Ga} and *EL2* are expected to be quite similar, consistent with the results of our study. The rest of the 10^{20} – 10^{21} - cm^{-3} As-related defects are probably electrically inactive As clusters or precipitates, which are often seen in annealed GaAs materials.

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