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Defect production in electron-irradiated, *n*-type GaAs

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Temperature-dependent Hall-effect measurements have been performed on pure, *n*-type, vapor-phase epitaxial GaAs, irradiated by 1-MeV electrons at room temperature. The energies and production rates of two dominant defect centers, C_2 and C_3 , are as follows:

$E_2 = E_C - 0.148$, $E_3 = E_C - 0.295 \pm 0.002$ eV, $\tau_2 = 2.0$, and $\tau_3 = 0.5 \pm 0.2$ cm⁻¹, in good agreement with deep level transient spectroscopy (DLTS) data. However, the most important result of this study is a very high production rate, $\tau_{AS} \approx 4 \pm 1$ cm⁻¹, for "shallow" acceptors (C_{AS}) lying below E_3 . In fact, C_{AS} is produced at a much higher rate than *all* of the DLTS traps observed in this energy range, proving that close to half of the primary defects in electron-irradiated GaAs are evidently not seen by DLTS. The high C_{AS} production rate has important implications for microscopic models of C_1 and C_2 , rendering unnecessary the assumption that one of these centers must be an acceptor in order to explain the Hall-effect results. Finally, we show that all available Hall-effect and DLTS data are consistent with C_{AS} representing Ga-sublattice damage, which has not been observed before.

INTRODUCTION

Electron-irradiation experiments in GaAs have been carried out extensively since the early 1960's. The results prior to 1977 have been reviewed by Lang,¹ and those up to 1985 by Pons and Bourgoin.² In spite of the results available from a wide variety of experimental techniques, no defect produced in quantity by 1-MeV irradiation has been conclusively identified, although the involvement of the As vacancy (V_{As}) is fairly certain.^{2,3} The primary characterization tool used for electron-irradiation investigations in the last decade is deep-level transient spectroscopy (DLTS),⁴ which allows many deep-level centers to be seen in the same sample. One disadvantage of DLTS, however, is that it requires either a Schottky barrier or a *p-n* junction diode in order to have a region in which the majority carrier concentration is adjustable by applying a voltage pulse. Thus, the main usefulness of DLTS lies in the study of majority-carrier traps, i.e., electron traps in *n*-type material and hole traps in *p*-type material. Although minority carriers can sometimes be injected, by voltage or light, still only qualitative information may be obtained on minority-carrier traps in most cases.⁵ In GaAs, it is easy to make a good Schottky diode on *n*-type material, and therefore, most of the DLTS work has been carried out on electron traps in *n*-type GaAs. At present, there are two major dilemmas concerning the DLTS data. The first is that a reconciliation of Hall-effect and DLTS results seems to demand that at least one of the two major electron traps, E_1 and E_2 (C_1 and C_2 in our notation) must be an acceptor.^{2,6} But C_1 and C_2 have also been identified with the As vacancy,² which intuitively should be a double donor. The second dilemma is that all of the electron and hole traps observed by DLTS have been attributed, with good evidence, to As sublattice damage.² If so, then the Ga sublattice damage, which certainly is created in roughly the

same amount, is missing. We show that the results of this study can resolve both dilemmas.

EXPERIMENT AND ANALYSIS

Most of the data presented here were obtained from one sample, a pure, vapor-phase epitaxial layer, 97 μ m thick, with a 77 K mobility of 157 000 cm²/V s, and ionized donor and acceptor concentrations of 2×10^{14} and 4×10^{13} cm⁻³, respectively. The thickness was low enough that the 1-MeV electron damage was quite uniform throughout the depth, but high enough that depletion effects were negligible. Other samples showed similar behavior, but were not investigated as extensively. Irradiations were performed at room temperature with a 1-MeV, 1- μ A/cm² electron beam from a van de Graaff accelerator. The sample was first irradiated in steps of 1×10^{14} e/cm² to a total of 2×10^{14} e/cm², then annealed at 450 °C for 3 min, and finally again irradiated up to 2.6×10^{14} e/cm², in 0.2×10^{14} e/cm² steps, and ultimately to about 10×10^{14} e/cm² in larger steps. The first anneal, as expected,^{1,2} returned the electrical properties to their preirradiation conditions. Hall-effect measurements were carried out in a magnetic field of 4.5 kG, from 77 to 420 K, except where sample resistances limited the lower end.

The carrier concentration data were obtained from the Hall-coefficient R at each temperature by using the relationship $n = r/eR$, where r is the Hall factor. To obtain high accuracy, the r 's were determined from the mobility data by iteratively solving the Boltzmann transport equation.⁷ The resulting n vs T data were then fitted to the usual "change-balance" equation derived from statistical principles.⁸ A generalized version of this equation can be obtained from Eq. B59 of Ref. 8:

$$n = p + \sum_{\substack{k,l,m \\ (\text{all } A,D)}} (l_k - l) n_{klm} - \sum_k l_k N_k, \quad (1)$$

where

$$n_{klm} = N_k \left/ \left(1 + \sum_{l',m' \neq l,m} \frac{g_{kl'm'}}{g_{klm}} \right) \right. \times e^{[\epsilon_{klm} - \epsilon_{kl'm'} - (l-l')\epsilon_F]/kT} \quad (2)$$

The energies here are measured with respect to the valence band, and all other symbols are defined in Ref. 8. For pure donors or acceptors, l_k denotes the number of ionizable electrons or holes, respectively, and the index l runs from 0 to l_k . (For amphoteric centers, not considered here, l_k denotes the number of donor states only, and l runs from 0 to the combined number of donor and acceptor states.) It is easy to show that the values of the major fitted parameters, the N 's, g 's, and ϵ 's of centers which have a varying occupation (charge state) within the temperature range of interest, do not depend on the last term of Eq. (1), since it is temperature-independent. But all the other terms in Eq. (1) are independent of the donor/acceptor (D/A) nature of a particular center, and thus an important conclusion is reached: the fitted values of N_k , ϵ_k , and g_k are the same, whether center k is assumed to be a donor or an acceptor. Thus, only one least-squares fit of the data need be carried out. However, an unfortunate corollary to this analysis is that there is no way, from an n vs T fit alone, to tell whether a particular center is a donor or an acceptor.⁹

The fitting procedure was as follows. For fluences ϕ from 1.0 to 2.4×10^{14} e/cm², the centers C_2 and C_3 dominated the temperature-dependent Hall-effect (TDH) data, and an assumption of single-charge states (i.e., $l_k = 1$ and $l = 0, 1$) gave good fits. Under these conditions, Eq. (1) can be written

$$N_C e^{-E_F/kT} = \sum_{i=2,3} \frac{N_i}{1 + (g_1/g_0)_i e^{-\alpha_i/k} e^{(E_0 - E_F)/kT}} + K, \quad (3)$$

where N_C is the effective conduction-band density of states (nondegenerate statistics apply here), E_i and E_F are the defect and Fermi energies, respectively (now with respect to the conduction band), α_i is defined by $E_i = E_0 - \alpha_i T$, g_1 and g_0 are the occupied and unoccupied state degeneracies, respectively, and K is a temperature-independent term. For example, if C_1 , C_2 , and C_3 are all donors, then $K = N_{DS} + N_1 - N_{AS}$; or if C_1 and C_3 are donors, and C_2 an acceptor, then $K = N_{DS} + N_1 - N_{AS} - N_2$. Other cases are listed in Table I. The fitting parameters, besides K , are N_2 , N_3 , E_2 , E_3 , g_2 , and g_3 , where

$$g_i \equiv (g_1/g_0)_i \exp(-\alpha_i/k).$$

Then, if $N_1 = N_2$, as is known from DLTS data, the value of N_{AS} can be calculated from K , N_2 , and N_3 , according to Table I.

For the low fluence data, $\phi = 0, 2$, and 4×10^{13} e/cm², the empirical method of Wolfe, Stillman, and Lindley¹⁰ is applicable to the 77 K mobility and carrier concentration data to determine N_D^+ and N_A^- , where $N_D^+ = N_{DS} + N_1^+ + N_2^+ + N_3^+$, and $N_A^- = N_{AS} + N_1^-$

TABLE I. Calculation of N_{AS} from Eq. (3) for various fluences ϕ (10^{14} e/cm²), and various donor/acceptor combinations of C_1 , C_2 , and C_3 .

C_1	C_2	C_3	N_{AS} (for $\phi = 1.2, 1.4$) ^{a,b}	N_{AS} (for $\phi = 2.4$) ^{a,c}
D	D	D	$N_{DS} + N_1 - K$	$N_{DS} + N_1 + N_2 - K$
A	D	D	$N_{DS} - K$	$N_{DS} + N_2 - K$
D	D	A	$N_{DS} + N_1 - N_3 - K$	$N_{DS} + N_1 + N_2 - K$
A	D	A	$N_{DS} - N_3 - K$	$N_{DS} + N_2 - K$
D	A	D	$N_{DS} + N_1 - N_2 - K$	$N_{DS} + N_1 - K$
A	A	D	$N_{DS} - N_2 - K$	$N_{DS} - K$
D	A	A	$N_{DS} + N_1 - N_2 - N_3 - K$	$N_{DS} + N_1 - K$
A	A	A	$N_{DS} - N_2 - N_3 - K$	$N_{DS} - K$

^a K is a fitting parameter (negative for all ϕ); N_{DS} is determined from τ_{DS} , measured at lower fluences; N_1 is assumed equal to N_2 .

^b N_2, N_3 are fitting parameters.

^c N_3 is a fitting parameter; N_2 is determined from τ_2 measured at lower fluences.

+ $N_2^- + N_3^-$. Further information can be obtained from the difference, $n(296) - n(77)$. To calculate the parameters N_{DS} , N_{AS} , N_1 , N_2 , and N_3 , from N_D^+ and N_A^- , we must first assume the respective D/A natures of C_1 , C_2 , and C_3 , and then use the energies and degeneracies determined from the higher-fluence data described previously. Actually, C_3 has little influence on the results because of the depth of the level and also its low production rate. Furthermore, it is known² that $N_1 = N_2$, and that $E_1 \approx 0.045$ eV, and we assume that $g_1 \approx 0.5$, since the midfluence fits also give $g_2 = g_3 \approx 0.5$. In this way, the parameters N_{DS} , N_{AS} , N_1 , and N_2 (but not N_3) can be determined at low fluences for all possible D/A cases.

RESULTS

The general behavior of the room-temperature resistivity ρ and mobility μ as a function of fluence are given in Fig. 1. The resistivity approaches that of a semi-insulating sample at fluences above about 4×10^{14} cm⁻². At the same time, the mobility begins to fall precipitously to values which are much too low to represent ionized-impurity scattering, and are undoubtedly due to inhomogeneity. This assertion is supported by previous results, which show that a sample can actually change type,¹¹ usually in the range of $n_0/\phi \approx 0.4$ cm⁻¹. Since our $n_0 \approx 1.5 \times 10^{14}$ cm⁻³, the effect should be taking place near $\phi \approx 4 \times 10^{14}$ cm⁻², which is consistent with the data. Until a sample is fully transformed to p type, the current conduction will be quite nonuniform, and the electrical results will be nonohmic and difficult to analyze.

In Fig. 2, we show how the low-temperature Fermi level varies with ϕ . It is seen that the shallow levels, C_{DS} and C_1 , dominate at low fluences, then C_2 at moderate fluences, next C_3 , and finally deeper levels of unknown origin at the highest fluences. Reliable n vs T fits could be obtained only when one level or another was clearly dominant, because of the inhomogeneity problem, as discussed above. The best fits in the C_2 regime were obtained for $\phi = 1.0, 1.2$, and 1.4×10^{14} e/cm², and these fits gave consistent values for E_2 , E_3 , g_2 , and g_3 . The same values of E_3 and g_3 were obtained from the fit at $\phi = 2.4 \times 10^{14}$ e/cm² (C_2 was too shallow to be seen). The rest of the data could not be well fitted with these parameters, as would have been predicted from Fig. 2. To illustrate,

FIG. 1. Resistivity (ρ) and mobility (μ) as a function of fluence.

we show the fitted n vs T curves for $\phi = 0.8, 1.0, 1.2, 1.4$, and 2.4×10^{14} e/cm² in Fig. 3. The 0.8 curve is not well fitted, probably because parts of the sample are dominated by C_{DS} , and other parts by C_2 .

The final N vs ϕ results are shown in Fig. 4. The notation is as follows: "DAD" means that C_1 and C_3 are donors, and C_2 is an acceptor; "AXX" means that C_1 is an acceptor, and the results are independent (or nearly so) of C_2 and C_3 ; etc. It is seen that N_2 vs ϕ is quite linear, with a slope (production rate) of $\tau_2 = 2.0 \pm 0.2$ cm⁻¹. The production rate of C_3 is

FIG. 2. Low-temperature Fermi level as a function of fluence. The energy levels for C_{DS} , C_2 , and C_3 are also shown.

FIG. 3. Carrier concentration as a function of temperature for various fluences. The solid lines are theoretical fits with the following common parameters: $E_2 = 0.148$, $E_3 = 0.295$ eV, and $g_2 = g_3 = 0.5$. The fit at $\phi = 0.8 \times 10^{14}$ is very poor due to inhomogeneity.

FIG. 4. Concentrations of N_{DS} , N_2 , N_3 , and N_{AS} as a function of fluence. The solid points were from an earlier irradiation. The three characters in quotation marks designate the assumed donor (D), acceptor (A), or either (X), nature of C_1 , C_2 , and C_3 , respectively.

TABLE II. Defect production rates^a in *n*-type GaAs for various donor/acceptor combinations of C_1 , C_2 , and C_3 .

C_1	C_2	C_3	τ_{DS}	τ_2^b	τ_3^c	τ_{AS}
<i>D</i>	<i>D</i>	<i>D</i>	0.2	2.0	0.5	5.4
<i>A</i>	<i>D</i>	<i>D</i>	1.3	2.0	0.5	4.5
<i>D</i>	<i>D</i>	<i>A</i>	0.2	2.0	0.5	4.5
<i>A</i>	<i>D</i>	<i>A</i>	1.3	2.0	0.5	3.6
<i>D</i>	<i>A</i>	<i>D</i>	0.2	2.0	0.5	3.5
<i>A</i>	<i>A</i>	<i>D</i>	1.3	2.0	0.5	2.6
<i>D</i>	<i>A</i>	<i>A</i>	0.2	2.0	0.5	2.7
<i>A</i>	<i>A</i>	<i>A</i>	1.3	2.0	0.5	2.1–2.7 ^d

^aUnits of cm^{-1} ; typical errors: $\pm 15\%$ or $\pm 0.2 \text{ cm}^{-1}$, whichever is greater.

^bRate for N_1 is assumed equal to that for N_2 .

^cRate at low fluence is unknown.

^dPlot is very nonlinear.

more uncertain, because of the wide scatter near $\phi \approx 1 \times 10^{14} \text{ e/cm}^2$, and also because low-fluence information could not be obtained. The possible values of τ_3 range from 0.4 to 0.7 cm^{-1} . The shallow (hydrogenic) donor concentration N_{DS} depends strongly on whether C_1 is assumed to have a donor or acceptor nature. Other evidence² suggests that τ_{DS} cannot be nearly as high as that calculated for the *AXX* case, and, if true, then C_1 must be a donor. Finally, the N_{AS} (i.e., acceptors below E_3) values are calculated as explained earlier, and three representative plots are shown in Fig. 4. Of the eight possibilities listed in Table I, the *AAA* case gives a decidedly nonlinear plot, and thus *AAA* is probably incorrect. The production rates for various cases are summarized in Table II.

DISCUSSION

The final TDH results for the energies and production rates are compared with DLTS results in Table III. The agreement for C_2 and C_3 is very good,² especially considering that other DLTS references^{1,3} give values for τ_2 of about 1.8 cm^{-1} , closer to our value of 2.0 cm^{-1} . The Hall-effect value for τ_{AS} , $4 \pm 1 \text{ cm}^{-1}$, covers all the reasonable possibilities

TABLE III. Energies and production rates of primary defects produced by 1-MeV electrons in GaAs.

Defect		Energy (eV)		Production rate (cm^{-1})	
Hall ^a	DLTS ^b	Hall ^a	DLTS ^b	Hall ^a	DLTS ^b
...	E_1^c	...	$E_C - 0.045$	...	1.5
C_2^c	E_2^c	$E_C - 0.148$	$E_C - 0.14$	2.0 ± 0.2	1.5
C_3^c	E_3^c	$E_C - 0.295$	$E_C - 0.30$	0.5 ± 0.2	0.4
...	E_4^c	...	$E_C - 0.76$	...	0.1
...	E_5^c	...	$E_C - 0.96$	...	0.1
...	H_0^d	...	$E_V + 0.06$	...	0.8
...	H_1^d	...	$E_V + 0.29$	...	0.1
C_{AS}^c	...	below $E_C - 0.3$	...	4 ± 1	...

^aThis work.

^bFrom Ref. 2.

^cMeasured in *n*-type GaAs.

^dMeasured in *p*-type GaAs.

listed in Table II, as discussed below.

First of all, an acceptor assignment for C_1 requires a high shallow-donor (i.e., $E_D \lesssim 0.01 \text{ eV}$) production rate, which has been judged improbable from other evidence.² Furthermore, C_2 is also most likely a donor, since the electron capture cross section is large, $1 \times 10^{-13} \text{ cm}^2$. Since both of these centers have been associated with the As vacancy,² and since $N_1 \approx N_2$ from DLTS results, it is quite possible that N_1 and N_2 are two charge states of V_{As} or a V_{As} -defect complex (no impurity involved). For example, E_1 could be the $(0/+)$ transition, and E_2 the $(+/++)$ transition. If, indeed, C_1 and C_2 are both donors, then from Table II, $\tau_{AS} \approx 5.0 \pm 0.5 \text{ cm}^{-1}$. However, we choose to quote a more conservative range, $4 \pm 1 \text{ cm}^{-1}$, which includes all cases in Table II (except *AAA*) to integer accuracy. It is virtually certain that these C_{AS} defects are not impurity related because they can obviously be created in numbers ($\geq 10^{15} \text{ cm}^{-3}$) far larger than the initial shallow donor and total acceptor concentrations, which account for most of the impurities incorporated in vapor-phase epitaxial (VPE) GaAs.

To explain the origin of the C_{AS} , it is natural to first consider the DLTS hole traps, H_0 and H_1 , which may well be acceptors. However, a glance at Table III suggests that the combined production rates of H_0 and H_1 are far too low to account for τ_{AS} . On the other hand, the H_0 and H_1 production rates listed in Table III are for *p*-type material, and it is entirely possible that these defects are more stable in *n*-type material. One problem with this model, however, is that H_0 and H_1 are associated with As-sublattice damage, and if they were being produced at $\sim 5 \text{ cm}^{-1}$ (the most probable value for τ_{AS} , since C_1 and C_2 are likely both donors), then the combined rates for C_1 , C_2 , C_3 , H_0 , and H_1 are $> 7 \text{ cm}^{-1}$, i.e., greater than the total rate expected for each sublattice.² This may not be a serious flaw in the H_0 , $H_1 \equiv C_{AS}$ model, but should be kept in mind.

A more intriguing model, which satisfies most, if not all, of the experimental and theoretical observations, is that C_{AS} is mainly Ga-sublattice damage (GSLD), which has not been identified up to now.² The GSLD would probably consist primarily of V_{Ga} , or the Frenkel pairs V_{Ga} -Ga_i, just as the As-sublattice damage (ASLD) is expected to be mostly V_{As} and/or V_{As} -As_i pairs. First, the ASLD is known, from DLTS data,² to have a total production rate of about 5 cm^{-1} . The GSLD would be expected to have nearly the same rate, and, indeed, the rate for C_{AS} is $4 \pm 1 \text{ cm}^{-1}$. Second, the GSLD defects, i.e., V_{Ga} and/or V_{Ga} -Ga_i, should have primarily acceptor nature,¹² and we have shown that the C_{AS} are acceptors. Third, the GSLD might be expected to be unstable in *p*-type GaAs, since V_{Ga} can then transform to the more stable configuration As_{Ga}- V_{As} by a simple nearest-neighbor hop,¹³ and V_{Ga} -Ga_i should have a high probability of recombination if the Ga_i is positively charged. In agreement, the C_{AS} must be unstable in *p*-type material, or else the Fermi level would not move up, toward midgap, in electron-irradiated *p*-type GaAs.^{1,11} Also, the nonobservation by DLTS of any high-production-rate ($\tau \sim 4 \text{ cm}^{-1}$) centers in *p*-type GaAs is consistent with this model. It would be interesting to make a more concerted DLTS effort to find and

quantify the C_{AS} centers in n -type GaAs by applying injection techniques. Furthermore, detailed isothermal annealing experiments should be performed to determine the thermal stability.

Finally, the τ_{AS} results have important implications for any theoretical attempts to identify C_1 and C_2 . A starting point for many of the current models is that either C_1 or C_2 must be an acceptor;^{2,6} otherwise, as seen in Table III, there are no other electron or hole traps with a high enough production rate to explain the well-known fact that electron irradiation depletes n -type GaAs of electrons. Since there is now good evidence to associate C_1 and C_2 with the As vacancy, it has even been claimed⁶ that these centers represent the two double-acceptor states of V_{As} , a model which seems to violate intuition, as well as the observed capture cross sections. We do not wish to debate the relative merits of this model or any other, but simply to point out that the high measured value of τ_{AS} makes it entirely unnecessary to assume that either C_1 or C_2 has an acceptor nature; i.e., the electron depletion in n -type GaAs can be basically explained by the C_{AS} acceptors alone.

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