9-1-1987

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David C. Look
Wright State University - Main Campus, david.look@wright.edu

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High acceptor production rate in electron-irradiated n-type GaAs: Impact on defect models

D. C. Look
University Research Center, Wright State University, Dayton, Ohio 45435

(Received 26 May 1987; accepted for publication 17 July 1987)

Defect production rates have been studied in electron-irradiated GaAs by temperature-dependent Hall-effect (TDH) measurements. The TDH results agree well with deep level transient spectroscopy (DLTS) results for the well-known electron traps $E_1$, $E_2$, and $E_3$, but conclusively demonstrate a much higher production rate ($4 \pm 1 \text{ cm}^{-1}$) of acceptors below $E_3$ than the total of all other DLTS traps. These findings strongly affect current defect models, and, e.g., are consistent with the existence of Ga sublattice damage, not seen before.

The effects of 1 MeV electron irradiation in GaAs have been studied since the early 1960s, and have been reviewed in 1977 and 1985. Although many characterization techniques have been employed during this time, most of the data during the last decade have been obtained by deep level transient spectroscopy (DLTS), largely because of its ability to observe different centers in the same sample. However, it is generally not possible with DLTS to accurately measure the concentration of both electron and hole traps in the same sample, and furthermore there is no way to know whether the traps are donors or acceptors. Temperature-dependent Hall-effect (TDH) measurements, on the other hand, give detailed results on only one or two centers in a given sample, but can accurately determine the concentration of compensating centers, e.g., acceptors in an n-type sample. In our study, we show that the three dominant radiation-induced defects in n-type GaAs, i.e., $C_{1s}$, $C_{2s}$, and $C_{2p}$ ($E_1$, $E_2$, and $E_3$ in the DLTS notation), are found at roughly equal energies and concentrations in both the DLTS and Hall-effect data, but that the total "shallow" acceptor concentration $N_{AS}$ (below $E_i$) is much higher than the total concentration of all traps observed by DLTS in this energy range. These results have an important impact on current irradiation-defect models.

The samples used here were grown by the vapor phase epitaxial technique in a (100) orientation, and were thin enough (97 $\mu$m) that the defect production was uniform, but thick enough that surface and interface depletion effects were negligible. The initial shallow donor concentration $N_{DS}$ was about $2 \times 10^{14}$ cm$^{-3}$ and the total acceptor concentration $N_A$ was about $4 \times 10^{15}$ cm$^{-3}$. The 1 MeV electron fluences ($\sim 1 \mu$A/cm$^2$) ranged from 0 to $2.4 \times 10^{14}$ e/cm$^2$, at which point the total defect concentration was $> 10^{15}$ cm$^{-2}$, i.e., much larger than the initial donor and acceptor concentrations. Free-electron concentrations were determined from the relationship $n = e/2R$, where $R$ is the measured Hall coefficient and $r$ is the Hall factor. To obtain maximum accuracy, $r$ was calculated by fitting the mobility with an iterative solution of the Boltzmann equation. For low fluences ($0.4 \times 10^{13}$ cm$^{-2}$), the empirical Wolfe-Stillman relationship could be used to determine $N_{DS}$ and $N_{AS}$ since the shallow donor still dominated at 77 K. For higher fluences, the full TDH curves had to be fitted according to a generalized "change-balance" equation, which can be derived from Eq. B59 of Ref. 4:

$$n = p + \sum_{k \in A} (l_k - l) n_{k,m} - \sum_{k \in A} l_k n_k,$$

where

$$n_{k,m} = N_k \left[ \frac{1 + \sum_{l \in A} \frac{g_k l_m}{g_k l_m}}{kT} \right] \exp \frac{\epsilon_{k,m} - \epsilon_{l,m} - (I - I') \epsilon_p}{kT}.$$

Here $l_k$ is the number of ionizable electrons or holes, respectively, for a pure donor center $k$ or a pure acceptor center $k$. Amphoteric centers can easily be included, but are not here. The index $l$ ranges from 0 to $l_k$ and other symbols are defined in Ref. 4. The utility of Eq. (1) is that all terms except the last are independent of the donor or acceptor nature of a particular center $k$, and the last term is temperature independent and thus does not affect the determination of the major fitting parameters $N_k, \epsilon_k$, and $g_k$. Therefore, all centers can initially be treated as donors (last term zero) and the temperature-independent term then adjusted for other cases. For fluences between 0.8 and $1.6 \times 10^{14}$ e/cm$^2$, our data can be fitted with two single-charge-state defects, $C_i$ and $C_j$, responsible for the temperature dependence. Then Eq. (1) becomes ($l_k = 1; l = 0, 1; m$ suppressed)

$$N_i e^{-\epsilon_i/kT} = \sum_{l \in A} \frac{N_i}{1 + [g_i/g_0] e^{-\epsilon_i/kT}} + K,$$

where $N_i$ is the effective conduction-band density of states (nondegenerate statistics apply); $g_i$ and $g_0$ are the unoccupied and occupied state degeneracies, respectively; and $\alpha_i$ is defined by $E_i = E_0 - \alpha_i T$, where all energies are measured with respect to the conduction band. The constant $K$ is determined from the donor/acceptor $(D/A)$ nature assumed for the defects $C_i, C_j$, and $C_k$. For example, if all three are assumed to be acceptors, then $K = N_{DS} - N_{AS} - N_2 - N_3$, and thus $N_{AS}$ can be determined, since $K, N_2$, and $N_3$ are fitting parameters, and $N_{DS}$ is known from its production rate calculated at lower fluences. The values of $N_{AS}$ for other possible D/A cases of $C_i, C_j$, and $C_k$ are given in Table I.

In performing the irradiations, the low-temperature Fermi level dropped rapidly at fluences of $\phi = 0.6, 1.8,$ and...
$2.8 \times 10^{14}$ e/cm$^2$, as the centers $C_2$, $C_3$, and then deeper centers, respectively, became dominant. For fluences near these transition points, the electrical properties were often inhomogeneous, as expected. Good fits could be obtained in the $C_2$ region at $\phi = 1.0, 1.2$, and $1.4 \times 10^{14}$ e/cm$^2$, and in the $C_3$ region at $\phi = 2.4 \times 10^{14}$ e/cm$^2$. As shown in Fig. 1, these four plots were well fitted by Eq. (3) with the following common parameters: $E_2 = 0.148$, $E_3 = 0.295 \pm 0.002$ eV, and $(g_2/g_3)\exp(-\alpha/k) = 0.5 \pm 0.2$, for both centers. The values of $E_2$ and $E_3$ are almost exactly the same as those given by DLTS. The fit at $\phi = 0.8 \times 10^{14}$ e/cm$^2$ is very poor, due to the inhomogeneity mentioned above. For the low fluences, $\phi = 0.2$, and $4 \times 10^{14}$ e/cm$^2$, the Wolfe-Stillman mobility analysis could be applied to the 77 K data, and further information could be obtained from the difference $n(796 \text{ K}) - n(77 \text{ K})$. With the $E_2$ and $E_3$ determined above, along with $E_1 = 0.045$ eV and $N_1 = N_3$, known from DLTS results, it was possible to calculate $N_{\text{DS}}$, $N_2 = N_1$, and $N_{\text{AS}}$ (but not $N_3$) at each of the low fluences.

The $N$ vs $\phi$ results are plotted in Fig. 2. Note that the $N_{\text{DS}}$ data are highly dependent on whether $C_1$ is assumed to be a donor or an acceptor, but independent of $C_2$ and $C_3$, which are deeper. The $N_2$ data, on the other hand, are only very slightly dependent on the value of $N_1$ at low fluences, and independent of all assumptions at the higher fluences. In contrast, the values of $N_{\text{AS}}$ are highly dependent on the $D/A$ natures of $C_1$, $C_2$, and $C_3$ at all fluences, as outlined in Table I. Three representative $D/A$ cases are plotted in Fig. 2, and each is seen to be quite linear. In fact, the only decidedly nonlinear $N_{\text{AS}}$ vs $\phi$ plot is for case $AA(\text{not shown})$, and this case is thus probably not correct.

The production rates deduced from the slopes of the various $N$ vs $\phi$ plots are listed in Table II. The values of $\tau_2 = 2.0 \pm 0.2$ and $\tau_3 = 0.5 \pm 0.2$ cm$^{-1}$ are very consistent with the DLTS results, 1.5–1.8 and 0.4–0.7 cm$^{-1}$, respectively. However, the high value of $\tau_{\text{DS}}$, required if $C_1$ is an acceptor, is inconsistent with other data, and thus $C_1$ is probably a donor. Also, $C_1$ is almost certainly a donor, since its electron capture cross section is quite large, $\sim 1 \times 10^{-13}$ cm$^2$. In fact, the identification of $C_1$ and $C_2$ as the double-donor states of the As vacancy fits well with all experimental evidence, except the fact that the free-electron concentration diminishes in irradiated $n$-type GaAs while there are no other DLTS traps of a sufficient concentration to provide the necessary acceptors. This dilemma is immediately resolved by our data. From Table II, if $C_1$ and $C_2$ are donors, then $\tau_{\text{AS}} \approx 5.0 \pm 0.5$ cm$^{-1}$. However, we prefer to quote a more con-

![FIG. 2. Concentrations of $N_{\text{DS}}$, $N_2$, $N_1$, and $N_{\text{AS}}$ 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 $X$ character of $C_1$, $C_2$, and $C_3$, respectively.](image_url)

![FIG. 1. 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, $g_2 = g_3 = 0.5$. The fit at $\phi = 0.8 \times 10^{14}$ e/cm$^2$ is very poor due to inhomogeneity.](image_url)

![TABLE I. Calculation of $N_{\text{AS}}$ from Eq. (3) for various fluences $\phi$ ($10^{14}$ e/cm$^2$), and various donor/acceptor combinations of $C_1$, $C_2$, and $C_3$.](table)

<table>
<thead>
<tr>
<th>$C_1$</th>
<th>$C_2$</th>
<th>$C_3$</th>
<th>$N_{\text{AS}}$ (for $\phi = 0.8-1.4$)abc</th>
<th>$N_{\text{AS}}$ (for $\phi = 2.4$)abc</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D$</td>
<td>$D$</td>
<td>$D$</td>
<td>$N_{\text{DS}} + N_1 - K$</td>
<td>$N_{\text{DS}} + N_1 + N_2 - K$</td>
</tr>
<tr>
<td>$A$</td>
<td>$D$</td>
<td>$D$</td>
<td>$N_{\text{DS}} - K$</td>
<td>$N_{\text{DS}} + N_2 - K$</td>
</tr>
<tr>
<td>$A$</td>
<td>$D$</td>
<td>$A$</td>
<td>$N_{\text{DS}} + N_1 - N_2 - K$</td>
<td>$N_{\text{DS}} + N_1 + N_2 - K$</td>
</tr>
<tr>
<td>$A$</td>
<td>$A$</td>
<td>$D$</td>
<td>$N_{\text{DS}} - N_1 - K$</td>
<td>$N_{\text{DS}} + N_2 - K$</td>
</tr>
<tr>
<td>$A$</td>
<td>$A$</td>
<td>$A$</td>
<td>$N_{\text{DS}} + N_1 - N_2 - K$</td>
<td>$N_{\text{DS}} + N_1 - K$</td>
</tr>
<tr>
<td>$A$</td>
<td>$A$</td>
<td>$A$</td>
<td>$N_{\text{DS}} - N_1 - K$</td>
<td>$N_{\text{DS}} - K$</td>
</tr>
<tr>
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<td>$A$</td>
<td>$A$</td>
<td>$N_{\text{DS}} + N_1 - N_2 - K$</td>
<td>$N_{\text{DS}} + N_1 - K$</td>
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<tr>
<td>$A$</td>
<td>$A$</td>
<td>$A$</td>
<td>$N_{\text{DS}} - N_1 - K$</td>
<td>$N_{\text{DS}} - K$</td>
</tr>
</tbody>
</table>

* $K$ is fitting parameter (negative for all $\phi$); $N_{\text{DS}}$ determined from $\tau_{\text{DS}}$, measured at lower fluences; $N_1$ assumed equal to $N_2$.

$N_2$, $N_1$ are fitting parameters.

$N_1$ is a fitting parameter; $N_1$ determined from $\tau_1$ measured at lower fluences.
TABLE II. Defect production rates* in n-type GaAs for various donor/acceptor combinations of $C_1$, $C_2$, and $C_3$.

<table>
<thead>
<tr>
<th>$C_1$</th>
<th>$C_2$</th>
<th>$C_3$</th>
<th>$\tau_{10}$</th>
<th>$\tau_1$</th>
<th>$\tau_2$</th>
<th>$\tau_{3s}$</th>
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</thead>
<tbody>
<tr>
<td>$D$</td>
<td>$D$</td>
<td>$D$</td>
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<td>2.0</td>
<td>0.5</td>
<td>5.4</td>
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<tr>
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<td>$D$</td>
<td>$D$</td>
<td>1.3</td>
<td>2.0</td>
<td>0.5</td>
<td>4.5</td>
</tr>
<tr>
<td>$D$</td>
<td>$D$</td>
<td>$A$</td>
<td>0.2</td>
<td>2.0</td>
<td>0.5</td>
<td>4.5</td>
</tr>
<tr>
<td>$A$</td>
<td>$D$</td>
<td>$A$</td>
<td>1.3</td>
<td>2.0</td>
<td>0.5</td>
<td>3.6</td>
</tr>
<tr>
<td>$D$</td>
<td>$A$</td>
<td>$A$</td>
<td>0.2</td>
<td>2.0</td>
<td>0.5</td>
<td>3.5</td>
</tr>
<tr>
<td>$A$</td>
<td>$A$</td>
<td>$A$</td>
<td>1.3</td>
<td>2.0</td>
<td>0.5</td>
<td>2.6</td>
</tr>
<tr>
<td>$A$</td>
<td>$A$</td>
<td>$A$</td>
<td>0.2</td>
<td>2.0</td>
<td>0.5</td>
<td>2.7</td>
</tr>
</tbody>
</table>

*Units of cm$^{-1}$, typical errors: ± 15% or ± 0.2 cm$^{-1}$, whichever is greater.

Plot very nonlinear.

servative value for $\tau_{3s}$, $4 \pm 1$ cm$^{-1}$, which covers every reasonable $D/A$ case in Table II to integer accuracy. The important point is that a very high rate of acceptors $C_{4s}$, lying below $E_a$, is being produced, and it is entirely unnecessary to require either $C_1$ or $C_2$ to be an acceptor. It is rather unfortunate that many of the models proposed in the past for $C_1$ or $C_2$, whether right or wrong, have been influenced by this unnecessary requirement.²,³

We postulate that the $C_{4s}$ acceptors could well be Ga sublattice damage (GSLD), i.e., perhaps $V_{Ga}^-$ or the Frenkel pair $V_{Ga^-}Ga_0$, for the following reasons. (1) The GSLD should be produced at about the same rate as that of the measured As sublattice damage² (ASLD), i.e., about $5 \pm 1$ cm$^{-1}$. The $C_{4s}$ rate is $4 \pm 1$ cm$^{-1}$. (2) The GSLD should be mainly acceptor in nature, since $V_{Ga}$ and $V_{Ga^-}Ga_0$ are probably dominated by acceptor states.⁹ Of course, $C_{4s}$ is also an acceptor. (3) The GSLD may well be unstable in p-type materials, since the Ga, can become positively charged, leading to a recombination, or the $V_{Ga^-}$ can, by a single As hop, be transformed to $V_{Ga^-}As_{Ga}$, which is known to be more stable in p-type material.¹⁰ This instability explains both the low production rate of DLTS hole traps in p-type material, and the upward movement of $E_F$ in p-type material, as observed by Hall effect.

In spite of the consistency of the GSLD model with experimental and theoretical results, we cannot rule out the possibility that the $C_{4s}$ consist of the hole traps $H0$ and/or $H1$, which are produced at a combined rate of only about $1 \text{ cm}^{-1}$ in p-type material, but might have a much higher rate in n-type material. In this case, we would not need to invoke GSLD, since $H0$ and $H1$ are presumably associated with ASLD.² One problem here is that the total ASLD would then be larger than $7 \text{ cm}^{-1}$, which is the maximum expected rate per sublattice.² In any case, more work, including careful isothermal annealing experiments, will be necessary to finally identify the $C_{4s}$, which is the existence of the $C_{4s}$, which must be taken into account in any future defect modeling.

This work was performed at Wright-Patterson AFB under USAF contract F33615-86-C-1062. We wish to thank J. Sizelove for data analysis, T. Cooper for electrical measurements, and P. Schwenke for manuscript preparation.