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New As_{Ga} Related Center in GaAs

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A new center related to As_{Ga} has been found at relatively high concentrations (10^{17} cm^{-3}) in semi-insulating ($2 \times 10^7 \Omega \text{ cm}$) molecular beam epitaxial GaAs grown at 400°C . Although the ir photoquenching and thermal recovery characteristics are nearly identical to those of $EL2$, the thermal activation energy is only $0.65 \pm 0.01 \text{ eV}$, much lower than the $EL2$ value of $0.75 \pm 0.01 \text{ eV}$. Other properties which are different include the electron-capture barrier energy, hyperfine constant, and magnetic circular dichroism spectrum.

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The arsenic antisite As_{Ga} in GaAs, and specifically its manifestation known as $EL2$ [from early deep-level transient spectroscopy (DLTS) studies], is perhaps the most studied defect in the history of semiconductors, because of both its technological importance and its interesting physical phenomena [1]. Thus it is surprising that there is still controversy over the microscopic structure of $EL2$, i.e., whether it is an isolated As_{Ga} center [2,3] or a complex with another defect, say, the arsenic interstitial As_i [4]. Over the years there has been evidence usually from a single experiment of one type or another that there is more than one “ $EL2$ -like” defect. However, as far as we know, no investigations have ever found a proven As_{Ga} related center with a thermal activation different from the $EL2$ value of 0.75 eV [5,6]. In this work we introduce such a defect and show that it is nearly identical to $EL2$ in several ways, but significantly different in several other ways.

This new center is found in GaAs layers grown by molecular beam epitaxy (MBE) at a substrate temperature of 400°C , well below the usual growth temperature range of 580 – 600°C . Actually, a new field of research and technology has grown around 200 and 250°C MBE material in the last several years because of important electronic and photoelectronic device applications for that material [7]. In light of the large volume of literature on the 200 and 250°C materials, it is useful to contrast their properties with those of the 400°C samples. In Fig. 1 we show the dark current under 20 V bias for several samples of approximate dimensions $6 \text{ mm} \times 6 \text{ mm}$ with In contacts on the corners. Two of the samples, LEC066 and LEC113, were from typical commercial liquid-encapsulated Czochralski (LEC) semi-insulating (SI) wafers of about $650 \mu\text{m}$ thickness, while LT282 was a $2\text{-}\mu\text{m}$ -thick MBE sample grown at 250°C and annealed at various temperatures, and LT283 and LT284 were $5\text{-}\mu\text{m}$ -thick MBE layers grown at 350 and 400°C , respectively, and not annealed. A critical factor here is that all the LTMBE layers were removed from their substrates

before measurement, because otherwise the substrate would have carried most of the current, and accurate measurements would have been impossible [8].

The first thing to note in Fig. 1 is that the 250°C grown samples, even after annealing, have strong conduction at low temperatures. This conduction is due to hopping between the dense As_{Ga} centers, a phenomenon which has been investigated earlier [9]. Strong, thermally activated band conductivity is observed only above 300 K in the sample annealed at 600°C . On the other hand, the layers grown at 350 and 400°C , as well as the LEC samples, show *only* band conductivity, because the As_{Ga} centers are too far apart to sustain hopping. An important point is that the dark-current activation energies of

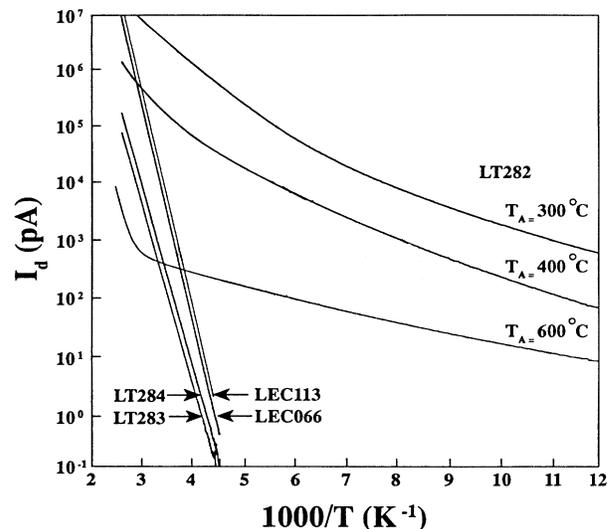


FIG. 1. The dark current I_d vs inverse temperature T^{-1} for several samples: LT282 (grown by MBE at 250°C and annealed at 300 , 400 , and 600°C); LT283 (grown by MBE at 350°C); LT284 (grown by MBE at 400°C); LEC066 and LEC113 (bulk, semi-insulating wafers).

the LEC and LTMBE samples are different: 0.74 eV for both of the LEC wafers, and 0.63 eV for both the 350 and 400°C MBE layers. (The slope of the line above 300 K for the 250°C layer annealed at 600°C cannot be easily determined because of too short a straight-line portion.)

A more accurate value of the thermal activation energy E_D can be found from Hall effect measurements. The electron concentration n is given by [5]

$$n = \frac{2(2\pi m^* k)^{3/2}}{h^3} \frac{g_u}{g_o} e^{a/kT} T^{3/2} \left[\frac{N_D}{N_A} - 1 \right] e^{-E_{D0}/kT}$$

$$= CT^{3/2} \left[\frac{N_D}{N_A} - 1 \right] e^{-E_{D0}/kT}, \quad (1)$$

where $E_D = E_{D0} - \alpha T$ and the other symbols have their usual meanings. For LEC066, LT283, and LT284, we find that $E_{D0} = 0.745$, 0.644, and 0.654 eV, respectively. For LT284 (400°C), $C(N_D/N_A - 1) \approx 5.7 \times 10^{15}$, which gives $N_D/N_A \approx 4$ if we use Blakemore's suggested value of $C = 1.85 \times 10^{15}$ for EL2 [10]. However, we can also determine N_D and N_A individually by applying a novel technique in which electron (or hole) transfer from an adjacent n -type (or p -type) layer is measured by the Hall effect and analyzed by Poisson's equation [see Eq. (6) of Ref. [11]]. This technique, to be published in detail elsewhere, gives $N_D = 1.4 \times 10^{17}$ and $N_A = 2.7 \times 10^{16} \text{ cm}^{-3}$, or $N_D/N_A \approx 5$, close to the value estimated above. If $g_u/g_o = 2$ (unoccupied/occupied degeneracies for the $+/0$ transition), then the temperature factor α is about $1.8 \times 10^{-4} \text{ eV/K}$, a reasonable number for a deep level.

From a practical point of view, we might also note that the 300-K resistivity for LT284 is about $2 \times 10^7 \Omega \text{ cm}$, which is the highest resistivity ever measured, to our knowledge, in epitaxial GaAs. Furthermore, the material has a high mobility ($> 5 \times 10^3 \text{ cm}^2/\text{Vs}$ at 300 K) and is not strongly dependent on growth conditions (e.g., growth temperatures of 350 and 400°C give similar results).

Two other experiments prove that this center is an As_{Ga} related defect. The first is optically detected electron paramagnetic resonance (ODEPR) which shows the characteristic As_{Ga} four-line spectrum. This work is described in detail elsewhere [12]. The second experiment is the quenching and recovery of the photocurrent (PC), which is shown in Fig. 2. In the unquenched case, the sample is cooled to 82 K in the dark, and then the PC is measured under weak 1.46 eV illumination as the temperature is swept upward at a constant rate β (0.2 K/s in this case). The PC decreases as temperature increases because the electron must overcome a barrier E_a to be recaptured, which is more easily accomplished at higher temperatures [5]. For EL2, E_a has been determined from capacitance by one group [13] to be 0.075 eV over the range $150 < T < 245 \text{ K}$, and, by another group [14], 0.066 eV over the range $50 < T < 275 \text{ K}$. For sample LEC066, our PC data from 80 to 180 K (not shown) are

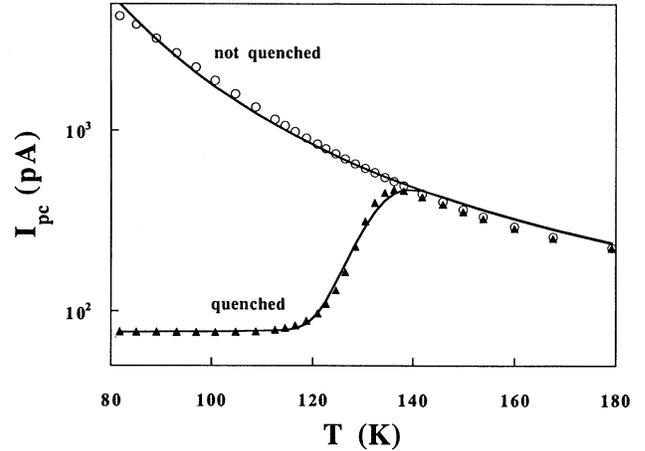


FIG. 2. The photocurrent I_{pc} vs temperature T for LT284, grown at 400°C, after initial ir illumination at 82 K (triangles), and without such initial illumination (circles). The solid lines are theoretical fits.

reasonably well fitted with $E_a = 0.073 \text{ eV}$ [see Eq. (4) below]. On the other hand, the LTMBE data in Fig. 2 are quite well fitted (solid line) over the range $80 < T < 180 \text{ K}$ with $E_a = 0.040 \text{ eV}$, clearly smaller than the EL2 value.

We next consider quenching of the PC. The PC from 1.46 eV light, which is subband gap at the temperatures of this experiment, is probably mainly due to electron excitation from neutral As_{Ga} centers, of concentration N_D^0 , where $N_D^0 = N_D - N_D^+ \approx N_D - N_A$ in the dark, or in weak light. However, if we illuminate the sample with strong 1.1 eV light at 82 K, then $N_D^0 \rightarrow N_D^*$, the metastable state, and the PC will greatly decrease [1], as shown. In the quenched state, the PC under weak 1.46 eV light will probably be a hole current due to photoexcitation of holes from the unoccupied As_{Ga} centers of concentration N_A . (In fact, preliminary Hall effect measurements confirm p -type conduction.) Such holes must be recaptured on nonmetastable neutral centers which are now of a concentration only equal to the number of holes. Since hole capture is evidently not activated [14], the current will remain relatively constant as temperature swept upward, until the metastable As_{Ga} centers begin recovering and producing more neutral centers. As $N_D^* \rightarrow N_D^0$, the hole current will rapidly decrease because of the greatly increased numbers of capture sites (neutral As_{Ga} centers). However, the electron current will now increase because of increased photoexcitation from the N_D^0 .

This process can be modeled as follows. As temperature is swept at a rate β the N_D^* obey

$$\frac{dN_D^*}{dT} = \frac{1}{\beta} \frac{dN_D^*}{dt} = \frac{v_0}{\beta} e^{-E_b/kT} N_D^*, \quad (2)$$

where E_b is the metastability recovery barrier and v_0 is the prefactor. From a simple rate equation the electron

concentration n at steady state is given by

$$n = \frac{I_0 \sigma_{vn} (N_D - N_A - N_D^*)}{N_A \sigma_n v_n} = \frac{I_0 \sigma_{vn}}{\sigma_n v_n} \left[\frac{N_D}{N_A} - 1 \right] \left[1 - \frac{N_D^*}{N_D - N_A} \right], \quad (3)$$

where I_0 is the light intensity, σ_{vn} the photoionization cross section for electrons, σ_n the capture cross section for electrons, and v_n the thermal velocity. For the unquenched sample at all temperatures, and the quenched sample above 140 K, holes are negligible and the fit gives $I_{PC} = C \exp(E_a/kT)$, where $C \approx 18$ pA and $E_a \approx 0.040$ eV. Thus, the electron current is given by

$$I_{PC}^e = C e^{E_a/kT} \left[1 - \frac{N_D^*}{N_D - N_A} \right] = C e^{E_a/kT} \left[1 - \frac{N_D^*(T_i)}{N_D - N_A} F(T) \right], \quad (4)$$

where T_i is the initial temperature (82 K), C is a constant depending on bias and geometrical factors, and

$$F(T) = \exp \left\{ - \int_{T_i}^T \frac{v_0}{\beta} e^{-E_b/kT} dT \right\}. \quad (5)$$

As stated earlier, when all of the neutral As_{Ga} centers are

quenched, then the hole current will become important and considerations similar to those leading to Eq. (4) give

$$I_{PC}^{hole} = D \left\{ \left[1 + K^2 \left(1 - \frac{N_D^*(T_i)}{N_D - N_A} F(T) \right)^2 \right]^{1/2} - K \left[1 - \frac{N_D^*(T_i)}{N_D - N_A} F(T) \right] \right\}, \quad (6)$$

where $K = (N_D - N_A)(\sigma_p v_p)^{1/2} / 2(I_0 \sigma_{vp} N_A)^{1/2}$, and D is a constant depending on bias and geometrical factors. The excellent fit of $I_{PC} = I_{PC}^e + I_{PC}^{hole}$ to the LTMBE data of Fig. 2 gives $v_0 = 2.5 \times 10^8 \text{ s}^{-1}$ and $E_b = 0.26$ eV. If we leave out the hole current, Eq. (6), then $v_0 = 2.8 \times 10^8 \text{ s}^{-1}$, or if we make the hole current a constant, then $v_0 = 1.6 \times 10^8 \text{ s}^{-1}$, with E_b remaining at 0.26 eV in either case. Of course, the overall fits are much worse in these latter two cases, but the important point is that only Eq. (4) is of much consequence for determining E_b and v_0 . To compare with *EL2*, recently Mohapatra and Kumar [15] used PC measurements in SI GaAs to calculate $v_0 = 2.5 \times 10^8 \text{ s}^{-1}$ and $E_b = 0.26$ eV, exactly the same as our best-fit values for the LTMBE data. Fischer [16] has used ir absorption data to find $E_b = 0.25\text{--}0.30$ eV in various semi-insulating samples. For sample LEC066, containing only *EL2*, we fitted our quenched PC data with

TABLE I. Comparison of *EL2* and new As_{Ga} center.

Property	<i>EL2</i>	New center
Thermal activation energy (Hall)	0.75 ± 0.01 eV ^{a,b}	0.64 ± 0.01 eV ^a
Metastability recovery barrier (PC)	0.26 ± 0.01 eV ^{a,c,d}	0.26 ± 0.01 eV ^a
Recovery prefactor (PC)	$(2.5 \pm 1) \times 10^8 \text{ s}^{-1}$ ^{a,c,d}	$(2.5 \pm 1) \times 10^8 \text{ s}^{-1}$ ^a
Electron capture barrier energy (PC)	$0.066\text{--}0.075$ eV ^{a,e,f}	0.040 ± 0.002 eV ^a
Magnetic resonance spectrum (EPR or ODEPR)	Four line (As_{Ga}) ^{g,h}	Four line (As_{Ga}) ^h (broader lines)
<i>g</i> factor (ODEPR)	2.04 ± 0.01 ^g	2.03 ± 0.01 ^h
Hyperfine constant (ODEPR)	2600 ± 70 MHz ^{g,h} (isotropic)	2300 ± 70 MHz ^h (isotropic)
Magnetic circular dichroism spectrum	Standard <i>EL2</i> ^{h,i}	Much different in 0.8 to 1.2 eV region ^h

^aThis work.

^bReferences [5,6].

^cReference [15].

^dReference [1].

^eReference [13].

^fReference [14].

^gR. J. Wagner, J. J. Krebs, G. H. Stauss, and A. M. White, *Solid State Commun.* **36**, 15 (1980).

^hReference [12].

ⁱK. Krambrock, J-M. Spaeth, C. Delerue, G. Allan, and M. Lannoo, *Phys. Rev. B* **45**, 1481 (1992).

$\nu_0 = 1.1 \times 10^8 \text{ s}^{-1}$ and $E_b = 0.26 \text{ eV}$. Thus, there is little doubt that the metastability recovery kinetics are nearly identical for *EL2* and the new As_{Ga} center in our LTMBE sample.

The LTMBE results presented here and in Ref. [12] are summarized in Table I, along with *EL2* data from our work and from the literature. It is obvious that the new center is As_{Ga} related, and very similar in some aspects to *EL2*, but not in other aspects. The similarities result from experiments which are evidently influenced primarily by the core As_{Ga} , and the differences from experiments which are sensitive to the local environment. Speculation on the microscopic structure of the new center is perhaps unwarranted in the light of the fact that even the structure of *EL2* is still controversial. However, we may note that LTMBE GaAs grown at 200 or 250 °C has a high native acceptor concentration ($\sim 10^{18}$ – 10^{19} cm^{-3}) [8], thought to be V_{Ga} related because of the strong As-rich stoichiometry [17] and our 400 °C material may have some of the same centers, even though of much lower concentration ($3 \times 10^{16} \text{ cm}^{-3}$). Thus, the new defect described here may be a complex involving As_{Ga} and V_{Ga} , which could form via a Coulomb attraction. Although centers involving As_{Ga} and V_{Ga} have already been proposed [18], there is no compelling evidence as to their existence.

In summary, we have for the first time established the existence of an As_{Ga} related center with a thermal activation energy different from that of *EL2*. Also different are the electron-capture barrier energy, the hyperfine constant, and the shape of the magnetic circular dichroism spectrum, whereas the *g* factor and metastability recovery kinetics are nearly identical. The microscopic structure is not yet known.

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