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# Double carriers pulse DLTS for the characterization of electron–hole recombination process in GaAsN grown by chemical beam epitaxy

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A nitrogen-related electron trap (E1), located approximately 0.33 eV from the conduction band minimum of GaAsN grown by chemical beam epitaxy, was confirmed by investigating the dependence of its density with N concentration. This level exhibits a high capture cross section compared with that of native defects in GaAs. Its density increases significantly with N concentration, persists following post-thermal annealing, and was found to be quasi-uniformly distributed. These results indicate that E1 is a stable defect that is formed during growth to compensate for the tensile strain caused by N. Furthermore, E1 was confirmed to act as a recombination center by comparing its activation energy with that of the recombination current in the depletion region of the alloy. However, this technique cannot characterize the electron – hole (e–h) recombination process. For that, double carrier pulse deep level transient spectroscopy is used to confirm the non-radiative e–h recombination process through E1, to estimate the capture cross section of holes, and to evaluate the energy of multi-phonon emission. Furthermore, a configuration coordinate diagram is modeled based on the physical parameters of E1.

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## 1. Introduction

InGaAsN, having a bandgap of 1.0 eV and lattice matched to both GaAs and Ge, can be obtained with the incorporation of around 3% nitrogen (N) and 9% indium (In) in the host material. Such an alloy has been identified as an ideal candidate for ultra-high efficiency multi-junction tandem solar cells, such as the Ge/InGaAsN/GaAs/InGaP structure [1]. However, the alloy's optoelectronic properties are significantly degraded with increasing N concentration [2–4]. One evident reason for such degradation is the increased density of N-related recombination and/or scattering centers [5]. Their formation has been attributed to the small atomic size of N compared with that of arsenic (As) as well as to the large miscibility between GaAs and GaN. In related literature, some N-related lattice defects were theoretically predicted from first principles calculations and investigated experimentally. For example, the summary of experimental results reported by Geisz and Friedman [6] provides a basic knowledge of these lattice defects in InGaAsN that has been grown by various techniques. Although they are important results that come from these studies, no N-related recombination center has yet been confirmed using a direct method and the recombination process in the material is still unclear. Therefore, identifying and clarifying the recombination process in InGaAsN are important steps to improving the electrical properties of the film.

Recently, we have confirmed the existence of a N-related deep electron trap (E1), located around 0.33 eV from the conduction band minimum (CBM), in p-type and n-type GaAsN grown by chemical beam epitaxy (CBE) [7]. This level exhibits a high capture cross section compared with that of native defects in GaAs. Its density increases significantly with N concentration, persists following thermal annealing, and is found to be quasi-uniformly distributed. These results indicate that E1 is a stable defect that is formed during growth to compensate for the tensile strain caused by N. Furthermore, E1 is observed with approximately the same density in GaAsN grown by both molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD) [8,9]. Such results confirm the independence of E1's formation from growth conditions, essentially the concentration of impurities and dopant atoms. Concerning its origin, proton implantation and As flow rate dependence of E1's density were used to investigate the possible atomic structure of E1. We have found that E1 has a high probability of associating with the split interstitial $(N–As)_{As}$ in a single As site, which is in conformity with the first-principles calculation [10,11]. In addition, E1 is confirmed to act as a recombination center by correlating between its thermal activation energy and that of the reverse bias current in the depletion region of GaAsN [12]. However, this method can only provide the nature of E1, without clarifying the recombination process through it. For that, a direct and efficient tool is required, such as the double

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carrier pulse deep level transient spectroscopy (DC-DLTS) technique [13,14].

In the present paper, we have confirmed the nature of E1 using DC-DLTS. The non-radiative recombination process is verified by investigating the temperature dependence of the capture cross section of electrons, which is evaluated at room temperature to be $\sigma_n(300\,\text{K}) \sim 8.89\times10^{-12}\,\text{cm}^2$. Furthermore, the physical parameters of E1 are summarized in a configuration coordinate diagram (CCD).

## 2. Experimental procedure

### 2.1. Growth conditions and sample preparation

An unintentionally doped n-type GaAsN Schottky junction ($\sim$1 μm) was grown on a p-type GaAs (0 0 1) substrate, 2° off cut towards (0 1 0), by the CBE method at a growth temperature and pressure of 420 °C and $2\times10^{-2}$ Pa, respectively. Triethylgallium ($(C_2H_5)_2Ga$; $TEGa$=0.1 sccm), tridimethylaminoarsenic ($((CH_3)_2N)_3As$; $TDMAAs$=1.0 sccm), and monomethylhydrazine ($H_3N_2CH_3$; $MMHy$=7.0 sccm) were used as the gallium (Ga), As, and N source materials, respectively. This structure is not commonly used for DLTS measurements; however, the absence of a p-type doping source and the existence of deep acceptors in unintentionally doped p-type GaAsN prevented us from obtaining a $p^+$–n junction for all the scanning temperature range. The p-type substrate was used as source of minority carriers in the sample. The N concentration of the GaAsN layer was determined by X-ray diffraction (XRD) to be $\sim$0.25%, using Vegard's law. The bandgap ($E_g$) was evaluated by transmittance measurements to be 1.35 eV. Al dots with a diameter of 1 mm and an Au–Zn alloy (95%:5%) were deposited at the top and bottom surfaces as Schottky and Ohmic contacts, respectively. The free electron concentration $N_d$ is evaluated at room temperature to be $\sim 2.62\times10^{16}\,\text{cm}^{-3}$, using the capacitance–voltage ($C$–$V$) method [15]. DLTS spectra are collected using a BIO-RAD digital DLTS system DL8000. The activation energy $E_a(E_C-E_T\ (\text{eV}))$ and the capture cross section $\sigma_n$ (cm$^2$) are determined from the slope and the intercept values of the Arrhenius plot of the DLTS spectra, respectively [13]. The density of each trap was evaluated using a conventional DLTS method [13].

### 2.2. Fundamental of DC-DLTS method

The DC-DLTS method is used in asymmetric $n^+$–p or $p^+$-n junctions. Its basic operation is illustrated in Fig. 1; it aims to check whether an electron trap is a recombination center or not. Two pulsed biases are applied to the sample, in turn, to inject majority and minority carriers to an electron trap. At the initial state, the junction is under reverse bias, and the energetic level ($E_T$) of the trap is higher than the Fermi level ($E_{Fn}$). When the first pulse voltage is applied to the sample, $E_{Fn}$ is higher than $E_T$, which allows the trap to capture electrons. During the second forward biased pulse,with a duration $t_{ip}$, holes are injected to the depletion region from the p-side of the junction. After the junction pulse is turned off, electrons and holes are thermally emitted. The amount of trapped carriers can be observed as a change in the DLTS peak height of the trap. If the trap captures both minority and majority carriers, the DLTS maximum of the corresponding level decreases compared with that in conventional DLTS. Such a decrease is explained by the electron–hole (e–h) recombination process, which indicates that the level is a recombination center. Quantitatively, the relationship between the densities of the defect $N_T$ and ionized traps $n_T(t)$ in the n-side of the junction can be expressed according to

$$\frac{dn_T(t)}{dt} = -e_p n_T(t) + \langle p \rangle C_p[\mathrm{N_T}-\mathrm{n_T}(t)] - N_d C_n n_T(t) + e_n[\mathrm{N_T}-\mathrm{n_T}(t)] \tag{1}$$

where $e_{n,p}$ and $C_{n,p}$ denote the emission and capture rates for electrons ($n$) and holes ($p$), respectively; $\langle p \rangle$ and $N_d$ are the average of injected minority carriers and the free electron concentration in the n-side of the sample, respectively. As a solution of Eq. (1), we have

$$n_T(t_{ip}) = n_T(\infty) + [n_T(0) - n_T(\infty)]\exp(-\tau^{-1} t_{ip}) \tag{2}$$

where $t_{ip}$ is the width of the injected pulse, $n_T(\infty) = \frac{\langle p \rangle C_p + e_n}{\tau^{-1} N_T}$ and $\tau^{-1} = \langle p \rangle C_p + N_d C_n + e_p + e_n$. Considering the $I_{DLTS}$ and $I_{DCDLTS}$ the peak heights of the defect in conventional and double carrier pulse DLTS, respectively. Eq. (2) can be rewritten properly as

$$\frac{n_T(t_{ip})}{N_T} = \frac{I_{DLTS} - I_{DCDLTS}}{I_{DCDLTS}} = \langle p \rangle \sigma_p v_{thp} t_{ip} \tag{3}$$

where $\sigma_p$ and $v_{thp}$ denote the capture cross section of holes and thermal velocity of holes at the peak's temperature of the defect, respectively.

## 3. Results and discussion

### 3.1. Conventional DLTS results

Presented in Fig. 2 and in its inset are the current–voltage ($I$–$V$) characteristics of the heterojunction and the Mott–Schottky plot of the GaAsN layer. These plots show the high quality of the heterojunction, the low leakage current for applied reverse bias, and the quasi-uniform doping in the n-type GaAsN junction over the reverse bias range from 1 to 0 V. This structure is then suitable for carrying out DLTS measurements in the n-type GaAsN layer. As given in Fig. 3(a), the DLTS signal of the majority carriers was recorded under a reverse bias voltage $V_R$=1 V, a pulse voltage $V_p$=0 V, a filling pulse width $t_p$=5 ms, and an emission rate window $e_n = 10\,\text{s}^{-1}$. Two electron traps, E1 and E2, were observed at approximately 0.33 and 0.45 eV from the CBM of GaAsN, respectively. The Arrhenius plot of the DLTS

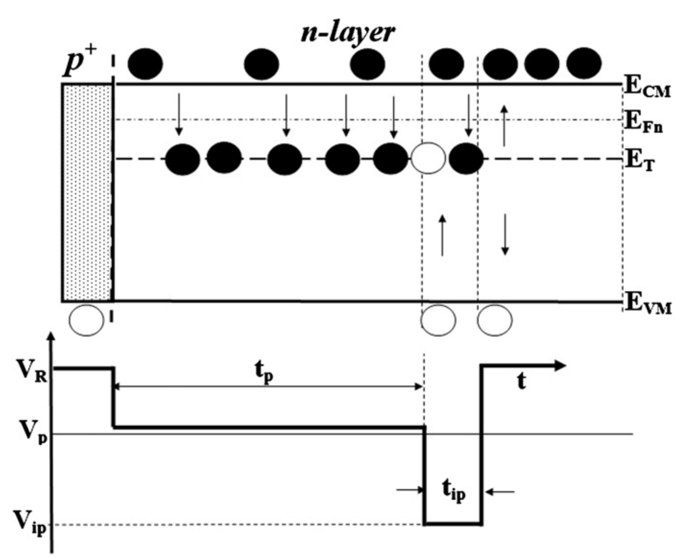


**Fig. 1.** Basic concept of capture and thermal emission processes from an electron trap located at an energy level $E_T$ in $p^+$–n junction. A saturating injection pulse is applied to the reversed biased junction to fill the trap with holes. Turning off the pulse returns the junction to equilibrium, resulting in a thermal emission of holes from the trap to the valence band.

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