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Effect of Thermal Stress on a N-related Recombination Center in GaAsN Grown by Chemical Beam Epitaxy

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1. Introduction

Insuring the current matching in the multijunction solar cell structure Ge (0.76 eV)/InGaAsN (1.04 eV)/GaAs (1.42 eV)/InGaP (1.89 eV) will push the conversion efficiency to over 40 % under AM0 illumination and will contribute greatly in the photovoltaic market. This objective can only be achieved if the diffusion length of minority carriers in InGaAsN is improved to the same level as in the host material. One key factor to increase such parameter is by dropping the density of recombination centers in the alloy and by recovering the lifetime of minority carriers. Recently, we have confirmed the existence of N-related non radiative recombination center, localized between 0.3 and 0.4 eV below the conduction band minimum (CBM) of GaAsN grown by chemical beam epitaxy (CBE) [1, 2]. The important properties of this lattice defect are the high trapping density (> 10^{16} cm^{-3}) and the extremely large capture cross section for electrons at room temperature (~ 10^{-13} cm^2) [2]. These parameters have been used to obtain the lifetime of electrons from the CBM to around ~0.2 ns, using the Shockley–Read–Hall model for generation–recombination [2]. The origin of this energy level was investigated experimentally and suggested to be the split interstitial $(N\text{-}As)_{As}$ formed from one N and one As in a single As site [3]. Actually, our preoccupation is to investigate the formation process of this defect to limit its density, aiming to improve the lifetime of minority carriers. In this paper, the evolution of the N-related recombination center under annealing of GaAsN grown by CBE is reported.

2. Experimental Procedure

Si-doped GaAsN film was grown by CBE on n-type GaAs (001), 2° off toward (010) substrates (2 cm x 2 cm), under a growth temperature and a pressure of 440 °C and ~ 2 x 10^{-2} Pa, respectively. Triethylgallium (TEGa = 0.1 sccm), tridimethylaminoarsenic (TDMAAs = 1.0 sccm), and monomethylhydrazine (MMHy = 9.0 sccm) were used as Ga, As, and N chemical compound sources, respectively. The N concentration [N] in the film was calculated from the X-ray diffraction method. The sample was treated by post thermal annealing for 5 min at a temperature TA (°C) = $(550 + n \times 50)_{1\leq n\leq 4}$, using GaAs cap layers and N_2 gas. The as grown and the annealed samples will be labeled S_0 and $(S_n)_{1\leq n\leq 4}$. Al dots with a diameter of 1 mm were evaporated as schottky contacts through a metal mask on the front side of the samples, under a vacuum of 10^{-4} Pa and an alloy of Au-Ge (88:12) was deposited on the bottom surface as ohmic contact. The ionized donor concentrations (N_D) at 300 and 155 K were calculated from the fitting of the Mott-Schottky plot using the capacitance-voltage method.

3. Results and Discussion

The DLTS spectra recorded on annealed samples S_1 to S_5 along with as grown sample are shown in Fig.1. In the reference sample, three major electron traps are observed with activation energies of 0.291, 0.407, and 0.874 eV below the CBM of the alloy, respectively. The two last defects are native defects, commonly observed in GaAs, and are identified as EL5 and EL2 under the classification of Martin, Mitonneau, and Mircea [4]. The first deep level (E1) is observed only after addition of N to GaAs. it was recorded in MOCVD, MBE, and CBE grown GaAsN [1]. As mentioned in the introduction, E1 is a N-related electron tarp with high density and large capture cross section. Furthermore, it was confirmed to act as a non-radiative recombination center and suggested to be the main cause of poor minority carrier lifetime in GaAsN films.

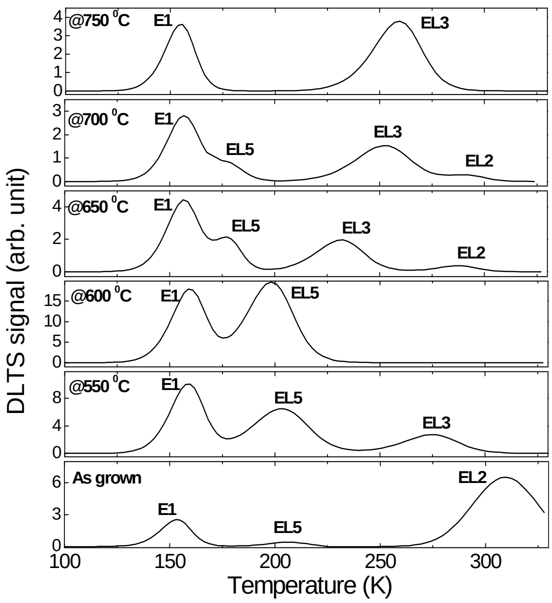

Fig. 1 DLTS spectra recorded on Al/Si-GaAsN Schottky Junctions with a reverse bias voltage of -1 V, a pulse voltage of 0 V, a rate-window time of 100 ms and a filling pulse width of 100 μs.

After annealing, a new defect is discontinuously enhanced between EL2 and EL5 at activation energy between 0.427 and 0.532 eV below the CBM, which is identified using again Martin, Mitonneau, and Mircea classification to EL3 [4]. The fluctuation of activation energy of all defects is due to the effect of electric filed on the thermal emission of electrons from the traps, which so called the Poole-Frenkel emission [1]. For the next, we will only focus on the evolution of the properties of E1 under post thermal cycle annealing. The physical properties of the samples, the signatures, and the trapping densities before and after adjustment of E1 are summarized in Table I.

Table I Physical properties of S_0 to S_5, signatures, and trapping densities before and after adjustment of E1

	[N] (%)	N_D (cm^{-3})	Ea (eV)	σ_n(E1) (cm^2)	N(E1) (cm^{-3})	N_{adj}(E1) (cm^{-3})
S_0	0.49	1.5×10^{17}	0.291	1.6×10^{-14}	5.9×10^{15}	1.9×10^{16}
S_1	0.49	3.4×10^{17}	0.277	2.6×10^{-15}	2.2×10^{16}	6.9×10^{16}
S_2	0.52	4.7×10^{17}	0.277	2.3×10^{-15}	3.9×10^{16}	1.2×10^{17}
S_3	0.45	2.2×10^{17}	0.287	7.4×10^{-15}	9.0×10^{15}	2.8×10^{16}
S_4	0.42	1.9×10^{17}	0.291	9.9×10^{-15}	7.5×10^{15}	2.4×10^{16}
S_5	0.47	5.9×10^{16}	0.299	2.3×10^{-15}	4.1×10^{15}	1.4×10^{16}

As shown in Fig.1 and Table I, no significant departure from the as grown DLTS signature of E1 occurs in annealed samples. The activation energy of E1 fluctuates in a range of 22 meV, which affects the capture cross section that was previously confirmed to have a thermal energy barrier [2]. Furthermore, the peak of E1 continues to appear clearly after annealing, which confirms our previous analyses about the stability of this recombination center and its uniform distribution in the bulk of GaAsN, as a reaction to compensate the tensile strain caused by the small atomic size of N compared with that of As. on another hand, the trapping density of E1 is adjusted according to the λ-effect factor to take in account the depletion region width, which fluctuates from one sample to another owing to the change in carrier concentration after annealing [1]. Here, an average adjustment factor of 3.2 is calculated and used. As shown in Fig. 2, the density of E1 changes dramatically and shows a peaking behavior. It increases significantly under annealing temperature of 550 and 600 °C. Then, it drops continuously to achieve, under annealing temperature of 750 °C, a density less than that in as grown sample. However, this density remains quietly high and can degrade the lifetime of electrons because the capture cross section is too large. To bring an explanation to the behavior of E1 under post thermal annealing, we have to consider the possible atomic structure of this recombination center. As mentioned in the introduction, our previous experimental results favorites the split interstitial $(N\text{-}As)_{As}$, as a possible origin of E1, since the density of this defects showed a clear sensitivity to As source flow rate [3]. Therefore, our analyses will repose on the effect of As out-diffusion on the density of E1. Similar to the growth of GaAs, our GaAsN films are grown under As-rich conditions. Experimentally, this leads to the existence of plenty interstitial As (As_i) in lattice sites [4, 5]. The thermal excitation of GaAsN lattice increases the mobility of As_i, which causes the accumulation of As surrounding other point defects and/or ideal lattice atoms such as N. a possible process can arise and leads to the formation of more $(N\text{-}As)_{As}$ is by interaction between N and As. Hence, we suggest that the thermal excitation under 550 and 600 °C has only produced more E1. However, the new formed centers may not have the same stability as for that in as grown sample, since the epitaxial growth under continuous temperature gives more stability and atomic arrangement better than under sudden thermal excitation. After that, the increase of the thermal stress in the lattice forces the As_i to out-diffuse. This can be supported by the recovering of [N] and the increase of EL3 concentration, which was related to As vacancy (V_{As}) [5].

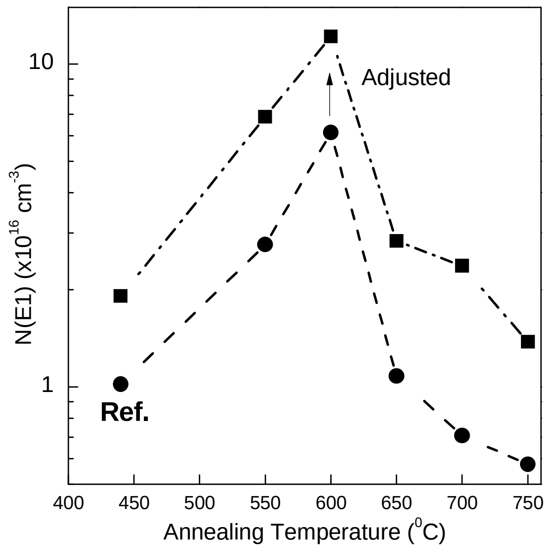

Fig. 2 Trapping density of E1 before and after adjustment as function of annealing temperature.

4. Conclusions

The main N-related non-radiative recombination center in GaAsN grown by CBE is a stable defect. Its density shows a peaking behavior with annealing. This behavior is explained by the dynamics of As out-diffusion under thermal excitation, since the defect is sensitive to the As atom.

Acknowledgements

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