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Defects in Electron-Irradiated and Hydrogenated GaAsN Grown by Chemical Beam Epitaxy

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1. Introduction

III-V-N dilute nitride is a promising material for ultra-high efficiency tandem solar cell, since its bang gap markedly decreases with increasing the N concentration in the film and without a significant change of lattice parameter. With only 3% of N and 9% of In, GaInAsN is a potential candidate to be introduced in the lattice–matched tandem Ge (0.76 eV)/InGaAsN (1.04 eV)/GaAs (1.42 eV)/InGaP (1.89 eV) cell, which could convert the sunlight over 40% to electricity [1]. Meanwhile, incorporating a small atomic fraction of N in the alloy degrades markedly the lifetime of electrons [1]. An obvious reason of such degradation is the presence of high electrically active recombination centers in the alloy [2]. For that, several studies on lattice defects in GaInAsN or GaAsN were carried out. Indeed, various growth techniques were performed to enhance the incorporation of N and to insure its uniform distribution. Furthermore, lattice defects in the alloy were extensively studied by using various characterization tools [2]. Nevertheless, further results are still required to understand the recombination process in GaInAsN.

Lattice defects in GaAsN grown by chemical beam epitaxy (CBE), were recently characterized using deep level transient spectroscopy (DLTS) and some other related methods [3]. An electron trap, E1, with thermal activation energy between 0.3 and 0.4 eV below the conduction band minimum (CBM) was confirmed to act as a N–related nonradiative recombination center [3]. It exhibits a high capture cross section (~ 10^{-13} cm^2) and a high trapping density (> 10^{16} cm^{-3}). Using these two parameters and the Shockley–Read–Hall model for generation–recombination, the lifetime of electrons was calculated to less than 0.30 ns. This result suggests that E1 is the main cause of short minority carrier lifetime in GaAsN grown by CBE. Therefore, it is essential to investigate its origin and understand its formation mechanism. The theoretical studies based on first-principle calculations predicted that the split interstitials $(N\text{-}As)_{As}$ and $(N\text{-}N)_{As}$ could act as two electron traps, localized at approximately 0.42 and 0.66 eV below the CBM of InGaAsN with a band gap of 1.04 eV, respectively [4]. Given the accuracy of the techniques of measurements, the calculated activation energy for $(N\text{-}As)_{As}$ is practically identical to that of E1. Moreover, the trapping density of E1 showed a dependence on the fluxes of N and As sources [5]. These two results expect that the split interstitial $(N\text{-}As)_{As}$ could be a possible origin of E1. However, this expectation requires more experiments and analysis to be confirmed.

In this work, electron irradiation and hydrogenation are used to study the evolution of E1 upon electron and proton irradiations. These experiments have a particular importance in the study of lattice defects in semiconductor materials. Indeed, the light mass of electrons, their high speed, and penetration power make from electron irradiation a powerful tool to change the structure of defects. Furthermore, hydrogenation showed a fascinating effect on dilute nitride semiconductors, especially the passivation of N-related defects and the limitation of their recombination activity.

2. Experimental Procedure

N-type GaAsN epi-layer was grown by CBE on n-type GaAs (2 cm x 2 cm), under a growth temperature and a pressure of 460 °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. A silane (SiH_4) source was used as n-type doping. The sample was sliced into several identical pieces to carry out hydrogenation and electron irradiation. For hydrogenation, H ions with multi-energy from 10 to 48 keV were implanted at room temperature into GaAsN films with peaks concentration of 5×10^{18} (HI1) and 1×10^{19} atom/cm^3 (HI2), respectively. For electron irradiation, two identical GaAsN pieces EI1 and EI2 were irradiated at room temperature with an energy of 2 MeV and with two different fluence doses of $\varphi_{EI1} = 9.0 \times 10^{14}$ ē/cm^2 and $\varphi_{EI2} = 9.0 \times 10^{15}$ ē/cm^2, respectively. The N concentrations of all samples were evaluated by X-ray diffraction method. The detail of fabrication of contacts could be found elsewhere [3]. The ionized donor concentration at room temperature was calculated using the capacitance-voltage method. The DLTS spectra were collected using a BIO-RAD (DL8000) digital DLTS system. The trapping density of each defect was adjusted according to the λ–effect factor [2].

3. Results and Discussion

DLTS spectra recorded on electron-irradiated, hydrogenated GaAsN samples along with as grown sample are shown in Fig.1. In the reference, only one main electron tap, E1, localized at 0.33 eV below the CBM of GaAsN, was recorded. The electronic properties of this recombination center have been published elsewhere [3]. Upon electron irradiation, E1 was also observed in DLTS spectra with a dependent trapping density N_T(E1) on irradiance fluencies.

N_T(E1) showed a little decrease with $\varphi_{EI1} = 9.0 \times 10^{14}$ ē/cm^2, which can be neglected by taking in account the accuracy of measurements. This means that this fluence is not effective, since electron irradiation often changes the properties defects. Increasing the fluence irradiance to $\varphi_{EI2} = 9.0 \times 10^{15}$ ē/cm^2, rises N_T(E1) with a factor of 1.68. The increase of N_T(E1) could be explained through two possible ways. First, the electron irradiation with enough fluence activates the E1-related atoms, which are electrically inactive. If E1 is the split interstitial $(N\text{-}As)_{As}$, the N and /or As atoms were transformed from their ideal sites to the interstitials N_i and As_i in the lower lattice and they interact with other N or As to form $(N\text{-}As)_{As}$. This scenario enhance the generation of N (V_N) and As (V_{As}) vacancies. However, the stability of N concentration upon irradiation and the smaller atomic size of N than of As make more probable the transformation of As atom than of N. Second, E1 involves interstitial or substitutional atoms in its atomic structure or a combination with other point defects, which are activated upon irradiation.

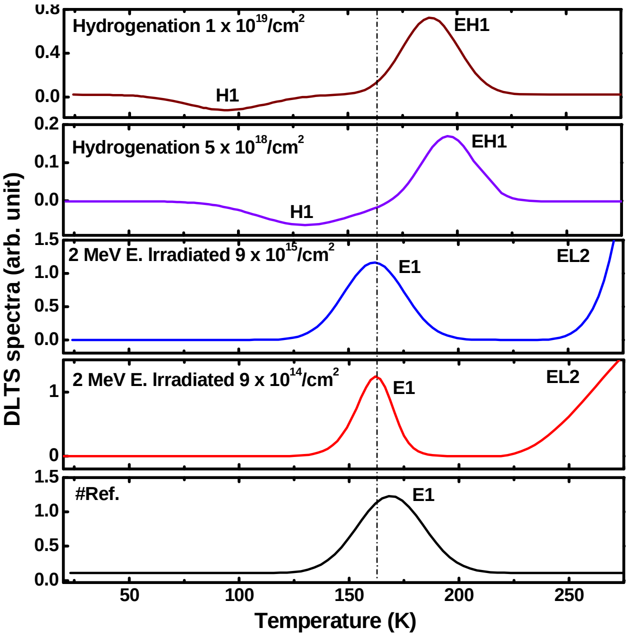

Fig. 1 DLTS spectra recorded on as grown, electron irradiated, and hydrogenated Si-GaAsN Schottky junctions with a reverse bias voltage of 3 V, a pulse voltage of 0 V, a rate-window time of 100 ms and a filling pulse width of 100 μs.

On another hand, E1 disappeared completely in the two hydrogenated samples, whereas two new lattice defects appeared. This result is totally in contrast to the effect of electron irradiation. Besides, it is an interesting perspective for the deactivation of the recombination activity of E1; however, the hydrogenation introduces damage to the semiconductor epilayer and degrades the performances of GaAsN based solar cells. The new defects are an electron, EH1, localized at average activation energy of 0.41 eV below the CBM of GaAsN and exhibits a capture cross section of 8.20×10^{-13} cm^2, and a hole trap H1, situated at 0.11 eV above the valence band maximum and has an average capture cross section of 8.20×10^{-17} cm^2. EH1 is commonly observed in GaAs, and is identified as EL5 on the basic of the classification by Martin et al. [6]. Its structure was extensively discussed and the common result deals with a complex defect free from impurities and dominated by As_i [7]. Concerning H1, we did not find a hole trap in the literature related to energy states in GaAs, which has the same electronic signature of H1. However, our previous studies on hole traps in GaAsN grown by CBE showed a hole trap, H2, which appears at the same peak temperature of H1 and nearly exhibits the same activation energy [3]. In addition, H2 was tentatively suggested to be related to N-H complex and in strong relationship with the high background doping in GaAsN [3]. Here, we suggest that H1 is responsible for the passivation of E1 and its formation could be the result of dissociation of N-As bound, followed by the formation of N-H complex. The free As atom could be involved in the formation of EL5.

4. Conclusions

In summary, the N-related nonradiative recombination center E1 in GaAsN grown by CBE increases in density with sufficient electron irradiation fluence; however, it is passivated upon hydrogenation. The passivation of E1 is suggested to be the result of the dissociation of N-As bond, followed by the formation of N-H complex, which plays the role of a hole trap. Therefore, the hydrogenation experimentation could be an essential perspective to limit the recombination activity of E1.

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