# N-H Defect Formation Mechanism in GaAsN Grown by Chemical Beam Epitaxy

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## 1. Introduction

(In)GaAsN is one of the candidate materials to realize the 4-junction solar cell with the conversion efficiency over 40 %. However, the concentration of the residual acceptor is too high  $(\sim 10^{17} \text{ cm}^{-3})$  to realize the expected value and it should be decreased. Theoretical analyses indicates that defects consisting of nitrogen-hydrogen bonding (N-H related defects) act as acceptors [1]. The existence of N-H bonds in GaAsN was experimentally shown by the Fourier transform infrared spectroscopy (FT-IR) [2]. However, those electrical properties, structures and formation mechanisms are still unknown. In this paper, the formation mechanisms of N-H structure were studied. Especially, the contribution of each source to the formation of the N-H defect was investigated. Since all sources used in the chemical beam epitaxy (CBE) have hydrogen atoms in the molecules, they are expected to form N-H defects. GaAsN films were grown by using the deuterated sources, in which all hydrogen was substituted with deuterium (D). Hence, the amounts of N-H and N-D in GaAsN were analyzed.

#### 2. Experimental

GaAsN films were grown by CBE on semi-insulating GaAs substrate. The Ga, As and N sources were triethylgallium [TEG, Ga(C2H5)3] or deuterated TEG trisdimethylaminoarsenic (D-TEG), [TDMAAs, As(N(CH3)2)3] and monomethylhydrazine [MMHy, N2H3(CH3)] or deuterated MMHy (D-MMHy). The concentrations of N-H and N-D were estimated by FT-IR. Since the atomic masses of H and D are smaller than those of Ga, As and N, the frequencies of the N-H and N-D are localized in both the real and frequency domains as local vibration modes (LVMs). The frequencies of N-H ( $v_H$ ) and N-D  $(v_D)$  vibrations in GaAs are known to have the ratio as  $v_{\rm H}/v_{\rm D} = 1.34$  [3]. The N-D peaks in FT-IR measurement were identified by using that ratio. The concentrations of the N-H-related defects are proportional to the integrated absorption intensities (IAs). The H and D concentrations in GaAsN film were obtained by secondary ion mass spectrometry (SIMS).

#### 3. Results and discussion

IR absorption spectra of the GaAsN films are shown in Fig. 1. When the normal sources were used, there were two peaks at 2952 and 3098 cm<sup>-1</sup>, which correspond to the N-H structures [2]. On the other hand, when deutertated MMHy

was used, new two peaks at 2206 and 2301 cm<sup>-1</sup> appeared. The ratio of 2952 to 2206 cm<sup>-1</sup> and 3098 to 2301 cm<sup>-1</sup> are  $\sim$ 1.34. This indicates that the peaks around 2200 cm<sup>-1</sup> are from N-D. Therefore, N-H in MMHy contributes to the formation of both N-H structures in GaAsN. However, since there were also N-H signals in the GaAsN film, -CH3 in MMHy is indicated to contribute to the formation of N-H in GaAsN. When GaAsN film was grown by deuterated TEG, the N-D peaks were not obtained. Hence, TEG does not contribute to the formation of the N-H. Then, the following results are obtained. (1) TEG does not contribute to the N-H formation, (2) the N-H is generated from H of N-H in MMHy, (3) -CH3 in TDMAAs and/or MMHy causes the N-H formation. Under the present growth condition, half of them are generated from H of N-H in MMHy and the other from -CH3 (Table in Fig. 1). The ratio of contribution of -CH3 to the formation of N-H in each defect and the IAs of N-H are shown in Fig. 2 as a function of growth temperature. Although the N-H concentration drastically decreased as increasing growth temperature, the ratio was almost constant independent of growth temperature. However, the ratio was different between each defect. The relationship between TDMAAs flow rate and IA of N-H and N-D is shown in Fig. 3. IAs at 2952 and 2206 cm<sup>-1</sup> increased as increasing the TDMAAs flow rate. On the other hand, IAs at 3098 and 2301 cm<sup>-1</sup> were constant with respect to the flow rate. This result suggests that -CH3 in TDMAAs contributes to the formation of defects at 2952(2206) cm<sup>-1</sup>. These results indicate that the formation mechanism of N-H at 3098 is different from that at 2952 cm<sup>-1</sup>.



Fig. 1 Infrared absorption spectra of GaAsN film. Upper table shows the contribution of N-H and -CH3 to N-H for-mation in each defect.



Fig.2. Ratio of contribution of  $-CH_3$  to the N-H formation (left axis) and integrated absorption intensity of N-H (right axis) as a function of growth temperature. The other contribution in N-H formation assumes to be N-H in MMHy.  $\mu$  is reduced mass.



Fig.3. Relationship between the TDMAAs flow rate and integrated absorption intensities of N-H and N-D.

#### 3. Conclusions

The formation mechanisms of H in N-H structure were studied by using deuterated sources. The N-H in MMHy and -CH3 in TDMAAs and/or MMHy molecules contributed to the formation of both N-H structures at 3098 and 2952 cm<sup>-1</sup> in GaAsN. On the other hand, H in the grown film did not come from TEGa. The dependence of the IAs of both N-H in FT-IR spectrum on the growth temperature and TDMAAs flow rate indicated that the formation mechanisms of them were different and that the -CH3 in TDMAAs contributed to the formation at 2952 cm<sup>-1</sup>.

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## References

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