Metal-Organic Molecular-Beam Epitaxy with a New Arsenic Precursor As[N(CH₃)₂]₃: Characterization of the Decomposition Processes and Growth of GaAs

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The decomposition process of a new alkyl-As source, tris-dimethylaminoarsenic (TDMAAs) was studied with RHEED and quadrupole-mass spectrometer (QMS) measurements together with a theoretical modeling. TDMAAs is decomposed efficiently on a GaAs surface and the Ga-stabilized surface turned to the As-one with very-low TDMAAs pressure. Grown GaAs films show small carbon-acceptor peak in PL measurements and were n-type. The origin of the n-type conductivity is discussed.

1. INTRODUCTION

There have been many trials to replace toxic high-pressured AsH₃ with alkyl-As sources. Although growth of high-purity films is reported with MOCVD, MOMBE growth of GaAs in high vacuum using alkyl-As sources usually tend to be p-type by the C incorporation as a shallow acceptor. In this paper, we propose the use of a new As precursor As[N(CH₃)₂]₃ (TDMAAs) in MOMBE, where As is bonded with N not directly with C as in other precursors. The decomposition processes of this precursor were studied with RHEED and QMS studies together with a theoretical modeling. The grown films show small carbon-acceptor peak in PL measurements and were n-type.

2. DECOMPOSITION MECHANISM OF TDMAAs

TDMAAs was supplied on a GaAs substrate surface without thermal cracking. Study of the QMS cracking patterns indicates that TDMAAs decomposes easily above 300° C. In the temperature range of $450\sim550^{\circ}$ C shown in Fig. 1, the Ga-stabilized surface turned to As-stabilized surface with the very-low TDMAAs pressure. This is a marked contrast to conventional As(C₂H₅)₃ with which the As-stabilized surface was never realized in high-vacuum system without precracking. The solid line is the fit of the theoretical modeling which assumes an intermediate species $As[N(CH_3)_2]$ in the decomposition process. This analysis indicated that the desorption rate of this intermediate species increases above 500°C as shown in Fig. 2. Figure 3 shows the substrate temperature dependence of the QMS transient response of the dominantly-observed intermediate species. The increase above 500°C indicates the increase of the desorption rate of this species, which is consistent with the above modeling of the steady state.



Fig. 1 Minimum TDMAAs pressure necessary to turn the initial Ga-stabilized surface to Asstabilized surface on (100) GaAs surface. The solid line is the fit with the modeling.

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3. GROWTH OF GaAs

The growth rate of GaAs using TEGa and TDMAAs was almost temperature independent in the range of 350~500°C as shown in Fig. 4. The growth rate was linear to the TEGa supply as shown in Fig. 5. The grown films



Fig. 2 Ratio of decomposition to the total reaction rate on the intermediate species $As[N(CH_3)_2]$. The decrease of the ratio above 500°C is due to the increase of the desorption from the surface.



Fig. 3(a) We propose a transient method of QMS to exclude the background signal and to study its dependence on the substrate temperature for estimating the activation energy in each decomposition process.



Fig. 3(b) Measured temperature dependence of $As[N(CH_3)]^+$ signal.

were n-type and the carrier concentration increases for the larger TEGa supply as shown in Fig. 6. The corresponding electron mobility at room temperature decreases from 2000 to 1000 cm²/Vsec with the increase of the TEGa supply.



Fig. 4 Dependence of growth rate of GaAs on substrate temperature.



Fig. 5 Dependence of growth rate of GaAs on TEGa pressure at substrate temperature of 450°C.



Fig. 6 GaAs grown at 450°C is n-type and the carrier concentration increases with TEGa supply.

Figure 7 shows the measured PL spectra at 40K. The carbon-acceptor peak at 830nm remains small for the wide range of the TEGa supply. This is consistent with the measured n-type conductivity. But the C-acceptor peak increases for the higher TEGa supply and the compensation was indicated.

The chemical reaction during the growth was studied with QMS by examining the spectral change with the TEGa supply. Clear spectral change was observed in the mass range shown in Fig. 8. The most distinct increase with the TEGa supply was observed on the intensities of 104 and 134 amu. As for the mass range of 101-106 and 120, AsNCH_n and AsC₂H_n are possible. But only AsC₂H_n is possible for 134 amu. Therefore 104 and 134 amu will correspond to As[C₂H₅] and As[C₂H₅]₂H, respectively. This indicates that the ethyl radicals decomposed from TEGa is bonded to As on the GaAs surface.







Fig. 8 During growth of GaAs, increase of the QMS intensity was observed in the mass range shown in this figure. Most distinct increase was observed on 104 amu $(As[C_2H_5])$ and 134 amu $(As[C_2H_5]_2H)$.

4. DISCUSSION AND CONCLUSIONS

The study of the TDMAAs decomposition on a GaAs surface indicated that TDMAAs is efficiently decomposed to supply As on the GaAs surface above 300°C. The n-type carrier concentrations in the grown GaAs films seem to be mainly due to the As-C bonds originating from the TEGa supply. SIMS measurements is now under study. Similar n-type conductivity was also observed in the growth of GaAs with TEGa and metallic

As in the temperature range below 550°C.¹⁾ Therefore higher purity will be possible by studying other alkyl-Ga sources. The promoted understanding of the decomposition processes in this study will serve for the low-temperature high-quality film growth with MOMBE using safe source materials.

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REFERENCE

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