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Nature and Formation of Non-Radiative Defects in GaNAs And InGaAsN

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ABSTRACT

The optically detected magnetic resonance (ODMR) technique has been employed to examine the nature and formation mechanism of non-radiative defects in GaNAs and InGaAsN. In both alloys, two defects were observed and were shown to be deep-level, non-radiative recombination centers. One of the defects has been identified as a complex involving an As_{Ga} antisite. These two defects gain more importance with increasing N composition up to 3%, presumably due to an increase in their concentration. With a further higher N composition, the defects start to lose importance in carrier recombination that is attributed to an increasingly important role of other new non-radiative channels introduced with a high N composition. On the other hand, effect of In composition up to 3% seems to be only marginal. Both defects were shown to be preferably introduced in the alloys during low-temperature growth by molecular beam epitaxy (MBE), but can be rather efficiently removed by post-growth rapid thermal annealing.

INTRODUCTION

N-containing III-V alloys, such as InGaAsN and GaNAs, are known to exhibit intriguing fundamental properties including giant bandgap bowing, that has attracted much interest in potential application for near infrared optoelectronic devices [1]. Unfortunately, radiative efficiency of the alloys has been shown to degrade rapidly with N incorporation largely attributed to the formation of competing non-radiative defects. However, very little is so far known about the nature of these defects and mechanism for their formation in the alloys. In this paper we shall provide some physical insight to grown-in non-radiative defects in GaNAs and InGaAsN derived from optically detected magnetic resonance (ODMR) studies.

EXPERIMENTAL DETAILS

Both GaNAs and InGaAsN alloys (either thick epilayers or multiple quantum well structures) studied in this work were grown by gas-source molecular-beam epitaxy (GS MBE). The thick GaNAs (typically 1100 Å) and InGaAsN epilayers (5000 Å) were grown at low temperatures, i.e. 420 °C and 440 °C, respectively. The N composition varies over the range 0-2.3% whereas the In composition is either 0% or 3%. The 7-period GaAs/GaNAs (200Å/70Å) multiple quantum wells (MQW), on the other hand, were grown either at a low temperature of 420 °C or at a high temperature of 580 °C. To study the effect of post-growth annealing, a piece of each low-temperature grown sample was treated by rapid thermal annealing (RTA) at 750-850 °C for 10-30 seconds in a flowing N_2 ambient.

ODMR experiments were performed at 2-5 K and with two microwave frequencies, i.e. X-band (9.28-GHz) and W-band (95-GHz), to cross-examine the contributions from various defects. The samples were illuminated by the 351 nm line of an Ar-ion laser or a tunable Ti:sapphire laser, to provide optical excitation above the GaAs bandgap or below the GaAs bandgap (resonant excitation of the GaNAs and InGaAsN alloys). The resulting photoluminescence (PL) was detected by a Ge-detector. The modulation of the PL intensity induced by the microwave radiation upon spin resonance was detected by a lock-in technique, giving rise to the ODMR signal [2].

EXPERIMENTAL RESULTS AND DISCUSSION

Nature of the non-radiative defects

Figure 1 shows typical ODMR spectra from the GaNAs and InGaAsN, obtained at two microwave frequencies. In both alloys, two non-radiative defects have been detected. The arguments for the non-radiative nature are: 1) both ODMR signals are negative in sign, meaning a spin-resonance induced decrease in PL intensity as a consequence of enhanced non-radiative recombination [2-3]; 2) the ODMR signal could be detected via any PL emissions of different origin in the alloys [2-3]. One of the ODMR signals (i.e. "1") exhibits resolved hyperfine structure, characteristic for a sizable interaction between an unpaired electron spin $S=1/2$ and a nuclear spin $I=3/2$. This provides unambiguous proof for the involvement of a defect atom with a

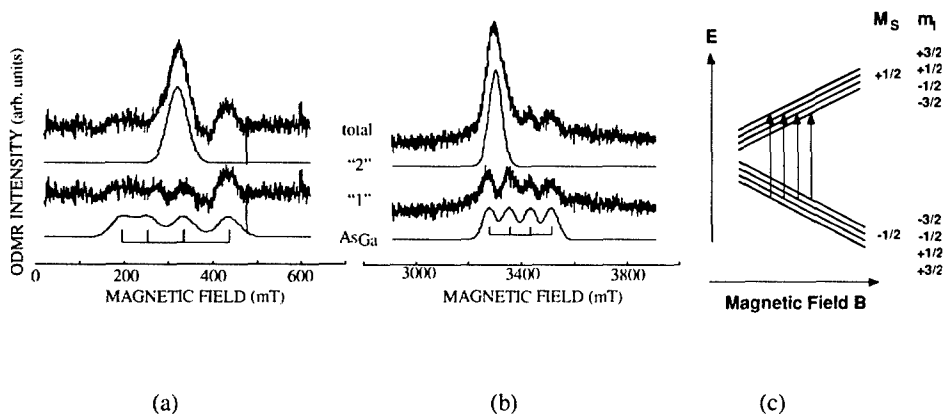


Figure 1. Typical ODMR spectra from the GaNAs and InGaAsN alloys, obtained at microwave frequencies of (a) 9.28 GHz and (b) 95 GHz. From the ODMR spectra, two ODMR signals (denoted by "1" and "2") can be deconvoluted. The former contains a four-line hyperfine structure, characteristic for a defect with an electronic spin $S=1/2$ ($M_S = -1/2, +1/2$) and a nuclear spin $I=3/2$ ($m_I = -3/2, -1/2, +1/2, +3/2$) as illustrated in (c) where the vertical arrows depict the spin resonance transitions. The ODMR signal "2" arises from a deep-level defect with $S=1/2$ and a g-factor of 2.03. The lowest curves are simulated ODMR spectra assuming the involvement of an As_{Ga} antisite complex.

nuclear spin $I=3/2$ in the defect core. The only plausible candidates in the studied materials are As or Ga atoms, both having $I=3/2$ from the isotope(s) with 100% natural abundance. Judging from the strength of the hyperfine interaction by comparing to earlier results from the parent binary compound GaAs and related ternary alloys [4-10], however, it can be concluded that the ODMR signal "1" arises most likely from a complex defect involving an As_{Ga} antisite. The other non-radiative defect has an effective electronic spin $S=1/2$ and gives rise to an isotropic ODMR signal "2". The exact chemical identity could not be obtained, unfortunately, due to the lack of resolved hyperfine structure. The g -value of the two defects was determined to be both around 2 [10], which strongly deviates from the known values for shallow-level impurities or defects and can therefore be regarded as support for the deep-level nature of the defects. Such type of defects is known to act as efficient recombination centers, very often non-radiative and harmful for optical quality of semiconductors. The results from the ODMR investigation under resonant excitation of the GaNAs and InGaAsN confirmed that both defects reside within the alloys.

Formation of the non-radiative defects

A series of GaNAs samples (both epilayers and MQW) were studied by ODMR to reveal the effect of N incorporation on the formation of the non-radiative defects. The results from the low temperature grown GaNAs MQW are presented in Fig.2. It is rather apparent that the ODMR

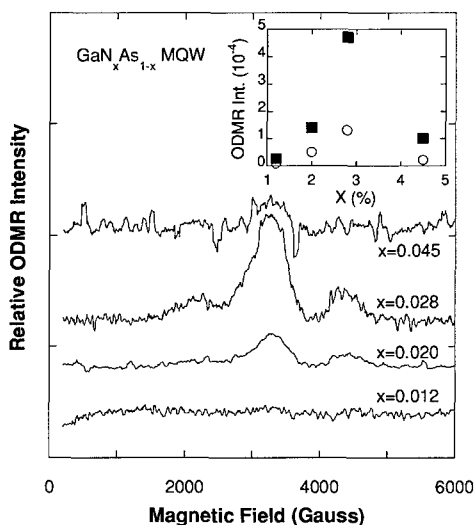


Figure 2. ODMR spectra at 9.28-GHz from the $\text{GaN}_x\text{As}_{1-x}$ MQW grown at 420 °C, as a function of N composition x . The normalized intensities of the ODMR signals "1" (the As_{Ga} antisite complex) and "2" vs. x are plotted in the inset, by the squares and circles, respectively.

intensity of both defects increases with increasing N composition up to about 3 %. This observation is believed to be largely due to an increase in the concentration of the corresponding defects. With a further increase in the N composition above 3%, the ODMR signals start to decrease. We believe that this is more likely due to a decreasing importance (rather than a decrease in the concentration) of the studied non-radiative defects, as a result of new, more efficient non-radiative defects being introduced with increasing N composition. The exact turning-point value of the N composition depends critically on growth conditions, such as growth temperature and strain.

The effect of In incorporation in the alloys on the formation of the studied non-radiative defects has, on the other hand, been found to be rather marginal at least within the range studied (<3%). This can be seen from Fig.3 where the ODMR spectrum from the InGaAsN epilayer is in a close comparison with that from the GaNAs epilayer.

In contrast, the effect of growth temperature on the formation of the studied defects is quite dramatic as shown in Fig.4. A significant reduction of the ODMR signals related to the studied non-radiative defects was observed in the MQW samples grown at 580 °C as compared to that with a similar N composition but grown at 420 °C. This provides strong evidence that these defects are preferably introduced during non-equilibrium growth at the lower temperature [11,12]. The studied non-radiative defects can be rather efficiently removed from the GaNAs and

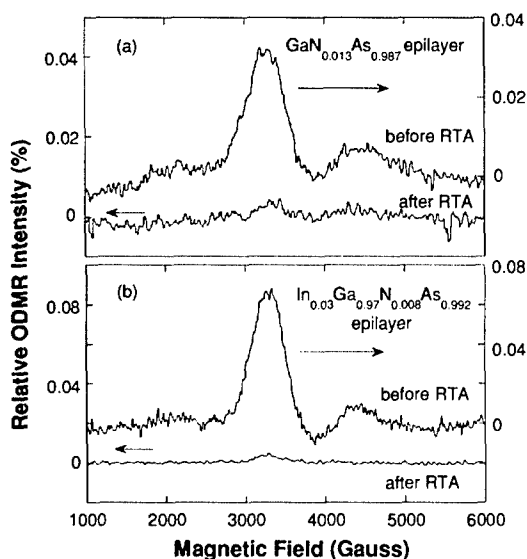


Figure 3. ODMR spectra at 9.28-GHz from the thick GaNAs and InGaAs epilayers grown at 420 °C and 440 °C, respectively. The ODMR intensity has been normalized to the total PL intensity monitored in the ODMR experiments, and is given in percentages. The ODMR spectra from both as-grown and after the post-growth RTA treatment are shown.

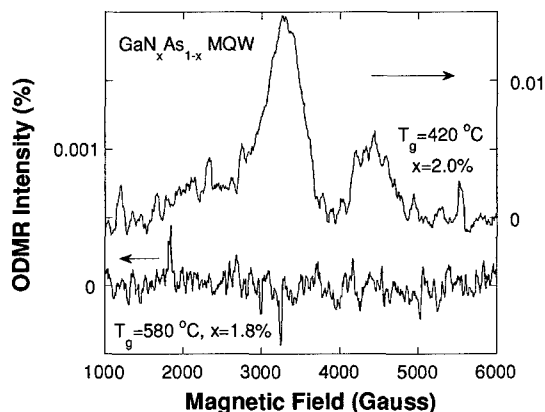


Figure 4. ODMR spectra at 9.28-GHz from two GaNAs MQW structures (with a close-match in N composition) grown at 420 °C and 580 °C, demonstrating the strong effect of growth temperature on the formation of the studied non-radiative defects.

InGaAsN alloys by RTA, see Fig.3. This is accompanied by a significant improvement in the optical quality of the alloys [13-16].

The observed anti-correlation between the ODMR intensity and the PL intensity of the alloys seems to point out that the studied defects may be among the important non-radiative recombination centers, at least within the ranges of the N compositions investigated in this work, that compete and degrade the radiative recombination processes crucial for optoelectronic device applications.

CONCLUSIONS

Two deep-level, non-radiative defects residing in the GaNAs and InGaAsN alloys were detected in the ODMR experiments. One of them exhibits characteristic hyperfine structure, arising from $S=1/2$ and $I=3/2$, which suggests a complex involving the As_{Ga} antisite as being the most likely candidate. We have shown that the introduction rate of these defects increases with decreasing growth temperature and with increasing N composition, leading to an increasingly important role in carrier recombination and thus in degrading optical quality of the material. With a further increase in nitrogen composition to 4.5% in the GaNAs/GaAs MQW structures grown at low temperature, the ODMR signals start to decrease probably due to the introduction of other competing defects that overshadow the role of the studied defects in carrier recombination. Post-growth rapid thermal annealing can significantly suppress the influence of the studied non-radiative defects in both GaNAs and InGaAsN alloys, accompanied by a drastic improvement in the efficiency of light emission.

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