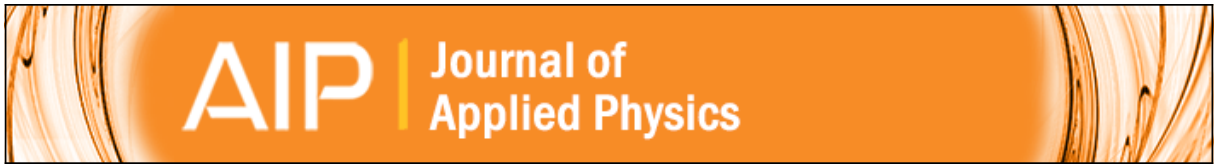

Epitaxially grown GaAsN random laser

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Epitaxially grown GaAsN random laser

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We have studied the photoluminescence properties of as-grown $GaAs_{1-x}N_x$ epitaxial layers grown on GaAs containing 0.6%, 1.77%, and 2.8% nitrogen. We found laser emission from thick ($d > 200$ nm) $GaAs_{0.972}N_{0.028}$ layers exhibiting the characteristic lasing properties of random lasers. This is unusual because random lasers have so far only been associated with highly disordered or random media. We believe that high gain in combination with structural inhomogeneities that are evident in these $GaAs_{0.972}N_{0.028}$ layers, can explain the random lasing in such epitaxial layers. [DOI: 10.1063/1.1568533]

I. INTRODUCTION

The dilute Ga(In)AsN alloy system exhibits some unique and unexpected characteristics that make it an important group of materials for optoelectronic applications.[1] Their most interesting attribute is the strong reduction of the band gap with increasing nitrogen content, a quality that makes these alloys a serious candidate for vertical cavity surface emitting lasers in the 1.3–1.55 μm spectral range. Despite some material challenges, the results are so far very encouraging. Several research groups have reported the fabrication of laser diodes with excellent characteristics.[2–4]

In this publication, we report another interesting and potentially significant observation related to this group of materials. We have observed efficient optically pumped laser emission from $GaAs_{0.972}N_{0.028}$ layers grown on GaAs by metalorganic chemical vapor deposition (MOCVD), layers that do not contain any obvious or apparent optical cavities. Based on the experimental evidence to be described in this paper, we believe the lasing is due to the formation of a random laser in this epitaxial material. Random lasers have been associated with disordered (or random) media with gain, in which recurrent optical scattering provides the coherent feedback.[5–7] Random lasing was observed, for example, in some semiconductor powders, polycrystalline layers, and laser dye solutions containing nanoparticles.[5–9] While epitaxial GaAsN is known to be a high gain medium, random lasing can only occur if sufficient light scattering is also present within the layer. Since we only observed random lasing in samples containing 2.8% nitrogen when the layer thickness was above the critical layer thickness, we propose that random lasing in these layers is due to optical scattering by structural inhomogeneities.

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II. SAMPLES AND EXPERIMENTAL SETUP

We have used low temperature continuous wave (cw) and pulsed photoluminescence (PL) measurements, as well as femtosecond time resolved PL spectroscopy to study the light emission from a range of GaAsN alloys containing 0.6%, 1.77%, and 2.8% nitrogen. The samples were grown on semi-insulating GaAs (100) substrate by low-pressure MOCVD. Trimethylgallium (TMGa) and trimethyindium (TMIn) were used as group III source, arsine (AsH_3), phosphine (PH_3) and 1.1-dimethylhydrazine (DMHy) were used as group V sources. GaAsN epilayers between 200 and 800 nm thick were grown at 76 Torr on a 200 nm undoped GaAs buffer. The nitrogen composition was estimated from double-crystal x-ray rocking curves using (004) reflection. Rapid thermal annealing was performed on all the GaAsN samples at 750 °C for 30 s using GaAs proximity capping in N_2 ambient. Samples were cut into 4×4 mm^2 platelets for the optical and structural examinations. The structural properties of the layers were investigated using x-ray diffraction, cross-sectional transmission electron microscopy (XTEM), and scanning electron microscopy (SEM).

Low temperature cw PL was excited by the 514 nm line of an Ar ion laser, dispersed with a 0.33 m monochromator. The pulsed luminescence measurements were excited by a Q-switched frequency doubled Nd:YAG laser ($\lambda=532$ nm, 5 kHz repetition and 1 μs pulse width) and dispersed with a 0.75 m monochromator and detected by a cooled Ge detector. The time resolved PL measurements were based on the PL upconversion technique using an Ar ion laser pumped femtosecond self-mode locked Ti:sapphire laser. The Ti: sapphire laser was tunable between 750 and 900 nm, the pulse width was 80 fs, the repetition rate 85 MHz, and the output power was 180 mW at $\lambda=780$ nm. The samples were mounted in a variable temperature, closed cycle, He cryostat,

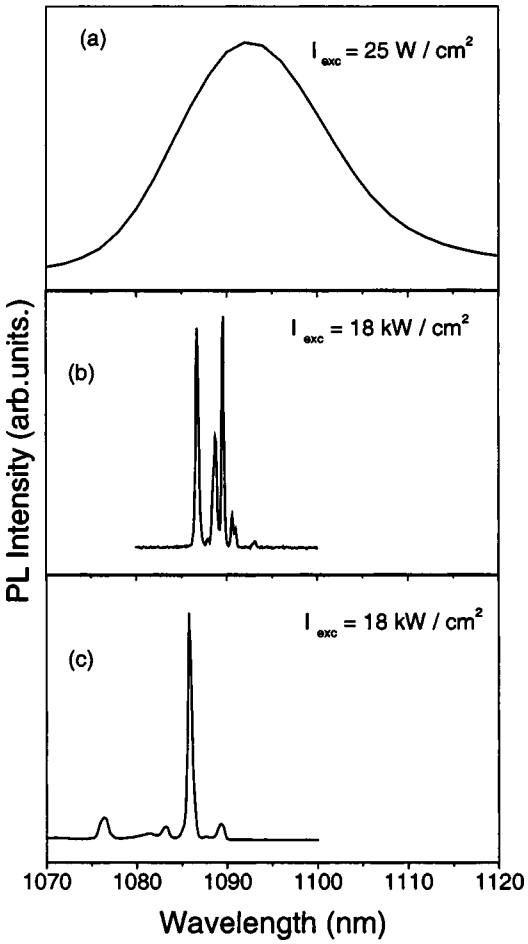

FIG. 1. (a) Low temperature ($T=10$ K) photoluminescence spectrum of $GaAs_{0.972}N_{0.028}$ layers grown on GaAs below threshold excitation. (b), (c) The spectra at two different points on the sample above threshold.

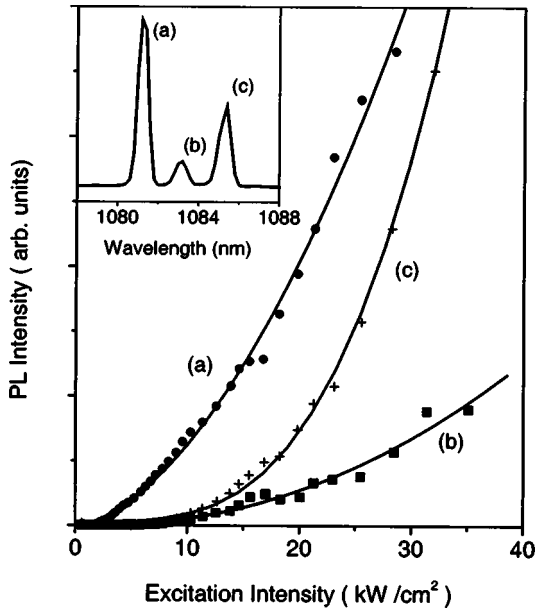

FIG. 2. Low temperature photoluminescence of a $GaAs_{0.972}N_{0.028}$ layer as a function of the excitation intensity for three laser modes, as shown in the insert, indicating the different threshold behavior for each of the modes of the emission.

the temperature of which could be varied between 10 K and room temperature.

III. RESULTS AND DISCUSSION

At low excitation levels and low temperatures ($T=10$ K) the PL spectra displayed the familiar features of this alloy system: an asymmetric PL line shape with an exponential low-energy tail, as shown in Fig. 1(a).[10] The PL peak energy exhibits a strong shift to lower energies with increasing nitrogen content, which has been shown to correspond to the decrease in the band gap energy of this alloy due to the bowing effect. These characteristics have been well documented in the past.[11–13] Under higher intensity pulsed excitation, the PL from the samples containing 2.8% nitrogen and layer thicknesses greater than 200 nm, changed radically. Above a certain threshold excitation intensity, several narrow, laser-like peaks appeared. The spectra measured at two different locations on the sample are shown in Figs. 1(b) and 1(c). Unlike conventional laser emission from semiconductor samples, the mode spacings of the lines were broad, irregular, and changed with excitation position and intensity. Moreover, unlike typical laser emission from semiconductor samples of similar dimensions (4×4 mm^2) and gain bandwidth, in these samples only few modes (<10) could be observed, even at the highest excitation intensities. This is consistent with optical cavities embedded in the samples that are much smaller than the actual dimensions of the samples.

The emission intensity of each mode displayed distinctive threshold behavior, as shown in Fig. 2, indicating that lasing does indeed occur. The inset of Fig. 2 shows the three modes for which the threshold was measured. The lasing threshold was found to depend on the position of excitation on the sample, with the thresholds varying between 0.1 and 10 kW/cm^2 for the various modes at different positions on a given sample. These characteristics reveal that the lasing modes are related to different optical cavities.

Another essential feature of these laser spectra is that they could be detected from any angle of observation. We have detected spectral similar to those shown in Figs. 1(b) and 1(c), irrespective of the direction of observation. In Fig. 3, we display the emission spectra collected in the three orthogonal directions. Although the spectral positions of the lines and the mode separations fluctuated as a function of angle of observation, laser-like emission was observed from all directions. It is also interesting to note that the emission emerging from the edges of the samples was linearly polarized in a direction perpendicular to the sample surface but was unpolarized when emerging perpendicular to the top surface of the sample. The laser-like emission (line spectra) disappeared upon heating the sample to temperatures above approximately $T=100$ K, and the broad, spontaneous emission remained.

A further distinctive feature of the laser emission from these samples is the excitation volume dependence of the threshold intensity. For example, at a given point on the sample and a given excitation intensity of 1 kW/cm^2, lasing occurred when the excitation spot size was *larger* than

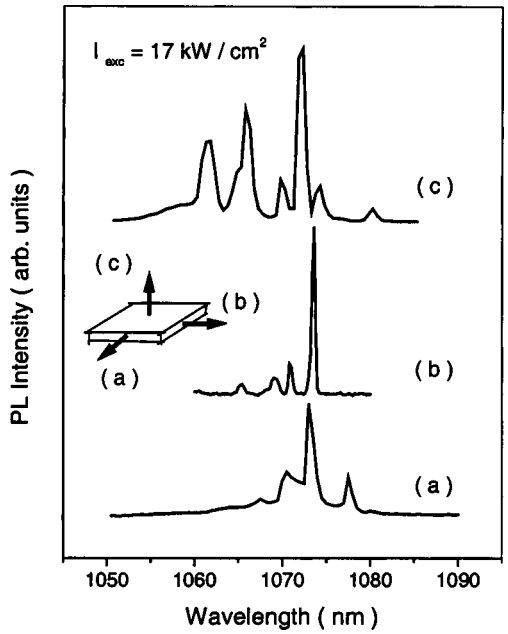

FIG. 3. Laser-like emission with similar mode structure detected from three orthogonal directions for a $GaAs_{0.972}N_{0.028}$ layer grown on GaAs.

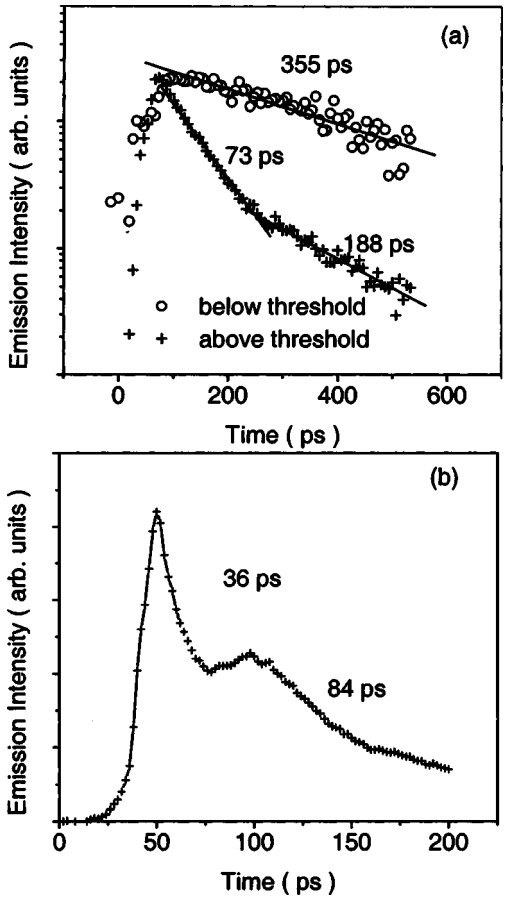

FIG. 4. (a) Low temperature photoluminescence decay as a function of time measured below and above threshold excitation, demonstrating the reduction of the decay time constant above lasing. (b) Photoluminescence relaxation oscillations observed at some positions on the sample.

2500 μm^2 but disappeared when the spot size was reduced below this value.

We have also measured the time evolution of the luminescence using PL upconversion measurements. The high temporal resolution of our measurements allowed us to detect the rise time and the decay time of the luminescence with great precision.[14] The rise time in these samples was found to be approximately 30 ps both below and above threshold, a value that is similar to the rise times measured for other GaAsN samples.[14] The decay time, on the other hand, was significantly reduced above the threshold excitation intensity. While below threshold the decay time was of the order of 400 ps or more, above threshold it was reduced to between 20 and 80 ps depending on the sample position and excitation intensity. The decay often indicated two decay processes with differing decay constants, as shown in Fig. 4(a). In addition, at some positions on the sample, relaxation oscillations were observed, as can be seen in Fig. 4(b). The significant reduction in the luminescence decay time above threshold is consistent with the nature of (stimulated) emission in these samples, and the variation of the decay constant with position can be an indication of the existence of different cavities with differing losses (gain).

Overall, these results are very similar to those reported recently by a number of research groups describing the observation of laser emission from random or disordered media of various types. For example, Cao *et al.*[6] reported laser emission from polycrystalline ZnO films, and ZnO and GaN powders, samples that did not contain any obvious optical cavities with mirrors. Above threshold, the spectra could be observed in all angular directions, and varied with excitation level and excitation volume.[6] Soukoulis *et al.*[15] reported relaxation oscillations and position-dependent decay times in ZnO random lasers, very similar to those found in our experiments. These results are also characteristic of other ''random lasers,'' such as those first reported by Lawandy *et al.*[8] on the emission from laser dye solutions containing microparticles, Frolov *et al.*[7] on organic dye-doped gel films, and opal crystals saturated with polymer and laser dye solutions. In all these systems, the key to laser emission is the existence of a high gain medium and efficient light scattering within the sample to produce the necessary coherent feedback.

Our experimental results indicate that the underlying mechanism for the laser-like emission from thick $GaAs_{0.972}N_{0.028}$ epitaxial layers is similar to that found in random lasers. While gain is known to be present in high quality GaAsN layers, the existence of efficient light scattering is uncertain. A number of previous studies have called attention to the inherent inhomogeneous structural properties of this group of alloys.[16,17] For example, optical absorption and Raman studies have suggested that alloy ordering may take place in these alloys, while other studies indicate the existence of nitrogen enriched GaAsN clusters due to the significant differences in atomic size and electronegativity between As and N.[16,17] In addition, for GaAsN layers with thickness above the critical layer thickness, strain relaxation occurs and misfit dislocations develop.[18] Although the exact value of the critical layer thickness is still a mater of controversy, our 500-nm-thick, 2.8% nitrogen containing layers are well above all estimates for the critical thickness for this

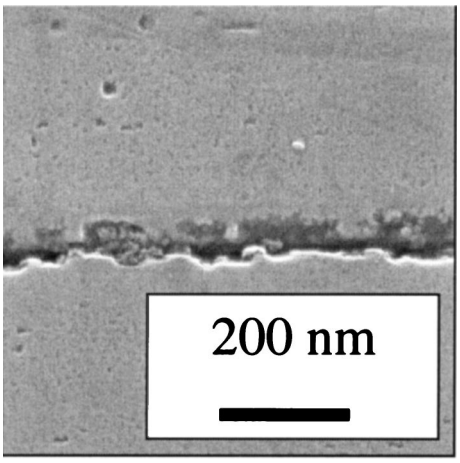

FIG. 5. SEM image of a 500-nm-thick $GaAs_{0.972}N_{0.028}$ layer, illustrating the type of random microscopic cracks that could be observed on the sample surface.

alloy. Such structural inhomogeneities could well explain light scattering in this group of alloys.

We have investigated the structural properties our samples using x-ray diffraction, XTEM, and SEM. For layer thicknesses below approximately 200 nm, the x-ray measurements indicated high homogeneity and single-phase material for all the samples (GaAsN alloys containing 0.6%, 1.77%, and 2.8% nitrogen). However, for the samples with 2.8% nitrogen, as the layer thickness above 200 nm, random microscopic cracks were observed on the sample surface, as shown in Fig. 5. The image taken using field emission SEM (Hitachi S900 FESEM) shows rough cracks with width of the order of 10–50 nm. These types of microcracks appeared randomly across these samples, often as two mutually perpendicular sets of parallel lines, and are the result of the likely strain relaxation and misfit dislocations. There could also be other sources of light scattering in this alloy system. Gwo *et al.*,[16] for example, found evidence for nanocrystals embedded at the interface between GaAsN and GaAs, which they assumed were due to the segregation of GaN in this alloy. These obvious structural inhomogeneities could very well explain the efficient light scattering necessary for random lasing.

IV. CONCLUSION

In summary, we have grown $GaAs_{1-x}N_x$ epitaxial layers on GaAs, containing 0.6%, 1.77%, and 2.8% nitrogen. We found that thick $GaAs_{0.972}N_{0.028}$ layers exhibited the characteristic emission properties of random lasers. We believe that structural inhomogeneities that are evident in the SEM images of these layers can explain the efficient light scattering necessary for random lasing.

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