PHYSICAL REVIEW B **73**, 195112 (2006)

Photon localization and lasing in disordered GaN_xAs_{1-x} optical superlattices

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(Received 1 March 2006; revised manuscript received 18 April 2006; published 15 May 2006)

We propose an approach to fabricate a disordered optical superlattice using microcracking faces in GaN_xAs_{1-x} epilayers. Laser action is observed and the emission exhibits random laser behaviors. A transfer-matrix simulation suggests photon localization occurs at the lasing modes.

DOI: 10.1103/PhysRevB.73.195112 PACS number(s): 78.45.+h, 78.67.Pt, 78.55.−m, 78.66.−w

Letokhov first theoretically discussed the laser action in random media.[1] Since then, extensive theoretical and experimental studies have been focused on the light propagation in disordered gain media due to the interest in random laser action.[2–6] Random lasers have been realized from various media, such as semiconductors powders,[7] laser dye solution containing microparticles,[8] and organic dye-doped gel films.[9] In all these systems, the key factor for laser emission is the existence of a high gain medium and efficient light scattering within it to produce the necessary coherent feedback. Also light amplification and localization behaviors in disordered layered media have been studied using the transfer-matrix and analytical approaches.[2–5] An inverted opal structure with randomly stacking faults has been presented, showing that the stacking faults can give rise to localized states within the absolute frequency gap of light in disordered medium.[10] Usually defect states are introduced in designed photon crystal for studying photon localization and laser action.[11,12] In this case, the amplified emission could be observed by pumping the defect states.[13] It is therefore interesting to realize the lasing emission experimentally from the disordered layered gain media and study the photon localization due to a possible low threshold.[6]

In this paper, we present a systematic study of a GaN_xAs_{1-x} disordered gain optical superlattice (SL). A random laser based on photon localization has been observed. A disordered optical SL was fabricated by randomly distributed parallel microcracking faces on a GaN_xAs_{1-x} epilayer, naturally formed due to the larger lattice mismatch between the epilayer and substrate. GaN_xAs_{1-x} samples were grown by metal-organic chemical-vapor deposition (MOCVD). The growth details were presented elsewhere.[14] The structural properties were analyzed using double-crystal x-ray diffraction and scanning electron microscopy (SEM). Three GaN_xAs_{1-x} epilayers with $x=0.6\%$ (sample A), 1.77% (sample B), and 2.8% (sample C) show high homogeneity in a single phase when the film thickness is below 200 nm. However, random microscopic cracking lines are observed on the XY surface plane of the sample C when the GaNAs layer is thicker than ~200 nm as a result of tensile strain relaxation, as indicated in Fig. 1(a), where a cracking line with the width of ~50 nm is also shown. The microcracking lines appear randomly across the 4×4 mm film studied in the experiment. They show a set of parallel faces along the Y direction and the depth of the cracks is of the order of the film thickness ~500 nm. The distance between the faces is a random distribution of ~1–5 μm, being of the same order as the lasing emission wavelength. Therefore, the microcracks in sample C may be considered as randomly stacked GaN_xAs_{1-x} slabs along the X direction with air gaps, as shown schematically in Fig. 1(b). The substrate of GaAs has a thickness of 0.5 mm, the top layer of GaNAs has a thickness of 500 nm, and the air gap of strip lines is ~50 nm in width. Each GaN_xAs_{1-x} slab is an amplifying medium with a dielectric constant of 12. In the Y and Z directions the medium is taken to be uniform due to the fact that the refractive indices of GaAs and GaN_xAs_{1-x} are almost the same as the concentration of nitrogen is low in the GaN_xAs_{1-x} alloy. Therefore, the GaN_xAs_{1-x} film of sample C may be considered to be a disordered SL along the X direction. In the experiment the pump light is incident normal to the sample surface along the Z direction, and the emission signals are collected from $-Z$, X, and Y directions in three configurations, as indicated in Fig. 1(b).

Light emission from the samples A, B, and C has been studied using low temperature pulsed photoluminescence (PL) measurements. The pulsed luminescence was excited by

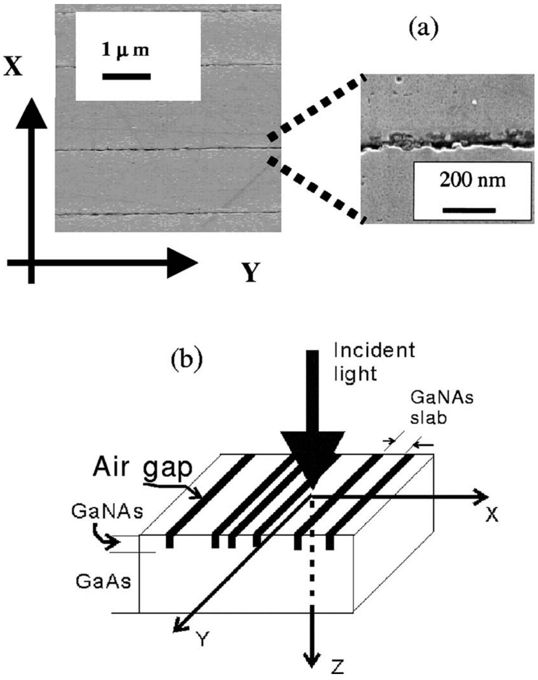

FIG. 1. (a) SEM image of sample C, illustrating the randomly distributed microcracking lines parallel to the Y axis. The right side is the amplified SEM image of one crack line. (b) Schematic diagram of the investigated GaN_xAs_{1-x} film structure grown on GaAs and the defined coordinates.

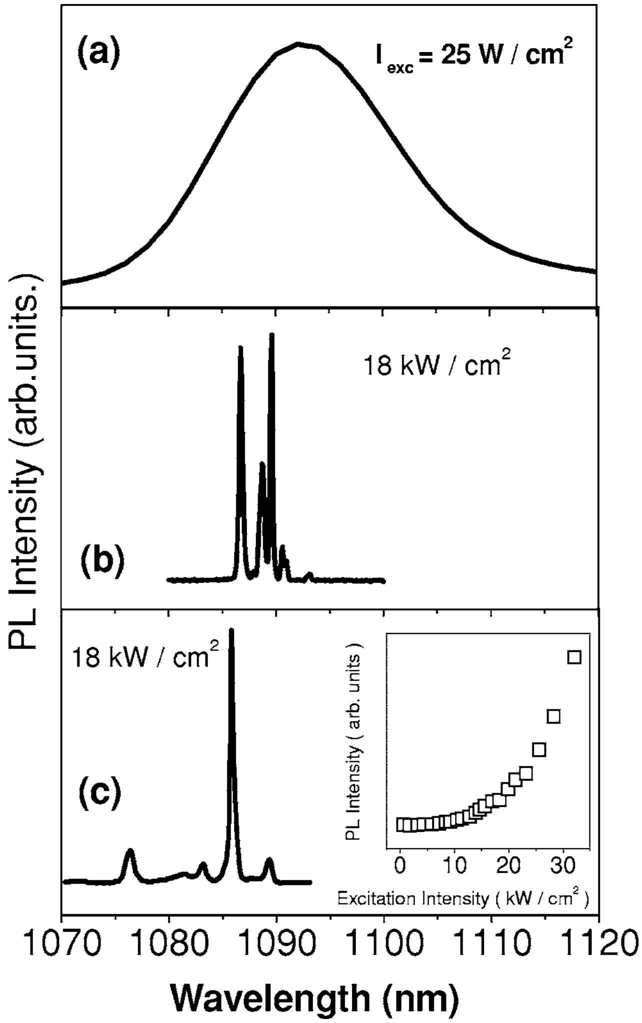

FIG. 2. Low temperature photoluminescence spectrum of sample C measured below the threshold excitation intensity (a) and above the threshold (b) and (c). PL spectra (b) and (c) were taken at two different positions of the excitation spot on the film. The inset in (c) displays the dependence of the PL intensity on the excitation intensity, showing a threshold behavior.

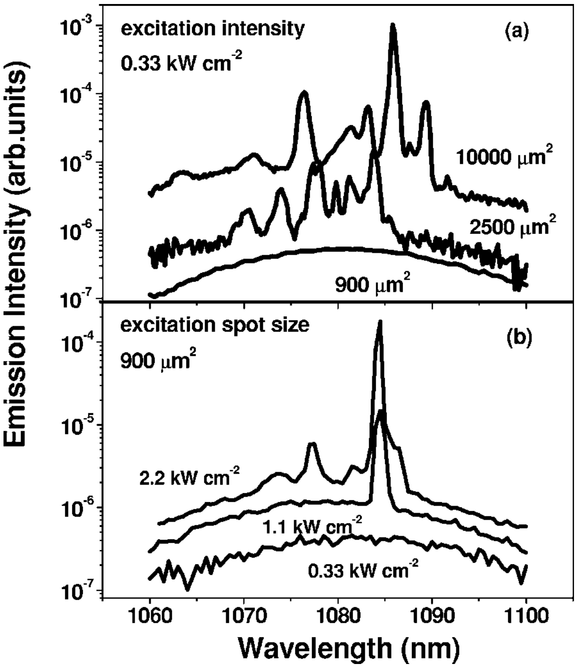

FIG. 3. (a) Emission intensity as a function of the excitation spot size measured at a fixed excitation intensity of 0.33 kW cm^{-2}. (b) Emission intensity as a function of the excitation intensity at a fixed excitation spot size of 900 μm^2.

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 then detected by a cooled Ge detector. At a low excitation level of 25 W/cm^2 and a low temperature (T=10 K) the broad PL spectrum of sample C displays emission features typically for a GaN$_x$As$_{1-x}$ alloy,[15] as shown in Fig. 2(a), where the emission signal is collected from the $-Z$ direction. Under higher intensity pulsed excitation, the PL spectrum from the sample C changes radically, but the PL spectra of samples A and B do not change so much. Above certain threshold excitation intensity, several narrow laser linelike peaks appear for sample C, as shown in Fig. 3(b) and 3(c). The abrupt change in the output power (PL integrated intensity) with the increasing pump intensity shown in the inset of Fig. 2(c) is characteristic of the lasing transition. Note that at different excitation spots emission spectra display different modes with different lasing thresholds. In the experiment the pump spot size with a local population inversion is $\sim 100 \times 100$ μm^2 around the center of the sample, which is smaller than the actual size of the sample dimension $\sim 4 \times 4$ mm^2, meaning that the optical cavities should be embedded inside the sample. Compared with the PL spectra of the samples A and B, the laser emission only happens in the sample C, which strongly suggests that randomly distributed cracking faces play an important role in lasing emission and photon localization.

A distinctive feature of laser emission from the sample C is an excitation spot size dependence of the threshold intensity. In the experiment edge emission signal is collected along the X direction with a strip pump beam incident on an area of $20 \times L(x)$ μm^2 at a constant excitation intensity, where $L(x)$ increases along the $-X$ direction. At a given excitation position and excitation intensity of 0.33 kW/cm^2, lasing occurs when the excitation spot size is larger than ~ 2500 μm^2 but disappears when the spot size is reduced below this value to a value such as 900 μm^2, as shown in Fig. 3(a). Emission modes of Fig. 3(a) show a spot size dependence. This means that lasing is initiated when the gain region spatially overlaps the peaks of long-lived localized modes.[6] Laser emission appears again for a spot size of 900 μm^2 when the excitation intensity increases, as shown in Fig. 3(b) at a pump intensity of 1.1 kW cm^{-2}. A new laser emission peak appears when the intensity is further increased as indicated for a pump intensity of 2.2 kW cm^{-2}. Thus lasing also occurs at shorter-lived modes when the pump intensity is increased. For example, lasing in one or more than one modes at an excitation intensity of 1.1 kW cm^{-2} or 2.2 kW cm^{-2} are observed as indicated in Fig. 3(b). The photon localized mode and the excitation spot size dependence of the lasing have strongly suggested that random laser emission has been observed in GaNAs film with cracking faces.[6]

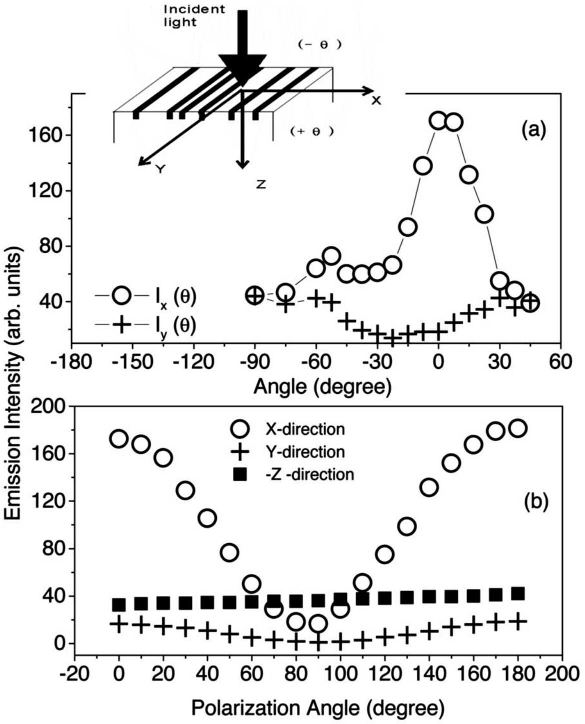

FIG. 4. (a) Angular distribution of the emission intensity $I_x(\theta)$ (open circle) and $I_y(\theta)$ (cross) at the fixed pump intensity of 18 kW/cm^2. The defined coordinates are shown in the inset of (a). (b) Polarization angle dependence of the emission intensity measured along the X axis (open circle), Y axis (cross), and $-Z$ direction (solid square).

These properties also demonstrate that lasing action is only relevant to the cracking faces in the GaN$_x$As$_{1-x}$ film, not dependent on the vertical cavity of Z direction formed by the top and bottom surfaces.

In order to study the influence of parallel cracking faces on the spatial distribution of laser emission and the mechanism of photon localization, we have measured the emission intensity as a function of orientation, such as emission intensity $I_x(\theta)$ and $I_y(\theta)$ measured from 45° to −90°, as indicated by the geometric configuration in the inset of Fig. 4, where $\pm\theta$ represents the angle between the direction of light collection and the X or Y axis. The emission intensity as a function of θ shown in Fig. 4(a) clearly displays a maximum of emission along the X direction and a weak emission intensity along the Y and $-Z$ directions. This behavior suggests that light is transmitted through and reflected from the barriers of the cracking faces into GaNAs slabs as would be the case in the Fabry-Pérot cavities along the X direction. The width of each Fabry-Pérot slab is about several micrometers with a dielectric constant of 12. The air gaps are about 50 nm in width with a dielectric constant of 1. Thus, there is a large dielectric difference along the X direction, which will effectively enhance the localization of photons. However, due to the narrow barrier width of ~50 nm between GaN$_x$As$_{1-x}$ slabs there is a strong coupling between the neighboring GaN$_x$As$_{1-x}$ slabs. The calculated reflectivity and transmissivity, are 17% and 83% at a lasing wavelength of 1.088 μm, respectively, when light tunnels across a barrier. Therefore, the light needs to travel across many interfaces to form multiple reflections before the light is localized. The medium can be described as a disordered distributed feedback random cavity. This arrangement is consistent with the observation of the spot size and intensity dependence of the lasing threshold. The emission observed from other directions is attributed to the light scattering off the X direction by rough interfaces due to Rayleigh scattering.[16] This is confirmed by the polarization measurement of emission intensity, as indicated in Fig. 4(b). Linearly polarized emissions with the polarization along the Z direction in a TM mode are observed in the X and Y directions. However, the lasing emission in the $-Z$ direction has not any polarization as expected in a TM mode, i.e., the observed emission comes from the light scattering on rough interfaces when the light travels in a GaN$_x$As$_{1-x}$ film along the $\pm X$ direction. Here a TM mode emission is due to the radiative recombination between electrons and light holes because the tensile strain in bulk GaNAs alloy lifts the degeneracy between heavy- and light-hole states at the Γ point.

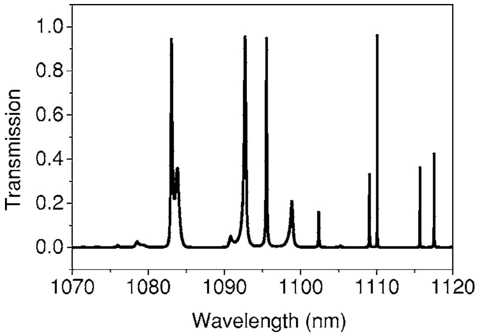

FIG. 5. Calculated transmission spectrum as a function of wavelength for an effective Fabry-Pérot cavity structure with distributed feedback and in absence of gain.

When the sample is pumped using a laser wavelength of 532 nm from the top side of the GaN$_x$As$_{1-x}$ film, the photogeneration electrons and holes only occur in the GaN$_x$As$_{1-x}$ layer due to the large absorption coefficient of ~8000 cm^{-1}. Hot electrons and holes will fast relax to the band edge of GaN$_x$As$_{1-x}$ within a time of picosecond order and then recombine giving rise to the emission as indicated in Fig. 1 with a decay time of ~355 ps below the threshold and ~30 ps above the threshold[14] Usually, the radiative recombination from the sample travels in all directions in three dimensions. However, the spot size dependence of the lasing threshold gives strong evidence that light is multiply reflected and amplified when light travels along the $\pm X$ direction. Laser emission occurs in the X direction when the longest lived modes in the X direction overlap the gain region. In other directions modes spread and do not overlap the gain region.[6] Effective Fabry-Pérot cavities with distributed feedback are formed inside the sample without any need to have edge reflections in the sample, as shown by a recent theoretical analysis and transfer matrix numerical simulations.[2–5]

As analyzed above, the radiation of the lasing modes is basically confined in the X direction while the polarization is in the Z direction. We can simulate our system by a transfer-matrix approach. In the absence of gain, the calculated transmission spectrum is shown in Fig. 5 for a stack of 40 slabs of random thicknesses between 1 and 5 μm. The mean linewidth of the spectra modes, $\delta\lambda$, and mean spacing between modes, $\Delta\lambda$, are 0.24 and 1.7 nm, respectively. This gives the Thouless number $\delta = \delta\gamma / \delta\nu \approx 0.14 < 1$. This seems to suggest that the light localization occurs at the spectra modes.[6,17–20]

In conclusion, photon localization and lasing is realized in a system based on randomly distributed parallel microcracking faces on a GaN_xAs_{1-x} film. The cracking faces form a random array of effective Fabry-Pérot cavities filled with gain material. Our results will help to provide guidance in the experimental exploration for photon localization and random lasing in epitaxial semiconductor materials.

The author would like to thank Z. Q. Zhang at HKUST for enlightening discussions. The national Nature Science Foundations of China under Grants No. 10474095 and No. 10334040 support this work.

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