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Photoluminescence investigation of high quality GaAs_{1-x}Bi_x on GaAs

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Photoluminescence (PL) of GaAs_{0.97}Bi_{0.03} alloy was measured over a wide range of temperatures and excitation powers. Room temperature PL with peak wavelength of 1038 nm and full-width-half-maximum of 75 meV was observed which is relatively low for this composition. The improved quality is believed due to reduced alloy fluctuations by growing at relatively high temperature. The temperature dependence of PL peak energy indicated significant exciton localization at low temperatures. Furthermore, the band gap temperature dependence was found to be weaker than GaAs. An analysis of dominant carrier recombination mechanism(s) was also carried out indicating that radiative recombination is dominant at low temperature. © 2011 American Institute of Physics. [doi:10.1063/1.3565244]

Bismuth (Bi) containing semiconductors have attracted increasing interest in recent years due to the large band gap reduction possible with small concentrations of Bismuth, thus promising for long wavelength devices.¹⁻³ Introducing Bi into GaAs reduces the semiconductor band gap by 88 meV/% Bi, which is much larger than the 16 meV/% and 21 meV/% of In and Sb, respectively, and only lower than the 125 meV/% N alloying.² Furthermore, it has been suggested that incorporation of Bi increases the valence band maximum due to a valence band anticrossing interaction.^{4,5} Concentrations of up to 4.5% have been shown to improve the optical quality of this low growth temperature alloy.⁶ This is in contrast with N alloying which reduces the conduction band minimum⁵ and degrades optical quality. GaAs_{1-x}Bi_x also has a giant spin-orbit bowing making it potentially suitable for spintronic applications.⁷

While Bi containing alloys hold great promise, wafer growth of this material is not straight forward. Bi tends to surface segregate during film growth and form droplets on the surface under conventional GaAs growth conditions.⁸ Recently, GaAs_{1-x}Bi_x with Bi content up to 0.11 was grown by molecular beam epitaxy (MBE).^{6,9} However, the photoluminescence (PL) data reported to date have significantly larger full-width-half-maximum (FWHM) than more established InGaAs,^{2,6,10} suggesting significant alloy composition fluctuations in the GaAs_{1-x}Bi_x samples. GaAs_{1-x}Bi_x also has a smaller temperature coefficient than GaAs ranging from 0.1 to 0.4 meV/K (Refs. 11–13) for Bi content between 0.019 and 0.05. In this letter, we report a detailed study of PL measured from our GaAs_{1-x}Bi_x samples with particular attention to $x=0.03$. The study focuses on the band gap temperature dependence, PL FWHM, and dominant carrier recombination mechanism(s) at different temperatures and excitation powers.

Growth was carried out in an Omicron MBE-scanning tunneling microscopy (STM) system with gate-valve isolated MBE and variable temperature STM chambers. The semi-

insulating on-axis GaAs (100) substrate used was cleaved into 11.0 × 3.5 mm² pieces (to fit into the substrate holder). A piece was then dipped in HCl to thin the native oxide, rinsed with de-ionized water and wiped with IPA, before being mounted on the substrate holder. After degassing at 400 °C, oxide on the substrate was removed via Ga-assisted oxide desorption, providing a flat growth surface and eliminating the requirement for thick buffer layers.¹⁴

A 80 nm GaAs buffer layer was grown at 585 °C using 20:1 As:Ga beam equivalent pressure ratio (BEPR). The active region, a 160 nm GaAs_{1-x}Bi_x layer was grown at 400 °C at a rate of 160 nm/h. This was followed by a 80 nm GaAs capping layer grown also at 400 °C. The same temperature was used to grow the GaAs_{1-x}Bi_x layer and the GaAs cap to avoid the unintentional annealing which may lead to the diffusion of Bi atoms to the GaAs cap. The GaAs_{1-x}Bi_x growth temperature used in our letter is higher than those reported in the literature^{2,8,10,15} (270–380 °C). Based on eye inspection, our sample surface is between a mirrorlike and a rough finish. The surface roughness strongly depends on the Bi content, thickness and the growth rate of the epilayer.

In order to determine the Bi content in our samples, x-ray rocking curves ($\theta/2\theta$ scans) diffracted off the (004) plane of the as-grown sample were measured. The peak split between the GaAs substrate peak and the GaAs_{1-x}Bi_x peak was used to estimate the Bi content when using 6.324 Å for the theoretical GaBi lattice constant.¹⁶ The analysis assumed tetragonal distortion to the GaAs_{1-x}Bi_x lattice using Poisson's ratio for GaAs. It also assumed there was no relaxation in the strained GaAs_{1-x}Bi_x layer based on relatively narrow epilayer x-ray diffraction peak and well-defined fringes.

For the PL measurements, the sample was excited with a continuous-wave 532 nm wavelength light emitted by a diode-pumped solid state laser. PL from the sample was dispersed in wavelengths using a monochromator, before being detected by a liquid nitrogen-cooled germanium detector. Phase-sensitive detection of the PL signal from the germanium detector was implemented with a lock-in amplifier for

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FIG. 1. Temperature dependence data (symbols) of PL peak energy with different P_e . The data are fitted using the standard and modified Varshni expression (dashed and solid lines). The inset shows the RT PL spectra for samples grown at 400 °C (solid line) and 325 °C (dashed line).

the PL signal and a mechanical chopper for the 532 nm laser light serving as the reference.

PL was measured at temperatures ranging from 10 K to room temperature (RT) as a function of excitation power density, P_e for $\text{GaAs}_{0.97}\text{Bi}_{0.03}$. Temperature dependence data of the PL peak energy obtained using $P_e=34$ and 340 W cm^{-2} are compared in Fig. 1. Both sets of data displayed S-shape behavior, particularly at the lower pump intensity, indicating exciton localization at the low temperatures. Similar observations were also reported in Ref. 13.

For the $P_e=34 \text{ W cm}^{-2}$ data, as temperature increases from 10 to $\sim 80 \text{ K}$, the PL peak energy decreases as excitons transfer to lower energy states nearby. From 80 to $\sim 150 \text{ K}$, the PL peak energy increases as the increasing thermal energy increasingly mobilizes previously localized excitons. Above 160 K, the more usual redshift in the PL peak is observed, which mainly depends on the temperature dependence of the band gap since PL at these temperatures originates from the band edges.

The temperature dependence of the band gap in unperturbed semiconductors is often described by the Varshni equation,¹⁷

$$E_g(T) = E_0 - \alpha T^2 / (T + \beta), \quad (1)$$

where E_0 is the band gap at 0 K, α and β are fitting parameters. For perturbed semiconductors with band-tail states contributing to radiative recombination, as in the case of $\text{GaAs}_{1-x}\text{Bi}_x$, Eq. (1) is modified into¹⁸

$$E_g(T) = E_0 - \alpha T^2 / (T + \beta) - \sigma^2 / kT, \quad (2)$$

where σ is the localization parameter which indicates the degree of localization and k is Boltzmann's constant. Large value of σ indicates strong localization effects. Equation (2) is only valid for $T > 70 \text{ K}$ where the assumption of nondegenerate occupation is satisfied.¹⁸

Fitting our data in Fig. 1 using Eq. (1) yielded $\alpha = 0.42 \text{ meV/K}$ and $E_0 = 1.275 \text{ eV}$ [using $\beta = 204 \text{ K}$ from GaAs (Ref. 19)]. For Eq. (2), we obtained $\alpha = 0.49 \text{ meV/K}$, $E_0 = 1.325 \text{ eV}$, $\beta = 120 \text{ K}$, and $\sigma = 22.5 \text{ meV}$. The α values are similar, which is expected because exciton localization due to band-tail states is not pronounced at high temperatures. While these values are larger than the reported values

FIG. 2. (a) FWHM vs temperature data for different P_e of this letter. (b) Comparison of RT FWHM of this letter and reports in the literature.

of 0.4 meV/K and 0.27 meV/K in Ref. 1 ($x=0.012$) and Ref. 13 ($x=0.04-0.05$), respectively, they are still smaller than the temperature coefficient from a control GaAs sample which is 0.56 meV/K (not shown in Fig. 1). Hence $\text{GaAs}_{0.97}\text{Bi}_{0.03}$ does have a weaker temperature dependence of band gap than GaAs.

FWHMs of the PL data are plotted against temperature in Fig. 2(a). The $P_e=34 \text{ W cm}^{-2}$ data exhibited a strong maximum at $\sim 100 \text{ K}$, while the peak for the $P_e=340 \text{ W cm}^{-2}$ data was much less pronounced. This difference is expected from the more pronounced S-shape for $P_e=34 \text{ W cm}^{-2}$ data in Fig. 1. This is because at higher pump intensity, the higher photogenerated carrier density blurs out carrier localization effects. The abrupt increase in the FWHM with temperature is explained by the increase in exciton's mobility with temperature.²⁰ As temperature increases, the excitons become more mobile, thus recombining from (and producing PL with) a broader (nonequilibrium) energy distribution. The FWHM then drops when excitons come into thermal equilibrium. The subsequent FWHM increase for $T > 160 \text{ K}$ is due to increasing thermal distribution.

At RT, the PL peak wavelength and FWHM do not vary much with excitation power density. The PL data are shown in the inset of Fig. 1. The FWHM is 75 meV , which is narrow for this composition, as shown in the comparison of FWHM versus Bi content of Fig. 2(b). Sample grown at lower growth temperature of 325 °C (with lower Bi to As BEPR) showed significantly weaker PL intensity and broader

FIG. 3. IPL intensity vs excitation power density at different temperatures for Bi=0.03. The dashed and solid lines refer to fittings with gradient of 1 and greater than 1, respectively.

FWHM (see Fig. 1 inset). The relatively small changes in the BEPR (to obtain approximately the same Bi composition, $x=0.036$) do not affect the FWHM significantly and the broadening observed can be attributed to the severe alloy fluctuations due to limited solubility and lack of thermal energy of the Bi species. Other GaAs_{1-x}Bi_x samples grown at 400 °C with $x=0.014$ and 0.021, included in Fig. 2(b), also exhibited relatively narrow FWHMs. Despite the much reduced FWHM, they all still exceed the expected value due to thermal distribution broadening at RT, which is 26 meV.

Figure 2(b) shows the introduction of small amount Bi in GaAs causes a sudden linewidth broadening from 28 meV for GaAs to 69 meV for GaAs_{0.986}Bi_{0.014}. However, the FWHM only increase slightly with further increases in Bi content for composition up to 0.03. This suggests that the crystal quality was mainly affected by the incorporation of Bi (which induced significant alloy fluctuations) rather than the amount of Bi in the sample. The large 10 K FWHM showed in Fig. 2(a) indicates significant alloy fluctuations and Bi clustering. These are large compared to the typical linewidth of GaAs and InGaAs at 10 K which is ~13 meV and ~5 meV, respectively.

Analyzing the dependence of integrated PL (IPL) versus excitation power density can provide information of the dominant carrier recombination mechanism.²¹ Since dominance of nonradiative recombination is closely related to defect density in the bismide layer, such analysis will also be valuable in comparative PL study of future bismide samples. Using our data for the GaAs_{0.97}Bi_{0.03} sample, IPL versus P_e data are shown as a log-log plot in Fig. 3. Based on their gradients, m , the data in Fig. 3 can be grouped into three categories, namely, $m \sim 1$, $1 < m < 2$, and $m \sim 2$. They corresponds to the condition where the dominant carrier recombination mechanism is, radiative ($m \sim 1$), a mixture of radiative and monomolecular nonradiative (associated with defects) ($1 < m < 2$), or monomolecular nonradiative ($m \sim 2$).²¹ At 10 K, $m=1$ was observed at all P_e , indicating that radiative recombination is dominant over the entire excitation range investigated. At 77 K, all P_e above 2 mW cm⁻² had $m=1$, but low P_e 's data showed $m=1.7$. As the tempera-

ture increases, more of the IPL dependence on P_e tended toward $m \sim 2$ for all data points. By 220 K, all the data have a gradient of 2 suggesting that with increasing temperature the more mobile carriers increasingly recombine via defect states. The other two samples also show similar trend of IPL against P_e with nonradiative recombination dominating at higher temperatures.

Temperature dependent and power dependent PL measurements were carried out on series of samples with particular attention to the GaAs_{0.97}Bi_{0.03}. The temperature dependence data indicated exciton localization and showed that the band gap of the GaAs_{0.97}Bi_{0.03} alloy varies more weakly with temperature than in GaAs. At 75 meV, the RT FWHM for this sample was significantly smaller than others reported in the literature. This improvement is thought to be due to reduced alloy composition fluctuations in our sample achieved by using relatively high growth temperature.

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