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Transient Grating Measurements of Ambipolar Diffusion and Carrier Recombination in InGaP/InAIP Multiple Quantum Wells and InGaP Bulk

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The ambipolar diffusion coefficient and carrier recombination lifetime in InGaP/ InAlP multiple quantum wells and InGaP epitaxial layers grown by gas source molecular beam epitaxy have been determined by measuring the diffraction efficiency decay of transient gratings induced by picosecond laser pulses. The multiple quantum well room temperature ambipolar diffusion coefficient of carrier transport parallel to the growth plane was measured to be approximately half that of the bulk material.

Key words: Ambipolar diffusion, carrier recombination, InGaP/InAlP multiple quantum well

InGaP/InAlP multiple quantum wells (MQWs), which have a bandgap energy in the range of 1.9-2.2 eV, are utilized in visible semiconductor lasers1,2 and are of potential interest for the design of fast optical modulators for this region of the spectrum. The characterization of carrier transport behavior and carrier lifetime in these materials is important for device design. In this communication, we report the first measurements of the ambipolar diffusion coefficient and the carrier recombination time in InGaP/InAlP MQWs and compare the results with those corresponding to a bulk InGaP sample grown under the same conditions. These measurements were conducted utilizing the optical transient grating technique.3-9

Laser induced transient grating techniques have been previously used to study carrier dynamics of Si<sup>3</sup> and Ge4 and of III-V compounds like GaAs,5 InP,5 and InGaAsP.6 This technique has also been utilized to characterize the transport in MQWs structures in the plane<sup>7,8</sup> and in the direction of growth.<sup>9</sup> An advantage of this ultrafast laser technique over other pump and probe methods in which the contributions of recombination and diffusion to the carrier density decay are often difficult to separate, is that by controlling the grating spacing, the process dominating the grating decay can be selected. For small grating spacings, diffusion dominates while for large grating spacings carrier recombination dominates, therefore allowing for a more accurate determination of the individual rates.

The samples studied were grown on a GaAs substrate by gas source molecular beam epitaxy. The MQW sample consisted of a stack of 30 InGaP wells 85A thick separated by 225A InAlP barriers and the bulk sample consisted of a 0.4 µm epitaxial film. Both samples had an impurity concentration  $N_d - N_s = 10^{16}$ cm<sup>-2</sup>. The samples were of high quality as evidenced by the results of x-ray diffraction, transmission electron microscopy, and cw photoluminescence studies previously described. 10,11 To perform the transient grating transmission measurements discussed herein the substrate was removed by selective chemical etching and the samples were placed on a sapphire substrate.

The experimental setup utilized in the measurements is similar to transient grating pump and probe setups described in the literature<sup>4,7</sup> and makes use of a Rhodamine 590 dye laser synchronously pumped by a frequency doubled mode locked ND:YLF laser operating at 76 MHz. Two trains of 2 picosec-

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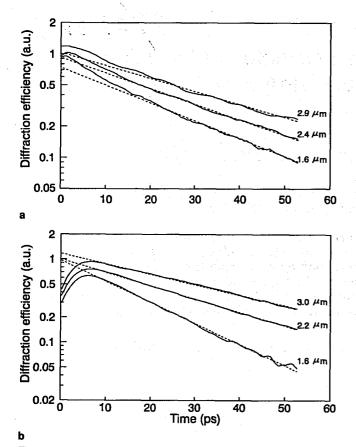


Fig. 1. Decay of the diffraction efficiency for three different grating spacings for (a) multiple quantum well structure and (b) bulk sample. Each curve is an average of five runs. The dotted line corresponds to the fit of the data with a single exponential function.

onds (ps) pump pulses from the dye laser set to operate at a wavelength of 600 nm were arranged to impinge on the sample with the same angle of incidence on each side of the normal to the surface, and set to coincide upon the sample in space and in time. The interference between these two pump pulses produces a sinusoidal intensity pattern on the plane of the wells and consequently, a spatially modulated free carrier density. 4,5 For an average power of 9 mW for each pump beam and taking into account the reflection losses at the sample surfaces, the maximum carrier density in the grating was computed to be  $7 \times 10^{12}$  cm<sup>-2</sup>. The decay of this laser created carrier grating, which is caused by carrier recombination and diffusion from regions of high carrier concentration to the regions of low carrier concentration, was monitored by measuring the variation of the intensity of the diffracted signal from a weak third probe beam as a function of delay with respect to the pump pulses. The delay between the pulses of the different beams was controlled by motorized translational stages. To separate the contribution of diffusion and recombination, the time decay of the diffracted signal was measured for various grating spacings ranging from 1 to 3 µm. The grating spacing was changed by varying the angle of incidence of the pump beams into the sample. This was achieved over a narrow range by

changing the spacing between the two parallel pump beams and over a broader range by utilizing different achromatic lenses with a focal length of 3, 4, and 5 cm. Conventional pump and probe measurements were also performed in these samples by blocking one of the pump beams.

Figure 1a shows the temporal evolution of the grating efficiency in the MQW sample as a function of delay with respect to the pump pulses for three grating spacings: 1.6, 2.4, and 2.6 µm. Figure 1b shows similar data for the bulk sample. In both cases, the decay rate of the diffracted signal is observed to decrease for larger grating spacings as a consequence of the reduced importance of diffusion. In the case of the InGaP bulk sample, the entire decay of the diffraction efficiency can be fitted well with a single exponential, indicating that the dynamics of the grating can be described with a linear diffusion-recombination model where the time variation of the free carrier density is described by<sup>4,7</sup>

$$\frac{d}{dt}n(x,z,t) = G(x,z,t) - n(x,z,t) / \tau_r + D_a \nabla^2 n(x,x,t)$$
 (1)

and where G(x,z,t) is the excitation rate,  $\tau_r$  is the linear carrier recombination lifetime, and D is the ambipolar diffusion coefficient. In the MQW, both the diffraction efficiency and the pump and probe transmission decays showed an initial fast decay followed by a slower single exponential decay for delay times larger than 25 ps. At the shorter delay times when the free carrier density is larger, the observed enhanced decay rate is likely to be associated with an initially non-negligible contribution of bimolecular and Auger recombination. A fit of pump and probe signals yielded values of these recombination coefficients which are of the same order of magnitude than those measured in other MQW materials.12 However, since the uncertainty associated with these fits is large, no attempt was made to determine the bimolecular and Auger recombination coefficients, and we limited our analysis to fitting the decays for times greater than 25 ps. At these delay times, the diffusion-recombination model of Eq. (1) applies, and the data can be fitted by a single exponential. The solution to Eq. (1) gives the temporal evolution of the carrier concentration from which the evolution of the diffracted signal can be obtained. The decay rate of the diffracted signal  $2\Gamma$ , in the linear recombination model is expressed as a function of the grating spacing  $\Lambda$  by<sup>4,8</sup>

$$2\Gamma = 8\frac{\pi^2}{\Lambda^2}D_a + \frac{2}{\tau_r}$$
 (2)

and is equal to twice the grating decay rate.

To determine the ambipolar diffusion coefficient and the carrier recombination lifetime, the decay rate of the diffracted signal,  $2\Gamma$ , was plotted as a function of  $8\pi^2/\Lambda^2$ . Figures 2a and 2b are the plots correspond-

ing to the MQW and bulk samples, respectively. As indicated by Eq. (2), the slope of such a plot is the ambipolar diffusion coefficient and the intercept at the ordinate axis is twice the inverse of the recombination lifetime. A least square fit of the data yielded values of the ambipolar diffusion coefficient of  $(3.4 \pm$ 0.6)cm<sup>2</sup>/s and  $(6.9 \pm 0.5)$ cm<sup>2</sup>/s for the MQW and bulk samples, respectively. The recombination lifetime of the bulk sample was determined to be  $(90 \pm 5)$ ps. slightly larger than the value of  $(70 \pm 5)$ ps corresponding to the MQW sample. The quoted value of the ambipolar diffusion coefficient in the InGaP/InAlP MQW is to our knowledge the first measurement of this quantity in these material, while the value for the bulk sample is consistent with a previous estimate based on pump and probe transmission measurements that gave a lower limit of 2.8cm<sup>2</sup>/s.<sup>13</sup>

The ambipolar diffusion coefficient in the bulk can be calculated from the electron and hole diffusivities, which are directly related to the electron and hole mobilities respectively through the Einstein relationship.14 When the hole mobility is much smaller than the electron mobility, as is the case in InGaP,15 the ambipolar diffusion constant is approximately equal to twice the diffusivity of holes. We calculated the hole diffusivity in the relaxation time approximation in bulk InGaP considering all the relevant scattering mechanisms. At room temperature, the hole mobility is limited by polar optical phonon scattering; however, alloy and deformation potential scattering, which have scattering rates about half that of polar optical phonon scattering, are also important. The nonpolar optical scattering rate, which in binary III-V semiconductors was found to be of the same order of magnitude than the polar optical scattering rate at room temperature, 16 for the ternary InGaP it was calculated to be an order of magnitude smaller. Taking into account the dominant scattering mechanisms, a value of the ambipolar diffusion coefficient of 10 cm<sup>2</sup>/s was obtained, in reasonable agreement with our measured value. Similar calculations of the hole mobility for the MQW cannot be easily performed due to the complicated structure of the valence band. However, the smaller ambipolar diffusion coefficient measured for the MQW is consistent with calculations based on an increased polar optical scattering rate in MQW structures as compared with bulk material. Hess<sup>17</sup> calculated an increase of the optical phonon scattering rate by a factor of 1.2–2 in GaAs/AlGaAs heterostructures as compared to bulk GaAs material. The presence of interface roughness scattering in the InGaP/InAlP MQW structure is also expected to contribute to the observed decrease in the ambipolar diffusion coefficient.

## **SUMMARY**

In summary, we have measured the ambipolar diffusion coefficient and carrier recombination lifetime in InGaP/InAlP MQW and in bulk InGaP utilizing the optical transient grating technique. Room temperature values of  $(3.4 \pm 0.5)$ cm<sup>2</sup>/s and  $(6.9 \pm 0.5)$ cm<sup>2</sup>/s

0.5)cm²/s for the ambipolar diffusion coefficient and recombination lifetimes of 70 and 90 ps were obtained for the MQW and bulk samples, respectively. The difference in the ambipolar diffusion coefficients between the two structures is attributed to an increase of the polar optical phonon scattering rate and the presence of interface roughness scattering in the MQW structure.

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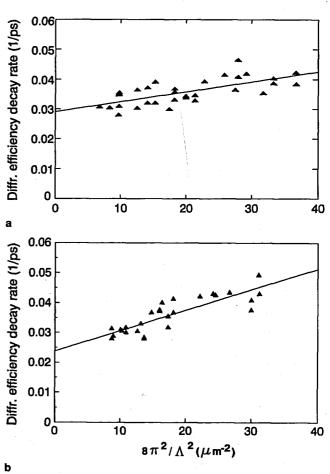


Fig. 2. Variation of the diffraction efficiency decay rate with the inverse of the square of the grating spacing for (a) multiple quantum well and (b) bulk sample. Each experimental point is the average of five runs.

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