Picosecond photoluminescence nonlinearity of two-dimensional plasma in GaAs-(Al_{0.3}Ga_{0.7}As) multiple quantum wells

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CW and sub-picosecond excitation correlation photoluminescence techniques were utilized to characterize the 2D-plasma modulation doped in the GaAs-Al_{0.3}Ga_{0.7}As multiple quantum wells. The gain or loss of photoluminescence intensity was found to be closely related to the excitation intensity. In addition, the relaxation time as a function of excitation intensity was determined with sub-picosecond precision, and theoretically can be attributable to phonon reabsorption.

The advent of ultrafast laser pulses ($< 10^{-12}$ s) has made it possible to observe some of the dynamic behaviour of nonequilibrium, hot, photoexcited carriers in multiple quantum wells (MQW) structures in real time, which is essential for a better understanding of the physical mechanisms involved in ultrafast high speed MQW devices. Photoluminescence (PL) techniques which include PL emission, PL excitation, and the sub-picosecond excitation correlation method were utilized to characterize the two-dimensional (2D) charged plasma in GaAs-(Al_{0.3}Ga_{0.7}As) multiple quantum wells. The samples were prepared by a molecular-beam-epitaxy system equipped with a multiple sample interlock [1]. Multiple layers of GaAs-Al_{0.3}Ga_{0.7}As were grown on a semi-insulating GaAs (100) substrate at a temperature of about 650 °C. A GaAs buffer layer was grown first, followed by the required number of alternat-

ing layers of Al_{0.3}Ga_{0.7}As and GaAs. In these modulation-doped structures, only Al(GaAs) layers are intentionally doped, leaving the GaAs layers essentially free of impurities. Electrons transfer from the higher-energy conduction band of (AlGa)As barrier layer to the lower-energy band of GaAs layer, achieving spatial separation of electrons from their parent ionic donors. Thus, a 2D charged plasma is formed inside the GaAs layer [2]. Our MQW heterostructures had 30 layers with a typical well width of 20 nm and undoped (AlGa)As spacer layers adjacent to Si-doped barrier layers. The carrier concentration, n_0 , was in the range of $1.0\text{--}2.0 \times 10^{12} \text{ cm}^{-2}$ in our samples.

The sample was cut with a wire saw so that its long axis was parallel to (110) to facilitate mounting, strain-free, in an exchange L-He cryostat. The sample was cooled to approximately 5 K. A standard Spex 1870B 0.5 m spectrometer equipped with a cooled GaAs photomultiplier was used to obtain the spectra. The laser source consisted of a dye laser pumped by a 5 W all-lines

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krypton-ion laser. For the PL excitation measurement, the birefringent filter unit in the dye laser cavity was coupled to a synchronous stepping motor system to provide continuous varying excitation energies while the spectrometer was set at a fixed wavelength (energy). Oxazine 750 dissolved in DMSO (dimethylsulfoxide) was employed giving a variable excitation energy, $\hbar\omega_{\text{exc}}$, from 1.50 to 1.64 eV. The laser beam was polarized along (110) and entered the sample along (001) nearly normal to the layers. An angle of incidence on the sample of 15° resulted in a nearly backscattering geometry inside the sample. The experimental setup for the picosecond excitation correlation photoluminescence relaxation measurements is shown in fig. 1. The excitation source is a synchronously pumped Rh 6G dye laser operating at 590 nm, which provided the output in the form of 0.6–0.8 ps temporal width with a repetition rate of 7.6 MHz controlled by a cavity dumper. The output beam is equally split into two for establishing a typical pump and probe optical arrangement. The rotator gave orthogonal polarized pump and probe beams to avoid parametric interference [3].

Figure 2 shows a typical PL emission spectra of our samples. The highest peak, which occurs at 8190 Å, is associated with a $\Delta n = 0$ transition, E_0 , i.e., the ground conduction subband to

Fig. 1. Schematic of the picosecond excitation correlation spectroscopy apparatus.

Fig. 2. Photoluminescence emission spectra from samples A and B MQW heterostructures with carrier concentration of 1.82×10^{12} and $1.5 \times 10^{12} \text{ cm}^{-2}$ respectively.

ground-state heavy hole subband. The $\Delta n = 1$ transition (E_1) from the first excited conduction subband to ground valence subband states, although forbidden by the selection rule, is visible in some MQW spectra (fig. 2(b)). One representative PL excitation spectrum on sample A (same sample as in fig. 2(b)) at 5 K is shown in fig. 3 by setting the spectrometer on the band-gap energy (ground state energy), E_0 . The 2D electron density can be determined from the energy separation obtained in the corresponding PL excitation spectrum, which turns out to be consistent with the Hall density. In addition, the concentration homogeneity revealed from layer to layer is useful in selecting high quality modulation doped MQW samples [4], which proved to be essential in the forthcoming photoluminescence nonlinearity investigations.

Fig. 3. Photoluminescence excitation spectrum of sample A. The 2D electron density as well as its uniformity were determined from the energy separation between the attenuated laser line and the absorption edge.

As the sample temperature was increased from 4 to 25 K, all the PL peaks from the GaAs layers were diminished dramatically and barely visible beyond 30 K (figs. 4, 5), while much less sensitivity to temperature for the PL peak from the AlGaAs barrier layers was found. In the picosecond excitation correlation technique applied at 4 K, the laser pulses enhanced the 2D-plasma temperature. Thus, the later arriving laser pulse can sense the non-equilibrium state set up by the first pulse. The time delay was continuously varied by scanning the path difference between two pulses. As expected, a dip of the PL peak intensity (E_0) as a function of time delay was found, as shown in the lower half of fig. 6. As the laser excitation intensity was increased, the dip leveled up and gradually evolved into a broad spike. The broad spike is similar to a feature previously reported on undoped GaAs-AlGaAs MQW [5,6]. The excited carrier density is given by the following equation:

$$n_{\text{ex}} = \frac{P}{1.602 \times 10^{-19} \times f E_e \pi r^2 (1 - R)(1 - e^{-\alpha x})}, \quad (1)$$

where P is the laser power, f the repetitive rate of the laser pulse, E_e the exciting photon energy in eV, r the radius of the laser beam spot, χ the width of GaAs layer, and $R = 0.35$ [7], $\alpha = 1.4 \times$

Fig. 4. Photoluminescence spectrum of sample B as a function of temperature.

Fig. 5. Photoluminescence intensity of the 8190 Å peak (sample B) as a function of sample temperature. The solid line is a guide to the eye.

Fig. 6. Delay time scans of photoluminescence intensity at the 8190 Å peak (sample B), with excitation density (a) $n_{\text{ex}} = 2.6 \times 10^{12}$; (b) 4.6×10^{11} ; (c) 1.4×10^{11} ; (d) 6.5×10^{10} ; (e) 2.5×10^{10} ; (f) 9.3×10^9 ; (g) 4.4×10^9 ; (h) $1 \times 10^9 \text{ cm}^{-2}$.

10^4 cm^{-1} [8] are the reflection and absorption coefficient of GaAs respectively. Taking into consideration the power loss by the optical components, the excited carrier density in our measurements ranged from about 1×10^9 to about $2.6 \times 10^{12} \text{ cm}^{-2}$.

At lowest n_{ex} , the correlated photoluminescence intensity remained the same. As n_{ex} was increased to $4.4 \times 10^9 \text{ cm}^{-2}$, a dip centered at zero delay started to occur, and had its maximum depth at $n_{\text{ex}} = 9.3 \times 10^9 \text{ cm}^{-2}$. As n_{ex} was further enhanced, the dip leveled up and gradually evolved into a spike which became stronger and broader (longer relaxation time) with increasing n_{ex} .

Obviously, there were two different kinds of effects involved in the time resolved spectra observed; the non-equilibrium thermal effect and the excitation correlation nonlinear effect from the two beams. Intense photoexcitation of a semi-

conductor leads to a heating of the carrier system and results in a higher carrier temperature than the lattice temperature, and the carrier temperature increases with increasing excitation intensity. Because of the fast relaxation of carriers, the excited carrier density decreases with delay time, which leads to a decrease of the carrier temperature. The “dip” was the result of the decrease of radiative recombination rate with an increase of the carrier temperature. Furthermore, it was notable that the temperature effect was more sensitive at low temperature than at high temperature, as shown in fig. 5. Consequently the “dip” effect became less prominent as the excitation density increased. In contrast to a recent report about a “dip” superimposed on the top of a “spike”, which was observed on undoped $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ single strained quantum wells due to the band refilling effect [6], we believe that the “dip” (with no background of broad “spike” profile) shown in our experimental data of modulation doped 2D plasma in MQWs is primarily caused by thermal heating.

On the other hand, contrary to the thermal effect, the nonlinear intensity I_s (“spike”) was only observable at higher excitation density. This was because that the nonlinear photoluminescence intensity, I_s , was proportional to $n(t)p(t + \gamma)$ (γ is the delay time; n and p are the electron states and hole states, respectively), which was much weaker than the background intensity $I_B \propto (n_0 + n(t))p(t)$ at low n_{ex} due to the large n_0 in the samples. But as n_{ex} was enhanced to be comparable with n_0 , I_s started increasing superlinearly and consequently a “spike” occurred [9].

Evidently, the “dip” and “spike” are two different phenomena. However, it is found that the relaxation of both is actually due to a common factor: the relaxation of hot carriers. To determine the relaxation time τ from the time delay spectra, it was found that an exponential function yielded good fit to the experimental data where ΔI , a fitting parameter, was

$$\left[I(t) = I_B + \Delta I \exp\left(\frac{-|t|}{\tau}\right) \right], \quad (2)$$

as a function of carrier temperature T_c and n_{ex} ;

I_B was the background intensity ($\gamma = \infty$), which was obtained as the sum of the two separately measured intensities due to individual beams. Figure 7 shows some results for the 8190 Å peak, where the smooth curves are fitted to eq. (2). The relaxation time as a function of excitation density is plotted in fig. 8. It is about 65 ps at low n_{ex} , and reaches about 210 ps at maximum n_{ex} , indicating that the carriers with a higher n_{ex} have slower relaxation.

Under an intense excitation, not only a high density of hot carriers is created, but also a significant non-equilibrium phonon population is built up, because the LO-phonon lifetime (~ 7 ps) is much longer than the electron-photon scattering rate (~ 0.17 ps). The build-up of these non-equilibrium phonons leads to a photon temperature, T_p , higher than the lattice temperature, T_L , and can substantially reduce the energy relaxation rate of carriers by phonon reabsorption.

It has been shown that in polar materials such as GaAs, longitudinal optical phonon scattering is

Fig. 7. Fitting of time delay spectra. The solid lines are fitted to eq. (1).

Fig. 8. Relaxation time as a function of excitation density. The solid line is a guide to the eye.

the dominant mechanism of scattering at temperature above 40 K, and acoustic phonon scattering is only important at low temperature [10]. In the temperature range of our experiment, the electrons emit LO-phonons with wave vectors in the range between about 1×10^6 and 5×10^6 cm⁻¹ [11] and the relevant phonon states have a sheet density of about $n_{LO}^{rel} = 2.0 \times 10^{12}$ cm⁻² [12]. We can describe the excess population in this phonon vector range by a phonon temperature T_p which is higher than the lattice temperature T_L , but lower than the carrier temperature T_c . In order to have a quantitative estimate of how these hot phonons influence the carrier energy loss rate (ELR), let us consider a simplified case in the limit of Maxwell-Boltzmann statistics and for T_L , T_c , and $T_p < \Theta = \hbar\omega_{LO}/k$, without considering screening and other effects. The reduced ELR is given by

$$\left[\frac{dE}{dt} \right]_{red} = - \frac{\hbar\omega_{LO}}{\tau_s} \left[\exp\left(\frac{-\Theta}{T_c} \right) - \exp\left(\frac{-\Theta}{T_p} \right) \right], \quad (3)$$

where τ_s is the carrier-LO-phonon scattering time. The second term on the right side of the equation represents phonon absorption.

The undisturbed ELR ($T_p < T_c$) is

$$\left[\frac{dE}{dt} \right]_{un} = - \frac{\hbar \omega_{LO}}{\tau_s} \exp\left(\frac{-\Theta}{T_c} \right), \quad (4)$$

which we related to the reduced ELR by a reduction parameter η as follows

$$\eta \left[\frac{dE}{dt} \right]_{red} = \left[\frac{dE}{dt} \right]_{un}. \quad (5)$$

We now first consider a steady-state equilibrium, where the excitation and annihilation of LO-phonons are equal. Under this condition, N electron ($N = n_0 + n_{ex}$) have a net emission of LO-phonons

$$\begin{aligned} \frac{N}{\tau_s} \left[\exp\left(\frac{-\Theta}{T_c} \right) - \exp\left(\frac{-\Theta}{T_p} \right) \right] \\ = \frac{n_{LO}^{rel}}{\tau_{LO}} \exp\left(\frac{-\Theta}{T_p} \right), \end{aligned} \quad (6)$$

where n_{LO}^{rel} is the relevant LO-phonon density mentioned earlier, τ_{LO} the lifetime of the LO-phonon, and the right-hand side describes the decay of available relevant LO-phonons into acoustic phonons. After some calculation, from eqs. (3)–(6) we get

$$\eta = 1 + \frac{N}{n_{LO}^{rel}} \frac{\tau_{LO}}{\tau_s}. \quad (7)$$

It is interesting to note that the reduction of η is a function of density only, but not T_c or T_p , which allows us to estimate the reduction of ELR in non-steady-state situation. By using $n_{LO}^{rel} = 2.0 \times 10^{12} \text{ cm}^{-2}$, $\tau_{LO} = 7 \text{ ps}$, $\tau_s = 0.17 \text{ ps}$, we found that η changed from about 31 to 87 in the density range of our work, which indicated a reduction of the ELR by a factor near 3, a reasonably good agreement with our experimental results.

In summary, we have observed the evolution of loss (dip) to gain (spike) for the PL signals of modulation doped GaAs-Al_xGa_{1-x}As MQW samples with sub-picosecond precision. For the

low excitation intensity, the loss is primarily due to the thermal effect set up by the first pulse. As the excitation intensity is increased, the product of np becomes the predominant factor which manifests the photoluminescence nonlinearity (spike). The relaxation time, as a function of excitation density, was precisely determined and was quite successfully interpreted by a hot-photon model.

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