Photoluminescence-excitation-correlation spectroscopic study of a high-density two-dimensional electron gas in GaAs/$Al_{0.3}Ga_{0.7}As$ modulation-doped quantum wells

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We reported the photoluminescence properties of a high-density electron gas in GaAs-$Al_{0.3}Ga_{0.7}As$ modulation-doped quantum wells by using cw and subpicosecond excitation correlation photoluminescence techniques. The gain or loss of photoluminescence intensity was found to be closely related to the excitation intensity vs carrier concentration. There is a significant distinction between the relaxation processes of minor carriers and of the doped two-dimensional electron gas in the wells. The former takes about a constant time duration ($\sim$70 ps), while the latter is sensitive to the excitation intensity, and theoretically can be attributable to phonon reabsorption.

I. INTRODUCTION

The modulation-doped two-dimensional electron gas (2DEG) exhibits a variety of important physical phenomena as well as attractive features for fabricating high-speed electro-optical devices.[1-5] The advent of ultrafast laser pulses ($<10^{-12}$ s) has made it possible to observe some of the dynamic behavior of nonequilibrium, hot, photoexcited carriers in multiple-quantum-well (MQW) structures in real time, which is essential for a better understanding of the physical mechanism involved in ultrafast high-speed MQW devices. The photoluminescence-excitation-correlation spectroscopic technique was chosen to serve that purpose based upon the simplicity of its experimental setup and the best possible temporal resolution, which is limited only by the laser-pulse width. In this paper, we present the photoluminescence properties of high-density 2DEG in GaAs-$Al_{0.3}Ga_{0.7}As$ by using photoluminescence emission, photoluminescence excitation, and subpicosecond photoluminescence-correlation techniques. The charge density, interband transitions, Fermi enhancement, and breakdown of the parity selection rule of the samples were analyzed. Most of the discussion will be focused on the gain or loss of photoluminescence intensity as a function of excitation intensity in the temporal domain, which reflects some dynamic properties of 2DEG in the wells.

II. EXPERIMENT

The modulation-doped GaAs-$Al_{0.3}Ga_{0.7}As$ multiple quantum wells have 15 periods with a typical well width of 200 Å and Si-doped barrier width of 600 Å.[6] The electron concentration obtained independently from Hall measurement and photoluminescence-excitation spectra is approximately 1.8×10^{12} cm^{-2} for sample A and 1.5×10^{12} cm^{-2} for sample B. The sample was cut with a wire saw so that its long axis was parallel to (100) to facilitate mounting, strain free, in an exchange L-He cryostat. The sample was cooled to approximately 5 K. A standard Spex 1870 B 0.5-m spectrometer equipped with a cooled GaAs photomultiplier was used to obtain the spectra. The experimental setup for the picosecond excitation correlation photoluminescence measurement is illustrated 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 time delay is continuously changing by scanning the path difference between two

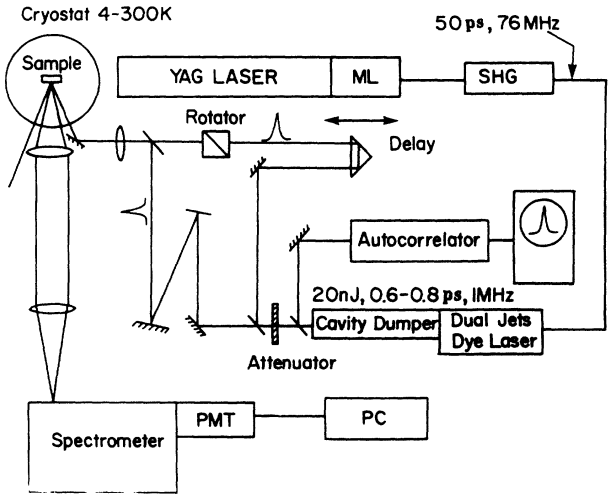

FIG. 1. Schematic of picosecond excitation correlation spectroscopy apparatus.

pulses. The rotator gave orthogonal polarized pump-and-probe beams to avoid parametric interference.

III. RESULTS AND DISCUSSION

Figure 2 shows a typical photoluminescence (PL) emission spectra of our samples. Four dominant peaks, indicated as $A-D$, are present. According to our previous investigations, peak D, at an energy of 1.49 eV, is due to the carbon acceptor (D^0, C_{As}) transition in the GaAs substrate.

Peaks $A-C$ are due to 2DEG, evidenced by a kind of group behavior as the temperature varies, and are much more sensitive compared to the photoluminescence signals from the $Al_xGa_{1-x}As$ barrier layer below 100 K. Peak A, which occurs at 8190 Å, is primarily associated with a $\Delta n = 0$ transition, E_0, i.e., the ground conduction subband to ground-state heavy-hole subband $(e\text{-hh})_1$. Strictly speaking, peak A is formed by two components attributed to $(e\text{-}A^0)$ and an $(e\text{-hh})_1$ transition at the renormalization band gap E_g'. The $\Delta n = 1$ transition (E_1) from the first excited conduction subband to the ground-state valence subband, although forbidden by the selection rule, is visible in some MQW spectra. The 13-meV energy separation between peaks B and C can be picked in resonant Raman scattering out of intersubband transition and, thus, gets further support for the corresponding assignments.[7] Peak A, identified as the Fermi-edge singularity, results from the strong correlation and multiple scattering of electrons near the Fermi edge by the localized holes.[8] Due to the strong electron-electron correlation, the scattering is multiple electron-hole scattering and results in the enhancement of the oscillator strength near the Fermi edge. Photoluminescence excitation was undertaken to determine the Fermi energy and, thus, the carrier concentration under laser illumination. We also found that the energy difference, ΔE, between peaks A and C is almost equal to the Fermi energy of 37.5 meV. Therefore, the PL spectrum, to a great extent, reflects the distribution of the two-dimensional electrons from the bottom of the conduction band to the Fermi level. Detailed discussions of the optical and electrical properties of the samples have been published elsewhere.[7,8] When the sample temperature was increased from 4 to 25 K, all the PL peaks from GaAs layers were diminished dramatically (Fig. 3), while much less sensitivity to temperature for the PL peak from the $Al_xGa_{1-x}As$ barrier layer was displayed (not shown). In the picosecond excitation correlation technique, the nonequilibrium condition set up by the first pulse varies the PL intensity generated by the second laser pulse, which arrived picoseconds later. Under the low-excitation condition, 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. 4. 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-$Al_xGa_{1-x}As$ MQW.[9] The excited carrier density

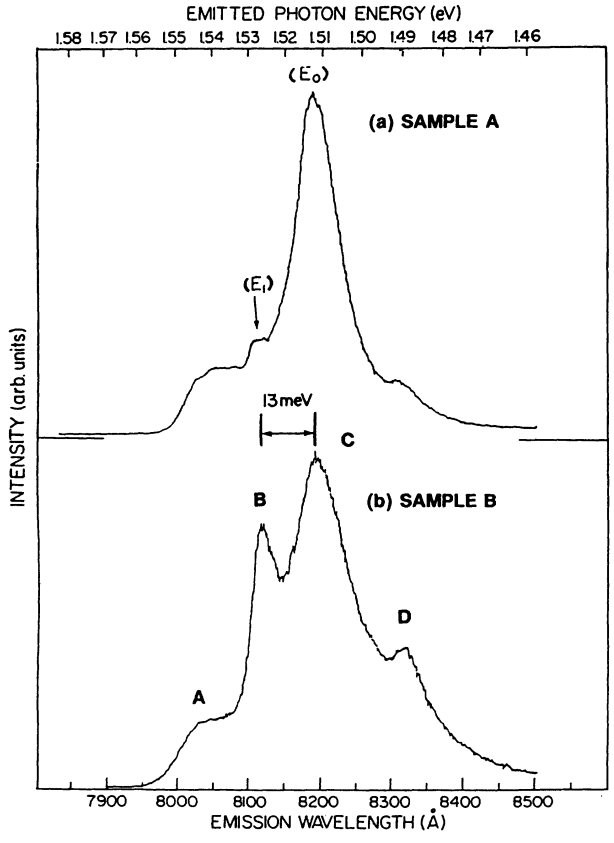

FIG. 2. Photoluminescence emission spectra of modulation-doped GaAs-$Al_{0.3}Ga_{0.7}As$ MQW's for samples A and B with a carrier concentration of 1.8×10^{12} and 1.5×10^{12} cm^{-2}, respectively, measured at 5 K.

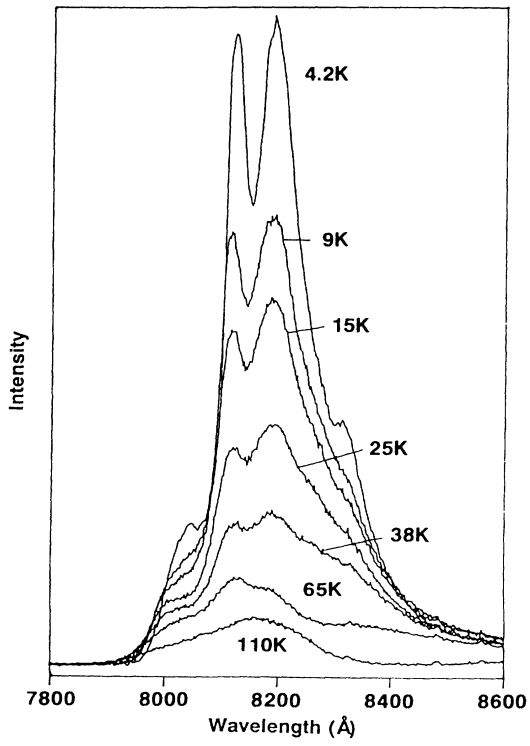

FIG. 3. Photoluminescence spectrum of sample B as a function of temperature.

is given by the following equation:

$$n_{ex}=\frac{P}{1.602\times10^{-19}\times fE_e\pi r^2}(1-R)(1-e^{-\alpha\chi}), \quad (1)$$

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 the GaAs layer, and $R=0.35$,[10] and $\alpha=1.4\times10^4$ cm^{-1} (Ref. 11) are the reflection and absorption coefficient of GaAs, respectively. Taking into account the power loss by the optical components, the excited carrier density in our measurements ranged from 1×10^9 to about 2.6×10^{12} cm^{-2}.

At lowest n_{ex}, the correlated photoluminescence intensity remained the same. As n_{ex} was increased to 4.4×10^9 cm^{-2}, a dip centered at zero delay started to occur, and had its maximum depth at $n_{ex}=9.3\times10^9$ 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}.

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

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

as a function of carrier temperature T_c and n_{ex}. I_B was the background intensity ($\gamma=\infty$, γ is the delay time), which was obtained as the sum of the two separately measured intensities due to individual beams. Figure 5 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 intensity is plotted in Fig. 6. It is about 70 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 low excitation, excitation-correlation signals mainly came from the PL spectral region of band-to-band transitions, which is shown in Fig. 7(b). Such a charac-

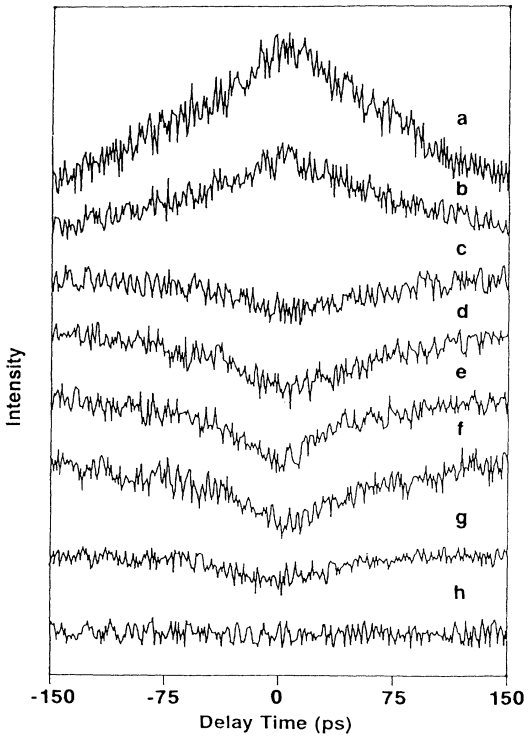

FIG. 4. Delay-time scans of photoluminescence intensity at the 8190-Å peak (sample B) with excitation density (a) $n_{ex}=2.6\times10^{12}$; (b) 4.6×10^{11}; (c) 1.4×10^{11}; (d) 6.5×10^{10}; (e) 2.5×10^{10}; (f) 9.3×10^9; (g) 4.4×10^9; (h) 1×10^9 cm^{-2}.

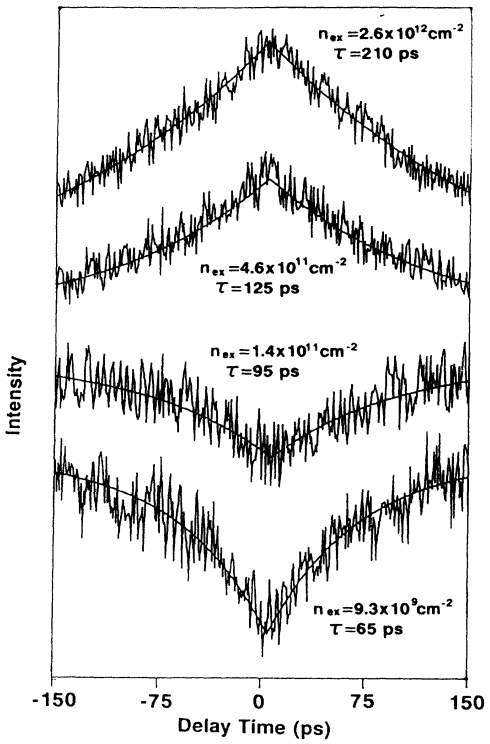

FIG. 5. Fitting of time delay spectra. The solid lines are fitted to Eq. (1).

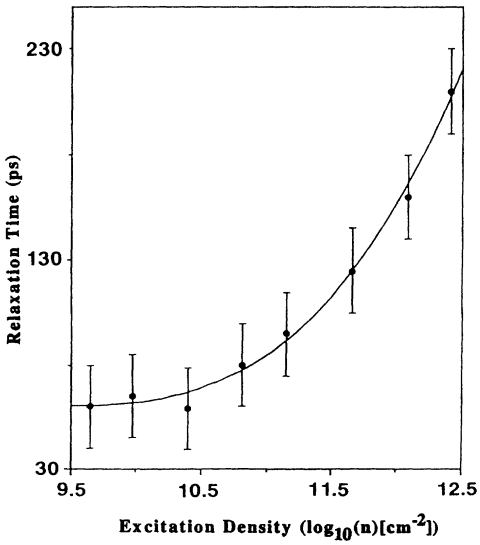

FIG. 6. Relaxation time as a function of excitation density. The solid line is a guide to the eye.

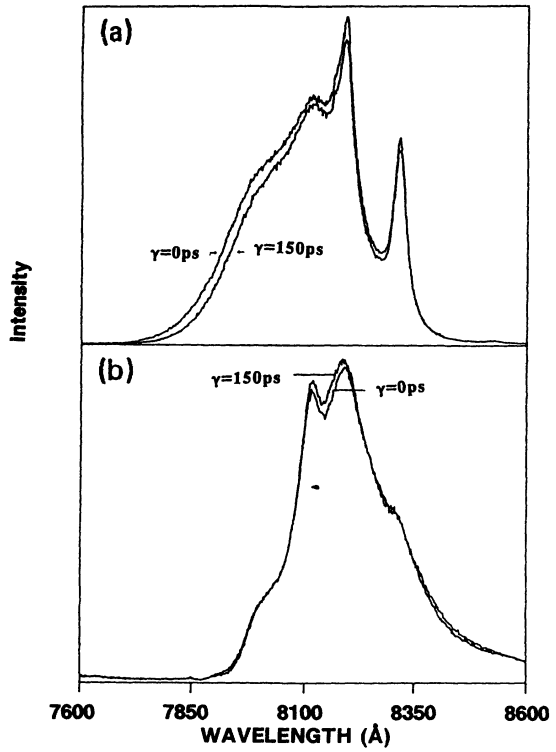

FIG. 7. Time-integrated PL spectra at a delay time of 0 and 150 ps with excitation density $n_{ex}=2.6\times10^{12}$ (a) and 2.5×10^{10} cm^{-2} (b) at 5 K.

teristic rules out that the "dip" is caused by thermal heating, which manifests the drop of the signals in the full-emissive spectral region (Fig. 3). The almost intact nature of the Fermi enhancement peak (peak A) and the Fermi-edge profile excludes that the 2DEG is the major factor for the "dip" present. Rather, it is most likely that the dip is attributed by the relaxation of the photogenerated holes in the valence band. The 70-ps relaxation time, which is almost insensitive to the excitation density under low excitation, is comparable with the value of 100 ps reported on the lifetime of hole and electron associated with the parallel recombination process in semi-insulating GaAs.[12] On the other hand, the relaxation time of the spike is closely related to the excitation intensity, as shown in Fig. 6. Furthermore, such a relaxation occurs in the full emissive spectral region [Fig. 7(a)], which can reasonably imply that the relaxation of the nonequilibrium situation of 2DEG perturbed by the first pulse affects the later arrived pulse in terms of its photoluminescence efficiency and consequently leads to photoluminescence nonlinearity of the spike observed. The relaxation time τ of the spike reveals the time duration required for the 2DEG to get back to equilibrium after being "disturbed" by the first laser pulse. Since the thermal effect on the sample can only drop the PL intensity, the spike is ruled out to be due to the higher lattice temperature as $\tau=0$. The nonlinear intensity I_s was proportional to $n(t)p(t+\gamma)$ (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 (n_0 is the doped electron concentration) in the sample. But as n_{ex} was enhanced to be comparable with n_0, I_s started increasing superlinearly and consequently a spike occurred.

Under an intense excitation, not only a high density of hot carriers is created, but also a significant nonequilibrium 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 buildup of these nonequilibrium phonons leads to a phonon 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, LO-phonon scattering is the dominant mechanism of scattering at temperatures above 40 K, and acoustic-phonon scattering is only important at low temperature.[13] 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} (Ref. 14) and the relevant phonon states have a sheet density of about $n_{LO}^{rel}=2.0\times10^{12}$ cm^{-2}.[15] 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 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 electrons ($N=n_0+n_{ex}$) have a net emission of LO phonons,

$$\frac{N}{\tau_s}\left[\exp\left[\frac{-\theta}{T_c}\right]-\exp\left[\frac{-\theta}{T_P}\right]\right]=n_{LO}^{rel}\exp\left[\frac{-\theta}{T_P}\right], \quad (6)$$

where n_{LO}^{rel} is the relevant LO-phonon density mentioned earlier, τ_{LO} is the lifetime of LO phonon, and the right-hand side describes the decay of available phonons into acoustic phonons. After some calculation, from Eqs.

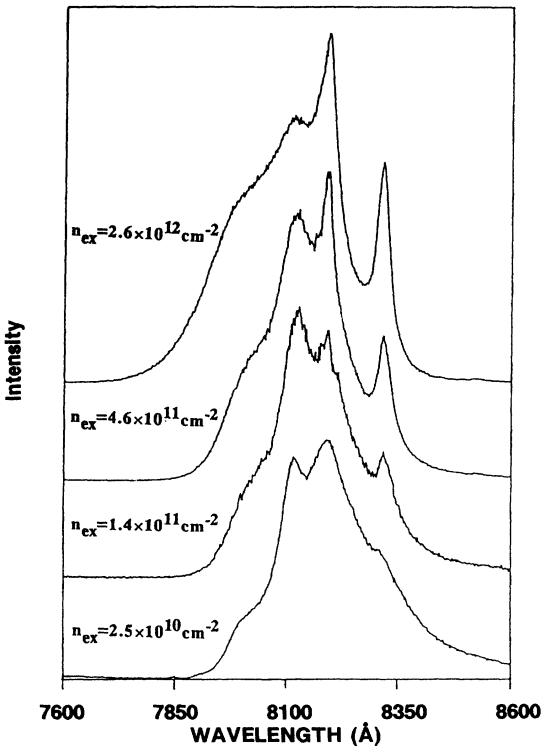

FIG. 8. Excitation-intensity-dependent photoluminescence spectra of sample B measured at 5 K.

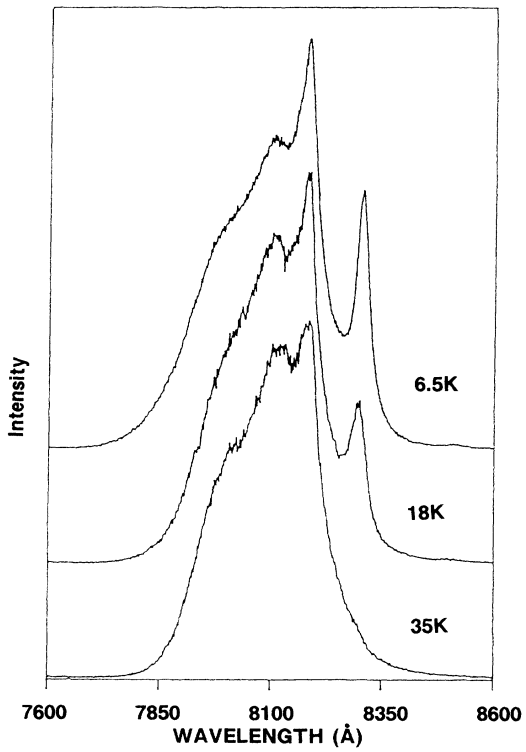

FIG. 9. Photoluminescence spectra of sample B at sample temperature 6.5, 18, and 35 K with excitation density $n_{ex}=2.6\times10^{12}$ cm^{-2}.

(3)–(6) we get

$$\eta=1+\frac{N}{n_{\mathrm{LO}}^{\mathrm{rel}}}\frac{\tau_{\mathrm{LO}}}{\tau_s}\ . \tag{7}$$

It is interesting to note the reduction of the ELR in the nonsteady-state situation. By using $n_{\mathrm{LO}}^{\mathrm{rel}}=2.0\times10^{12}$ cm^{-2}, $\tau_{\mathrm{LO}}=7$ ps, $\tau_s=0.17$ ps, we found that η changes from 43 to 85 in the density range from 1.95×10^{12} to 4.1×10^{12} cm^{-2}, which indicates a reduction of the ELR by a factor near 2, in reasonably good agreement with the relaxation time, from 125 to 210 ps obtained experimentally.

As mentioned earlier, the peak C in Fig. 2 is actually formed by two components—intrinsic and extrinsic emission. The enhancement of the extrinsic emission observed as the excitation intensity increases can be taken to resolve the components, as shown in Fig. 8. The extrinsic peak vanishes very rapidly as the temperature is above 30 K due to thermal ionization (Fig. 9)—a typical behavior of $(e\text{-}A^0)$ transition in GaAs.[16] The resolution of the peak is relevant to the analysis of band-gap renormalization of $Al_xGa_{1-x}As$-GaAs modulation-doped quantum wells reported recently.[17]

IV. CONCLUSION

In summary, cw and subpicosecond excitation correlation photoluminescence techniques were utilized as complementary tools to characterize 2DEG modulation doped in the GaAs-$Al_{0.3}Ga_{0.7}As$ multiple quantum wells. The relaxation time of loss (dip) and gain (spike) for the PL signals was determined with subpicosecond precision. For low excitation, the loss is primarily due to the relaxation of minority carriers. As the excitation population approaches the doped carrier concentration, the product of np becomes the predominant factor which manifests the photoluminescence nonlinearity (spike). The relaxation time as a function of excitation intensity can be quite successfully described by a hot-phonon model.

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