Effect of interparticle interactions on radiative lifetime of photoexcited electron–hole system in GaAs quantum wells

L. V. Kulik, A. I. Tartakovskiĭ, A. V. Larionov, E. S. Borovitskaya, and V. D. Kulakovskiĭ

Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow Region, Russia

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The paper reports on an investigation of changes in the photoluminescence linewidth and lifetime of excitons and electron–hole plasma over a wide range of densities between 3×10^7 and 3×10^{12} cm^{-2} at a temperature of 77 K in GaAs/AlGaAs quantum wells. The roles played by thermal ionization of excitons at low densities of nonequilibrium carriers, exciton–exciton and exciton–electron collisions, and ionization of excitons at high pumping power densities have been studied. © *1997 American Institute of Physics.* [S1063-7761(97)03107-7]

1. INTRODUCTION

Radiative recombination of excitons in quasi-two-dimensional semiconducting structures is quite different from the three-dimensional case. The lower dimensionality of the system leads to radical changes in the interaction between excitons and electromagnetic waves. Owing to the translational symmetry of a bulk crystal, this interaction results in formation of stationary excitonic polaritons, which can decay only through phonon scattering or conversion on the crystal surface.[1,2] In the case of excitons in quantum wells, the translational symmetry in the direction perpendicular to the quantum well plane is broken, which results in a very fast (of order 10 ps) decay of excitons with very small in-plane quasimomenta ($k<k_0=n\omega_x/c$). Here $\hbar\omega_x$ is the exciton energy, n is the refraction index, and c is the speed of light. Excitons with $k>k_0$ do not recombine.[2]

The cause of the fast recombination of excitons with $k<k_0$ is the phase coherence of the excitonic states. The loss of coherence due to either localization of excitons, or scattering by phonons, electrons or other quasiparticles leads to a sharp increase in the electron lifetime.[1] Partial ionization of excitons at higher temperatures also leads to an increase in the excitonic system lifetime.[3]

In the present work, we have studied the effect of interparticle interactions in the excitonic system in GaAs/AlGaAs quantum wells on the luminescence linewidth and lifetime over a wide range of densities of nonequilibrium carriers, including the region of the transition from excitonic gas to electron–hole plasma. Experiments have been conducted at a relatively high temperature of 77 K, when excitons and electron–hole plasma coexist in equilibrium and the effect of exciton localization on potential irregularities is negligible. Under these conditions, it is possible to reliably determine both the total density of photoexcited carriers and the system composition, which allows us to analyze on a quantitative level the effect of exciton–electron collisions on the decay of excitonic states and on the radiative annihilation of excitons, and also to study the radiative recombination time in a quasi-two-dimensional system in the region of high densities, where the transition from excitons to electron–hole plasma occurs.

2. EXPERIMENTAL TECHNIQUE

We selected for our experiments an undoped GaAs/$Al_{0.18}Ga_{0.82}As$ heterostructure grown by the MBE technique and containing a single quantum well of width $L=5$ nm. Excitons were generated by a pulsed picosecond R6G dye laser operating at a wavelength of 590 nm with a 70-ps pulse with a repetition rate of 4 MHz. The sample was placed in a cryostat. Pumping radiation was conducted to the sample and luminescence was fed from the cryostat via an optic fiber with a diameter of 0.6 mm, adjacent to the sample surface (within 0.5 mm). In order to prevent the spread of nonequlibrium carriers from the optically excited area, we used samples with 0.5-mm mesas selectively etched on their surfaces. Luminescence was detected by a photomultiplier tube operating in the time-correlated photon-counting mode. The density N of nonequilibrium carriers in the quantum well at high pumping powers, when a dense $e-h$ plasma was produced, was determined using two methods, namely, by analyzing the luminescence line shape[4] and by deriving it from the pumping power density (under conditions of the experiment, the lifetime of nonequilibrium carriers was always much longer than both the laser pulse width and the width of the time gate during which luminescence was detected). The values of N determined by the two methods agreed within 10%, which indicates that nonequilibrium carriers were effectively contained in the quantum well. At lower pumping power densities, when the excitonic line dominated in the luminescence spectrum, the concentration was derived from the pumping power density under the assumption that the fraction of carriers contained in the quantum well was constant as a function of the pumping power density.

3. EXPERIMENTAL RESULTS

Figure 1 shows luminescence spectra of a GaAs/AlGaAs quantum well recorded over a wide range of pumping densities at an ambient temperature of 77 K. For comparison, an excitonic spectrum recorded at 4.2 K is shown by a dashed line in Fig. 1a. This curve demonstrates that the full width at half maximum (FWHM) Δ_x of the exciton line at liquid helium temperatures is 1.4 meV. This linewidth is due to local-

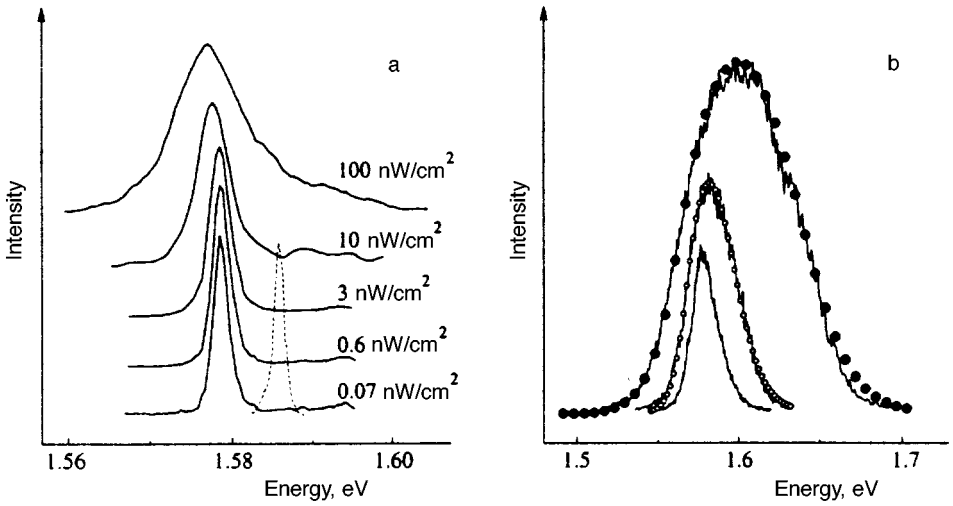

FIG. 1. Luminescence spectra of a GaAs/AlGaAs quantum well at 77 K. For comparison, the dashed line in Fig. 1a plots the exciton luminescence spectrum at 4.2 K.

ization of excitons at potential fluctuations caused by the inhomogeneities in the quantum well width and the content of Al in AlGaAs.[5] Inhomogeneous broadening of the exciton line of about 1 meV is typical of high-quality quantum wells with $L=5$ nm.[5] As the temperature is increased to 77 K at a low pumping power density, the excitonic spectral line broadens to $\Delta_x=2.2$ meV. In the range of low pumping powers, the linewidth is almost constant for photogenerated carrier densities of up to $N\approx 10^9$ cm^{-2}. This leads us to a conclusion that the increase in Δ_x in the temperature range between 4 K and 77 K is mainly caused by the increase in the exciton state damping due to the exciton–phonon scattering, i.e., it is a manifestation of the increase in the homogeneous linewidth of the excitonic line.[1,6]

Figure 1 shows that the exciton line monotonically broadens with the pumping power density W for $W>3$ nW/cm^2. This broadening is an indication of additional exciton damping due to collisions among particles, primarily exciton–electron collisions. The exciton binding energy in the quantum well is comparable to kT at 77 K, and the excitonic gas is highly ionized in the range of densities up to fairly high values. Besides, the exciton–exciton interaction (involving two neutral particles) is notably weaker than the exciton–electron interaction.

At pumping power densities $W>100$ nW/cm^2, the carrier concentration is higher than the critical value for the Mott transition from the excitonic gas to the $e-h$ plasma. Figure 1b clearly shows that the shape of the recombination line in this case is in a good agreement with calculations based on the plasma approximation,[3] and the density and temperature of the $e-h$ plasma can be derived from the shapes of experimental curves.[7] Our fits of calculations to the experimental line shapes indicate that the temperature of the $e-h$ plasma increases from 90 K at $N=5\times 10^{11}$ cm^{-2} to 150 K at $N=2\times 10^{12}$ cm^{-2}. Figure 1 also clearly demonstrates that there are no peaks in the behavior of the linewidth in the density range corresponding to the transition from the excitonic gas to an $e-h$ plasma. This should have been expected, because the excitonic line broadening just below this transition due to interaction between particles is approximately equal to the excitonic Rydberg, which is, in turn, comparable near this transition to the Fermi energy in the $e-h$ plasma.[8]

Figure 2 displays a decay of luminescence from the quantum well, $I(t)$, at various pumping power densities. The luminescence decay time constant τ falls monotonically with the density of carriers in the quantum well. In a general case, when the radiation lifetime τ_r of the excitonic system recombination is density-dependent, the time constant τ is related to τ_r by the equation

$$\tau=\eta\tau_r/(1+\eta d\tau_r/dt), \tag{1}$$

where $\eta=\tau_{nr}/(\tau_r+\tau_{nr})$ is the luminescence quantum efficiency and τ_{nr} is the nonradiation lifetime. Equation (1) is derived from the relations

$$dI/dt=-I/t, \tag{2}$$

$$I=N/\tau_r, \tag{3}$$

$$dN/dt=-N(1/\tau_r+1/\tau_{nr}). \tag{4}$$

Measurements of the quantum efficiency as a function of electron–hole pair density in the quantum well at 77 K are plotted in Fig. 3. In the range of high densities ($N=10^{10}$–10^{11} cm^{-2}) the quantum efficiency is constant. Measurements taken at lower temperatures indicate that η is

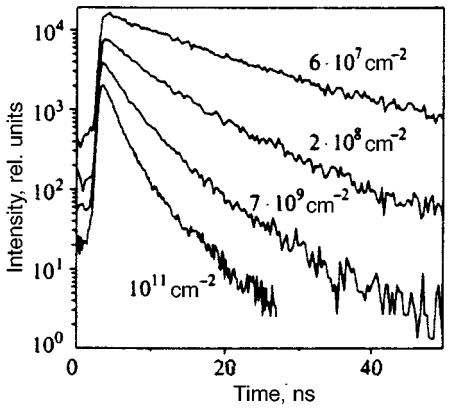

FIG. 2. Luminescence decay curves for the quantum well at different densities.

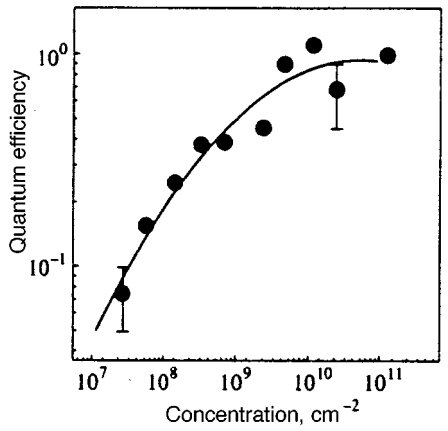

FIG. 3. Quantum efficiency as a function of $e-h$ pair density in the quantum well at 77 K.

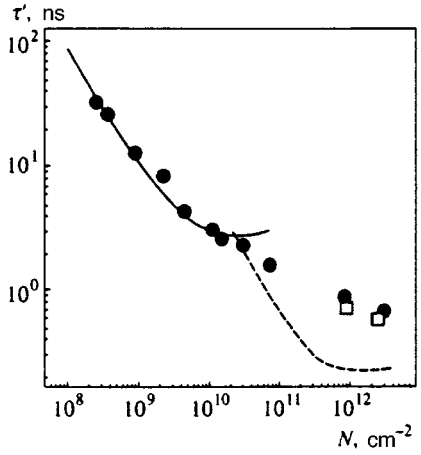

FIG. 5. Lifetime τ' as a function of $e-h$ pair concentration. Measurements are plotted by full circles, calculations by Eqs. (1), (9)–(12) by the solid line. The dashed line shows the calculated $e-h$ plasma lifetime at 77 K. Open squares show calculations at $N=10^{12}$ cm^{-2}, $T=150$ K and $N=3\times10^{12}$ cm^{-3}, $T=280$ K.

also constant in this density range as the temperature drops down to 2 K, although the lifetime in this range drops more than threefold. Therefore, in this range of densities we assume $\eta\sim1$. Figures 2 and 3 show that τ increases and the quantum efficiency η drops as the density falls below 10^{10} cm^{-2}, so the radiation lifetime becomes comparable to τ_{nr} in this density range. In the range $N<10^8$ cm^{-2}, the quantum efficiency η is lower than 0.1, i.e., the nonradiational channel of recombination dominates.

4. DISCUSSION

The exciton line FWHM as a function of the density of e-h pairs is given in Fig. 4. In the range $N<10^{11}$ cm^{-2} the function $\Gamma(N)$ is linear. It can be approximated using the expression[9]

$$\Delta_x(N_e)=\Delta_{x0}+\Gamma_e(N_e), \tag{5}$$

where

$$\Gamma_e(N_e)=g_e\pi R a_x^2 N_e,$$

R and a_x are the exciton binding energy and Bohr radius, respectively, N_e is the number of decoupled $e-h$ pairs, and g_e is the constant of electron–exciton interaction. The density of free electrons can be determined from the equality between chemical potentials of excitons and free electrons and holes in equilibrium:

$$\mu_x=\mu_e+\mu_h, \tag{6}$$

where $\mu_{x,e,h}$ are the chemical potentials of excitons, electrons, and holes, respectively.

It follows from the approximation that $g_e=9.5$. This value is in agreement with earlier estimates of g_e based on the four-wave mixing measurements:[9] $g_e=10.2$. The effect of exciton–electron collisions on the width of the luminescence line has been quantitatively estimated using the formalism developed by Feng and Spector.[8] The homogeneous linewidth is[8]

$$\Gamma_e=\frac{4\hbar^2}{\pi M}\int_0^\infty dk k^2 Q f\left(\frac{2m_e+m_h}{m_e+m_h}k\right),$$

where $M=m_e(m_e+m_h)/(2m_e+m_h)$, $m_e(m_h)$ is the electron (hole) effective mass, and f is the Fermi distribution function. The scattering cross section in this case is expressed as

$$Q=4\pi\left(\frac{M}{m_e}\right)^2 k^{-1}\int_0^\pi d\theta K^{-2}\left(\left[1+\left(\frac{m_e K}{4m_h}\right)^2\right]^{-3/2}-\left[1+\left(\frac{K}{4}\right)^2\right]^{-3/2}\right)^2,$$

where $K=2ka_x\sin(\theta/2)$ and θ is the scattering angle. As a result, we have derived the constant of the exciton–electron interaction $g_e=12.4$, which is in a fair agreement with the experimental data.

Figure 5 shows the function $\tau'(N)=\eta\tau(N)$ obtained by processing the data plotted in Figs. 2 and 3. It follows from Eq. (2) that $\tau'(N)$ equals the radiation lifetime divided by the factor $1+\eta d\tau_r/dt$, which is, as will be shown below, approximately equal to 2. Figure 5 clearly shows that in the range $N=3\times10^7-10^9$ cm^{-2} the time $\tau'(N)$ decreases by more than one order of magnitude in inverse proportion to

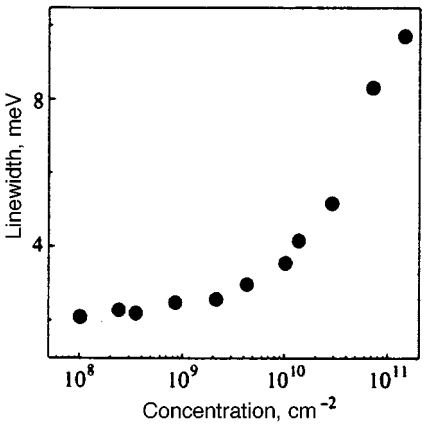

FIG. 4. FWHM of the exciton line versus density of $e-h$ pairs.

the density. At higher excitation densities this dependence is weaker. The increase in the radiation lifetime at low N is caused by ionization of excitons.

The fraction of nonionized excitons in the photoexcited system is

$$\alpha=\frac{N_x}{N}=1-\frac{K}{2N\sqrt{(1+4N/K)}}, \tag{7}$$

where

$$K=\frac{N_eN_h}{N_x}=\frac{m_em_hkT}{\pi\hbar^2m_x}\exp\left(-\frac{R}{kT}\right), \tag{8}$$

and m_x is the exciton mass. Since the exciton Rydberg in the quantum well (11.5 meV) is comparable to kT at 77 K, the ionization degree of excitons at low densities is high and $\alpha \propto 1/N$ (Fig. 5). Luminescence due to free electrons and holes is negligible, so the radiation lifetime of the two-dimensional system can be expressed by

$$\tau_r(N)=\tau_x(T)/\alpha(N). \tag{9}$$

Here $\tau_x(T)$ is the exciton radiation lifetime. It is related to the reduction lifetime τ_0 for excitons with $k<k_0$ by

$$\tau_x(T)=3m_xkT\tau_0/\hbar^2k_0^2. \tag{10}$$

In the range of densities $N>10^{10}$ cm^{-2}, when the exciton line FWHM increases (Fig. 4), the effect of collisions among particles on τ_x should be also taken into account. In a general case, the time τ_x is related to the homogeneous broadening Γ_h of the luminescence line as follows:[2,6]

$$\tau_x\propto\frac{\hbar\Gamma_h}{1-\exp(-\hbar\Gamma_h/kT)}\tau_0. \tag{11}$$

At low carrier concentrations $\hbar\Gamma_h\ll kT$, the lifetime satisfies $\tau_x \propto kT$ and is independent of N. However, the contribution of exciton–electron collisions becomes important at higher densities, when the exciton damping increases and becomes comparable to kT. In particular, estimates based on Eq. (11) yield an increase in τ_x by nearly half at $N=3\times10^{11}$ cm^{-2} when Γ increases to 8 MeV as derived from the luminescence line FWHM.

In addition, note that at high densities the exciton wave function is modified by interparticle interaction, which leads to a change in the exciton oscillator strength f_x, and therefore in τ_0 (since $\tau_0 \propto 1/f_x$, see Ref. 10). In fact, an electron or hole, either free or bound in an exciton, can be scattered only to an unoccupied cell of the k-space. Therefore the scattering rate of carriers decreases at high density. The modification of the exciton wave function results in a smaller oscillator strength since it satisfies $f_x \propto |\Psi(r=0)|^2$, hence longer τ_0 and τ_x.

The contribution of interparticle interaction to the oscillator strength can be taken into account in perturbation theory with the Coulomb interaction treated as a perturbation. A similar technique was used by Schmitt–Rink *et al.*,[10] who studied the many-body effects on excitonic absorption. To first order, the oscillator strength is given by

$$f_x\propto|\Psi_0(r=0)|^2\left[1-\pi a_x^2N_x-(\pi^2-\pi)a_x^2N_e+\frac{\pi\ln 2}{2\sqrt{3}}(k_0a_x)^2\frac{\mu}{m_x}\right], \tag{12}$$

where

$$k_0=\frac{4\pi N_ee^2}{\epsilon_0kT},$$

μ is the exciton reduced mass, and ϵ_0 is the material dielectric permittivity. The ratio between N_x and N_e is determined by Eq. (7). This result has been obtained in the limit $k_0a_x\ll1$ and $R>kT$, i.e., Eq. (12) is valid for $N_e<10^{11}$ cm^{-2}. The second condition corresponds to the case $R=11.5$ meV and $kT=6.6$ meV.

The first two terms in Eq. (12) are due to the exchange interaction between excitons, and between an exciton and free carriers, respectively. The third term is the contribution of recombination of a free electron (hole) with a hole (electron) bound in an exciton. This term is positive, i.e., it decreases the exciton lifetime, and its contribution is significant at $N\sim10^{10}$ cm^{-2}.

The solid trace in Fig. 5 corresponds to the function $\tau'(N)$ calculated by Eqs. (1), (9)–(12) with due account of exciton ionization, exciton–electron collisions, and changes in f_x. The only adjustable parameter is the radiation time τ_0. It was selected to fit the calculations to experimental data in the range of low densities, where interparticle interaction can be neglected. The value $\tau_0=24$ ps derived from this fitting is in a fair agreement with the calculations ($\tau_0=16$ ps for a AlGaAs/GaAs quantum well with $L=50$ Å) based on Ref. 2.

The calculation of $\tau'(N)$ shown in Fig. 5 by the solid trace is in a good agreement with measurements for carrier densities of up to $N\approx7\times10^{10}$ cm^{-2}. At higher densities, the calculations of $\tau'(N)$ are notably different from measurements, which indicates that in this region excitonic correlations are no longer dominant and the lifetime of $e-h$ pairs should be calculated in the plasma approximation.

The lifetime of dissociated $e-h$ pairs in the quantum well can be calculated similarly to that of excitons.[2] The inverse lifetime of a pair can be expressed as follows:

$$\tau_{eh}^{-1}=\frac{1}{N}\frac{2\pi}{m_0c_0\sqrt{\epsilon_0}}e^2mE_{eh}\sum\mu_{vc}\left|\int\psi_c\psi_vdV\right|^2\times\frac{1}{\hbar^2}\frac{1}{2\pi^2}\int n_kn_{k'}\delta(\mathbf{k}-\mathbf{k}')d^2kd^2k', \tag{13}$$

where n_k is the fermion distribution function, and $\psi_{c(v)}$ is the envelope of the wave function in the conductance (valence) band. It follows from Eq. (13) that the usually accepted density dependence of the $e-h$ pair lifetime $\tau \propto n^{-2}$ is valid only in the case of nondegenerate fermion distribution, whereas in the limit $N\pi\hbar^2/m\gg kT$ of degenerate fermion statistics, the lifetime of $e-h$ pairs becomes independent of the density and approaches a constant which equals 0.24 ns for the parameters of the quantum well in question. This time is about an order of magnitude longer than the lifetime of

excitons for $k<k_0$, but considerably shorter than the exciton lifetime at 77 K: $\tau_x=0.8$ ns. The measured and calculated radiative lifetimes in the region of high densities are compared in Fig. 5, where the calculated dependence is indicated by a dashed line. Even at very high densities, $N>10^{12}$ cm^{-2}, the lifetime of the $e-h$ plasma calculated for $T=77$ K is notably shorter than measured. This is not surprising because, as was noted above, the temperature of the photoexcited $e-h$ plasma in a quantum well is higher than 200 K at such densities. At so high temperatures, holes are nondegenerate, which increases the radiative lifetime of the $e-h$ pairs. The two points indicated by open squares in Fig. 5 correspond to calculations for $N=10^{12}$ cm^{-2} at $T=150$ K and for $N=3\times10^{12}$ cm^{-2} at $T=280$ K, i.e., at temperatures derived from luminescence spectra of the $e-h$ plasma. These calculations are in good agreement with experimental values.

It is obvious that the lower boundary of the region where the plasma approximation can be used in calculating τ' is determined by the emergence of exciton-like correlations in the plasma. Excitonic correlations lead to a larger overlap between the electron and hole wave functions, hence to a shorter lifetime. Figure 5 indicates that this transition occurs near the density $N\propto10^{11}$ cm^{-2}, which corresponds to the nondimensional parameter $r_s=1/\pi a_x^2 N\approx3$, in agreement with the expected value for the exciton-to-plasma transition in a dense electron–hole system.

5. CONCLUSION

Under conditions of equilibrium between excitons and electron–hole plasma, we have studied the effect of interparticle interaction in a GaAs/AlGaAs quantum well on the width of the luminescence line and carrier lifetime. At low excitation densities, the role of thermal ionization of excitons has been considered. At higher excitation levels, including those corresponding to the region of the transition from the exciton gas to electron-hole plasma, the effects of exciton–electron and exciton–exciton collisions and ionization of excitons have been analyzed.

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