Kinetics of indirect photoluminescence in $GaAs/AI_xGa_{1-x}As$ double quantum wells in a random potential with a large amplitude

L. V. Butov,*) A. V. Mintsev, and A. I. Filin

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

K. Eberl

Max-Planck-Institut für Festkörperforschung, 70569 Stuttgart, Germany (Submitted 10 November 1998) Zh. Éksp. Teor. Fiz. **115**, 1890–1905 (May 1999)

The kinetics of indirect photoluminescence of GaAs/Al_xGa_{1-x}As double quantum wells, characterized by a random potential with a large amplitude (the linewidth of the indirect photoluminescence is comparable to the binding energy of an indirect exciton) in magnetic fields $B \le 12$ T at low temperatures $T \ge 1.3$ K is investigated. It is found that the indirectrecombination time increases with the magnetic field and decreases with increasing temperature. It is shown that the kinetics of indirect photoluminescence corresponds to singleexciton recombination in the presence of a random potential in the plane of the double quantum wells. The variation of the nonradiative recombination time is discussed in terms of the variation of the transport of indirect excitons to nonradiative recombination centers, and the variation of the radiative recombination time is discussed in terms of the variation of the population of optically active excitonic states and the localization radius of indirect excitons. The photoluminescence kinetics of indirect excitons, which is observed in the studied $GaAs/Al_xGa_{1-x}As$ double quantum wells for which the random potential has a large amplitude, is qualitatively different from the photoluminescence kinetics of indirect excitons in AlAs/ GaAs wells and GaAs/Al_xGa_{1-x}As double quantum wells with a random potential having a small amplitude. The temporal evolution of the photoluminescence spectra in the direct and indirect regimes is studied. It is shown that the evolution of the photoluminescence spectra corresponds to excitonic recombination in a random potential. © 1999 American Institute of Physics. [S1063-7761(99)02605-0]

1. INTRODUCTION

The neutral system consisting of spatially separated twodimensional layers of electrons and holes in double quantum wells has been widely studied in recent years.^{1–12} This system is of interest primarily because of the possibility of constructing structures with the required architecture and a low rate of indirect (interwell) recombination. Since the effective carrier temperature is determined by the ratio of the relaxation and recombination times, the low indirectrecombination rate makes it possible to obtain a neutral electron-hole system of a high density with a low effective temperature. A number of theoretical treatments have shown that in a system of spatially separated layers of electrons and holes in double quantum wells at low temperatures collective states can be observed, including a condensate of indirect excitons similar to the Bose-Einstein condensate bosons.^{13–20} An interesting particular case is a system of spatially separated layers of electrons and holes in a strong magnetic field perpendicular to the plane of the well. A number of theoretical studies have shown that the critical conditions for condensation of excitons are improved in a strong magnetic field as a result of complete quantization of the energy spectrum of electrons and holes^{15,16} and as a result of the lifting of spin degeneracy. Theory predicts that when the distance between the electron and hole layers is small, $d \leq l_B$ ($l_B = \sqrt{\hbar c/eB}$ is the magnetic length), the ground state of the system is determined by the electron-hole interaction and is an exciton condensate, while for large distances, $d \geq l_B$, the ground state is determined by electron-electron and hole-hole interaction and is an incompressible Fermi liquid or Wigner crystal of electrons and holes.^{18,19}

An inevitable property of semiconductor quantum wells and double quantum wells is the existence of a random potential produced in the plane of a well by irregularities of the interfaces, composition fluctuations, defects, and impurities. A random potential qualitatively affects the properties of the system. Specifically, a strong random potential destroys possible collective states (see Ref. 12 and citations there). No theory of a system of spatially separated electron and hole layers in the presence of a random potential is yet available. We shall parametrize the magnitude of the potential by the ratio of the binding energy of an indirect exciton to the width of the indirect luminescence line, determined by the amplitude of the random potential, E_I/Δ_{PL} . In terms of the parameters d, l_B , E_I , and Δ_{PL} (in a zero magnetic field the analog of l_B is the Bohr radius of an indirect exciton), four

classes of spatially separated electron-hole systems in double quantum wells can be distinguished. Class B1 consists of double quantum wells with a small effective distance between the layers and a weak disorder $(d \leq l_B, E_I \geq \Delta_{PL})$. According to theoretical studies, $^{13-20}$ for this class of double quantum wells the ground state of the system at low temperatures should be an excitonic condensate, and the critical conditions for condensation of excitons are improved in a strong magnetic field.^{15,16} Class B2 consists of double quantum wells with a small distance between the layers and strong disorder $(d \leq l_B, E_I \leq \Delta_{PL})$. Class A1 consists of double quantum wells with a large distance between the layers and weak disorder $(d \ge l_B, E_I \ge \Delta_{PL})$. According to theory,^{18,19} for this class of double quantum wells the ground state of the system at low temperatures should be an incompressible Fermi liquid or a Wigner crystal of electrons and holes. Class A2 consists of double quantum wells with a large distance between the layers and strong disorder (d $\geq l_B$, $E_I \leq \Delta_{PL}$). This classification must be supplemented by the carrier density and the temperature, which determine the phase boundaries. Interclass transitions between the classes are probably, continuous. Thus a transition between the classes B1 and A1 and a transition between the classes B2 and A2 can be accomplished by increasing the magnetic field, while a transition between the classes B1 and B2 and between the classes A1 and A2 can be followed by studying double quantum wells with various degrees of disorder.

At experimental investigation of $\Gamma - X_{\tau}$ AlAs/GaAs double quantum wells, characterized by a small distance between the electron and hole layers, $d \approx 3-4$ nm, $E_I \sim 10$ meV, and Δ_{PL} from 3 to 6 meV, which therefore belong to the classes B1 and B2, implying condensation of indirect excitons in strong magnetic fields at low temperatures have been observed: an anomalous increase of the diffusion coefficient and radiative recombination rate of excitons, interpreted as the appearance of superfluidity of excitons and superradiance of an excitonic condensate,¹² and anomalously large fluctuations of the total intensity of the photoluminescence of excitons, interpreted as critical fluctuations near a phase transition, which are associated with instability of condensate domains.⁸ As the disorder in the experimental AlAs/ GaAs wells increased, these anomalies became weaker and disappeared, which corresponded to a transition from class B1 to class B2.¹²

The kinetics of photoluminescence in double quantum wells belonging to the class A1 has been investigated in a zero magnetic field. Specifically, double quantum wells GaAs/Al_xGa_{1-x}As with $d \approx 12$ nm, $E_I \sim 5$ meV, and $\Delta_{PL} = 1.3$ meV have been investigated.²¹ A sharp increase of intensity and narrowing of the photoluminescence line of indirect excitons were found after the pulsed laser excitation was switched off at low temperatures and high exciton densities. The effect was described by a rapid increase in the population of optically active excitonic 2D states.²¹

In the present work we investigate the optical properties of a system consisting of spatially separated layers of electrons and holes in class A2 wells. Specifically, we investigate GaAs/Al_xGa_{1-x}As double quantum wells with $d \sim 11$ nm and $E_I \sim \Delta_{PL} \sim 6$ meV. The experimental data are compared with the analogous dependences for class B1, B2, and A1 double quantum wells.

2. SAMPLE AND EXPERIMENTAL PROCEDURE

In gate voltage tunable $n^+ - i - n^+$ heterostructure with a single GaAs/Al_xGa_{1-x}As double quantum well, which was adjusted by varying the gate voltage, was grown by molecular-beam epitaxy on an n^+ -GaAs substrate. The *i* layer consists of two, 5 nm wide, GaAs quantum wells separated by a 5.5 nm thick Al_{0.35}Ga_{0.65}As barrier and surrounded by 55 nm thick Al_{0.35}Ga_{0.65}As barriers. The band diagram of the *i* layer of the structure in the indirect regime is shown in Fig. 1. The 1100 nm thick n^+ layers on the substrate side and the 110 nm thick layer on the surface side were doped with Si to a density $N_{\rm Si} = 5 \times 10^{17} \, {\rm cm}^{-3}$. To improve the electrical contact, δ doping with $N_{\rm Si} = 10^{13} \, {\rm cm}^{-2}$ was performed at 10 nm from the surface. As a result of the high doping density the n^+ layers are of a metallic character, and the gate voltage V_g applied between the substrate and the surface decreases in the *i* layer. The front gate consisted of a frame around a mesa with a 200 \times 200 μ m² window.

The sample was placed in a helium cryostat with a superconducting solenoid. Excitation and detection were performed through a 200 μ m in diameter optical light guide, placed 300 μ m from the mesa surface. The carriers were excited by a pulse semiconductor laser ($\hbar \omega = 1.85$ eV). The laser pulse was approximately square with ~50 ns duration and ~1 ns edges. The temporal resolution of the detection system was 0.5 ns. A double grating monochromator, a photomultiplier, and a time-correlated photon counting system with time resolution were used to detect the signal.

3. PHOTOLUMINESCENCE KINETICS IN THE INDIRECT REGIME

The indirect regime in the $n^+ - i - n^+$ structure of the $GaAs/Al_xGa_{1-x}As$ double quantum wells occurs for finite values of V_g . The V_g dependence of the photoluminescence spectra and kinetics are shown in Figs. 1a and 1c. For V_g ≤ 0.3 V the energy of the photoluminescence line is essentially independent of V_g , and the decay of this line is characterized by a short lifetime. Therefore the ground state of the system for $V_g \leq 0.3$ V is a direct exciton. For $V_g \geq 0.4$ V, increasing V_g produces an approximately linear energy shift of the principal photoluminescence line and increases the decay time of the line. Therefore for $V_{g} \gtrsim 0.4$ V electrons and holes in the ground state of the system occupy different quantum wells; this corresponds to an indirect regime. The magnitude of the shift of the indirect photoluminescence line is determined by the electrostatic energy eFd, where e is the electron charge and F is the electric field in the z direction. A transition from the direct regime into the indirect regime occurs in a nonzero electric field F_{D-I} . This corresponds to excitonic recombination with direct and indirect exciton energies $\mathcal{E}_D = E_g - E_D$ and $\mathcal{E}_I = E_g - E_I - eFd$, where E_g is the energy gap, including the electron and hole quantization energy in the double quantum well, E_D and E_I are the binding energies of the direct and indirect excitons. For $F = F_{D-I}$ the energy difference between the single-particle direct and in-



FIG. 1. Time-integrated photoluminescence spectrum (a, b) and photoluminescence kinetics, measured at the maximum of the principal line (c, d), as a function of the gate voltage (a, c) and magnetic field (b, d) at T=1.3 K and $W_{ex}=10$ W/cm². The dashed line corresponds to the trailing edge of the 50 ns laser excitation pulse. The spectra and kinetics are shifted along the ordinate for clarity. The direct and indirect photoluminescence lines are labeled by D and I, respec-The band tively. diagram of the GaAs/Al_xGa_{1-x}As double quantum well is shown in the inset.

direct pair states is equal to the difference between the binding energies of the direct and indirect excitons: $eF_{D-I}d$ $=E_D-E_I$ (see Refs. 7 and 9 and the references cited there).

The width of the indirect-exciton line is determined by the random potential in the plane of the double quantum well. Several types of disorder, making the main contribution to the inhomogeneous broadening of the indirect photoluminescence line, can be distinguished: a) interfacial fluctuations; b) electric-field fluctuations in the z direction; c) charged impurities (other types of disorder, such as composition fluctuations, neutral impurities, and defects also exist, but their contribution to disorder is, as a rule, smaller). Fluctuations of the electric field in the z direction give rise to in-phase fluctuations of the potential for an electron and for a hole, so that they can be treated as fluctuations of the potential for the indirect exciton center of mass. Charged impurities give rise to antiphase fluctuations of the potential for an electron and for a hole. Strong fluctuations due to charged impurities can result in breakup of the exciton and independent localization of an electron and a hole in a local minimum of the random potential.¹¹ Interfacial fluctuations give rise to in-phase fluctuations of the potential for an electron and a hole in single quantum wells; for an indirect exciton (electron-hole pair) in GaAs/Al_xGa_{1-x}As wells interfacial fluctuations produce independent fluctuations of the potential for an electron and a hole. Fluctuations of the electric field in the z direction are determined primarily by fluctuations of the extent of the section where gate voltage drops. To reduce such fluctuations to a minimm the n^+ layers should possess good conductivity, and the *i* layer should be a good insulator. Then the region where gate voltage drops is clearly determined—it is the *i* layer.

Since fluctuations of the electric field in the z direction are specific to indirect excitons in double quantum wells, their relative contribution to the inhomogeneous broadening of the indirect photoluminescence line can be estimated by comparing the widths of the direct and indirect photoluminescence lines. In the GaAs/Al_xGa_{1-x}As double quantum well studied the indirect photoluminescence line (6.5 meV for $V_g = 0.8$ V) is even narrower than that of the direct photoluminescence line (14.7 meV in the direct regime with $V_o = 0$ and 9.4 meV in the indirect regime with $V_o = 0.8$ V), indicating that fluctuations of the electric field make a negligibly small contribution to the broadening of the line. The quantum wells in the experimental structure are narrow: 5 nm corresponds to 18 monolayers. In narrow wells interface fluctuations make the main contribution to inhomogeneous broadening of the line. Thus, in the experimental double quantum well a fluctuation of the well width by one monolayer with an infinite terrace produces a change $\delta_m \sim 5 \text{ meV}$ in the electron energy and $\sim 2 \text{ meV}$ in the hole energy. The finiteness of the terraces results in quantization of the electron and hole energies in the plane. This produces states in the entire energy interval from 0 to δ_m . The observed width of the indirect photoluminescence line corresponds to δ_m (Fig. 1a). This confirms that interfacial fluctuations make the dominant contribution to the broadening of the line. Therefore the large magnitude of the random potential in the experimental GaAs/Al_xGa_{1-x}As double quantum wells is due primarily to the small width of the quantum wells. The shape of the photoluminescence line reflects the energy distribution of excitons over local energy minima in the random potential. The direct-photoluminescence line is probably broadened in part because the widths of the two quantum wells are different and the direct photoluminescence line includes two spectrally unresolved lines from the two quantum wells.

In the experimental structure in the indirect regime both radiative and nonradiative recombination contribute to the recombination of indirect excitons. The observed decrease of the recombination rate with delay time (see, for example, Fig. 1) is characteristic for both radiative and nonradiative recombination of indirect excitons. The radiative recombination rate of an exciton is proportional to the population of the optically active 2D excitonic states (with quasimomenta $k \leq k_0 = \mathcal{E}/\hbar c$, where *c* is the velocity of light in the medium) and it increases with the extent of the wave function of the exciton center of mass in the plane, called the coherent area of the exciton (the radiative recombination rate saturates when the coherent length of the reciprocal of the wavelength of the emitted light is reached). $^{22-26}$ As a result of the spread of the localization radius in a random potential, the radiative recombination time of excitons is nonuniform over the plane of the double quantum well. As a result, the radiative recombination rate decreases with increasing delay time, since it is excitons with a large localization radius that are the first to recombine in the photoluminescence decay process.

Moreover, as the delay time increases, electrons and holes independently localized in the local minima of the random potential and having, as a result of the spatial separation in the plane (in addition to separation in the z direction for indirect electron-hole pairs), a low radiative-recombination rate make an increasingly larger contribution to the intensity of photoluminescence. Since the independently localized electrons and holes with the smallest separation in the plane are the first to recombine in the process of photoluminescence decay, the radiative recombination rate of independently localized electrons and holes also decreases with increasing delay time.¹¹ In narrow double quantum wells, characterized by a low diffusion coefficient of indirect excitons, nonradiative recombination is determined by exciton transport toward nonradiative recombination centers.12,27-29 The decrease of the nonradiative recombination rate with the delay time is due to the monotonic decrease of the exciton diffusion coefficient. As a result, more and more localized excitons, which have not had enough time to reach the centers of nonradiative recombination and to recombine, predominate in the spectrum.

The magnetic-field and temperature dependences of the indirect-photoluminescence spectra and kinetics are displayed in Figs. 1b, 1d and 2. The temperature dependence of the indirect-photoluminescence spectra and kinetics in strong magnetic fields, $B \leq 12$ T, is qualitatively the same as the analogous dependence for B=0. The corresponding initial decay time of photoluminescence, τ , and the integrated intensity of indirect photoluminescence, I_{PL} , are shown in Fig. 3. The time τ increases with magnetic field (Figs. 1d and 3a) and decreases with increasing temperature (Fig. 2b, 3b, and 3c).

The observed photoluminescence kinetics in GaAs/Al_xGa_{1-x}As double quantum wells is qualitatively different from the kinetics in the B1-, B2-, and A1-class double quantum wells investigated: in contrast to GaAs/Al_xGa_{1-x}As double quantum wells with weak disorder (A1 class),²¹ in the experimental GaAs/Al_xGa_{1-x}As double quantum wells a sharp increase in the intensity of photoluminescence after the laser excitation pulse ceases is not observed; in contrast to AlAs/GaAs double quantum wells (B1 class),^{8,12} in the experimental GaAs/Al_xGa_{1-x}As wells a sharp decrease of τ in strong magnetic fields is not observed. The intensity of pho-



FIG. 2. Time-integrated photoluminescence spectrum (a) and indirect photoluminescence kinetics, measured at the maximum of the indirect line (b), as a function of temperature with $V_g = 0.8$ V, $W_{ex} = 10$ W/cm², and B = 0. The spectra and kinetics are shifted along the ordinate for clarity. The dashed line corresponds to the trailing edge of the 50 ns laser excitation pulse. The direct and indirect photoluminescence lines are labeled by *D* and *I*, respectively.

toluminescence does not exhibit a sharp increase in the studied double quantum well with a large random potential after the laser excitation pulse ceases because the large potential smears the boundary between the optically active (with $k \le k_0$) and passive ($k > k_0$) excitonic states³⁰, so that the possible sharp increase in the population of optically active states does not result in a higher photoluminescence intensity. The absence of a sharp decrease of τ in strong magnetic fields, which is observed in AlAs/GaAs double quantum wells (B1 class) and indicates the appearance of superfluidity of excitons,¹² indicates the absence of collective effects in the studied double quantum wells with a large random potential and a low binding energy of the indirect exciton (A2 class), as expected (see Sec. 1).

The observed monotonic increase of the lifetime with the magnetic field is characteristic for radiative and nonradiative single-exciton recombination in a random potential. The change in the radiative lifetime of excitons with increasing magnetic field is determined by the ratio of the increase in the oscillator strength of the exciton as a result of a decrease in the exciton radius³¹ and the decrease in the oscillator





FIG. 3. Measured indirect-recombination times τ (\blacksquare) and the integrated intensity of indirect photoluminescence together with the deduced radiative and nonradiative indirect recombination times τ_r (\bigcirc) and τ_{nr} (\triangle) versus the magnetic field with $V_g = 0.8$ V and $W_{ex} = 10$ W/cm².

strength of an exciton as a result of a decrease in the localization radius of the exciton $^{22-26}$ in a magnetic field as a result of an increase in the mass of the magnetoexciton.^{31,32} The increase in the nonradiative lifetime of an exciton (determined by transport toward nonradiative recombination centers) with the magnetic field is due to a decrease in the diffusion coefficient caused by an increase in the magnetoexciton mass.³³ A decrease of the diffusion coefficient with increasing magnetic field has also been observed for indirect excitons in AlAs/GaAs double quantum wells in weak magnetic fields¹² and for direct excitons in single $GaAs/Al_xGa_{1-x}As$ double quantum wells.³⁴ We note that a monotonic increase of the radiative and nonradiative lifetimes with increasing magnetic field is also characteristic for independently localized electrons and holes and is due to a decrease of the carrier localization radius.

The method described in Ref. 12 was used to distinguish the radiative and nonradiative lifetimes. The radiative lifetime τ_r can be directly extracted from the measured total lifetime τ and the time-integrated photoluminescence intensity I_{PL} . For single-exponential decay of photoluminescence $\tau_r = (G/I_{PL})\tau$, where G is the generation rate of electronhole pairs in double quantum wells (weak nonexponentiality introduces negligible quantitative corrections¹²). The quantity G is unknown; to estimate it the quantum yield with parameters corresponding to maximum I_{PL} was taken to be 1. Then $G = I_{\text{max}}$ and $\tau_r = (I_{\text{max}}/I_{PL})\tau$, where I_{max} is the maximum integrated photoluminescence intensity observed in the experimental double quantum well with $V_g = 0.3$ V (Fig. 1a). The formula $\tau^{-1} = \tau_r^{-1} + \tau_{nr}^{-1}$ was used to find the nonradiative lifetime au_{nr} using the measured value of au and the calculated value of τ_r . The values of τ_r and τ_{nr} found in this manner are shown in Fig. 3.

Note that the parameter dependence of τ_r found by the method indicated above is correct if *G* does not depend on the given parameter. This condition was satisfied for the AlAs/GaAs double quantum wells studied in Ref. 12. However, it is not satisfied in the present investigations of GaAs/Al_xGa_{1-x}As double quantum wells. The photon en-

ergy for photoexcitation is only 100 meV lower than the energy of the Al_{0.35}Ga_{0.65}As barrier, so that as a result of the Franz-Keldysh effect and the tails of the density of states in $Al_{r}Ga_{1-r}As$, the absorption in the barrier layers was substantial. The collection of carriers generated in the barrier layers to double quantum wells made an appreciable contribution to G, comparable to the generation in GaAs layers. Thus the increase in the total photoluminescence intensity with a small applied gate voltage (Fig. 1) is due to an increase in G as a result of the drift of carriers generated in the barrier layers (this effect is absent for photoexcitation with photon energy much less than the gap width in the barrier layers). The drift of carriers generated in the barrier layers in double quantum wells can depend on the magnetic field and temperature. For this reason G is not independent of the parameters, and the method described above for finding the dependence of τ_r on the magnetic field and temperature from the measured values of τ and I_{PL} is not entirely correct for the present experiment. Moreover, an error in estimating the quantum yield for parameters corresponding to maximum I_{PL} will enter in the absolute value of τ_r and τ_{nr} as well as in the magnetic-field and temperature dependences of τ_{nr} . Nonetheless the method employed makes it possible to follow the qualitative variations of τ_r and τ_{nr} as a function of magnetic field and temperature.

It is evident in Fig. 3 that τ_r is virtually independent of the magnetic field and τ_{nr} increases monotonically with the field. This corresponds to the single-exciton behavior described above. As temperature increases, τ_r increases and τ_{nr} decreases (Fig. 3; the opposing behavior of τ_r and τ_{nr} could cause a weak nonmonotonicity of the temperature variations of τ , and the possible increase of τ at low temperatures falls within the experimental error, Figs. 2 and 3). The decrease in τ_{nr} with increasing temperature is characteristic for both single-exciton recombination and recombination of independently localized electrons and holes and is due to the increase in the diffusion of excitons (electrons and holes) toward nonradiative recombination centers as a result of their thermal activation from local minima of the random poten-

tial. The increase in τ_r with temperature is characteristic for excitonic recombination because the population of optically active excitonic states decreases (for a Boltzmann distribution of excitons the fraction of optically active excitons with $k \le k_0$ is $1 - \exp(-E_0/kT)$, where $E_0 = \hbar^2 k_0^2/2m \sim 1$ K; for $T \gg E_0$ the Boltzmann distribution leads to a linear increase of the radiative lifetime of excitons with increasing temperature).^{22–26} However, the observed increase of τ_r with temperature contradicts the model of recombination of independently localized electrons and holes, on the basis of which an increase in temperature should enhance the overlapping of the electron and hole wave functions due to their delocalization. Hence it follows that the random potential in the double quantum well under study is not strong enough to break up excitons, and the fraction of independently localized electrons and holes is low. Note that the magnetic-field and temperature dependences of τ_r and τ_{nr} in the studied GaAs/Al_xGa_{1-x}As double quantum well correspond qualitatively to the analogous dependences for AlAs/GaAs wells (B1 class) in weak magnetic fields.¹² In both cases these dependences correspond to single-exciton recombination.

The photoluminescence spectra and kinetics of indirect excitons are virtually independent of the exciton density, fixed by the laser excitation density $W_{\rm ex}$ (Fig. 4). For $W_{\rm ex}$ = 10 W/cm^2 the density of indirect excitons with lifetime τ =100 ns is estimated to be several times 10¹⁰ cm⁻². As $W_{\rm ex}$ increases, a shift of the indirect exciton line in the direction of high energies is observed. This behavior corresponds to the theoretically predicted increase of the energy of indirect excitons with increasing density^{18,20} and is explained by the repulsive dipole-dipole interaction between indirect excitons for low exciton densities and by the energy shift for high electron-hole densities due to the electric field between the separated electron and hole layers. Moreover, since the degeneracy of the zero-dimensional excitonic state in a local minimum of the random potential is finite (neglecting the exciton-exciton interaction, the degeneracy is $\sim S/a_B^2$, where S is the area of the local minimum and a_B is the Bohr radius of an indirect exciton), an increase of the exciton density results in an increase of the average exciton energy. This effect should also contribute to the observed increase in the indirect-exciton energy with density.

A small decrease of the recombination time of indirect excitons is observed with increasing W_{ex} (Fig. 4b). This behavior is characteristic for single-exciton recombination in a random potential: the exciton localization radius increases with exciton density (since at low density excitons are strongly localized in deep local minima of the potential). This decreases both the radiative-recombination time of excitations^{22–26} and the nonradiative-recombination time of excitons due to transport toward nonradiative-recombination centers.

4. EVOLUTION OF PHOTOLUMINESCENCE SPECTRA IN DIRECT AND INDIRECT REGIMES

In this section we examine the temporal revolution of the photoluminescence spectra. This is the evolution of the energy distribution of excitons (electron-hole pairs) with



FIG. 4. Time-integrated photoluminescence spectrum normalized to the laser excitation density (a) and indirect-photoluminescence kinetics measured at the maximum of the indirect line (b) as a function of the laser excitation density with $V_g = 0.8$ V, T = 1.3 K, and B = 0 and 12 T. The spectra and kinetics are shifted along the ordinate for clarity. The dashed line corresponds to the trailing edge of the 50 ns laser excitation pulse. The direct and indirect photoluminescence lines are labeled by *D* and *I*, respectively.

weight proportional to the radiative-recombination probability. The dependence of the photoluminescence spectra on the time delay is shown in Fig. 5a and 5b, respectively, for the direct and indirect regimes. After the laser excitation pulse ends, both the direct-photoluminescence line in the direct regime and the indirect-photoluminescence line in the indirect regime shift monotonically in the direction of low energies as the time delay increases (Fig. 5). Similar behavior is also observed in strong magnetic fields. The shift of the photoluminescence line with increasing time delay is typical both for excitonic recombination and for recombination of independently localized electron-hole pairs as well as for radiative and nonradiative recombinations. In the first place, as the delay increases, the average energy of photoexcited excitons (electron-hole pairs) in a random potential decreases as a result of the energy relaxation of the carriers: excitons (electrons and holes) migrate in the plane of the double quantum well in search for lower-energy local minima of the potential with the emission of acoustic



FIG. 5. Direct-photoluminescence spectrum in the direct regime with $V_g = 0$ (a) and the indirect-photoluminescence spectrum in the indirect regime with $V_{g} = 0.8$ V (b) versus the delay time, T = 1.3 K, B = 0, and $W_{ex} = 10$ W/cm². The spectra were measured in the time intervals shown in panels c and d. The time-integrated spectra are shown in the top. All spectra are scaled to roughly the same intensity. The curves of photoluminescence kinetics versus energy are shown for the same parameters (c, d); the energies of the detected signal are shown in panels a and b by dashed lines. The spectra and kinetics are shifted along the ordinate for clarity. The dashed line in panels c and d corresponds to the trailing edge of the 50 ns laser excitation pulse. The direct and indirect photoluminescence lines are labeled by D and I, respectively.

phonons. This mechanism of energy relaxation of excitons in a random potential was theoretically examined in Ref. 35 and has been observed for indirect excitons in a double guantum well.^{10,36} In the second place, as the delay time increases, the average energy of the excitons (electron-hole pairs) in a random potential decreases because the higherenergy excitons (independently localized electrons and holes) have higher radiative and nonradiative recombination rates. As the energy of the excitons in a random potential increases, their localization radius increases,³⁷ which results in a higher radiative recombination rate^{22–26} and higher nonradiative recombination rate, due to transport of excitons toward nonradiative-recombination centers. For independently localized electron-hole pairs, the higher-energy pairs also have a higher recombination rate because of their larger localization radius and the corresponding larger overlap between the electron and hole in the plane.¹¹

Another aspect of the dependence of the photoluminescence spectra on the time delay is the energy dependence of the photoluminescence kinetics, shown in Figs. 5c and 5d. As energy decreases, the photoluminescence decay becomes slower and slower, which corresponds to the time dependence of the spectra (Fig. 5a and 5b) and was discussed above.

As a result of the inhomogeneous broadening of the direct and indirect photoluminescence lines, for the appropriate gate voltages a mixed regime in which the direct and indirect photoluminescence energies overlap can be obtained. Such a regime of energy resonance between direct and indirect excitons was considered in Ref. 38 in a study of the photoluminescence of zero-dimensional excitonic states in the local minima of the random potential (natural quantum dots). In our double quantum well the mixed regime appears for $V_g \sim 0.2-0.5$ V; this is evident from the V_g dependence of the photoluminescence spectra (Fig. 1). In the studied double quantum well, characterized by a larger width of the direct-

photoluminescence line than the width of the indirect line, a specific energy dependence of the photoluminescence kinetics is observed in the mixed regime (Fig. 6). This dependence is different from the monotonic decrease of the recombination rate with decreasing energy, as is observed in the direct and indirect regimes. Specifically, in the mixed regime the recombination rate on the initial times of photoluminescence decay depends nonmonotonically on the energy, reaching a minimum at energies corresponding to indirect photoluminescence (kinetics 4-6 in Fig. 6b). The high recombination rate of direct photoluminescence at energies below the indirect-photoluminescence energy (8, 9 in Fig. 6b) indicates that the electron-hole distance in the plane is less than the distance in the z direction. Since the latter distance is ~ 11 nm, the electron-hole distance in the plane is less than the radius of a direct exciton (≈ 10 nm) and especially an indirect exciton, which has a larger radius as a result of the lower binding energy (≈ 20 nm).⁷ A small electron-hole distance in the plane indicates that excitonic recombination predominates over recombination of independently localized electrons and holes.

5. CONCLUSIONS

We have investigated the kinetics of indirect photoluminescence in GaAs/Al_xGa_{1-x}As double quantum wells characterized by a random potential with a large amplitude (the width of the photoluminescence line is comparable to the binding energy of an indirect exciton) and a large distance between the electron and hole layers ($d \sim 11$ nm, which is greater than the magnetic length for $B \ge 5.5$ T) in magnetic fields $B \le 12$ T at low temperatures $T \ge 1.3$ K. It was found that the indirect recombination time τ increases with the magnetic field and decreases with temperature. Analysis of the variation of τ and the total intensity of photoluminescence gave the radiative and nonradiative indirect recombi-



FIG. 6. Energy dependence of the photoluminescence kinetics in the mixed regime corresponding to overlapping of the direct and indirect photoluminescence lines ($V_g = 0.4$ V) at T = 1.3 K, B = 0, and $W_{ex} = 10$ W/cm² (b). The energies of the detected signal are shown in panel a, which shows the time-integrated spectrum. The spectra are shifted along the ordinate for clarity. The dashed line in panel b corresponds to the trailing edge of the 50 ns laser excitation pulse.

nation times, τ_r and τ_{nr} . It was found that τ_{nr} increases with the magnetic field and decreases with increasing temperature, while τ_r is essentially independent of the magnetic field and increases with temperature. It was shown that the kinetics of indirect photoluminescence corresponds to the single-exciton recombination in the presence of a random potential in the plane of the double quantum well. The variation of the nonradiative recombination time was discussed in terms of the variation of the transport of indirect excitons toward nonradiative recombination centers, and the variation of the radiative recombination times was discussed in terms of the variation of the population of optically active excitonic states and the localization radius of indirect excitons. The photoluminescence kinetics of indirect excitons, which was observed in the studied GaAs/Al_xGa_{1-x}As double quantum wells with a random potential having a large amplitude, is qualitatively different from the photoluminescence kinetics of indirect excitons in AlAs/GaAs and GaAs/Al_xGa_{1-x}As double quantum wells when the amplitude of the random potential is small.

The temporal evolution of the photoluminescence spectra in the direct and indirect regimes was examined. It was found that after the laser excitation pulse ceases both the direct photoluminescence line in the direct regime and the indirect photoluminescence line in the indirect regime shift monotonically in the direct of lower energies as the delay time increases. It was shown that the evolution of the photoluminescence spectra corresponds to excitonic recombination in a random potential and is determined by the energy relaxation of excitons and by the energy dependence of the recombination rate.

We thank V. D. Kulakovskiĭ and S. G. Tikhodeev for a discussion of the results obtained in this work as well as the Russian Fund for Fundamental Research (Project 98-02-18371) and the program "Physics of Solid-State Nanostructures" (Project 97-1067) for financial support.

*)E-mail: butov@issp.ac.ru

- ¹Y. J. Chen, Emil. S. Koteles, B. S. Elman, and C. A. Armiento, Phys. Rev. B **36**, 4562 (1987).
- ²M. N. Islam, R. L. Hillman, D. A. B. Miller, D. S. Chemla, A. C. Gossard, and J. H. English, Appl. Phys. Lett. **50**, 1098 (1987).
- ³S. R. Andrews, C. M. Murray, R. A. Davies, and T. M. Kerr, Phys. Rev. B **37**, 8198 (1988).
- ⁴S. Charbonneau, M. L. W. Thewalt, E. S. Koteles, and B. Elman, Phys. Rev. B 38, 6287 (1988).
- ⁵C. C. Phillips, R. Eccleston, and S. R. Andrews, Phys. Rev. B **40**, 9760 (1989).
- ⁶T. Kukuzawa, E. E. Mendez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990); A. Alexandrou, J. A. Kash, E. E. Mendez, M. Zachau, J. N. Hong,
- T. Kukuzawa, and Y. Hase, Phys. Rev. B **42**, 9225 (1990); J. A. Kash, M. Zachau, E. E. Mendez, J. M. Hong, and T. Fukuzawa, Phys. Rev. Lett.
- **66**, 2247 (1991).
- ⁷M. M. Dignamn and J. E. Sipe, Phys. Rev. B **43**, 4084 (1991).
- ⁸L. V. Butov, A. Zrenner, G. Abstreiter, G. Böhm, and G. Weimann, Phys. Rev. Lett. **73**, 304 (1994).
- ⁹L. V. Butov, A. Zrenner, G. Abstreiter, A. V. Petinova, and K. Eberl, Phys. Rev. **52**, 12153 (1995); A. B. Dzyubenko and A. L. Yablocnskii, Phys. Rev. B **53**, 16355 (1996).
- ¹⁰J. E. Golub, S. D. Baranovskii, and P. Thomas, Phys. Rev. Lett. **78**, 4261 (1997).
- ¹¹ V. B. Timofeev, A. I. Filin, A. V. Larionov, J. Zeman, G. Martinez, J. M. Hvam, D. Birkedal, and C. B. Sorensen, Europhys. Lett. **41**, 535 (1998); V. B. Timofeev, A. V. Larionov, A. S. Iozelevich, J. Zeman, G. Martinex, J. Hvam, and K. Sorensen, JETP Lett. **67**, 613 (1998).
- ¹²L. V. Butov and A. I. Filin, Phys. Rev. B **58**, 1980 (1998).
- ¹³ Yu. E. Lozovik and V. I. Yudson, Zh. Eksp. Teor. Fiz. **71**, 738 (1976) [Sov. Phys. JETP **44**, 389 (1976)].
- ¹⁴S. I. Sevchenko, Fiz. Nizk. Temp. **2**, 505 (1976) [Sov. J. Low Temp. Phys. **2**, 251 (1976)].
- ¹⁵ I. V. Lerner and Yu. E. Lozovik, JETP Lett. **27**, 467 (1978); I. V. Lerner and Yu. E. Lozovik, J. Low Temp. Phys. **38**, 333 (1980); I. V. Lerner and Yu. E. Lozovik, Zh. Eksp. Teor. Fiz. **80**, 1488 (1981) [Sov. Phys. JETP **53**, 763 (1981)].
- ¹⁶Y. Kuramoto and C. Horie, Solid State Commun. 25, 713 (1978).
- ¹⁷T. Fukuzawa, S. S. Kano, T. K. Gustafson, and T. Ogawa, Surf. Sci. 228, 482 (1990).
- ¹⁸D. Yoshioka and A. H. MacDonald, J. Phys. Soc. Jpn. 59, 4211 (1990).
- ¹⁹X. M. Chen and J. J. Quinn, Phys. Rev. Lett. 67, 8895 (1991).
- ²⁰X. Zhu, P. B. Littlewood, M. S. Hybersten, and T. M. Rice, Phys. Rev. Lett. **74**, 1633 (1995).
- ²¹L. V. Butov, A. Imamoglu, A. V. Minstev, K. L. Campman, and S. C. Gossard, Phys. Rev. B **59**, 1625 (1999).
- ²² J. Feldman, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliot, Phys. Rev. Lett. **59**, 2337 (1987).
- ²³E. Hanamura, Phys. Rev. B **38**, 1228 (1988).
- ²⁴L. C. Andreani, F. Tassone, and F. Bassani, Solid State Commun. **77**, 641 (1991).
- ²⁵ B. Deveaud, F. Clerot, N. Roy, K. Satzke, B. Sermage, and D. S. Katzer, Phys. Rev. Lett. **67**, 2355 (1991).
- ²⁶D. S. Citrin, Phys. Rev. B 47, 3832 (1993).

- ²⁷F. Minami, K. Hirata, K. Era, T. Yao, and Y. Matsumoto, Phys. Rev. B 36, 2875 (1987).
- ²⁸M. Maaref, F. F. Charfi, D. Scalbert, C. Benoir a la Guillaume, and R. Planet, Phys. Status Solidi B **170**, 637 (1992).
- ²⁹G. D. Gilliland, A. Antonelli, D. J. Wolford, K. K. Bajaj, J. Klem, and J. A. Bradley, Phys. Rev. Lett. **71**, 3717 (1993).
- ³⁰W. Zhao, P. Stenius, and A. Imamoglu, Phys. Rev. B 56, 5306 (1997).
- ³¹I. V. Lerner and Yu. E. Lozovik, Zh. Éksp. Teor. Fiz. **78**, 1167 (1980) [Sov. Phys. JETP **51**, 588 (1980)].
- ³²Yu. E. Lozovik and A. M. Ruvinskiĭ, Zh. Éksp. Teor. Fiz. **112**, 1791 (1997) [JETP **85**, 979 (1997)].
- ³³A. V. Dzyubenko and G. E. W. Bauer, Phys. Rev. B **51**, 14524 (1995).

- ³⁴ M. Jiang, H. Wang, R. Merlin, D. G. Steel and M. Cardona, Phys. Rev. B 48, 15476 (1993).
- ³⁵T. Takagahara, Phys. Rev. B **31**, 6552 (1985).
- ³⁶L. V. Butov and A. I. Filin, Zh. Éksp. Teor. Fiz. **114**, 1115 (1998) [JETP 87, 608 (1998)].
- ³⁷Zh. S. Gevorkyan and Yu. E. Lozovik, Fiz. Tverd. Tela (Leningrad) 27, 1800 (1985) [Sov. Phys. Solid State 27, 1079 (1985)].
- ³⁸A. Zrenner, L. V. Butov, M. Hagn, G. Abstreiter, G. Böhm, and G. Weimann, Phys. Rev. Lett. **72**, 3382 (1994).

Translated by M. E. Alferieff