Pis'ma v ZhETF, vol.67, iss.8, pp.580 - 585 © 1998 April 25

INTERWELL RADIATIVE RECOMBINATION IN THE PRESENCE OF RANDOM POTENTIAL FLUCTUATIONS IN GaAs/AlGaAs BIASED DOUBLE QUANTUM WELL

V.B.Timofeev, A.V.Larionov, A.S.Ioselevich+, J.Zeman, G.Martinez*, J.Hvam▽, K.Soerensen▽*

Institute of Solid State Physics RAS, 142432 Chernogolovka, Moscov.reg., Russia

+Landau Institute for Theoretical Physics RAS, 142432 Chernogolovka, Moscov.reg., Russia

**High Field Magnetic Laboratory, MPI-FKF and CNRS, 38042 Grenoble, France*

▽Microelectronic Centre, Lyngby, Denmark

Submitted 23 March 1998

The interwell radiative recombination from biased double quantum wells (DQW) in *pin* GaAs/AlGaAs heterostructures is investigated at different temperatures and external electrical fields. The luminescence line of interwell recombination of spatially separated electron–hole pairs exhibits systematic narrowing with temperature increase from 4.5 to 30 K. A theoretical model is presented which explains the observed narrowing in terms of lateral thermally activated tunneling of spatially separated *e–h* pairs localized by random potential fluctuations in quantum wells.

PACS: 71.23.-k, 78.60.-b, 78.66.-w

Introduction. Biased double quantum wells (DQWs) have attracted considerable interest in the last decade [1–4]. These structures provide a unique possibility to optically excite electrons and holes spatially separated in adjacent quantum wells. These electrons and holes can be bound into interwell excitons by the Coulomb interaction. The interwell excitons have long lifetimes compared to intrawell ones, because their recombination requires tunneling through the interwell barrier. The interwell excitons can therefore be accumulated to rather high densities and cooled down to low temperatures. Existing theories predict an interesting collective behavior of interwell excitons at high densities and low enough temperatures [5–8].

In this letter we point out that in real biased DQWs, *pin* or *nin* structures, a random potential arising due to charged residual impurities and other imperfections can modify the interwell radiative process quite dramatically. At low temperatures and reduced excitation rates the photoexcited electrons and holes, besides having a *vertical* spatial separation in adjacent wells, are strongly *laterally* localized by random potential fluctuations in quantum wells (see Fig.1). Under conditions of such lateral localization the interwell luminescence spectrum should involve a distribution function of random "wells" and "hills" of the fluctuating potential, which are occupied by electrons and holes, respectively. The interwell radiative probability in this case is controlled not only by vertical tunneling through the interwell barrier but also by lateral tunneling of electrons and holes localized at randomly disributed extrema of the fluctuating potential in the respective quantum wells. One can expect the interwell luminescence spectrum of localized *e–h* pairs to be a rather broad line, with a width that depends on the amplitude of the random potential fluctuations, on the filling of these fluctuations by electrons and holes, and on the (thermally activated) lateral tunneling. In this letter we report both experimental

and theoretical studies of the interwell luminescence line shape and its evolution with temperature at low excitation levels.

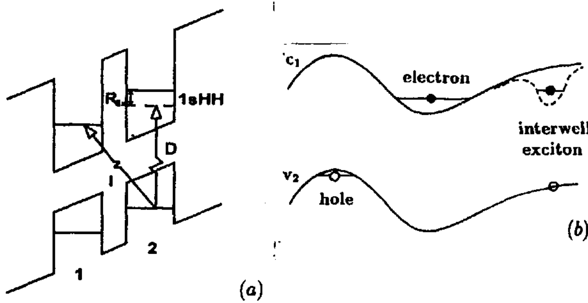

Fig.1. (*a*) Direct (*D*) and indirect (*I*) radiative transitions in a biased double quantum well. (*b*) An electron and a hole, laterally localized at extrema of a random potential. c_1 and v_2 are the profiles of the bottom of the conduction band in the 1st QW, and the top of the valence band in the 2nd QW, correspondingly. An interwell exciton is also shown

Experimental structures. In the experiments we used GaAs/AlGaAs *pin* DQW structures with the following architecture: a substrate of n-type GaAs (Si-doped to 10^{18} cm^{-3}) is covered by a buffer layer of GaAs (Si-doped to 10^{17} cm^{-3}) followed by a layer of $Al_{0.35}Ga_{0.65}As$ (n-type Si doped to 10^{18} cm^{-3}) with a width of 0.5 μm. The intrinsic part of the samples consists of a 0.3 μm thick undoped layer of $Al_{0.35}Ga_{0.65}As$ followed by the first well (undoped GaAs layer 80 Å wide), an undoped $Al_{0.35}Ga_{0.65}As$ barrier layer 50 Å wide, the second well (undoped GaAs layer 80 Å wide), and by a 0.3 μm thick undoped $Al_{0.35}Ga_{0.65}As$ barrier. Then comes the p-type part of the structure, consisting of a 0.5 μm thick layer of p-type $Al_{0.35}Ga_{0.65}As$ doped with Be to 10^{18} cm^{-3}, covered by a 40 Å undoped GaAs cap layer. A 1×1 cm^{-2} rectangular structure with Ohmic contacts to the p- and n-type regions was made for applying external bias to the structure. The structures used exhibited typical I–V characteristics for *pin* diodes in the dark and at low excitation powers. The photoluminescence (PL) spectra were excited either with a He–Ne laser (6328 Å) or with a semiconductor injection laser (6750 Å).

Experimental results. Fig.2 illustrates the PL spectra of the *pin* DQWs measured at a given external field applied to the structure (forward bias $U = +0.4$ V) under cw excitation (6750 Å) at different temperatures. Besides the narrow line $1sHH$, corresponding to the ground state of the intrawell exciton with a heavy hole, one can see at low T a several-times-broader line (the I line), corresponding to interwell recombination. The I line is linearly tuned on the spectral scale under variation of the applied bias, a property which is characteristic of interwell luminescence in *pin* DQWs (see Fig.3). From Fig.2 one can see a strong narrowing of the interwell luminescence line as the temperature increases. The violet tail of this line is close to exponential, with a width equal to T. A systematic narrowing of the interwell luminescence line (by around a factor of four) with increasing temperature was observed at low excitation power for different external bias, forward or reverse, and in the built-in-field. Therefore this is a general property of interwell PL which, in our opinion, has to do with the random potential fluctuations.

The phenomenon of strong lateral localization of photoexcited electrons and holes at random potential fluctuations is confirmed independently by direct measurements of the interwell luminescence kinetics [9]. First of all, this kinetics is nonexponential within the I line at low T. The kinetics exhibit shorter decay times at the violet boundary and longer ones (a few hundred nanoseconds) on the red side of the I line. At higher T, when the FWHM of the I line approaches $2T$, the luminescence decay kinetics becomes, to high accuracy, exponential.

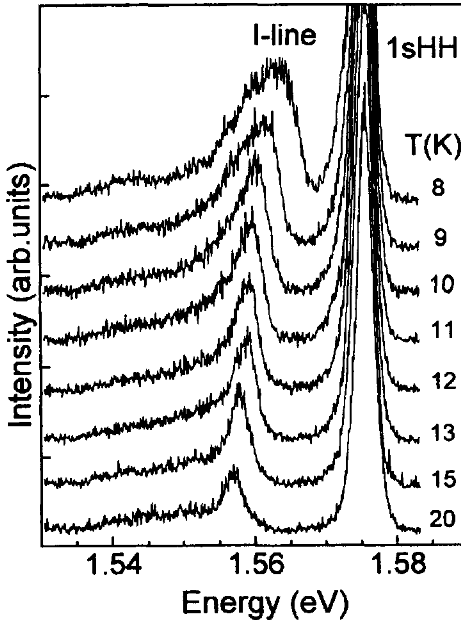

Fig.2. PL spectra measured at forward bias $U = +0.4V$ and different temperatures under CW excitation (6328 Å) of $\sim 5\,\mathrm{mW/cm^2}$

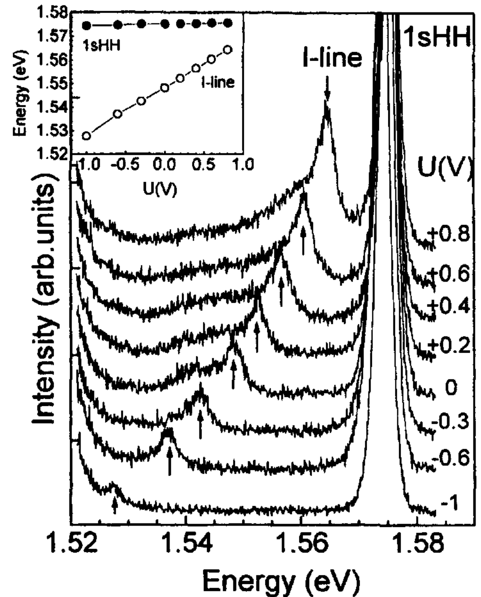

Fig.3. PL spectra measured at temperature 20K and different bias under cw excitation (6328 Å) of $\sim 1.5\,\mathrm{mW/cm^2}$. The inset shows the evolution of the positions of $1sHH$ and I lines

Theory. Let us discuss the theory of the interwell luminescence in the presence of a random potential. We estimate the luminescence intensity within the "exponential approximation", totally disregarding all pre-exponential factors, and neglect the Coulomb interaction (i.e., the effects of the interwell excitons), which do not qualitatively change the results. Consider two quantum wells, the energy difference between the averaged (with respect to the random potential) position of the bottom of the conduction band in the first well and that of the top of the heavy hole band in the second well being ω_0. We assume that a smooth Gaussian random potential energy $u(\rho) = -|e|\varphi(\rho)$ (ρ is a lateral coordinate) is present in the wells, so that the local positions of the bottom of the conduction band and the top of the valence band depend on ρ. If the correlation radius r_c of the random potential is larger than the distance (~150 Å) between the wells, then $u(\rho)$ is identical in both wells. The assumption that r_c is large looks plausible, since charged impurities are localized relatively far from the quantum wells. It is also supported by the magnetic field dependence of the luminescence spectrum [9]. The temperature T is much lower than the characteristic amplitude u_0 of the random potential, so that the photoexcited electrons (holes) are normally localized near the minima (maxima) of the random potential $u(\rho)$ (Fig.1*a*).

From the very low excitation power used (5 $\mathrm{mW/cm^2}$) and the value of the interwell decay time (100 ns) measured directly from the luminescence kinetics under pulsed excitation, one can estimate an upper bound on the average density of nonequilibrium e–h pairs as $10^9\ \mathrm{cm^{-2}}$. Therefore in this experiment the filling of potential fluctuations by electrons (holes) (i.e., the average number of photoexcited carriers per area $\sim r_c^2$) is much less than unity if the correlation length r_c of the random potential is around 1000

Å or less. Thus at such low excitations the classical Boltzmann distribution is likely to be established in the vicinity of each extremum (i.e., minimum i or maximum j):

$$n_i^{(e)}(\epsilon_e) = \exp\{-(\epsilon_e - \mu_i^{(e)})/T\}, \quad n_j^{(h)}(\epsilon_h) = \exp\{-(\mu_j^{(h)} - \epsilon_h)/T\}, \tag{1}$$

where the energies of electrons (holes) $\epsilon_e(\epsilon_h)$, and their local chemical potentials $\mu_i^{(e)}$ ($\mu_j^{(h)}$) are reckoned from the average position of the bottom of the conduction band (top of the valence band). The interwell luminescence intensity can be written in the form ($\hbar = 1,\ k_B = 1$)

$$I(\omega) \propto \Big\langle \sum_{ij} \int n_i^{(e)}(\epsilon_e)\nu_i^{(e)}(\epsilon_e)d\epsilon_e n_j^{(h)}(\epsilon_h)\nu_j^{(h)}(\epsilon_h)d\epsilon_h w_{ij}[\epsilon_e, \epsilon_h, \{u\}] \times$$
$$\times w_0\delta[(\epsilon_h - \epsilon_e) + \delta\omega]\Big\rangle, \tag{2}$$

where $\delta\omega \equiv \omega - \omega_0$, and $\nu_i^{(e)}(\epsilon_e)$ and $\nu_j^{(h)}(\epsilon_h)$ are the densities of states for electrons near the minimum i and for holes near the maximum j of the random potential, respectively. Since the probability of recombination is proportional to the square modulus of the overlap integral of the envelope wave functions of an electron and a hole, expression (2) involves two factors: w_0 is the probability of tunneling between the two quantum wells (vertical tunneling), and $w_{ij}[\epsilon_e, \epsilon_h, \{u\}]$ is the the square modulus of the overlap integral of the lateral envelope functions of an electron and a hole. The latter factor is exponentially small if the classically accessible regions for the electron and the hole do not overlap: then it involves the probability of lateral tunneling (Fig.4). The sum in (2) runs over the pairs of adjacent minima (i) and maxima (j) of the random potential, and the angle brackets stand for the average over the random potential.

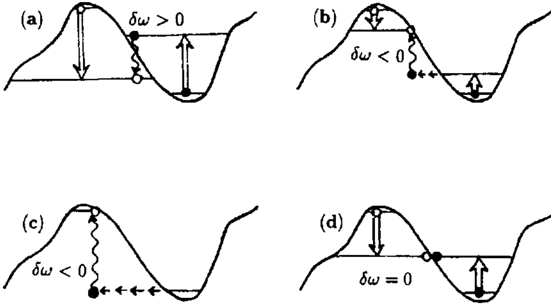

Fig.4. Recombination processes for different energies of the emitted photon. The curves c_1 and v_2 of Fig.1*b* are vertically shifted to coincide. Double arrows show activation, broken arrows show tunneling, and wavy arrows show recombination. (*a*) $\delta\omega > 0$, (*b*) $\delta\omega < 0$, (*c*) The dominant process for low T: pure tunneling, $\delta\omega = U_i - U_j < 0$, (*d*) The dominant process for high T: pure activation, $\delta\omega \approx 0$

In the main part of the present paper we adopt the assumption of *global equilibrium*: $\mu_i^{(e)} = \mu^{(e)}$, $\mu_j^{(h)} = \mu^{(h)}$, which means that the photocarriers escape from the quantum wells (due to recombination, leakage to the leads through the bulk, or whatever) so slowly that an equilibrium between different local extrema has time to be established. Under this assumption one can rewrite (2) in the form:

$$I(\omega) \propto e^{-\delta\omega/T} \left\langle \sum_{ij} \int_{U_i}^{\infty} d\epsilon_e \int_{-\infty}^{U_j} d\epsilon_h w_{ij}[\epsilon_e, \epsilon_h, \{u\}]\delta[(\epsilon_h - \epsilon_e) + \delta\omega] \right\rangle, \tag{3}$$

where U_i (U_j) is the value of the ith (jth) minimum (maximum). In writing the expression (3) we have omitted all ω-independent (though, may be, T-dependent) factors, like w_0, or average concentration of carriers: These factors do not affect the lineshape we are interested in.

In principle, the average (3) may be evaluated for general $\delta\omega$ with the aid of the optimal fluctuation method. In this letter, however, we restrict consideration to several simple limiting cases, in which the calculations are transparent and elementary.

Violet tail. For $\delta\omega > 0$ the classically accessible regions for the electron and the hole overlap (Fig.4a); therefore the function $w_{ij}[\epsilon_e, \epsilon_h, \{u\}]$ does not introduce any exponential factor into the integrand of (3); the only exponential factors come from the Boltzmann distribution functions. As a result, for $\delta\omega > 0$ one obtains

$$I(\omega) \propto \exp(-\delta\omega/T), \tag{4}$$

in accordance with experimental observations.

Far red tail. For large negative $\delta\omega$ the dependence of the lateral tunneling probability w on the realization of the random potential is of only secondary importance: the bottleneck of the process is finding a proper "antisymmetric" fluctuation of the random potential with sufficiently high maximum and deep minimum (namely, $U_j - U_i > \hbar|\delta\omega|$ is required). The probability P for such a fluctuation to occur can be estimated as $P \approx P(U_j = |\delta\omega|/2, U_i = -|\delta\omega|/2) \approx P(u = |\delta\omega|/2)P(u = -|\delta\omega|/2) = \exp\left\{-(\hbar|\delta\omega|/2u_0)^2\right\}$, where $u_0^2 = \langle u^2(\rho)\rangle$. Thus, in the case of large negative $\delta\omega$ one can, roughly, write:

$$I(\omega) \propto \exp\left\{-\delta\omega/T - (\delta\omega/2u_0)^2\right\}. \tag{5}$$

Taking into account the lateral tunneling introduces only a relatively small correction to the exponential in (5), we do not consider this correction in the present letter.

Near red tail. If $|\delta\omega|$ is small (smaller than u_0), then the tunneling path is shorter than the correlation radius r_0. This means that the lateral tunneling proceeds, basically, in the constant field $\mathbf{F}(\rho) = \nabla u(\rho)$, and we can write

$$w_{ij}[\epsilon_e, \epsilon_h, \{u\}] \propto \exp\left\{-4\sqrt{2m}|\delta\omega|^{3/2}/3|\mathbf{F}(\rho)|\right\}, \tag{6}$$

so that, for Gaussian random potential, the procedure of averageing in (3) can be reduced to the integration over fields $\mathbf{F}$ with the Gaussian weight: $\propto \exp\left\{-(\mathbf{F}/F_0)^2\right\}$, where $F_0^2 = \langle \mathbf{F}^2(\rho)\rangle$. Performing the integration by the method of steepest descent , we finally arrive at

$$I(\omega) \propto \exp\left\{-\delta\omega\left[(1/T) - (1/T_c)\right]\right\}, \quad T_c = \left(F_0^2/24m\right)^{1/3} \equiv u_0(\Delta/24u_0)^{1/3}, \tag{7}$$

where $\Delta = F_0^2/mu_0^2 \sim 1/mr_0^2$ is a typical level-spacing for the size-quantized electronic levels in the random potential. Thus, the semiclassical condition $\Delta \ll u_0$ for the random potential ensures that $T_c \ll u_0$.

The overall line shape. Combining all the results obtained in the above limiting cases, we can draw the following conclusions concerning the shape of the I line:

(i) For $T \ll T_c$ the spectrum of the I line should have a broad symmetric Gaussian peak having a full width $\Gamma = \sqrt{2}u_0$ and centered at $\delta\omega = 2u_0^2/T$.

(ii) For $T \approx T_c$ (more specifically, for $|T - T_c| < T_c^2/u_0$) the spectrum should be highly asymmetric: the red tail is Gaussian with a width $\Gamma_r = u_0/\sqrt{2}$, while the violet tail is exponential with a width $\Gamma_v \sim T$.

(iii) For $T > T_c$ the spectrum should show a quite narrow peak centered at $\delta\omega = 0$; the red wing is exponential with a width $\Gamma_r \sim TT_c/(T - T_c)$, while the violet wing is exponential with a width $\Gamma_v \sim T$.

Discussion. The physical mechanism underlying the narrowing of the I line is the switching between the tunneling mode and the activational mode of barrier penetration, a phenomenon which is common to all systems where thermally activated tunneling plays a

part (see, e.g., [10]). At low temperatures $T < T_c$ the optimal mode of barrier penetration for the system (in our case penetration of the lateral potential barrier for electrons) is tunneling: the electrons and holes take the energetically most favorable positions (minima and maxima of $u(\rho)$, respectively); the electron starts tunneling from the very bottom U_i of the potential minimum and meets a hole only upon reaching a nearby potential maximum U_j (Fig.4*c*). Such a process contributes to luminescence with energy $\delta\omega = U_i - U_j$, and therefore at low temperatures the I line is subject to inhomogeneous broadening, with a width $\Gamma \sim u_0$. At high temperatures $T > T_c$ the optimal mode of barrier penetration is activation: the system prefers to wait for a thermal fluctuation, such that both the electon and hole are excited from their equilibrium positions at the extrema of $u(\rho)$, while the classically accessible regions for the electron and hole overlap in this excited state, so that no lateral tunnel is required (Fig.4*d*). Such a process contributes most to luminescence with an energy $\delta\omega \approx 0$, and therefore the inhomogeneous broadening of I line does not occur at $T > T_c$. Note that the switching takes place at $T_c \ll u_0$, so that most of photocarriers are still localized near the extrema of $u(\rho)$.

The above physical picture predicts the high-T narrow I peak to appear at the violet edge of the broad low-T I band. Experimentally, however, this peak arises, roughly, in the center of the I band. One reason for this discrepancy might be a systematic shift of the I line, due to different screening conditions for the external electric field at different temperatures. The other reason might be lack of *global* equilibrium at low T. Under certain (rather general) conditions the *local* equilibrium is characterized by there being a roughly equal number of electrons (holes) captured by different minima (maxima). This means that $\mu_i^{(e)} \approx U_i+\text{const}$, $\mu_j^{(h)} \approx U_j+\text{const}$ at local equilibrium. A detailed analysis of this nonequilibrium case will be published elsewhere; here we only mention that under the above conditions, an intermediate T-independent near violet tail of luminescence arises at low temperatures:

$$I(\omega) \propto \exp\{-(\delta\omega/2\tilde{u}_0)^2\} \quad \text{for } 2\tilde{u}_0 < \delta\omega < 4\tilde{u}_0^2/T, \tag{8}$$

where $\tilde{u}_0 \sim u_0$. This tail should, however, be easily destroyed with increase of temperature, because of the efficient onset of the global equilibrium for relevant fluctuations. The above suppression of the violet wing of the I-band might be partly responsible for the shift of the position of the narrow high-T I-line towards the middle of the broad low-T band.

V.B.T. and A.V.L. acknowledge the partial support of INTAS, RFFI, and Physics of Nanostructures grants.

---

1. Y.J.Chen, E.S.Koteles, B.S.Elman and C.A.Armiento, Phys. Rev. B **36**, 4562 (1987).
2. T.Fukuzava, E.E.Mendes and J.M.Hong, Phys. Rev. Lett. **64**, 3066 (1990).
3. J.A.Kash, M.Zachau, E.E.Mendes et al., Phys. Rev. Lett. **66**, 2247 (1991).
4. L.V.Butov, A.Zrenner, G.Abstreiter et al., Phys. Rev. Lett. **73**, 304 (1994).
5. Yu.E.Lozovik and V.I.Yudson, Zh. Éksp. Teor. Fiz. **71**, 738 (1976) [Sov. Phys. JETP **44**, 389 (1976)].
6. D.Yoshioka and A.H.MacDonald, J. Phys. Soc. Japan, **59**, 4211 (1990).
7. X.M.Chen and J.J.Quinn, Phys. Rev. Lett. **67**, 895 (1991).
8. H.Chu and Y.C.Chang, Europhys. Lett. **35**, 535 (1996).
9. V.B.Timofeev, A.I.Filin, A.V.Larionov et al., , Europhys. Lett. **41**, 535 (1998).
10. A.S.Ioselevich, E.I.Rashba, in: *Quantum Tunneling in Condensed Media*, Eds. Yu.Kagan, A.J.Leggett, North-Holland, Amsterdam, 1992, Chapter 7, pp. 347.

Edited by Steve Torstveit