Resonances in the excitonic transfer in biased double quantum wells

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We report evidence of resonances in the assisted transfer of *excitons* in a biased asymmetrical GaAs-(Ga,Al)As double quantum well. The excitonic transfers, i.e., the conversions of a "direct" exciton into a "crossed" one, are such that the initial and final hole locations are identical but the electron locations are different before and after the transfer. The calculated excitonic energies and transfer times provide a good description of the energy locations and intensities of the photoluminescence lines.

The physics of resonant tunneling has recently given rise to an abundant literature (see, e.g., Refs. 1 and 2). Biased double barriers,[3] double wells,[4–6] or superlattice[7] structures have been investigated. The biased asymmetrical double quantum wells offer many opportunities to study in detail, by use of optical techniques, the resonances in the carrier transfer from one well to the other when the field lines up two electron or hole levels that are mainly localized in different wells under nonresonant conditions. At low temperature the excitons are quickly formed and fairly stable entities and it is natural to inquire whether there is a possibility that an exciton transfers from one well to the other as a whole entity. The purpose of this Brief Report is to report a detailed analysis of the *excitonic* transfer times in biased double quantum wells. We shall show that resonances, i.e., considerable shortenings of the transfer times, take place when the external electric field lines up two interacting exciton transitions. We shall also show that our evaluation of the field dependence of the exciton transfer times allows, via simple rate equations, for a satisfactory description of the field dependence of the various excitonic luminescences observed on a biased 78 Å–55 Å–35 Å GaAs-$Ga_{0.7}Al_{0.3}As$ asymmetrical double quantum well. The experimental data (electric field dependences of the energy peak positions and integrated line intensities of the $T=2$ K photoluminescence and photoluminescence excitation spectroscopy) have been obtained on a biased single double-well structure grown by molecular-beam epitaxy (for details, see Ref. 8).

Figure 1 (inset) shows the band-edge profiles of the biased double quantum well. To be specific, we shall use primed (unprimed) notations for the double-well eigenstates that are mostly localized in the narrow (wide) well and we shall call "direct" ("crossed") excitons the excitons that are formed by pairing an electron and a hole, which are mostly localized in the same (different) well. Negative electric fields correspond to tilting the narrow conduction well below the wide one, allowing E_1-E_1' anticrossings while the ground hole state is always HH_1. It

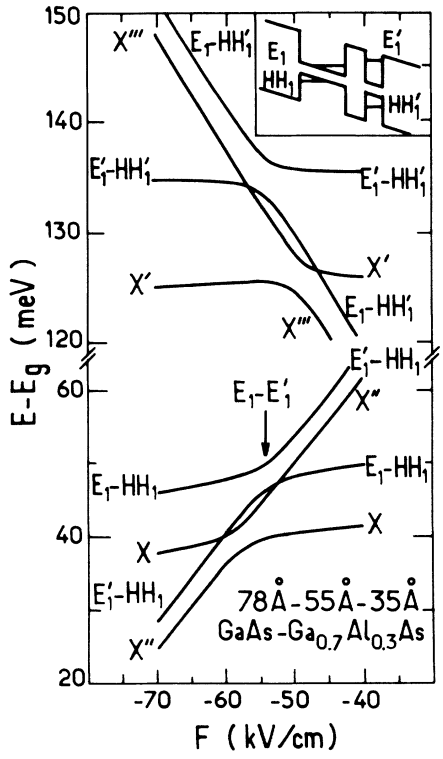

FIG. 1. Inset: Band-edge profile of the negatively biased double quantum well. Lower panel: Calculated electric field dependence of the band-to-band transitions (e.g., E_1-HH_1) and 1*S* excitonic transitions (e.g., X,X').

is possible to form four different types of excitons between the E_1, E_1', HH_1 and HH_1' electron and hole states; E_1-HH_1 1S, E_1'-HH_1' 1S, E_1'-HH_1 1S, and E_1-HH_1' 1S. They are labeled, respectively, X, X', X'', and X'''. The observation by photoluminescence (PL) of these various excitons (the PL lines are labeled, respectively, I, I', I'', and I''') depends on the relative orders of magnitude of their radiative and nonradiative lifetimes. The latter include the assisted transfer times from one well to the other and are therefore strongly dependent upon the electric field strength. The calculated electric field dependence of the four band-to-band and excitonic transitions energies are shown in Fig. 1 for a biased 78 Å–55 Å–35 Å GaAs-$Ga_{0.7}Al_{0.3}As$ double well. The electron, hole, and exciton[9] energy levels in the biased double well have been calculated in the envelope-function approximation using parabolic hosts' bands.

In the vicinity of an anticrossing between a "direct" and a "crossed" exciton transition, we have used the following trial exciton wave function:

$$\psi(z_e,z_h,\rho,\mathbf{R})=S^{-1/2}\exp(i\mathbf{K}\cdot\mathbf{R})\Sigma_{\pm}a_{\pm}\psi_{\pm}(z_e,z_h,\rho)\ , \quad (1)$$

$$\psi_{\pm}(z_e,z_h,\rho)=\sqrt{2/\pi\lambda_{\pm}^2}\,\phi_{\pm}(z_e)\xi(z_h)\exp(-\rho/\lambda_{\pm})\ , \quad (2)$$

where only 1S exciton states have been considered, the valence-band mixing neglected, and where $\rho,\mathbf{R}$ denote the in-plane electron-hole reduced distance and center of mass, respectively, S the sample area, $\xi(z_h)$ the hole eigenfunction in the biased double well which remains unchanged during the transfer (say that of the HH_1 state), and $\phi_{\pm}(z_e)$ the two electron eigenfunctions of the double well that correspond to the two anticrossing electron states (say those associated with E_1 and E_1'). The variational parameters λ_+ and λ_- are first determined by taking, respectively, $a_-=0$ and $a_+=0$. This corresponds to two decoupled exciton states; E_1-HH_1 1S "direct" exciton and E_1'-HH_1 1S "crossed" exciton, respectively. Mixing effects due to the direct-crossed excitons interaction are accounted for by taking subsequently $a_{\pm}$ as variational parameters for fixed $\lambda_{\pm}$. Figure 2(a) shows the comparison between the calculated excitonic transitions in the decoupled and full exciton calculations. The large flexibility of the trial wave function in the latter model leads to larger anticrossing gaps. The calculation of the z_e dependence of the resulting exciton wave function (after averaging over the in-plane and hole coordinates) shows that the Coulombic potential in effect prevents the electron delocalization at the electron resonance because some binding energy is gained by keeping the electron in the same well as the hole. It is remarkable that the direct-cross exciton anticrossing is field shifted with respect to the single electron anticrossing. This is clearly associated with the difference in the binding energies between both kinds of excitons (e.g., 3.5 and 8.3 meV for X' and X, respectively, at −40 kV/cm). It is also important to stress that the two exciton anticrossing (X-X'' and X'-X''' corresponding, respectively, to keeping either the HH_1 or HH_1' unchanged) are field shifted in opposite ways with respect to the E_1-E_1' anticrossing of the undressed electron.

We can now proceed with the calculation of the excitonic transfer times. In one kind of process the electron dressed by the hole undergoes a resonance in its assisted transfer or, equivalently, the direct exciton is converted into a crossed exciton (or vice versa). There also exists a symmetrical process where the hole dressed by the electron transfers from one well to the other. Finally, the conversion of a direct (say X') into another direct exciton (say X) has been found negligible (i.e., the transfer times are much too long) and this case will no longer be discussed here. An external agent is needed to take care of

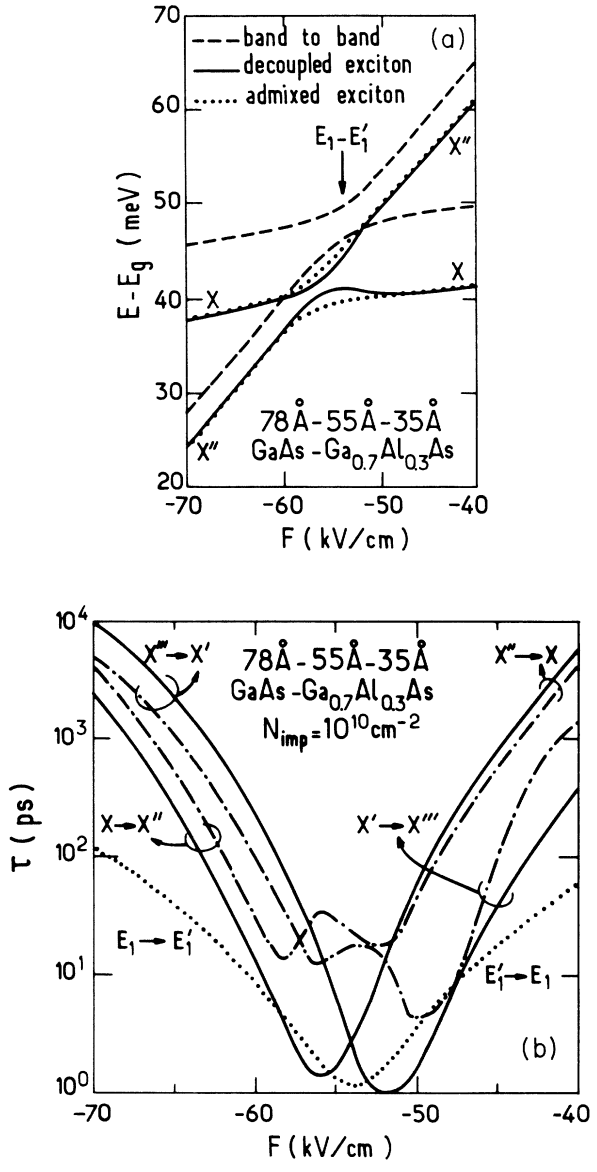

FIG. 2. (a) Comparison between the calculated electric field dependence of the X-X'' excitonic transitions in the decoupled (solid lines) and full (dotted lines) exciton calculations. The band-to-band transition energies are shown as dashed lines. (b) Comparison between the field dependence of the impurity-assisted transfer times of electrons (dotted line), excitons in the decoupled exciton model (solid lines), and in the full exciton model (dashed-dotted lines).

the difference between the in-plane wave vectors of the exciton center of mass in the initial and final states. Since our experiments are performed at low temperature, the impurity participation is more likely. Optical phonon scattering has to be discarded because it is too inelastic compared with the energy differences between the involved excitons. Acoustical phonons cannot be ruled out, but in any event the trends to be discussed below are to a large extent independent of the precise nature of the external elastic or quasielastic agent that induces the exciton transfer. The initial state is always taken to be a $1S$ state with a zero **K** in the upper exciton branch and we compute at the Born approximation the transfer time for such an exciton to make impurity-induced transitions to all possible states of the lower exciton branch that conserve the total exciton energy. We have investigated two different sorts of final states, i.e., those corresponding either to the $1S$ bound state or to dissociated excitons (plane waves for the in-plane reduced motion). Figure 2(b) shows a comparison between the calculated $1S \rightarrow 1S$ exciton impurity-assisted transfer times using both the decoupled (solid lines) and the full exciton model (dash-dotted lines). The impurities are assumed to sit on the two inverted interfaces of the structure with an areal concentration of 10^{10} cm^{-2}. We notice that both exciton calculations predict the occurrence of two distinct exciton resonances located on each side of the electron resonance (dotted line). We also note that the shifts in the exciton resonances are more pronounced in the full exciton calculation as a result of the wider exciton anticrossing gaps. Comparing the exciton and free electron transfer times, we find that the free electrons transfer more easily, and more so when the full exciton model is used. This had to be expected on the grounds that in the full exciton model the Coulombic interaction prevents the delocalization of the electron, thereby reducing the spatial overlap of the initial and final exciton states, an adverse factor of the exciton transfer. Finally, we find that this Coulombic "relocalization" gives rise to a secondary maximum in the $\tau(F)$ curves in the vicinity of the minimum transfer time. Detailed calculations of the field-induced deformation of the exciton wave functions have revealed that the X-X'' resonance and X'-X''' resonances take place at the electric field where the $\tau(F)$ curves admit an absolute minimum.

We have inserted the various exciton transfer times into simple rate equations which incorporate pump rates for the E_1-HH$_1$ and E_1'-HH$_1'$ excitons, radiative recombination in both wells (with a radiative lifetime of 1 and 0.5 ns, respectively), and direct↔crossed exciton transfers. Other nonradiative processes have been neglected. The transfer times for the direct↔crossed exciton transitions which involve the hole transfer cannot be evaluated without the valence-band mixing effects in the exciton states. Otherwise the transfer times are orders of magnitude too long. Thus, we have approximated these transfer times by a constant: 330 ps. This value represents the averaged free hole impurity-assisted transfer time[10] over the field range (-70 kV/cm, 0). In such a field range the free-hole-assisted transfer time exhibits two very narrow (<1 kV/cm) resonances and it is likely that inhomogeneities in the well thicknesses will wash out the resonant features. In Fig. 3 we compare the calculated and measured field dependences of the I, I', and I'' PL intensities (in arbitrary units). I''' was not observed as a result of the fast hole transfer compared to the radiative lifetime of the crossed exciton transitions. In the calculations we have taken into account both the $1S \rightarrow 1S$ and $1S \rightarrow$continuum exciton-assisted tunnelings. This explains the occurrence of double minima in the I and I' calculated curves, the farther away from the purely electronic resonance corresponding to the $1S \rightarrow$continuum contributions. We notice that I'' is extremely small unless $F \leq -58$ kV/cm, in good agreement with experiment. This is again due to the competition between radiative and nonradiative channels: the I'' PL is observable only when X'' is the exciton ground state. At lower fields, the calculated I'' is constant while the experimental one decreases. Field-induced sweeping out of the electron participating in the weakly bound X'' exciton is likely to explain this discrepancy. The $I(F)$ curve is fairly well reproduced by the calculations, the lack (or uncertainty) of data right near the resonance being due to the spectral overlap of the I and I'' PL lines. The experimental I' curve displays a single minimum near the field corresponding to the (E_1'-HH$_1'$, $1S$)→(E_1-HH$_1'$, continuum) resonance. In reality, the theoretical curve should comprise an infinite number of resonances ($1S \rightarrow nS$, nP, nD, . . .) between the $1S \rightarrow 1S$ and $1S \rightarrow$continuum resonances. The allowance for these multiple and increasingly dense resonances may eventually suppress the sawtooth aspect of the theoretical $I'(F)$ curve and bring it closer to the experimental results. Altogether, we believe that our rough steady-state modeling provides a very satisfactory description of the experimental findings: despite our neglect of nonradiative processes, the relative

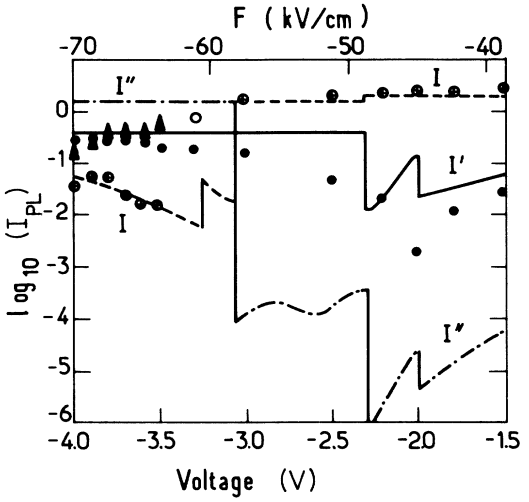

FIG. 3. Calculated (lines) and measured (symbols) electric field dependence of the X (dashed line and circled crosses, respectively), X' (solid line and solid circles, respectively), and X'' (dashed-dotted lines and triangles, respectively) PL intensities in arbitrary units. The open circle corresponds to the case of a spectral overlap between the two X and X'' PL lines.

intensities I, I', and I'' are well reproduced by the calculations. More importantly, the location of the observed resonances are well predicted by our calculations. This leads us to conclude that we have obtained the first convincing evidence of resonances in the excitonic transfer in biased double quantum wells. These findings are important to the understanding of the vertical transport of excitations in a variety of heterostructures, noticeably in superlattices.

During the writing of this paper, evidence of excitonic behavior at anticrossing in biased asymmetric quantum wells was reported.[11]

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[1]F. Capasso, K. Mohammed, and A. Y. Cho, IEEE J. Quantum Electron. **QE-22**, 1853 (1986).

[2]E. E. Mendez, in *Physics and Applications of Quantum Wells and Superlattices,* Vol. 170 of *NATO Advanced Study Institute Series B: Physics* (Plenum, New York, 1987).

[3]V. J. Goldman, D. C. Tsui, and J. E. Cunningham, Phys. Rev. Lett. **58**, 1256 (1987); Phys. Rev. B **35**, 9387 (1987).

[4]R. Sauer, K. Thonke, and W. T. Tsang, Phys. Rev. Lett. **61**, 609 (1988).

[5]Y. J. Chen, Emil S. Koteles, B. S. Elman, and C. A. Armiento, Phys. Rev. B **36**, 4562 (1987).

[6]H. W. Liu, R. Ferreira, G. Bastard, C. Delalande, J. F. Palmier, and B. Etienne, Appl. Phys. Lett. **54**, 2082 (1989).

[7]H. Schneider, K. v. Klitzing, and K. Ploog, Superlatt. Microstruct. **5**, 305 (1989).

[8]R. Ferreira, H. W. Liu, C. Delalande, J. F. Palmier, and B. Etienne, Surf. Sci. **229**, 192 (1990).

[9]I. Galbraith and G. Duggan, Phys. Rev. B **40**, 5515 (1989).

[10]R. Ferreira and G. Bastard, Europhys. Lett. **10**, 279 (1989).

[11]R. P. Leavitt and J. W. Little, Bull. Am. Phys. Soc. **35** (3), 346 (1990).