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## Picosecond photoluminescence of resonantly-excited excitons in GaAs quantum wells

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**ABSTRACT:** We show that the *initial* dynamics of resonantly-excited excitons in quantum wells is controlled by *several processes* such as radiative recombination, spin relaxation of excitons, electrons and holes, and scattering between different in-plane momentum states of excitons. We present a *unified* analysis of the results, which provides quantitative information about these important rates, and a good physical understanding of the initial dynamics of non-equilibrium excitons. In particular, we show that the enhanced radiative recombination rate of excitons makes an important contribution to the early decay of the photoluminescence.

Picosecond dynamics of excitons in quantum wells has been extensively investigated for both resonant and non-resonant excitations. For non-resonant excitation the dynamics of exciton formation and their relaxation to the optically active state plays an essential role. For resonant excitation, a cold distribution of excitons is created in the optical active states and the temporal profile of the photoluminescence decay provides information on how this non-thermal distribution reaches thermal equilibrium with the lattice. The dynamics of resonantly excited excitons is determined by various processes, such as scattering in large momentum ( $K$ ) states, exciton spin relaxation, electron and hole spin relaxation and exciton radiative recombination. All the previous studies under resonant excitation [1,2] focussed only on one or two of these processes. Besides, most of these investigations, both resonant and non-resonant, concentrated on the long-time, equilibrium behavior of excitons [3]. As a consequence, the dynamics of resonantly-excited, non-equilibrium excitons is not well understood, and is the main focus of our work. Since most of the processes contributing to the initial dynamics occurs on the same timescale, the full complexity of the non-equilibrium exciton dynamics can only be understood by a *simultaneous* investigation of all aspects, and a *unified* analysis of the results. This is the main aim of our work.

We have investigated picosecond dynamics of excitons in GaAs quantum wells at low temperatures, resonantly exciting the heavy-hole (HH) exciton. Using an up-conversion technique, we improved the time resolution (5 ps), compared to previous studies of this topic and nearly completely suppressed the scattered laser light from the detected photoluminescence (PL). Therefore we were able to extract significant information about non-equilibrium exciton dynamics (<100 picoseconds) not possible previously. Our results show that the initial dynamics of resonantly-excited excitons is controlled by several different dynamical processes. We present a unified analysis which takes into account the enhanced radiative recombination of two-dimensional (2D) excitons, the spin relaxation of excitons, holes and electrons, and scattering between different  $K$  (in-plane momentum) states of excitons and we determine these rates. In particular we show that the fast radiative recombination of

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excitons in 2D systems makes an important contribution to the early dynamics. From our data we deduce the exciton radiative rate  $\Gamma_0$  for several samples with different well thickness in the range 80-175 Å, finding a good agreement with theoretical predictions. The details about the experimental apparatus have been discussed elsewhere [4].

High quality GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As multiple quantum well structures are studied in the temperature range 10-40 K at low excitation density (typically  $5 \times 10^9 \text{ cm}^{-2}$ ). CW characterization of the samples in the temperature range 10-40 K shows that the luminescence is due to free exciton recombination. Circularly polarized light is used in the experiments;  $I^+$  and  $I^-$  denote the PL intensity from the  $|+1\rangle$  excitons when the optical pulse excites the  $|+1\rangle$  and  $|-1\rangle$  excitons, respectively. S and D denote  $(I^+ + I^-)$  and  $(I^+ - I^-)$ , respectively. The homogeneous linewidth  $\Gamma_h$ , for each sample, is deduced from four-wave mixing (FWM) experiments and it varies between 0.2 and 0.5 meV, depending on the sample, at 12 K.

Typical experimental results are shown in Fig.1 for a 130 Å QW. After a rapid rise,  $I^+$  decays of more than a decade in less than one hundred ps, with a characteristic profile much longer than the cross correlation signal (dotted line, Fig.1d).  $I^-$  slowly increases and, after few hundreds ps,  $I^+$  and  $I^-$  are almost equilibrate. At longer time the decay of S is monoexponential, with a slope decreasing with increasing temperature, indicating a thermal distribution of excitons at longer time.

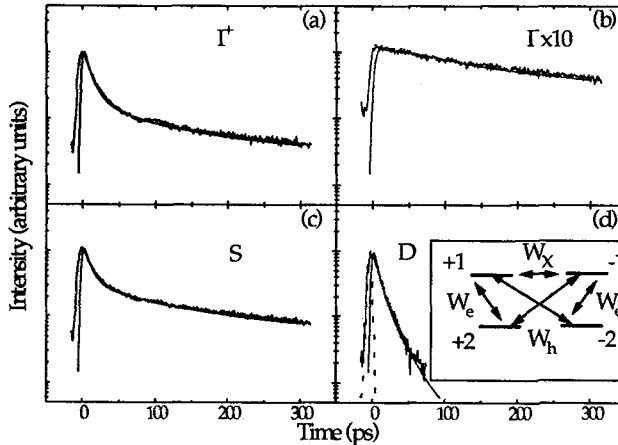


Fig.1: Temporal evolution of resonantly-excited HH exciton PL from a 130 Å GaAs QW at T=12 K for different polarizations and calculated fits as discussed in the text. The dotted line in Fig.1d is the cross-correlation signal, showing the time resolution ( $< 5$  ps). The inset in Fig.1d is the model as discussed in the text.

The starting point of our analysis is the long-time behavior of the PL. In fact, under the assumption that excitons can recombine as long as their in-plane momentum  $K$  is less or equal than the photon momentum  $K_0$  (or kinetic energy  $\leq E_0 \approx 0.1$  meV), all excitons within  $\Gamma_h$  of  $E_0$  can radiatively recombine. When thermal equilibrium is reached the fraction  $f$  of excitons that can recombine is given by  $f = (1 - \exp(-\Gamma_h / kT)) / (1 + \exp(\Delta / kT))$  [5] where  $\Delta$  is the exchange splitting (0.08 meV) between the  $|\pm 1\rangle$  and  $|\pm 2\rangle$  exciton states. Therefore, the decay rate at equilibrium  $W_D$  is given by:

$W_D = W_R \times f$ , where  $W_R = \frac{4E_0}{3\Gamma_h} \Gamma_0$  [5] with  $\Gamma_0$  exciton radiative rate. As shown in Table I for the

different samples, the experimentally determined  $\Gamma_0^{-1}$  is of the order of few tens ps in good agreement with theoretical estimates [6]. Therefore, because initially all the excitons are created in a radiative state,

the early dynamics which occurs in the first 100 ps, is strongly affected by the radiative recombination of excitons, even at higher temperature.

Dynamical processes other than radiative recombination also influence the initial exciton dynamics. These processes are spin relaxation of excitons between  $|+1\rangle$  and  $|-1\rangle$  states, spin relaxation of electrons and holes between  $|\pm 1\rangle$  states and  $|\pm 2\rangle$  dark states, scattering with acoustic phonon in large  $K$ -states. We want to emphasize that the initial slope of the PL decay is given by  $(W_R \times f)^{-1}$  and, because  $f$  is changing until equilibrium is reached, the initial decay is *not* exponential and therefore  $W_R$  cannot be extracted from the initial slope as previously done [2].

We fit the experimental data in the framework of a model which takes into account all the processes previously mentioned. We simplify the model considering only two manifolds of spin-states: the ones at  $K=0$  and an equivalent set at  $K\neq 0$ , introducing appropriate thermal factors for the population between different states and allowing only spin-conserving transition between the two sets of states at different  $K$  [7]. In the inset of Fig. 1d the model is shown only for the set of states at  $K=0$ . From the fit to four different curves (I<sup>+</sup>, I<sup>-</sup>, S and D) various scattering rates can be estimated (Table 1) and because each parameter affects differently each curve there is not much flexibility and we can put reasonable error bars on the parameters ( $W_k$ ,  $W_x$ ,  $W_e$  and  $W_h$ ) determined from the fits. In table I we report the results for the measured decay rate at thermal equilibrium  $W_D$ , the experimentally deduced  $\Gamma_0$ , compared with the theoretical prevision  $\Gamma_{0th}$ , and for  $W_k$  and  $W_x$ .

**Table I:** Results for the thermal equilibrium decay rate  $W_D$ , the radiative rate  $\Gamma_0$ , the scattering rate  $W_k$  and the exciton spin relaxation rate  $W_x$  for different QWs.  $\Gamma_0$  is compared with the theoretical value  $\Gamma_{0th}$ .  $W_D$ ,  $W_k$  and  $W_x$  are the values at 12 K.

QW (Å)	$W_D$ ( $\times 10^9$ s <sup>-1</sup> )	$\Gamma_0$ ( $\times 10^{10}$ s <sup>-1</sup> )	$\Gamma_{0th}$ ( $\times 10^{10}$ s <sup>-1</sup> )	$W_k$ ( $\times 10^{10}$ s <sup>-1</sup> )	$W_x$ ( $\times 10^{10}$ s <sup>-1</sup> )
80	2.8	4.5	4.6	10±2	2.5±0.5
130	1.8	2.6	3.1	8±1.5	2.0±0.5
175	1.54	2.2	2.5	5.5±1.5	1.5±0.5

We have already discussed the results on  $\Gamma_0$ . The other interesting results concern the dependence of the effective scattering rate  $W_k$  and the exciton spin-relaxation  $W_x$  on the well-width. These results agree with theoretical predictions; in fact an increase of the scattering rate  $W_k$  is expected reducing the well width [8] and similarly for the increase of  $W_x$  due to an increase in the exchange interaction [7]. Under the assumption that  $W_h$  is faster than  $W_e$ , the estimate of  $W_h$  is  $(1_{-0.3}^{+1}) \times 10^{10}$  s<sup>-1</sup>, while the fits are not very sensitive to  $W_e$  ( $3 \times 10^8 < W_e < 3 \times 10^9$  s<sup>-1</sup>). The dependence on temperature shows that the product  $\Gamma_h W_R$  is constant in agreement with predictions [5], indicating that the contribution of radiative recombination at the initial dynamics is important also at higher temperature. The scattering rate  $W_k$  increases with temperature less than expected, but this is easily understood because  $W_k$  is an effective scattering rate and the value that we extract from the fit is somehow weighted by the value of the homogeneous linewidth, which also increases with T.

We want to emphasize that our results do not show any superradiant effect and in fact, no coherence is required for the fast radiative recombination of excitons. We do not find any signal decaying with  $T_2/2$ , where  $T_2$  is the dephasing time as measured from FWM experiments. In our model we only require that excitons remain within the homogeneous linewidth for recombining. This is confirmed by the measured temperature dependence that is strongly different from the expected in the presence of coherent effects. This is not surprising because in our picture the loss of interband coherence, due for instance at any kind of dephasing process which elastically or quasi-elastically dephases the coherently excited exciton population, does not affect the population within the

homogeneous linewidth.

From our results an important conclusion can be drawn: the thermalization time for a cold exciton distribution is quite long ( of the order of hundred ps). Therefore, under resonant excitation, the fast exciton recombination cannot be neglected at short times and we believe that previous analysis from different experiments (like picosecond pump and probe), in which this effect was completely ignored, have to be reconsidered in the light of our results. In particular the picosecond bleaching at the exciton transition can be dramatically modified, considering the exciton recombination and the scattering in large  $K$ -states. Besides, the spin relaxation rates for electrons and holes cannot simply be equated to the initial and final slopes of the differential transmission in the picosecond time-scale.

In conclusion, we have shown that several different processes control the initial dynamics of non-equilibrium excitons. We have simultaneously investigated these processes and presented a unified analysis to obtain the rates for radiative recombination, spin relaxation and momentum relaxation. The fundamental exciton recombination rate for different well-width has been deduced, in good agreement with theoretical previsions, considering the evolution of the exciton distribution toward thermal equilibrium.

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