Coherent vs. Incoherent Emission in Quantum Wells studied by Polarisation- and Time-Resolved Spectroscopy

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Abstract We present a new method to discriminate between coherent and incoherent light emission from a two dimensional exciton gas. It is based on the resonant creation of spin-polarized excitons by means of ultrashort light pulses and the subsequent analysis of the polarized emission using dynamic rate equations. Introducing two dephasing mechanisms, radiative and nonradiative, we found that increasing the excitation density leaves the latter unaffected while it enhances considerably the radiative one.

Due to the advances in femtosecond spectroscopy during the last few years, the very early stages of light emission dynamics in semiconductor heterostructures became an object of intense study. To investigate the processes in these structures at times <10 ps several techniques have been used: time resolved secondary emission [1], four wave mixing [2], speckle resolved spectrospcopy [3] and spectral interferometry [4]. We present here an alternative method: light emission from the two spin components of a circularly polarised exciton gas are detected separately with a time resolution of 150 fs. The spin dynamics is explained by an existing rate equation model [5], which was extended by including a coherent exciton population and exciton-exciton (XX) scattering effects.

The measurements were performed on a 50 period, multi quantum well (MQW) consisting of 77 Å GaAs wells separated by 72 Å wide AlAs barriers. The sample was held at 8 K. The excitation with a σ^+ circularly polarised pulse was done in resonance with the excition energy with a two-colour up-conversion set-up, detecting σ^+ (from spin +1 excitons, X^{+1}) and σ^- (from spin -1 excitons, X^{-1}) emission separately.

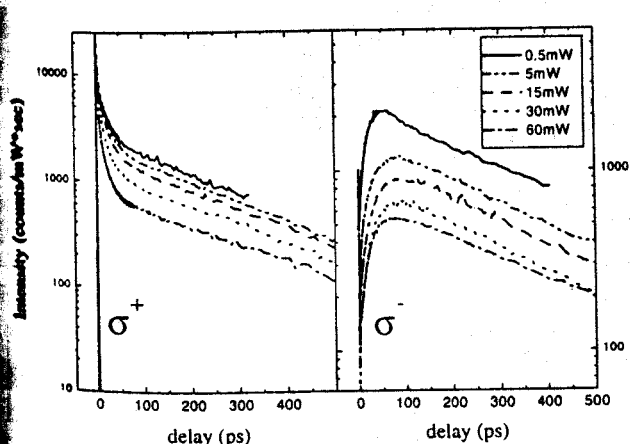

Fig. 1 Emission dynamics for +1 (a) and -1 (b) excitons for several exciatation densities. Intensities are normalised to the excitation power.

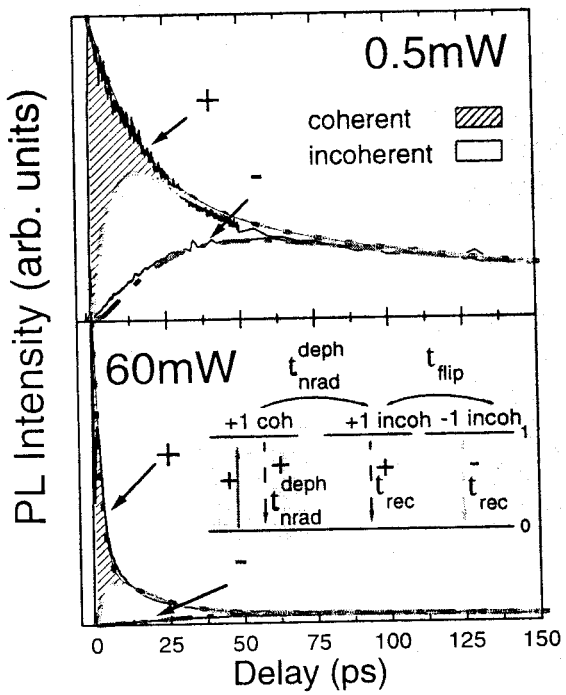

Fig. 2 Fits to the experimental data for 0.5 mW (a) and 60 mW (b) excitation power. The inset shows a simplified scheme of the rate equation model.

After resonant σ^+ excitation, the majority X^{+1} component decays from its maximum while the minority X^{-1} component becomes populated by spin-flip processes reaching its maximum value after ~100 ps. The striking effect, shown in Fig. 1a, is that on increasing the excitation density, the initial decay of the X^{+1} becomes steeper, while the X^{-1} population is rising at about the same rate and its maximum population is decreasing. Exponential fits of the slow decay of σ^+ and σ^-, and on the rise of σ^- gives the following results: a long emission decay time $\tau_{dec}^{(\pm 1)} = 245 \pm 15$ ps independent on excitation density; rise time of σ^-, $\tau_{rise}^{(-1)}$=23 ps at 0.5 mW, which

becomes constant $\tau_{rise}^{(-1)} = 42 \pm 4$ ps at higher excitation densities. Finally a separate fit of the initial fast decay of the σ^+ emission yields a value of $\tau_{fdec}^{(+1)}$=18 ps at 0.5 mW, which decreases to $\tau_{fdec}^{(+1)} \simeq 2$ ps at 60 mW. In resonant Rayleigh scattering experiments the behavior of $\tau_{fdec}^{(+1)}$ is generally explained by an enhanced dephasing rate due to XX interaction, whereas in spin-dynamic measurements this fast initial decay is attributed to the hole spin-flip and momentum scattering to optically inactive high-K states. Vinattieri *et al.* [5] presented a model which explaines the fast initial decay in terms of this hole flip and momentum scattering. They could obtain good agreement with experiment at low densities having a time resolution of ~ 5 ps. However, it was not possible to reproduce our observations with this model. Adjusting the fast decay leads to an overestimation of the σ^- emission. Since in the model of Ref. [5] the initial coherent emission or resonant Rayleigh scattering is neglected we added a new decay path to the model. Assuming that the created X^{+1} exciton population is initially in phase with the laser pulse and coherent, we introduce two dephasing mechanisms: a radiative dephasing characterizing the coherent emission and a nonradiative one responsible for the transfer of excitons from coherent to incoherent $X^{\pm 1}$ populations, which then gives rise to PL and further spin-flip processes. The fast decay of the σ^+ emission is characterized by the radiative dephasing time τ_{rad}^{deph} and the rise of the incoherent σ^+ emission by the nonradiative time τ_{nrad}^{deph}. The basic scheme of the model is drawn in the inset of Fig. 2. Exciton spin-flip (with τ_{flip}) allows a transfer between the incoherent X^{+1} and X^{-1} components. A detailed description of the incoherent dynamics is given in Ref. [5]. Additionally, since a density-dendent energy splitting between X^{+1} and X^{-1} exists, a Boltzman factor was added to the exciton spin-flip [6,7]. The mechanism responsible for direct flip from ± 1 to ± 2 states due to inter-excitonic exchange was also included [8,9]. As in any fit procedure with many parameters, some of these can be interdependent, making difficult the separation of the influence of different mechanisms. However, the dominant role of the coherent part in the total exciton population and its dynamic is not influenced by that fact. In all the fits we fixed the hole flip time to 1 ns [10] and the electron flip time to 300 ps [5]. The Boltzman factor for the exciton flip was taken from measured values [7] and the inter-excitonic exchange scattering rates from Ref. [9]. The exciton recombination time, τ_{rec}, was 120 ps, which differs a factor of 2 from the one that was fitted directly to the emission by exponential decay ($\tau_{dec}^{(\pm 1)} = 245 \pm 15$ ps). This is not surprising since the population of the optically dark $X^{\pm 2}$ states contributes to the long decay of the emission signal.

In Figs. 2a and b the resulting fits for two excitation densities are shown. The assumption of an initial coherent population and the two dephasing mechanisms is crucial to obtain a good agreement with the measurements. Now the initial rise of σ^- and the overall X^{-1} population is fitted satisfactorily. The hatched area in Fig. 2 between the total σ^+ and the fit for the incoherent σ^+ emission, represents the part of emitted coherent light in the total emission. On increasing the excitation by two orders of magnitude, the initial decay is considerably faster whereas the time to populate the incoherent X^{+1} component, τ_{nrad}^{deph}, is practically unchanged. The dephasing times obtained with the extended model for several excitation densities are shown in Fig. 3. The radiative decay time, τ_{rad}^{deph}, is decreasing very quickly between 5 and 15 mW and only reduces by a factor 2 from 15 to 60 mW, whereas the non-radiative time, $\tau_{nrad}^{deph} = 11 \pm 1$ ps, is constant. Dispite this clear behaviour further experiments are needed, and are under way, to confirm the nature of the initial emission. In principle, emission in angular directions which were not detected, non-radiative processes or the different dynamics of localised and extended excitons could contribute to the initial, density dependent fast decay.

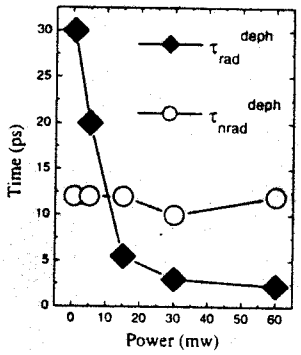

Fig. 3 Times for emission, τ_{rad}^{deph}, and dephasing, τ_{nrad}^{deph}, as obtained from the fit with the rate equation model.

This work was partially supported by the European Union through the Training and Mobility of Researchers Ultrafast Quantum Optoelectronics Network, the austrian BMWF, the spanish DGICYT (PB96-0085), the CAM (07N/0026/1998) and the Fundacion Ramon Areces.

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