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Kinetics of dark excitons and excitonic trions in InGaAs single quantum well

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Abstract. Magnetooptical studies performed on a wide InGaAs/GaAs single quantum well indicate that optically non-active (dark) excitons with total angular momentum $M = \pm 2$ play the role of a reservoir for the creation of free multiparticle excitonic complexes. After analyzing the magnetic field evolution of the circularly polarized components of the low energy structure appearing in the main excitonic luminescence line we assign this feature to the excitonic trion formation. The binding energy of the excitonic trions was estimated to be of the order of 1 meV.

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1 Introduction

In direct-gap semiconductors with a four-fold degenerate valence band (particularly in GaAs and related direct-gap compounds) in addition to optically active (radiative) excitons, X_A, there exist non-radiative (dark) excitons X_D with a total spin angular momentum of electron and hole $M = \pm 2$ which correspond to a dipole-forbidden interband transition. Due to the non-radiative nature of dark excitons, their direct spectroscopic observation is not, in principle, an easy task [1], especially in GaAs where the value of the exchange energy between the electron and the hole in the exciton is very small [2]. Their energy position is not therefore spectroscopically distinguishable from that of an X_A-exciton, even in confined systems, such as quantum wells. Nevertheless, the existence of non-radiative excitonic states may lead to observable consequences in the radiative kinetics of the luminescence. The role that dark excitons play in the dynamics of the exciton spin polarization in GaAs quantum wells has been discussed in a series of recent publications [3,4]. In particular, time-integrated studies of the magneto-luminescence polarization were used to determine indirectly the exchange splitting between dark and optically active excitons [3]. The existence of dark excitations has also been exploited in modeling the exciton spin kinetics [4] in order to explain earlier time-resolved luminescence polarization measurements in two-dimensional quantum wells [5]. Another role that dark excitons may play in the recombination kinetics in confined systems results from their non-radiative nature and, therefore, from their relatively long life-time, as compared to that of optically active excitons. Therefore, in experiments where dark excitons are created with densities comparable to the radiative ones, a non-equilibrium accumulation of non-radiative entities can be achieved with respect to short-living optically active excitons. As a result, the observation of phenomena requiring high density of cooled excitonic gas may become possible. In this report we present experimental results which can be interpreted in terms of the formation of free excitonic complexes, such as trions X^- or excitonic molecule (biexciton), resulting from collisions between residual equilibrium electrons and dark excitons accumulated in the well after the photoexcitation of the electron-hole pairs in the bulk (barrier) semiconductor. The difficulty of accumulating and observing free excitonic complexes in direct gap semiconductors under usual circumstances (without the help of a vast reservoir of non-radiative particles) lies in the fact that radiative decay of excitons is so fast that the accumulation of an appropriate density is not easily achievable. In quantum wells, the situation is even worse due to the increase of the radiative recombination rate due to the enhanced overlap of the confined electron and hole wave functions, so that most of the experiments aiming at the observation

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of trions were performed in relatively high electron density structures [6–8].

After a brief review of the qualitative aspects related to this problem in Section 2, the experimental set-up and results will be detailed and analyzed (Sects. 3 and 4).

2 Basic qualitative aspects

Being optically passive, dark excitons have a life-time limited by the spin-relaxation process, which converts them into optically active excitons and which involves either the electron or hole spin flip due to the spin-orbit coupling with a characteristic time τ_{SO}. Alternatively, the inter-excitonic collisions involving an exchange between differently polarized electrons may convert a pair of dark particles into optically active ones through the process: $X_D(e\uparrow, h\uparrow) + X_D(e\downarrow, h\downarrow) \Rightarrow X_A(e\uparrow, h\downarrow) + X_A(e\downarrow, h\uparrow)$. In both cases the lifetime of dark excitons is reduced as well as their density. The latter process is somewhat similar to that discussed in the problem of optically inactive ortoexcitons in strained Cu_2O [9]. The accumulation of dark excitons can also be limited by another kind of collisions between dark excitons and residual free electrons through the process: $X_D(e\uparrow, h\uparrow) + e\downarrow \Rightarrow X_A(e\downarrow, h\uparrow) + e\uparrow$. In those processes the arrow indicates an electron spin $\pm 1/2$ and a hole spin $\pm 3/2$ with an orientation perpendicular to the plane of a quantum well. In pure structures and at low temperatures, the time τ_{SO} can be quite long compared to the radiative lifetime τ_A of optically active excitons. Assuming that $\tau_A \ll \tau_{SO}$, one can estimate the dark exciton density as $n_D = G\tau_S$, with $\tau_S^{-1} = \tau_{SO}^{-1} + n_D\sigma_D$. In this relation, σ_D is the cross-section of one of the inter-excitonic collision process and G is the generation rate of free electron-hole pairs which can form (after relaxation) both kinds of excitons. As mentioned above, despite the non-equilibrium accumulation of dark excitons, their direct manifestation in luminescence spectra of GaAs-based structures is hardly possible because of the very small value of the X_A and X_D energy splitting. On the other hand, the presence of a relatively high density of cold non-radiative excitons may induce radiative processes involving the recombination of excitonic complexes–molecules or trions, which are improbable events in a low-density excitonic system. These complexes can be formed *via* excitonic collisions assisted by a phonon interaction giving rise to the formation of singlet optically active complexes. For instance one can have $X_D(e\uparrow, h\uparrow) + X_D(e\downarrow, h\downarrow) \Rightarrow X_2$ (singlet) + phonon or $X_D(e\uparrow, h\uparrow) + e\downarrow \Rightarrow X^-$ (2 e^- singlet) +phonon (where X_2 is a singlet state of the excitonic molecule and X^- is a trion with a singlet electron configuration). *A priori*, none of possibilities can be excluded because in many cases a weak residual n-type doping in the bulk surrounding material is present, and in the presence of a sufficient number of electrons the trionic features in the luminescence spectra can be observed [6,7]. The role of dark entities is to provide a reservoir for creating complexes, which later recombine giving an additional recombination line shifted to the low energy side of the original excitonic line. The splitting between these two lines is the sum of the complex's binding energy and of the energy of the recoil particle. This latter particle is an exciton after an excitonic molecule decay or an electron after the decay of a trion. Because both X_2 or X^- are weakly bound, the cross-section of an inter-excitonic collision or of a scattering between a low energy electron and dark exciton has the same characteristic features like that of a scattering by a shallow bound state [10], which means that it exceeds many times the excitonic Bohr radius and may efficiently promote a bound complex.

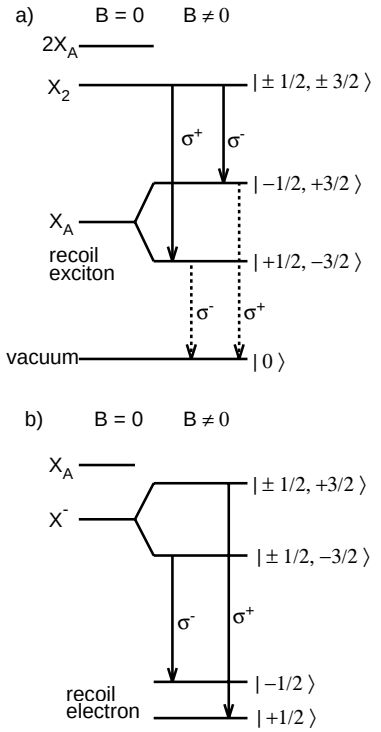

Fig. 1. Schematic presentation of the radiative decay of an excitonic molecule (a) and an excitonic trion (b) subjected to a magnetic field perpendicular to the quantum well plane. The full arrows correspond to the observed transitions.

In the material we study in this experiment, we do not have a precise estimate of the binding energies of a two-dimensional biexciton or of a trion, which both strongly depend on the electron/hole mass ratio and the carriers' wave functions inside the quantum well. We only list below some qualitative features which are expected for their magneto-optical properties. In the low magnetic field regime orbital magnetic field effects can be ignored and the discussion will focus on spin effects. Qualitative schemes illustrating the expected spin splittings of different terms for radiative decay of an excitonic molecule and trion, for a magnetic field perpendicular to the QW plane, are presented in Figures 1a and b, respectively. The recombination from a complex which is a singlet with respect to the electron spin is allowed in both polarization, σ^+ and σ^-, with relative intensities determined by the occupation numbers of spin $\pm 3/2$ of holes. In a trion (Fig. 1b) the

latter occupancies would be the same as the occupancies of holes in the excitons, whatever is its radiative activity, and therefore one expects the intensities of the σ^+ and σ^- components of a trionic satellite to be in the same ratio as those of an optically active exciton line. The Zeeman splitting of trion lines in different polarizations is determined by the polarization of the hole in the initial state and of the electron in the final state and follows the corresponding splitting of the main excitonic line. The biexciton which, in the ground state, is singlet for both electron and hole spins will exhibit a luminescence line with equal intensities of the σ^+ and σ^- components, which may be considered as a characteristic signature of this entity (see Fig. 1a). We emphasize that the latter feature would be a universal proof of the molecular nature of complexes observed in optical experiments. The spectral positions of its luminescence lines in both polarizations are determined by the energetics of the electron and a hole Zeeman splitting in the final state, and their splitting also follows the splitting of the σ^+ and σ^- components of the luminescence of an optically active exciton.

When a magnetic field $B_\parallel$ is applied parallel to the plane of a well, its effect on the formation of complexes is different. Since the generation rate of trions or biexcitons formed in the system is determined by the number of accumulated dark excitons, the enhanced efficiency of the conversion mechanism of X_D into X_A would drastically reduce the intensity of the complex's satellite. This is due to the fact that a parallel magnetic field induces a spin precession of the electron around the field direction with the Zeeman frequency $\mu g_e B_\parallel$ whereas, to the lowest order in $B_\parallel$ the spin state of a 3/2 heavy hole in the exciton remains unchanged (*i.e.* perpendicular to the QW plane). Therefore when $B_\parallel$ is applied, one expects a strong reduction of the luminescence intensity related to the excitonic complexes and this can be considered as a specific manifestation of the role played indirectly by dark excitons in the magneto luminescence spectra of a quantum well.

3 Experimental set-up and samples

In order to quantify the aspects described above, we have investigated the photoluminescence (PL) and the photoluminescence excitation (PLE) spectra of InGaAs/GaAs single quantum well (SQW) in a magnetic field, perpendicular and parallel to QW's planes. To optimize the parameters of the investigated structure, one has to choose a wide enough undoped quantum well, so that interfacial effects would not mix the heavy- and light-hole states breaking the selection rules for the optical transitions and would not enhance the spin-orbit relaxation of carriers induced by a surface roughness scattering. For this purpose an InGaAs/GaAs heterostructure with a 20 nm InGaAs width (9.5% of In content) has been grown by MBE-technique on a (001)-surface of insulating GaAs substrate. The substrate or buffer layer imposes a relatively large compressive strain in the well layers. This splits off the quantum well light-hole state from the heavy-hole and as a result essentially simplifies the analysis of the system. We have performed both PL and PLE experiments on such a sample. The PL experiments have been performed exciting the sample with an Ar^+ laser. For the PLE measurements a tunable Ti-sapphire laser excited by all lines of the Ar^+ laser has been used. All spectroscopy investigations including analysis of the circular polarization of the luminescence have been performed using fiber optics. The high quality of the investigated SQW is confirmed by the observation of a narrow 1*s*HH-exciton line in luminescence (FWHM around 0.8 meV at $T = 1.5$ K and $B = 0$) and a small Stokes shifts (less than 0.3 meV) between PL and PLE spectra.

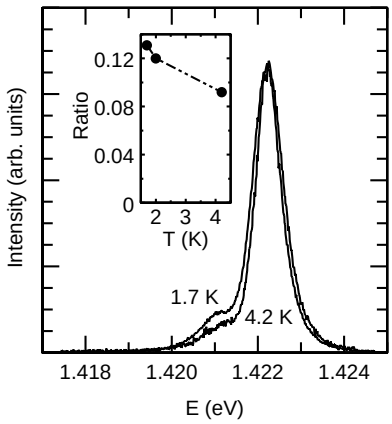

Fig. 2. Photoluminescence spectra measured at low excitation power in the range 1.7−4.2 K. The inset shows the decrease of the relative intensity of the excitonic complex with temperature.

4 Experimental data and their interpretation

Figure 2 shows the typical PL spectra measured at low photoexitation power (1 mW cm^{-2} at the output of the fiber with a 0.6 mm diameter core). At the low energy side of the optically active ground state of the heavy hole exciton 1*s*HH, a low intensity satellite line is clearly seen shifted by about 1.5 meV. In contrast to the ground state exciton line, this line is asymmetric in shape and has a relative strength with respect to the main line which increases when temperature decreases in the range 4.2−1.7 K (see inset of Fig. 2). What is more important, since it does not fit to any model of localized exciton on impurity sites, this satellite intensity behaves superlinearly with respect to the excitation intensity and only saturates at relatively high excitation levels. In PLE spectra, when the detection is fixed at the red side of the free exciton line (around 4 meV below the 1*s*HH-exciton maximum), besides the ground exciton state, no other feature is observed in absorption in the region of this satellite line. On the base of these properties, we assume that the luminescence channel of the observed satellite is connected to the radiative decay of free excitonic complexes, accompanied by the emission of a photon and a recoil particle-exciton in the case of excitonic molecule, or electron in the case of excitonic trion radiative decay.

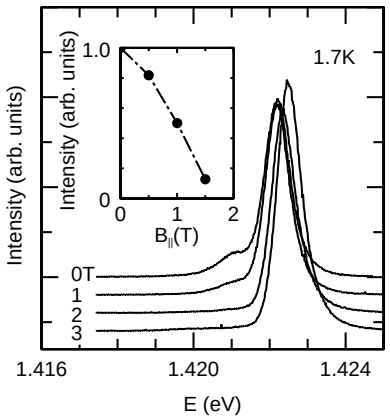

Fig. 3. Photoluminescence spectra, measured at $T = 1.7$ K, for different values of the magnetic field parallel to the QW plane. The inset shows the decay of the excitonic complex intensity with $B_\parallel$.

In a magnetic field $B_\parallel$, the PL intensity of the satellite strongly decreases, and at magnetic fields around 1.5−2 T this line completely disappears (see Fig. 3). In principle, such a satellite line on the red side of the main exciton line could be assigned to excitons trapped on any kind of impurity located in the barrier or in the well. It is known that in this case the corresponding optical transition does not disappear with the magnetic field. Therefore the observed variation with $B_\parallel$ definitively excludes that this channel is somehow connected with localized or bound excitonic states. We interpret this as a confirmation that the parallel magnetic field destroys the accumulation of dark excitons, and therefore the source for the efficient creation of free exciton complexes.

We first try to estimate the magnitude of $B_\parallel$, which makes a significant conversion of dark excitons into optically active ones. The electron spin precession of a dark exciton in $B_\parallel$ results in a finite radiative decay time $\tau_S^{-1}(B_\parallel)$ such that:

$$\tau_S^{-1}(B_\parallel) = \tau_S^{-1}(0) + \frac{x^2}{(1+\sqrt{1+x^2})^2 + x^2}\tau_A^{-1}$$

with

$$x = \frac{\mu_B g_e B_\parallel}{\delta}$$

and δ is the electron-hole exchange energy in the exciton (which provides the zero-field splitting between dark and optically active excitons [1–3]). Therefore, the number of accumulated dark excitons decreases with $B_\parallel$ since $n_D\ (B_\parallel) = G\tau_S^{-1}(B_\parallel)$ and the intensity of the complex satellite I_x varies like:

$$\frac{I_x(B_\parallel)}{I_x(0)} = \frac{1}{1 + \frac{x^2}{[1+\sqrt{1+x^2}]^2} \times \frac{\tau_S(0)}{\tau_A}}\,.$$

The magnetic field, for which I_x is halved can be estimated as $B_{1/2} \approx \frac{2\delta}{\mu_B g_e}\sqrt{\frac{\tau_A}{\tau_S(0)}}$, for $\tau_S(0)/\tau_A \gg 1$. For

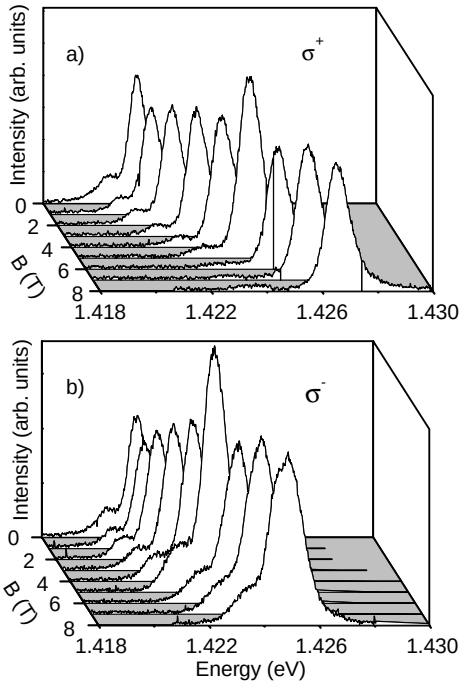

Fig. 4. Photoluminescence spectra, measured at $T = 1.7$ K, for different values of the magnetic field perpendicular to the QW plane. (a) σ^+ and (b) σ^- polarized spectra.

$\delta \approx 0.1$ meV [3], $\tau_S(0)/\tau_A \approx 10$ which corresponds to $\tau_A \approx 20$ ps and $\tau_S(0) \approx 200$ ps, and an absolute value of the electronic Landé factor ≈ 3.5 [12], we estimate $B_{1/2}$ around 0.3 T which is sufficiently close to observation.

Figures 4a and b show the results of the polarization studies of the satellite line in a configuration where the magnetic field is perpendicular to the plane of the quantum well. These studies have been performed to distinguish between the two kinds of free excitonic complexes – molecules or trions – which may be responsible for the radiative process giving rise to the satellite line in the spectrum. As mentioned above, the radiative decay of an excitonic molecule under perpendicular magnetic field should be accompanied by the emission of σ^+ and σ^- components of equal intensities, and simultaneously by the creation of two optically active recoil excitons $|-1/2;\ +3/2\rangle$ and $|+1/2;\ -3/2\rangle$ (see Fig. 1a). Besides, in a sufficiently strong enough magnetic field, when the Zeeman splitting in the exciton becomes larger than the EM binding energy, the molecular state is unstable at equilibrium owing to the alignment of hole spins and, as a result, both polarized components should disappear from the PL spectra.

On the other hand, the radiative decay of excitonic trion should be accompanied by the emission of corresponding σ^+ and σ^- components with intensities which are determined by the population of spin states of a hole having a large Zeeman splitting, and which are in the same ratio as those of an optically active exciton (see Fig. 1b). Besides, a recoil electron with spin $|+1/2\rangle$ or $|-1/2\rangle$ should accompany the radiative decay of a trion.

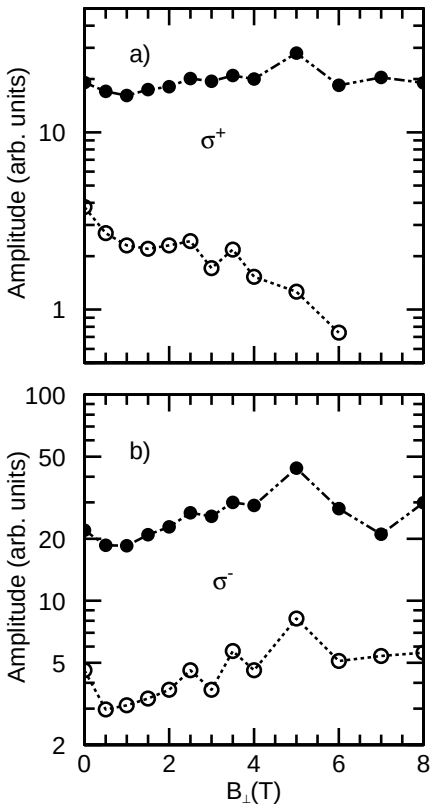

Fig. 5. Variation of the intensity of the exciton line (black dots) and of the trion line (empty dots) as a function of the magnetic field perpendicular to the QW plane for the σ^+ (a) and σ^- (b) polarization conditions.

The experimental PL spectra of the polarized components of excitons and excitonic complexes in perpendicular magnetic field demonstrate that the ratio of the intensities of the lower energy component σ^- of the exciton and a complex remains about the same when B increases (see Figs. 4a and b). At the same time the intensity of the high energy component σ^+ of the complex is strongly reduced as compared to the intensity of σ^+ exciton component (Figs. 5a and b). So these findings do not support the concept of the excitonic molecule decay and lead us to propose that, in the present experiment, the radiative decay of excitonic trions is responsible for the low energy satellite of the luminescence line. With this interpretation, the strong reduction of the σ^+ component of the trion luminescence in a perpendicular magnetic field can be used at least to estimate the trion binding energy. This component disappears when the spin splitting of X^- is around the trion binding energy (see Fig. 1b) which indeed occurs at $B = 6$ T (Fig. 5a). Thus the estimated trion binding energy is equal to $\Delta_T \approx 1$ meV the value found for the exciton spin splitting from PL and PLE experiments.

The interpretation of the present data in terms of excitonic trions assumes that some number of equilibrium electrons should exist in the investigated quantum well. These electrons can originate from residual n-type impurities present in the GaAs barriers surrounding the QW. This is a typical situation encountered in quantum wells with persistent electron concentrations of the order of $10^9 - 10^{10}$ cm^{-2} as already pointed out in reference [6].

5 Conclusion

The present magneto-optical studies of an InGaAs/GaAs single quantum well have shown that optically non-active, or dark excitons with a total angular momentum $M = \pm 2$ play the role of a reservoir for the creation of free excitonic complexes. Based on the intensity variation with magnetic field of the different polarized components of the structure found on the low energy side of the main exciton peak, this has been assigned to the recombination of an excitonic trion. Different techniques could be used to study this kind of complexes. Among others, a very attractive one seems to realize an optical detection of the electron spin resonance for dark excitons through the corresponding reduction of the intensity of the complex satellite.

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