Femtosecond study of the interplay between excitons, trions, and carriers in (Cd,Mn)Te quantum wells

P. Plochocka, P. Kossacki,1 W. Maślana,1,2 J. Cibert,2 S. Tatarenko,2 C. Radzewicz,1 and J. A. Gaj1,∗

1Institute of Experimental Physics, Warsaw University, Hoża 69, 00-681 Warsaw, Poland
2Laboratoire de Spectrométrie Physique, CNRS et Université Joseph Fourier-Grenoble, B.P.87, 38402 Saint Martin d’Hères Cedex, France

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We present an absorption study of the neutral and positively charged exciton (trion) under the influence of a femtosecond, circularly polarized, resonant pump pulse. Three populations are involved: free holes, excitons, and trions, all exhibiting transient spin polarization. In particular, a polarization of the hole gas is created by the formation of trions. The evolution of these populations is studied, including the spin flip and trion formation processes. The contributions of several mechanisms to intensity changes are evaluated, including phase space filling and spin-dependent screening. We propose a new explanation of the oscillator strength stealing phenomena observed in p-doped quantum wells, based on the screening of neutral excitons by charge carriers. We have also found that binding heavy holes into charged excitons excludes them from the interaction with the rest of the system, so that oscillator strength stealing is partially blocked.

Since the first experimental observation of charged excitons (trions) in semiconductor quantum wells (QW) it has been commonly recognized that optical spectra of moderately doped QWs contain neutral- and charged exciton lines, whose parameters are influenced by the presence of free carriers. Amongst the effects of carriers on the oscillator strength of the exciton, phase space filling (PSF) and carrier-carrier interaction are usually distinguished. PSF arises due to carriers with the same spin as that of the carrier entering the exciton; thus, in the case of photocreated carriers, the pump and probe beams have to be co-polarized. Carrier - carrier interaction (screening) takes place between carriers of any spin. However, as pointed out in [2], a cut-off of the screening is expected at the closest approach of like particles, as given by the size of the exchange-correlation hole. This effect, due to the Pauli exclusion principle, makes the screening by carriers of opposite spins more efficient. This applies to the neutral exciton when the free carrier and the carrier within the exciton have opposite spins (e.g., with photocreated carriers when the pump and probe beams are cross polarized), but not to the trion, which contains one carrier of each spin. In the same spirit, diffusion of the exciton on carriers was considered in [2] to be responsible for the polarization-selective broadening of the exciton line due to the presence of a polarized electron gas in GaAs-based quantum wells. A decrease of the exciton oscillator strength is systematically observed in transmission spectra of CdTe-based QWs in the presence of carriers with opposite spins (either electrons or holes). As it is the presence of these carriers which makes the formation of trions possible, this effect produces a balance between the decrease of the neutral exciton intensity and the increase of the trion intensity, which is often referred to as "oscillator strength stealing" (OSS). Theoretical efforts have been undertaken to describe OSS but no commonly accepted model has been devised to establish the existence of a "sum rule" between the intensities of the neutral and charged excitons with the same polarization. In what follows we reexamine the origin of OSS thanks to a sub-picosecond pump-probe experiment on a (Cd,Mn)Te/(Cd,Mg)Te QW. We first show that under our experimental conditions, PSF plays a minor role in the variation of the oscillator strength of (neutral) excitons, thus confirming a previous observation from CW experiments on (Cd,Mn)Te QWs where the hole gas was spin polarised thanks to the giant Zeeman effect. Then we show that spin-dependent carrier-carrier interaction is at the origin of the OSS. To this purpose, we use a pump-probe transmission experiment where we establish a controlled population of free carriers, neutral excitons, and charged excitons. Actually, we demonstrate a clear mechanism for the creation of a spin polarization in a 2D hole gas. Previously reported studies of the creation of a spin polarization in a 2D carrier gas were focused on n-type doping, and obtained thanks to the fast relaxation of the holes within the photocreated electron-hole pair. Here we show that the formation of charged excitons is an efficient way of inducing a polarization of the free carriers.

The time resolved study was carried out on a modulation doped structure consisting of a single 80 Å quantum well (QW) of Cd_{1−x}MnxTe (x ≈ 0.0018) embedded between Cd_{0.07}Zn_{0.012}Mg_{0.93}Te barriers grown pseudomorphically on a (100) Cd_{0.88}Zn_{0.12}Te substrate. Due to strain and confinement, only heavy holes have to be considered. Modulation p-type doping was assured by a nitrogen-doped layer at 200 Å from the QW. The density of the hole gas in the QW was controlled by an additional illumination with photon energy above the gap of the barriers, provided by a tungsten halogen lamp with a blue filter: the mechanism and its calibration are described in detail in [2]. The sample was mounted strain-free in a superconducting magnet and immersed in su-
perfluid helium at 1.8 K. The pulses were generated by a Ti$^{3+}$:Al$_2$O$_3$ laser tuned at 765 nm (1620 meV), at a repetition rate of 100 MHz. The 100 fs duration of the laser pulse assured a spectral width of about 40 nm (80 meV), much broader than the splitting between the neutral and the charged excitons. The pump and probe pulses were focused on the sample to a common spot of diameter smaller than 100 µm, and the spectrum of the probe pulse transmitted through the sample was recorded as a function of the pump-probe delay. The power of both pulses was controlled independently, the pump-to-probe ratio being at least 20 : 1. The average intensity and an increase of the neutral exciton one. I.e., photocreating ($\sigma^+$) trions (excitons bound to spin-down holes) has the same effect as decreasing the density of excitons by the carriers of photocreated excitons appears to dominate over PSF in such a (Cd,Mn)Te QW. A theoretical estimate of the effect of PSF [11], predicts it to be proportional to the area occupied by the exciton in the quantum well. Substituting the parameters of our QW we obtain a decrease of the oscillator strength by a few percent. This is much smaller than the experimental value (about 20 percent), supplying an additional argument for a minor role of PSF.

We turn now to the case of the QW with carriers, so that both the neutral exciton and the trion lines are visible. On transmission spectra before and shortly after the pump pulse (Fig. 1), we observe a decrease of the trion intensity and an increase of the neutral exciton intensity. The results shown in Fig. 1c, obtained from Gaussian functions fitted to transmission spectra, will be explained below assuming that PSF is negligible, and that screening by free carriers is more efficient than that by carriers engaged in excitons (neutral or charged). We first focus on the most pronounced effect, which is observed in the co-polarized configuration with the pump beam tuned to the trion line (Fig. 1b). We observe a decrease of the trion intensity and an increase of the neutral exciton one. I.e., photocreating ($\sigma^+$) trions (excitons bound to spin-down holes) has the same effect as decreasing the density of excitons by the carriers of photocreated excitons appears to dominate over PSF in such a (Cd,Mn)Te QW. A theoretical estimate of the effect of PSF [11], predicts it to be proportional to the area occupied by the exciton in the quantum well. Substituting the parameters of our QW we obtain a decrease of the oscillator strength by a few percent. This is much smaller than the experimental value (about 20 percent), supplying an additional argument for a minor role of PSF.

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FIG. 3: (a) Optical density for QW with hole concentration $p \approx 2.5 \times 10^{10} \text{ cm}^{-2}$. Solid line denotes negative delays (-8ps), dash line is for short positive delays (0.5ps) for co polarized when the pump pulse was tuned on $X^+$ absorption. (b) evolution of neutral $X$ and charged $X^+$ exciton oscillator strength if pump pulse is tuned to charged exciton. (c) evolution of $X$ and $X^+$ if pump pulse is tuned to neutral exciton.

The same spin-down holes in CW experiments where line intensities have been studied as a function of background carrier density. We propose a common description of the effects observed in the present pump probe experiment and those of the CW experiments. Then we observed a linear dependence of the trion oscillator strength on the population of holes with the appropriate spin, at constant total hole density. In the pump-probe experiment, a significant amount of the free carriers with spin down become bound into trions. They are no more available to form new trions in $\sigma^+$, so that the $X^+$ intensity decreases in this polarization. For the neutral exciton, in CW experiments, a linear decrease was observed and attributed to the OSS. We propose to interpret this OSS as the spin-dependent part of screening of the neutral exciton by free holes. As mentioned above, due to the Pauli exclusion principle, this reduction is stronger in $\sigma^+$ polarization for free holes with spin down. In the pump-probe experiment, those holes are used to form the trions and therefore are excluded from the exchange interaction with neutral excitons, so that the screening of $X$ is reduced. Our interpretation removes the conceptual difficulty of the sum rule, which can be invoked to explain the CW results by arguing that introduction of holes into the quantum well creates new trion states at the expense of neutral exciton states. In our pump-probe experiments the number of the trion states does not change, only their occupation is modified by the pump pulse, nevertheless the OSS effect occurs. The proposed interpretation explains coherently both types of experiments. Note that when the pump beam is tuned to the neutral exciton line, Fig.3, similar effects are observed, with a delay. The rise time of $X$ decreases with the initial hole density (Fig.4).

We identify this rise time with the trion formation time, estimated using suitable rate equations (not shown) as 5 ps, 2 ps, and 1 ps for hole densities $2.5 \times 10^{10} \text{ cm}^{-2}$, $3 \times 10^{10} \text{ cm}^{-2}$, and $4 \times 10^{10} \text{ cm}^{-2}$ respectively. As expected, these values are shorter than the value of 65 ps reported before for similar QWs with smaller hole densities (the $X$ and $X^+$ lines were of similar intensities in [4, 12], while in the present sample the $X$ line is absent in CW photoluminescence spectra excited nonresonantly, not shown, proving a much higher hole density). Besides, a higher temperature due to strong excitation may contribute to the faster trion formation.

It is interesting to discuss now the weaker effects observed in the cross-polarized configuration. When pumping into the $(\sigma^+)$ neutral exciton, we create spin-up holes, thus increasing the screening of the $(\sigma^-)$ charged and neutral excitons. This results in an intensity decrease of both the neutral and charged excitons, which is small due to the binding of these holes within excitons, and even weaker for the charged exciton due to the Pauli exclusion principle. This is observed in Fig.4. When pumping into the trion however, in addition to the former mechanism of screening by spin-up photocreated holes, we trap free holes of spin-down into trions, thus reducing their efficiency in the screening of the excitons. In the case of the neutral exciton, we thus have a balance between two weak effects of opposite sign (spin-up holes because they are bound in excitons and spin-down holes due to the Pauli exclusion principle), so that a very small effect, if any, is seen. In the case of the trion, screening by the photocreated spin-down holes is even further reduced by Pauli exclusion, so that one observes a fast increase of the $(\sigma^-)$ trion intensity when pumping into the trion, due to the
reduction of screening by the spin-down free holes.

At longer pump-probe delays we observe a slow recovery of the oscillator strength, which we attribute to spin relaxation. At time delays longer than the trion formation time and smaller than its recombination time, no matter whether the pump beam created excitons or trions, we deal with two populations: trions and free holes. The total population of these two species is constant, but both exhibit a spin polarization. In particular, a nonequilibrium spin polarisation of the free hole gas has been created by optical excitation of the trions, which trap spin-down holes. Both polarisation relax, and two spin flip processes have to be considered: (i) spin flip of the free holes from the spin-up free hole gas unaffected by the pump to the spin-down hole gas depleted by the formation of $X^+$; (ii) spin flip of the $X^+$, which actually is that of the electron bound in the trion. However, as previously, we expect that the dominant contribution to screening comes from the free holes. Thus, fitting the difference between the exciton intensities measured in the two polarizations with an exponential function, we determine the spin relaxation time of free heavy holes equal to 8 ps, weakly dependent on the hole density. These values are comparable to the spin relaxation times determined for holes bound in exciton and charged exciton complexes. The photoluminescence experiments with much lower excitation power and very similar heterostructures gave values from about 3ps through 20 ps up to 35ps. The difference might be a result of different Mn content, temperatures and density of photo-created carriers. In particular, increase of k-vector and scattering processes lead to shortening of spin flip time, as predicted theoretically. It was shown experimentally for a similar n-type sample that for the hole in $X^-$ the spin relaxation time may be decreased from 35ps to below 5ps by varying the excitation energy. To conclude, the evolution of the charged and neutral exciton line intensities (including oscillator strength stealing) is well explained by neglecting phase space filling and assuming that: - the trion intensity increases with the population of free carriers with the opposite spin - neutral exciton screening is spin dependent, trion is not, in accordance with the Pauli exclusion principle - screening by free carriers is more efficient than screening by carriers engaged in excitons.

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* Electronic address: {Jan.Gaj@fuw.edu.pl}

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FIG. 4: Evolution of the X oscillator strength for different hole densities, as indicated, with the pump beam tuned to X line.