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Oscillator strengths of charged excitons: combining magnetoabsorption and photoluminescence dynamics in semimagnetic quantum wells

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Abstract

We present a systematic study of the oscillator strength of the positively charged excitons (X^+) in $Cd_{1-x}Mn_xTe$ quantum wells. CW-absorption and time-resolved photoluminescence measurements were combined as two approaches for the determination of the oscillator strength. By varying (in a small magnetic field) the spin subband hole distribution at constant total concentration, we observe an increase of the oscillator strength in absorption, proportional to the hole concentration in one spin subband (the one which allows the X^+ formation). The measurements done for different total hole concentrations show an important decrease of the X^+ *oscillator strength per carrier* when increasing the total concentration. On the contrary the radiative lifetime, measured in time-resolved PL experiment, is found to be constant over the whole range of hole gas concentrations, and equal to the value which we deduce from transmission in the limit of vanishing hole concentration.

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Interest in optical effects resulting from the exciton-carrier interactions in the presence of a 2D-carrier gas of low density, has been renewed recently by the observation of charged excitons [1–3] in several types of quantum wells (QW). Several aspects of this new quasiparticle have been extensively studied. One of them is the oscillator strength which has been analyzed on the basis of reflectivity [4–6] and absorption measurements [7]. Also, the radiative lifetime was measured and discussed for several systems [8–11] and is usually considered to be linked to the oscillator strength per exciton. In this communication we combine the

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determination of the oscillator strength from the CW transmission spectra, with the direct measurement of the charged exciton radiative lifetime from time resolved photoluminescence (PL).

Additionally, choosing a semimagnetic material ($Cd_{1-x}Mn_xTe$, with very low Mn concentration $x = 0.0018$), we exploited the unique opportunity to tune independently the total hole concentration (through barrier illumination) and its distribution among the two Zeeman split $\pm\frac{3}{2}$ spin subbands (by applying a weak magnetic field) [12]. Therefore, we were able to analyze independently the effects of carrier polarization and of total carrier concentration, as opposed to the direct influence of the magnetic field present in most studies of similar systems. In particular, we can distinguish spin-independent effects from spin-dependent ones. This is especially important for the analysis of the oscillator strength because it allows us to distinguish the true population effects from screening and electrostatic field variation, which are usually dominant in nonmagnetic structures [3,8,9].

In our experiment, we used samples grown by molecular beam epitaxy, consisting of a single 80 Å QW of $Cd_{1-x}Mn_xTe$ embedded between $Cd_{0.66}Mg_{0.27}Zn_{0.07}Te$ barriers. The nitrogen-doped region in the front barrier was located at a distance of 200 Å from the QW. The nominal hole concentration in the QW was $2\times10^{11}\,cm^{-2}$.

To control the carrier concentration we used above-barrier illumination, which reduces the concentration of the hole gas in the QW due to the diffusion of electrons photocreated in the barrier into the QW [13]. These electrons recombine with the holes and under continuous illumination the system approaches a stationary state with a decreased hole concentration. The efficiency of this process was calibrated in the high concentration range by measuring the Moss–Burstein shift (the distance between the PL line and the energy of the absorption edge) and is discussed in detail in Ref. [12].

All magnetooptic measurements were performed in the Faraday configuration with the magnetic field applied perpendicular to the sample surface. The sample was mounted strain-free in a superconducting magnet and immersed in liquid helium. Transmission spectra were obtained with a halogen lamp conveniently filtered to avoid hole depletion. We used an $Al_2O_3:Ti$ laser providing about $2\,mW\,cm^{-2}$ for CW PL, and for time-resolved PL a picosecond tunable $Al_2O_3:Ti$ laser with a 2 ps pulse width, a repetition rate of 80 MHz, and an averaged power density of less than $100\,mW\,cm^{-2}$. The PL signal was collected through a spectrometer by a 2D streak camera with 10 ps resolution.

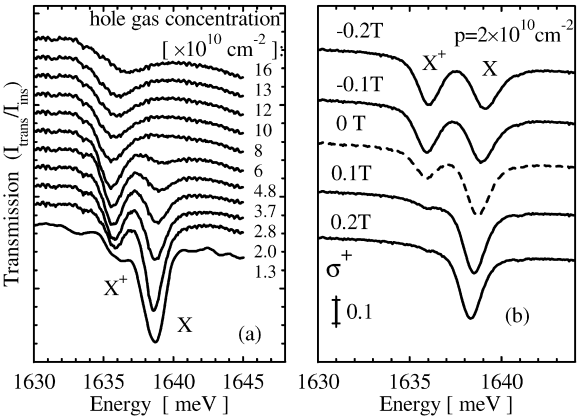

Fig. 1. Transmission spectra (transmitted intensity/incident intensity) at a temperature of 1.3 K: (a) at zero-field for different hole gas concentrations (controlled by additional blue light illumination) (b) at a density of $2\times10^{10}\,cm^{-2}$ as a function of magnetic field.

Typical transmission spectra as a function of the hole gas concentration and magnetic field are presented in Fig. 1 (a and b, respectively). At zero field the two lines related to neutral (X) and positively charged exciton (X^+) are observed. At the lowest concentration, $\approx 1.3\times10^{10}\,cm^{-2}$, both are sharp and the X line is much stronger than X^+. When the carrier concentration increases, the X^+ line becomes more intense while the X line weakens and finally disappears at a hole concentration of about $8\times10^{10}\,cm^{-2}$. For low hole concentrations the distance between the two lines approaches 2.7 meV, i.e., the same value as that reported in CdTe–CdZnMgTe QWs of similar characteristics [2]. These findings support the identification of these lines. The unambiguous test is given by studying quantitatively the influence of a magnetic field. Fig. 1b shows typical transmission spectra at constant hole concentration, taken at several magnetic fields in both circular polarizations. We observe a

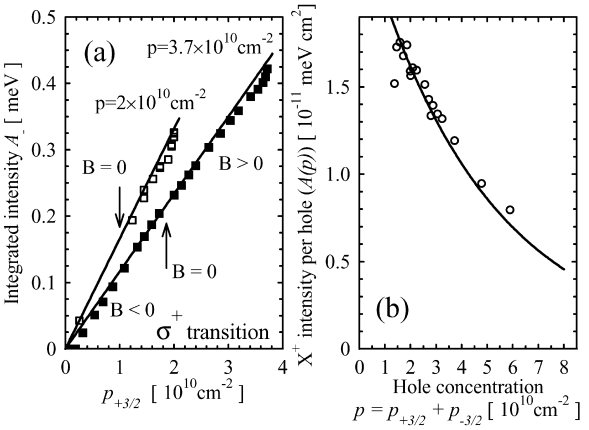

Fig. 2. (a) Integrated intensity of X^+ transmission line versus $p_{+3/2}$ hole concentration, keeping the total hole concentration constant. The $p_{+3/2}$ hole concentration was controlled by a magnetic field varying from -0.5 to 0.5 T. The temperature was 1.3 K. Squares represent experimental data and solid lines are a fit of X^+ intensity per $p_{+3/2}$ hole (with Eq. (1)). (b) $A(p)$ as a function of the total hole gas concentration.

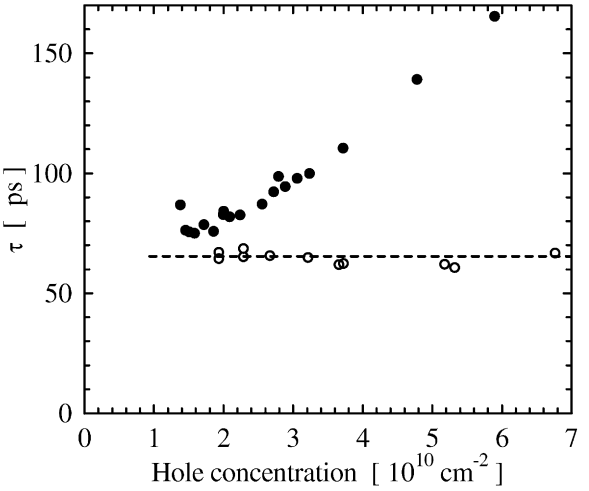

Fig. 3. X^+ PL decay time versus hole gas concentration: (open symbols) – experimental (measured at 1.5 K); (closed) – values obtained from absorption intensity per hole (Eq. (3)).

characteristic population effect on the X^+ line. Its intensity increases with field in σ^- polarization, while it decreases in σ^+ to disappear at a field of about 0.2 T. In σ^+ polarization, the creation of an X^+ in its singlet state involves a photocreated spin-up hole and a pre-existing spin-down hole. Our observation reflects the disappearance of such spin-down holes [1] at large enough field.

Quantitatively, we analyze the intensity of both transitions by fitting two Gaussian functions to the absorption spectra. The integrated intensity of the X^+ line when varying the magnetic field is shown in Fig. 2a, as a function of the concentration of holes with spin $+\frac{3}{2}$ $(p_{+3/2})$. The hole distribution between the Zeeman split subbands was obtained by using a Maxwell–Boltzmann distribution, with the valence band Zeeman splitting deduced from the directly measured splitting of the exciton line by assuming the ratio 1:4 of the conduction- to valence band splitting [14]. Note that we used the Maxwell–Boltzmann distribution to describe the population ratio at low hole density, as done in Ref. [2] for a CdTe QW. At higher density the Fermi–Dirac distribution should be used [4–6]. As examples, experimental results obtained for two different total hole concentrations p are displayed in Fig. 2a. At constant p, we find that the intensity $A_\pm$ of the X^+ line in $\sigma^\pm$ polarization is proportional to the population of holes with the appropriate spin, i.e., $p^\pm$ respectively,

$$A_\pm = A(p)p_\mp, \tag{1}$$

where the absorption intensity per carrier $A(p)$ is proportional to the oscillator strength. Similarly, good fits were achieved at 4.2 K or at other hole densities. The slope [i.e., $A(p)$] decreases when the total hole density increases, as shown in Fig. 2b.

We now turn to the time-resolved PL results. The X^+ lifetimes were extracted from the monoexponentional decay of the PL signal. Under non-resonant excitation (with a laser energy higher than the X transition) and hole gas concentration $p < 1.3\times10^{10}\,\mathrm{cm}^{-2}$, X was at least 10 times less intense than X^+. When excited resonantly with X^+, no neutral exciton line was detected in PL, and therefore the process of X^+ dissociation could be neglected (details of the internal dynamics of the X–X^+ system in a CdTe QW are reported in Refs. [10,11]). In both cases, the PL dynamics was dominated by the charged exciton radiative recombination and we measured the same decay time, shown in Fig. 3 as a function of hole gas concentration. Within experimental accuracy it is constant over

the whole range of concentrations for which sharp lines were observed in absorption. The measured value of 65 ps is in good agreement with lifetimes reported for similar structures [8–11]. Also, applying a magnetic field (up to 0.7 T) did not change this value: this agrees with the fact that the absorption coefficient, for a given total hole concentration, is proportional to the concentration of preexisting holes in the relevant spin subband. The radiative character of the observed PL decay (absence of nonradiative processes) was tested by a PL measurement under a strong blue light illumination, which further decreased hole gas concentration and increased its temperature. No decrease of the total energy- and time-integrated PL signal was observed even when the PL spectra were completely dominated by X line and the decay time increased by a factor of 2.5.

The quantitative comparison of both sets of data (PL and absorption) can be done using an atomic-like formula [7,15], which has been successfully applied for excitons bound to impurities. Such an approach leads to the following relation between lifetime and absorption:

$$\frac{\hbar}{\tau} = \frac{n^2\omega^2}{3\pi^2 c^2} A(p), \tag{2}$$

where n is the refractive index, c the velocity of light, and ω the frequency.

The expected lifetime calculated using absorption data (Fig. 2b) and taking into account the effect of the cap layer [12,16] is shown in Fig. 3. One notices a good agreement between both experiments for vanishing hole density, while for the higher ones the difference increases and reaches a factor of two for $6 \times 10^{10}\,\mathrm{cm}^{-2}$.

The observed difference has no obvious explanation. It suggests that the states observed in absorption are different from the one participating in PL. The calculation of Stébé et al. [17] shows that at high density, the absorption involves preexisting carriers with higher wave vectors, for which the transition matrix element is smaller. Therefore, one may expect the reduction of the oscillator strength with respect to that at $\boldsymbol{k} = 0$ which is involved in PL. Another mechanism could be based on the localization of the trions.

In conclusion, we have combined the determination of the oscillator strength of trions from CW transmission experiments, with the direct measurement of the charged exciton radiative lifetime. Varying the spin subband hole distribution (by magnetic field) we observe an increase of charged exciton oscillator strength proportional to the hole concentration in one spin subband. The measurements done for a fully polarized hole gas (in magnetic field up to 0.7 T), show an important decrease of the X^+ *oscillator strength per carrier of a given spin* when increasing the total hole gas density. On the other hand, the charged exciton radiative lifetime, measured in time-resolved PL experiment, is found to be constant over the whole range of hole gas concentrations, and equal to 65 ps. This value corresponds to the oscillator strength per hole obtained from transmission in the limit of vanishing hole concentration.

The direct comparison of the measurement of the oscillator strength in absorption and time-resolved luminescence leads to interesting conclusions. In particular it shows, e.g., that due to phase space filling and/or localization, charged excitons may have very different characteristics when observed in transmission and in PL.

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