Oscillator strength transfer from X to X^+ in a CdTe quantum-well microcavity

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We report on a study of oscillator strength transfer from the neutral exciton X to the positively charged exciton X^+, in a modulation doped CdTe quantum well embedded in a microcavity. The strong-coupling regime between X or X^+ and the cavity photon mode has been analyzed for various hole densities p and the oscillator strengths f_X and f_{X^+} are derived accurately from polariton line energies. For increasing p, f_{X^+} increases and f_X is shown to decrease linearly with f_{X^+}. We propose a model that accounts for those oscillator strength variations. This model relies on an effective cross section $S_{X^+}=6.6\times10^4$ Å^2 for the creation of X^+ in the vicinity of a hole, and a saturation density equal to 10^{11} cm^{-2} for phase-space filling effect.
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I. INTRODUCTION

The existence in semiconductor materials of trions or charged excitons (X^-, X^+) resulting from the binding of an electron or a hole with an exciton (X) was predicted by Lampert in 1958.[1] The additional carrier has a weak binding energy, so that the observation of these charged excitons in bulk materials is only possible at very low temperatures. However, in two-dimensional (2D) structures, the binding energy of the additional carrier is dramatically increased,[2] and in 1993 Kheng *et al.* reported the first observation, to our knowledge, of 2D X^- in a n-type modulation doped CdTe quantum well (QW).[3] It was followed by the observation of both kinds of trions in III-V 2D systems,[4–6] and X^+ was also observed in II-VI QW samples.[7] In the pioneering experimental papers pointing out the existence of X^- (X^+) in 2D structures, optical lines were identified as charged excitons on the basis of magneto-optical study, temperature dependence, and selection rule considerations. Among the numerous experimental works that have followed on the effect of 2D carrier gas on excitonic properties, a few papers deal with the relative strength of X and X^- versus electron-gas density. Shields *et al.*[8] observed, in GaAs QW's, a quenching of neutral exciton photoluminescence (PL) and the emergence of X^- PL line when the electron density increases. However, PL studies are difficult to interpret since they involve both a complicated thermal equilibrium between trions and neutral excitons, and dynamics. Recently, Miller *et al.*[9] showed quantitatively by absorption measurements on multi-quantum-well structures that there is both a sharing of the absorption strength between X and X^- and a decrease of their summed absorption when the 2D electron-gas density increases. Note that the idea that the trion seems to steal oscillator strength from the neutral exciton was already proposed by Kheng *et al.*[3] However, the efficiency of this transfer of oscillator strength is still an open question.

We present an experimental investigation of this phenomenon of oscillator strength transfer from X to X^+ in a single p-type modulation doped CdTe quantum well. The QW we use is identical to one period of the multi-QW in which Haury *et al.*[7] identified X^+. The key point of our work is that we insert this QW in the middle of a microcavity. Hence, the oscillator strengths of X and X^+ are extracted from their coupling with the confined photon mode when the cavity mode is brought into resonance with exciton states. This effect, known as strong-coupling regime between QW excitons and cavity modes, was observed in a semiconductor microcavity by Weisbuch *et al.*[10] in 1992. The main feature of this regime is an anticrossing between exciton and photon states giving rise to mixed states called cavity polaritons.[11] Strong coupling is achieved when the coupling effect is significantly larger than both the exciton and cavity mode linewidths. In this case the energy levels of the system can be described as the eigenvalues of the following Hamiltonian:

$$H=\begin{pmatrix} E_{\mathrm{ph}} & V_X \\ V_X & E_X \end{pmatrix}, \tag{1}$$

where E_{ph} and E_X are the energies of the photon and exciton modes, and V_X is defined by

$$V_X=\hbar\sqrt{\frac{e^2}{4\pi\epsilon_0\epsilon_r}\frac{2\pi}{m_0L_{\mathrm{eff}}}f_X}. \tag{2}$$

ϵ_r is the dielectric constant in the cavity, m_0 is the free electron mass, L_{eff} is the effective photon length in the cavity, which depends on refractive index contrast of the heterostructure materials, and f_X is the 2D exciton oscillator strength.[12] When the detuning δ between exciton and photon energies vanishes ($\delta=E_{\mathrm{ph}}-E_X$), the two polariton lines are split by the so-called Rabi splitting $\hbar\Omega_{\mathrm{Rabi}}=2V_X$. The oscillator strength f_X (per unit area) is then directly proportional to the square of the Rabi splitting energy measured on optical spectra [Eq. 2].

In the present case polariton states arise from coupling of the photon mode with both X and X^+ simultaneously, and the aim of this work is to extract, for various hole densities, f_X and f_{X^+} from polariton line energies measured on reflectivity spectra as a function of the exciton-photon detuning. The interest of this approach for a quantitative oscillator strength study is that all the information is contained in optical line energy separations with no need of absolute absorp-

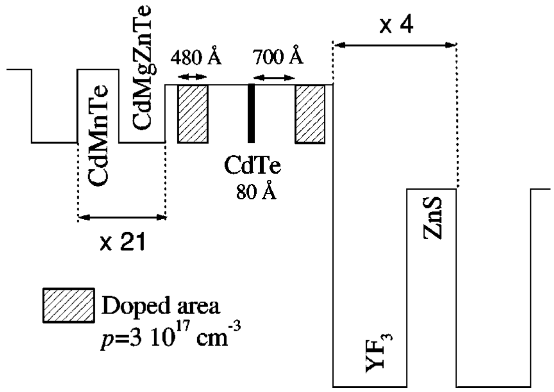

FIG. 1. Scheme of the sample on a refractive index scale. First, a 21-pair $Cd_{0.69}Mn_{0.31}Te/Cd_{0.38}Zn_{0.02}Mg_{0.60}Te$ Bragg mirror has been deposited on a $Cd_{0.88}Zn_{0.12}Te$ substrate (not shown). Then the active layer consists of a 80-Å-thick CdTe QW located in the middle of a $Cd_{0.69}Zn_{0.08}Mg_{0.23}Te$ λ cavity. 480 Å p-doped regions are located 700 Å from the QW on both sides. The surface of the sample is covered by a 4-pair YF_3/ZnS Bragg mirror.

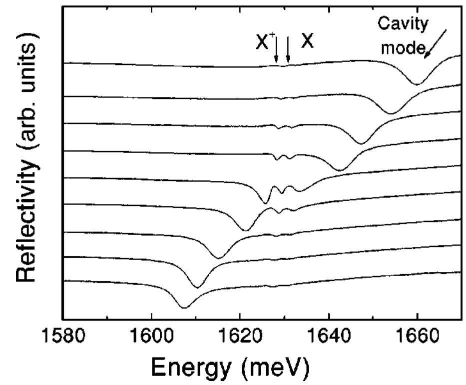

FIG. 2. Reflectivity spectra at 1.7 K corresponding to different positions along the wedged-shape cavity. The 2D hole concentration in the quantum well is $p_0=8\times10^{10}$ cm^{-2}. Three polariton lines arising from the strong coupling between the exciton states and the cavity mode are observed.

tion measurement and no difficulty with integrated absorption and baseline determination. This technique is also very sensitive and allows us to investigate a single QW instead of a multi-QW, thus avoiding effects of QW width or carrier gas density fluctuations. The drawback is that the sample growth and design are more complicated.

II. SAMPLE GROWTH AND EXPERIMENTAL DETAILS

The microcavity sample used in this study was elaborated by two different techniques: a 21-period $Cd_{0.38}Zn_{0.02}Mg_{0.60}Te/Cd_{0.69}Mn_{0.31}Te$ Bragg mirror and the active layers (λ cavity and QW) were first grown by molecular-beam epitaxy on a $Cd_{0.88}Zn_{0.12}Te$ substrate. Then a 4-period YF_3/ZnS Bragg mirror was evaporated on top of the sample (Fig. 1). The active layer is a single 80-Å-thick CdTe QW with p-type doped barriers. The QW is located at an antinode of the confined electromagnetic field in the middle of the λ cavity. An electron-cyclotron-resonance nitrogen plasma source was used to obtain a doping level of about 3×10^{17} cm^{-3} in the barriers on a thickness of 480 Å, and the QW is separated from the doped layers by 700-Å-thick undoped spacer layers. The doped regions are located symmetrically on each side of the QW to avoid electric-field effects and subsequent oscillator strength reduction by a spatial separation of electrons and holes inside an asymmetric QW. The barrier material is $Cd_{0.69}Mg_{0.23}Zn_{0.08}Te$, which has a large band gap, and a large enough valence-band offset with CdTe to ensure a good hole transfer from nitrogen acceptors into the well.[13] The 2D hole density in the QW has been evaluated by a C-V measurement at room temperature[14] before evaporating the top mirror, and is equal to $p_0=8\times10^{10}$ cm^{-2}. The relatively high doping level of the barriers was obtained thanks to a structure matched with the $Cd_{0.88}Zn_{0.12}Te$ substrate, which prevents nitrogen diffusion.[14] Moreover, the sample was wedged during the growth by positioning the substrate off the effusion cell axis. Hence, by moving the observation spot along the sample, the strong variation of the photon mode energy with the cavity thickness enables us to study exciton-photon coupling as a function of the exciton-photon energy detuning. We will focus only on coupling with the fundamental excitonic state, which involves heavy holes because compressive strain in the QW splits the valence band into light- and heavy-hole subbands. In the following, for the sake of simplicity, the heavy-hole exciton will be referred to as exciton.

Reflectivity measurements were performed, at normal incidence, in a helium bath cryostat, at 1.7 K, with a tungsten lamp. The incident light was filtered to reject photons with energies higher than the barrier material band gap, and the reflected light was analyzed with a monochromator coupled to a CCD camera. The carrier density in the QW can be decreased from $p=p_0$ to $p\approx0$ by recombination of the hole gas with electrons photocreated in the barriers[16] by the 5145 Å line of an Ar^+ laser, whose intensity was increased from zero to about 1 mW/cm^2. The dielectric materials YF_3 and ZnS of the top mirror were chosen because they are transparent at the laser wavelength.

III. RESULTS AND DISCUSSION

For a hole concentration $p\approx0$, the experimental data show two polariton lines, arising from the strong coupling between the QW exciton and the cavity photon mode. This effect, briefly introduced in Sec. I is fully developed in Ref. 11, and references therein. The Rabi splitting measured between the two polariton lines when exciton and photon mode energies are brought into resonance is: $\hbar\Omega_{\mathrm{Rabi}}=2V_X=8.2$ meV. For $p=p_0$ the situation is somewhat different: we observe three polariton states resulting from the coupling between X , X^+, and the cavity mode. This is shown in Fig. 2 by a series of reflectivity spectra corresponding to different positions along the wedged-shape sample. To analyze these data, we take into account not only a variation of the photon mode energy E_{ph} with the sample thickness but also slight variations of the uncoupled exciton energies E_X and E_{X^+}. For this purpose, the sample thickness is represented by the detuning δ of the cavity mode defined with respect to the X exciton energy at resonance E_0: $\delta=E_{\mathrm{ph}}-E_0$, with $E_0=1630.7$ meV. We assume that the energies of X and X^+ are given by $E_X(\delta)=E_0+\alpha\delta$ and $E_{X^+}(\delta)=E_0-W+\alpha\delta$ with $\alpha=0.042$. The charged exciton binding energy W slightly

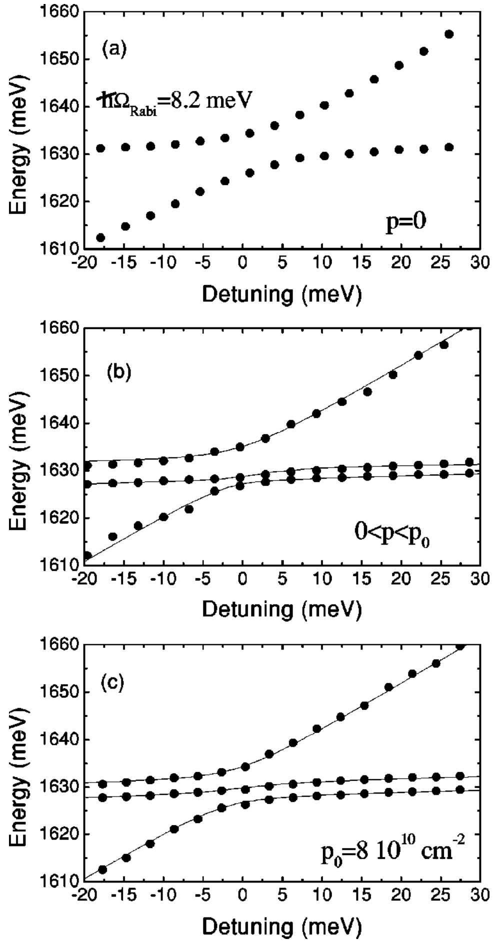

FIG. 3. Polariton energies as a function of the exciton-photon detuning (defined in the text). Dots are experimental results and lines represent model calculations. The 2D hole density vanishes in (a), is equal to $p_0 = 8\times10^{10}$ cm^{-2} in (c), and has an unknown intermediate value in (b).

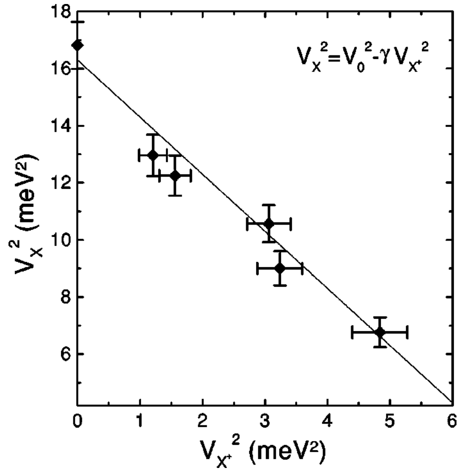

FIG. 4. V_X^2 vs $V_{X^+}^2$ for hole densities varying from $p=0$ to $p_0 = 8\times10^{10}$ cm^{-2}, where V_X and V_{X^+} are coupling matrix elements between exciton and photon states. This demonstrates a linear relation between the two oscillator strengths $f_X(p)$ and $f_{X^+}(p)$ which are respectively proportional to V_X^2 and $V_{X^+}^2$.

varies with hole density ($W = 3.5$ meV for $p = p_0$). The slope α is obtained from optical measurements far below and above the anticrossing region at the two edges of the sample. The slight variation of excitonic energies with δ results from the dependence of the electron and hole confinement energies on the QW thickness. Energies of the polariton lines as a function of δ and for different hole densities are plotted in Fig. 3. Figure 3(a) shows the particular case obtained for $p = 0$ when the hole gas density vanishes. In Fig. 3(c) the hole density is maximum (no laser beam), and in Fig. 3(b) the intensity of the Ar^+ laser has been attenuated so that $0 < p < p_0$. The important feature is that polariton energies around $\delta = 0$ are shifted compared to the uncoupled states, and the shift depends on p because the oscillator strength of X and X^+, namely, their coupling with the photon mode, depend on p.

We describe the energies of this three level system by the eigenvalues of the Hamiltonian:

$$H(\delta) = \begin{pmatrix} E_{\mathrm{ph}}(\delta) & V_X & V_{X^+} \\ V_X & E_X(\delta) & 0 \\ V_{X^+} & 0 & E_{X^+}(\delta) \end{pmatrix}. \tag{3}$$

This model is valid unless polariton lines are too broad compared to their separation, but in our case polariton lines are well resolved whatever the detuning. To extract the coupling terms V_X and V_{X^+}, we have calculated numerically the eigenvalues of $H(\delta)$. For each value of the carrier density p, we compared experimental measurements and calculated curves of polariton energies versus δ. The parameters V_X and V_{X^+} were adjusted to obtain the best fit between experiment and calculations (Fig. 3). For example, for $p = p_0$, the best fit is obtained with $V_X(p_0) = 2.6$ meV and $V_{X^+}(p_0) = 2.2$ meV [Fig. 3(c)]. In Fig. 3(b) the best fit corresponds to $V_X = 3.4$ meV and $V_{X^+} = 1.1$ meV.

In Fig. 4, V_X^2 is plotted versus $V_{X^+}^2$ for hole concentration p varying from $p=0$ to $p_0 = 8\times10^{10}$ cm^{-2}. We observe a linear behavior $V_X^2(p) = V_0^2 - \gamma V_{X^+}^2(p)$, with $V_0 \approx 4$ meV and $\gamma \approx 2$. According to Eq. 2, this demonstrates also a linear dependence of $f_X(p)$ versus $f_{X^+}(p)$: $f_X(p) = f_X^0 - \gamma f_{X^+}(p)$. f_X^0 is the oscillator strength of the exciton X in the CdTe QW free of hole gas ($f_X^0 = 2.2\times10^{13}$ cm^{-2}). It corresponds to a Rabi splitting of 8.2 meV, which is typical for a single CdTe QW and agrees with previous data from undoped CdTe QW's in a microcavity.[15] The fact that γ is found to be larger than 1 shows that the summed oscillator strength decreases with p.

We propose a simple model that accounts for this linear dependence. The model, valid for a dilute hole gas, assumes both a sharing of the QW surface between hole sensitive or insensitive areas and a quenching of X and X^+ transitions when the 2D hole gas density increases. Within this approach, the oscillator strength, per surface unit, for X and X^+, respectively, have the expressions:

$$f_X(p) = f_X^0\left(1 - \frac{p}{2} S_{X^+}\right)\rho(p), \tag{4}$$

$$f_{X^+}(p) = f_{X^+}^0 \frac{p}{2} S_{X^+} \rho(p). \tag{5}$$

p has been divided by 2 because of the spin degeneracy of the heavy-hole valence subband. $\rho(p)$ stands for screening effects $[\rho(p)\leqslant 1]$. f_X^0 is the oscillator strength of X at vanishing hole densities or far from a hole, and $f_{X^+}^0$ is the analogous for X^+ when holes are available. We also define an effective cross section per hole S_{X^+} for the creation of X^+. This model is quite intuitive if we assume a localization of holes and charged excitons in potential fluctuations. Brinkmann *et al.* evidenced such localization effects, in p-type modulation doped CdTe QW's, from a determination of the exciton and trion dephasing rates by four-wave mixing.[17]

Up to that point, $\rho(p)$ could stand for all kinds of screening effects. It has been found in 2D systems that phase-space filling (i.e., Pauli exclusion principle) dominates over Coulomb screening and is described by

$$\rho(p)=\frac{1}{1+p/p_c}, \tag{6}$$

where p_c is a critical hole density.[18] We assume this expression, although we have no direct evidence here that Coulomb screening is negligible. Combining Eqs. (4), (5), and (6), we obtain a linear relationship between f_X and f_{X^+}, $f_X=f_X^0-\gamma f_{X^+}$, with

$$\gamma=\frac{f_X^0}{f_{X^+}^0}\left(1+\frac{2}{p_cS_{X^+}}\right). \tag{7}$$

The ratio $f_X^0/f_{X^+}^0$ has been evaluated recently by magneto-optical measurements[19] in very similar QW's, except a few percent Mn in the well to enhance the valence-band Zeeman splitting for low magnetic-field studies. Their experimental result is $f_X^0/f_{X^+}^0\approx 0.5$. With this ratio and the experimental determination of γ $(\gamma\approx 2)$, we get $p_cS_{X^+}\approx 0.66$. Moreover, considering the case $p=p_0$ [Fig. 3(c)], we get from Eqs. (4) and (5):

$$\frac{f_{X^+}(p_0)}{f_X(p_0)}=\frac{f_X^0}{f_{X^+}^0}\left(\frac{1-(p_0/2)S_{X^+}}{(p_0/2)S_{X^+}}\right).$$

Experimentally $f_{X^+}(p_0)/f_X(p_0)=V_{X^+}^2(p_0)/V_X^2(p_0)\approx 1.4$ and $p_0=8\times 10^{10}$ cm^{-2}. We get $S_{X^+}\approx 6.6\times 10^4$ Å^2 and $p_c\approx 10^{11}$cm^{-2}. The radius of this cross section is 145 Å , that is about twice the 2D exciton Bohr radius in CdTe QW's, and it is clearly smaller than the mean hole distance for $p=p_0$, which is about 355 Å. The saturation density expressed in units of reciprocal exciton surface is $p_c\approx 0.11/\pi a_0^2$ with $a_0=60$ Å. This dimensionless critical density is twice the saturation density calculated by Schmitt-Rink *et al.* for screening by free-carrier pairs in the low-temperature limit.[18] Their model, purely bidimensional, takes into account phase-space filling and exchange interaction.

IV. CONCLUSION

A microcavity containing a modulation doped CdTe QW was used to measure the oscillator strengths f_X and f_{X^+} as a function of the 2D hole gas density p in the QW. We have shown that f_X decreases with increasing p whereas f_{X^+} increases with a linear relationship between these two oscillator strengths. The effect of oscillator strength transfer and reduction of the summed oscillator strength is consistent with a simple model, valid for small 2D hole density, where the efficiency of holes for the creation of a positively charged exciton is represented by a cross section. The radius of this effective surface is estimated to be 145 Å. The screening effects, mainly phase-space filling effects in our case, are characterized by a critical hole density of 10^{11} cm^{-2}. The decrease of the summed oscillator strength results of a combined effect of the ratio $f_X^0/f_{X^+}^0$ and of screening: it would increase if there were no screening $(p_c\rightarrow\infty)$ because $f_X^0/f_{X^+}^0<1$.

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