# Trion and exciton dephasing measurements in modulation-doped quantum wells: A probe for trion and carrier localization

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To investigate the properties of a two-dimensional carrier gas at intermediate densities, we perform picosecond transient four-wave mixing experiments on trions (charged excitons) and neutral excitons in modulation *p*-doped $CdTe/Cd_{1-x-y}Mg_xZn_yTe$ quantum wells. The determination of trion and exciton dephasing rates reveals a localization of both trions and holes in potential fluctuations induced by the ionized remote acceptors. We demonstrate that trions can be efficiently used as a charged optical probe sensitive to *electrostatic* potential fluctuations which are imperceptible for neutral excitons. [S0163-1829(99)15731-X]

Modifications of optical spectra due to the introduction of carriers into a semiconductor quantum well (QW) recently have been of considerable interest, in particular as they provide information on the electronic properties in the vicinity of the metal-insulator transition, and on the role of disorder.[1] It has been known for a long time that excitons characterize the optical spectra of undoped semiconductors while band-to-band transitions are observed on strongly doped samples. It has been recognized more recently that spectra of moderately doped QW's feature also charged excitons, either positively or negatively according to the type of doping—the so-called trions.[1–5] Extensive investigations have been reported on the binding energy of trions,[2–5] on their polarization in a magnetic field,[2,3] and on their behavior in an electric field[6] and at different carrier densities.[1,3,4,7] Up to now, however, only very few papers have been devoted to the dynamics of trions, including transport and relaxation.[7–9]

Actually, there has been some controversy in the literature about the possible localization of trions in modulation doped QW's (MDQW), in which carrier gases are formed in the QW's by introducing doping layers in the barriers. It was argued that free carriers would screen the Coulomb interaction thus forbidding the formation of excitons and trions.[1,2] Hence the charge carriers should be localized by electrostatic potential fluctuations caused by the ionized remote donors or acceptors. Recent near-field photoluminescence experiments[10] directly evidence the localization of trions in n-doped GaAs QW's and conclude indirectly to the localization of the carriers themselves. Other authors[3,11] have reported the observation of free trions in high-quality MDQW's with relatively thick spacer layers, which decreases the fluctuation depth.

In this paper we demonstrate that the study of the coherence properties of trions and excitons by degenerate four-wave mixing (FWM) is a very efficient method to investigate a possible localization of both trions and carriers in a MDQW. The idea is as follows: First, localized quasiparticles have a strongly reduced scattering efficiency as compared to free ones. Second, this scattering plays an important role in the dephasing of the quasiparticles. Consequently, the determination of exciton and trion dephasing rates under different conditions (exciton, trion and carrier densities, temperature) allows us to test the possible localization of excitons, trions, and carriers. To show this, we apply FWM to high-quality *p*-type $CdTe/Cd_{1-x-y}Mg_xZn_yTe$ multiple MDQW's, in which positive trions have been identified by magneto-optical spectroscopy.[12] Thereby, we perform the first, to the best of our knowledge, direct measurements of dephasing dynamics of trions. The results obtained in this way for the considered sample directly point to a localization of both trions and charges (in our case: holes) whereas the neutral excitons are found to be delocalized. Thus, through a study of their dephasing dynamics, trions and excitons are used as an optical probe of disorder. In particular, unlike neutral excitons, charged excitons experience electrostatic potential fluctuations, as those induced by the remote dopants.

The sample we chose contains 5 CdTe QW's of 8-nm thickness enclosed between $Cd_{0.69}Mg_{0.23}Zn_{0.08}Te$ barriers. It was grown by molecular-beam epitaxy on a $Cd_{0.88}Zn_{0.12}Te$ substrate which is transparent at the QW optical gap. At a distance of 50 nm on both sides of each QW the barriers are doped with $7\times10^{11}$ $cm^{-2}$ nitrogen acceptors.[12] The holes tunnel from the acceptors into the QW's (inset of Fig. 1) creating a two-dimensional (2D) hole density of about $10^{11}$ $cm^{-2}$ per QW. *C-V* measurements confirm these doping characteristics.[12,13] For our FWM experiments, we used a sample with a hole density $n_h$ in the QW's such that the resonances of the heavy-hole 1*s* exciton ($X$) and the positive trion ($X^+$) have comparable transition probabilities. The low temperature (5 K) absorption spectra in Fig. 1 presents a doublet structure with the $X$ peak at 1628 meV and the $X^+$ at 2.7 meV below. This splitting represents the binding energy of the trion in such CdTe QW's. The high optical quality of the sample is demonstrated by the rather small linewidths (1.4 meV for $X$ and 1.6 meV for $X^+$).

 

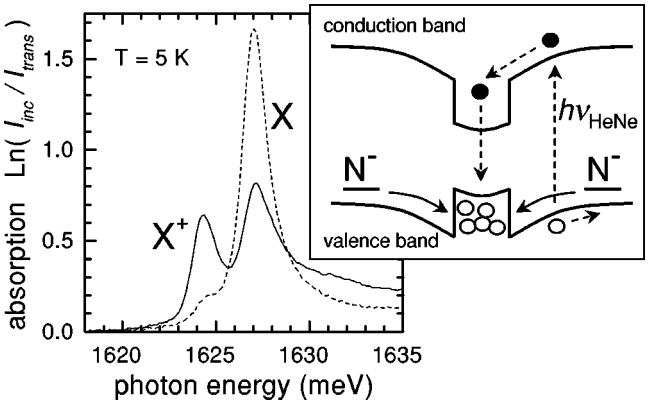


FIG. 1. Absorption spectra at 5 K showing the $X$ and $X^+$ peaks for two different hole densities, i.e., without (solid line: $n_h \approx 10^{11}$ cm$^{-2}$) and with (dashed line: $n_h \approx 10^{10}$ cm$^{-2}$) excitation of the sample by a cw He-Ne laser. Inset: Band structure of the QW's showing the hole transfer from the acceptors N to the QW's (solid arrows) and the effect of the cw HeNe laser excitation (dashed arrows, see text).

Our transient FWM experiments are performed in the usual two-beam self-diffraction configuration.[14] The FWM signal is measured as a function of the delay time between the two exciting pulses and detected by a photomultiplier and a lock-in amplifier. The laser pulses emitted by a tunable self-mode-locked Ti:Sapphire laser have a duration of 1.7 ps and a spectral width [full width at half maximum (FWHM)] of 0.7 meV, narrow enough to excite selectively the $X^+$ or the $X$ resonance and to prevent the excitation of continuum states. Unless explicitly mentioned, the sample is kept in a helium bath cryostat at 5 K.

Figures 2(a) and 2(b) display the FWM signal as a function of the delay time with the laser centered on the $X$ and $X^+$ resonances, respectively, for different excitation intensities. At equivalent $X^+$ and $X$ densities we observe that the exciton dephases much faster than the trion.[15] Assuming the trions to move freely in the QW's, their charge would force them to strongly scatter with each other and with the likewise charged holes. This scattering would be by far more efficient than the exciton-exciton ($X$-$X$) or exciton-hole ($X$-$h$) interaction. Consequently, free trions should dephase much faster than excitons at equivalent densities. The fact that we observe the contrary suggests that the $X^+$ FWM signal originates from localized trions.

As both resonances are inhomogeneously broadened, the dephasing rates are obtained from the decay times $T_D$ by $\Gamma = \hbar/(2T_D)$.[14] Since the exciton decay times at high densities are comparable to the pulse duration we determine the $X$ dephasing rates by fitting the results of a numerical integration of the usual optical Bloch equations[14] to the experimental transients [solid curves in Fig. 2(a)].

The dephasing rates $\Gamma_X$ and $\Gamma_{X^+}$ at 5 K are plotted in Fig. 3 as a function of the exciton and the trion densities $n_X$ and $n_{X^+}$, respectively. In both cases we find a linear increase of $\Gamma$ with increasing densities, fitted by

$$\Gamma_X(n_X) = \Gamma_X(n_X = 0) + \beta_{XX} n_X, \tag{1}$$

$$\Gamma_{X^+}(n_{X^+}) = \Gamma_{X^+}(n_{X^+} = 0) + \beta_{X^+X^+} n_{X^+}, \tag{2}$$

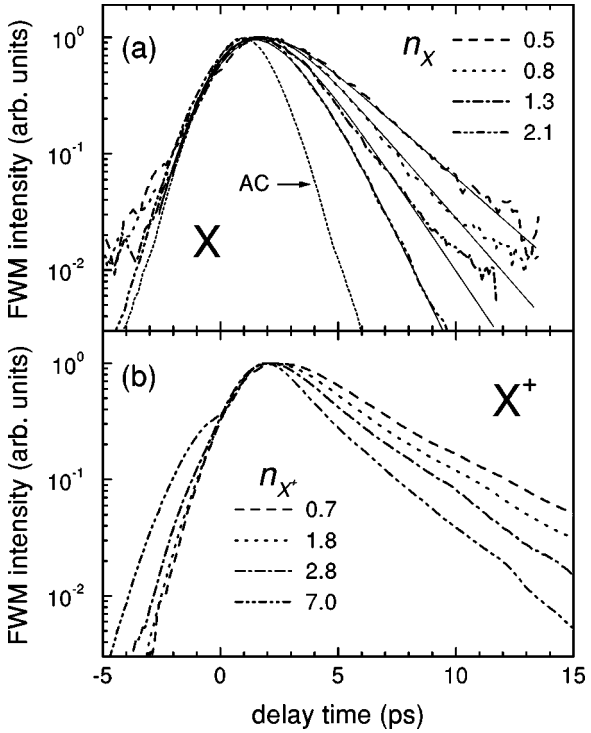


FIG. 2. FWM signals for a spectrally narrow picosecond excitation centered on the exciton (a) and trion (b) resonances measured at different densities $n_X$ and $n_{X^+}$ (as indicated in $10^{10}$ cm$^{-2}$). The autocorrelation (AC) of the picosecond pulses is also depicted in (a). The solid lines correspond to the numerically calculated response of an inhomogeneously broadened two-level system.

with the parameters $\beta_{XX}$ and $\beta_{X^+X^+}$ describing the exciton-exciton and trion-trion scattering, respectively. The quasiparticle densities $n_X$ and $n_{X^+}$ created by the two subsequent laser pulses are estimated from the measured spot size and the absorption coefficients of the two resonances.

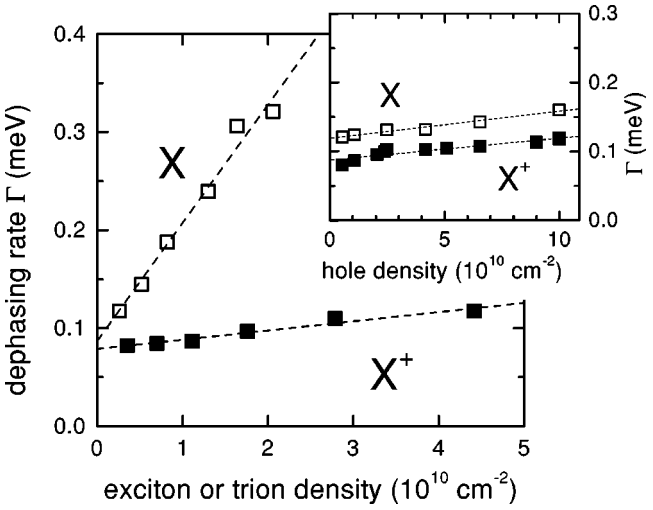


FIG. 3. Dephasing rates for excitons (open squares) and trions (filled squares) at 5 K as a function of the $X$ and $X^+$ densities. The inset shows the variation of the dephasing rates for excitons and trions as a function of the hole density $n_h$ in the QW's. In this latter measurement the constant picosecond excitation intensity creates for maximum $n_h$ an $X$ density of $0.6\times10^9$ cm$^{-2}$ and an $X^+$ density of $4.4\times10^{10}$ cm$^{-2}$.

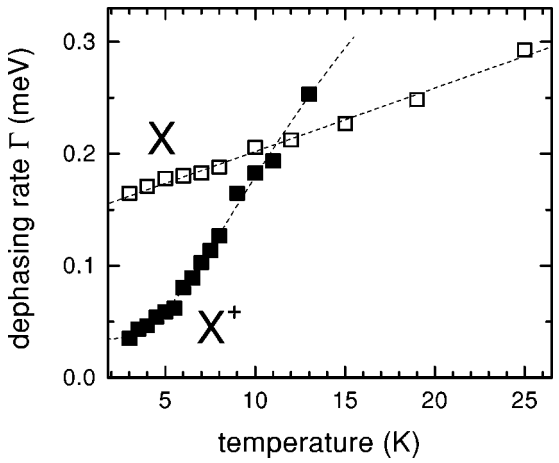


FIG. 4. Dephasing rates for excitons (open squares) and trions (filled squares) as a function of temperature.

We obtain $\beta_{XX}=(1.2\pm0.2)\times10^{-11}$ meV cm$^2$. Using the exciton binding energy $E_B=20$ meV (Ref. 16) and a Bohr radius $a_B=6$ nm we define, as usual, a dimensionless parameter $\gamma_{XX}=\beta_{XX}/(a_B^2E_B)=1.7$. This value is only slightly smaller than $\gamma_{XX}=2.5$ observed for the scattering of free excitons in an undoped 6-nm-thick $CdTe/Cd_xZn_{1-x}Te$ QW.[17] Moreover, values reported for $CdTe/Cd_xMn_{1-x}Te$ QW's, in which excitons are subject to efficient localization, are one order of magnitude smaller than our $\gamma_{XX}$.[18] We infer, that the excitons in our sample are delocalized. On the other hand, the trion-trion interaction parameter $\beta_{X^+X^+}=(0.9\pm0.3)\times10^{-12}$ meV cm$^2$ is one order of magnitude smaller than $\beta_{XX}$. This again disproves the conception of free trions strongly interacting through Coulomb forces and confirms the localization of trions.

In order to investigate the interaction of excitons and trions with phonons we measure the temperature dependence of the $X$ and $X^+$ dephasing rates (Fig. 4). In these measurements constant laser intensities photocreate $X$ and $X^+$ densities of $n_X=7\times10^9$ cm$^{-2}$ and $n_{X^+}=2\times10^9$ cm$^{-2}$. In contrast to the exciton, the trion excitation lies in the low-density limit so that the dephasing rates contain no contribution due to $X^+-X^+$ scattering. As expected for free excitons at low temperature, the $X$ dephasing rate depends linearly on the lattice temperature $T$. This is typical for the scattering of free excitons by acoustic phonons.[19] In the considered temperature range we can neglect the dephasing contribution due to longitudinal-optical phonons. Fitting the relation

$$\Gamma_X(n_X,T)=\Gamma_X(n_X,0)+\beta_{AX}T \tag{3}$$

to the experimental data we find the acoustic-phonon scattering parameter $\beta_{AX}=(5.7\pm0.4)$ $\mu$eV/K and the low temperature limit of the dephasing rate $\Gamma_X(n_X,0)=(0.15\pm0.01)$ meV. Our $\beta_{AX}$ is in very good agreement with the value of 5 $\mu$eV/K found in undoped $CdTe/Cd_xMg_{1-x}Te$ multiple QW's (Ref. 20) and close to values obtained in $GaAs/Al_xGa_{1-x}As$ QW's of comparable widths.[22]

In contrast to the exciton case, the temperature dependence of the $X^+$ dephasing rate is nonlinear and presents the characteristic shape of a thermal activation law

$$\Gamma_{X^+}(n_{X^+},T)=\Gamma_{X^+}(n_{X^+},0)+A\exp(-E_A/k_BT). \tag{4}$$

$E_A$ is the activation energy and $A$ measures the strength of this contribution to $X^+$ dephasing. Similar behaviors have been observed for the temperature-dependent dephasing of bound and localized excitons.[20,21] The fit depicted as a dashed curve in Fig. 4 yields $E_A=(1.5\pm0.2)$ meV and $A=(0.8\pm0.2)$ meV. Since $E_A$ is only about one-half of the $X^+$ binding energy $E_B=2.7$ meV, the observed activation process cannot consist in the dissociation of a trion leaving a localized hole and a free exciton. On the contrary, the absorption of thermal energy from the lattice leads to a delocalization of a trion as a whole. $E_A$ thus corresponds to the localization energy of the trions and constitutes a good estimation of the depth of the electrostatic potential fluctuations. As $E_A$ is only slightly smaller than the $X^+$ linewidth we can exclude the existence of a mobility edge within the inhomogeneously broadened $X^+$ line[20] above which the trions are delocalized.

By extrapolation, we determine the low-density low-temperature limit of the $X$ and $X^+$ dephasing rates. They include the residual contributions to the dephasing due to scattering with holes, interface roughness, impurities, and alloy fluctuations in the barriers. We obtain $\Gamma_X$ $(n_X=0, T=0)=(0.06\pm0.02)$ meV in good agreement with the values obtained for the undoped $CdTe/Cd_xZn_{1-x}Te$ QW's of Ref. 17 and with those reported for $GaAs/Al_xGa_{1-x}As$ QW's (Ref. 22) free of any carrier gas. Moreover, our $\Gamma_X$ are much smaller than dephasing rates found on excitons immersed in free 2D carrier gases of comparable density.[23,24] Consequently, the dephasing induced by collisions between excitons and holes cannot be predominant compared to the exciton-exciton or exciton-phonon scattering.[25] This indicates that the holes themselves are localized. The trions can thus be considered as excitons bound to localized holes. For the trions we find $\Gamma_{X^+}$ $(n_{X^+}=0,\ T=0)=(0.03\pm0.02)$ meV. As expected, it is smaller than the exciton value since the scattering of localized trions by holes, phonons, impurities, interface roughness, and alloy fluctuations is less efficient than that of free excitons.

To get further insight into the contribution of the hole gas to the scattering of excitons and trions we determine their dephasing rates for different hole densities $n_h$. To reduce $n_h$ we photocreate electron-hole pairs in the doped barriers by exciting the sample above the optical gap of the barriers with a cw He-Ne laser. The electrons are attracted into the QW's where they recombine with the hole gas. Finally, the system is found in a steady state with decreased $n_h$ resulting in an enhanced $X$ oscillator strength and a weakened $X^+$ transition (Fig. 1). We roughly estimate the reduced $n_h$ by assuming that the $X^+$ oscillator strength per hole is approximately constant and thus that $n_h$ is proportional to the integrated $X^+$ absorption coefficient.[26]

Honold *et al.*[23] state that the dephasing of excitons by collisions with free electron-hole pairs is 10 times more efficient than the dephasing due to $X$-$X$ scattering. Koch *et al.*[24] found that the scattering of excitons by a gas of free electrons is two times stronger than by other excitons. In contrast, we observe that $\Gamma_X$ and $\Gamma_{X^+}$ only decrease by about 0.04 meV when $n_h$ is reduced by one order of magnitude (inset of Fig. 3). Comparing this result to the main part of Fig. 3 we infer that in our case the scattering of excitons and trions with holes is by far less efficient than $X$-$X$ scattering.

This represents a direct confirmation of the localization of the holes themselves. It is worthwhile to note that in this measurement at constant picosecond excitation intensity the variation of $\Gamma_X$ and $\Gamma_{X^+}$ is not only caused by the reduction of the hole density but also by an increase of $n_X$ and a decrease of $n_{X^+}$ due to the change of the absorption spectrum. This contribution, however, is small compared to the total variation of $\Gamma_X$ and $\Gamma_{X^+}$ and thus does not alter our interpretation.

In summary, by a FWM study of the dephasing of trions and excitons in a modulation *p*-doped $CdTe/Cd_{1-x-y}Mg_xZn_yTe$ QW sample, we have evidenced that this method is a very powerful tool to investigate the localization of the charged quasiparticles (excess holes and trions) in the fluctuations of the electrostatic potential induced by the ionized remote acceptors. In this manner, we have demonstrated that trions can be used as a charged optical probe for electrostatic disorder.

The hole localization is deduced from the low-temperature $X$ and $X^+$ dephasing rates. Despite the presence of a hole concentration of up to $10^{11}$ $cm^{-2}$, we observe $X$ dephasing rates comparable to values reported for free excitons in undoped QW's and smaller than rates measured for excitons in free carrier gases. The dephasing of the charged trions, which should suffer an even stronger scattering by free holes, is slower than that of uncharged excitons. In addition, both $X$ and $X^+$ dephasing rates are only slightly affected by changes of the hole density.

Consequently, while the excitons are delocalized, the trions must be considered as excitons bound to localized holes. Confirmation of these hypothesis is provided by experiments performed for different photocreated $X^+$ and $X$ densities and for different temperatures. We obtain $X$-$X$ and $X$ acoustic-phonon collision rates in good agreement with measurements for free excitons in similar undoped QW structures. The $X^+$-$X^+$ collision rate is one order of magnitude smaller than the $X$-$X$ one, showing that trion localization prevents efficient $X^+$-$X^+$ scattering. The temperature dependence of the $X^+$ dephasing rate shows a thermal detrapping of localized trions with an activation energy of 1.5 meV.

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