Damping of Rabi oscillations in intensity-dependent photon echoes from exciton complexes in a CdTe/(Cd,Mg)Te single quantum well

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We study Rabi oscillations detected in the coherent optical response from various exciton complexes in a 20 nm-thick CdTe/(Cd,Mg)Te quantum well using time-resolved photon echoes. In order to evaluate the role of exciton localization and inhomogeneous broadening we use selective excitation with spectrally narrow ps-pulses. We demonstrate that the transient profile of the photon echo from the localized trion (X−) and the donor-bound exciton (D0X) transitions strongly depends on the strength of the first pulse. It acquires a non-Gaussian shape and experiences significant advancement for pulse areas larger than π due to non-negligible inhomogeneity-induced dephasing of the oscillators during the optical excitation. Next, we observe that an increase of the area of either the first (excitation) or the second (rephasing) pulse leads to a significant damping of the photon echo signal, which is strongest for the neutral excitons and less pronounced for the donor-bound exciton complex (D0X). The measurements are analyzed using a theoretical model based on the optical Bloch equations which accounts for the inhomogeneity of optical transitions in order to reproduce the complex shape of the photon echo transients. In addition, the spreading of Rabi frequencies within the ensemble due to the spatial variation of the intensity of the focused Gaussian beams and excitation-induced dephasing are required to explain the fading and damping of Rabi oscillations. By analyzing the results of the simulation for the X− and the D0X complexes we are able to establish a correlation between the degree of localization and the transition dipole moments determined as μ(X−)=73 D and μ(D0X)=58 D.

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Introduction. Coherent control of excitonic states in semiconductor nanostructures under resonant excitation with intense optical pulses attracts a lot of attention in relation with possible applications in quantum information [1]. These ideas exploit coherent rotations of the Bloch vector in the photoexcited two-level system (TLS), which depends on the area of the exciting pulse via Rabi oscillations [2,3]. Since stronger localization of excitons is in favor of longer decoherence times, most of the studies of coherent control have concentrated on quantum dots (QD) [3,4,5,6]. However, the strong localization in QDs is accompanied by large variations in QD size, shape, and composition which consequently leads to the large inhomogeneous broadening of the optical transitions when an ensemble of emitters is used. Therefore, most Rabi oscillation studies were performed on single QDs [3,4,5,6].

In semiconductor quantum well (QW) structures the inhomogeneous broadening of the optical transitions is significantly smaller as compared to QD systems, i.e. it is possible to selectively address different exciton complexes, such as free and localized excitons, localized charged excitons (trions, X−), and donor-bound excitons (D0X). Therefore, QW structures can be considered as a model system for the investigation of Rabi oscillations and their damping for optical excitations with different degree of localization and inhomogeneity. In spite of this fact, so far only few studies on Rabi oscillations have been performed in QW structures [7,8,9]. This is mainly due to the presence of many-body interactions, which can, however, be suppressed by using spectrally-narrow optical pulses [10,11,12]. Another important issue is related with the detection of Rabi oscillations. One of the approaches is based on reading out of the excited state population [13,14,15], while another exploits direct measurements of the coherent optical response [6,12]. Due to the inhomogeneous broadening of optical transitions in the TLS ensemble, the coherent optical response in a four-wave-mixing (FWM) experiment is represented by photon echoes [16]. In contrast to single pulse excitation, in FWM the amplitude and the transient profile of the detected signal depend sensitively on the optical field amplitude Ai (pulse area Θi) of both exciting (i = 1) as well as rephasing (i = 2) pulses which, as is demonstrated below, provides valuable information on the relevant physical effects.

In this work, we study the coherent optical response for intense resonant excitation of various exciton complexes with different degrees of localization and inhomogeneity in a single CdTe/(Cd,Mg)Te QW structure. In our low temperature measurements we observe photon
The sample immersed in liquid helium and cooled down to the temperature of 1.8 K was excited by a sequence of ps-pulses which destroy the coherence and diminish the photon echoes 

$P_{det}(t_{\text{Ref}}) \sim \int_{-\infty}^{+\infty} P_{FWM}^*(\tau_{\text{Ref}} - t') \cdot \exp(-t'/2\tau_P^2)dt'$

Here, $2\sqrt{\ln 2}\tau_P = 2.2$ ps is the laser pulse duration in the intensity scale.

To study the coherent optical dynamics of the localized systems we chose a structure with a high quality 20-nm thick single CdTe QW grown by molecular beam epitaxy. The structure was grown on (100)-oriented GaAs substrate followed by a 4.5 $\mu$m-thick Cd$_{0.76}$Mg$_{0.24}$Te buffer and 5 short-period superlattices separated by 100 nm Cd-MgTe spacers. This is followed by the QW sandwiched between the 100 nm CdMgTe barriers. The structure was not intentionally doped with donors. The resident electron density due to an unavoidable background of impurities is estimated to be $n_e \sim 10^{10}$ cm$^{-2}$. Recent studies performed on this sample have demonstrated the presence of two very closely located optical transitions corresponding to the negatively charged trion ($X^-$) and the neutral donor-bound exciton ($D^0X$) at 1.5980 eV and 1.5972 eV, respectively, which are clearly seen in the photoluminescence (PL) spectrum measured at a temperature of 2 K, see Fig. 2(a). Additionally, it displays the neutral exciton line ($X$) located at 1.6005 eV. In order to increase the density of resident electrons, which are required for the excitation of the trion, an additional weak above-barrier-illumination from a spectrally broad white light source was applied.

Experimental results. Figure 2(b) displays a typical FWM transient measured at $\tau_{12} = 40$ ps, in which a single PE peak at $2\tau_{12} = 80$ ps is seen. To obtain the optical coherence time $T_2^0$, the delay $\tau_{12}$ was varied and the PE amplitude was detected at $\tau_{\text{Ref}} = 2\tau_{12}$ in the regime of weak optical excitation. This procedure was performed for a number of spectral positions, and Fig. 2(c) displays three PE decays measured...
responding homogeneous linewidth of $\hbar$ which provides the following values of the coherence time: $\hbar$ respectively:

*a very short free excitons located at energies above 1.6005 eV reveal trions have long coherence times up to 100 ps, while the exciton and the trion show an increase of $T_2^0$ with decreasing excitation energy. The degree of the exciton and the trion localization on the potential and composition fluctuations increases with the decrease of the energy. Thus, longer $T_2^0$ times are expected for lower energy excitons and trions [13]. For $\text{D}^0\text{X}$, the transition energy is known to be affected by other factors, in particular, the interaction between neighboring donors [15]. Thus, the spectral dependence of $T_2^0$ for $\text{D}^0\text{X}$ can be more complex and is clearly different from that of the localized exciton and trion.

In order to observe Rabi oscillations, PE transients similar to that shown in Fig. 2(b) were recorded as a function of the amplitude of one of the two exciting pulses, $A_i (i = 1, 2)$, which we define as the square root of the energy per pulse, while that of the other pulse was kept constant. Thus, we vary the exciting pulse areas $\Theta_i (i = 1, 2)$ and keep the pulse duration $\tau_P$ fixed. Figure 3 summarizes the experimental results obtained at the three spectral positions indicated above.

The excitation of the localized exciton at $E(\text{X}_L)=1.5990$ eV results in a PE positioned at $2\tau_{12} \approx 53$ ps, as shown in Fig. 3(b). Here, the first pulse amplitude $A_1$ was scanned using a constant $A_2 = 3.5 \text{ pJ}^{1/2}$. The PE amplitude becomes damped at $A_1 \approx 7 \text{ pJ}^{1/2}$ and no oscillatory behavior can be observed. This behavior is to be expected since the nonlinear response of free and weakly localized excitons is very strongly affected by many-body interactions that lead to rapid dephasing and thus prevent the appearance of oscillatory intensity-dependent echo signals [20–22].

The same type of measurement performed on the trion at $E(\text{X}_L)=1.5980$ eV and shown on the upper panel of Fig. 3(c) displays a drastically different result. When the first pulse amplitude $A_1$ is varied at $A_2 = 3.5 \text{ pJ}^{1/2}$, the PE transients reveal a complex two-dimensional picture with the split PE profile similar to that observed recently in (In,Ga)As quantum dots [17]. The first maximum of the Rabi oscillations for $A_1 = 3.6 \text{ pJ}^{1/2}$ is centered at $2\tau_{12} \approx 53$ ps and well described by a single pulse of Gaussian shape, which is expected for a Hahn echo [23]. However, the second maximum at $A_1 = 13 \text{ pJ}^{1/2}$ is significantly advanced. The tail of the third maximum of Rabi oscillations at $A_1 > 20 \text{ pJ}^{1/2}$ appears to be non-shifted. When the amplitude of the second pulse $A_2$ is varied at $A_1 = 3.5 \text{ pJ}^{1/2}$, the PE maximum has no shift, as can be seen from the upper panel of Fig. 3(d).

Here, however, only the first maximum of Rabi oscillations at $A_2 = 7.5 \text{ pJ}^{1/2}$ is pronounced, while the second one around $A_2 = 20 \text{ pJ}^{1/2}$ is very weak. This is better visible in Fig. 3(a), where the cross-sections of the two-
FIG. 3: (Color) Rabi oscillations measured on the various localized exciton complexes in the studied CdTe/(Cd,Mg)Te QW. (a) Dependence of PE amplitude measured on the trion (E=1.5980 eV) detected at τ_{Ref} = 2τ_{12} = 53.3 ps on both pulse amplitudes. Measurements of PE transients as a function of pulse amplitude: localized exciton at E(\text{X}_L)=1.5990 eV, A_1 is varied at A_2 = 3.5 pJ^{1/2} (b); localized trion at E(\text{X}^-)=1.5980 eV, A_1 is varied at A_2 = 3.5 pJ^{1/2} (c), A_2 is varied at A_1 = 3.5 pJ^{1/2} (d); donor-bound exciton at E(D^0\text{X})=1.5972 eV, A_1 is varied at A_2 = 3.4 pJ^{1/2} (e). Upper and lower panels of (c)-(e) are the experimental data (upper row) and the numerical simulations (lower row), respectively. The scales in panels (b)-(e) are normalized to unity.

dimensional plots of Rabi oscillations according to the variations of both pulse amplitudes are plotted.

Figure 3(e) displays the measurement of Rabi oscillations performed on the donor-bound exciton at E(D^0\text{X})=1.5972 eV. Here, when A_1 is varied at A_2 = 3.4 pJ^{1/2}, the PE transients appear somewhat broader in time as compared to those measured on the trion. This means that the optically-excited inhomogeneous ensemble of D^0\text{X} is narrower than that for the trion. Also, the transition dipole moment of D^0\text{X} is smaller than that of \text{X}^-, since the Rabi frequency is smaller.

Theoretical model and discussion. The experimental measurements clearly manifest a rich intensity-dependent coherent behavior of the donor-bound exciton and the trion, which can be analyzed by modeling them as inhomogeneous ensembles of two-level systems interacting with the optical field. This approach is, however, unlikely to be applicable to the localized excitons X whose dynamics is more strongly influenced by complex many-body correlations [20–22] that may not be described properly by the modeling used here. In the following we develop an adapted theoretical model that is able to describe the observed intensity-dependence of PEs measured on D^0\text{X} and \text{X}^- and study the mechanisms responsible for the observed damping of the Rabi oscillations.
To simulate the photon echo transients, we numerically solve sets of the optical Bloch equations \cite{22,24}, i.e., the coupled equations of motion for the microscopic polarization \( p \) and the occupation of the excited state \( n \), considering excitation parameters corresponding to the experimental situations. Since an adequate explanation of the experimental results requires the incorporation of a number of effects, which are described in more detail below, the numerically-solved system of equations contains two indices \( f \) and \( s \):

\[
\frac{\partial}{\partial t} p_{f,s}(t) = \left( -i \omega_f - \gamma(t) \right) p_{f,s}(t) + i \frac{\mu}{\hbar} E_s(t) \left( 1 - 2n_{f,s}(t) \right), \tag{2}
\]

\[
\frac{\partial}{\partial t} n_{f,s}(t) = -2 \frac{\mu}{\hbar} \text{Im} \left[ p_{f,s}(t)^* E_s(t) \right]. \tag{3}
\]

Here, \( \omega_f \) is the optical transition frequency of the TLS, \( \mu \) is the dipole matrix element which is taken to be constant for the ensemble, \( E_s \) the total electric field including both laser pulses, \( E(t) = E_1(t) + E_2(t) \), and \( \gamma(t) \) is the excitation and thus time-dependent dephasing rate. Even a qualitative explanation of the experimental findings requires to incorporate three effects into the theoretical analysis: (i) An inhomogeneous broadening of the resonance, i.e., a Gaussian distribution of the optical frequency within the TLS ensemble which is described by the \( f \); (ii) the spatial profile of the incident laser pulses which is described as a Gaussian leading to the additional index \( s \); and (iii) excitation-induced dephasing, which is caused by the amount of optical excitation and, therefore, leads to the time-dependent dephasing rate \( \gamma(t) \).

By solving the system of equations (2) and (3) we compute the total FWM polarization describing photon echoes as

\[
P_{FWM}(t) = \sum_{f,s} \alpha_f \beta_s p_{f,s}(t), \tag{4}
\]

where \( \alpha_f \) and \( \beta_s \) are weight coefficients described further below. To compare with the experimentally-detected signal we convolute Eq. (4) with the Gaussian shaped reference pulse, as described by Eq. (1).

When setting up and solving Eqs. (2)-(3), we take a number of experimental requirements into account: (i) In order to obtain a photon echo, an inhomogeneous distribution of the optical transition energies is necessary. We define the spectral density of oscillators by a Gaussian distribution \( \alpha_f \propto \exp\left[-(\omega_f - \omega_0)^2/2\sigma^2\right] \) centered at \( \omega_0 \), which is coincident with the optical field frequency. Here, we adjust the width \( \sigma \), such that the temporal profile of the echo calculated at both exciting pulse areas \( \Theta_i = \int \frac{dt}{T} E_i(t) dt = \pi/2, (i = 1, 2) \) \cite{22}, and the temporal profile of the PE measured at \( A_1 \approx A_2 \approx 3.5 \text{ pJ}^{1/2} \) have equal temporal widths.

(ii) The spread of Rabi frequencies within the ensemble. A statistical distribution of the dipole moments in the ensemble can result in a variation of the pulse area. However, in contrast to strongly inhomogeneous QD ensembles, this effect is unlikely to take place in a QW structure \cite{22,26}. Here, we, however, have to consider the spatial variation of the pulse within the excitation spot which is modeled by a Gaussian profile \( E_s \sim \exp(-r^2/2\sigma_R^2) \) in real space \( r \) is the distance from the laser spot center), with \( \sigma_R \) characterizing the size of the focused laser beam. This spatial excitation profile defines also a weight \( \beta_s \sim r \), which corresponds to the amount of oscillators, located in the sample area with the radius \( r \) and excited by the field amplitude \( E_s \). In other words, due to the nonlinear characteristics of the Bloch equations \cite{22-24}, various Rabi frequencies are superimposed and enter the total signal with according weight \( \beta_s \). This spatial integration has a profound effect on the \( \Theta_1 \) and \( \Theta_2 \) dependencies and leads to an additional decrease of the Rabi oscillation amplitude for the higher pulse areas. This is visualized in Fig. 4(a) and (b), where numerically simulated Rabi oscillations are shown for the cases, when all dephasing mechanisms are deactivated and only the spatial integration is considered, respectively. Additionally to the damping, the effective frequency of the Rabi oscillations is also affected. It should be noted that the result of the calculations does not depend on the value of the beam size \( \sigma_R \), when the spatial integration is performed for a large enough area \( (\text{max}(r) \gtrsim 3\sigma_R) \).

(iii) The dephasing rate \( \gamma(t) = \gamma(E(t)) \), which depends on the state of the optical excitation of the system and thus on the exciting electric fields, is a crucial factor in modeling the different experimental situations. It is known that Rabi oscillations can be damped depending on the excitation strengths of the driving fields and quite a few different mechanisms have been identified. The damping might be due to phonons, population leakage into the delocalized states, Auger capture or the transfer of the excitation to other states \cite{12,27,29}. For the trion and donor-bound exciton heating of the electronic ensemble in the ground state is also one of the relevant mechanisms \cite{30}. Nevertheless, it has been shown that the concrete mechanism is less important than the non-Markovian behavior of a reservoir \cite{31,32}. Here, we assume that for our conditions the intensity-dependence of the dephasing rate is governed by excitation-induced dephasing (EID) \cite{20,21}. In our simulations we describe this dependence by

\[
\gamma(t) = \frac{1}{T_2^1} + a \int_{-\infty}^t dt' E^2(t'). \tag{5}
\]

The main idea is that the laser field off-resonantly excites populations that via scattering effects lead to a damping of the polarization. The effect of EID can be seen in Fig. 4(c), where Rabi oscillations are simulated with activated EID without integrating over the spatial coordinate.
By adjusting the model parameters for the effects (i)-(iii), using \( T_2^0 \) obtained from the PE decay measurements with weak pulses as well as the proper laser pulse characteristics, one can model and understand the measured echo signals shown in Fig. 3(c)-(e). Results of the Rabi oscillation simulations are shown in this figure in the lower panels under the according experimental plots. In these simulations, the adjusted spectral widths of the X\( ^- \) and D\(^0\)X ensembles are \( 2\sqrt{2}\ln 2\sigma = 1.0 \pm 0.1 \) meV and 0.5\( \pm 0.1 \) meV, respectively. This ratio of linewidths seems reasonable, since the potential for donor-bound exciton is less affected by the fluctuations of composition and QW width. The adjusted EID parameter \( a \) is different in the two simulations, so that \( a(X^-)/a(D^0X)=2.5 \) giving significantly different EID for the two considered exciton complexes. It can be seen from Fig. 4 where the calculated dephasing rate \( \gamma \) and the dephasing time \( T_2 = 1/\gamma \) are plotted as functions of the first pulse area \( \Theta_1 \) at \( \Theta_2 = 0 \). As a result, the Rabi oscillations for the case of donor-bound excitons are more robust. This qualitative conclusion correlates with the assumption that D\(^0\)X is stronger localized than the trion and, hence, many-body interactions leading to EID are weaker for this transition.

Finally, from the Rabi oscillation simulations we can obtain the transition dipole moments for both resonances. In order to calculate them we use the expression for the Rabi frequency, \( \Omega_R = \mu E/\hbar \), and take into account the laser spot size, the pulse duration, and the scale of the Rabi oscillations in units of the pulse amplitude known from the experimental data. As a result, we evaluate \( \mu(X^-) \approx 73 \) D and \( \mu(D^0X) \approx 58 \) D. This gives a ratio of \( \mu(X^-)/\mu(D^0X) = 1.26 \) which can also be deduced from the period of the Rabi oscillations in Fig. 4(c) and (e). We can compare this value with another estimation based on first-principles calculations, from which we know that \( \mu^2 \sim \tau_0 \), where \( \tau_0 \) is the radiative lifetime of the optical excitation. Values of the optical lifetime \( T_1 \) for X\(^- \) and D\(^0\)X were measured earlier in the same sample at low temperature: \( T_1(D^0X)=66 \) ps, \( T_1(X^-)=55 \) ps \cite{18}. Comparing these values with \( T_2^0(D^0X)=96 \) ps and \( T_2^0(X^-)=75 \) ps measured here we have the ratio \( 2T_1 \approx T_2^0 \) roughly fulfilled for both transitions for weak optical excitation. This means that the energy decay of both the trion and the donor-bound exciton is mainly radiative, i.e. \( \tau_0 \approx T_1 \). Thus, we can estimate \( \mu(X^-)/\mu(D^0X) \approx \sqrt{\Omega_1(D^0X)/\Omega_1(X^-)} = 1.10 \), which is in qualitative agreement with the previous estimation. We can therefore conclude that the transition dipole moment of the trion and the donor-bound exciton correlates with the degree of localization of these
particles.

**Conclusion.** We have observed and analyzed optical Rabi oscillations by measuring photon echoes from the trion and the donor-bound excitons in a CdTe/(Cd,Mg)Te QW structure. The photon echoes detected from the localized exciton states exhibit strong excitation-induced dephasing, which rapidly quenches the Rabi oscillations. Our experimental findings together with the proposed model provide a spectroscopic method by which the coherent evolution of various optical excitations can be studied in detail. By comparing the results of numerical simulations for the D^0X and the X^- complexes we establish a correlation between the degree of localization and the transition dipole moment of the exciton complexes: the stronger localized donor-bound exciton has a smaller µ than the trion (58 D and 73 D, correspondingly). It follows that the influence of EID on the coherent response of D^0X is significantly weaker as compared to the localized trion (hγ=9 µeV and 14 µeV, respectively, at Θ_1 = 3π/2, Θ_2 = π/2). The inhomogeneous broadening extracted from the simulations amounts to 0.5 meV and 1.0 meV for D^0X and X^-, respectively. The experiment and the simulations demonstrate that the Rabi oscillations are strongly smoothed due to the spatially inhomogeneous optical excitation spots. This averaging can be significantly reduced or overcome by using a spatial mask to define a more homogeneously excited sample area, which we consider as a prospect for future studies.

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