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Coupling of exciton transitions associated with different quantum-well islands

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We show by means of time-resolved four-wave-mixing experiments that the simultaneous excitation of energetically distinct and inhomogeneously broadened exciton transitions associated with spatially separated quantum-well islands leads to a beating photon echo. This shows that the exciton transitions cannot be described by uncoupled two-level systems. We discuss bleaching, screening, and dipole-dipole interaction as possible coupling mechanisms.

The coherence properties of exciton transitions in semiconductor quantum-well structures have recently attracted considerable interest.[1] Besides resonant light scattering,[2] transient four-wave mixing[3] (FWM) has been proven as a most powerful experimental tool. In these experiments a beating of the nonlinear signal often is observed due to the fact that, in particular with short (i.e., spectrally broad laser pulses), more than one excitonic transition is excited simultaneously.[2-7] However, since transient FWM experiments usually employ time integrated detection of the nonlinear signal, it has often been very difficult to extract information about the underlying multilevel system. For example, a three-level system as well as two uncoupled two-level systems lead to a beating behavior of the transient four-wave-mixing signal. However, in the case of two uncoupled two-level systems, the beating can be understood as an external interference phenomenon of two independently emitted FWM signals and is thus called polarization interference (PI) instead of quantum beating (QB).

The work by Göbel *et al.*[4] showed that the simultaneous excitation of exciton transitions associated with spatially separated islands of a quantum-well (QW) structure differing in thickness by one monolayer, leads to a beating behavior of the transient FWM signal. The authors attributed the observed periodic modulations to quantum beats. Subsequently, it was argued[5] that this oscillatory structure should not be called QB, since it originates from two spatially separated oscillators, i.e., from two uncoupled two-level systems. Thereafter, this "island" beating has been attributed to PI.[8,9] Recently, it has been shown that two uncoupled two-level systems indeed fully describe the temporal evolution of the FWM signal when excitons localized in different spatially separated quantum wells are simultaneously excited.[10] However, a definite determination in the case of excitons associated with different spatially separated islands within the plane of the same quantum well has not been possible so far.

In this paper we demonstrate that the previously observed periodic temporal modulation of the FWM signal from excitonic transitions associated with spatially separated islands[4] is in fact due to *real quantum beats* and cannot be explained by assuming two uncoupled two-level systems. For the experimental demonstration we employ time resolved detection of the FWM signal. As has been shown recently, this technique allows us to distinguish between a three-level system (3-LS) causing real QB's and two two-level systems (2×2-LS) causing PI.[10] We discuss the extension of this method for the case of considerable inhomogeneous broadening which governs the excitonic linewidth in the present sample. Finally, we discuss possible mechanisms which might cause the coupling of the different exciton transitions.

The experimental technique which allows the distinction between a 3-LS and 2×2-LS is based on the fact that the real-time behavior of the nonlinear FWM signal is distinctly different for the two cases.[10] The experimental setup is schematically illustrated in Fig. 1(a): Two successive laser pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are superimposed on the sample. If the temporal delay τ between the pulses is smaller than or on the order of the phase relaxation time T_2, this gives rise to a diffracted nonlinear signal (due to the third-order polarization) in a direction corresponding to $2\mathbf{k}_2 - \mathbf{k}_1$. Measuring the diffracted signal time integrated, i.e., by employing a slow detector, one might observe an oscillatory structure as a function of the delay time τ in the case that two (or more) optical transitions are coherently excited. By up-converting this diffracted signal in a nonlinear crystal using a third delayed reference beam (delay time t equal to "real time") one can study the real-time behavior of the third-order polarization [time resolved FWM (Refs. 11–14)]. Again, if two optical transitions are involved, this *real-time signal* exhibits an oscillatory structure as shown in Fig. 1(b). As can be derived from the optical Bloch equations the real-time positions of the interference maxima as a function of τ will be different for a 3-LS compared to a 2×2-LS.[10] In the case of a 3-LS maxima appear for $t = \tau + nT_B$ (T_B is the beat period, n is an integer), while in the case of a 2×2-LS the peak positions are given by $t = 2\tau \pm nT_B$. By plotting the positions of the maxima of the time resolved signal versus t and τ one can unambiguously distinguish between

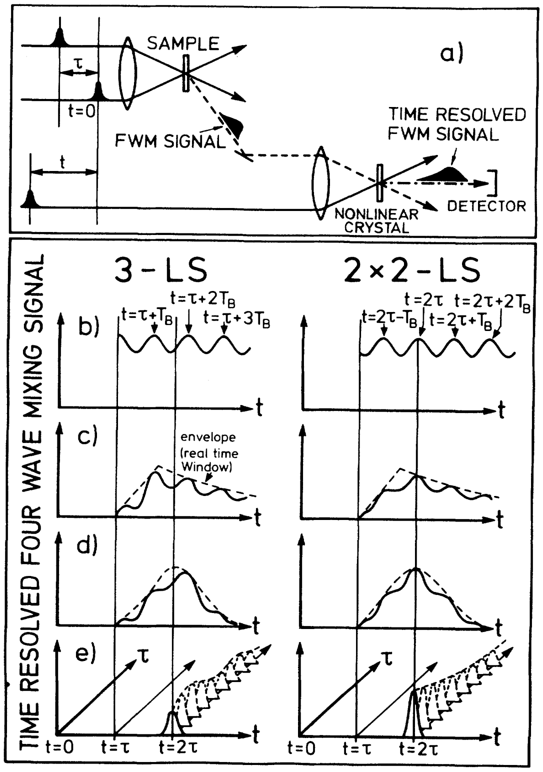

FIG. 1. Upper part: (a) Experimental setup for a time resolved FWM experiment. The diffracted FWM signal (dashed line) is time resolved via up-conversion in a nonlinear crystal. Lower part: Time resolved FWM signal (schematically) for the case of a 3-LS (left-hand side) and for a 2×2-LS (right-hand side). (b) Neglecting dephasing, many-particle Coulomb effects, and inhomogeneous broadening; (c) including dephasing, and many-particle Coulomb effects, neglecting inhomogeneous broadening; (d) including dephasing, many-particle Coulomb effects, and weak inhomogeneous broadening; (e) including dephasing, many-particle Coulomb effects, and strong inhomogeneous broadening.

a 3-LS (QB) and a 2×2-LS (PI).[10]

As depicted in Fig. 1(b), this beating is observable in real systems in a more or less broad "real-time window," which serves as an envelope superimposed on the beating signal. This envelope is determined by different effects like dephasing, inhomogeneous broadening, and many-particle Coulomb effects. While the dephasing is responsible for an exponential decay of the envelope, many-particle Coulomb effects manifest themselves in a gradual increase of the real-time signal,[14–18] which is considerably slower than the exciting laser pulses [Fig. 1(c)]. Finally, inhomogeneous broadening will cause a further narrowing of the time window [Fig. 1(d)], which in the case of strong inhomogeneous broadening will result in the emission of a sharp photon echo at $t = 2\tau$ [Fig. 1(e)]. If in the case of considerable inhomogeneous broadening the temporal width of the window is smaller than the beat period, no beating can be observed in the real-time (t) behavior of the diffracted signal for a fixed delay time τ. However, in this case the distinction between a 3-LS and a 2×2-LS becomes possible due to the characteristic behavior of the echo intensity as a function of τ. For a 2×2-LS the photon echo amplitude decays with increasing τ and exhibits no modulation since the maximum of the envelope is always coincident with a beat maximum at $t = 2\tau$ [Fig. 1(e)].[19] In contrast, in the case of a 3-LS the echo envelope at $t = 2\tau$ alternately coincides with a beat maximum or a beat minimum and any value in between when τ is varied. Therefore the echo intensity will exhibit a beating with the beat period T_B as a function of the delay time τ.[20]

The above arguments are quantitatively supported by a simple numerical calculation within the frame of the model developed by Yajima and Taira[21] based upon experimental parameters as applicable to the case discussed here (see below). The results of these calculations are shown in the left inset of Fig. 2, where we have plotted the echo amplitude as a function of the delay time τ for the case of considerable inhomogeneous broadening. It can be seen that the oscillatory structure totally vanishes for the 2×2-LS (dashed line), while there is still a pronounced beating for the 3-LS (solid line). Thus, in the case of inhomogeneous broadening a beating of the photon echo amplitude is a definite indication that two simultaneously excited excitonic resonances cannot be described by two uncoupled, i.e., independent two-level systems but rather by a level system that gives rise to quantum beats, an example of which is a 3-LS.[22]

The sample investigated is the same as the one studied in Refs. 3 and 4. It is grown by molecular-beam epitaxy and consists of a 70-Å GaAs QW followed by

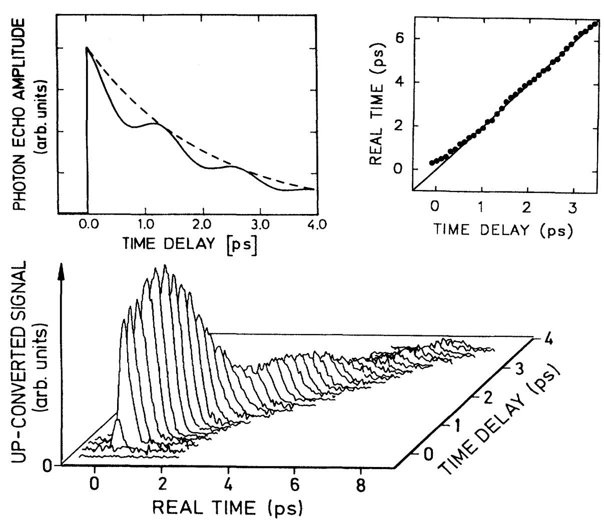

FIG. 2. Intensity of the up-converted FWM signal as a function of both the time delay τ between the pump laser pulses, and of the real time t. The right inset shows the (t, τ) positions of envelope maxima in the time resolved nonlinear signal (dots). The solid line corresponds to $t = 2\tau$. The left inset shows the calculated photon echo intensity as a function of time delay τ between the pump pulses for the case of a 3-LS and a 2×2-LS, respectively. The calculation is based on the experimental parameters for the sample studied here: $T_B = 1.33$ ps, $T_{\mathrm{echo(FWHM)}} = 0.8$ ps, and $T_2 = 4.2$ ps.

a 48-Å-thick $Al_{0.35}Ga_{0.65}As$ barrier and a second 45-Å-thick GaAs QW. Ten periods separated by 150-Å $Al_{0.35}Ga_{0.65}As$ barriers are grown with growth interruptions of 60 s at each interface. The photoluminescence excitation spectrum in Ref. 4 and the absorption spectrum in Ref. 3 show three distinct peaks with an energy splitting of 2.7 meV which could be attributed to excitonic recombination from spatially separated islands of the 70-Å QW with one-monolayer thickness difference. Due to the dependence of the exciton energies on well thickness this results in different transition energies for the exciton states, which can be resolved for sufficiently thin QW's provided the inhomogeneous broadening is not too strong. Performing a deconvolution of the inhomogeneous lines in Ref. 4 (the inhomogeneous broadening is about 2.6 meV) one obtains the correct energy splitting of $\Delta E = 3.1$ meV corresponding to one-monolayer thickness difference. The oscillatory structure observed in the previous transient FWM experiments had a time period of 1.33 ps. This corresponds exactly to $\Delta E = 3.1$ meV. Therefore the oscillatory structure could be clearly attributed to an interference between the optical transitions of the differently confined excitons.[4] The present experiments are performed at a sample temperature of 10 K. The laser source is a mode-locked Ti:sapphire laser emitting pulses of typically 110 fs duration and 22 meV [full width at half maximum (FWHM)] spectral width. The central laser photon energy is slightly detuned below the resonance of the lowest 1*s* exciton transition, to ensure that higher exciton states (2*s*, ...) are not excited at the same time. The excitation density is estimated to be about 9×10^9 cm^{-2}. The experimental setup is described in more detail in Ref. 22.

The experimental findings are shown in Fig. 2. This three-dimensional plot shows the time resolved diffracted FWM signal as a function of the real time t for different time delays τ between the exciting pulses. For each time delay τ, the time resolved signal exhibits an unmodulated, nearly symmetric shape with a temporal halfwidth of about 800 fs. This time resolved signal can be classified as a "broad" photon echo, as can be seen more clearly in the inset, where the position of the envelope maximum (dots) is plotted versus t and τ. The temporal position of the maximum follows the condition $t = 2\tau$ (solid line) as expected for a photon echo. From the temporal width of the echo we determine the inhomogeneous broadening[12] to approximately 3.3 meV. Thus, the temporal width of the nonlinear signal is smaller than the beat period. Consequently, the real-time signal is unmodulated and a beating can only be observed as a function of τ in the amplitude of the broad echo. This in fact can be seen very clearly in the three-dimensional plot shown in Fig. 2. The beat period is 1.33 ps and hence exactly the same as observed before in the time integrated FWM experiment.[4] As discussed above, the observed beating of the photon echo amplitude shows that the two excitonic transitions associated with spatially separated islands having different quantum-well thicknesses cannot be described by two uncoupled two-level systems. Accordingly, the observed signal beats do not arise from polarization interference but are due to quantum beats as claimed by Göbel *et al.*[4]

Further understanding of what is meant by "coupling" can be obtained by considering how it modifies the underlying level system. The left-hand side of Fig. 3 illustrates the uncoupled 2×2-LS in the one-particle picture, whereas the right-hand side of Fig. 3 shows the 2x2-LS in the two-particle picture. In the ground state $|g\rangle = |00\rangle$ of the crystal none of the excitons are excited. The states $|10\rangle$ and $|01\rangle$ indicate that one of the respective exciton transitions is excited, whereas $|11\rangle$ indicates that both excitons are excited. The calculation of the FWM signal using the right-hand level system leads to the same result (namely, PI behavior) as the calculation using the left-hand one-particle picture provided that transitions A and A' as well as B and B' are identical in terms of transition frequency, transition strength, and dephasing rate.[23] The excitation of one excitonic transition then does not affect the other excitonic transition, i.e., the two excitonic transitions are independent or *uncoupled.* In the opposite case, when excitation of one transition does affect the excitation of the other one, i.e., when A and A' or B and B' are not identical, the right-hand level system does not lead to polarization interference but to quantum beats. In this case, the two excitonic transitions are not independent and the beating behavior of the FWM signal can no longer be calculated in the simple one-particle 2×2-LS. In this sense, the excitonic transitions associated with different and spatially separated quantum-well islands are *coupled.*

There are several possible mechanisms which might lead to a coupling, i.e., to an inequality of the A and A' or the B and B' transitions. First of all, the excitation of one excitonic transition might lead to a bleaching of the other transition, since both exciton states might use the same one-particle k states. Second, screening of the Coulomb interaction might lead to a coupling of the two exciton transitions. The coupling may also be caused by Coulomb-exchange interaction. This can be seen analytically, when the Coulomb renormalized electric laser field is introduced into the optical Bloch equations.[24] In this case, three additional terms can be identified, which may cause a nonlocal coupling. Two of these terms are cubic in the amplitude of the laser field and describe many-particle Coulomb effects.[25,15,16] The third term, however, is linear in the laser field and describes dipole-dipole interaction.[26,27] Recent theoretical considerations indicate that dipole-dipole interaction between localized excitons indeed can cause quantum beats.[28] However,

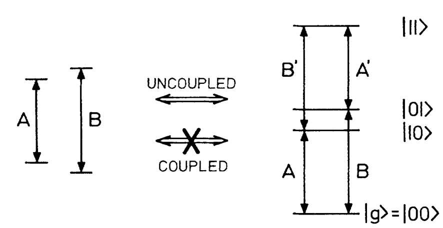

FIG. 3. Level scheme for a 2×2-LS in the one-particle (left-hand side) and the two-particle picture (right-hand side). The equivalency between both pictures only holds in the uncoupled case, where $A = A'$ and $B = B'$.

dipole-dipole interaction should not only couple excitonic transitions associated with different islands within the same well but also excitonic transitions stemming from spatially separated quantum wells. Since exciton transitions from spatially separated wells only generate PI, dipole-dipole interaction cannot be the main coupling mechanism in case of "island excitons." Further experimental and theoretical work is clearly needed to obtain a definite answer on the origin of the coupling mechanism.

In summary, we have shown that excitonic transitions associated with spatially separated in-plane islands of a QW having different thicknesses cannot be described by two uncoupled two-level systems (in contrast to excitonic transitions associated with spatially separated QW's). This is unambiguously inferred from time resolved transient FWM experiments, where we observe a beating in the photon-echo-like signal. We propose bleaching and screening as possible coupling mechanisms.

After completion of this work it came to our attention, that Matsusue *et al.*[29] have recently studied so-called island-inserted quantum wells by time integrated FWM experiments. They also observe a modulation of the FWM signal due to the simultaneous excitation of excitons localized in different islands, which they interpret as real quantum beats due to the strong modulation of the diffracted signal. Although this in fact is a strong criterion for existence of QB's, the final decision should be made by performing time resolved FWM, like in our case. Nevertheless these data may provide another example for the importance of nonlocal coupling mechanisms as discussed here.

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