Coherent effects in resonant quantum-well emission

N. Garro, M. J. Snelling, S. P. Kennedy, and R. T. Phillips
Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, United Kingdom

K. H. Ploog
Paul-Drude-Institut für Festkörperelecktronik, D-10117 Berlin, Germany
(Received 18 August 1998)

We demonstrate that resonant Rayleigh scattering can dominate the secondary emission of quantum wells for much longer times than previously reported. Coherent manipulation of the quantum state of the system allows cancellation of the beats preserving subpicosecond time resolution. The main features of the rise and decay of the emission are unmasked showing that the latter can be governed by excitonic dephasing. [S0163-1829(99)01631-8]

Semiconductor quantum wells (QW's) are extensively used in optoelectronic devices and the dynamics of excitons in these systems is an active field of study.[1] Recently, QW emission following resonant excitation by an ultrashort pulse has been attracting a great deal of interest. An ideal system would emit coherent radiation only in the reflection and transmission directions. The intensity of the signal would rise with the leading edge of the excitation pulse and decay as the excitonic coherence is lost, i.e., with a characteristic time $T_2/2$, where T_2 is the transverse relaxation time.[2] Simultaneously incoherent photoluminescence (PL)—a product of the radiative recombination of the exciton population—builds up in the whole solid angle. This is a long-lived signal that lasts for hundreds of picoseconds. In real QW systems, however, a strong coherent component—resonant Rayleigh scattering (RRS)—accompanies the incoherent PL. RRS therefore becomes central to a full understanding of QW emission.

To study RRS one needs to overcome two major difficulties: (a) ultrafast RRS is spectrally undistinguishable from PL; (b) RRS arises from complex static disorder, such as well width and alloy fluctuations, about which there is almost no information. One possible way of separating both components is by working at low exciton densities and low temperatures, where RRS dominates the emission at early times and shows the quadratic rise[3] predicted theoretically.[4] However, this feature is not always visible as the shape of the emission is most often obscured by beats.[5] A different approach was taken by Birkedal and Shah[6] who probed RRS using ultrafast spectral interferometry. This technique is sensitive to the spatial and spectral disorder of the system. Thus the experiment had to be performed on a single speckle of the emission.[7] Current models, based on ensemble averaging,[4] are not valid for these experimental conditions and complete understanding of these results is still absent. Also, the rather coarse spectral resolution limited the temporal range accessible with this technique to less than 10 ps. Problems caused by the spectral disorder of the sample are avoided when the interference is set to take place at the sample in a two-pulse excitation experiment.[8–10] Gurioli *et al.*[8] proved the presence of a coherent component in the emission employing time-integrated detection. More information can be extracted in a time-resolved experiment.[10] Very recently it has been demonstrated that the degree of coherence of the secondary emission can be deduced from a statistical analysis of the speckle pattern.[11] This is possible employing a streak camera that allows for a single speckle to be resolved but with poor time resolution.

In this Brief Report we explore the emission of QW's following resonant excitation by an ultrashort pulse, when both heavy-hole (hh) and light-hole (lh) excitons are photogenerated. The quantum state of the system is manipulated by arranging an interaction with two phase-locked pulses. This is monitored by the changes in the amplitude and phase of the hh-lh beats in the time-resolved emission collected over a wide solid angle. We demonstrate destruction of either excitonic species and cancellation of the beats while preserving sub-ps time resolution. The rise and the decay of the emission are then unmasked and their main features can be studied and compared with recent theoretical predictions.[12] RRS is found to dominate the emission for much longer times than predicted in past investigations.[3,6,10] Dephasing times of hh and lh excitons can be deduced from the decay of the emission. These values are in good agreement with the decay of the beat visibility and with the interferograms obtained in the two-pulse excitation experiment.

We have studied a sample containing 10×20-nm GaAs QW's separated by GaAs/AlAs superlattice barriers. Steady-state PL measurements gave narrow lines [full width at half maximum (FWHM) is 0.7 meV] at 4.5 K. The sample was excited in resonance with the hh and lh exciton transitions by a pair of collinear 130-fs pulses from a Ti:sapphire laser, with their relative phase locked by an actively stabilized Michelson interferometer. The excitation was spectrally centered 4 meV below the hh exciton transition to minimize free-carrier generation. The two pulses were linearly copolarized and entered the sample at 35° from the surface normal. The secondary emission was collected from a cone of 0.17 sr near the backscattering direction, avoiding the specular direction, and mixed in a BBO crystal with a 130-fs pulse at 1.5 μm generated in a parametric oscillator synchronously pumped by the same source. The sum-frequency signal was recorded by a charge-coupled device. All measurements were performed with the sample mounted on a coldfinger

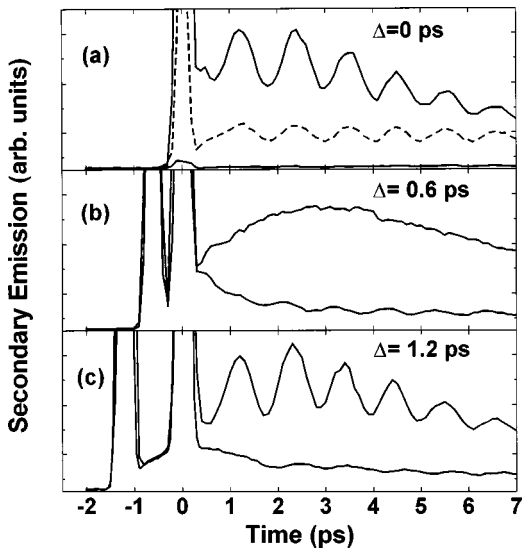

FIG. 1. Secondary emission after excitation by two phase-locked pulses. In (a) and (c) the second pulse coincides with successive peaks in the hh-lh quantum beats, and with the intermediate trough in (b). Upper curves are when the second pulse arrives in phase ($\tau = mT_{hh}$) lower when in antiphase [$\tau = (m+1/2)T_{hh}$] with the hh exciton. The dashed curve in (a) corresponds to the time-resolved emission after single-pulse excitation. The average laser power was 1 mW for each arm of the Michelson.

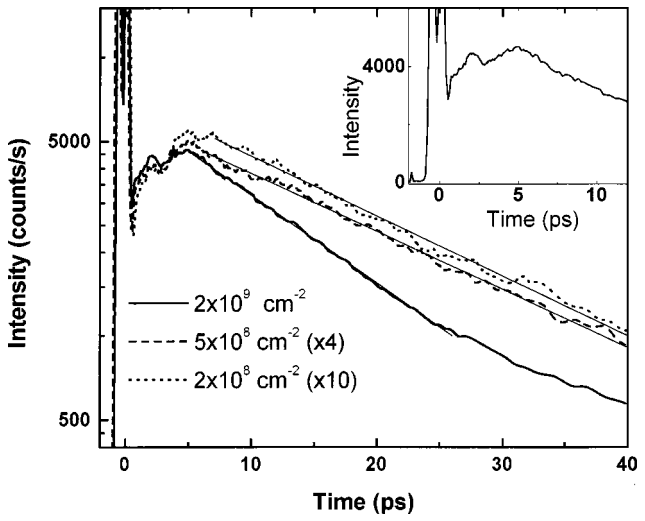

FIG. 2. Exponential decay of the hh exciton secondary emission for different exciton densities. The Michelson delay of the two excitation pulses was fixed at $\Delta = 0.6$ ps and $\tau = 0$. The inset shows the detail of the rise of the hh emission for an excitation density of 2×10^9 cm^{-2}.

cryostat at a temperature of 4.5 K. The excitation power varied from 1 to 0.1 mW for each arm of the Michelson focused onto a 100-μm-diam spot which, assuming a fractional absorption of 0.07 per well[13] and taking into account the transition and the excitation linewidth ratio, gives estimated ''exciton densities'' (n_X) varying between 5×10^8 and 5×10^7 cm^{-2}.

Figure 1 shows the time-resolved QW emission after resonant excitation by a pair of phase-locked pulses. The results of different temporal separations between the pulses τ_m are displayed in graphs 1(a)–1(c). We define $\tau_m = \Delta + \tau$ where Δ is a multiple of the hh-lh beat period T_{12} (Ref. 14) [$T_{12} = \hbar/(E_{lh} - E_{hh}) = 1.20$ ps] and τ is an additional number of periods of the hh exciton oscillation T_{hh} ($T_{hh} = \hbar/E_{hh} = 2.72$ fs). Figure 1(a), with $\Delta = 0$, corresponds to the case when the two pulses interfere directly. In Fig. 1(c), $\Delta = T_{12}$, the second pulse reaches the sample when the hh and lh exciton polarizations induced in the well by the first pulse are in phase. The electric field of the pulse interacts with both of them in the same manner. When τ_m coincides with a complete number of hh oscillations, $\tau = mT_{hh}$, the final polarization is enhanced over twice the single-pulse value (upper curve). In the destructive interference case (lower line), $\tau = (m+1/2)T_{hh}$, the emission after the second pulse is reduced as the polarizations are destroyed by it. The separation of the Michelson pulses can be set so that the second pulse coincides with a minimum in the hh-lh exciton beats, $\Delta = T_{12}/2$ [Fig. 1(b)]. In this case the hh and lh polarizations are in antiphase when the second pulse arrives. When $\tau = mT_{hh}$ the hh polarization experiences constructive interference while the lh excitons interfere destructively. The converse is true when $\tau = (m+1/2)T_{hh}$. In each case beating ceases as only hh or lh excitons remain after the interaction with the second pulse.

Now that we have demonstrated our ability to perform coherent control of the excitonic ensemble[9,15] let us benefit from it in order to study the emission process. We use this technique to stop the emission from beating in a physical way, without any added processing such as Fourier filtering[3] and, most importantly, without losing any time resolution. Figure 2 displays the time-resolved emission after excitation by a pair of pulses with a time separation identical to that in the upper curve of Fig. 1(b). The emission rises in the first few picoseconds, as it can be seen more clearly in the inset of Fig. 2. Notice the double-peak structure occurring in the first 5 ps. The presence of the second maximum depended on the position on the sample, see Fig. 1(b), which corresponds to a different region. Either situation can be understood with the recent theoretical predictions of Savona and Zimmermann for the rise of RRS.[12] They have found that, depending on the correlation in the disorder potential, the time-resolved RRS can show a single or a double peak in the initial rise. Their calculations were done performing ensemble averaging which is valid for our experimental conditions as we are collecting the emission over a wide solid angle. Thus regional differences in the fluctuating well width are likely to cause the change observed in the rise from different spots.

The decay in the intensity of the emission for different exciton amplitude (density) was investigated (see Fig. 2). The exciton densities were calculated assuming perfect constructive interference, i.e., densities four times the single-pulse densities, so they can be slightly overestimated. In all cases the measured decay in the emission could be well fitted by an exponential decay. For the higher n_X (solid line in Fig. 2) the initial regime truncates after 25 ps and it is followed by a slower decay, also exponential. This is not apparent at lower n_X (dotted and dashed curves) where the decay rate is slower and density independent. The rise of the emission, on the other hand, does not change considerably for different n_X: the position of the two peaks remained constant and only their relative intensities reversed for decreasing n_X. The fact that the emission decay slows down for lower n_X could

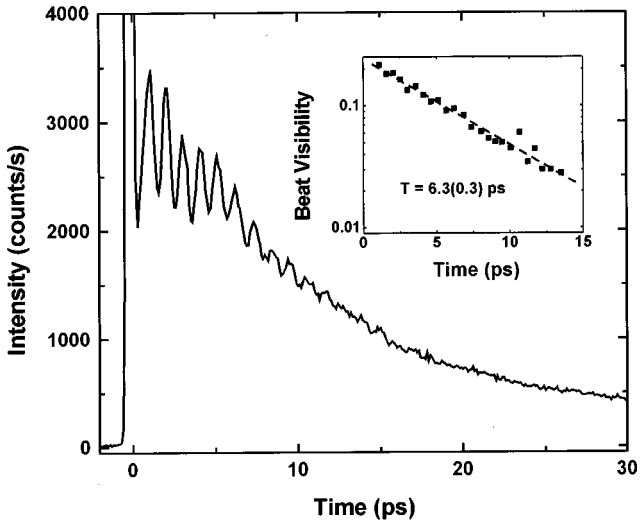

FIG. 3. Secondary emission after excitation by a single pulse. The injected exciton density is $5\times10^8\,\mathrm{cm}^{-2}$. The decay of the beat visibility is plotted in the inset of the figure for the same experimental conditions.

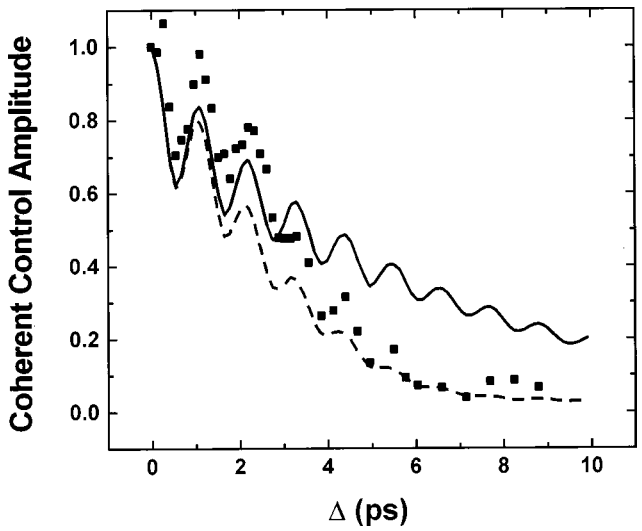

FIG. 4. The evolution of the coherent control amplitude is related to interpulse separation, with the grating time fixed at 2.2 ps after the second pulse. The solid line shows the calculation for homogeneously broadened excitons while the dashed line corresponds to the inhomogeneously broadened case. The ratio of the hh to lh density is 4 to 1.

be related to exciton dephasing and therefore the emission would be still dominated by RRS, even after 20 ps from excitation. This is once more in good agreement with the theoretical predictions in Ref. 12. We have then measured hh exciton dephasing times T_2^{hh} of 27.4(0.1) ps for $n_X=2\times10^9\,\mathrm{cm}^{-2}$ and 41(1) ps and 39(2) ps for $n_X=5\times10^8$ and $2\times10^8\,\mathrm{cm}^{-2}$, respectively. At high density (solid curve) we also observe the transition between coherent and incoherent (after 25 ps) emission. These long dephasing times are not surprising as RRS rises due to excitonic localization in the plane of the well. The measured T_2^{hh} at the highest n_X is comparable to the 24(1) ps obtained in photon-echo experiments for similar well width and exciton density conditions.[16] The advantage of our linear technique is that we can reach much lower exciton densities. Dephasing induced by exciton-exciton interaction reduces for lower densities and causes T_2^{hh} to increase. Below a certain density threshold exciton dephasing occurs only through interaction with phonons and crystal imperfections. We found that threshold at $5\times10^8\,\mathrm{cm}^{-2}$ in our sample.

Direct observation of the main features of QW emission is possible only after suppression of the beats. Figure 3 shows the secondary emission after single-pulse excitation. The spot on the sample was the same as in Fig. 2 and the power of the laser was increased to inject $n_X=2\times10^9\,\mathrm{cm}^{-2}$. Yet the behavior looks very different and it is hard to discern a double peak in the rise of the emission. We measured T_2^{lh} as we did for the hh excitons and found it to be considerably shorter than T_2^{hh} [$T_2^{\mathrm{lh}}=6.1(0.2)$ ps]. Consequently the combined emission from hh and lh excitons has a different shape from the emission of either of the species alone. The difference between T_2^{hh} and T_2^{lh} also explains the decay in the beat visibility, which is characterized by a time constant T given by $1/T\approx1/T_2^{\mathrm{lh}}-1/T_2^{\mathrm{hh}}$.[17] According to our measured dephasing times the beat visibility should decay with $T=7.8(0.3)$ ps, which is close to the measured value of $T=6.3(0.3)$ ps (inset in Fig. 3). Hence it is clear that hh-lh beats are a manifestation of the exciton or ''optical'' coherence.

Our observations of RRS decaying with $T_2/2$ are in open contradiction with previous interferometric investigations[6,10] where a decay mainly governed by destructive interference between different emitters within the inhomogeneous linewidth of the transition has been claimed. In an identical experiment as that to Ref. 10, we have investigated the evolution of the coherent control amplitude (the difference between emission for the constructive and destructive conditions) for increasing pulse delay Δ. Figure 4 shows the amplitude of the coherent control at a fixed gating time of 2.2 ps after the arrival of the second pulse (this corresponds to the same spot as the data from Fig. 1 and is different than Figs. 2 and 3). As discussed in the first part of this paper, in this kind of experiment the second pulse interferes with the polarization induced in the sample by the first pulse. The oscillations in the amplitude shown in Fig. 4 are due to the periodic change of the relative phase of hh and lh excitons. We have calculated the effect of increasing the pulse separation in a coherent control experiment with a model where excitons are treated as a three-level system and the laser pulses are approximated by δ pulses. Such a model succeeds in reproducing the main features of the coherent control.[18] Dephasing is included in the model through the measured exponential dephasing times. The result of this calculation is shown in Fig. 4 (solid line: homogeneous case; dashed line: with Gaussian inhomogeneous broadening). The calculations give good agreement with the experimental results only when inhomogeneous broadening is taken into account. After excitation, the different evolution of the components in the inhomogeneous transitions means that the second pulse interferes either constructively or destructively, depending on the energy of the states. This reduces the magnitude of the coherent control after a time inversely proportional to the inhomogeneous broadening. It is clear that the decay of the

coherent control amplitude in this, or other similar experiments,[9] cannot simply be interpreted in terms of T_2. If we monitor the interference amplitude at a gating time of 0 ps, i.e., at the arrival of the second pulse as in Ref. 10, although there may be an additional interference of the emitted light with the nonresonantly scattered light of the second pulse, we will still be sensitive to the interference with the excitonic polarization and therefore still limited by the inhomogeneous broadening of the transition.

In conclusion, we have presented a consistent picture of coherent emission from excitons in quantum wells which successfully explains the main features of the secondary emission in the first 50 ps after excitation. RRS rises due to static disorder and its decay is exponential and only mediated by exciton dephasing. We performed coherent control to manipulate hh-lh exciton beats, to isolate either species, and hence to measure the individual dephasing times even at exciton density conditions inaccesible for other techniques. The measured dephasing times can also explain the decay in the hh-lh exciton beats visibility. We also showed how other interferometric techniques are strongly limited by the inhomogeneous broadening.

This work has been supported by EPSRC. We are grateful to B. Deveaud, S. Haacke, G. Hayes, and V. Savona for many stimulating discussions. We also benefited from many hours of discussions with P. Eastham, A. Ivanov, and P. Littlewood.

[1] For a review see M. J. Kelly, *Low-Dimensional Semiconductors* (Clarendon, Oxford, 1995).
[2] R. Loudon, *The Quantum Theory of Light*, 2nd ed. (Clarendon, Oxford, 1983).
[3] S. Haacke *et al.*, Phys. Rev. Lett. **78**, 2228 (1997).
[4] R. Zimmermann, Nuovo Cimento D **17**, 1801 (1995).
[5] H. Wang *et al.*, Phys. Rev. Lett. **74**, 3065 (1995).
[6] D. Birkedal *et al.*, Phys. Rev. Lett. **81**, 2372 (1998).
[7] J. C. Dainty, *Laser Speckle and Related Phenomena* (Springer, New York, 1984).
[8] M. Gurioli *et al.*, Phys. Rev. Lett. **78**, 3205 (1997).
[9] X. Marie *et al.*, Phys. Rev. Lett. **79**, 3222 (1997).
[10] M. Woerner *et al.*, Phys. Rev. Lett. **81**, 4208 (1998).
[11] W. Langbein *et al.*, Phys. Rev. Lett. **82**, 1040 (1999).
[12] V. Savona *et al.*, Phys. Rev. B (to be published).
[13] Y. Masumoto *et al.*, Surf. Sci. **170**, 635 (1986).
[14] Δ is rounded to the nearest integer number of hh periods.
[15] A. Heberle *et al.*, Phys. Rev. Lett. **75**, 2598 (1995).
[16] A. Honold *et al.*, Phys. Rev. B **45**, 6010 (1992).
[17] The beat visibility is defined as $(I_{\max}-I_{\min})/(I_{\max}+I_{\min})$.
[18] N. Garro *et al.*, J. Phys. C (to be published).