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Coherent control of resonant quantum well emission

N Garro†, M J Snelling†, S P Kennedy†, R T Phillips† and K H Ploog‡

† Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, UK
‡ Paul-Drude-Institut für Festkörperelectronik, D-10117 Berlin, Germany

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Abstract. We have investigated the manipulation of the phase and amplitude of the state of a three-level exciton system by excitation with pairs of phase-locked optical pulses. We demonstrate coherent control over the phase of quantum beating between the light- and heavy-hole excitons which are the components of the system. The state of the system is monitored by time-resolved detection of the resonant secondary emission coupled out of the system in non-phase-matched directions by virtue of the static disorder. We show that the amplitude of the coherent manipulation must diminish as a function of time delay between the components of the pulse pair in a way which takes into account both the dephasing of the excitons and their unphasing which arises from inhomogeneous broadening.

1. Introduction

A short pulse propagating through a transparent medium induces a polarization in the material. After the driving field disappears, the polarization decays to zero in a time of the order of the period of the field. If the pulse frequency coincides with a real electronic transition in a semiconductor, such as an exciton, then it induces a coherent coupling of the ground and excited states of the crystal. This corresponds to a coherent polarization which stays in the medium long after the optical pulse has left, the phase of which may be stochastically scattered, for example, by exciton–exciton or exciton–phonon interactions. In the limit in which there is no memory in the reservoir of scattering excitations this leads to an exponential decay of the coherent terms with time, described by the optical transverse relaxation time T_2. This is typically several ps in the case of excitons in semiconductor quantum wells (QWs) at low temperatures. What happens if the optical field encounters a medium already bearing a coherent polarization? This fundamental problem has been addressed in several ways since the development of sub-ps lasers, such as by four-wave mixing or photon echo experiments [1]. These techniques generally provide information about the spectral and temporal variations of the amplitude of the polarization but not about its phase. The phase of the polarization is directly probed in an alternative technique based on excitation by a pair of phase-locked pulses. The coherent polarization induced in the medium by the first pulse can interfere with the electric field of the second pulse leading to spectacular changes in the optical response of the system. This technique is usually known as *coherent control*.

The use of phase-controlled pulse sequences was originally applied to manipulate the quantum state of atoms and molecules [2]. Planken *et al* [3, 4] introduced this technique to the study of THz emission from semiconductor QWs. The shape and magnitude of the THz transient generated by the second pulse depended on the phase difference between the exciting pulses in a two-pulse experiment. They identified the origin of these coherent effects in the

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interaction of the electric field of the second pulse with the coherence excited in the medium by the first pulse. Consistently with this explanation, the coherent control disappeared after phase-breaking events destroyed the coherence. Further development of the coherent control technique has been carried out by Baumberg and Heberle and co-workers [5–8]. The phase-dependent changes in the excitonic density and polarization induced by the second pulse were monitored in reflectivity and four-wave mixing measurements, respectively. The coherent control of the excitonic polarization is also measurable off the phase-matched direction [9]. The loss of coherent control with time has been interpreted in terms of dephasing of the excitonic ensemble and its decay fitted to obtain T_2 [9].

We pursue this form of coherent experiment in this paper and show that in general it is not possible to interpret the decay of the coherent control in terms of T_2. We have exerted coherent control of the excitonic polarization when heavy-hole (hh) and light-hole (lh) excitons are excited simultaneously, giving effectively a three-level system. The state of the system is monitored by time-resolved detection of the secondary emission which is emitted other than in the reflection direction. The secondary emission of a three-level system exhibits beats that disappear as the coherence of the excitonic ensemble is lost. We show that coherent control can be achieved in both excitonic polarizations independently as can be seen by recording changes in the amplitude and phase of the beats. It is possible to chose the separation in time of the two exciting pulses such that the hh exciton polarization is enhanced and the lh exciton polarization is destroyed, and vice versa. In this way it is possible to separate the emission from the two excitonic species. We also study for the first time the effects of the inhomogeneous broadening of the excitonic transitions and show how its magnitude also limits the magnitude of the coherent control with time.

The organization of this paper is as follows. A description of the experimental set-up and the samples studied is given in section 2. The manipulation of the hh/lh beating is demonstrated in section 3. Section 4 discusses the experimental results in terms of the solutions of the optical Bloch equations for a simple three-level system. The effects of the inhomogeneous broadening on the magnitude of the coherent control are discussed in section 5. Finally we summarize the main conclusions of this work.

2. Experiment

We have used a standard upconversion photoluminescence apparatus employing a tunable Ti:sapphire laser for excitation of the specimen, and gating the emission with synchronous pulses from a parametric oscillator (figure 1). The excitation beam consisted of two collinear

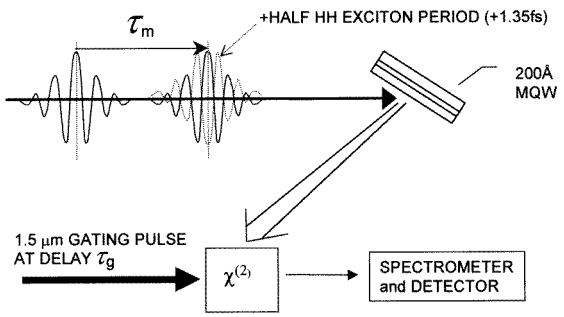

Figure 1. A schematic diagram of the experimental set-up.

pulses whose separation (τ_m) was locked to a fraction of a fs in an actively stabilized Michelson interferometer. The 130 fs pulses from the Ti:sapphire laser also pumped the parametric oscillator which generated 130 fs pulses at 1.5 μm wavelength, which were mixed with the emission in a 1 mm thick BBO crystal, at a time delay τ_g. This time delay was measured with respect to the second excitation pulse. The upconverted signal was recorded by a charge-coupled device. The excitation pulses were linearly copolarized and entered the sample at 35° from the surface normal. The secondary emission was collected from a wide cone (0.17 sr) far from the specular reflection.

We have studied a high-quality GaAs multiple-QW sample containing ten periods of 20 nm GaAs wells separated by GaAs/AlAs superlattice barriers. The sample was mounted on a cold finger at 4.5 K; at this temperature it showed a sharp PL line due to hh exciton recombination with 0.8 meV FWHM. For time-resolved experiments the excitation was spectrally centred 4 meV below the hh exciton transition to excite both hh and lh excitons simultaneously and minimize the generation of carriers in the continuum. The average power of the excitation was kept at 1 mW for each arm of the interferometer. This power over a spot of 100 μm of diameter, assuming a fractional absorption of 0.07 per well [10] and taking into account the ratio between the transition and the excitation linewidths, gives an estimated exciton density of 5×10^8 cm^{-2}.

3. Coherent control of beats in emission intensity

The time-resolved secondary emission after resonant excitation with a single pulse is shown in figure 2 (dashed curve). The initial spike at $\tau_g = 0$ is due to the laser scattering at the sample's surface. This is followed by a rise of the secondary exciton emission that peaks at

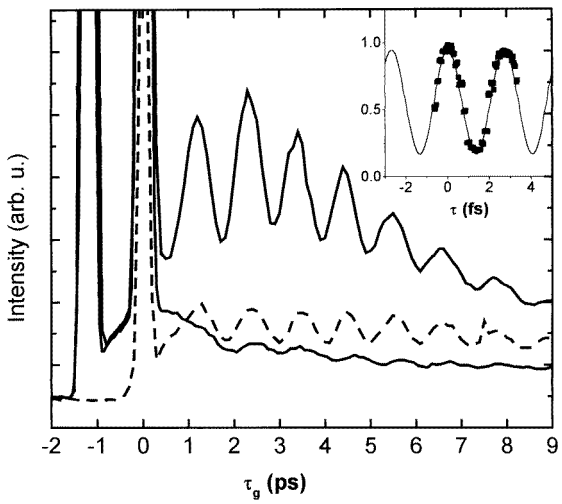

Figure 2. Time-resolved secondary emission following excitation by a pair of phase-locked pulses when the second pulse coincides with the first peak of the emission hh/lh beats ($\Delta = 1.2$ ps). The dashed curve corresponds to the emission after single-pulse excitation. The inset shows the intensity of the emission for a fixed gating time of 2.4 ps as a function of the fine-scale Michelson separation τ. Maximum emission occurs when the second pulse reaches the sample in phase with the hh exciton ($\tau = m\ T_{hh}$) while the troughs in the emission correspond to the second pulse arriving in anti-phase ($\tau = (m + 1/2)\ T_{hh}$).

$\tau_g = 3$ ps and then decays smoothly. At these early times (usually during the first 10 ps after excitation) the shape of the secondary emission is dominated by the hh/lh exciton beats. They are associated with the coherent nature of the early secondary emission which is mostly due to resonant Rayleigh scattering [11, 12]. The differentiation of resonant Rayleigh scattering from the incoherent luminescence in the secondary emission is an issue still being debated intensely [11–14]. We are not going to discuss the implications of the coherent control technique in the recognition of coherent emission in this paper, but will deal with that in a separate publication.

Let us now consider the excitation by a pair of collinear pulses that reach the sample with a time delay τ_m. For convenience, τ_m is defined as $\tau_m = \Delta + \tau$. Δ is a coarse parameter which is a multiple of the hh/lh exciton beat period T_{12} ($T_{12} = \hbar/(E_{lh} - E_{hh}) = 1.20$ ps, in the case of our sample) and it is rounded to the nearest integer number of hh exciton periods T_{hh} ($T_{hh} = \hbar/E_{hh} = 2.72$ fs). τ, on the other hand, is a fine-scale parameter defined in terms of T_{hh}. Figure 2 shows the time-resolved emission after resonant excitation by a pair of phase-locked pulses delayed by $\Delta = 1.2$ ps, i.e. the second pulse coincides with the first maximum of the hh/lh exciton beats. The upper curve represents the case where $\tau = m\ T_{hh}$, and shows a marked enhancement of the emission compared to the single-pulse experiment. The lower curve corresponds to $\tau = (m + 1/2)\ T_{hh}$ and shows a considerable extinction. For intermediate values of τ the emission evolves periodically in between these two extremes with a period of T_{hh}. This is shown in the inset of figure 2 where the emission at a fixed gating time $\tau_g = 2.4$ ps is plotted against τ.

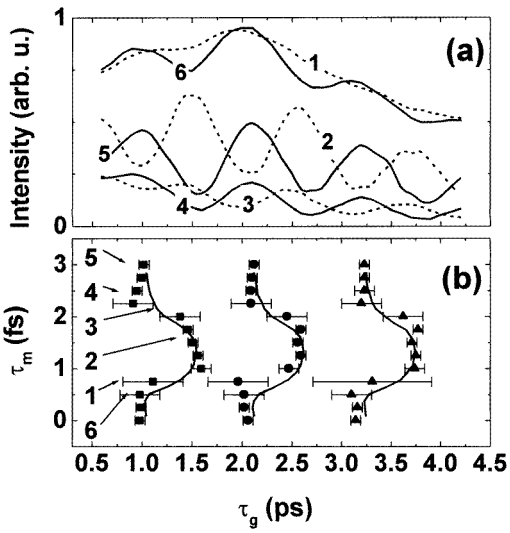

Figure 3. In (a) the amplitude and phase evolution of beats is shown for various τ within a hh cycle at $\Delta = 0.6$ ps. Curves in (b) show the positions of the first three beat peaks. Experimental results are indicated by squares and the solid curves are the result of the perturbative calculation.

The evolution of the emission at $\Delta = 0.6$ ps, when the second pulse arrives at a trough in the emission beats, is presented in figure 3(a) for different values of τ. The beats evolve in a periodic manner through one cycle of the hh exciton. For the first half of the cycle, from dotted curve 1 to 3, the intensity of the emission decreases while the amplitude of the beats increases to a maximum indicated by curve 2, then decreases again to near to its former value. In the second half of the period, from solid curve 4 to 6, the emission increases while the beat amplitude increases to a maximum value indicated by curve 5 then decreases again to

complete the cycle. Note the almost complete disappearance of the beats in curves 1 and 3. It is also noticeable that between the two halves of the period, which occurs between curves 3 to 4 and 6 to 1, the beats experience a phase shift of π. This shift can be seen more clearly in figure 3(b) where the positions of the first three peaks of the emission are plotted as functions of the Michelson fine delay τ.

4. Theoretical model

In order to interpret the experimental results we model the excitons as a three-level system, an approximation that has often been often found to retain the essential aspects of the physics [15]. The optical Bloch equations (OBE) for the case of excitation by two δ-pulses separated by a time τ_m, namely

$$E(t) = E_0\delta(t) + E_0 e^{i\omega\tau_m/h}\delta(t-\tau_m) \tag{1}$$

to first order in perturbation theory can be solved analytically. The linear polarization obtained in this way is

$$\begin{aligned} \boldsymbol{p} = -\frac{2e}{\hbar}\epsilon\Im\Big\{&\Theta(t)e^{iE_{hh}t/\hbar}[|\boldsymbol{r}_{hh}|^2 + |\boldsymbol{r}_{lh}|^2 e^{i(E_{lh}-E_{hh})t/\hbar}] \\ &+ \Theta(t-\tau_m)e^{iE_{hh}(t-\tau_m)/\hbar}[|\boldsymbol{r}_{hh}|^2 + |\boldsymbol{r}_{lh}|^2 e^{i(E_{lh}-E_{hh})(t-\tau_m)/\hbar}]\Big\} \end{aligned} \tag{2}$$

where Θ is the Heaviside function and $\boldsymbol{r}_{hh}$ and $\boldsymbol{r}_{lh}$ are the matrix elements for the interband transitions. To simplify the expression, dephasing effects have not been taken into account, but they could be included easily by introducing a phenomenological exponential decay for hh and lh excitons [15]. The terms in $\Theta(t)$ and $\Theta(t-\tau_m)$ come from the first and the second pulse, respectively. Each of these terms includes contributions from the hh exciton (the term in r_{hh}) and from the lh exciton (the term in r_{lh}). These beat together with a frequency determined by the difference in energy between the two transitions.

This simplified treatment gives very good qualitative agreement with our experimental results. We interpret the optical Bloch equations as giving a picture of the phase evolution of the beating system, which is visible in our detection of the secondary radiation as a result of the modification of the magnitude of the total polarization. See, for instance, that when $\Delta = n\,T_{12}$ as reported in figure 2, the second pulse finds the hh and the lh exciton polarizations oscillating in phase ($e^{i(E_{lh}-E_{hh})\tau_m/\hbar} \approx 1$ in equation (2)). In this case, the second pulse interferes with the two polarizations in the same way. Thus when the second pulse arrives in phase with the hh exciton polarization, $\tau = m\,T_{hh}$, it drives the total polarization to twice the initial value. In the same way, the polarization is destroyed when the pulse arrives in anti-phase with the hh exciton polarization, $\tau = (m+1/2)\,T_{hh}$. For a general τ the amplitude of the linear polarization is driven by the second pulse to $\boldsymbol{p}(1+e^{iE_m\tau/\hbar})$.

Let us concentrate now on the time-resolved emission shown in figure 3 where the second pulse arrives at a trough of the hh/lh exciton beating, $\Delta = (n+1/2)\,T_{12}$. At this Δ, hh exciton and lh exciton polarizations are oscillating in anti-phase when the second pulse reaches the sample. If τ_m coincides with a complete number of hh exciton cycles, i.e. $\tau = m\,T_{hh}$, the hh exciton polarization experiences constructive interference at the same time that the lh excitons interfere destructively. The complete opposite occurs when $\tau = (m+1/2)\,T_{hh}$. In both cases the beats are destroyed by the second pulse as only one of the polarizations remains in the sample. The intermediate curves of figure 3 correspond to intermediate values of τ. In these cases, and according to equation (2), the individual polarizations are rotated by $E_{hh}\tau/(2\hbar)$ and $(E_{hh}\tau/\hbar+\pi)/2$, respectively. Thus the phase of the hh/lh exciton beats is shifted by $\pi/2$ for τ

taking values in the first half of the hh exciton cycle and by $-\pi/2$ for values in the other half. These results are in good agreement with what was observed experimentally (see figure 3).

Other effects involving many-body processes as well as effects of band dispersion are beyond the scope of the independent three-level model. Semiconductor Bloch equations [16] offer a more realistic treatment of the problem. Nevertheless, for low excitation densities, where exciton–exciton interaction can be neglected, the OBE constitute a good first approximation, and give a good guide to the physical interpretation of the experimental results.

5. Decay of coherent control

So far we have shown how the coherent control technique is a powerful tool for manipulation of the emission from semiconductor QWs. Nevertheless it also provides extremely useful information about the coherence of the excitonic ensemble and its loss. In this sense Marie *et al* claim that the decay of the coherent control amplitude (the difference between the emissions at maximum constructive and destructive interference conditions) is determined by the optical dephasing time T_2 of the transition and therefore it can be used as an alternative technique to four-wave mixing for the determination of T_2 [9]. In the course of their argument they do not take into account that in real QW systems the imperfections in the well interfaces in the form of alloy and well-width fluctuations are responsible for the inhomogeneous broadening of the spectral content of the excitonic ensemble. Such fluctuations are in fact essential for the emission of light at short times in non-phase-matched directions. We have performed a calculation of the coherent control amplitude as a function of τ_m for an ensemble of three-level systems with different hh exciton energies (the hh/lh exciton splitting has been considered constant) and giving to each contribution a Gaussian weight. The results obtained are plotted in figure 4. Notice that the amplitude of the coherent control exhibits beats with the period of the hh/lh exciton beats. See as well that the amplitude decays with increasing Δ. No dephasing has been taken into account, so the decay of the coherent control has to be attributed to the inhomogeneous energy distribution. An intuitive explanation of this effect is the following: some time after the initial excitation, due to the different time evolution of the distinct energy components in the inhomogeneous transition, the second pulse can interfere constructively with

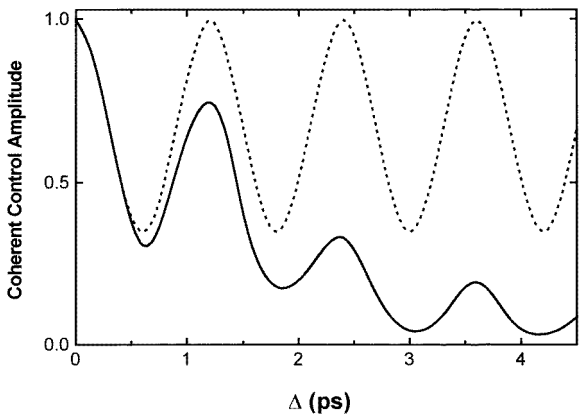

Figure 4. Calculated coherent control amplitude as a function of the pulse separation τ_m for an inhomogeneous excitonic ensemble (solid curve) in comparison to the homogeneously broadened case (dotted curve). The inhomogeneous distribution of energies has been modelled with a Gaussian of 0.8 meV width.

some components while interfering destructively with others. This reduces the magnitude of the coherent control after a time inversely proportional to the inhomogeneous broadening, as we see in figure 4. It is clear that the decay of the coherent control in this or other similar experiments [9] cannot simply be interpreted in terms of T_2.

6. Conclusions

We have presented a systematic study of the coherent control of an excitonic ensemble in which hh and lh excitons have been simultaneously excited. We showed how the coherent control technique allows the manipulation of the phase and amplitude of the beats of the secondary emission. Total cancellation of the beats can be achieved when either excitonic species is destroyed by the excitation pulse. We have developed a simple model based on the OBE which explains satisfactorily the coherent control of the polarization. Finally we have studied the effects of the inhomogeneous energy distribution of the excitonic ensemble and concluded that the coherent control is reduced faster than the homogeneous dephasing rates and is limited by the inhomogeneous broadening.

Acknowledgments

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