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Coherent Control Experiments as a Probe of the Carrier Scattering Processes

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Abstract

We report on coherent control experiments of excitons in GaAs/AlGaAs quantum wells. This technique, which relies on the linear response of the crystal, is used to investigate the spin-dependent exciton-exciton scattering.

The coherent control of a quantum system by light relies on the possibility to control both the amplitude and the phase of its photoexcited states. It consists in producing interferences between different excitation quantum paths, each one resulting from the interaction of the electromagnetic field with the system [1,2]. Among the different kinds of control investigated, the use of a sequence of two time delayed ultrashort optical pulses allows the creation of two temporally separated excitation paths. The coherence decay of optically excited electronic systems such as excitons in semiconductors provides one of the most powerful tools to investigate interaction processes of excited states [3]. From an applied physics point of view, the coherent manipulation of carriers is of great interest since it can remove one of the main obstacles to ultrafast optoelectronics and all optical-switching device performances which are degraded by long-lived carriers in the active region. We demonstrate in this paper that the coherent control of electronic excitations in semiconductors can be monitored through the secondary emission following the excitation, yielding a direct measurement of the optical dephasing time. In dense and polarized exciton systems, this technique evidences the spin dependent mutual interactions between excitons.

We present the results on a GaAs/AlGaAs Multiple Quantum Well (MQW) which consists of 30 periods of non-intentionally doped 10 nm GaAs wells and 20 nm $Al_{0.6}Ga_{0.4}As$ barriers grown by molecular beam epitaxy on a (100) substrate. The cw photoluminescence linewidth is 0.9 meV at 1.7 K and the shift between the heavy-hole exciton (XH) absorption and the luminescence peaks is about 0.1 meV denoting the high quality of the sample.

A sequence of two optical pulses of opposite helicities σ^+ and σ^-, split from a mode locked Ti: Sapphire laser beam (pulse width 1.5 ps) resonantly excites the heavy hole excitons at energy E_{XH}. This sequence is produced by a Mach-Zender type interferometer [Fig. 1(a)]. The temporal separation between the two pulses is controlled on two different time scales : a coarse tuning sets the delay t_1 between the two pulses on a picosecond scale; a fine tuning adds the delay t_2 on a femtosecond scale, allowing a very accurate control of the relative phase. The t_2 variation is achieved through the symmetrical rotation of two glass plates in opposite directions resulting in the variation of the optical path of the beam which travels across. The exact delay between the two pulses is then $t_1 + t_2$, and it is convenient to calibrate the time scale so that t_1 is an exact multiple of λ/c, where λ is the excitation wavelength. The Time Resolved Secondary Emission (TRSE) kinetics are recorded by up-converting the emission signal in a $LiIO_3$ non linear crystal with the output from an optical parametric oscillator (OPO) synchronously pumped by the same Ti:Sapphire laser which is used for the sample excitation. All the measurements were carried out at a temperature of 10 K and the photogenerated exciton density is varied from about 10^9 cm^{-2} to $3 \times 10^{10} cm^{-2}$. For a (100)-grown QW, the relevant symmetry is D_{2d}. The growth direction Oz is taken as the quantization axis for the angular momentum. The conduction band is s-like, with two spin states $s_z = \pm 1/2$. The upper valence band is split into a heavy-hole band with the total angular momentum projection $j_z = \pm 3/2$ and a light-hole band with $j_z = \pm 1/2$. The appropriate basis for the exciton states is then $\{|J_z\rangle \equiv| j_z + s_z\rangle\}$ i.e. $\{|+1\rangle, |-1>, |+2\rangle, |-2\rangle\}$. A circularly polarized light $\sigma^{\pm}$ creates excitons on states $|\pm 1\rangle$ (the "circular excitons" in the following), and a linearly σ^X or σ^Y polarized light, creates excitons on the coherent states $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$ and $|Y\rangle = (|1\rangle - |-1\rangle)/i\sqrt{2}$ respectively (the "linear excitons").

First, when the main delay between the two excitation pulses is $t_1 = 0$ and the intensities are strictly equal, their optical interference results in a linearly-polarized light excitation. The polarization direction in the QW plane depends on t2. Figure 1(b) shows the time dependence of the two linearly polarized luminescence components $I^X(t)$ and $I^Y(t)$ and the resulting linear polarization $P^l(t) = (I^X - I^Y)/(I^X + I^Y)$ for $t_2 = m\lambda/c$ (where m is an integer), i.e. when the interference of the two laser pulses results in a linearly σ^X-polarized optical excitation. The recorded linear polarization, initially almost equal to 1, decays with a characteristic time $T_{s2} \approx 20$ ps, the so-called "transverse spin-relaxation time" [4].This transverse spin relaxation time is the decay time of the quantum spin coherence. It is generally longer than "the optical dephasing time" T_2 of excitons [5]. Figure 1(c) displays the linear polarization dependence versus the fine temporal separation between the two pulses $P^l(t_2)$. The oscillations of the luminescence polarization observed in Figure 1(c) at the period $T = h/E_{XH}$ merely reflect the rotation of the excitation light polarization in the QW plane driven by t_2 which results in the photogeneration of linear excitons in states: $\left|\psi^l(t_2)\right\rangle = \cos(\omega t_2/2)\left|X\right\rangle + \sin(\omega t_2/2)\left|Y\right\rangle$ As a matter of fact, the linear polarization of the exciton luminescence is $P^l = \cos \omega t_2$ where $\omega = E_{XH}/\hbar$.

Now, the delay between the two excitation pulses is set to 6.6 ps, so that there is no temporal overlap between the two pulses. Figure 2(a) presents the secondary emission dynamics. The excitation with the second laser pulse results in a sharp rise of the linear

polarization of the excitonic luminescence which then decays with the characteristic time T_{s2}. Obviously, this linear polarization originates from the interaction of the second pulse with the coherent excitonic polarization created in the crystal by the first pulse. The linear polarization (measured again 4 ps after the second pulse) is displayed as a function of t_2 in Fig. 2(b). The clear oscillations which are observed again are interpreted as follows.

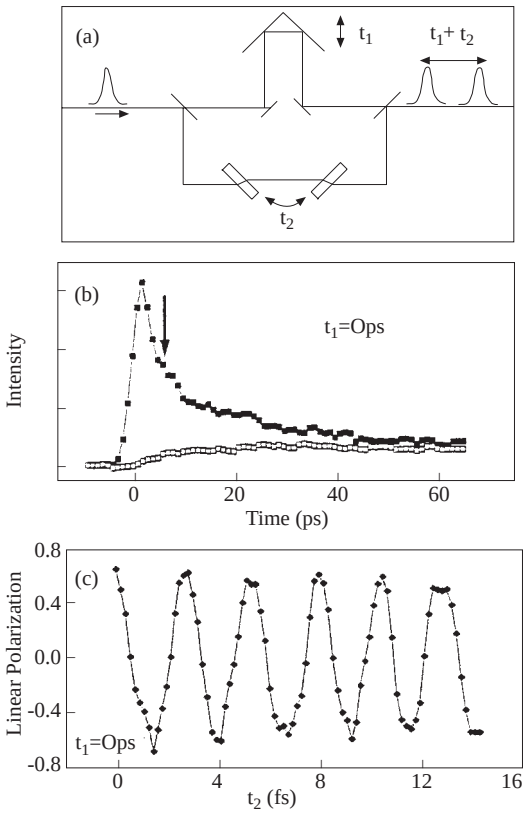

Figure 1. (a) Schematic excitation arrangement. In (b) and (c) the sequence configuration is (σ^+, σ^-) and $t_1 = 0$. (b) The time evolution of I^X(■) and I^Y (I^Y(□) for $t_2 = m\lambda/c$. (c) Linear polarization P^l, measured 4 ps after the excitation [arrow in (b)], as a function of the fine temporal separation t_2 between the two excitation pulses.

The first optical pulse (σ^+-polarized) sets up a material polarization in the crystal, built with $|+1\rangle$ excitons, which is coherent with the laser electromagnetic field. The interference of the second optical pulse (σ^--polarized) with this material polarization at time t_2 results in a coherent polarization of linear excitons on $\left|\psi^l(t_2)\right\rangle$ states. The oscillation as a function of time t_2 of the linear polarization of the luminescence reflects the rotation of the orientation of these linear excitons in the QW plane. The emission amplitude arising

from these excitons $|\psi^l(t_2)\rangle$ is a decreasing function of the delay t_1 between the two pulses, which reflects the decay of the coherent polarization of the matter. As a consequence, the amplitude of the oscillations of the linear polarization of the luminescence observed in Fig. 2(b), proportional to the fraction

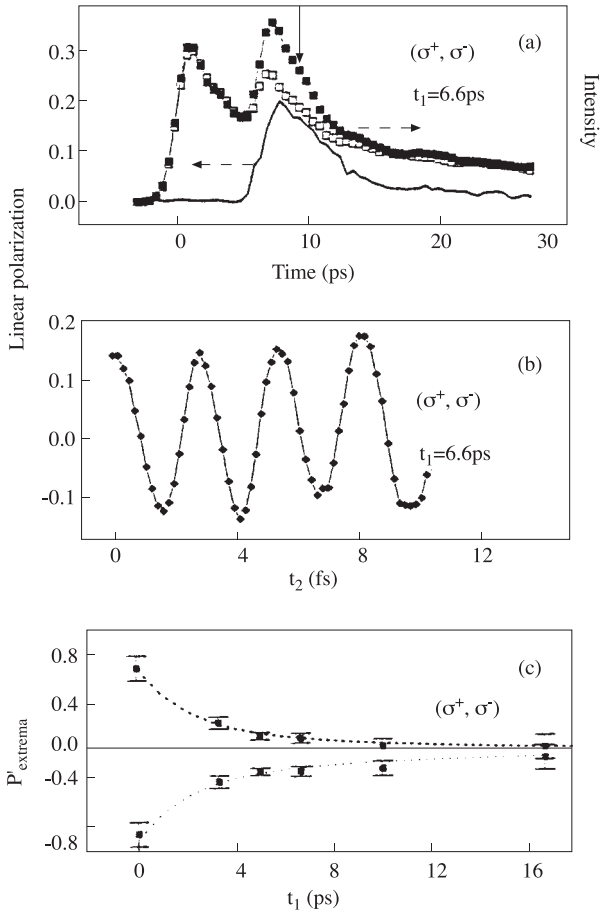

Figure 2. The configuration is (σ^+, σ) and $t_1 = 6.6$ ps. (a) The time evolution of I^X(■), I^Y(□) and the linear polarization P^l (full line) for $t_2 = m\lambda/c$ (the back-scattered laser light from the sample surface is negligible). (b) The linear polarization P^l measured 4 ps after the second excitation pulse [vertical arrow in (a)], as a function of the fine temporal separation t_2 between the two excitation pulses. (c) The maxima and minima of the linear polarization oscillations as a function of t_1 (the dotted line is a guide for the eye).

of excitons promoted on states $|\psi^l(t_2)\rangle$, is directly proportional also to the fraction of the excitons created by the first pulse which still oscillate in phase with their photogenerating optical field at time t_1. Figure 2(c) displays the minima and maxima of the linear

polarization oscillation as a function of t_1. Thus the amplitude decay of these oscillations follows the decay of the coherent exciton population created by the first pulse; it directly reflects the optical dephasing of excitons in the time interval $[0, t_1]$ even in the presence of inhomogeneous broadening [6]: the decay time is the so-called "optical dephasing time" (T_2). We measure $T_2 = 6 \pm 1$ps. The result is in agreement with previous measurements by the Four Wave Mixing technique [3].

Mutual interactions in dense exciton gas was first investigated by Four-Wave Mixing experiments using linearly polarized pulses [7,8]. In both references, the homogeneous exciton broadening was found to increase with the exciton density N, with a law which can be linearized in a moderate density range ($N \leq 2 \times 10^{10} cm^{-2}$) according to: $\Gamma(N) = \Gamma_0 + \gamma_{XX} N$, where Γ_0 is the non-density dependent homogeneous broadening which includes all dephasing mechanisms except the mutual exciton scattering, and γ_{XX} is the collision broadening parameter resulting from exciton-exciton scattering. This linear behaviour was predicted in Ref. [9], but a discrepancy remains since the theoretical value for γ_{XX} is much lower than the experimentally determined one. In addition, we have shown previously on the basis of photoluminescence experiments [10], that the mutual exciton interactions were strongly dependent on their polarization state, due to the effect of exchange interaction between their constitutive elements. We investigate here, as an application of our coherent control technique, the influence of the exciton density on the optical dephasing time $T_2 = 2\hbar/\Gamma$, taking into account the spin state of the excitons. We recall that using a (σ^+, σ^-) sequence we measure the dephasing time of a circular exciton population, whereas with a (σ^X, σ^Y) sequence we determine the dephasing time of a linear exciton population. At low photocreated exciton densities ($N \leq 10^9$ cm^{-2}), we find the same value for T_2 for both sequences ($T_2 = 6 \pm 1$ ps), i.e. for the two types of exciton populations. However, when the total photogenerated density increases, the measured phase coherence decay time decreases much faster in the (σ^X, σ^Y) configuration than in the (σ^+,σ^-) one. Figure 3 displays the density dependence of $1/T_2$ for circular or linear exciton populations. It demonstrates the spin dependence of the exciton mutual interactions. The fit with the experimental data for linear excitons yields $\gamma_{XX} = 0.15$ meV$\times 10^{-10}$ cm^{-2} and $\Gamma_0 = 0.2$ meV. The γ_{XX} value is similar to the collision broadening parameters measured in [7,8]. In contrast to the case of linear excitons, we see in Figure 3 that the optical dephasing time of circular excitons depends very slightly on the exciton density in the range $10^9 - 3 \times 10^{10}$ cm^{-2}. This behaviour difference between the circular and linear excitons is interpreted as a manifestation of the spin dependent mutual exciton interactions [10]. The stability of the pure circularly polarized exciton phase, with respect to these interactions, results in an optical dephasing time quasi-independent of the density. This behaviour difference can explain the discrepancy mentionned previously by Honold et al. [7] between the broadening parameter measured in FWM experiments (with linearly polarized pulses) and the calculated value in a many body approach by Manzke et al. [9]. The experimental γ_{XX} value for linear excitons is about four times larger than the theoretical one. The latter however agrees well with the broadening parameter we measure for circular excitons : the full line in Figure 3 corresponds to the calculated broadening parameter by Manzke et al. [9].

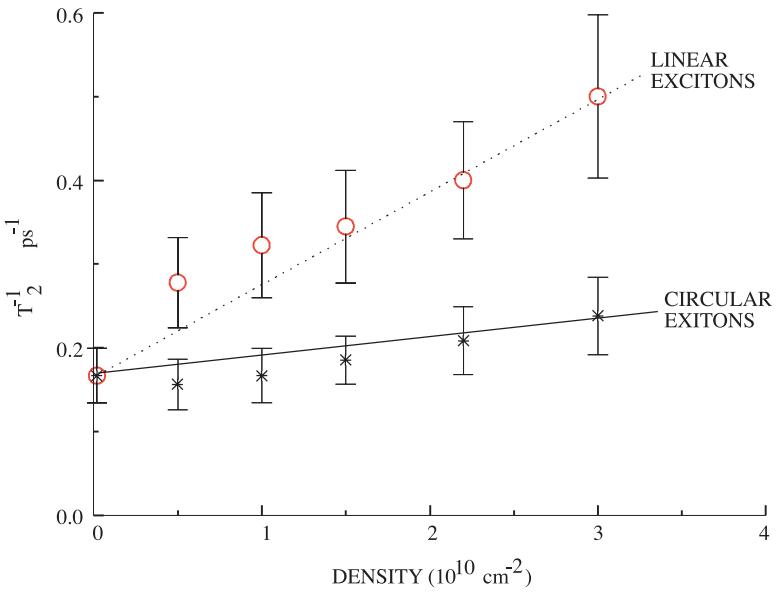

Figure 3. Inverse of the optical dephasing times T_2 of circular (*) or linear ($\circ$) excitons as a function of the exciton density. Dashed line:linear fit of the experimental data for linear excitons. Full line:linear fit of the experimental data for circular excitons taking the broadening parameter calculated in [9] ; the zero density value of T_2 is the low density experimental one.

The agreement between the theory and the experiment is here quite convincing. This is a satisfactory result since Manzke et al. did not include in their calculations the spin-dependent mutual exciton exchange interactions. In other words, their calculations is appropriate for circular and not for linear excitons.

In summary, we demonstrate in this work that coherent superposition of exciton states can be achieved by using a sequence of two phase-controlled optical excitation pulses. We show that the coherent control of excitons in semiconductors can be monitored by their secondary emission, yielding direct measurements of the optical dephasing time T_2. At high density, mutual exciton exchange interaction is evidenced, leading to a spin dependent collisionnal broadening parameter.

References

[1] M. M. Salour and C. Cohen-Tannoudji, Phys. Rev. Lett. **38**, 757 (1977).

[2] N. F. Sherer, A. Ruggiero, M. Du, and G. R. Fleming, J. Chem. Phys. **93**, 856 (1990)

[3] For a review, see Coherent Optical Interactions in Semiconductors, edited by R. T. Philips, NATO ASI Ser. B, Vol. 330 (Plenum Press, New York, 1994).

[4] M. Z. Maialle, E Andrada e Silva, and L. J. Sham, Phys. Rev. B **47**, 15776 (1993).

[5] E. O. Göbel, K. Leo, T. C. Damen, and J. Shah, S. Schmitt-Rink and W. Schäfer, J. F. Müller, K. Köhler, Phys. Rev. Lett. **64**, 1801 (1990).

[6] S. Ceccherini, F. Bogani, M. Gurioli, M. Colocci, Opt. Commun. **132**, 77 (1996).

[7] A. Honold, L. Schulteis, J. Kuhl, C. W. Tu, Phys. Rev. B **40**, 6442 (1989).

[8] R. Eccleston, B. F. Feuerbacher, J. Kuhl, W. W. Rühle, and K. Ploog, Phys. Rev B **45**, 11403 (1992).

[9] G. Manzke K. Henneberger, and V. May, Phys. Status Solidi (b) **130**, 233 (1987)

[10] T. Amand, D. Robart, X. Marie, M. Brousseau, P. Le Jeune, and J. Barrau, Phys. Rev. B. **50**, 9880 (1997).