Coherent Control of the Optical Orientation of Excitons in Quantum Wells

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We demonstrate that the optical orientation of excitons can be coherently controlled and directly observed in a resonant time resolved photoluminescence experiment. The optical dephasing time of excitons, their longitudinal and transverse spin relaxation times, and their radiative lifetime, are measured with strictly the same experimental conditions. The measurements rely on the linear response of the crystal. [S0031-9007(97)04222-1]

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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 [1]. Two types of coherence phenomena must be distinguished. One is the *optical coherence* between the ground and excited states whereby the electronic excitations are coupled to the phase of an electromagnetic wave. The other one is *quantum coherence* between the excited electronic states that are closely adjacent in energy. The optical coherence phenomena in solids are usually investigated by nonlinear techniques as four wave mixing [2–4], photon echo [5], and more recently reflectivity [6] experiments. The coherent control of a quantum mechanical system by a laser light which allows one to manipulate the quantum states is a kind of ultimate challenge in the laboratory. However, the direct monitoring by luminescence of such a coherent control is so far restricted to atoms and molecules. We demonstrate in this Letter that the coherent control of electronic excitations in semiconductors can also be monitored by luminescence, yielding direct measurements of the *optical dephasing* time. We show that the optical orientation of excitons can be coherently controlled and directly observed in a time resolved luminescence (TRL) experiment. In contrast to recent reflectivity experiments [6] or resonant Rayleigh interferometric experiments [7] which demonstrate the coherent destruction of carriers, the present work deals with a pure phase of cold excitons photogenerated under resonant excitation. As shown recently this is a condition for the stability of the exciton states [8], a decisive point in the present study which deals with the manipulation of these states.

We present the results on a GaAs/AlGaAs multiple quantum well (MQW) which consists of 30 periods of nonintentionally 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 luminescence peaks is about 0.1 meV, denoting the high quality of the sample. Similar observations have been made in other high quality GaAs/AlGaAs MQWs with sufficiently narrow linewidths.

A sequence of two 1.4 ps optical pulses of opposite helicities σ^+ and σ^-, split from a mode locked Ti: sapphire laser beam, resonantly excites the XH 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 resolution on the optical path difference between the two interferometer arms is better than $\lambda/20$ where λ is the excitation wavelength. 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. The detection direction is set along the normal to the sample surface, 20° from the excitation specular reflection. The TRL kinetics are recorded by upconverting the luminescence signal in a $LiIO_3$ nonlinear 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. The temporal pulse width of the OPO measured from a cross correlation is 1.5 ps. This two colors up-conversion technique is necessary to record the dynamics of excitons photocreated resonantly [9]. We used picosecond pulses rather than femtosecond ones in order to selectively excite the XH excitons: This is a crucial difference with previous experiments in which light-hole excitons and free carriers are also excited simultaneously [6,9,10]. It is now well known that the exciton dephasing is strongly modified with the presence of free electrons and holes [4]. All the measurements were carried out at a temperature of 10 K and the photogenerated exciton density is about 10^9 cm^{-2}.

For a (100)-grown QW, the relevant symmetry is D_{2d}. The growth direction oz is taken as the quantization

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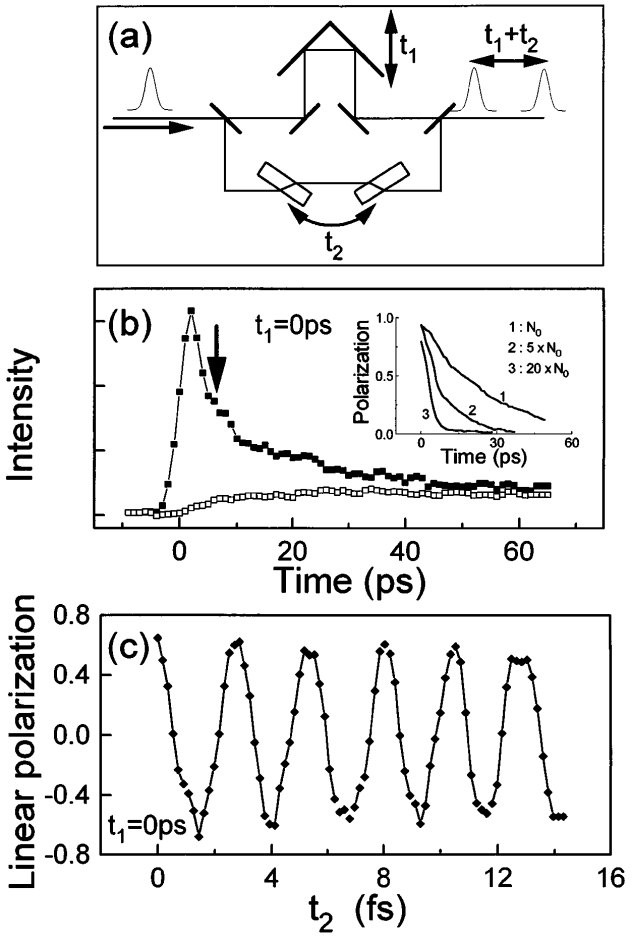

FIG. 1. (a) Schematic excitation arrangement. In (b) *and* (c) the sequence configuration is (σ^+, σ^-) and $t_1 = 0$. (b) The time evolution of I^X (■) and I^Y (□) for $t_2 = m\lambda/c$. Inset: Dependence of the linear polarization decay (characteristic time T_{s2}) as a function of the photocreated exciton density, $N_0 \simeq 10^9$ cm^{-2}. (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.

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 $j_z = \pm 1/2$. As the heavy-hole/light-hole splitting in the investigated sample is greater than the exciton binding energy, the XH exciton states can be described using the heavy-hole subspace only. The appropriate basis is $\{|J_z\rangle \equiv |j_z + s_z\rangle\}$, i.e., $\{|1\rangle, |-1\rangle, |2\rangle, |-2\rangle\}$. In the time domain investigated in the Letter ($\lesssim$25 ps) only the optically active subspace $\{|1\rangle, |-1\rangle\}$ is explored by the excitons due to the much longer electron *and hole* single particle spin-flip times [8,11]. A circularly polarized light $\sigma^\pm$ creates excitons on states $|\pm 1\rangle$ (the "circular excitons" in the following), and a linearly X- or Y-polarized light creates excitons on the coherent states $|X\rangle = (|1\rangle + |-1\rangle)/\sqrt{2}$ and $|Y\rangle = (|1\rangle - |-1\rangle)/i\sqrt{2}$ (the "linear excitons") [12].

First, the main delay between the two excitation pulses is $t_1 = 0$ and the intensities are strictly equal. In this case, the optical interference results in a linearly polarized light excitation with a polarization direction in the QW plane depending on t_2. 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 in the QW plane results in a linearly X-polarized optical excitation. The recorded linear polarization, initially almost equal to 1, decays with a characteristic time $T_{s2} \simeq 20$ ps, the so-called "transverse spin-relaxation time." This transverse spin-relaxation time is the decay time of the quantum spin coherence. It is generally longer than "the optical dephasing time" T_2^0 of excitons [3,4]. It is recognized now that, at low density, T_{s2} is driven by the intraexciton exchange interaction [13]. It has been demonstrated recently that, when the density increases, a much more efficient spin-relaxation process takes place, driven by the interexciton exchange interaction [14]. This effect of the density on the linear polarization decay is illustrated in the inset of Fig. 1(b). The exciton population lifetime measured in Fig. 1(b) ($T_1 \simeq 15$ ps) is consistent with other measurements and theoretical calculations of the free exciton radiative recombination time on high quality MQWs [15].

Figure 1(c) displays the linear polarization dependence versus the fine temporal separation between the two pulses $P^l(t_2)$. This recording, as all the similar recordings in this paper, is systematically taken 4 ps after the second pulse to avoid any perturbation due to backscattered laser light from the sample surface (as a consequence, a systematic small loss of polarization occurs). The oscillations of the luminescence polarization observed in Fig. 1(c) at the period $T = h/E_{\mathrm{XH}}$ 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,

$$|\psi^l(t_2)\rangle = \cos(\omega t_2/2)\,|X\rangle + \sin(\omega t_2/2)\,|Y\rangle .$$

As a matter of fact, the linear polarization of the exciton luminescence is $P^l = \cos \omega t_2$ where $\omega = E_{\mathrm{XH}}/\hbar$.

Now, the delay t_1 between the two excitation pulses is set to 6.6 ps. There is no temporal overlap between the two pulses. Figure 2(a) presents the luminescence 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.

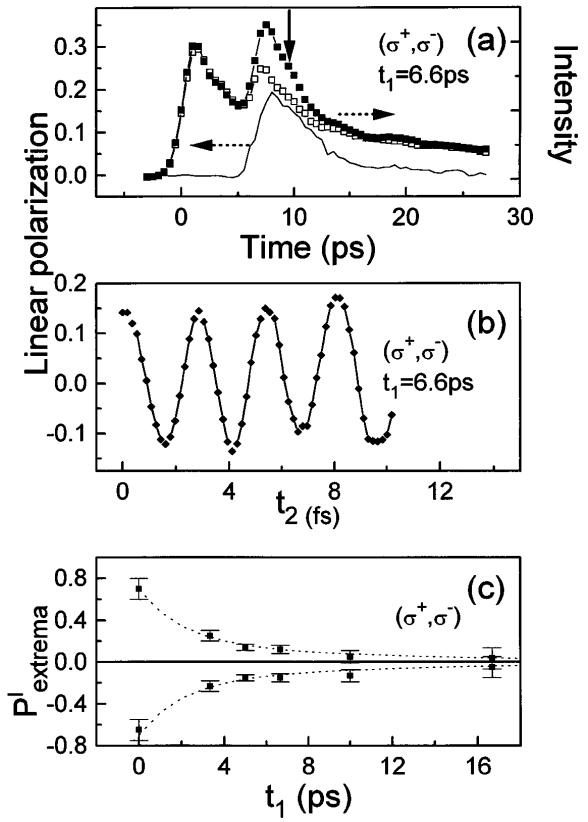

FIG. 2. The configuration is (σ^+, σ^-) and $t_1 = 6.6$ ps. (a) The time evolution of I^X (■), I^Y (□), and the linear polarization P^l (full line) for $t_2 = m\lambda/c$ (the backscattered laser light from the sample surface is negligible). (b) The linear polarization P^l measured 4 ps after the second excitation pulse [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 eyes).

The first optical pulse (σ^+ polarized) sets up a macroscopic 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 macroscopic polarization of linear excitons on states $|\psi^l(t_2)\rangle$. The oscillations 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 luminescence 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 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 oscillations 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, as can be inferred from [7,16]: The decay time is the so-called "optical dephasing time" (T_2^0). We measure $T_2^0 = 6 \pm 1$ ps [17]. The result is in agreement with previous measurements by the four wave mixing technique [1].

At this point it is useful to be more precise about the true nature of what we call in this Letter "the luminescence arising from the excitons $|\psi^l(t_2)\rangle$." Following recent works by Zimmermann [18], Citrin [19], and Haacke *et al.* [20], the secondary emission after the coherent excitation of excitons by an ultrafast optical pulse includes both disorder-induced resonant Rayleigh scattering, which is coherent with the resonant excitation, and incoherent luminescence from excitons which have experienced energy or phase relaxation. Actually excitons $|\psi^l(t_2)\rangle$ are detected 4 ps after their generation at time t_1, when a fraction of them has lost the optical phase. However, this has no consequence for their contribution to the linear polarization of the light emission. This means that "the luminescence of excitons $|\psi^l(t_2)\rangle$" includes both coherent and incoherent contributions.

Consider now a different experimental configuration in which we use a sequence of two linearly cross-polarized optical pulses, X and Y successively. Here, the interference of the second pulse with the coherent macroscopic polarization created by the first one results in a coherent macroscopic polarization of "elliptic excitons" which occupy the states [14],

$$|\psi^{\varepsilon}(t_2)\rangle = \cos(\omega t_2/2 - \pi/4)\,|1\rangle - i\,\sin(\omega t_2/2 - \pi/4)\,|\bar{1}\rangle .$$

These excitons, which are characterized by a circular polarization $P^c = \sin \omega t_2$, contribute to the circular polarization of the luminescence. This effect can be compared to the observation of Faraday rotation of a linearly polarized light probe after the excitation by a sequence of two cross-polarized pump pulses [10]. This Faraday rotation demonstrates that a quantum superposition of exciton states is achieved. Here the circular polarization P^c decays in a characteristic time T_{s1}, the so-called "longitudinal spin-relaxation time" of excitons [11,13]: We measure $T_{s1} = 40$ ps. It is shown in Fig. 3(a) that this circular polarization oscillates at the pulsation ω as a function of t_2 when $t_1 = 6.6$ ps. In this configuration also the envelope of minima and maxima of the circular polarization oscillations decay when t_1 increases with the characteristic time T_2^0, providing again the direct measurement of the optical dephasing time of excitons. We find again $T_2^0 = 6 \pm 1$ ps [Fig. 3(b)].

Finally we consider the experimental configuration in which we use a sequence of two linearly copolarized optical pulses. In contrast to the previous configurations which relied on coherent control of the exciton

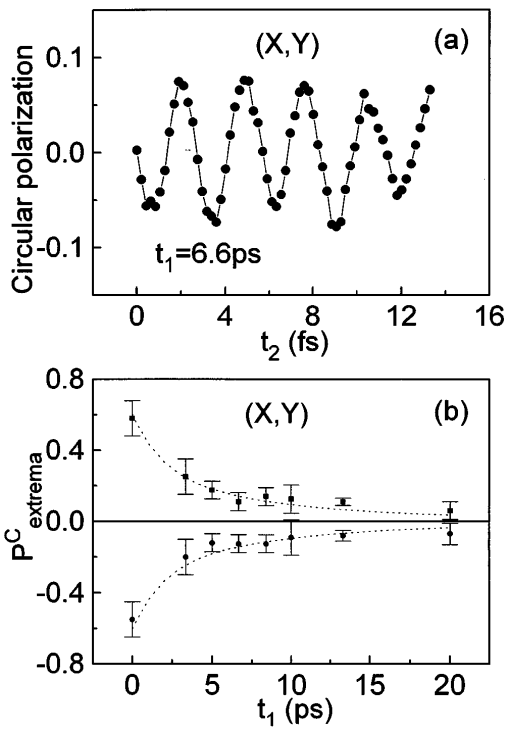

FIG. 3. The configuration is (X, Y) and $t_1 = 6.6$ ps. (a) The circular polarization $P^c = (I^+ - I^-)/(I^+ + I^-)$, as a function of the fine temporal separation t_2 between the two pulses ($I^{\pm}$ are the circularly polarized luminescence components). (b) The maxima and minima of the circular polarization oscillations as a function of t_1 (the dotted line is a guide for the eyes).

polarization, it is now the exciton density which is coherently controlled, as in Ref. [6]. Constructive and destructive interferences between the second optical pulse and the coherent exciton surviving from the first pulse modulate the total exciton density, as demonstrated by the oscillations of the luminescence intensity as a function of t_2 at a fixed delay t_1. This is illustrated in Fig. 4. The decay of this modulation amplitude as a function of t_1 reflects the decay of the coherent exciton population after its generation by the first optical pulse; the decay time is in agreement with the T_2^0 measured in the other configurations. Similar observations have been done when the sample is excited by a sequence of two circularly copolarized optical pulses.

In summary, we demonstrate in this Letter that the coherent control of electronic excitations in semiconductors can be monitored by luminescence, yielding direct measurements of the *optical dephasing* time T_2^0. In contrast to four wave mixing techniques in which the principle of measurement of T_2^0 is based on the third order nonlinear polarization effect, the present measurement relies on the linear response of the system. We have shown that the optical orientation of excitons can be coherently controlled on time scales shorter than the optical dephasing

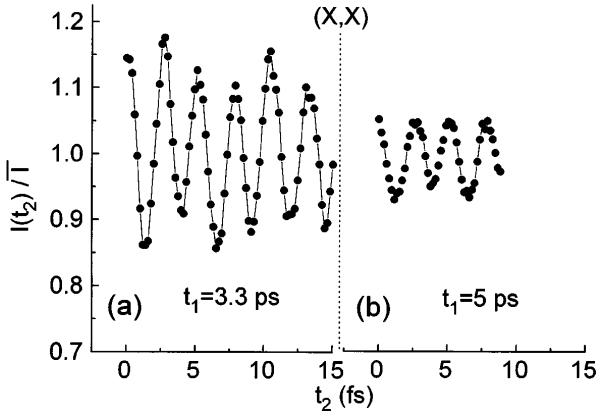

FIG. 4. The configuration is (X, X). The total luminescence intensity $I(t_2)$ when (a) $t_1 = 3.3$ ps and (b) $t_1 = 5$ ps. In order to show the damping of the oscillations as a function of t_1, the total luminescence intensity has been divided by its average value $\bar{I} \equiv \overline{I(t_2)}$.

time and directly observed in a TRL experiment. The optical dephasing time T_2^0 of excitons, their longitudinal and transverse spin-relaxation times T_{s1} and T_{s2}, and their radiative lifetime T_1 are measured in the same experiment with strictly the same experimental conditions for the first time.

We are grateful to Professor B. Girard for fruitful discussions.

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