Long exciton spin relaxation in coupled quantum wells

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Spatially indirect excitons in a coupled quantum well structure were studied by means of polarization and time-resolved photoluminescence. A strong degree of circular polarization (>50%) in emission was achieved when the excitation energy was tuned into resonance with the direct exciton state. The indirect transition remained polarized several tens of nanoseconds after the pumping laser pulse, demonstrating directly a very long relaxation time of exciton spin. The observed spin relaxation time exceeds the radiative lifetime of the indirect excitons. © *2010 American Institute of Physics*. [doi:10.1063/1.3458703]

Semiconductor heterostructures with tunable spatial separation of electrons and holes attract a great attention for their possible applications in optoelectronic devices.[1–4] While the storage of excitons for several microseconds has been experimentally evidenced,[1,5–8] it is still an open question how to exploit the spin of indirect excitons for information storage. Recent experiments performed on coupled quantum wells (CQWs) reported exciton spin transport in the range of micrometers, suggesting indirectly a long exciton spin relaxation time of nanoseconds.[9] Here, we exploit a confocal optical scheme in order to study the spin dynamics of indirect excitons at the location of their excitation. Hereby, we resolve the spin relaxation of indirect excitons in CQWs via time-resolved and polarization-resolved photoluminescence (PL) studies after resonant and nonresonant excitation of the direct and indirect exciton states. We observe that under conditions of resonant excitation a highly efficient initialization of exciton spin takes place. The PL remains strongly circularly polarized long after the laser pulse and nearly constant during the lifetime of excitons. Our results directly confirm a long spin relaxation time of >80 ns for indirect excitons. This time scale is an order of magnitude longer than the one obtained by read-out schemes with a larger focus spot.[10]

The studied heterostructure consists of two 8 nm wide GaAs/AlGaAs CQWs separated by a 4 nm $Al_{0.3}Ga_{0.7}As$ barrier and it was fabricated as a field-effect device.[11–13] The shape of the confining potential is controllably adjusted by the bias (V_g) applied between a semitransparent metal gate and a deep Ohmic contact, allowing the direct manipulation of the exciton lifetime via a modified electron–hole separation [Fig. 1(a)]. The PL excitation and collection are performed through the semitransparent part of the top Schottky gate using a confocal microscope based on a short focal length aspheric lens. The luminescence is dispersed by a 0.5 m double monochromator and detected with an intensified charge coupled device detector. The configuration of the setup allows us to perform measurements under identical (co) and orthogonal (cross) polarizations of excitation and detection. The resulting extinction ratio is below 0.0015 for the two opposing circular polarizations. All measurements are performed at 4.2 K.

In order to precisely define the initial spin polarization of excitons we optimize the process of resonant excitation. In CQWs the bright excitons form a two level system and the eigenstates $|\pm 1\rangle$ are coupled to circularly polarized photons [see scheme in Fig. 1(b)]. As a strictly resonant excitation and PL measurements are rather incompatible, we take ad-

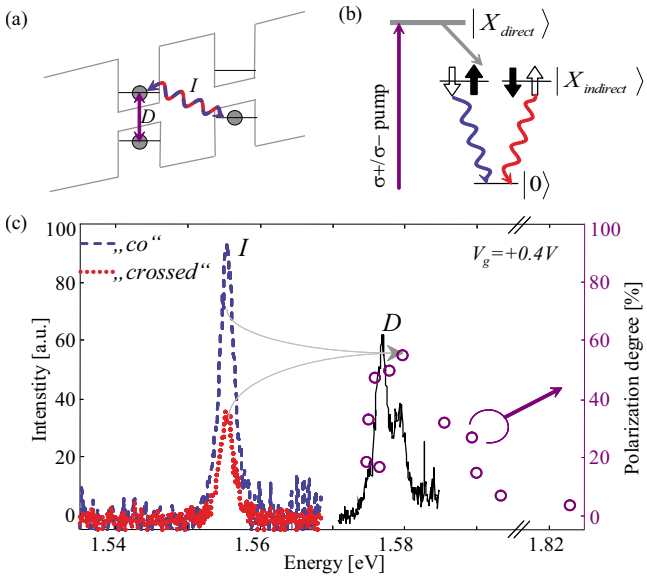

FIG. 1. (Color online) (a) Scheme of GaAs CQW under applied electric field showing formation of direct (D) and indirect (I) excitons. (b) Diagram of resonant polarized excitation in direct excitonic states and emission from two optically active indirect states. (c) Left scale: PL spectra for direct excitons (D, solid black line) under nonresonant excitation and for indirect excitons (I) under resonant 1.58 eV excitation, polarization resolved in co (dashed blue) and cross (dotted red) configurations. Right scale: degree of circular polarization for indirect excitons vs excitation laser energy (open circles). The gray arrows indicate the point obtained from the spectra shown in the left part of the figure.

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vantage of the quasiresonant excitation into spatially direct exciton states, which has proven to be very efficient in similar structures.[9] Such a quasiresonant and circularly polarized excitation should provide an efficient transfer of light polarization into exciton spin polarization. The two quantum wells in the heterostructure are identical. Therefore a quasi resonant excitation results in pumping simultaneously the direct transitions in both wells. Since the direct PL recombination lifetime is typically 1 ns,[14] the conversion of direct into indirect states is likely related with a fast carrier tunneling between the wells, which is of the order of tens of picoseconds (estimated from Wentzel–Kramers–Brillouin approximation[15]). Then, the exciton recombination transforms the spin polarization of the tunneled charged carriers into the polarization of the emitted photon. Thus, the optical polarization of the indirect excitons provides a direct and very sensitive tool to study spin related phenomena in CQWs [Fig. 1(b)].

We employ a 1.823 eV (680 nm) laser diode to examine a nonresonant excitation. The scheme is used to measure the PL of the direct excitons and to determine the optimal excitation conditions for indirect emission [black line in Fig. 1(c)]. Then, the PL from indirect excitons is measured for different voltages and under "co" and "cross" configurations of the circular polarization in the excitation and the detection paths. In the former part of the experiment, a continuous wave laser diode is used. Tuning systematically the laser diode energy in the range 1.57–1.59 eV (780–790 nm) enables us to drive the excitation through the direct excitonic states. We measure the degree of circular polarization of indirect excitons $P_{\mathrm{circ}}=\frac{I_{\mathrm{co}}-I_{\mathrm{cross}}}{I_{\mathrm{co}}+I_{\mathrm{cross}}}$ as a normalized difference between PL intensities I_{co} and I_{cross} in co and cross configurations, respectively. Figure 1(c) shows the two spectra of indirect excitons taken in co and cross polarization configurations under strictly resonant excitation (1.58 eV) into the direct excitons. The right hand side of the figure illustrates the degree of circular polarization for indirect excitons as a function of excitation laser energy. For nonresonant excitation at 1.82 eV no significant polarization of indirect excitons PL is measured, whereas a clear enhancement of the circular polarization is observed for a quasiresonant excitation of the direct excitonic states. The maximum value of P_{circ} is about 55% for the excitation at 1.58 eV. Such efficient polarization conservation suggests a long relaxation time of the exciton spin, which exceeds the radiative recombination time.

The PL circular polarization can be written as:[16,17]

$$P_{\mathrm{circ}}=\frac{\tau_s}{\tau_s+\tau_r}P^0_{\mathrm{circ}}, \tag{1}$$

where $\tau_s(\tau_r)$ is the spin relaxation time (radiative lifetime) of excitons and P^0_{circ} is the effective initial circular polarization determined by the laser polarization and losses during the formation of an indirect exciton. Hereby, we estimate that the spin relaxation under strictly resonant excitation is at least 1.2 times longer than the exciton lifetime, when neglecting any polarization loss in the excitation process $P^0_{\mathrm{circ}}=1$. The radiative lifetime for indirect excitons reaches several tens of nanoseconds, thus the spin relaxation time must be even longer.

To confirm straightforwardly this hypothesis we apply a pulsed excitation at 1.577 eV and co and cross polarized PL spectra are recorded for different delays after the 400 ns long

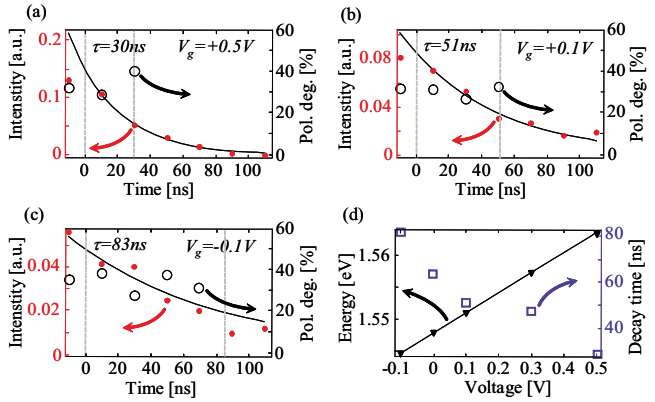

FIG. 2. (Color online) [(a)–(c)] For three different applied biases under 1.577 eV excitation: left scale—PL decay vs delay time after the excitation pulse (filled circles—experimental data, line—exponential fit), and right scale—circular polarization degree (open circles). Vertical dashed lines mark zero delay and the one corresponding to the PL decay time, respectively. (d) Dependence of transition energy on applied bias (black triangles—experimental data, line—linear fit of the Stark shift), and of PL decay time (opened blue squares).

laser pulse.[18] Figures 2(a)–2(c) presents the time evolution of the PL after the laser pulse (filled circles), measured for different gate voltages. The emission decay rate, obtained from the exponential fit,[18] shows its increase with applied electric field[19] as pictured in Fig. 2(d). The obtained circular polarization degree P_{circ} is plotted up to delay times comparable with radiative decay time in Figs. 2(a)–2(c). The degree of circular polarization for the indirect exciton emission remains constant several tens of nanoseconds after the laser pulse. For an applied bias of −0.1 V the radiative lifetime is measured to be as long as 83 ns and PL polarization is constantly ~35% during this period [see Fig. 2(c)]. This suggests that the measured spin relaxation time is limited by the PL decay time.

Generally, the spin polarization of excitons is maintained only for very short times in a single quantum well. In such case the spin relaxation is dominated by dephasing phenomena which results from carriers exchange interaction.[20–22] The large electron–hole separation in CQWs suppresses this interaction and enables much longer relaxation time for excitonic spin. If the dipolar excitons exhibit a high mobility, their spin relaxation time might be shortened due to Dyakonov–Perel mechanism (DP).[9] We note that τ_s in CQW obtained from our studies exceeds few times the one estimated from the other experiments.[9,10] In Ref. 10 only very short <1 ns relaxation times were measured but the authors noted that high-energy part of the spectrum conserved the exciton spin for much longer time scales (few nanoseconds) than the lower energy tail. As their signal was investigated only in the time domain but without confocal scheme, it averaged over big area of drifting excitons. The authors of Ref. 9 used the PL polarization degree and decay time to estimate the spin relaxation time using an equation similar to Eq. (1). Exciton transport was included in the modeling, which improved the accuracy of such estimate. Their method gave $\tau_s \sim 10$ ns. Our technique allows to directly measure the time evolution of the spin polarization. In our confocal microscope the emission and collection aperture is limited to ~1 μm. Generally, studying the excitons in the excitation spot does not eliminate the spin relaxation caused by DP mechanism. Thus, if excitons in the excitation spot are mo-

bile, the DP mechanism can induce a strong reduction in τ_s. If exciton transport is suppressed, i.e., excitons are localized, the DP relaxation mechanism does not influence much τ_s.

To conclude, the polarization and time-resolved PL of the indirect emission from coupled GaAs/AlGaAs quantum wells is investigated. Quasiresonant excitation is employed to effectively initialize the exciton spin population via the direct excitons and a subsequent spin transfer and spin storage into indirect excitons. Our findings from continuous and time-resolved experiments directly demonstrate a long spin relaxation time of indirect excitons in CQWs exceeding 80 ns.

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