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Optical control over electron *g* factor and spin decoherence in (In,Ga)As/GaAs quantum dots

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We have studied the dependence of the electron in-plane *g* factor and spin decoherence time on the built-in electric field (E_i) at the position of a single layer of self-assembled (In,Ga)As/GaAs quantum dots (QDs). Control of E_i is achieved by inducing screening charges in a *p-i-n* GaAs matrix with a continuous wave (cw) laser. Using a time-resolved pump-probe technique to measure the spin dynamics via the magneto-optical Kerr effect, we observe a large hole spin decoherence time of 440 ps. Measurements as function of the cw laser power and, thus, of E_i show that the electron spin decay time in the QDs depends strongly on E_i and decreases from 310 to 110 ps with increasing E_i. We attribute this effect to increasing tunneling rates of electrons out of the QDs at high E_i. We observe a slight increase of the electron *g* factor from 0.40 ± 0.03 to 0.46 ± 0.04 with increasing E_i, which might be a result of a changing wavefunction as a result of a different confinement potential due to E_i. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2838156]

INTRODUCTION

Semiconductor quantum dots (QDs) are good candidates for buildings blocks of future spintronic devices, such as single photon emitters for quantum information applications[1] and qubits for quantum computation.[2] In the latter application, the electron spin in a quantum dot forms a two-level quantum system that is the basis of the qubit. Two key ingredients for full functionality of the qubit are a long spin decoherence time and the ability to manipulate each spin. Spin decoherence times should be longer than the time needed for performing spin operations. One way to enable local spin manipulation that might lead to scalable qubits is *g*-factor engineering in a solid state environment. Methods reported for *g*-factor engineering include the use of parabolic quantum wells and a gate voltage,[3,4] strain engineering of the quantum dot growth,[5] and electrical tunability of the molecular spin state *g* factor in quantum dot molecules.[6] Besides this application driven research, some fundamental issues still exist, such as the precise dependence of the electron and hole *g* factor and spin decoherence time on the size, shape, and composition of the QDs.[7,8] In this paper, we study the dependence of the electron *g* factor and spin decoherence time on the built-in electric field (E_i) at the position of a single layer of self-assembled (In,Ga)As/GaAs QDs by a time-resolved magneto-optical technique. Control over E_i is achieved by screening E_i with photoexcited carriers by continuous wave (cw) laser excitation. The tunability of the electron *g* factor and spin decoherence time will be discussed.

EXPERIMENTAL DETAILS

The self-assembled (In,Ga)As/GaAs QDs are grown by molecular beam epitaxy at a temperature of 525 °C. The single layer of QDs is situated at the center of the intrinsic region of a GaAs *p-i-n* heterostructure which is grown on a *p*-doped GaAs substrate, as shown in Fig. 1(a). The heavily p^{++} and n^{--} regions have a doping concentration of 5×10^{18} cm^{-3}, which results in a built-in electric field over the 60 nm intrinsic region of ~240 kV/cm. The (In,Ga)As QDs are grown on GaAs via the Stranski–Krastanov growth method. Atomic force microscope images taken at the same sample right after the quantum dot growth indicated that the QDs have a height of 6.7 ± 1.5 nm and an average diameter of 24.8 ± 5.0 nm. The dot density was determined to be 4.5×10^{10} cm^{-2}. Photoluminescence (PL) spectra were taken with a cw laser operating at 1.55 eV as function of excitation power (P_{exc}). A PL spectrum taken with $P_{exc}=0.4$ kW/cm^2 is shown in Fig. 1(b). It has a peak at 1.269 eV and a full width

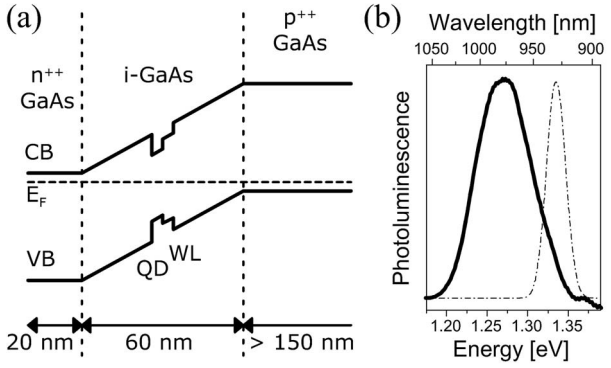

FIG. 1. (a) Schematic energy diagram of the *p-i-n* heterostructure with the quantum dots (QDs) and wetting layer (WL) at the center of the intrinsic region. The position of the conduction band (CB), valence band (VB), and Fermi level (E_F) is indicated. (b) Photoluminescence spectrum of the QDs with P_{exc} 0.4 kW/cm^2. Also, the (low) emission from the wetting layer at 1.37 eV can be seen. The dashed line represents the spectral profile of the laser pulses used in the time-resolved experiments.

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at half maximum of 80 meV owing to the size distribution and, therefore, the ground state energy levels of the QDs. The PL spectra exhibit a Stark shift as function of P_{exc}, with saturation when $P_{exc} > 0.2$ kW/cm^2. The saturation indicates the full quenching of E_i and by mapping E_i via the Stark shift to P_{exc}, we are able to tune E_i from 0 to 240 kV/m. These PL results will be published elsewhere.[9]

We have studied electron and hole spin dephasing by optical means, using time-resolved magnetization modulation spectroscopy (TiMMS). The quantum dot sample is placed in a magneto-optical cryostat at a temperature of 5 K and in a magnetic field (up to 0.35 T) in the Voigt geometry (parallel to the sample plane). A mode-locked Ti:sapphire laser, operating at 80 MHz, produces ~100 fs laser pulses with a wavelength of 930 nm (1.335 eV, which is *below* the GaAs bandgap). The spectral width of the laser pulses is typically 10 nm (14.4 meV) so we probe a relative large portion of the QDs that emit in the high energy tail of the PL spectrum [see Fig. 1(b)]. Consequently, the obtained decoherence times and g factors are averages over the probed ensemble of QDs. In order to perform stroboscopic time-resolved measurements, the laser beam is divided in a strong pump beam (~5 mW) and a weaker probe beam (~1.5 mW). The use of these relatively strong laser powers is justified by the low absorption of the thin wetting layer and QDs. The time delay between pump and probe beams is controlled by a motorized translation stage. The pump beam passes through a photoelastic modulator, producing left- and right-circularly polarized light at a frequency of 50 kHz for optical spin injection. Both pump and probe beams are focused to an overlapping spot of ~12 μm on the quantum dot sample and the reflected probe light is collected with a Si photodiode, after passing a quarter wave plate ($\alpha_{qwp}=45°$) and analyzer ($\alpha_{ana}=0°$). It can be shown[10] that the measured signal is proportional to the magneto-optical Kerr rotation, which originates from differences in the joint density of states of spin up and spin down carriers. It is thus proportional to the electron and hole spin in the system. We note that a TiMMS measurement yields the electron and hole spin decay time (τ_e and τ_h, respectively). The electron-hole recombination time (τ_r) should be used to extract the true electron and hole spin decoherence times τ_{se} and τ_{sh}, respectively, via $1/\tau_e = 1/\tau_r + 1/\tau_{se}$.

Additionally, we use a cw HeNe laser with $\lambda = 632.8$ nm (1.92 eV, *above* the GaAs bandgap) and a laser power of 0–3.2 mW ($P_{HeNe}=0-0.4$ kW/cm^2). The laser beam is also focused to an overlapping spot of ~16 μm diameter in order to create charge carriers in the GaAs that screen E_i. We note that the capture of carriers excited by the pump pulse is much larger than those excited by the HeNe laser.

RESULTS AND DISCUSSION

TiMMS measurement taken with $P_{HeNe}=0.38$ kW/cm^2 and $B=0.35$ T is shown in Fig. 2. The observed time response consists of several parts. First, there is a peak in the TiMMS signal around zero delay, which we attribute to coherent higher order processes during temporal overlap of pump and probe beams. Second, after the coherent peak, the TiMMS signal is negative and rises exponentially toward a maximum positive value with a characteristic time much larger than the temporal width of the laser pulses. This exponential rise of the signal can be attributed to the capture of electron and holes, first excited in the wetting layer, into the quantum dots. Finally, the signal decays to zero and consists of an exponentially decaying part and an oscillatory component. In bulk materials, where the spin relaxation time of holes (τ_{sh}) is typically very short (<1 ps) (Ref. 11) due to the mixing of heavy and light hole states, one would expect a damped sine wave oscillating around zero due to electron spin precession and dephasing (with characteristic time τ_{se}) when $B>0$. Here, however, a striking difference is observed, as the signal does not oscillate around zero. Such a response may arise when τ_{sh} is of the same order as τ_{se}. This is very well possible in QDs, where similar to the situation in quantum wells,[12] due to quantum confinement effects, the degeneracy between heavy and light holes is lifted. Therefore, the hole spins do not precess and τ_{sh} is enhanced up to several orders of magnitude.[13–15] The fact that the first minimum reaches nearly zero signal indicates that equal amounts of electron and hole spins are present in the system with comparable decay times. Apart from the coherent peak, the full time response can be well described by the following fitting function:

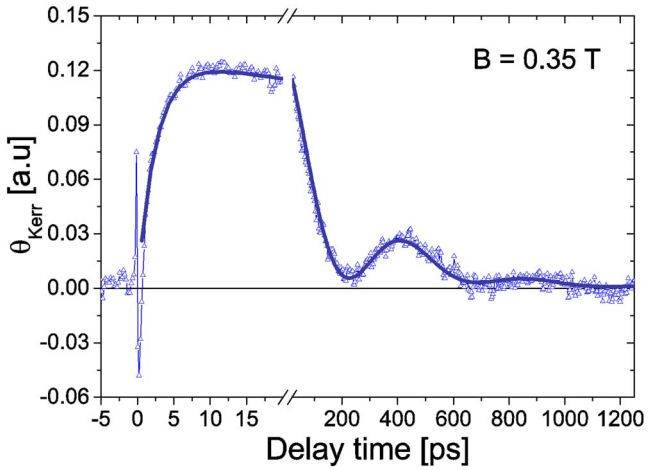

FIG. 2. (Color online) TiMMS signal with $B=0.35$ T and $P_{HeNe}=0.38$ kW/cm^2. The solid line is a fit of the data using Eq. (1).

$$\theta(t) = A[1 - e^{-t/\tau_c}][e^{-t/\tau_e}\cos(\omega t) + e^{-t/\tau_h}]. \quad (1)$$

Here, A is the amplitude of the signal, τ_c the capture time (taken to be equal for both electron and holes, assuming charge neutrality), τ_e and τ_h the spin decay time time of electrons and holes, respectively, and ω the angular frequency of the precessing electrons. Fitting the response using Eq. (1), gives the following parameters: $\tau_c = 2.0 \pm 0.1$ ps, $\tau_e = 240 \pm 9$ ps, $\tau_h = 307 \pm 23$ ps, and $\omega = 13.8 \pm 0.3$ GHz. Indeed, τ_e and τ_h are of the same order of magnitude. Correcting for a recombination time of 1 ns (obtained from time-resolved PL measurement[9]) yields $\tau_{se}=316$ ps and $\tau_{sh}=443$ ps and, thus, a large hole spin decoherence time is observed. From $\omega = g_{\parallel}\mu_B B/\hbar$, with μ_B the Bohr magneton, we find an absolute value of the in-plane electron g factor of

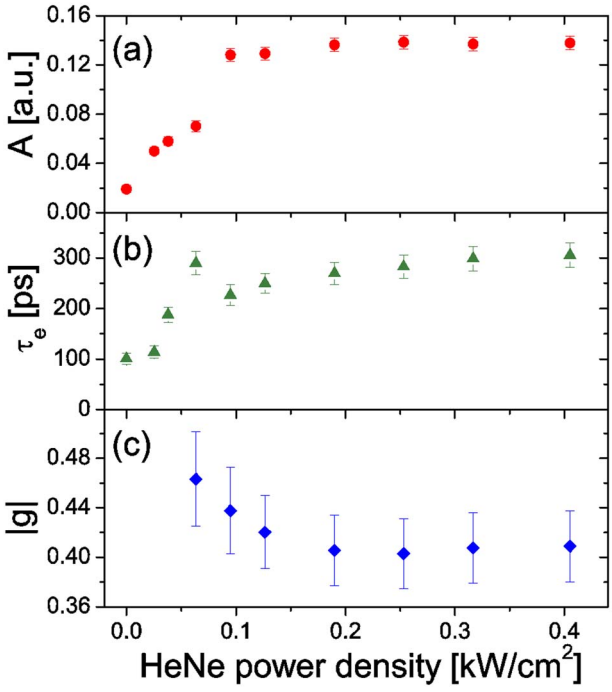

FIG. 3. (Color online) (a) The amplitude of the TiMMS signal, (b) electron spin decay time, and (c) electron g factor as function of P_{HeNe}. Error margins result from the errors of the fitting procedure as well as from variations in different measurements.

$g_{\parallel}=0.44$, which is in good agreement with recent theoretical predictions.[7,8] We note that although we cannot determine the sign of the g factor, and although the absolute value is similar to the bulk g factor of GaAs, the observed capture time and the large hole spin decoherence time are indications that we are truly sensitive to spins in the quantum dots.

We have performed similar measurements as a function of P_{HeNe}, from which we have extracted the amplitude A of the TiMMS signal, the electron spin decay time τ_e and the in-plane electron g-factor $g_{\parallel}$, as shown in Fig. 3. Discussion of the hole spin decay times is beyond the scope of this paper. We note that in the analysis of some measurements, we omitted the exponential rise part of Eq. (1). We observe that A strongly depends on P_{HeNe} [Fig. 3(a)], rising from 0.02 at low P_{HeNe} (high E_i) to 0.14 when E_i is fully quenched. A similar dependence on P_{HeNe} is found for τ_e. These observations can be attributed to the increased loss of electron spin due to tunneling of electrons from the QDs to the GaAs at high E_i. As shown in Fig. 1, at high E_i, a triangular barrier between the QDs and the GaAs exists through which electrons can tunnel. As it is difficult to correct for the exact contribution of tunneling, we cannot conclude that the observed decrease in τ_e indicates a decrease in the electron spin decoherence time τ_{se}. Figure 3(c) shows the dependence of the electron g factor on P_{HeNe}. Although the uncertainty in the determination of $g_{\parallel}$ is rather large as a result of the combination of relative low B and small $g_{\parallel}$, these results indicate a slight increase of $g_{\parallel}$ from 0.40 ± 0.03 at high P_{HeNe} to 0.46 ± 0.04 at smaller P_{HeNe} (thus, increasing E_i). It is known that the electron g factor can change when the overlap of the wavefunction with the surrounding material changes, however, it is not trivial to predict what the change will be for these particular QDs.

In conclusion, we have performed time-resolved magneto-optical measurements on a single layer of self-assembled (In,Ga)As/GaAs QDs as function of the built-in electric field E_i. We have observed electron spin precession and large hole spin decoherence times in the QDs. The decrease of the observed electron spin decay time with increasing E_i can be attributed to enhanced tunneling of electrons out of the QDs. Also, we have observed a slight increase of the electron in-plane g factor from 0.40 ± 0.03 to 0.46 ± 0.04 with increasing E_i, however, additional experiments are needed to fully resolve the behavior of $g_{\parallel}$ at high E_i.

ACKNOWLEDGMENTS

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