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Dispersion of electron g-factor with optical transition energy in (In,Ga)As/GaAs self-assembled quantum dots

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The electron spin precession about an external magnetic field was studied by Faraday rotation on an inhomogeneous ensemble of singly charged, self-assembled (In,Ga)As/GaAs quantum dots. From the data the dependence of electron g-factor on optical transition energy was derived. A comparison with literature reports shows that the electron g-factors are quite similar for quantum dots with very different geometrical parameters, and their change with transition energy is almost identical. © 2011 American Institute of Physics. [doi:10.1063/1.3588413]

A long spin coherence time is an essential ingredient for spintronic devices, in which it is required to store, manipulate, and detect the electron’s spin state. In a singly charged quantum dot (QD), the coherence time of the resident electron spin is in the microseconds range, exceeding by two orders of magnitude the coherence time measured in bulk semiconductors. Therefore, singly charged QDs have attracted much interest as promising candidates for the construction of practical spintronic devices.

Among the various QD fabrication methods, the growth by self-assembly has the advantage of producing high density dot structures free from defects. However, an intrinsic property of such a heterostructure is the dispersion in composition and size of the dots it contains. As a prominent consequence the optical transition energy in the QD ensemble varies, as evidenced by the inhomogeneous broadening of photoluminescence (PL) emission. This inhomogeneity is also translated into the spin properties of confined carriers, resulting in a dispersion of the electron g-factor, which leads to fast dephasing, i.e., loss of measurable magnetization associated with spin polarization of the ensemble when monitored through the spin precession in a perpendicularly applied magnetic field.

Still, the dependence of the electron g-factor on the parameters of the confining QD structures has not been established in full detail. A corresponding understanding is required for tailoring g-factors, which to a limited extent can be obtained by application of external electric fields. The main adjustment, however, has to be obtained already during QD fabrication.

In this letter, we investigated (In,Ga)As/GaAs QD samples grown by molecular beam epitaxy on (100)-oriented GaAs substrates. One sample was grown at the University of São Paulo, Brazil (sample I), and the results on this structure are compared to those of a sample grown at the Ruhr-University of Bochum, Germany (sample II), that were reported in detail already in Refs. 2, 7, and 12. Sample I contained 10 QD layers, separated by 30 nm wide GaAs barriers, and Si δ-doping sheets 15 nm above each QD layer with a doping density equal to the dot density. Sample II, on the other hand, contained 20 QD layers, spaced by 60 nm wide GaAs barriers; here 20 nm above each QD layer, the Si δ-doping was incorporated.

The dot density per layer and dot dimensions were estimated using atomic force microscopy of samples grown under the same conditions but without capping. These parameters are given in Table I, which shows that sample I contains QDs much smaller in size than sample II. Naturally overgrowth changes the dot geometry as does subsequent rapid thermal annealing (which was done for 30 s at almost identical temperatures of 950 °C for sample I and 945 °C for sample II). Nevertheless, due to similar treatments, the difference in dot parameters should be maintained. Despite of this difference, we find that the samples possess similar spintronic properties.

Thermal annealing causes intermixing between dot and barrier materials, resulting in an increase in the QD band gap, and a blueshift in the PL band associated with electron-hole QD ground state recombination as compared to the as-grown sample. Figure 1 shows the PL spectra of both QD samples, using excitation light of wavelength 532 nm (above the GaAs band gap). Sample I shows a PL maximum at somewhat lower photon energies than sample II, despite of the much smaller dot size, suggesting a considerably higher In-content in these dots. The PL linewidth is larger in sample I than in sample II, which is caused by the combined effects of different sizes and compositions, determining width and height of the carrier confinement potential. Reduced size and higher In-content as in sample I lead to a stronger variation in the optical transition energies as result of fluctuations of these parameters in the dot ensemble.

The spin dynamics of the electrons confined in the QDs were probed by pump-probe time-resolved Faraday rotation (TRFR) at $T=6 \text{K}$. The electron spins were oriented by exciting the trion transition with circularly polarized pump light. The data for both samples are shown in Fig. 2. The electron spin precession is shown in Fig. 3, revealing a full dephasing of the spin observable within a few nanoseconds. The spin lifetime, extracted from the data, is 120 ns for sample I and 50 ns for sample II.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Base (nm)</th>
<th>Height (nm)</th>
<th>Density (cm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>20</td>
<td>5</td>
<td>$4 \times 10^{10}$</td>
</tr>
<tr>
<td>II</td>
<td>30</td>
<td>7–8</td>
<td>$1 \times 10^{10}$</td>
</tr>
</tbody>
</table>

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pulses (see Ref. 2 for details). The pulse duration of 1.5 ps corresponds to a linewidth of about 1 meV, much smaller than the inhomogeneous PL linewidths. The sample is placed in a transverse magnetic field, so that the spins precess around the field. A probe pulse, of same energy and duration as the pump, but polarized linearly and variably delayed relative to the pump, traverses the sample. The rotation of its polarization plane due to the Faraday effect is measured, which characterizes the instant orientation of the spin ensemble along the optical axis. Therefore, by scanning pump and probe across the PL band, the energy dispersion of spin properties can be determined, as only the small fraction of the QD ensemble whose absorption falls within the 1 meV range laser width is explored.

Here, we present the results obtained for sample I (results for sample II are given in Ref. 7 and serve as reference here). For pump light resonant with the PL maximum, Fig. 2 shows TRFR as a function of the delay time, \( t \), between pump and probe for different magnetic fields \( B \). At \( B = 0 \), the FR signal after spin orientation at \( t = 0 \) shows a fast decrease up to 500 ps, followed by a slow decrease for later times. The fast component is mostly due to the decay of spin-polarized excitons in charge-neutral QDs in the sample. For times longer than 500 ps, the electron-hole pairs have recombined, and the detected magnetization is reduced to the contributions arising from electrons resident in charged QDs only. The FR decrease in this time range is due to spin precession about the arbitrarily oriented effective magnetic field \( B_{\text{NSF}} \) of the nuclei spin fluctuations. The strength of this field can be estimated by \( B_{\text{NSF}} = h/(\mu_B g_e T_2^*) \). Here, \( \mu_B \) is the Bohr magneton and \( g_e \) is the electron \( g \)-factor (see below). \( T_2^* \) is the spin dephasing time determined from an exponential fit to the \( B = 0 \) trace for times exceeding 0.6 ns, from which we find \( T_2^* = 830 \) ps, which corresponds to \( B_{\text{NSF}} = 25 \) mT. Note that this is considerably larger than for sample II for which we measured \( B_{\text{NSF}} = 7.5 \) mT, giving a ratio of \( B_{\text{NSF}}^2 \) (sample I) greater than \( B_{\text{NSF}}^2 \) (sample II) by an order of magnitude.

According to Ref. 8, for a total number \( N \) of nuclei in the dot

\[
B_{\text{NSF}}^2 = \frac{16}{3N \mu_B^2 g_e^2} \sum_j A_j^2 I_j(I_j + 1),
\]

where the sum runs over all nuclei in a unit cell, with \( A_j \) being the hyperfine interaction energy (38 \( \mu_eV \), 56 \( \mu_eV \), and 46 \( \mu_eV \) for Ga, In, and As, respectively) for an electron with a particular nucleus \( j \) with nuclear spin \( I_j \) (3/2 for Ga and As, 9/2 for In). From this equation, we attribute the larger NSF field partly to the four-times smaller number \( N \) of nuclei in the sample I-dots compared to sample II-dots, as estimated from the QD volume using parameters of Table I. The remaining difference is attributed to the higher In-content, which we estimate to be roughly twice as large in sample I than in sample II-dots.

Next, we discuss TRFR at \( B > 0 \). At positive delays pronounced oscillations are observed, which are associated with the resident electron spin precession for \( t > 0.6 \) ns. The precession frequency, \( \omega_p = (\mu_B g_e B)/h \), increases with magnetic field, as does the damping due to dephasing. The traces for these late delays can be analyzed by the form \( \exp(-t/T_2^*) \cos \omega_p t \), from which we can extract the average \( g \)-factor \( g_e \) within the optically excited spin ensemble as well as the dephasing time \( T_2^* \). Figure 3 shows \( T_2^* \) obtained in this way for both samples as function of magnetic field. The decrease in \( T_2^* \) with increasing field can be described by a
\( \frac{1}{B} \)-dependence. Clearly the dephasing is much stronger for sample I than for sample II.

For magnetic fields exceeding the NSF field, the dephasing is dominated by variations in the electron \( g \)-factor, caused by variations in the dot parameters within the ensemble. For a \( g \)-factor dispersion of half-width \( \Delta g_e \), the dephasing time is given by \( T_2^e = \hbar/\mu_B \Delta g_e \). This explains the \( \frac{1}{B} \)-dependence and allows one to estimate \( \Delta g_e \) from fitting the data. Such fits, shown by the lines in Fig. 3, render transition energy the difference in parameters, in agreement with theoretical predictions.13 This cent is surprisingly small based on the quite different dot samples is at most 0.025. This difference of only a few percent between the samples I and II.

We see a greater dispersion of values for sample I, in agreement with theoretical predictions.13 This indicates that also in QDs the electron \( g \)-factor is mostly determined by the magnitude of the band gap, as expected from the Roth–Lax–Zwerdling relation

\[
g_e = g_0 \left[ 1 - \frac{1}{n_e} \right] \Delta \tag{2}\]

Here, \( g_0 = 2 \) is the free electron \( g \)-factor, \( n_e \) is the conduction band effective mass, and \( E_g \) is the effective band gap (given by the emission energy) and \( \Delta \) is the split-off gap.

This formula should then also describe the variation in the electron \( g \)-factor across the PL bands. The corresponding experimental data versus photon energy are shown in Fig. 1 and demonstrate that the variation in the \( g \)-factor magnitude with optical transition energy shows basically the same slope for the two samples. The black dashed lines in Fig. 1 represent fits by Eq. (2), for a split-off energy fixed at \( \Delta = 0.4 \) eV and using the effective mass as variable parameter to fit the data. Best agreement is obtained with effective masses of 0.0598 for sample I and 0.0591 for sample II, which are basically the same. The observed dependence of the \( g \)-factor on optical transition energy suggests a universal dependence to be checked further.
