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Spin and carrier relaxation dynamics in InAs/GaAs quantum-dot spin-LEDs

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Abstract. We investigate the dynamics of electrons injected into InAs/GaAs quantum dots by initializing and further observing the spin state of the electrons. For this purpose, we use spin polarized light-emitting diodes where the electron spin is set in a semimagnetic ZnMnSe layer. We find that the degree of optical polarization depends strongly on the ground state energy of the quantum dot. A dependence of polarization on dopant concentration in the spin aligner suggests an influence of residual electrons in the quantum dots.

To make use of electronic spin states in semiconductor quantum dots (QD) for quantum information processing, it is necessary to reliably initialize such states. Two main approaches exist: Optical initialization by exciting spin-polarized carriers resonantly in the QD using optical orientation [1] or electrical injection by making use of a spin-aligner to orient the electron spin. The latter method is very elegant since it does not need complex laser setups. Still it is very challenging because reliable and error-free initialization of the quantum dots must be given.

We investigate here spin polarized light-emitting diodes (spin-LEDs) based on III-V/II-VI heterostructures. The electron spins are initialized in a semimagnetic ZnMnSe layer. In the latter, the s-d exchange interaction between the electrons and the Mn²⁺ ion spins at low temperatures and in an external magnetic field leads to a huge Zeeman splitting and due to efficient spin relaxation into the ground state the electron spins become spin polarized. Then, the electrons relax through an undoped spacer layer into InAs/GaAs QDs. From the other side of the spin-LED, unpolarized holes are injected into the QD layer. Since the contributing valence band states are solely the heavy hole states (the light-hole states are well separated due to strain and confinement), selection rules yield that the electron spin orientation determines the polarization of the emitted photons. This is only valid if we observe recombination of an electron which has been polarized previously in the spin aligner. If residual electrons are present in the quantum dot, this needs to be taken into account.

The samples were grown by combined III-V/II-VI molecular-beam epitaxy, details are provided elsewhere [2, 3]. For the investigations presented here, we discuss four different samples, which are similar except that the quantum dots exhibit different sizes and compositions (for samples A and B) and different doping of the spin aligner layer (C and C*), respectively.
Table 1. Sample overview.

| Sample | InAs | QD Density | QD peak emission energy | spin aligner doping |
|--------|------|------------|-------------------------|---------------------|
| A      | 2.25 ML | 2.5 \( \cdot \) \( 10^{11} \) \( cm^{-2} \) | 1.2 eV | \( 1 \cdot 10^{18} \) \( cm^{-3} \) |
| B      | 1.85 ML | 1.2 \( \cdot \) \( 10^{10} \) \( cm^{-2} \) | 1.3 eV | \( 1 \cdot 10^{18} \) \( cm^{-3} \) |
| C      | 1.85 ML | 1.2 \( \cdot \) \( 10^{10} \) \( cm^{-2} \) | 1.3 eV | \( 1 \cdot 10^{18} \) \( cm^{-3} \) |
| C*     | 1.85 ML | 1.2 \( \cdot \) \( 10^{10} \) \( cm^{-2} \) | 1.3 eV | \( 2 \cdot 10^{17} \) \( cm^{-3} \) |

p-i-n diode structures were grown on p-doped GaAs substrates. Below the quantum dots, a 100 nm thick undoped GaAs layer was deposited. On top of the QD layer, a 50 nm spacer layer was grown. This is needed to separate the QD spins from the magnetic influence of the Mn-containing layer. Then, a thick (750 nm) ZnMnSe layer and a higher doped ZnSe contact layer are deposited on top in a second MBE chamber. Here, for samples C and C*, pieces of the same III-V heterostructure were overgrown with differently doped ZnMnSe:Cl. Table 1 summarizes the basic parameters of the spin-LEDs.

Figure 1 shows the low-temperature photoluminescence of the samples A and B for optical excitation into the barrier. These spectra as well as Indium distribution profiles obtained by CELFA (composition evaluation by lattice fringe analysis, [4, 5]) evaluation of high-resolution transmission electron microscopy [6] were taken as input parameters for extensive numerical calculations of the quantum-dot states. We find that excited states, where the wave functions are confined in the quantum dot, exist for all quantum dots with a ground state transition energy below roughly 1.35 eV.

For electroluminescence measurements, the samples are mounted in Faraday geometry in a magneto-cryostat with broadband polarization-selective detection. If not otherwise mentioned, all data given here are taken at \( T = 5 \) K. In Fig. 2, the degree of circular polarization of the emitted photons is shown for some external magnetic field strengths. The CPD is determined from the intensities of the \( \sigma^+ \) and \( \sigma^- \) [7] polarized luminescence, \( I_+ \) and \( I_- \), respectively, using \( \text{CPD} = (I_+ - I_-)/(I_+ + I_-) \). A general trend is the increase of the CPD with higher QD emission energy. We observe this trend over a large set of samples with different quantum-dot morphology.
In all-optical measurements the latter effect has been attributed to excited-state luminescence due to Pauli-blocking of the ground state [8, 9]. By injecting electrons with 50% polarization (excited by polarized light into the GaAs barrier), a higher degree of polarization from the excited state was observed than from the ground state since due to Pauli-blocking the ground state can only be populated with electrons with anti-parallel spin. Therefore, at matching carrier densities, the additional electron in the excited state leads to higher optical polarization. In our studies, this effect seems to play a minor role, but needs to be considered as our ensemble luminescence could partially come from excited states. We have measured the CPD of the three samples while increasing the driving current by several orders of magnitude and see no increase of the CPD on the high-energy side of the QD ensemble. In Fig. 3 this is exemplarily shown for sample A. Additionally, the strong robustness of the spin injection is obvious from these measurements as the polarization degree does not significantly decrease.

Figure 2. Circular polarization degree of samples A and B shown as a function of the emission energy for various magnetic field strengths.

Figure 3. CPD spectra for sample B for different driving currents. The current is given as electrons per quantum dot per nanosecond as estimated from the QD sheet density, not taking into account non-radiative recombination.

An important prerequisite for the photons emitted from the quantum dots having a high circular polarization degree is that only electrons injected via the spin aligner recombine in the dots. If residual electrons are present in the quantum dots, their spin can determine the photon polarization. In single quantum dot experiments we found that the g-factor in the quantum dots is of opposite sign than in the spin aligner. That means the electrons in the quantum dot are injected into the upper state of the ground-state doublet [6]. But since spin-relaxation within the doublet is inefficient, only long living residual electrons can populate the lower spin state.
and can thus lead to a reduced optical polarization. Presence of residual electrons in the QDs should be influenced by the electronic structure of the device. To investigate the influence of the spin-aligner doping, we fabricated a sample with reduced carrier density of this layer. A comparison of the CPD for samples with differently doped spin aligner layers is shown in Fig. 4. A strong increase of the CPD at the low-energy side of the quantum-dot ensemble is clearly visible.

**Figure 4.** CPD spectra for samples incorporating the same type of quantum dots as sample B with two different doping levels of the ZnMnSe spin aligner.

![Figure 4](image1.png)

We also find a much faster build-up and saturation of the CPD with increasing magnetic field for the sample with smaller doping of the ZnMnSe layer. This is shown in Fig. 5, where the CPD has been calculated from the spectrally integrated $\sigma^+$ and $\sigma^-$ polarized luminescence intensities.

**Figure 5.** Integrated circular polarization degree of samples C and C* as a function of the magnetic field.

![Figure 5](image2.png)

In conclusion, we have shown that electrical spin injection into quantum dots can be achieved with high efficiency. But a high and homogeneous initialization of electron spin in an ensemble of quantum dots is still challenging and depends on various sample parameters. One major issue seems to be the presence of residual electrons in the dots. First results show that a careful adjustment of sample growth, especially reducing the spin aligner dopant concentration leads to significant improvement of the polarization degrees of the photons emitted from the spin-LEDs.

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