Spin-polarization dynamics in electrically excited single InGaAs quantum dots

Pablo Asshoff, Michael Hetterich, Jochen Zimmer, Heiko Füser, Wolfgang Löffler and Heinz Kalt
Institut für Angewandte Physik, Universität Karlsruhe (TH) and DFG Center for Functional Nanostructures (CFN), 76131 Karlsruhe, Germany
E-mail: pablo.asshoff@physik.uni-karlsruhe.de

Abstract. Potential applications of semiconductor quantum dots include spin memory and quantum information processing devices. For these, a rapid spin-initialization process would be desirable. In this contribution, we demonstrate nanosecond-pulsed electrical spin-injection into quantum dots embedded in a spin light-emitting diode. As inferred from time-resolved electroluminescence experiments, the circular polarization degree of the emitted light exhibits a peak at the beginning of the electroluminescence signal, indicating that the spin-initialization is more efficient in pulsed operation than under constant-current excitation. Results for the quantum-dot ensemble, the wetting layer states, and for single quantum dots are presented. We attribute these observations to energy-dependent spin-relaxation processes in the device.

1. Introduction
A reliable initialization of electron spins in semiconductor quantum dots (QDs) constitutes a sine qua non to realize a semiconductor-based spin quantum information processing [1]. For QDs embedded in spin light-emitting diodes (spin-LEDs), spin-initialization with near-unity fidelity was achieved by constant-current electrical pumping [2, 3]. However, to integrate these spin-based devices into high-frequency electronics, the spin-initialization process has to be accomplished using short electrical pulses. Besides, pulsed electrical operation of spintronic devices is essential for other next-generation applications, e.g. spin memory devices [4]. Hence, we explored the characteristics of initializing electron spins in QDs embedded in spin-LEDs by nanosecond electrical pulses. Time-resolved electroluminescence (TREL) experiments show that at the beginning of the light emission a high circular polarization degree (CPD) occurs.

2. The spin-LED structure
For the experiments, a $p-i-n$ diode with InGaAs quantum dots (QDs) is employed. The semimagnetic $n$-type ZnMnSe acts as a spin-aligner for electrons, caused by the giant Zeeman splitting of the conduction band in an external magnetic field [5]. An overview of the layer sequence is depicted on the left panel of Fig. 1.

The spin-LED was produced in two molecular-beam epitaxy (MBE) facilities. In the first, the III–V part of the structure was fabricated. On a GaAs:Zn(001) substrate ($p \sim 1 \times 10^{19}$ cm$^{-3}$), $\sim$500 nm GaAs:Be ($p \sim 1 \times 10^{19}$ cm$^{-3}$) and 100 nm $i$-GaAs were grown. Then, we deposited 0.6 nm InAs (0.0057 ML/s growth rate), followed by a growth interruption of 10 s.
Due to the Stranski-Krastanov growth mode this results in the formation of InGaAs QDs and a corresponding wetting layer (WL) [6]. A QD sheet density of $5 \times 10^{10} \text{ cm}^{-2}$ was determined by plan-view transmission electron microscopy. On top of the QDs, a 25 nm thick $i$-GaAs spacer was deposited. In the second MBE facility, the II–VI part consisting of 750 nm of the spin aligner material $\text{Zn}_{0.95}\text{Mn}_{0.05}\text{Se:Cl}$ ($n \sim 10^{18} \text{ cm}^{-3}$) and a 200 nm layer of $\text{ZnSe:Cl}$ ($n = 5 \times 10^{18} \text{ cm}^{-3}$) were grown. The ZnSe:Cl layer improves the quality of the In contact which is finally evaporated. Subsequently, the lateral extension of the spin-LED was reduced using photolithography. Moreover, gold apertures were fabricated on parts of the surface with electron beam lithography, allowing an easy identification of the emission lines from single quantum dots [2, 3]. The remaining surface was left uncovered for QD ensemble measurements.

3. Setup for time-resolved electroluminescence experiments
The experimental setup for the time-resolved electroluminescence measurements is sketched in Fig. 1 (right). The spin-LED is excited with electrical pulses of 125 ns width, 5 ns rise and fall time, a repetition frequency of $f = 4 \text{ MHz}$ ($T = 250 \text{ ns}$) and a peak voltage of 4.4 V by an electrical pulser. A coaxial cable designed for low-temperature environments transmits the signal to the spin-LED located in a magneto-optical cryostat, where a 50-Ω resistor is connected in parallel for impedance matching. The light emitted from the spin-LED is filtered by a quarter-wave plate and a linear polarizer, allowing only light corresponding to one spin orientation to pass, i.e., to distinguish between $\sigma^+$- and $\sigma^-$-polarized light. After traveling through this filter setup, a fiber guides the light pulse to a spectrometer, where the wavelength of interest is selected. The photons then impinge on an avalanche photodiode (APD), which converts the light signal to an electrical pulse. A time-correlated single-photon counting module (TCSPCM) records the time difference between the APD signal and the trigger signal from the electrical pulser. The data is transmitted to a computer, which maps the events. Thereby, the time-dependent circular polarization degree (CPD) can be determined, defined as $(I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$, where $I_{\sigma^+(-)}$ denotes the intensity of $\sigma^+(-)$-polarized light.

4. Ensemble dynamics
With the TREL setup, we pumped the sample with electrical pulses and studied the ensemble spin-polarization dynamics for several wavelengths. The electroluminescence (EL) spectrum is shown on the left panel of Fig. 2, the time-dependent CPD corresponding to five different wavelengths is depicted on the right panel of Fig. 2.
Figure 2. Left panel: EL spectrum of the spin-LED at $T = 5$ K. The GaAs-related emission is centered at 840 nm. The wetting layer emission exhibits a peak at 885 nm, the QD emission can be observed at higher wavelengths. Right panel: Time-dependent CPD for several wavelengths at time $t$ after the beginning of the EL signal ($T = 5$ K, $B = 6$ T). Lower curves represent higher wavelengths. The data points are obtained through ten-point averaging over the original data and are plotted for statistically significant intervals only. Solid lines are exponential fits. The CPD exhibits an initial peak and the subsequent relative decrease is wavelength-dependent.

| $\lambda$ [nm] | CPD($t_0$) | CPD($t_E$) | $\Delta$CPD | $\tau$ [ns] |
|---------------|------------|------------|-------------|-------------|
| 870           | 0.41       | 0.38       | -7%         | 13.6        |
| 880           | 0.40       | 0.32       | -20%        | 9.9         |
| 890           | 0.35       | 0.22       | -37%        | 8.3         |
| 900           | 0.32       | 0.17       | -47%        | 7.6         |
| 930           | 0.24       | 0.12       | -50%        | 7.8         |

Table 1. Parameters obtained from the exponential fits to the data point distributions in Fig. 2 for each wavelength $\lambda$. The initial and the final points of the fitted curves are denoted by CPD($t_0$) and CPD($t_E$), respectively. $\Delta$CPD represents the relative change of the CPD and $\tau$ the time when the CPD dropped to $1/e$ of its initial value.

A distinctive decrease of the CPD during the first nanoseconds can be observed. Then, a constant plateau value is reached. Decaying exponential functions were utilized to approximate the data point distribution. The parameters obtained from the fits are displayed in Table 1. Comparing the time-resolved CPD for the light pulses associated with the WL and QD states, it is apparent that the CPD drop is higher for the QD emission. The relative drop increases monotonically from -7% for 870 nm to -50% for 930 nm. The characteristic time $\tau$, denoting the instant when the CPD value dropped to $1/e$ of the initial value, also exhibits a trend. It decreases from 13.6 ns at 870 nm to 7.8 ns at 930 nm.

The result that the plateau value of the CPD is higher for lower wavelengths is consistent with observations reported in Ref. [3, 7] using constant current excitation, where mechanisms responsible for this effect are discussed. For the initial CPD peak, the most straightforward explanation would be the fact that the CPD depends on current [7]. However, simulations and measurements showed that when the voltage pulse is applied across the spin-LED, the initial current flowing through it is high. This should result in an initial decrease of the CPD rather than the increase found experimentally.
We ascribe the CPD peak at the beginning of the pulse to spin-relaxation mechanisms, which the experiments prove to be energy-dependent. First, spin-loss channels during spin-transport to the target states (WL, QDs) should play a role. Initially, the QD state population is low and spin-injection quite efficient. Later on, however, part of the dot states are always filled. Therefore, electron injection is hampered and slows down, thus giving more opportunity for spin scattering outside the QDs and resulting in a lower CPD. Another point to consider are spin-flip processes inside the dots, although there is experimental evidence that spin scattering outside the QDs should be the dominant effect for steady-state operation [3, 7]. Assuming longer spin relaxation times for the holes than for the electrons of quantum-dot excitons [8], it can easily be shown by numerical simulations that at the beginning of the pulse a higher CPD is expected when polarized electrons and unpolarized holes are injected.

5. Spin-polarization dynamics of single quantum dots
While the ensemble measurements provide aggregated information, the analysis of the spin-dynamics in single QDs opens up the possibility to study single electron spins. Utilizing the gold apertures on top of the spin-LEDs, we measured the temporal evolution of the CPD for several individual QD emission lines. Due to the faint emission from a single QD and the high temporal resolution of the setup, these experiments required long integration times.

Some of the results strongly resemble those of the ensemble measurements described before, i.e., a CPD peak at the onset of the EL with a subsequent plateau value is found. However, in some cases we also observed an initial minimum of the CPD, followed by a higher constant plateau value. In the ensemble dynamics, these differences appear to cancel out. Moreover, values for the characteristic time $\tau$ are similar to the ensemble measurements. A closer analysis of the processes involved and experiments on shorter timescales are currently performed [9].

6. Conclusion
We presented time-resolved electroluminescence measurements of quantum-dot spin-LEDs excited with short electrical pulses. Results for the quantum-dot ensemble, the wetting layer states, and single quantum dots were shown. For all of these, the CPD exhibits a strong time-dependence during the first nanoseconds. These properties are attributed to energy-dependent spin-scattering processes.

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References
[1] DiVincenzo D P 2000 Fortschritte der Physik 48 771
[2] Löffler W, Hetterich M, Mauser C, Li S, Passow T and Kalt H 2007 Appl. Phys. Lett. 90 232105
[3] Hetterich M, Löffler W, Asshoff P, Passow T, Litvinov D, Gerthsen D and Kalt H 2009 Electrical spin injection into single InGaAs quantum dots Advances in Solid State Physics vol 48 ed Haug R (Springer) p 103
[4] Awschalom D D and Flatté M E 2007 Nature Physics 3 153
[5] Furdyna J K 1988 J. Appl. Phys. 64 R29
[6] Marzin J Y, Gérard J M, Izrael A, Barrier D and Bastard G 1994 Phys. Rev. Lett. 73 716
[7] Hetterich M, Löffler W, Fallert J, Hörncke N, Burger H, Passow T, Li S, Daniel B, Ramadout B, Lupacchomer J, Hetterich J, Litvinov D, Gerthsen D, Klingshirn C and Kalt H 2006 phys. stat. sol. (b) 243 3812–3824
[8] Tsitsishvili E, Baltz R v and Kalt H 2005 Phys. Rev. B 72 155333
[9] Asshoff P, Zimmer J, Füser H, Merz A, Hetterich M and Kalt H unpublished