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Electrical spin injection into InGaAs quantum dots: single dot devices and time-resolved studies

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In the context of a potential future quantum information processing we investigate the concurrent initialization of electronic spin states in InGaAs quantum dots (QDs) via electrical injection from ZnMn(S)Se spin aligners. Single dots can be read out optically through metallic apertures on top of our spin-injection light-emitting diodes (spin-LEDs). A reproducible spin polarization degree close to 100% is observed for a subset of the QD ensemble. However, the average polarization degree is lower and drops with increasing QD emission wavelength. Our measurements suggest that spin relaxation processes outside the QDs, related to the energetic position of the electron quasi-Fermi level, as well as defect-related spin scattering at the III–V/II–VI interface should be responsible for this effect, leading us to an improved device design. Finally, we present first time-resolved electroluminescence measurements of the polarization dynamics using nanosecond-pulsed electrical excitation. The latter should enable us to gain a more detailed understanding of the spin relaxation processes in our devices. They are also the first step towards future time-resolved spin manipulation experiments.

1 Introduction In recent years, the idea of a spin-based quantum information processing has attracted considerable attention [1]. Some of the prerequisites for the realization of the latter are the ability to spin-polarize electrons, store them at defined sites and address them individually, e.g., to read out the spin state. In this contribution, we investigate electrical spin injection from semimagnetic *n*-type ZnMn(S)Se spin aligners [2] into III–V *p*-*i*-*n* diodes containing InGaAs/GaAs quantum dots (QDs). As shown in Sect. 2, the advantage of this concept is that spin states in different dots can be initialized *simultaneously* with high fidelity (which would be more difficult to achieve utilizing non-resonant optical excitation of the spectrally broad QD ensemble due to the excitonic relaxation processes involved). On the other hand, individual spin states in *single* QDs can be accessed optically through metallic micro- or nano-apertures on top of our devices. The spin loss mechanisms in the realized spin-injection light-emitting diodes (spin-LEDs) as well as possible design optimizations are discussed in the second part of Sect. 2. Finally, we present first time-resolved electroluminescence (EL) measurements of the polarization dynamics using nanosecond-pulsed electrical excitation in Sect. 3. The latter should not only enable us to better understand the spin and carrier relaxation processes in our devices, they are also the first step towards future time-resolved spin manipulation experiments.

2 Single quantum-dot spin-LEDs The basic layer sequence of the devices investigated as well as the corresponding band structure for typical operating conditions is shown in Fig. 1 (see [3, 4] for further details). Electrons supplied through an In contact and an *n*-type ZnSe:Cl layer (not shown) are injected into a dilute magnetic *n*-$Zn_{0.95}Mn_{0.05}S_zSe_{1-z}$:Cl spin aligner. In an externally applied magnetic field B and at low temperatures the conduction band of the latter shows a giant Zeeman splitting, resulting from the strong (s, p)–d exchange interaction of the charge

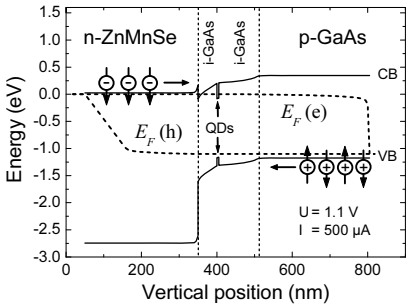

Figure 1 Spin-LED band structure for typical operating conditions.

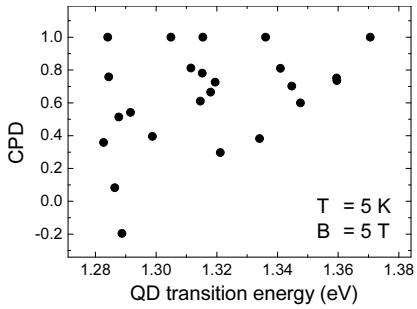

Figure 3 Circular polarization degree (*CPD*) of the electroluminescence for different QDs in the same spin-LED.

carriers with the half-filled *d*-shell of the Mn atoms. As a result, the electrons fed into the spin aligner quickly thermalize into the lower $m_s = -1/2$ spin state. The obtained spin-polarized carriers are then injected into InGaAs/GaAs QDs. The sign of the *g* factor in the dots is opposite to that in ZnMn(S)Se, i.e., the electrons are injected into the upper spin state [3]. To optically read out the spin orientation, unpolarized holes are fed into the QDs from the bottom *p*-GaAs layer. Due to the strain- and quantization-induced heavy-hole/light-hole splitting only the $m_J = \pm 3/2$ heavy-hole QD states are populated and lead to optical transitions. Electrons with spin polarization −1/2 (+1/2) can only recombine with −3/2 (+3/2) holes, emitting circularly polarized σ^+ (σ^-) photons in Faraday geometry. Therefore, the circular polarization degree

$$CPD = \frac{I_{\sigma^+} - I_{\sigma^-}}{I_{\sigma^+} + I_{\sigma^-}} \qquad (I_{\sigma^\pm}: \text{polarized EL intensities})$$

of the emitted light directly indicates the sign and degree of electron spin polarization in the dots.

Figure 2 shows micro-EL spectra of a single QD in a spin-LED, measured through a gold aperture for different applied magnetic fields. At $B = 0$ T, a sharp emission peak (line width resolution-limited) with no circular polarization is observed. For non-zero magnetic fields, the Zeeman splitting of the QD transition can be observed. With increasing *B* the electrons injected into the dot become more and more spin-polarized. As a result, the σ^+ transition, corresponding to the injected spin-down electrons, grows, while the spin-up related σ^- peak drops strongly. Finally, at about $B = 7$ T, the σ^- emission nearly disappears. This clearly indicates that electronic spin states in QDs can be initialized with high fidelity using electrical spin injection. Furthermore, the robustness of the spin states involved becomes evident, because spin polarization is conserved during the whole process of electrical injection, transport, storage in QDs, and final optical readout. In particular, no significant spin relaxation takes place in the dots, although the electrons are injected into the upper Zeeman level.

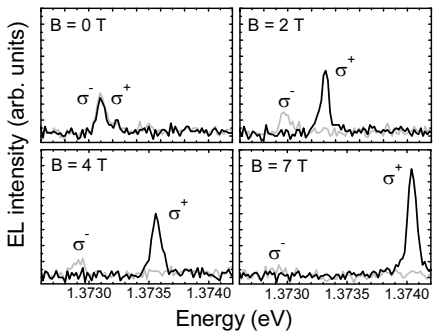

Figure 2 *B*-dependent µ-EL spectra of a single QD spin-LED ($I = 4$ mA, $U = 2.1$ V, $T = 5$ K). For each field value, the σ^+ and σ^- component of the QD emission have been measured.

Repeating the described measurements for different QDs in the same device, we could demonstrate the *simultaneous* initialization of spin states with fidelity close to 100% in at least five dots [5]. However, as shown in Fig. 3, the achieved *CPD* generally varies strongly from one QD to another (although the values obtained for a given dot are reproducible), i.e., a *local* loss of spin polarization occurs. It is tempting to ascribe this variation to structural differences between the QDs that might influence the spin relaxation time. On the other hand, all-optical measurements suggest no significant spin relaxation during exciton lifetime in QDs [6], i.e., spin polarization should be essentially preserved in the dot, even if a hole is injected to read out the spin state. This expected robustness is also confirmed by the measurements shown in Fig. 2. Indeed, we have strong experimental evidence that spin polarization must already be lost *before* the electrons are captured in the dots (for details see [3] and following discussion).

An obvious source for local spin relaxation processes outside the QDs is dislocations at the III–V/II–VI interface [7]. For that reason we have optimized the Mn concentration in our $Zn_{1-x}Mn_xSe$ spin aligners in order to obtain a low density of stacking faults/misfit dislocations and other defects while preserving a sufficient Zeeman splitting [3, 8–10]. However, despite the good quality achieved [3, 4], misfit dislocations due to the lattice mismatch between ZnMnSe and GaAs cannot be avoided. Therefore, we have recently started to develop *lattice-matched* $Zn_{1-x}Mn_xS_zSe_{1-z}$ spin aligners. First results of these investigations are shown in Fig. 4, where QD *ensemble* measurements of two spin-LEDs are compared. The devices are identical apart from the spin-aligner material used (750 nm $Zn_{0.95}Mn_{0.05}Se$ and

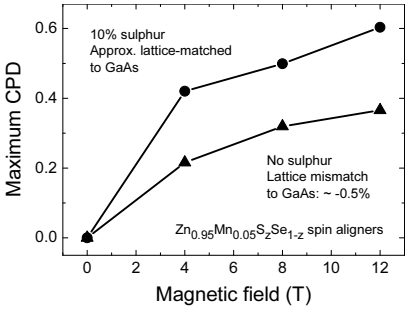

Figure 4 Magnetic-field dependent *CPD* of the QD ensemble EL ($T = 5$ K) for two spin-LEDs. The devices are identical apart from the spin-aligner material used. The *CPD* has been measured at its spectral maximum (at the short-wavelength end of the ensemble emission).

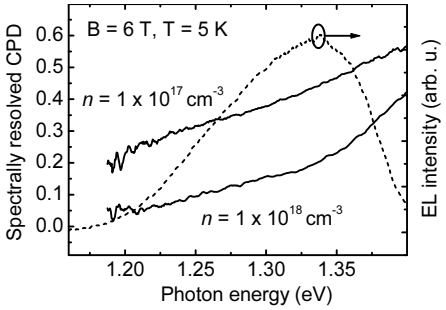

Figure 5 Spectrally resolved *CPD* for two spin-LEDs with identical structure but different spin aligner doping levels. The EL spectrum (dashed line) is the same for both devices.

$Zn_{0.95}Mn_{0.05}S_{0.1}Se_{0.9}$, respectively). As can clearly be seen, the utilization of a ZnMnSSe spin-aligner layer approximately lattice-matched to GaAs leads to a significant *CPD* improvement. Based on these preliminary results, the development of lattice-matched spin aligners with higher Mn concentrations seems feasible as well. The latter would provide a larger Zeeman splitting and thus enable operation of the devices at lower magnetic fields and/or higher temperatures.

As can be seen in Fig. 3, the achieved *average CPD* depends strongly on QD transition energy. This effect can be traced back to some peculiarities in the band structure of our devices (see [3, 5, 11] for a detailed discussion of this aspect). Essentially, as shown in Fig. 1, the band bending and the position of the electronic quasi-Fermi level E_F (e) give rise to the formation of a two-dimensional electron gas (2DEG) at the III–V/II–VI interface. Furthermore, the injected spin-polarized electrons have to tunnel through a potential barrier in order to reach the wetting layer (WL) and the QDs. Since the average effective barrier is larger for low-energy dots, tunnelling will be slower, thus giving more opportunity for spin relaxation, i.e., the *CPD* should drop for these QDs, as observed experimentally. However, in reality, the problem is more complex. Both the effective tunnel barrier and E_F(e) fluctuate locally, e.g., due to spatial variations in the properties of the spin aligner (Mn mole fraction, defects, doping) as well as the QDs (density, morphology, and composition, all modifying the density of states) but also due to the influence of nearby impurities and defects. As a result, the *CPD* achieved should depend on the local environment of the individual dot under consideration. In particular, QDs with high spin polarization would be expected to be found in the whole spectral emission range of the spin-LEDs, although the probability to obtain efficient injection should drop with decreasing transition energy, which is indeed observed experimentally (Fig. 3). If the outlined model is correct, spin relaxation should essentially occur in the GaAs spacer layer between the spin aligner and the QDs (see Fig. 1) as a result of only slow tunnelling into the dots. Indeed, this hypothesis could be confirmed by a number of experimental results [3]. For instance, the *CPD* drops strongly with increasing spacer thickness as a consequence of the larger tunnel barrier. Furthermore, temperature-dependent measurements reveal a *CPD* improvement in a certain spectral range, when the device is heated up from $T = 5$ K to values ≤ 30 K. This can be attributed to phonon-assisted tunnelling and motional narrowing processes (typical for the D'yakonov-Perel' mechanism). Therefore, mobile states (in the GaAs spacer but also the WL) should indeed play a major role in the spin relaxation process, consistent with the outlined model.

Based on these results, the obvious strategy to improve our devices must be to shift the electron Fermi level to lower energies in order to prevent the formation of a 2DEG at the III–V/II–VI interface and the injection of poorly polarized electrons into the QDs. One possibility to achieve this is to raise the effective QD density of states via an increased dot density. In fact, preliminary experiments seem to indicate an improved injection efficiency when this approach is used. Alternatively, the spin aligner doping can be reduced to lower the electron Fermi level and potentially avoid the formation of a 2DEG altogether. Figure 5 compares the spectrally resolved *CPD* of two spin-LEDs with identical structure but different spin aligner doping levels (~1×10^{17} cm^{-3} and ~1×10^{18} cm^{-3}, respectively). As can be seen, the predicted improvement in spin polarization is indeed observed, in particular for low QD transition energies [12]. An additional advantage of the lower Fermi level in the spin aligner is the fact, that a smaller Zeeman splitting in the latter suffices to prevent population of the upper conduction band state, i.e., to enable efficient spin polarization. As a result, high *CPD* values can already be achieved for relatively weak magnetic fields [12] or for increased temperatures.

3 Time-resolved measurements In order to gain a more detailed understanding of the spin relaxation processes in our devices but also as a first step towards future spin manipulation experiments we have recently started to perform time-resolved EL studies of the polarization dynamics using pulsed (~20 ns) electrical excitation and time-correlated single photon counting. First results of these measurements are shown in Fig. 6. Surprisingly, the

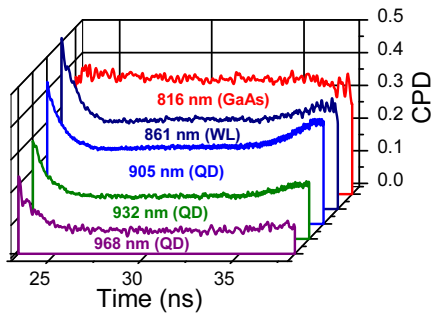

Figure 6 Time-resolved *CPD*, measured at $B = 6$ T and for different emission wavelengths of the same device, corresponding to the spectral range of GaAs, the WL, and the QDs, respectively.

determined *CPD* is not constant but shows a pronounced maximum at the rising edge of the pulse (to a lesser degree also at the falling edge), while the polarization degrees found in the plateau region of the traces essentially agree with those obtained in continuous current measurements. The observed feature only appears for confined states (QDs, WL), but not for the GaAs emission, thus excluding experimental artefacts like, e.g., delay effects in the measurement of the differently polarized EL components. As shown in previous work, the *CPD* drops with increasing current through the spin-LED [3]. However, although there is certainly a connection between this effect and the described phenomena, the initially lower current through the device alone cannot explain the observed features, because the *CPD* peak at the end of the pulse is much less pronounced for equally low currents. Moreover, the current dependence of the polarization degree is too weak to fully account for the increased *CPD*. Currently we assume that many-carrier and non-equilibrium effects in the device should play an important role in the explanation of our findings. Initially, at the rising edge of the pulse, the carrier density in the spin-LED is still low. That means, the spin-polarized electrons can easily tunnel into the empty QD states and thus retain their polarization. However, later on, in dynamic equilibrium, a certain fraction of the dots will always be filled by electrons. Therefore, tunnelling will only be possible when an empty target state becomes available, i.e., after injection of a hole into an occupied dot and subsequent optical recombination. The resulting retardation should give more opportunity for spin relaxation in the GaAs spacer, which might explain the lower *CPD* observed under steady-state conditions. The GaAs luminescence would hardly be affected by this mechanism, because a high number of empty states are always available, and tunnelling processes are less important for the majority of electrons contributing to this emission.

Although the described effects may provide a partial explanation for our measurements, it should be emphasized, that further detailed investigations are required and currently under way in order to reach a better understanding of the observed phenomena.

4 Conclusions We demonstrated the concurrent initialization of electronic spin states in InGaAs QDs with near 100% fidelity based on electrical injection from a ZnMn(S)Se spin aligner. Individual QD states could be read out through metallic apertures. Based on our results suggesting that spin relaxation mainly occurs outside the QDs, we realized optimized devices with improved electron quasi-Fermi level and lattice-matched spin aligners. Finally, we presented first time-resolved EL measurements of the polarization dynamics using ns-pulsed electrical excitation. The latter should enable us to gain a more detailed understanding of the spin relaxation processes in our spin-LEDs. Furthermore, they are the first step towards future time-resolved spin manipulation experiments.

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