A spintronic source of circularly polarized single photons

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We present a spintronic single photon source which emits circularly polarized light, where the helicity is determined by an applied magnetic field. Photons are emitted from an InGaAs quantum dot inside an electrically operated spin light-emitting diode, which comprises the diluted magnetic semiconductor ZnMnSe. The circular polarization degree of the emitted light is high, reaching 83% at an applied magnetic field of 2 T and 96% at 6 T. Autocorrelation traces recorded in pulsed operation mode prove the emitted light to be antibunched. The two circular polarization states could be used for representing quantum states |0> and |1> in quantum cryptography implementations.

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The most established materials for enabling spin-polarization of electronic carriers are ferromagnets, half-metals and magnetic semiconductors [1]. Excellent spin-injection efficiencies into adjacent semiconductor structures have been achieved with magnetic semiconductors [2,3], resulting from their similar physical properties and the high interface quality. Due to this outstanding performance, it makes magnetic semiconductors a material of choice for designing an electrically operated light source for photons with defined helicity. A spin-polarized electrical current created by the magnetic semiconductor and injected into a quantum dot (QD) will result in emission of circularly polarized light, assuming the ideal case that the spin state of the carriers is preserved until optical recombination [4]. Furthermore, in order to generate single photons on demand, when a voltage pulse is applied to the device, the light emitted will need to be antibunched [5]. A classical structure for the implementation of these design prerequisites is a spin light-emitting diode (spin-LED), which usually serves as test device for determining the spin-polarization degree of injected carriers. However, as a single photon source with σ+ or σ−-polarized emission, it could be employed for securely coding and transmitting data for applications in quantum cryptography. In this paper we will show that such a spin-LED can be realized, which emits circularly polarized light exhibiting very high polarization degrees, that the helicity of the light can be controlled by the direction of an external magnetic field, and that by applying voltage pulses antibunched light is generated.

The basic design layout of the specific sample presented is sketched in Fig. 1, where the most important functional elements are marked in color. The single photon source is a single QD emitting circularly polarized photons at ~1.348 eV, which is located in the optically active region (orange layer) of a spin-LED with a surface area of (400 μm)². The diluted magnetic semiconductor (DMS) ZnMnSe is integrated into the heterostructure (blue layer), it renders traversing electrons spin-polarized when a magnetic field is applied. This is due to electrons relaxing into the energetically lower of the two spin-split conduction bands, which are separated through a giant Zeeman splitting [6]. Thus, applying a voltage across the spin-LED results in spin-polarized electrons reaching the QD, where they recombine with unpolarized holes injected from the bottom part of the spin-LED. Due to optical selection rules, these transitions can only take place under the emission of circularly polarized light. A submicron aperture on top of the heterostructure (within the yellow top layer) helps to minimize spurious emission from other nearby QDs.

Fabrication and processing of the sample were carried out as follows. A GaAs:Zn(001) wafer (ρ ~ 1 × 10¹⁹ cm⁻³) was used as substrate. On the substrate a ~500 nm layer of GaAs:Be (ρ ~ 1 × 10¹⁹ cm⁻³) was grown using a III–V molecular-beam epitaxy (MBE) facility, followed by 100 nm i-GaAs, the InGaAs QDs / wetting layer (WL) (see Ref. [7] for details) and a 25 nm thick i-GaAs spacer. Plan-view transmission electron microscopy revealed a QD sheet density of ~5 × 10¹⁰ cm⁻².

From optical characterization we found that the QDs

![FIG. 1: Schematic of the spintronic single photon emitting diode. InGaAs QDs are sketched in the optically active region and an aperture on top of the heterostructure is shown.](image-url)
have a negligible in-plane asymmetry. The heterostructure was then transferred to a second MBE facility, designed for the growth of II–VI materials, where 750 nm of the DMS Zn$_{0.95}$Mn$_{0.05}$Se:Cl ($n \sim 10^{18}$ cm$^{-3}$) followed by a 200 nm layer of ZnSe:Cl ($n = 5 \times 10^{18}$ cm$^{-3}$) were deposited. The latter layer improves the ohmic contact to the subsequently evaporated In contact pad. Then, a thin gold layer was thermally evaporated wherein apertures were defined by electron beam lithography. Finally, optical lithography was employed to obtain square-shaped spin-LEDs.

We now analyze our device optically. First, we show that the QD emission is spectrally isolated and determine its polarization degree for varying magnetic fields. As measure of the photon polarization state we define the circular polarization degree $P_C = (I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$, with $I_{\sigma^+(-)}$ denoting the intensity of $\sigma^+$(-)-polarized light. $P_C$ has been shown to correspond to the spin-polarization degree of the injected electrons, since when electrons and holes recombine in the QD, only heavy hole states contribute to optical transitions [2]. Experiments were carried out in a magneto-optical cryostat with the sample temperature set to $T = 5$ K. The magnetic field was applied in Faraday geometry ($k \parallel B$) and swept from -6 T to +6 T. A 60× microscope objective mounted inside the cryostat allowed to guide light from one aperture of the device, positioned by piezoelectric actuators at the focal point, to the outside of the cryostat. To differentiate between $\sigma^+$- and $\sigma^-$-polarized photons, a quarter-wave plate and a linear polarizer were inserted in the beam path. This array was arranged such that either $\sigma^+$- or $\sigma^-$-polarized photons were remaining after having passed the filters. Light with intensity $I_{\sigma^+/-}^\text{exc}$ was then focused into a multi-mode fiber and transmitted to a double spectrometer with two 1200 grooves/mm gratings, where it was spectrally dispersed. A charge-coupled device was used to detect the diffracted light. Using this setup, a spectral resolution in the order of 20 μeV per pixel was achieved.

In Fig. 2 the obtained electroluminescence spectra of the selected QD during continuous excitation with a current of 0.29 mA are shown. Selectively recorded $\sigma^+$- and $\sigma^-$-polarized spectral components were integrated in this graph. The degeneracy of the spin-up and spin-down sublevels in the QD is lifted when a magnetic field is applied (Zeeman splitting), shifting the $\sigma^+/\sigma^-$-polarized excitonic emission from the spin-down/spin-up conduction band sublevel to higher/lower energies. The spectral position of the emission from the spin sublevels ($\uparrow, \downarrow$) shifts according to $E_{\sigma^\pm} = E_0 \mp \frac{1}{2} g_{\text{exc}} \mu_B B + \gamma B^2$ with $g_{\text{exc}} = 2.22$, where the Zeeman splitting and the diamagnetic shift are considered. The upper spin subband is filled with spin-down electrons from the DMS, which occupy a higher energy state in the QD [4]. Therefore, spin relaxation within the QD would populate the lower spin sublevel. The spectra show that the net spin-polarization $P_C$ increases strongly (corresponding to an increase of the normalized value of $I_{\sigma^+} - I_{\sigma^-}$) along with magnetic field to high values. At high magnetic fields, it can be seen that the emission is originating preferably from the upper spin subband and the minority component vanishes almost completely at $B = +6$ T. As the upper spin sublevel is populated, it is clear that spin-polarization originates from the DMS [3]. Significant circular polarization degrees $P_C$ are already reached at much lower

![FIG. 2: Spectrally resolved emission from the QD single photon source at $T = 5$ K and $B \geq 0$ T during excitation with a continuous current of 0.29 mA. As the magnetic field is increased, emission splits into two components from the spin-up and spin-down levels, which are indicated by the markers.](image)

![FIG. 3: Main characteristics of the spintronic single photon source ($T = 5$ K): (a) The average circular polarization degree $P_C$ of the emitted photons from the QD (with a continuous current of 0.29 mA) reaches high values. Typical error bars obtained from repeated measurements are indicated. (b) Autocorrelation measurement of $\sigma^+$-polarized photons emitted from the QD at an applied field of $+2$ T. The sample is excited with sub-nanosecond pulses from a fast electrical pulser. From the zero delay peak area we obtain a value of $\gamma(0) = 0.38$ for the correlation signal, giving evidence of single photon emission.](image)
fields. The corresponding circular polarization degrees are shown in Fig. 3(a), where also results for reversed magnetic fields are given. It can be seen that without an applied magnetic field the DMS does not spin-polarize electrons, leading to a net spin-polarization of the electrons recombining with holes in the QD of zero. This is due to the paramagnetic nature of the specific DMS used here, which requires an external magnetic field to effectively polarize the electron spins in the conduction band [6]. As noticed before, when the magnetic field is turned on, the circular polarization degree increases rapidly. At an applied field of +2 T a circular polarization degree of 83% is observed, while at +6 T it approaches 96%, indicating emission is almost completely \(\sigma^+\)-polarized. When the magnetic field is reversed, the circular polarization degree changes sign, but reaches the same magnitude. This corroborates that the high values of \(P_c\) are not caused by experimental artifacts. With reversed field, emitted photons now are predominantly \(\sigma^-\)-polarized. The desired circular polarization can thus be chosen by the direction of the applied magnetic field. Furthermore, the value of \(P_c\) can be further increased by reducing the current density through the spin-LED. For instance, with \(I = 0.26\) mA and \(B = +6\) T we obtained \(P_c = 98\%\). However, along with a lower current density the overall emission intensity is further reduced and close to the detection limit of the spectroscopic setup. During pulsed operation of the device, the circular polarization degree should exhibit a time-dependency, and its time-averaged value should be higher than in continuous operation [9].

In a second experiment, used to show antibunching of the circularly polarized photons from the QD, the previously described setup was modified. The magnetic field inside the magneto-optical cryostat was set to \(B = +2\) T. A pulse generator excited the spin-LED with sub-nanosecond electrical pulses. The pulses had an offset slightly below the threshold voltage of the spin-LED, a repetition frequency of 106.5 MHz and a nominal peak width of 0.5 ns. The signal was transmitted to the spin-LED with a low-temperature coaxial cable. To reduce spurious effects from impedance mismatch, parallel to the spin-LED we mounted a 1 nF capacitor in series with a 50 \(\Omega\) resistor. Emitted light was again collected by a 60 \(\times\) microscope objective and left the cryostat through an optical window as parallel beam. In the optical detection path, the quarter-wave plate and the linear polarizer were removed to allow more of the signal to pass. After being transmitted to the spectrometer by a fiber, the light left the double spectrometer at the first exit port, thereby being diffracted by only one of the two 1200 grooves/mm gratings. The spectrometer was adjusted such that only the more intensive \(\sigma^+\)-polarized excitonic emission line emitted from the QD reached the exit port. Then, the light was collimated into a 1\(\times\)2 multi-mode fiber optical splitter with a nominal splitting ratio of 50/50. The splitted signals were outcoupled from the two terminal fiber connectors and focused onto single photon counting modules (nominal photon detection efficiency of 33% at relevant wavelength, 25 dark counts/s). This fiber-based Hanbury-Brown Twiss setup was chosen to prevent false coincidence signals from cross-talk effects [10]. Antibunching was detected by measuring coincidence counts at zero time delay between the two single photon counters. The second order correlation function \(g^{(2)}(t)\) obtained from this experiment is shown in Fig. 3(b). From the value of the zero delay peak area \(g^{(2)}(0) = 0.38\) it clearly results that the emission is non-classical, proving the layer sequence, compositions and thicknesses of the design are suitable for obtaining single photons by applying voltage pulses.

Although during pulsed excitation WL emission adds to the overall sample emission to a larger extent than under continuous current excitation, WL states do contribute only faintly to the spectrally filtered signal reaching the single photon counters. This is due to the WL emission being centered at 1.40 eV, such that the QD emission lines are only marginally influenced. We assume that the value of the zero delay peak area can be significantly reduced by varying the excitation parameters, e.g., by depleting the spin-LED of carriers with a short negative voltage pulse immediately after the excitation to further suppress multiphoton emission. Evidence of this for an electrically driven single photon source with an InAs QD was reported in Ref. [11].

To conclude, we have demonstrated an electrically operated spintronic device based on a magnetic semiconductor which emits single photons with a selected circular polarization degree. The measured circular polarization degrees are very high, indicating the effectiveness of our approach. The helicity of the emitted photons can be controlled by the direction of an externally applied magnetic field. The presented design is limited by the magnetic field and temperature requirements. Further development of ferromagnetic semiconductor materials may lift these requirements. However, the material used in this study may serve as a model system demonstrating a possible design, and suggests that magnetic semiconductors are most promising for the realization of light sources for single photons with defined helicity.

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