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Direct observation of all-optical injection of spin-polarized currents

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Ballistic currents of spin-polarized carriers are generated by quantum interference of one and two photon absorption. Polarization and relative phase of the exciting femtosecond laser fields control the magnitude and direction of the currents as well as the electron spin orientation. These spin-polarized currents are experimentally detected by a phase-dependent spatial shift of the circularly analyzed photoluminescence in cubic ZnSe.

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1 Introduction The manipulation of carrier spins in semiconductors currently attracts great attention since it is a prerequisite to spintronics [1]. Besides the extension of today's electronics and optoelectronics, spintronics are of particular interest in the context of quantum information processing since spin coherence often largely exceeds the phase coherence of the carriers, and long coherence times are necessary for quantum data processing. One of the main problems of spintronics is the efficient and controlled injection of carrier spin currents.

The spin polarization of carriers generated by excitation with circularly polarized light is due to the optical selection rules and is known for a long time [2]. The generation of directed currents due to the preferential occupation of carriers in specific k directions via quantum interference of one and two photon absorption has also been already theoretically and experimentally demonstrated [3]. Bhat and Sipe predicted [4] that the quantum interference of one and two photon absorption can, under suitable polarization and phase conditions, also lead to injection of *ballistic, spin-polarized currents*.

Directed currents due to intersubband re-distribution of spin-polarized carriers have already been observed [5] and the quantum interference of one and two photon absorption of circularly polarized laser beams has been reported [6]. However, the spin polarization in both of these cases was only theoretically inferred and was not experimentally proven. We report here on photoluminescence (PL) experiments to directly evidence the injection of fast, ballistic, spin-polarized electrons in an unbiased semiconductor.

2 Theory The relation between spin orientation and current direction for coherent excitation with one and two photons is sketched in Fig. 1 (from Ref. [4]): in part a) the one and two photon beams are co-circularly polarized and in part b) the beams are cross-linearly polarized. The direction of laser propagation is parallel to the z axis and the sample surface lies in the xy plane. Thin arrows denote the

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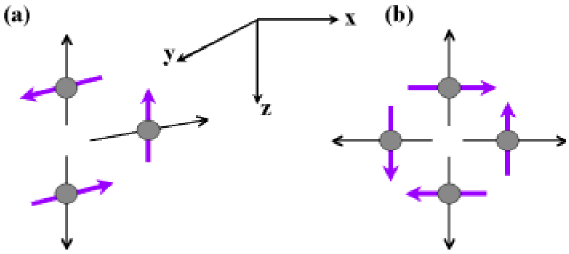

Fig. 1 (online colour at: www.interscience.wiley.com) Scheme of the electron motion (thin arrows) and their spin orientation (thick arrows) for the excitation beams in z direction. Case a) is for co-circular and case b) for cross-linear polarization.

current direction and thick arrows the spin orientation. A major difference between the two cases is that a real net current in the xy plane perpendicular to the light propagation direction is present for the co-circular polarization whereas only spin currents are present in case of the cross-linear polarization. We have chosen the excitation geometry of case b). The ω laser pulses are linearly polarized along the x axis, whereas the 2ω pulses are polarized along the y axis. The theoretical model predicts that the direction of the in-plane currents are parallel to the orientation of the linear polarization of the ω excitation. The currents in $\pm z$ direction are neglected in the following because we detect the photoluminescence in $-z$ direction and their spin polarization is not detectable in this geometry. The current magnitudes and their spin polarization are determined by the relative phase between the two light fields. Only the electron spins are considered since the spin-flip times of holes are very short [7]. The maximum value for the spin current injection depends mainly on the excess energy and the mobility of the carriers.

3 Experiment

Cubic ZnSe with a band gap of 2.72 eV at 100 K was selected for practical reasons: A Ti:sapphire laser can be used for two photon excitation and its frequency-doubled mode for one photon excitation. A 290 nm thick ZnSe layer was grown by molecular beam epitaxy on a GaAs substrate. The substrate is afterwards in the area of the excitation spot completely removed by selective chemical etching.

The linearly polarized Ti:sapphire laser has a repetition rate of 80 MHz, a pulse width of 150 fs and a wavelength of 800 nm. Frequency-doubled light with a perpendicular linear polarization is generated in a lithium-borate crystal (LBO). The time and, in particular, the phase delay between the ω and the 2ω beam is controlled in a Michelson set-up as depicted in Fig. 2.

The two laser beams are focused with a mirror of focal length of 5 cm to a spot size of about 4 µm (full width at half maximum) on the sample, which is kept at 100 K in a temperature-controlled micro-

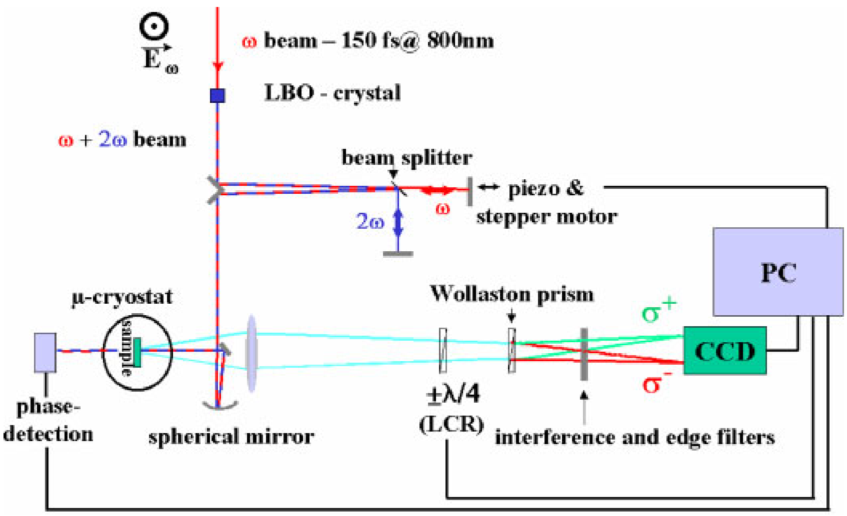

Fig. 2 (online colour at: www.interscience.wiley.com) Experimental set-up.

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cryostat. A carrier density of about 10^{18} cm^{-3} is generated with both beams for powers of 350 mW for the ω and of 80 µW for the 2ω beam. The carriers excited in the conduction band have an initial excess energy of 305 meV. The phase relation and the wave-front accuracy of the two beams are checked with the laser light passing the thin ZnSe layer: The ω beam is frequency doubled and then the interference pattern with the 2ω beam is monitored with a CCD camera behind the cryostat.

The magnified PL spot is separated with a liquid crystal retarder (LCR) and a Wollaston prism into σ^+ and σ^- polarized light and detected as two spots on a high resolution CCD-camera. Laser stray light is suppressed by appropriate interference and edge filters.

The movement of the circularly polarized PL spots is small in comparison with the diameter of the excitation spot, therefore we apply a doubly differential technique: We measure the distance of the σ^+ and σ^- polarized PL spots for the liquid crystal retarder (LCR in Fig. 2) set to + and – $\lambda/4$ in subsequent measurements. The immediate interchange of the position of the σ^+ and σ^- polarized PL spots removes pointing instabilities of the exciting laser beams. The centers of gravity of the PL spots are determined by gaussian fits.

The detailed experimental procedure is sketched in Fig. 3: The electrons move parallel to the polarization of the ω beam (left part). The current magnitude depends on the phase difference ($2\Phi_\omega - \Phi_{2\omega}$). The solid black circle in the middle part is the position of the PL spot for $2\Phi_\omega - \Phi_{2\omega} = 0$, whereas the dashed circles are the positions of the PL spots for σ^+ and σ^- polarization for $2\Phi_\omega - \Phi_{2\omega} = \pi/2$. The determination of the distance $L(+\lambda/4)$ (when the LCR is set to $+\lambda/4$) of the PL spots on the CCD camera is depicted in the right part of Fig. 3. The final data are obtained by determining the differential distance $\Delta = L(+\lambda/4) - L(-\lambda/4)$. This quantity Δ (dots in the upper part) is plotted in Fig. 4 as a function of the voltage applied to the piezo crystal in one of the arms of the Michelson interferometer. The simultaneously measured central intensity of the interference pattern of the frequency-doubled ω beam with the 2ω beam (as measured behind the cryostat) is shown as a blue line in the lower part of Fig. 4. This measurement fixes the relation between the piezo voltage and the phase difference, i.e., the period in the periodic movement of the PL spots. A sine fit (with this fixed period) to the experimental dots is shown as a red line in the upper part of Fig. 4. We deduce that the PL spots move by ±6 nm.

4 Discussion We measured the carrier lifetime with a streak camera system to be only 6 ps (at 100 K). This short lifetime is certainly caused by recombination at the surface of the thin ZnSe layer. The spin lifetime, as determined by the polarization dependent decay of the PL, is with 150 ps much longer. Therefore spin relaxation has not to be included when modelling the data. An initial spin-polarization up to 50% can be obtained with our experimental conditions, where electrons are excited from heavy- and light-hole valence bands, respectively. The effect of space charge fields due to the faster moving electrons and the slower moving holes and dispersion within the thin sample is neglected. The average displacement between the spin-up and the spin-down electrons is then calculated according the theory in

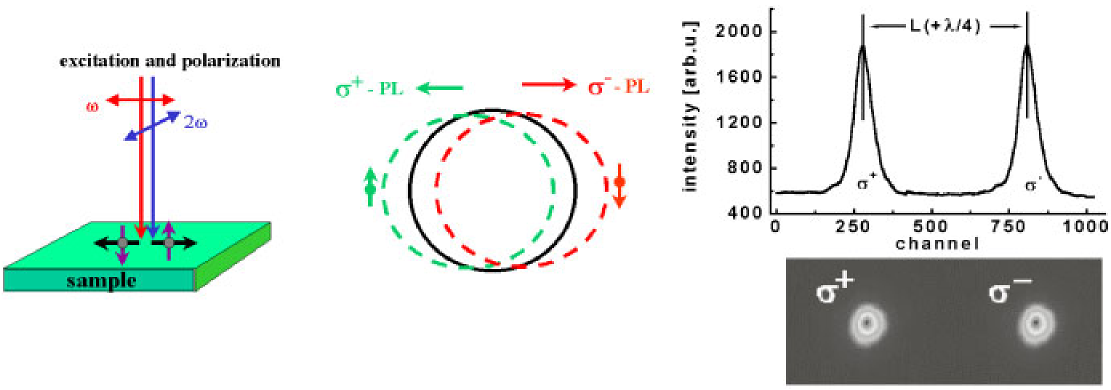

Fig. 3 (online colour at: www.interscience.wiley.com) Determination of the PL-spot positions for σ^+ and σ^- polarization.

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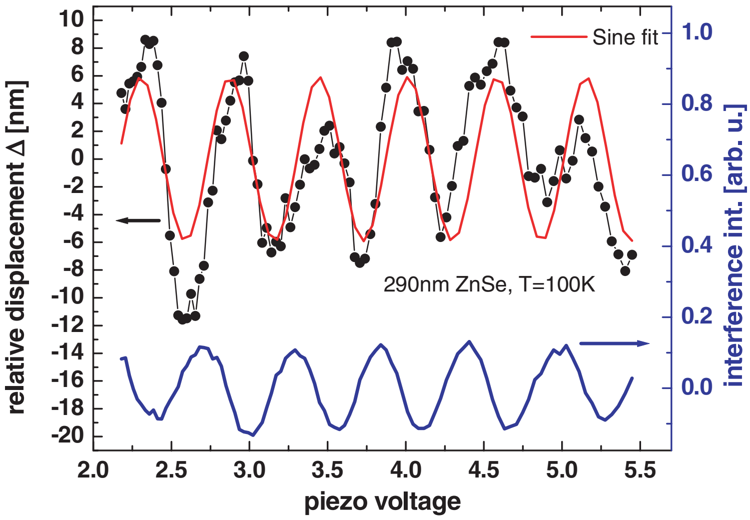

Fig. 4 (online colour at: www.interscience.wiley.com) Relative displacement Δ of the σ^+ and σ^- polarized PL spots (upper part) and the intensity of the interference of the frequency-doubled ω with the 2ω laser beam after the cryostat (lower part) as a function of voltage applied to the piezo crystal in the Michelson interferometer.

reference [4]. We assume an isotropic distribution of carriers in k space for calculating the spatial PL signals for σ^+ and σ^- PL since momentum relaxation time is with about 100 fs (scattering with optical phonons) much faster than the carrier lifetime. We obtain a displacement of the PL spots (which is smaller than the displacement of the electrons due to the optical selection rules in bulk material) of ±9.5 nm. We consider the agreement with experiment satisfactory since optimum conditions as, e.g., optimum balancing of the excitation power of one and two photon excitation, are assumed in the model.

5 Conclusion We have provided experimental evidence that all-optical injection of spin currents via the quantum interference of one and two photon excitation-paths is indeed possible. Similar results on GaAs QWs are reported on this conference. The initially ballistic spin currents could be used for efficient spin injection in nanostructures as in e.g. quantum wells and quantum dots. This all-optical control of spin currents is extremely fast, and the steering mechanism itself for the spin currents requires only minor power.

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