Direct Observation of Optically Injected Spin-Polarized Currents in Semiconductors

J. Hübner and W.W. Rühle
Department of Physics and Materials Science Center, University of Marburg, Renthof 5, 35032 Marburg, Germany

M. Klude and D. Hommel
Institute of Solid State Physics, Semiconductor Epitaxy, University of Bremen, P.O. Box 330440, 28334 Bremen, Germany

R. D. R. Bhat, J. E. Sipe, and H. M. van Driel
Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario M5S 1A7, Canada
(Received 20 December 2002; published 30 May 2003)

Quantum interference of one- and two-photon excitation of unbiased semiconductors yields ballistic currents of carriers. The magnitudes and directions of the currents *and* the spin orientations of the carriers are controlled by the polarization and relative phase of the exciting femtosecond laser fields. We provide direct experimental evidence for the spin polarization of the optically injected spin currents by detecting a phase-dependent spatial shift of the circularly polarized photoluminescence in cubic ZnSe.

DOI: 10.1103/PhysRevLett.90.216601 PACS numbers: 72.25.Dc, 42.65.–k, 72.25.Fe

Macroscopic manifestations of pure quantum mechanical phenomena are fascinating. One of many examples is the generation of macroscopic currents via quantum mechanical interference of the optical transitions for one- and two-photon absorption in semiconductors [1,2]. Here the initial state (the top valence band) and the final state (the lowest conduction band) are connected by two different pathways: a direct one with one-photon excitation and a second one with two-photon excitation via an intermediate virtual state close to the middle of the band gap. According to quantum mechanics, the transition probabilities do not simply add, however; constructive or destructive interference of the transition *amplitudes* leads to an asymmetric distribution of carriers in quasimomentum k space which then results in macroscopic, initially ballistic currents in real space. The quantum interference control (QUIC) can, under carefully chosen experimental conditions, lead to currents of the order of several $\mathrm{kA/cm^2}$, and the magnitude and direction of these QUIC currents can be directly controlled by the relative phase of the exciting two-color light fields [2].

A second example is the generation of a macroscopic magnetization by optical generation of spin-polarized carriers via excitation with circularly polarized light due to spin-orbit coupling and the optical selection rules [3]: The transition matrix element from the heavy hole band ($J = \frac{3}{2}, m_j = \pm\frac{3}{2}$) is about 3 times larger than that from the light hole band ($J = \frac{3}{2}, m_j = \pm\frac{1}{2}$). Electron populations with a spin polarization, $(n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow)$, of about 50% are thus generated in bulk semiconductors yielding a macroscopic magnetization. The carrier and spin distribution in k space are in this case symmetric, no macroscopic currents are generated.

Recently, Bhat and Sipe [4] combined these two schemes to predict the possible generation of spin currents. Spin currents are carried by electrons with preferential spin orientation. They need not, however, be connected with a net electrical current: A symmetric distribution of carriers in k space with an asymmetric distribution of spin orientation yields a pure spin current without a corresponding net electrical current. This extension was achieved by considering various possibilities of the polarization of the incident two-color light fields with frequencies ω and 2ω. The photon energy $\hbar 2\omega$ is larger than the band gap energy E_g. Then electrons with excess energy are generated, leading to ballistic currents before scattering occurs.

The current directions as well as the carrier spin orientations can be directly controlled via the relative phase $(2\phi_\omega - \phi_{2\omega})$ and polarization $\vec{\mathbf{E}}_\omega, \vec{\mathbf{E}}_{2\omega}$ of the optical two-color light fields $\vec{\mathbf{E}}(t)$:

$$\vec{\mathbf{E}}(t) = \vec{\mathbf{E}}_\omega e^{i(\phi_\omega - \omega t)} + \vec{\mathbf{E}}_{2\omega} e^{i(\phi_{2\omega} - 2\omega t)} + \text{c.c.} \quad (1)$$

This all-optical control of spin currents can be made extremely fast and the control process via the relative phase shift of the exciting lasers requires only minor power, since it relies on the *coherent* control of two light fields. This all-optical method also requires no electrical contacts and no external bias. Current magnitudes, directions, and spin orientations can be controlled. Therefore these possibilities of coherent optical generation of spin currents should attract great attention of the *spintronics* [5] community since it is a completely new means to inject spin-polarized carriers to control their injection rate and degree of spin polarization.

Some groups have already experimentally observed directed currents due to intersubband redistribution of spin-polarized carriers [6] or quantum interference of one- and two-photon absorption using circularly polarized laser beams [7]; however, the spin polarization of

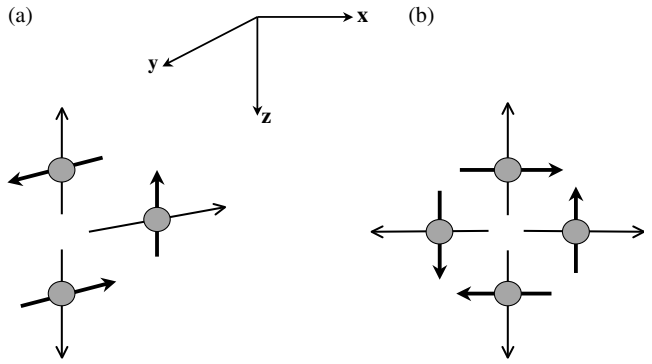

FIG. 1. Schematic drawing of the electron motion and their corresponding spins (from [4]). The light propagation direction is parallel to z and the sample surface lies in the xy plane. Thin arrows denote the current direction and thick arrows the spin orientation. Case (a) corresponds to cocircular and case (b) to cross-linear polarization.

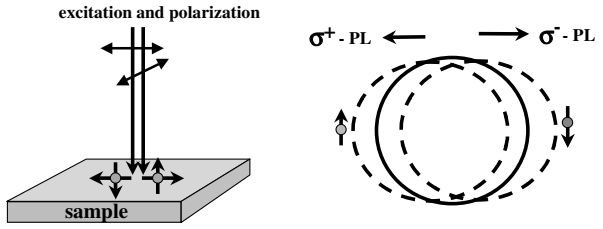

FIG. 2. Schematic drawing of the PL-spot movement. The solid line marks the excitation spot, which also marks the position of the resulting photoluminescence (PL) *without* spin currents [e.g., for $(2\phi_\omega - \phi_{2\omega}) = 0, 2\pi, 4\pi, \ldots$]. The dashed lines mark the position of the PL spots for spin-up and spin-down electrons for *maximum* spin current [$(2\phi_\omega - \phi_{2\omega}) = \frac{\pi}{2}, \frac{5\pi}{2}, \frac{9\pi}{2}, \ldots$]. For $(2\phi_\omega - \phi_{2\omega}) = -\frac{\pi}{2}, \frac{3\pi}{2}, \frac{7\pi}{2}, \ldots$ the spin-up current flows to the right and the spin-down current to the left; i.e, the positions of the σ^+ and σ^- PL are interchanged.

these currents was always inferred only theoretically and not proven experimentally.

The relation between spin orientation and current direction for excitation by coherent one- and two-photon absorption is sketched in Fig. 1 for two possible cases. In 1(a), the ω and 2ω beams are cocircularly polarized. Pure spin currents without net electrical currents flow in the $\pm z$ directions, and a spin-polarized macroscopic net electrical current is present in the xy plane. The direction in the xy plane of the latter current depends on the relative phase $(2\phi_\omega - \phi_{2\omega})$, and the spin orientation is given by the helicity of the exciting ω and 2ω light fields.

In 1(b) the ω laser light is linearly polarized along the x axis, whereas the 2ω light is polarized along the y axis. The theoretical model predicts that only pure spin currents are present. The direction of the in-plane spin currents are parallel to the orientation of the linear polarization of the ω excitation. Additionally, pure spin currents flow in $\pm z$ directions. The current magnitude and the spin polarization are determined by the relative phase $(2\phi_\omega - \phi_{2\omega})$ of the ω and 2ω light fields. The maximum value for the spin-current injection depends mainly on the excess energy and on the mobility of the carriers. We concentrate here on electron spins only, because the spin-flip times of holes are rather short [8].

In this Letter we present first experimental evidence for the pure spin currents flowing in $\pm x$ directions in Fig. 1(b). In particular, we prove that their magnitudes and spin polarization can be directly controlled with the relative phase of the exciting ω and 2ω light fields: Spin currents lead to a relative displacement of electrons with spin-up and spin-down orientation. The displacements can be visualized by a relative displacement of the σ^+- and σ^--polarized photoluminescence after coherent excitation with ω and 2ω femtosecond laser pulses (see Fig. 2). The currents in $\pm z$ directions are omitted from now on, since they play no role in our experiment.

We have chosen ZnSe in our experiment for practical reasons: the band gap of 2.72 eV (at 100 K) [9] allows the use of a Ti:sapphire laser for two-photon (ω) excitation and its frequency-doubled mode for one-photon (2ω) excitation, respectively. ZnSe is also an interesting material since it has long spin coherence times, in particular, at elevated temperatures [10], which makes it a promising candidate for noncryogenic spintronic applications. The sample is a 290 nm thick layer of cubic ZnSe with a high crystalline quality grown by molecular beam epitaxy on a GaAs substrate.

An 80 MHz Ti:sapphire laser generates 150 fs pulses at a wavelength of 800 nm ($\hat{=}\,\omega$) with linear polarization (Fig. 3). The laser light is partially frequency doubled in a lithium-borate (LBO) crystal which generates the 2ω light fields at 400 nm with perpendicular linear polarization. The split-off band is not excited at this

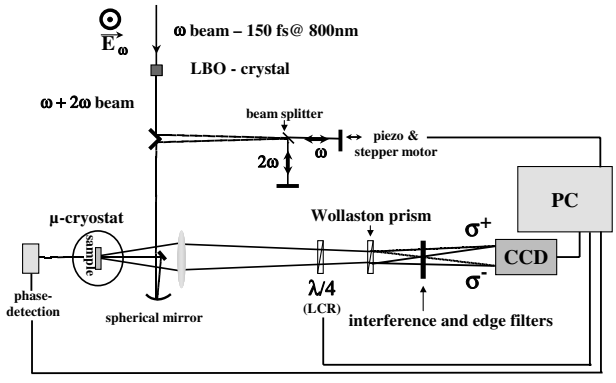

FIG. 3. Experimental setup. The ω-light beam is delayed relative to the 2ω-light beam by a delay stage with coarse and fine adjustment.

wavelength. The electrons excited from the heavy hole band have an excess energy of about 300 meV. The delay between the ω and 2ω light fields is achieved by a Michelson setup with a dichroic beam splitter. The Michelson separates the ω and 2ω beams into the two arms of the interferometer, where dielectric mirrors suitable for the proper wavelengths are used. The relative phase is varied with a piezo. The reunified two-color light fields are focused by a spherical mirror ($f = 5$ cm) onto the sample to an excitation spot with a diameter of about 4 μm. The power of the ω beam is 350 mW at the sample. The power of the 2ω beam is adjusted such that the excitation with each of the beams leads to a comparable carrier density as monitored by the strength of the photoluminescence (PL). This procedure is chosen to balance the powers of the two beams such that a maximum modulation for their interference is expected. The carrier density is about $\sim 10^{18}$ cm^{-3} for excitation with both beams. The absorbing GaAs substrate is removed by selective chemical etching, so that a fraction of the two-color light fields passes the thin ZnSe layer. This allows the phase relation to be measured on the transmitted beams *in situ*: the ω beam is frequency doubled behind the sample and brought into interference with the 2ω beam. Additionally, the shape of the interference pattern of the frequency-doubled ω and 2ω beams is monitored with a charge-coupled device (CCD) camera. This allows the parallelism and quality of the wave fronts to be checked. The PL spot on the sample is imaged onto a high resolution CCD camera. Bandpass interference filters and edge cut filters completely suppress scattered light of the exciting lasers.

Differential measurements have to be applied, since the movement of the spot for a given circular polarization as a function of the phase $(2\phi_\omega - \phi_{2\omega})$ is expected to be much smaller than the optical resolution. First, both circular polarizations of the same spot are detected simultaneously but spatially separated on the CCD camera. The spatial separation is achieved by a combination of a liquid crystal retarder acting as $\lambda/4$ plate and a Wollaston prism. The CCD camera thus detects two spots, where one corresponds to the σ^+- and the other to the σ^--polarized part of the PL. Their movement against each other is measured by the relative change of the centers of gravity of the spots which are determined by Gaussian fits. Second, noise and artifacts caused by long term beam instabilities are strongly reduced by switching the wavelength retardance of the liquid crystal retarder from $+\lambda/4$ to $-\lambda/4$ and vice versa after each measurement, thus interchanging the σ^+ and σ^- detection on the CCD camera in sequential data sets. The final data are given by the difference between the determined distances $\Delta_{\text{signal}} = \Delta_{\text{LCR}(+\lambda/4)} - \Delta_{\text{LCR}(-\lambda/4)}$, eliminating first the offset displacement by the Wollaston prism, second apparent displacements, e.g., due to inhomogeneities on the CCD camera, and third pointing instabilities of the exciting laser. The results of this double differential method are shown in Fig. 4.

The lower solid line in Fig. 4 displays the measured interference intensity of the frequency-doubled ω and the 2ω light beams as a function of delay, which yields the relative phase relation between the ω and the 2ω light fields. The upper curve shows the measured displacement between the centers of gravity of the photoluminescence spots. The measured data are fitted with a sine function with the amplitude as fit parameter [11]. The period of the sine is fixed by the simultaneously recorded lower curve in Fig. 4. We checked our results with the following experiments: There is no phase-dependent movement of the spots in the perpendicular direction to the polarization of the ω beam. The movement of the spots disappears (a) if only one laser beam is used for excitation, (b) if the two beams do not temporally overlap, and (c) if the wave fronts at the sample are not well shaped.

A sample temperature of 100 K was chosen since then spin lifetime is longer and carrier scattering (emission of LO phonons) is still not much faster than at lower temperatures [12]. The carrier lifetime ($\tau_l \sim 6$ ps) and spin lifetime ($\tau_s \sim 150$ ps) for this sample are determined by measuring the decay time of the PL and the decay of its polarization for excitation with circularly polarized 2ω light with a streak camera system. The short lifetime is caused by nonradiative recombination at the surfaces of the thin ZnSe layer. This lifetime gets shorter with increasing temperatures, setting an upper limit of 100 K to our experiments. Please note that this temperature limit is set by our experimental method. Spin currents can also be generated at room temperature.

We model the transport of the conduction band electrons using the Boltzmann equation in the relaxation-time approximation, with a relaxation-time τ:

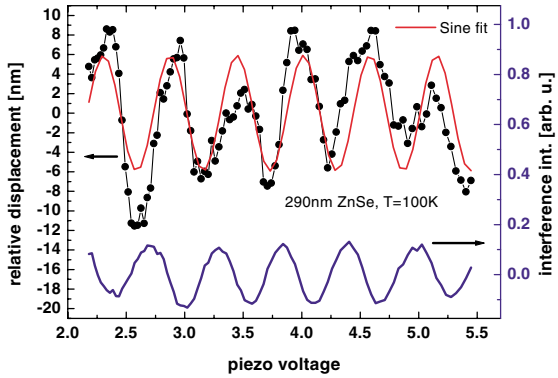

FIG. 4 (color online). Results of the measured displacement in ZnSe at 100 K with excitation with 400 and 800 nm light, respectively. The lower curve shows the relative phase relation $(2\phi_\omega - \phi_{2\omega})$ between the ω and 2ω light beams as a function of the delay. The upper curve displays the displacement of spot due to the spin currents.

$$\frac{\partial \rho}{\partial t} + \mathbf{v}(\mathbf{k}) \cdot \nabla_r \rho + \mathbf{E} \cdot \nabla_k \rho = -\frac{1}{\tau}(\rho - \rho_{\mathrm{eq}}). \quad (2)$$

Here, $\rho(\mathbf{k}, \mathbf{r}, t)$ is the 2×2 density matrix for electrons in the conduction band with velocity $\mathbf{v}(\mathbf{k})$ at position $\mathbf{r}$, $\rho_{\mathrm{eq}}(\mathbf{k}, \mathbf{r}, t)$ is a quasiequilibrium density matrix with no net velocity and the same local density as ρ (i.e., $\int \rho_{\mathrm{eq}} d^3k = \int \rho d^3k$), and $\mathbf{E}$ is the space-charge field that builds up due to the separation of the faster electrons from slower moving holes. The space-charge field is very small. By neglecting it, Eq. (2) can be solved for the average position of the electrons in terms of moments of the initially injected density matrix.

It is a good approximation to assume that ρ has become isotropic in $\mathbf{k}$ before recombination, since the carrier lifetime is much larger than the momentum relaxation time. We calculate the luminescence using band-edge matrix elements [3]. The distance between σ_+- and σ_- spots, Δ, is proportional to the expectation value of the product of the x component of velocity and the z component of spin of the electrons, which is K_e^{xz} in the notation of Ref. [4]. Specifically,

$$\Delta = \frac{2\tau}{\hbar} \frac{\dot{K}_e^{xz}}{\dot{n}_{(1)} + \dot{n}_{(2)}}, \quad (3)$$

where the spin-current injection rate $\dot{K}_e^{xz}$ and the one- (two-) photon injection rate $\dot{n}_{(1)}$ ($\dot{n}_{(2)}$) are evaluated using the peak optical fields. The difference in the in-plane spatial dependencies for one- and two-photon absorption processes is considered in a noncongruent intensity distribution of the incident fields arising from the first nonlinear frequency doubling process.

We have evaluated Eq. (3) using an eight band Kane model [4] using ZnSe parameters from [13]. We obtain $\Delta = 19$ nm assuming a momentum relaxation time of 100 fs (scattering with longitudinal optical phonons [14]) and for optimally balanced beam intensities. This theoretical value corresponds well to the experimental result of $\Delta = 12$ nm. A shorter scattering time would improve the agreement. Further reduction of the shift in the experiment can be attributed to the fact that the phase front matching is not perfect and the ω and 2ω beams are not ideally balanced in power [15]. The Coulomb attraction of the slower moving holes on the electrons also reduces the overall PL-spot movement.

The initial displacement between spin-up and spin-down electrons during their ballistic motion is larger than the distance between the measured σ^+- and σ^--luminescence spots for several reasons: First, electrons with a certain spin polarization emit both σ^+- and σ^--polarized PL. Second, the displacement of the carriers has to be averaged over a distribution in k space. Third, carrier-carrier scattering and diffusion randomize the spin currents after their initial ballistic phase. It should be noted that the spin-polarized ballistic electrons actually move up to 100 nm in ZnSe, and up to 100% spin polarization of ballistic electrons can be achieved in principle in quantum wells.

In conclusion, we have given unambiguous experimental evidence that the all-optical injection of spin currents is possible by using the quantum interference of two-color laser fields with cross-linear polarization.

We acknowledge the financial support by the German Ministry for Education, Technology and Research (BMBF), the Optodynamics Center Marburg, the Natural Sciences and Engineering Research Council, Photonics Research Ontario, and the Defense Advanced Research Projects Agency.

Note added.—Just before submitting the paper we became aware that similar experimental results were obtained in GaAs quantum wells [16].

[1] R. Atanasov, A. Haché, J. L. P. Hughes, H. M. van Driel, and J. E. Sipe, Phys. Rev. Lett. **76**, 1703 (1996).
[2] A. Haché, Y. Kostoulas, R. Atanasov, J. L. P. Hughes, J. E. Sipe, and H. M. van Driel, Phys. Rev. Lett. **78**, 306 (1997).
[3] M. I. Dyakonov and V. I. Perel, in *Optical Orientation*, edited by F. Meier and B. Zakharchenya (Elsevier Science Publishing, Amsterdam, 1984).
[4] R. D. R. Bhat and J. E. Sipe, Phys. Rev. Lett. **85**, 5432 (2000).
[5] D. D. Awschalom and J. M. Kikawa, Phys. Today **52**, No. 6, 33 (1999).
[6] S. D. Ganichev, E. L. Ivchenko, S. N. Danilov, J. Eroms, W. Wegscheider, D. Weiss, and W. Prettl, Phys. Rev. Lett. **86**, 4358 (2001).
[7] M. J. Stevens, A. L. Smirl, R. D. R. Bhat, J. E. Sipe, and H. M. van Driel, J. Appl. Phys. **91**, 4382 (2002).
[8] D. J. Hilton and C. L. Tang, Phys. Rev. Lett. **89**, 146601 (2002).
[9] L. Malikova, W. Krystek, F. H. Pollak, N. Dai, A. Cavus, and M. C. Tamargo, Phys. Rev. B **54**, 1819 (1996).
[10] D. Hägele, M. Oestreich, W. Rühle, J. Hoffmann, S. Wächter, H. Kalt, and K. Ohkawa, Physica (Amsterdam) **272B**, 338 (1999).
[11] The phase offset with respect to the lower curve of Fig. 4 is caused by dispersion of the ω and 2ω beams after passing the sample.
[12] H. Kalt, J. Hoffmann, D. Tröndle, S. Wachter, D. Lüerßen, D. Hägele, W. Rühle, M. Oestreich, I. Blewett, and I. Galbraith, J. Cryst. Growth **214/215**, 630 (2000).
[13] H. P. Wagner, M. Kühnelt, W. Langbein, and J. M. Hvam, Phys. Rev. B **58**, 10 494 (1998).
[14] A. Leitensdorfer, C. Fürst, A. Lauberau, W. Kaiser, G. Tränkle, and G. Weimann, Phys. Rev. Lett. **76**, 1545 (1996).
[15] The phase front displacement of ω and 2ω beams in the light propagation direction due to dispersion within the sample can be neglected because of the small sample thickness.
[16] A. Smirl (private communication).