PHYSICAL REVIEW B **72**, 201302(R) (2005)

Injection of ballistic pure spin currents in semiconductors by a single-color linearly polarized beam

Hui Zhao, Xinyu Pan, and Arthur L. Smirl*
Laboratory for Photonics & Quantum Electronics, 138 IATL, University of Iowa, Iowa City, Iowa 52242, USA

R. D. R. Bhat, Ali Najmaie, J. E. Sipe, and H. M. van Driel
Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario, Canada M5S 1A7
(Received 6 September 2005; published 16 November 2005)

We demonstrate the injection and control of pure spin currents in [110]-oriented GaAs quantum wells at room temperature by one-photon absorption of a single linearly polarized optical pulse. These currents result from the interference of absorption processes associated with the right and left circularly polarized components of the pulse, with the current direction determined by their relative phase. The current generation process differs from the circular photogalvanic effect, which relies only on the intensity of circularly polarized beams. By using spatially resolved pump-probe techniques, we obtain signatures for the currents by measuring the resulting spin separations of 1–4 nm. The separation decreases with increasing excitation fluence, consistent with a reduction in the momentum relaxation time with increasing carrier density.

DOI: 10.1103/PhysRevB.72.201302 PACS number(s): 72.25.Fe, 42.65.−k, 72.25.Dc

The single-photon absorption of linearly polarized, monochromatic radiation by atoms, molecules, and solids is widely known to change the population of electronic states. In the case of a semiconductor, if the photon energy is higher than the fundamental band gap, electrons are created in the conduction band and holes in the valence band. Numerous experiments conducted on semiconductors over the past few decades have involved such simple absorption processes, and it is widely believed that only populations are changed and that no currents of any type are generated.

Recently, however, it has been predicted[1] that *ballistic* pure spin currents (PSC), which involve spin transport but no net charge transport, can be injected in a semiconductor that lacks inversion symmetry using a *single-color* linearly polarized beam by taking advantage of the sample symmetry and the spin-orbit coupling. In this paper we demonstrate such a current in [110]-oriented GaAs multiple quantum wells. The quantum wells weakly reduce the T_d symmetry of bulk GaAs to C_{2v}, but they play no fundamental role, and thus, the PSC should also be observable in bulk materials. This effect is in direct contrast to the previous[2,3] injection of ballistic PSC by the quantum interference between two-photon absorption of a linearly polarized fundamental pulse and one-photon absorption of an orthogonally polarized second harmonic pulse, which required the control of the polarizations of two harmonically related beams, which did not require a lack of inversion symmetry, and where the symmetry was broken by the superposition of the fundamental and second-harmonic fields. This observation can have important consequences for a wide variety of optical experiments on semiconductors and could also have specific implications for the field of spintronics,[4] which is based on the use of spin as an alternate, or additional, degree of freedom in information technology.

The geometry that we use is shown schematically in Fig. 1. The sample consists of 20-periods of 8-nm-wide GaAs wells alternating with 8-nm-thick $Al_{0.3}Ga_{0.7}As$ barriers, grown on a (110)-oriented GaAs substrate, which has been removed by chemical etching to allow transmission measurements. The band gap of the GaAs wells is 1.485 eV at 300 K. A single 70 fs [full width at half maximum (FWHM)] optical pulse at 750 nm (obtained by frequency doubling the 1.5 μm signal from an optical parametric oscillator operating at 80 MHz) is normally incident on the sample surface traveling along the $z=[110]$ growth direction and is focused onto the sample to a spot with a diameter of 2 μm (FWHM). The pump pulse excites electrons from the valence to the conduction band by one-photon absorption with an excess energy of ~148 meV.

For the C_{2v} symmetry appropriate for quantum wells in a [110]-oriented sample and for our excitation conditions, we use a rate equation analysis to calculate the charge and spin currents injected by a single color beam, which leads to essentially the same results as those calculated earlier[1] using the semiconductor optical Bloch equations. The pump pulse is of the form $\mathbf{E}=\mathbf{E}_{\boldsymbol{\omega}}\exp[-i(\omega t-kz)]+\text{c.c.}$, where the complex field amplitude $\mathbf{E}_{\boldsymbol{\omega}}$ allows for an arbitrary polarization and can be written either in terms of the x and y amplitudes (E_x and E_y) and phases (ϕ_x and ϕ_y) or the right and left circular amplitudes (E_+ and E_-) and phases (ϕ_+ and ϕ_-), respectively, as

$$\begin{aligned}\mathbf{E}_{\boldsymbol{\omega}} &= E_x\exp(i\phi_x)\hat{\mathbf{x}}+E_y\exp(i\phi_y)\hat{\mathbf{y}}\\ &= E_+\exp(i\phi_+)\hat{\boldsymbol{\sigma}}^+ + E_-\exp(i\phi_-)\hat{\boldsymbol{\sigma}}^-,\end{aligned}\tag{1}$$

where $\hat{\boldsymbol{\sigma}}^+=(\hat{\mathbf{x}}+i\hat{\mathbf{y}})/\sqrt{2}$ and $\hat{\boldsymbol{\sigma}}^-=(\hat{\mathbf{x}}-i\hat{\mathbf{y}})/\sqrt{2}$ denote the unit vectors for right and left circularly polarized light, respectively. This leads to a charge current injection rate $\dot{J}^y$ along the y direction given by

$$\dot{J}^y=-2(\text{Im}\ \eta^{yxy})E_xE_y\sin(\phi_x-\phi_y)=(\text{Im}\ \eta^{yxy})[E_+^2-E_-^2]\tag{2}$$

and a spin current injection rate $\dot{K}^{yz}$ along y with the spin polarized along z given by

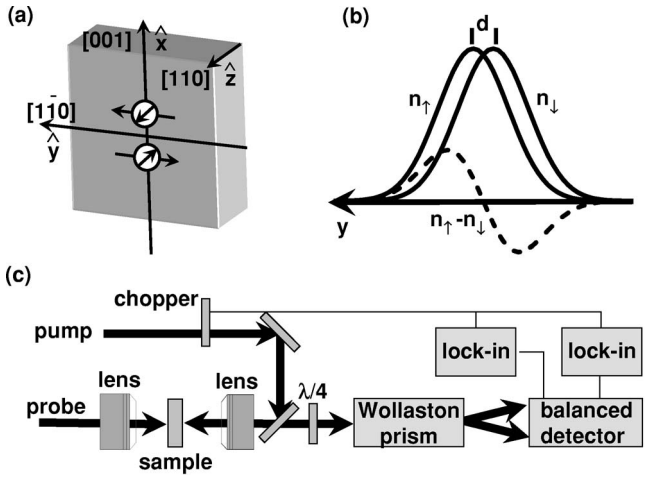

FIG. 1. (a) Sample orientation and definition of a laboratory coordinate system schematically depicting the injection of a PSC along the y direction by one-photon absorption of an x polarized field propagating along z. Small arrows inside spheres show the direction of net electron spin polarization, while the larger arrows intersecting the spheres show the direction of ballistic electron propagation. The spin-up (spin-down) electrons are shown moving in the $+y$ ($-y$) direction, yielding no net charge current. (b) Schematic representation of the spin-up and spin-down electron spatial distributions (solid lines) produced by the currents in (a), immediately following momentum relaxation. The dashed line shows the corresponding difference in the spin-up and spin-down electron densities resulting from this motion. (c) Experimental geometry for producing and measuring pure spin currents: $\lambda/4$ represent a quarter-wave plate.

$$\begin{aligned}\dot{K}^{yz} &= \mu^{yzxx}E_x^2 + \mu^{yzyy}E_y^2 \\ &= \frac{1}{2}(\mu^{yzxx} + \mu^{yzyy})[E_+^2 + E_-^2] \\ &\quad + (\mu^{yzxx} - \mu^{yzyy})E_+E_-\cos(\phi_+ - \phi_-), \end{aligned} \tag{3}$$

where $K^{yz} \equiv \langle v^y s^z \rangle$, $\langle\ \rangle$ denotes the expectation value, v^y and s^z are the y and z components of the velocity and spin, respectively, and η^{ijk} and μ^{ijkl} are the tensor and pseudotensor associated with the charge current and spin current, respectively, as defined in Refs. 5 and 1, and i, j, k, and l denote the Cartesian coordinates shown in Fig. 1(a).

For x- or y-polarized light, there is no charge current along $\hat{y}(\dot{J}^y = 0)$, but there is a spin current ($\dot{K}^{yz} \neq 0$). The expected electron dynamics accompanying the PSC injected by the one-photon absorption of linearly x-polarized light are schematically illustrated in Figs. 1(a) and 1(b). Initially, absorption creates equal numbers of spin-down and spin-up electrons: $n_\downarrow(y, t=0) = n_\uparrow(y, t=0)$, each having the same Gaussian spatial profile. However, each electron is injected with an initial ballistic velocity of $\sim$900 km/s. In addition, more of the spin-up electrons move in the $+y$ direction than in the $-y$ direction, while the opposite is true for the spin-down electrons. In this way, two oppositely directed currents with opposite spin are produced, but the sum of the two charge currents is exactly zero. Note that the optically injected holes move in the opposite direction to the electrons, but lose their spins on a time scale of $\sim$100 fs at room temperature;[6] therefore, the holes do not contribute to the spin currents and are ignored in subsequent discussions.

After injection, the spin current decays through momentum relaxation. Before the momentum is randomized, however, this pure spin current causes the spin-up electron spatial profile to shift in the $+y$ direction and the spin-down profile to shift in the $-y$ direction [see Fig. 1(b)]. This shift decays by spin relaxation, carrier diffusion, and recombination, and therefore persists long after momentum relaxation has destroyed the ballistic current. The magnitude of this shift is determined by the fraction of the total injected density that is spin polarized along $\hat{z}$ and travels along $\hat{y}$, the initial injection velocity, and the momentum relaxation time. At room temperature for the carrier densities used here ($\sim 10^{18}$ cm^3 for absorption of a 10 μJ/cm^2 pump), the shift is expected to be only a few nanometers.[1] For such small shifts, the difference in the spin populations, $n_\uparrow(y,t) - n_\downarrow(y,t)$, is given by the product of the spatial derivative of the original Gaussian profile and the separation d, and the sign gives the direction of spin flow. Specifically, the ratio between the peak heights of the $n_\uparrow - n_\downarrow$ and $n_\uparrow$ (or $n_\downarrow$) spatial profiles is equal to the ratio between d and the width of the Gaussian profile w.

We measure $n_\uparrow(y) - n_\downarrow(y)$ using the spatially resolved pump-probe apparatus shown in Fig. 1(c). A 100 fs probe pulse at 815 nm is taken from the output of the Ti:sapphire laser after it pumps the parametric oscillator. The probe pulse is initially linearly polarized along $\hat{y}$ and is focused on the sample to a diameter of 1.8 μm, as shown. The probe arrives at a fixed time delay of $\sim$10 ps after the pump, which is long enough to allow for carrier thermalization and momentum relaxation and for the spin relaxation of the holes, but short compared to recombination and spin relaxation of the electrons. The probe is tuned near the band edge of GaAs to allow for the efficient sampling of the spin-up and spin-down populations, but also far enough above it ($\sim$40 meV) to probe both heavy-hole and light-hole transitions. The σ^+ component of the linearly polarized probe interacts more strongly (approximately in the ratio of 3/1) with the spin-down electrons, while the σ^- component interacts more strongly with the spin-up electrons. Consequently, the net spin polarization of the carriers present in the sample at the position of the probe can be readily deduced from the difference in the absorption of the σ^+ and σ^- components of the probe.

We measure this circular dichroism by using an optical bridge that consists of a quarter-wave ($\lambda/4$) plate, a Wollaston prism, and a balanced detector. The quarter-wave plate converts the right circularly polarized component of the transmitted probe to x polarized light and converts the left circularly polarized component to y polarized light. The Wollaston prism separates the x and y components, which are measured by two photodiodes connected to a common electronic bridge. Two signals are independently, but simultaneously, recorded. One lock-in amplifier records the change in transmission (ΔT_+) induced by the pump in the σ^+ component of the probe, which for small ΔT_+ is a measure of $n_\downarrow$ at a given probe position. The other lock-in amplifier moni-

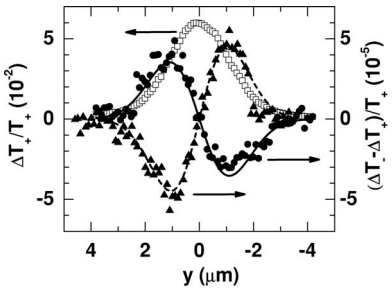

FIG. 2. Measurement of $\Delta T_+/T_+ \propto n_\downarrow(y)$ (open squares) and $\Delta T_-/T_+ - \Delta T_+/T_+ \propto n_\uparrow(y) - n_\downarrow(y)$ for an x polarized (solid circles) and a y polarized (solid triangles) pump pulse at room temperature for a pump fluence of 10 μJ/cm^2. The lines are the fits to the data for spin separation $d=2.8$ nm.

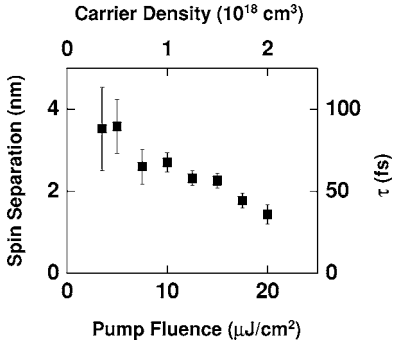

FIG. 3. Separation of the spin-up and spin-down profiles associated with the injection of PSC by single photon absorption as a function of pump fluence. The estimated momentum relaxation times τ extracted from these separations (right axis) and the approximate carrier density (top axis).

tors the difference between the differential transmissions for σ^- and σ^+ (i.e., $\Delta T_- - \Delta T_+$), and consequently, produces a signal that is proportional to $n_\uparrow - n_\downarrow$. Finally, $n_\downarrow(y)$ and $n_\uparrow(y) - n_\downarrow(y)$ are measured as a function of position by scanning the probe across the pump profile in the y direction by mechanically tilting a mirror in the pump path over a very small range.

The results of measuring $n_\downarrow(y) \propto \Delta T_+/T_+$ (open squares) and $n_\uparrow(y) - n_\downarrow(y) \propto \Delta T_-/T_+ - \Delta T_+/T_+$ (solid circles) for an x polarized pump pulse using the procedure described above are shown in Fig. 2 (where T_+ denotes the linear transmission of the σ^+ component of the probe with the pump blocked). Clearly, the $n_\uparrow(y) - n_\downarrow(y)$ signal is consistent with a PSC. Specifically, it follows the spatial derivative of the $n_\downarrow(y)$ spatial profile, consistent with a spin-up spatial profile $n_\uparrow(y)$ that has shifted to the left and a spin-down profile $n_\downarrow(y)$ that has shifted to the right. The separation d between these two spin profiles following momentum relaxation can be extracted by fitting the open squares to a Gaussian then shifting two identical Gaussians by d until the difference fits the solid circles in Fig. 2. The solid curve in Fig. 2 corresponds to a fit for $d=2.8$ nm.

The results of repeating the measurements for a y polarized pump are represented by the solid triangles in Fig. 2. Two features are important. First, the sign of the derivative is opposite to the x polarized case, indicating that the spin-up currents and spin-down currents have reversed directions. Thus, the PSC depend upon the relative phase of the right and left circular components of the linearly polarized light, which suggests that the second term of the second equality in Eq. (3) dominates. Second, the magnitudes of the derivatives for the x polarized and the y polarized cases are approximately the same, which implies that $\mu^{yzxx} \cong -\mu^{yzyy}$. Consequently, both features indicate that $\dot{K}^{yz} \cong 2\mu^{yzxx} E_+ E_- \cos(\phi_+ - \phi_-)$. In this sense, the PSC can be viewed as arising from an interference between the absorption processes for the right and left circular components of the linearly polarized light.

For circularly polarized light, Eq. (2) predicts a charge current in the y direction ($\dot{J}^y \neq 0$). This current is sometimes referred to as the circular photogalvanic effect[7,8] (CPGE) and can be viewed as arising from the interference between the absorption of the x and y components of the field. The PSC that we measure here cannot be a CPGE current, since $\mu^{yzxx} \cong -\mu^{yzyy}$ requires that $\dot{K}^{yz}=0$ for circularly polarized light (i.e., for $E_x^2 = E_y^2$). Moreover, our PSC cannot be viewed as a simple superposition of two CPGE charge currents of equal magnitude: spin-down carriers generated by right circularly polarized light moving in one direction and spin-up carriers generated by left circularly polarized light moving in the opposite direction, since in general such a superposition requires no specific phase relationship between E_+ and E_-. In principle, the CPGE can indeed produce PSC through such a superposition with the appropriate lowering of the sample symmetry, but it makes a negligible contribution to the signals measured here.

We also measured the spin separation resulting from these PSC as a function of pump fluence (i.e., carrier density). The results are shown in Fig. 3. Despite the large uncertainties in the data (particularly at low injection densities), a decrease of the spin separation with increasing pump fluence and, therefore, with carrier density is clearly observed. We also show approximate momentum relaxation times extracted from the measured separations by modeling the carrier transport at the Boltzmann level in the relaxation time approximation and using values for the initial PSC injection calculated with a bulk 14 band $k \cdot p$ model.[1] Since each carrier is injected with an initial velocity of $\sim$900 km/s, these values can be crudely viewed as suggesting that less than 10% of the carriers are moving in the y direction with spins aligned along the z direction.

In summary, we have shown how interference between absorption pathways associated with opposite circularly polarized components of a single-color linearly polarized pulse can be used to generate and control a PSC in (110) GaAs at room temperature. We have measured the net displacements of the ballistically injected spins and find that they are in the range of 1–4 nm, depending on the excitation fluence.

This work was supported by DARPA, ONR, and NSERC. We thank Eric Gansen, Scot Hawkins, Fred Nastos, and Martin Stevens for stimulating discussions and suggestions.

*Email address: art-smirl@uiowa.edu

[1] R. D. R. Bhat, F. Nastos, Ali Najmaie, and J. E. Sipe, Phys. Rev. Lett. **94**, 096603 (2005).

[2] M. J. Stevens, A. L. Smirl, R. D. R. Bhat, A. Najmaie, J. E. Sipe, and H. M. van Driel, Phys. Rev. Lett. **90**, 136603 (2003).

[3] J. Hubner, W. W. Rühle, M. Klude, D. Hommel, R. D. R. Bhat, J. E. Sipe, and H. M van Driel, Phys. Rev. Lett. **90**, 216601 (2003).

[4] *Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, D. Loss, and N. Samarth (Springer-Verlag, Berlin, 2002).

[5] N. Laman, A. I. Shkrebtii, J. E. Sipe, and H. M. van Driel, Appl. Phys. Lett. **75**, 2581 (1999).

[6] D. J. Hilton and C. L. Tang, Phys. Rev. Lett. **89**, 146601 (2002).

[7] S. D. Ganichev and W. Prettl, J. Phys.: Condens. Matter **15**, R935 (2003).

[8] V. V. Bel'kov, S. D. Ganichev, P. Schneider, C. Back, M. Oestreich, J. Rudolph, D. Hägele, L. E. Golub, W. Wegscheider, and W. Prettl, Solid State Commun. **128**, 283 (2003).