Coherence Control of Hall Charge and Spin Currents

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Using two-color optical coherence control techniques in intrinsic GaAs at 80 K with orthogonally polarized 70 fs, 1430 and 715 nm pulses, we generate a *pure spin* source current that yields a transverse Hall *pure charge* current; or alternatively, with parallel polarized pulses, we generate a pure charge source current that yields a pure spin current. By varying the relative phase or polarization of the incident pulses, one can effectively tune the type, magnitude and direction of both the source and transverse currents without application of electric or magnetic fields.

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The possibility that an electron's spin may offer advantages to charge for the development of new devices that might be used in information technology has given birth to the field of spintronics [1]. If this effort is to succeed beyond well-established storage or memory applications, it will be necessary to develop new ways to generate, control, and otherwise manipulate spin in semiconductors. Much of the recent emphasis has been on engineering suitable materials with high spin injection efficiency and long spin lifetime that could be used with conventional, dc-field-driven devices; however, increasing attention is focusing on pure spin transport and current generation. In particular, following a prediction 35 years ago [2] and substantial additional theoretical effort [3], within the last two years two groups [4,5] have reported observations of the elusive spin Hall effect, whereby an electrical current produces a transverse spin current without a magnetic field. In the context of spintronics, besides its fundamental nature, the spin Hall effect provides new manipulation techniques for converting electrical currents into pure spin currents. However, there remains considerable debate over the existence of, and distinction between, intrinsic and extrinsic spin Hall effects [6,7] and, *inter alia*, the role of dc fields, impurities, strain and sample structure. In addition, the reverse effect, the production of a pure charge current from a pure spin current, has not been observed.

To date, the source currents for *all* reported Hall experiments, including the spin Hall experiments, have consisted of quasithermalized carriers drifting in response to dc applied fields, with spin accumulation measured at the edge of a sample. Here we show that coherence techniques and the phase or polarization of optical beams can provide new controls for generating and manipulating transverse or Hall currents in *intrinsic* semiconductors with a spin-orbit interaction. In particular, employing two-color quantum interference control (QUIC) techniques [8–15], we use 70 fs, 1430 and 715 nm pulses with bulk or quantum well GaAs samples to generate either pure spin or pure charge source currents in intrinsic GaAs, demonstrating how the former yields a transverse pure charge current and the latter, a transverse pure spin current. These currents occur in the absence of any external fields, electrical contacts, or material strain with the current produced away from a sample edge. Unlike drift currents consisting of thermalized carriers produced by dc electric fields, the carrier currents generated by QUIC are ballistic (i.e., carriers are not accelerated, but decelerate via scattering, following injection with a speed of $\sim$1000 km/s) and, therefore, will typically experience only one scattering event in establishing the Hall currents during their momentum scattering lifetime. The optical techniques can provide control on a time scale limited by the optical pulse duration and in locations and areas dictated by the pulse focal spot. By altering optical phase or beam polarization, we can control/tune the current type (spin, charge, QUIC, Hall) as well as their magnitude and direction. The use of optical phase in QUIC experiments may also provide new, beneficial links between optics and information technologies. Overall, QUIC currents may therefore provide a unique platform for studying current generation, manipulation, and detection, thus providing essential information necessary for any spin-based technology.

Figure 1(a) shows the configuration that allows a pure spin source current to be generated in a semiconductor crystal with band gap E_g. The inset illustrates the quantum interference between absorption pathways for one- and two-photon absorption connecting the same initial and final states that is responsible for current generation [8,9]. Specifically, a coherent pulse centered at ω with phase ϕ_ω is normally incident along $\hat{z}$, linearly polarized along an $\hat{x}$ direction that can be arbitrary since the effects discussed here are not strongly sensitive to crystal orientation [9]. A copropagating 2ω pulse with phase $\phi_{2\omega}$ is linearly polarized along the orthogonal $\hat{y}$ direction. Excited spin-up electrons ($\uparrow$) are polarized along $\hat{z}$ and move preferentially in one direction along $\hat{x}$, while spin-down electrons ($\downarrow$) move in the opposite direction. Together they generate a spin current $K^x \propto \cos(\Delta\phi)$ where

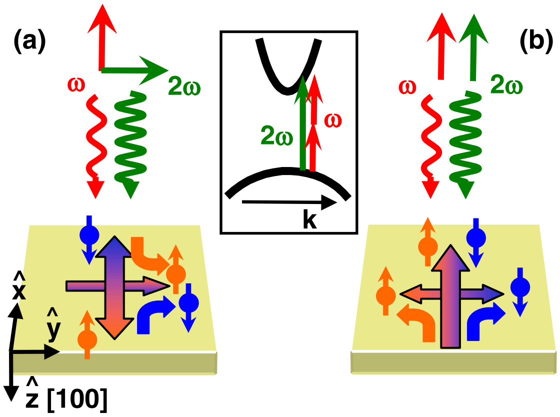

FIG. 1 (color online). (a) Illustration of orthogonally polarized ω and 2ω pulses producing a pure spin current (double headed straight arrow) along ω beam polarization direction ($\hat{x}$). A Hall charge current (curved arrows) and a transverse QUIC charge current along $\hat{y}$ lead to electron accumulation near one edge of the illuminated region. (b) Same as (a) except for collinearly polarized pulses producing a QUIC charge current along $\hat{x}$ and leading to a Hall pure spin current (curved arrows) and a transverse QUIC pure spin current (double headed arrow) resulting in up and down spin accumulations along $\pm\hat{y}$. Inset: schematic showing how ω and 2ω pulses connect valence and conduction band states for current generation.

$\Delta\phi = 2\phi_\omega - \phi_{2\omega}$. We define $K^x > 0$ if $\uparrow$ electrons preferentially move along $+\hat{x}$ with $\downarrow$ electrons preferentially moving along $-\hat{x}$ when $\Delta\phi = 0$. Optically injected holes move in opposition to electrons but lose their spin on a time scale <100 fs [16]; therefore, hole spin is ignored in subsequent discussions. Spin-dependent skew, side-jump [17,18], and perhaps other types of asymmetric scattering send $\uparrow$ and $\downarrow$ in the same direction along $\hat{y}$, yielding a Hall charge current, as shown by the curved arrows in Fig. 1(a). Since the spin current is the source for this Hall current, these two currents have the same $\Delta\phi$ dependence with the transverse current taking the form $J^y_{\rm Hall} = -A_{\rm Hall}\cos(\Delta\phi)$. Along with the Hall current, a transverse current produced by the QUIC process [straight transverse arrow in Fig. 1(a)] has been predicted [9], but not yet observed, with $J^y_{\rm QUIC} = A_{\rm QUIC}\sin(\Delta\phi)$, where $A_{\rm QUIC}$ and $A_{\rm Hall}$ are positive coefficients. The spin and charge currents decay via momentum relaxation, although the latter is also influenced by space charge effects. Note that, in comparison with other observations of the spin Hall effect, our sample is intrinsic, with the holes playing the role of spin-scattering "impurities"; the currents are ballistic and can be produced anywhere on the sample. Given that the spin-dependent scattering time is much longer than a typical momentum relaxation time, one can expect that $\sim$ a single scattering event will lead to a particular electron contributing to the Hall current.

For the experiments, a sample consisting of 10 periods of 14 nm wide GaAs wells ($E_g = 1.542$ eV at 80 K) alternating with 14 nm thick $Al_{0.3}Ga_{0.7}As$ barriers was grown on a (100)-oriented GaAs substrate (removed for the experiments). All effects reported here have also been observed in bulk, intrinsic GaAs. An optical parametric oscillator produces 70 fs (full width at half maximum) ω pulses centered at 1430 nm, with 2ω pulses centered at 715 nm obtained by frequency doubling. The $\Delta\phi$ is controlled using a scanning interferometer and pulse polarizations are selected independently. The pulses are cofocused to a $w \sim 3$ μm diameter spot on the sample. For $2\hbar\omega - E_g \sim 193$ meV, the electrons are injected with a speed $\sim 10^3$ km/s. For a 2ω pulse fluence of 1.5 μJ/cm^2, the injected electron density is $\sim 10^{17}$ cm^{-3}; the ω pulse fluence is set to generate approximately the same density. For this density the momentum relaxation time is ~ 100 fs, governed by phonon and carrier-carrier scattering [19].

The source spin current is detected via spin accumulation that follows spin motion [12]; such accumulation is proportional to the source current. Upon injection, $\uparrow$ and $\downarrow$ electrons have the same $\sim$ Gaussian spatial profile with the same peak density, i.e., $N_\downarrow(x, y, t = 0) = N_\uparrow(x, y, t = 0)$. The two profiles move in opposite directions and separate by an effective distance L_S by the time momentum and skew scattering are complete. This spin separation persists long after the currents have vanished, decaying eventually by spin relaxation, carrier diffusion, and recombination as we have independently verified. The magnitude of L_S is determined by: (i) the fraction of the total injected density that is spin polarized and that travels along $\hat{x}$; (ii) the initial injection velocity [20]; and (iii) the momentum relaxation time. For our experimental conditions, L_S is expected to be <100 nm, and therefore, we use a derivative technique to achieve sub-optical-wavelength resolution [12]. When $L_S \ll w$, the net spin accumulation $\Delta S = N_\uparrow(x, y) - N_\downarrow(x, y) \sim L_S dN_\uparrow/dx$, with the sign giving the spin flow direction. The ΔS, centered on (x, y), is measured using a 100 fs, 808 nm, linearly polarized probe pulse that is focused onto the sample with a ~ 3 μm (FWHM) diameter. The probe arrives ~ 10 ps after the pump pulses by which time carrier thermalization and momentum relaxation are complete, but not electron recombination or spin relaxation. The probe spectrum is centered on the heavy-hole exciton for efficient sampling of $N_\downarrow$ and $N_\uparrow$. The efficiency of this sampling is the reason for choosing to present results for the quantum well sample. Circular dichroism, i.e., absorption difference for right and left circularly polarized probe beam components in the presence of $\uparrow$ and $\downarrow$ electrons, is used to deduce [12] ΔS.

The transverse charge current is similarly measured via electron (and hole) accumulation [11]. For the charge current, $\uparrow$ and $\downarrow$ electrons move in the same direction and so the entire charge profile moves by L_C, where $\Delta N(x, y) = L_C dN/dx$ and $N(x, y)$ denotes the total electron density. In contrast to L_S, the magnitude of L_C is influenced by space charge fields produced by electron-hole separation; thus, L_C and L_S cannot be directly compared without additional information. The $N(x, y)$ is measured through differential transmission $\Delta T(N)$ (i.e., the transmission with and without pump beams) of a linearly polarized probe pulse, for which $\Delta T(N)/T \propto N$ when $\Delta T(N) \ll 1$, where T is the linear transmission. Finally, ΔN is measured by monitoring the phase-dependent component of the differential transmission $\Delta T(\Delta\phi)/T \propto \Delta N$ with a lock-in amplifier slaved to the interferometer scanning frequency. Using these procedures, we simultaneously measure ΔS and ΔN at each (x, y) in the spot as a function of $\Delta\phi$.

Figures 2(a) and 2(b) show the measured $\Delta\phi$ dependence of ΔS and ΔN as the probe is scanned first along $\hat{x}$ (for $y = 0$), then along $\hat{y}$ (for $x = 0$) with the pump focal spot center defining $x = y = 0$. The scans along $\hat{x}$ [Fig. 2(a)] show $\uparrow$ accumulation for one sign of x, $\downarrow$ accumulation for the other sign, and periodic reversal with $\Delta\phi$. No charge accumulation is measured anywhere along $\hat{x}$. These observations confirm a pure spin source current along $\hat{x}$. The scans along $\hat{y}$ [Fig. 2(b)], which show regions of charge accumulation that oscillate with $\Delta\phi$, confirm a transverse charge current along $\hat{y}$. The charge current has a different $\Delta\phi$ dependence from the spin current, suggesting the presence of in-phase (Hall) and quadrature (QUIC) components.

The Hall and QUIC contributions to the transverse current can be unambiguously distinguished by measuring the charge and spin accumulations as a function of (x, y) for $\Delta\phi = 0$ and $\Delta\phi = \pi/2$, as shown in Fig. 3. For the data in Figs. 3(a) and 3(b) we adjusted $\Delta\phi$ so that the source current is maximal ($\Delta\phi = 0, \pi, 2\pi \ldots$), with the transverse QUIC current therefore zero. Under these conditions, the ΔN shown in Fig. 3(b) results from the Hall current generated by the source spin current displayed in Fig. 3(a). For Figs. 3(c) and 3(d) $\Delta\phi$ is chosen so that the spin source current, and therefore, the Hall current is zero. Figure 3(d) is thus a contour plot of the ΔN produced by the transverse QUIC current predicted theoretically [9]. Through the appropriate choice of $\Delta\phi$ one can "tune" the source current magnitude and direction as well as that of the two transverse currents.

By polarizing both beams along the same ($\hat{x}$) direction as depicted in Fig. 1(b) one can produce a QUIC source charge current of the form [8] $\mathbf{J}^x \propto \sin(\Delta\phi)$ that is capable of producing the ballistic analogue to the previously observed spin Hall current [4,5]. In this case, spin-dependent scattering deflects $\uparrow$ and $\downarrow$ electrons in opposite directions along $\hat{y}$ to produce a pure spin current given by $K^y_{\rm Hall} = -B_{\rm Hall}\sin(\Delta\phi)$. Also along $\hat{y}$, a current of the form $K^y_{\rm QUIC} = -B_{\rm QUIC}\cos(\Delta\phi)$ is generated by the QUIC process, as earlier predicted [9], but not yet observed. The procedures used to measure the charge and spin currents via accumulation effects are analogous to those used to

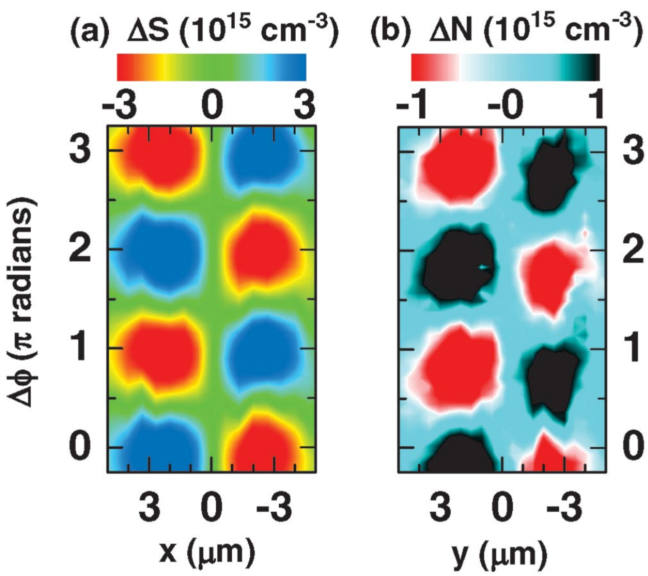

FIG. 2 (color). (a) Contour map showing dependence of ΔS induced by a source spin current on $\Delta\phi$ and position along $\hat{x}$ ($y = 0$). (b) Corresponding contour map of combined Hall and QUIC ΔN measured along $\hat{y}$ ($x = 0$).

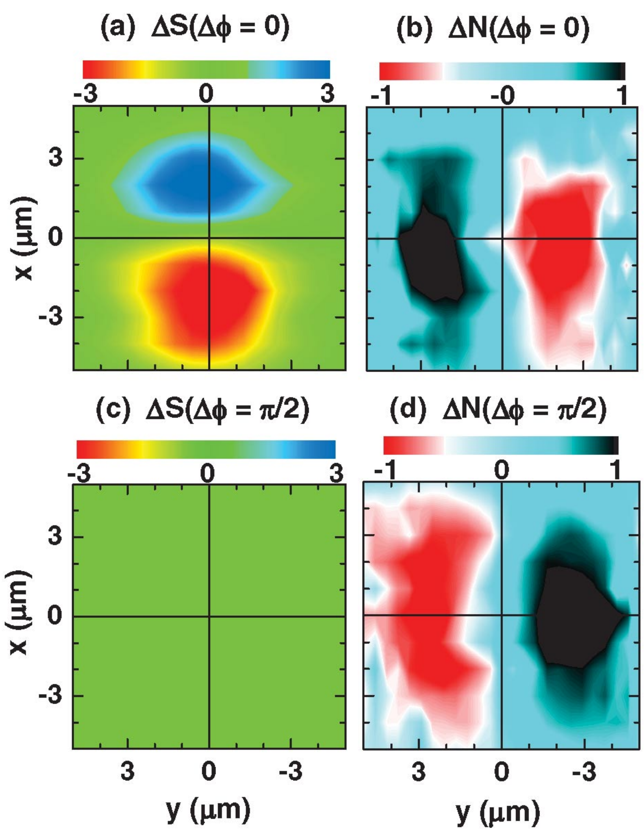

FIG. 3 (color). Spatial contour maps showing: (a) the spin ΔS and (b) the charge ΔN accumulations for $\Delta\phi = 0$ associated with the pure spin source and Hall charge currents, respectively; (c) and (d) show ΔS and ΔN for $\Delta\phi = \pi/2$, when the pure spin source current is zero and only a transverse QUIC charge current is present. Approximate spin and charge densities can be obtained by multiplying ΔS and ΔN by 10^{15} cm^{-3}.

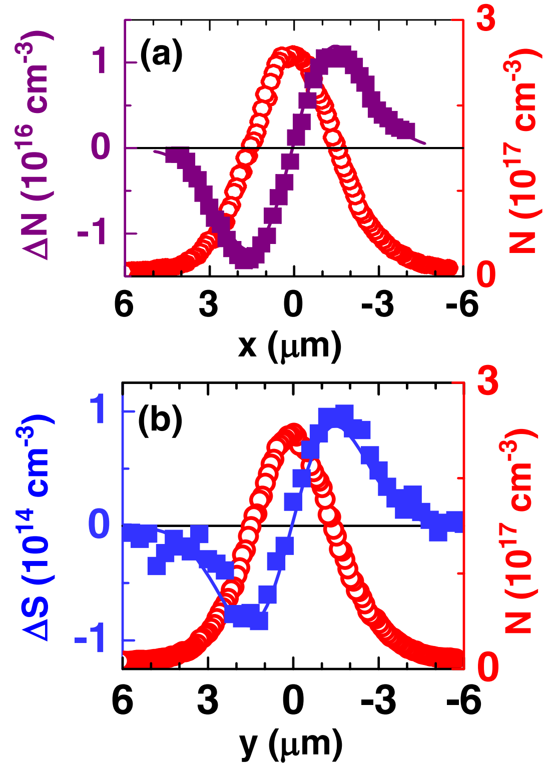

FIG. 4 (color online). (a) N (circles) and ΔN (squares) caused by the source charge current and measured along $\hat{x}$ with $\Delta\phi = \pi/2$, for which the source current is maximum. (b) N (circles) and ΔS (squares) caused by the transverse pure spin current and measured along $\hat{y}$ with $\Delta\phi = 0.12\pi$, for which ΔS is a maximum.

obtain the data shown in Figs. 2 and 3. Specifically, the $\Delta\phi$ dependence of the ΔN and ΔS associated with the source and transverse currents are simultaneously measured at each spatial position. The charge accumulation and the spin accumulation produced by their respective currents are shown in Figs. 4(a) and 4(b), respectively, together with the x and y cross sections of the carrier spatial profile.

In conclusion we have shown that optical quantum interference effects can be used to generate and tune pure spin or pure charge source currents on fast time scales via optical phase and polarization. Associated with each of these are two types of transverse currents, one due to spin-dependent scattering or a spin Hall effect and the other a QUIC current. In particular, the source spin currents lead to transverse charge currents and *vice versa*. Optical phase and polarization can be used to control or tune the current type (spin, charge, Hall, QUIC), including their direction and magnitude and between *spin* and *charge* Hall effects. While these effects may be of interest for future spintronic applications, they also, with further study, may provide insight into the nature of spin-dependent scattering processes since, unlike the limited data which exists from measurements based on steady-state experiments in samples containing impurities, our results are based on pulsed sources, an intrinsic semiconductor, and injected ballistic currents with electrons traveling initially ~1000 km/s.

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