Optical injection and detection of ballistic pure spin currents in Ge

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Ballistic pure spin currents are injected into Ge at 295 K using quantum interference between one and two photon absorption processes for 1786 and 893 nm, 200 fs optical pulses. The spin currents are spatially and temporally detected using polarization- and phase-dependent differential transmission techniques with nanometer spatial and femtosecond temporal resolution. We interpret the dynamics in terms of the fast spin relaxation of the holes and intervalley transfer of electrons. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3222869]

The generation of spin currents is one of the goals of spintronics. Using electronic techniques, it has been possible to generate spin polarized charge currents in ferromagnetic metals or magnetic semiconductors,[1] and pure spin currents (PSCs) in quantum dots[2] or via the spin Hall effect in bulk GaAs.[3] However, PSCs have not been produced in a group IV semiconductor, and to date very little research in spintronics has involved such materials although they form the platform for much of electronics technology.

In this letter, we report the generation and detection of a transient PSC in germanium at 295 K. As demonstrated earlier in GaAs,[4–8] we produce and detect spin currents using a purely optical technique in which ballistic PSC are injected through the interference between two photon absorption of a linearly polarized pulse with frequency ω and the single photon absorption of an orthogonally polarized second harmonic pulse at 2ω. This quantum interference and control (QUIC) technique is noninvasive and allows precise control of the injected currents.[8] Once injected, however, the currents quickly decay by spin momentum relaxation on subpicosecond time scales. The spin motion is monitored[8] by measuring the dichroism of the *phase-dependent* differential transmission in space and time, a sensitive technique for measuring spin currents.

Earlier demonstrations[6–8] of QUIC induced PSC injection employed GaAs since it is a direct band gap material and is the semiconductor most frequently used for photonic applications. On the other hand, group IV are the most common materials for electronic (i.e., transport) applications, but are not widely used in photonics since they are centrosymmetric, indirect band gap materials with $\chi^{(2)} \equiv 0$. Nonetheless, considerable effort has been expended to render them so.[9] QUIC techniques are based on third order optical processes, which are allowed in centrosymmetric and noncentrosymmetric materials, and they relate transport and photonic effects, which may lead to applications in group IV materials. Germanium is chosen because, like GaAs, it allows the resonant injection of ballistic currents across the *direct* gap [see Fig. 1(b)] and because it has a strong spin-orbit coupling, offering the prospect of injecting and controlling spin.

The procedure that we use to inject PSCs into Ge is similar to that described previously[8] for GaAs. As suggested schematically in Fig. 1, an ~200 fs (full width at half maximum, FWHM) fundamental pulse ($\lambda = 1786$ nm, $\hbar\omega = 0.6945$ eV) with a phase ϕ_ω and a 2ω pulse with a phase $\phi_{2\omega}$ copropagate along the z-direction ([111] direction) and are tightly focused to produce an excited carrier profile with a diameter $W \sim 2$ μm (FWHM) in a 40 Ω cm and 1-μm-thick layer of bulk Ge. The sample was prepared from the same boule using the same technique described previously.[10] Instead of the piezoelectric transducer used in our previous measurements,[8] a scanning phase modulator controls the phase difference $\Delta\phi = 2\phi_\omega - \phi_{2\omega}$ in the 2ω arm of a dichroic interferometer. (The dispersion of the modulator broadens the 2ω pump to ~300 fs, FWHM.)

In Ge, as in GaAs,[8] the one-photon absorption of 2ω and two-photon absorption of ω connect the same states in the heavy-hole or light-hole valence bands and the direct conduction band valley ($\hbar\omega < E_\Gamma < 2\hbar\omega$, where the direct gap at Γ, $E_\Gamma = 0.805$ eV at 295 K). However, in contrast to GaAs, these processes also couple the split-off valence band (spin orbit splitting $\Delta = 0.295$) to the conduction band at Γ

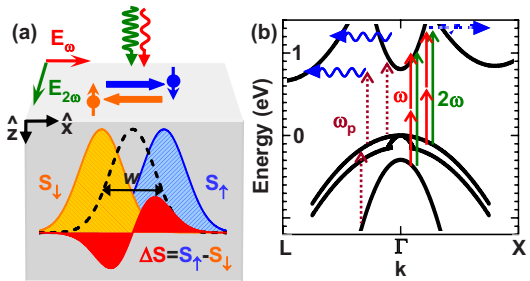

FIG. 1. (Color online) (a) Schematic showing ω and 2ω pump pulses with orthogonal linear polarizations injecting a PSC: spin up (down) carriers move to the right (left) causing a change in the spin density (red filled curve). (b) Corresponding excitation scheme showing the ω and 2ω pump pulses coupling the same initial states in the heavy-hole, light-hole, and split-off valence bands and final states in the conduction band of Ge, and showing the direct, indirect, and intervalence band transitions associated with the absorption of the probe pulse (ω_p).

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[Fig. 1(b)]. The peak 2ω fluence is restricted to $\sim 100\ \mu J/cm^2$, corresponding to a peak carrier density of $\sim 10^{19}\ cm^{-3}$. The ω pulse produces a density that is three times as large.

The PSC injection process and subsequent spin dynamics in Ge are expected to be similar to those in GaAs. Orthogonally polarized ω and 2ω pulses[4] inject identical Gaussian spatial profiles of spin up ($S_\uparrow$) and spin down ($S_\downarrow$) polarized carriers (electrons and holes) with oppositely directed net average velocities $v_\uparrow \cos\Delta\phi$ and $v_\downarrow \cos\Delta\phi$, respectively [Fig. 1(a)]. The two spin polarized carrier profiles move apart (a distance L_s) until spin momentum relaxation is complete. Equal numbers of electrons (and holes) move in each direction; therefore, there is no net charge transport, and no space charge field is formed. The profiles remain separated until diffusion, recombination, or spin relaxation destroys them. If $L_s \ll w$ (the width of $S_\uparrow$ or $S_\downarrow$), the net spin density, $\Delta S = S_\uparrow - S_\downarrow$, is proportional to the derivative of the original profile.[4,8]

The redistribution of spin ΔS is monitored using a linearly polarized probe pulse at 1450 nm ($\hbar\omega_p = 0.855$ eV). For the carrier densities injected here, details[10] of the probe absorption are complicated, since direct,[11] indirect,[11] free carrier,[12,13] and intervalence band[10,14,15] (split-off to heavy and light hole) transitions are all allowed [Fig. 1(b)] and band gap narrowing[16,17] contributes to the change in absorption. Nevertheless, in the regime where the differential transmission is separately shown to be approximately proportional to the electron and hole densities, ΔS is proportional to the *circular dichroism* in the phase-dependent differential transmission: $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$, where $\delta T^\pm(\Delta\phi) = [T^\pm(\Delta\phi) - T^\pm(\Delta\phi = \pi/2)]$ are the differential transmissions for the right (+) and left (−) circular components of the probe and where $T^\pm(\Delta\phi)[T^\pm(\Delta\phi = \pi/2)]$ are the transmissions for the right and left circular components with [without] current injection.[8] $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$ is measured by using an optical bridge consisting of a 1/4 wave plate, Wollaston prism and a balanced detector, as described previously.[8]

The spatial dependence of $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$ ($\propto \Delta S$) is illustrated in Fig. 2 for a fixed phase of $\Delta\phi = \pi$ (where PSC injection is maximum) and a fixed time delay τ between the pumps and probe. The phase dependence of $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$ is shown in the inset for the same fixed τ and for two fixed positions on the sample (one on the right side; the other on the left). Together, these two figures demonstrate that for a fixed phase (e.g., $\Delta\phi = \pi$) spin down polarized carriers accumulate on one side of the sample while spin up accumulate on the other and that the positions of the spin up and spin down carriers are smoothly exchanged as the phase is varied by multiples of π. The cosinusoidal dependence of $\delta T(\Delta\phi)^+/T - \delta T(\Delta\phi)^-/T$ on phase and the derivativelike spatial profiles provide convincing evidence of PSC injection.

The dynamics of the spin density, ΔS, are investigated by measuring $\delta T(\Delta\phi)^+/T - \delta T(\Delta\phi)^-/T$ as a function of τ at a fixed position and phase (Fig. 3). Notice that the temporal profile of ΔS has roughly the same width as the autocorrelation of the ω pump pulse, suggesting that it decays on a time scale comparable to the optical pulse widths. This behavior is in contrast to observations in GaAs,[8] where $\delta T(\Delta\phi)^+/T - \delta T(\Delta\phi)^-/T$ persists for hundreds of picoseconds. In the GaAs experiments, the probe was primarily sensitive to the electrons, and the separation of the spin profiles (or ΔS) remained until *electronic* spin relaxation, recombination and diffusion, all of which occur on 100 ps time scales, were complete.

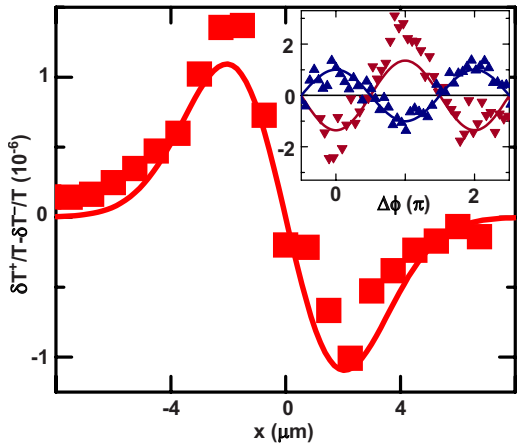

FIG. 2. (Color online) The spatial profile of the circular dichroism of the phase-dependent differential transmission, $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$ ($\propto$ the spin density ΔS), as a function of x with $y=0$ (filled squares) for $\tau = 122$ fs and for a fixed $\Delta\phi \approx \pi$. The solid line is a fit to the data using the derivative of a Gaussian. The inset shows the phase ($\Delta\phi$) dependence of the same quantity for the same fixed τ and at two fixed positions $x = 2.25\ \mu$m, $y=0$ (blue up triangles) $x = -2.25\ \mu$m, $y=0$ (red down triangles). The solid lines are cosinusoidal fits to the data.

The fast [compared to GaAs (Ref. 8)] decay of ΔS in Ge is either a consequence of the rapid scattering of the electrons to the side valleys or of the ultrafast spin relaxation of the holes; however, the extent of the work presented here does not allow us to determine their relative roles. If, for example, $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$ is dominated by bleaching of the probe absorption between heavy-hole valence and Γ-conduction band, then the probe absorption change is: $\Delta\alpha_{hh-\Gamma} \propto -f_e(E^\Gamma_{hh-\Gamma}) - f_h(E^{hh}_{hh-\Gamma})$, where $f_{e(h)}$ is the electron (hole) distribution function evaluated at the optically coupled

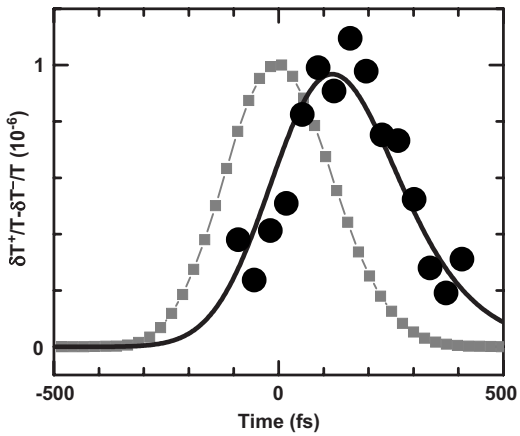

FIG. 3. The temporal dynamics (solid circles) of $\delta T^+(\Delta\phi)/T - \delta T^-(\Delta\phi)/T$ at a fixed position $x = 2.25\ \mu$m, $y=0$ and phase $\Delta\phi \approx \pi$. The separately measured autocorrelation of the ω pulse (solid squares) is shown for comparison. The solid line is primarily intended as a guide to the eye, but is also the result of a simulation assuming a rigid shift of the spin profiles (Ref. 8) and taking the momentum and hole spin (or electron intervalley) relaxation times to be 100 fs. Spatial and temporal pump-probe convolutions and finite carrier generation are taken into account. The only fit parameter is the separation of the spin profiles L_s.

energy $E^{\Gamma}_{\mathrm{hh}-\Gamma}(E^{\mathrm{hh}}_{\mathrm{hh}-\Gamma})$ in the Γ conduction band valley (heavy-hole band). Electrons and holes are injected with a great deal of excess energy ($2\hbar\omega - E_{\Gamma} \cong 600$ meV). If the electrons thermalize and/or relax to the bottom of the Γ-valley *before* they scatter to the *X* or *L*-valleys, $f_e > f_h$, and the electrons will dominate the probe differential transmission. Following injection, the spin profiles move apart and remain separated, but the electrons rapidly scatter to the side valleys, where they no longer directly influence the probe transmission. In this case, the decay of $\delta T(\Delta\phi)^+/T - \delta T(\Delta\phi)^-/T$ is a measure of the intervalley scattering time for the electrons.

Indeed, intervalley scattering times in the range of 200–300 fs have been measured[17] for carrier densities of $\sim 10^{17}$ cm^{-3} injected near the Γ-band edge, consistent with the decay of ΔS shown in Fig. 3. However, our electron density is 100 times larger, and electrons are injected much higher in the Γ-valley than in Ref. 17. Both are expected to decrease the scattering and thermalization times. Moreover, electrons must emit ~20 optic phonons to reach lattice temperature. Thus, it is likely that the electrons will scatter to the side valleys, while thermalizing, before occupying the near-band edge states interrogated by the probe. Under these circumstances (assuming a nondegenerate distribution at 295 K), only $\sim 10^{-4}$ of the initially injected electrons remain in the central valley, and $f_h \sim 10^3 f_e$. In this case, $\delta T(\Delta\phi)^+/T - \delta T(\Delta\phi)^-/T$ is strongly dominated by the holes, and its decay is determined by the spin relaxation of holes. The hole spin relaxation in Ge has not been directly measured, but is expected to occur on a 100 fs time scale,[18] consistent with our observations. At the carrier densities encountered here, intervalence band absorption will also contribute,[10,15] and once the carriers cool, the distribution will be degenerate.[15] These considerations make quantitative differences in ΔS, but do not change the qualitative nature of the arguments above.

In summary, we have demonstrated all-optical spin injection in Ge using quantum interference processes and have detected the signatures of the spin current through measurement of the induced circular dichroism. As in GaAs, the currents remain for a time of the order of the spin momentum relaxation time. Moreover, the efficiency of generating, and the lifetime of, spin currents in Ge via the QUIC scheme might be comparable to that of GaAs; however, because the electron intervalley scattering and the hole spin relaxation times are short, the spin signatures (as detected by circular dichroism) also are short lived and weaker in Ge than in GaAs. Finally, because these two relaxation times are similar in magnitude, it is not possible at present to determine the relative roles of electrons and holes in defining the detected signal or the spin currents.

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