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Quantum interference injection and control of spin-polarized transient current gratings in GaAs

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Abstract
We demonstrate the creation of a transient spin-polarized current grating in bulk GaAs at room temperature. The spin current is injected through quantum interference between two-photon absorption of fundamental (1.55 μm) and one-photon absorption of second harmonic (775 nm) pulses. Because the fundamental and second harmonic pulses do not propagate collinearly, a spin current grating is formed, which decays by electron diffusion.

Control and manipulation of spin have attracted a great deal of attention in the last two decades [1]. The inherently long relaxation time of spin and its quantum mechanical nature are two of the main reasons for the new emerging field of spintronics. One challenge facing this field is learning to inject and control spin-polarized currents in semiconductors. In addition to the electrical injection of spin currents [2, 3], recently, it has been shown that it is also possible to use optical pulses and quantum interference and control (QUIC) techniques to generate spin currents in semiconductors without applying an electrical bias [4].

Typically, QUIC currents are generated through the quantum interference between the pathways for one- and two-photon absorption in a semiconductor. This quantum interference can cause the density or the spin (or both) of the injected carriers to be asymmetric in k-space, resulting in a charge current or a spin current. The magnitude, direction and spin of the currents produced by QUIC depend on the relative phase between the two incident optical pulses, their polarizations and on the orientation and symmetry of the crystal. For example, unpolarized charge currents [5] have been produced using two pulses with parallel linear polarizations; spin-polarized charge currents [6] have been observed using the same circular polarizations; and pure spin currents [7, 8] (with no accompanying charge currents) have been reported using orthogonal linear polarizations.

In all previous QUIC experiments, the 2ω (second harmonic) and ω (fundamental) pulses were collinearly propagating, and the relative phase between the two pulses was temporally controlled by a scanning dichroic Michelson interferometer. However, here, we automatically and periodically vary the relative phase across the sample surface by using non-collinearly propagating pulses. Specifically, by using non-collinearly propagating ω and 2ω pulses with orthogonal linear polarizations, we are able to produce pure spin current gratings, without any accompanying charge or population gratings, by quantum interference in bulk [001]-oriented GaAs at room temperature.

The experimental geometry that we use is shown schematically in figure 1. The ω and 2ω pulses make an angle θ with the normal to the sample (z axis), and the ω beam is s-polarized and the 2ω beam p-polarized. For this geometry, the total optical electric field can be written

$$\mathbf{E}(t) = E_\omega \exp(-\mathrm{i}\omega t + \mathrm{i}\mathbf{k}_\omega \cdot \mathbf{r} + \mathrm{i}\phi_\omega)[\cos\theta\,\hat{\mathbf{x}} + \sin\theta\,\hat{\mathbf{z}}] + E_{2\omega}\exp(-\mathrm{i}2\omega t + \mathrm{i}\mathbf{k}_{2\omega}\cdot\mathbf{r} + \mathrm{i}\phi_{2\omega})\hat{\mathbf{y}} + \mathrm{cc}.$$

where E_ω ($E_{2\omega}$), $\mathbf{k}_\omega$ ($\mathbf{k}_{2\omega}$) and ϕ_ω ($\phi_{2\omega}$) are the field envelope, propagation vector and phase, respectively, of the fundamental (second harmonic) pulse. Under these conditions, theory [9] predicts that initially, at each point, two *ballistic* currents will be produced with equal magnitudes, opposite propagation directions and opposite spins:

$$\dot{\mathbf{J}}^{\pm z}_{\pm x} = \pm \dot{J}_1 \cos(2\pi x/\Lambda + \Delta\phi)\hat{\mathbf{x}}$$

where $\Delta\phi = 2\phi_\omega - \phi_\omega$; $\Lambda = \lambda_\omega/4\ \sin\theta$ is the spatial period of the spin-polarized current grating; and λ_ω is the wavelength

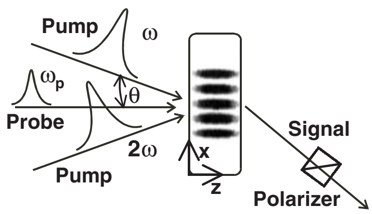

Figure 1. Geometry for producing spin-polarized transient spin current gratings by quantum interference and control using pump beams at frequencies ω and 2ω and monitoring the grating formation and decay using probe pulses at ω_p.

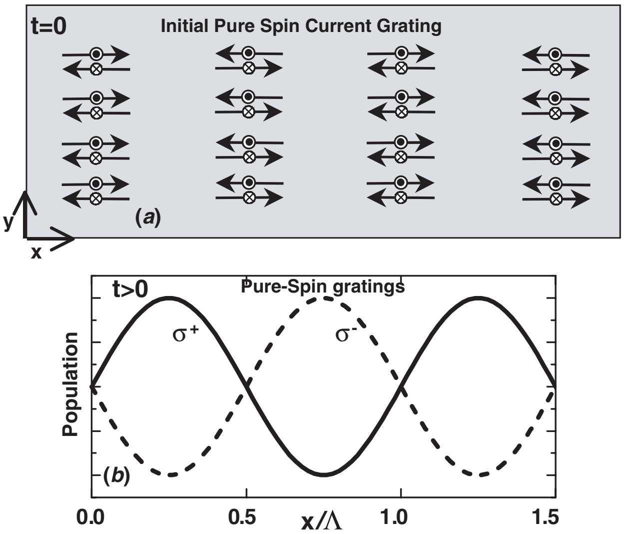

Figure 2. Schematic diagram depicting (a) the initial ($t = 0$) spin grating consisting of a periodic modulation of the amplitude of equal and opposite spin-polarized currents (no charge current) at each position in space produced by pump pulses with orthogonal linear polarizations and (b) the pure spin-polarized population gratings ($t > 0$) produced by this spin current. The larger arrows show the directions of current flow. The smaller circles indicate the direction of the net spin polarization of the electrons (into or out of the page), and σ^+ (σ^-) denotes the spin 'down' (up) population. Note that there is no modulation of the total electron population.

of the ω beam. $\dot{J}_1$ can be expressed in terms of the material parameters and the intensity of the pulses.

Note that the magnitude of each of these currents varies periodically across the sample surface due to the change in relative phase between the non-collinearly propagating pulses. However, since one current, $\mathbf{J}^{+\mathbf{z}}_{+\mathbf{x}}$, always travels along the $+x$ direction (with its spin orientation along $+z$) and the other, $\mathbf{J}^{-\mathbf{z}}_{-\mathbf{x}}$, travels in the opposite direction $-x$ (with the spin orientation along $-z$), there is no spatial modulation of the net charge current, the population or the spin immediately following the incident pulses. Consequently, two *spin-polarized ballistic current* gratings are produced that are exactly out of phase spatially, as depicted in figure 2(a).

Until momentum relaxation destroys these currents, ballistic transport will cause electrons with one spin to accumulate at some positions and electrons with the opposite spin to accumulate at other positions. This transport will cause the formation of two spin-polarized population gratings that are exactly out of phase by 180°, as illustrated in figure 2(b). It should be noted that the total carrier density is still uniform

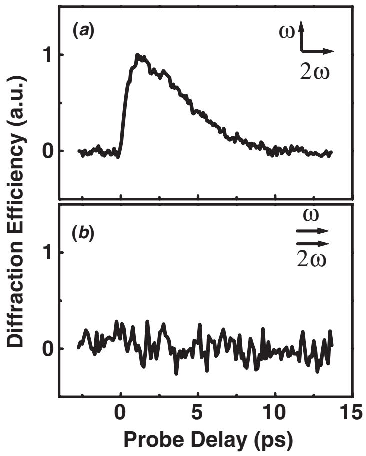

Figure 3. The p-polarized component of the diffracted signal from an s-polarized probe (a) when the ω pump pulse is p-polarized and the 2ω pump pulse is s-polarized and (b) when the ω and 2ω pump pulses are both s-polarized. A spin grating is formed in (a), but not in (b). The larger noise in (b) compared to (a) is due to the use of different beam modulation techniques.

across the sample, and therefore, no net population grating is formed.

We then use a linear s-polarized time-delayed probe pulse to monitor the dynamics of the population gratings produced by the two pump beams. An s-polarized probe would be expected to become p-polarized upon diffraction from two spatially-orthogonal spin-polarized population gratings. To understand this, it is perhaps simplest to think of the s-polarized probe in terms of its right and left circularly polarized components. The right (left) circular component will couple to, and diffract from, the spin down (up) population grating (respectively). The spatial shift between the two gratings produces a 180° phase shift between the diffracted right and left circular polarized components, thus, causing a 90° rotation of the incident polarization.

To experimentally demonstrate these spin current gratings in bulk [001]-oriented GaAs at room temperature (E_g = 1.42 eV), a Ti:sapphire laser, which is regeneratively-amplified at 250 kHz, is used to pump an optical parametric amplifier tuned to produce 150 fs signal pulses at 1550 nm and idler pulses at 1650 nm. A BBO crystal is used to generate 775 nm pulses (2ω) from the signal beam (ω), and another BBO crystal is angle-tuned to generate 840 nm probe pulses (ω_p) from the idler beam. The two orthogonal linearly polarized pumps and the probe are focused onto the sample. The pump pulses make a half angle $\theta = 10°$ with respect to the normal to the sample. Appropriate wave plates and polarizers enable us to control the polarization state of all three beams. The irradiances of 2ω and ω pump beams ($\sim$550 MW cm^{-2} and $\sim$7 GW cm^{-2}, respectively) have been chosen so that the rates of carrier generation by one- and two-photon absorptions are nearly equal and the peak carrier concentration is $\sim 10^{18}$ cm^{-3}.

Figure 3(a) shows the p-polarized diffracted component of the s-polarized incident probe as a function of the time delay between the probe and the pump beams. Three features are evident. First, the polarization of the diffracted signal is orthogonal to the polarization of the incident probe pulse, consistent with scattering from two oppositely spin-polarized population gratings that are spatially out-of-phase by π. Second, the diffracted signal initially follows the integral of the pump pulse envelope, confirming that the grating formation (determined by the momentum relaxation time) is short compared to our pulse width. Third, this pulse-width-limited grating formation process is followed by a decay of $\sim$3.2 ps, which is consistent with an electronic diffusion coefficient of $D_e = 390$ cm^2 s^{-1} and a mobility of $\mu_e = 7700 \pm 800$ cm^2 V^{-1} s^{-1}, a value that is in reasonable agreement with the electron mobilities that have been reported previously [10]. Since the hole spin relaxation time is very fast [11], the decay of a pure spin grating is expected to be determined by electron diffusion, since electron spin relaxation and electron–hole recombination are slower. Thus, these gratings have all of the characteristics expected of pure spin gratings. By comparison, in figure 3(*b*), it can be seen that no grating is formed for parallel linearly polarized beams as predicted by theory [9].

To ensure that the grating measured in figure 3(*a*) is a pure spin grating, in a separate experiment, we generated a pure spin grating by interfering two non-collinear and orthogonally polarized pump beams *with the same frequency* using the same geometry. As expected [12], we measure the same decay time as in figure 3(*a*).

In addition, we have carried out several tests to ensure that the grating measured in figure 3(*a*) is actually formed from the quantum interference between ω and 2ω and not, for example, from classical interference between 2ω in one arm and the leakage of 2ω in the other arm. It is also important to show that this spin grating is not generated by population control as opposed to current control. If the grating is formed by population control, it can be shown that the grating amplitude will depend strongly on the sample orientation [13]. Experimentally we have not observed a strong dependence, a clear indication that the grating is not formed by population control.

In summary, we have shown that the quantum interference between the probability amplitudes for the one- and two-photon absorptions of two non-collinearly propagating 2ω and ω pump pulses with orthogonal linear polarizations produces two spin-polarized ballistic current gratings. It is important to emphasize that in previous conventional transient grating experiments, e.g., [13], the interference that produced the grating was a classical interference between the two coherent optical fields, and it was the carrier population (spin polarized, or not) that was periodically modulated. Here, by contrast, the interference is between quantum mechanical wavefunctions, and it is the magnitudes and directions of the ballistic spin *currents* that are spatially modulated. These results are a significant first step in using quantum interference and control techniques in concert with transient grating techniques to inject and control spin-polarized currents in semiconductors and to study the transport associated with those spin polarized carriers.

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