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Pure spin current gratings in semiconductors generated by quantum interference

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We demonstrate that the quantum mechanical interference between the probability amplitudes for the two-photon absorption of a fundamental $(1.55\ \mu\mathrm{m})\sim 150$ fs pulse and for the one-photon absorption of a *noncollinearly* propagating second-harmonic (775 nm) pulse can create transient, ballistic, purely spin-polarized current gratings in bulk GaAs at room temperature. For fundamental and second-harmonic pulses having orthogonal linear polarizations, two periodically modulated ballistic spin-polarized current gratings are injected that have opposite spins and opposite propagation directions at each point along the grating. Consequently, there is no initial modulation of the charge current, carrier population, or net spin. Before the carrier momentum relaxes, the transport associated with these spin currents forms two oppositely spin-polarized population gratings that are exactly out of phase spatially and that decay by electronic spin diffusion in a time of 3.2 ps. In addition, charge density gratings are directly produced by the quantum interference process, and they decay by ambipolar diffusion and recombination (~17.6 ps). The polarization selection rules and sample orientation are used to separate the contributions of the current and density gratings. [DOI: 10.1063/1.1757033]

I. INTRODUCTION

In the last two decades, there has been growing interest in controlling the spin of electrons and holes in semiconductors.[1] The inherently long coherence time of spin as well as its quantum mechanical nature offer opportunities for the development of spin-based quantum information devices. Consequently, it is important to understand and control the generation, transport, and detection of spin-polarized carriers. Here, we report the use of a two-frequency-induced transient grating technique, which is based on the interference in the quantum mechanical transition amplitudes for the one- and two-photon absorption processes, to coherently inject ballistic spin-polarized currents and, subsequently, to monitor the transport of the spin-polarized carriers.

All-optical quantum interference and control (QUIC) techniques have been used previously to inject and control both electron–hole carrier populations and electrical charge currents.[2] For example, recently, the polarization and phase of the incident light and the crystal symmetry have been used to independently control both the carrier density and the spin of the optically created carriers.[3] Such techniques have also been used to produce a ballistic *spin-polarized* current in an unbiased semiconductor, with[4] and without[5] an accompanying electrical charge current. In most previous experiments,[2–5] the quantum mechanical interference was between the different pathways associated with two-photon absorption of ultrashort fundamental (ω) pulses and one-photon absorption of the corresponding second-harmonic (2ω) pulses. In such experiments, the injection process is dependent on the relative phase of the ω and 2ω pulses, and typically, the phase was controlled by a scanning Michelson interferometer. In all cases, the 2ω and ω pulses were collinearly propagating, and no population or current gratings were formed, spin polarized or not.

By comparison, conventional gratings are formed in semiconductors when two degenerate noncollinearly propagating pulses resonantly produce a spatially modulated carrier distribution.[6] The carrier transport is then studied by monitoring the diffraction of a time-delayed probe pulse from the decaying grating. Such techniques[6] have been used to study in-well[7,8] and cross-well transport[7,9,10] and the decay of spin gratings[11] in multiple quantum wells. We emphasize that all previous gratings were created by the absorption of two interfering pump beams having approximately the same frequency. The interference 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, we use two *noncollinear* propagating 2ω and ω pulses to produce a periodic spatially modulated quantum mechanical interference between the one- and two-photon absorption processes in GaAs. The interference is between quantum mechanical transition amplitudes, and for the polarizations chosen here, it is the magnitudes and directions of *spin-polarized currents* that are spatially modulated. Initially, there is no modulation of the carrier population, the spin, or the electrical charge current.

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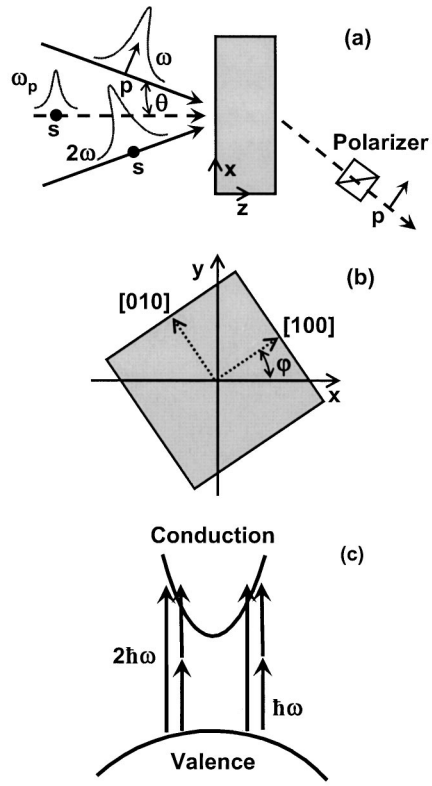

FIG. 1. Geometry (a) for producing a pure spin-polarized transient current grating by QUIC by using a *p*-polarized (in the plane of the figure) pump pulse with frequency ω and an *s*-polarized pump (perpendicular to the plane of the figure) with frequency 2ω and for monitoring the grating formation and decay by detecting the *p*-polarized diffracted component of an *s*-polarized probe with frequency ω_p and (b) for rotating the sample. (c) A schematic illustrating 2ω and ω connecting the same initial and final states by one- and two-photon absorption, respectively.

II. GENERATION AND MEASUREMENT OF PURE SPIN GRATINGS

The geometry that we use is shown schematically in Fig. 1. The ~150 fs ω pulse with wavelength centered at 1550 nm and the 2ω pulse at 775 nm are temporally and spatially coincident on the sample with a half-angle of $\theta=10°$ between the two pump pulses. A third time-delayed probe pulse at 830 nm measures the diffraction efficiency of the grating produced by the two pump beams. The fundamental (i.e., the ω pulse) is produced by an optical parametric amplifier (OPA) that is pumped by a regeneratively amplified Ti:sapphire laser operating at 250 kHz. The second harmonic of this pulse (i.e., the 2ω pulse) is produced by frequency doubling in a beta barium borate (BBO) crystal. The probe pulse at 840 nm is generated by separately frequency doubling the idler from the OPA. Wave plates and polarizing components (not shown) allow the independent control of the polarization of each pulse.

The sample is a 1-μm-thick sample of [001]-grown bulk GaAs, and the measurements are performed at room temperature. At this temperature, $\hbar\omega<E_g<\hbar 2\omega<E_{SO}$, where E_g (E_{SO}) denotes the heavy-hole (split-off) valence-to-conduction-band energy gap. Consequently, the 2ω and ω pulses acting independently will generate carriers through one- and two-photon absorption respectively. The irradiances of the 2ω and ω pump pulses (~550 MW/cm^2 and ~7 GW/cm^2, respectively) are adjusted such that each pulse will independently generate a peak carrier concentration of $\sim 10^{18}$ cm^{-3}.

In this article we consider the polarization configuration in which the ω pump pulse is *p*-polarized and the 2ω pump pulse is *s*-polarized, and the total incident pump field can be written

$$\mathbf{E}(t)=E_\omega \exp(-i\omega t+i\mathbf{k}_\omega\cdot\mathbf{r}+i\phi_\omega)[\hat{\mathbf{x}}\cos\theta+\hat{\mathbf{z}}\sin\theta] + E_{2\omega}\exp(-i2\omega t+i\mathbf{k}_{2\omega}\cdot\mathbf{r}+i\phi_{2\omega})\hat{\mathbf{y}}+\text{c.c.}, \quad (1)$$

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.

The theory given by Bhat and Sipe[12] to describe the currents produced by the quantum interference between one- and two-photon absorption of collinearly propagating 2ω and ω pulses can be readily extended to the case of *noncollinearly* propagating pulses. For the orthogonal linear polarizations given by Eq. (1), this theory predicts that quantum interference is expected to produce a current traveling in the $+x$ direction with electronic spins oriented in the $+z$ direction ($\mathbf{J}_{+x}^{+z}$) and a current with equal magnitude traveling in the $-x$ direction with electronic spins along $-z$ ($\mathbf{J}_{-x}^{-z}$). The injection rates for these two spin currents traveling along x are given by

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

where $\Delta\phi\equiv 2\phi_\omega-\phi_{2\omega}$, $\Lambda\equiv\lambda_\omega/4\sin\theta$ is the spatial period of the spin-polarized current gratings, and λ_ω is the wavelength of the ω pulse.

In terms of the parameters given in Ref. 12,

$$\dot{J}_1=4e\hbar^{-1}E_\omega^2E_{2\omega}D(A_1\cos^2\theta-A_2\sin^2\theta+A_3),$$

where D and A_i are functions of material parameters, but are independent of $\Delta\phi$. It should be noted that Ref. 12 gave an isotropic expression for the currents. However, by properly taking into account the zinc-blende symmetry, one finds that the cubic anisotropy to the spin currents is less than 7%.[13] Consequently, we are justified in using Eq. (2) and assuming that the spin currents depend only weakly on the sample orientation φ.

We reiterate that in previous QUIC experiments,[2] the 2ω and ω pulses were collinearly propagating ($2\pi x/\Lambda=0$) and the relative phase $\Delta\phi$ was varied as a function of time (typically by scanning one arm of a Michelson interferometer). Here, in contrast, we hold the temporal phase constant ($\Delta\phi=$constant), and the *spatial* phase variation ($2\pi x/\Lambda$) is provided automatically (and periodically) by the propagation directions chosen for the two pump pulses. Consequently, when QUIC techniques are combined with transient grating techniques, there is no need to explicitly control the phase between the 2ω and ω pulses, and a temporal variation in phase is replaced by a spatial variation.

The initial transient spin current gratings produced when the ω and 2ω pump pulses are orthogonally polarized [and represented by Eq. (1)] are depicted in Figs. 2(a) and 2(b).

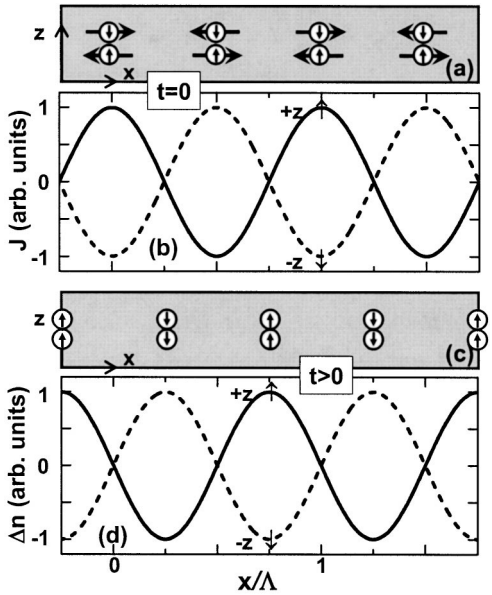

FIG. 2. (a) Schematic diagram depicting the initial ($t=0$) pure spin current grating produced by pump pulses with orthogonal linear polarizations. The grating consists of a periodic modulation of two oppositely directed currents with equal magnitudes, but opposite spins. The larger solid horizontal arrows with the smaller arrows pointing up (down) indicate the directions of the spin-up (spin-down) electron velocities at selected positions along x. (b) A plot of the corresponding magnitudes and directions of the x-directed spin-up (solid) and spin-down (dashed) electron currents as a function of position along the sample surface at $t=0$. (c) A schematic of the spin-up (arrows up) and spin-down (arrows down) electron densities as a function of position resulting from the ballistic transport depicted in (a) and (b). (d) A plot of the corresponding modulation of the spin-up (solid) and spin-down (dashed) electron densities.

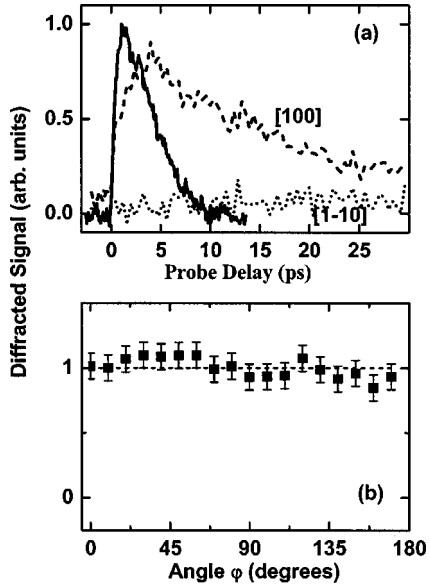

FIG. 3. The diffracted signal from an s-polarized probe when the ω pump pulse is p-polarized and the 2ω pump pulse is s-polarized: (a) the solid curve is the p-component of the diffracted signal for $\varphi=\pi/4$ (i.e., x along $[1\bar{1}0]$), and the dashed and dotted curves are the s-components for $\varphi=0$ (i.e., x along [100]) and $\varphi=\pi/4$, respectively, and (b) is the peak of the p-component of the diffracted signal as the sample is rotated by an angle φ about the z axis.

(In drawing this figure, we have assumed a fixed phase difference $\Delta\phi$, which without loss of generality, we have taken to be $\Delta\phi=0$). Immediately following the two pump pulses, at each point on the sample, two currents are produced with equal magnitudes, but opposite propagation directions and opposite spins. Consequently, initially there is no net spin grating, no net charge current grating, and no population grating. These currents always point along the $+x$ or $-x$ directions; however, their magnitudes vary cosinusoidally with phase $2\pi x/\Lambda$ along the x direction, but are out of phase by π, as shown in Fig. 2(b).

As time progresses, these currents cause the spin "down" electrons (spin vector in the $-z$ direction) to accumulate at certain positions (e.g., at $x=\Lambda/4$ and $5\Lambda/4$ in Fig. 2) and the spin "up" electrons (spin in the $+z$ direction) to accumulate at others (e.g., at $x=3\Lambda/4$). In this way, the two spin-polarized current gratings will evolve (by ballistic transport) into two spin-polarized electron population gratings (one with spin up and the other spin down) out of phase by exactly π [see Fig. 2(c)]; however, there still will be no modulation of the total electron density. The quantity that is modulated is the spin of the electrons. The ballistic formation of the two oppositely spin-polarized electron population gratings is determined by (and limited by) the momentum relaxation time, and it is too rapid to be resolved with our pulse widths.

In contrast to the electrons, the holes are injected with oppositely directed spins and velocities; however, the spin of the holes relaxes rapidly compared to the electrons.[14] Consequently, there will be no modulation of the hole population or spin.

Once formed, these electronic spin gratings will decay by the diffusion of the *spin-polarized electrons*, by the relaxation of the electronic spin, and by electron–hole recombination. Electronic diffusion is expected to be much faster than spin relaxation or recombination and is expected to dominate the grating decay. Notice that this decay by the diffusion of spin-polarized electrons is in contrast to conventional population gratings (where the electron and hole densities are modulated) that decay by ambipolar diffusion and recombination.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The results of measuring the diffracted signal from an s-polarized probe pulse as a function of time delay before and after the sample is irradiated with a p-polarized ω pump pulse and an s-polarized 2ω pump pulse are shown as the solid curve in Fig. 3(a). The signal shown was obtained with the polarizer in Fig. 1 oriented to pass p-polarized light. Therefore, 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.[11] 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.

This pulse-width-limited grating formation process is followed by a decay of the diffraction efficiency in $(2\Gamma)^{-1}$

$=3.2$ ps, where Γ is the decay rate for the grating. An electronic diffusion coefficient of $D_e = 195 \pm 40$ cm^2/s can be extracted from this decay by assuming that the grating decay is dominated by diffusion (i.e., $\Gamma = 4\pi^2 D_e/\Lambda^2$ (Ref. 6)) and by using $\Lambda = 2.23$ μm for our experimental conditions. From the Einstein relation, one can then obtain an electronic mobility of $\mu_e = 7700 \pm 800$ cm^2 V^{-1} s^{-1}, a value that is in reasonable agreement with the electron mobility (9000 cm^2 V^{-1} s^{-1}) reported previously.[15]

To further confirm that we are measuring the decay of two oppositely spin-polarized concentration gratings of the type portrayed in Figs. 2(c) and 2(d), in a separate experiment, we generated such spin gratings directly by using two resonant orthogonal linear polarized pump beams at frequency 2ω.[11] We measured the same decay time as in Fig. 3(a), providing additional evidence that the dynamics illustrated in Fig. 2 are correct.

Charge and spin population gratings (as opposed to current gratings) also can be formed directly for the geometry shown in Fig. 1, and it is important to establish that they are not responsible for the signal reported here. One can readily extend the calculation of charge and spin population control outlined in Refs. 4 and 12 to show that in addition to the current gratings given by Eq. (2) that one obtains a charge density grating given by

$$\dot{n}_I = 2\xi_I E_{2\omega} E_\omega^2 \cos(2\varphi)\sin(2\theta')\cos(2\pi x/\Lambda + \Delta\phi), \quad (3)$$

and a spin-polarized density grating given by

$$\dot{S}_I^z = 2\zeta_{\mathrm{IB}} E_{2\omega} E_\omega^2 \sin(2\varphi)\sin(2\theta')\sin(2\pi x/\Lambda + \Delta\phi), \quad (4)$$

where θ' is the angle of incidence inside the sample, $\dot{S}_I^z$ and $\dot{n}_I$ are the injection rates for the z component of the spin and the carrier density, respectively, associated with the quantum interference terms, and where ξ_I and ζ_{IB} are materials parameters defined in Ref. 4, which do not depend upon the sample orientation φ or phase difference $\Delta\phi$. Notice that when both pumps are normally incident, neither the charge nor the spin density grating is produced. Most importantly, both of these gratings undergo a 100% modulation of the grating amplitude as the sample is rotated about the z axis (see Fig. 1). The result of measuring the peak of the diffracted p-polarized signal as a function of sample orientation φ is shown in Fig. 3(b). No resolvable dependence on φ is observed, again, providing additional confirmation that the signal is dominated by the pure spin current gratings, not charge population gratings or spin-polarized population gratings.

Nevertheless, the charge density (or population) gratings predicted by Eq. (3) are present and can be isolated by monitoring the s-polarized diffracted component of an s-polarized probe. The results of two representative measurements for sample orientations of $\varphi = 0$ (x along [100]) and $\varphi = \pi/4$ (x along $[1\bar{1}0]$) are shown in Fig. 3(a) by the dashed and dotted curves, respectively. For $\varphi = 0$, the signal is maximum, and for $\varphi = \pi/4$, the signal is not measurable, consistent with Eq. (3). Moreover the signal at $\varphi = 0$ decays in $\sim$17.6 ps, consistent with a grating decay determined by ambipolar diffusion and recombination. Although we have demonstrated in Fig. 3(b) that the *peak* diffracted signal from the current grating (i.e., the peak p-component) is independent of sample orientation, nevertheless, the solid curve in Fig. 3(a) was measured with $\varphi = \pi/4$ to ensure that the charge density grating was "turned off" and that leakage from its s-polarized signal did not distort measurement of the pure spin grating.

Finally, it should be noted that a grating could also be generated by the phase-dependent energy transfer between ω and 2ω as the result of frequency up (down) conversion in the GaAs, but this process [which is associated with the real part of the second-order susceptibility $\mathrm{Re}(\chi_2)$] is also expected to result in a 100% modulation of the grating amplitude as the sample is rotated. No such angular dependence is observed.

IV. SUMMARY

Consequently, we conclude (i) that the diffracted light signal represented by the solid curve in Fig. 3(a) arises from two spin-polarized population gratings that are shifted spatially by π with respect to one another, (ii) that these population gratings are produced by the ballistic transport associated with two oppositely directed spin-polarized current gratings with opposite spins, and (iii) that these pure spin gratings, in turn, are created by the interference between the quantum mechanical probability amplitudes for one and two photon absorption of noncollinearly propagating 2ω and ω pump pulses with orthogonal linear polarizations. 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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