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Coherent control of non-equilibrium population and spin in GaAs

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Abstract
We report independent coherent control of the density and spin of a non-equilibrium electron population in (111)-oriented GaAs. The symmetry and orientation of the GaAs combined with the polarizations and phases of the excitation fields provide a number of control parameters that allow for controlling either the density or spin—or both or neither—of the population. We show that coherent control arises through interference processes involving one- and two-photon absorption.

1. Introduction

The injection into and control of electron spin in semiconductors remains a major challenge of semiconductor spintronics [1, 2]. Recently, it was shown [3, 4] that the carrier *density* can be controlled by using interference between quantum mechanical transition amplitudes induced by harmonically related optical fields, but no spin control was demonstrated. Here, we demonstrate all-optical injection and coherent control of a *spin-polarized* non-equilibrium carrier population through interference processes involving optical fields of frequencies ω and 2ω. We present the results of macroscopic symmetry analysis that give predictions of both spin and population control, and we demonstrate that these predictions are consistent with experimental observations.

2. Macroscopic symmetry predictions

When ω and 2ω pulses are present simultaneously and satisfy the relation $\hbar\omega < E_g < 2\hbar\omega$, where E_g is the band gap energy, it is possible to establish quantum interference of one photon absorption of 2ω and two photon absorption of ω photons connecting the same initial valence band and final conduction band states in bulk GaAs. The geometry we consider is shown in figure 1. The light propagates in the [111] direction, and ω and 2ω fields are linearly polarized with an angle θ between them and an angle δ between the ω polarization and the $[01\bar{1}]$ axis. The interference term in the injection rate of carrier population ($\dot{n}_I$) is proportional to the imaginary part of $\chi_2(-2\omega, \omega, \omega)$ [3]. In a crystal with zincblende symmetry,

$$\dot{n}_I = -\frac{\varepsilon_0 4\sqrt{2}}{\hbar\sqrt{3}} \operatorname{Im} \chi_2^{abc} |E_{2\omega}||E_\omega|^2 \cos(\Delta\phi) \sin(3\delta - \theta) \quad (1)$$

where E_ω and $E_{2\omega}$ are the field amplitudes at ω and 2ω, respectively, ε_0 is the permittivity of free space, the phase parameter $\Delta\phi \equiv 2\phi_\omega - \phi_{2\omega}$ and a, b and c denote components along the cubic axes [100], [010] and [001], respectively.

The interference term in the injection rate of spin along the z-axis (the [111] axis) ($\dot{S}_I^z$) can be described by a fourth rank pseudotensor that in zincblende crystals has two independent components $\operatorname{Im} \zeta^{abba}$ and $\operatorname{Im} \zeta^{abab}$ [5]. For these polarizations,

$$\dot{S}_I^z = \sqrt{\frac{2}{3}} \zeta |E_{2\omega}||E_\omega|^2 \sin(\Delta\phi) \cos(3\delta - \theta) \quad (2)$$

where $\zeta = \operatorname{Im} \zeta^{abba} + 2\operatorname{Im} \zeta^{abab}$ is the only combination of the two independent components that appears for light normally incident on a (111) surface.

According to equations (1) and (2), the population and spin are 90° out of phase in terms of the phase difference $\Delta\phi$. They are also in quadrature in terms of the angle between the two polarizations (θ) and the orientation of the sample (δ).

Simulations using these equations are shown in figure 2. Figure 2(*a*) illustrates what is expected to happen to the maximum phase-dependent amplitude when the sample orientation is fixed with respect to the ω polarization and the 2ω polarization is rotated through one revolution. Regardless

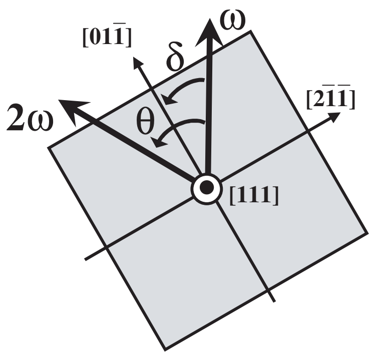

Figure 1. Sample geometry showing sample orientation angle δ and angle between polarizations θ for linearly polarized fields that propagate in the [111] direction.

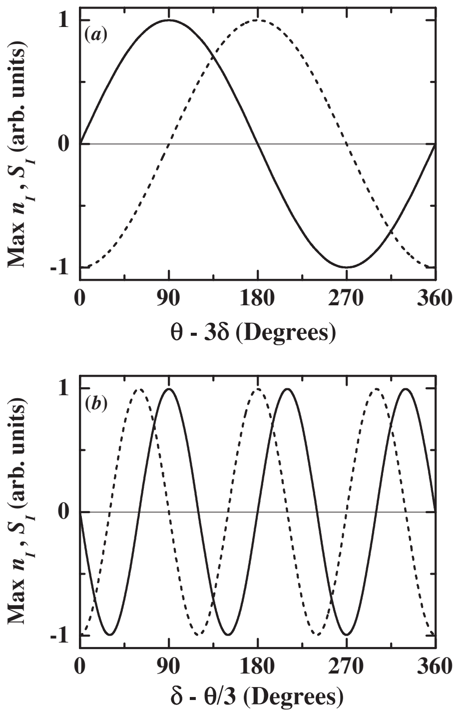

Figure 2. Predicted maximum population control (solid curve) and spin control (dashed curve) as a function of sample orientation angle δ and the angle between polarizations θ. (*a*) Fix δ, rotate θ. (*b*) Fix θ, rotate δ.

of the sample orientation δ, there are polarization angles θ for which we expect maximum spin control but no population control, and other values of θ for which we expect maximum population control but no spin control. There are also configurations where it is possible to control both population and spin. Conversely, by fixing the polarization and rotating the sample, figure 2(*b*) illustrates that the phase-dependent population and spin go through three cycles in quadrature—consistent with the fact that (111)-GaAs exhibits three-fold rotational symmetry.

For circularly polarized excitation pulses, the dependence on sample orientation disappears, since circularly polarized light has no preferred direction in the transverse plane. When ω and 2ω have opposite circular polarizations, we expect coherent control of *both* population and spin, and the magnitudes should be larger than the peak magnitudes for linear excitation by a factor of $\sqrt{2}$.[3] For these polarizations, the macroscopic symmetry analysis also predicts that population and spin control should vanish at the same values of $\Delta\phi$, as both are proportional to $\cos(\Delta\phi)$. When ω and 2ω have the same circular polarization, we expect neither population control nor spin control.

3. Experiment

In our experiments, ω (wavelength 1.43 μm) and 2ω (0.715 μm) pulses of $\sim$100 fs in duration are temporally overlapped, propagate collinearly and are focused at normal incidence onto an optically thin (111)-grown GaAs wafer at room temperature. With these photon energies, we generate a non-equilibrium carrier population with up to 275 meV excess energy. The phase difference $\Delta\phi$ is controlled with a scanning Michelson interferometer, and the polarization of each pulse is independently controlled. A third pulse (wavelength 0.81 μm) probes the population and spin of pump-injected carriers at lower energy in the band, arriving 0.9 ps after the pumps to allow for carrier thermalization. To monitor the fractional change in population ($\Delta n/n_0$), we measure the phase-dependent differential transmission [$\Delta T(\Delta\phi)$] of a linearly polarized probe. To measure the fractional spin change ($\Delta S/S_0$), we monitor $\Delta T(\Delta\phi)$ as the probe polarization is periodically modulated between right and left circular polarizations with a photoelastic modulator.

4. Results and discussion

We have performed an extensive set of experiments which demonstrate independent control of the population and of the spin of the population by systematically varying the polarizations of the two incident beams and by rotating the sample. Some of these results are plotted in figure 3. We have observed, for example:

(i) For selected fixed sample orientations and linearly polarized ω and 2ω pump pulses, parallel polarizations allow the *control of the spin without modulating the population* (figure 3(*a*)); conversely, orthogonal linear polarizations allow the *control of the population, but not the spin* (figure 3(*b*)). For other specific sample orientations, the dependences of the spin control and of the population control on the polarizations are reversed (not shown).

(ii) For fixed parallel (or orthogonal) linear polarizations, the sample can first be oriented so that the phase $\Delta\phi$ can be used to control the spin, but not the population. The sample can then be rotated so that $\Delta\phi$ controls the spin, but not the population (not shown).

(iii) For opposite circular polarizations, both the spin and the population can be simultaneously controlled, independent of sample orientation (figure 3(*c*)), in agreement with theoretical predictions.

(iv) When ω and 2ω have the same circular polarization, a highly spin-polarized background population is produced, but $\Delta\phi$ controls neither the spin nor the population (not shown), also in agreement with our predictions.

[3] Note that in equation (6) of [5], the numerical factor should be $\sqrt{2/3}$ instead of $1/\sqrt{3}$.

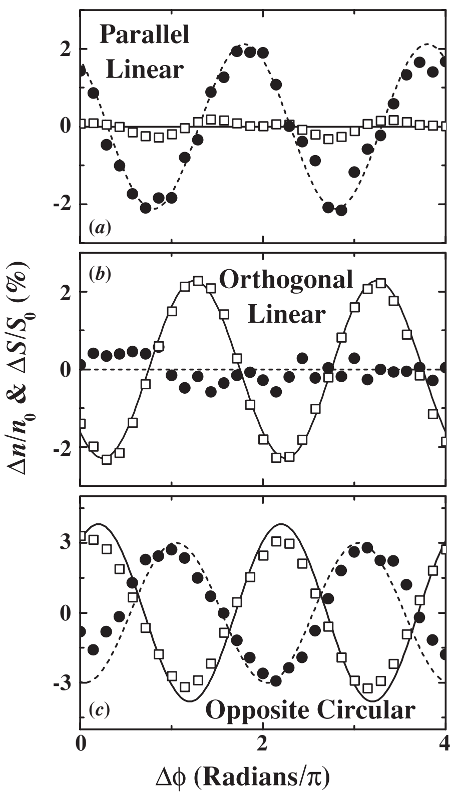

Figure 3. Fractional change in spin (solid circles) and population (open squares) as a function of relative phase $\Delta\phi$ between ω and 2ω beams for various polarization combinations, all at a fixed sample orientation of $\delta = 0$. Solid lines (dashed lines) show simulations of population (spin) control. (*a*) Parallel linear polarizations (along [01$\bar{1}$]), showing spin control without population control. (*b*) Orthogonal linear polarizations, showing population control without spin control. (*c*) Opposite circular polarizations, showing both spin and population control.

All of these observations agree with the macroscopic theory presented above, as evidenced in part by the excellent agreement between data and simulations in figure 3. Note that $\Delta\phi$ has an arbitrary offset for each of the simulations in figure 3.

In addition to the direct sources of population and spin control discussed above, there are also contributions from cascaded processes. In the cascaded processes, χ_2 causes frequency conversion between the ω and 2ω pulses which can then modulate the one photon carrier- and spin-injection. In zincblende-type crystals, the cascaded and direct contributions to population control have the same symmetry properties and thus they cannot be distinguished by varying the polarization states or crystal orientation. For spin control, on the other hand, the cascaded and direct contributions have different symmetry properties, although the difference vanishes for light normally incident on a (111) surface. As discussed by Fraser *et al* [3], the cascaded and direct processes can, however, be separated by their different dependences on sample thickness; for thin samples, the direct process dominates. For our sample, which is the same one used by Fraser *et al* [3], we expect that the cascaded and direct processes contribute roughly the same to population control.

In summary, we have demonstrated the use of the phase together with either the polarization of the light or the crystallographic symmetry to independently control the spin and density of a non-equilibrium carrier population. We have also shown that results agree with a theory based on macroscopic symmetry analysis.

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