Cascaded second-harmonic coherent control of carrier density and spin in a [110]-oriented semiconductor

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Abstract: We demonstrate independent coherent control of carrier density and spin in [110]-oriented GaAs/AlGaAs quantum wells by using the phase-dependent energy transfer between ~100-fs ω and 2ω pulses, as opposed to quantum interference.

OCIS codes: (190.5970) Semiconductor nonlinear optics including MQW; (320.7130) Ultrafast processes in condensed matter, including semiconductors

Two-color all-optical Quantum Interference and Control (QUIC) techniques have been used previously to inject and control the densities [1,2] and spins [2] of carriers in [111]-oriented semiconductors. The coherent control was attributed to the interference between the quantum mechanical probability amplitudes for the two-photon absorption of a fundamental (ω) pulse and the one-photon absorption of a second harmonic (2ω) pulse ($\hbar\omega < E_g < 2\hbar\omega$, where E_g is the bandgap). Since these probability amplitudes depend upon the phase and polarization of the incident light and on the crystal symmetry and orientation, each of these parameters can be used to control both the carrier density and the spin [2]. QUIC of population can be shown [1,2] to be related to the imaginary part of the second order nonlinear susceptibility, $\mathrm{Im}\chi_2$, and the control of the spin to the difference between "spin-up" and "spin-down" pieces of $\mathrm{Im}\chi_2$.

In contrast, here, we provide the first demonstration of ω–2ω coherent control dominated by a cascaded second harmonic generation (SHG) process, and the first demonstration of ω–2ω coherent control of either the *population* or *spin* in [110]-oriented material (in this case a GaAs/AlGaAs multiple quantum well (MQW)). Here, the symmetries of population and spin control are quite different than for the (111)-GaAs used earlier [2], and we investigate them in more detail. The cascaded SHG process takes advantage of the net frequency up or down conversion that occurs via $\mathrm{Re}\chi_2$ as the ω and 2ω pulses propagate collinearly through the material. The direction of energy transfer and the relative phase of the field associated with that transfer are determined by the phase and polarization of the ω and 2ω pulses and by the crystal symmetry and orientation: If the *intensities* of the ω and 2ω pulses are modified by SHG, then the carrier *density* will be modulated. If the *polarization state* is modified, the carrier *spin* can be modulated.

This process can be regarded as cascaded second harmonic generation (SHG) in two senses. First, it requires two sequential SHG processes: one in an external frequency doubler to generate the incident phase-controlled ω- and 2ω-pulses, and another within the material. Second, within the material, an SHG up or down conversion process is followed by one or two-photon absorption. We emphasize that this phase-dependent modulation of the carrier spin and density results directly from a modification of the ω and 2ω fields by the SHG—not from the quantum interference. We note that this SHG process has been discussed previously in the context of population control, but not spin control, and was found to make a small contribution for the sample used [1]. Here, for our experimental conditions (which are significantly different from those in [1]), we demonstrate that the phase and polarization of the ω and 2ω pulses, as well as the crystal symmetry and orientation, can be used to independently control the carrier density and spin via this process.

In our experiment, the ~100 fs fundamental pulse (ω), with wavelength 1.43 μm, is generated in an optical parametric amplifier. SHG in BBO produces the 2ω pulse at 0.715 μm. These two pump pulses are temporally overlapped, propagate collinearly, and are focused at normal incidence onto the sample, which is a [110]-oriented MQW with twenty periods of 8 nm GaAs wells and 8 nm $Al_{0.3}Ga_{0.7}As$ barriers, topped with a ~0.5 μm thick $Al_{0.3}Ga_{0.7}As$ etch-stop layer (Fig. 1(d)). The phase parameter $\Delta\phi \equiv 2\phi_\omega - \phi_{2\omega}$ (where ϕ_ω and $\phi_{2\omega}$ are the phases of the ω and 2ω fields, respectively) is controlled with a scanning Michelson interferometer, and the polarization of each pulse is independently controlled with waveplates and polarizers. A third pulse, with a photon energy just above the bandgap (at a wavelength of 0.81 μm), probes the changes in sample transmission (ΔT) to give information about the density and spin of pump-injected carriers [2].

Figure 1(a)-(b) illustrates the use of the incident polarizations to independently control the population and spin for a fixed sample orientaion ($\alpha = 56°$). When 2ω and ω have parallel linear polarizations along x, there is a large population control signal, but little or no modulation of the spin. By contrast, for orthogonal linear polarizations (ω along x, 2ω along y), the phase $\Delta\phi$ controls the spin, but not the population. Additionally, when 2ω and ω have opposite circular polarizations, there is strong modulation of both population and spin. Thus, at this fixed sample orientation, we can choose pump polarizations to control the population only, the spin only, or both.

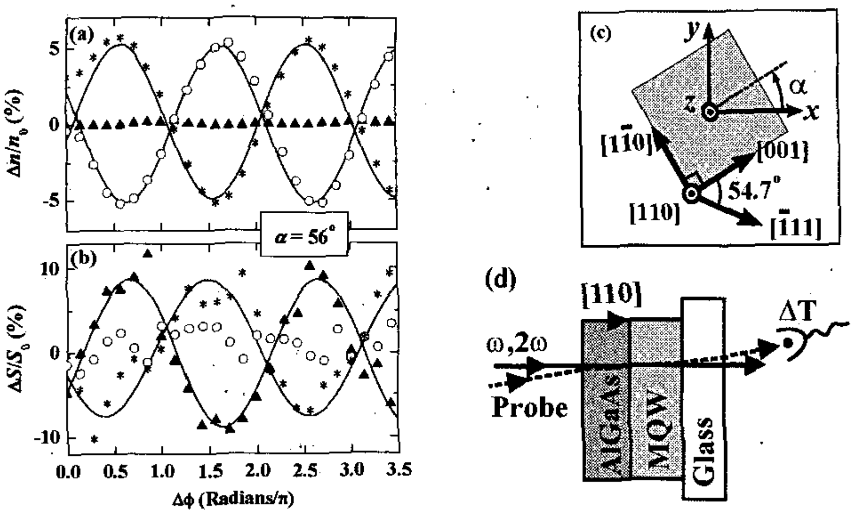

FIG. 1. Measured fractional change in (a) population and (b) spin at the sample angle $\alpha = 56°$ for parallel linear (open circles), orthogonal linear (solid triangles) and opposite circular (asterisks) excitation. Solid curves are sinusoidal fits to the data. (c) Normal view of the sample, showing crystal directions and laboratory coordinate system; ω and 2ω fields propagate along the z-axis. (d) Side view of the sample.

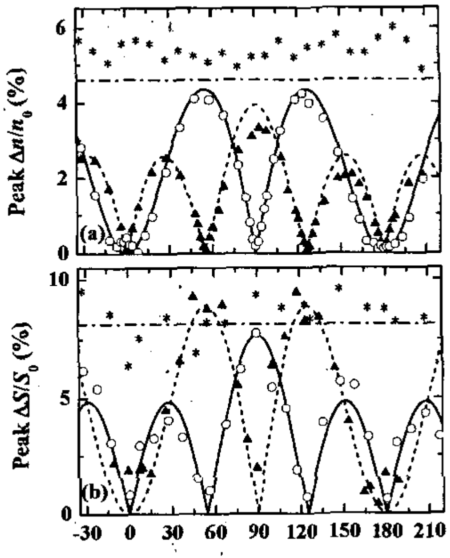

FIG. 2. Peak measured change in (a) population and (b) spin as a function of sample angle α for parallel linear (open circles), orthogonal linear (solid triangles) and opposite circular (asterisks) excitation. Also shown are simulations of cascaded population and spin control for parallel linear (solid line), orthogonal linear (dash) and opposite circular (dash-dot) excitation.

Figure 2 illustrates the use of *sample orientation* to control the population and spin. For fixed parallel (or orthogonal) linear pump polarizations, a sample orientation can always be chosen so that the phase $\Delta\phi$ controls the population, but not the spin, or a different orientation can be chosen so that $\Delta\phi$ controls the spin, but not the population. By contrast, when 2ω and ω have opposite circular polarizations, both the population and the spin are controlled independent of sample orientation.

To verify that our signal is dominated by the SHG cascaded process, we repeated our measurements with the sample flipped so that the beams entered the MQW from the glass side in Fig. 1(d). The population and spin control signals were three and four times smaller, respectively. Since SHG, but not QUIC, occurs in the AlGaAs layer, the "amplifying" effect of this layer suggests that the cascaded process is dominant under the conditions used here. This conclusion is supported by the fact that all of the tendencies displayed in Fig. 1 and Fig. 2 are consistent with a simple symmetry analysis for the SHG cascaded process; however, QUIC alone can not account for the observed tendencies in the spin control using accepted values for the GaAs materials parameters. Finally, detailed measurements of the difference between the phase of the control signals and the phase of the transmitted 2ω pulse of the type described in [1] are consistent with a coherent control process dominated by cascaded SHG, although QUIC is expected to contribute at some level. In fact, we have found that whether the cascaded SHG or QUIC dominates depends upon sample geometry and excitation conditions.

In summary, we have demonstrated that cascaded SHG coherent control allows the use of the phase together with the polarization of the light and the crystallographic symmetry to independently control carrier spin and density. We find that a cascaded effect, which was thought to play a small role in previous measurements [1], can actually *dominate* population and spin control in this sample under the given excitation conditions.

References

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