Optical Orientation and Femtosecond Relaxation of Spin-Polarized Holes in GaAs

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Optical orientation of spin-polarized heavy and light holes followed by relaxation to other valence subband states has been observed unambiguously in undoped bulk GaAs in spite of the extremely short spin relaxation time. The measured relaxation time for the heavy holes is 110 fs $\pm 10\%$. The results are relevant for applications such as interpretation of spin-polarized transport in semiconductors as well as the assessment of feasibility of hole-based spin-transport devices which relies on precise knowledge of the hole-spin relaxation time.

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There has been a great deal of interest in the physics of spin relaxation in III-V semiconductors [1–3]. Much of the attention has been focused on spin-polarized conduction band electrons, which is undoubtedly motivated primarily by potential applications [4–6]. It is also partly because electron spins relax relatively slowly in comparison with spin-polarized holes and are, therefore, more accessible to experimental studies. Because of the strong spin-orbit interaction and coupling of the quasimomentum and the angular momentum of the holes in III-V compounds [1,2], the spin relaxation of holes in an undeformed cubic crystal of the GaAs type is extremely fast—on the scale of the momentum relaxation time or in the femtosecond time domain. Direct observation of optical orientation and relaxation of spin-polarized holes in bulk GaAs is, therefore, very difficult. On the other hand, in quantum wells [7–10], because of the modification of the valence band structure, a significant change in the hole-spin relaxation dynamics near the zone center leading to a substantial increase in the spin relaxation time is expected. Hole spin relaxation times in quantum wells ranging from 4 ps [9] to as long as 1 nsec [10], depending upon the doping levels, temperature, and quantum well structures, have been reported.

Information on the relaxation dynamics of spin-polarized holes is particularly important for understanding the relaxation of electron spins. First of all, relaxation of hole spins often plays a direct role in the relaxation of electron spins through electron-hole exchange interaction [such as the Bir-Aronov-Pikus (BAP) process [11]], especially in heavily doped and quantum well [12] samples. Second, much of the information on various electron spin relaxation processes, such as the Elliott-Yafet [13], the Dyakonov-Perel' [14], or the BAP process, is obtained on the basis of the conduction-to-valence-band luminescence studies which involve both the electrons and the holes. It is always assumed that the hole spins relax instantaneously [1–3,7] and can, therefore, be ignored, at least within the time resolution of the experiments. The question of just exactly how fast the hole spins relax in pure GaAs single crystals has never been resolved experimentally. In this Letter, we report the direct observation of optical orientation and the subsequent relaxation of spin-polarized holes in undoped bulk GaAs at room temperature, using time- and polarization-resolved femtosecond spectroscopy. These results are relevant for applications such as interpretation of spin-polarized transport in semiconductors as well as the assessment of feasibility of hole-based spin-transport devices which relies on precise knowledge of the hole-spin relaxation time.

In the experiment, spin-polarized holes in a thin $\sim 5\ \mu$m antireflection coated intrinsic GaAs single crystal on a semi-insulating GaAs substrate are first created near the zone center of the heavy- and light-hole (hh and lh, respectively) subbands through valence-to-conduction-band (v-c) absorption of circularly polarized ($\hat{\sigma}_+$) femtosecond pulses (width $\sim$90 fs) at $\sim$800 nm from a mode-locked Ti:sapphire laser. The same Ti:sapphire laser is used to pump a periodically poled lithium niobate femtosecond optical parametric oscillator [15] generating precisely synchronized signal and idler pulses in the $\sim$1.1 to 1.5 μm range and in the midir in the $\sim$2.6 to 5.4 μm range ($\sim$ 80 fs), respectively. Suitably delayed tunable midir pulses are used to probe selectively the spin-aligned hh and lh through excitation of electrons from the fully occupied splitoff (so) subband (See Fig. 1). The dynamics of the spin-polarized hh and lh can then be studied by measuring the circular dichroism of the probe beam.

To do so, a linearly polarized incident probe beam is used, which contains an equal mixture of right and left circularly polarized components. For the probe beam transmitted through the sample, the degree of circular polarization ρ is defined in the usual way as the ratio of the difference to the sum of the intensities of the right and left circular components of the transmitted beam,

$$\rho = \frac{I_+ - I_-}{I_+ + I_-}. \qquad (1)$$

If the hh or lh spin in the sample is aligned, it will preferentially absorb the corresponding circular

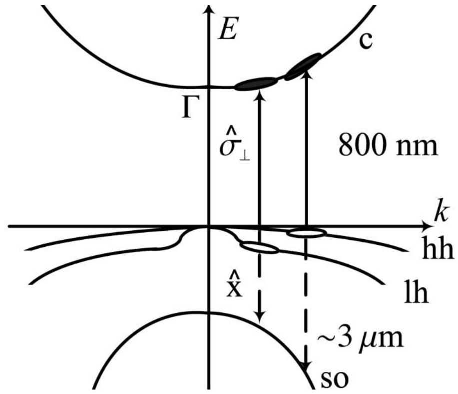

FIG. 1. The femtosecond output of a Ti:sapphire laser at $\lambda_{\text{pump}} = 800$ nm (solid line) generates oriented holes in the hh and lh. The change in the degree of circular polarization of a transmitted, time-delayed $\lambda_{\text{pr}} \sim 3$ μm synchronized probe pulse (dashed line) corresponding to the so-hh and so-lh transitions is proportional to the spin state at that value of the delay.

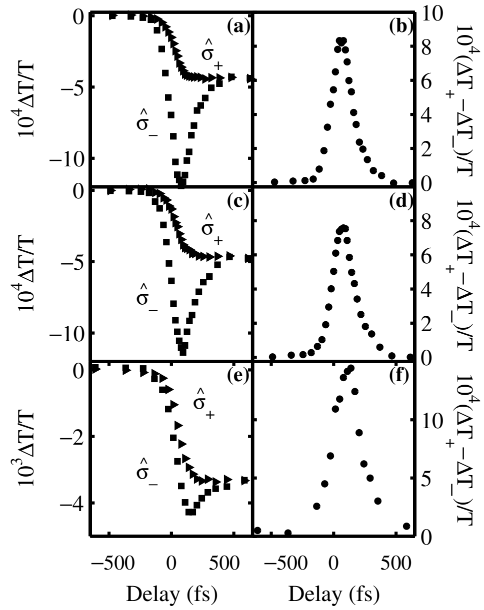

FIG. 2. Time and polarization resolved differential transmission at $\lambda_{\text{probe}} = 3200$ nm (top), 3000 nm (middle), and 3800 nm (bottom).

component of the probe beam and change its degree of circular polarization accordingly. ρ is proportional to the average of the spins in the probed valence subband in the direction of propagation of the probe beam [2], since the splitoff subband is initially fully occupied by electrons. Thus, the time dependence of ρ gives the time dependence of the hole-spin alignment. The time dependence of the relaxation dynamics of the spin-polarized hh and lh can, thus, be determined from the measured values of $I_+(\tau)$ and $I_-(\tau)$ where τ is the delay of the midir probe pulse from the 800 nm pump pulse.

We focus on the hh first. A standard femtosecond pump and probe measurement technique is used, and is shown in Fig. 1. The setup is essentially the same as that described in Ref. [16] with the use of suitable additional wave plates and detectors to resolve and measure simultaneously the right and left circularly polarized components of the transmitted probe beam.

Defining $\Delta T/T$ as the differential transmission, the measured value (at a carrier concentration of 10^{17} cm^{-3}) of $\Delta T_+/T = \Delta I_+/(I_+ + I_-)$ and $\Delta T_-/T = \Delta I_-/(I_+ + I_-)$ for two so-hh probe wavelengths ($\lambda_{\text{probe}} = 3.2$ and 3.0 μm) are shown in Figs. 2(a) and 2(c), respectively. Also shown in Figs. 2(b) and 2(d) are the corresponding difference $(\Delta T_+ - \Delta T_-)/T = (\Delta I_+ - \Delta I_-)/(I_+ + I_-) = (I_+ - I_-)/(I_+ + I_-)$. The experimental results show unambiguously that, first of all, *optical orientation and relaxation of the heavy holes are clearly observed,* in spite of the extremely short momentum relaxation time of the holes.

Similar results for the so-lh wavelength ($\lambda_{\text{probe}} \sim 3.8$ μm) corresponding to the lh-c transition point have also been obtained and shown in Figs. 2(e) and 2(f). Qualitatively, the hh and lh results are similar. The difference in the magnitudes of the two sets of results are primarily due to the difference in the joint density of states of the so-hh and so-lh probe transitions and relaxation of the holes due to thermalization toward the zone center within the pump and probe time duration. The resulting lh relaxation dynamics are also expected to be more complicated.

A quantitative interpretation of the results is complicated due to the complexity of the valence band structure and the fact that the pump and probe pulse widths are not short compared with the spin relaxation time. For the hh state directly excited by the pump wave (corresponding to $\lambda_{\text{probe}} \sim 3.2$ μm) and assuming a single exponential decay $\sim \exp(-t/\tau_s)$ for the spins, the degree of circular polarization of the transmitted probe beam is, in the context of the present experiment, proportional to the convolution of this exponential decay factor and the cross correlation of the pump and probe pulse profiles:

$$\rho(\tau) \propto \int_0^\infty \exp\left(\frac{-t}{\tau_s}\right) C(t-\tau)dt, \tag{2}$$

without the negligible nonresonant coherent artifact contribution [17]. $C(\tau)$ is the normalized cross correlation of the pump and probe pulses as a function of the delay τ,

$$\int_{-\infty}^{\infty} C(\tau)d\tau = 1$$

and

$$C(\tau) = \int_{-\infty}^{\infty} g_{\text{probe}}(t')g_{\text{pump}}(t'-\tau)dt', \tag{3}$$

where $g_{\text{probe}}(t)$ and $g_{\text{pump}}(t)$ are the normalized intensity profiles of the appropriate circularly polarized components of the probe and the pump pulses, respectively. For a Gaussian cross-correlation function with a full width at half maximum $\Delta\tau_c = 2(\ln 2)^{1/2}\sigma$,

$$C(\tau) = \frac{1}{\sigma\sqrt{\pi}}\exp(-\tau^2/\sigma^2). \tag{4}$$

It follows from Eqs. (2) and (4) that

$$\rho(\tau) \propto \frac{1}{2}\exp\left(\frac{-\tau}{\tau_s} + \frac{\sigma^2}{4\tau_s^2}\right)\left[1 - \mathrm{erf}\left(\frac{\sigma}{2\tau_s} - \frac{\tau}{\sigma}\right)\right]. \tag{5}$$

The measured cross-correlation trace is shown in Fig. 3. As can be seen, it can be represented quite precisely by a Gaussian function. With the help of Eq. (5), deconvolution of the measured $\rho(\tau)$ on the basis of Eq. (2) yields a *best-fit* ($\pm$ 10%) *value of 110 fs for* τ_s. The resulting fit between the calculated and measured shapes of $\rho(\tau)$ is excellent, as shown in Fig. 3. Because of the more complicated subband structure and relaxation process near the zone center for the lh case, no single exponential relaxation time could be extracted from the lh data.

The Hamiltonian describing the hh subband in the spherical approximation is of the form [2]

$$\hat{H} = \frac{\hbar^2}{2m_0}\left(\gamma_1 + \frac{5}{2}\gamma\right)k^2 - \frac{\hbar^2}{2m_0}\gamma(\vec{k}\cdot\vec{J})^2, \tag{6}$$

where m_0 is the free electron mass, the γ's are the appropriate Luttinger parameters, and J_x, J_y, and J_z are 4×4 matrices corresponding to the value of angular momentum being equal to 3/2. Because of the strong spin-orbit interaction and the direct coupling of the quasimomentum and the angular momentum $(\vec{k}\cdot\vec{J})$, τ_s is expected to be on the order of the momentum relaxation time τ_p, which is usually taken to be "instantaneous" [7]. To our knowledge, there does not seem to be any quantitative theory on either τ_p or τ_s for holes in undoped bulk GaAs, even though there are a large number of experimental and theoretical studies [4–12] of τ_p or τ_s for GaAs/Al_xGa_{1-x}As quantum wells or superlattices. In any case, τ_p is expected to be smaller than the energy relaxation time τ_e. Femtosecond spectroscopic studies [16,18] of the hole-burning effects in GaAs suggest that the energy relaxation time of hh and lh is well over a hundred femtoseconds. On the basis of the commonly used range of values for the hole mobility of GaAs near the zone center ($\sim$ 400 cm^2/V s or less), a hh momentum relaxation time on the order of 110 fs is not unreasonable. Clearly, only a detailed numerical calculation based upon the appropriate Boltzmann equation can account for such a number theoretically.

Because of the pump-beam induced circular dichroism, the transmitted probe beam becomes elliptically polarized and has a polarization component, $I_\perp$, perpendicular to that of the linearly polarized incident probe beam. In addition, the concomitant difference in the index of refraction for the right and left circularly polarized components can lead to a small rotation of the axes of the polarization ellipse of the transmitted probe beam leading to an additional contribution to $I_\perp$. If the right and left circularly polarized transmitted probe beams, $\hat{\sigma}_+$ or $\hat{\sigma}_-$, are perfectly coherent and the intensity is the exact perpendicular component, $I_\perp(\theta_\perp = 0°)$, it can be shown that, for weak absorption of the probe beam, $I_\perp(\theta_\perp = 0°)$ is proportional to the square of the pump-beam induced spin population difference, ΔN^2, of the $M_J = +3/2$ and $M_J = -3/2$ components of the hh subband. However, at a small angular deviation, $\theta_\perp = \delta$, from the true perpendicular direction, $\theta_\perp = 0°$, or if there is a small incoherence between the $\hat{\sigma}_+$ and $\hat{\sigma}_-$ components of the transmitted probe beam, the experimentally measured $I_\perp(\theta_\perp = \delta)$ contains a term linearly proportional to ΔN and, hence, ρ as shown in Fig. 4. Thus, the time dependence of the relaxation dynamics of the spin-polarized hh and lh can also be inferred from the more easily measured values of $I_\perp(\theta_\perp = \delta; \tau)$.

We have made a series of such measurements corresponding to various so-hh and so-lh probe wavelengths to study the scattering of the optically excited hole spins to other valance band states. The results are summarized in Fig. 5. Because of rapid spin relaxation due to the coupling of the quasimomentum and the angular momentum of the holes, the shapes of the $\rho(\tau)$ traces at all the probe wavelengths are similar. The slight variation with

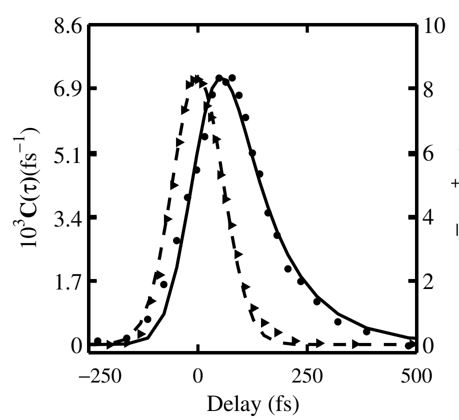

FIG. 3. Cross correlation and measured circular dichroism at 3200 nm in GaAs. ▶: measured $C(\tau)$; dashed line: fit to Eq. (4); •: measured $(\Delta T_+ - \Delta T_-)/T$; solid line: fit to Eq. (5)

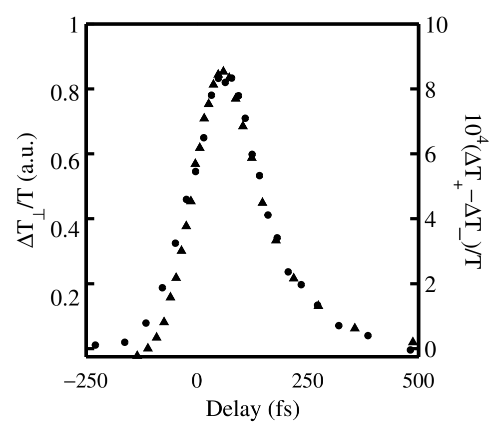

FIG. 4. Comparison of the normalized $\Delta T_\perp/T = I_\perp/(I_+ + I_-)$ (▲) and $(\Delta T_+ - \Delta T_-)/T$ (•) results for $\lambda_{probe} = 3200$ nm.

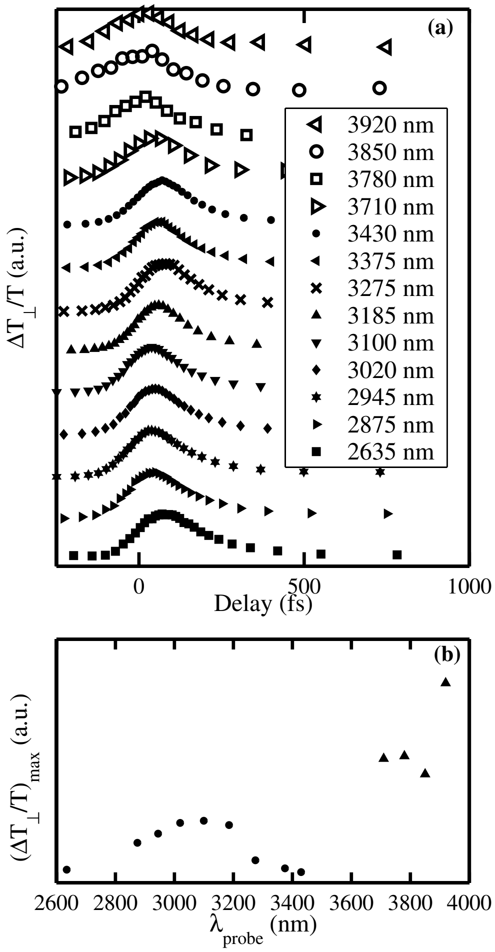

FIG. 5. Variation of $I_\perp$ with λ for both hh and lh in (a), where the traces are normalized and displaced vertically for clarity. The unnormalized relative peak amplitudes of the traces in (a) are shown in (b); the point near 4 μm in (b) is discussed in further detail in the text.

wavelength of the delay time at which the maximum signal occurs shown in Fig. 5(a) is due to two sources: (i) the difficulty in determining experimentally the precise position of the zero delay time, $\tau = 0$, and (ii) the actual delay of the arrival time at the probe wavelengths of the holes scattered from the excitation point. The only significant trend is the change of the peak values of $\rho(\tau)$ with respect to the wavelength as shown in Fig. 5(b), which is due primarily to the variation in the joint density of states of the so-hh and so-lh transitions. Note that the $I_\perp$ results shown in Fig. 5 are consistent with the circular dichroism results shown in Figs. 2(b), 2(d), and 2(f). For example, as shown in both sets of data, the peak values of $\rho(\tau)$ at 3.0 and 3.2 μm are similar and are a factor of $\sim$2 smaller than that at 3.8 μm. The significant change in the peak value of ρ near 4 μm is due to the drastic increase in the joint density of states (Van Hove singularity) for the so-lh transition because the corresponding E-versus-k surfaces for the so and the lh subbands are nearly parallel to each other at this wavelength [16].

In conclusion, we have observed unambiguously the optical orientation of spin-polarized heavy and light holes and their subsequent relaxation in undoped GaAs in spite of the extremely short spin relaxation time. The measured relaxation time for the heavy holes is approximately 110 fs ($\pm$ 10%). Scattering of the optically excited hole spins to the other heavy- and light-hole states and their subsequent relaxation have also been observed.

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