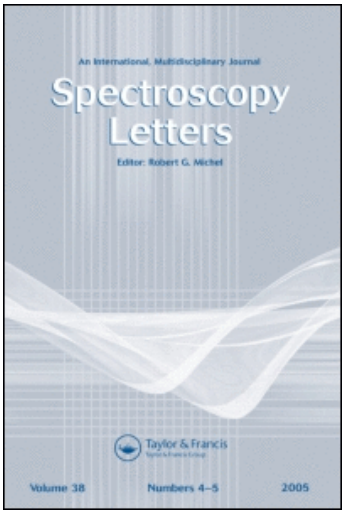

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Bandgap Shifting Effect in Spin Injection

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ABSTRACT A polarization-resolved optical pumping investigation in a nonmagnetic semiconductor is performed. Spins are injected optically by a circularly polarized two-photon pumping in the samples. The injection of the electron spins is probed by measuring the polarization-resolved transmission of the samples. From the transmission data, the degree of spin-polarization of electrons is measured as a function of the time delay of the probe as well as of the temperature. The results show that the degree of spin-polarization is decreased with increasing temperature. The polarization decrease at higher temperatures might be due to the band-narrowing effect.

KEYWORDS semiconductor, spectroscopy, spin-polarization

INTRODUCTION

Electronic spin states and their transport in semiconductors have been extensively studied because of their potential applications in spintronic devices.[1–5] In these studies, spin injection, which is an injection of spin-polarized carriers into a nonmagnetic semiconductor with the conservation of the spin-polarization, is a crucial subject, since it is a key for the manipulation of the carrier spin in semiconductors.

The injection of spin-polarized electrons in semiconductors has been obtained by either optical or electrical techniques.[2,4] However, the actual advantages of these techniques in practical devices have not yet been clearly established.[2,5] To fully understand the reliable transport (without loss of the spin-polarization over distances that are comparable to the device dimensions) of the injected spins in semiconductors, and to successfully implement spintronic device concepts, substantial research effort in this area is required.[5]

Multiphoton pumping has been widely used for a long time to investigate the optical nonlinear processes, particularly in semiconductors and insulators.[6,7] It has also been demonstrated that the technique can be used to inject spin-polarized carriers. For example, in an earlier theoretical study,[8] the two-photon spin injection in lead chacogenides was observed, and high spin-polarization in these cubic materials having fundamental band gaps was predicted. Recently, it was suggested that high spin-polarization could be achieved in unstrained bulk GaAs from two-photon pumping.[9] For this class of semiconductors, similar predictions were also observed in the earlier theoretical investigations[10,11] using two-photon excitation.

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Here, a polarization-resolved optical pumping experimental investigation is performed in a non-magnetic semiconductor. Spins are injected optically by a circularly polarized two-photon pumping in the samples. The injection of the electron spins is probed by measuring the time- and polarization-resolved transmission of the samples. The degree of spin-polarization of electrons is measured in the dependences of the time delay of the probe as well as of the temperature. In this article, the results obtained in the present investigation are reported. A detailed discussion on the results is also presented.

EXPERIMENTAL PROCEDURES

Samples were n-type ($1 \times 10^{21}\,\mathrm{m}^{-3}$) doped [001]-grown bulk gallium arsenide. The sample thickness was ~1 μm. A polarization-resolved two-photon pump-probe experiment was performed, where the differential transmission was $\Delta T/T = (T - T_0)/T$, where $T(T_0)$ is the transmission with (without) the pump, of the probe pulses was measured as a function of the delay between circularly polarized pump and probe pulses. The probe beam was resonant with the bandgap energy (E_g), and the pump beam (ω_{Pump}) had an excess energy $\hbar 2\omega_{\mathrm{Pump}} - E_g$ of 100 meV. An illustration of the two-photon pump-probe pumping scheme showing the pump pulse coupling the initial and final states in the valence band (VB) and conduction band (CB), and the probe pulse tuned to the bandgap excitation resonance is shown in Fig. 1. The measurements were performed at various temperatures by placing the samples in a temperature-regulated cryostat and using femtosecond (fs) pulses from an optical parametric amplifier pumped by a regeneratively amplified Ti:sapphire laser operating at 250 kHz. The laser system was tuned to produce ~150 fs pulses (signal and idler). A beta barium borate crystal was used to generate pulses from the signal and idler. The second harmonic and fundamental pulses were then separated using a dichroic beamsplitter. Pulses of ~150 fs were used to excite the sample by two-photon absorption, and 825-nm pulses were used to probe the transmission of the sample. Broadband quarter wave plates were used to transform pump and probe beams from linear to (right/left) circular polarized light.

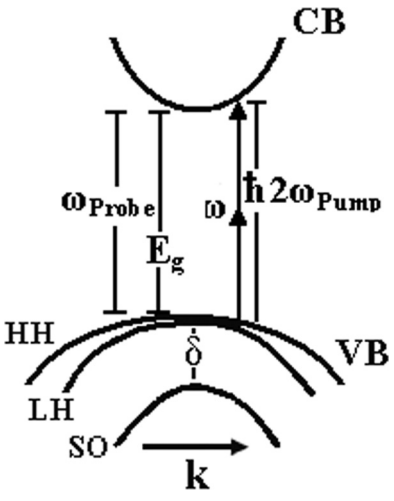

FIGURE 1 **An illustration of the two-photon pump-probe pumping scheme showing the pump pulse (ω_{Pump}) coupling the initial and final states in the VB and CB and the probe pulse (ω_{Probe}) tuned to the band gap (E_g) excitation resonance ($\hbar\omega_{\mathrm{Probe}} \approx E_g$). The scheme also shows the HH, LH, and SO bands of GaAs. The pumping condition $\Delta E_{2\omega_{Pump}} = \hbar 2\omega_{Pump} - \hbar\omega_{Probe} \square \hbar 2\omega_{Pump} - E_g < \delta$ (where δ is the spin-orbit splitting) for the excess photon energy was maintained in the experiments.**

RESULTS AND DISCUSSION

In optical pumping, the production of a carrier population with a net spin by direct absorption of a circularly polarized light is a consequence of the optical selection rules ($\Delta m_j = \pm 1$) for the heavy-hole (HH) and light-hole (LH) VB to CB transitions.[12] Since hole spin-polarization in bulk semiconductors is known to relax in ≤100 fs, one typically obtains only the electron spin-polarization on a longer time scale.

For a circularly polarized light, direct transitions from the HH VB produce electrons with the opposite spin from those from the LH band. However, the HH transition in unstrained bulk GaAs is three times stronger than the LH band transition, leading to a 3:1 ratio of $N_\downarrow$ to $N_\uparrow$ ($N_\uparrow$ to $N_\downarrow$) for right (left) circularly polarized light σ^+ (σ^-), where $N_\uparrow$ ($N_\downarrow$) is the density of spin-up (spin-down) CB electrons. Consequently, optical excitation with σ^+ (σ^-) injects spins along the direction parallel (antiparallel) to the direction of the light propagation and produces an initial $P_0 = P(0,0) = 50\%[P(r,t) = \{N_\downarrow(r,t) - N_\uparrow(r,t)\}/\{N_\downarrow(r,t) + N_\uparrow(r,t)\} = \pm 0.5$ at $r = 0$ and $t = 0$ for $\sigma^\pm]$ spin-polarized carrier population, provided the photon energy of the two-photon pumping ($E_{2\omega_{Pump}} = \hbar 2\omega_{Pump}$) is $E_g < E_{2\omega_{Pump}} < E_g + \delta$ (where δ is the spin-orbit splitting), that is, low enough to avoid exciting carriers from the split-off (SO) band to the CB.

We measured the differential transmission excited by right circularly polarized pump pulses at various temperatures. Figure 2 shows the results probed with a right circularly polarized pulse at the liquid nitrogen temperature. The same differential transmission signal measured with the left circularly polarized light pulse is shown in Fig. 3. As can be seen from Figs. 2 and 3, there is a difference between the different polarization conditions (right and left), which is caused by spin-dependent phase-space filling.[13,14]

Since, for probe pulses near the band edge of the semiconductor material, $(\Delta T/T)_{\sigma^+\sigma^+} \propto 3N_\downarrow + N_\uparrow$ and $(\Delta T/T)_{\sigma^+\sigma^-} \propto 3N_\uparrow + N_\downarrow$, as a result of the selection rules discussed above, one can determine the degree of circular polarization P from the relation $P = 2[(\Delta T/T)_{\sigma^+\sigma^+} - (\Delta T/T)_{\sigma^+\sigma^-}]/[(\Delta T/T)_{\sigma^+\sigma^+} + (\Delta T/T)_{\sigma^+\sigma^-}]$, where $(\Delta T/T)_{\sigma^+\sigma^+}$ and $(\Delta T/T)_{\sigma^+\sigma^-}$ are the differential transmissions for pump and probe pulses having the same and opposite circular polarizations respectively.

We determine P from the measured $(\Delta T/T)_{\sigma^+\sigma^+}$ and $(\Delta T/T)_{\sigma^+\sigma^-}$ using the above relation. The resulting P as a function of the time delay is shown in Fig. 4. The P decays with time giving a value for the spin relaxation time of ~410 picoseconds. The decay of P might be due to the randomization of the initial spin-polarization P_0 by the Dyakonov–Perel (DP) spin relaxation mechanism.[4,15] The DP spin relaxation occurs in semiconductors lacking inversion symmetry due to the spin precession about an intrinsic magnetic field, $\vec{b}(\vec{k})$, induced by the presence of the spin-orbit interactions in a zinc-blende structure,

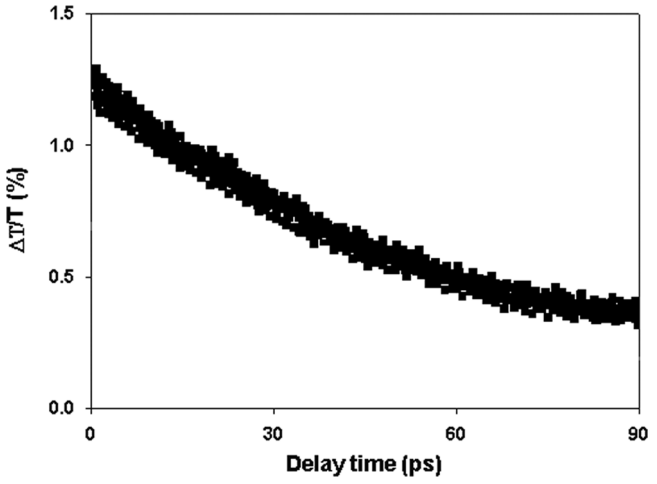

FIGURE 2 Measured differential transmission using probe pulses with the same circular polarization.

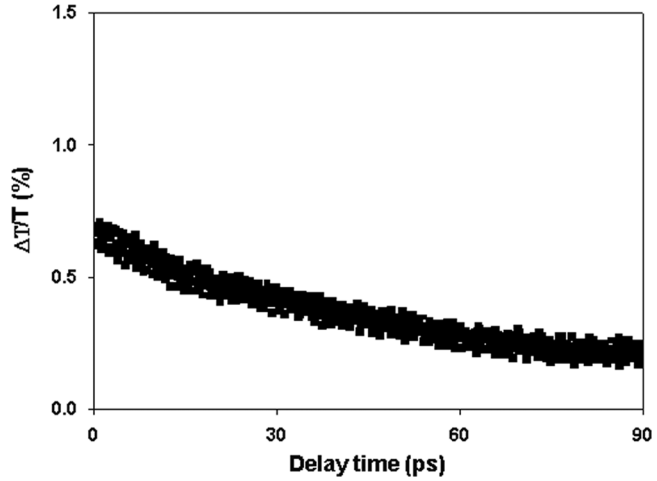

FIGURE 3 Measured differential transmission using probe pulses with the opposite circular polarization.

acting on the spin with its magnitude and orientation depending on $\vec{k}$ and results in spin precession with intrinsic frequency $\omega(\vec{k})$ during the time between collisions, according to the relation $d\vec{S}/dt = \omega(\vec{k}) \times \vec{S}$, where $\omega(\vec{k}) = (e/m^*)\vec{b}(\vec{k})$, $\vec{S}$ is electron spin-polarization vector, $m^* = \eta m_0$ ($\eta = 0.076$ for GaAs) is the effective electron mass and m_0 is the free electron mass. The corresponding Hamiltonian term due to spin-orbital splitting of the CB describing the precession of electrons in the CB[12] is

$$H_{DP}(\vec{k}) = \frac{\hbar}{2}\vec{\sigma} \cdot \omega(\vec{k}), \qquad (1)$$

where $\vec{\sigma}$ is the vector of Pauli spin matrices. For bulk materials, the spin-orbit interaction is only due to the

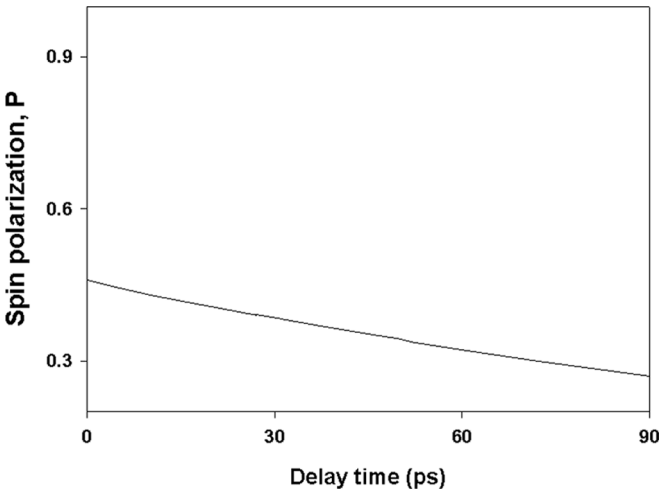

FIGURE 4 Measured spin-polarization of the CB electrons. The curve is drawn (fit) through the mean of the experimental data. The data taken from Figs. 2 and 3 are excluded for clarity.

bulk inversion asymmetry, and the DP Hamiltonian, expressed in Eq. (1), is composed of the Dresselhaus term only. However, in a quantum well, for example, the DP Hamiltonian is composed of the Dresselhaus and Rashba terms.[12] The Rashba term appears if the self-consistent potential within a quantum well is asymmetric along the growth direction and is, therefore, referred to as structural inversion asymmetry contribution.

Figure 5 shows the crystal temperature (T_C) dependence of the initial spin-polarization P_0. As can be seen, the maximum value of P_0 is obtained at the liquid helium temperature, and P_0 decreases with increasing T_C. The observed temperature dependence might be due to the temperature-dependent bandgap shift of GaAs.

To see the bandgap-shift effect, we calculate the bandgap energy as a function of T_C for GaAs. The temperature dependence of the direct bandgap (determined from the absorption edge) of a zinc-blende semiconductor, such as GaAs, can be described[16] by

$$E_g(T_C) = E_g(0) - \frac{AT_C^2}{B + T_C}, \qquad (2)$$

where $E_g(0)$ is the bandgap at 0 K, and A and B are the temperature independent parameters. We calculate E_g as a function of T_C using the parameter values for GaAs.[17] Figure 6 shows the results. As can be seen, the bandgap energy decreases as the crystal temperature increases, and there seems to be a correlation with the temperature dependence of the polarization (Fig. 5). Thus, the circularly polarized light with photon energy $E_{2\omega_{Pump}} < E_\delta$ (where E_δ is the SO band to the CB energy gap) might be able to excite some of the carriers from the SO VB due to the band-narrowing effect at higher temperatures, and consequently, the polarization would decrease. The results are consistent with those obtained in another investigation,[18] where the authors studied the spin relaxation of electrons during transport in the similar material by the photoluminescence (PL) polarization measurements and showed that the initial PL polarization under drift increased with decreasing T_C. It is worthy to note that the drift field played an additional role in their temperature dependence of P_0. The observed results also agree with those of the PL measurements in a semiconductor heterostructure.[19]

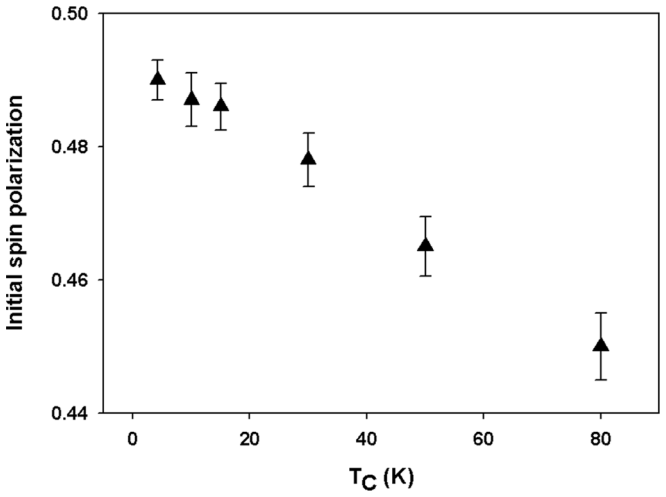

FIGURE 5 **Measured spin-polarization of the CB electrons as a function of T_C. Bars show the deviations of means of data from several experiments.**

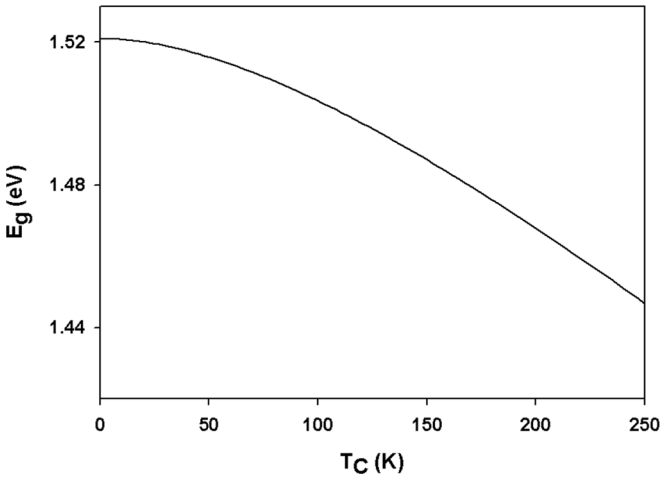

FIGURE 6 **Crystal temperature-dependent bandgap shift of the material, calculated using the parameter values for GaAs.**

As detailed above, although the maximum optical spin-polarization for an unstrained bulk sample is expected to be 50% in theory, the maximum has experimentally been observed to be much less than that.[2] In the earlier polarization measurements, they used one-photon excitation, and hence, there were some background unpolarized electrons on the bottom of the bulk sample. The two-photon excitation enhances the spin-polarization because it takes the advantages over one-photon spin generation due to a much longer absorption depth, which allows spin excitation in the deep level, that is, throughout the volume of a thin bulk sample. The 1-μm thick samples used for this study are reasonable as thin

enough, and the absorbance change induced by the pump is considered to be small, that is, $l(\alpha - \alpha_0) << 1$, where l is the sample thickness and $\alpha(\alpha_0)$ is the absorption coefficient with (without) the pump. The results discussed above demonstrate that due to a much longer absorption depth, highly spin-polarized CB electrons can be generated optically by the two-photon HH and LH states pumping of the bulk semiconductors.

CONCLUSIONS

A polarization-resolved optical pumping investigation in a nonmagnetic semiconductor was performed. Spins were injected optically by a circularly polarized two-photon pumping in the samples. The injection of the electron spins was probed by measuring the time- and polarization-resolved transmission of the samples. The degree of spin-polarization of electrons was measured in dependences of the time delay of the probe as well as of the temperature. The results showed that the degree of spin-polarization was decreased with increasing temperature. It was demonstrated that the polarization decrease at higher temperatures might be due to the bandgap shift of the semiconductor because of the band-narrowing effect.

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