Subpicosecond spin relaxation in GaAsSb multiple quantum wells

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Spin relaxation times in $GaAs_xSb_{1-x}$ quantum wells are measured at 295 K using time-resolved circular dichroism induced by 1.5 μm, 100 fs pulses. Values of 1.03 and 0.84 ps are obtained for samples with $x=0$ and 0.188, respectively. These times are >5 times shorter than those in InGaAs and InGaAsP wells with similar band gaps. The shorter relaxation times are attributed to the larger spin-orbit conduction-band splitting in the Ga(As)Sb system, consistent with the D'yakonov–Perel theory of spin relaxation [M. I. D'yakonov and V. I. Perel, Sov. Phys. JETP **38**, 177 (1974)]. Our results indicate the feasibility of engineering an all-optical, polarization switch at 1.5 μm with response time <250 fs. [S0003-6951(99)02549-8]

Ultrafast spin relaxation is of interest for applications such as all-optical polarization switches which utilize optically induced circular dichroism.[1] Such switches have been demonstrated at a wavelength of 800 nm using GaAs quantum wells and spin-relaxation times of >7 ps.[2–4] Because of the possible relevance to optical communications technology, InGaAs(P) quantum wells with a band gap near 1.55 μm have also been investigated[1,5] and typical spin-relaxation times are >5 ps. Recently, the GaAsSb system has gained attention because such structures can also be grown for device applications in the 1.55 μm region.[6–11] As we show below, from the D'yakonov–Perel (DP) mechanism of spin relaxation,[12] the GaAsSb system is expected to exhibit >6 times shorter relaxation times than the InGaAs system, pointing to a possible subpicosecond response. Since this ultrafast relaxation is related to an intrinsic property of the system, devices with a switching time <250 fs would be attainable, without the need to use low-temperature growth or defect-induced recovery time reduction, which have the undesirable consequence of degraded optical nonlinearity at the band edge.[13]

We present optical measurements of the electron spin relaxation time in $GaAs_xSb_{1-x}/AlSb$ multiple quantum wells near the $n_z=1$ heavy-hole-to-conduction-band transition for $x=0$ and 0.188. Our measurements reveal a subpicosecond decay of spin-polarized carriers, consistent with the D'yakonov–Perel mechanism of spin relaxation. The multiple quantum wells were grown by molecular beam epitaxy in a VG V80 machine equipped with a valved cracked As source and a cracked Sb source. The structures were designed to provide a band-gap transition wavelength of 1.55 μm, taking into account the effects of As fraction, quantum confinement, and strain. Due to the presence of bowing in the valence bands of the ternary system Ga(As)Sb,[14] and because the quantum well layers experience tensile strain, confinement tuning of the band gap to 1.55 μm requires that the quantum well layer thickness be lowered with increasing As fraction for concentrations below $x=0.40$. For samples with $x=0$ and 0.188, the quantum well thicknesses are 8 and 5.1 nm, respectively, with AlSb barrier layer thicknesses of 8 nm for both samples. From photoluminescence experiments, the band-gap wavelengths were determined to be 1.55 μm ($x=0$) and 1.52 μm ($x=0.188$). The multiple quantum wells, which contain 6 periods ($x=0$) or 60 periods ($x=0.188$), were grown on a Bragg mirror with a peak reflectivity of 80% at 1.55 μm. The mirror consists of 5 periods of $Ga_{0.68}Al_{0.32}Sb/AlSb$ $\lambda/4$ layers on a GaSb substrate.

Pump–probe experiments were carried out at 295 K using circularly polarized pulses, as shown in Fig. 1. The optical source is a 250 kHz repetition rate optical parametric amplifier (Coherent OPA 9800), which provides 100 nJ, 100 fs pulses, tunable from 1.2 to 2.4 μm. For each sample, the pulse center wavelength (1.53 μm; $x=0$, 1.49 μm; $x=0.188$) was chosen to correspond to 15 meV above the $n_z=1$ heavy-hole-to-conduction-band transition. A pump pulse creates spin-polarized carriers and the resulting dynamics are probed using a delayed, weaker pulse. From the selection rules in quantum wells, excitation of the $n_z=1$ heavy-hole-to-conduction-band transition using circularly polarized

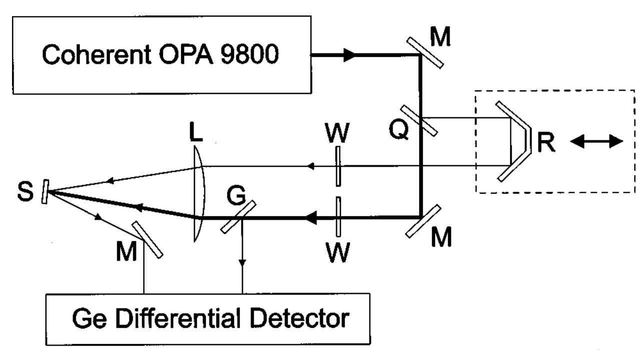

FIG. 1. Apparatus used for pump–probe differential reflection measurements. The pump beam is indicated by a thick line, the probe by a thin line. S, sample; W, quarter wave plate; Q, quartz window; M, mirror; R, retroreflector; G, glass slide for pickoff of a reference beam for subtraction; and L, lens, f=25 cm.

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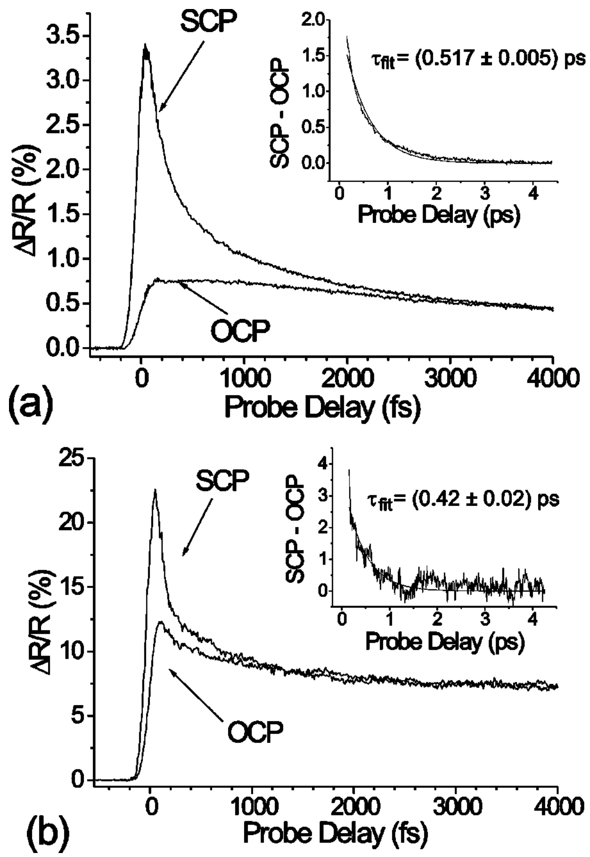

FIG. 2. Results of pump–probe measurements of $GaAs_xSb_{1-x}$/AlSb multiple quantum wells with conditions of the same (SCP) and opposite circular (OCP) polarizations in pump and probe beams, for (a) $x=0$, (b) $x=0.188$. Inset: single-exponential fit to the difference between the decay curves for SCP and OCP.

light produces carriers with pure spin states, with holes and electrons which are both spin up (down) for excitation with right (left) circular polarized light. Absorption bleaching occurs due to state filling, which is sensitive to the spin state of the excited carriers, and Coulomb screening, which only depends on the total carrier density. The decay of spin polarization is determined by comparison of the absorption bleaching measured by the probe pulse when it has the same circular polarization (SCP) and opposite circular polarization (OCP) state as the pump pulse. Because the quantum wells are on top of Bragg mirrors, a reflection geometry was used. Changes in the probe pulse absorption are linearly proportional to reflectivity changes. The differential reflectivity, which is the pump-induced change in the probe reflectivity expressed as a percentage of the unsaturated probe reflectivity, was measured as a function of probe delay. To reduce noise from laser power fluctuations, lock-in detection was used in conjunction with a differential amplifier. In all cases, the pump fluence at the samples was 22 nJ/cm^2, producing a quantum well carrier density of 4.5×10^{12} cm^{-2}.

Figure 2 shows the differential reflectivity as a function of probe delay for (a) $x=0$ and (b) $x=0.188$ samples. A positive signal indicates a reduction in probe pulse absorption. The bleaching signals for SCP and OCP beams clearly converge in <4 ps for both samples, indicating rapid spin-polarization decay. In Fig. 2(a), the peak in the bleaching signal at zero delay for SCP is completely absent in the OCP data, indicating that this peak is due to state filling alone. The

TABLE I. Values of the spin splitting coefficients and effective masses in selected semiconductor systems (see Refs. 5, 17, and 18).

Materials	γ (a.u.)	m^*/m_0	$(m^*)^3\gamma^2$
GaAs	6.3	0.067	0.012
GaSb	46	0.041	0.148
$GaAs_{0.188}Sb_{0.812}$	38.5	0.046	0.145
$In_{0.53}Ga_{0.47}As$	16.5	0.044	0.023

small peak which exists in the OCP data at zero delay in (b) is likely due to imperfect polarization states in the two beams. This will reduce the difference between the OCP and SCP curves for the $x=0.188$ sample, but will not affect the extraction of spin-relaxation times. [The 1.49 μm wavelength for the measurements in (b) is 60 nm removed from the design wavelength of the zero order, λ/4 plates.] The steady-state signal in (a) is only 23% of the mean value at zero delay, compared to a value of 46% in (b). This difference is attributed to the presence of intervalley scattering ($\Gamma\rightarrow L$) processes which are energetically allowed for the injected carriers in the $x=0$ sample, but not in $x=0.188$ material due to an increase in the $\Gamma-L$ separation in the ternary system.[15] Spin-relaxation times were determined using the rate equation analysis of Ref. 1. The difference between the differential reflection signals for SCP and OCP configurations was fit to a single-exponential decay, as shown in the insets of Fig. 2. The fits were restricted to delay values >150 fs to avoid contributions from coherence effects. The spin-relaxation time is related to the fitting time constant by $\tau_s/2=\tau_{\rm fit}$. For the $x=0$ and 0.188 samples, we obtain $\tau_s=(1.03\pm0.01)$ ps and $\tau_s=(0.84\pm0.04)$ ps, respectively.

It has been proposed[1,5] that the dominant mechanism for spin relaxation of electrons in quantum wells at room temperature is that of D'yakonov and Perel.[12] Spin relaxation of holes is expected to be much faster than that of the electrons due to mixing in the valence-band states, and so electrons will provide the dominant contribution to our observed spin-relaxation times. The DP mechanism of spin decay is present in zinc-blende semiconductors due to the lack of a center of inversion symmetry, which leads to a spin-orbit splitting of the conduction band. This splitting is given by $\Delta E=\gamma k^3$, where k is the electron wave number and γ is the spin splitting coefficient. The spin-orbit-induced conduction-band splitting provides an effective magnetic field causing the electron spins to flip. If one neglects the energy dependence of the momentum relaxation time, the DP mechanism gives a spin-relaxation rate in quantum wells of[5]

$$\frac{1}{\tau_s}=\frac{16k_BT(m^*)^3(\gamma E_{1e})^2\tau_v}{\hbar^8}, \tag{1}$$

where E_{1e} is the electron confinement energy for the $n_z=1$ subband, τ_v is the momentum relaxation time, and m^* is the electron effective mass. The spin-decay rate is proportional to the square of the confinement energy in the quantum well and the cube of the effective mass, predicting a faster spin decay in narrow quantum wells and lower-band-gap materials. (The electron effective mass scales inversely with the band-gap energy in the III–V semiconductors.[16]) Values for the spin splitting coefficient and effective masses in GaSb and GaAsSb are given in Table I,[5,17,18] along with the corresponding results in GaAs and InGaAs for comparison. Linear interpolation was used for both the mass and the spin splitting parameter in $GaAs_{0.188}Sb_{0.812}$. If we assume that the electron confinement energies and the momentum relaxation times are the same in the different systems, the relaxation rates are governed by the values of $m^{*3}\gamma^2$. As Table I shows, this quantity is 6.3 times larger in the Ga(As)Sb quantum wells than in the InGaAs system. Tackeuchi, Wada, and Nishikawa[5] observed a spin-relaxation time of 5.2 ps in $In_{0.53}Ga_{0.47}As$ quantum wells with well widths of 7 nm. These well widths are similar to those in our GaSb quantum wells, for which we observed a spin-decay time of 1.03 ps. Our results, which reveal an enhancement of the spin- relaxation rate in GaSb/AlSb quantum wells of 5 times relative to the InGaAs system are, therefore, consistent with what one expects from the DP theory of spin relaxation.

The observed spin decay rate for the sample with $x=0.188$ is a factor of 1.2 larger than that in the sample with $x=0$, which has a 36% larger quantum well thickness. Although the DP theory indicates that the spin-relaxation rate increases for decreasing well widths, in agreement with our results, the degree of enhancement in the spin-relaxation rate due to the narrower wells in the As-containing sample is expected to be larger than that observed here. A direct comparison of these samples is not entirely appropriate since, e.g., the momentum relaxation times are not known. Indeed, the differences may be explained by a reduction in the momentum relaxation time in the ternary system due to the presence of defects.

In summary, spin-relaxation times have been measured in Ga(As)Sb/AlSb multiple quantum wells using pump–probe techniques with 100 fs pulses at 1.5 μm. Samples with As fractions of 0 and 0.188 give spin recovery times of 1.03 and 0.84 ps, respectively. The subpicosecond spin relaxation observed in these Ga(As)Sb quantum wells makes these structures excellent candidates for all-optical polarization switching devices operating at 1.55 μm. Such devices would provide switching times $\leqslant$250 fs with high-quality structures and good optical properties. The repetition rate is limited by the lifetime of carriers in the device active region, an issue which may be addressed using an applied electric field.[19] The much shorter spin-decay times in these GaSb-based heterostructures compared to InGaAs(P)/InGaAsP and InGaAs/InP quantum wells, which exhibit similar-sized band gaps, are attributed to the increased spin-orbit interaction in GaSb, consistent with the DP mechanism of spin relaxation.

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