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Room temperature spin relaxation in GaAs/AlGaAs multiple quantum wells

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We have explored the dependence of electron spin relaxation in undoped GaAs/AlGaAs quantum wells on well width (confinement energy) at 300 K. For wide wells, the relaxation rate tends to the intrinsic bulk value due to the D'yakonov–Perel (DP) mechanism with momentum scattering by phonons. In narrower wells, there is a strong dependence of relaxation rate on well width, as expected for the DP mechanism, but also considerable variation between samples from different sources, which we attribute to differences in sample interface morphology. © *1998 American Institute of Physics.* [S0003-6951(98)02541-8]

The mechanisms of carrier spin relaxation in quantum-confined semiconductor systems are of interest, in part because of the possible applications of polarization selective optical nonlinearities,[1] and also because it frequently occurs under ''motional narrowing'' conditions,[2] so that the spin-relaxation rate is proportional to the momentum relaxation time, in which case spin relaxation is fastest in the best quality materials. Another reason for interest is the very slow ($\sim$1 ns) room temperature electron spin relaxation recently observed in *n*-type $Zn_{0.8}Cd_{0.2}Se$ quantum wells.[3] Spin relaxation of resonantly excited $n=1$ heavy-hole excitons,[4] holes,[5] and electrons[6] in GaAs/AlGaAs quantum wells has been systematically investigated at low temperatures. However, there has been relatively little systematic investigation at room or intermediate temperatures.[7,8] In this letter we investigate the dependence of the room temperature electron spin relaxation rate on confinement energy in samples with different thicknesses. Such measurements require careful interpretation because of possible variations due to sample imperfections.[9] We therefore compare results from a variety of molecular beam epitaxy (MBE)-grown samples studied by ourselves and published previously.[7]

All the samples were grown on (100)-oriented substrates and were without intentional doping. In each case a spin-polarized population of photogenerated carriers was produced by a circular polarized optical pump pulse at the $n=1$ exciton resonance, and the evolution of the spin orientation and population were examined via induced nonlinear changes in absorption or reflection of a degenerate, delayed probe pulse. We use here the standard definition of spin relaxation time τ_s,[2] as the characteristic decay time for the population difference of spin-up and spin-down states. This differs by a factor of 2 from the definition used in Ref. 7.

For our experiments both the pump and probe were derived from a mode-locked Ti:sapphire laser with pulse duration $\sim$1 ps, giving pump excitation density $\sim 10^{10}$ cm^{-2} and probe intensity $\sim 10^{-2}$ that of the pump. Reflection measurements carried out at Southampton (see Fig. 1) employed a modulation method to determine simultaneously the pump-induced difference in reflectivity for left and right circularly polarized light (the ''difference'' signal) and the pump-induced change in total reflectivity (the ''sum'' signal).[10] Transmission measurements made at St. Andrews[11,12] employed an etched window in the substrate and involved a sequence of two runs with the probe beam similarly circularly polarized (SCP) and oppositly circularly polarized (OCP) to the pump. A third run was made with pump and probe beams having orthogonal linear polarizations (OLP). In the two techniques the ''sum'' and OLP signals indicate the total population of photoexcited carriers, while the ''difference'' and SCP and OCP traces indicate the evolution of spin alignment.

Details of our samples appear in Table I, which includes the low temperature photoluminescence (PL) linewidth and Stokes shift as some measure of sample quality. Residual doping levels were $\sim 2\times 10^{14}$ cm^{-3} *p* type except in KLB269 for which it was $\sim 10^{16}$ cm^{-3} *p* type. With the exception of GWS196 all samples were multiple quantum wells with at least 15 repeats and barrier thicknesses greater

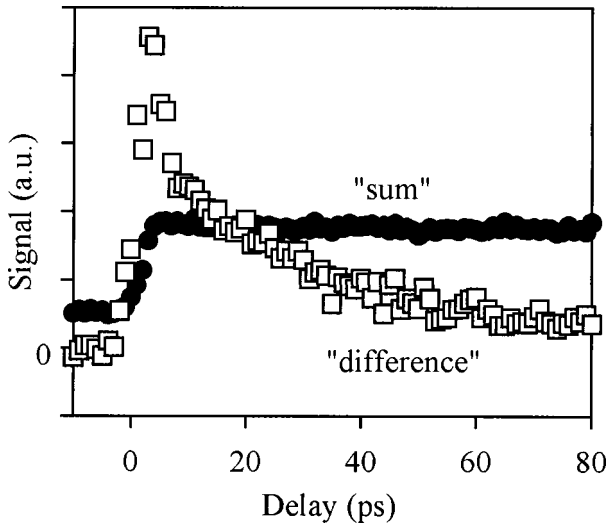

FIG. 1. Time dependence of pump-induced change in reflectivity (''sum'') and spin alignment (''difference'') for sample G51, pumped at the $n=1$ exciton resonance.

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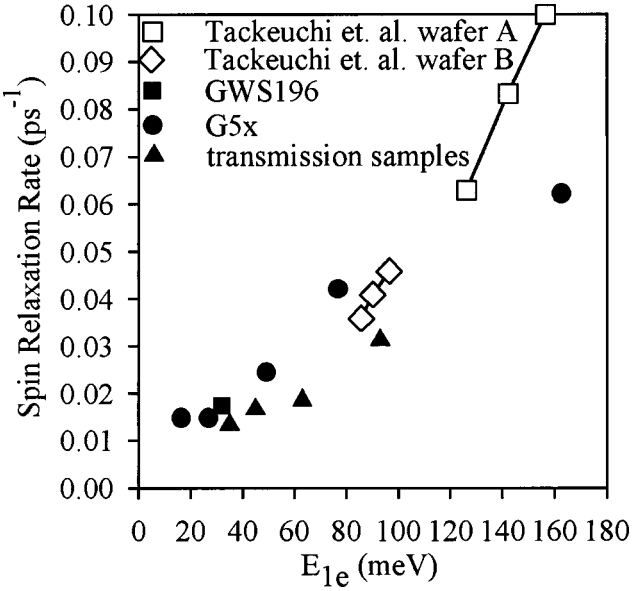

FIG. 2. Spin relaxation rate vs confinement energy for all samples investigated here (solid symbols) and from the work of Tackeuchi *et al.* (Ref. 7) (open symbols). The latter are increased $\times 2$ relative to the published values to conform to the standard definition of spin-relaxation rate (Ref. 2).

than 100 Å; GWS196 contained three separate quantum wells of different width, the central one being studied here. The electron mobilities of two samples, S51 and KLB269, were measured[11,12] to be 0.46 and 0.48 $m^2\,V^{-1}\,s^{-1}$, respectively, and may be compared with the value for high purity bulk material of be 0.85 $m^2\,V^{-1}\,s^{-1}$ at 300 K.

For both reflection and transmission measurements the pulse repetition frequency was about 70 MHz. Carrier lifetime at room temperature is up to 70 ns[12] in our samples, considerably greater than the interpulse spacing ($\sim$14 ns), so that the experiments were made with a significant ambient population of photoexcited electrons and holes. We estimate this to be $\leqslant 10^{11}$ cm^{-2} (i.e., $\leqslant 10^{17}$ cm^{-3}). Preliminary transmission measurements on samples with a 7 MHz repetition frequency, and therefore with greatly reduced ambient background, gave the same spin-relaxation rates, within experimental error, indicating that spin relaxation is not strongly dependent on the ambient population.

At room temperature thermal dissociation of excitons to free electrons and holes takes place on a timescale of order 300 fs.[13] It has been observed that hole-spin relaxation takes place on a subpicosecond timescale in type II multi quantum well (MQW) at room temperature,[14] and we expect it to be similar in our type I samples. Hence, the slower decays observed in Fig. 1 and in Ref. 12 the ''difference,'' SCP and OCP traces may be attributed to electron spin relaxation.

TABLE I. Characteristics of GaAs/AlGaAs MQW samples. PL Stokes shift and PL width are photoluminescence characteristics measured at $T<10$ K.

Samples	Well width (Å)	Al fraction	Pl Stokes shift (meV)	PL Width (meV) FWHM
G50	25.7	0.36	6.0	8.0
G51	73.4	0.35	1.5	2.20
G52	149	0.36	$\sim$0	1.1
G55	112.5	0.36	0.8	1.40
G57	56.0	0.36	3.0	3.0
GWS196	102	0.33	0.2	0.7
S51	44	0.33		
KLB269	65	0.4	9	11
G1273	80	0.3		
FK141	90	0.2		

In Fig. 2 we show extracted values of spin relaxation rate (τ_s^{-1}) against electron confinement energy E_{1e}. The values for τ_s^{-1} were obtained by fitting single exponential functions to the tails of the ''difference'' or SCP-OCP graphs. This gives the spin-relaxation rate directly because electron recombination is much slower than the spin relaxation, and hence will not significantly affect the observed decay rate. The slow recombination time is indicated by the step-like behavior of the ''sum'' (Fig. 1) and OLP[11,12] signals. The values of E_{1e} were calculated using the effective mass approximation, assuming a conduction to valence band offset ratio of 57:43 and making allowance for variations of Al content of the barriers. Also included in Fig. 2 are data from Tackeuchi *et al.*[7]

Three mechanisms can contribute to the electron spin relaxation in bulk material; these are due to Bir, Aharonov, and Pikus (BAP), Elliott and Yafet (EY), and D'yakonov and Perel (DP).[2] The EY mechanism arises out of spin-orbit interaction and is unlikely to be significant due to the weak spin-orbit coupling of the conduction band.[15] The BAP mechanism comes from interaction of the electron spins with a population of unpolarized holes via exchange interaction, and is generally only significant in strongly *p*-type samples above a certain threshold concentration which increases with temperature and is of the order 10^{18} cm^{-3} in bulk GaAs at 300 K.[2] Our samples are undoped but in principle this mechanism might be important since our measurements are made in the presence of a background of photoexcited electrons and holes. However, the low-repetition-rate experiments on a selection of our samples clearly show that the background population has little or no effect on the spin relaxation, and furthermore the estimated ambient hole population is below the bulk threshold, so we do not believe that the BAP mechanism is significant in our measurements. Consequently we expect the (DP) interaction discussed below to be dominant at room temperature.

The DP mechanism in a zincblende structure semiconductor results from the lack of inversion symmetry and the spin-orbit coupling which gives a *k*-dependent splitting between the spin components of the conduction band for $k \neq 0$. The splitting can be regarded as an effective magnetic field acting on electron spin, leading to precession of the spin. Momentum scattering events cause fluctuations in this magnetic field, on a timescale fast compared to the spin precession so that spin reorientation occurs in many small random steps and is much slower than the precession period. This ''motional narrowing'' situation gives a spin relaxation rate proportional to the momentum relaxation time (τ_p). In bulk material the electron spin relaxation rate due to the DP mechanism is given by[2,16]

$$\frac{1}{\tau_s} = A\alpha_c^2 \tau_p \frac{(k_B T)^3}{\hbar^2 E_g}, \tag{1}$$

where A is a numerical factor dependent on the scattering

mechanism causing momentum relaxation, α_c is a parameter related to the spin splitting of the conduction band, and E_g is the band-gap energy. In quantum wells, for electron confinement energy $E_{1e} \gg k_B T$, and for relaxation of the spin component along the growth direction in a (100)-oriented sample, the rate becomes[17]

$$\frac{1}{\tau_s} = B\alpha_c^2 \tau_p \frac{E_{1e}^2 k_B T}{\hbar^2 E_g}, \tag{2}$$

where $B \approx A$. In high purity and moderately doped *p*-type bulk GaAs at room temperature τ_p is determined by phonon scattering, and published measurements[2,15] on a variety of samples have given a common value of τ_s^{-1} (bulk) $\sim 1.3 \times 10^{10}$ s^{-1}. This value is in agreement with the theoretical estimates based on the DP mechanism, Eq. (1). If phonon scattering, which is not strongly well width dependent,[18] remained the dominant mechanism in our quantum well samples we would expect from Eq. (2) to find, for $E_{1e} \gg k_B T$, that

$$\tau_s^{-1}(\mathrm{QW}) \approx \frac{E_{1e}^2}{(k_B T)^2}\, \tau_s^{-1}(\mathrm{bulk}). \tag{3}$$

The presence of additional scattering mechanisms in the quantum wells will tend to reduce τ_s^{-1} (QW) below this theoretical maximum.

The data in Fig. 2 converge on a value of τ_s^{-1} between 1×10^{10} and 2×10^{10} s^{-1} for low E_{1e}, in good agreement with the bulk value. For higher E_{1e}, τ_s^{-1} does follow a generally quadratic dependence in accordance with Eq. (2), but there is a fairly large spread, and indeed all the points lie a factor of 2 or more below the ideal upper limit suggested by Eq. (3). Thus it is clear that additional scattering mechanisms in the quantum wells play a significant role. The spread in the data is somewhat reduced if we concentrate on the two particular subsets of the data which we might expect to have similar strength for the additional momentum relaxation. Tackeuchi *et al.*'s data (open symbols) come from two different wafers where the well thickness varied across the wafer. The two sets of three points were each found to fit curves of the form $\tau_s^{-1} = \beta \times E_{1e}^{\gamma}$, where $\gamma = 2.2$,[7] and taken together the two sets show an approximately parabolic dependence on E_{1e}. Similarly, the G-series samples (solid circles) appear to follow a smooth parabolic trend except for the point at highest E_{1e}. As a further test of Eq. (2) our measurements of mobility for KLB269 and S51 give values of τ_p of 1.83×10^{-13} and 1.75×10^{-13} s, respectively. Using these values we obtain $B\alpha_c^2 \hbar^{-2} = 1.4 \times 10^3$ and 1.2×10^3 eV^{-2} ps^{-2}, respectively, which are in reasonable agreement with each other, as required by the theory, Eq. (2).

We conclude that the behavior of the spin-relaxation rate in these samples is consistent with the dominance of the DP mechanism, and that momentum relaxation mechanisms in addition to optical phonon scattering are important in these QWs. These mechanisms are sample dependent, but allow clear quadratic behavior with E_{1e} to be observed for samples from common growth runs, as suggested by the theoretical expressions for constant momentum relaxation rate. MBE-grown material is generally of high quality, with very low point defect concentration and in this case the wells are pure GaAs, so it is natural to conclude that while phonon scattering is still dominant, scattering by interface roughness in the structures is important and that variations in interface morphology cause the significant differences between samples from different growth runs having a given value of E_{1e}. The strong deviation of the narrowest of our samples from a parabolic dependence on E_{1e} suggests that mobility is strongly reduced in the narrowest quantum wells, perhaps due to the increasing importance of interface roughness scattering in narrow wells.

Finally we point out that the very long relaxation times of electrons in *n*-type $Zn_{0.8}Cd_{0.2}Se$ quantum wells and ZnSe epilayers[3] are also consistent with the DP mechanism, taking into account the much lower mobilities of these materials.[19] For example, to scale the spin-relaxation time of sample KLB269 to 1 ns would require a change of mobility to the order of 0.02 m^2 V^{-1} s^{-1} which would be reasonable for these doped II–VI materials.

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