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Hanle Effect Measurements of Spin Lifetime in $Zn_{0.4}Cd_{0.6}Se$ Epilayers Grown on InP

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We use the Hanle effect to study spin relaxation in $Zn_xCd_{1-x}Se$ epilayers grown on lattice-matched InP substrates. We study three samples with a fixed composition ($x = 0.4$) and with varying levels of n-doping, as well as an undoped sample with $x = 0.5$. Our measurements show that the spin relaxation time changes non-monotonically as a function of carrier density, with a maximum transverse spin lifetime of ∼10.5 ns at low temperatures for a sample doped near the metal–insulator transition.

KEY WORDS: Hanle effect; spin relaxation; (Zn,Cd)Se.

Recent time-resolved Faraday rotation (TRFR) measurements have revealed relatively long inhomogeneous transverse spin lifetimes (T_2^*) in a variety of n-doped semiconductors [1–4]. Since these measurements establish a lower limit for the intrinsic spin coherence time T_2, they have revived interest in the fundamental physics of spin relaxation in semiconductors [5] with a view towards quantum information processing [6]. The wide band gap semiconductors ZnSe and $Zn_xCd_{1-x}Se$ are of particular relevance in this context since T_2^* has a weak temperature dependence, with values ranging from tens of nanoseconds at liquid helium temperatures to a few nanoseconds at room temperature [1,7]. However—unlike GaAs where extensive measurements of spin relaxation have been reported as a function of n-doping [2,8–11]—there is still a need for systematic studies of spin relaxation in ZnSe-based alloys over a broad range of parameters, such as carrier density, alloy composition and strain. In GaAs, detailed studies have shown that T_2^* increases dramatically upon the slightest n-doping, reaches a maximum plateau in the vicinity of the metal-insulator transition (MIT) and then decreases again at higher carrier densities [2,11].

Qualitatively similar behavior is suggested by TRFR measurements of spin lifetimes in modulation-doped $Zn_{0.8}Cd_{0.2}Se$ quantum wells and in n-doped ZnSe epilayers: in both cases, n-doping results in a dramatic increase in T_2^* compared with undoped samples [1,7]. The longest values of T_2^* in these II–VI materials have been found for n-ZnSe epilayers at a carrier density $n \sim 5 \times 10^{16}$ cm^{-3}, varying from ∼60 ns at 4.2 K to ∼10 ns at 80 K. Here, we use steady state measurements of the magnetic field depolarization of photoluminescence (the "Hanle effect") to probe transverse spin lifetimes in high quality $ZnCd_{1-x}Se$ epilayers with a large Cd content ($x =$ 0.5–0.6) grown on lattice-matched InP substrates. These samples provide access to a different regime of electronic structure parameters: for instance, we estimate that the ratio of the spin–orbit coupling to the band gap in $Zn_{0.4}Cd_{0.6}Se$ is ∼25% smaller than in ZnSe. If spin relaxation in these II–VI materials occurs via the Dyakonov-Perel (DP) [12] mechanism, then—all else being equal—we might expect the smaller conduction band spin splitting to result in longer values of T_2^* in $Zn_{0.4}Cd_{0.6}Se$ compared to ZnSe [13]. The large Cd content may also allow the exploration of possible coupling to nuclear spin since

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Table I. Physical Properties of $Zn_xCd_{1-x}Se$ Epilayers Used in the Current Study

Sample	n (cm^{-3})	μ (cm^2/Vs)	X (eV)	T_2^* (ns)	T_S (ns) at 4.2 K	T_S (ns) at 200 K
A	8.0×10^{16}	250	2.137	7.5	4.7	1.4
B	4.3×10^{17}	310	2.154, 2.143	10.5	2.95	1.03
C	1.1×10^{18}	170	2.138	2.0	—	—
D	—	—	2.179	0.7	—	—

a significantly larger fraction (~25%) of naturally occurring Cd isotopes have a nuclear spin compared to Zn and Se isotopes.

In the Hanle effect, we measure the magnetic field-dependent depolarization of photoluminescence (PL) excited using circularly polarized light in the Voigt geometry. The excitation creates spin-polarized electrons and holes with angular momentum perpendicular to an external magnetic field B. Since the holes relax their spin very rapidly, the precessing spin-polarized electrons recombine with unpolarized holes, emitting photons whose degree of circular polarization p provides information about the transverse electron spin relaxation. Measurements of the magnetic field dependence of the PL polarization—defined as $p = (I_+ - I_-)/(I_+ + I_-)$, where I_+ and I_- are the right and the left circular polarization components of the PL—yield a Lorentzian depolarization curve $p(B)$ given by [5]:

$$p(B) = \frac{p(0)}{1 + (\Omega T_S)^2}. \tag{1}$$

In this expression, $p(0)$ is the circular polarization in zero magnetic field, $\Omega = g\mu_B B/\hbar$ the angular Larmor precession frequency for electrons (g is the electron g-factor and μ_B is the Bohr magneton) and T_S an effective electron spin lifetime. Under steady-state conditions, T_S depends on both the transverse electron spin lifetime T_2^*, as well as on an electron "lifetime" $\tau_J = G/n$:

$$\frac{1}{T_S} = \frac{1}{\tau_J} + \frac{1}{T_2^*} = \frac{G}{n} + \frac{1}{T_2^*}, \tag{2}$$

where n is the equilibrium electron concentration and G the rate of electron–hole pair generation rate per unit volume. Thus, T_2^* can be estimated from power-dependent Hanle measurements by extrapolating to the limit of zero laser power [8]. We note that bound exciton emission usually dominates the PL spectra in lightly doped semiconductors; excitonic effects are even more important in the wide band gap alloys studied here. However, the linewidth measured in the Hanle effect under nonresonant excitation conditions essentially reflects the spin relaxation characteristics of the equilibrium electron population.

We study a set of 1 μm-thick $Zn_xCd_{1-x}Se$ epilayers grown by molecular beam epitaxy on InP(100) substrates. The epilayers are closely lattice matched to the substrate and are of high crystalline quality as verified by narrow X-ray rocking curve (full width at half maximum (FWHM) ~60 arcsecond) and a low defect density ($<10^5$ cm^{-2}) [14]. Three $Zn_{0.4}Cd_{0.6}Se$ samples are doped n-type with Cl to the level of 8.0×10^{16} cm^{-3} (A), 4.3×10^{17} cm^{-3} (B), and 1.1×10^{18} cm^{-3} (C) and have a room-temperature mobility of 250 cm^2/Vs (A), 310 cm^2/Vs (B), and 170 cm^2/Vs (C) (Table I). Measurements of electrical transport in n-ZnSe and n-CdSe have shown that the metal–insulator transition occurs at a critical carrier density of $n_C = 3.7 \times 10^{17}$ and 2.8×10^{17} cm^{-3}, respectively [15,16]. An interpolation to the alloy composition in this work suggests that sample A is on the insulating side of the MIT, sample B is near the boundary of the MIT, and sample C is on the metallic side of the MIT. An undoped $Zn_{0.5}Cd_{0.5}Se$ epilayer (D) is also included for comparison.

Hanle effect measurements are carried out in a liquid He continuous flow cryostat with the sample placed on a cold finger in vacuum. A table-top electromagnet is used to apply a magnetic field in the sample plane. PL is excited at near-normal incidence using circularly polarized photons of wavelength 488-nm line from a continuous wave mixed gas Ar-Kr laser. We note that our present set up is constrained to excitation that is ~0.3–0.4 eV above the band gap of the material (2.18 eV). This probably results in additional spin scattering as energetic carriers cascade down to the band edge before recombining. PL is collected in a back scattering geometry and passed through a circular polarization analyzer composed of a photo-elastic modulator (PEM) and a linear polarizer (the PEM is used at a quarter wave plate mode to switch the polarization between opposite circular polarizations at a 50 kHz frequency). PL is spectrally dispersed with a spectrometer and detected by a photo multiplier tube connected to a lock-in amplifier working synchronously with the PEM.

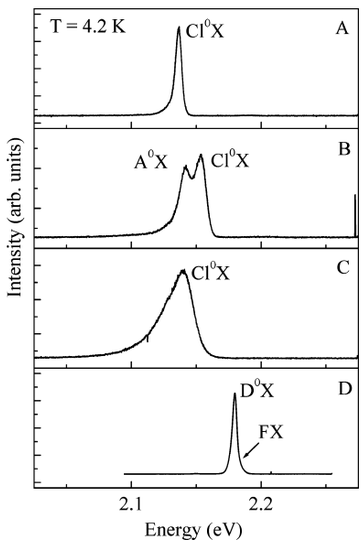

Fig. 1. $Zn_xCd_{1-x}Se$ photoluminescence spectra taken at 4.2 K. The excitation wavelength and intensity are 488 nm and ~0.5 W/cm^2, respectively.

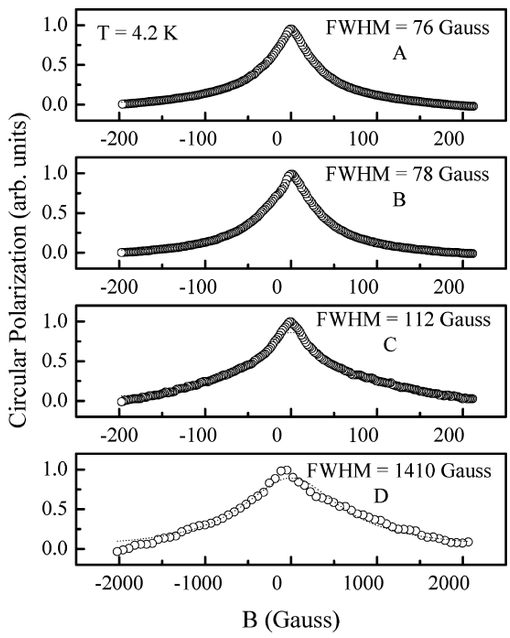

Fig. 2. $Zn_xCd_{1-x}Se$ Hanle depolarization curves taken at 4.2 K. The excitation wavelength and intensity are 488 nm and ~2 W/cm^2, respectively. Symbols correspond to the data; the dotted lines are fits to a Lorentzian.

In Fig. 1, we show the low-temperature (4.2 K) PL spectra. All the spectra are dominated by a strong, fairly sharp peak in the band edge region. Using power-dependent measurements we determine that 2.137 eV PL from sample A (FWHM of 5 meV) is due to the recombination of neutral donor bound excitons (D^0X). The PL from sample B is a sum of D^0X at 2.154 eV (FWHM of 7 meV) and acceptor-bound exciton (A^0X) at 2.143 eV (FWHM of 11 meV) lines. The PL from sample C is at 2.138 eV; it is broader (FWHM of 15 meV) and is a sum of a few bound exciton lines. The PL from sample D is at a slightly higher energy of 2.179 eV (due to a larger Zn content); it has a FWHM of 4 meV and has contributions from both free exciton and D^0X lines [17].

We measure the degree of circular polarization in the high-energy shoulder of PL. As is well known, electric dipole selection rules determine a maximum circular polarization of 25% in cubic semiconductors [5]. In practice, the measured polarization is far smaller due to spin scattering of the photo-excited carriers during thermalization. This is certainly the case in our experiments where the photo-created electrons have to lose ~200 meV before recombining. In the samples studied, the polarization increases with excitation laser intensity and reaches ~1.2% at the maximum excitation intensity (~2 W/cm^2). Since we do not observe saturation, we expect that a higher value of polarization can be obtained with a higher pumping intensity.

In Fig. 2, we show Hanle depolarization curves for all the samples at 4.2 K. The Lorentzian shape of the Hanle curves suggests that the spin orientation is homogeneous throughout the layer thickness and that spin diffusion effects are negligible. We fit the experimental data using Eq. (1) using an electron g-factor $g \sim 1$, estimated from direct TRFR measurements in lower composition $Zn_{1-x}Cd_xSe$ ($g = 1.1$) [1,7] and indirect magneto-optical measurements in bulk (wurtzite) CdSe ($g = 0.7$) [18]. These fits yield values of the effective spin lifetime $T_S = 2.95$, 2.93, 2.00, and 0.16 ns, for samples A, B, C, and D, respectively. As mentioned earlier, we anticipate additional spin scattering due to the high energy photo-excitation; hence, the values of T_S reported here provide a lower limit to the intrinsic values. These expectations are consistent with the observation of significantly narrower Hanle linewidths (ranging from 10 to 20 G) measured on two of these samples using longer wavelength excitation (532 nm) (Stern and Awschalom, private communication).

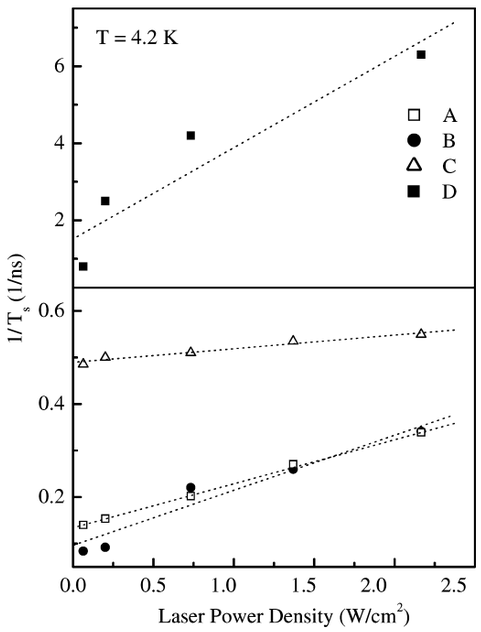

Fig. 3. Inverse of the effective spin lifetime plotted as a function of excitation laser power. The symbols correspond to the data and the dotted lines are linear fits. The data are measured at 4.2 K.

In Fig. 3 we show plots of $1/T_2^*$ versus excitation laser intensity (symbols). From a linear fit to the data (dotted lines), we determine $T_2^* = 7.5$, 10.5, 2.0, and 0.7 ns for samples A, B, C and D, respectively (Table I). As reported for n-doped GaAs [2,8] and n-ZnSe [1,7], we find that even with moderate n-doping ($N_d = 8.0 \times 10^{16}$ cm^{-3}), T_2^* is greatly enhanced compared with the undoped sample. This increase in spin lifetime is probably due to the suppression of the Bir-Aronov-Pikus (BAP) [19] mechanism in which spin coherence is lost due to the spin–flip transitions caused by electron–hole exchange. In studies of spin relaxation in n-GaAs, it has been suggested that the hyperfine interaction between localized electrons and lattice nuclei spins also contributes to spin relaxation [20,21]. Hopping electron transitions from one site to another become more probable with increasing donor density; this averages out the hyperfine interaction, decreases the effect of nuclear-spin fluctuations, and increases the spin lifetime. However, oblique Hanle effect measurements in our samples have thus far not shown any indications of nuclear polarization by optical orientation (Stern and Awschalom, private communication). This suggests that—despite the large fraction of Cd nuclei with nuclear spin–spin relaxation via the hyperfine interaction is perhaps not significant.

We have also measured the temperature dependence of the Hanle effect in samples A and B over the temperature range 4.2–200 K. These measurements (currently carried out only at a fixed pump intensity) show that T_S is nearly constant between 4.2 and 25 K and then decreases linearly at higher temperatures, becoming three times shorter when the temperature is raised from 4.2 to 200 K (Table I). However, since the carrier lifetime changes as a function of temperature, either power-dependent Hanle effect measurements at different temperatures or time-resolved optical measurements are necessary to measure the temperature dependence of T_2^*.

In summary, we have used the Hanle effect to study spin relaxation in $Zn_xCd_{1-x}Se$ epilayers with a high Cd concentration ($x = 0.5$–0.6). Power-dependent measurements at low temperatures reveal that a moderate level of n-type doping significantly increases the transverse spin lifetime from 0.7 ns for an undoped epilayer to 10.5 ns for an epilayer with a doping level close to the MIT.

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