Electron spin lifetimes in long-wavelength $Hg_{1-x}Cd_xTe$ and InSb at elevated temperature

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We have made direct pump-probe measurements of spin lifetimes in long-wavelength narrow-gap semiconductors at wavelengths between 4 and 10 μm and from 4 to 300 K. In particular, we measure remarkably long spin lifetimes $\tau_s \sim 300$ ps even at 300 K for epilayers of degenerate n-type InSb. In this material the mobility is approximately constant between 77 and 300 K, and we find that τ_s is approximately constant in this temperature range. In order to determine the dominant spin relaxation mechanism we have investigated the temperature dependence of τ_s in nondegenerate lightly n-type $Hg_{0.78}Cd_{0.22}Te$ of approximately the same band-gap as InSb and find that τ_s varies from 356 ps at 150 K to 24 ps at 300 K. In this material lattice scattering dominates giving a $T^{-3/2}$ dependence for the mobility, and we expect a strong temperature dependence of τ_s. There are two main models that have been invoked for describing spin relaxation in narrow-gap semiconductors: the Elliott-Yafet (EY) model which gives a $T^{-7/2}$ dependence of τ_s in this limit and the D'yakonov-Perel model which gives a $T^{-3/2}$ dependence. Our results, both in magnitude and temperature dependence of τ_s, imply that the EY model dominates in these materials.

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I. INTRODUCTION

We have used the Dutch free-electron laser (entitled FELIX, free-electron laser for infrared experiments) to measure spin lifetimes in the long-wavelength narrow-gap semiconductors (NGS's) $Hg_{1-x}Cd_xTe$ and InSb at wavelengths between 4 and 10 μm and from 4 to 300 K. In particular, we measure remarkably long spin lifetimes even at 300 K in degenerate n-InSb. We note that apart from one report of spin relaxation in InAs (Ref. 1) at 300 K, these are the first measurements of spin lifetime to be made in NGS's at temperatures above nitrogen temperature. These first results are being extended to a range of compositions, mobilities, and temperatures, both to explore the physics determining spin relaxation in NGS's and to establish conditions required to optimize the spin lifetime.

In bulk semiconductors three main spin relaxation processes have been found to be important in optical orientation experiments: the Elliott-Yafet[2] (EY), D'yakonov-Perel[3] (DP), and the Bir-Aronov-Pikus[4] (BAP) mechanisms. The BAP mechanism is thought to be particularly important in p-type wide-gap materials and is based on the electron-hole exchange interaction. By contrast with the other two processes, it depends directly on the concentration of holes and is not thought to be important in n-type narrow-gap semiconductors.

The EY mechanism results from the fact that in real crystals Bloch states are not spin eigenstates because of the strong spin-orbit coupling induced by the lattice ions. In the presence of strong spin-orbit coupling, the valence-band states have mixed spin character. In NGS's the conduction-electron states, in turn, are strongly mixed with the valence states through the $\mathbf{k}\cdot\mathbf{p}$ interaction across a narrow energy gap. In this case spin-independent interactions with impurities, boundaries, phonons, etc., can connect spin-up and -down electrons, leading to a relaxation whose rate $1/\tau_s$ is proportional to $1/\tau_p$ where τ_p is the orbital momentum (mobility) scattering time. This process is thought to be important in NGS's.[5,6]

The DP mechanism is expected to dominate in large-gap n-type semiconductors that lack inversion symmetry so that the $\mathbf{k}$-dependent spin-orbit interaction lifts the spin degeneracy even in the absence of a magnetic field.[3] Somewhat counterintuitively and in contrast with EY, the spin relaxation rate $1/\tau_s$ is subject to motional narrowing and is therefore directly proportional to the orbital (mobility) scattering time τ_p. In NGS's both the EY and DP mechanisms may be important, as shown by reports of spin relaxation in InSb at helium temperature[5–7] and InAs at room temperature[1]—the former being interpreted in terms of EY and the latter of DP. Typically spin lifetimes in the range 1–10 ns were obtained from spin resonance in InSb at helium temperatures, whereas a spin lifetime of 20 ps was reported for InAs at 300 K. It therefore has been unclear whether enhancement of the spin lifetime may be achieved with higher or lower mobility samples, which is affected by, e.g., layer thickness through interface scattering, as well as by sample temperature. Additional factors influence the spin relaxation, and we report its

measurement as a function of doping, mobility, and temperature in order to investigate systematically the dominant mechanisms.

We find, in accordance with the earlier work,[5,6] that at least for small-band-gap HgCdTe and InSb it is indeed the EY mechanism that dominates, giving good agreement with both the magnitude and temperature dependence of τ_s up to 300 K (see below).

A key step in controlling spin relaxation has been the observation of a large enhancement of the spin memory of electrons by *n*-type doping in both bulk (GaAs) semiconductor and quantum wells (ZnCdSe): τ_s is found to increase by several orders of magnitude over corresponding intrinsic material.[8,9] We have found a similar effect in *n*-HgCdTe and *n*-InSb, where we obtain more than an order of magnitude enhancement of τ_s between intrinsic and *n*-type material. The present tentative explanation for this is that in intrinsic samples, photoexcited spin-polarized holes (of concentration $>1\times10^{16}$ cm^{-3} in our case) efficiently scatter electron spins,[4,8–11] leading to a short spin relaxation time, which dominates the measured value. In the presence of a degenerate Fermi sea, the effect of the photoexcited holes is removed by screening, and a longer electron spin relaxation time is measured. It is also true that by going to a degenerate distribution one replaces kT by the Fermi level E_F in the theoretical expression for τ_s, thereby removing some of the strong temperature dependence of τ_s.

II. EXPERIMENTAL METHOD

We have developed a setup at FELIX to utilize both the polarization pump-probe method[1,12,13] and the Faraday rotation method[9,11,13] in the 5–10 μm region in order to examine the spin relaxation times of bulk epilayers of long-wavelength $Hg_{1-x}Cd_xTe$ and InSb as a function of temperature, doping, and mobility. The principal modification we have to make is that we are using FELIX to generate the long-wavelength (ps) pump-probe radiation, with corresponding long-wavelength plane and circular polarizers. We use the three-beam balanced pump-probe setup described elsewhere.[14] The only difference here is that we have a circular polarizer in the pump beam consisting of a variable quarter-wave plate and utilizing the plane polarization of FELIX. The probe beam was either circularly polarized for the polarization pump-probe technique or plane polarized for the Faraday rotation technique. The short-wave limit of our measurements (~4 μm) and the pulse duration ($\sim1-5$ ps) are determined by FELIX. A varitemp cryostat enables measurements in the range 4–300 K.

Both methods rely on the fact that because of the different transition strengths associated with heavy hole and light hole to conduction-band transitions from a complex (spin-orbit mixed) valence band, a partially spin-polarized distribution will be created by pumping with circularly polarized radiation. We note that the spin relaxation will be dominated by electrons, as the fourfold degeneracy of the valence band at $\mathbf{k}=0$ and the large spin-orbit coupling are expected to result in extremely rapid decay of the hole spin polarization.

If we take the case of (e.g.) a left circularly polarized pump pulse (σ), the relative concentration of optically generated spin-up to spin-down electrons is 3:1, leading to a maximum spin polarization of $P_s=0.5$ where[1,12,13]

$$P_s=(n\uparrow-n\downarrow)/(n\uparrow+n\downarrow). \tag{1}$$

In the circularly polarized pump-probe method one determines the difference between probe transmission for the same circularly polarized pump and probe (SCP) and opposite circularly polarized pump and probe (OCP). The actually measured probe transmission is proportional to the absorption change $\Delta\alpha$. A σ_- probe will experience $\Delta\alpha_-\propto3n_{\mathrm{opt}\uparrow}+n_{\mathrm{opt}\downarrow}$, while a σ_+ probe will experience $\Delta\alpha_+\propto3n_{\mathrm{opt}\downarrow}+n_{\mathrm{opt}\uparrow}$. Thus the optical polarization signal becomes

$$\begin{aligned}P_{\mathrm{opt}}&=(\mathrm{SCP}-\mathrm{OCP})/(\mathrm{SCP}+\mathrm{OCP})\\&=(\Delta\alpha_--\Delta\alpha_+)/(\Delta\alpha_-+\Delta\alpha_+)=0.5P_s,\end{aligned} \tag{2}$$

which has maximum value 0.25.

If there is a background concentration of unpolarized electrons, n_b, then this will be correspondingly reduced,[1]

$$P=0.25/(1+n_b/n_{\mathrm{opt}}). \tag{3}$$

In the Faraday rotation method, the probe is plane polarized. Then, the excess population of $n\uparrow$ over $n\downarrow$ electrons produced by σ pumping gives a differential dispersion between the left and right circularly polarized components of the plane polarized probe, and hence a Faraday rotation. We measure this by setting an analyzer at $+$ or $-45°$ to the polarizer and determining the difference in transmitted signal intensity I_{+45} and I_{-45}, respectively. Then for small angles we have for the Faraday rotation, θ,

$$2\theta\approx(I_{+45}-I_{-45})/(I_{+45}+I_{-45}). \tag{4}$$

At the present time it has not been possible to set up the differential optical bridge method of measurement[8–11] owing to the difficulty of obtaining a fast matched detector system at these long wavelengths. Our signals are sufficiently large to overcome this, but obviously the method would have a great advantage from the point of view of data retrieval speed and consistency. In the absence of the optical bridge, it is more convenient for us to use the polarization pump-probe method where possible, because we do not have any wavelength restrictions resulting from imperfect plane polarizers. We utilize the plane polarized beam from FELIX in conjunction with a variable quarter-wave plate (optimized for each wavelength by simply adjusting the plate tilt angle), so that we are not restricted to any particular wavelength range.

We have measured the set of InSb and $Hg_{1-x}Cd_xTe$ samples shown in Table I, produced (either from the in-house stock or purpose grown) by QinetiQ and BAE Systems, specifically to study the effect of carrier type and concentration, and carrier (Hall) mobility on electron spin lifetimes of NGS's.

TABLE I. Sample characteristics of InSb and HgCdTe.

Sample code	Material	T (K)	n_d (p) (cm^{-3})	μ ($cm^2\,V^{-1}\,s^{-1}$)
ME1654	InSb	77	2.6×10^{15}	7870
		300	1.5×10^{16}	68700
ME1655	InSb	77	2.8×10^{15}	29900
		300	1.4×10^{16}	62700
ME1629	InSb	77	2.08×10^{17}	47700
		300	2.26×10^{17}	41500
ME1722	InSb	77	2×10^{17}	45000
G256	n-$Hg_{0.78}Cd_{0.22}Te$	150	3×10^{16}	50000
		220		18000
		300		7000
GVG763	p-$Hg_{0.7}Cd_{0.3}Te$	150	$p_a=2\times10^{16}\ cm^{-3}$	40000
GVG775	p-$Hg_{0.78}Cd_{0.22}Te$	150	$p_a=1\times10^{17}\ cm^{-3}$	20000

III. EXPERIMENTAL RESULTS

A. InSb

Chronologically, our first experiments were on lightly doped n-InSb ($n_d\sim1\times10^{15}\ cm^{-3}$ at 77 K), ME1654 and ME1655. Results are shown for sample ME1655 in Fig. 1. A surprisingly short spin lifetime was obtained (measured decay time $\tau_s\sim16$ ps between 77 and 300 K). However, the fact that the initial degree of polarization was $P\sim0.2$, which is close to the theoretical limit of Eq. (2), gives confidence to the measurement (given imperfect circular polarization at these long wavelengths). In order to test whether these measurements were being dominated by photoinduced holes under FELIX excitation ($n_{opt}\sim1\times10^{16}\ cm^{-3}$ at the intensities used), we repeated the measurements for more heavily doped n-type material (sample ME1629, $n_d=2\times10^{17}\ cm^{-3}$). A substantially longer spin lifetime was obtained ($\tau\sim300$ ps at 300 K), in broad agreement also with the prediction (200 ps, for a smaller carrier concentration) in Ref. 15. This sample was difficult to measure using this technique because the size of the polarization achieved was reduced in the presence of a high n_b, by a factor of ~5 in agreement with the expectation of Eq. (3). We have estimated $n_{opt}\sim5\times10^{16}\ cm^{-3}$ in this case. (N.B. There was a significant Burstein-Moss shift measured in the doped sample, necessitating pumping further above the band edge, with correspondingly higher absorption coefficient and resultant n_{opt} than for ME1655.) In order to check that this result was not some artifact of the experimental method used, we repeated it using the Faraday rotation method and obtained consistent results, Fig. 2. We also measured the temperature dependence for the n-type material (both ME1629 and ME1722) where we obtained an approximately constant value for τ_s. N.B. In this case the mobility was more or less constant between 77 and 300 K (47 700 and 41 500 $cm^2\,V^{-1}\,s^{-1}$, respectively—the ionized impurity and lattice scattering processes approximately cancel in this range), and degenerate statistics apply.

A feature which becomes apparent on inspection of Figs. 1 and 2 is that with increasing n_{opt}, the decay of population as a whole by Auger recombination becomes more pro-

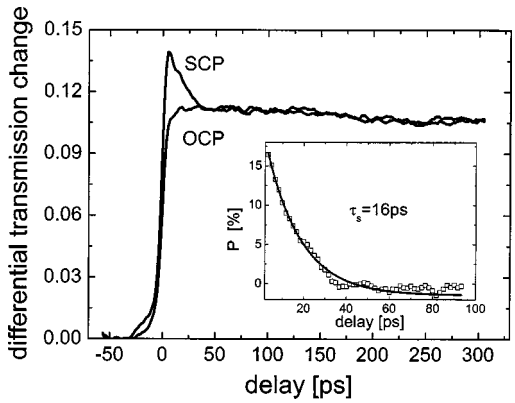

FIG. 1. Pump-probe transmission change as a function of probe delay time for same-same and same-opposite pump-probe circular polarization (SCP and OCP, respectively) for lightly doped n-InSb (sample ME1655, $n_d=2\times10^{15}\ cm^{-3}$). On the right-hand side is shown the spin polarization as a function of probe delay, giving a measured lifetime of $\tau_s=16$ ps at 150 K.

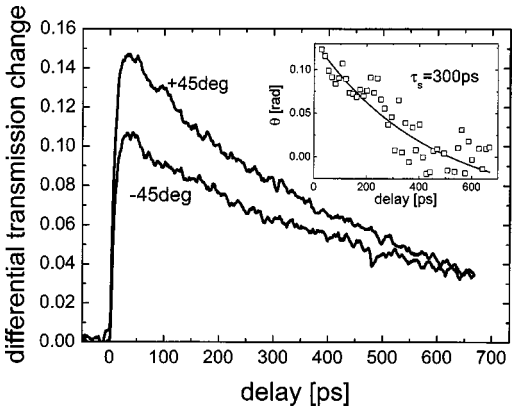

FIG. 2. Lifetime determination by Faraday rotation method for n-InSb (sample ME1629, $n=2\times10^{17}\ cm^{-3}$) at 300 K. The pump utilized the plane polarization of FELIX, and the probe was plane polarized at an angle of $+$ or $-45°$ to the pump. The fractional transmission change is proportional to θ. On the right-hand side is shown the decay of Faraday rotation, giving a measured lifetime of $\tau_s=300$ ps at 300 K.

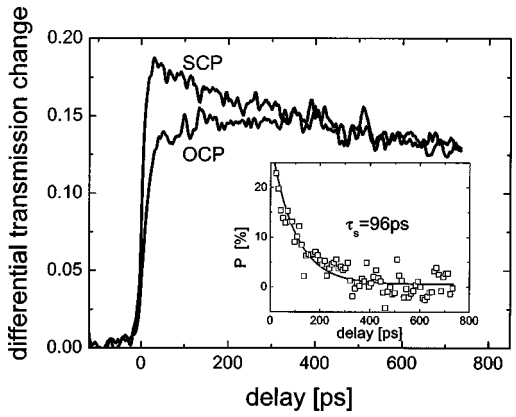

FIG. 3. Pump-probe transmission change as a function of probe delay time for SCP and OCP, respectively, for n-$Hg_{0.78}Cd_{0.22}Te$ (sample G256, $n_d=3\times10^{16}$ cm^{-3}) at 220 K. On the right-hand side is shown the spin polarization as a function of probe delay, giving a measured lifetime of $\tau_s=96$ ps at 220 K.

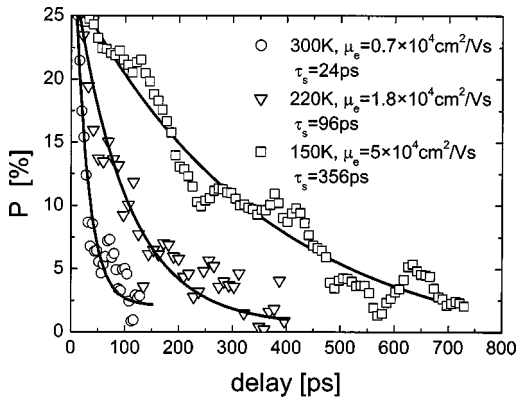

FIG. 4. Spin polarization as a function of probe delay for n-$Hg_{0.78}Cd_{0.22}$ (sample G256, $n_d=3\times10^{16}$ cm^{-3}) at 150, 220 and 300 K.

nounced. The Auger rate is $1/\tau_A \approx C(n_b+n_{opt})^2$, where $C \sim 2\times10^{-26}$ cm^6 s^{-1} for these materials.[16] This gives Auger recombination times in good agreement with the average experimental decay times, $(\Delta\alpha_+ + \Delta\alpha_-)/2$, in all cases (20 ns and 800 ps for Figs. 1 and 2, respectively).

B. HgCdTe

In parallel with the above we have been investigating $Hg_{1-x}Cd_xTe$ (of composition $x=0.2-0.3$) with a band gap comparable to that of InSb. N.B. The band gap of InSb increases as the temperature is lowered, whereas that of $Hg_{1-x}Cd_xTe$ has the opposite temperature dependence, so that it is not possible to get a perfect match. Our first results for moderately doped p-type $Hg_{1-x}Cd_xTe$ (GVG763) gave short lifetimes $\tau\sim20$ ps, consistent with the results for lightly doped InSb. Furthermore, when we repeated the measurements for heavily doped p-$Hg_{1-x}Cd_xTe$ (GVG775, $p \sim 1\times10^{17}$ cm^{-3}) we obtained an even shorter lifetime, probably limited by the resolution of our method (~5 ps). However, when we then moved to the moderately n-type sample (G256, $n\sim3\times10^{16}$ cm^{-3}) we obtained a substantially longer lifetime, Fig. 3, in accordance with the InSb results. Finally, we again measured the temperature dependence of the lifetime, Fig. 4, where we expect a pronounced effect since lattice scattering is dominant for HgCdTe and we have a nondegenerate distribution. We found that the spin lifetime scaled approximately as $T^{-7/2}$ as expected for the EY model for a nondegenerate distribution with lattice scattering dominant, as described in the next section.

IV. ANALYSIS AND DISCUSSION

As already pointed out the scattering models that can contribute to spin relaxation in NGS's are the EY and DP models as follows.

Elliott-Yafet[2,5,6] (EY). The conduction–valence-band interaction through $\mathbf{k}\cdot\mathbf{p}$ terms, in the presence of nonparabolicity and strong spin-orbit coupling, leads to the following spin relaxation rate for ionized impurity scattering and degenerate statistics:

$$\frac{1}{\tau_s}\approx\frac{32}{27}\left(\frac{\gamma E_F}{E_G}\right)^2\left(\frac{1-\gamma/2}{1-\gamma/3}\right)^2\frac{1}{\tau_p}, \tag{5}$$

where $\gamma=\Delta/(E_G+\Delta)$ and Δ is the spin-orbit splitting of the valence band.

In the other limit of lattice scattering and nondegenerate statistics the expression becomes

$$\frac{1}{\tau_s}\approx 2\left(\frac{\gamma kT}{E_G}\right)^2\left(\frac{1-\gamma/2}{1-\gamma/3}\right)^2\frac{1}{\tau_p}. \tag{6}$$

D'yakonov-Perel[3,6] (DP). The conduction band k^3 inversion asymmetry splitting $(\Delta E=\beta k^3)$ gives a pseudofield about which electrons precess, with relaxation rate for lattice scattering and nondegenerate statistics:

$$\frac{1}{\tau_s}\approx 0.8\beta^2\frac{(kT)^3}{\hbar^2 E_G}\tau_p. \tag{7}$$

For the case of degenerate statistics one obtains a similar expression with kT being replaced by E_F.

By comparing Eqs. (6) and (7) one immediately sees that for the case of moderately doped HgCdTe in the limit of lattice scattering (τ_p proportional to $T^{-3/2}$) and nondegenerate statistics one expects τ_s to depend on $T^{-7/2}$ on the EY model and $T^{-3/2}$ on the DP model. For the temperature range 150–300 K on the EY model one expects τ_s to decrease by a factor of ~12, which is in good agreement with the experimental finding of 14.8. (N.B. The DP model would give a decrease by a factor of ~3.) We obtain the material parameters of Eq. (6) from well-known expressions[17] for the energy gap E_G, the conduction-band edge effective mass, m_c, and electron mobility $(\mu=e\tau_p/m_c)$:

$$E_G(x,T)=-0.302+1.93x-0.81x^2+0.823x^3+5.35\times10^{-4}(1-2x)(T^3-1822)/(T^2+255.2), \tag{8}$$

$$m_0/m_c=1+(E_p/3)\{(2/E_G)+[1/(E_G+\Delta)]\}, \tag{9}$$

where $E_p = 19$ eV and $\Delta = 1.0$ eV.[17] The actual values obtained from Eq. (6)—$\tau_s(300) = 13$ ps, $\tau_s(220) = 67$ ps, $\tau_s(150) = 360$ ps—are in remarkably good agreement with those obtained experimentally as shown in Fig. 4.

Finally, taking the degenerate case with ionized impurity scattering dominant for the InSb sample ME1629 (Fig. 2), the nonparabolicity of the conduction band is important at $n = 2 \times 10^{17}$ cm^{-3} (both for the effective mass m_c/m_0 and the Fermi level), and we take the material parameters at 300 K from previous work:[18] $E_G = 0.18$ eV, $\Delta = 0.8$ eV, $E_F = 0.03$ eV, and $m_c/m_0 = 0.027$. We obtain a theoretical value of $\tau_s \sim 100$ ps from Eq. (5), in satisfactory agreement with the measured value of 300 ps considering the simplifying approximations of the model.

In summary, we have measured an approximately constant spin lifetime of ~ 300 ps for degenerate n-InSb at elevated temperatures up to 300 K. This is of the same order of magnitude as the earlier results at liquid helium,[5,6] but we emphasize that this is the first time that such long lifetimes have been established at 300 K in NGS's. Both the magnitude and the dependence of τ_s on temperature are consistent with the EY model for n-$Hg_{0.78}Cd_{0.22}Te$ and n-InSb.

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