Dynamic nuclear Overhauser shifts in Larmor beats from a quantum well

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Abstract

The significance of nuclear spin polarisation in time-resolved optical studies of III-V semiconductors is addressed. Electron Larmor beats in pump-probe reflectivity from a GaAs/AlGaAs quantum well show Overhauser shift of 0.7 T due to accumulated nuclear polarisation \( \langle I \rangle / I = 0.065 \). This leads to precision values of electron g-factor, elucidates nuclear spin pumping and diffusion mechanisms in quantum wells and informs discussion of implications for spin-electronics and transport.

The proposed application of electron spin transport and coherence in semiconductors to spin electronics and perhaps quantum computing \cite{1,2} will involve a high degree of conduction electron spin polarisation and extended electron spin lifetimes \cite{3}. Recently free Larmor precession continuing for \( \geq 100 \text{ ns} \) \cite{4} and spin transport over microns \cite{1,5} were observed in time-resolved studies of n-type GaAs and quantum wells using circularly polarised optical pulses. On the other hand, since all naturally occurring isotopes in III-V semiconductors have non-zero nuclear spin \( \hbar \), hyperfine interaction \( A_{I,S} \), can be expected to result in an equivalent level of polarisation of the nuclear spins \( \hat{I} \) with, conversely, profound effects on the behaviour of the electrons. These include built-in effective magnetic fields (Overhauser effects) and nonlinear or bistable response to applied external fields \( \hat{B} \). Such effects have long been known in cw optical phenomena in bulk semiconductors \cite{7,8,10} and in quantum wells \cite{11}, and in quantum magneto-transport \cite{11}, but consequences for time-resolved optical measurements and spin applications have been neglected. In this letter we report measurements of nuclear Overhauser shift \( \hat{B} \) in ps time-resolved Larmor beats \cite{12,13} in a GaAs/AlGaAs quantum well, revealing the dynamics and long timescales of nuclear spin polarisation by optical pumping and the change of electron spin precession frequency induced by a polarised nuclear spin system. In this experiment the effective internal field approaches 1 T although the nuclear polarisation is only 6.5%, emphasising the significance of nuclear phenomena. Proper account of the Overhauser shift gives excellent agreement between measured conduction electron Larmor frequencies and anisotropic k.p theory \cite{14}, while the dynamics provides new insights to nuclear spin diffusion.
in quantum well systems. We suggest that such effects may commonly influence and could be fruitfully exploited in time-resolved polarised measurements.

In our experiment (see inset to figure 1) a circularly polarised optical pump pulse from a mode-locked laser is absorbed in the quantum well (QW) at time $t_0$ and generates a transient population of conduction electrons with spin-polarisation $S(t_0)$, parallel (antiparallel) to the incident beam for $\sigma - (\sigma +)$ polarisation \[7\]. Time-evolution of $S$ is monitored through rotation, on reflection, of the plane of polarisation of a weak, delayed probe pulse, almost colinear with the pump, the rotation being proportional to the population difference of the electron spin-states \[8\]. Magnetic field, $B_{ext}$, applied at angle $\theta$ to the beam, causes the spins to precess while $S(t)$ decays comparatively slowly by spin-relaxation and recombination so that there is a non-precessing component of electron spin, $S_{av}$, parallel to the field. Integrated over many laser pulses, nuclear spin polarisation, $\langle I \rangle$, parallel to $S_{av}$, will build-up through mutual spin flip-flops with lattice nuclei driven by the hyperfine interaction. The polarisation $\langle I \rangle$ reacts back on the electron spins as an effective (Overhauser) magnetic field \[8\],

$$B_N = \frac{A(I)}{g_e \beta} \approx \frac{\langle I \rangle}{T} \text{tesla (1)}$$

where $g_e$ is the electron g-factor and $\beta$ is the Bohr magneton. The numerical value is calculated for a 9.6nm GaAs quantum well with $\theta=45^\circ$ (see below), using an average of $A$ over the isotopes $^{75}\text{As}$, $^{69}\text{Ga}$ and $^{71}\text{Ga}$ each with $I=3/2$ \[8\]. This is a significant field even when the nuclear polarisation is low. When $g_e$ is negative, as for bulk GaAs and GaAs/AlGaAs quantum wells wider than 5.5 nm \[8\], $B_N$ opposes $S_{av}$ (see fig. 1) and the Larmor frequency is $\Omega = g_e \beta (B_{ext} + B_N)/\hbar$. Reversal of the polarisation of the pump reverses $S_{av}$ and therefore also $\langle I \rangle$ and $B_N$, so increasing the Larmor frequency by $2 \frac{g_L \beta B_N}{\hbar}$.

For small nuclear polarisation and for $B_{ext} \gg B_L$ the internuclear dipolar field ( $10^{-4}$T), the steady state value is \[8\]

$$\langle I \rangle = \frac{5}{3} \left( \frac{\tau_s}{\tau_s + \tau_r} \right) \left( \frac{T_1^*}{T_1^* + T_{1e}} \right) \left( \frac{S(t_0)}{S} \right) \cos \theta (2)$$

where $t_s$ and $t_r$ are respectively electron spin-relaxation and recombination times; $T_1^*$ and $T_{1e}$ are respectively times characterising nuclear spin relaxation and angular momentum transfer from electronic to nuclear spins. As we shall see, the latter may involve spin diffusion as well as hyperfine coupling. We neglect the equilibrium electron spin due to applied field, which is of order 1% in the highest fields used in this experiment. Equation 2 shows that accumulation of nuclear polarisation can be avoided if $\theta=90^\circ$. The electron spins then precess on a disc, not a cone. $\langle I \rangle$ is also zero if $B_{ext} \ll B_L$ \[8\], often experimentally impractical. The main conditions, readily met, for build-up of $\langle I \rangle$ are $t_s \geq t_r$ so that photoelectrons remain spin-polarised during their lifetime, and $T_{1e} \leq T_1^*$ so that angular momentum transferred from electron to nuclear spin systems is not rapidly dissipated to the lattice. In III-V semiconductors, values of $t_s$ vary
from $\sim5$ ps to $\sim100$ ns \cite{4, 7, 15, 16, 17, 18, 19} while typically $t_r \geq 100$ ps. For Bloch states, $T_1$ has been estimated as $10^4$ s in GaAs for excitation density $10^{15}$ cm$^{-3}$ \cite{6} but it is reduced by a factor $10^5$ for nuclei in contact with electrons localised, for example, by interface roughness or donors \cite{7}. The nuclear spin-lattice relaxation time is of order minutes or hours at low temperatures, driven by phonon-modulation of electric field gradient in intrinsic material \cite{20} or mediated by hyperfine coupling to electron spins in n-type systems \cite{21}.

Our sample consisted of three undoped single GaAs quantum wells of widths 5.1 nm, 9.6 nm and 19 nm with 34 nm Al$_{0.33}$Ga$_{0.67}$As barriers and grown on an (001)-oriented n+ (10$^{18}$ cm$^{-3}$ Si-doped) substrate. A bias applied between the substrate and an ITO electrode on the top of the structure controlled the carrier type and concentration in the wells. Most of the measurements described here were performed at 10 K on the 9.6 nm well with $0.5 \times 10^{11}$ cm$^{-2}$ heavy holes injected to ensure that the effects were dominated by the photoexcited electrons \cite{13}. In fact, biasing the sample flat-band was found to make no significant difference to the results. This structure was also chosen to allow tests of inter-well spin diffusion. Pump and probe pulses were derived from a mode-locked Ti-Sapphire laser giving 2 ps pulses at 80 MHz repetition frequency and tuned to the $n=1$ heavy-hole absorption edge \cite{15}. We estimate pump excitation density $1.0 \times 10^{10}$ cm$^{-2}$ per pulse in the quantum well. The electron lifetime and spin relaxation time were determined to be $t_r \approx 240$ ps \cite{8} and $t_s \approx 200$ ps respectively. In the actual experimental geometry, dictated by the configuration of the magnet, the beams intersected at $3^\circ$ on the sample, at $45^\circ$ to the quantum well normal giving propagation close to the normal inside the sample, due to the high refractive index. The pump-induced probe polarisation rotation (typically $\leq 0.1^\circ$) was measured by a sum-frequency lock-in technique \cite{15}. Magnetic field, $B_{ext}$, was applied perpendicular to the incident beams, at $45^\circ$ to the normal to the quantum wells.

Data for linear pump polarisation, fig.1(a), i.e. nominally equal electron spin populations, shows a background resulting from nonidealities of the experimental arrangement. Circular pump polarisation, fig.1(b), gives Larmor beating, which decays due to electron spin-relaxation and recombination. The spin vector, initially parallel to the pump beam, precesses on a cone of half-angle $\theta \sim 45^\circ$ (see inset) and, after an odd number of half periods is perpendicular to the pump, corresponding to an equal coherent superposition of spin-states and giving zero probe polarisation rotation. After an even number of half periods the spin state is pure $\sigma^+$ or $\sigma^-$, so rotation should be a maximum absolute value \cite{13}. The observed beats are consistent with this expected behaviour, superimposed on the background of fig. 1(a). Solid curves in fig. 1(b) are numerical fits used to obtain the Larmor frequency.

A frequency shift between the Larmor beats for the two pump polarisations, is clearly apparent in fig. 1(b). The only change between the traces is that of pump polarisation, unambiguously demonstrating that the effect is due to an internal field associated with the electron spin orientation and ruling out any interpretation in terms of stray applied fields. The shift corresponds to an Overhauser field $B_N \sim 0.2$ T opposite to $B_{ext}$ for $\sigma^+$ pump and hence to
The data of fig. 1 were taken after 100 s of exposure to pump light. Repeating the experiment after longer exposures to pump light showed a slow time evolution in the Larmor frequency, corresponding to a slow increase of $B_N$ to a saturation value $B_N = 0.7$ T after about 3000 s, corresponding to $(I)/I = 0.065$ (eqn.1). Following saturation, with the beams blocked the value of $B_N$ fell by 30% over a period 600 s, giving $T_1 \gg 1.8 \times 10^3$ s. Inserting this and the known values of $t_s$ and $t_r$ into eqn. 2 and assuming that $S(t_0)/S \sim 0.8$, we find $T_1 \approx 10^4$ s. This is comparable to values calculated for hyperfine coupling to Bloch electrons [7,8], but, as discussed below, is almost certainly a value associated with polarisation by localised electron states and limited by nuclear spin diffusion.

Figure 2(a) shows Larmor frequencies for $\sigma^- $ pump polarisation as a function of $B_{ext}$ after 100 s of pumping (open circles) and with $B_N$ saturated after 4000 s of pumping for each point (filled circles). The fitted lines give $|B_N| = 0.2$ T after 100s and $|B_N| = 0.71 \pm 0.05$ T after 4000s and $g_e(\theta= 45^\circ) = -0.213 \pm 0.004$. The triangles are data for $\theta = 90^\circ$ and the dashed fit yields $|B_N| = 0.04 \pm 0.03$ T and $g_e(\theta = 90^\circ) = -0.178 \pm 0.001$. From these values we calculate [13] $g_e(\theta = 0) = -0.243 \pm 0.004$ for $B_{ext}$ along the growth direction. Calculations based on k.p theory by Ivenchenko and Kiselev [14] give $g_e(\theta = 90^\circ) = -0.166$ and $g_e(\theta = 0) = -0.248$ for 9.6 nm wide quantum wells, in close quantitative agreement with the measurements.

Figure 2(b) shows a measurement tracking the approach to saturation for $\sigma^+ $ pumping, at $B_{ext} = 4$ T, by setting the pump-probe delay to 129 ps, a steeply changing portion of the signal in fig.1(b) (The sign of the signal has been reversed with respect to fig.1 for clarity). The initial rise includes the switch-on of the signal for $B_N = 0$ and the establishment of the "instantaneous" part of $B_N$ within 10 s. As $B_N$ increases, the Larmor frequency increases, and the signal observed at a fixed pump-probe delay changes according to the gradient of the Larmor oscillations at that delay. The observed change in signal yields a time-constant for the buildup of $B_N$ of $T_B \sim 900$ s. Combining this data with the determination of $(I)/I$ after $\sim 100$ s and after saturation as described above, we can plot the form of pumping dynamics shown in fig. 2(c). The two-timescale behaviour can be assigned to effects of nuclear spin diffusion. The hyperfine interaction rapidly polarises nuclei near to electron localisation centres giving the "instantaneous" component of $(I)/I \sim 0.017$. The local nuclear polarisation then spreads into the intervening regions by diffusion, to build up additional polarisation at a slower rate. This rate, neglecting relaxation of the nuclear spins, is given by $T_B \sim D n_{loc}$ where $n_{loc}$ is the concentration of localised electronic states and D is the nuclear spin diffusion coefficient. Taking $D=10^{-13}$ cm$^2$ s$^{-1}$, measured for As in GaAs [22], and our value of $T_B = 900$ s, we obtain $n_{loc} \sim 10^{10}$ cm$^{-2}$.

Establishment of equilibrium involves in-well spin-diffusion over $\sim 100$ nm and if the diffusion coefficient in the 34 nm barriers were similar there would be significant inter-well spin transfer. We investigated diffusion across the barriers by, first, optically pumping the 19 nm quantum well in the sample for 6000 s to produce saturation of $B_N$. This required tuning the laser to the band edge for
the 19 nm well, a photon energy not absorbed in, so not directly pumping, the 9.6 nm well. The laser beams were then blocked, retuned to the band edge of the 9.6 nm well and unblocked to make a rapid measurement of $B_N$ for that well; this gave only the 'instantaneous' value. Therefore little, if any, nuclear polarisation had diffused through the 34 nm AlGaAs barrier during prolonged pumping of the 19 nm well. An upper limit for spin-diffusion coefficient in the barrier is therefore $1 \times 10^{-14} \, \text{cm}^2\text{s}^{-1}$ (assuming the dark relaxation time, $T_1^*$ is not significantly lower in the barriers). This finding complements those from optically pumped NMR [10], which show $^{71}\text{Ga}$ signal build-up from nuclei in the barrier over $\sim 200$ s, due to (limited) spin-diffusion into the barriers. Spin diffusion occurs mainly via mutual spin flips of like isotopes which conserve the Zeeman energy of the spin system. Therefore inter-well diffusion in GaAs/AlGaAs should, indeed, be much slower than diffusion within the wells because $^{27}\text{Al}$ ($I=5/2$) gives magnetic spin disorder in the Ga sublattice and also introduces disorder in the As sublattice due to random quadrupole splittings.

In conclusion our results show the importance of nuclear spins in time-resolved spectroscopy of III-V semiconductors. Among possible applications we find that it is essential to take account of such polarisation in measurements of electron g-factors and that new insights are obtained into nuclear spin diffusion in quantum structures. The time-resolved reflection technique allows investigations in situations inaccessible to traditional cw measurements [7, 8] where luminescence may be short-lived compared to spin memory. In our GaAs/AlGaAs sample, only 6.5% nuclear spin polarisation is generated in the quantum well, causing an effective internal field approaching 1 T, limited by the comparatively short spin relaxation time of the electrons. Much greater nuclear polarisations, beyond this essentially linear (low-polarisation regime) should occur in weakly n-type GaAs under strong pumping conditions, because of the long spin relaxation times [4] and donors which will enhance electron-nuclear spin transfer. We note that it is just this type of material which is under consideration for future applications in spin-devices.

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Figure 1: Representative results for pump-induced probe polarisation rotation at 4 T in 9.6 nm single GaAs/AlGaAs quantum well containing $0.5 \times 10^{11}$ cm$^{-2}$ heavy holes at 10 K. (a) Signal for linearly polarised pump shows background associated with nonideality of experimental setup; (b) signals for circularly polarised pump, showing Larmor beats and frequency shift due to Overhauser field following about 100 s of exposure to pump light. Inset shows principle of optical pumping of nuclear spin polarisation $I$ and associated Overhauser field $B_N$ in a quantum well (QW).
Figure 2: (a) Larmor frequencies observed for $\sigma-$ polarised pump. Open circles and filled circles are for $\theta=45^\circ$ with pumping times 100 s and 4000 s respectively. Triangles are for $\theta=90^\circ$ and pumping time 100 s. The solid and dashed lines are fits giving $B_N$ and $g_\epsilon(\theta)$ (see text). (b) Time-dependence of Larmor beat signal in fig. 1 at 129 ps delay, midway between turning points, illustrating the approach of the Overhauser field to a saturation value with a time-constant $T_B \sim 900$ s. (c) Inferred time-evolution of nuclear spin polarisation.