Ultrafast Manipulation of Electron Spin Coherence

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A technique is developed with the potential for coherent all-optical control over electron spins in semiconductors on femtosecond time scales. The experiments show that optical “tipping” pulses can enact substantial rotations of electron spins through a mechanism dependent on the optical Stark effect. These rotations were measured as changes in the amplitude of spin precession after optical excitation in a transverse magnetic field and approach π/2 radians. A prototype sequence of two tipping pulses indicates that the rotation is reversible, a result that establishes the coherent nature of the tipping process.

Multiple pulse sequences in time-domain nuclear magnetic resonance and electron spin resonance (ESR) experiments are widely used to study spin-spin interactions and spin dephasing in inhomogeneous magnetic environments (1). A canonical sequence consists of a π/2 pulse to generate a nonequilibrium transverse spin polarization followed by a π-pulse that may enact a rephasing (“spin echo”) of transverse spin if inhomogeneous broadening dominates the ensemble spin dynamics (2). Current technology limits the number of systems to which pulsed-ESR experiments can be applied because the minimum achievable pulse length of ~10 ns should be much smaller than the spin coherence time (1). To apply pulse sequences to study conduction-band electron spin dynamics in a variety of semiconductors where spin lifetimes can vary from ~3 ps (3) to ~130 ns (4), a complementary ESR technique capable of much shorter pulse widths is desirable. Application of spin echo sequences might be limited by the optical Stark effect (5). Such a technique could also be useful for spin-based implementations of quantum computing in solid-state systems (6), where it is necessary to perform many operations (>10^10) on a quantum bit within the coherence time to realize full computation with error correction (7). The ability to perform spin operations on femtosecond time scales would satisfy this need in a number of semiconductor systems and may be applicable to single quantum bits using optical probes with high spatial resolution.

Here we present time-resolved Faraday rotation experiments that extend a technique first applied in atomic sodium (8) to semiconductor nanostructures by using ultrafast laser pulses to produce coherent rotations of electron spins (9). In our experiments, a pump pulse optically excites spin-polarized electrons that precess about a static magnetic field. A second below-band gap “tipping” pulse produces an additional effective magnetic field that can reach 20 T through the optical Stark effect (10). Effective field strengths are calculated from measurements of Stark shifts and depend on the pulse intensity, polarization, and energy. This field is used to coherently rotate electron spins by angles that approach π/2, as monitored through the Faraday rotation imparted to a probe pulse. A sequence of two tipping pulses suggests that spin coherence is preserved during the tipping process. Because the tipping pulses additionally excite a small number of real carriers, a variety of checks were performed to identify resultant background contributions. Although the tipping effects can be qualitatively interpreted as rotations about a light-induced effective field, quantitative comparisons of the tipping angle from Faraday rotation data with that expected from measured Stark shifts reveal a significant discrepancy.

Samples were chosen that illustrate a variety
of electronic and magnetic environments in which this technique can be applied. The samples consist of 10 150 Å–wide Zn$_{1-x}$Cd$_x$Se ($x \sim 0.33$) quantum wells (QWs) fabricated with an all-digital approach that uses a short-period ZnSe/CdSe superlattice as the confining region (11). In addition to modulation-doped ($n = 4.1 \times 10^{11}$ cm$^{-2}$) and undoped QW samples, a magnetic QW was studied in which submonolayer MnSe planes were inserted into the confining region that increase the effective electron $g$ factor (3). Synchronized optical parametric amplifiers (250-kHz repetition rate) produce two independently tunable (2.6 to 1.7 eV) pump pulses and probe pulses of a white light continuum (each ~150 fs in duration) that were focused to a ~100-μm-diameter spot on the sample. The substrate was removed from a circular region of the sample, permitting measurements in transmission on free-standing epilayers.

First studied in atoms (12), the optical Stark effect in semiconductors has been explained as a repulsive interaction between real and virtual excitons that are created by probe and below–band gap pump photons, respectively (13). The end result is a shift $\Delta E$, of the absorption spectrum toward higher energy that lasts for the duration of the pump-probe correlation time (typically 200 to 500 fs). $\Delta E$ is proportional to the pump intensity and inversely proportional to the pump detuning (13), in our case given by $\Delta = E_p - E_{\text{hh}} - E_p'$, where $E_{\text{hh}}$ and $E_p$ are the heavy-hole exciton and pump energies. Stark shifts in these QWs follow optical selection rules that govern the coupling of pump photons to heavy-hole (hh) and light-hole (lh) exciton transitions, visible as peaks in the linear absorption spectra (Fig. 1A). Illustrated in Fig. 1B, the solid lines show the conduction and valence band levels shifted from their unperturbed values (dotted lines) in the presence of a below–band gap, $\sigma^+$–polarized pump pulse (14). When tuned to the hh excitation energy, co- or counterpolarized probe pulses (blue or red arrows in Fig. 1B) measure shifts of $\sigma^+$ and $\sigma^-$ hh excitons. The $\sigma^+$ hh exciton shift reflects coupling of pump photons to the $\sigma^+$ hh exciton, whereas the $\sigma^-$ hh exciton shift arises indirectly through the shift of the $\langle +1/2 \rangle$ conduction band level produced by coupling of the pump to the $\sigma^+$ hh exciton. Because of both optical selection rules and larger pump detuning from the lh exciton energy, the $\sigma^-$ lh exciton shift is smaller (14).

Shifts were measured as pump-induced changes in the absorption spectrum given by $\Delta \alpha L \equiv \left( \alpha_0 - \alpha_L \right)$, where $\alpha_0$ is the absorption coefficient without the pump, $\alpha$ is the coefficient with the pump as measured by a time-delayed probe, and $L$ is the sample thickness. In these initial two-beam experiments, the pump and probe were both circularly polarized with Soleil-Babinet compensators, and the probe was spectrally resolved and detected at an energy $E_p$ (15). Absolute values of the Stark shift were calculated from the relation $\Delta E = (\Delta \alpha L) (d\alpha/dE)^{-1}$ (15).

Figure 1C shows the pump-intensity dependence of Stark shifts for co- and counterpolarized probe pulses ($\Delta E_{\text{co}}$ and $\Delta E_{\text{counter}}$) taken at two values of detuning for the undoped QW sample. There is a departure from linearity at higher intensity that is consistent with previous studies (15), in addition to the expected increase in shift as the pump detuning is reduced. The data are indicative of a net conduction-band spin splitting given by $\delta_E = 1/2 \Delta E_{\text{co}} - \Delta E_{\text{counter}}$ (14) that may be described as an effective magnetic field along the laser direction, $H_{\text{Stark}} = \delta E (g_e \mu_B)^{-1}$, where $g_e$ = 1.1 is the electron $g$ factor in this sample. This field is plotted versus intensity in Fig. 1D where a plateau value of ~19 T ($\delta_E = 1.2$ meV) with $\Delta = 64$ meV results from a change in the relative intensity dependence of $\Delta E_{\text{co}}$ and $\Delta E_{\text{counter}}$ at high intensity. The ratio $\Delta E_{\text{co}}/\Delta E_{\text{counter}}$ can be compared with the theoretical expectation of ~6.5 calculated with measured values of hh and lh exciton energies (14). Data at higher detuning and low intensity agree with this value; differences with lower detuning and higher intensity may indicate a transition to a non-perturbative regime for which the theory is inappropriate.

In order to clearly observe the Stark shift, the pump energy must be adjusted to minimize the excitation of real carriers. Figure 1E shows that when the pump energy is higher than the hh exciton ($\Delta < 0$), real-carrier generation results in state-filling effects that...
bleach the QW absorption, visible as a red “streak” that persists for the carrier recombination time (∼100 ps, not shown in figure). When the pump energy is tuned below the band gap (Δ > 0), the generation of real carriers is energetically unfavorable, and a ∼250-fs-wide feature appears in Fig. 1F that corresponds to a 5.1-meV shift. The lobed spectral shape reflects the change in sign of dα/du that occurs when the detection energy is scanned across the hh exciton peak. Tipping pump energies in the following experiments were chosen that represented a compromise between state-filling effects and magnitude of the Stark shift.

In time-resolved Faraday rotation experiments (Fig. 2A), a circularly polarized pump pulse (with Δ = 0) excites spin-polarized electrons and holes along x̂, perpendicular to an applied magnetic field HV. Previous studies indicated that hole spins are either pinned along the QW growth direction or dephase rapidly (3). As a result, the Faraday rotation, θP, of linearly polarized probe pulses is proportional to S(t), the projection of electron spin S ̃ along x̂. Because the electron spins comprise a coherent superposition of eigenstates quantized by the field, θP oscillates as a function of probe delay at the Larmor spin precession frequency vL = gμBB. A circularly polarized tipping pump (TP) pulse (usually with Δ > 0) was incident on the sample after the pump. The relative time delays of the three pulses incident on the sample could be independently adjusted with ∼10-fs resolution, and data were collected by scanning either the probe or tipping pump delay. The three optical beams were modulated at different kHz-scale frequencies, and lock-in amplifiers were used to record changes in the total Faraday rotation induced by the pump and tipping pump pulses. This scheme was necessary for separately identifying signals initiated by the pump and TP alone, as well as signals due to their interaction. Data shown here were taken with reference to the pump modulation frequency as an added correction.

Figure 2B depicts the rotation of spin polarization away from the x̂-γ plane caused by the TP. The torque exerted by the TP is given by τ = gμBB × HStark and is largest when S ̃(t) is perpendicular to HStark, a situation that occurs whenever θP(t) crosses zero. Spin precession after the tipping event describes a cone about HStark, because θP only measures S ̃, the amplitude is reduced. This is realized in Fig. 2C, where spin precession in the undoped QW is shown with the TP position indicated by the arrow. A priori, the tipping angle θTP can be simply calculated from the relation θTP = Arccos(A ̂ 0), where A and A ̂ 0 are the reduced and unperturbed amplitudes as shown in Fig. 2C.

However, the undesirable excitation of a small number of real carriers by the TP (∼100 times fewer than the pump) complicates this analysis. Some real-carrier excitation was observed for the entire range of detuning studied (up to Δ = 120 meV) and appears as a faint white streak in Fig. 1F. No net improvement was observed by increasing Δ, because both tipping effects and real-carrier generation were comparably reduced. Although energetically unfavorable, absorption of TP photons may occur because of low-energy states in the QW (perhaps related to impurity sites) (16) or phonon-assisted absorption (17); two-photon processes are unlikely because of the preservation of spin polarization and the dependence on detuning (not shown in figure). Because the lock-in detection scheme is “bidirectional,” the signal detected at the pump modulation frequency can reflect both (i) TP-induced changes in θP from electron spins excited by the pump (the desired signal) and (ii) pump-induced changes in θP from electron spin excited by the TP (due to undesired real-carrier generation). The latter are detected because the TP excites fewer carriers when preceded by the pump than if the pump were absent, a change that is picked up in lock-in measurements at the pump modulation frequency as an added background. This background contribution to the measured signal does not reflect a change in the dynamics of the pump-excited spins but is merely superimposed on the desired tipping results. These state-filling effects are strongest when the spins excited by the pump and TP are parallel but generally exist to some degree at any relative polarization.

To study the consequences of these effects, Fig. 2D shows the signal ΔθP = θP − θTP corresponding to the difference between red and black curves in Fig. 2C with the TP (polarized so HStark || x̂) positioned at three different points in the spin precession cycle. As expected, the difference signal is largest when the TP is positioned where S ̃ is perpendicular to HStark (red circles in Fig. 2D). State-filling effects result in the nonzero difference signal when S ̃ is parallel to HStark (blue squares) and are largely absent when S ̃ is antiparallel to HStark (black squares). To identify the optical Stark effect as the underlying mechanism for spin rotation, we did the same comparison with the TP energy chosen to excite an additional spin population S ̃ || x̂ directly (Δ < 0). State-filling effects are dominant for this situation, so that ΔθP is expected to be largest when S (t) is parallel to S ̃. Figure 2E shows that the magnitude of ΔθP becomes progressively larger as the TP is moved from a position where S ̃ and S ̃ are antiparallel to a position where S and S ̃ are parallel. The phase differences present in the oscillations of ΔθP reflect the initial time and polarization of spins excited by the TP relative to those previously excited by the pump.

Figure 3 summarizes an effort for the magnetic QW to subtract real-carrier background contributions from the calculated value of θTP by studying the dependence of the tipping angle on TP intensity. Spin precession in this sample is indicative of an enhanced effective g factor (ge ~ 20) and decreased spin lifetime characteristic of QWs with a large exchange interaction between electron spins and paramagnetic Mn2+ ions (3). The reduction in amplitude caused by the TP (HStark || x̂, Δ = 46 meV) increases with TP intensity (Fig. 3A), eventually causing a reversal in sign (heavy solid curve) that in the absence of state-filling effects would indicate a tipping angle greater than π/2. To quantify contributions from state-filling effects, we...
took data by fixing the probe time delay at a peak in the spin precession curve [dotted line in Fig. 3, B and C, with $\hat{S}(10\text{ ps}) = \pi$] and monitoring the change in amplitude as a function of TP time delay, $\Delta_{\text{tip}}$. Figure 3B shows spin precession data taken in the absence of the TP, included as a reference. Peaks in the signal detected at 10 ps (Fig. 3C) occur when $\hat{S}(\Delta_{\text{tip}}) \perp \hat{H}_{\text{Stark}}$ (for example, dotted line ii). Consistent with Fig. 2, D and E, the amplitude does not return to its unperturbed value $A_i$ when $\hat{S}(\Delta_{\text{tip}}) \parallel \hat{H}_{\text{Stark}}$ (dotted line iii) because of state-filling effects. These situations appear as valleys in Fig. 3C that are closer to $A_i$ when $\hat{S}(\Delta_{\text{tip}}) \perp \hat{H}_{\text{Stark}}$ are antiparallel than when they are parallel. The valley at $\Delta_{\text{TP}} = 10$ ps is associated with the Stark shift itself and marks the point where probe and TP overlap in time.

To calculate $\Theta_{\text{tip}}$, we recorded the extrema in Fig. 3C, yielding values of $A_{\text{RC}}$ and $A_i$, where $A_{\text{RC}}$ is a measure of the background contribution due to real-carrier effects. Because only real-carrier effects are present at the valleys in Fig. 3C, we subtract this contribution to $\Theta_{\text{tip}}$ using the relation $\Theta_{\text{tip}} = \text{ArcCos}(\langle \hat{S} - (A_{\text{RC}} - A_i) \hat{A}_i \rangle) / 2$ (18). Figure 3D compares the results from this calculation averaged over all peaks and valleys from $0 < \Delta_{\text{TP}} < 9$ ps in Fig. 3C with values for $\Theta_{\text{tip}}$ expected from Stark shift data:

$$\Theta_{\text{Stark}} = \frac{1}{\hbar} \int_{-\infty}^{+\infty} \delta_{\text{CB}}(t) \, dt,$$

where $\hbar$ is Planck’s constant divided by $2\pi$. Here the conduction-band split splitting is integrated over the temporal profile of the TP (Gaussian with a width of $\sim 400$ fs). Although the two plots have a similar functional dependence on intensity, there is a significant discrepancy in magnitude. Earlier studies of hole spin dynamics have shown that in-plane magnetic fields can modify optical selection rules because of valence-band mixing (9, 19). Although we do not observe much change in the Stark shift with magnetic field, it may be that the relative valence and conduction-band contributions to the shift change with field, effectively causing an increase in $\delta_{\text{CB}}$. It is perhaps more likely that the simple calculations presented here are inadequate for describing the highly nonadiabatic, impulsive effects of these belowband gap tipping pulses.

In addition to possible systematic errors not accounted for in the subtraction routine above, the difference between expected and measured tipping angles could be due to an incoherent reduction in amplitude caused by the tipping process itself. We investigated this possibility by further extending the technique to include two tipping pumps (TP1 and TP2), to establish the reversibility and coherent nature of the tipping process (20). To begin with, TP1 arrives at a zero crossing in $\Theta_{\text{tip}}$ (co- and counterpolarized $\hat{H}_{\text{TP}}$, causing spins to be tipped away from the x-y plane and resulting in a reduction of amplitude as before (Fig. 4B)). The spins then are allowed to precess a half-cycle before they are tipped again by TP2, resulting in a second torque of opposite sign for co- and counterpolarized TPs (Fig. 4C) and the same sign for counterpolarized TPs (Fig. 4A). The data in Fig. 4D were taken with equal-intensity TPs and show a reversal or enhancement of the net tipping angle as the TP2 helicity is varied, indicating that spin coherence is largely preserved during the tipping process. Data were taken with various combinations of relative circular and linear polarizations to verify that the dependence on TP2 helicity existed only when preceded by a circularly polarized TP1. Figure 4E shows one such check, where data taken without TP1 show a similar amplitude reduction for the two TP2 helicities ($\Theta_{\text{tip}} < \pi$). We expect that spin echoes could occur for imperfect tipping pulses if $\pi/2 < \Theta_{\text{tip}} < \pi$. Because the echo amplitude should sinusoidally increase as the tipping angle approaches $\pi$, the current degree of rotation is just at the limit where echoes may start to become observable. However, an additional requirement for echo observation is that inhomogeneous dephasing in the sample limits the spin lifetime. Although this condition was not clearly met in the present samples, model systems may be realized by specifically engineering inhomogeneous dephasing into magnetic heterostructures based on the one studied here. In conclusion, these experiments demonstrate a principle by which $\pi$-pulses may be constructed to coherently control spins in semiconductors on femtosecond time scales.

Fig. 4. (A to C) Illustration of the net tipping angle with two co- and counterpolarized tipping pumps. Parameters for the tipping pumps in (D and E): $\Delta = 51$ meV, $t_{\text{TP}} = 0.2$ GW cm$^{-2}$. (D) Data taken with tipping pumps positioned at consecutive zero crossings, the situations in (A) (red) and (C) (blue). (E) The dependence of the net tipping angle on TP2 helicity vanishes in the absence of TP1.