Pump–probe Faraday rotation magnetometer using two diode lasers

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A time-resolved Faraday rotation magnetometer using externally triggered pulsed diode lasers is described. This device permits measurement of the dynamic properties of polarized electronic spins in semiconductors. A nonequilibrium spin polarization is created in the conduction band electrons of *n*-type GaAs using a circularly polarized laser pulse generated by a pulsed laser diode. A subsequent linearly polarized pulse from a second laser diode probes the time evolved electronic polarization via the Faraday effect. Since two different laser diodes are used for the pump and probe process, the dynamics of optically pumped spins can be directly observed at arbitrarily long pump–probe delays with a temporal resolution of 75 ps and a spatial resolution of 25 μm. The signal-to-noise of the laser diodes is sufficient to achieve a sensitivity on the order of 3000 spins. [DOI: 10.1063/1.1912688]

Time-resolved Faraday rotation (TRFR) is a powerful technique for studying the dynamics of spin-polarized electrons in the conduction band of semiconductors.[1,2] With this technique, two laser pulses are used to, respectively, pump and probe electronic spins in a material. First, a circularly polarized pump pulse generates a nonequilibrium population of spin-polarized carriers in the conduction band of the sample. Subsequently, a linearly polarized probe pulse tuned to the absorption band edge is transmitted through the sample. Due to the circular birefringence induced by the spin-polarized conduction band, the polarization axis of the probe pulse rotates in proportion to the magnitude and direction of electronic spin polarization.

Most TRFR magnetometers rely upon mode-locked Ti:sapphire lasers to provide the pump and probe pulses.[3] Recently, Bauer *et al.* developed a time-resolved magnetometer for studying ferromagnetic materials that used a single pulsed diode laser synchronized with magnetic field pulses.[4] We have extended this concept and developed a TRFR magnetometer that uses two externally triggered diode lasers to study magnetization dynamics in paramagnetic materials. Our technique has the advantages of reduced cost, smaller size, and improved flexibility.

We examined spin dynamics in bulk *n*-type GaAs [2×10^{16} Si/cm^3, (100) surface]. Similar materials have been extensively studied with TRFR and other optical orientation methods,[5,6] making it an excellent reference material to assess instrument performance. The 1 cm$\times$1 cm sample was bonded to a glass substrate with ultraviolet-cured epoxy and mechanically polished to a thickness of $\approx$75 μm.

The sample was mounted in a continuous flow liquid helium cryostat that cooled the sample down to 4.3 K (see Fig. 1). An external magnetic field was applied in the sample plane along the $\hat{x}$ direction using a conventional air-cooled *C*-frame electromagnet. The field inhomogeneity was less than 0.5% over the entire area of the sample and essentially zero over the optically sampled area. The applied field range was ±0.3 T. Due to the relative positions of the gaussmeter and sample, there is a 0.5% uncertainty in the absolute value of the measured field.

TRFR measurements were performed as follows: Two externally triggered pulsed diode lasers provided the pump and probe pulses, as shown diagrammatically in Fig. 1. The lasers were produced by Advanced Photonic Systems, models PiL 080G-TE (pump) and PiL 082Q (probe).[7] These lasers are not spectrally tunable; they were specifically chosen to operate near the band gap of GaAs at cryogenic temperatures. As specified by the manufacturer, the pump and probe pulses have minimum temporal widths (full width at half maximum) of 36 and 15 ps; however, the pulse width varies with output power. The spectral widths of the pump and probe lasers are 2.5 nm (4.6 meV) and 4 nm (7.7 meV), respectively. A digital pulse-delay generator separately triggered each laser so that the pulses were temporally separated by a period t. Trigger rates up to 1 MHz were possible; single pulses could also be applied. A 500 kHz trigger rate was employed for the measurements presented here. The trigger jitter for each diode laser was 6 ps; the trigger jitter for the digital pulse-delay generator was 70 ps. Pump pulses with a photon energy of 1.55 eV (800 nm), chosen to be slightly above the absorption band edge of the sample, were triggered at $t=0$. Each pump pulse contained 16 pJ of energy. The pump beam was circularly polarized using a linear polarizer in combination with a Soleil–Babinet compensator with its retardance adjusted to $\lambda/4$. The beam was then focused onto the surface of the sample with a spot size of $\approx$5000 μm^2 and an angle of incidence $\approx$10° from the surface normal. The two-dimensional intensity profile of the focused pump spot (which was very non-Gaussian) was measured by positioning a charge-coupled device (CCD) array in the focal plane of the optics. The linearly polarized probe beam, triggered at time t, had a photon energy of 1.51 eV (822 nm) and a total energy of 0.45 pJ/pulse. The probe was focused on an oblong spot with an area of $\approx$700 μm^2, as

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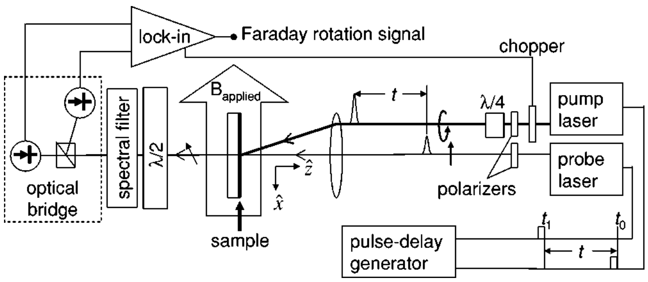

FIG. 1. Diagram of the time-resolved Faraday rotation magnetometer.

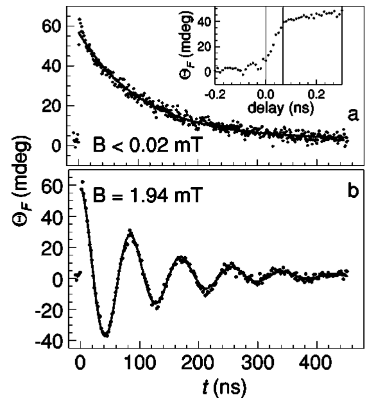

FIG. 2. A pair of TRFR scans of bulk GaAs with varying applied magnetic fields. The solid lines are fits of Eq. (3) to the data: (a) $B=0$ mT, $T_2^*=107$ ns. (a, inset) $B=0$ mT. A closeup illustrating the risetime of the Faraday rotation. To optimize the laser pulse width, a weaker pump pulse was used for the inset; hence, it has a smaller Θ_F than is seen in the primary figure; (b) $B=1.94$ mT, $T_2^*=110$ ns, $g=0.43$.

measured with the CCD array. The average fluence f of the pump beam at the position of the probe spot was $40\ \text{nJ cm}^{-2}/\text{pulse}$. The probe beam was normal to the sample.

The probe pulses were detected with an opto-electronic bridge to measure the Faraday rotation angle Θ_F. The detector consisted of a Glan–Foucault polarizing beam splitter and a pair of cooled, amplified, Si photodiodes (detector size $1\ \text{cm}\times 1\ \text{cm}$, 130 Hz bandwidth). Using these large area photodiodes greatly simplifies the alignment of the probe beam. The difference between the outputs of the two diodes was detected with a lock-in amplifier synchronized to an optical chopper (20 Hz) set in the pump beam path. A rotatable $\lambda/2$-plate immediately in front of the detector was used to rotate the polarization of the probe beam to balance the signal between the two diodes. Since GaAs has an absorption depth on the order of 1 μm for energies above the band gap,[8] the sample was sufficiently thick to act as a spectral filter to prevent pump beam light from reaching the detector. A low-pass spectral filter was placed in front of the detector to reduce fluorescence from the sample and remove any scattered pump light. The complete system had a polarization rotation sensitivity of approximately 10^{-3} deg (17 μrad).

Figure 2 presents the results of a series of measurements of Θ_F versus t performed with the magnetic field nominally zero ($|B|<0.02$ mT) and $B=1.94$ mT. In Fig. 2(a), when $t<0$, the probe pulse arrived before the pump; with no pumped spins in the sample, Θ_F was nominally zero. At $t=0$, Θ_F suddenly rose and thereafter exponentially decayed with a time constant given by the spin dephasing time T_2^*

$$\Theta_F(t)=\Theta_0 e^{-t/T_2^*}=Q_F N e^{-t/T_2^*}. \tag{1}$$

Here, N is the areal density of polarized spins in the sampled area, and Q_F is the Faraday rotation per unit spin density. Fitting the data from Fig. 2(a) to Eq. (1) yields $T_2^*=107\pm 2$ ns and $\Theta_0=53.7\pm 0.4$ mdeg. (Unless otherwise noted, all uncertainties are given in terms of standard uncertainty, i.e., the square root of the sum of the squares of the individual deviations.) The relatively long period (2 μs) between the arrival of sequential pump pulses prevented spins from earlier pump pulses affecting the measurement.

Reflectivity measurements show that 70% of the pump pulse was absorbed by the sample; the other 30% was reflected off of the sample. Selection rules for GaAs dictate that the spin polarization efficiency ε be equal to 50%.[9] We can now estimate an upper bound for the spin density in the area sampled by the probe beam:

$$N=\frac{(f\times 0.70)\varepsilon}{\hbar\omega}=5.6\times 10^{10}\ \text{cm}^{-2}. \tag{2}$$

Thus, $Q_F=\Theta_F/N=10^{-12}$ deg cm^2. This is only a lower bound for the strength of the Faraday effect, understanding that there is some uncertainty in the area of the pump pulse spot, estimated to be approximately 20% of the measured spot size. This uncertainty results from the difficulty in superimposing the pump and probe beams. We also note that Q_F is a strong function of the probe wavelength and the size of the band gap, thereby implying that the measured sensitivity is specifically for the particular probe beam parameters described above. We can estimate the absolute number of spins detected: $N\times$ probe area $\approx 4\times 10^5$. From the angular rotation sensitivity given above, this implies that the minimum number of detectable spins $(0.4/53.7)\times 4\times 10^5\approx 3000$. Such absolute sensitivity to the spin polarization could be even further enhanced by using a smaller probe pulse diameter, as the net Faraday rotation is not affected by the probe pulse intensity. Reduction of the probe pulse size to the diffraction limit of $\approx 1\ \mu\text{m}^2$ might improve the sensitivity by another factor of 100–700.

If a magnetic field was present [as in Fig. 2(b)], the spins immediately precessed in the y–z plane upon the arrival of the pump pulse, with a rate given by the Larmor frequency $\omega_L=|g|\mu_B B/\hbar=\gamma B$, where g is the Landé g-factor, μ_B is the Bohr magneton, and γ is the gyromagnetic ratio. Hence, the spin polarization along the probe direction oscillated as it dephased. The observed Faraday rotation is well described by

$$\Theta_F(B,t)=Q_F N\cos(\gamma B t)e^{-t/T_2^*}. \tag{3}$$

For the data shown in Fig. 2(b), $B=1.94$ mT. Fitting the data to Eq. (3) yields an amplitude $Q_F N=56.7\pm 0.7$ mdeg, $T_2^*=110\pm 2$ ns and $|g|=0.429\pm 0.002$. (Since the instrument is sensitive only to the magnetization along the direction of the probe beam, it is insensitive to the sign of g.) This is in good agreement with a previously published value of $g=-0.44\pm 0.02$.[10] When measured over a wide range of applied fields from 0 to 100 mT, the fitted values for T_2^* and $Q_F N$ varied by roughly 10%; this is possibly due to longterm noise in the laser output or drift in the position of the focused laser beams. However, the g-factor is remarkably consistent over this range. While the absolute value of the g-factor is limited by uncertainty in the value of the applied field due to the positioning of the sample, this uncertainty is a constant percentage for all field values. $|g|$ is invariant with applied field within 0.1%.

The most important advantage of this system over earlier TRFR magnetometers is financial, with the cost of each pulse diode laser over 1 order of magnitude less than a Ti:sapphire laser. However, this system also provides several inherent technical advantages. Most previous magnetometers have used a single Ti:sapphire laser to provide both the pump and probe beams (although a few systems have been developed that use two Ti:sapphire lasers,[11] albeit at significantly increased expense). Using different photon energies for the pump and probe pulses permits spectroscopic removal of the pump beam from the detector, greatly simplifying probe detection. With Ti:sapphire systems, probe delays are typically performed with mechanical optical delay stages. While these devices do permit femtosecond resolution, employing them for delays of more than a few nanoseconds is impractical due to the large physical length of the delay line. Use of a digital pulse-delay generator permits time-resolved measurements over a wide range of pump–probe delays from ≈ 50 ps to several minutes. The third advantage is that external triggering enables synchronization of the optical pulses with electronic signals without the complication of slaving the external systems to the laser. For Ti:sapphire systems, there exist technological solutions for each of these problems, but they are technically or financially expensive to implement.

The principal shortcoming of the pulsed diode laser system is the inability to adjust the wavelength of the lasers. While the pump energy may differ from the absorption band edge by 0.1 eV or more and still efficiently pump spins, the absorption band edge of the sample must be within the bandwidth of the probe beam (≈ 0.01 eV in our case) to observe a detectable Faraday rotation. Some control of the band gap is possible by controlling the temperature of the sample. The absorption band edge in our sample increased ≈ 0.025 eV from 4 to 120 K.

Other shortcomings are the limited optical power and the temporal resolution of the system. The maximum average power for each laser is ≈ 10 μW, and the maximum peak powers are 450 and 750 mW for the pump and probe lasers, respectively. If the output power is adjusted to minimize the pulse width of the lasers, the temporal resolution is limited by the following: the width of each pulse (≈ 8 ps and ≈ 18 ps rms), the jitter on each laser (6 ps rms), and by the jitter in the digital pulse-delay generator (70 ps rms). (These are quoted nominal uncertainties for each of these components.) Hence, the net rms temporal uncertainty is 73 ps. The measured rise time (time to change from 0.1 to 0.9 of the final value) of the Faraday rotation signal is 75 ps, indicating that this process is limited by the temporal resolution of the magnetometer [see the inset of Fig. 2(a)]. Thus, the approximate 3 dB bandwidth of the magnetometer is 0.35/75 ps $=5$ GHz.[12] For GaAs, this limits the range of applied field to $|B| < 0.8$ T.

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