Measurement of Electron Spin Lifetime and Optical Orientation Efficiency in Germanium Using Electrical Detection of Radio Frequency Modulated Spin Polarization

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(Received 11 May 2011; revised manuscript received 10 August 2011; published 12 October 2011)

We propose and demonstrate a technique for electrical detection of polarized spins in semiconductors in zero applied magnetic fields. Spin polarization is generated by optical injection using circularly polarized light which is modulated rapidly using an electro-optic cell. The modulated spin polarization generates a weak time-varying magnetic field which is detected by a sensitive radio-frequency coil. Using a calibrated pickup coil and amplification electronics, clear signals were obtained for bulk GaAs and Ge samples from which an optical spin orientation efficiency of 4.8% could be determined for Ge at 1342 nm excitation wavelength. In the presence of a small external magnetic field, the signal decayed according to the Hanle effect, from which a spin lifetime of 4.6 ± 1.0 ns for electrons in bulk Ge at 127 K was extracted.

DOI: 10.1103/PhysRevLett.107.166603 PACS numbers: 72.25.Dc, 78.20.Ls, 78.40.Fy, 85.75.−d

Optical spin orientation is a method of exciting spin-polarized electrons and holes in semiconductors such as GaAs by absorption of circularly polarized light [1]. Optical detection techniques such as photoluminescence and Faraday or Kerr rotation have also played a key role in the experimental investigation of spin dynamics in semiconductors [2]. However these techniques have been mostly applied to direct band-gap semiconductors such as GaAs [3], CdTe [4], and ZnSe [5] which have large optical absorption coefficients. Indirect gap semiconductors such as Si and Ge are unique since, being crystals with inversion symmetry, the D'yakonov-Perel spin relaxation mechanism is absent [6]. In addition the dominant ($> 90\%$) isotope does not possess nuclear spin [7] thus reducing the hyperfine interaction. Therefore the effective spin relaxation mechanism is the Elliot-Yafet mechanism [6] and the spin lifetimes are expected [2] to be long (1–10 ns). However it is difficult to use optical techniques for spin injection in these materials [8]. Optical orientation in combination with electrical detection technique has been recently reported [9] for Si. More recently [10,11] an all-optical spin polarization signal was detected in Ge although the intensity was about 2 or 3 orders of magnitude lower than that of III-V semiconductors. The absolute optical orientation efficiency could not be determined in these experiments. Optical injection in AlGaAs combined with spin transport in Ge at room temperature has also been reported [12]. The motivation of this work is to develop a spin detection technique that can be used for indirect band-gap semiconductors without application of external magnetic fields. It is based on sensing the magnetic fields of the polarized spins directly by using a sensitive radio-frequency (rf) coil. We first validate this technique using GaAs in which optical spin injection and detection has been well studied. We later apply it to detect polarized spins in Ge and determine the optical orientation efficiency. Using a weak external magnetic field, we observe the Hanle effect from which the spin lifetime was determined.

The experimental setup is shown in Fig. 1 (inset). The excitation laser ($\lambda = 850$ nm diode laser module for GaAs and $\lambda = 1342$ nm DPSS module for Ge) with built-in collimation optics was first linearly polarized using a Glan-Taylor Calcite polarizer with 10^5:1 extinction ratio. A zero-order quartz half-wave plate was then used to rotate the plane of polarization of the light with respect to the axis of the electro-optic modulator (EOM). The EOM consists of a lithium tantalate crystal with a half-wave voltage of 183 V and 100:1 extinction ratio at 1064 nm. It is driven by a function generator through a high voltage amplifier with bandwidth 8 MHz. The dc bias and rf high voltage drive to the EOM were adjusted to achieve the desired degree of polarization modulation. The ~0.4 mm thick and ~2 mm diameter sample of bulk GaAs or Ge was placed at the

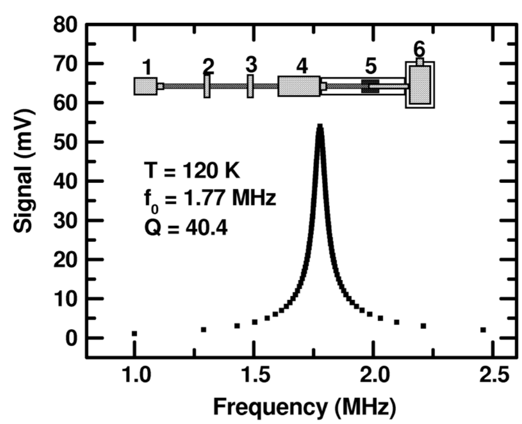

FIG. 1. Frequency response of rf pickup coil as measured by the rf lock-in amplifier, when excited by a current carrying coil in place of the sample. Inset shows experimental setup: (1) laser, (2) linear polarizer, (3) half-wave plate, (4) EOM, (5) rf coil with sample inside, (6) cryostat.

center of the pickup coil and the light beam of $\sim$1 mm diameter was incident almost normally to the (100) sample surface. The sample was mounted on a thermally conducting, but electrically insulating cold finger of a liquid nitrogen cryostat for low temperature measurements. The entire space between the EOM output window and sample stage is evacuated to avoid an additional window in the beam path. The pickup coil consisted of 300 turns of 4-stranded 110 μm diameter Litz wire with a winding length of 2.2 mm, inner radius 1.8 mm and outer radius 3.4 mm. It was connected through a coaxial feedthrough to an rf lock-in amplifier with input impedance of 1 MΩ whose frequency was tuned to the resonance of the pickup coil as shown in Fig. 1. The resonance frequency was typically in the 1–2 MHz range and shifted slightly with temperature. The Q factors were about 30–50, depending on the pickup coil dimensions, number of turns and temperature. Rare-earth permanent magnets were used to apply external magnetic fields.

In order to validate the system, experiments were first carried out with semi-insulating GaAs at room temperature. If circularly polarized incident laser power of 100–200 mW is completely absorbed by GaAs to generate electrons with rate G, the estimated number of spin-polarized electrons ($n_s = \eta G \tau_s$) is about 10^7, assuming an orientation efficiency (η) of 50% and spin lifetime (τ_s) of $\sim$100 ps at room temperature. Given the dimensions, number of turns, and Q factor of the pickup coil, the theoretical sensitivity γ, defined as the output voltage generated per unit magnetic moment modulated at the resonance frequency, is estimated to be $\sim 10^8$ VT/J at the resonant frequency. The expected output signal (M) is therefore only $\sim$10 nV. If the EOM is driven by a high voltage signal at the same frequency, it is extremely difficult to avoid spurious signals picked up by the sensitive detector, despite extensive rf shielding and grounding precautions. In addition, spurious signals can also be caused by the small ($< 1\%$) intensity modulation of the laser beam. To overcome these difficulties, the EOM was driven at half the resonance frequency with an amplitude and dc bias such that the phase difference ϕ between the two orthogonal components of the beam varied sinusoidally as $\phi(t) = \pi/2 + \phi_m \cos\omega t$. Since ϕ crosses 90 degrees twice in every cycle, the circularity of the polarization is modulated at double the excitation frequency, i.e., the resonance frequency of the coil. In this way the spurious pickup due to the high voltage excitation is rejected by the rf lock-in amplifier. Keeping the phase difference modulation constant, the plane of polarization of the incident beam (θ_0) was rotated with respect to the EOM axis using the half-wave plate. When the polarization plane is at 45 degrees with respect to the EOM axis, the spin polarization signal is maximum. As the half-wave plate is rotated, the polarization signal decreases and goes to zero when the incident beam is aligned with either of the EOM axes. Further rotation of the plate will cause the signal to reach a maximum with opposite sign. The theoretical (rms) signal measured by the lock-in amplifier in zero magnetic field is given by:

$$M_0 = \sqrt{2}\gamma g \mu_B n_s \sin(2\theta_0) J_2(\phi_m), \tag{1}$$

where μ_B is the Bohr magneton, g is the Lande g factor, $J_2(x)$ is the Bessel function, and other parameters defined above.

This dependence on θ_0 was clearly observed in GaAs as shown in Fig. 2. The inset shows the raw data which are the output of the lock-in amplifier sampled every 3 seconds. After every 10 minutes, the half-wave plate was rotated by 22.5 degrees, changing the incident beam polarization plane by 45 degrees. Note that the signal levels are in nanovolts, in agreement with the theoretical estimates given above. It can also be noted that the drift in the background is less than 5 nV over 3 hours of data collection. The time-averaged values for each position of the half-wave plate are shown in Fig. 2, which illustrates the expected sinusoidal dependence. In this way spin polarization signals can be measured in zero external magnetic field. In order to further validate the origin of this signal, an external field was applied parallel to the sample surface. As expected from the Hanle Effect, the polarization signal (M) decayed with increasing field (B) as given by the Hanle equation [1]:

$$M(B) = \frac{M_0}{1 + (\Omega \tau_s)^2}, \tag{2}$$

where $\Omega = \frac{g\mu_B B}{\hbar}$. In this expression, M_0 is the signal in zero magnetic field and τ_s is the spin lifetime, i.e., rate at which the net spin n_s ($= n_\uparrow - n_\downarrow$) relaxes. Note that the

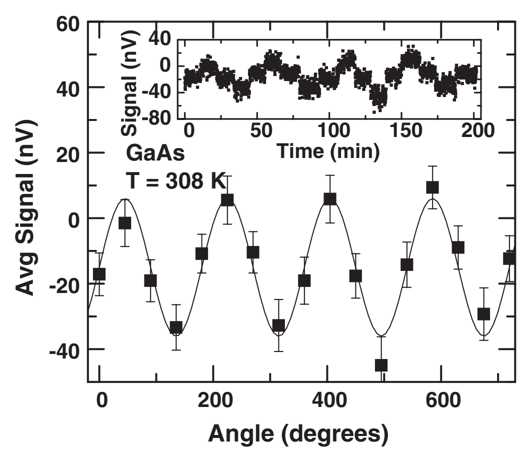

FIG. 2. Time-averaged signal from semi-insulating GaAs in zero magnetic field plotted against the change of angle between plane of incident polarization and the optical axis of the EOM. Inset shows the raw data acquired from the lock-in amplifier every 3 seconds for a total of $\sim$3 hours. The half-wave plate was rotated every $\sim$10 min to change the polarization axis. The solid line is a sinusoidal fit to the data.

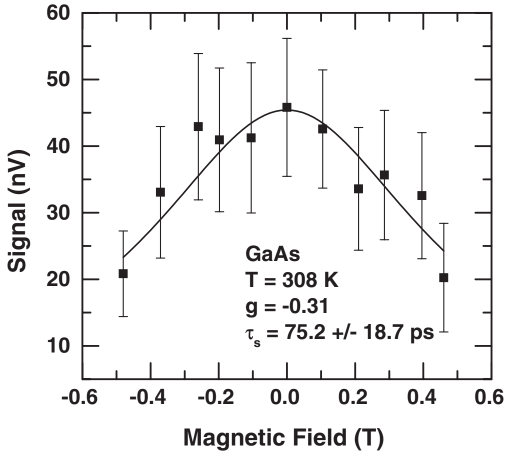

FIG. 3. Dependence of output signal (peak-to-peak value of the sinusoidal fit shown in Fig. 2) in GaAs on a magnetic field applied parallel to sample surface. Sample temperature was slightly higher than room temperature due to the high power laser beam. The data were fitted to the Hanle equation to extract the spin relaxation lifetime of 75.2 ± 18.7 ps.

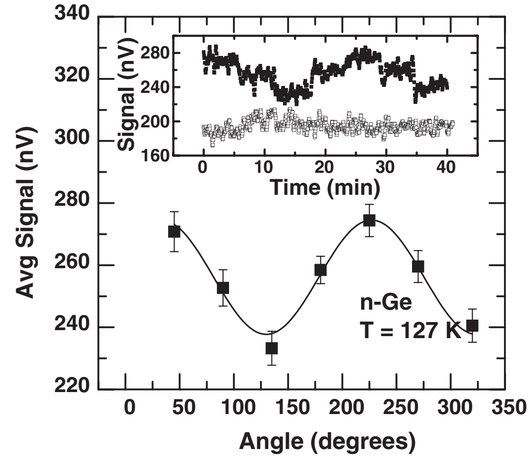

FIG. 4. Time-averaged signal from n-Ge in a zero magnetic field plotted against the change of angle between the plane of incident polarization and the optical axis of the EOM. Inset shows the raw data acquired from the lock-in amplifier every 3 seconds for a total of ~40 min at 127 K (filled squares) and 300 K (open squares). The half-wave plate was rotated every ~6 min to change the polarization axis in both cases. The null result at room temperature confirms that there is no spurious modulation of the background signal. The solid line is a sinusoidal fit to the data.

electron-hole recombination lifetimes are much longer than the τ_s for both GaAs and Ge. By fitting the measured data to the above equation (using the Lande g factor [1] of -0.31 for electrons in GaAs at 300 K) a spin lifetime of ~75 ps was obtained as shown in Fig. 3, in agreement with established values [13].

The technique was then applied to an indirect gap semiconductor, germanium. Samples were cut from a (100) n-Ge Sb-doped 0.01 Ω cm 350–400 μm thick bulk wafer. The laser $\lambda = 1342$ nm was incident normally with power 130 mW and beam diameter ~1 mm at the sample surface. Polarization modulation was carried out as before with $\phi_m = 0.36\pi$. At room temperature there was no change in the signal when the half-wave plate was rotated. However, when cooled to the base temperature of the cryostat ($T = 127$ K at the sample position in the presence of the laser) a clear signal was observed as shown in Fig. 4. The sinusoidal variation with the plane of polarization establishes the origin of the signal to be due to the spin polarization. For confirmation, a weak external magnetic field was applied parallel to the sample surface as before. As shown in Fig. 5, the signal decreases rapidly with magnetic field due to the Hanle Effect, thus excluding any spurious origin of the signal. It should be noted that, in contrast to GaAs, magnetic fields as low as a few millitesla are sufficient to observe the Hanle depolarization in Ge, indicating that the spin lifetimes are large. A fit to the experimental data using Eq. (2) with the experimental [14] Lande g factor of 1.6 yields a spin lifetime of 4.6 ± 1.0 ns. As mentioned previously, such large lifetimes are expected due to the absence of the D'yakonov-Perel spin relaxation mechanism in Ge. The g factor for electrons in each ellipsoidal L valley is actually anisotropic [15] with the theoretical values $g_{\parallel} = 0.9$ and $g_{\perp} = 2.04$ where parallel ($g_{\parallel}$) and perpendicular ($g_{\perp}$) refer to the [111] symmetry axis. In our experiment, the magnetic field was in the (100) plane; therefore the calculated g factor varies from 1.39 to 2.04 depending on the valley and direction of the field in the (100) plane. The total response of electrons in all the valleys to the magnetic field $M^{\text{tot}}(B) = \sum M_0^i/[1 + (\Omega_i B)^2]$ is close to the fitted curve in Fig. 5 using a single average g factor of 1.6, and cannot be distinguished by the data.

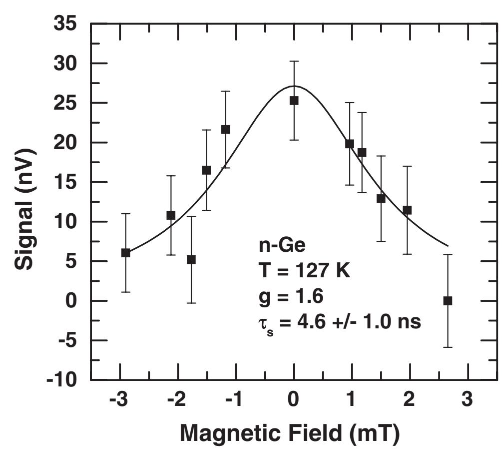

FIG. 5. Dependence of output signal (peak-to-peak value of the sinusoidal fit shown in Fig. 4) in Ge at 127 K on a magnetic field applied parallel to sample surface. Note the magnetic field scale compared to Fig. 3. The data was fitted to the Hanle equation to extract the spin relaxation lifetime of 4.6 ± 1.0 ns.

To determine the optical spin orientation efficiency, the absolute sensitivity of the detection system was calibrated to be 3.8×10^7 VT/J by using a small current carrying coil of known magnetic moment in place of the sample. The incident laser power of 130 mW at 1342 nm, after accounting for 30% reflection loss, was completely absorbed by the optically thick sample to generate about 6×10^{17} electrons per second (G) in steady state. If all these electrons were spin polarized by the circularly polarized radiation (i.e., orientation efficiency $\eta = 100\%$) and the measured spin lifetime (τ_s) is 4.6 ns at 127 K, the number of spin-polarized electrons ($n_s = \eta G \tau_s$) will be 2.7×10^9. From Eq. 1, the maximum signal (at $\theta_0 = \pi/4$) is expected to be 315 nV. However the measured signal is 15 nV and therefore it is concluded that the spin orientation efficiency (η) is only 4.8% at this wavelength. It was previously reported [11] that the efficiency does not depend strongly on direct or indirect gap excitation in Ge. Theoretical calculations of the orientation efficiency are not available for Ge, but recent calculations [16] for Si indicate that this number is ~2%–20% depending on the photon energy. If we assume that the spin orientation and detection efficiency [17] are similar in optical pump-probe experiments, then the signals in Ge should be lower by a factor $\sim(0.05)^2$, i.e., 2 or 3 orders of magnitude when compared to GaAs, which is consistent with the observations [11]. The modulation spin polarization experiments were also attempted with p-Ge samples, but the observed signals at 300 K and 127 K were too weak to be resolved. This indicates that the electron spin lifetime in p-Ge is much smaller, possibly due to strong Bir-Aronov-Pikus [2] spin relaxation.

In summary we have demonstrated a sensitive rf detection technique which directly measures the weak magnetic field generated by a small number of optically generated spin-polarized electrons. We have applied the technique to n-Ge and obtain a spin lifetime of 4.6 ns and spin orientation efficiency of 4.8% at 127 K using 1342 nm optical excitation. The technique can be readily applied to other indirect semiconductors such as Si in bulk or quantum well samples, and even to direct gap semiconductors where the optical detection signals (especially at higher temperatures) are weak.

The authors would like to thank Dr. Arjun Joshua for assistance with the experimental setup and Professor Vasant Natarajan for several optical components. The authors would also like to thank the Department of Science and Technology (DST), Government of India for funding this project.

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