Nuclear Magnetic Resonance in a Ferromagnet-Semiconductor Heterostructure

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We report the observation of nuclear magnetic resonance (NMR) in a ferromagnet-semiconductor heterostructure in the presence of a spin-polarized current. Spin-polarized electrons injected from a metallic ferromagnet generate a large nuclear spin population in a GaAs quantum well by dynamic polarization. The characteristic time for the polarization process is approximately 20 sec, and the nuclear polarization can persist for several minutes after the current is turned off. Resonant depolarization is observed in the presence of an AC magnetic field or when the injection current is modulated at the NMR frequency.

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A detailed understanding of the hyperfine interaction is a prerequisite for the coherent control of electron and nuclear spin systems in semiconductor heterostructures. Electrical spin injection provides a simple means of introducing spin-polarized carriers into a semiconductor, but it has proven more difficult to achieve the degree of tunability over the combined electron and nuclear spin systems demonstrated in optical pumping experiments. For example, Poggio et al. have recently shown that it is possible to electrically control the spatial profile of optically-pumped nuclear polarization within a single quantum well. One of the advantages of optical pumping is the efficiency of dynamic nuclear polarization (DNP), in which a non-equilibrium nuclear polarization is generated by the spin relaxation of electrons. In principle, the DNP process can also be driven by a spin-polarized current. This was inferred in recent experiments on Fe/Al$_x$Ga$_{1-x}$As heterostructures, although an explicit dynamical signature of DNP was not demonstrated.

In this letter we report the direct observation of nuclear magnetic resonance (NMR) in a GaAs quantum well (QW) in the presence of a spin-polarized current. We demonstrate that DNP can be partially suppressed by resonant depolarization of the nuclear spins. The polarization can be achieved either by the traditional approach of applying a resonant AC magnetic field or by modulating the spin-polarized current at the NMR frequency. The time dependence of the DNP process is measured, indicating a characteristic build-up time of $\approx$ 20 sec and persistence of nuclear polarization up to eight minutes after the spin-polarized current has been turned off.

The heterostructure is grown by molecular beam epitaxy on a p$^+$ GaAs (100) substrate and consists of p-Al$_{0.1}$Ga$_{0.9}$As/QW/n-Al$_{0.1}$Ga$_{0.9}$As/Fe/Al. The QW comprises 100 Å GaAs between intrinsic Al$_{0.1}$Ga$_{0.9}$As barriers, and the Fe film is 50 Å thick. The metal layers are grown in situ at $\approx 0^\circ$ C, and a $3 \times 10^{13}$ atoms/cm$^2$ $\delta$-doped layer of Si is grown 25 Å from the Fe/Al$_x$Ga$_{1-x}$As interface to create a thin Schottky tunnel barrier. The devices are fabricated into bars with an active region 80 $\mu$m wide (see schematic drawing in Fig. 1), and the experiments are carried out in a magneto-optical cryostat at temperatures ranging from 2 K to 40 K.

The measurements are performed in the Voigt geometry with the applied field $H_0$ in the plane of the QW. Electroluminescence (EL) is collected along the growth direction, indicated by $k$ in Fig. 1, which coincides with the direction of the AC magnetic field $H_1$ generated by a four-turn coil placed on top of the sample. The EL is due to the recombination of electrons tunneling from the Fe layer with heavy holes flowing from the substrate. The circular polarization of the EL, referred to as the ELP signal, is equal to the component of electron spin polarization along $k$ at the time of recombination.

Typical ELP data as a function of applied field are shown in Fig. 1. All of the measurements under discussion here were conducted with $H_0$ nearly parallel to the [011] direction, which corresponds to a magnetic hard axis. As $H_0$ decreases below 500 Oe, the magnetization $M$ rotates away from [011] towards the [011] (easy) direction. The non-zero angle between the injected spin $S_0$ and $M$ leads to precession of the spin in the QW, resulting in a steady-state component of $S$ along $k$ and hence the observed ELP signal. The ELP hysteresis loops shown in Fig. 1 can be modeled extremely well provided that the total field $B = H_0 + B_N$, where $B_N$ is assumed to be the hyperfine field due to nuclei polarized by DNP. As discussed in Ref. 12, the magnitude of $B_N$ depends on the injection current density and the angle $\theta$ between $S_0$ and $H_0$ and reaches several kilogauss for current densities $\approx 10$ A/cm$^2$ and $\theta < \pi/2$.

The large nuclear magnetic field is due to the long spin-relaxation time for nuclei and the large difference between the electron and nuclear magnetic moments. Since the nuclear spin-lattice relaxation time can be many minutes, we expect the field $B_N$ to build up on laboratory time scales when the current is turned on and then decay slowly when the current is turned off. Figure 2 shows the ELP signal as the device current is switched on and off for variable amounts of time. In this case, the current is off for ten minutes prior to the start of data collection at $t = 0$. The LED is then turned on.
and the ELP signal is measured for three minutes. The increase in the ELP signal is due to the increase in total internal magnetic field and the consequent increase in the angle of precession of the injected spin polarized electrons. To probe the lifetime of the nuclear polarization in the absence of continuous pumping, the current is turned off for a time $t_{\text{off}}$ and then on again for three minutes. Figure 2 shows a series of these measurements with $t_{\text{off}}$ varying from 1 min to 9 min. The first data point taken after each “off” period indicates the degree of nuclear polarization that has been retained. By repeating this procedure with increasing $t_{\text{off}}$, we observe a decay time of approximately 5 min. Although both the build-up and decay times for the ELP signal are long, it is difficult to make a direct quantitative mapping between the magnitude of the ELP signal and the actual nuclear polarization. In particular, the relationship between the ELP signal and $B_N$ is very non-linear. The ELP signal increases rapidly at small nuclear fields and then saturates, and this is responsible in part for the large difference between the build-up and decay times for the ELP signal.

Although the nuclear polarization can relax slowly through coupling to the lattice, the most effective way of destroying DNP is to apply an oscillating magnetic field $\mathbf{H}_1$ at the NMR frequency along a direction perpendicular to $\mathbf{H}_0$. The transverse field couples directly to the nuclear spin system, leading to depolarization of the nuclei that are in resonance. $\mathbf{H}_1$ is generated by a 1 cm diameter, four turn coil driven by the sinusoidal output of a function generator. By sweeping the frequency of $H_1$ at fixed $H_0$, depolarization signatures for each of the three isotopes in the GaAs QW $^{75}$As ($\gamma = 0.731 \text{ kHz/Oe}$), $^{69}$Ga ($\gamma = 1.025 \text{ kHz/Oe}$), and $^{71}$Ga ($\gamma = 1.302 \text{ kHz/Oe}$) are observed, as seen in Fig. 3(a) for $H_0 = 330 \text{ Oe}$. In each case, the linewidth of the depolarization signature is 10 - 15 kHz, with no systematic dependence on magnetic field. The linewidth at these fields ($< 500 \text{ Oe}$) is therefore not limited by inhomogeneous dephasing.

Frequency sweeps were conducted at several different magnetic fields. The peaks of the depolarization features for all three isotopes are plotted versus $H_0$ in Fig. 3(b). Linear fits of $\nu = \gamma H_0$ were obtained for each isotope, yielding gyromagnetic ratios $\gamma^{75}\text{As} = 0.71 \pm 0.1 \text{ kHz/Oe}$,
As described by previous studies, nuclear resonances have also been observed in sub-harmonics such as $\nu/3$. Some of the additional peaks can be identified by comparing the data taken under AC magnetic field (upper curve). In the alternating device current (lower curve) with the data obtained with an AC magnetic field, and we attribute them to depolarization is resonant modulation of the device current. In this case, the coil is not used ($H_1 = 0$), and the DC bias voltage is set above the threshold for light emission. An AC voltage is then added so that the average bias voltage corresponds approximately to the value used in the measurements discussed above. In this case, the bias is being used only to modulate the injection current. Given the square well design, we cannot control the relative position of the nuclear and electronic spins as achieved recently in parabolic quantum wells. Fig. 4 shows the ELP signal as a function of bias voltage frequency for $H_0 = 330$ Oe, $V_{DC} = 2.1$ V, and $V_{AC} = 0.5$ V. The principal resonances are clearly visible, as can be seen by comparing the data taken under alternating device current (lower curve) with the data taken under AC magnetic field (upper curve). The reason for this non-intuitive result is that for sufficiently large nuclear fields, the average angle through which the electrons precess during their lifetime becomes larger than $\pi/2$. An increasing nuclear field therefore results in a decrease in the magnitude of the ELP signal because the electrons are actually precessing further away from the measurement axis. Although a complete quenching of $B_N$ would result in nearly total suppression of the ELP signal, only a single isotope is depolarized at each resonance. We have confirmed that for low $B_N$, corresponding to applied fields less than 60 Oe, resonant depolarization results in a decrease in the ELP magnitude as expected.

Since the nuclear polarization in these devices is generated by the injected electrons, an alternative approach to depolarization is resonant modulation of the device current. In this case, the coil is not used ($H_1 = 0$), and the DC bias voltage is set above the threshold for light emission. An AC voltage is then added so that the average bias voltage corresponds approximately to the value used in the measurements discussed above. In this case, the bias is being used only to modulate the injection current. Given the square well design, we cannot control the relative position of the nuclear and electronic spins as achieved recently in parabolic quantum wells. Fig. 4 shows the ELP signal as a function of bias voltage frequency for $H_0 = 330$ Oe, $V_{DC} = 2.1$ V, and $V_{AC} = 0.5$ V. The principal resonances are clearly visible, as can be seen by comparing the data taken under alternating device current (lower curve) with the data taken under AC magnetic field (upper curve). The reason for this non-intuitive result is that for sufficiently large nuclear fields, the average angle through which the electrons precess during their lifetime becomes larger than $\pi/2$. An increasing nuclear field therefore results in a decrease in the magnitude of the ELP signal because the electrons are actually precessing further away from the measurement axis. Although a complete quenching of $B_N$ would result in nearly total suppression of the ELP signal, only a single isotope is depolarized at each resonance. We have confirmed that for low $B_N$, corresponding to applied fields less than 60 Oe, resonant depolarization results in a decrease in the ELP magnitude as expected.

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Sub-harmonics of the primary resonances are never observed with an AC magnetic field, and we attribute them to the non-sinusoidal time dependence of the injection current as the bias voltage is modulated. This is a consequence of the diode structure of the device. The origins of harmonics of the fundamental resonances are less clear. These peaks are also observed in AC magnetic field experiments at high amplitudes ($H_1 > 0.4$ Oe). The observation of the $\Delta m = \pm 2$ peaks in the AC magnetic field experiments at high amplitudes indicates that in this experiment they originate from some intrinsic non-linearity. Dipole-dipole coupling can lead to both $\Delta m = \pm 2$ and $\Delta m = \pm 3$ transitions and may play a more significant role because of the large nuclear polarization in these experiments.

The spectroscopic measurements presented here unambiguously demonstrate dynamic nuclear polarization by electrical spin injection. The ability to modulate the nuclear polarization with a spin-polarized current provides a unique means of controlling the coupled electron-nuclear spin system. This capability, in combination with the large nuclear polarization achieved at low fields, provides a new approach to NMR spectroscopy of heterostructures.

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$$\gamma^{\text{Ga}} = 1.00 \pm 0.04 \text{ kHz/Oe}, \quad \text{and} \quad \gamma^{\text{Ga}} = 1.27 \pm 0.04 \text{ kHz/Oe},$$

in agreement with the accepted values. The deviations from the fits are smaller than the limit set by the trapped flux in the (≈ 10 Oe).

Fig. 4 shows the ELP signal as a function of bias voltage at $T = 20$ K, and $H_0 = 330$ Oe corresponding to the three isotopes present in the GaAs QW. The upper curve shows the ELP signal measured with an AC magnetic field $H_1 \approx 0.2$ Oe. The lower curve shows the ELP signal measured under alternating device current ($H_1 = 0$ Oe). The filled symbols identify principal nuclear transition frequencies and their sub-harmonics. The empty symbols indicate harmonics of the primary resonances as well as their sub-harmonics.

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