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Collective single-mode precession of electron spins in an ensemble of singly charged (In,Ga)As/GaAs quantum dots

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We show that the spins of all electrons in an ensemble of singly charged (In,Ga)As/GaAs quantum dots can be driven into a single mode of precession about a magnetic field. This regime is achieved by allowing only this single mode within the electron-spin precession spectrum of the ensemble to be synchronized with a train of periodic optical excitation pulses. Under this condition a nuclei-induced frequency focusing leads to a shift of almost all spin precession frequencies into the synchronized mode. The macroscopic magnetic moment of the electron spins that is created in this regime precesses free of dephasing induced by inhomogeneous distribution of g factors.

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Solid-state implementations of quantum information processing promise scalability toward large numbers of qubits.[1,2] However, they are typically impeded by the nonideal crystal environment leading to a wide dispersion of the properties of elementary excitations envisaged as qubits. This gives rise to a number of complications: a single excitation needs to be isolated, which often requires high resolution in space, energy, etc. The read-out signal of such an excitation is typically weak, so measurements may require times comparable to the decoherence time.[3] These problems would be overcome if one had access to an ensemble of identical quantum bits, all prepared in the same quantum state. However, this is prevented by the unavoidable inhomogeneities.

A qubit candidate with promising features is an electron spin confined in a quantum dot (QD).[1–8] Its decoherence time T_2 at cryogenic temperatures is in the microseconds range, as determined by a spin-echo measurement on a single GaAs/(Al,Ga)As gated QD.[9] This property, which should allow one to perform many operations coherently, is, however, obscured in a QD ensemble by fast dephasing of electron-spin polarization due to the frequency dispersion for precession about a transverse magnetic field.[10,11] The dephasing could be suppressed for particular spin subsets by synchronizing their precession with the repetition rate of the periodically pulsed laser used for generation of spin polarization.[12] The precession frequencies in these subsets satisfy the mode-locking condition: $\omega_K=2\pi K/T_R$, where T_R is the pulse repetition period and K is an integer. Fulfillment of this condition gives rise to bursts in the Faraday rotation (FR) signal measured from an (In,Ga)As/GaAs QD ensemble right before excitation pulse arrival. The FR signal decay allowed us to measure $T_2=3\ \mu$s.[12]

The majority of electrons in the ensemble would not satisfy the mode-locking condition if the electron-spin precession frequency in an individual dot was determined just by the external magnetic field B and the electron g factor g_e. In most *III-V* compound QDs, however, an electron is also exposed to the collective hyperfine field of the dot nuclei. As a result, the electron-spin precession frequency, $\omega=\mu_B g_e B/\hbar+\omega_{N,x}$, contains the nuclear contribution, $\omega_{N,x}$, which is proportional to the projection of the nuclear spin polarization on the external field ($\boldsymbol{B}\|\boldsymbol{x}$). Here μ_B is the Bohr magneton. The magnetic field suppresses magnetodipole interactions between nuclei and, in darkness, the projection of the nuclear spin polarization does not change for hours or even days. The resonant optical excitation of the QDs leads, however, to light-assisted flip-flop processes between electron and nuclei. The consequent random fluctuation of $\omega_{N,x}$ eventually drives almost all electron spins in the ensemble into synchronized modes, corresponding to a nuclei-induced frequency focusing effect.[13] For the experimental conditions in Refs. 12 and 13 still a few tens of mode-locked frequencies were excited. Their superposition results in a damped FR signal with oscillations given by the central frequency of synchronized modes. Therefore, the FR traces show dephasing of spin coherence on a ns time scale. Complete suppression of the dephasing would require precession of all electron spins in the QD ensemble on a single frequency, i.e., focusing of the spins to a single mode.

In this Rapid Communication we demonstrate a regime where 95% of about a million QD electrons processes at a single frequency. The regime is achieved by proper tailoring of the laser excitation protocol and magnetic field strength, and it allows controlled switching between single and double-mode precessions in the magnetic field range from about 50 mT to 1 T.

The time-resolved pump-probe FR measurements were performed on singly negatively charged (In,Ga)As/GaAs self-assembled QDs (see Ref. 14 for details). The sample was held at a temperature $T=6$ K in a superconducting split coil, and magnetic fields were applied perpendicular to the sample growth axis. For optical excitation we used a mode-locked Ti:Sapphire laser emitting 4 ps pulses at a rate of 75.6

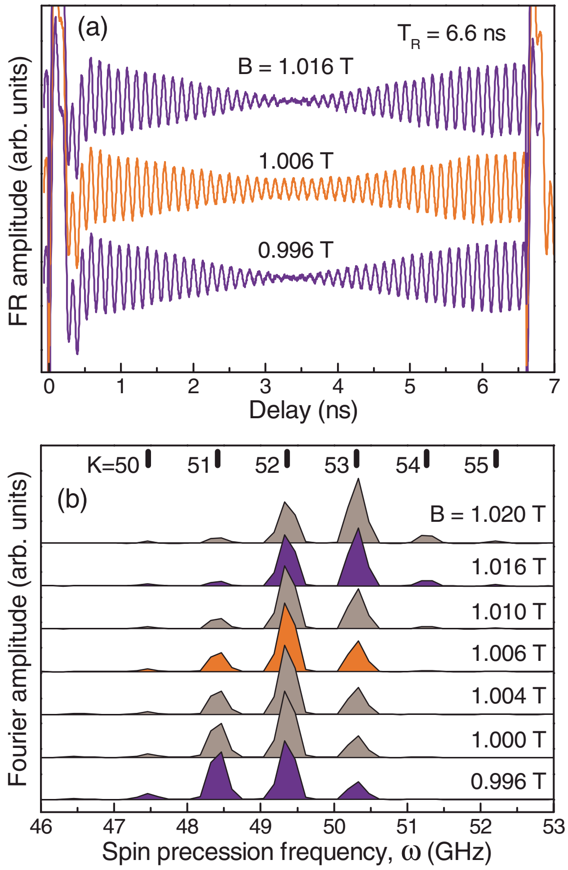

FIG. 1. (Color online) (a) Faraday rotation signals recorded around $B=1$ T. Pump and probe with degenerate photon energies at 1.38 eV have powers of 23 and 13 W/cm^2, respectively. (b) Fourier transforms extracted from FR signals measured over an eight period time interval of 54 ns. Positions of phase-synchronized modes ω_K are marked by vertical lines.

MHz (13.2 ns pulse separation). The pump pulse repetition rate was doubled to 151.2 MHz, corresponding to $T_R=6.6$ ns, by splitting the pump into two beams with 50:50 intensity ratio and delaying one beam by 6.6 ns. The photon energy was tuned to the QD ground-state optical transition. The circular polarization of the pump pulses was modulated at 50 kHz. The probe pulses were linearly polarized, and their energy was either equal to the pump (degenerate FR) or different from it (nondegenerate FR).

Figure 1(a) shows FR traces for magnetic fields around 1 T. The pump pulses hit the sample at times $t=0$ and 6.6 ns. Over a narrow range of magnetic fields the FR traces undergo strong modifications, although the oscillation frequencies appearing in them do not change significantly. At $B=0.996$ and 1.016 T the signal amplitude shows a strong decay after the first pulse, hits a node in the middle between the pumps, and afterwards increases symmetrically toward the second pulse. However, at the intermediate field of 1.006 T, deviating by 10 mT only from the two other traces, the signal decay after the first pulse is weaker and, in particular, it does not show a node. This nonmonotonic behavior of the FR signal with B is repeated every 0.02 T and suggests involvement of only a few precession modes.

Indeed, the Fourier spectra in Fig. 1(b) confirm that the

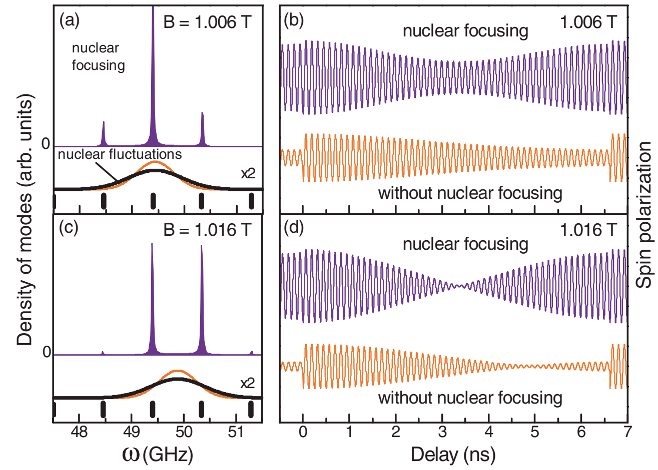

FIG. 2. (Color online) Calculations of the density of electron-spin precession modes $\rho(\omega)$ and corresponding FR signals at $B=1.006$ and 1.016 T. Panels (a) and (c) show the density of modes created by g-factor dispersion (red/gray line), by nuclear fluctuations (black line), and by nuclei-induced frequency focusing. Positions of phase-synchronized modes ω_K are marked by vertical lines. Panels (b) and (d) show the spin polarization which is proportional to the FR signal magnitude, calculated with and without frequency focusing. Parameters for calculations: $|g_e|=0.556$, $\Delta g_e=0.004$, $\Delta\omega_{N,x}=0.37$ GHz, and pump pulses with π area (Ref. 15).

FR signal in this interval of magnetic fields is created by four to six electron-spin precession modes, out of which two to three have strong weight. These Fourier transforms were obtained by integrating over a time interval equal to eight repetition periods T_R to improve the resolution. With increasing magnetic field the center of the contributing precession frequencies shifts to higher values with respect to the discrete mode-locked frequency spectrum, $\omega_K=2\pi K/T_R$, where $K=50-55$ around 1 T. This shift changes the relative contribution of the different modes to the Fourier spectra. If for a magnetic field two strong modes of the same weight dominate the spectrum, the FR signal shows a node between the pump pulses (see $B=0.996$ and 1.016 T). For the intermediate field of $B=1.006$ T, the FR signal is determined by a central mode which is accompanied, however, by strong symmetric satellites. Therefore, the FR signal has a considerable amplitude in between the pulses.

Under resonant optical excitation the nuclei drive the electron-spin precession frequencies in all dots to modes satisfying the phase synchronization condition (PSC) (Ref. 13): $\omega_K=2\pi K/T_R$. This redistribution removes the nonsynchronized background in the precession mode density, $\rho(\omega)$, by focusing on a few leading modes for the chosen conditions. In Fig. 2 we show the theoretical mode density and the corresponding FR spectra created by a train of π pulses with repetition period of $T_R=6.6$ ns at $B=1.006$ and 1.016 T. The calculations[13] are in good agreement with the experimental data. For comparison we also show the corresponding dependencies without nuclei-induced frequency focusing. The significant deviation from the experimental observations underlines the importance of the nuclear contribution in the mode locking.

Let us estimate the number of mode-locked modes that contribute to the FR signal for a particular magnetic field strength. The separation between these modes is $2\pi/T_R$.[12] Consequently, the number of such modes, M, is

$$M = 2\Delta\omega(\Delta g_e, \Delta\omega_{N,x})T_R/2\pi, \tag{1}$$

where $\Delta\omega(\Delta g_e, \Delta\omega_{N,x})$ is the half-width at a half-maximum dispersion of electron-spin precession frequencies in the QD ensemble,

$$\Delta\omega(\Delta g_e, \Delta\omega_{N,x}) = \sqrt{[\mu_B \Delta g_e B/\hbar]^2 + \Delta\omega_{N,x}^2}. \tag{2}$$

Here Δg_e is the dispersion of electron g factors in the ensemble of optically excited dots and $\Delta\omega_{N,x}$ is the nuclear contribution to the dispersion of electron-spin precession frequencies for each specific dot. The magnitude of $\Delta\omega_{N,x}$ is determined by statistical fluctuations of the nuclear spin polarization projection onto the magnetic field in the dot volume.[10] For our dots $\Delta\omega_{N,x}=0.37$ GHz.[15]

Equations (1) and (2) define a clear strategy for achieving the single-mode precession regime in a QD ensemble. The number of mode-locked modes can be reduced: (a) by minimizing Δg_e, (b) by reducing T_R, (c) by decreasing B, and (d) by decreasing $\Delta\omega_{N,x}$.

(a) Generally, the dispersion Δg_e in a QD ensemble is connected to variations in dot shape and size. For (In,Ga)As dots a systematic dependence of the electron g factor on energy of the band edge optical transitions has been observed.[14] Δg_e can then be controlled by the laser spectral width, which is inversely proportional to the pulse duration. However, as one is interested in fast spin initialization, the duration should not exceed ~10 ps (spectral width of ~0.1 meV). Otherwise the efficiency of spin polarization initialization drops when the pulse duration becomes comparable with the times of hole spin precession and electron-hole recombination.[14]

(b) A reduction in the repetition period is generally limited by the trion decay time and the time scale that multiple coherent operations would require. For practical reasons, to observe the single-mode regime, T_R should be longer than the ensemble dephasing time of a few ns.[14]

(c) A reduction in the magnetic field strength is possible to an extent that it is still considerably larger than the randomly oriented effective field of the nuclei. Otherwise the nuclei would induce fast dephasing.[10,17] In our dots the random nuclear fluctuation field has an amplitude of about 7.5 mT.[15] Also, to conserve nuclear spin polarization, which is needed for frequency focusing, the magnetic field should exceed the hyperfine field of the electron acting on the nuclei (Knight field), which is about 1–3 mT in our dots.

In Fig. 3(a) we show the magnetic field dependence of precession frequency dispersion $\Delta\omega(\Delta g_e, \Delta\omega_{N,x})$ and mode number M calculated for our dots under the applied experimental conditions. For $B<0.8$ T, we estimate $0.75<M\leq 1$, giving a lower limit for the number of mode-locked frequencies in the FR signal. In this range the Δg_e contribution to $\Delta\omega$ (dash-dotted line) is smaller than the nuclei-induced dispersion (dashed line). The total dispersion is therefore approximately equal to $\Delta\omega_{N,x}$, which is independent of magnetic field. This results in periodic switching between almost pure single and double-mode regimes for $B<0.8$ T.

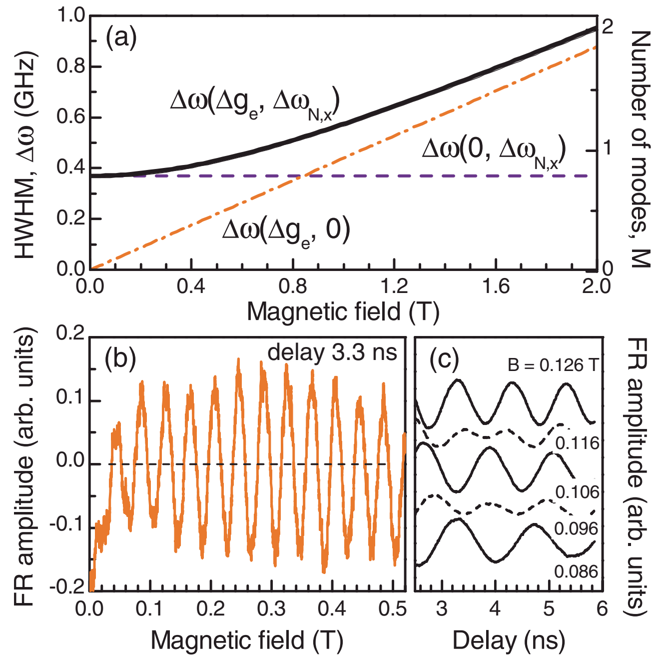

FIG. 3. (Color online) (a) Magnetic field dependence of dispersion of electron-spin precession frequencies, $\Delta\omega(\Delta g_e, \Delta\omega_{N,x})$ (left scale) and of number of mode-locked frequencies (right scale) contributing to the FR signal (solid line). Dashed line shows the nuclear contribution to the dispersion, $\Delta\omega_{N,x}$, in our QDs. The contribution by the electron-spin g-factor dispersion is shown by dash-dotted line. Parameters are the same as in Fig. 2. (b) Magnetic field dependence of the FR signal at 3.3 ns delay for pump and probe powers of 13 W/cm^2 each. (c) FR spectra recorded at different magnetic fields. The dashed lines correspond to zero FR amplitude at panel (b) and the solid lines to its maximal and minimal values.

Whether one or two modes fall within the dispersion can be adjusted by the magnetic field, which shifts the spectrum of optically excited electron-spin precession frequencies in the QD ensemble relative to the spectrum of phase-synchronized modes.

The magnetic field dependence of the FR amplitude midway between the pump pulses (at a delay of 3.3 ns, where the node is observed) oscillates symmetrically around zero value illustrating the mode switching [Fig. 3(b)]. It is partly contributed by the change in central precession frequency by the field scan as the harmonic oscillation of spin precession is tuned through the fixed delay. But the dominating effect comes from the overall amplitude oscillation due to mode interference, as seen from panel (c). The strongest signal in panel (b), either minimum or maximum, is reached for an odd number of modes, while zero signal is caused for an even mode number.

Magnetic fields larger than 0.8 T increase the dispersion of spin precession frequencies, see Fig. 3(a), and allow $\Delta\omega$ to cover more than three mode-locked frequencies. This increases the amplitude of side modes significantly, as seen in Fig. 1(b) for $B=1$ T, and, consequently, leads to considerable dephasing.

To address experimentally the single-mode regime we apply very weak magnetic fields in order to minimize the contribution of Δg_e. In Fig. 4(c) the spectral broadening due to

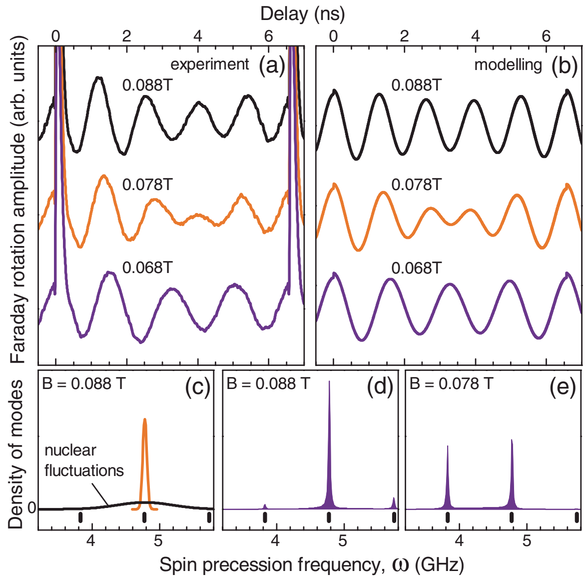

FIG. 4. (Color online) (a) FR traces measured at weak magnetic fields to demonstrate the switching from single-mode to double-mode regime. Pump energy 1.3837 eV at power 130 W/cm^2, probe energy 1.3842 eV at power 13 W/cm^2. (b) Modeled FR signals. (c) Calculated density of electron-spin precession modes due to g-factor dispersion (red/gray) and nuclear fluctuations (black). (d) and (e) Calculated density of modes accounting for the nuclei-induced frequency focusing. Parameters are the same as in Fig. 2. Positions of phase-synchronized modes ω_K are marked by vertical dashes.

Δg_e is much smaller than the distance between neighboring mode-locked modes, but it becomes strongly broadened by the nuclei-induced dispersion $\Delta\omega_{N,x}$. The experimental spectra shown in Fig. 4(a) were measured around 0.078 T by nondegenerate pump probe to increase the signal contrast. A spectrum with a node in the middle is seen at 0.078 T. However, for fields of 0.068 and 0.088 T, the FR amplitude shows virtually no decay between the pump pulses. Indeed, the calculated mode density in Fig. 4(d) is dominated by a strong central peak at 4.78 GHz (corresponding to $K=5$). Two satellites, which arise from the small overlap of $\rho(\omega)$, determined by the nuclear fluctuations, with the three phase-synchronized modes [panel (c)], are hardly visible. It is remarkable that under this condition about 95% of precession frequencies is focused on the single mode.

(d) For a true single-mode regime the condition $2\Delta\omega_{N,x}<2\pi/T_R$ should be satisfied. This can be reached by increasing the dot size because $\Delta\omega_{N,x}$ is controlled by the statistical fluctuations of the nuclear spin polarization in the dot volume, V, given by $\Delta\omega_{N,x}\sim 1/\sqrt{V}$. Otherwise, quantum dots with other nuclear composition, e.g., with nuclei having smaller nuclear spins, might be studied.

In summary, we have demonstrated that mode locking, combined with nuclei-induced frequency focusing, allows us to drive an entire ensemble of electron spins, confined in singly charged quantum dots, into coherent single-mode precession. The coherently synchronized precession of a million spins in the QD ensemble represents a macroscopic magnetic moment. Therefore, this regime will be very useful for studying various coherent phenomena, such as electromagnetically induced transparency or control-NOT gate operations.

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