Stabilizing nuclear spins around semiconductor electrons via the interplay of optical coherent population trapping and dynamic nuclear polarization

A. R. Onur, J. P. de Jong, D. O’Shea, D. Reuter, A. D. Wieck, and C. H. van der Wal

Zernike Institute for Advanced Materials, University of Groningen, 9747AG Groningen, The Netherlands
Angewandte Festkörperphysik, Ruhr-Universität Bochum, D-44780 Bochum, Germany

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We experimentally demonstrate how coherent population trapping (CPT) for donor-bound electron spins in GaAs results in autonomous feedback producing stabilized states for the spin polarization of nuclei around the electrons. CPT was realized by excitation with two lasers to a bound-exciton state. Transmission studies of the spectral CPT feature on an ensemble of electrons directly reveal the statistical distribution of prepared nuclear spin states. Tuning the laser driving from blue to red detuned drives a transition from one to two stable states.

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Following the emergence of electron spins in quantum dots and solid state defects as candidates for spin qubits, it has become a major goal to realize control over the nuclear spins in such nanostructures. In many experimental settings, interaction with disordered nuclear spins in the crystal environment is detrimental to the coherent evolution of carefully prepared electron spin states. Preparation of nuclear spins in a state that has reduced spin fluctuations with respect to the thermal equilibrium state will help to overcome this problem. Proposals to achieve this goal by electron spin resonance (ESR) have been put forward for one- and two-electron quantum dots, as well as by optical preparation techniques that either rely on a quantum measurement technique or a stochastic approach. Experimental advancements have been made in single quantum dots by ESR and optical excitation techniques, with nitrogen-vacancy-centers and with quantum dot ensembles. Several of these works make use of the optical response of the electronic system near the coherent population trapping (CPT) resonance because it is highly sensitive to perturbations from nuclear spins. Notably, these experiments have focussed on quantum dots where, due to the particular anisotropic confinement, hole-spin hyperfine coupling is dominant. In recent work, we discussed how the interplay between hyperfine coupling and CPT influences the stochastics of the nuclear spin bath for a class of systems where electron-nuclear spin interaction dominates.

Here we demonstrate an all-optical technique to stabilize the nuclear spin bath near silicon donors in gallium arsenide into a non-thermal state under conditions of two laser optical pumping. We show that the spin system is directed either towards a single stable state or (probabilistically) towards either of two stable states, depending on laser detuning from the excited state. Our results show how feedback control arises from the interplay between coherent population trapping (CPT) and dynamic nuclear spin polarization (DNP). In particular, we suggest that this interplay may be used to create stable states of nuclear polarization with potentially reduced fluctuations.

We perform measurements on the nuclear spin dynamics in an epitaxially grown film of GaAs doped with Si donors at a concentration of $3 \times 10^{13}$ cm$^{-3}$, which is well below the metal-insulator transition around $10^{16}$ cm$^{-3}$. The wafer is cleaved in 2-by-2 mm parts along the (110) crystal axes. The film is lifted off of its GaAs substrate by removing an AlAs buffer layer with a hydrofluoric acid wet-etch. After lift-off the film is transferred to a sapphire substrate, which allows us to do transmission measurements in a cryogenic microscope. Measurements are performed at a temperature of 4.2 K and a magnetic field of 5.9 T. The sample is mounted such that the magnetic field direction is along the (110) crystal axis. Light from tunable continuous wave lasers (Coherent MBR110) is delivered to the sample by polarization maintaining fibers and passes through the sample (along the (100) crystal axis). Transmitted light is collected in a multimode fiber and detected by an avalanche photodiode outside the cryostat.

The optical transitions that we address are from the bound electron spin states $|\uparrow, \downarrow\rangle$ to a level of the bound trion $|D^{0}X\rangle$, consisting of two electrons plus one hole together bound to the silicon donor. The magnetic field is applied perpendicular to the light propagation direction (Voigt geometry) such that the optical transitions have polarization selection rules discriminating between horizontally ($\sigma_{+-}$, coupling to $|\uparrow\rangle - |D^{0}X\rangle$) and vertically ($\pi$, coupling to $|\downarrow\rangle - |D^{0}X\rangle$) polarized light. These three localized states form a Λ-type energy level configuration, depicted in Fig. 1(a).

We first demonstrate CPT in our system by driving...
with two laser fields. In transmission this appears as a narrow window of increased transparency within the broader absorption dip when one laser is scanning while the other remains locked, as shown in Fig. 1(b). The optical transition frequencies are $\omega_{13}$ and $\omega_{23}$. Particularly of interest is the position of resonance where the lasers directly drive two-photon (Raman) transitions between the electron spin states. Here the lineshape of the CPT resonance reveals important information about the electron spin states. This can be obtained by fitting the curve in Fig. 1(b) to the Lindblad equation for the A-system [24]; our n-GaAs samples yield an inhomogeneous dephasing time $T_2^* \approx 3$ ns [25]. However, the homogeneous dephasing time, $T_2$, has been estimated via the spin-echo technique [26] to be approximately $7 \mu s$. The discrepancy between $T_2$ and $T_2^*$ is largely due to dephasing caused by the disordered nuclear spins.

Due to the Fermi contact hyperfine interaction, any non-zero nuclear spin polarization exerts an effective magnetic (Overhauser) field on the electron and hence causes a shift of the electron spin splitting, denoted by $\delta$ in figure Fig. 1(a). The value of $\delta$ is proportional to the nuclear spin polarization, $p \in [-1,1]$, such that $\delta = p\delta_{\text{max}}$ where $\delta_{\text{max}}$ is the maximum shift set by the hyperfine interaction strength. For the donor electron in GaAs its value is $\delta_{\text{max}} = 24.5$ GHz [27].

The thermal equilibrium properties of the nuclear spin bath are well approximated by considering $N$ non-interacting spins $I$. The polarization $p$ and its variance $\sigma_p^2$ in the high temperature limit, $h\gamma B/k_B T \ll 1$ (valid in our experimental conditions), are $p = h\gamma B(I + 1)/3k_B T \approx 0$ and $\sigma_p^2 = (I + 1)/(3I - 1)$. Because nuclear spin dynamics are slow as compared to the electron’s light interacting with the system sees a snapshot of the Overhauser shift taken from the distribution $P(\delta)$. Any measurement on an ensemble of these systems should include an average over $P(\delta)$. The CPT lineshape of Fig. 1(b) arises from the transmittance, taking into account that the susceptibility is averaged over $P(\delta)$.

$$T(\omega_i) = \exp\left(-d\rho_i\frac{\omega_i}{c} \int_{-\infty}^{+\infty} P(\delta)\chi_i''(\omega_i, \delta) d\delta\right),$$

where $d$ is the thickness of the medium, $\rho$ is the density of donors, $c$ is the speed of light, $i = 1,2$ indicates the laser fields and $\chi_i$ is the susceptibility for the laser field at fixed $\delta$ which can be calculated from the Lindblad equation and depends on other system parameters implicitly. At thermal equilibrium $P(\delta)$ is a Gaussian centered at zero with variance $\sigma_\delta^2 = \delta_{\text{max}}^2 \sigma_p^2$. For $I = 3/2$ and $N = 10^5$ this yields a FWHM of $2\sqrt{2\ln(2)}\sigma_\delta = 136$ MHz, which roughly corresponds to the width of the measured CPT. However, $P(\delta)$ can undergo changes when the electron spin is brought out of thermal equilibrium by optical orientation. An optically induced electron spin polarization will in turn induce nuclear spin polarization via a hyperfine mediated cross-relaxation process known as dynamic nuclear polarization. In Ref. [22] it was described how the interplay between the laser induced electron spin polarization near CPT resonance and dynamic nuclear polarization can change the shape of $P(\delta)$ by autonomous feedback control, leading to the formation of stable states for the nuclear spin polarization and offering the potential of reducing the variance $\sigma_\delta^2$. The essence of this method is pictured schematically in Fig. 1(c), it shows two distinct control regimes (color coded, red and blue) where both lasers are either red ($\Delta < 0$) or blue ($\Delta > 0$) detuned from the excited state. The change in laser coupling strength with $\delta$ is asymmetric when $\Delta \neq 0$ (one laser approaches resonance while the other moves away from it). For a single system with particular Overhauser shift this causes a sharp change in the optically
induced electron spin polarization, \( \langle S_z \rangle - \langle \overline{S}_z \rangle \) (where the overbar implies that the expectation value is taken at thermal equilibrium), as a function of \( \delta \) as shown in the middle panels (the Overhauser shift is expressed as fraction of the excited state decay rate \( \Gamma_3 \)). The blue and red dots indicate stable points, where \( \langle S_z \rangle = \langle \overline{S}_z \rangle \) and \( \partial / \partial \delta \left( \langle S_z \rangle - \langle \overline{S}_z \rangle \right) < 0 \). We thus expect \( P(\delta) \) to evolve from the initial Gaussian to either a distribution with two maxima, or to a distribution with one maximum. Such steady state distributions are non-thermal and can thus have reduced fluctuations if the system’s feedback response (slope of \( \langle S_z \rangle - \langle \overline{S}_z \rangle \) near the stable point) is strong enough.

We investigate this interplay between CPT and DNP for the donor bound electrons in GaAs by monitoring the changes in the CPT lineshape induced by two laser optical pumping, with both lasers at equal intensity, near two-photon resonance. Figure 2(a,b) show zoomed views of the CPT lineshape before (gray lines) and after 10 minutes of optical pumping with blue and red detuned lasers. The scanning (probe) laser effectively probes a range of \( \delta \)-values and the transmission is proportional to the number of electron spins experiencing a particular Overhauser shift \( \delta \), hence reflecting the underlying nuclear spin distribution. The nuclear spin distribution stabilizes as predicted in both cases. In Fig. 2(a) the CPT line stays centered at 0 MHz, a situation that can be contrasted to the CPT line after 10 minutes of optical pumping with a single laser on the \( |\uparrow\rangle \) − \( |D^0X\rangle \) transition (inset) which shifts the CPT line by \( \sim 400 \) MHz, indicating a net nuclear spin polarization. The lineshape remains similar, whereas if the width of \( P(\delta) \) would have been reduced an increase in transmission at 0 MHz would have been measured. In Ref. 22 it was pointed out that in an open system nuclear spin diffusion takes place which competes with the narrowing effect of the feedback mechanism. For the donor in GaAs this is of particular importance since, as opposed to a quantum dot, there is no material barrier surrounding the defect that can prevent spin diffusion. In Fig. 2(b) the other control regime, with red detuned pump lasers, shows the expected splitting of the CPT line after pumping, indicative of a doubly peaked \( P(\delta) \). Figure 2(c,d) show the transition between the two control regimes, Fig. 2(c) shows the CPT traces for a range of detunings \( \Delta \) and Fig. 2(d) shows the splitting in these traces obtained by fitting two Gaussians to each CPT line. Whenever this fit does not improve with respect to fitting a single Gaussian, we take the splitting to be identically zero. The data reproduces the essential features of the model (22, black line) showing a discontinuous transition and a maximum splitting when the pump lasers are tuned to the slope of the resonance at \( \sim -5 \) GHz, where the asymmetry in laser detuning as a function of \( \delta \) is largest. In the transition region around \( \Delta = 0 \) there is no good match, but we also detect a large error from the fitting, we attribute this to inhomogeneous broadening in the optical transitions (causing an effective spread in detunings \( \Delta \)) which prevents all systems from making the transition between the two regimes simultaneously.

We now focus on the control regime \( \Delta < 0 \) to examine the dependence of the splitting on the control parameters during the optical pumping phase. Figure 3(a) shows the sensitivity of the splitting to detuning and to laser power. Figure 3(b) shows the importance of carefully tuning the relative frequencies such that the distribution is balanced, a minor detuning of 31 MHz in one of the lasers shifts the weight of \( P(\delta) \) to either one of the stable states. Fig. 3(b) shows the power dependence when vary-
FIG. 3. (a) CPT traces after DNP pumping with two lasers with $\Delta \approx -3.5$ GHz. The trace labeled $\omega_1 = 0$ MHz is taken after pumping on exact two-photon resonance. The blue (red) trace is measured after pumping with $\omega_1 = 31$ MHz (-31 MHz) detuned from exact two-photon resonance (see Fig. 1(c)). (b) The peak splitting in CPT traces after pumping with $\Delta \approx -4$ GHz and exact two-photon resonance, as a function of the intensity of the two lasers (keeping the intensity ratio fixed near 1). The gray background shows the range where the CPT peak shape was analyzed as a single peak. Significant double-peak character was observed for the total laser intensity above $\sim 4$ Wcm$^{-2}$. CPT traces were all taken with both lasers intensities at $\approx 3$ Wcm$^{-2}$. Black line: simulation with same parameters as in Fig. 2. Top axis shows Rabi frequency corresponding to the simulation.

In Figure 4 the time evolution of the effect is shown. The peak splitting in CPT traces after DNP pumping ($\Delta = -4$ GHz) was obtained from CPT scans of 1 s in between periods of 30 s DNP pumping with two lasers fixed at two-photon resonance ($\Delta = -4$ GHz). Panel (b) presents CPT scans of 1 s, taken after a fully dark period of 1 s (top trace) to 10 s (bottom trace) after DNP pumping ($\Delta = -4$ GHz). The results in (c) are from continuously taking CPT scans of 1 s (only four traces shown), after a DNP pumping period at $\Delta = -2$ GHz. All data was taken with both laser intensities stabilized at values of about 3 Wcm$^{-2}$ for DNP pumping and CPT probing.

FIG. 4. Time evolution of the nuclear spin stabilization during (a) and after (b, c) the DNP pumping period (subsequent traces top to bottom, as labeled). Panel (b) gives a reference for the CPT peak before pumping. The data in (a) is obtained from CPT scans of 1 s in between periods of 30 s DNP pumping with two lasers fixed at two-photon resonance ($\Delta = -4$ GHz). Panel (b) presents CPT scans of 1 s, taken after a fully dark period of 1 s (top trace) to 10 s (bottom trace) after DNP pumping ($\Delta = -4$ GHz). The results in (c) are from continuously taking CPT scans of 1 s (only four traces shown), after a DNP pumping period at $\Delta = -2$ GHz. All data was taken with both laser intensities stabilized at values of about 3 Wcm$^{-2}$ for DNP pumping and CPT probing.

In Figure 4 the time evolution of the effect is shown. Figure 4(a) shows the build up of the splitting, obtained by taking CPT traces during the optical pumping phase every 30 seconds (each CPT trace is collected within 1 seconds). The splitting stabilizes after approximately 4 minutes. Figure 4(b) shows the decay of the splitting, it consists of CPT traces collected after the optical pumping phase. After 10 minutes of optical pumping (repeated before each trace) the system is kept in the dark for a time $t$, where $t$ ranges from 1 to 10 seconds. The splitting fades away in seconds, consistent with the relaxation of the lattice nuclear spins by spin diffusion, which occurs on the timescale of seconds for this system [28]. When CPT scans are taken continuously after the optical pumping phase the splitting is seen to decay slowly and at least persist up to 20 minutes (Fig. 4(c)). We attribute this to a suppression of the spin diffusion while the system is illuminated: under optical excitation (so during CPT scans) the electron spin is significantly polarized and it has been demonstrated that spin polarization of the electron suppresses spin diffusion because it creates an inhomogeneous Knight field for the surrounding nuclear spins [29, 30]. In fact we propose that this could even be used to improve the strength of the feedback control to possibly observe narrowing. If the temperature of the experiment would be lower or the magnetic field stronger (such that $\langle S_z \rangle$ is increased) the thermal equilibrium electron spin polarization can suppress nuclear spin diffusion, ef-
fectively protecting the system from spin diffusion to the environment (outside the range of the electron) nuclear spins.

These results open the possibility to use the interplay between CPT and DNP to operate a mesoscopic spin system as a feedback loop that converges towards a well defined steady state, determined by applied laser power and detuning, with the possibility of achieving reduced nuclear spin fluctuations and hence providing protection to electron spin dephasing. The mechanism is generally applicable to localized spin systems where DNP is dominated by electron-nuclear spin hyperfine coupling and can also be used in other paramagnetic defects, in ensemble configuration as well as for single electron spins. As a notable example we suggest using this technique on the fluorine donor in ZnSe [31, 32]. In a II-VI material in which the lattice nuclear spins are more dilute (in GaAs every lattice site carries a nett nuclear spin) spin diffusion, mediated by nuclear dipole-dipole interaction, which is inversely proportional to distance between non-zero nuclear spins to the power 6, will be much less prominent.

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