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Nuclear spin cooling using Overhauser field selective coherent population trapping

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Hyperfine interactions with a nuclear spin environment fundamentally limit the coherence properties of confined electron spins in the solid-state. Here, we show that a quantum interference effect in optical absorption from two electronic spin states of a solid-state emitter can be used to prepare the surrounding environment of nuclear spins in well-defined states, thereby suppressing electronic spin dephasing. The evolution of the coupled electron-nuclei system into a coherent population trapping state by optical excitation induced nuclear spin diffusion can be described in terms of Lévy flights, in close analogy with sub-recoil laser cooling of atoms. The large difference in electronic and nuclear time scales simultaneously allow for a measurement of the magnetic field produced by nuclear spins, making it possible to turn the lasers that cause the anomalous spin diffusion process off when the strength of the resonance fluorescence reveals that the nuclear spins are in the desired narrow state.

The phenomenon of coherent population trapping (CPT) in three-level emitters [1] is at the heart of a number of key advances in quantum optics, such as sub-recoil cooling of atoms [2] and slow-light propagation [3–5]. In these experiments, optical excitation from two low energy (spin) states to a common optically excited state vanishes due to a quantum interference effect, leading to the formation of a *dark resonance* whenever the two driving laser fields satisfy the two-photon resonance condition. The fundamental limit on how well quantum interference eliminates optical absorption is provided by the decoherence rate of the two low-energy spin states. Typically, this decoherence rate is assumed to be induced by a reservoir which could be treated using the usual Born-Markov approximation, implying that the reservoir has a short correlation time and its density operator is not influenced by the interactions.

Unlike their atomic counterparts, solid-state spins are in general subject to non-Markovian dephasing [6–8] due to their coupling to reservoirs with long correlation times. In particular, hyperfine coupling to nuclear spins constitutes the most important source of decoherence for spin qubits. It has been proposed that polarizing or cooling nuclear spins could alleviate this decoherence process [6], which prompted theoretical [9, 10] as well as experimental efforts aimed at narrowing down the Overhauser field distribution [11–13]. These schemes could be considered as a form of *reservoir engineering*; remarkably, recent experiments showed that the substantial manipulation of the nuclear spins (reservoir) could be achieved by using the electron spin (system) itself [12–17].

In this Letter, we show that CPT in the spin states of a solid-state emitter could be used to prepare a nuclear spin environment in states with a near-deterministic Overhauser field. The preparation of an ultra-narrow nu-

clear spin distribution is achieved by optical excitation induced anomalous diffusion processes [2]. As a consequence of the anomalous diffusion, the coupled electron-nuclei system dynamically switches back and forth between a trapped regime where nuclear spin diffusion slows down drastically due to the formation of a dark state, and a non-trapped regime where optical excitation leads to fast diffusion [18]. We find that for a range of system parameters, the coupled system finds the dark state via this diffusion process and then remains trapped in it for long times, ensuring a narrow nuclear spin distribution with a standard deviation that is close to the single-spin limit. An additional remarkable feature of the scheme that we analyze is the possibility of using resonantly scattered photons to measure which regime the coupled system is in at a given time [19]; turning the laser fields off after determining the coupled system to be in the dark state can then be used to further narrow down the Overhauser field distribution to the sub-single-spin regime. Such a feedback mechanism is enabled by the large difference between the time scales corresponding to electronic light scattering and nuclear spin-flip processes.

Nuclear-spin selective coherent population trapping

We consider a solid-state emitter where the two ground electronic spin states, denoted by $|\uparrow_x\rangle$ and $|\downarrow_x\rangle$, are coupled by two laser fields to a common optically excited state $|t\rangle$ (Fig. 1a). The laser field with frequency ω_p (ω_c) that couples the $|\uparrow_x\rangle - |t\rangle$ ($|\downarrow_x\rangle - |t\rangle$) transition with Rabi frequency Ω_p (Ω_c) is referred to as the probe (coupling) field. The state $|t\rangle$ decays in turn via spontaneous emission back to the two ground spin states with an equal rate $\Gamma_{t\uparrow} = \Gamma_{t\downarrow} = \Gamma/2$. Denoting the Zeeman energy of the electron spin due to the external field B_x with ω_x and the energy of the optically excited state with ω_t , we express the bare optical detunings relevant for the CPT system as $\Delta\omega_p = \omega_t - \omega_p$ and $\Delta\omega_c = \omega_t - \omega_x - \omega_c$. In the absence of any spin interactions or decoherence,

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laser fields satisfying the two-photon resonance condition ($\delta = \Delta\omega_p - \Delta\omega_c = 0$) pump the electron spin into the dark state $|D\rangle = \frac{\Omega_c}{\sqrt{\Omega_p^2 + \Omega_c^2}} |\uparrow_x\rangle - \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_c^2}} |\downarrow_x\rangle$, which is decoupled from optical excitation. When $\Delta\omega_c = 0$ and $\Omega_p, \Omega_c \ll \Gamma$, the absorption lineshape of the emitter appears as a Lorentzian with a quantum interference induced transparency dip in the center, with a width $\delta\nu_{\text{trans}} \sim (\Omega_p^2 + \Omega_c^2)/\Gamma \ll \Gamma$.

In practice, the electronic spin states of most solid-state emitters are mutually coupled via hyperfine interaction with a nuclear spin ensemble consisting of N nuclei

$$H_{\text{hyp}} = g \sum_i^N g_i \left(I_x^i \sigma_x + \frac{1}{2} (I_+^i \sigma_- + I_-^i \sigma_+) \right). \quad (1)$$

Here, g_i defines the normalized hyperfine coupling constant between the emitter electron and the i^{th} nucleus ($\sum g_i^2 = 1$). In this convention $g = A_H / \sum g_i$ quantifies the collective hyperfine coupling strength, with A_H denoting the hyperfine interaction constant of the material. σ_k and I_α^i ($\alpha = +, -, x$) are the electronic and nuclear spin operators, respectively; $\sigma_+ = |\uparrow_x\rangle \langle \downarrow_x|$.

Our analysis of CPT in the presence of hyperfine interactions with a nuclear spin reservoir starts with the master equation, obtained by eliminating the radiation field reservoir using a Born-Markov approximation:

$$\dot{\rho} = \frac{\Gamma}{2} (\mathbb{1}_S \otimes \rho_{tt} - \{ |t\rangle \langle t|, \rho \}_{+}) - i[H_0 + H_{\text{laser}} + H_{\text{hyp}}, \rho], \quad (2)$$

where $\rho_{tt} = \langle t|\rho|t\rangle$ acts on the Hilbert space of nuclear spins and $\mathbb{1}_S = |\uparrow_x\rangle \langle \uparrow_x| + |\downarrow_x\rangle \langle \downarrow_x|$. We assume that in the absence of optical excitation, the electron spin is well isolated from all reservoirs other than the nuclear spins [20], and spin-flip co-tunneling or phonon emission rates are negligible within the timescales of interest.

In the limit of a large external field ($\omega_x \gg g$), the direct electron-nuclei flip-flop processes $I_+ \sigma_- + I_- \sigma_+$ (collective spin operators are defined as $I_\alpha = \sum_i g_i I_\alpha^i$) are strongly suppressed due to the large mismatch in the electronic and nuclear Zeeman splitting. In contrast, optical excitation does allow for energy conservation in an optically assisted electron-nuclear spin-flip process. We take the higher order processes into account by applying a Schrieffer-Wolff transformation to eliminate the direct hyperfine flip-flop interaction. The master equation then reads

$$\begin{aligned} \dot{\rho} &= \frac{\Gamma}{2} (\mathbb{1}_S \otimes \rho_{tt} - \{ |t\rangle \langle t|, \rho \}_{+}) - i[H_0 + H_{\text{laser}} + \tilde{H}_{\text{hyp}}, \rho] \\ &\quad + \epsilon^2 \frac{\Gamma}{4} \mathbb{1}_S \otimes D(\rho_{tt}) \\ &= \mathcal{L}_0(\rho) + \epsilon^2 \mathcal{L}_1(\rho_{tt}), \end{aligned} \quad (3)$$

where the new term containing

$$D(\rho) = I_+ \rho I_- + I_- \rho I_+ - \frac{1}{2} \{ I_+ I_- + I_- I_+, \rho \}_{+} \quad (4)$$

describes an optically induced random nuclear diffusion process caused by the optically assisted hyperfine flip-flop processes, that are lowest order in the parameter $\epsilon = g/(2\omega_x)$. In Eq. (3) we have neglected terms $\propto \epsilon^2$ that only affect the electron evolution [21].

After the Schrieffer-Wolff transformation the Hamiltonian relevant for electron spin dynamics (to highest order in ϵ) is $\tilde{H}_{\text{spin}} = \tilde{H}_{\text{hyp}} + \delta\sigma_x = g\sigma_x(I_x + \epsilon I_+ I_- + \delta/g)$. The electron experiences an effective magnetic field, which is composed of the two-photon detuning δ , as well as a contribution originating from the nuclei, which we refer to as the generalized Overhauser field $\tilde{I}_x = I_x + \epsilon I_+ I_-$. For a given laser detuning δ each eigenvector of the generalized Overhauser field $\tilde{I}_x |\lambda\rangle = \lambda |\lambda\rangle$ corresponds to a steady state $\rho_\lambda = \rho^e(\lambda) \otimes |\lambda\rangle \langle \lambda|$ of the unperturbed evolution $\mathcal{L}_0(\rho_\lambda) = 0$. Here, $\rho^e(\lambda)$ is given as the solution of the optical Bloch equations (OBE) found after projection of the unperturbed master equation on the respective nuclear state $|\lambda\rangle$; in the OBE $\delta_{\text{eff}} = g\lambda + \delta$ gives the effective two-photon detuning that determines the CPT condition. The lifetime of such quasi-steady states ρ_λ under the full dynamics of Eq. (3) is determined by hyperfine assisted scattering events, which are described by the term \mathcal{L}_1 . The corresponding nuclear spin flip rate in positive (negative) direction D^+ (D^-) can be directly deduced from Eq. (3): $D^\pm = \epsilon^2 \frac{\Gamma}{2} \rho_{tt}^e(\lambda) \langle I_\mp I_\pm \rangle$, where $\rho_{tt}^e(\lambda) = \langle t | \rho^e(\lambda) | t \rangle$ is the population in state $|t\rangle$. Each nuclear spin flip event of this kind changes $\langle \tilde{I}_x \rangle$ by a value of order g_i .

For nuclear states with $g\lambda = -\delta$ the system is in two-photon resonance and the electronic system is transparent such that $\rho_{tt}^e = 0$: as a consequence, the nuclear spin diffusion vanishes and the system is trapped in a dark state. Since the generalized Overhauser field in an electronic-nuclear dark state is locked to a fixed value, its variance will be strongly reduced (nuclear state narrowing) suppressing hyperfine-induced electron spin decoherence. Strikingly, by narrowing the generalized Overhauser field, even electron-mediated nuclear spin diffusion is suppressed, thus eliminating the second order contribution to hyperfine-induced electron spin decoherence as well. For all nuclear states satisfying $g\lambda \approx -\delta$, the excited electronic state population will remain small ($\rho_{tt}^e \propto \delta_{\text{eff}}^2$), ensuring that the spin diffusion rate will remain vanishingly small: we refer to this subspace as the trapping region.

In contrast, nuclear states with $g\lambda \not\approx -\delta$ render the electron optically active and the generalized Overhauser field experiences random diffusion (recycling region). To illustrate the dynamics allowing the nuclei to move from the recycling to the trapping region, we consider an electron that is optically excited to state $|t\rangle$: as it decays, it could induce a nuclear spin flip event (with probability $\sim \epsilon^2$) in either direction. Through successive spin-flip events, the nuclear reservoir probes different spin configurations with distinct generalized Overhauser shifts. When the diffusion allows the nuclei to reach a configuration that yields $\delta_{\text{eff}} \approx 0$, the electron becomes trapped

in the dark state; further optical excitation is then inhibited and nuclear spin flips are strongly suppressed.

Owing to the quasi-continuous nature of the generalized Overhauser field spectrum, the dark-state condition $\delta_{\text{eff}} = 0$ can be satisfied for a wide range of initial detunings $\Delta\omega_p$. This leads to a drastic change in the CPT signature in absorption spectroscopy: instead of exhibiting a narrow transparency dip at (bare) two-photon-resonance ($\delta = 0$), the coupled electron-nuclei system displays a broad transparency window.

The operator valued correction \tilde{I}_x to the two-photon detuning δ and the optically induced diffusive dynamics of \tilde{I}_x described by the second line of Eq. (3) are at the heart of the nuclear-spin cooling scheme we analyze in this work. The predictions we outlined hold in general for any nuclear operator \tilde{I}_x with a sufficiently large density of states around $g\lambda + \delta = 0$. In the Methods we show that this requirement is fulfilled for the generalized Overhauser field $\tilde{I}_x = I_x + \epsilon I_+ I_-$ and that its properties are very similar to those of I_x for the parameters we consider. Therefore, for the sake of simplicity, we will proceed by neglecting the ϵ correction. As a further simplification we will constrain our analysis to nuclear spin 1/2 systems. While our results apply to a broad class of solid-state emitters, ranging from various types of quantum dots to NV centers, we will focus primarily on a single electron charged quantum dot (QD) where the optically excited state is a trion state consisting of an electron singlet and a valence-band hole (Fig. 1a) [19, 22, 23]. For most QD systems, the assumptions we stated earlier are realized in Voigt geometry where B_x is applied perpendicular to the growth direction.

Semiclassical analysis

We first consider the semiclassical limit to numerically confirm the principal striking features of the coupled electron-nuclei system – altered CPT signatures and the drastic nuclear state narrowing – for inhomogeneous electron-nuclear coupling. To obtain a semiclassical description of the coupled electron-nuclei dynamics, we start by assuming that the electron (ρ^e) and the nuclear (ρ^n) spins remain unentangled throughout the system evolution ($\rho = \rho^e \otimes \rho^n$). Since, as discussed earlier, the electron dynamics takes place on a timescale that is faster by a factor $\epsilon^{-2} \gg 1$ than the nuclear dynamics, it is justified to solve the OBE in steady state to determine the trion population ρ_{tt}^e for a given nuclear spin configuration (and the associated effective magnetic field).

To describe the nuclear spin dynamics semiclassically, we assume that the nuclear density operator ρ^n is diagonal in the basis of individual nuclear spin eigenstates. This assumption is justified for QDs in which either strongly inhomogeneous hyperfine coupling or inhomogeneous quadrupolar fields lead to large variations in the splitting of the nuclear spin states; when this is the case, the nuclear superposition states will effectively dephase, justifying the assumption of a diagonal density operator. In this limit, the master equation Eq. (3) reduces to rate

equations which can be numerically solved using Monte Carlo techniques (see Methods).

Figure 2 shows the result of the Monte Carlo simulations of the coupled electron-nuclei evolution. To obtain the probe field absorption lineshape as well as the Overhauser field variance, we assume that for each probe field detuning, we start out from a completely mixed ρ^n , take $\Delta\omega_c = 0$ and evolve the coupled system to its steady state for a range of probe laser detunings. We find that the transparency window that has a width of $\sim 0.12\Gamma$ ($\sim 0.48\Gamma$) for $\Omega_c = \Omega_p = 0.2\Gamma$ ($\Omega_c = \Omega_p = 0.4\Gamma$) in the absence of hyperfine coupling (Fig. 2a, red dashed curve) is drastically broadened and assumes a width $\delta\nu_{\text{trans}} > \Gamma$ (Fig. 2a, solid curves). This *dragging of the dark resonance* effect is in contrast to Faraday geometry experiments where nuclear spin polarization ensures that the applied laser field remains locked to a detuning that ensures maximal absorption [13]. Concurrently, the Overhauser field distribution is narrowed dramatically from its value in the absence of optical excitation (Fig. 2b, black dashed line) such that its standard deviation σ_{OF} is smaller than the change induced by flipping one nuclear spin of the most weakly coupled class (Fig. 2b, solid curves). These simulations show all the striking features that are a consequence of the optically induced nuclear spin diffusion [second line of Eq. (3)] which leads to a uni-directional evolution into the electronic-nuclear dark state $\rho_D = |D\rangle\langle D| \otimes \rho_D^n$, where ρ_D^n is a nuclear spin density operator that yields $\delta_{\text{eff}} = 0$.

We remark that the narrowing of the Overhauser field distribution could be measured by using the same two-laser set-up and scanning the probe laser on timescales short compared to those required to polarize the nuclear spins, thanks to the large separation between the electronic and nuclear dynamical timescales. Figure 2c shows the simulation of the absorption lineshape obtained by such probe laser scans. Starting out with 100 random nuclear spin configurations, we first let the coupled electron-nuclei system evolve to its steady-state under initial laser detunings $\Delta\omega_c = 0$ and $\Delta\omega_p = -0.2\Gamma$. We then scan the probe laser in either direction to obtain the absorption lineshape (solid blue curve) that directly reveals information about the narrowing of the Overhauser field distribution. If we repeat the numerical experiment by assuming that the laser fields were initially completely off resonance, we find that the absorption lineshape is nearly Lorentzian (dashed green curve).

Quantum model of the nuclear spin dynamics

Next, we study the homogeneous coupling limit using a full quantum treatment. To capture the full quantum dynamics, we derive a master equation which depends only on nuclear degrees of freedom, allowing for both an analytical steady state solution and the comparison between the quantum and the semiclassical limit. To this end, we assume the homogeneous limit ($g_i = 1/\sqrt{N}$). First, we eliminate the state $|t\rangle$ in the limit $\Omega_p, \Omega_c \ll \Gamma$, giving a master equation involving the nuclear and

electronic spins only. We also assume $\Omega_c = \Omega_p = \Omega$, which ensures that the relevant electron spin states in the rotating frame are $|D\rangle = |\downarrow_z\rangle$ and $|B\rangle = |\uparrow_z\rangle$, and choose $\delta = 0$, for simplicity. In the interaction picture we then obtain from Eq. (3) the reduced master equation

$$\begin{aligned} \dot{\rho} = & \Gamma_{\text{eff}}(\sigma_-^z \rho \sigma_+^z - \frac{1}{2}\{\sigma_+^z \sigma_-^z, \rho\}_+) \\ & + \frac{\Gamma_{\text{eff}}}{2}[\sigma_z, [\sigma_z, \rho]] - igI_x[\sigma_x, \rho] \\ & + \epsilon^2 \mathbb{1}_S \otimes D(\Gamma_{\text{eff}} \rho_{\uparrow_z \uparrow_z}), \end{aligned} \quad (5)$$

where σ_{\pm}^z are the electron spin matrices in the z -basis and $\Gamma_{\text{eff}} = \frac{\Omega^2}{(\Gamma/2)^2 + (gI_x/2)^2} \frac{\Gamma}{2}$ is an operator valued effective (electron) spin decay rate. The last line of Eq. (5) describes the nuclear spin diffusion determined by the nuclear operator $\rho_{\uparrow_z \uparrow_z} = \langle \uparrow_z | \rho | \uparrow_z \rangle$, cf. Eq. (4) [25].

In order to eliminate the electronic degrees of freedom from Eq. (5) we once again make use of the fact that on the timescales of the electron evolution, the nuclear field can be considered as quasi-static and hence the electron settles quickly (on nuclear timescales) to its interim steady state. We find that on this coarse grained timescale $\rho_{\uparrow_z \uparrow_z} = \frac{1}{2}[1 - (\frac{\Gamma_{\text{eff}}}{|\Delta_{\text{eff}}|})^2] \rho_n$, with $|\Delta_{\text{eff}}|^2 = \Gamma_{\text{eff}}^2 + (gI_x)^2$. Using this relation, the electron spin can be eliminated from Eq. (5), yielding

$$\dot{\rho}^n = \text{Tr}_S(\dot{\rho}) = D(\Gamma_{\text{nuc}} \rho^n), \quad (6)$$

where we defined the nuclear spin flip rate $\Gamma_{\text{nuc}} = \epsilon^2[1 - (\frac{\Gamma_{\text{eff}}}{|\Delta_{\text{eff}}|})^2] \Gamma_{\text{eff}}$. Note that since Γ_{nuc} vanishes for all zero eigenstates of I_x , Eq. (6) implies, in accordance with the considerations above, that every state in the kernel of the collective nuclear spin operator I_x – i.e. a state of vanishing Overhauser field – is a steady state of the dynamics. We plot the nuclear spin flip rate in Fig. 3a.

Equation (6) can be used to directly compare the quantum mechanical and semiclassical diffusion rates in the homogeneous limit (see Methods). Surprisingly, the two opposite regimes of semiclassical and quantum mechanical description show both qualitative (evolution can be fully characterized by rate equations) and quantitative (for the relevant states the calculated rates are comparable) agreement (cf. Fig. 3a); this result is particularly interesting since we would expect the semiclassical description to fail in the homogeneous limit.

In order to calculate the achievable Overhauser field standard deviation σ_{OF} we numerically compute the exact steady state solution of master equation (2) for homogeneously coupled nuclei. To this end, we explicitly consider all orders of the hyperfine interaction including processes that result in a (small) finite decay rate out of the dark state [26]. Figure 3b shows σ_{OF} as a function of Ω , where we find that σ_{OF} decreases with decreasing Ω until it reaches a minimum of $\sigma_{OF} \simeq 2$ ($\sigma_{OF} \simeq 0.7$) for $\Omega \simeq 0.2\Gamma$ and an electron spin decoherence rate of $T_2^{-1} = 100s^{-1}$ ($T_2^{-1} = 0$). This result can be understood by recalling that the width of the transparency dip

in CPT scales as Ω^2/Γ , implying that the range of Overhauser field values yielding transparency can be narrowed simply by reducing Ω . For $\Omega < 0.2\Gamma$, we find that σ_{OF} increases rapidly; for such small values of Ω , the coupled electron-nuclei system spends substantial amount of time outside the narrow transparency region, leading to the observed increase in steady state value of σ_{OF} . As we will argue below, this increase does not constitute a fundamental limitation for the attainable narrowing and values $\sigma_{OF} < 1$ are possible by using a feedback mechanism. Clearly though, such a remarkable level of narrowing could only be observed if it is achieved on timescales short compared to those imposed by electron spin decoherence and optical excitation independent nuclear spin decay processes. We now turn to the question *how quickly* the nuclear spins reach this narrowed state.

Evolution of the nuclear spins as Lévy flights

There is close analogy between the problem of CPT in the presence of inhomogeneous hyperfine interactions with a slow nuclear spin ensemble and that of a one-dimensional velocity selective CPT [2, 18]; the role of atomic momentum in the latter case is assumed by the nuclear Overhauser field I_x in the present problem. Just like the atomic momentum along the direction of interest could change by any value up to the full recoil momentum upon light scattering, the Overhauser field could change by any value, thanks to an inhomogeneous distribution of hyperfine interaction constants $0 \leq g_i \leq g_i^{\text{max}}$. The two models differ in two important aspects: first, there is a maximum value of the Overhauser field $\langle gI_x \rangle = A_H$ given by full polarization of the nuclei, and second, only a small fraction $\epsilon^2 \ll 1$ of light scattering events give rise to a change in the nuclear spin configuration.

It is known from the velocity selective CPT problem that the timescales for sub-recoil cooling of the atomic momentum distribution could be understood using Lévy flight analysis [18]. We apply this method to determine the timescale over which we expect the nuclear spins to reach a configuration with a mean Overhauser field that is smaller than a prescribed value (trapping region). Consider the random walk of the Overhauser field in time for $\Delta\omega_p = \Delta\omega_c = 0$, $\Omega_p = \Omega_c = \Omega$: this random walk is characterized by periods of diffusion followed by long intervals where the Overhauser field is restricted (trapped) at a value close to $\langle I_x \rangle = 0$. The duration of the longest trapping interval is typically on the order of the interaction time – a signature of Lévy statistics. The probability distribution functions $P(t)$ for the trapping time t , and $\hat{P}(\hat{t})$ for the recycling time \hat{t} for which the Overhauser field diffuses within the recycling region before returning to the trap characterize the Lévy flights [18].

We are mainly interested in the time required for an Overhauser field initially in the recycling region to diffuse to a value that is within a prescribed interval that defines the trapping region. Once again, this simplification is a consequence of the fact that a drop in scattered light intensity could reveal whether or not the nuclear spin distribution has the prescribed value on timescales smaller

by ϵ^2 than those needed to flip another nuclear spin. A feedback mechanism could therefore ensure that laser excitation is turned off and the desired/attained σ_{OF} is preserved. To simplify the estimation of the trapping time, we consider a limiting case where $\Omega^2/\Gamma < A_H/N$; i.e. a typical single nuclear spin flip will take the system out of the transparency window.

Around the transparency point, the rate at which nuclear spins flip is given by $D^\pm \propto \langle I_x \rangle^2$. This dependence yields $P(t) \propto t^{-\frac{3}{2}}$, which in turn leads to infinite average trapping times [18]. If we assume that the width of the recycling region is determined by $\frac{A_H}{\sqrt{N}} \approx \frac{\Gamma}{4}$, then the light scattering rate outside the transparency window could be taken to be constant with value Ω^2/Γ [27]. The nuclear spin flip rate in the recycling region is then given by $\tau_0^{-1} \approx \epsilon^2 \Omega^2/\Gamma$.

In this simplified model the random walk is confined and unbiased so in the limit of many nuclear spin flips, the number of steps required to return to the trap is given by $\langle M \rangle = \frac{A_H/\sqrt{N}}{\Omega^2/\Gamma} \approx \frac{A_H/\sqrt{N}}{A_H/N} = \sqrt{N}$. Since the time for a single spin flip is taken to be independent of the Overhauser field, the time to return to the trap is given by

$$\langle \hat{t} \rangle = \langle M \rangle \tau_0 = \frac{A_H/\sqrt{N}}{\Omega^2/\Gamma} \frac{\Gamma}{\Omega^2} \frac{1}{\epsilon^2} \approx \frac{N^{3/2}}{A_H \epsilon^2}. \quad (7)$$

For $\omega_x \approx A_H$ this expression simplifies to $\langle \hat{t} \rangle \approx N^{5/2}/A_H$.

Given the strong N dependence of $\langle \hat{t} \rangle$ corresponding to the timescale needed to establish $\sigma_{OF} \sim 1$, it is important to consider nuclear spin dynamics arising from optical-excitation-independent nuclear spin diffusion or decay processes, as well as the electron spin decoherence. The ultimate limit for the latter is due to spin-orbit mediated spin-flip phonon emission with a rate $\sim 10^{-7}\Gamma$ for $\omega_x \approx A_H$ [28]; as seen in Fig. 3b the resulting increase in σ_{OF} is a factor ~ 3 as compared to the case with no electron spin decay. Physical processes leading to nuclear spin diffusion in the dark state include (a) nuclear spin diffusion mediated by exchange coupling of the QD electron spin to a degenerate electron gas or by phonon emission/absorption [8], (b) electric field fluctuations in the QD environment leading to spatial shifts in the electron wave-function, (c) nuclear quadrupolar fields with axes not parallel to B_x . If we denote the optical excitation independent single nuclear spin diffusion rate that can arise from any of these mechanisms with γ_n and assume that $N\gamma_n \ll \Omega^2/\Gamma$, then we could write the steady-state standard deviation of the Overhauser field as

$$\sigma_{OF} \simeq \tilde{\delta} \frac{\langle t \rangle}{\langle t \rangle + \langle \hat{t} \rangle} + \frac{A_H}{\sqrt{N}} \frac{\langle \hat{t} \rangle}{\langle t \rangle + \langle \hat{t} \rangle} \quad (8)$$

where the average time spent in the trapping region $\langle t \rangle = (N\gamma_n)^{-1}$ and the effective width of the trap $\tilde{\delta} = \epsilon^{-1}\Omega\sqrt{N\gamma_n/\Gamma}$. The optimal condition for measurement independent Overhauser field narrowing is obtained when the two contributions to σ_{OF} are comparable. As

we have argued earlier, the use of feedback from the resonance fluorescence intensity should allow for reaching $\sigma_{OF} \sim \tilde{\delta}$.

Prospects for experimental realization

We have seen that the strength γ_n of optical excitation independent nuclear spin diffusion processes determines the degree of attainable Overhauser field narrowing. In this context, we remark that experimental observations reported by the Bayer group [29], obtained by driving an ensemble of single-electron charged QDs using periodic ultra-short optical pulses in the Voigt geometry, demonstrated that optically prepared nuclear spin states could survive for ~ 10 minutes [30]. Such long nuclear spin lifetimes in principle allow for reaching $\sigma_{OF} \sim 1$ using the proposed CPT scheme. We also note that the basic signatures of CPT have been observed in both single electron [22] and hole [23] charged QDs.

Even though we have concentrated on nuclear spin diffusion associated with the ground-state hyperfine coupling, the conclusions of our work remain unchanged if the solid-state emitter has hyperfine coupling leading to nuclear spin diffusion in the optically excited state. This would be the case for example in QDs with vanishing heavy-light hole mixing leading to near-resonant hole-mediated nuclear spin-flips in the excited state due to the dominant $S_z^h I_z$ term in the hole-hyperfine interaction Hamiltonian.

While prior experimental results on pulsed excitation of an ensemble of QDs strongly suggest the feasibility of our proposal in self-assembled QDs, we expect our findings to be relevant for a wider range of solid-state emitters. Of particular interest is nitrogen-vacancy (NV) centers in diamond where CPT has also been previously observed [31]. The small number of nuclear spins coupled to the optically excited spin in the case of NV-centers should make it possible to reduce the time needed for the system to find the dark state drastically. A principal difference with respect to the large N limit we analyzed is the fact that only a small set of optical detunings will allow the NV system to find a dark state. Finally, extensions to other solid state systems such as superconducting qubits may be possible [32].

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METHODS

Hamiltonian of the laser driven coupled electron-nuclei system.

The Hamiltonian of the solid-state CPT system we

consider is $H_{CPT} = H_0 + H_{\text{laser}} + H_{\text{hyp}}$ with

$$H_0 = \omega_x \sigma_{\downarrow\downarrow} + \omega_t \sigma_{tt}, \quad (9)$$

$$H_{\text{laser}} = [\Omega_p \sigma_{t\uparrow} e^{-i\omega_p t} + \Omega_c \sigma_{t\downarrow} e^{-i\omega_c t} + h.c.]. \quad (10)$$

Here, \uparrow (\downarrow) denotes \uparrow_x (\downarrow_x). The definition of H_{hyp} as well as the quantities appearing in H_0 and H_{laser} are given in the main text.

Rate equation description of nuclear spin dynamics.

The semiclassical limit can be derived from the master equation by replacing the collective spin decay by independent decay of individual spins. This is readily accomplished by making the substitution $I_+ \rho^n I_- = \sum_{ij} g_i g_j I_+^i \rho^n I_+^j \rightarrow \sum_i g_i^2 I_+^i \rho^n I_+^i$ [and correspondingly for other terms in Eq.(4)]. We coarse grain the nuclear motion with regard to the electron dynamics and from the new master equation we obtain a rate equation. We introduce a shell model of the QD with M different classes of nuclear spins (Fig. 1b); the nuclei in class (ν) have identical g_ν and their net spin polarization is $m_\nu = \frac{1}{2}(N_\nu^+ - N_\nu^-) = \langle \sum_{i \in \nu} I_x^i \rangle$, where N_ν^+ (N_ν^-) denote the total number of up (down) spins in class (ν). The derived rate equation for the joint probabilities $\mathcal{P}(\{m_\mu\})$ associated with the nuclear spin configuration $\{m_\mu\}$ is given by

$$\begin{aligned} \frac{\partial \mathcal{P}(\{m_\mu\})}{\partial t} &= \sum_{\nu}^M \mathcal{P}(\{\tilde{m}_\mu\}) N_\nu^- (\{\tilde{m}_\mu\}) \Gamma_+^\nu (\{\tilde{m}_\mu\}) \\ &+ \sum_{\nu}^M \mathcal{P}(\{\tilde{m}_\mu\}) N_\nu^+ (\{\tilde{m}_\mu\}) \Gamma_-^\nu (\{\tilde{m}_\mu\}) \\ &- \sum_{\nu}^M \mathcal{P}(\{m_\mu\}) [N_\nu^- \Gamma_+^\nu (\{m_\mu\}) + N_\nu^+ \Gamma_-^\nu (\{m_\mu\})] \end{aligned}$$

where $\Gamma_\pm^\nu (\{m_\mu\}) = (\frac{gg_\nu}{4\omega_x})^2 \frac{\Gamma}{2} \rho_{tt} (\{m_\mu\})$ are the rates at which nuclear spins of the ν th class are flipped if the nuclear spin polarizations in each class are given by $\{m_\mu\}$. $\{\tilde{m}_\mu\}$ ($\{\tilde{m}_\mu\}$) denotes the nuclear spin configuration that differs from the configuration $\{m_\mu\}$ only in the ν th class, with polarization $m_\nu - 1$ ($m_\nu + 1$).

We numerically simulate the evolution of the nuclear spins with a Monte Carlo method. We assume in our numerical simulations that the QD contains 100 nuclear spins. We group these spins into five concentric shells ($M = 5$) with different hyperfine coupling constants that are determined by the 3D Gaussian electronic envelope function (Fig. 1b). The coupling constants gg_i for these shells are 0.0934 Γ , 0.0828 Γ , 0.0678 Γ , 0.0513 Γ , 0.0358 Γ and the corresponding total numbers of nuclear spins in each shell are 2, 8, 16, 28, 46. The coupling constants

are chosen to ensure that the standard deviation of the Overhauser field seen by the QD electron for nuclei in a completely mixed state satisfies $\sigma_{OF}(\rho) = \frac{\Gamma}{4}$. We do not keep track of the exact configuration within each class (ν) of nuclear spins and assume that any configuration of spins leading to the same m_μ is equally likely and that the nuclear spin distribution in each shell is independent of the other shells.

Nuclear spin flip rates for homogeneous hyperfine coupling

For any eigenstate $|m\rangle$ of I_x with $I_x |m\rangle = m/\sqrt{N} |m\rangle$ ($\Gamma_{\text{nuc}} |m\rangle = \Gamma_{\text{nuc}}^m |m\rangle$) the nuclear spin flip rate in negative (positive) direction is given by $D^- = \langle I_+ I_- \Gamma_{\text{nuc}} \rangle_m$ ($D^+ = \langle I_- I_+ \Gamma_{\text{nuc}} \rangle_m$). In the semiclassical limit under the assumption $\langle I_+^i I_-^j \rangle = 0$ ($i \neq j$) the rates are simply given as $D_{sc}^\mp = (\frac{1}{2} \pm \frac{m}{N}) \Gamma_{\text{nuc}}^m$. For the quantum description the characterization via the spin projection quantum number m is not sufficient; the rates also depend on the symmetry of the nuclear state, quantified by the total spin $J \in \{0, \dots, N/2\}$. For a Dicke state $|J, m\rangle$ the rates are given as $D_{qm}^\mp = \langle I_\pm I_\mp \Gamma_{\text{nuc}} \rangle_{J,m} = \frac{1}{N} [J(J+1) - m(m \mp 1)] \Gamma_{\text{nuc}}^m$. For the statistically relevant J -subspaces [$J = \mathcal{O}(\sqrt{N})$] we find that D_{qm}^\mp are in good agreement with those obtained in the semiclassical limit for small m values.

Generalized Overhauser field

For the main part of the nuclear Hilbert space – namely the domain where the operator $I_x + \delta/g$ is large (recycling region) – the ϵ correction to the Overhauser field represents a negligible perturbation to the hyperfine interaction. However, in the domain of small eigenvalues of $I_x + \delta/g$ (trapping region) this perturbative picture is not trivially justified. However, for the diffusive dynamics we are mainly interested in the number of eigenstates in a region $L = (-\delta - \eta, -\delta + \eta)/g$ around $-\delta/g$. For I_x the number of eigenstates with small eigenvalues is very large (exponential in the number of spins), which is favorable for our scheme. For the generalized Overhauser field it is given by $D(\eta, \epsilon) = \int_{-\delta-\eta}^{-\delta+\eta} dE \text{Tr}(\delta(E - I_x - \epsilon I_+ I_-))$ and deviates from the number of unperturbed eigenstates:

$$D(\eta, \epsilon) = D(\eta, 0) + \sum_{n=1}^{\infty} U^{(n)}. \quad (11)$$

The sum $\sum_{n=1}^{\infty} U^{(n)}$ can be upper bounded by $\sim D(\eta, 0)$ for large trapping regions $\eta \gg \epsilon$, i.e. the number of eigenstates changes at most by a factor of order 1. Numerical calculations for $N = 10^4$ inhomogeneously coupled spins show that even for $\eta \sim \epsilon$ the number of states in both the perturbed and unperturbed case differ only by a few percent.

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- [26] An example of such a process is off-resonant hyperfine-assisted laser scattering $\propto \epsilon^2 (\Gamma/\omega_x)^2$.
- [27] This assumption is justified by the fact that the degeneracy of the the available Overhauser field configuration drops for larger $\langle I_x \rangle > A_H/\sqrt{N}$. In addition, the random walk of the Overhauser field is biased since the rates of flipping a spin up or down come with a factor proportional to the number of available final configurations. This bias becomes large for $\langle I_x \rangle > A_H/\sqrt{N}$ and prevents the Overhauser from exploring extreme polarizations.
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Figure Captions

Figure 1 The energy level diagram of a solid-state emitter. (a) The electron spin state $|\uparrow_x\rangle$ ($|\downarrow_x\rangle$) is resonantly coupled to a trion state with an x (y) polarized laser field with Rabi frequency Ω_p (Ω_c). In Voigt geometry the oscillator strengths of the two transitions are identical, leading to spontaneous emission rates with equal strength $\Gamma/2$. Optical excitation allows for energy conserving hyperfine flip-flop transitions that result in nuclear spin diffusion; these second-order processes are depicted using dashed curves. (b) The confined electron wave-function leads to inhomogeneous hyperfine coupling with the nuclei. In the simulations, we assume that the dot can be described as consisting of 5 different classes of nuclei. All nuclei within a class have identical hyperfine coupling, with strength determined by the electron wave function.

Figure 2 Nuclear spin state selective coherent population trapping. (a) The absorption lineshape in the presence of hyperfine interactions with quantum dot nuclei for Rabi frequencies $\Omega_p = \Omega_c = 0.2 \Gamma$ (red) and $\Omega_p = \Omega_c = 0.4 \Gamma$ (blue): in stark contrast to the standard coherent population trapping profile (dashed lines), the dark resonance is drastically broadened (solid lines). The broadening of the dark resonance is a consequence of the fact that optical excitation induced nuclear spin diffusion allows the coupled electron-nuclei system to find a Overhauser field configuration that satisfies the dark state condition for a broad range of initial laser detunings. (b) The standard deviation of the Overhauser field σ_{OF} for $\Omega_p = \Omega_c = 0.2 \Gamma$ (red line) and $\Omega_p = \Omega_c = 0.4 \Gamma$ (blue line) is reduced to the level below that of a single nuclear spin flip (green dashed line). The dashed black line shows the standard deviation in the absence of laser drive. (c) The final Overhauser field distribution could be determined by using a fast scan of the probe laser across the optical resonance. The solid (blue) curve shows the absorption lineshape obtained when the system is initially prepared in a dark state by setting $\Delta\omega_p = -0.2\Gamma$: the width and the depth of the dark resonance reveals information about the Overhauser field distribution. If the experiment is carried out by starting out in a random nuclear spin state, the observed lineshape is close to a Lorentzian (dashed green curve).

Figure 3 Homogeneously coupled electron-nuclei system. (a) Nuclear spin diffusion rates depending on the nuclear spin projection m assuming homogeneous coupling (cf. Eq. (6)). Parameters are $N = 4 \times 10^4$, $\Gamma = 1 \text{ GHz}$, $A = \omega_x = 100 \mu\text{eV}$ and $\Omega = 0.1 \text{ GHz}$. The quantum mechanical rates are taken for the subspace $J = \sqrt{N}/2$. (b) The dependence of the steady state value of the nuclear Overhauser field standard deviation as a function of Ω , calculated using the fully quantum model. The increase in standard deviation for $\Omega_p = \Omega_c < 0.2\Gamma$ is

a consequence of the fact that the coupled electron-nuclei system spends a substantial amount of time outside the transparency region. The inset shows the steady state population of I_x eigenstates for $\Omega_p = \Omega_c = 0.02\Gamma$. Despite the finite standard deviation $\sigma \approx 9$ the system is strongly peaked around $m = 0$.





