Quantum description of nuclear spin cooling in a quantum dot

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We study theoretically the cooling of an ensemble of nuclear spins coupled to the spin of a localized electron in a quantum dot. We obtain a master equation for the state of the nuclear spins interacting with a sequence of polarized electrons that allows us to study quantitatively the cooling process including the effect of nuclear spin coherences, which can lead to "dark states" of the nuclear system in which further cooling is inhibited. We show that the inhomogeneous Knight field mitigates this effect strongly and that the remaining dark-state limitations can be overcome by very few shifts of the electron wave function, allowing for cooling far beyond the dark-state limit. Numerical integration of the master equation indicates that polarizations larger than 90% can be achieved within a millisecond time scale.

I. INTRODUCTION

Nuclear spins are one of the best studied quantum systems, and highly developed techniques such as NMR have allowed detailed studies of the properties and dynamics of molecular and solid-state systems.[1] Due to their very long decoherence time, nuclear spins (and hyperfine levels) have also played a central role in many approaches to the implementation of quantum information processing (QIP).[2–6]

Recently, the localized ensemble of nuclear spins in a quantum dot (QD) has received special attention in the context of QIP with electron spins in QDs: the nuclei couple via a Fermi contact interaction to the electron spin[7] and, as predicted by theory,[8–12] have been shown in recent experiments to constitute the major source of decoherence of electron spin qubits in some of the most promising QD-based implementations.[13,14] The vice of this strong coupling is turned into a virtue when the electron is used to manipulate the state of the nuclear ensemble. This has long been exploited in dynamical nuclear polarization[15–18] (DNP) in bulk systems and has afforded many insights into the spin dynamics in solids.[17,19]

DNP *in quantum dots* has come into focus more recently in the context of QIP, since strongly polarized nuclei could lead to much longer electron spin dephasing times,[12] provide strong local magnetic field gradients required in quantum information proposals,[20,21] and even allow one to utilize the nuclear spins themselves as long-lived quantum memory.[22,23] More generally, a highly polarized nuclear spin ensemble in a QD provides, together with the electron spin, a strongly coupled, well-isolated mesoscopic quantum system with close similarities to the Jaynes-Cummings model in quantum optics,[23–25] with the fully polarized state corresponding to the vacuum in all cavity modes. Thus ultrahigh DNP in QDs may open the door to realize cavity-QED in quantum dots and implement tasks such as state engineering.

Experimentally, significant nuclear polarization in self-assembled QDs has been achieved.[26–30] However, the degree of polarization in these experiments was still too low to improve electron spin coherence times considerably and still far from the ground state.

Theoretically, cooling dynamics has mostly been considered in the spin temperature approximation,[1,17,31,32] in which coherences among the nuclear spins are neglected. This is appropriate if, as in bulk or quantum well systems, there is no fixed electron wave function and many motional states are involved, or if the nuclear dephasing rate is large. In quantum dots, however, the nuclei interact collectively with an electron in the motional ground state of the QD and the higher motional levels are far detuned. Therefore the coupling strength of each nucleus is fixed, and well-defined phase relationships between the nuclear spins can build up, necessitating a quantum treatment of the process, which was first pointed out by Imamoğlu *et al.*[33] who showed that the cooling process can be inhibited by so-called dark states, which trap excitations and potentially result in serious constraints on the achievable polarizations. While it was pointed out in Ref. 33 that inhomogeneities (either inherent in the system or introduced actively by modulating the wave function of the electron) can mitigate this problem, these ideas were put to numerical test only in very small one-dimensional (1D) systems of ten nuclear spins. However, the effect of inhomogeneities is expected to be reduced for realistic larger systems,[22] and thus limitations due to dark states are more severe.[60]

We consider the cooling of N nuclear spins in a QD through interaction with polarized electrons. One cooling cycle consists of (a) initialization of the electron spin in a well-defined direction and (b) evolution of the combined system for a "short" time. In this way the electron spin acts effectively as a $T=0$ reservoir for the nuclear spin bath and pumps excitation out of it.

We derive in a consistent manner a full quantum model of this process, which allows us to numerically study particle numbers of up to $N \sim 10^3$. We show that a sufficient inhomogeneity of the couplings leads to a dephasing of nuclear spin states and thus limitations due to dark states are partially lifted. We demonstrate that enhanced cooling protocols involving only a few (≤ 10) modulations of the electron wave function allow one to fully overcome these limitations, indicating that Overhauser fields above 90% of the maximal value can be created within the nuclear spin diffusion time.

The paper is organized as follows: In Sec. II we present the generic cooling protocol and analyze its performance in Sec. III; the applicability of the scheme to some specific physical systems is studied in Sec. IV.

II. COOLING SCHEME

Interaction. The Fermi contact interaction between an (s-type conduction band) electron spin $\mathbf{S}$ and the spins $\mathbf{I}_i$ of the lattice nuclei leads to a Heisenberg-like coupling $A\alpha_i\mathbf{I}_i \cdot \mathbf{S}$ to the nuclear spin at lattice site i, where A sets the overall strength of the hyperfine interaction and the factor $0 < \alpha_i < 1$ is determined by the probability to find the electron at site i and the gyromagnetic ratio of the ith nucleus.[7] In the presence of an external magnetic field B_{ext} we write the Hamiltonian of the spin system with the collective nuclear spin operators $A^\mu = \Sigma_i g_i I_i^\mu$ ($\mu = \pm, z$) as ($\hbar = 1$)

$$H = \frac{g}{2}(A^+ S^- + S^+ A^-) + gA^z S^z + g^* \mu_B B_{ext} S^z, \quad (1)$$

where we have defined $g = A\sqrt{\Sigma_i \alpha_i^2}$ and $g_i = \alpha_i/\sqrt{\Sigma_i \alpha_i^2}$, such that $\Sigma_i g_i^2 = 1$, and denoted the electron g factor by g^* and the Bohr magneton by μ_B.

We do not consider the Zeeman energy of the nuclear spins, because for typical QDs it is much (10^3 times) smaller than the electron's Zeeman energy,[7] and similarly we neglect the even smaller dipolar interaction between the nuclei. The effects of these are briefly discussed at the end of Sec. III. Finally, we restrict the analysis to nuclear spins $I = 1/2$ and one nuclear species only in this article.

The first part of the above Hamiltonian exchanges spin excitation between the electron and the nuclei, and it is this mechanism that is used to create polarization. The second part of the Hamiltonian constitutes a "quantum" magnetic field, the Overhauser field, for the electron spin generated by the nuclei.

The cooling scheme. We assume initially the electron spin to be pointing in the $-z$ direction $|\psi_{e-}\rangle = |\downarrow\rangle$. In the absence of a magnetic field this initial state defines the axis of quantization. The cooling cycle we consider is an iteration between evolution with Hamiltonian, Eq. (1), and reinitialization of the electron to $|\downarrow\rangle$. The nuclei effectively "see" a large cold reservoir of electron spins, and the concatenated evolution of the nuclear spin density matrix becomes

$$\rho \to \cdots U_t \text{tr}_e [U_t(\rho \otimes |\downarrow\rangle\langle\downarrow|)U_t^\dagger] \otimes |\downarrow\rangle\langle\downarrow| U_t^\dagger \cdots . \quad (2)$$

Here $U_t = \exp(-iHt)$ is the time evolution operator, tr_e denotes the trace over the electron, and here and in the following ρ will denote the state of the nuclear spin system only. Spin-polarized currents or optical pumping with polarized light give rise to a polarized electron bath, but also the fast electrical control available in double QDs[13] allows for the creation of nuclear spin polarization without the need for preprepared electrons, as we will detail in the last section of this article.

Considering small times for the evolution in each individual step of the cooling protocol, we expand the time evolution operators in Eq. (2) to second order. The standard deviation of the $A^{\pm,z}$ terms scales as $A\sqrt{\Sigma_i \alpha_i^2} = g \sim \mathcal{O}(A/\sqrt{N})$ for the initially totally mixed nuclear spin state, and thus for $\Delta t \ll g^{-1} \sim \sqrt{N}/A$ we neglect higher orders. The readily obtained master equation

$$\rho_{t+\Delta t} - \rho_t = i\frac{g\Delta t}{2}[A^z, \rho_t] - \frac{g^2(\Delta t)^2}{8}[A^z, [A^z, \rho_t]]$$
$$- \frac{g^2(\Delta t)^2}{8}(A^+ A^- \rho_t + \rho_t A^+ A^- - 2A^- \rho_t A^+) \quad (3)$$

contains a Hamiltonian part arising from the Overhauser field and a contribution in Lindblad form. The latter generates the nuclear spin polarization and has been studied in the limit of homogeneous coupling constants in the context of superradiance.[34–36]

As polarization builds up and $g\langle A^z\rangle \gg A/\sqrt{N}$ the Hamiltonian terms on the right-hand side of Eq. (3) may become large (for fixed time step Δt). To preserve the validity of the master equation one can either reduce the interaction time $\Delta t < A^{-1}$ or assume that the Overhauser field $\langle A^z\rangle$ is approximately compensated by an applied magnetic field, so that $\langle gA^z - g^*\mu_B B_{ext}\rangle\Delta t \ll 1$ for all times. In the latter case Δt is short enough to ensure quasiresonant hyperfine flips despite the random detunings stemming from the fluctuating Overhauser field and at the same time large enough to guarantee a fast cooling rate.[61] This is the situation we investigate in the following. Without retuning the system in this manner the polarization rate becomes dependent on the polarization itself and the emerging nonlinearities give rise to the bistability effects observed in Refs. 14, 30, and 37–41 and limit the final polarization.

Homogeneous coupling. Before we discuss general inhomogeneous couplings, consider for a moment the homogeneous case $\alpha_i \propto 1/N$ as a demonstration of some interesting features of the above master equation. In this case, the operators $A_{\pm,z}$ appearing in Eq. (3) form a spin algebra $I_{\pm,z}$ and the collective angular momentum states (Dicke states) $|I, m_I, \beta\rangle$ provide an efficient description of the system dynamics:[42,22] the total spin quantum number I is not changed by $A_{\pm,z}$ and the effect of Eq. (3) is simply to lower [at an (I, m_I)-dependent rate] the I_z quantum number. If $m_I = -I$ is reached, the system cannot be cooled any further, even if (for $I \ll N/2$) it is far from being fully polarized. These dark states[22,33] are a consequence of the collective interaction, Eq. (1). Thus spin excitations are trapped and cooling to the ground state prevented. We evaluate the steady-state polarization $\langle I^z\rangle_{ss} = \langle\Sigma_i I_i^z/\sqrt{N}\rangle_{ss}$ as

$$\frac{\langle I^z\rangle_{ss}}{\langle I^z\rangle_0} = \frac{2}{2^N N}\sum_{I=0}^{N/2} I(2I+1)D_I = \sqrt{\frac{8}{\pi N}} + \mathcal{O}(1/N); \quad (4)$$

i.e., for a mesoscopic number of particles the obtained polarization is negligible. In the above equation $\langle I^z\rangle_0$ is the expectation value in the completely polarized state, $D_I = \binom{N}{N/2-I} - \binom{N}{N/2-I-1}$ is the degeneracy of the subspaces of different total angular momentum, and the last equality has been obtained by employing the Stirling formula.

Evolving the nuclei according to Eq. (3), we find the exact time evolution of the polarization as shown in Fig. 1. In these and the following simulations $g\Delta t = 0.1$—i.e., $\Delta t = 0.1g^{-1} \sim 0.1\sqrt{N}/A$. As expected the polarization decreases as $1/\sqrt{N}$ as N increases, which underlines the importance of the nuclear spin coherences. In particular this shows that an in-
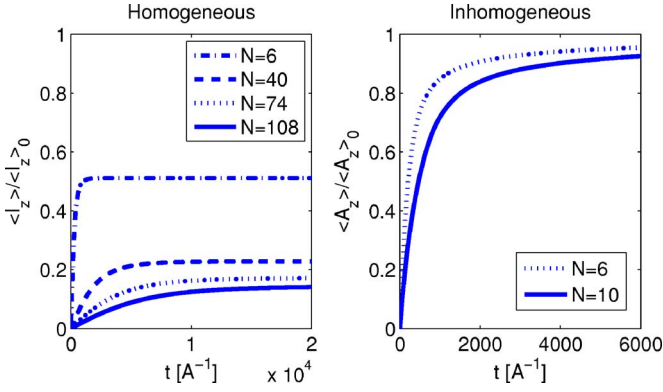
FIG. 1. (Color online) Exact polarization dynamics. Left: Homogeneous case, $g_j = 1/\sqrt{N}$. Right: In the inhomogeneous case, $g_j \propto \exp(-(j-N/2-1/4)^2/w^2)$. The term $1/4$ is added to account for asymmetry between electron wave function and the lattice and avoid symmetry effects for this small scale system.

coherent spin temperature description of the process would give even qualitatively wrong results. The time scale over which the steady state is reached is $\sim N/(g \Delta t A)$.

Inhomogeneous coupling. Consider now an inhomogeneous wave function. The results for the exact evolution of the quantity of interest, $\langle A^z \rangle$, are shown in Fig. 1. The coupling constants g_j in this example are taken from a 1D Gaussian distribution with width $N/4$.[62] The most important and striking feature is that in this situation almost complete polarization is obtained.

The reason that this is possible here is *not* that there are no dark states in the case of inhomogeneous coupling constants. On the contrary it has been shown that there exists a one-to-one mapping[22] from the familiar homogeneous dark states ($|I,-I,\beta\rangle$ in the Dicke basis) to their inhomogeneous counterparts, defined by $A^-|D\rangle=0$. The reason for obtaining high polarization beyond the homogeneous limit is the Hamiltonian part of the master equation (3). To illustrate this point, consider two spins with coupling constants $g_1 \neq g_2$. Then the dark state $|\Psi_D\rangle \propto g_2|\uparrow\downarrow\rangle - g_1|\downarrow\uparrow\rangle$ evolves due to the A^z term in Eq. (3) to $e^{i\delta_g t}g_2|\uparrow\downarrow\rangle - e^{-i\delta_g t}g_1|\downarrow\uparrow\rangle$, where δ_g is proportional to g_1-g_2. Obviously this state will become "bright" again after a time $\propto 1/|g_i-g_j|$ and $A^-|D\rangle \neq 0$. This process is first order and, as we will detail later, "delivers" coolable excitations sufficiently fast to maintain a high cooling rate.

III. POLARIZATION DYNAMICS

The polarization dynamics of the nuclear ensemble is governed by Eq. (3). While for homogeneous systems the collective angular momentum Dicke basis enables an efficient description of the problem, for realistic large and inhomogeneous systems more effort is required.

To study the evolution of the nuclear polarization, we are interested in the individual spin expectation values $\langle \sigma_i^+ \sigma_i^- \rangle$. These depend, via Eq. (3), on all the elements of the covariance matrix

$$\gamma_{ij} = \langle \sigma_i^+ \sigma_j^- \rangle,$$

which, in turn, depend on higher-order correlations as seen from the equations of motion

$$\frac{\Delta \gamma_{ij}}{\Delta t} = \xi_{ij}\gamma_{ij} - \kappa \sum_k g_k(- g_i \langle \sigma_k^+ [\sigma_i^+, \sigma_i^-] \sigma_j^- \rangle$$
$$+ g_j \langle \sigma_i^+ [\sigma_j^-, \sigma_j^+] \sigma_k^- \rangle), \tag{5}$$

where $\xi_{ij} = ig(g_j-g_i)/2 - g^2\Delta t(g_j-g_i)^2/8$ and $\kappa = g^2\Delta t/8$ and the σ_i^μ refer to the Pauli matrices at site i.

The simultaneous solution of the ensuing hierarchy of equations is only feasible for very small particle numbers N, and further approximations are needed to treat the large systems of interest. We introduce several ways, labeled (i)–(v), of closing this set of equations and discuss their validity and implications in detail below.

In the strongest approximation (i) all coherences between different spins are neglected, yielding independent rate equations for each individual nuclear spin. This reproduces essentially the spin-temperature description commonly employed in the discussion of bulk DNP[1,17] (each subset of spins with identical coupling strengths g_i is assigned its own effective temperature). This approach cannot reproduce the quantum effects we want to study, but it can serve as a benchmark for how strongly these are influencing the cooling process.

The simplest approximations that take quantum coherences between nuclear spins into account close the hierarchy of equations at the level of second-order correlations. Our approximation (ii) is motivated by the generalized Holstein-Primakoff description,[43] which in lowest order treats the nuclei as bosonic modes $\sigma_i^- \to a_i$. The bosonic commutation relations $[a_i, a_j^\dagger] = \delta_{ij}$ yield a closed set of equations for the elements of the covariance matrix γ. The bosonic description is known to be accurate for highly polarized and moderately inhomogeneous systems[25] and allows one to bring results and intuition from quantum optics to bear in the spin system discussed here. Dark states are included in the form of the vacuum of the collective mode $b = \sum_i g_i a_i$ coupled to the electron in Eq. (1). For unpolarized systems (with on average $1/2$ excitations per bosonic mode a_i), this description provides a lower bound on the performance of the cooling protocol, since in the absence of an inhomogeneous Knight field cooling is limited to $\mathcal{O}(1)$ excitations per mode rather than the $\mathcal{O}(\sqrt{N})$ coolable excitations expected at the beginning of the cooling process for spins; cf. Eq. (4). In the two limiting cases discussed so far, Eq. (5) simplifies to

$$\frac{\Delta \gamma_{ij}}{\Delta t} = \begin{cases} -2\kappa\delta_{ij}g_i^2\gamma_{ii} & \text{(i) spin temp.,} \\ \xi_{ij}\gamma_{ij} - \kappa\sum_k g_k(g_i\gamma_{kj} + g_j\gamma_{ik}) & \text{(ii) bosonic.} \end{cases}$$

One can take into account more aspects of the spin algebra by replacing some higher-order expectation values by lower orders using the properties of Pauli matrices $[\sigma_i^+, \sigma_i^-] = \sigma_i^z$ and $\sigma_i^z \sigma_i^\pm = \pm \sigma_i^\pm$, obtaining

$$\frac{\Delta \gamma_{ij}}{\Delta t} = \xi_{ij}\gamma_{ij} - \kappa\delta_{ij}\sum_k g_k(g_i\gamma_{kj} + g_j\gamma_{ik})$$
$$- \kappa(1-\delta_{ij})\left(-\sum_{k\neq i} g_k g_i \langle \sigma_k^+ \sigma_i^z \sigma_j^- \rangle + g_i^2\gamma_{ij}\right.$$
$$\left.- \sum_{k\neq j} g_k g_j \langle \sigma_i^+ \sigma_j^z \sigma_k^- \rangle + g_j^2\gamma_{ij}\right). \tag{6}$$
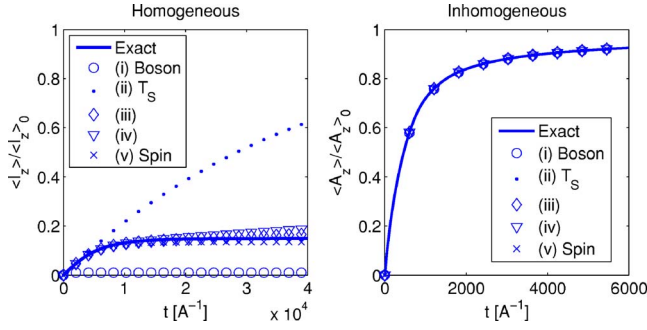
FIG. 2. (Color online) Comparison of different approximation schemes for the homogeneous situation with $N=100$ (left) and the case of Gaussian couplings (as in Fig. 1) and $N=10$ nuclear spins (right).

The remaining higher-order expectation values (now having distinct indices $i \neq j, j \neq k$) can be approximated in a Hartree-like way[44] (iii) or, having the bosonic limit in mind, by the Wick theorem (iv),

$$\frac{1}{2}\langle \sigma_k^+ \sigma_i^z \sigma_j^- \rangle = \begin{cases} \left(\gamma_{ii} - \dfrac{1}{2}\right)\gamma_{kj} & \text{(iii)}, \\[2mm] -\dfrac{1}{2}\gamma_{kj} + \gamma_{ki}\gamma_{ij} + \gamma_{kj}\gamma_{ii} & \text{(iv)}. \end{cases}$$

The fifth and final approximation scheme we invoke has been introduced in the context of superradiance as a Wick-type factorization, which takes into account the partly bosonic, partly fermionic properties of spin-1/2 operators.[36] In contrast to the last two factorization schemes, it does not rely on a distinction of cases. It is directly based on the exact equation (5) and approximates the three-operator-expectation values in the following way:

$$\frac{1}{2}\langle \sigma_k^+ \sigma_i^z \sigma_j^- \rangle = -\frac{1}{2}\gamma_{kj} - \gamma_{ki}\gamma_{ij} + \gamma_{kj}\gamma_{ii} \quad \text{(v) "spin."}$$

Direct comparison of the approximation schemes (i)–(v) with the exact solution for both homogeneous and inhomogeneous couplings is shown in Fig. 2. In the homogeneous case the spin temperature description (i) is clearly qualitatively wrong, because it neglects correlations in the bath. The bosonic description (ii) captures the feature of dark states, but it overestimates their influence: Instead of $\sim\sqrt{N}$, only one excitation can be removed. The two schemes based on a distinction of cases, (iii) and (iv), give very good results initially, until roughly $\sqrt{N}$ spins have been flipped. Then, however, the polarization keeps increasing on a slow time scale and does not reach a steady state in the correct time. The (v) "spin" approximation gives very good results and gets both the polarization time scale and the finally obtained value of the polarization right within a few percent.

The comparison of the different approaches to the exact solution for inhomogeneous couplings is restricted to small particle numbers (see Fig. 2). In this regime all introduced approximation schemes reproduce the exact dynamics correctly. The reason for the good correspondence is the strong
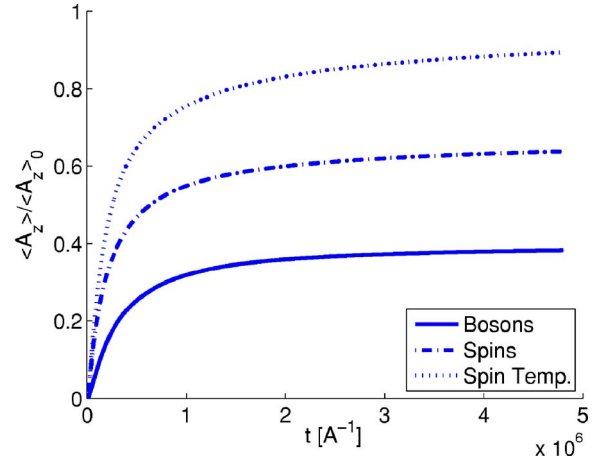

FIG. 3. (Color online) The polarization dynamics for $N=1000$ spins coupled with a 2D Gaussian wave function, which is shifted from the origin by $1/3$ in the x and y directions.

dephasing of dark states and generally coherences between nuclear spins for small inhomogeneous systems.

Using these approximations we present the polarization dynamics for $N=10^3$ spins coupled through a 2D Gaussian wave function in Fig. 3. For the data presented in this and the following figure, we considered the spins in a 2D square lattice geometry, with the lattice constant set to unity. The bosonic description displays the lowest final polarization and polarization rate (for the same reasons as in the homogeneous case) and is expected to give lower bounds on the performance on the polarization procedure. Of particular interest are the predictions of the (v)-"spin"-approximation scheme, because its good performance in the completely homogeneous situation gives us confidence that also partial homogeneities are correctly accounted for. The achieved polarizations of $\sim60\%$ in this setting show the importance of the intrinsic dephasing due to the inhomogeneity (homogeneous coupling would allow for $<5\%$ polarization). However, the intrinsic inhomogeneity alone does not allow for ultrahigh polarizations and we are thus led to investigate more sophisticated cooling schemes. As shown later, in these enhanced protocols all approximation schemes lead to the same conclusions.

To gain a better understanding of the presented phenomena in the inhomogeneous situation, we go to an interaction picture $\rho_I = U_0 \rho U_0^\dagger$, with $U_0 = \exp(-iA^z t/2)$, which shows very clearly the oscillating coherences between spins with $g_i \neq g_j$:

$$\frac{\Delta \rho_I}{\Delta t} = -\kappa \left[\sum_{ij} g_i g_j e^{-ig(g_i - g_j)t/2} \sigma_i^+ \sigma_j^-, \rho_I \right]_+ + 2\kappa \sum_{ij} g_i g_j e^{-ig(g_i - g_j)t/2} \sigma_j^- \rho_I \sigma_i^+. \tag{7}$$

In the rotating-wave approximation (RWA), the rotating terms ($g_i \neq g_j$) are neglected and in the absence of exact symmetries the above equation reduces to the spin temperature description. A partial rotating-wave approximation neglects only the coherences between spins with considerably differ-

ent coupling constants; i.e., the ratio between dephasing and polarization rate is required to be large $[4|g_i-g_j|/(g\Delta tg_ig_j) > 1]$. This procedure gives a block-diagonal Liouvillian which allows for the extension of the numerical studies to particle numbers up to $N=10^4$.

In the RWA we evaluate the buildup time τ_p for the polarization as the inverse of the weighted average of the individual spin decay times,

$$\tau_p = \left(\frac{\sum_i g_i\kappa_i}{\sum_i g_i}\right)^{-1} = \frac{4\sum_i g_i}{g(g\Delta t)\sum_i g_i^3} = \mathcal{O}\left(\frac{4N^{3/2}}{A(g\Delta t)}\right), \quad (8)$$

and find good agreement with the numerically obtained time scale to reach the steady state in all discussed schemes. For example, for the data presented in Fig. 3 we find times of 3.4×10^5 (spin temp.), 4.6×10^5 (bosonic), and 3.3×10^5 ("spin") in units of A^{-1} to reach $(1-e^{-1})\approx0.63$ of the quasi-steady-state Overhauser field. This agrees well with the analytical estimate $\tau_p\approx2.4\times10^5/A$, despite the differences in the final polarizations obtained in the different approximation schemes. This correspondence between the RWA-based estimate and the numerically obtained polarization times for the coherent evolution indicates that the inhomogeneous Knight field provides coolable excitations at a rate larger than the polarization rate, thus not slowing down the process.

When the inhomogeneity of the coupling is large enough to justify the rotating-wave approximation, each spin evolves with its own Liouvillian and the nuclei remain in a product state during the whole evolution. To keep the errors in the derivation of the master equation [due to higher-order terms of the expansion of the time evolution operators in Eq. (2)] small, it is sufficient to do so for each spin individually in this case. This allows a larger time step $\Delta t \ll (A\alpha_{\max})^{-1} = \mathcal{O}(N/A)$ in each cycle, and therefore the cooling rate can be significantly enhanced. The cooling time effectively scales only linearly in the particle number

$$\tilde{\tau}_p = \mathcal{O}\left(\frac{4N}{A(\Delta tA/N)}\right). \quad (9)$$

Taking $A=100$ μeV ~ 40 ps, a value typical for GaAs QDs, and 0.1 as the value for the terms $g\Delta t$ and $A/N\Delta t$ in the denominators of Eqs. (8) and (9), respectively, we find that approximately 4×10^3 and 3×10^5 spins can be cooled to more than 90% of the steady-state value $\langle A^z\rangle_{ss}$ within a millisecond.

We now study enhanced cooling protocols that lift the dark-state limitations and which rely solely on the ability to shift the center of the electron wave function. These shifts can be effected by applying dc gate voltages to the QD. After such a shift only very few spins will have the same coupling constants for *both* wave functions and therefore singletlike coherences are broken up. We confirm this expectation numerically as shown in Fig. 4 for some exemplarily chosen shifts of the electron wave function. The shifts range from a few lattice sites to roughly the width of the electron wave function. The timing of the shifts we have performed for obtaining the data presented in Fig. 4 can be inferred from
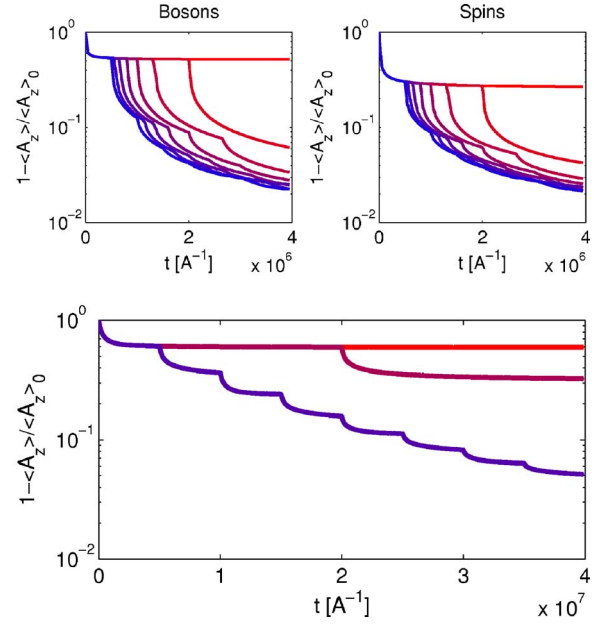

FIG. 4. (Color online) Polarization dynamics in the enhanced cooling protocol for $N=196$ (upper plots) and $N=1000$ (lower plot). In the upper plots approximation schemes (ii) (left) and (v) (right) have been invoked; the lower plot is based on the bosonic model and the partial rotating-wave approximation (see text). In all plots the different lines are representing cooling procedures with different numbers of mode changes. In the upper plots the randomly chosen Gaussian modes with width $w=N/4$ are defined by the centers $\{(1/3,1/3),(1.35,-0.81),(0.32,-0.04),(1.17,0.79),(-0.13,-1.44),(0.96,-0.17),(0.35,0.88),(1.27,0.71)\}$. In the lower plot only two modes with centers $\{(1/3,1/3),(-3.15,-1.5)\}$ have been iterated.

the plots, as it is accompanied by a rapid increase in the cooling rate.

Regarding the approximation schemes, we have found that all schemes taking into account coherences, (ii)–(v), predict the same behavior and the spin-based factorization (v) offers the quantitatively best description. It is important to note that all these descriptions coincide at the end of the cooling protocol [shown in Fig. 4 only for (ii) and (v)]. In particular the limiting bosonic model predicts the same high ($\geq95\%$) polarizations and cooling rates as the other schemes, which leads us to conclude that the $\mathcal{O}(10)$-mode changes are sufficient to achieve near-ground-state cooling for realistically large numbers of nuclei in QDs.

Despite being a radical approximation at low polarization, the bosonic scheme (ii) captures the cooling dynamics qualitatively and we remark that it can be generalized to provide an accurate and conceptually simple description of the electron-nuclear spin dynamics at high polarizations.[25]

The cooling schemes we have presented are governed by the optimal time scale set by the hyperfine interaction constant A, but the schemes themselves leave room for optimization: The cooling rate can be tuned by choosing Δt adaptively during the cooling process. The mode changes can be optimized by a careful choice of the size and the timing of the shifts, and through more sophisticated deformations of the electron wave function. These and further modifications

are implementation dependent and will be the topic of future work.

In using the Hamiltonian, Eq. (1), we have neglected a number of weak interactions that are present in actual systems and, while being much smaller than the dominant hyperfine term, may become important on the long time scales required to reach high polarization. We argue in the following that these terms do not affect the quantitative conclusions obtained. While nuclear Zeeman energies are large enough to cause additional dephasing between the nuclear spins, similar to the inhomogeneous Knight fields, this will only be effective between nuclei of *different* Zeeman energy—i.e., belonging to different nuclear species. This leads to two to three mutually decohered subsystems (in a partial rotating-wave approximation), each of which is described by our model.

The nuclear dipole-dipole interaction[45] can lead to both diffusion and dephasing processes, both of which are of minor importance as shown below. Dipolar processes that change A^z are off-resonant and hence expected to be slow, as indicated by the nuclear spin diffusion rates measured, e.g., in Ref. [46], and should not significantly affect the polarizations reached. Resonant processes such as terms $\propto I_i^z I_j^z$ affect the cooling process only insofar as they can cause dephasing of dark states similar to the inhomogeneous Knight shift. The rate at which coolable excitations are provided is set by the energy difference for two nuclear spins in a dark pair. The interaction energy for two neighboring spins is about $\sim 10^{-5} \ \mu\text{eV}$ (Ref. [7]); hence, a singlet of neighboring spins can dephase in $\sim 100 \ \mu\text{s}$ (or slower if all surrounding spins are polarized). Even widely separated spins interacting with differently polarized environments dephase only up to a few 10 times faster than this (depending on the geometry). Thus we see that the dipolar dephasing is considerably slower than that caused by the inhomogeneous Knight field, and only if the latter becomes inefficient due to homogeneities (towards the end of cooling a given mode) can the dipolar dephasing contribute coolable excitations, but at a much slower rate than what can be achieved by changing the electron wave function and the ensuing return to a situation of strong Knight inhomogeneity. Thus, one does not expect the cooling process to be affected except for a slight additional dephasing. However, on much longer time scales of tens of milliseconds the dipole-dipole interaction provides the depolarizing mechanism (mainly affecting nuclei with a weak hyperfine interaction) that needs to be considered, e.g., when cooling much beyond 90% polarization is studied.

Clearly a polarization <100% of the electron "reservoir" directly translates into limitations on the final polarization of the nuclei. A quantification of this necessarily needs to refer to the details of a concrete physical realization of our model, which is not the topic of this article. The limitations can be minute—e.g., in the case of the double-dot setup presented in the next section.

IV. ADAPTING THE MODEL TO CONCRETE PHYSICAL SETTINGS

The generic model of a single spin-1/2 particle coupled inhomogeneously to an ensemble of N nuclear spins can readily be adapted to various experimental settings.

If a source of spin-polarized electrons is available, single electron tunneling into the QD provides the initialization. Controlled tunneling into and out of the QD with rates $>10 \ \text{ns}^{-1}$ appears feasible,[47,48] justifying the description of the dynamics by a suddenly switched on and off interaction.

For self-assembled QDs, optical pumping with polarized light has been shown to provide a spin-polarized bath of electrons that cools the nuclei.[26–30] However, in this setup the average dwell time of a single polarized electron in the dot is large and the detuning due to the z component of the Overhauser field leads to instabilities[39–41] in the nuclear polarization which are avoided in our scheme.

In double QDs in the two-electron regime[49,50] the role of the states $|\downarrow\rangle, |\uparrow\rangle$ is played by the two-electron singlet $|\widetilde{S}\rangle$ and one of the triplet states; in the following, we consider $|T_+\rangle = |\uparrow\rangle|\uparrow\rangle$. Tunnel coupling between the two dots and the external magnetic field is chosen such that the other triplet states are off-resonant and cause only small corrections to the dynamics sketched here.

As discussed in more detail in Refs. [49–51], the hyperfine interaction in this system is described by the Hamiltonian $\Sigma_l \mathbf{S}_l \cdot \mathbf{A}_l$, where $l = L, R$ refers to the orbital state of the electron. Coupling between $|\widetilde{S}\rangle$ and $|T_+\rangle$ is mediated by the *difference* $\delta A^{\pm} = (A_L^{\pm} - A_R^{\pm})/2$ of the collective nuclear spin operators of the two dots L, R, while the effective Overhauser field is given by the sum $(A_L^z + A_R^z)/2$. Thus we have that the analysis of the previous sections applies to the double-dot case in this regime (to zeroth order; cf. Ref. [52]) with the replacements

$$|\downarrow\rangle \rightarrow |\widetilde{S}\rangle, \quad |\uparrow\rangle \rightarrow |T_+\rangle,$$

$$A^{\pm} \rightarrow -\sqrt{2}(\cos\theta)\delta A^{\pm}, \quad A^z \rightarrow \frac{1}{2}(A_L^z + A_R^z).$$

The adiabatic singlet has contributions from both the delocalized (1,1) and localized (0,2) charge states, and with $\cos\theta$ we denote the amplitude of the (1,1) contribution[50] [with (m,n) we denote a state with m electrons on the left and n electrons on the right dot]. The effect of higher-order terms (e.g., of the nuclear spin components $\delta A^z, A_L^{\pm} + A_R^{\pm}$) merits more detailed analysis.

This system is of particular interest since fast electrical control of gate voltages can provide a highly spin-polarized electron system through near-unity fidelity initialization of a singlet in the right-hand dot $|S(0,2)\rangle$.[13,53] Starting from this singlet, rapid adiabatic passage [1 ns (Ref. [13])] by means of tuning the asymmetry parameter ϵ between the dots, initializes the electrons to the adiabatic singlet $|\widetilde{S}\rangle$ and brings the system to the S-T_+ resonance.

The transitions from the singlet to the other two triplets $T_{0,-}$ are detuned by an external magnetic field (of order 100 mT in the experiments of Ref. [13]). After a time Δt the system is ramped back to the (0,2) charge region and the electrons relax to the singlet ground state, completing one cooling cycle. If relaxation to the state $S(0,2)$ is fast, the limiting time scale for this cycle is given by the hyperfine

coupling constant A, showing that here the polarization rate is governed by the natural and optimal time scale (and not other, slower time scales, like, e.g., cotunneling in Refs. 32 and 37).

In the GaAs double-dot setup the sudden approximation is justified for typical tunnel couplings $\sim 10~\mu$eV, which have to be compared to the typical time scale for a hyperfine flip $\lesssim 0.1~\mu$eV and the fact that additionally all spin flip transitions are off-resonant during the adiabatic ramp. At the S-T_+ resonance selecting a suitable combination of external magnetic field and time step Δt detunes the unwanted transitions and at the same time ensures resonance for the polarizing transition. Note also that the Overhauser field increases the external magnetic field in materials with negative electron g factor, like GaAs ($g^* \approx -0.44$), thus further suppressing unwanted transitions and requiring retuning of the end point of the adiabatic ramp. Given the availability of fast (100 ps) voltage pulses, the reinitialization of $|S(0,2)\rangle$ via a $(0,1)$ charge state is likely to be limited by the tunneling rate from the reservoir to the QD. For optimal cooling efficiency this rate should and could be made large $\gtrsim 10A/\sqrt{N}$.[47,48]

Since in the double-dot setup the "polarized" state is a spin singlet, there is no inhomogeneous Knight field to dephase the dark states and DNP will be severely limited. However, there are many ways of providing it—for example, by extending the cooling cycle to include a third step in which a single-electron state of the double dot is realized or by increasing the time spent at the S-T_+ resonance in each cooling cycle [the latter would require a reformulation of the master equation (3) not presented here]. At the same time it would be interesting to find evidence for quantum coherence between nuclear spins in QDs by comparison of the obtained Overhauser field in the case of strong and weak inhomogeneous Knight fields.[54]

V. CONCLUSIONS AND OUTLOOK

In summary we have presented a quantum treatment of a dynamical nuclear spin polarization scheme in single-electron quantum dots that takes into account quantum coherences between nuclei and allows numerical study of the cooling dynamics for thousands of spins. We have quantified limitations due to dark states and shown that these limits are overcome by the inhomogeneous Knight shift and active mode changes. From this we conclude that cooling to more than 90% (of the maximal Overhauser field) is feasibly faster than typical nuclear spin diffusion processes. Setups for the experimental realization of our scheme have been proposed.

In order to go beyond the presented results to polarizations larger than 99%, which would bring the system of coupled nuclei close to a pure state and significantly reduce electron spin decoherence, the presented scheme can be optimized, both in terms of timing (length of the individual cooling step and wave function changes) and in terms of the electron wave functions chosen. A further enhancement may be achieved by combining the polarization scheme with A^z measurements[56–58] to reduce the A^z variance and to tailor the interaction times and the external field to the measured A^z value. Dipolar interactions and other depolarizing processes will become more important in later stages of the cooling and need to be considered carefully in the development of ground-state cooling techniques. More detailed studies of these processes may, in addition, lead to schemes to monitor the intrinsic (dipolar) nuclear dynamics via the hyperfine interaction.

The combination of high polarization and long coherence times makes the nuclear spin ensemble itself a candidate for an active role in quantum computation. Like the actively explored single-nucleus-spin qubits,[5] collective excitations of a polarized ensemble of spins could also be used for quantum information purposes.[23] Similar to their atomic counterparts,[59,63] the ensembles might become more suited than their isolated constituents for certain quantum information tasks.

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[60] In a fully homogeneous system only a fraction of $\mathcal{O}(1/\sqrt{N})$ spins can be cooled before the system is trapped in dark states.

[61] Ensuring the validity of Eq. (3) for all times by retuning B_{ext} assumes that the standard deviation of gA^z remains bounded by $\mathcal{O}(A/\sqrt{N})$ thus keeping the error in each cooling step small. Computing $\text{Var} A^z$ in each step and choosing Δt accordingly guarantees correctness. In general, the polarizing process is expected to *decrease* $\text{Var} A^z$ from the initial value in the maximally mixed state. This is confirmed by exact numerical calculations for small particle numbers. We are confident that this holds for large N, too, since the generic states exhibit standard deviation $\leqslant \mathcal{O}(A/\sqrt{N})$ (as evidenced by the variance of the maximally mixed state). Moreover, the standard deviation in the maximal entropy state of total polarization P is $\mathcal{O}((1-P^2)A/\sqrt{N})$ for all P. Similar reasoning holds for the x and y directions.

[62] In this article we focus on Gaussian electron wave functions, which approximate experimental conditions well. For the coherent phenomena we discuss, the distribution of the groups of similarly coupled spins is of major importance. This property is generally mainly determined by the width and dimensionality of the wave function, and only to a smaller extent by its exact functional form.

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