Spin-bath polarization via disentanglement

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Abstract
The occurrence of any physical process is restricted by the constraints imposed by the laws of thermodynamics on the energy and entropy exchange involved. A prominent class of processes where thermodynamic constraints are crucial involve polarization of nuclear spin baths that are at the heart of magnetic resonance imaging, nuclear magnetic resonance (NMR), quantum information processing. Polarizing a spin bath, is the key to enhancing the sensitivity of these tools, leading to new analytical capabilities and improved medical diagnostics. In recent years, significant effort has been invested in identifying the far-reaching consequences of quantum modifications to classical thermodynamics for such processes. Here we focus on the adverse role of quantum correlations (entanglement) in the spin bath that can impede its cooling in many realistic scenarios. We propose to remove this impediment by modified cooling schemes, incorporating probe-induced disentanglement or, equivalently, alternating non-commuting probe–bath interactions to suppress the buildup of quantum correlations in the bath. The resulting bath polarization is thereby exponentially enhanced. The underlying quantum thermodynamic principles have far-reaching implications for a broad range of quantum technological applications.

1. Introduction
Thermodynamics permeates many aspects of daily life and technology, from machines to information. In particular, the thermodynamics of cooling, such as, the cooling of spins in a sample to be measured using magnetic resonance imaging (MRI), can have significant implications on the achievable sensitivity of the progress, by reducing its noise [1–8]. Quantum considerations may have direct consequences for spin cooling [9, 10]. The focus of this work is on the limitations of spin cooling due to quantum inter-spin correlations (entanglement), and on the proposed solution of introducing disentanglement in order to overcome these limitations. This concept can be generalized to diverse systems and applications, and specifically to emerging quantum technologies.

In the quantum domain [7, 11, 12], energy exchange between spins is maximized when they undergo resonant exchange (flip–flop), known as the Hartmann–Hahn (HH) effect [13, 14]. Under HH conditions, a low-polarized (hot) spin can be cooled through polarization swapping with a higher-polarized (cold) spin. If the cold spin is a probe that is continuously polarized by an external source, i.e., if the probe entropy is nearly-instantly removed, this probe may be expected to fully polarize the entire spin bath coupled to it. This concept of polarizing a spin bath is referred to in the fields of nuclear magnetic resonance (NMR) and...
MRI as hyperpolarization [14]. While the energy exchange interactions involved are described by quantum Hamiltonians, the cooling rates and bounds of hyperpolarization are usually calculated by classical thermodynamics [14]. The issue we deal with is that such treatments may not apply in common cases where the quantum correlations within the spin-bath have a very long decay time [21].

Cooling a spin-bath along with quantum correlations would lead to an effective steady state wherein the polarization of each spin in the bath is much smaller than that of the spin probe [15, 16]. Such steady-state behavior cannot be described within the classical rate equation model [14], and one has to rely on its full quantum description. Previously explored spin-bath cooling procedures by a single qubit [17, 18] have shown that the stationary states of such cooled spin-bath could be highly correlated, but the thermodynamic consequences of these correlations are less understood. The fact that quantum correlations reduce the purity at the single-spin level in a cooling process becomes clear when we analyze the process in the collective (angular-momentum) basis of a bath composed of $N$ spin-$1/2$ particles. When polarizing such a bath, its collective coupling to a quantum spin probe causes the spin-bath to evolve into disjoint manifolds of the collective spin observables. If all spins in the bath were identically coupled to the probe spin (which is rarely the case), they would evolve into Dicke [19] (collective-spin) states. Yet, even though the couplings of the individual spins to the probe may vary significantly, the spin bath still possesses collective variables whose eigenstates pertain to distinct manifolds (subspaces) that remain invariant under resonant exchange with the probe [20]. In particular, the isotropic central spin model possesses symmetries that cause initial correlations in the bath [21] to persist indefinitely. This is true even in the limit of an infinite spin bath, and these persistent quantum correlations are not linked to the integrability of the model. For initial thermal states such correlations are induced by the interaction of the spin bath with a polarized central spin. As shown here, these persistent inter-bath correlations may form a bottleneck in steering the spin-bath evolution toward desired targeted states, particularly the ground state. These symmetry-imposed invariant subspaces form a bottleneck for cooling, since they block heat and entropy removal from each subspace via resonant swap with the probe.

Here we wish to polarize all spins in the bath to their ground state. One option is to have strong inhomogeneous broadening so as to assign a different resonance to each spin [15]. This regime, however, precludes the HH resonant swap and results in limited effectiveness of the cooling. Alternatively, we here advocate a dynamic symmetry-breaking control that repeatedly destroys the correlations induced by the collective coupling, allowing much more population to be pumped into the uncorrelated single-spin states. We show that, as a result, it is possible to nearly fully polarize the spins in the bath to their ground state, efficiently and rapidly, by alternating resonant and off-resonant (dispersive) coupling of the probe to the bath, thereby inducing in turn flip-flop and decohering (disentangling) interactions in the spin-bath. Previous attempts to break such collectivity resorted either to wave-function modulation of the central spin so asto controllably vary the interaction of the central spin with the bath [18], or harnessed the spin–spin interactions among the bath spins [22]. The former control is however not applicable to such solid-state spin systems as NV centers. The latter control was shown to enhance polarization only by 30% [22]. We here prescribe a general framework for destroying the collectivity and enhancing the spin-polarization in a spin bath toward unity. We show that our novel control scheme applies to both dipolar spin-coupled systems such as NV centers and to hyperfine-coupled quantum dot systems.

The proposed spin-bath cooling protocol can take a completely unpolarized (thermalized) ensemble toward a pure state, namely $(1/2^N) I \rightarrow |00 \cdots 0\rangle |00 \cdots 0\rangle$, via entropy-raising decohering operations that alternate with entropy-lowering resonant-swap (HH) steps. Such counterintuitive use of decoherence, which tends to increase the state entropy, as a means of cooling down an unpolarized spin ensemble, opens a new vista into the effects of quantum correlations on entropy changes in an open many-body system. This general protocol is illustrated for the cooling of a nuclear spin ensemble by an electron spin of a defect center in a diamond [1] or in a semiconductor quantum-dot [23–25]. The advantages and conceptual implications of the protocol are presented, particularly, its ability to help elucidate the thermalization of many-body quantum systems [26–31].

2. Concept

We now describe the physical limits of cooling a spin-bath, and show that decoherence (dephasing) can be the key to circumventing the HH cooling bottleneck described above. To this end, we first consider a scenario wherein a spin probe is coupled to $N$—identical spins. On resonance, we then obtain an energy-exchange flip-flop Hamiltonian (see appendix A)

$$H = \sum_k g_k \left( S^+ I_k^- + S^- I_k^+ \right).$$
Such a resonant coupling is optimal for the exchange of polarization (excitations) between single spins, but due to multi-spin symmetries it alone may not be well suited for polarization exchange among spins, as we show below. For example, in the case of identical couplings, i.e., \( g_k = g \), we find that the total spin \( I \) of the ensemble remains conserved and the Hamiltonian assumes a block-diagonal form so that dynamics takes place independently in each block (manifold) with fixed \( I \). Consequently, no population exchange can take place among manifolds corresponding to collective-spin subspaces with different \( I \). Resonant exchange of the spin ensemble via Hamiltonian (1) with a frequently polarizable spin probe \( S \), allows each manifold to be polarized (purified) independently. As mixing among spin-manifolds with different \( I \) is not allowed by total spin conservation, the total purity of the spin-bath is restricted to values well below unity, even though the spin probe is able to transfer its entire polarization to the spin-bath. The resulting purity of the spin-bath (for even \( N \)) under such resonant exchange takes the form

\[
\mathcal{P} \equiv \text{Tr}[\rho_I^2] = \sum_{I=0}^{N/2} \lambda_I \left( \frac{2I+1}{2^N} \right)^2 \sim e^{-2N/3},
\]

where \( \lambda_I = N_{N/2-I} \frac{2^{I+1}}{2^{2I+1}} \) is the multiplicity of the different \( I \)-manifolds. Here \( N_{N/2-I} \equiv N! / [((N/2-I)!(N/2+I)!)] \) is the combinatorial factor. Such restricted purification of the bath holds even when the couplings \( g_k \) are non-identical. Since the purity \( \mathcal{P} \) decays exponentially with \( N \) under resonant exchange (figure 1), an exponential enhancement of the purity, \( \mathcal{P} \), is needed for complete polarization. We show that such enhancement is achievable by decohering (disentangling) operations.

The resonant interaction in equation (1) guarantees that all the subspaces (manifolds) are independently purified. However, one must also redistribute the polarization among the subspaces during the cooling process. To this end, one may resort to decoherence/dephasing of the bath spins. This dephasing would lead to population exchange among total-\( I \) subspaces, thereby increasing the mixedness in each subspace. Through repeated dephasing, followed by heat exchange of the probe with a very cold bath, we effectively pump all the population to the highest spin subspace \( I = N/2 \) and only to states with \( \langle I_z \rangle = \pm N/2 \).

Namely, by destroying repeatedly the quantum correlations in the bath, we can fully populate either of the states with \( \langle I_z \rangle = \pm N/2 \), which are the only disentangled eigenstates of the total-spin operator \( I \), corresponding to a fully polarized (purified) spin-bath. Removal of off-diagonal elements through dephasing in the energy-basis is near optimal disentangling operation as it conserves the diagonal basis. Disentangling (decohering) in any other basis also redistributes the population along the diagonal resulting in lower cooling efficiencies (see appendix A.1).

Perhaps counter-intuitively, maximal purity is achieved for any \( N \) through exponential enhancement of \( \mathcal{P} \) as a function of \( N \) by erasing rather than generating quantum correlations. We now show in detail how to apply our approach, consisting of alternating unitary and non-unitary operations, in order to completely polarize spin-baths in solid-state media.

**Figure 1.** The bath purity (log \( \mathcal{P} \)) exhibits an exponential decay with the bath size \( N \). The analytical formula equation (2) is verified by numerical diagonalization (N.D) for \( N \leq 10 \) (green diamonds). Inset: a central spin (NV center) interacting with a spin-bath (consisting of \( ^{13} \)C nuclear spins). The central spin is optically pumped (O.P) by a green laser to dispose of its entropy and coherently manipulated by microwave (MW) pulses. The color bar indicates the strength of coupling between the NV and bath spins.
3. Spin polarization enhancement control in solids

Our approach is quite generally applicable to solid-state spin baths coupled to a spin probe. Full polarization is achievable in two cases of probe–bath coupling, as discussed in the appendices: (i) a nitrogen vacancy (NV) center in diamond acting as a spin probe that is dipole-coupled to a bath of neighboring nuclear spins that are negligibly coupled to each other [3–5, 7, 32] and (ii) an electron in a quantum dot that is hyperfine-coupled to its nearest nuclear spins that have appreciable dipolar interactions [18, 23–25, 33]. For the sake of concreteness, we discuss the NV center case in what follows.

The NV spin–probe (central spin) is a two-level system (TLS) at frequency $\omega_0$ that plays the role of the ‘working fluid’ of the cooling machine. A radio-frequency (RF) or microwave (MW) field at resonance frequency $\omega_0$ drives the TLS with the Rabi-frequency $\Omega_0$ in the rotating frame according to the Hamiltonian

$$H_0(t) = \frac{1}{2}\omega_0 S_z + \Omega_0 \left( S^- e^{i\omega_0 t} + S^+ e^{-i\omega_0 t} \right),$$

where $S_z, S^\pm = \frac{1}{2}(S^x + iS^y)$ are, the respective Pauli spin operators.

The surrounding spins labeled by ‘$k$’ are viewed as a hot bath to be cooled down. They are all identical, two-level systems with energy-splitting $\omega_L$, and are dipole-coupled to the probe via the Hamiltonian

$$H_{SB} = S \otimes B,$$

where the spin-bath operator $B = \sum_k \vec{g}_k \cdot \vec{I}_k$ is the sum over individual spin operators $\vec{I}_k$ weighted by their coupling coefficients $\vec{g}_k$. The coupling between $S$ and $\vec{I}_k$, being magnetic-dipolar, inherently involves both $S_z$ and $S_{x,y}$ terms. However, owing to the large energy mismatch in the Larmor frequencies between the probe (electron) spin, $\omega_0$, and bath (nuclear) spins, $\omega_L$, the dipolar coupling tensor is predominantly oriented along the quantization ($z$)-axis of the probe, i.e., one can rewrite the probe–bath coupling [13, 14, 34] as $H_{SB} = S_z \otimes \vec{B}$.

In the rotating frame of the probe, the probe–bath interaction is then given by

$$H = H_0 + H_{SB}, \quad H_0 = \Omega_0 S_x + \omega_L \sum_k \vec{I}_k^h; \quad H_{SB} = S_z \sum_k \vec{g}_k \cdot \vec{I}_k.$$

By restricting the probe–bath coupling to the $x$–$z$ plane, i.e., setting $\vec{g}_k \cdot \vec{I}_k = g_k^x I_k^x + g_k^z I_k^z$, (we have here omitted the coupling along $y$-direction for simplicity, and will not affect the overall polarization process shown below) and by transforming to the interaction picture [34], where the above the Hamiltonian (equation (4)) simplifies to

$$\hat{H} \equiv e^{-iH_0 t} H e^{iH_0 t} = \sum_k \vec{g}_k \left( e^{-i\Omega_0 \omega_L t} S^z I_k^+ + e^{-i\Omega_0 \omega_L t} S^+ I_k^- + h.c. \right) + \sum_k \vec{g}_k \left( e^{-i\Omega_0 \omega_L t} S^z + h.c. \right) I_k^z.$$

From the above equation one can see that the HH-condition for polarization transfer (swap) between the probe and the bath is fulfilled in the dressed basis by setting (the second resonance condition) $\Omega_0 = \omega_L$, known as the double-resonance transfer (DRT) (figure 2) [34]. In contrast, if we choose a dispersive (off-resonant) coupling between the probe and the bath in the bare basis by setting $\Omega_0 = 0$, then probe-induced energy shifts of the bath spins occur without any polarization transfer. Thus, we can have two contrasting regimes for the combined probe–bath dynamics governed by equation (5)

$$\Omega_0 = \omega_L \Rightarrow \hat{H} \equiv H_{res} = \sum_k \vec{g}_k \left( S^+ I_k^- + S^- I_k^+ \right),$$

$$\Omega_0 = 0 \Rightarrow \hat{H} \equiv H_{disp} = S_z \sum_k \vec{g}_k^z I_k^z.$$

We show here that alternating evolutions governed by equations (6) or (7), respectively, which we dub alternating dispersive-resonant transfer (ADRT) (figure 2(a)) are required to maximize the polarization of the spin bath, as opposed to DRT that only employs resonant exchange (equation (6)).

In addition to the RF/MW control described above, the probe ($S$) spin is subject to optical (laser) pumping: a transition to an electronic excited state followed (through a cascade of vibrational and electronic transitions) by entropy dumping via the coupling of this state to an electromagnetic (EM) bath. This process results in the rapid relaxation (decay) of the probe spin to its ground state, since at optical frequencies the EM-bath is effectively zero-temperature (empty) i.e., has Boltzmann factor $\beta \omega_{\text{opt}} \to \infty$, $\beta$ being the inverse temperature.

As the thermalization of the probe with the cold (EM) bath can occur much faster than that of the spin bath, we can treat these processes separately [34]. Namely, we repeatedly reset the probe to its ground state by optical pumping, and let the spin dynamics be induced either by equations (6) or (7) between
occupied, the evolution should involve all the (2

\[ \frac{\Gamma}{\Gamma} \]

2) evolutions described above. As we start from an initial thermal state, where all the spin states of the bath are exact numerical diagonalizaton results for the cooling process through the coherent and incoherent probe yield the ADRT pulse sequence for spin polarization shown in figure 2(a). In figure 2 we show the interactions of the bath spins with the probe in equation (6), lead to relaxation (polarization) at the

\[ \Gamma_{\text{res}} \] and \[ \Gamma_{\text{disp}} \] respectively. The \[ S^+I^+ \], \[ S^-I^- \] and \[ S^zI^z \] elements of the density matrix, in both the exact numerical analysis and the master equation analysis that we discuss below. Our main point is that the bath correlation time is long enough (in typical systems considered here) to warrant the destruction of the symmetry subspaces by controlled dephasing. Such destruction becomes evident only when we consider the full density matrix, both numerically and analytically by means of our master equation.

In figure 2, we plot the purity of the spin bath as a function of the number of optical pumping cycles that reset the probe spin to its ground state. We see that under the DRT in the absence of microwave-field modulation of the probe, the bath purification is strongly inhibited. In contrast, under the ADRT obtained by modulating the field acting on the probe-spin such that \[ H_{\text{res}} \] and \[ H_{\text{disp}} \] alternately govern the evolution, we attain nearly-complete purification of the bath. These numerical findings concur with our anticipated and analytically-supported insights that correlations generated among the bath spins under the action of \[ H_{\text{res}} \] hinder the polarization exchange with the probe spin, and that switching \[ H_{\text{res}} \] off and \[ H_{\text{disp}} \] on causes their disentanglement, thereby allowing them to gain more polarization from the probe spin (see appendix ).

To gain further insight into these processes, we note that the polarization dynamics of the spin probe becomes quasi-static as the bath polarization saturates, so that the spin probe can no longer transfer polarization, rendering its state unchangeable. Due to the repeated resetting of the probe spin by optical pumping, its correlations to the spin bath are erased, leaving the total state \[ S + B \] uncorrelated at any time \( t \), i.e., \( \rho(t) = \rho_S(t) \otimes \rho_B(t) \), where \( \rho_B \) denotes the spin-bath state. Hence, the probe spin can be traced out to obtain an effective non-Markovian master equation (ME) for the spin-bath state \( \rho_B(t) \) to second order in the couplings \( g \) (see appendix ).

4. DRT and ADRT dynamics of non-interacting spin bath

During the alternating driving of the bath by \( H_{\text{res}} \) and \( H_{\text{disp}} \), the ME dissipators are constructed from the collective spin operators of the bath associated with equations (6) and (7) respectively. The \( S^+I^+ \), \( S^-I^- \) and \( S^zI^z \) interactions of the bath spins with the probe in equation (6), lead to relaxation (polarization) at the respective rates \( \Gamma^+ \) and \( \Gamma^- \), whereas the interaction in equation (7) yields decoherence at the rate \( \Gamma^z \). By resonantly coupling a spin bath to a probe spin under the DRT, a collective quantity akin to the total spin of the ensemble (hereafter denoted as quasi-spin) remains conserved, even though the individual
spin couplings $g^a_1, g^a_2$ may be different. Hence, no population exchange can take place among various manifolds labeled by the total spin $I$, that are associated with the collective-spin angular-momentum operators,

$$\tilde{I}_{\text{res}}^{\pm} = \frac{1}{g_\perp} \sum_k g_k^\pm \tilde{I}_k^\pm; \quad g_\perp = \sqrt{\sum_i (g_i^\pm)^2};$$  \hspace{1cm} (8)

In contrast, upon switching $H_{\text{res}}$ (6) off and $H_{\text{disp}}$ (7) on, the conserved operator becomes,

$$\tilde{I}_{\text{disp}}^{\pm} = \frac{1}{g_\parallel} \sum_k g_k^\pm \tilde{I}_k^\pm; \quad g_\parallel = \sqrt{\sum_i (g_i^\pm)^2};$$  \hspace{1cm} (9)

instead of total quasi-spin. The dynamics that is alternately governed by either $H_{\text{res}}$ or $H_{\text{disp}}$ then leads to population mixing among various quasi-spin manifolds that conserve either $\tilde{I}_{\text{disp}}^{\pm}$ or $\tilde{I}_{\text{res}}^{\pm}$, since the respective invariant manifolds are different.

The collective quasi-spin operators conserve the total quasi-spin $\tilde{I}$, and there exists a complete set of bases $\{|\tilde{I}, \tilde{m}\}$ in which $\tilde{I}$ is diagonal, i.e.,

$$\tilde{I} |\tilde{I}, \tilde{m}\rangle = \tilde{I} |\tilde{I}, \tilde{m}\rangle; \quad \tilde{I} = 0, \ldots, \tilde{I}_{\text{max}}.$$  \hspace{1cm} (10)

Thus, for each $\tilde{J}, \tilde{m}$ may take $2\tilde{J} + 1$ values. The total number of elements is $N = \sum_{\tilde{J},\tilde{m}} d_{\tilde{J}}$, where $d_{\tilde{J}}$ corresponds to $2\tilde{J} + 1$, i.e., there are $d_{\tilde{J}}$ states with the same eigenvalue $\tilde{J}$.

The master equation in this case can be rewritten as a sum of two parts, similarly to what is done in quantum jump approaches [35–37]: one that commutes with the total quasi-spin, and hence is block-diagonal in the basis where $\tilde{I}$ is diagonal, conserving the number of excitations and the other that does not conserve the excitation number

$$\mathcal{L}\rho_h(t) = \mathcal{C}\rho_h(t) + \mathcal{B}\rho_h(t),$$  \hspace{1cm} (11)

where

$$\mathcal{C}\rho_h(t) = \frac{1}{i\hbar} \left( A\rho_h(t) - \rho_h(t) A^\dagger \right),$$  \hspace{1cm} (12)

with the effective non-Hermitian operator

$$A = H_0 - i\hbar \sum_{\tilde{m},\tilde{J} = \pm} \Gamma^m_{\tilde{J}} \tilde{I}_{\text{res}}^{\pm},$$  \hspace{1cm} (13)

and

$$\mathcal{B}\rho_h(t) = 2 \sum_{\tilde{m},\tilde{J} = \pm} \Gamma^m_{\tilde{J}} \rho_h(t) \tilde{I}_{\text{res}}^{\pm}.$$  \hspace{1cm} (14)

We first analyze the term $\mathcal{C}\rho_h(t)$. The operator $A$, as defined in equation (13), clearly commutes with the total quasi-spin $\tilde{I}$. Since $\tilde{I}$ is diagonal in the basis $|\tilde{J}, \tilde{m}\rangle$, we have

$$\langle \tilde{J}, \tilde{m} | A | \tilde{J}', \tilde{m}' \rangle = \tilde{J} \delta_{\tilde{m}} \delta_{\tilde{m}'}.$$  \hspace{1cm} (15)

It then follows from $[A, I] = 0$ that

$$\langle \tilde{J}, \tilde{m} | A | \tilde{J}', \tilde{m}' \rangle = \delta_{\tilde{m}} A_{\tilde{J}}^{\tilde{J}', \tilde{m}'}.$$  \hspace{1cm} (16)

Equation (16) suggests that $A$ is also block-diagonal in the $|\tilde{J}, \tilde{m}\rangle$ bases and satisfies the relation

$$A |\tilde{J}, \tilde{m}\rangle = \sum_{\tilde{m}' = 1}^{d_{\tilde{J}}} A_{\tilde{J}, \tilde{m}}^{\tilde{J}', \tilde{m}'} |\tilde{J}, \tilde{m}\rangle.$$  \hspace{1cm} (17)

In this block-diagonal structure, each block is given by a matrix of size $d_{\tilde{J}} \times d_{\tilde{J}} \equiv (2\tilde{J} + 1) \times (2\tilde{J} + 1)$, where blocks with different $\tilde{J}$ values are not coupled to each other.

The action of $A$ on the elements $|\tilde{J} + \tilde{\nu}, \tilde{m}\rangle$ $|\tilde{J}, \tilde{\mu}\rangle$ can then be evaluated to be [36]

$$\mathcal{C}|\tilde{J} + \tilde{\nu}, \tilde{m}'\rangle \langle \tilde{J}, \tilde{\mu}| = \sum_{\tilde{m}'' = 1}^{d_{\tilde{J} + \tilde{\nu}}} c_{\tilde{m}' \tilde{m}''} \langle \tilde{J} + \tilde{\nu}, \tilde{m}' | \tilde{J} + \tilde{\nu}, \tilde{m}'' \rangle |\tilde{J}, \tilde{\mu}|,$$  \hspace{1cm} (18)
where

\[ \tilde{m} = 1, \ldots, d_{l+\nu}; \quad \tilde{\mu} = 1, \ldots, d_j; \]
\[ \tilde{j}, (\tilde{j} + \tilde{\nu}) = 0, \ldots, \tilde{j}_{\text{max}}. \]

(19)

Since \( A \) does not couple elements with different \( \tilde{j} \), \( C \) also does not couple elements with different pairs of excitations \((\tilde{j} + \tilde{\nu}) \) and \( \tilde{j} \). The operator \( C \) is thus represented by the uncoupled blocks \( C^{(\nu, j)} \), where each block is a tensor of rank 4 with dimension \( d_{l+\nu} \times d_j \times d_{l+\nu} \times d_j \).

To simplify our analysis, we introduce a bijective mapping effected by a two dimensional matrix of size \( D_{\nu, j} \times D_{\nu, j} \) where \( D_{\nu, j} = d_{l+\nu}d_j \). Then, we may show that if \( B = 0 \), the evolution caused only by \( C \rho_B(t) \) results in a block diagonal structure with each block of size \( D_{\nu, j} \times D_{\nu, j} \).

The quantum jump operator \( B \) in equation (11) modifies the dynamics in the following way: the blocks of the jump operator \( B^{(\nu, j)} \) in the eigenbasis of \( C \) form a matrix of dimension \( D_{\nu, j1} \times D_{\nu, j1} \) that connect matrices of dimension \( D_{\nu, j1} \times D_{\nu, j1} \). The number of excitations is changed by \( \pm 1 \), in each step but due to the conserved value of \( \tilde{I} \), the population is confined to each invariant manifold, thus limiting the purity achievable from an initially unpolarized bath to \( e^{-2N/3} \) (equation (2)).

In order to obtain complete mixing among manifolds, one needs to switch off \( H_{\text{res}} \) and switch on \( H_{\text{disp}} \). The master equation for the dispersive interaction is then given by

\[ \dot{\rho}_B(t) = \frac{1}{i\hbar} \left[ H_0, \rho_B(t) \right] + \Gamma_\uparrow(t) \left\{ \left[ \tilde{T}_{\text{disp}}, \rho_B(t) \tilde{T}_{\text{disp}}^\dagger \right] + \left[ \tilde{T}_{\text{disp}}^\dagger \rho_B(t), \tilde{T}_{\text{disp}} \right] \right\}, \]

(20)

where

\[ \tilde{T}_{\text{disp}} = \frac{1}{g_0} \sum_k g_k^\nu \tilde{I}_k^\nu; \quad g_0^\nu = \sqrt{\sum_i (g_i^\nu)^2}, \]

(21)

commutes with the total spin. As the dipolar-couplings \( g_k^\nu \) and \( g_k^\nu \) of a \( k \)th spin to the probe depend on the spatial location and orientation with respect to the probe, they are in general different. Due to this the dephasing operation permits mixing between different blocks (manifolds) and thus overcomes the bottleneck of cooling for resonant swap. As a result, it becomes possible to nearly (fully) polarize the spin bath to its ground state by alternating the resonant and off-resonant (dispersive) coupling of the probe to the bath.

To sum up, the invariant correlation-induced manifolds in the spin bath are periodically destroyed (erased) under ADRT because the alternating dominance of \( \Gamma_{\downarrow}(t) \) or \( \Gamma_{\uparrow}(t) \) effectively projects \( \rho_B \) onto the collective basis-state of \( \tilde{T}_{\text{res}}^\nu \) or \( \tilde{T}_{\text{disp}}^\nu \) respectively. If the couplings \( g_k^\nu \) and \( g_k^\nu \) were the same, the alternation of Hamiltonians (6) and (7) would merely rotate the collective states but do not destroy their collectivity.

Yet, because of the inequivalence of the two collective bases, the ADRT modulation of the spin-probe level distance by \( \Omega(t) \) between \( \omega_d \) and 0 allows unrestricted population mixing among all bath manifolds, followed by the cooling down of the spin-bath via HH resonant transfer and spin–probe entropy dumping. In the long-time limit, ADRT allows the spin-bath to reach a steady-state. The final effective bath-temperature and the rate at which it is cooled depend on the inverse temperature of the cold (EM) bath to which the probe spin is coupled. Since the cold bath satisfies \( \beta \omega_{\text{disp}} \rightarrow \infty \), and a steady constant rate of polarizing the probe spin is reached, the cooling rates of the bath also reach their asymptotic limits, i.e., \( \Gamma_{\downarrow} \rightarrow g_{\downarrow}, \Gamma_{\uparrow} \rightarrow g_{\uparrow} \), whereas \( \Gamma_{\downarrow} \rightarrow 0 \) (see appendix ). The master equation then yields decay to the ground state of the spin-bath.

The superiority of ADRT over DRT is conspicuous (figure 2(b)). The effect of two competing alternating quasi-spin bases that are formed by the probe–bath alternating resonant and dispersive interactions can purify the spin-bath to their common eigenstate \( |N/2; \pm N/2 \rangle \) i.e., to a fully polarized state.

5. Interacting spin-bath dynamics

Competing bases may also be obtained when considering an interacting spin system. Let us consider a scenario where a central spin is uniformly coupled to a spin-bath, wherein the spins interacts with each other. In the rotating frame with \( \omega_0 = \omega_L \), we then obtain the resonant exchange Hamiltonian, which now includes the intra-bath spin–spin interactions i.e.,

\[ H = S^+ \sum_k g_k \tilde{I}_k^- + \sum_{ij} \tilde{l}_{ij} \tilde{l}_{ij}^\dagger + \text{h.c.} \]

(22)

For either of the terms in this Hamiltonian, the fully polarized state \( |N/2; \pm N/2 \rangle \) is an eigenstate. The symmetrized basis for the evolution of the spin-bath depends on the relative strengths of \( g_k \) and \( \tilde{l}_{ij} \).
Polarization transfer is incomplete when either of these terms dominate the dynamics, since the basis consists of collective states, that compete with each other, so that their only common eigenstate is the ground state \( |N/2, -N/2 \rangle \). This anticipation is confirmed by exact numerical diagonalization of the above Hamiltonian in figure 3. The role of spin–spin interactions among the nuclear spins in the bath may thus be similar to that of ADRT, namely, spin–spin interactions enable full purification. As the interactions lead to the formation of different total-\( I \) subspaces, the symmetry imposed by DRT is invariably broken. In comparison with ADRT, the purity gain of interacting spins is slow, but eventually saturates to the fully polarized state of the bath.

We note that the key requirement for the interaction-enhanced polarization of the bath is that the target state be a common eigenstate of both the spin-probe and spin–spin Hamiltonian. Only then can the polarizing probe–bath interaction Hamiltonian and the spin–bath Hamiltonian progressively evolve the bath to the true steady state where no further dynamics occurs, as shown in figure 3. The inhomogeneous spin–spin interactions have been chosen here to be near optimal for attaining this steady-state efficiently. Any randomly picked spin–spin interaction may still enhance cooling, but with a much lower efficiency.

6. Conclusions

We have considered here the common scenario wherein quantum correlations (entanglement) induced among the bath spins by their common coupling to a spin probe persist over long times. These correlations lead to a strong deviation of the spin bath from the initial thermal state as the spins resonantly interact with the probe, giving rise to complex many-body dynamics. The spins are driven into invariant manifolds that inhibit any further mixing among these manifolds due to their coupling to the spin probe, thus preventing the bath cooling. Hence, only by frequently obliterating the correlations among the bath spins may one achieve maximal cooling of the bath. To this end, we have introduced the unconventional sequence we dub alternate dispersive-resonant transfer (ADRT) wherein disentanglement of the spin bath (by modulating the spin-probe energy) alternates with the common flip-flop regime (the HH double resonance transfer—DRT). While DRT leads to a spin–bath polarization (purity), that decreases exponentially with the size of the bath, ADRT can nearly-fully polarize it. The present insights hold for interacting spin baths, where spin–spin interactions have been shown to yield effects similar to ADRT. Thus, in the currently trendy \(^{13}\)C nuclear spin-bath around a single NV center, only incomplete polarization, up to \( \sim 25\% \), has been reported [38]. In addition to technical reasons, such as pulse errors and magnetic field fluctuations, we have here identified and suggested the bypass of a fundamental bottleneck caused by the inevitable quantum correlations among the spins in the ensemble (bath). Further, we anticipate the incomplete polarization of external spins in recent NV experiments for non-interacting spins [39] can be drastically increased through ADRT shown here.

Figure 3. (a) Comparison of spin bath purification for (i) interacting spins (blue-dashed), (ii) non-interacting spins under ADRT (red-dotted), and (iii) non-interacting spins under DRT (black solid), obtained numerically are shown. (b) The role of spin–spin interactions among the nuclear spins in achieving the maximal purity, is numerically shown to be similar to that achieved by ADRT. We consider a bath of six spins, where the \( S–I \) and \( I–I \) interactions (see SI [https://stacks.iop.org/NJP/22/083035/mmedia]) are given by \( g_{ij} = g_{ij} [1, 1, 1, 1, 1, 1] \), and nearest-neighbor coupling among the bath spins with \( J_{ij} = \alpha [0, 8, 1, 0, 1, 2, 1, 0, 1, 2, 1, 0, 1, 2, 1] \). The strength of coupling \( \alpha \) in the three cases shown are respectively (denoting \( \alpha \sim \alpha \) \( J_{ij} = 1 \) \( g \sim J \), \( \alpha = 0.1 \) \( g > J \)), \( \alpha = 10 \) \( g >> J \), and only DRT interaction between the bath and the probe. With the inhomogeneous coupling chosen among the bath spins, the unique common eigenstate of the Hamiltonian is \( \uparrow \downarrow \cdot \cdot \cdot \cdot \cdot \) i.e., a fully polarized bath, whose polarization is similar to that of the central spin. (c) Numerical simulations of similar protocols (shown in figure 2) applied to the case of nuclear spins that are hyperfine-coupled to either electron (E)- or hole (H)-spins in a semiconductor quantum dot. The protocol analogous to ADRT is realized by modulating optical pulses, thereby causing nuclear spins to evolve with random phases through hyperfine coupling to both electron- and hole-spins (red line). The analog of DRT is realized when the nuclear spins evolve only under coupling to the electron spin (blue dashed-line). The analog of ADRT shows great improvement over the analog of DRT.
We have used exact numerical diagonalization techniques to analyze the evolution of the cooling process, and a non-Markovian master equation to generalize the cooling procedure to diverse baths (bosons, magnons, fermions), and obtained analytically the cooling and dephasing rates in terms of Hamiltonian parameters. While the density matrix evolution can be solved numerically for \( N \sim 10 \) spins, the calculation can be extended to a larger number of bath spins by using MonteCarlo random wave-function methods [47], but the computation complexity involved for initial thermal states still remains the same.

Controlled destruction of correlations (disentanglement) among quantum systems can be key to understanding the thermalization of quantum systems coupled to finite baths, and the observed equilibration of quantum systems. The proposed strategy may be highly useful in reducing the noise produced by the surrounding electronic or nuclear spin bath on a probe spin by polarizing these spin baths in probed samples. Further, the proposed polarization of random spin ensembles can have impact on understanding the role of quantum correlations in charging and discharging of quantum batteries [26]. As the sensitivity of the NV magnetometer can be boosted by a locally polarized spin-bath [40], attaining full polarization would have important consequences for quantum sensing, magnetic imaging and spectroscopy, metrology and quantum information processing schemes [1–4, 7, 32, 41–46].

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Appendix A. Purity of the spin-bath

For the ideal case scenario wherein a spin probe is coupled to \( N \)-identical spins homogeneously, the resonance Hamiltonian \( H_{\text{res}} \) can be diagonalized in the total-\( I \)-basis of the spin-bath. Due to this the spin-bath is polarized in each \( I \) sector independently. The purity, \( \langle \text{Tr} \rho^2 \rangle_I \) of the bath in each spin sector is given by

\[
P_I = \frac{(2I + 1)^2}{(2N)^2}.
\]

Summing over all the spin sectors gives the total purity of the bath

\[
P = \sum_I \lambda_I P_I.
\]

For example for \( N = 2 \), there are four spin states and two spin sectors \( I = 1 \) and \( I = 0 \). The maximum achievable purity of the bath in each of these spin sectors is respectively \( 9/16 \), \( 1/16 \), and hence the total purity is \( P = 0.625 \). Similarly for \( N = 4 \) there are 6 spin sectors with one \( I = 2 \), three \( I = 1 \) and two \( I = 0 \). Similar to the above analysis, the respective purities in each of the spin sectors is \( P_2 = 25/256 \), \( P_1 = 9/256 \), and \( P_0 = 1/256 \), giving the final purity of \( 25/256 + 3 \times 9/256 + 2 \times 1/256 = 54/256 \).

A.1. Short-time behavior

To see how correlation hinders the polarization of any \( (k) \)th spin in the bath we perform a short-time analysis of the spin-bath operators. The time-evolution of any spin-bath operator \( O_I \) induced by Hamiltonian \( H \), can be expanded in powers of \( t \) as

\[
O_I(t) \equiv e^{iHt} O_I e^{-iHt} \approx O_I + it[H,O_I] - \frac{t^2}{2}[H,[H,O_I]] + \cdots .
\]

The general Hamiltonian leading to the resonant-exchange interaction among the electron and nuclear spins given in equation (1) can be obtained, for example, starting from

\[
\mathcal{H} = \omega_e S^z + \sum_k \omega_n F^z_k + \sum_k g_k S^z I^z_k,
\]

where \( \omega_e \) and \( \omega_n \) are the spin-resonance frequencies of the electron and nuclear spins respectively. By transforming to the interaction picture and setting \( \omega_e = \omega_n \), we obtain the flip-flop (HH) Hamiltonian given
in equation (3) \cite{48}. Using it, we find the short-time evolution of the single-spin and the two-spin operators to be

\[ O_i = I_i^z(t) \approx I_i^z - t^2\left(g_{kx}^z(I_i^z - S^z) + \sum_{m \neq k} g_{km}^z g_{km}^z S^z(I_m^z + h.c)\right), \]

\[ O_{ii} = I_i^z I_m^z(t) \approx I_i^z I_m^z + g_{km}^z g_{km}^z t^2 \left[S^z(I_k^z + I_m^z) + I_k^z I_m^z\right]. \]  \hspace{1cm} (26)

The short-time probability of the \(k\)th bath-spin polarization, \(P_k(t) = \langle \hat{F}(t) \rangle\) and the two-spin correlator, \(\langle I_i^z I_m^z \rangle\), is then

\[ P_k(t) \approx P_k(0) + g_{km}^z g_{km}^z t^2(P_k(0) - P_k(0))) - g_{km}^z t^2 \left[P_k(0) \sum_{m \neq k} \left(g_{km}^z g_{km}^z I_i^z I_m^z + h.c\right)\right]_0, \]  \hspace{1cm} (27)

\[ \langle I_i^z I_m^z \rangle \approx \langle I_i^z I_m^z \rangle_0 + g_{km}^z g_{km}^z t^2(S^z(I_k^z + I_m^z) + I_k^z I_m^z) \]  \hspace{1cm} (28)

where \(P_k(0) \approx 1\) is the polarization of the probe spin after each resetting, and the subscripts 0 and \(t\) refer to averages with respect to the bath state at times \(t = 0\) and later \(t\). If the bath spins have nonzero polarization, \(P_m > 0\), then the two-spin correlators of the bath \(\langle I_i^z I_m^z \rangle\) do not vanish (to fourth order) and modulate the single-spin polarization \(P_k(t)\). Two competing effects arise: that of the first term in the square brackets of (26) enhances the spin-bath polarization (as \(P_k(0) > P_k(0)\)), and that of the second term therein tries to reduce it. We thus find a saturation of the polarization transfer from the spin probe to the bath spins at a value \(< P_k(0)\). Only upon erasing these correlations by the inhomogeneous dispersive interaction \(H_{\text{disp}}\), we may monotonically increase the polarization of the bath spins toward their maximum value \(P_k(0) \approx 1\), as shown in figure 1.

### A.2. Disentanglement basis

As argued in the main text, disentangling (dephasing) operations in the energy basis are optimal for achieving a high degree of polarization in the energy basis of the spin-bath. While disentanglement can be achieved by dephasing in various other bases, such processes may not satisfy the optimal cooling conditions described here. As an illustration, we may consider a bath composed of spins and show that the asymptotic cooled state of the bath depends on disentangling operations in various bases:

\[ \rho_0 = \frac{1}{4} |S_0\rangle \langle S_0| + \frac{3}{4} |\uparrow\!\downarrow\rangle \langle \uparrow\!\downarrow| \] (no disentanglement)

\[ \rho_Z = |\uparrow\!\downarrow\rangle \langle \uparrow\!\downarrow| \] (disentanglement in \(Z\) basis)

\[ \rho_X = \frac{1}{4} I \] (disentanglement in \(X\) basis)

\[ \rho_{ZX} = |\uparrow\rangle \langle \uparrow| \otimes \frac{1}{2} I \] (disentanglement in \(ZX\) basis) \hspace{1cm} (29)

where \(|S_0\rangle = \frac{1}{2} \left(|\uparrow\!\downarrow\rangle - |\downarrow\!\uparrow\rangle\right)|\rangle\) is the singlet state. Clearly, disentanglement (decoherence) in the \(Z\)-basis is optimal for polarizing both spins. One may choose any random bases for both spins, and easily verify that the polarization is incomplete except in the energy (\(Z\)) basis.

### Appendix B. Non-Markovian master equation analysis for a non-interacting spin-bath

The non-Markovian behavior of the spin-bath can be found using the time-convolutionless (TCL) master equation \cite{49}. Due to the rapid thermalization of the spin by the cold bath, one can neglect its correlations with the spin-bath. Under this approximation we can write down the total state of the S–B system as \(\rho(t) = \rho_S(t) \otimes \rho_B(t)\). Further, the state of the S spin at any \(t\) is determined by its Markovian dynamics with the cold bath i.e., \(\rho_S(t) = e^{-\gamma_C t} |1\rangle \langle 1| + \left(1 - e^{-\gamma_C t}\right) |0\rangle \langle 0|\), where \(\gamma_C\) is the probe relaxation to the cold bath. Using these approximations, one can obtain the master equation for the spin bath in the form

\[ \dot{\rho}_B(t) = -\text{Tr}_S \int_0^t ds [H_i(t), [H_i(s), \rho_S(t) \otimes \rho_B(t)]]]. \]  \hspace{1cm} (30)

Expanding the above equation to second order in the coupling strength and substituting the state \(\rho_B(t)\), we get the following master equation in the Schrödinger picture
spins are also interacting. The Hamiltonian describing this interaction is given by

\[ \rho_\text{p}(t) = \mathcal{L}_\text{p}(t) = \frac{1}{i t} \{ H_\text{p}, \rho_\text{p}(t) \} + \sum_{m\pm k=\pm} \Gamma_m(t) \{ [\vec{I}_\text{res}^m, \rho_\text{p}(t)], \vec{I}_\text{res}^m \} + \Gamma_z(t) \{ [\vec{I}_\text{disp}, \rho_\text{p}(t)], \vec{I}_\text{disp} \}. \]

In the above equation

\[ \vec{I}_\text{res}^\pm = \frac{1}{g_{\perp}} \sum_k g_k^\pm I_k^\pm; \quad \vec{I}_\text{disp} = \frac{1}{g_\parallel} \sum_k g_k^\parallel I_k^\parallel, \]

where \( g_{\perp} = \sqrt{\langle g^\parallel \rangle^2}; g_\parallel = \sqrt{\langle g^\parallel \rangle^2} \). Interaction with the probe leads to both dissipation and dephasing within the spin ensemble at rates given by

\[ \Gamma_+(t) \approx g_{\perp} \text{Re} \int_0^t dt' \exp[i\omega(t - t')]|\epsilon(t)|\epsilon'(t')(1 \pm P_3(t)), \]

\[ \Gamma_z(t) \approx 2g_\parallel \text{Re} \int_0^t dt' \exp[i\omega(t - t')] \]

where \( \epsilon(t) = \int_0^t dt' \exp[i\Omega(t')]; \Omega(t) \) being the time-dependent Rabi-frequency of the driven probe spin.

**Appendix C. Cooling down an interacting spin bath**

We have shown earlier that the effect of two competing angular momentum bases with a common eigenstate would allow the spin-bath to purify to that state when interacting with a polarizable central spin to which the spin-bath is coupled. We have considered the two competing bases formed by the resonant and dispersive interaction and have shown that the spin-bath would purify to their common eigenstate \( |N/2; \pm N/2 \rangle \) i.e., to a fully polarized state. Such competing bases may also be obtained when considering an interacting spin system, as detailed below.

Let us consider a scenario where a central spin is interacting uniformly with a spin-bath, wherein the spins are also interacting. The Hamiltonian describing this interaction is given by

\[ H = \omega_S S^z + \omega_I \sum_i I_i^z + \sum_k g_k S^z I_k^z + \sum_{i,j} J_{ij} I_i^+ I_j^-. \]

In the rotating frame with \( \omega_S = \omega_{1/2} \), we obtain the resonant exchange Hamiltonian, which now also includes the spin–spin interactions i.e.,

\[ H = S^+ \sum_k g_k I_k^- + \sum_{i,j} J_{ij} I_i^+ I_j^- + \text{h.c.} \]

For either of the terms in this Hamiltonian, the fully polarized state \( |N/2; \pm N/2 \rangle \) is an eigenstate. The symmetrized basis for the evolution of the bath is dominated by the diagonal bases of either of the terms in the above equation, depending on the relative strengths of \( g_k \) and \( J \). Hence, polarization transfer is expected to be incomplete when either of these terms dominate the dynamics. In contrast, perfect polarization is anticipated when these terms are of the same order. The values of \( J \) and \( g \) used here are experimentally adjustable, as shown in [50]. These couplings, for example in the case of NV centers are determined by the \( ^{13}\text{C} \) concentration in the diamond, which can be manipulated during the growth process [50]. The distributions in the \( g \)-couplings range from 500 kHz to 10 Hz, and in order to achieve a nuclear spin-bath with strong dipolar couplings kHz, one can use a thick diamond layer consisting of \( ^{13}\text{C} \) nuclear spins doped with nitrogen-vacancy centers (NV) that are embedded in a spin-free \( ^{12}\text{C} \) crystal matrix [50].

**Appendix D. NV center in diamond: dipole coupled probe–bath** [3–5, 7, 32, 42, 46]

Any NV center consists of a substitutional nitrogen atom and a vacancy occupying adjacent lattice sites in the diamond crystal. The electronic ground state of the NV center is a spin triplet, in which the \( m_\text{s} = 0 \) and \( \pm 1 \) sublevels are separated by a \( \omega_0 \sim 2.87 \text{ GHz} \) zero-field splitting. A magnetic field can lift the \( m_\text{s} = \pm 1 \) level degeneracy. Decay from \( m_\text{s} = -1 \) to the \( m_\text{s} = 0 \) level is induced by a three-step process: optical excitation, then inter-system crossing to a singlet-state, followed by phononic relaxation. As this process is irreversible one can assign a Markovian decay rate for the initialization of the NV center to the \( m_\text{s} = 0 \) state. Optically-induced initialization of the NV to the \( m_\text{s} = 0 \) sublevel of the electronic ground state followed by microwave driving of the \( m_\text{s} = 0 \leftrightarrow m_\text{s} = -1 \) transition restricts the dynamics to an effective TLS with resonance frequency \( \omega_0 \). It can be controlled by a (green) continuous-wave or pulsed laser field, effectively...
inducing decay from the $m_s = -1$ to the $m_s = 0$ level in the TLS. We have assumed instantaneous resetting of the probe since the relevant time-scale for optical polarization of the probe is experimentally $\sim 1000$ times faster than the probe–bath dynamics.

A typical diamond sample has a natural abundance of 1.1% of $^{13}$C atoms which form the nuclear spin bath for the NV center. This results in an average distance of 2–3 nm between any two $^{13}$C atoms, and an average dipolar coupling of $\sim 1–10$ Hz. Clearly, the intra-nuclear interaction is much weaker and can be safely neglected. The average dipolar coupling between the NV centers and its nearest $^{13}$C spin can range from $1–100$ kH, and is much larger than the intra-nuclear coupling in the spin-bath that may lead to diffusion of spin excitation. The accessible number of bath spins that interact with the NV center well within their lifetime is in the range of $10^5$. As the nuclear spins are randomly distributed, their coupling to the NV center contains transverse couplings, $g_{kS}^T$, that lead to polarization transfer between the NV and the nuclear spins while the longitudinal couplings determined by $g_{kS}^L$ lead to dephasing of the bath.

The time-scales relevant for the experimental implementation are determined by the exchange interaction given by equation (6) and the dispersive interaction given by equation (7). The exchange interaction time ($\tau_e$) can be fixed to be well within the $T_2$ time of the NV center. We aim at erasing all the coherences among the nuclear spins, via the inhomogeneous coupling between the NV and the spin bath to mimic the random gradient fields used in the NMR experiments. To enhance the effect, we randomize the evolution time $\tau_d$ under $H_{\text{disp}}$ instead of choosing a constant time for evolution. This truly mimics randomizing the coupling and leads to perfect dephasing. The time-scale of the dispersive interaction ($\tau_d$) may be chosen randomly in the range $T_2 \leq \tau_d \leq T_1$, as the electron spin-coherence is not required during the evolution with $H_{\text{disp}}$.

D.1. Experimental implementation of ADRT

Our improved spin polarization scheme uses the alternating double resonance transfer (ADRT) wherein the system dynamics is alternating between two Hamiltonian evolutions by varying the strength of the microwave power that alters the resonance condition. In the simulations shown in figures 2 and 3, we have used a periodic step-function for the microwave power $\Omega$, where it is switched on/off between the coherent and incoherent parts of the evolution. While this is more experimentally friendly, as most of the pulse control employs stroboscopic switching of the fields. As the microwave can be switched on/off on nanosecond time scales, they can be considered step-like functions given the microsecond time scale for spin-bath evolution and probe polarization. While a smooth variation of $\Omega$ (cosine or sine functions) would still work, simulating its effect requires solving the time-dependent Schrödinger equation. For the bath of 10–12 spins considered here it is computationally demanding, and hence only a periodic on–off pulses are used. Further to optimize the speed of polarization transfer, we set the time scale to the value for which the polarization transfer between the probe and the bath is maximized. This value is given by the inverse root mean square coupling of the bath to the probe.

Appendix E. Quantum dot: contact hyperfine-coupled probe–bath [18, 23–25, 33]

The transfer of optical polarization from the electron to the nuclear spins in quantum dots is more complex than for the case of the NV centers in diamond as it involves both the electron and hole states in the optical excitation process. The optical excitation couples the hole states to the trion (electron) states whose contact hyperfine interaction with the nuclear spins allows them to be polarized by flip-flop terms. The trion states rapidly decay to the hole state and then re-excited by the next optical pulse to the trion state with the electron spin polarized. This polarization is again transferred to the nuclear spins by the hyperfine interaction with the nuclear spins that may lead to diffusion of spin excitation. The accessible number of bath spins that interact with the NV center well within their lifetime is in the range of $10^5$. As the nuclear spins are randomly distributed, their coupling to the NV center contains transverse couplings, $g_{kS}^T$ that lead to polarization transfer between the NV and the nuclear spins while the longitudinal couplings determined by $g_{kS}^L$ lead to dephasing of the bath.

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