

Spin coherence in strongly coupled spin baths in quasi-two-dimensional layers

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We investigate the spin-coherence decay of NV^- spins interacting with the strongly coupled bath of nitrogen defects in diamond layers. For thin diamond layers, we demonstrate that the spin-coherence times exceed those of bulk diamond, thus allowing to surpass the limit imposed by high-defect concentrations in bulk. We show that the stretched-exponential parameter for the short-time spin-coherence decay is governed by the hyperfine interaction in the bath, thereby constraining random-noise models. We introduce a method based on the cluster-correlation expansion applied to strongly interacting bath partitions instead of individual spins. Our results facilitate material development for quantum-technology devices.

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Spin coherence is key for quantum-technology applications but the interaction of a spin with the surrounding bath typically leads to the loss of its spin coherence, i.e., its transverse polarization [1,2]. Generally, the dynamics in the bath creates a noisy magnetic field for the spin and dephases it. For weakly coupled spin baths, where the dynamics of the bath is slow compared to the central-spin dynamics, suitable theoretical approaches include (Gaussian) random-noise models [3–7], approximate analytical descriptions [8–13], and numerical simulations [10,14–19]. In strongly coupled spin baths, however, where the intrabath and the center-to-bath interactions are of similar strength, the nonequilibrium many-body spin-coherence dynamics is not easily accessible by analytical or numerical techniques. The reasons are the absence of small parameters for a perturbative series expansion, the exponential scaling of resources for exact numerical simulations and the complex, even non-Markovian, bath dynamics. Such central-spin problems become increasingly important due to their relevance for spin-based quantum technologies [20–23] as, for example, quantum sensing [24].

For the nitrogen-vacancy (NV^-) center in diamond [25], which offers immense potential for quantum technologies [26–38], the spin-coherence decay is mainly governed by the interaction with spins of single-substitutional nitrogen defects (called N_s^0 or P1 centers) [10] and the ^{13}C nuclear spins [39]. The P1 centers, each comprising an electron spin and a nuclear spin, usually dominate the spin-coherence decay of NV^- centers even at comparatively low concentrations

of about 1 ppm due to the large electron-spin gyromagnetic ratio. To suppress the interaction with P1 centers, the material properties of diamond samples can be enhanced [40], for example, by using delta-doping diamond-growth techniques [41–43] which allow for a precise depth placement of

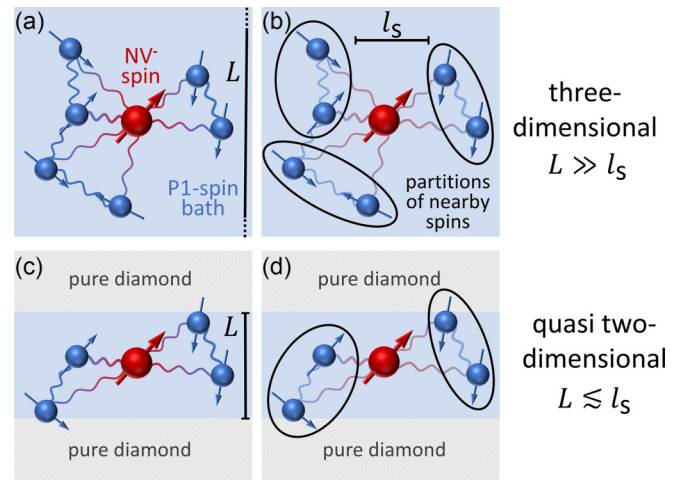


FIG. 1. Partitioning the P1-center spin bath of different spatial dimensionality around the NV^- center. Sketch of a three-dimensional (a) and quasi-two-dimensional (c) P1-center spin bath, which differ only by the layer thickness L . The relation between the layer thickness L and the mean nearest-neighbor spin distance l_s determines the dimensionality of the bath: $L \gg l_s$ for the three-dimensional and $L \lesssim l_s$ for the quasi-two-dimensional case. Wavy lines indicate the interaction between the spins. For the pCCE method, we first partition the bath into local strongly interacting indivisible partitions. As an example, two-spin partitions are indicated by the ovals in parts (b) and (d). Then, the conventional CCE method is applied to the partitions. Only the electron spin of the P1 centers is indicated in the figure, nuclear spins are omitted.

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defects with layer thicknesses in the nanometer range. In such thin layers, the P1-center spin bath is typically quasi-two-dimensional, leading to longer Hahn-echo T_2 times [23,44–46]. The theoretical analysis of such quasi-two-dimensional strongly coupled baths is particularly difficult due to large fluctuations of dipolar interaction coefficients [47–49].

In this letter, the results can be divided into three main parts: (i) In the effort to improve diamond samples for quantum-technology devices, we numerically simulate the spin-coherence decay of NV[−]-spin ensembles in strongly coupled P1-center spin baths in diamond layers of different thickness and P1-center concentration, thereby identifying parameters for extended spin-coherence time. In particular, we calculate the spin-coherence times T_2 in quasi-two-dimensional diamond layers, when the mean nearest-neighbor spin distance l_s satisfies $L \lesssim l_s$ (cf. Fig. 1), and show that they exceed those in bulk diamond. (ii) We show that the stretched-exponential parameter p for the short-time spin-coherence decay is governed by the hyperfine interaction of the P1 centers, which strongly modifies the bath-spin dynamics (see, for example, Refs. [10,50]). In particular, predictions from bath-dynamics models based on the Ornstein-Uhlenbeck process [23] are valid only when neglecting the nitrogen nuclear spins of the P1 centers in the numerical simulations. These results indicate the complexity of the spin dynamics in P1-center baths and offer direct means to identify more accurate effective magnetic-noise models for the description of P1-center spin baths [51]. Moreover, these results are essential for efforts to suppress the magnetic noise generated by the spin bath. (iii) To overcome difficulties of available methods when applied to strongly coupled baths of different dimensionality, we introduce a numerical method used in the above calculations, which we name partition-CCE (pCCE), cf. Fig. 1. This method divides the spin bath into local strongly interacting subsystems, which we refer to as partitions and treat as indivisible units. Then, the cluster-correlation expansion (CCE) [16] is applied to the partitions instead of individual spins. In this way, the method takes into account important higher-order spin correlations leading to improved accuracy, broader range of applicability, and enhanced convergence properties.

We consider ensembles of NV[−]-center electron spins (we omit the minus sign in the following) in isotopically purified ¹²C diamond layers with a sparse disordered P1-center spin bath of concentration ρ_{P1} . A strong magnetic field parallel to the NV axis defines the z axis. In the rotating reference frame, the P1-center electron spins interact via the secular part of the dipolar Hamiltonian [1,2]

$$\mathcal{H}_{P1} = \sum_{i>j\geq 1} J_{ij} \left(I_{iz} I_{jz} - \frac{1}{4} [I_i^+ I_j^- + I_i^- I_j^+] \right), \quad (1)$$

where I_{jz} is the z -projection operator for the j th P1-center electron spin, I_j^+ (I_j^-) is the corresponding spin raising (lowering) operator, and $J_{ij} \cong 1/r_{ij}^3$ is the dipolar coupling constant with r_{ij} being the distance between spins i and j . For each P1 center, the hyperfine interaction with the nitrogen nuclear spin reads $\mathcal{H}_{hf,j} = A_j I_{jz} S_{jz}$, where S_{jz} is the spin-1 z -projection operator of the nuclear spin and A_j is the hyperfine interaction constant, which depends on the Jahn-Teller axis alignment [10,50]. An energy mismatch generally suppresses the

flip-flop transitions between the NV- and P1-center electron spins such that the corresponding interaction Hamiltonian reads [1,2] $\mathcal{H}_{int} = \sum_j J_{0j} I_{0z} I_{jz}$, where the index 0 corresponds to the NV spin. We quantify the spin coherence of the NV spin by $\langle M_x(2\tau) \rangle \equiv \langle I_{0x}(2\tau) \rangle / \langle I_{0x}(0) \rangle$, where $\langle I_{0x}(2\tau) \rangle$ is the x magnetization of the NV spin after a Hahn echo sequence [52]. For a more detailed description of the system, see Supplemental Material [53].

The interaction with the P1-center spin bath described by $\mathcal{H}_{int}$ leads to a time-dependent magnetic field for the NV spin $B_z^{P1}(t) \cong \sum_j J_{0j} \langle I_{jz}(t) \rangle$, where $\langle I_{jz}(t) \rangle$ is the expectation value of I_{jz} . The characteristics of $B_z^{P1}(t)$ are governed by the dynamics of the P1-center bath, which is in turn dominated by the flip-flop transitions described in Eq. (1). Generally, the Hahn echo sequence cannot fully refocus an initial polarization of the NV spin for such time-dependent magnetic fields. Instead, the NV spin dephases in the (x, y) plane corresponding to the decay of the spin coherence $\langle M_x(2\tau) \rangle$.

The CCE method approximates the spin-coherence evolution $\langle M_x(2\tau) \rangle$ of a central spin in a large spin bath by including the contributions of *all possible* spin clusters in the bath of size $n \leq N$ (CCEN) $\langle M_x(2\tau) \rangle \approx \langle \langle \tilde{\mathcal{L}}_0 \tilde{\mathcal{L}}_1 \tilde{\mathcal{L}}_2 \cdots \tilde{\mathcal{L}}_N \rangle \rangle$, where $\langle \langle \dots \rangle \rangle$ denotes the average over mean-field spin configurations [53], $\tilde{\mathcal{L}}_n \equiv \prod_i \tilde{\mathcal{L}}_{ni}$ (i counts all possible n -spin clusters) and

$$\tilde{\mathcal{L}}_{ni} \equiv \frac{\langle M_x(2\tau) \rangle_{ni}}{\prod_{B \subset C_{ni}} \tilde{\mathcal{L}}_B}, \quad (2)$$

where B runs over all possible subclusters of C_{ni} . $\langle M_x(2\tau) \rangle_{ni}$ is obtained by calculating the quantum dynamics only of spins in the cluster C_{ni} , while all other bath spins are included on the mean-field level, i.e., they are static and point either up or down along the z direction [54–56].

In practical calculations of the spin coherence of an NV electron spin in the weakly coupled bath of nuclear spins, CCE2 is often used (see, for example, Refs. [44,57]), which includes the dynamics of single spins and spin pairs in the bath. However, in the strongly coupled P1-center spin bath, the dynamics in the bath takes place on the same timescale as the spin-coherence decay of the NV spin and, hence, correlated bath dynamics of higher order than for the nuclear-spin bath must usually be taken into account. Therefore, higher order CCEN ($N > 2$) need to be applied, which, however, often show unphysical behavior for strongly coupled spin baths [54,55], when, for example, $|\langle M_x(2\tau) \rangle| > 1$ for an initially fully polarized NV spin. This can occur, for example, when clusters in the CCE method include *only a part of a strongly-interacting spin group*. In principle, this issue can be corrected for by further increasing N but, in practice, this in turn introduces even more clusters with unphysical behavior.

The main idea of the pCCE method is to divide the disordered spin bath into strongly interacting local subsystems (partitions) and to apply the conventional CCE method to the partitions instead of individual spins, cf. Fig. 1. The partitions are chosen such that the interaction inside the partitions is maximized. These partitions act as indivisible units, when applying the CCE method [53], and thereby facilitate capturing the correct spin dynamics. We choose for simplicity partitions with an equal number of spins K (partition size) and adopt the

notation $\text{pCCE}(N, K)$, when applying CCEN after partitioning the bath into K -spin partitions. Using $\text{pCCE}(N, K)$, the clusters formed are unions of up to N partitions. For example, in $\text{pCCE}(2, 2)$, CCE2 is applied to partitions of strongly interacting spin pairs as illustrated in Fig. 1, and in $\text{pCCE}(2, 4)$, each partition contains 4 spins such that the clusters considered are either the 4-spin clusters or the union of two 4-spin clusters [53]. Important to notice is also that $\text{pCCE}(N, 1)$ is equivalent to CCEN.

The pCCE method offers several advantages by construction. At sufficiently large K , each strongly interacting spin group is included in a single partition, thereby capturing important higher-order spin correlations. Moreover, in contrast to the CCE method, the number of samples for the mean-field average decreases with increasing K , cf. Fig. 2 (for details about the mean-field average, see Supplemental Material [53]). Further, $\text{pCCE}(N, K + 1)$ is not based on results of $\text{pCCE}(N, K)$ because the partitioning of the bath significantly changes. Therefore, to increase the accuracy of results at larger K , the pCCE method does not rely on correcting inaccuracies of results at smaller K . Hence, the order K is effectively unrestricted. Another advantage is the significantly reduced number of clusters included. For example, in a simple comparison between $\text{pCCE}(N, K)$ and $\text{CCEN}K$ [53], the number of largest clusters of size NK is $\binom{N_s/K}{N}$ and $\binom{N_s}{NK}$, respectively, where N_s is the total number of bath spins. For example, for $N_s = 80$, $N = 2$, and $K = 4$, the number of clusters is reduced by the factor 10^{-8} .

To demonstrate the performance of the pCCE method, $\langle M_x(2\tau) \rangle$ for the NV spin interacting with a small disordered quasi-two-dimensional bath of 20 electron spins-1/2 is shown for different values of K in Fig. 2. With increasing partition size K , the pCCE results converge towards the exact solution and become almost indistinguishable from the exact solution at $K = 4$. This demonstrates the power of the pCCE method to capture the relevant dynamics and even approach the exact solution with a relatively low partition size K .

Extensive tests of the $\text{pCCE}(2, K)$ performance for large spin baths show that $K = 4$ is required for the method to be sufficiently accurate for all systems considered. The most demanding systems requiring $K = 4$ are quasi-two-dimensional layers of P1 centers without the hyperfine interaction, i.e., electron spins-1/2, which implies that the correlated spin dynamics extends over local spin subsystems of at most $NK = 8$ spins on the relevant timescales, thereby establishing an indirect measure of correlations in the bath. For P1-center spin baths (including the hyperfine interaction), pCCE converges already at $K = 1$ corresponding to CCE2, which we attribute to the complex substructure in the bath suppressing flip-flop transition as discussed below (for convergence tests, see Supplemental Material [53]). We use $K = 4$ for all systems in the following. In principle, the order N could also be increased for pCCE but, for our calculations, this was not necessary. For the results described below, we perform disorder average over 100 randomly chosen spin distributions and average over 20 mean-field spin configurations for each system.

Let us now discuss pCCE results for the spin-coherence $\langle M_x(2\tau) \rangle$ decay of NV spins interacting with the P1-center spin bath. On short timescales [53], $\langle M_x(2\tau) \rangle$ is generally

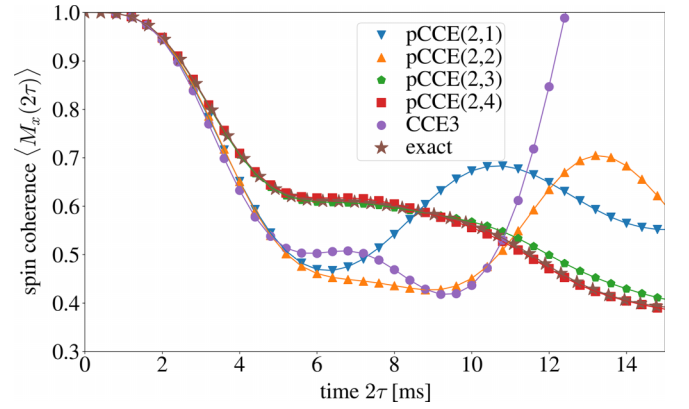


FIG. 2. Performance of the pCCE method. The time evolution of the spin coherence $\langle M_x(2\tau) \rangle$ of the NV electron spin is shown for a quasi-two-dimensional bath of 20 randomly distributed electron spins-1/2. Lines connect points to guide the eye. When increasing the partition size K , $\text{pCCE}(2, K)$ converges towards the exact solution obtained using the Suzuki-Trotter expansion [58] (for details, see Supplemental Material [53] and also Refs. [59–61]). The number of samples for the mean-field average decreases for pCCE with larger K , whereas it increases for CCE with larger N . For example, the CCE3 results shown here have not converged after averaging over 1000 mean-field configurations, while $\text{pCCE}(2, 4)$ converged after averaging over 10 mean-field configurations. For details about the mean-field averaging, see Supplemental Material [53].

expected to follow a stretched-exponential function [4]

$$\langle M_x(2\tau) \rangle \approx \exp \left[- \left(\frac{2\tau}{T_2} \right)^p \right]. \quad (3)$$

For a well-defined stretched-exponential parameter p , the function $-\ln(\langle M_x(2\tau) \rangle)$, when plotted on a double-logarithmic scale, is expected to exhibit a linear behavior with slope p . At short times, the results obtained indeed indicate a linear behavior, see Fig. 3. Fitting the function in Eq. (3) to the numerical results in this regime, we obtain p and T_2 for different values of the diamond-layer thickness L and the P1-center concentration ρ_{P1} , see Fig. 4. There are two different scalings of the T_2 time as a function of ρ_{P1} , which we attribute to different spatial dimensionalities of the spin bath: $T_2 \propto (\rho_{\text{P1}})^{-1}$ for bulk diamond at larger layer thicknesses (e.g., the slope is -1.02 for $L = 240$ nm) and $T_2 \propto (\rho_{\text{P1}})^{-3/2}$ for quasi-two-dimensional layers (e.g., the slope is -1.46 for $L = 30$ nm) [49]. The quasi-two-dimensional nature of the $L = 30$ nm layer at the concentrations ρ_{P1} considered is confirmed by relating L to the mean nearest-neighbor spin distance $l_s = l_s(\rho_{\text{P1}})$: at $L = 30$ nm, L/l_s varies approximately between 1.5 and 3 depending on ρ_{P1} and, for $L = 240$ nm, $L/l_s > 10$. For intermediate L , both regimes can be observed in Fig. 4: at larger ρ_{P1} , the spin bath tends to the three-dimensional case and, for smaller ρ_{P1} , to the two-dimensional case. These results highlight the broad range of applicability of the pCCE method and open new avenues to optimized material properties.

The T_2 times obtained are consistent with experimentally measured values. We obtain for the coefficient c in $T_2 = c/\rho_{\text{P1}}$ for bulk diamond ($L = 240$ nm) $c = 253 \pm 13$ ppm and the experimentally measured value is 160 ± 12 ppm

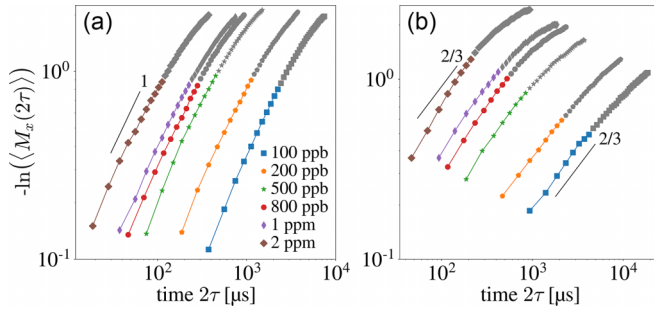


FIG. 3. Evidence for stretched-exponential behavior of the spin coherence $\langle M_x(2\tau) \rangle$ on short timescales. The function $-\ln(\langle M_x(2\tau) \rangle)$ plotted on a double-logarithmic scale at layer thickness $L = 240$ nm (a) and $L = 60$ nm (b) for various P1-center concentrations ρ_{P1} as indicated in the legend. The slope of the initial linear behavior corresponds to the stretched-exponential parameter p . In part (a), the layer thickness $L = 240$ nm corresponds to bulk diamond for all ρ_{P1} considered ($p \approx 1$). In part (b), the slope is changing corresponding to a transition from quasi-two-dimensional ($p \approx 2/3$) towards three-dimensional bath when increasing ρ_{P1} ($p > 2/3$). The values used for fitting are indicated by the lines connecting the colored points. For $\langle M_x(2\tau) \rangle$, this range approximately corresponds to the decay from 0.9 to 0.5. The standard deviation of the mean is smaller than the symbol size [53].

[33,62]. The calculated T_2 times are generally considered upper bounds because of other potential sources of dephasing in experiments, such as other nitrogen-related defects [63] or surface defects for shallow NV centers [45]. Suppressing the coupling to such defects, for example, by means of decoupling pulse sequences or optimized sample growth, could lead to a further increase of the measured T_2 time. Further, for an accurate comparison between theory and experiment, the fitting procedures need also to be aligned [53].

The corresponding values for p are shown in the inset of Fig. 4. We obtain $p = 1.03 \pm 0.03$ for bulk diamond ($L = 240$ nm) and $p = 0.64 \pm 0.03$ for quasi-two-dimensional layers ($L = 30$ nm) [64]. For intermediate diamond layer thicknesses L , we obtain a smooth transition between these two cases as we vary ρ_{P1} . Interestingly, if we leave out the nuclear spins of the P1 centers in our simulations, such that each P1 center is an electron spin-1/2, we obtain on average $p = 1.56 \pm 0.04$ and $p = 1.05 \pm 0.05$ for the bulk and the quasi-two-dimensional case, respectively, see Table I. These values are consistent with those analytically predicted by

TABLE I. Evidence for the dependence of the stretched-exponential parameters p on the hyperfine interaction in the P1-center bath. The parameter p is calculated including (P1 center) and excluding (no hyperf.) the hyperfine interaction for various spin concentrations ρ . For the error estimate, see Supplemental Material [53].

		ρ	2 ppm	1 ppm	0.1 ppm
two-dim.	P1 center		0.65 ± 0.05	0.63 ± 0.05	0.67 ± 0.05
($L \leq 60$ nm)	no hyperf.		0.95 ± 0.06	1.09 ± 0.07	1.05 ± 0.07
three-dim.	P1 center		0.95 ± 0.06	1.04 ± 0.06	1.08 ± 0.06
($L = 240$ nm)	no hyperf.		1.62 ± 0.09	1.55 ± 0.09	1.51 ± 0.08

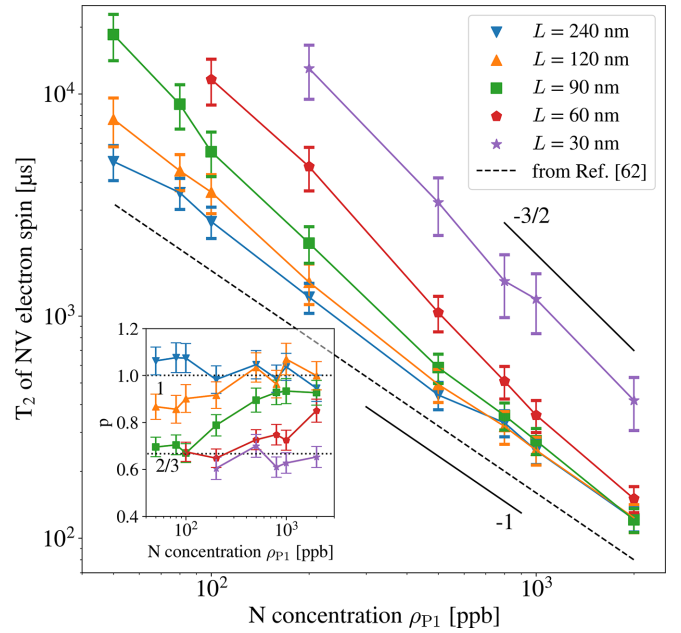


FIG. 4. Coherence time T_2 and the stretch-exponential parameter p signaling the spatial bath dimensionality. The T_2 time of the NV-center electron spin in a P1-center spin bath is shown for various P1-center concentrations ρ_{P1} and layer thicknesses L as indicated in the legend. Different scalings $T_2 \propto (\rho_{P1})^{-1}$ and $T_2 \propto (\rho_{P1})^{-3/2}$ are indicated by short black lines. The dashed black line is a fit to experimental results from Ref. [62]. Inset: The corresponding stretched-exponential parameters p with two distinguished values $p \approx 1$ for bulk diamond and $p \approx 2/3$ for quasi-two-dimensional layers. At intermediate L , we observe a smooth transition from the quasi-two-dimensional case ($p \approx 2/3$) to the bulk diamond ($p \approx 1$) when increasing ρ_{P1} . Lines connect points to guide the eye. Fluctuations are mainly due to uncertainties in the choice of the time window for fitting [53].

bath-dynamics models based on the Ornstein-Uhlenbeck process [23]. We attribute these findings to the complex internal substructure of the P1-center spin bath caused by the hyperfine interaction described by $\mathcal{H}_{\text{hf},j} = A_j I_{jz} S_{jz}$, where A_j depends on the orientation of the Jahn-Teller axis of the P1 defects ($A_j = 114$ MHz for the [111] axis and $A_j = 86$ MHz otherwise) [65]. The energy splittings induced by this interaction are much larger than the dipole-interaction constants at the concentrations ρ_{P1} considered [53]. Therefore, the P1-center bath is effectively divided into five subgroups depending on the orientation of the nuclear spins and the Jahn-Teller axes. Flip-flop transitions between P1-center electron spins from different subgroups are strongly suppressed (see, for example, Refs. [10,50]). Effectively, this situation corresponds to a NV spin in presence of five coupled spin baths with different characteristic times [53]. Even if the baths were uncoupled, the sum of Ornstein-Uhlenbeck processes describing the individual baths would typically not correspond to an Ornstein-Uhlenbeck process. We ascribe the dependence of the stretched-exponential parameter on the hyperfine interaction to this complexity in the bath.

To conclude, we investigate the spin-coherence decay of NV^- spins interacting with the strongly coupled disordered

bath of the single-substitutional nitrogen defects in diamond layers. For quasi-two-dimensional diamond layers, we obtain T_2 times which exceed those in bulk diamond, hence allowing to surpass the limit imposed by the defect concentrations in bulk diamond. We show that the stretched-exponential parameter p for the short-time spin-coherence decay is governed by the hyperfine interaction of the P1 centers, which is particularly important for understanding the underlying spin dynamics and for the effective modeling of the magnetic noise generated by the spin bath. We introduce the pCCE method based on the cluster-correlation expansion applied to local strongly interacting partitions of the bath, which includes important high-order spin correlations leading to better accuracy, applicability to longer timescales and enhanced convergence

properties. We expect this method to be applicable beyond the setting considered here: for example, to NMR systems, i.e., nuclear spins on a lattice, to longer pulse sequences such as the Carr-Purcell-Meiboom-Gill pulse sequence, and perhaps even to non-Markovian spin baths.

The source code is published in a GitHub repository [66].

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