

Microscale sensing with strongly interacting NV ensembles at high fields

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Advances in sensing devices that utilize nitrogen-vacancy (NV) center ensembles in diamond are driving progress in microscale nuclear magnetic resonance spectroscopy. Utilizing quantum sensing techniques in the high-field regime significantly boosts sensitivity by increasing thermal polarization and improves spectral quality via enhanced energy shifts. Compatible with the latter, a straightforward manner to further raise sensor sensitivity is to increase NV concentration, although this intensifies detrimental dipole-dipole interactions among NVs. In this Letter, we present a method for detecting NMR signals in high-field scenarios while effectively suppressing dipole-dipole couplings in the NV ensemble. Thus, this approach enhances sensitivity by combining highly doped diamond substrates and elevated magnetic fields.

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Introduction. Quantum sensors have emerged as tools for detecting magnetic signals in different regimes, spanning pure and applied science [1]. Notably, in the realm of nuclear magnetic resonance (NMR) spectroscopy [2,3], conventional inductive detection techniques struggle with inherent low sensitivity, limiting their application to millimeter-sized samples [4]. On the contrary, quantum sensor-based NMR shows great potential for applications involving minimal sample sizes [4–8], such as high-throughput chemistry [9,10], single-cell biology [11], and two-dimensional material science [12]. In this context, nitrogen-vacancy (NV) color defects in diamond [4,5,13] have gained popularity owing to their performance at ambient conditions, making microscale ensembles of NVs a viable solution to investigate picoliter samples via NMR techniques.

Recently, efforts have concentrated on synthesizing diamonds with a large NV concentration to further enhance NV-based NMR sensitivity [14–23]. Having more NV centers lowers the system's noise floor, resulting in a higher signal-to-noise ratio (SNR) [24,25]. In light of this, researchers are designing various mechanisms based on dynamical decoupling (DD) techniques to mitigate detrimental dipole-dipole interactions that arise in highly NV-doped diamonds [19–22,26]. However, these techniques do not extend to the high-field regime, which is a particularly relevant domain for various applications. Among other benefits, elevated fields amplify the sample-emitted signal due to increased nuclear thermal polarization and also improve the spectral resolution of chemical shifts [27–31].

In this Letter, we introduce SHIELD (Sensing at High field Improved via ELusion of Dipole-dipole), a microwave DD sequence specifically designed for densely packed NV ensembles, capable of operating at high-field conditions. Our sequence simultaneously suppresses dipolar interactions among NVs while coupling the sensor to the high-field NMR signal. Besides, it is robust against common experimental errors, such as strain in the diamond lattice, couplings with other impurities in the host substrate, and errors on the applied control drivings. Additionally, our sequence can be incorporated to quantum heterodyne protocols that enable high-resolution sensing [6,7].

Methods.

a. Theoretical framework. We consider a nuclear spin sample placed on top of a diamond with embedded NV centers, in the presence of a large external magnetic field B_z (see Fig. 1). The target NMR signal $B(t)$ is generated by driving the sample with radio frequency (RF). In this manner, even in the high-field regime, the target signal oscillates at a slow rate, in particular, at the nuclear Rabi frequency (~ 100 kHz) [27–30]. Our goal then is the detection of this slowly oscillating $B(t)$ with a highly dense NV ensemble.

The Hamiltonian describing the system is

$$\begin{aligned} \frac{H}{\hbar} = & \sum_{i=1}^N [D(S_i^z)^2 + (|\gamma_e|B_z + \xi_i)S_i^z] \\ & + \sum_{i=1}^N |\gamma_e|B(t)S_i^z + \sum_{i<j}^N H_{dd}^{ij} + \sum_{i=1}^N \sqrt{2}\Omega \cos(\omega_c t)S_i^\phi, \end{aligned} \quad (1)$$

where $S_i^\phi = \cos\phi S_i^x + \sin\phi S_i^y$, while $S_i^{x,y,z}$ are the spin-1 operators. The first term in Eq. (1) accounts for the free energy of N NV centers, determined by the zero-field splitting $D/(2\pi) = 2.88$ GHz, the electron gyromagnetic ratio $|\gamma_e|/(2\pi) = 28.024$ GHz/T, the external magnetic field B_z ,

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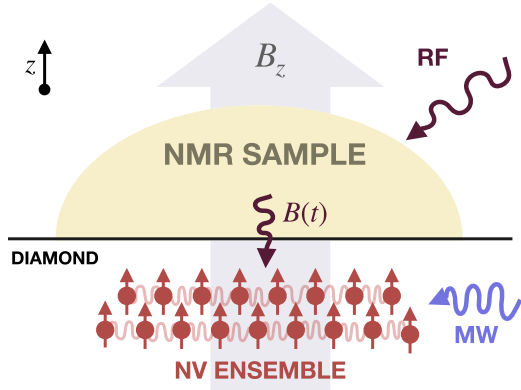


FIG. 1. Illustration of the setup: A [111] cut diamond hosts a dense ensemble of NV centers, exhibiting strong dipole-dipole interactions. A target sample containing nuclear spins is placed on the diamond surface, with a strong magnetic field on the order of teslas is applied along the z axis. Nuclear spins rotate at moderate speed due to an RF field resulting into a slowly oscillating signal $B(t)$ that encodes sample information. Simultaneously, NV centers are controlled using an MW driving.

and the site-dependent energy shifts ξ_i , which can introduce variations up to MHz [32] in the resonance energy of the distinct NV centers in the ensemble. The second term describes the interaction between the NV ensemble and the NMR signal $B(t)$, which we define as $B(t) = B_t \cos(\omega t + \phi_t)$. The third term, $\sum_{i<j}^N H_{dd}^{ij}$, corresponds to the dipole-dipole interaction among NVs; see the Supplemental Material [33] for an explicit expression of H_{dd}^{ij} . The final term corresponds to the microwave (MW) control driving with amplitude Ω and frequency ω_c .

The Hamiltonian of the system [Eq. (1)] in a rotating frame with respect to $H_0 = \sum_{i=1}^N \omega_c S_i^z$ reads

$$\frac{H_I}{\hbar} = \sum_{i=1}^N \xi'_i S_i^z + \sum_{i=1}^N [\Delta S_i^z + \Omega S_i^\phi] + \sum_{i=1}^N \gamma_e B(t) S_i^z + \sum_{i<j}^N \bar{H}_{dd}^{ij}, \quad (2)$$

where $\Delta = (D - |\gamma_e| B_z) - \omega_c$ is the detuning on the MW control and $\bar{H}_{dd}^{ij} = d_{ij} [s_i^z s_j^z - (s_i^x s_j^x + s_i^y s_j^y)]$, while $\xi'_i = \xi_i + \sum_{i \neq j} d_{ij}/2$. Note that spin operators $s_i^{x,y,z}$ are now spin-1/2 operators; for simplicity, the Hamiltonian is restricted to the subspace spanned by $\{|b_1\rangle \otimes \dots \otimes |b_N\rangle \mid b_j \in \{0, -1\}\}$, neglecting spin occupations in $|1\rangle_j$ (see the Supplemental Material [33] for details).

We start by analyzing the role of dipole-dipole interactions among NV detectors in the widely used Carr-Purcell-Meiboom-Gill (CPMG) protocol [34]. As in NV-based NMR experiments, the expectation value s^z of NV centers is inferred; we compute the latter via Eq. (2). In particular, for the CPMG protocol, the system dynamics is described with Eq. (2) by setting $\Delta = 0$ while Ω is deployed stroboscopically. Then, after applying CPMG over a cluster of q interacting NVs, we have (see the Supplemental Material

[33])

$$\left\langle \sum_{i=1}^q s_i^z \right\rangle \approx \frac{2\gamma_e B_t t_s}{\pi} \cos(\phi_t) \left\langle \sum_{i=1}^q s_i^x \right\rangle_{\rho^*}, \quad (3)$$

where t_s is the CPMG sequence duration and ρ^* the NV cluster state evolved only under dipole-dipole interactions. Hence, the term $\langle \sum_{i=1}^q s_i^x \rangle_{\rho^*}$ depends on the dipole-dipole interactions and it can be bounded as $|\langle \sum_{i=1}^q s_i^x \rangle_{\rho^*}| \leq q/2$, achieving its maximum $q/2$ in the absence of dipole-dipole couplings. To illustrate this concept, we analytically solved the case involving two NV centers (see the Supplemental Material [33]).

In a scenario with an NV ensemble composed of Q clusters, each containing q_k NV centers (where $k = 1, \dots, Q$), the total NV signal is the sum from all clusters: $\sum_{k=1}^Q \langle \sum_{i=1}^{q_k} s_i^z \rangle$. Then, the contribution of each cluster to the total NV signal has a random factor in the range $[-q_k/2, q_k/2]$, depending on the structure of dipole-dipole terms within the cluster. This means that (1) each cluster, most likely, generates a weaker signal compared to that without dipole-dipole interactions and (2) In the total NV ensemble signal different clusters can interfere destructively, making signal detection challenging. This underlines the need for protocols that suppress dipolar interactions to boost NV-based quantum sensing.

b. The protocol. Our protocol—SHIELD—integrates MW driving blocks that continuously mitigate the effect of NV-NV interactions. These blocks are interspaced by π pulses, which suppress disorder terms ξ'_i while simultaneously coupling the NV ensemble to $B(t)$; refer to Fig. 2(a) for a scheme of our sequence.

The MW driving blocks generate concatenated 2π rotations of the NV centers around the axes A , B , $\bar{A} = -A$, and/or $\bar{B} = -B$, realized by fixing the appropriate phase ϕ and setting the MW detuning to $\Delta = \pm\Omega/\sqrt{2}$ [see Eq. (2)]. This specific choice of detuning continuously symmetrizes the dipole-dipole interaction, converting $\bar{H}_{dd}$ into $H_{dd}^{\text{sym}} = \sum_{i<j}^N d_{ij}/3 \cdot [s_i^z s_j^z + s_i^x s_j^x + s_i^y s_j^y]$, provided that $d_{ij} \ll \Omega$ for the rotating wave approximation (RWA). For further detail refer to the Supplemental Material [33].

Rotations along A , B , $\bar{A}$, and $\bar{B}$ can be combined in different manners; these include the frequency-switched LG sequence [35,36], which corresponds to the block $A\bar{A}\bar{A}\bar{A}$ and the LG4 sequence $A\bar{A}\bar{B}\bar{B}$ [37], where if A uses phase $\phi = 35^\circ$, B uses $180^\circ - \phi = 145^\circ$. This is, if rotations around A are exerted with $\sum_{i=1}^N [\Delta s_i^z + \Omega s_i^{35^\circ}]$, for B , $\sum_{i=1}^N [\Delta s_i^z + \Omega s_i^{145^\circ}]$. Tuning the MW control in Eq. (2) for either the $A\bar{A}\bar{B}\bar{B}$ or $A\bar{A}\bar{A}\bar{A}$ block results in the average Hamiltonian

$$H_C = \sum_j^N [\xi'_i + \gamma_e B(t)] f_r s_j^C + \sum_{i<j}^N H_{dd}^{\text{sym}} \quad (4)$$

(see the Supplemental Material [33] for the derivation). In Eq. (4), f_r and s_j^C are the reduction factor and the spin operator of the j th NV projected along axis C , which bisects A and B [see Fig. 2(b)]. Both quantities— f_r and s_j^C —depend on the chosen configuration for the MW control block (see the Supplemental Material [33] for analytical expressions). In particular, the reduction factor has a value of $f_r = 0.4292$ for the MW block $A\bar{A}\bar{B}\bar{B}$ and $f_r = 0.5774$ for the block $A\bar{A}\bar{A}\bar{A}$.

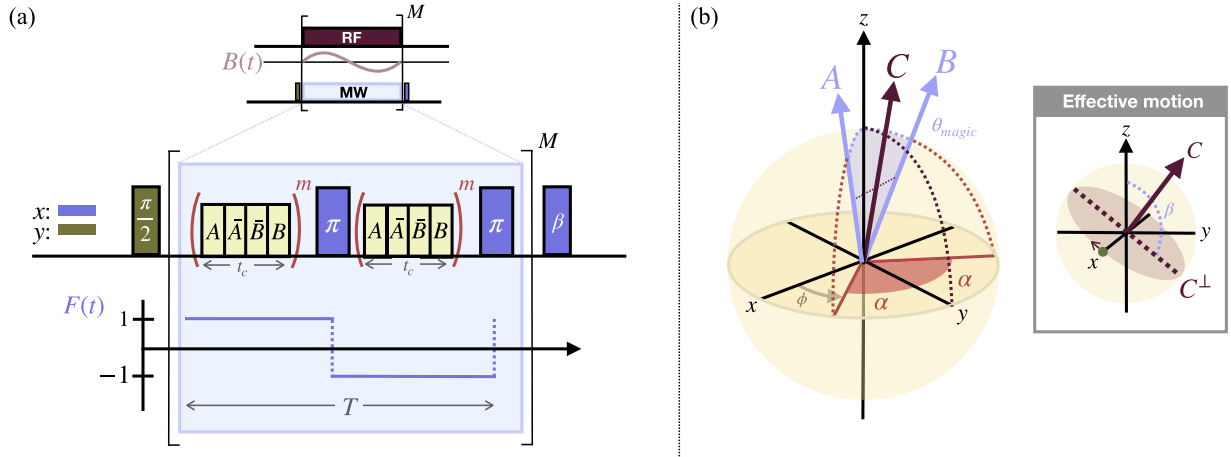


FIG. 2. Scheme of the protocol. (a) At the top, continuous RF driving induces Rabi oscillations of the sample nuclei, which results in the NMR signal $B(t)$. At the bottom, the blue square represents the MW pattern of our sequence SHIELD, which simultaneously captures $B(t)$ while suppressing dipole-dipole interactions. SHIELD includes an initial and a final pulse, and in between, M repetitions of two m blocks of rotations around the axes A , $\bar{A}$, $\bar{B}$, and B , interspersed by π pulses along the x axis. These pulses imprint the function $F(t)$ with periodicity T on the effective operators σ_j^C . (b) Relevant direction and angles. In the XY plane, the axes A and B are rotated by angles $-\alpha$ and α from the y axis about the z axis, and both axes— A and B —form the magic angle with the z axis. The bisector of A and B defines direction C . The gray box inset illustrates the effective dynamics of the NV spins: The initial pulse polarizes the NV centers along the x axis (green dot), and then the MW scheme slightly rotates the initial state around C , inducing changes in $\langle S^{C^\perp} \rangle$, where $C^\perp = C \times x$. For readout, the accumulated spin polarization along $C^\perp$ is transferred to z axis via the final pulse of angle β along the x direction.

The sequence is completed by introducing equidistant π pulses along a direction perpendicular to axis C , for example, along x . The corresponding pulse propagator is $U_\pi^x = \prod_{j=1}^N (-2i) s_j^x$. Consequently, the full sequence reads

$$U = \prod_{k=1}^M U^k, \quad U^k = U_\pi^x \left[\prod_{l=1}^m U_C(t) \right] U_\pi^x \left[\prod_{l=1}^m U_C(t) \right], \quad (5)$$

where $U_C(t)$ denotes the propagator $U_C(t) = e^{-i \int_t^{t+tc} H_C(\tau) d\tau}$, with t corresponding to the instant at which the given MW block starts, either $A\bar{A}\bar{B}B$ or $A\bar{A}A\bar{A}$; then, m defines the number of repetitions of that MW block between π pulses, while M is the number of sequence repetitions. In the toggling frame, the π pulses imprint a modulation function $F(t) = +1, -1$ on s_j^C , alternating the sign of the operator with each pulse; hence, within the average Hamiltonian theory approach, the effective first-order Hamiltonian is

$$\begin{aligned} \bar{H} &= \sum_j^N \gamma_e F(t) \cdot B(t) f_r s_j^C + \sum_{i<j} H_{dd}^{sym} \\ &= H'_{\text{CPMG}} + \sum_{i<j} H_{dd}^{sym}, \end{aligned} \quad (6)$$

where we define $H'_{\text{CPMG}} = \sum_j^N \gamma_e F(t) \cdot B(t) f_r s_j^C$ to retain parallelism with the development for the CPMG protocol presented in the Supplemental Material [33]. From Eq. (6), it follows that the modulation $F(t)$ serves the dual purpose of coupling the NV ensemble to $B(t)$ and averaging out the disorder ξ_j' . The latter occurs because the protocol spans M full periods of $F(t)$, during which $F(t)$ takes the values $+1$ and -1 for equal duration, while ξ_j remain constant in time.

Then, the propagator corresponding to SHIELD protocols reads

$$U = e^{-i\bar{H}t_s} = e^{-iH'_{\text{CPMG}}t_s} e^{-i(\sum_{i<j} H_{dd}^{sym})t_s}, \quad (7)$$

where $t_s = MT$ is the duration of the sequence. Recall the decomposition of the propagator, which is permitted due to the commutation relation $[\sum_j^N s_j^C, \sum_{i<j} H_{dd}^{sym}] = 0$.

We can now compute the spin population for q interacting NVs after the application of a SHIELD protocol. For that, note that the initial $\frac{\pi}{2}$ pulse in Fig. 2(a) prepares the NV ensemble in the state $\rho_0 = \bigotimes_j^N (\mathbb{1} + \sigma_j^x)$, while a final pulse of an angle β along x transfers the NV population from $C^\perp$ to the z axis for measurement [see Fig. 2(b)]. Thus, following a procedure similar to that in Eqs. (S10)–(S18) in the Supplemental Material [33],

$$\begin{aligned} \left\langle \sum_{i=1}^q s_i^z \right\rangle &= \text{Tr} \left[U \rho_0 U^\dagger \sum_j^q s_j^{C^\perp} \right] \\ &= \text{Tr} \left[\rho_0 \left(e^{iH'_{\text{CPMG}}t_s} \sum_j^q s_j^{C^\perp} e^{-iH'_{\text{CPMG}}t_s} \right) \right] \\ &\approx \frac{2\gamma_e B_t t_s}{\pi} f_r \cos(\phi_t) \frac{q}{2}, \end{aligned} \quad (8)$$

where the dipole-dipole coupling is absent, as the residual interaction does not generate any dynamics; i.e., since $[\rho_0, \sum_{i<j} H_{dd}^{sym}] = 0$, it follows that $e^{-i(\sum_{i<j} H_{dd}^{sym})t_s} \rho_0 e^{i(\sum_{i<j} H_{dd}^{sym})t_s} = \rho_0$.

Comparing the latter to the CPMG signal in Eq. (3), we observe two key differences. First, as a direct consequence of the continuous MW driving, our protocol introduces the re-

duction factor f_r in the signal amplitude; second, eliminating the dipole-dipole interaction from the output signal enables all clusters to contribute constructively to the total signal. In other words, under the continuous MW driving, the term $\langle \sum_{i=1}^q s_i^z \rangle_{\rho^*}$ in Eq. (3) converges to the upper bound $\frac{q}{2}$ for each cluster, resulting in $\frac{N}{2}$ when accounting for the whole NV ensemble. Therefore, SHIELD, at the cost of introducing the f_r factor, enables to leverage highly doped diamonds for increased sensitivity through the symmetrization of the dipole-dipole interaction, which leads to commutation relations that inhibit the detrimental expression of them.

Besides, the protocol is versatile: Adjusting the number m of MW blocks between π pulses in the sequence of Fig. 2(a) enables the detection of target signals across a broad frequency range. From a technical perspective, the delivered π pulses induce a modulation function $F(t)$ with a period $T = 4m/\Omega$, which can be tuned changing m [38]. Then, via the first harmonic of $F(t)$, NVs couple to NMR signals of frequency $\sim 1/T$, which can take values from tens of kHz to MHz. In particular, the lower range (kHz) allows for high-resolution NMR signal detection in a high-field scenario, where the sample-NV interaction is mediated by an RF field [27–30], while the higher range (MHz) corresponds to medium-field NMR, where SHIELD excels integrated in heterodyne schemes [39]. In contrast, other methods for highly doped diamonds [18–23] are limited to a narrower frequency range in the MHz regime, limiting their application to medium field NMR.

Results and Discussion. To assess the performance of SHIELD, we conducted numerical simulations for the particularly interesting high-field scenario. Constrained by computational resources, simulations are limited to clusters of up to six dipolar-coupled NVs, over which distinct MW detection schemes are applied. In particular, the clusters couple to an NMR signal via (1) SHIELD with an $A\bar{A}\bar{B}\bar{B}$ MW block, (2) SHIELD with an $A\bar{A}\bar{A}\bar{A}$ MW block, and (3) a standard CPMG sequence. To carry out the comparison between these schemes, we evaluate the average NV spin population across hundreds of clusters, i.e., $Q \geq 300$) as

$$\bar{s}^z = \frac{1}{\sum_{k=1}^Q q_k} \sum_{k=1}^Q \sum_{i=1}^{q_k} \langle s_i^z \rangle, \quad (9)$$

for systems with different NV densities. Further details are provided in the Supplemental Material [33].

The simulations are performed using Eq. (2) as a starting point, dressed with realistic noise effects. Specifically, the MW driving has an amplitude of $\Omega/(2\pi) = 20$ MHz, with a 1% constant Rabi error applied to both the π pulses and MW blocks. Detuning errors in the driving are incorporated into the disorder shifts ξ_i , which also account for strain in the diamond. Since all NV centers within a cluster are spatially localized, we assume that they share the same ξ_i , drawn from a Gaussian distribution with zero mean and standard deviation of 4 MHz [32]. In addition, each NV acquires an additional shift due to dipole-dipole interactions, so that the total site-dependent shift is $\xi'_i = \xi_i + \sum_{i \neq j} \frac{d_{ij}}{2}$. On the other hand, the NMR signal $B(t)$ has frequency $\omega_t/(2\pi) = 95.68$ kHz and an amplitude $B_t = 1$ nT. The latter corresponds to the thermal polarization of protons in a microscale pure ethanol sample under $B_z \sim 2$ T [27]. Additionally, $m = 32$ and $M = 2$, as the NV center

decoherence time under dynamical decoupling ($T_2 \sim 20$ μ s) permits the detection of two oscillations of $B(t)$, which last $t_s = 21$ μ s.

In Fig. 3, we present the numerical outcomes. The first plot (a) shows the amplitude of $\bar{s}^z$ at various NV concentrations and the second plot (b) illustrates the corresponding SNR, accounting for shot noise and projection noise. The SNR is computed because it reflects the advantages of higher NV concentration; namely, a larger number of NV sensors significantly reduces projection noise. Details on the methodology are provided in the Supplemental Material [33].

Analyzing the outcome for the CPMG case, in Fig. 3(a), we observe a rapid decay in signal amplitude as NV concentration increases. This decay occurs because at higher density, NV centers are closer together, leading to stronger dipole-dipole interactions [see Fig. 3(c)]. Consequently, in Fig. 3(b), we observe a sensitivity sweet spot for the CPMG. This is, for low NV density, the SNR improves with NV concentration, benefiting from the increasing of NV sensors, until the dipole-dipole interaction among NVs becomes overly strong, causing a decay at 0.4 ppm. Then, according to our numerical results, 0.4 ppm is the optimal concentration for the CPMG scheme; nevertheless, further numerical analysis suggests that this would serve as an upper bound (see the Supplemental Material [33]).

Concerning SHIELD protocols, in Fig. 3(a), they exhibit a gradual decay in signal amplitude as NV density increases. This is because dipole-dipole interactions become stronger, making the RWA less valid. Nevertheless, this decay is slow, indicating significant suppression of dipole-dipole interactions within the considered NV densities. In this regard, note that the SHIELD sequence incorporating the $A\bar{A}\bar{A}\bar{A}$ block exhibits greater amplitude compared to the $A\bar{A}\bar{B}\bar{B}$ configuration, although the latter demonstrates superior robustness, characterized by a slower decay. Moving to Fig. 3(b), this shows that the reduction factor f_r makes SHIELD protocols unsuitable in scenarios, where dipole-dipole interactions among NVs are negligible. However, as the NV concentration increases, the amplitude penalty from f_r is compensated by the dipole-dipole cancellation provided by SHIELD. This enables to operate with a large number of NV detectors, resulting in sensitivity levels that exceed those achieved by standard CPMG at its peak performance.

To further illustrate the comparison, we focus on the case of 3 ppm, where dipole-dipole interactions of 1.5 MHz are observed [see Fig. 3(c)]. At this concentration, CPMG suffers a drastic reduction in the signal amplitude, while SHIELD sequences are barely affected. This leads to sensitivity rates for SHIELD protocols that are five to seven times greater than that of the CPMG sequence. From a general point of view, curves in Fig. 3(b) indicate a growing SNR for SHIELD at sensible NV concentrations, while standard schemes fail.

Conclusion. We present SHIELD, an ac signal detection scheme tailored for highly concentrated NV-based quantum detectors. Our findings indicate that even at common NV concentrations, just above 0.4 ppm, dipole-dipole interactions detrimentally affect standard schemes; SHIELD continuously suppresses these interactions while operating over a wide range of target signal frequencies that spans from tens of kHz to MHz. In this window, the lower frequency

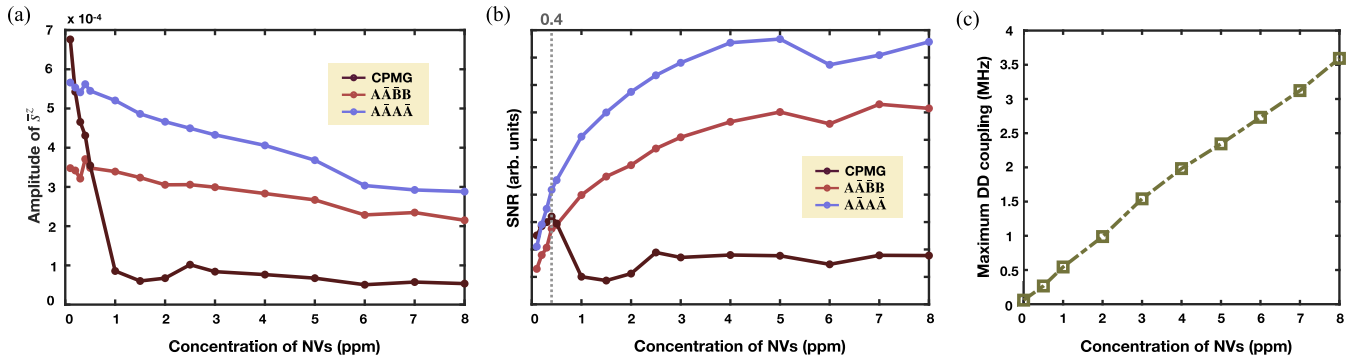


FIG. 3. Numerical results. In every graph, the NV density in the x axis corresponds to the concentration of NV centers aligned with the external magnetic field. This direction is the preferential alignment in [111] diamond cuts [40–44]. (a) Oscillation amplitude of $\bar{s}^z$ for different NV concentrations. Purple curve corresponds to the CPMG protocol, while red and blue curves to SHIELD using the $A\bar{A}\bar{B}\bar{B}$ and $A\bar{A}\bar{A}\bar{A}$ MW blocks, respectively. (b) Expected SNR considering $\sqrt{N}$ scaling of the noise. For CPMG sequence, at 0.4 ppm is reached the best sensitivity, while SHIELD protocols exhibit an increasing tendency of SNR for sensible concentrations. (c) Maximum dipole-dipole coupling, i.e., $d_{ij}/(2\pi)$, at different NV concentrations. This is computed by averaging the maximum dipole-dipole interaction over thousands of distinct configurations, each constituted, on average, by four NVs.

range is particularly interesting, as it enables the detection of high-field signals through RF mediation. Thus, SHIELD harnesses the sensitivity gains of dense ensembles and high fields, while further dipole-dipole cancellation resulting in enhanced sensitivities is possible by introducing stronger MW drivings.

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Data availability. The data generated in this study (via numerical simulations) is available from the contact author upon request.

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