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Dynamical nuclear polarization using multi-colour control of color centers in diamond

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Abstract

Dynamical nuclear polarization (DNP) transfers the polarization of electron spins at cryogenic temperatures to achieve strong nuclear polarization for applications in nuclear magnetic resonance. Recently introduced approaches employ optical pumping of nitrogen-vacancy (NV) centers in diamond to achieve DNP even at ambient temperatures. In such schemes microwave radiation is used to establish a Hartmann-Hahn condition between the NV electron spin and proximal nuclear spins to facilitate polarization transfer. For a single monochromatic microwave driving field, the Hartmann-Hahn condition cannot be satisfied for an ensemble of NV centers due to inhomogeneous broadening and reduces significantly the overall efficiency of dynamical nuclear polarization using an ensemble of NV centers. Here, we adopt generalized Hartmann-Hahn type dynamical nuclear polarization schemes by applying microwave driving fields with (multiple) time-modulated frequencies. We show that it is possible to enhance the effective coupling between an ensemble of NV center spins with inhomogeneous broadening and nuclear spins, thereby improving significantly the overall efficiency of dynamical nuclear polarization. This approach can also be used to achieve dynamical nuclear polarization of an ensemble of nuclei with a distribution of Larmor frequencies, which would be helpful in magnetic resonance spectroscopy using a single NV spin sensor.

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Keywords: dynamical nuclear polarization; quantum imaging; color centers in diamond; nuclear magnetic resonance spectroscopy

1 Introduction

With the appealing advantages of operating at room temperature and ambient conditions as well as providing nanoscale spatial resolution, the applications of nitrogen-vacancy (NV) centers in diamond [1] as a novel type of quantum sensor have extended from the precise measurement of magnetic field, electric field, pressure and temperature [2–9] towards the detection of single proteins [10, 11] and even the detection of individual nuclear spins with potential applications to single molecule magnetic resonance spectroscopy [12–31]. The main techniques underlying the NV-based quantum sensing can be grouped into pulsed schemes [12–25] and continuous driving schemes [26–31]. The basic idea of both schemes is to establish a resonance condition between the induced effective frequency of

a driven NV spin and the Larmor frequency of nuclear spins, via tuning either the interpulse time interval (in pulsed schemes) or the Rabi frequency of microwave driving field (in continuous driving schemes).

More specifically, the continuous driving scheme uses resonant microwave radiation to induced dressed spin states of NV centers. The energy gap between the dressed states of the driven NV spin is specified by the Rabi frequency of the continuous driving field. By tuning the intensity of microwaves, one can establish an effective resonant exchange interaction between the dressed states of the NV center spin and the spin-up and spin-down states of nuclei when the Rabi frequency Ω matches the Larmor frequency ω_L of nuclei, i.e. the so called Hartmann-Hahn condition [32]. This idea was exploited in proposals for determining the position of nuclei in single molecule magnetic resonance spectroscopy [26, 30, 31] and dynamical nuclear polarization, which has shown very high efficiency in achieving substantial nuclear polarization [29].

In realistic scenarios, a bulk diamond or diamond powder will contain a large number of NV centers within the laser and microwave focus. Consequently, it is worthwhile considering DNP experiments using an ensemble of NV centers. This leads to the challenge that the transition frequency of NV centers may have a distribution of values, for example due to the magnetic dipole-dipole interactions with the surrounding impurity electron spins, or due to the misalignment between the spin preferential (quantization) axis (e.g. the NV symmetry axis) and the direction of the magnetic field, e.g. an ensemble of NVs in diamond powder [33, 34]. In these scenarios, using one single microwave driving field at a specific frequency, the Hartmann-Hahn condition would only be satisfied for a small portion of NV center spins or nuclear spins, thus severely limiting the efficiency of dynamical nuclear polarization protocols.

In this work, we address this challenge by the design of microwave fields with time-dependent frequencies (and/or amplitudes), similar to integrated solid effect [34, 35], to improve the effective exchange couplings between an inhomogenously broadened ensemble of NV centers and nuclear spins. In contrast with the standard continuous driving scheme, which uses a single frequency that corresponds to the electronic transition frequency of NV center spin and a constant amplitude to match the Larmor frequency of nuclei, the frequency-modulated microwave driving fields enable the Hartmann-Hahn resonance to be achieved for an ensemble of NV centers. As a result the efficiency of dynamical nuclear polarization can be significantly improved. In the following, we first illustrate this approach with a simple case in which an ensemble of NV centers has a Lorentzian shape of frequencies. Then we proceed to apply our idea to the scenario of dynamical nuclear polarization using NV centers with random orientations of their symmetry axes. In a similar manner, the present idea can be extended easily to achieve dynamical nuclear polarization of an ensemble of nuclei on diamond surface with inhomogeneous linebroadening using one single NV center by sweeping the Rabi frequency of microwave field (while with one single fixed frequency), e.g. in magnetic resonance spectroscopy using a single strong coupling NV center spin sensor [18]. The potential advantage of our protocol is that it is a simple extension of the standard dynamical nuclear polarization based on Hartmann-Hahn resonance, and thus may be easily achievable in experiments.

2 Basic principles

The NV-center has the ground state $^3A_2$ level which exhibits a zero field splitting of $D = (2\pi)2.87$ GHz between the $m_s = 0$ and $m_s = \pm 1$ spin sub-levels, see Figure 1. The NV center

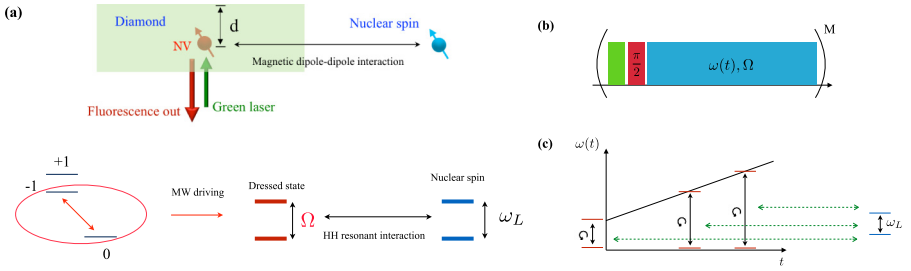

Figure 1 Protocol of DNP using multi-color control of NV centers in diamond. (a) Standard dynamical polarization transfer based on Hartmann-Hann resonance. A nitrogen-vacancy center spin in diamond is coupled with nuclei via magnetic dipole-dipole interaction. A microwave driving field satisfying the Hartmann-Hahn condition $\Omega = \omega_L$ induces a resonant exchange interaction between NV center spin and nuclei, which facilitates polarization transfer from NV center spin to nuclei. **(b)** The repetitive dynamical nuclear polarization cycle: (1) the NV centers are initialized by a green laser pulse; (2) a $(\frac{\pi}{2})_y$ pulse with the frequency $\omega(t)$ prepares the NV centers into the dressed spin down state; (3) the frequency-modulated microwave fields are applied to facilitate the polarization transfer from the NV center to the nuclei. **(c)** The frequency sweep strategy for dynamical nuclear polarization using an ensemble of NV centers. The frequency of the microwave driving field sweeps linearly in time, namely $\omega(t) = \omega(0) + \nu t$. The NV centers with different transition frequencies are resonantly driven sequentially when the microwave frequency $\omega(t)$ comes close to its individual transition frequency.

spin Hamiltonian is given by [1]

$$H = D\mathbf{S}_z^2 + E\left(\mathbf{S}_x^2 - \mathbf{S}_y^2\right) + \gamma \mathbf{B} \cdot \mathbf{S}, \tag{1}$$

where $\gamma = (2\pi)28\ \mathrm{MHz} \cdot \mathrm{mT}^{-1}$ is the electron gyromagnetic ratio, $\mathbf{B}$ is the applied external magnetic field, and E (on the order of MHz) represents the strain effect of the diamond lattice, $\mathbf{S}_x$, $\mathbf{S}_y$, $\mathbf{S}_z$ are the three components of the NV center spin operator. We assume that the magnetic field B is not very large (while being much larger than the lattice strain effect), e.g. B = 50-200 G, so that the NV axis is still the good quantization axis of the NV center spin even when the magnetic field is not perfectly aligned with the NV axis, thus allowing high fidelity initialization and readout of NV center spin state. In this weak magnetic field regime, the Hamiltonian of the NV center spin can be approximated as

$$H_{NV} = D\mathbf{S}_z^2 + \gamma B \cos(\theta)\mathbf{S}_z, \tag{2}$$

where θ is the relative angle between the NV axis and the magnetic field. The magnetic dipole-dipole interaction with the surrounding NVs or electron impurities, as well as the strain effects, will cause line-broadening of NV centers. Taking these effects into account, the Hamiltonian of an ensemble of NV centers with inhomogeneous line broadening can be described as in the following general form

$$H_{NV} = (D + \delta_{+1})|{+1}\rangle\langle{+1}| + (D + \delta_{-1})|{+-1}\rangle\langle{-1}|, \tag{3}$$

where the values of δ_{+1} and δ_{-1} vary between different NV centers. Without loss of generality, we take $m_s = 0$ and $m_s = -1$ to carry the dressed spin sensor, and the electronic transition frequency is $\omega_0 = D + \delta_{-1}$.

We consider weakly coupled distant nuclear spins, e.g. assuming that the distance between the NV center and the nucleus is $r \geq 1.5$ nm (corresponding to a hyperfine coupling

of ∼5 kHz). This includes ^{13}C inside diamond or nuclei in biomolecules on the surface of diamond with shallow implanted NV centers. As we assume that the magnetic field is weak so that the NV axis remains a good quantization axis of the NV centers, the magnetic dipole-dipole interaction between the NV center spin and the nuclear spin can thus be written as follows

$$H_I = g\mathbf{S}_z \otimes \left[(\mathbf{I}\cdot\hat{r}_0) - 3(\hat{r}\cdot\hat{r}_0)(\mathbf{I}\cdot\hat{r})\right], \tag{4}$$

where we choose the coordinate system with the direction of the magnetic field as the $\hat{z}$ axis, and $\hat{r}$ is the unit vector that connects the NV center and the nucleus, $\hat{r}_0$ is the unit vector that characterizes the NV axis, and $g = (\mu_0\gamma_e\gamma_c)/(4\pi r^3)$ with r the distance from the NV center to the nucleus, $\mathbf{I}$ represents the nuclear spin operator. The Hamiltonian for a nuclear spin is

$$H_L = \omega_L\mathbf{I}_z = (\gamma_L B)\mathbf{I}_z, \tag{5}$$

where the magnetic field direction specifies the quantization axis of the nuclear spin, and γ_L is the gyromagnetic ratio of the nuclear spin, $\omega_L = \gamma_L B$ is the Larmor frequency.

In the standard dynamical nuclear polarization based on Hartmann-Hahn resonance [32], we apply a microwave driving field

$$H_m(t) = \Omega_0\cos(\omega_0 t)\mathbf{S}_x \tag{6}$$

to induce a dressed spin-1/2. Here, we choose the frequency of the microwave driving field close to the electronic transition frequency of the NV center between the spin sublevels $m_s = 0$ and $m_s = -1$, while remaining large detuning from the electronic transition frequency between the spin sublevels $m_s = 0$ and +1. The effective Hamiltonian of a dressed spin-1/2 of NV center is

$$H_e = \frac{\Omega}{2}\sigma_x, \tag{7}$$

where σ is the Pauli operator. And thus the total Hamiltonian of the NV center spin and the nuclear spin turns out to be

$$H_T = H_e + H_L + H_I. \tag{8}$$

When the Hartmann-Hann condition $\Omega = \omega_L$ is satisfied, the interaction between the NV center dressed spin $\{|\uparrow_x\rangle, |\downarrow_x\rangle\}$, which are the eigenstates of the NV spin effective Hamiltonian in Eq. (7), and the nuclear spin $\{|\uparrow_L\rangle, |\downarrow_L\rangle\}$ becomes resonant. This allows for the polarization to be transferred efficiently from the NV center spin to the nuclear spin, when the NV center is initially prepared in the dressed spin down state $|\downarrow_x\rangle$, see Figure 1(a). As the NV center spin can be re-polarized by a green laser pulse, the polarization transfer process can be repeated, see Figure 1(b), and thus allowing us to achieve high efficiency dynamical nuclear polarization [29]. We now consider an ensemble of NV centers, the transition frequencies of which have a certain distribution instead of a unique value of ω_0.

This leads to the fact that only a part of NV centers can match the Hartmann-Hahn resonance condition with the nuclear spin if we apply microwave continuous driving field with one fixed frequency ω and amplitude Ω, while most of the other NV centers would not contribute to dynamical nuclear polarization. These NV centers that approximately match the Hartmann-Hahn resonance condition shall satisfy $|\omega_0 - \omega| \ll g$. This means that $\gamma B(1 - \cos\theta) \ll g$. Thus, we can estimate that the overall dynamical nuclear polarization efficiency $\bar{P} \ll g/(2\gamma B)$, which is negligible for $g \sim 10$ kHz and $B \sim 50$-100 G. To overcome this obstacle, we apply the following microwave driving fields with multiple time-dependent frequencies and amplitudes

$$H_m(t) = \left[\sum_k \Omega_k(t)\cos\left[\omega_k(t)t\right]\right]\mathbf{S}_x. \tag{9}$$

For simplicity, we require that the values of $\omega_k(t)$ for different k are sufficiently separated from each other at each time, so that they will not effectively drive the same NV simultaneously. By optimizing $\Omega_k(t)$ and $\omega_k(t)$, one would be able to improve the efficiency of dynamical nuclear polarization.

3 DNP using a Lorentzian ensemble of NV centers

We first consider a simple case of NV center spins whose spectrum have a Lorentzian distribution

$$f(\omega) = \frac{1}{\pi}\frac{\Delta}{(\omega - \omega_c)^2 + \Delta^2}, \tag{10}$$

with the linewidth Δ. This may correspond to the scenario of an ensemble of NV centers with linebroadening arising from magnetic dipole-dipole interactions between NVs, where Δ is on the order of a few MHz [36]. The simplest choice of $\omega(t)$ is a linearly increasing function as

$$\omega_1(t) = \omega_c + \nu t, \tag{11}$$

$$\omega_2(t) = \omega_c - \Delta + \nu t, \tag{12}$$

where $\omega_1(0) = \omega_c$ and $\omega_2(0) = \omega_c - \Delta$ are the starting frequencies respectively, and ν is the frequency changing speed. For simplicity, in order to show the effect of the frequency sweep rate ν on the efficiency of dynamical nuclear polarization, we only consider the driving field with the frequency $\omega_1(t)$ and NVs with the transition frequency $\omega \geq \omega_c$. The effect of the other driving field with the frequency $\omega_2(t)$ on the NVs with $\omega \leq \omega_c$ is similar.

As an example to demonstrate how well the scheme would work, we consider the polarization dynamics of a ^{13}C at a distance of 1.5 nm from the electron spin. In this case, the interaction with the NV center spin is $\sim$5 kHz. For ^{13}C in the proximity of NVs, it is possible to polarize them using the other methods, see e.g. [33]. In Figure 2(a), we assume that an NV center has a transition frequency $\omega = (D - \gamma B) + (2\pi)5$ MHz, and the microwave frequency starts from $\omega(0) = D - \gamma B$ and increases linearly with a speed ν. It can be seen that at time $t = \Delta/\nu$, the microwave driving becomes resonant with the NV transition, the Hartmann-Hahn condition is satisfied and the nuclear spin is being polarized (i.e. the nuclear spin down population increases). For a smaller rate of change of the driving field

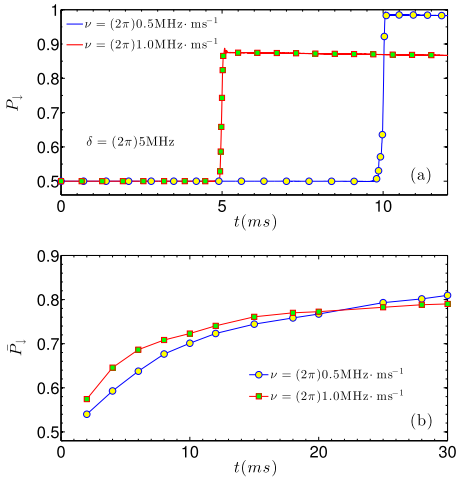

Figure 2 Dynamical nuclear polarization using the frequency-modulated microwave driving field. (a) The polarization dynamics of a ^{13}C at a distance of 1.5 nm from the NV center with the transition frequency $\omega = (D - \gamma B) + \delta$, where $\delta = (2\pi)5$ MHz, $P_\downarrow$ is the nuclear spin down state population, and the frequency of microwave field changes at the speed of ν. **(b)** The average nuclear spin down state population $\bar{P}_\downarrow$ as a function of the total polarization time for the frequency sweeping speed $\nu = (2\pi)1$ MHz $\cdot$ ms^{-1} and $\nu = (2\pi)2$ MHz $\cdot$ ms^{-1}, using an ensemble of NV centers with a Lorentzian transition frequency distribution, the corresponding linewidth is $\Delta = (2\pi)10$ MHz. In both **(a)** and **(b)**, the magnetic field is $B = 200$ G and is aligned along the NV axis, and the Rabi frequency of microwave field is $\Omega = \gamma_C B$ where γ_C is the gyromagnetic ration of ^{13}C, and the NV center is re-polarized every 120 μs.

frequency, a longer sweep time to reach the resonant point is required, while the resonance condition may remain approximately satisfied for a longer time. As a consequence we obtain a longer interaction time and thus a higher spin polarization, see Figure 2(a). In Figure 2(b), we consider an ensemble of NV centers, which have a Lorentzian spectrum (Eq. (10)) with a linewidth Δ, and plot the average nuclear spin down population achieved $\bar{P}_\downarrow$ that characterizes the overall dynamical nuclear polarization. For a shorter total polarization time, the polarization efficiency is better with a faster microwave frequency sweep, as more NV centers will become resonant in the same total time of the experiment. In contrast, if a longer polarization time is allowed, it is preferable to sweep the microwave frequency at a lower speed to enable more sufficient polarization transfer for each NV center.

4 DNP using NVs of random axis orientation

Given a specific direction of magnetic field, the transition frequency of the NV center spin is

$$\omega(\theta) = D - \gamma B \cos(\theta). \tag{13}$$

Here, we assume that the applied magnetic field is much smaller than the zero field splitting D. The transition frequencies of NV centers with random NV axis orientations, e.g. in diamond powder [33, 34], are in the range $[D-\gamma B, D+\gamma B]$. Using the standard Hartmann-Hahn condition based dynamical nuclear polarization, the achievable overall nuclear polarization is negligible. Starting from one single frequency ω_0, and using a microwave sideband frequency of Δ_0, we can obtain another two frequency component $\omega_1^{(1)} = \omega_0 + \Delta_0$ and $\omega_1^{(2)} = \omega_0 - \Delta_0$ by adding another frequency component Δ_0, where $\Delta_0 = \gamma B$. In a similar way, we can get another two frequency components $\omega_2^{(1)} = \omega_0 + \Delta_0 - \Delta_1$ and $\omega_1^{(2)} = \omega_0 - \Delta_0 + \Delta_1$ within the above range. Particularly, we choose $\Delta_1 = \Delta_0/2$, and thus obtain four equally separated frequency components that we would use for frequency-modulated dynamical nuclear polarization: $\omega_0(t) - \Delta_0$, $\omega_0(t) - \Delta_0/2$, $\omega_0(t)$, $\omega_0(t) + \Delta_0/2$ with the initial value $\omega_0(0) = D$. More generally, following the same procedure, we can use K microwave frequencies and get 2^{K-1} equally separated frequency components, see

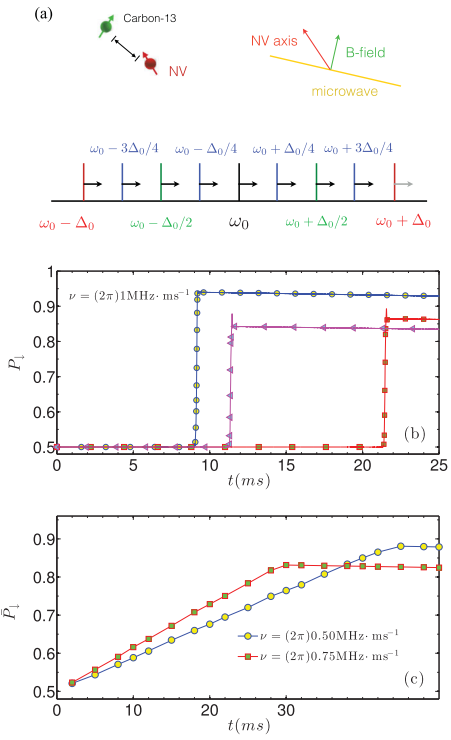

Figure 3 Efficient DNP using NVs of random axis orientation. (a) The parallel dynamical nuclear polarization dynamics with microwave driving fields of multiple time-dependent frequencies. The microwave wire is assumed to be perpendicular to the direction of the magnetic field. **(b)** The polarization dynamics of a ^{13}C at a distance of 1.5 nm using single NV center with different symmetry axis (NV axis) orientation. $P_\downarrow$ is the nuclear spin down state population, and the central frequency of microwave field $\omega_0(t)$ changes at the speed of $\nu = 1$ MHz $\cdot$ ms^{-1}. **(c)** The average nuclear spin down state population $\bar{P}_\downarrow$ as a function of the total polarization time t for the frequency changing speed $\nu = 0.75$ MHz $\cdot$ ms^{-1} and $\nu = 0.5$ MHz $\cdot$ ms^{-1}, using an ensemble of NV centers with random NV axis orientations. In both **(a)** and **(b)**, the magnetic field is $B = 250$ G, the amplitude of microwave field is $\Omega = \gamma_C B$, where γ_C is the gyromagnetic ratio of ^{13}C, and the NV center is re-polarized every 100 μs.

Figure 3(a) for an example of $K = 4$. An alternative and simpler way is to use an arbitrary waveform generator. Therefore, we generate the following microwave driving fields with 2^{K-1} time-dependent frequencies

$$H_m(t) = \left[\sum_{k=1}^{2^{K-1}} \Omega_k(t)\cos\left[\omega_k(t)t\right]\right] S_x, \tag{14}$$

where

$$\omega_k(t) = \omega_0(t) - \Delta_0 + \frac{k}{2^{K-1}}\Delta_0, \tag{15}$$

and

$$\omega_0(t) = D + \nu t, \tag{16}$$

with ν the frequency sweeping speed. We assume that the microwave wire is oriented perpendicular to the direction of the magnetic field, see Figure 3(a). Note that only the microwave component that is orthogonal to the NV axis will drive the NV spin transition, the effective Rabi frequency is thus $\Omega_k^{eff}(t) = \Omega_k(t)\cos(\theta)$. It is the aim that at the time t, the corresponding microwave will drive the NVs with the transition frequency $\omega_k(t) = D + \gamma B\cos(\theta)$ resonantly and establish the Hartmann-Hahn condition between those NVs and nuclei. To this end we require $\Omega_k(t)\cos(\theta) = \omega_L = \gamma_L B$, which leads to

$$\Omega_k(t) = \gamma_L\gamma\frac{B^2}{\omega_k(t) - D}. \tag{17}$$

To avoid the divergence of microwave power, we put the following constraint on the maximum value of the individual Rabi frequency $|\Omega_k(t)| \leq (2\pi)1$ MHz.

In Figure 3(b), we plot the polarization dynamics of ^{13}C with $K = 7$ using three NV centers respectively with random orientations. It can be seen that, as these NV centers have different NV symmetry axis orientations, their transition frequencies are different (see Eq. (13)), the NV centers start to polarize ^{13}C at different times when the frequency of the microwave fields matches the transition frequency individually. In Figure 3(c), we calculate the overall efficiency for dynamical nuclear polarization using an ensemble of NV centers with random NV axis orientations as a function of the total running time. For a given frequency sweeping speed ν, the optimal time is $t_o = \Delta_c/\nu$, where $\Delta_c = \Delta_0/2^{K-1}$ is the interval between two sweeping frequencies, see Figure 3(a). One can also see that given the experiment time $t \geq \Delta_c/\nu$, the lower the frequency sweeping speed the higher the overall dynamical nuclear polarization efficiency.

5 Conclusion

We have generalized the scheme of dynamical nuclear polarization using NV centers in diamond based on the Hartmann-Hann condition to the scenarios when an ensemble of NV centers having inhomogeneous linebroadening. By sweeping the frequencies of microwave driving fields, it is possible to enable a large amount of NV centers make contribution to dynamical nuclear polarization, and thus significantly improve the overall dynamical nuclear polarization efficiency. With detailed numeric calculations, we demonstrate that the scheme performs well in two specific and important cases, namely dynamical nuclear polarization using an ensemble of NV centers with a Lorentzian spectrum, and using an ensemble of NV centers with random NV axis orientations in diamond powder. The present results will be helpful for the implementation of dynamical nuclear polarization based on NV centers in diamond and its applications in magnetic resonance imaging and spectroscopy.

Competing interests
The authors declare that they have no competing interests.

Authors' contributions
All authors contributed equally to the writing of this paper. All authors read and approved the final manuscript.

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