

Time-optimal rotation of a spin $\frac{1}{2}$: Application to the NV center spin in diamond

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We study the applicability of the time-optimal bang-bang control designed for spin $\frac{1}{2}$ [Boscaïn and Mason, *J. Math. Phys.* **47**, 062101 (2006)] to the rotation of the electron spin of a nitrogen-vacancy (NV) center in diamond. The spin of the NV center is a three-level system, with two levels forming a relevant qubit subspace where the time-varying magnetic control field performs rotation, and the third level being idle. We find that the bang-bang control protocol decreases the rotation time by 20%–25% in comparison with the traditional oscillating sinusoidal driving. We also find that for most values of the bias field, the leakage to the idle level is very small, so that the NV center is a suitable testbed for experimental study of the time-optimal protocols. For some special values of the bias field, however, the unwanted leakage is greatly increased. We demonstrate that this is caused by the resonance with higher-order Fourier harmonics of the bang-bang driving field.

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I. INTRODUCTION

Studying the dynamics of quantum spins helps to better understand many fundamental issues of quantum physics, and it is important for applications in quantum-information processing,¹ coherent spintronics,² and high-precision metrology.³ Recently, spins of the nitrogen-vacancy (NV) impurity centers in diamond have emerged as a particularly strong candidate for long-range quantum communication,⁴ scalable quantum computation,^{5,6} and high-sensitivity magnetometry with nanoscale resolution.^{7–9} NV centers also present an excellent platform for exploring such fundamental problems as the dynamics of quantum spins coupled to their environment,^{10–13} and quantum control and dynamical decoupling of single solid-state spins.^{13–16} This system has become very popular due to a favorable combination of properties: a NV electronic spin has a long coherence time,^{10,11,17,18} a single NV center can be optically initialized and read out,^{19,20} and it can be manipulated both optically²¹ and magnetically.²²

In this paper, we consider fast manipulation of the spin of a single NV center by an externally applied magnetic field, which is important for many prospective applications. The magnetic field can be quickly varied in time with modern electronics, but its magnitude is always limited, so the problem of time-optimal spin control naturally arises, namely how to shape the time profile of the control field so that the desired spin evolution is accomplished within the shortest time. Time-optimal control is a well-developed science nowadays,²³ and the optimal control of quantum spins has been studied extensively in various contexts, from general mathematical considerations of the optimal control of two-level systems^{24–32} to numerous applications, e.g., in the areas of magnetic resonance,^{25,33,34} and quantum-information processing.^{35–39} In most cases, when the system under consideration is sufficiently complex, one has to use numerical methods to find the optimal control. There are, however, rare cases in which the exact solution is known, and its optimality is rigorously proven.^{26,27} It is of interest to see whether NV centers in diamond may present a suitable system for experimental test of such fundamental results of the control theory. Besides basic

interests, such protocols can speed up control of NV spins in future applications.

We consider the exact solution for time-optimal control of a spin $\frac{1}{2}$, placed in a static magnetic field along the z axis, with the corresponding energy splitting $2E$ between the states $|\uparrow\rangle$ and $|\downarrow\rangle$. As in standard resonance experiments,⁴⁰ a driving field $B_x(t)$ of limited amplitude, $|B_x(t)| \leq M$, can be applied along the x axis to rotate the spin from $|\downarrow\rangle$ to $|\uparrow\rangle$. The traditional solution is to use driving, which oscillates with the spin's Larmor frequency $2E/\hbar$: for small driving, $M \ll E$, within the rotating-wave approximation (RWA), it implements the desired rotation. However, this approach is not optimal: only half of the oscillating control field (the co-rotating component⁴⁰) is used to rotate the spin, while the other half (the counter-rotating component) is wasted. Also, at strong driving, the RWA breaks down, making the spin dynamics intricate.^{16,41,42} It has been proven²⁷ that for a single-axis driving, the bang-bang control is optimal, with the driving field $B_x(t)$ switching between its extremal values $\pm M$.

Here we investigate whether this exact solution is applicable to the spin of a NV center. A NV center has spin $S = 1$ with the anisotropy splitting $D = 2.88$ GHz between the levels $m_S = \pm 1$ and 0. Static external field applied along the anisotropy axis further splits the levels $m_S = +1$ and -1 . In most applications, the states $m_S = 0$ and -1 form the relevant two-level system (qubit, or a pseudo-spin $\frac{1}{2}$) while the third level $m_S = +1$ remains idle. However, when the optimal control designed for a spin $\frac{1}{2}$ is applied to the NV spin $S = 1$, the idle third level can become occupied. Protocols minimizing the leakage to higher levels have been developed before, using numerical tools, for different physical systems and situations.^{38,39,43,44} But numerical optimization does not guarantee global optimality, and is model-dependent. Thus, it is interesting to consider the opposite approach, and study the applicability of the known, rigorously optimal spin- $\frac{1}{2}$ protocol, to a NV spin rotation (although such protocols could be suboptimal when the full three-level system is considered). We show that for most regions of the relevant parameter space, the leakage to the idle level is negligible, so that the NV center does present a good testbed for the spin- $\frac{1}{2}$ protocol.

This also means that the known time-optimal protocol can be safely used to manipulate the NV center's (pseudo)spin, and can noticeably, by $\sim 25\%$, speed up its rotation compared to the RWA. Moreover, we investigate the cases in which the known solution for spin $\frac{1}{2}$ is not applicable, and we explain the reasons for the increased leakage to the idle NV spin state. The bang-bang control considered here may present an alternative to the two-axis driving,^{26,45} where the magnetic field of maximum possible amplitude M rotates in the x - y plane, and which requires two independent fields along the x and y axes with precisely locked directions, phases, and amplitudes.

The rest of the paper is organized as follows. In Sec. II, we describe in more detail the spin Hamiltonian of the NV center, the time-optimal spin- $\frac{1}{2}$ protocol, and our results. Discussion and conclusions are given in Sec. III.

II. RESULTS: BANG-BANG CONTROL OF THE NV CENTER AND ITS PERFORMANCE

The nitrogen-vacancy color center in diamond consists of a substitutional nitrogen atom with an adjacent vacancy. Its many-electron orbital ground state has a total spin⁴⁶ $S = 1$. The combined effect of the spin-orbit and spin-spin interactions leads to a single-axis spin anisotropy, with the anisotropy axis directed along the $\langle 111 \rangle$ crystallographic direction (symmetry axis of the NV center), which we take as the z axis. As a result, the states $m_S = \pm 1$ and 0 are split by $D = 2.88$ GHz (everywhere in this paper, we assume $\hbar = 1$). The external static field applied along the z axis further splits the states $m_S = 1$ and -1 , as shown schematically in Fig. 1(a), and the total Hamiltonian of the system is

$$H_0 = DS_z^2 + B_0 S_z + B_x(t) S_x, \quad (1)$$

where $B_x(t)$ is the control field to be optimized. When NV spin is used, for instance as a qubit for quantum-information processing, only the two levels $m_S = 0$ and -1 are employed. The control field $B_x(t)$, whose amplitude is bounded $|B_x(t)| \leq M$, is applied to drive the spin from the initial state $m_S = 0$ to the final state $m_S = -1$. Since the heating of the sample is not a significant issue in current experiments, the total driving power does not pose a limitation. But the bounded amplitude

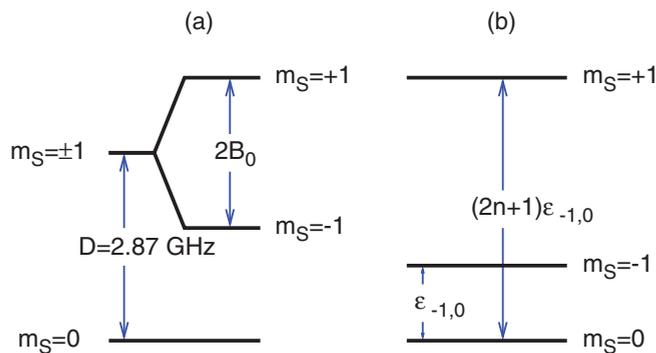


FIG. 1. (Color online) (a) Energy levels of the NV center spin $S = 1$. The states $m_S = 0$ and -1 form the relevant qubit subspace. (b) The position of the energy levels when the higher-order harmonics in the control field can induce the resonant leakage to the idle level $m_S = +1$.

of the driving field, determined by the details of the amplifier, presents a major restriction.

Within the relevant subspace spanned by these two eigenstates, the system is equivalent to a (pseudo)spin $s = \frac{1}{2}$. The Hamiltonian for this spin, omitting the irrelevant energy shift, is

$$H = 2Es^z + \sqrt{2}B_x(t)s^x, \quad (2)$$

where $E = (D - B_0)/2$, and $B_x(t)$ should steer the spin from $|\downarrow\rangle$ to $|\uparrow\rangle$. The optimal control for this problem has been thoroughly investigated in Ref. 27. The time-optimal rotation of the (pseudo)spin is implemented by the bang-bang control, where the control field $B_x(t)$ switches between $+M$ and $-M$. The term *bang* here, following the standard terminology, denotes the interval between two neighboring switchings, i.e., during a bang the control field is constant. When the maximal control field is strong, $M > \sqrt{2}E$, the time-optimal rotation is achieved with exactly one switching,²⁷

$$B_x(t) = \begin{cases} M, & 0 \leq t < t_1, \\ -M, & t_1 \leq t \leq T, \end{cases} \quad (3)$$

where the switching time $t_1 = [\pi \pm \arccos(2E^2/M^2)] / (2\sqrt{E^2 + M^2/2})$, and the optimal control time is

$$T = \frac{\pi}{\sqrt{E^2 + M^2/2}}. \quad (4)$$

The corresponding spin trajectory on the Bloch sphere is rather simple: the spin vector starts from the south pole of the sphere ($|\downarrow\rangle$ state), rotates about the axis $2E\hat{z} + \sqrt{2}M\hat{x}$ until it hits the equator, then continues to rotate about the axis $2E\hat{z} - \sqrt{2}M\hat{x}$ all the way to the north pole ($|\uparrow\rangle$ state). Here \hat{x} and \hat{z} are the unit vectors along the x and z axes, respectively. Note that the point $E = 0$ is singular: a static field of maximum amplitude would perform the necessary rotation twice as fast as the protocol of Ref. 27. However, this can be used only in a very special situation, in the nearest vicinity ($E \ll M$) of the point $E = 0$, so we do not consider this singular case below.

For weak driving $M < \sqrt{2}E$, the duration of all bangs is the same, except for the initial and final bangs:

$$B_x(t) = (-1)^{k-1} M, \quad [0, t_i], k = 1 \\ t \in [t_i + (k-2)t_m, t_i + (k-1)t_m], k = 2, 3, \dots, n_{\text{swt}} \quad (5) \\ [t_i + (k-2)t_m, t_i + (k-2)t_m + t_f], k = n_{\text{swt}} + 1$$

where t_i , t_m , and t_f denote the durations of the initial, middle, and final bangs, respectively. n_{swt} is the number of times the field switches between $+M$ and $-M$, and $k = 1, 2, \dots, n_{\text{swt}} + 1$ denotes the bangs. There are also theorems relating t_m and t_f to t_i , and limitations on the number of field switchings.²⁷

Instead of the protocol of Ref. 27, a traditional approach for spin rotation employs a driving field along the x axis oscillating at Larmor frequency, $B_x(t) = M \cos(2Et)$. This control works well in the weak-driving regime, $M \ll E$. Within the rotating-wave approximation (RWA),⁴⁰ when only the secular terms are retained in the control field, the oscillating field is equivalent to a driving field with magnitude $M/2$, which rotates in the x - y plane in step with the Larmor precession of

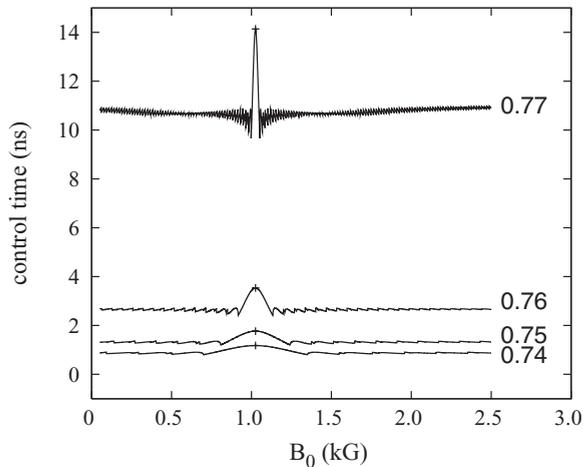


FIG. 2. Spin rotation time as a function of the static field, B_0 , for different amplitudes of the control field, $M = 50, 200, 400$, and 600 MHz from top to bottom, respectively. The rotation time at the peak of each curve is (accidentally) equal to t_{RWA} , and is denoted by the cross on each curve. The number on the right of each curve indicates the ratio of the control time to t_{RWA} at large bias B_0 .

the spin s . The corresponding rotation time is $t_{RWA} = \sqrt{2}\pi/M$. In a general situation, when the control field can be directed anywhere in the x - y plane, the rotating field is known to provide time-optimal control.²⁶ However, in reality it may not be easy to create two perpendicular control fields with precisely locked directions, amplitudes, and phases. Although such a rotating field has been implemented,⁴⁵ the single-axis driving still remains more convenient for experiments. For sinusoidal single-axis driving, only half of the control field approximates the optimal two-axis rotating-wave solution, while the other half (corresponding to the omitted nonsecular terms in the RWA) is wasted. The solution of Ref. 27 utilizes the full magnitude of the single-axis control in an optimal way. For $M \ll E$, its approximately periodic structure resembles the oscillating driving, but the period (duration of the bangs) is slightly different, and its time shape is rectangular rather than sinusoidal.

In a typical experiment, the maximum magnitude of the driving field M is fixed, limited by the setup details, while the static external field B_0 is adjusted to provide the convenient conditions for driving the rotation of the NV center spin from $m_S = 0$ to -1 . Figure 2 shows the rotation time for the protocol of Ref. 27 as a function of the bias B_0 for different driving amplitudes $M = 50, 200, 400$, and 600 MHz. The results here and below have been obtained by directly solving the time-dependent Schrödinger equation for $S = 1$ via the second-order Runge-Kutta method. Overall, as expected, the rotation time decreases approximately as $1/M$. At $B_0 = 1028$ G, the distance $2E = D - B_0$ between $m_S = 0$ and -1 is zero (level crossing point), and the rotation time is maximal. The corresponding control time $2\pi/(\sqrt{2}M)$ is, accidentally, equal to t_{RWA} , although the RWA is not applicable here. Farther from the level crossing point, the rotation time decreases. At very large distance $2E$, the rotation time almost saturates at the value not very far from²⁷ $t_{RWA}/\sqrt{2}$. Therefore, the protocol of Ref. 27 can decrease the rotation time by about 25% in comparison with the standard oscillatory $B_x(t)$.

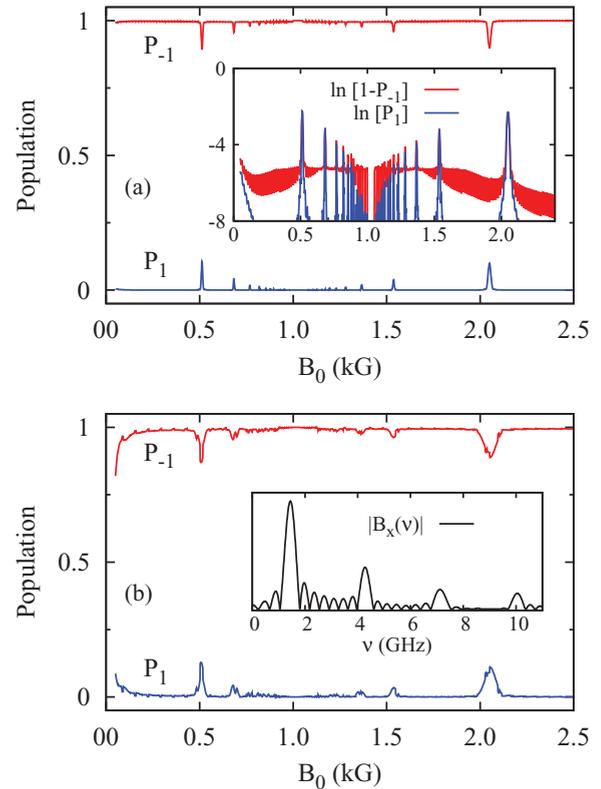


FIG. 3. (Color online) Dependence of the final population P_{-1} (red line) and P_1 (blue line) on the static field, B_0 , after the application of the optimal control for the corresponding two-level system. (a) The results for $M = 50$ MHz. The inset shows the values $\ln[1 - P_{-1}]$ (red) and $\ln[P_1]$ (blue) vs B_0 . (b) The results for $M = 200$ MHz. The inset shows the Fourier transform of the bang-bang control, $B_x(v)$, for static bias $B_0 = D/2$. The first peak at 1.435 GHz corresponds to the desired transition between levels $m_S = 0$ and -1 , while the second peak at 4.305 GHz drives the unwanted transition to the $m_S = +1$ state.

Figure 3 shows the fidelity of the spin- $\frac{1}{2}$ protocol when applied to a NV spin $S = 1$. Except for a few sharp dips at certain bias fields, the final population P_{-1} of the targeted level $m_S = -1$ stays high, and far from the level crossing it saturates at the values 0.999, 0.994, and 0.986 for $M = 50, 200$, and 600 MHz, respectively. Thus, the protocol of Ref. 27 provides good fidelity for $S = 1$ spin of the NV center for most values of B_0 . The loss of fidelity, including the neighborhood of the dips, is predominantly due to leakage into the idle state $m_S = +1$, as seen in Fig. 3, and the dips in P_{-1} are accompanied by the peaks of P_1 .

Such leakage is particularly prominent when the energy levels satisfy the condition

$$\epsilon_{+1,0} = (2n + 1)|\epsilon_{-1,0}|, \quad (6)$$

where $\epsilon_{\pm 1,0}$ are the energy splitting between $m_S = \pm 1$ and 0, respectively, and $n = 0, 1, 2, \dots$. In terms of B_0 , these conditions are

$$B_0 = \frac{n}{n+1}D \quad \text{or} \quad \frac{n+1}{n}D. \quad (7)$$

The increased leakage is determined by the (approximately) square-wave time profile of $B_x(t)$, with (approximate) period $2t_m$; see Eq. (5). The Fourier expansion of the square wave

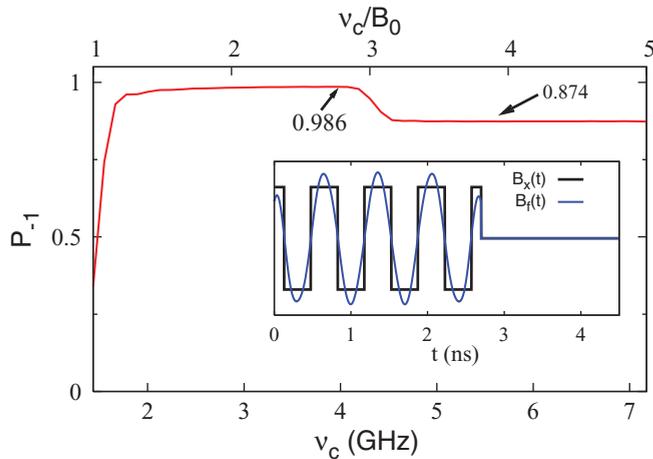


FIG. 4. (Color online) Final population P_{-1} of the $m_S = -1$ state as a function of the cutoff frequency ν_c , for $M = 200$ MHz and $B_0 = D/2 = 1.435$ GHz (where the energy gap between $m_S = 0$ and -1 equals B_0). When ν_c becomes smaller than $\epsilon_{+1,0}$, the leakage to $m_S = +1$ is suppressed, and P_{-1} abruptly increases. The inset shows the time profile of the original bang-bang [$B_x(t)$, black line] and filtered [$B_f(t)$, blue line] controls. The bang-bang control has $t_i = t_f = 0.1266$ ns, $t_m = 0.3497$ ns, with $n_{\text{swt}} = 8$ switchings.

contains all frequencies $(2n + 1)\pi/t_m$ with integer n , and these features are prominent in the Fourier transform of the bang-bang control $B_x(t)$ [inset of Fig. 3(a)]. The harmonics of the driving field come in resonance with the transition between $m_S = 0$ and $+1$, see Fig. 1, thus drastically increasing the leakage. To demonstrate this, we set the external field at the value $B_0 = D/2$ ($n = 1$) corresponding to the middle of the pronounced dip in Fig. 3. Instead of the actual bang-bang control $B_x(t)$, which leads to $P_{-1} = 0.874$, we used its filtered version $B_f(t)$ by cutting off the Fourier harmonics with frequencies above a certain value ν_c . Also, to make the comparison fair, the control time for filtered control was kept the same as the original bang-bang control time [by turning off the filtered control $B_f(t)$ at longer times; see Fig. 4]. Figure 4 shows the dependence of P_{-1} on the cutoff value: as soon as ν_c becomes smaller than $3D/2$, the value of P_{-1} demonstrates a clear jump, since the unwanted resonant harmonic in the driving field is filtered out almost completely, and the leakage to the $m_S = +1$ state is suppressed.

The analysis of the filtered control also shows that the formally infinite bandwidth of the bang-bang control does not present a serious problem for experiments. As long as the cutoff frequency is large enough (i.e., an amplifier with sufficiently large bandwidth is used), the control performance is not significantly affected by the high-frequency cutoff.

III. CONCLUSIONS AND DISCUSSIONS

NV centers in diamond represent a very suitable system for fundamental studies of quantum spin dynamics, and for

many quantum spin-based applications. In many applications so far, the spin of a NV center was used to represent a qubit, with the states $m_S = 0$ and -1 forming the relevant two-level subspace, and the state $m_S = +1$ remaining idle. We investigated whether the spin $S = 1$ of a NV center constitutes also a suitable testbed for studying the time-optimal control protocol designed for two-level systems. We found that by applying the optimal control protocol developed in Ref. 27, the rotation time from the $m_S = 0$ to -1 state can be reduced by $\sim 25\%$ in comparison with the standard sinusoidal oscillatory driving (within the RWA approximation). We checked how important is the unwanted leakage to the idle state $m_S = +1$. We found that this leakage, for most values of the external field, is negligible, so that the NV center indeed could be a suitable platform for studying the time-optimal control problems. We found, however, that near certain values of the bias field B_0 , the leakage to the idle state is drastically increased, and we demonstrated that this is induced by higher-order harmonics in the bang-bang driving. We show that by frequency filtering, this leakage can be noticeably suppressed.

Experimental implementation of the protocol studied above on NV centers seems reasonable. Currently, the magnetic fields that drive the desired rotation can be controlled on time scales of tens of picoseconds (or even faster with specialized techniques). The driving field amplitudes of hundreds of MHz have also been implemented in many experiments.^{13–16} Moreover, for faster rotation, it is important to move far away from the level crossing point at $B_0 = 1028$ G, so that negative external bias or very large external field is more favorable for the rotation speedup.

It is important to note that the focus of this work is to check whether a NV spin is a suitable testbed for experimental studies of the time-optimal protocols. Although the protocol studied above does speed up the NV spin rotation, it most probably presents a suboptimal solution for the full system $S = 1$. However, the detailed studies of the time-optimal control of three-level systems present a difficult mathematical problem, and are still lacking. In this case, using an optimal solution within a restricted subspace is a reasonable option, which may complement the numerical solution of the time-optimal problem.

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