

Time-optimal control of a  $\text{GdW}_{30}$  molecular spin quditMarc Alsina <sup>\*</sup>

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(Received 21 November 2025; accepted 31 March 2026; published 2 June 2026)

Quantum systems with more than two levels, known as qudits, offer enhanced information density and potentially more efficient quantum operations compared with traditional qubits. Among the various physical platforms under investigation, single-molecule magnets (SMMs) stand out due to their intrinsic magnetic anisotropy, long spin-relaxation times, and the natural presence of multiple accessible spin states. In this work, we explore two distinct control strategies for implementing state-to-state transitions within the multilevel structure of SMMs, viz., a resonant magnetic-field manipulation ( $\pi$ -pulse control) and the so-called quantum Zermelo navigation approach, which provides the time-optimal Hamiltonian that transforms a given initial state to a given target state. We present a comparative analysis of these two methods, evaluating their operational efficiency and feasibility for experimental realization. Our results show that, while  $\pi$ -pulse control is limited by strict timing constraints, the Zermelo method faces significant challenges in terms of its physical implementation.

DOI: [10.1103/c7yr-jrs4](https://doi.org/10.1103/c7yr-jrs4)

## I. INTRODUCTION

With the emergence of quantum computing, a wide range of highly impactful computational capabilities has been devised [1–7]. Quantum computers have potential advantages over classical computers in specific tasks, such as the factorization of large numbers with Shor's algorithm [7–9], the acceleration of unstructured search problems with Grover's algorithm [7,10,11], and the simulation of complex biological and chemical systems [12–14], to mention a few. Although most applications remain in their early stages of development, ongoing progress in quantum computing is widely regarded as

essential for future technological and scientific advancement.

One of the main challenges preventing quantum computing from becoming a viable solution is its high sensitivity to noise, particularly in the current generation of devices known as Noisy Intermediate-Scale Quantum (NISQ) Computers [7]. These systems are limited by short coherence times—commonly denoted as T1 (energy relaxation time) and T2 (de-phasing time)—which restrict the duration over which quantum information can be reliably processed [7,15]. To overcome these limitations, several approaches are under active development, including quantum error correction and mitigation [16,17], time-optimal quantum control methods that implement quantum operations faster than decoherence [18–21], and the design of novel quantum hardware architecture with improved coherence and scalability [22,23].

This work emphasizes the significance of time-optimal quantum control (TOQC) as a guiding principle in quantum computing research. Given that quantum operations must be completed within the limited coherence times of current hardware, TOQC focuses on identifying the fastest possible control strategies given a fixed energy resource for the control. This approach offers three key advantages. First, faster operations reduce the risk of decoherence, thereby increasing

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computational fidelity. Second, more operations can be performed within the available coherence window, enhancing the computational depth of NISQ devices. Third, by shortening execution times, TOQC can reduce the burden associated with quantum error correction, potentially easing the path toward practical quantum advantage. As such, TOQC is widely viewed as a promising avenue for advancing scalable quantum computing [24].

The field of TOQC traces its origins to early work in the calculus of variations, notably by Carathéodory [25], which introduced Zermelo's navigation problem: given a ship in a flowing medium (e.g., a sea current), what is the optimal steering strategy to reach a destination in the shortest possible time? An analytical solution was provided for the case of a medium with constant velocity. This classical framework has since inspired the formulation of analogous problems in the context of quantum control, where the goal is to steer a quantum system from an initial to a target state in minimal time under certain physical constraints [18,20,21]. In particular, the so-called Quantum Zermelo navigation problem adapts the original classical scenario to unitary evolution under a drift Hamiltonian and control terms, and has been shown to admit time-optimal solutions. Recent experimental realizations have demonstrated the practical implementation of these optimal navigation protocols in quantum systems [26–29], showing improved efficiency compared with conventional control methods. These findings underscore the relevance of time-optimal quantum control strategies as a promising approach for enhancing the performance of near-term quantum devices.

In this work, we focus on the implementation of Zermelo time-optimal control within a specific class of quantum systems: single-molecule magnets (SMMs). These are molecules that display magnetic bistability under certain thermodynamic conditions and have emerged as promising platforms for quantum information processing. SMMs span a broad chemical landscape, from radical organic compounds to organometallic complexes containing one or more magnetic centers. Their appeal in quantum computing stems from their exceptional tunability through careful molecular design, which allows us to precisely tailor their magnetic properties, reduce sources of magnetic noise, and thereby extend coherence times [30].

A particularly attractive feature of SMMs is their potential to function not only as qubits but also as qudits. Qudits enable more compact encoding of quantum information and may improve gate efficiency and error resilience [31,32]. This can be achieved by designing molecules with high-spin magnetic centers (e.g.,  $S > 1/2$ ) or by incorporating multiple coupled spin centers within a single molecular framework [30]. Another key advantage of SMMs is their energy level structure, which allows for significant splitting between spin states under low magnetic fields achievable at experimentally accessible cryogenic temperatures. Finally, many SMMs demonstrate good chemical and structural stability, even in solution, which is essential for scalable synthesis and integration into quantum hardware architectures [30].

Given these attributes, SMMs provide a convenient platform for exploring advanced quantum control strategies. In this study, we focus on two distinct approaches for implementing state-to-state transitions within the multilevel spin

structure of SMMs: (i) resonant magnetic-field manipulation, commonly referred to as  $\pi$ -pulse control, and (ii) the Quantum Zermelo Navigation method. These strategies are evaluated through a comparative analysis, with particular attention to their operational efficiency and feasibility under realistic experimental constraints.

Our analysis centers on the manipulation of spin transitions between eigenstates of the molecular spin Hamiltonian operations that are fundamental to qudit-based quantum computing. Efficient and precise control of these transitions is critical, as it directly impacts gate fidelity and error rates. In multilevel systems, the number of possible transitions increases rapidly with dimensionality, complicating control and increasing susceptibility to decoherence and leakage. While qudit architectures offer theoretical advantages such as reduced circuit depth and improved fault tolerance, these benefits can only be realized if control methods scale effectively and maintain high precision.

By comparing the conventional  $\pi$ -pulse protocol with the more recently proposed Zermelo-based time-optimal strategy, we aim to elucidate the trade-offs between accessibility and performance. The Zermelo approach, which minimizes transition time under physical constraints, holds promise for reducing decoherence and enhancing fidelity—particularly in systems with dense energy spectra, as is typical for SMMs. Through this comparative framework, we seek to highlight both the theoretical benefits and practical considerations of optimal control in high-dimensional quantum systems.

The comparison is designed to elucidate the operational meaning and physical attainability of Zermelo optimal control in molecular spin qudits, rather than to restate its formal optimality. By contrasting Zermelo dynamics with a standard, experimentally realizable  $\pi$ -pulse protocol, we identify which features of time-optimal control survive under physically constrained Hamiltonians.

Our analysis demonstrates that, while Zermelo control formally saturates the quantum speed limit, the corresponding optimal Hamiltonian requires control terms that cannot be generated by any physically available interactions in single-molecule magnets. Crucially, we show that in the long-time regime, the reachable dynamics under  $\pi$ -pulse control converge to those of the Zermelo solution in terms of state transfer, but at the cost of fundamentally different control resources. This establishes that the Zermelo advantage is not a realizable speedup under magnetic-field-only control, but rather an upper bound defined in an unconstrained control landscape.

The novel insight provided by this work is therefore a sharp separation between formal time optimality and physical controllability. We show that, despite full theoretical controllability, single-molecule magnets cannot attain the quantum speed limit when realistic control constraints are imposed. This positions Zermelo control not as a practically superior protocol, but as a benchmark that exposes intrinsic limitations of experimentally feasible quantum control in high-spin molecular systems.

The remainder of this paper is organized as follows: Section II introduces the theoretical model and control schemes under investigation. Section III presents the results and discusses the implications of Zermelo time-optimal control for state-to-state transitions in SMMs.

## II. THEORETICAL FRAMEWORK

### A. Single molecule magnets

As an example of a real SMM with applications in quantum computing, we study the K<sub>12</sub>Gd(H<sub>2</sub>O)P<sub>5</sub>W<sub>30</sub>O<sub>110</sub> · 27.5H<sub>2</sub>O molecule, hereafter abbreviated as GdW<sub>30</sub>, which has been previously characterized experimentally [33,34] and studied for the implementation as a qudit [35]. Such a molecule pertains to the inorganic polyoxometalate family of molecules, which are characterized by being composed of a magnetic center and a surrounding framework.

Throughout, we assume the temperature is low enough to prevent thermal excitation [36]. Additionally, because of the 4f<sup>7</sup> electronic configuration of the Gd<sup>3+</sup> ion, it has no net orbital momentum ( $\langle \hat{L} \rangle = 0$ ), and we consider no spin-orbit coupling. Such a molecule is attributed to have the largest spin multiplicity ( $S = 7/2$ ) attainable with a unique atom, which allows us to encapsulate three qubits in a single physical entity [33,34]. These kinds of systems exhibit the zero-field splitting phenomena, where spin states are pair degenerated but separated among different spin states [33,34,36]. Finally, the available magnetic anisotropy allows sufficient energy anharmonicity to manipulate the spin states using conventional radio-frequency technologies [33,34,36].

Since only the spin wave function is of interest, and it can be decoupled from orbital states, such systems, consisting of a unique magnetic center and with zero net orbital momentum, can be described by the following phenomenological Hamiltonian, also known as the giant spin approximation [33–37]:

$$\hat{H}_0 = \hat{H}_1 + \hat{H}_2 + \hat{H}_{\text{Zeeman}}, \quad (1)$$

$$\hat{H}_1 = D[\hat{S}_z^2 - \hat{\mathbf{I}}S(S+1)/3], \quad (2)$$

$$\hat{H}_2 = E(\hat{S}_x^2 - \hat{S}_y^2), \quad (3)$$

$$\hat{H}_{\text{Zeeman}} = -g\mu_B \vec{\hat{S}} \cdot \vec{B}, \quad (4)$$

where  $D$  is the axial zero-field splitting parameter and  $E$  is the rhombic zero-field splitting parameter [36]. Equation (2) describes the  $z$  axial symmetry assumed in the spin states, giving as a result a set of degenerate spin states [37]. Equation (3) introduces an implicit perturbation that admixes the spin states  $|S, m_S\rangle$  [37]. Such perturbation results in the introduction of magnetic anisotropy in the  $x$ - $y$  plane, and removes the symmetry imposed by  $\hat{H}_1$ . If  $E > 0$ , then spins in the  $x$ - $y$  plane will avoid the  $x$  plane, and thus  $x$  axis will become the easy axis, the  $y$  axis will become the intermediate axis, and  $z$  axis will become the hard axis [36,37]. Finally, Eq. (4), known as the Zeeman interaction, comes from the external magnetic perturbation ( $\vec{B}$  being the magnetic field vector), which allows us to increase the energy separation between eigenstates [36–38]. The resulting eigenstates of  $\hat{H}_0$  will consist of an admix of  $|S, m_S\rangle$  where each spin state will be coupled with  $|S, m_S \pm 2\rangle$  states.

For the studied molecule, Jenkins *et al.* characterized experimentally the different parameters needed to supply in Eq. (2)–(4), being  $D = 1281$  MHz and  $E = 294$  MHz [33]. Because of the positive value of the  $E$  parameter, its easy axis will be the  $x$  axis, whereas the hard axis will be the  $z$  axis.

### B. Control of spin states

#### 1. Monochromatic pulses

The use of radio-frequency monochromatic pulses is the *de facto* technique when it comes to the manipulation of spin states [35,39,40]. The basis of such a technique is to couple the system with an oscillating magnetic field, with the associated interaction Hamiltonian given by [39]

$$\hat{H}_{\text{MRF}} = \hat{H}_{\text{Zeeman}} A \cos(\omega t), \quad (5)$$

where  $A$  is the amplitude of the monochromatic pulse, and  $\omega$  is the associated frequency.

To obtain a transition from an initial state  $|\psi_i\rangle$  to a final state  $|\psi_f\rangle$  [being both eigenstates of the Hamiltonian of Eq. (1)], the frequency of the monochromatic pulse must obey  $\Delta\omega := |\omega - \omega_{fi}| \ll |\omega_{fi}|$ , being  $\omega_{fi}$  the Bohr frequency associated with the transition. Additionally, such a technique will only be effective if the system perceives a true oscillating field. Thus, this implies that the protocol time  $\tau$  must be larger than  $1/|\omega_{fi}|$ , setting a lower time bound [39].

To infer specific values of both  $A$  and  $\omega$  to perform state-to-state transitions, we consider first that  $\Delta\omega = 0$ ; that is, we work with the Bohr frequency associated with the specific transition. The probability transition can be approximated by the Rabi formula [38]:

$$\mathcal{P}_{if}(t) = \sin^2 \left( \frac{A |\mathbb{H}_{\text{Zeeman}}^{fi}| t}{2\hbar} \right), \quad (6)$$

being  $\mathbb{H}_{\text{Zeeman}}^{fi} = \langle \psi_f | \hat{H}_{\text{Zeeman}} | \psi_i \rangle$ . Such approximation is valid if we assume that each transition is totally independent of the others, which implies that  $\Delta\omega^k \gg 0$  for each transition  $k$  that is different from the targeted one. By setting the transition probability equal to one and a given transition time  $t = \tau$ , we obtain the associated amplitude of the radio-frequency pulse to perform such a transition:

$$A = \frac{\pi \hbar}{\tau g \mu_B \langle \psi_f | \vec{\hat{S}} \cdot \vec{b} | \psi_i \rangle}, \quad (7)$$

where  $\vec{b}$  is the unit magnetic field vector associated with the dynamic or control magnetic field. Amplitudes given by Eq. (7) are commonly known as  $\pi$ -pulse waves [35].

#### 2. Quantum Zermelo formalism

Given an initial state  $|\psi_i\rangle$  and a final target state  $|\psi_f\rangle$ , the objective is to identify a control Hamiltonian  $\hat{H}_c(t)$  such that the total Hamiltonian  $\hat{H} = \hat{H}_0(t) + \hat{H}_c(t)$  induces the desired transformation within the shortest time interval  $\tau$ , while adhering to an energy constraint imposed by the control field  $||\hat{H}_c||$  (such a measure of cost will be motivated later). Here,  $\hat{H}_0(t)$  represents the Hamiltonian governing the background time-dependent drift field, which cannot be modified (and can be understood as the wind in the classical counterpart) [20,21].

The dynamics of the process will be dictated by a unitary transformation  $\hat{U}(t)$  such that  $|\psi(t)\rangle = \hat{U}(t) |\psi_i\rangle$ , obeying the Schrödinger equation for unitary operators [38]:

$$i\hbar \frac{d\hat{U}(t)}{dt} = [\hat{H}_0(t) + \hat{H}_c(t)] \hat{U}(t), \quad \hat{U}(0) = \hat{\mathbf{I}} \quad (8)$$

Without loss of generality, we introduce the following separable ansatz [20,41]:

$$\hat{U}(t) = \hat{U}_0(t)\hat{U}_c(t), \quad (9)$$

where  $\hat{U}_0(t)$  is the evolution operator associated with the action of the drift Hamiltonian only, obeying the Schrödinger equation:

$$i\hbar \frac{d\hat{U}_0(t)}{dt} = \hat{H}_0\hat{U}_0(t), \quad \hat{U}_0(0) = \hat{1} \quad (10)$$

and  $\hat{U}_c(t)$  is the time evolution operator associated with the action of the control Hamiltonian only. Introducing Eq. (9) into Eq. (8), we obtain

$$i\hbar \frac{d\hat{U}_c(t)}{dt} = \hat{U}_0^\dagger(t)\hat{H}_c(0)\hat{U}_0(t)\hat{U}_c(t) \quad (11)$$

Note that the interaction picture or the “rotating frame” representation arises naturally in Eq. (11) [41], which allows us to reduce the problem to inferring an initial form of the control Hamiltonian.

The solution of Eq. (10) can be obtained analytically and is given by  $\hat{U}_0(t) = e^{-i\hat{H}_0 t/\hbar}$ , which is valid only when the drift Hamiltonian is time independent. A more general geometric framework for constructing geodesic paths between two fixed states—even in the presence of a time-dependent background Hamiltonian—can be found in Ref. [21]. Since in our case the background Hamiltonian is time independent, we adopt the formulation developed by Bofill *et al.* [20]. This simplification enables us to express the control Hamiltonian as  $\hat{H}_c(t) = e^{-i\hat{H}_0 t/\hbar}\hat{H}_c(0)e^{i\hat{H}_0 t/\hbar}$ , which, upon differentiation with respect to time, gives

$$\frac{d\hat{H}_c(t)}{dt} = -\frac{i}{\hbar}[\hat{H}_0, \hat{H}_c(t)]. \quad (12)$$

Equation (12) governs the system’s trajectory along the minimum-time path connecting the initial state  $|\psi_i\rangle$  to the final target state  $|\psi_f\rangle$ . The same result was first derived by Carlini *et al.* [19] and later reformulated in a different framework by Brody and Meier [42,43].

So far, we have assumed a structure of the control Hamiltonian, which has to be initially defined. For such purposes, Bofill *et al.* [20] assumed that the control Hamiltonian generates orthogonal directions to the drift Hamiltonian [42] in order to guide the state through the minimum time path (also known as the geodesic trajectory). After some manipulation and imposing maximum and constant travel velocity, they reach the functional form for the control Hamiltonian at the initial time:

$$\hat{H}_c(0) = i\sqrt{k/2}(|\overline{\psi_f'}\rangle\langle\psi_i| - |\psi_i\rangle\langle\overline{\psi_f'}|), \quad (13)$$

where  $|\overline{\psi_f'}\rangle$  is the orthonormal state with respect to the initial state  $|\psi_i\rangle$  in the interaction picture, i.e.,  $|\psi_f'\rangle = \hat{U}_0^\dagger(\tau)|\psi_f\rangle$ .

The parameter  $k$  can be understood as the velocity or energy resource of the control protocol, which is written as  $k = 2(\cos^{-1}(|\langle\psi_i|\hat{U}_0^\dagger(\tau)|\psi_f\rangle|)/\tau^2)$ . An alternative derivation of solution (13) can be found in the Supplemental Material, Sec. 1.4 [44].

The resulting control Hamiltonian is hermitic and drives the quantum system through the line that stationarises the duration of the protocol [19]. A subtlety has to be noted in this

formulation: we are not making any prior assumption about the control, either in its field or coupling shape. Contrary to  $\pi$ -pulses, or many other control alternatives [35], we formulate a control that induces the desired transformation within the prescribed time, without imposing any specific form of the control Hamiltonian.

### III. RESULTS AND DISCUSSION

In this work, we focus on qudits rather than qubits when examining the implementability of Quantum Zermelo navigation because, in two-level systems, the problem is essentially trivial. In single-qubit systems, every target unitary belongs to  $SU(2)$ , whose Lie algebra can be generated under standard laboratory controls consisting of a drift Hamiltonian and at least one noncommuting drive. As a result, the Zermelo navigation problem always yields a physically realizable time-optimal control sequence for qubits, making implementability largely guaranteed. In contrast, multilevel systems exhibit a richer and more constrained dynamical landscape: selection rules, anharmonic spectra, and limited control channels can restrict the reachable subset of unitaries. Studying Zermelo navigation in the qudit setting is therefore significantly more informative because it tests the practical limits of optimal control under experimentally realistic constraints and highlights cases where time-optimal solutions may fail to be directly implementable.

In addition, we restrict our analysis to state preparation tasks. Specifically, eigenstate to eigenstate transitions rather than full quantum gate implementation or the generation of arbitrary superposition (an exception to this can be found in the Supplemental Material, where the Zermelo control scheme is considered for transitions between superpositions of different eigenstates [44]). Eigenstate-to-eigenstate transitions provide a clean and physically transparent benchmark for assessing the feasibility of time-optimal control: they isolate population transfer dynamics, avoiding the complications of phase tracking. Focusing on these transitions allows us to probe the core question of implementability under realistic control constraints while avoiding unnecessary overhead. At the same time, state-preparation fidelities directly reflect the dominant sources of error in multilevel systems and are foundational building blocks for more complex gate operations. Thus, limiting our study to eigenstate preparation enables conceptual clarity in evaluating the practical advantages of Quantum Zermelo navigation.

To generate the results presented in this work, we integrated the system dynamics numerically using an adaptive-step Magnus integrator [45–47], selected for its reliability in capturing time-dependent quantum evolution with high accuracy. This method adapts the time-step during propagation to maintain numerical stability, which is particularly important when dealing with rapidly varying control fields and multilevel energy structures. In our simulations, the integrator was configured with a time-step of  $\Delta t = 10^{-5}$  ns,  $\varepsilon = 10^{-14}$  and a norm tolerance of  $10^{-8}$ ; further details regarding implementation and convergence checks are provided in Supplemental Material, Sec. 1.1 [44]. In addition, the reference control pulses based on standard radio-frequency driving were refined through a gradient-descent optimization routine to improve

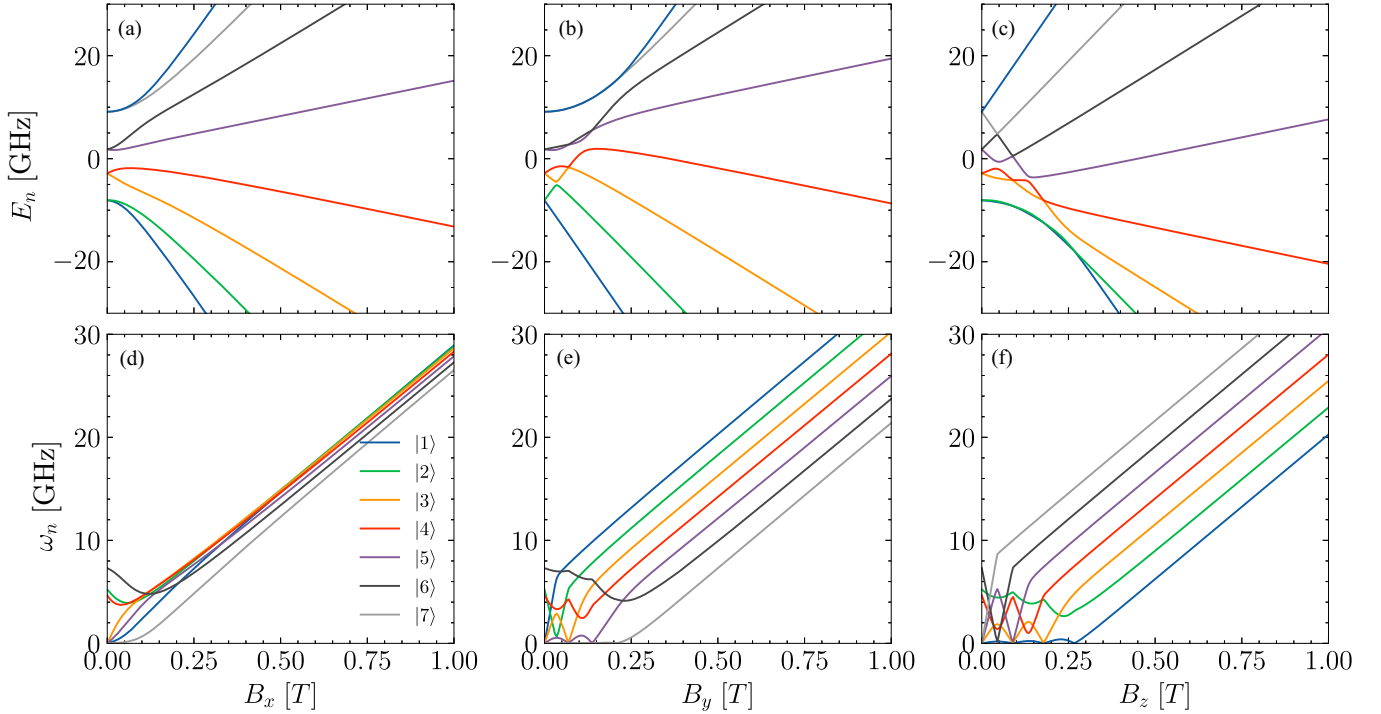


FIG. 1. (a)–(c) Eigenenergies of the associated eigenstates from Eq. (1), at different static magnetic fields and (d)–(f) Associated Bohr frequencies.

performance at short protocol durations where fidelity degradation is most pronounced. The optimization employed a learning rate of  $\eta = 10^{-3}$  and a maximum of 1000 iterations, which we found sufficient to achieve stable convergence across the set of transitions considered. This pre-optimization ensures a fair and meaningful comparison between conventional  $\pi$ -pulse strategies and the Zermelo-based time-optimal protocols analyzed in this work. A complete description of the optimization scheme, including cost-function design and convergence criteria, is provided in Supplemental Material Sec. 1.2 [44].

Depicted in Fig. 1, the different energies and frequencies associated with the eigenstate solutions of Eq. (1) as a function of the static magnetic field (for the three possible orientations) are shown. Since the easy axis of the  $\text{GdW}_{30}$  molecular complex is the  $x$  axis, we observe lower energetic transitions when the magnetic field points in that direction. To perform qubit operations effectively, it is desirable to have a well-separated set of transition frequencies. This minimizes spectral overlap, which can otherwise hinder selective control when using conventional techniques. While magnetization along the  $x$  axis yields a frequency spectrum that is experimentally accessible, it typically exhibits low anharmonicity. In contrast, magnetization along both the medium and hard axes can produce a more anharmonic spectrum, which is advantageous for distinguishing between transitions. To explore alternative control strategies, this study also considers magnetization along the  $z$  axis. By applying a sufficiently strong magnetic field in this direction, it is possible to achieve a more uniform and well-resolved frequency spectrum, which is beneficial for implementing precise quantum control protocols.

An important consideration is the choice of the static magnetic-field magnitude. This value must be sufficient to

fully lift the degeneracy among the system's eigenstates, ensuring well-resolved energy levels for controlled transitions. For magnetization along the  $x$  axis, magnetic fields exceeding 0.1 T are required for a significant energy splitting, while for the  $z$  axis, fields greater than 0.25 T are necessary to achieve the same effect. In this study, following the implementation by Castro *et al.* [35], we adopt a magnetic field of 0.15 T along the  $x$  axis. This configuration will be compared with a stronger field of 0.4 T applied along the  $z$  axis, allowing us to assess the impact of field orientation and strength on the system's spectral properties and control performance. We add these settings to explore a range of values where control can be obtained.

### A. State-to-state transitions

To evaluate the performance of the control schemes considered in this work, we target adjacent transitions between eigenstates of the drift Hamiltonian. The quality of each control protocol is quantified in terms of fidelity, defined as  $|\langle \psi(\tau) | \psi_f \rangle|^2$ . This metric provides a direct measure of how accurately the desired state transfer is achieved.

We begin our analysis with the resonant magnetic-field manipulation through  $\pi$ -pulses, which will serve as a baseline later for evaluating the performance and limitations of the quantum Zermelo approach. A dynamic magnetic field along the  $y$  direction has been considered, within the picture of having the static and dynamic magnetic fields orthogonal to each other. In this case, the  $y$  axis has been chosen, since principal magnetization will be done on the  $x$  or  $z$  axis. Note that the determination of its magnitude will be modified accordingly by Eq. (7). As a first step, and taking into account the fact that only monochromatic pulses can be used, an optimization

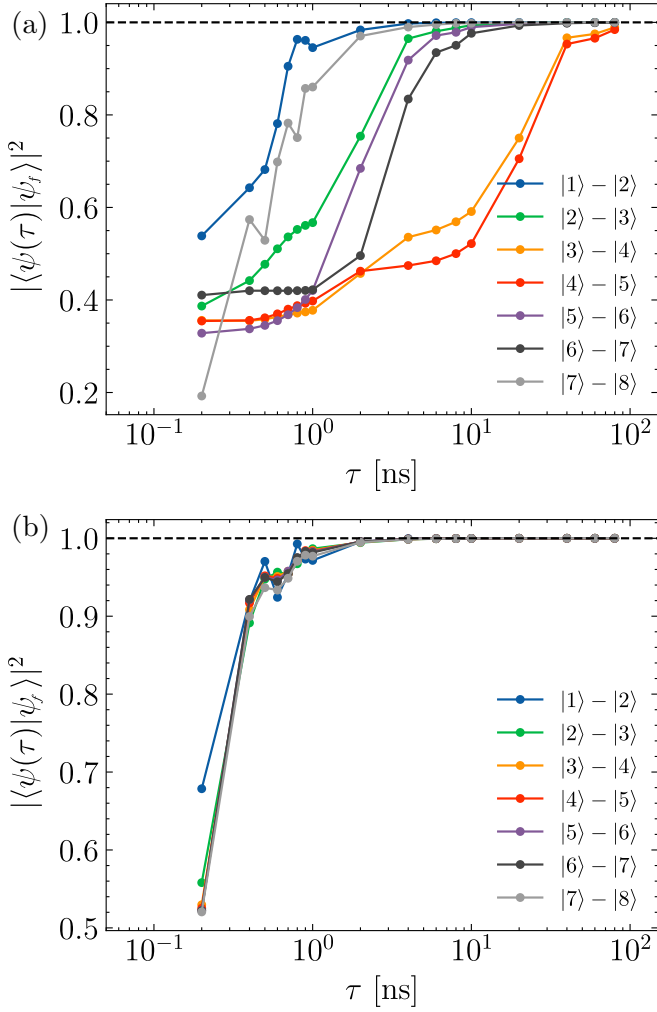


FIG. 2.  $\pi$ -pulse control scheme fidelities for multiple adjacent transitions and multiple protocol times for (a)  $B_x = 0.15$  T and (b)  $B_z = 0, 4$  T. The dashed black line refers to Zermelo's unit fidelity.

of the amplitude has been carried out by means of what is described in the Supplemental Material (see Sec. 1.2) [44].

Depicted in Fig. 2, are the optimized fidelities for the  $\pi$ -pulse control scheme for  $B_x = 0.15$  T and  $B_z = 0.4$  T. It can be seen that the longer the protocol time, the more fidelity we reach. This leads to a critical consideration in quantum control: the trade-off between fidelity and protocol duration. In quantum information processing, state transitions must be executed with near-perfect fidelity, ideally approaching unity. While this requirement may appear stringent, it becomes essential in scenarios involving multiple sequential transitions. This challenge highlights a fundamental limitation of control schemes based on  $\pi$ -pulses. Although conceptually simple and widely used,  $\pi$ -pulse protocols often require longer operation times, especially in multilevel systems where transitions must be selectively addressed. The increased duration of these protocols reduces the number of operations that can be performed within the system's coherence window. For instance, in the case of the GdW<sub>30</sub> molecular complex, the decoherence time ranges from approximately 470 to 600 ns, depending on the specific quantum state involved [33]. As a result, the time

overhead associated with  $\pi$ -pulse control can significantly constrain the computational depth achievable before coherence is lost.

Let us emphasize the critical role played by the frequency separation associated with each transition. A comparison between Figs. 2(a) and 2(b) reveals a noticeably more uniform fidelity profile when the transition frequencies are sufficiently well-separated, minimizing spectral overlap. This separation is essential for achieving selective and high-fidelity control. In particular, when magnetization is applied along the  $z$  axis, the resulting transition behavior between adjacent eigenstates aligns well with the requirements for quantum computing applications, enabling more predictable and reliable state operations.

Note that, for nonadjacent transitions (such as  $|101\rangle = |\psi_6\rangle \rightarrow |111\rangle = |\psi_8\rangle$ ), a chain of  $\pi$ -pulses must be employed (in this example, we would need to perform transition  $|\psi_6\rangle \rightarrow |\psi_7\rangle$  and after  $|\psi_7\rangle \rightarrow |\psi_8\rangle$ ), which greatly increases the qubit operation time [35].

To explore the potential of more advanced control techniques, we next consider the Zermelo time-optimal navigation framework. While the Zermelo approach enables unit-fidelity state transfer, it is equally important to assess the energy requirements—or the cost—associated with the protocol. Understanding this cost is essential for determining whether the implementation of such a control scheme is practically advantageous and worth pursuing in realistic quantum computing scenarios. The cost of implementation is measured as  $C(t) = \|\mathbb{H}_c(t)\|_F^2$ , where  $\|\cdot\|_F$  denotes the Frobenius norm [21]. To physically motivate this measure of cost, we can write any Hamiltonian as a linear combination of basis elements, such that  $\mathbb{H}(t) = \sum_j u_j(t)\mathbb{H}^j$  being  $\{\mathbb{H}^j\}$  a set of linearly independent Hermitian matrices. By computing the squared Frobenius norm over this expression, we get that  $\|\mathbb{H}(t)\|_F^2 = \sum_j u_j^2(t)\|\mathbb{H}^j\|_F^2$ . Since  $\|\mathbb{H}^j\|_F^2$  will correspond to the same number for every basis element, we obtain that the squared Frobenius norm can be understood as the sum of the square control fields. This can be easily visualized with the  $\pi$ -pulse control Hamiltonian, since  $\text{Tr}[\mathbb{H}_{\text{MRF}}(t)^2] = \text{Tr}[A^2 \sin^2(\omega t)\mathbb{H}_{\text{Zeeman}}^2] = A^2 \sin^2(\omega t)\text{Tr}[\mathbb{H}_{\text{Zeeman}}^2]$ . To better compare the obtained results obtained, it is convenient to take the average value of the cost along the protocol, as  $\bar{C} = \frac{1}{\tau} \int_0^\tau C(t)dt$ . In the specific case of the pulse, this is given analytically by  $\bar{C} = A^2 \text{Tr}[\mathbb{H}_{\text{Zeeman}}^2](\frac{1}{2} - \frac{\sin(2\tau\omega)}{4\omega\tau})$ .

Figure 3 illustrates the average energy cost associated with multiple adjacent transitions across varying protocol durations. For both control schemes, the cost exhibits an exponentially decreasing trend, approaching zero as the protocol time increases. This behavior reflects the trade-off between speed and energy efficiency in quantum control.

Another noteworthy aspect is the energy scale associated with each control scheme. In the case of  $\pi$ -pulse control, the energy required to perform transitions at short protocol durations is significantly higher than that of the Zermelo-based approach. Moreover, the Zermelo protocol exhibits a high degree of uniformity in energy consumption across different adjacent transitions (i.e., solid lines corresponding to Zermelo are superimposed), indicating a consistent and balanced control strategy. This homogeneity is not observed in the  $\pi$ -pulse scheme, where energy require-

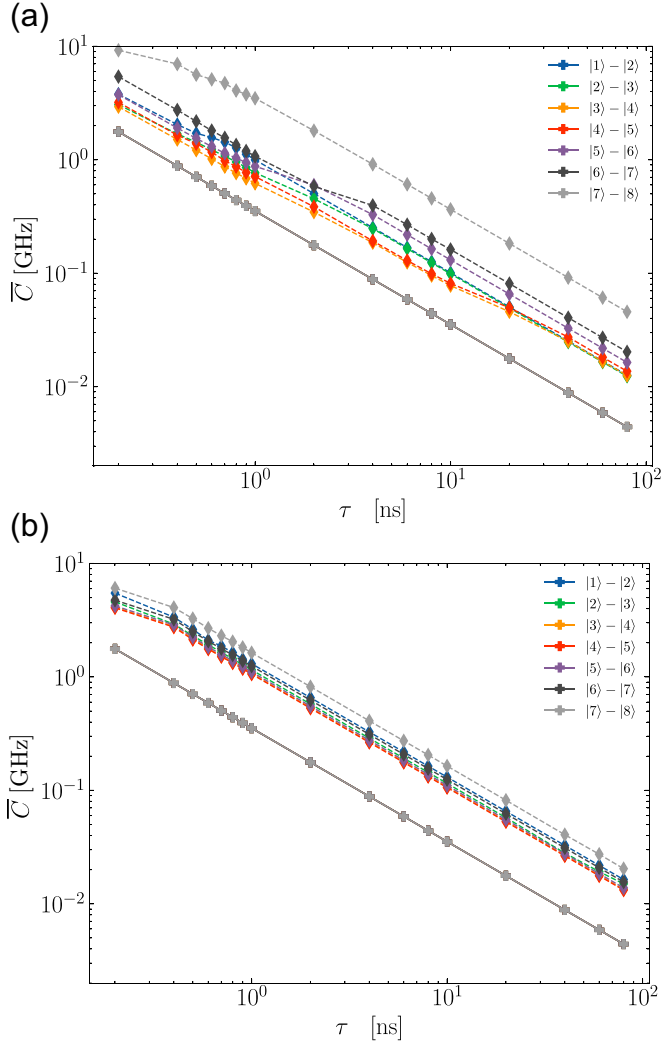


FIG. 3. Average cost for different protocol time  $\tau$  and different adjacent transitions for the two implemented control schemes for (a)  $B_x = 0.15$  T and (b)  $B_z = 0.4$  T. Lines refer to the Zermelo control protocol, dashed lines refer to  $\pi$ -pulse control protocol.

ments vary more noticeably between transitions. Interestingly, when the static magnetic field is applied along the  $z$  axis, the  $\pi$ -pulse control shows improved uniformity in energy cost compared with the  $x$  axis configuration, aligning with trends observed in earlier analyses.

### B. Interpretation of Zermelo's control Hamiltonian

The next step is to interpret the control Hamiltonian derived through the quantum Zermelo time-optimal control framework in physically meaningful terms. It is important to emphasize that, while mathematically well-defined, the resulting operator may possess a structure that lacks a clear physical realization. In such cases, the control Hamiltonian may involve components or interactions that are not naturally appearing and that are not implementable within known physical systems, posing a significant challenge for experimental realization.

Figure 4 presents the initial control Hamiltonian  $\hat{H}_c(0)$ , derived from the Zermelo time-optimal control protocol,

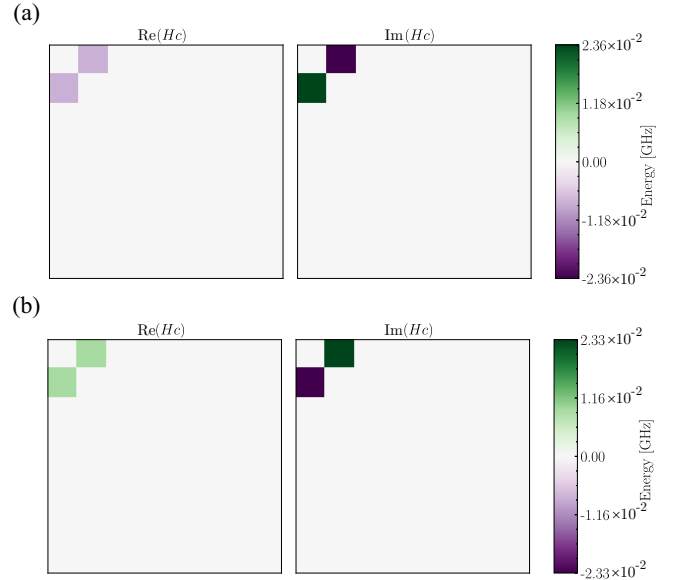


FIG. 4. Zermelo's  $\mathbb{H}_c(0)$  for transition  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $\tau = 10$  ns for the two different static magnetic.

projected onto the eigenbasis of the drift Hamiltonian for the transition  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  with a protocol duration of  $\tau = 10$  ns. Since similar qualitative behavior is observed across all transitions, we display only this representative case for clarity and conciseness. Readers interested in the full set of transitions are referred to Figs. S3 and S5 in the Supplemental Material [44]. Additionally, in Fig. S4 of the Supplemental Material the reader may find the Zermelo control Hamiltonian for transitions between superposition of eigenstates, thanks to the qudit structure of the background Hamiltonian. For comparison, the control Hamiltonian of the optimized  $\pi$ -pulse control scheme is also shown in Fig. 5.

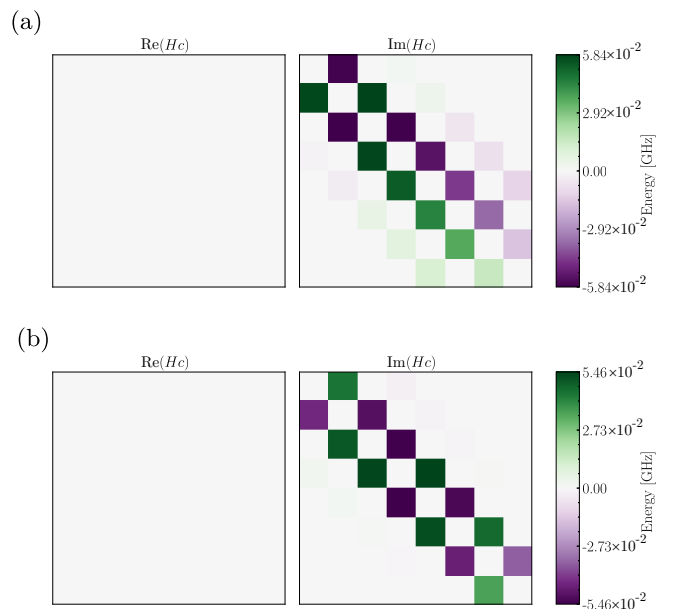


FIG. 5.  $\pi$ -pulse's  $\mathbb{H}_c(0)$  for transition  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $\tau = 10$  ns for the two different static magnetic fields.

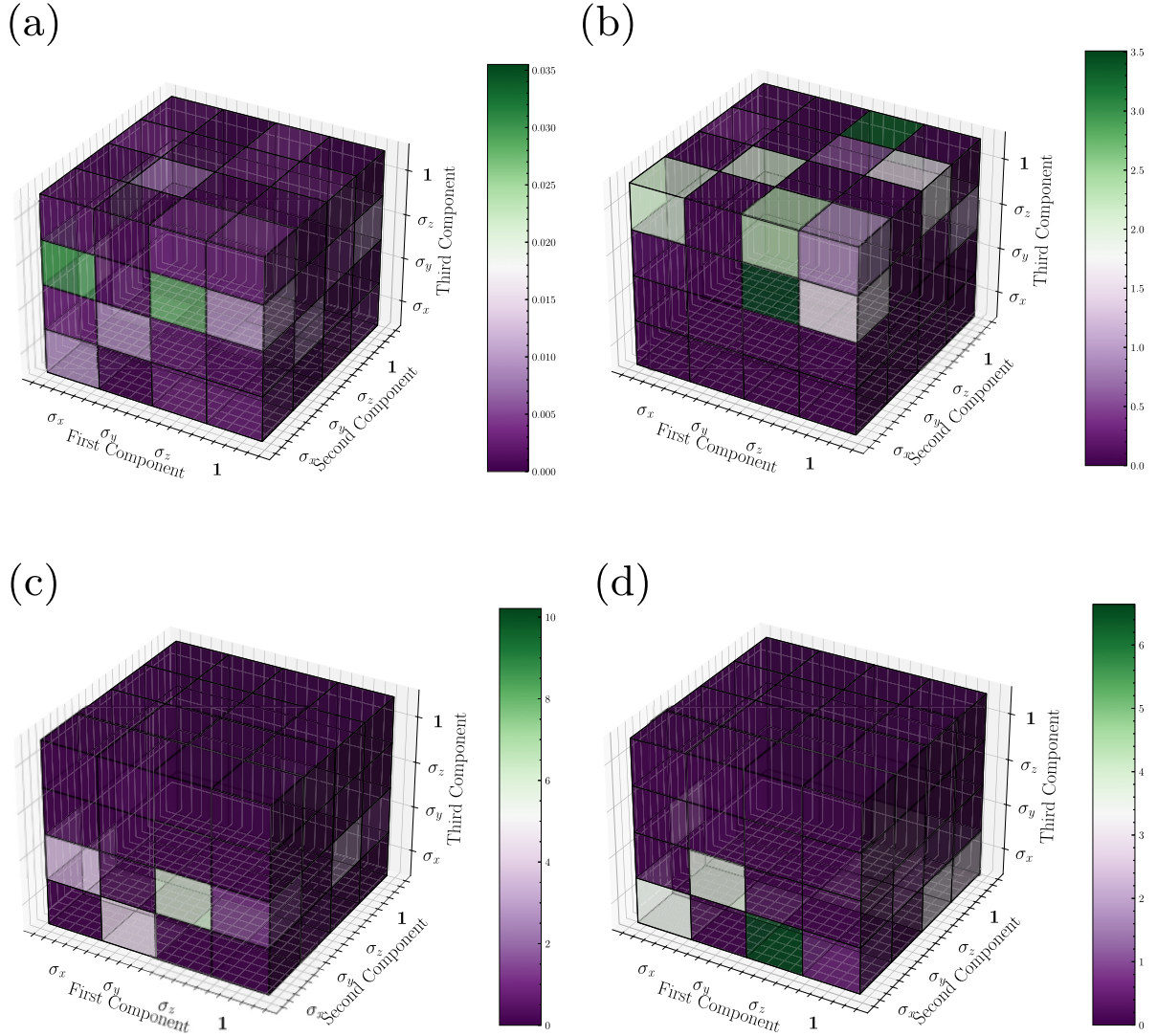


FIG. 6. Orthogonal decomposition of the (a) Zermelo Hamiltonian, (b)  $x$  component, (c)  $y$  component, and (d)  $z$  component angular-momentum operators in the Kronecker product of Pauli basis elements. The Zermelo Hamiltonian is built for the case of transitioning from  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $\tau = 10$  ns and with a background magnetic field  $B_x = 0.15$  T.

First, we do not observe any difference between Figs. 4(a) and 4(b), indicating that the structure of the time-optimal control Hamiltonian derived from the Zermelo approach is largely independent of the magnetization direction. In contrast, the  $\pi$ -pulse control scheme, shown in Figs. 5(a) and 5(b), reveals a noticeable reduction in the coupling of unwanted (parasitic) transitions when shifting the magnetization from the  $x$  axis to the  $z$  axis. The most notable difference resides when comparing the structure of the Zermelo and  $\pi$ -pulse Hamiltonian. The picture is clear, the optimum way to perform an eigenstate-to-eigenstate transition is to couple the departure and arrival states without acting on the other states.

To gain intuition about the physical structure of the Zermelo Hamiltonian, we project it onto the Kronecker product Pauli basis  $\{\sigma_i \otimes \sigma_j \otimes \sigma_k\} \forall i, j, k \in \{x, y, z, 1\}$ , which forms a natural operator basis for an  $N = 8$  Hilbert space. We also include projections onto the spin angular-momentum operators to assess whether any common subset of components reveals signatures of

effective magnetic interactions. Figure 6 presents a compact, rapid-inspection comparison of the projection coefficients:

$$\frac{\langle \hat{H}_k, \sigma_i \otimes \sigma_j \otimes \sigma_k \rangle}{\langle \sigma_i \otimes \sigma_j \otimes \sigma_k, \sigma_i \otimes \sigma_j \otimes \sigma_k \rangle}, \quad \hat{H}_k \in \{\hat{H}_{\text{Zermelo}}, \hat{S}_x, \hat{S}_y, \hat{S}_z\},$$

while a detailed, term-by-term comparison is provided in the Supplemental Material (Fig. S8) [44]. As can be observed, the  $\hat{S}_z$  operator shares several basis components with the Zermelo Hamiltonian, suggesting that the latter incorporates contributions reminiscent of an effective magnetic field along the  $z$  axis, although the signs of the coefficients differ. At the same time, the Zermelo Hamiltonian displays additional basis components absent from the expansions of the magnetic-field operators, indicating the involvement of interaction channels not captured by simple spin-field coupling. Notably, the structural pattern of these projections remains consistent across different molecular magnetizations, suggesting that these features are robust and common to all cases analyzed.

Given the impossibility of writing a Zermelo solution as a linear combination of spin operators, we aim to formulate a rigorous argument demonstrating why the Zermelo control Hamiltonian cannot be expressed as a magnetic-field interaction with the system, thus showing the limits of this physically feasible mechanism for manipulating this class of systems. From the decomposition of the drift Hamiltonian [Eq. (1)], it is evident that none of the constituent terms (2), (3), or (4) mutually commute. Consequently, the eigenstates of the composite Hamiltonian change with each successive addition, implying that the eigenbasis of the full Hamiltonian  $\hat{H}_0$  differs from those of its individual components.

Particular attention should be given to the terms  $\hat{H}_1$  and  $\hat{H}_2$ . The term  $\hat{H}_1$  introduces an easy-axis anisotropy along the  $z$  direction. The addition of  $\hat{H}_2$  perturbs this anisotropy, introducing a planar distortion in the  $x$ - $y$  plane. This perturbation complicates the controllability of the system because it disrupts the symmetry introduced by  $\hat{H}_1$ .

To reinforce this point, we note that the only physically realizable control Hamiltonians for this system are linear combinations of the spin operators  $\hat{S}_x$ ,  $\hat{S}_y$ , and  $\hat{S}_z$ . Since  $\hat{S}_z^2$  and  $\hat{S}_z$  commute, the operator  $\hat{S}_z^2$  can be expressed in terms of  $\hat{S}_z$  via spectral decomposition because they share a common eigenbasis. If the drift Hamiltonian consisted solely of  $\hat{H}_1$ , the eigenstates used to construct the Zermelo control Hamiltonian would align with those of  $\hat{S}_z$ , allowing the control Hamiltonian to be expressed as a linear combination of  $\hat{S}_z$  eigenstates. However, the presence of  $\hat{H}_2$  breaks this commutativity. The resulting Hamiltonian  $\hat{H}_1 + \hat{H}_2$  does not commute with all of  $\hat{S}_x$ ,  $\hat{S}_y$ , and  $\hat{S}_z$ , rendering its eigenstates incompatible with a representation based solely on these operators. As a result, the Zermelo control Hamiltonian cannot be synthesized as a linear combination of the physically implementable interactions, i.e., magnetic-field couplings.

While this limitation may appear to undermine the practical utility of the Zermelo control framework, its true value lies in a different insight. Specifically, the Zermelo solution can serve as a theoretical benchmark for identifying quantum platforms capable of achieving unit-fidelity, time-optimal transitions. More precisely, consider a hypothetical quantum system characterized by a drift Hamiltonian  $\hat{H}_0$  and a control Hamiltonian  $\hat{H}_c$ . If the Zermelo Hamiltonian commutes with the physical control Hamiltonian  $\hat{H}_c$  at all times during the protocol, then the system can, in principle, realize the desired transformation under optimal conditions.

To further identify the differences and similarities between the Zermelo and  $\pi$ -pulse control schemes, one feasible approach is to determine how they act upon the system. To do so, the spin projection (i.e.,  $\langle \hat{S}_x \rangle$ ,  $\langle \hat{S}_y \rangle$ , and  $\langle \hat{S}_z \rangle$ ) has been computed. As an example, take the transition  $|\psi_1\rangle \rightarrow |\psi_2\rangle$ , whose spin components are depicted in Fig. 7 for the Zermelo control scheme, and in Fig. 8 for the  $\pi$ -pulse scheme (the same conclusions can be drawn for other transitions). We observe that the dominant spin component undergoing change during the evolution is  $\langle \hat{S}_x \rangle$ . The remaining components exhibit smaller variations and appear to act as auxiliary channels or “momentum buffers” that support the primary transition dynamics.

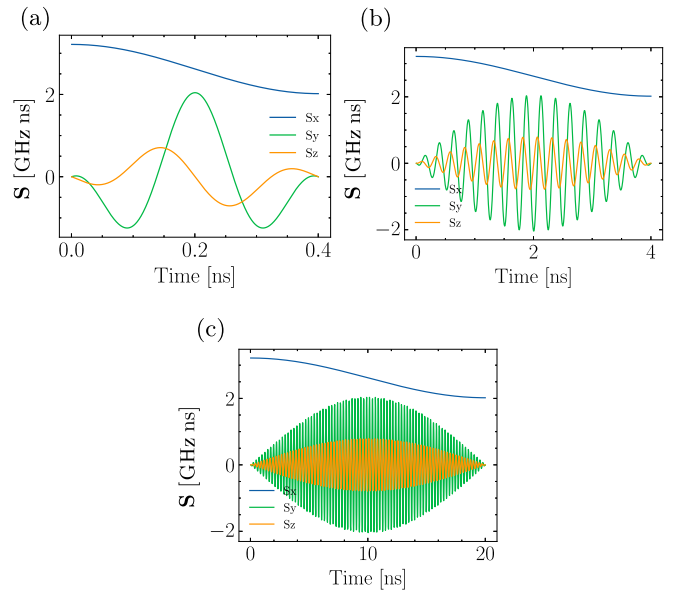


FIG. 7. Spin expected values for transitions  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $B_x = 0.15$  T for the Zermelo control scheme and protocol times (a)  $\tau = 0.4$  ns, (b)  $\tau = 4$  ns, and (c)  $\tau = 20$  ns.

A particularly striking observation is the strong similarity between Fig. 7(c) and Fig. 8(c). This resemblance suggests that both control schemes—Zermelo and  $\pi$ -pulse—manipulate the spin system in a remarkably similar manner when the protocol duration is sufficiently long. This convergence implies an effective equivalence between the two approaches in the long-time limit. This assumption is further supported by examining the population dynamics of the system’s eigenstates over the course of the protocol. As shown in Figs. 9 and 10, the population profiles for both control

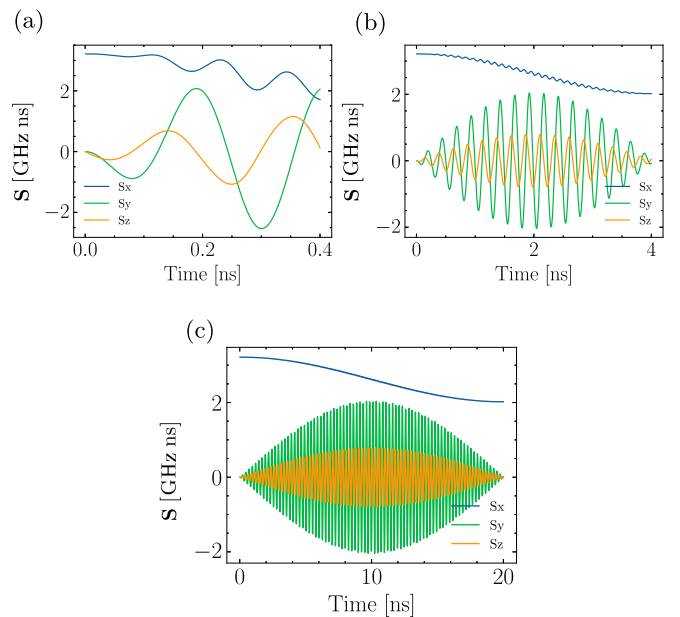


FIG. 8. Spin expected values for transitions  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $B_x = 0.15$  T for the  $\pi$ -pulse control scheme and protocol times (a)  $\tau = 0.4$  ns, (b)  $\tau = 4$  ns, and (c)  $\tau = 20$  ns.

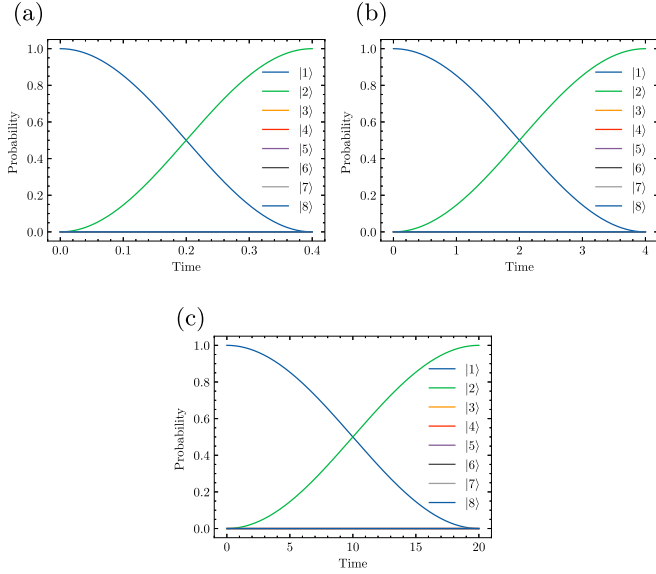


FIG. 9. Spin-state populations for transitions  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $B_x = 0.15$  T for the Zermelo control scheme and protocol times (a)  $\tau = 0.4$  ns, (b)  $\tau = 4$  ns, and (c)  $\tau = 20$  ns.

schemes become increasingly similar as the protocol time increases.

This effective equivalence between the dynamics generated by the Zermelo Hamiltonian and the dynamics generated by the  $\pi$ -pulse scheme can be explained from the theoretical derivation of such a control scheme. The first principal assumptions to derive the  $\pi$ -pulse control is that the control Hamiltonian is much smaller in magnitude compared with the background Hamiltonian, which by virtue of Eq. (7) happens to be true when the protocol time is large enough. The

second principal assumption is that, thanks to the resonant frequency of the pulse with the Bohr frequency of the transition, such control couples only the states present in the desired transition, thus masking other types of transitions present in the Zeeman Hamiltonian. When these two assumptions are met, then the associated matrix of the  $\pi$ -pulse control can be understood as a matrix coupling only the departure and arrival states, which is exactly the structure of the Zermelo Hamiltonian, thus explaining this observed convergence despite both methods producing different trajectories because they are generated with different Hamiltonians. Further details on the validity of the assumptions behind the  $\pi$ -pulse control can be found in the Supplemental Material, Sec. 1.3 [44].

Therefore, the Zermelo solution is a shortcut to apply such a mechanism, although for its particular Hamiltonian shape of  $\mathbb{H}_c(0)$  (Fig. 4), the exact translation into a physical interaction turns out to be impossible to deduce.

#### IV. CONCLUSIONS

A study of how to optimally control spin states of the Gadolinium based  $\text{GdW}_{30}$  molecular complex has been carried out. In this work, we compared two control models in order to perform adjacent transitions between eigenstates of the drift Hamiltonian describing the spin states of the molecule. To describe the particular system of interest, the giant-spin approximation was used. The  $\pi$ -pulse model, widely used in spin control, was evaluated against the Zermelo approach, which constructs a control Hamiltonian capable of driving quantum state transitions with unit fidelity, regardless of initial conditions. Our analysis of the system's dynamics revealed that  $\pi$ -pulse control is fundamentally limited by the resonance condition, which imposes a lower bound on the protocol duration. In the regime of short protocol times—where high-speed performance is critical— $\pi$ -pulse control suffers from reduced fidelity, making it less suitable for precise quantum operations.

In contrast, the Zermelo solution consistently achieves unit-fidelity transitions, even at short timescales. However, the resulting control Hamiltonian is mathematically complex and lacks a clear physical interpretation when it is represented in terms of spin components. Its structure does not correspond to any known expression of physical interaction Hamiltonian, raising questions about its experimental feasibility. Further research is needed to determine whether such Hamiltonian representation can be approximated or implemented using physically realizable interactions and quantum states. This fact was later confirmed by the careful study of the commutation properties of the drift Hamiltonian and the physical representation of the feasible control of this kind of system.

The orientation of the static magnetic field also plays a crucial role in the controllability of spin states. When magnetization is applied along the molecule's easy axis ( $x$  axis), the resulting Bohr frequency spectrum exhibits low anharmonicity, leading to overlapping transitions and more complex control trajectories. In contrast, magnetization along the hard axis  $z$  produces a broader and more anharmonic frequency distribution, which facilitates selective control. However, this configuration may require Bohr frequencies ranges that exceed current experimental capabilities. While the  $\pi$ -pulse

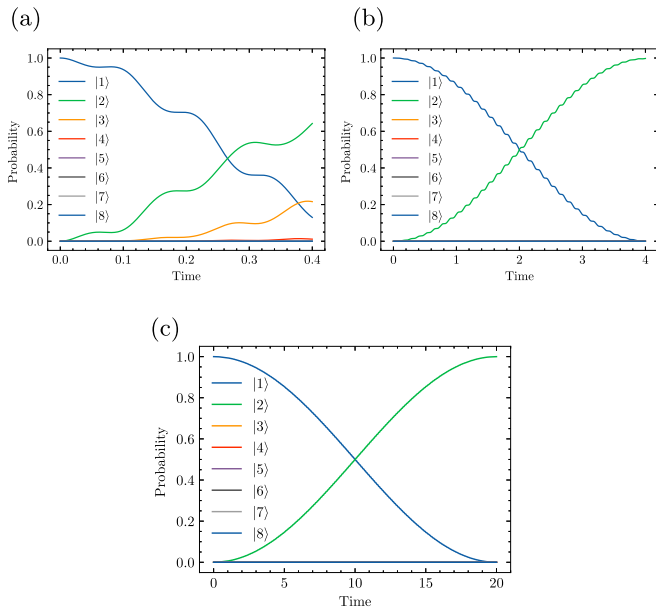


FIG. 10. Spin-state populations for transitions  $|\psi_1\rangle \rightarrow |\psi_2\rangle$  at  $B_x = 0.15$  T for the  $\pi$ -pulse control scheme and protocol times (a)  $\tau = 0.4$  ns, (b)  $\tau = 4$  ns, and (c)  $\tau = 20$  ns.

control scheme was sensible to the orientation of the applied static magnetic field, the Zermelo control scheme showed no difference among tested fields.

In summary, although the Zermelo control scheme provides clear theoretical advantages in terms of fidelity and time-efficiency, its practical realization remains fundamentally challenging. The Zermelo Hamiltonian contains interaction terms that cannot be generated using the physically available controls in this platform, i.e., it cannot be expressed as any linear combination of the three experimentally accessible magnetic-field operators, making its direct synthesis impossible in practice. This limitation would likely become even more pronounced if one were to apply numerical optimal control techniques such as CRAB [48] or GRAPE [49], as these methods would remain constrained by the same restricted control set. While such algorithms can, in principle, enhance fidelities at short protocol durations - sometimes outperforming the standard  $\pi$ -pulse scheme - they can only optimize within the limited space of implementable Hamiltonians and, therefore, cannot reconstruct the full structure required for genuinely time-optimal transitions. As a result, the gap between the theoretically optimal solution and the experimentally achievable dynamics persists even under advanced optimization, reflecting a fundamental incompatibility between the ideal Zermelo framework and the physical control resources available in this system. Future work should focus on bridging the gap between TOQC and experimentally accessible quantum states and interactions, potentially enabling time-optimal quantum operations in molecular spin

systems. Two possible lines of development could go in the direction of (1) identifying quantum platforms where such optimal conditions are naturally satisfied, and (2) engineering or optimizing physical systems to approximate the Zermelo and the natural control Hamiltonians as closely as possible, even under physical constraints. This might include the consideration of exploring control mechanism beyond global magnetic fields, or adding restrictions to the Zermelo Hamiltonian [50]. Given that the resulting Zermelo control Hamiltonian cannot be understood as a given physical interaction, this might be an indication of the limitations for the exact control to induce the desired transitions between spin states, for the qubit architecture considered here.

## ACKNOWLEDGMENTS

The authors would like to thank the financial support from the Spanish Structures of Excellence María de Maeztu Program, through Grant No. CEX2021-001202-M and the Agència de Gestió d'Ajuts Universitaris i de Recerca of Generalitat de Catalunya, Projecte 2021 SGR 00354. M.A. acknowledges financial support from the Agència de Gestió d'Ajuts Universitaris i de Recerca of Generalitat de Catalunya through the Doctorats Industrials 2023 DI 00020 program.

## DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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