

Long-distance coupling between magnetic centers in ultrafast spin transfer: Dynamic Goodenough-Kanamori-Anderson rules

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Using state-of-the-art quantum chemistry, we present a microscopic mechanism for ultrafast laser-induced long-range spin transfer in carbon chains to which two magnetic atoms (Ni) are attached. Here, the Ni atoms function as spin carriers, while the carbons provide the delocalized conjugated p_z -character orbitals to bridge them. First, we establish that the key to the whole process is the virtual excitations between the localized Ni atomic orbitals, and the conjugated orbitals, through which the two Ni atoms can indirectly couple to each other. Second, the efficiency of dynamic interaction depends on the orbital phases of carbon atoms bonded to Ni, with longer-distance transfers benefiting from favorable degeneracies and phase alignments. Third, even when conjugated orbitals interact with only one Ni atom, their coherent superposition facilitates long-distance spin coupling. Fourth, the different structural characters between *cis* and *trans* configurations result in distinct spin-transfer behavior. Fifth, the spin-transfer speed and efficiency are tunable based on the position of magnetic centers. We name this set of interaction mechanisms dynamical Goodenough-Kanamori-Anderson (GKA) rules, because it resembles the well-known static GKA rules, in which the nature of the indirect metal-to-metal coupling (i.e., ferromagnetic versus antiferromagnetic) is dictated by the occupancy of the degenerate p_z orbitals of the bridging oxygen. Unlike in the static case, the spin density here is dynamically transferred between two Ni atoms via well-designed laser pulses.

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I. INTRODUCTION

Significant progress has been made over the past decades in laser-induced magnetization dynamics and spin control [1–11], with growing evidence for their potential applications in memory and quantum logic devices [12–25]. In particular, advances in on-chip laser technology have enabled precise optical control in nanoscale magnetic clusters, further supporting these prospects [26–28]. Therefore, the mechanism underlying long-range, optically induced spin dynamics is of great importance and remains an interesting topic to debate.

The Goodenough-Kanamori-Anderson (GKA) rules describe interatomic spin interactions through bridging ligands,

mediated by superexchange and semicovalent exchange [29–32]. They provide a fundamental framework to understand magnetic coupling in molecular and extended systems [33–44]. Such static exchange interactions are inherently governed by coordination geometry and ligand orbital symmetry. In addition, the strength of spin interactions such as the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange critically depends on the overlap between the localized magnetic centers and the extended conduction channels, as well as the nonlocal spin susceptibility of the conduction electrons near the Fermi level [45,46]. A recent study in optical lattices also reveals the metal-insulator crossover accompanied by antiferromagnetic (AFM) spin correlations in the Hubbard perspective [47]. Beyond static magnetism, these factors are naturally expected to influence time-dependent spin behaviors. Actually, several studies have already indicated that spin-transfer processes follow similar orbital pathways as those governing virtual hopping mechanism in static interactions [48–52]. In addition, extending relevant tunable Hubbard interactions from optical lattices (ultracold gas systems) to conventional condensed-matter systems is of generic cross-disciplinary interest [53–55]. Here, we develop

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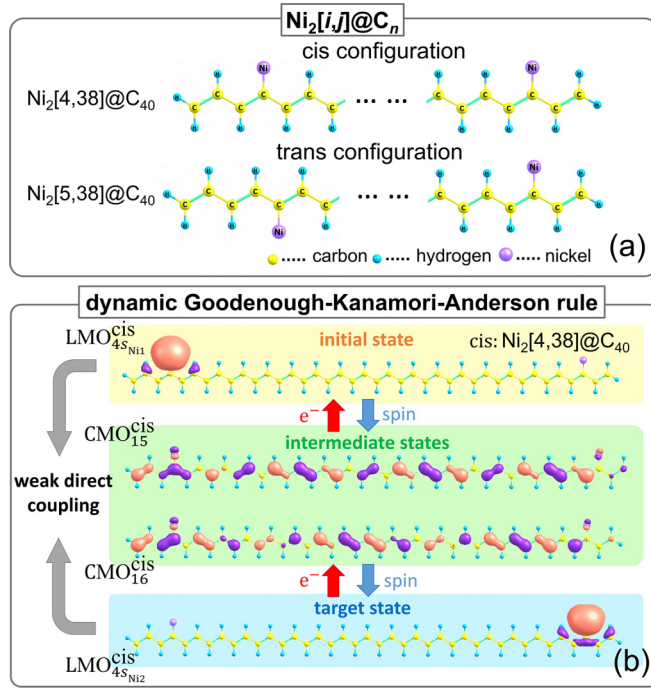


FIG. 1. (a) Geometry of *cis* and *trans* configurations of $\text{Ni}_2[i, j]@C_n$. The yellow, cyan, and purple spheres denote the carbon, hydrogen, and nickel atoms, respectively. (b) Schematic of dynamic Goodenough-Kanamori-Anderson rules. The colors of molecular orbitals denote positive (orange) and negative (purple), respectively. The initial and target states, localized at the Ni atoms, interact weakly with each other, but interact indirectly through the delocalized p_z orbitals, allowing long-distance spin transfer. Blue arrows show spin transfer and red arrows electron hopping.

a theoretical description of dynamic spin coupling in idealized condensed-matter systems and investigate how a similar tunable scheme may be implemented based on *ab initio* calculations.

In this work, we take carbon-chain structures [Fig. 1(a)] as a prototypical platform to explore the underlying mechanisms of long-range, optically induced spin dynamics via Λ processes [56–62]. By analyzing optical coupling among the many-body states, we find a clear pathway of spin transfer in a molecular-orbital perspective. Our systems, $\text{Ni}_2[i, j]@C_n$, have Ni1 and Ni2 attached to carbon atoms indexed by i and j , with n as the total carbon count. Hydrogen atoms passivate the carbons as needed. The system has C_s symmetry and two configuration classes: (i) *cis*, with Ni atoms on the same side, and (ii) *trans*, with Ni atoms on opposite sides.

II. THEORY AND METHODS

The highly correlated many-body ground and excited states are obtained with the Symmetry-Adapted-Cluster-Configuration-Interaction (SAC-CI) method with single and double excitations as implemented in the GAUSSIAN 09 package [63–67]. For each structure 61 spin states are obtained. For the subsequent dynamics spin-orbit coupling (SOC) and an infinitesimal magnetic field are perturbatively taken

into account. A one-electron integral between two states is given by $M^{(\text{SAC-CI})} = \text{Tr}(M^{(\text{MO})}\rho^{(\text{MO})})$, where $M^{(\text{MO})}$ is the one-electron integral matrix and $\rho^{(\text{MO})}$ is the one-electron reduced density matrix (one-matrix), both expressed in the basis of the MOs.

In a finite quasiperiodic carbon chain the molecular orbitals (MOs) resemble Bloch states. In certain energy ranges (loosely named “bands”), the MOs are similar, and can be uniquely labeled by the number of node planes (analogously to the wave vector k of periodic systems). For example, at low energies ($-1.056 \text{ eV} \leq E \leq -0.731 \text{ eV}$), the MOs mainly consist of the $2s$ orbitals of the carbon atoms, which combine into 26 delocalized σ states with the number of node planes increasing from 0 to 25. Around the Fermi level ($-0.432 \text{ eV} \leq E \leq 0.470 \text{ eV}$), the MOs mainly consist of conjugated carbon bonds (i.e., alternating single s - s and double s - s/p_z - p_z bonds). This opens a wide window for the charge-transfer states, thus strongly facilitating the long-distance optical coupling between two Ni atoms. We term the conjugated p_z molecular orbitals as $\text{CMO}_m^{\text{str}}$, where m is the number of node planes and “str” is either *cis* or *trans*. Generally the $4s$ of the nickels are closer to the Fermi level than the $3d$ orbitals (with an energy difference of 0.218 eV), and in the SAC-CI calculations the virtual excitations between the Ni $4s$ and the delocalized conjugated C orbitals play the most important role in the lowest triplet state. The corresponding localized MOs are denoted as $\text{LMO}_{4s\text{Ni}}^{\text{str}}$.

III. RESULTS AND DISCUSSION

A. Conjugated-orbital-mediated spin transfer

An excitation between two many-body states with spin localized on different Ni atoms translates into a long-distance spin-transfer process. In the absence of bridging ligands, direct coupling between the two Ni atoms becomes difficult over long distances. For instance, the direct excitation from $\text{LMO}_{4s\text{Ni}1}^{\text{cis}}$ to $\text{LMO}_{4s\text{Ni}2}^{\text{cis}}$ of $\text{Ni}_2[4, 38]@C_{40}$ is forbidden. Nevertheless, the conjugated bonds of the carbon chain enable a dynamic optical coupling between the two nickels, analogous to the virtual electronic transition between two manganese atoms mediated by the p orbitals of a bridging oxygen atom (GKA rules [29,31,32]). Taking $\text{Ni}_2[4, 38]@C_{40}$ as an example, from Table I, one sees that states $|1\rangle^{\text{cis}}$ and $|14\rangle^{\text{cis}}$ (states are labeled in ascending order of energy) exhibit strong spin localization on Ni1 and Ni2, respectively, due to the singly occupied orbitals $\text{LMO}_{4s\text{Ni}1}^{\text{cis}}$ and $\text{LMO}_{4s\text{Ni}2}^{\text{cis}}$, which is reflected in their dominant Slater excitation determinants, $|\Psi_{4s\text{Ni}1}^{19}\rangle$ and $|\Psi_{4s\text{Ni}2}^{20}\rangle$ ($|\Psi_m^{\pi}\rangle$ denotes a single-electron virtual excitation from MO_m to MO_n , where the overline indicates a spin flip from α to β). An optical excitation from $|1\rangle^{\text{cis}}$ to $|14\rangle^{\text{cis}}$, corresponding to a spin transfer from Ni1 to Ni2, is achieved by involving $|34\rangle^{\text{cis}}$ as an intermediate state. The time evolution of the population of the three states is governed by

$$\frac{\partial c_n(t)}{\partial t} = -i \sum_k \langle n | \hat{H} + \hat{\mathbf{D}} \cdot \mathbf{E}_{\text{laser}}(t) | k \rangle c_k(t) e^{i\omega_{nk}t}, \quad (1)$$

where c_n is the complex scalar coefficient of the n th state, ω_{nk} is the energy difference between $|n\rangle$ and $|k\rangle$, $\hat{\mathbf{D}}$ is the electric-

TABLE I. Many-body states of the *cis* Ni₂[4, 38]@C₄₀ and the *trans* Ni₂[5, 38]@C₄₀. The energy (ΔE) and spin density distribution of the states involved in the spin-transfer scenarios calculated by SAC-CI are listed. SOC and external magnetic field ($B = 2.351$ T, along the z axis) are included. The amplitudes are linear combination coefficients of the corresponding excitation Slater determinants. For the notation of, e.g., $|\Psi_{4sNi1}^{19}\rangle$, see text.

Configurations	States	ΔE (eV)	Spin density			Dominant determinants
			Ni1	Ni2	C Chain	
Ni ₂ [4, 38]@C ₄₀ (<i>cis</i> configuration)	Initial state: $ 1\rangle^{cis}$	0.000	0.483	0.000	0.507	$0.792 \Psi_{4sNi1}^{19}\rangle$
	Target state: $ 14\rangle^{cis}$	1.085	0.001	0.523	0.375	$0.613 \Psi_{4sNi2}^{20}\rangle$
	Intermediate state: $ 5\rangle^{cis}$	0.430	0.057	0.003	0.938	$-0.553 \Psi_{17}^{19}\rangle + 0.502 \Psi_{18}^{19}\rangle$
	Intermediate state: $ 34\rangle^{cis}$	1.824	0.014	0.016	0.974	$0.410 \Psi_{15}^{19}\rangle - 0.386 \Psi_{16}^{20}\rangle$
Ni ₂ [5, 38]@C ₄₀ (<i>trans</i> configuration)	Initial state: $ 5\rangle^{trans}$	0.692	0.595	0.001	0.370	$-0.582 \Psi_{4sNi1}^{20}\rangle + 0.452 \Psi_{4sNi1}^{19}\rangle$
	Target state: $ 8\rangle^{trans}$	0.784	0.001	0.573	0.377	$0.535 \Psi_{4sNi2}^{20}\rangle + 0.403 \Psi_{4sNi2}^{19}\rangle$
	Intermediate state: $ 17\rangle^{trans}$	1.259	-0.016	-0.012	0.973	$0.628 \Psi_{17}^{19}\rangle + 0.495 \Psi_{18}^{20}\rangle$
	Intermediate state: $ 35\rangle^{trans}$	2.313	0.107	0.025	0.871	$0.324 \Psi_{16}^{19}\rangle + 0.259 \Psi_{15}^{20}\rangle$

dipole-transition operator, and $\mathbf{E}_{\text{laser}}(t)$ is the electric field of the laser.

As shown in Figs. 2(a) and 2(b), with $|1\rangle^{cis}$ fully populated initially, the maximum population of the target state $|14\rangle^{cis}$ ($\text{Pop}_{\text{max}}^{14}$) reaches 99% under continuous laser with a frequency around 0.542 eV. The dominant Slater determinants of $|34\rangle^{cis}$ are $|\Psi_{15}^{19}\rangle$ and $|\Psi_{16}^{20}\rangle$. According to the Slater-Condon rules [68], the two Kramers' degenerate orbitals CMO_{15}^{cis} and CMO_{16}^{cis} [Fig. 1(b)], electronically coupled with LMO_{4sNi2}^{cis} and LMO_{4sNi1}^{cis} , give rise to nonzero transition dipole moments between $|34\rangle^{cis}$ and both $|1\rangle^{cis}$ and $|14\rangle^{cis}$ (Table II). In the molecular orbital picture, such three-level excitation $|1\rangle^{cis} \rightarrow |34\rangle^{cis} \rightarrow |14\rangle^{cis}$ proceeds via the two-step electron transition: $\text{LMO}_{4sNi1}^{cis} \rightarrow \text{CMO}_{15}^{cis}$ and $\text{CMO}_{16}^{cis} \rightarrow \text{LMO}_{4sNi2}^{cis}$. Similar processes are also found in different configurations, such as Ni₂[5, 38]@C₄₀, where a spin-transfer process $|5\rangle^{trans} \rightarrow |17\rangle^{trans} \rightarrow |8\rangle^{trans}$ is enabled by the conjugated orbitals

CMO_{17}^{trans} and CMO_{18}^{trans} (Table II). These conjugated MOs usually lie 0.3 eV below the HOMO and have 15–18 node planes (for the other configurations, see the supplemental Material [69]). Thus, we arrive at our first conclusion (1): the dynamical coupling (resulting in spin transfer) between two distant magnetic centers can be achieved through two Kramers' degenerate, conjugated molecular orbitals. Each orbital opens a transfer channel for one of the two spin species.

B. Phase-alignment-driven variable distance spin transfer

Regarding the influence of Ni-Ni distance, we find that, counterintuitively, a shorter distance does not necessarily mean a faster process: The transfers on Ni₂[10, 38]@C₄₀ and Ni₂[11, 38]@C₄₀, which have the shortest Ni-Ni distance, require the longest time [Fig. 2(c)]. We attribute this to phase modulations in the conjugated MOs. Due to their specific shape, distribution gaps are observed on specific sites, such

TABLE II. The dipole moment among the states involved in the spin-transfer processes of Ni₂[4, 38]@C₄₀ and Ni₂[5, 38]@C₄₀. Dominant excitation denotes the most dominant molecular orbital excitation, which characterizes the real excitation between the corresponding many-body states. Note that it is different from the dominant determinant in Table I, which denotes the virtual excitation.

Structures	Excitation	Dipole moment (D)			Dominant excitation
		D_x	D_y	D_z	
Ni ₂ [4, 38]@C ₄₀ (<i>cis</i> configuration)	$ 1\rangle^{cis} \rightarrow 34\rangle^{cis}$	0.000	0.000	0.221	$\text{CMO}_{15}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$
	$ 34\rangle^{cis} \rightarrow 14\rangle^{cis}$	0.000	0.000	0.153	$\text{LMO}_{4sNi2}^{cis} \rightarrow \text{CMO}_{16}^{cis}$
	$ 1\rangle^{cis} \rightarrow 5\rangle^{cis}$	0.000	0.000	0.694	$\text{CMO}_{17}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$ and $\text{CMO}_{18}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$
	$ 5\rangle^{cis} \rightarrow 14\rangle^{cis}$	0.000	0.000	0.000	weak interaction
	$ 5\rangle^{cis} \rightarrow 34\rangle^{cis}$	0.607	1.708	0.000	$\text{CMO}_{15}^{cis} \rightarrow \text{CMO}_{17}^{cis}$ and $\text{CMO}_{15}^{cis} \rightarrow \text{CMO}_{18}^{cis}$
Ni ₂ [5, 38]@C ₄₀ (<i>trans</i> configuration)	$ 5\rangle^{trans} \rightarrow 17\rangle^{trans}$	0.000	0.000	0.112	$\text{CMO}_{17}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$ and $\text{CMO}_{18}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$
	$ 17\rangle^{trans} \rightarrow 8\rangle^{trans}$	0.000	0.000	0.126	$\text{LMO}_{4sNi2}^{cis} \rightarrow \text{CMO}_{17}^{cis}$ and $\text{LMO}_{4sNi2}^{cis} \rightarrow \text{CMO}_{18}^{cis}$
	$ 5\rangle^{trans} \rightarrow 35\rangle^{trans}$	0.000	0.000	0.293	$\text{CMO}_{15}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$ and $\text{CMO}_{16}^{cis} \rightarrow \text{LMO}_{4sNi1}^{cis}$
	$ 35\rangle^{trans} \rightarrow 8\rangle^{trans}$	0.000	0.000	0.167	$\text{LMO}_{4sNi2}^{cis} \rightarrow \text{CMO}_{15}^{cis}$ and $\text{LMO}_{4sNi2}^{cis} \rightarrow \text{CMO}_{16}^{cis}$

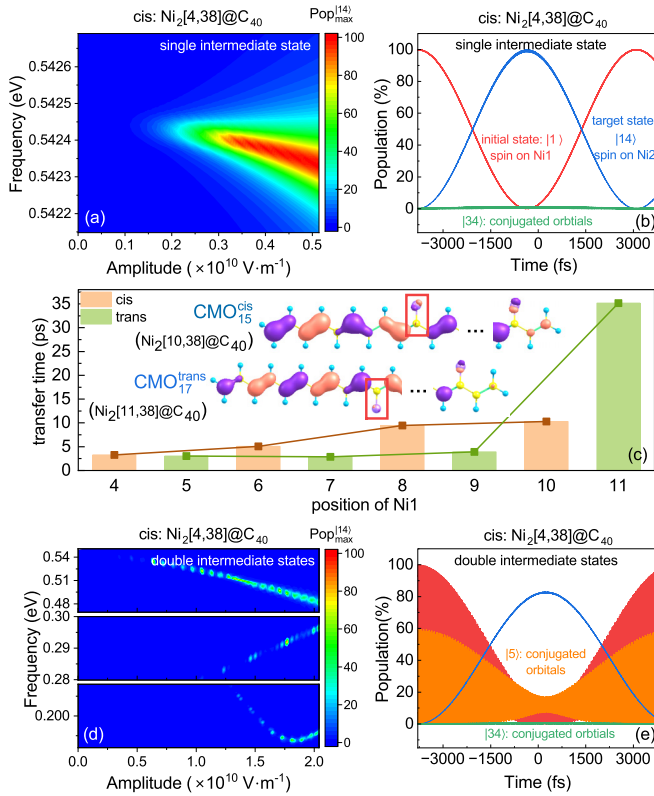


FIG. 2. (a) and (b) single-intermediate-state spin transfer $|1\rangle^{cis} \rightarrow |34\rangle^{cis} \rightarrow |14\rangle^{cis}$ on $\text{Ni}_2[4,38]@C_{40}$. (a) contour map of the largest population of the target state $|14\rangle$ through intermediate state $|34\rangle^{cis}$. (b) time evolution of the many-body states during the spin-transfer processes. (c) times required for spin-transfer processes on different *cis* and *trans* configurations. Note the shallow minimum at the Ni1 position 7 in the *trans* configuration. The two dominant conjugated molecular orbitals CMO_{15}^{cis} of $\text{Ni}_2[10,38]@C_{40}$ and CMO_{17}^{trans} of $\text{Ni}_2[11,38]@C_{40}$ are shown as well. Distribution gaps on the 10th and 11th carbon atoms are highlighted in the red frame. (d) and (e) double-intermediate-state scenario $|1\rangle^{cis} \rightarrow |5\rangle^{cis} + |34\rangle^{cis} \rightarrow |14\rangle^{cis}$ on $\text{Ni}_2[4,38]@C_{40}$. (d) contour map of the largest population of the target state $|14\rangle$ through intermediate states $|5\rangle^{cis}$ and $|34\rangle^{cis}$. (e) time evolution of the many-body states.

as the 10th carbon atom in CMO_{15}^{cis} and the 11th carbon atom in CMO_{17}^{trans} [Fig. 2(c)]. These gaps significantly weaken the overlap between Ni1 and the carbon chain, suppressing the spin transfer. This is also observed in shorter chains ($n = 32, 34, 36, 38$), where the distance between the two Ni atoms is the same as in $\text{Ni}_2[10,38]@C_{40}$ ($j - i = 28$), but the total number of carbon atoms varies and therefore some MOs also have distribution gaps on the carbon atoms attached to Ni1 (Fig. S6 [69]). This is our second conclusion (2): the efficiency of spin transfer is governed by the different phases of the conjugated orbitals on the carbon atoms connecting the Ni atom, rather than simply by the distance between two Ni atoms.

C. Coherent superposition spin transfer

There are certain states which exhibit strong interactions with either initial or target state, such as $|5\rangle^{cis}$ in $\text{Ni}_2[4,38]@C_{40}$. Its electronic configuration is characterized

by the two dominant Slater determinants $|\Psi_{17}^{19}\rangle$ and $|\Psi_{18}^{19}\rangle$. Therefore, it couples strongly with $|1\rangle^{cis}$ via transitions between $\text{LMO}_{45\text{Ni1}}^{cis}$ and both CMO_{17}^{cis} and CMO_{18}^{cis} , but only weakly with $|14\rangle^{cis}$ (Table II). Due to this “bias coupling,” spin transfer cannot proceed efficiently via $|5\rangle^{cis}$ alone. Nevertheless, when both $|5\rangle^{cis}$ and the aforementioned $|34\rangle^{cis}$ are involved, efficient spin transfer can be achieved. At a laser frequency of approximately 0.531 eV, $\text{Pop}_{\text{max}}^{14}$ reaches 82.9% [Figs. 2(d) and 2(e)]. In this scenario, the coherent superposition between $|5\rangle^{cis}$ and $|34\rangle^{cis}$ governs the spin transfer, while removing either of the two intermediate states completely blocks the transfer. Different from the single-intermediate-state scenario, the superposition state also increases the probability of successful spin transfer. As shown in Fig. 2(d), two additional frequencies facilitate this process. Thus, our third conclusion (3) is that optical spin coupling between two magnetic centers can arise from the coherent superposition of states with conjugated orbitals.

D. General case and symmetry-driven spin transfer

Following these rules, spin transfer can be generalized to all 61 SAC-CI states. A dedicated program using a genetic algorithm and linear iteration optimizes the laser parameters (details in the supplemental Material [69]). Instead of the continuous laser, a sech^2 pulsed laser with elliptical polarization is used to maximize the final population of the target state, termed “fidelity,” which serves as a measure of spin-transfer efficiency.

For the *cis* configurations, the fidelity on each configuration is 98.7% ($\text{Ni}_2[4,38]@C_{40}$), 96.2% ($\text{Ni}_2[6,38]@C_{40}$), 73.5% ($\text{Ni}_2[8,38]@C_{40}$), and 80.1% ($\text{Ni}_2[10,38]@C_{40}$), respectively (see the Supplemental Material [69] for details). All the four dynamic scenarios are mediated by multiple intermediate states. In order to elucidate the spin-transfer pathway, we need to evaluate the contributions of each intermediate state. We employ the kernel-based Hilbert-Schmidt Independence Criterion (HSIC) to assess the dependence of the fidelity on these states [70,71]. Taking $\text{Ni}_2[4,38]@C_{40}$ as an example [Figs. 3(a)–3(d)], the HSIC analysis identifies eight dominant intermediate states. The significance of each is evidenced by the drop in fidelity ($\approx 50\%$) when it is excluded. Among them, $|5\rangle^{cis}$ and $|50\rangle^{cis}$ have the strongest effect on the fidelity, with $|23\rangle^{cis}$ playing a secondary role. This behavior clearly reflects the influence of rule (3). From the perspective of molecular orbitals, the dominant contribution still arises from the conjugated orbitals, such as CMO_{15}^{cis} and CMO_{16}^{cis} , which is consistent with rule (1). These key MOs exhibit distribution gaps at the Ni1 site in the $\text{Ni}_2[8,38]@C_{40}$ and $\text{Ni}_2[10,38]@C_{40}$ configurations. As a result, the spin transfer has much lower fidelity than in the $\text{Ni}_2[4,38]@C_{40}$ and $\text{Ni}_2[6,38]@C_{40}$ [Fig. 4(a)], highlighting the role of rule (2).

In the four *trans* configurations, the fidelity of each spin transfer reaches 97.4% ($\text{Ni}_2[5,38]@C_{40}$), 95.1% ($\text{Ni}_2[7,38]@C_{40}$), 96.6% ($\text{Ni}_2[9,38]@C_{40}$), and 96.8% ($\text{Ni}_2[11,38]@C_{40}$), respectively. Different from the *cis* configurations, these dynamic scenarios are all single-intermediate-state processes. Actually, the *trans* configurations partially retain the C_s symmetry of a pristine carbon chain, leading to quasidegeneracy for the two spin-

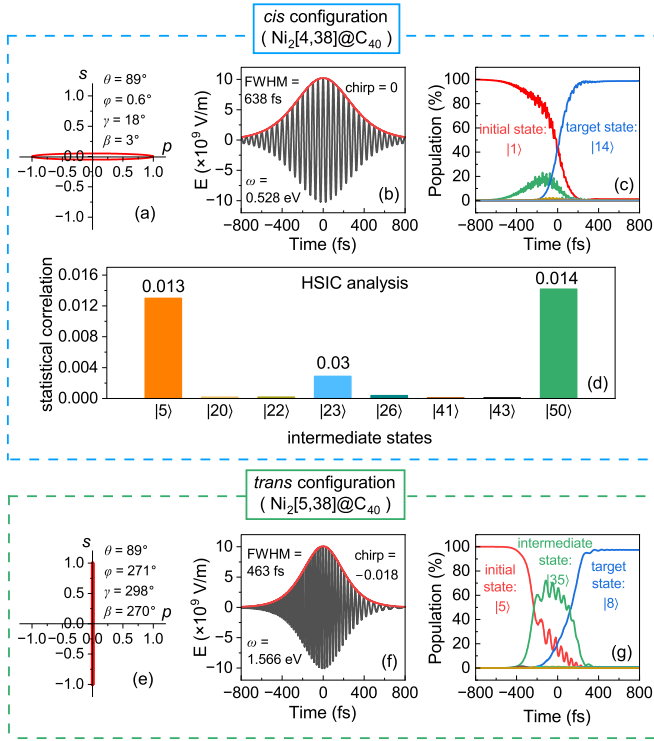


FIG. 3. spin-transfer processes on the *cis* configuration $\text{Ni}_2[4, 38]@C_{40}$ [(a)–(d)] and *trans* configuration $\text{Ni}_2[5, 38]@C_{40}$ [(e)–(g)]. (a) and (e) polarization ellipses of optimized laser pulses applied for spin-transfer processes. (b) and (f) laser envelopes and parameters. (c) and (g) spin-transfer process on $\text{Ni}_2[4, 38]@C_{40}$ ($|1\rangle^{cis} \rightarrow |14\rangle^{cis}$) and $\text{Ni}_2[5, 38]@C_{40}$ ($|5\rangle^{trans} \rightarrow |8\rangle^{trans}$), respectively. (d) HSIC analysis of the spin-transfer process on $\text{Ni}_2[4, 38]@C_{40}$.

localized states [red and blue lines in Fig. 4(b)]. By applying a chirped laser pulse, the intermediate state couples exclusively with both the initial and target states, despite the energy gap between them. For instance, state $|35\rangle^{trans}$ of $\text{Ni}_2[5, 38]@C_{40}$ [Figs. 3(e)–3(g)] provides a spin-transfer pathway through two Kramers’ degenerate orbitals CMO_{15}^{trans} and CMO_{16}^{trans} [Table II]. Due to the Jahn-Teller effect, a energy splitting

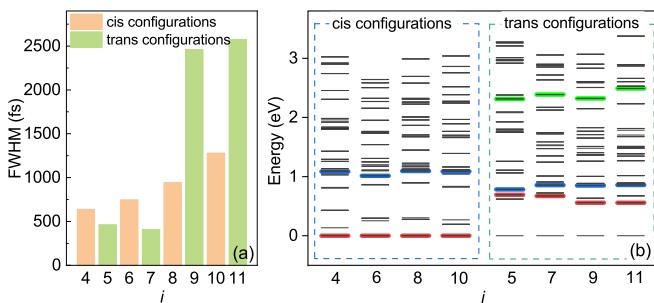


FIG. 4. (a) Full width at half maximum (FWHM) of the laser pulses applied on the *cis* and *trans* configurations. (b) energy levels of the many-body states obtained from SAC-CI calculations. The red and blue lines denote the initial and target states, respectively. The green lines denote the dominant states on *trans* configurations. i denotes the position of Ni_1 .

between the two spin-localized states occurs as the Ni-Ni distance decreases, slowing down the spin transfer on the corresponding configurations [Fig. 4(b)]. This is our fourth conclusion (4): By preserving the symmetry of systems, the approximate degeneracy of initial and target states facilitates the single-intermediate transfer process.

E. Tunability of spin transfer

In previous works we did not address the tunability of the spin-transfer speed and efficiency. As shown in Fig. 4(a), the transfer speed can be optimized by properly positioning the Ni. In the *cis* configurations, the speed increases almost linearly as the two Ni atoms get closer, while in the *trans* configurations, the fastest transfer occurs in $\text{Ni}_2[7, 38]@C_{40}$ ($i = 7$). Notably, different Ni-Ni distances necessitate different laser pulses (see also Supplemental Material [69]). This can be exploited to individually trigger specific spin-transfers (in lieu of magnetic gradients [48,49]). This is our fifth conclusion (5): Spin transfers are tunable on the conjugated carbon-chain systems through structural properties.

IV. SUMMARY

In summary, we systematically investigated the dynamic spin coupling between two Ni atoms mediated by quasidegenerate highly delocalized MOs of long carbon chains. For symmetry reasons, the $4s$ orbitals of the Ni atoms do not readily combine with the conjugated p_z orbitals of carbon, leading to strong spin localization on the Ni atoms. We identify five “dynamic Goodenough-Kanamori-Anderson rules” governing the interaction between the Ni atoms through a laser pulse. Rule (1): A well-optimized laser pulse, the two Ni atoms, despite being separated by a significant distance, can still interact ferromagnetically through the conjugated carbon orbitals. Rule (2): The phases of conjugated orbitals associated with the carbon atoms bonded to the Ni atoms influence the efficiencies of spin-transfer processes. Transfer across longer distances may be faster due to the favorable combination of degeneracies and phases. Rule (3): The dynamic spin coupling can be maintained by the coherent superposition of intermediate states, which interact with either of the two Ni atoms. Rule (4): the *cis* and *trans* configurations differ in their C_s symmetry, leading to distinct effects of laser pulse detuning and resulting in different spin-transfer scenarios. Rule (5): The speed and efficiency are affected by the position of the Ni atoms as a consequence of intrinsically tuning the electron-electron correlations. These findings offer a clear view of the dynamic interactions of magnetic centers through the long ligand chains, thus providing the design route for ultrafast magneto-optical devices.

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the calculations were performed on Tianhe new generation supercomputer.

DATA AVAILABILITY

The data that support the findings of this article are openly available [72].

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