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Phys. Rev. Lett. **126**, 037402 — Published 21 January 2021

DOI: [10.1103/PhysRevLett.126.037402](https://doi.org/10.1103/PhysRevLett.126.037402)

Long-distance ultrafast spin transfer over zigzag carbon chain structure

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(Dated: December 21, 2020)

Using high-level *ab initio* quantum theory we suggest an optically induced subpicosecond spin-transfer scenario over 4.428 nm, a distance which is directly comparable to the actual CMOS scale. The spin-density transfer takes place between two Ni atoms and over a 40-atom-long zigzag carbon chain. The suitable combination of the local symmetries of the participating carbon atoms and the global symmetry of the whole molecule gives rise to what we term the dynamical Goodenough-Kanamori rules, allowing the long-range coupling of the two Ni atoms. We also present local spin-flip scenarios, and compare spin-flip and spin-transfer with respect to their sensitivity against an external static magnetic gradient. Finally, we use two identical laser pulses, rather than a single one, which allows us to accurately control local (intrasite) vs. global (intersite) processes, and we thus solve the problem of embedding individually addressable molecular nanologic elements in an integrated nanospintronic circuit. Our results underline the great potential of carbon chain systems as building and supporting blocks for designing future all-optical magnetic processing units.

Nowadays, higher integration density and faster processing speeds become the major aim in the development of logical processing units [1–15]. However, traditional logic devices such as integrated circuits based on Complementary Metal Oxide Semiconductor (CMOS) technology are reaching their physical limits, so that new alternative technologies gradually emerge [16–19]. Especially, the idea of building logical-functionality networks with spintronics or magnetic heterostructures attracts more and more attention [20–26].

Integrating logic elements in a circuit needs a physical coupling between the participating magnetic molecules or clusters [27, 28]. Although several magnetic molecules have been proposed recently [29–35], an investigation of how to combine them into one functional circuit is still missing. Previous studies demonstrated the spin-transfer between nearby magnetic atoms [36–38], which can be regarded as a communication channel among the logical elements. Nevertheless, due to computational limitations, studying spin transfer from first principles on the mesoscopic scale remains a challenge. At the same time, with the substantial progress of the chip technology, the CMOS scale reaches 7 nm [39]. This provides a unique opportunity for quantum chemistry and device technology to shake hands with each other: the properties of elements of such sizes can be accurately predicted with state-of-the-art quantum chemistry. Therefore, we extend our studies to long-distance spin-transfer processes to propose a potential integration procedure.

Regarding real applications, recently there is substantial progress in on-chip-laser technology, already

allowing photonic integration. For example, a design methodology and experimental characterization of compact out-of-plane focusing grating couplers for integration with magnetoresistive random access memory technology was recently published [40], while Kim *et al.* introduced a single-chip optical phased array realized through wafer-scale 3D integration of silicon photonics and CMOS [41]. This development renders all-optical spin manipulation even more appealing.

Motivated by the spin dynamics and the calculability of endohedral metallofullerenes [33, 42, 43] and the striking magnetic properties of graphene [44], here we “unfold” the fullerenes to mimic the quasi-linear zig-zag graphene boundary and optically transfer the spin over distances comparable to the current CMOS scale. In addition, the sp^2 hybridization and the bond conjugation of such structure can also facilitate the long-distance spin transfer [45, 46]. Two attached Ni atoms accommodate the active spins. We denote our systems as $\text{Ni}_2[i, j]@C_n$, where n is the total number of carbon atoms, i and j indicate the site carbon numbers to which the nickels are attached, respectively. The number of carbon atoms between the two nickels is $k = j - i + 1$. As an example we present a 40-atom-long zigzag carbon chain [Fig. 1(a)]. Such zigzag configurations belong to the C_{2h} or C_s symmetry point groups, depending on whether there is an even or an odd number of carbon atoms. The Ni atoms (detoted as Ni1 and Ni2 respectively) are asymmetrically attached on carbons near the opposite ends of the chain, leading to spin localization. In $\text{Ni}_2[2,38]@C_{40}$ the Ni1-Ni2 distance is 4.428 nm.

The electronic states are obtained with the

equation-of-motion coupled cluster with single and double virtual excitations (EOM-CCSD) [47, 48] and the 6-31G basis set. We also compare with the symmetry-adapted cluster configuration interaction (SAC-CI) method [49–51], but due to computational limitations with smaller basis sets, i.e., STO-3G for C and the Los Alamos basis set plus double ζ (LanL2DZ) with relativistic effective core potentials (ECP) for the Ni [52]. These methods have already been successfully used in our previous investigations and agree nicely with many different experimental results [34, 37, 53–62]. After the perturbative inclusion of spin-orbit coupling (SOC) and a static magnetic field (with a magnitude of 0.235 T), we find clearly distinguishable many-body states with different spin directions or spin-density localization, which can be individually addressed with a laser pulse. Both EOM-CCSD and SAC-CI yield a triplet ground state with spin density highly localized on one Ni atom (Ni2 with spin density 1.003 for EOM-CCSD and Ni1 with spin density 0.957 for SAC-CI). The half-filled sp^2 orbitals of the carbon atoms allow them to share the rest of the spin density, similarly to the edge magnetism of graphene.

Next we optically transfer the spin between the two Ni atoms through the carbons with a Λ process [see Fig. 1(b)] [53, 55, 63, 64]. Details of the Λ process are given in the supplementary material [65]. To this end, two triplet states with spin density localized on different Ni atoms are selected as initial and target states. The carbons provide the intermediate charge-transfer states. The propagation of the many-body wave function is performed within the interaction picture:

$$\frac{\partial c_n(t)}{\partial t} = -\frac{i}{\hbar} \sum_k \langle \Phi_n | \hat{H}(t) | \Phi_k \rangle c_k(t) e^{-i(E_k - E_n)t/\hbar}, \quad (1)$$

where $|\Phi_n\rangle$ and $|\Phi_k\rangle$ are the eigenstates of the static Hamiltonian (including SOC and the magnetic field), c_n is the complex scalar coefficient of the state $|\Phi_n\rangle$, E_n and E_k are the energies of $|\Phi_n\rangle$ and $|\Phi_k\rangle$, respectively. $\hat{H}(t) = \hat{\mathbf{D}} \cdot \mathbf{E}_{\text{laser}}(t)$ describes the system-laser interaction, where $\hat{\mathbf{D}}$ and $\mathbf{E}_{\text{laser}}$ are the electric-dipole-transition operator and the electric field of the laser pulse, respectively. Eqs. (1) are solved numerically with an embedded fifth-order Runge-Kutta method combined with the Cash-Karp adaptive-step-size control [66]. The laser parameters [amplitude, energy, full width at half maximum (FWHM), chirp, polarization, and direction of propagation] are optimized with a specially developed genetic algorithm [67]. The fidelity of each process is defined as the final population of the desired final state. Additional details of the theory and the methods are given in the supplementary material [65].

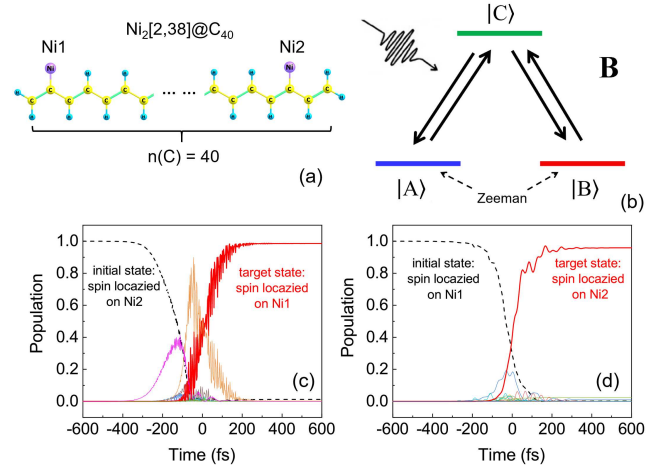


FIG. 1. (a) $\text{Ni}_2[2,38]@\text{C}_{40}$ geometry. (b) Λ process [65]. (c) and (d) time-dependent population of the initial (dashed black line), target (solid red line), and intermediate states (solid line in other colors) participating in the spin-transfer process for EOM-CCSD and SAC-CI, respectively (laser parameters shown in Tab. I).

In EOM-CCSD the ground state of $\text{Ni}_2[2,38]@\text{C}_{40}$ is the initial state, and a state at 3.486 eV with the same spin orientation but with spin localization on Ni1 is our target state, thus achieving a spin transfer over 4.428 nm, the longest distance in our results, within 750 fs. This is the first major result of this study. In SAC-CI a similar process is found, with a final state lying at 2.834 eV and a completion time of 700 fs with 95.9% fidelity. Both calculations indicate that a long-distance spin-transfer process can be achieved through a carbon-based channel system [see Figs. 1 (c), (d) and Table I], contrary to our unsuccessful previous attempts in which linear sodium chains were used. Due to the zigzag geometry of the carbon chain (sp^2 hybridization), the two Ni atoms couple to each other indirectly through the conjugated delocalized molecular orbitals. These, although energetically close, are still discrete leading to ferromagnetic coupling of the Ni atoms. This mechanism lies in-between the superexchange interaction [68] (described by the Goodenough-Kanamori rules [69, 70]) and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediated through conduction electrons in metallic systems [71, 72].

Integrating logic processors necessitates the individual addressability of each functional unit (in our case each Ni atom). In order to increase the integration density beyond the spatial optical limit imposed by the laser wavelength, we use an inhomogeneous magnetic field and potentially double pulses [schematically shown in Fig. 2(a)]. By varying the applied field, we can distinguish between local spin-flip and global

TABLE I. Optimized parameters of the laser pulse for each spin-dynamics process. ΔE is the energy difference between the initial and target states. θ and ϕ denote the angles of incidence in spherical coordinates, and γ is the angle between the polarization of the light and the optical plane. FWHM is the full width at half maximum of the laser pulse, E_{laser} denotes the laser energy. The chirp is linear and the tabulated values indicate the change of the frequency at times $\pm$ FWHM with respect to the peak (central) frequency.

<i>cis</i> configurations										
Structures	method	scenario	fidelity	ΔE (eV)	θ ($^\circ$)	ϕ ($^\circ$)	γ ($^\circ$)	FWHM (fs)	E_{laser} (eV)	chirp
Ni ₂ [2,38]@C ₄₀	EOM-CCSD	spin flip	97.1%	0.109×10^{-3}	135.0	95.5	322.9	484.035	3.092	1.017
Ni ₂ [2,38]@C ₄₀	EOM-CCSD	spin transfer	98.6%	3.486	96.4	77.4	27.4	250.041	3.757	1.000
Ni ₂ [2,38]@C ₄₀	SAC-CI	spin flip	99.9%	0.104×10^{-3}	19.2	5.8	2.5	463.376	2.076	1.000
Ni ₂ [2,38]@C ₄₀	SAC-CI	double-pulses	97.8%	0.104×10^{-3}	241.7	341.2	55.9	488.510	1.016	0.998
Ni ₂ [2,38]@C ₄₀	SAC-CI	spin transfer	95.9%	2.834	90.2	85.7	145.2	405.623	0.960	1.000
Ni ₂ [4,38]@C ₄₀	SAC-CI	spin transfer	81.0%	2.741	250.4	92.1	102.9	135.257	0.736	1.000
Ni ₂ [6,38]@C ₄₀	SAC-CI	spin transfer	61.8%	2.324	96.0	165.8	166.4	330.460	1.144	1.041
Ni ₂ [8,38]@C ₄₀	SAC-CI	spin transfer	58.9%	2.831	90.1	350.5	163.0	256.978	1.398	1.000
Ni ₂ [10,38]@C ₄₀	SAC-CI	spin transfer	62.2%	2.661	267.5	118.8	122.1	382.135	2.706	1.000
<i>trans</i> configurations										
Structures	method	scenario	fidelity	ΔE (eV)	θ ($^\circ$)	ϕ ($^\circ$)	γ ($^\circ$)	FWHM (fs)	E_{laser} (eV)	chirp
Ni ₂ [5,38]@C ₄₀	SAC-CI	spin transfer	98.5%	0.014	95.9	110.1	294.8	168.845	2.016	1.000
Ni ₂ [7,38]@C ₄₀	SAC-CI	spin transfer	88.2%	0.368	290.2	102.8	26.9	438.765	3.304	0.999
Ni ₂ [9,38]@C ₄₀	SAC-CI	spin transfer	70.6%	0.223	319.7	261.4	69.7	484.868	2.160	0.963
Ni ₂ [11,38]@C ₄₀	SAC-CI	spin transfer	45.7%	0.056	359.6	175.4	304.0	328.930	2.598	1.020

spin-transfer processes, by exploiting their tolerance against the strength of the local magnetic field. Local processes have a strong dependence, i.e., only the Ni atom for which the pulse is optimized responds to the laser pulse (a scenario which is desirable for local manipulation of information). Processes with little dependence can extend over a larger area (suitable for the transport of information [25, 26]). In Ni₂[2,38]@C₄₀ the spin flips on Ni1 and Ni2 are indeed local processes. To check this we take the $\langle S_z \rangle \approx -1$ substate of the triplet ground state as the initial state and the $\langle S_z \rangle \approx +1$ substate as the final state. The intermediate state is spin-mixed (due to SOC). The fidelity for the optimized laser pulses is 97.1% and 99.9%, for the EOM-CCSD and the SAC-CI calculations, respectively. We check the dependence on the magnetic field by varying its magnitude from 0 to 23.505 T and repeating the processes with the same laser pulses. Generally, with increasing B field, the fidelity of the spin-flip processes drops, since the necessary spin-mixed intermediate states gradually become spin-pure states, thus in the Paschen-Back limit the process is completely suppressed. Spin-transfer processes are, on the contrary, global processes [black line in Fig. 2(b)]. Since they do not need spin-mixed intermediate states, they are insensitive to the disentanglement of the spin components even in the Paschen-Back limit. The detuning of the laser pulse due to the Zeeman splitting is too weak compared to the laser frequency.

We furthermore propagate again after excluding the two major spin-mixed intermediate states (at 2.059 and 2.067 eV) for the SAC-CI calculations, which are

involved in both the spin-flip and transfer processes. The spin-flip gets almost completely suppressed, while the spin transfer survives (with lower fidelity) and remains independent of the magnetic field. If, however, we exclude resonant pure-spin intermediate states, then this transfer becomes suppressed too. Thus we conclude that in Ni₂[2,38]@C₄₀ the magnetic field can distinguish between local (intrasite) and global (intersite) processes. This constitutes our second major result. The more detailed description of the involved states is given in the supplementary material [65].

Next we investigate the spatial limit of selectively addressing single magnetic atoms in a grid (Fig. 2). Taking the maximal possible magnetic field gradient to be of the order of 1-10 T/ μm [73–78], the two nickels should be about 23.5 nm apart in order for a local spin-flip process to be successful on one and completely suppressed on the other in a weak magnetic field strength ($B < 0.235$ T). However, when $B > 0.235$ T, this distance will be extended to 0.910 μm . By using two identical laser pulses with a delay twice their FWHM rather than one, we can exploit the quantum interferences between the participating intermediate states and substantially decrease this distance, e.g., down to 21.9 nm for $B < 0.235$ T and 0.121 μm for $B > 0.235$ T. The fidelity of the optimized double-pulse spin-flip reaches 99.8% (for the SAC-CI calculations), but drastically drops for increasing magnetic fields [blue line in Fig. 2(b)]. However, it increases again, when the relative phase between the intermediate states becomes favorable again, leading to an oscillatory behavior. By adjusting the delay, one can additionally control the

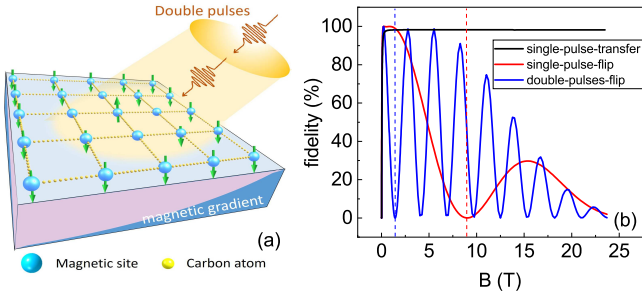


FIG. 2. (a) schematic of an optical integrated logic unit. The blue and yellow spheres denote magnetic atoms (e.g., Ni atoms) and carbon atoms, respectively. The green arrows denote the spin direction. The local spin flip and global spin transfer are realized by external laser pulses. An inhomogeneous magnetic field helps individually address the magnetic atoms. (b) dependence of different spin processes on the magnetic field (based on SAC-CI calculations). The black, red, and blue lines denote the single-pulse spin-transfer, single-pulse spin-flip, and double-pulse spin-flip processes, respectively. The vertical dashed lines indicate the magnetic field strength that totally suppresses the spin-flip processes.

spatial selectivity. Setting it to 50 times the FWHM reduces the distance down to 7.1 nm, exactly matching the current CMOS limit. The increased site selectivity with double laser pulses is our third major result.

Finally, we study the dependence of the spin-manipulation scenarios on the Ni-Ni distance. We look into eight different structures, with Ni1 attached to sites 4, 5, 6, 7, 8, 9, 10, and 11, respectively ($\text{Ni}_2[3,38]@C_{40}$ is not included, because in the C_{2h} symmetry the two Ni atoms are indistinguishable). They can be divided into two classes (the geometries are given in the supplementary material [65]): *cis* configurations (Ni atoms on the same side of the chain), namely $\text{Ni}_2[2,38]@C_{40}$, $\text{Ni}_2[4,38]@C_{40}$, $\text{Ni}_2[6,38]@C_{40}$, $\text{Ni}_2[8,38]@C_{40}$, and $\text{Ni}_2[10,38]@C_{40}$, and *trans* configurations (Ni atoms on opposite sides

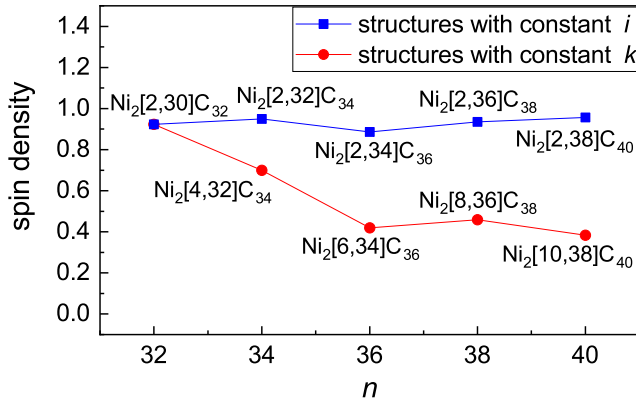


FIG. 3. Spin densities of Ni1 in some structures with constant i (blue line) and constant k (red line).

of the chain), namely $\text{Ni}_2[5,38]@C_{40}$, $\text{Ni}_2[7,38]@C_{40}$, $\text{Ni}_2[9,38]@C_{40}$, and $\text{Ni}_2[11,38]@C_{40}$. The ground state is a singlet in the *trans* configurations and a triplet in the *cis* configurations. For the latter ones, the spin localization on Ni1 decreases as the Ni1-Ni2 distance gets shorter due to the shorter distance between the Ni atoms and the increasing distance between Ni1 and the edge. To check this we also calculate the spin density in structures with the same Ni1-edge distance but different Ni1-Ni2 distances (structures with constant $i = 2$ but variable $k = 29, 31, 33, 35, 37$), and structures with the same Ni1-Ni2 distance but different Ni1-edge distances (structures with constant $k = 29$ but variable $i = 2, 4, 6, 8, 10$). We find that the Ni1-edge distance is the main factor affecting the spin localization (Fig. 3), analogous to the edge magnetization in graphene. In fact, the closer Ni1 is to the center of the chain, the stronger it interacts with the conjugated C-C bonds, inducing an electron delocalization, which, in turn, results in less spin density on it. In the *cis* configurations (with triplet ground states), shorter Ni1-Ni2 distances result in lower

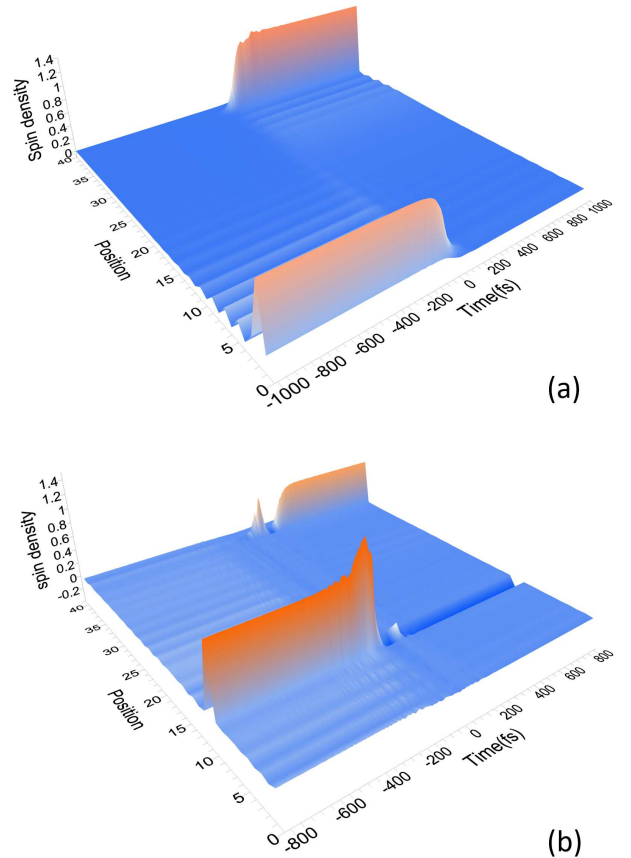


FIG. 4. The time evolution of expectation values of the spin density distributed on (a) $\text{Ni}_2[2,38]@C_{40}$ and (b) $\text{Ni}_2[11,38]@C_{40}$ during the spin-transfer processes. The colors and height both denote the expectation values of spin densities.

fidelties (Tab. I) and higher laser frequencies (all Λ scenarios are complex multi-photon processes). Only in $\text{Ni}_2[10,38]@\text{C}_{40}$ the laser frequency is in resonance with the excited state.

In the *trans* configurations the spin-flipping processes must start from an excited state (since the ground states are singlets) and generally have lower fidelity (Tab. I). Shorter Ni1-Ni2 distances also result in a deterioration of the Λ process. Extraordinarily, in $\text{Ni}_2[11,38]@\text{C}_{40}$ the spin transfer is accompanied by a spin flip, resulting in an asymmetric antiparallel spin configuration (Fig. 4). We attribute this special spin-transfer scenario to the strong conjugation effect of the carbon chain, which induces similar spatial distributions for both spin species. Generally all transfers are *not* a ballistic, but rather of a “communicating vessels” type: the spin density disappears on the one Ni and directly reappears on the other (Fig. 4). The details of the spin localization and spin transfer on the *cis* and *trans* configurations constitute our fourth result.

In conclusion, using high-level quantum chemistry we systematically investigate the ultrafast laser-induced spin-manipulation scenarios on a zigzag carbon chain, to which two Ni atoms are attached. We find that (i) the conjugated C-C bonds allow for spin transfer at a distance of 4.428 Å, comparable to today’s CMOS scale, (ii) a gradient of the magnetic field allows the distinction between local (intrasite) spin-flips and global (intersite) spin-transfer, (iii) using double laser pulses can substantially improve the spatial resolution and individual addressability of the local (intrasite) spin-flips, and (iv) in the *cis* configurations the ground state is a triplet, while in the *trans* configurations it is a singlet. We also find that in general the spin density on the Ni atoms decreases when they are closer to the center of the carbon chain, and that a shorter Ni-Ni distance necessitates higher laser frequencies. Last-but-not-least, the spin transfer is not ballistic, but happens in a “communicating vessels” way. Our results demonstrate the feasibility of individual intrasite and intersite spin manipulation and therefore represent a big step towards the integration of magnetic-logic elements in larger circuits, and ultimately building logical (nano)spintronic devices.

This work is supported by the National NSF (Grant Nos. 11872309, 11572251, and 11504223) of China, the National Science Basic Research Plan in Shaanxi Province (Grant Nos. 2020JM-120 and 2017JM1033), and the Fundamental Research Funds for the Central Universities (Grant Nos. 3102017JC01003 and GK201903015) of China.

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