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Optical spin manipulation for minimal magnetic logic operations in metallic three-center magnetic clusters

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We present a first-principles scenario where a realistic three-magnetic-center metallic cluster acts as a prototypic magnetic-logic element within the frame of a unified optically induced spin manipulation. We find that the spins of the energetically low-lying triplet states of a Ni_3Na_2 cluster are always localized at a single magnetic center and that controlled spin flips and transfers are possible within a hundred femtoseconds with suitable static external magnetic field and laser pulses. The magnetic state or the position of the spins and the static magnetic field can be used as input bits while the output bit is the final state of the magnetic centers, thus the gates AND, OR, XOR (CNOT), and NAND can be built.

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I. INTRODUCTION

In recent years due to the continuous speed upscaling and size downscaling of computers, new technologies to complement existing semiconductor electric-charge-based transistors are needed. Magnetic logic appears as an appealing alternative due to its nonvolatile character, which can boost up switching on/off, its possibility to reduce the size of the element down to the several-atoms scale (one spin per atom instead of one elementary charge per 10^4 atoms in semiconductors), and speed increase as a secondary size effect. To that end, several experiments have been performed which however deal with macroscopic magnetic effects such as magnetoresistive elements,¹ magnetic-domain-wall logic,² and majority logic gates for magnetic quantum dots.³ While these experiments are promising, they still move in the micrometer regime, thus not fully exploiting the possible quantum nature of molecular magnetism. A different approach toward smaller structures has been taken by de Silva and Uchiyama where small molecules perform logic operations using as input cation concentrations.⁴ The latter is fast with respect to the logic operation but slow with respect to repeatability. Thus a need for magnetic-logic devices on the molecular scale emerges. At the same time the experimental evidence of laser-driven ultrafast magnetic manipulation in (anti) ferromagnetic materials^{5,6} motivates the design of a cluster with more than one magnetic center which allows for spin manipulation both spectroscopically and spatially resolved, i.e., both spin switch and spin transfer.

Theoretical works on the other hand do present potentially smaller spintronics systems however with the use of model Hamiltonians that do not include all the effects of realistic magnetic materials,⁷ even though some of them describe complicated quantum-computing structures based on many-qubit states⁸ or submicrometer devices that perform logical NOT operations on magnetic-logic signals.⁹ Much progress has been achieved in the field of quantum computing and the realization of logic gates, e.g., the works of Troiani *et al.*¹⁰ where doped semiconductor double-quantum-dot molecules were proposed as qubit realization. There however the model

Hamiltonian is driven by a (relatively) slow Raman adiabatic passage.¹⁰ A realistic Cr_7Ni ring shows promising behavior, though being driven by a magnetic field it exhibits dynamics in the few hundred picoseconds regime.¹¹ At the same time the logical functionalization of model Hamiltonians has allowed realization of a CNOT gate in finite Heisenberg- and Ising-type spin chains.¹² Overall, to date no realistic materials have been shown to exhibit ultrafast full-fledged magnetic-logic functionalization.

II. MAGNETIC LOGIC

In order to perform logical operations on a molecular cluster, the structure must consist of a certain number of poles. In the case of one center, i.e., in our case one spin, this center needs to be used both as incoming and outgoing signals (the two poles are then temporally and not spatially separated, a fact that hinders the permanent connectivity to other elements) [see Fig. 1(a)]. Single magnetic centers are useful for conventional computer memories. Two magnetic centers already allow for signal transport [Fig. 1(b)] while three centers may in addition provide interference features [Fig. 1(c)]. Finally a fourth center can act as a control switch [Fig. 1(d)]. An important factor is also the symmetry of the structure. Symmetric molecules do not allow discrimination between different out poles, while asymmetry enables output-signal differentiation by means of pure population effects or quantum interference effects [Figs. 1(d) and 1(f)]. The cluster can be dissolved in a liquid suitable for spectroscopy, mass selected and optically probed in the gas phase, or be deposited on a surface. The energy separation of the electronic levels must delicately balance between being neither too far apart, in order to remain addressable, nor too close, to allow distinguishability and avoid mixing due to thermal broadening. The proximity of the poles renders spatial resolution very difficult which underlines the importance of distinction by exploiting the different resonance structure of the magnetic centers (geometric asymmetry).

Here we present a functioning spin-based nanologic unit, where the prerequisite is a unified *ab initio* picture of opti-

FIG. 1. (Color online) Several possible structures for magnetic logic (a) with one pole, (b) two poles, (c) three poles symmetric and (d) asymmetric, and (e) four poles symmetric and (f) asymmetric. White spheres indicate the input bit and the red (dark gray) and yellow (light gray) ones the spatially separated output bits. With four or more poles one can imagine a control pole as well (gray “ctrl” spheres).

cally induced switching and information transport with the use of high-level quantum chemistry, thus taking into account correlation effects that otherwise remain elusive. The proposed cluster, which consists of three magnetic centers (Ni), interconnected with Na atoms, can be synthesized, e.g., by soft landing of the atoms on an inert Cu surface, that serves as structure stabilizer. The Ni atoms are interconnected with Na chains and the interatomic distance is set to 3.6 Å, the lattice constant of a fictitious Cu(001) surface which acts as a structure-stabilizing substrate. An alternative environment of the clusters could be in the liquid phase. This would avoid the broadening due to surface effects but replace it with broadening due to stabilizing ligands. It would additionally exhibit the necessity of markers in order to detect the electronic state. Clusters in liquid phase might be more stable but the spectroscopic data of the solution would inevitably mix with the desired structure scheme. Here we derive a structure taking care (a) that the states are discrete, (b) the intermediate (Λ) state is energetically far enough so that the process is fast, and (c) that the energy difference between initial and final state is balanced between being small enough with respect to the intermediate state, so that the Λ process is achievable and direct relaxation processes between the states are slow, but still adequately large so that the initial and final state are energetically separable (e.g., their respective populations due to thermal distribution are not equal).

In previous works we have shown the possibility of local all-optical spin switching, i.e., the explicit addressing and local manipulation of the spins of a NiO cluster embedded in a well-defined chemical environment^{13–15} and two-magnetic-center metallic chains.^{16,17} In this paper we extend this idea further by including more magnetic centers so that an all-optical *spin transfer* as an additional scenario can be realized along with the local *spin-switching* mechanism, thus leading to an enhanced functionality. The driving force is in both cases (as discussed in our previous works) the spin-orbit coupling (SOC) that interlinks light helicity and spin angular

momentum. Not all Ni atoms lie on a straight line in order to (a) locally lift spatial and thus electronic symmetry and (b) simulate at a minimum level the branching of the propagation of a signal. The cluster must be asymmetric so that the many-body wave functions are nondegenerate and the respective localized spin densities are distinguishable. The chain branching ensures that the magnetic centers always “terminate” the chains. The Na atoms, although nonmagnetic, contribute an odd number of electrons and their number is chosen so that we arrive at an even total number and deal with singlets and triplets instead of doublets and quartets,¹⁸ so we can separate spin and charge dynamics.

III. THEORY AND RESULTS

Calculations are performed at three stages. First the highly correlated electronic structure of the system is obtained on a nonrelativistic level with the use of the symmetry-adapted cluster configuration-interaction method (SAC-CI) of Nakatsuji *et al.*¹⁹ incorporated in the GAUSSIAN 03 package.²⁰ Then SOC and an external static magnetic field are added by means of time-independent perturbation theory and finally the laser pulse is turned on as a time-dependent perturbation (semiclassical model); integration over time is done with the fifth order Runge-Kutta method and Cash-Karp adaptive step size control (see previous works^{13–15}). The expectation values of the various operators are calculated with the reduced-density-matrix formalism in order to determine their spatial localization as well.

The first unexpected result is that all energetically low-lying many-body magnetic states have spin localization at a single magnetic center. Subsequently we define the easy axis for every state as the direction of an infinitesimal external $\mathbf{B}$ field for which the energy reaches its minimum. This $\mathbf{B}$ field has a strength of 10^{-5} a.u., is homogeneous, and couples to all the atoms of the cluster. Note that the spins mostly point in plane and that the spins of two out of the three magnetic centers are almost (anti) parallel while the third one points in an orthogonal and out-of-plane direction (Fig. 2).

Typically (for a whole family of similar clusters) we find that the lowest-lying many-body states originate from triplets (after inclusion of SOC and Zeeman splitting) with their spin densities localized at a single magnetic center. For excitation energies below 1 eV we always find at least one “spin-up” and one “spin-down” state localized at any given magnetic center plus several nonmagnetic ones (Table I). Note that spin up or spin down merely means that although S is not a good quantum number, the expectation value of its projection along the respective easy axis $\hat{q}$ is in the vicinity of ± 1.8 . Moreover they do not refer to the spin of individual electrons but to the expectation value of the spin-density operator acting on the whole many-body wave function.

Three quantities can be used as input bits for our magnetic-logic unit: (a) the overall magnetic state (spin up or spin down), (b) the localized magnetic state (spin up, spin down or absent), and (c) the localization of the magnetic state (magnetic center 1, 2, or 3). The same quantities can be regarded as output bits as well. Clearly the idea of two spins localized at two centers as input which, after a logical opera-

FIG. 2. (Color online) Level scheme of Ni_3Na_2 structure (without SOC). The solid-black lines are spin triplets and the dashed red ones spin singlets. The six structures next to the level scheme show the spin localization. Large circles represent Ni atoms and small circles magnetically inert Na atoms. Solid circles indicate the spin localization of each state (arrows next to the sphere show its easy-axis direction). Note that the upper two states have the spin perpendicular to the molecular plane (xy plane).

tion, lead to a spin localized to another center has to be abandoned since the two spins are *always* localized at the same atom (at least for the 50 lowest states, however spin localization at a Na atom is possible at high energies, an idea that can be resumed for the case of more than one cluster—note also that in such an arrangement the clusters need not be parallel to each other). Finally three possible mechanisms naturally emerge in a unified picture from our first-principles theory, i.e., (a) local spin flip, (b) spin transfer, and (c) simultaneous flip and transfer of the spin, all possible with a suitable Λ process and an optimized laser pulse.¹⁵ All optimizations were performed with a specially developed genetic algorithm.¹⁶ As it turns out, however, not all mechanisms are possible: while spin flip is almost always feasible (although occasionally cumbersome), spin transfer can only be achieved between magnetic centers with (almost) parallel easy axes (note that the cluster as a whole is illuminated but only one center at a time is in resonance, which leads to an effective localization of the light pulse). Simultaneous flip and switch could not be achieved. The presence of the third “isolated” magnetic center facilitates the different processes, although it does not directly participate in them.

The most interesting findings are: (a) there exists a $\mathbf{B}$ -field orientation which allows a spin transfer but *no* spin flip. This can be used for controlling the localization of the spin in the logical process without loss of the overall spin orientation (see Fig. 4). (b) The local spin flip at the edge Ni atom is 5 times slower (approximately 450 fs) than at the middle atom (approximately 100 fs). This difference can be used to selectively flip the spin depending on its localization by simply using a pulse which is long enough to flip the spin only if it is located at the middle atom but not at the edge Ni.²¹ In similar investigated clusters (with Ni and Co centers) either

TABLE I. Some of the lowest levels of the Ni_3Na_2 cluster (with SOC) with a static external field almost parallel to the easy axis of the ground state (slightly out of plane with $\theta=77^\circ$ and $\phi=90^\circ$). Arrows indicate the approximate direction of the spin density. The states marked as bold are the ones used in the logic processes (compare to Fig. 2). Note that states 61–63 are where the spin density is mainly localized on the chain Na atoms. The lowest 40 states originate from triplets. The first “singlet” is state 41 (not shown here).

State	Energy (eV)	$\langle S \rangle$	Direction	Atom
63	1.4376	1.258	$\downarrow$	Na
62	1.4373	0.068	n.a.	Na
61	1.4370	1.250	$\uparrow$	Na
20	0.2552	0.006	n.a.	Middle Ni
19	0.2410	0.006	n.a.	Edge Ni
18	0.2384	0.004	n.a.	Edge Ni
17	0.2179	0.738	$\searrow$	Middle Ni
16	0.2164	0.736	$\swarrow$	Middle Ni
15	0.2038	1.872	$\swarrow$	Middle Ni
14	0.2024	1.872	$\nearrow$	Middle Ni
13	0.1922	1.050	$\downarrow$	Edge Ni
12	0.1912	1.058	$\uparrow$	Upper Ni
11	0.1812	1.918	$\downarrow$	Edge Ni
10	0.1800	1.918	$\uparrow$	Edge Ni
9	0.1780	0.102	$\downarrow$	Upper Ni
8	0.1665	0.214	$\odot$	Upper Ni
7	0.1642	0.238	$\otimes$	Upper Ni
6	0.0331	1.628	$\downarrow$	Middle Ni
5	0.0321	1.628	$\uparrow$	Middle Ni
4	0.0272	0.088	n.a.	Middle Ni
3	0.0063	1.640	$\downarrow$	Edge Ni
2	0.0052	1.642	$\uparrow$	Edge Ni
1	0.0000	0.088	n.a.	Edge Ni

the resonances of the two processes do not differ enough to make them controllable or close vicinity of excited states leads to destructive interference (only Co magnetic centers).

IV. DISCUSSION

By exploiting all the aforementioned processes we find combinations that lead to different logic operations where we typically think of the edge Ni as the input bit and the $\mathbf{B}$ field as the control bit (or in an alternative nomenclature a second input bit). For example, take an AND gate built with $\text{Ni}_2\text{Na}_2\text{Ni}$: one input bit is the spin orientation at the edge atom, i.e., 1 for spin up and 0 for spin down, the second input bit is the orientation of the $\mathbf{B}$ field, i.e., 1 for light polarization parallel to the propagation direction of the light and 0 for perpendicular ($\theta=0^\circ$) light polarization. We find that spin transfer is optimized with linearly polarized light (in line with previous findings¹⁵) and parallel field. After the pulse is over we search for a spin-up orientation at the middle atom: if we find it the state reads bit 1 and if not it reads bit 0. The possible outcomes of the operations match

TABLE II. AND gate. We put in the spin at the edge Ni and the **B** field and read the middle Ni “up” state. θ and ϕ are the angles with respect to the normal of the molecular plane and the short molecular axis, respectively (xy is the molecular plane).

Input 1	Input 2	Output
Spin	B field (ctrl)	Spin+position
1 (edge $\uparrow$)	1 ($\theta=0^\circ$)	1 (middle $\uparrow$)
0 (edge $\downarrow$)	1 ($\theta=0^\circ$)	0 (middle $\downarrow$)
1 (edge $\uparrow$)	0 ($\theta=78^\circ$ and $\phi=96^\circ$)	0 (edge $\uparrow$)
0 (edge $\downarrow$)	0 ($\theta=78^\circ$ and $\phi=96^\circ$)	0 (edge $\downarrow$)

exactly the truth table of the AND gate (Table II). Especially for the AND gate we are able to construct two different realizations with the one input bit being either the magnetic state of the edge Ni atom (Table II) or the localization of a spin-up state, i.e., whether the spin is at the edge or the middle atom (Table III). In both cases the output is interpreted as 1 if the spin is located at the middle Ni and points “up.” This is one of the reasons why the branching of the chain is needed in order to best spatially differentiate the input from the output bits.

In a more complicated scenario, we apply a spin-flipping pulse followed by a spin-transfer pulse and, depending on the magnetic state of the edge Ni and the magnetic field, we detect the magnetic state of the middle Ni atom again. This time however the truth table corresponds to an XOR gate (Table IV) which plays a role analogous to the famous quantum CNOT gate. Since by detecting the localized spins we “read” the result of an operation, one could think of putting several clusters together so that adjacent spins would take over the role of the **B** field. Thus the output bit of one cluster could act as the control bit of the next one. For carefully chosen distances the spin could be felt by the neighboring element without their respective wave functions getting combined (in order not to give one localized spin density only). Furthermore their respective orientations could lead to a different interpretation of spin-up and spin-down states, giving the possibility of reinterpreting the bits 0 and 1. Thus a redefinition of all the bits 0 as 1 and vice versa of an AND gate gives rise to an OR gate (Table V) and a redefinition of only the output bit of an AND gate results in an NAND gate (not

TABLE III. Another AND gate. One input bit is the position of the spin and the control bit is the **B** field. We read the middle Ni up state as output bit. θ and ϕ are the angles with respect to the normal of the molecular plane and the short molecular axis, respectively (xy is the molecular plane).

Input 1	Input 2	Output
Spin	B field (ctrl)	Spin+position
0 (edge $\uparrow$)	0 ($\theta=78^\circ$ and $\phi=96^\circ$)	0 (edge $\downarrow$)
1 (middle $\uparrow$)	0 ($\theta=78^\circ$ and $\phi=96^\circ$)	0 (middle $\downarrow$)
0 (edge $\uparrow$)	1 ($\theta=0^\circ$)	0 (edge $\uparrow$)
1 (middle $\uparrow$)	1 ($\theta=0^\circ$)	1 (middle $\uparrow$)

TABLE IV. XOR (CNOT) gate. We put in the spin at the edge Ni and the **B** field and read the middle Ni up state. θ and ϕ are the angles with respect to the normal of the molecular plane and the short molecular axis, respectively (xy is the molecular plane).

Input 1	Input 2	Output
Spin	B field (ctrl)	Spin
1 (edge $\uparrow$)	1 ($\theta=78^\circ$ and $\phi=96^\circ$)	0 (middle $\downarrow$)
0 (edge $\downarrow$)	1 ($\theta=78^\circ$ and $\phi=96^\circ$)	1 (middle $\uparrow$)
1 (edge $\uparrow$)	0 ($\theta=0^\circ$)	1 (middle $\uparrow$)
0 (edge $\downarrow$)	0 ($\theta=0^\circ$)	0 (middle $\downarrow$)

shown here). The fidelities of all the aforementioned processes (between 78% and 93%, see Figs. 3 and 4) can be very nicely compared to conventional semiconductor-based electronics, where for a 5 V input one counts with up to 0.7 V of signal loss, therefore logical values of 0 and 1 are typically assigned only to voltages below 1.5 or above 3.5 V, respectively.

Note that there are some operational dead times that originate from the time needed to switch on and off the external magnetic field. This time however is not decisive for the spin dynamics of the process and prevents the occurrence of secondary undesirable effects, e.g., during random access memory (RAM) reading or writing. It is important to note as well that the direct transitions between the initial and the final states would be forbidden if it were not for SOC. Hence the transition matrix elements between them are weak (typically between 10^{-1} and 10^{-3} a.u.), which for energy separations in the order of 70 meV (Fig. 2) leads, for the spontaneous emission, to half lives considerably longer than 1 ps. Thus the prerequisite that the coherence time be longer by a factor of 10^4 than the process time itself is fulfilled. On this time scale phonons can play a very important role among others because they can alter the selection rules for the electric dipole transitions.²²

V. CONCLUSIONS

In conclusion we have performed high-level first-principles quantum-chemical calculations of three-magnetic-center clusters and found that the spin is always localized at

TABLE V. OR gate. We put in the spin at the edge Ni and the **B** field and read the middle Ni up state. θ and ϕ are the angles with respect to the normal of the molecular plane and the short molecular axis, respectively (xy is the molecular plane).

Input 1	Input 2	Output
Spin	B field (ctrl)	Spin+position
0 (edge $\uparrow$)	0 ($\theta=0^\circ$)	0 (middle $\uparrow$)
1 (edge $\downarrow$)	0 ($\theta=0^\circ$)	1 (middle $\downarrow$)
0 (edge $\uparrow$)	1 ($\theta=78^\circ$ and $\phi=96^\circ$)	1 (edge $\uparrow$)
1 (edge $\downarrow$)	1 ($\theta=78^\circ$ and $\phi=96^\circ$)	1 (edge $\downarrow$)

FIG. 3. (Color online) Spin manipulation on $\text{Ni}_2\text{Na}_2\text{Ni}$. Left: local spin flip on the edge Ni. Right: spin transfer from the edge Ni to the middle Ni. The upper panels show the respective occupations of the relevant states, the middle panels the projections of $\langle M_s \rangle$ on the atoms, and the lower panels the pulse envelope. The laser is linearly polarized, has a maximum amplitude of 2.57×10^9 V/m, and propagates along the easy axis of the edge Ni (see Fig. 2).

one single atom. Furthermore we have shown that with the proper use of a homogeneous external magnetic field and laser pulses it is possible to explicitly manipulate the spin in two ways, i.e., to flip spins locally or to transfer spin density from one magnetic center to another. Finally and most importantly by using these controlling mechanisms we propose a prototypic magnetic-logic element based on a sufficiently realistic material.

FIG. 4. Spin flips and transfers in $\text{Ni}_2\text{Na}_2\text{Ni}$. Spheres indicate the magnetic centers and arrows the localization and direction of the spin. The numbers show the fidelity of the Λ processes. All four mechanisms are possible if the $\mathbf{B}$ field has $\theta=155^\circ$ and $\phi=270^\circ$ or $\theta=78^\circ$ and $\phi=96^\circ$ (solid arrows). If the $\mathbf{B}$ field has $\theta=0^\circ$, i.e., if it is perpendicular to the cluster plane, then only transfer is possible and *no* switch. Thus the orientation of the static $\mathbf{B}$ field opens and closes the spin-switch channel. A $\mathbf{B}$ field along the molecule axis allows for a spin flip at the edge Ni (process Λ_1) only with a much longer laser pulse (approximately 450 fs) while spin flip at the middle Ni (process Λ_3) and transfer can be achieved with shorter pulses (<100 fs).

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