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Switching of antiferromagnetic chains with magnetic pulses

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Recent experimental studies have demonstrated the possibility of information storage in short antiferromagnetic chains on an insulator substrate [S. Loth *et al.*, Science **335**, 196 (2012)]. Here, using the density functional theory and atomistic spin dynamics simulations, we show that a local magnetic control of such chains with a magnetic tip and magnetic pulses can be used for fast switching of their magnetization. Furthermore, by changing the position of the tip one can engineer the magnetization dynamics of the chains.

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Manipulation of spin switching at the nanoscale or even at the atomic level is the key for the miniaturization of storage devices. Usually, for ferromagnetic systems, their spins can be controlled by the magnetic field or spin transfer torque [1]. The first spin switching of a single adatom on the insulator surface was observed by Heinrich *et al.* [2]. Several studies have demonstrated that the spin states of single adatoms can also be controlled by exchange interactions [3–6] or the spin transfer torque [7,8]. Due to the lack of magnetic moment and the relatively strong exchange interactions in antiferromagnetic (AFM) systems, their spin states are hard to switch. Critical to switching the AFM system is to exert a local control of the devices [5,9–14]. At the atomic level, the exchange interaction between atoms decays exponentially with the distance and shows a localized character. Recently, it has been demonstrated in experimental and theoretical studies that the exchange interaction with an STM tip can be used to tune the spin states of an AFM atomic chain [6,15].

To manipulate spin states of AFM materials, a local control, such as the STM tip [14] or the exchange coupling [6,15], should be stationary, fixed at a certain position of the AFM materials. And the amplitude of the local control has to be varied to switch AFM systems. When the exchange interaction disappears or the STM tip moves to other places, however, the different spin states of the AFM system will become degenerated and indistinguishable. How to manipulate magnetization switching is still a challenge for the real application of AFM-based spintronic devices.

The rapid progress in laser technology makes it possible to reorient or destroy the spin order of ferrimagnetic and AFM materials [16,17]. Recently, it has been demonstrated that the ultrashort laser pulse, in the terahertz (THz), regime provides a new path to the excitation of magnetic materials [18,19]. The magnetic component of the THz wave can be directly coupled to the spin system through the Zeeman interaction. The precessional motion of the magnetic moment in AFM materials can be controlled or switched by a THz pulse. Up to now, however, laser-induced magnetic switching has mainly focused on bulk materials, such as NiO [18] and $YFeO_3$ [20].

Recent experiments by Loth *et al.* [14] have demonstrated that information can be stored in short antiferromagnetic chains. Local control of the magnetization by a spin-polarized STM was realized [6,15]. Using antiferromagnetic chains to record information requires fast switching of their magnetization and long lifetimes of Néel states. A recent theoretical study [21] predicts that Néel states can be made very stable if one or two dozen Fe spins are used.

The goal of the present paper is to demonstrate that a local magnetic control of atoms in AFM chains and magnetic pulses allow us to perform fast magnetization switching. We provide clear evidence that magnetic dynamics in AFM chains can be engineered by varying the tip-substrate distance and the lateral position of the tip. We concentrate on 3-Fe-atom chains on the Cu_2N surface as used in recent experiments by Yan *et al.* [6]

Our calculations are performed within the framework of density functional theory (DFT) as implemented in the Vienna *Ab Initio* Simulation Package [22,23] with projector augmented wave potentials and the generalized gradient approximation due to Perdew, Burke, and Ernzerhof. The basis set contained plane waves with a kinitec energy cutoff of 520 eV and the total energy was converged to 10^{-7} eV. All geometries were optimized without any symmetry constraint until all residual forces on each atom were less than 0.01eV/Å. Spin-orbital coupling with a fully relativistic effect was included in all calculations. A 3×7 supercell is employed in all calculations, and the distance between two neighboring supercells is larger than 10Å. The supercell consists of five Cu(100) layers containing 42 Cu atoms per layer with a $c(2 \times 2)$N–Cu(100) molecular network on one side. Magnetic adatoms are known to reside on top of Cu sites in Cu_2N. In our calculations, three Fe atoms were placed so as to form a finite chain with a distance of 7.2 Å between neighboring atoms. We mimic the influence of the STM tip on this chain by using nine Cu atoms and one Fe atom as the apex of the tip. The two possible geometric setups of the system are sketched in Figs. 1(a) and 1(b), in which arrows denote the magnetization direction of each atom and Δ is the distance between the tip and the nearest atom of the chain.

To understand the influence of the exchange interaction on the spin dynamics process of the system, we first fit the exchange interaction of the system with the Heisenberg model,

$$E = J_{ts}\hat{S}_t \cdot \hat{S}_s + \sum_{i \neq j} J_{ij}\hat{S}_i \cdot \hat{S}_j, \quad (1)$$

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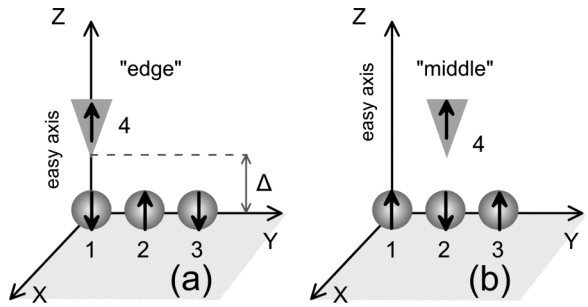

FIG. 1. Local magnetic control of the AFM chain. The tip position can be (a) above the edge atom ("edge") or (b) above the middle atom ("middle") of the chain. The magnetic Fe adatom is placed at the apex of the tip.

where E is the total energy of the system with different spin configurations. The first term on the right-hand side describes the exchange interaction between the tip and the atom just beneath the tip, and the second term is that between atoms in the chain. Since the exchange interaction decays exponentially with the distance, only exchange coupling between nearest neighbors has been considered. According to our calculations, the spin of Fe atoms in all cases was $S = 1.5$, which is used in the following.

We define the magnetic anisotropy energy (MAE) of the chain as $E_{\mathrm{MAE}} = E_{\leftarrow} - E_{\uparrow}$, where $E_{\leftarrow}$ and $E_{\uparrow}$ are the total energy of the system when spins are aligned, respectively, along and perpendicular to the chain axis and the surface plane.

To get the exchange interaction J_{ij} between atoms the total energy of the system with several different spin configurations has been calculated. Exchange interaction and uniaxial anisotropy (K_i) constants for both geometry types and for two distances, as an example, are summarized in Table I. Our DFT results show that the easy axis of the chain is out of plane, and the spin configuration between the tip and its underneath atom is always in an antiparallel alignment for the considered tip-chain distances [24].

Note that the best fit to the experimental results of inelastic electron tunneling spectroscopy of Fe adatoms and chains was found for spin $S = 2$ [25,26] The quantized spin S refers to the spin of many-body states that include both d electrons and ligand electrons. The DFT results clearly show that the charge and spin of the d shell are not quantized [27]. A spin on Fe atoms is found to be $S = 3/2$. However, the cell magnetic moment, which includes magnetic moments of surrounding atoms, is close to the free case [27]. We believe that using DFT results for a localized spin (only Fe) is a good approximation for describing the switching of Fe chains in magnetic fields.

TABLE I. Calculated exchange interaction constants (J_{ij}; in meV) and magnetic anisotropy (K_i) for a tip positioned over the edge or the middle atom in the chain at different tip-chain distances Δ.

	Δ = 5.0 Å		Δ = 4.5 Å	
	Edge	Middle	Edge	Middle
J_{12}	−3.00	−2.99	−2.81	−2.88
J_{13}	0	0	0	0
J_{14}	−0.53	0	−1.9	0
J_{23}	−3.00	−2.99	−2.81	−2.88
J_{24}	0.09	−0.63	0.05	−2.25
J_{34}	0	0.04	0	0
$K_{1,2,3}$	0.3	0.48	3.8	2.36

For theoretical studies of the evolution of a magnetic system the Landau-Lifshitz-Gilbert (LLG) equation is used [28]. This phenomenological equation was primarily implemented for the macroscopic magnetic system, but after some modifications it is also useful for analyzing the atomic spin dynamics [29–36]. We denote $\mathbf{S}_i$ as the unit vector of the ith atom with a magnetic moment μ_s ($\mathbf{S}_i \equiv \boldsymbol{\mu}_s/\mu_s$). Then the magnetization dynamics can be described by the LLG equation [29–31,37],

$$\frac{\partial \mathbf{S}_i}{\partial t} = -\gamma \mathbf{S}_i \times \mathbf{H}^i_{\mathrm{eff}} + \frac{\alpha}{\mu_s}\mathbf{S}_i \times \frac{\partial \mathbf{S}_i}{\partial t}, \tag{2}$$

where γ is the gyromagnetic ratio, α the damping parameter, and $\mathbf{H}^i_{\mathrm{eff}}$ the effective magnetic field acting on the magnetic moment of the ith atom. $\mathbf{H}^i_{\mathrm{eff}}$ is determined by the exchange interactions, the external magnetic field $\mathbf{H}$, and the magnetic anisotropy,

$$\mathbf{H}^i_{\mathrm{eff}} = \frac{\partial}{\partial \mathbf{S}_i}\left[\sum_{j(j\neq i)} \frac{J_{ij}}{\mu_s}\mathbf{S}_i\mathbf{S}_j + \mathbf{S}_i\mathbf{H} + \frac{K_i}{\mu_s}(\mathbf{S}_i\mathbf{e}_a)^2\right], \tag{3}$$

where J_{ij} is the exchange interaction between the ith and the jth atoms ($J_{ij} < 0$ for antiferromagnetic and $J_{ij} > 0$ for ferromagnetic), $K_i = DS^2$ is the anisotropy energy of the ith atom, D is the uniaxial anisotropy parameter, the transverse anisotropy E is negligible [14], and $\mathbf{e}_a$ denotes the direction of the easy axis. Values of the exchange interaction and the magnetic anisotropy energy from our DFT calculations are used, as listed in Table I.

In our system we have only four atoms with nonzero magnetization: three Fe atoms forming the chain and one Fe atom at the tip apex. According to our DFT calculations, in all cases the magnetic moments for these atoms are nearly the same, about $\mu_S = 3\mu_B$.

In the absence of the external magnetic field, the finite chain has two possible Néel states, $|1\rangle = |\downarrow,\uparrow,\downarrow\rangle$ and $|2\rangle = |\uparrow,\downarrow,\uparrow\rangle$, where arrows denote the magnetization orientation of a particular atom in the chain. These two Néel states have the same energy and are quantum mechanically indistinguishable. If we apply an external magnetic field along the easy axis, however, due to interactions between the external magnetic field and the magnetization of the chain atoms, the energy degeneracy of the AFM chain will be lifted and one Néel state will become more energetically favorable than the other Néel state. Thus, we can use the external magnetic field to switching the system from one state to the other.

The first term on the right-hand side of Eq. (2) describes the influence of the effective magnetic field on the magnetization dynamics. According to this and Eq. (3), we can conclude that the effect of the external magnetic field on the magnetization dynamics is vanishingly small as $\mathbf{S}_i \parallel \mathbf{H}$. Hence, it would be rather hard to switch the AFM chain from one Néel state to another Néel state using only the external magnetic field. Here, we propose that, with the help of ultrashort magnetic pulses which act as a sort of "trigger" for magnetization switching

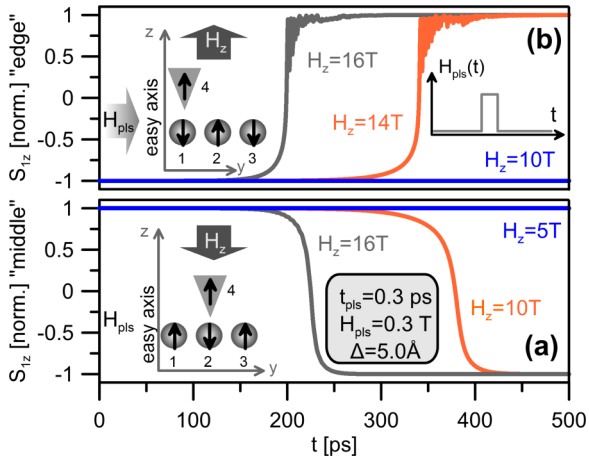

FIG. 2. Switching of the AFM chain. Projection of the magnetic moment unit vector **S** of atom 1 onto the easy axis of the Fe chain under different magnetic fields: for a tip (a) over the edge atom and (b) over the middle atom in the chain.

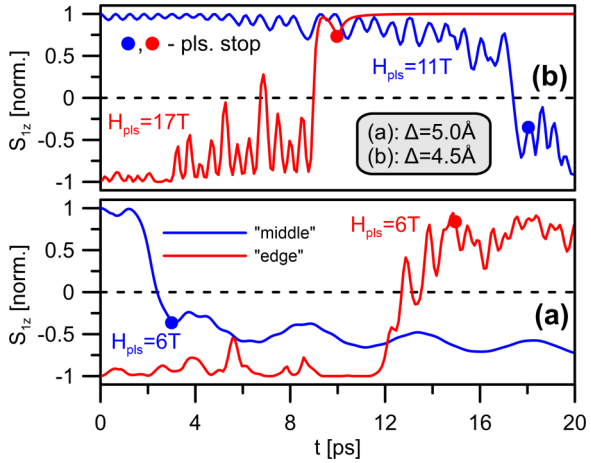

FIG. 3. Magnetization dynamics of atom 1 caused only by a magnetic pulse ($\mathbf{H}_{\rm pls} \perp \mathbf{e}_a$) at different tip-chain distances: (a) $\Delta =$ 5.0 Å and (b) $\Delta = 4.5$ Å.

of the system, the external magnetic field can be used to manipulate the magnetization of a finite AFM atomic chain.

For the magnetic pulse simulation, we used a rectangular magnetic field $H(t)$ with the amplitude $H_{\rm pls} = 0.3$ T and the width $t_{\rm pls} = 0.3$ ps, which are available with current technology [18]. Magnetic pulses can also correspond to THz radiation (magnetic component) as in [18]. Note that for tabletop setups, most THz generation schemes rely on intense femtosecond laser pulses from powerful laser systems [38].

Also, the generation of a magnetic-field pulse was done with relativistic electron bunches (at the Stanford Linear Accelerator Center). The magnetic field yields up to more than 3–5 T in amplitude at a duration of only 2–4 ps [39]. This magnetic pulse is among the shortest and most powerful pulses available worldwide [40]. In all spin dynamics calculations, the direction of the magnetic pulse $\mathbf{H}_{\rm pls}(t)$ is perpendicular to the easy axis of the system. The external magnetic field **H** is oriented along the easy axis, i.e., perpendicular to the surface as plotted in the inset in Fig. 2. Dynamical studies of the magnetic moment unit vectors were performed by using the homemade code for numerical solution of the LLG equation, (2). The Runge-Kutta fourth-order method with a time step of $\Delta t = 10^{-18}$ s and a damping parameter $\alpha = 0.01$ has been applied in all calculations. Due to conserving the antiferromagnetic order of the system, we can only consider the magnetization dynamics process of one atom. We took atom 1 in the chain as an example. In Fig. 2, we show a time dependence of the unit vector $\mathbf{S}(t)$ projected onto the easy axis of the system, i.e, $S_z(t)$ for the "edge" and the "middle" configurations.

When the STM tip is positioned over the chain ($\Delta = 5.0$ Å), the critical external magnetic field $\mathbf{H}_{\rm crt}$ to switch the chain is 14 T and it further reduces to 10 T when the tip moves to the middle atom of the chain. The switching time $t_{\rm sw}$ for both cases is about 350 ps. The relatively long time of the switching can be explained by the very small angles between $\mathbf{S}_i$ and **H** after the pulse action. Note that one can switch back chains by applying a magnetic pulse. We are interested in very fast magnetization dynamics (picosecond level), therefore we apply very short magnetic pulses. If the field remains on for a longer time, switching occurs considerably later, after the field has been turned off.

We have revealed that one can significantly reduce the switching time by increasing the amplitude of the magnetic pulse. In Fig. 3, we present the switching of the system magnetization under the action of only $\mathbf{H}_{\rm pls}$ perpendicular to the easy axis of the system for different tip positions and different Δ's. In this case the pulse length $t_{\rm pls}$ and magnitude $H_{\rm pls}$ should be sufficiently larger but the switching time is drastically decreased. In Fig. 3(a), $\Delta = 5.0$ Å, magnetization switching occurs for $H_{\rm pls} = 6$ T and $t_{\rm pls} = 15$ ps (edge) and $H_{\rm pls} = 6$ T and $t_{\rm pls} = 3$ ps (middle). The switching time is 13.5 and 2.5 ps, respectively. For $\Delta = 4.5$ Å, as shown in Fig. 3(b), the switching of the "middle" configuration is realized after $t_{\rm sw} = 16.8$ ps for $H_{\rm pls} = 11$ T, $t_{\rm pls} = 18$ ps. One should note that all atoms switch the magnetization direction at practically the same time (delay ≈ 0.5 ps). The pulses in this setup are rather large. This could be a major challenge for the experimental realization of switching Fe chains. However, real fields from laser radiation are neither rectangular nor single cycle. As noted by Wienholdt *et al.* simulations with more realistic pulses significantly reduce the amplitude of pulses [19]. We also can detect switching of the magnetization of atom 1 in the edge configuration at the same tip-chain distance, but in this case the magnetization of atoms 2 and 3 does not switch.

Finally, we want to comment on the limits of validity of the classical LLG approach to the investigated chains. The experiments by Loth *et al.* [14] showed that chains switch from one Néel state to the opposite Néel state via spontaneous temperature-dependent process. Two regimes of switching have been found: Arrhenius behavior at high temperatures (above 5 K) and low-temperature ($T < 5$ K) behavior consisting of quantum tunneling of magnetization. The study of the thermal stability of Néel states has allowed to Loth *et al.* to determine effective energy barriers and pre-exponential factors. The barriers change from 1.46 up to 8.2 meV, depending on the size and structure of the Fe chains. A recent theoretical study [21] has predicted that barriers strongly depend on the number of atoms in the

chain, changing from 3–4 meV for 2- to 4-atom chains to 16 meV for 10-atom chains. In a very recent study [41], Gauyacq and Lorente arrived at the unexpected conclusion that magnetization evolution in Fe chains is between complete decoherence-induced stability and unobservable fast quantum-tunneling switching. The spontaneous switching rate observed in the experiments is orders of magnitude slower than quantum tunneling. The combined effect of substrate-induced decoherence (which leads to the observation of classical magnetic states) and quantum oscillations has been proposed as a possible scenario of the switching process in chains. Gauyacq and Lorente [41] have found classical and quantum regimes for Fe chains. For example, Fe_6 and Fe_8 chains are found in the classical regime at temperatures used by Loth *et al.* [14]. In contrast, Fe_2 chains at 330 mK in experiments by Bryant *et al.* [12] appear to be in the quantum domain. According to the scenario proposed by Gauyacq and Lorente [41] our system can presumably be treated as classical for temperatures above 1 K (see Fig. 2 in [41]). Additionally, classical behavior can be realized due to strong coupling to other localized spins (in our case, to the magnetic tip), as demonstrated by Delgado *et al.* [42]. Therefore, we believe that application of the classical LLG equation in our work is well justified for classical regimes of Fe chains.

In summary, we have studied the spin dynamics of short AFM chains on the Cu_2N surface. We have found that short magnetic pulses can be used for switching the magnetization of AFM chains. Tuning the magnetization dynamics by vertical and lateral movement of the magnetic STM tip is demonstrated. We have presented a theoretical prediction of information storage in AFM chains using the STM tip and magnetic pulses.

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K.T. and O.P.P. contributed equally to this work.

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