



Spin Friction Observed on the Atomic Scale

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With the advent of scanning probe microscopy techniques that involve a tip and a sample in relative motion in the contact or noncontact regime, the microscopic aspects of friction have become a major branch of research called nanotribology. A significant number of recent studies in this field have concentrated on the distinction between electronic and phononic contributions to friction. Here, we are using the combination of spin-polarized scanning tunneling microscopy and single-atom manipulation in order to move individual magnetic atoms over a magnetic template. By monitoring the spin-resolved manipulation traces and comparing them with results of Monte Carlo simulations, we are able to reveal the characteristic friction force variations resulting from the occurrence of spin friction on the atomic scale.

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Friction is one of the oldest phenomena known and utilized by mankind. Frictional heating was already used by our prehistorical ancestors more than 400 000 years ago for the lighting of fires. While the force of friction had already been recognized and studied by Aristotle, it was Leonardo da Vinci who performed the first quantitative studies of interacting surfaces in relative motion, thereby pioneering the field of tribology [1]. Related phenomena such as triboluminescence, i.e., the emission of light when a material is stressed to the point of fracture [2], or tribochemistry, i.e., the acceleration of reaction rates by friction [3], have also been known since the 16th century. The first two laws of (macroscopic) friction, generally known as Amontons' laws, governed the field of tribology for a long time. Recently, advanced experimental methods, such as the quartz-crystal microbalance, the surface forces apparatus, or friction force microscopy, have allowed for deeper insight into the microscopic origin of frictional phenomena [4]. For instance, by making use of the transition between normal and superconducting states, it has been demonstrated that electronic friction is the main dissipative channel in the metallic state, while phononic friction dominates in the superconducting state [5–8]. Since electronic degrees of freedom govern frictional phenomena for normal metals, it is natural to consider a possible spin-dependence of frictional forces in the case of magnetic materials [9,10]. Because of the fact that the experiments used previously to measure friction-related forces on the atomic scale [11] cannot resolve spin structures, the experimental method of choice is then spin-polarized scanning tunneling microscopy (SP-STM) [12,13] involving a magnetic tip and a magnetic sample. By operating under extreme conditions, i.e., tiny tunnel gap resistances [14], atomic manipulation experiments have recently become feasible while still being sensitive to the spin degree of freedom [15–17]. Here, we present a combined experimental and theoretical study of frictional phenomena occurring when a single

magnetic atom is moved over a magnetic surface by means of an SP-STM tip, thereby revealing the importance of the spin degree of freedom in frictional phenomena.

Our experiments were carried out under ultrahigh vacuum conditions at a temperature of 8 K, using a home-built SP-STM setup [18]. As substrate we chose a Mn monolayer on W(110) [19], which exhibits a spin spiral with roughly 170° between adjacent atomic rows as visualized in Fig. 1(a). Figure 1(b) shows a spin-resolved constant-current STM image of this substrate obtained with a mag-

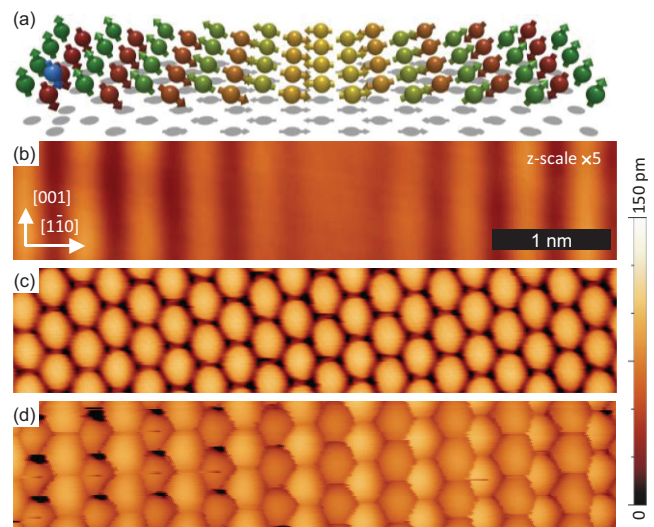


FIG. 1 (color). Magnetic template and STM measurements. (a) Visualization of the Mn/W(110) spin spiral. (b) SP-STM image of Mn/W(110). (c) and (d) Manipulation images of a Co adatom moved across the substrate (c) exhibiting the symmetry of the atomic lattice and (d) reflecting the magnetic order of the underlying spin spiral, obtained with a spin-polarized tip. (measurement parameters: (b) $I = 80$ nA, $U = -8$ mV, (c) $I = 50$ nA, $U = -5$ mV, (d) $I = 80$ nA, $U = -5$ mV).

netic tip. Bright and dark lines indicate atomic rows with parallel and antiparallel magnetization components relative to the tip magnetization. The magnetic signal vanishes for Mn atoms that have a magnetization direction perpendicular to the tip magnetization due to the cosine dependence of the spin-polarized part of the tunnel current on the angle between tip and sample magnetization [13].

To investigate the influence of the spin degree of freedom on friction we have moved single Co atoms over this surface. From a previous study [15] it is known that Co adatoms on Mn/W(110) couple ferromagnetically to the nearest Mn atoms and do not alter the spin spiral state. When small tip-adatom distances are realized, the Co adatoms can be moved by utilizing the attractive force of the tip. If an adatom is trapped in the tip potential and follows the tip during scanning, the constant-current images change drastically as shown in Fig. 1(c). We refer to this kind of image as a manipulation image, similar to the experiments reported by Strosio *et al.* [20]. It is important to note that the manipulation images do not represent the Mn atoms of the surface, but rather show the same Co adatom on different adsorption sites of the Mn monolayer while it is following the scanning tip. When a spin-polarized tip is used [Fig. 1(d)], we observe a superstruc-

ture which is intimately tied to the magnetic order of our substrate.

To study the role of the spin degree of freedom in the manipulation processes of a magnetic atom, we performed Monte Carlo simulations (see Supplemental Material [21], S1 and S2 for details). We approximate the spin spiral state by a row-wise antiferromagnetic order, as illustrated in Figs. 2(a) and 2(b), to reduce complexity. The surface Mn atoms, as well as the SP-STM tip, are treated as localized magnetic moments with a fixed spin direction. One layer of W atoms with their spins set to zero is included below the Mn layer. A magnetic adatom is placed on the substrate and its interaction with the surface atoms as well as the tip is modeled equivalently: To describe the chemical potentials we use Morse potentials with equilibrium binding energies U_{Surf} and U_{Tip} , and to model the magnetic interaction we consider pairwise Heisenberg exchange with exponential decay and coupling constants J_{Surf} and J_{Tip} . U_{Surf} keeps the adatom within the preferred adsorption sites [Fig. 2(a)], and $J_{\text{Surf}} = 75$ meV per Mn atom at nearest neighbor distance leads to an effective ferromagnetic coupling to the two nearest neighbors. The tip is scanned over the sample at constant height. After every increment of the tip movement, the adatom position

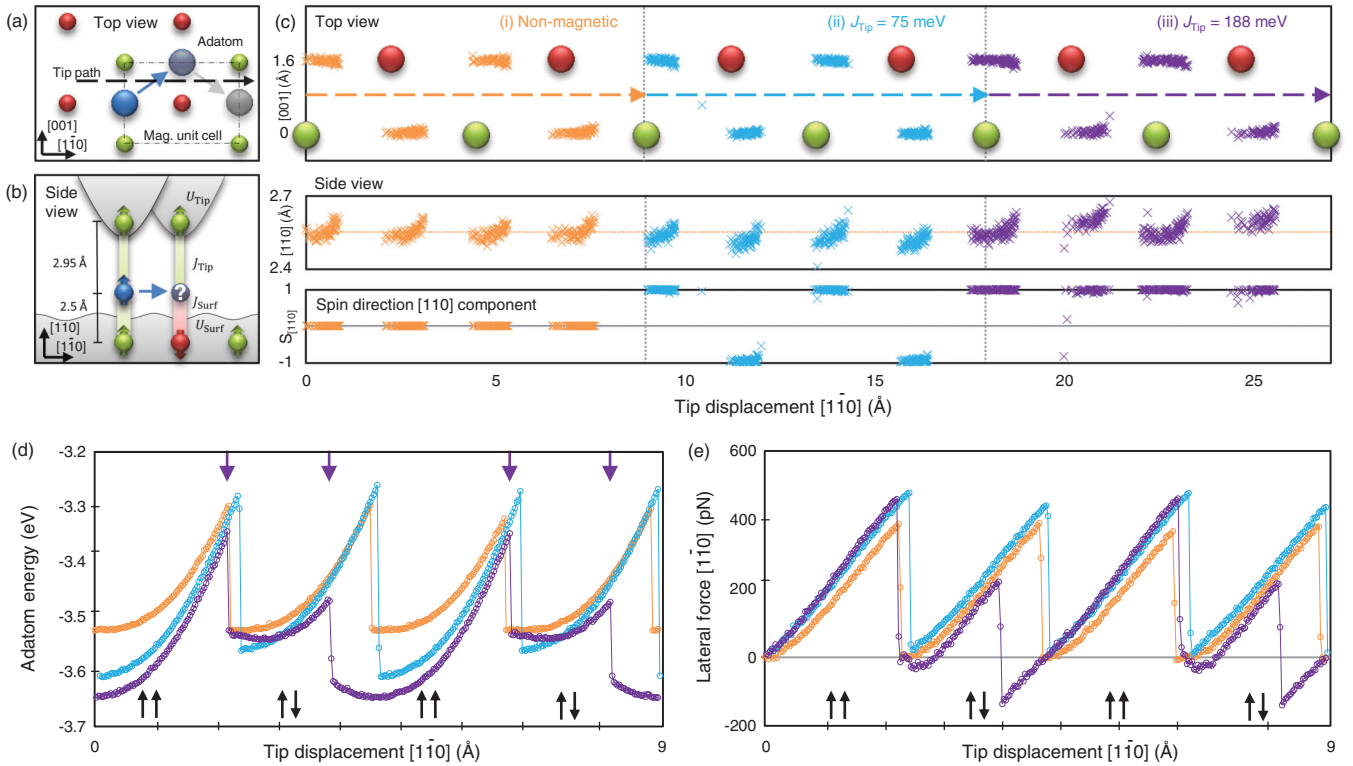


FIG. 2 (color). Setup and results of the Monte Carlo simulations. (a) Top view of the equilibrium adsorption sites of the Co adatom (blue). (b) Side view of the Monte Carlo simulation setup. (c) Top view (dashed arrows indicate tip path), side view of the adatom position (dotted orange line indicates average adatom height of the nonmagnetic case), and $[110]$ component (up or down) of the adatom spin during manipulation, for different strengths of the tip-adatom exchange interaction J_{Tip} , sampled at constant time intervals. (d) Adatom energy and (e) lateral force as a function of tip position, for the three cases presented in (c). The black arrows indicate the spin configuration of tip and adsorption site.

$\mathbf{r}_a$ and spin $\mathbf{S}_a$ are relaxed to the local energy minimum within the combined surface and tip energy landscape by applying the Metropolis algorithm at the experimental temperature of $T = 8$ K. While small changes of the chemical potentials and J_{Surf} do not alter the manipulation process dramatically, we found that the strength of the tip-adatom exchange interaction J_{Tip} has a considerable influence, which we will investigate in the following, keeping all other parameters fixed or set to zero.

First we will focus on the most symmetric manipulation trace as sketched by the dashed arrows in Fig. 2(c): When the tip moves along the $[1\bar{1}0]$ direction, the adatom moves within its current adsorption site due to the attractive tip potential, until it overcomes the diffusion barrier and jumps to the next site. The data points in Fig. 2(c) reflect the lateral and the vertical position as well as the spin of the adatom for three different scenarios sampled at constant time intervals during the manipulation. The three cases are: First a nonmagnetic adatom (orange), second a magnetic adatom with weak exchange interaction with the tip (blue), and third a magnetic adatom with strong exchange interaction with the tip (purple). In the first two cases the movement within the adsorption sites is very similar. The spin of the magnetic adatom in the second case flips at every jump to stay aligned with the spin of the nearest surface atoms. In the third case, the exchange coupling to the tip dominates and the spin of the adatom stays mostly aligned with that of the tip. While the behavior of the adatom is comparable to the first two cases on the parallel sites ($\uparrow\uparrow$, where the nearest neighbor surface atoms are parallel to the tip magnetization), it changes qualitatively for the antiparallel sites ($\uparrow\downarrow$). Here, the mobility of the adatom is increased due to a slightly lifted equilibrium position [cf. side view in Fig. 2(c)] and a shallower potential well caused by the magnetic frustration.

It is instructive to compare the energy of the adatom E_{Adatom} as a function of tip displacement x_{Tip} along $[1\bar{1}0]$ [Fig. 2(d)] for the three cases above. The manipulation mechanism (pulling-mode) is evident in the general shape of the curves: As long as the adatom stays within one site, its energy increases due to the progressing movement of the tip, until it eventually overcomes the diffusion barrier to the next site. The adatom jump is accompanied by the dissipation of energy (sharp drops in the energy curves). In the first case (orange), the process is independent of the magnetism of the adsorption site. However, in the two magnetic cases, the exchange coupling to the magnetic tip lifts the energetic degeneracy of adjacent sites due to opposing spin directions. For weak magnetic coupling to the tip (blue), the main effect is an alternation of the energy minima, which is accompanied by an alternation of the dissipated energy. The adatom also jumps earlier from the antiparallel to the parallel site than vice versa. These three effects are more pronounced in the strong exchange cou-

pling regime (purple), where the adatom spin stays mostly parallel to the tip's spin direction on all sites.

Figure 2(e) shows the lateral force $F_x = dE_{\text{Adatom}}/dx_{\text{Tip}}$ exerted by the tip onto the adatom, derived from the data in Fig. 2(d). The calculated threshold forces needed to move the atom are of the same order of magnitude (10^{-10} N) as measured experimentally for Co adatoms on Pt(111) [11]. In our case, they develop an increasing site dependence with rising J_{Tip} : In the third case (strong adatom-tip magnetic exchange, purple) only about half the force is necessary to move the adatom from the antiparallel to the parallel site than vice versa. We conclude that the spin degree of freedom has several implications for the manipulation process: It modifies the amount of dissipated energy, the threshold force needed to move the magnetic adatom, and the tip position at which the jump to the next site occurs.

For a direct comparison with the experimental SP-STM images, we have calculated the spin-polarized tunnel current for every lateral position of the tip. This is done using the spin-polarized extension of the Tersoff-Hamann model [22] with the polarization of the magnetic atoms and tip set to $P = 0.35$. The resulting simulated constant-height images (Fig. 3 left) can be compared qualitatively to the experimental constant-current images (Fig. 3 right) due to the known exponential relation between tip height and tunnel current. The simulated manipulation image for a nonmagnetic tip ($J_{\text{Tip}} = 0$ and $P_{\text{Tip}} = 0$) in Fig. 3(a) shows the adatom in each adsorption site. Abrupt transitions correspond to jumps between regions of stability; here, the size of these cells (cf. whitened area) is identical to the chemical unit cell. The line profile taken along the symmetric manipulation path (white arrow) shows a pulling-mode manipulation trace [14]. As expected, the characteristics of the current are identical for each adsorption site since the nonmagnetic tip causes no energetic asymmetry or spin-dependent current contribution. A comparable experimental image and line profile is displayed in Fig. 3(b): The round shape of the adatom indicates a pulling-mode in which the adatom jumps in front of the tip followed by a scan across its center.

A situation where the adatom's spin has an effect on the manipulation is shown in Fig. 3(c): The simulated manipulation image for a spin-polarized tip with $J_{\text{Tip}} = 188$ meV (cf. also Fig. 4, purple) shows alternating intensities as a result of spin-dependent tunneling. While the variation in dissipated energy and threshold forces cannot directly be extracted from such an image, the position of the tip at which the adatom jumps to the next site can be analyzed. As indicated by the blue arrows in the line profile, every jump from $\uparrow\downarrow$ to $\uparrow\uparrow$ occurs earlier. This is also reflected in the image by the alternating cell size. Qualitatively the same behavior is observed in the experimental data in Fig. 3(d): It displays an alternating height difference due to spin-polarized tunneling and an alternating cell size. The latter

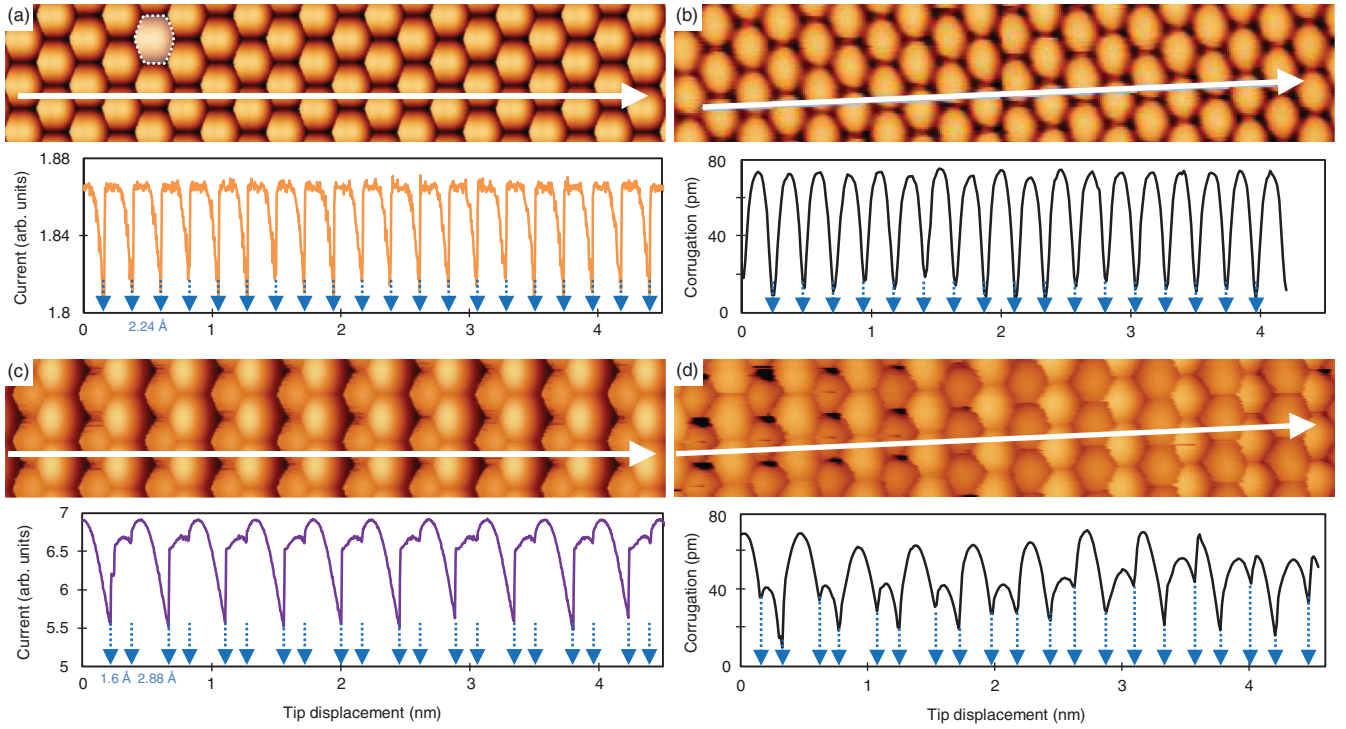


FIG. 3 (color). Comparison of simulation and experiment. (a) Simulated manipulation image for a nonmagnetic tip (tip height 6 Å). The discontinuities mark the positions at which the adatom jumps to the next site. (b) Experimental manipulation image comparable to (a). (c) Simulated manipulation image (spin-polarized tip with $J_{\text{Tip}} = 188$ meV and tip height 5.45 Å). (d) Experimental manipulation image comparable to (c). All line profiles are taken along the symmetric manipulation path (see also Supplemental Material [21] S3). The distance variation between consecutive adatom jumps in (c) and (d) reflects the occurrence of spin friction.

is especially pronounced on the left side of the image, fades out towards the middle and returns on the right side. This gradual transition is connected to the spin spiral order of the Mn layer, opposed to the collinear antiferromagnetic state in the simulation, and reveals an angular dependence of the spin-dependent friction forces.

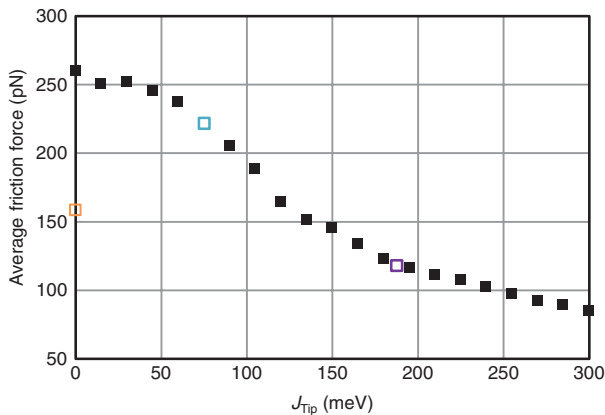


FIG. 4 (color). Spin-dependent average friction force. Average friction force $\langle F_x \rangle$ of a magnetic adatom on a row-wise antiferromagnetic surface [cf. Fig. 2(a)] in dependence of the tip-adatom exchange interaction strength J_{Tip} . The hollow data points correspond to the cases discussed in Figs. 2(c)–2(e).

It is surprising that the spin degree of freedom can play such a significant role in atom manipulation processes, given the orders of magnitude of difference between chemical and magnetic coupling energies. The reason is that the exchange energy does not have to compete with the adatom's total chemical binding energy (i.e. the energy necessary to completely remove the adatom from the surface), but only with the energy barriers between adjacent adsorption sites, which can be of similar magnitude as the exchange interaction, especially in manipulation experiments, in which the tip is utilized to reduce the natural diffusion barriers of the surface. We expect spin friction to be a general phenomenon occurring in a large class of systems.

Figure 4 shows the average friction force as a function of J_{Tip} for the antiferromagnetic system used in the simulations. The average friction force $\langle F_x \rangle$ can be calculated in a straightforward fashion from the lateral forces in Fig. 2(e), $\langle F_x \rangle = \frac{1}{np} \int_0^{np} F_x(x) dx$, or, alternatively, from the energy dissipated at the adatom jumps, $\langle F_x \rangle = E_{\text{Diss}}/np$, where E_{Diss} is the total energy loss over the interval np , which is a multiple of the distance p between two equivalent tip positions [i.e., several periods of the curves in Figs. 2(d) and 2(e)]. This means friction is intimately linked with dissipation losses: the energy pumped into the system by the tip movement dissipates

completely through atom jumps within one cycle. Changing the adatom from nonmagnetic (orange square in Fig. 4) to magnetic for $J_{\text{Tip}} = 0$ leads to an increase of roughly 60% in the average friction force for our simulation parameters. While the chemical contribution is identical for both cases, the magnetic adatom has to flip its spin at every jump, causing an additional dissipation of exchange energy and increasing the total friction by this magnetic contribution. For a magnetic adatom, the average friction force decreases with rising J_{Tip} . This is caused at first by the growing magnetic frustration, until the spin direction of the adatom stays aligned with that of the tip, practically reducing the degrees of freedom available to the adatom (at roughly $J_{\text{Tip}} = 180$ meV). For larger values of J_{Tip} , the equilibrium position of the adatom gets shifted towards the tip, which in turn reduces the strength of the interactions between adatom and surface and leads to the slow continuous friction decrease that follows.

Our experimental findings combined with our Monte Carlo simulations clearly indicate that not only electronic and phononic contributions to frictional phenomena must be taken into account, but also the spin degree of freedom in the case of magnetic atoms in relative motion. Moreover, our study of spin friction on the atomic scale indicates the importance of the spin degree of freedom in related surface phenomena, such as the diffusion of magnetic adatoms on magnetic substrates.

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