Experimental Evidence for Intra-Atomic Noncollinear Magnetism at Thin Film Probe Tips

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The energy-dependent spin-density orientation (SDO) at the apex of thin magnetic film tips is studied by spin-polarized scanning tunneling spectroscopy (SP-STS) at different bias voltages. At most energies the SDO is collinear with the tip magnetization resulting in a domain or domain-wall contrast in SP-STS images of out-of-plane magnetized samples measured with Gd or Fe coated tips, respectively. For some bias voltages, however, the SDO of the tip is found to be almost perpendicular to its magnetization. This result is explained in terms of intra-atomic noncollinear magnetism.

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In recent years noncollinear magnetism (NCM) has attracted considerable interest. NCM can be classified in two different types, namely, inter- and intra-atomic NCM. Interatomic NCM can be described within the well-known Heisenberg model [1]. Although in this model a local quantization axis is defined by an atomic magnetic moment, the moments of different atoms may be noncollinear [2]. In contrast, an unconstrained approach has already been considered in the original publication on spin-polarized density functional theory [3]. This concept treats the spin density as a vector field which, if integrated over the whole magnetic volume, gives the global magnetization, but which also allows that its orientation "varies on the length scale of an atom" either by "different quantization axes for different orbitals on the same atom … or by nontrivial spin mixture produced by the spin-orbit coupling" [4]. While the existence of interatomic NCM is well established we are not aware of any direct experimental evidence of intra-atomic NCM.

In this Letter we report on the investigation of the energy-dependent spin-density orientation (SDO) of magnetic thin film tips. This is accomplished by a detailed analysis of the magnetic contrast in spin-polarized scanning tunneling spectroscopy (SP-STS) images measured on perpendicularly magnetized Fe nanowires at different bias voltages. At most bias voltages Gd tips show a *domain* contrast while the use of Fe coated tips results in a *domain-wall* contrast indicating out-of-plane and in-plane SDO of the tip, respectively. These results can be understood on the basis of the global magnetization of the films as determined by their anisotropy. Surprisingly, we found that at some values of the applied bias voltage the SDO at the tip apex is almost perpendicular to the tip's global magnetization direction. This is interpreted in terms of tunneling from different orbitals of the probing atom at the tip apex which, as a result of intra-atomic noncollinear magnetism, exhibit different SDOs.

The experiments have been performed in an ultrahigh vacuum system with separate chambers for substrate preparation, sample transfer, metal vapor deposition, surface analysis, and cryogenic scanning tunneling microscopy (STM) equipped with a 2.5 T magnet [5]. The W(110) substrate preparation has been described in Ref. [6]. We used etched polycrystalline W tips which were flashed *in vacuo* at $T > 2200$ K to remove oxide layers. As revealed by scanning electron microscopy this results in a blunt tip with a diameter $D \approx 1\ \mu$m. The tips were magnetically coated with 7 ± 1 monolayers (ML) Gd or 5 ± 1 ML Fe while held at 300 K, subsequently annealed at $T \approx 550$ K for 4 min, and then transferred into the cryogenic STM. During the measurements, tip and sample were at a temperature $T = 15$ K.

Figure 1(a) schematically illustrates the structural and magnetic properties of the sample used in this study, i.e., 1.5 ML Fe epitaxially grown on a stepped W(110) substrate. As a result of the final annealing treatment which leads to step-flow growth, the Fe overlayer forms a self-organized grating of stripes with a thickness of alternating one and two ML. The easy magnetization direction of the Fe double-layer (DL) nanowires is out of plane. As originally found by spatially averaging Kerr-effect measurements [8,9] and recently confirmed by SP-STS [7] adjacent DL nanowires prefer an antiferromagnetic arrangement which is driven by the energy gain from reducing the stray field [see Fig. 1(a)]. Frequently, domain walls were observed by high resolution SP-STS imaging [7] which are oriented along the $[1\bar{1}0]$ direction and which exhibit a wall width of 6 ± 1 nm. For domain walls in ultrathin out-of-plane magnetized films it is energetically favorable to rotate the magnetization vector parallel to the wall plane (Bloch-type) thereby avoiding magnetic charges. In this particular case a Bloch-type wall is also supported by the anisotropy of the adjacent Fe monolayer which is magnetized in-plane along the $[1\bar{1}0]$ direction, too. Since, however, the magnetization may rotate either clockwise or counterclockwise the local magnetization in the center of the wall can point into either the $[1\bar{1}0]$ or the opposite, i.e., the $[\bar{1}10]$ direction.

As already discussed in previous publications [7,10,11] the spin-valve effect leads to a differential tunneling

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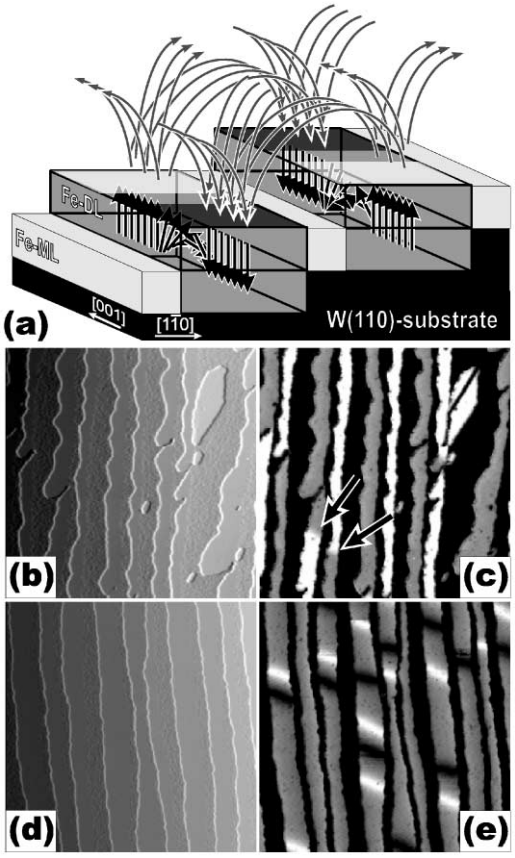

FIG. 1. (a) Schematic domain structure of 2 ML thick Fe nanowires grown on W(110). The easy magnetization axis points along the surface normal. Because of dipolar coupling between adjacent nanowires the magnetization direction periodically alternates. The domain walls are Bloch walls. Topography [(b) and (d)] and dI/dU signal [(c) and (e)] of Fe nanowires as measured with Gd and Fe coated probe tips, respectively (scan range: 200 nm $\times$ 200 nm). Because of its low differential conductivity dI/dU the Fe ML appears black at $U = +0.7$ V (cf. Ref. [7]). Two discrete levels of the dI/dU signal of the Fe DL can be recognized in the measurement performed with a Gd coated probe tip. These areas correspond to *domains* with the magnetization vector pointing up or down. In contrast, the in-plane magnetization within the *domain walls* is imaged by using an Fe tip.

conductivity dI/dU between the magnetically coated probe tip and the magnetic sample which depends on their relative SDO: while a parallel SDO leads to an enhanced dI/dU signal it is reduced in the case of an antiparallel orientation. It was theoretically proposed by Slonczewski [12] and experimentally confirmed by Miyazaki and Tezuka [13] that the spin-valve effect scales with $\cos\theta$ (from a global viewpoint θ is the angle between the magnetization vectors of tip and sample). Consequently, no effect is observed in the case of an orthogonal SDO between tip and sample, i.e., $\cos(\theta) = 0$. In the STM the spin-valve effect can be utilized to measure the sample's magnetic domain structure, i.e., the spatial distribution of the surface SDO, at high lateral resolution down to the atomic scale [7,10,11].

Before we come to the main point of this Letter, i.e., the intra-atomic NCM of magnetic thin film tips, we describe the *normal* properties of coated STM tips which are found at most bias voltages and which—to our interpretation—can be understood on the basis of collinear magnetism [14]. Figures 1(b)-1(e) show the topography [(b) and (d)] and the dI/dU signal ($U = +0.7$ V) [(c) and (e)] of Fe DL nanowires as measured with a Gd and Fe coated probe tip, respectively. The scan range of the images is 200 nm $\times$ 200 nm. Since at this particular bias voltage the differential conductivity of the Fe monolayer is much smaller than that of the Fe DL nanowires the former appears black in both dI/dU maps [7]. The dI/dU map as measured with a Gd coated tip [Fig. 1(c)] shows that the intensity of the dI/dU signal alternates between adjacent Fe DL nanowires. This observation is consistent with the proposed antiferromagnetic coupling [8,9]. As marked by arrows in Fig. 1(c) some of the Fe DL nanowires change the dI/dU intensity indicating the presence of domain walls [7]. Obviously, Gd coated probe tips are sensitive to the out-of-plane component of the magnetization thereby providing an image of the *domain* structure. In contrast, the data of Fig. 1(e) which have been measured with a Fe coated tip reveal a sensitivity to the in-plane component of the magnetization. This is indicated by a vanishing domain contrast due to the orthogonal magnetization between tip (in plane) and sample (out of plane). Within the domain walls, however, the magnetization locally possesses an in-plane orientation. As stated above, in the center of the domain wall the magnetization vector may point along either the $[1\bar{1}0]$ or the $[\bar{1}10]$ direction. Although we cannot control the azimuthal orientation of the tip magnetization it is clear that as long as the projection of the tip magnetization vector onto the $[1\bar{1}0]$ direction is nonvanishing we will be able to distinguish both cases due to the spin-valve effect leading to two different dI/dU signals. Indeed, numerous domain walls showing up as black or white lines along the $[1\bar{1}0]$ direction can be recognized in the dI/dU map of Fig. 1(e). Black and white domain walls were found to alternate within each particular nanowire.

On the first view, the strong dependence of the tip magnetization on the coating material might be surprising since the elongated shape of the tip should always lead to some shape anisotropy such that the magnetization vector—independent of the particular coating material—should always point along the tip axis, i.e., out of plane. However, we emphasize that compared to the thickness of the magnetic coating layer which typically amounts to a few atomic layers only, the tips are extremely blunt ($D \approx 1$ μm). This leads to an almost vanishing curvature at the very end of the tip which makes the contribution from the tip shape anisotropy very small. Instead, the results suggest that the anisotropy of thin film tips is dominated by surface and interface terms as can be illustrated by comparing the properties of thin film tips with equivalent magnetic films

deposited on a flat W(110) surface. This dense-packed W surface is most likely created at the tip apex after melting the tip during the flash. While it has been proposed that thin Gd films ($\Theta \leq 9$ ML) on W(110) exhibit a low-temperature phase with a perpendicular easy axis [15] it is well known that 5 ML Fe/W(110) are magnetized in plane [16] being consistent with the different sensitivities as found for thin film tips.

This general rule is, however, not always valid. For example, Fig. 2 shows (a) the topography and the dI/dU signal at (b) $U = -0.1$ V and (c) $U = +0.1$ V which have been *simultaneously measured* line by line in the forward and the backward scan direction, respectively, using an Fe coated probe tip. Again, Fig. 2(b) exhibits a pure domain wall contrast as expected from the in-plane sensitive Fe coated tip [cf. Fig. 1(e)]. In contrast, the dI/dU map of Fig. 2(c) is clearly dominated by a domain contrast, i.e., an out-of-plane spin contrast. In order to allow a more detailed analysis we have drawn line sections between the arrows along one particular nanowire in Figs. 2(b) and 2(c). On the basis of more dI/dU images (not shown here) which were imaged at the *same* location of the sample using the *same* tip and the *same* tunneling current $I = 0.3$ nA we have also analyzed equivalent line sections for many different bias voltages. The results are plotted in Fig. 3. Along the line section four domain walls can be found which, at in-plane spin contrast, show up as peaks or dips in the dI/dU signal corresponding to a parallel or antiparallel orientation of the sample spins with respect to the tip. This *normal* situation is observed at $U \leq 0$ mV and at $U \geq +200$ mV. Under these conditions no offset from the average dI/dU signal could be found above different domains indicating that we have no sensitivity to the out-of-plane component of the sample magnetization. At small positive sample bias, however, a significant offset is found. Although some contrast from the domain walls remains always visible an appropriate choice of the bias voltage ($U \approx +200$ mV) leads to a strongly dominating domain contrast [17]. In the following we discuss these observations which cannot be understood on the basis of simple collinear magnetism. We consider four different effects to explain the experimental facts at least qualitatively, namely, electric dipole forces, current-induced magnetic torque [18], coherent spin rotation at the interface [19], and intra-atomic noncollinear magnetism [4].

The apex of the tip is probably formed by a small cluster of magnetic material which protrudes from the spherical end of the tip. To our experience it is sufficiently sharp in order to routinely image step edges on the sample surface as long as their heights do not exceed several atomic layers. First, let us assume that the cluster is collinearly magnetized in equilibrium, i.e., with no electric field present within the vacuum gap. Since the magnetic anisotropy is only a small correction to the total energy of the system, it is very sensitive even to minor structural changes. Hence, we can speculate that the application of an electric field may lead to a slight change of the interlayer distance between the apex atom and the first subsurface layer at the tip. This may result in a field dependence of the anisotropy of the last atom of the tip. We believe, however, that this explanation is very unlikely mainly because of the small bias voltage value at which the effect is observed and because it vanishes again at higher voltages.

If a current-induced magnetic torque $\vec{L}$ would be responsible for the observations, the effect should increase linearly with the current density j [18]. Our results indicate, however, that the SDO of the tip is independent of the tunneling current. Coherent spin rotation is caused by the precession of the electron spin around the magnetic field [19]. In our case it might occur in the vacuum gap between the sample and the tip. Since, however, the angle of precession $\epsilon \propto E^{-1/2}$, it should be largest for a small bias voltage which is in contrast to our observations. In particular, for Gd coated tips the effect was largest at a relatively large bias voltage $U = -700$ mV. Therefore, we can exclude that either a current-induced magnetic torque or coherent spin rotation play an important role.

The most likely explanation for our observation is found in the concept of intra-atomic noncollinear magnetism. We probably observe tunneling from different tip orbitals, e.g., e_g- and t_{2g}-like d orbitals as has already been suggested in Ref. [2]. Under *normal* conditions, i.e., for negative and high positive sample bias, the tunneling current flows through orbitals which have a SDO being collinear with the magnetization direction, i.e., in plane for an Fe coated tip. At small positive sample bias, however, another occupied orbital of the tip with a SDO being almost orthogonal to

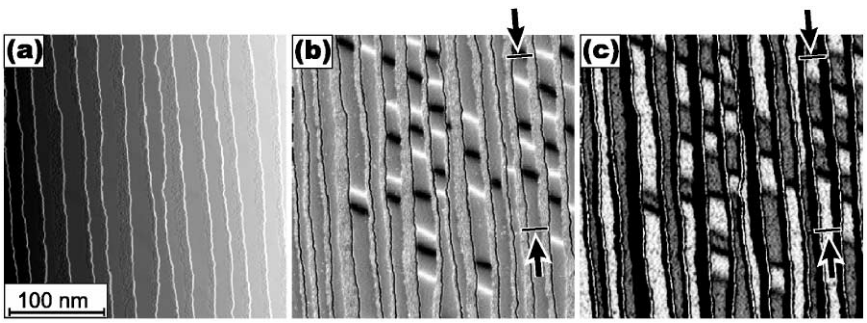

FIG. 2. (a) Topography and dI/dU signal of Fe nanowires as simultaneously measured at (b) $U = -0.1$ V and (c) $U = +0.1$ V with an Fe coated probe tip. While the dI/dU image at negative sample bias shows a pure domain wall contrast, the dI/dU image measured at positive bias exhibits a significant out-of-plane contribution to the signal which results in a clear domain contrast. Averaged line sections drawn between the arrows indicated in (b) and (c) are shown in Fig. 3.

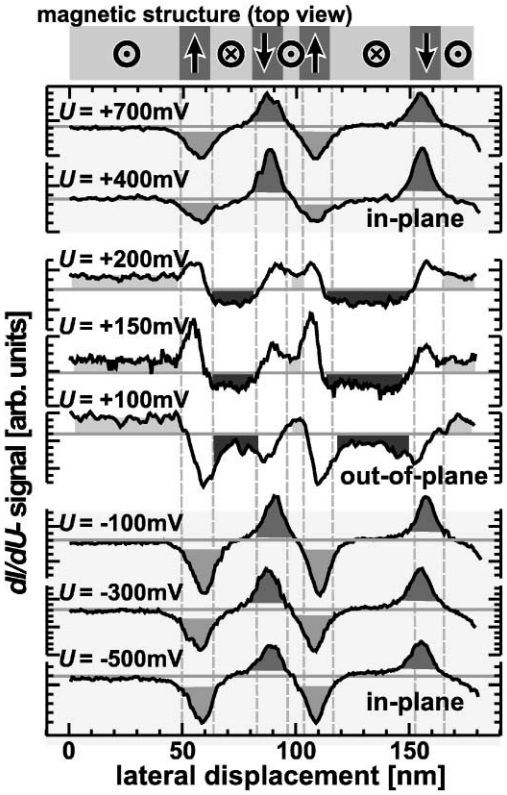

FIG. 3. Averaged line sections of the dI/dU signal as drawn between the arrows indicated in Figs. 2(b) and 2(c). A pure in-plane contrast in the magnetic dI/dU signal is found for $U < 0$ V and $U > +0.2$ V. In contrast, some out-of-plane contribution appears at $0\ \text{V} < U < +0.2$ V. All data were measured with the same tunneling current $I = 0.3$ nA.

the global tip magnetization probes the sample's density of states leading to an out-of-plane contrast. Besides the possibility that different orbitals of the probing atom at the tip apex may exhibit different SDOs [2] another mechanism which also produces intra-atomic NCM has recently been described by Nordström and Singh [4]. Since the physical origin of this mechanism is, however, spin-orbit coupling which is very small for transition metals like Fe it is very improbable that our observations are caused by this effect. In fact, it has been calculated for fcc Pu, a material with "substantial" spin-orbit coupling, that the maximal deviation of the local SDO from the global magnetization direction amounts to 5° [4]. Although symmetry breaking — a situation which is certainly fulfilled at the tip apex — may lead "to a further tipping" of the spin orientation [4] we do not believe that the resulting effect in an Fe tip can account for the observations.

In summary, we have studied the SDO of magnetic thin film tips by imaging the domain structure of Fe nano-wires. It is found that the *normal* SDO being collinear to the magnetization is determined by the anisotropy of the thin magnetic film which is dominated by surface and interface terms. This results in an out-of-plane and in-plane sensitivity for Gd and Fe coated probe tips, respectively. We could show that by choosing an appropriate bias voltage the SDO can point into a direction which is almost perpendicular to the easy magnetization axis of the tip. As an important implication this effect allows the simultaneous imaging of the domain and the domain-wall structure of the sample with a single probe tip.

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