Spin-polarized Scanning Tunneling Microscopy

In scanning tunneling microscopy (STM; see Scanning Tunneling Microscopy), the electron charge is used as information carrier to image the topography of a sample surface. The small tunneling current between the tip and the conductive surface is used as a feedback parameter to move the tip on lines of constant tunneling current. In first approximation, these lines correspond to lines of constant charge density of the sample surface probed by the tip apex (Tersoff and Hamann 1983). For ferromagnetic or antiferromagnetic materials, the charge density is spin split into majority and minority states. A net-spin polarization is present in the atoms and the individual atoms carry a magnetic moment. In spin-polarized STM (Sp-STM), the spin of the tunneling electrons is used as an additional information carrier to map the spin polarization of the sample surface. This way, Sp-STM allows one to image both the topography and the magnetic structure of a surface with nanometerlateral resolution. Due to the strong localization of the tip-sample interaction, the resolution is superior to magnetic force microscopy (MFM; see Magnetic Force Microscopy), which uses the long-range magnetic dipole interaction.

1. Fundamental Contrast Mechanism

When imaging ferromagnetic surfaces with STM, the tunneling current is spin-polarized as a consequence of the sample spin-polarization. This alone is not enough to obtain spin information in STM, as the spin-polarization of the tunneling current is not directly accessible. The only measured parameter is the size of the current. If, however, the tip is spin-polarized as well, the size of the tunneling current is influenced. The pioneering experiments of Jullière (1975) showed that when electrons tunnel between two ferromagnets, the size of the current depends on the relative orientation of the magnetization of both electrodes. For parallel orientation, the tunneling current I is usually higher than for antiparallel orientation. This effect is also called the tunneling magnetoresistance (TMR) effect. This finding can be explained on the basis of a simple model for tunneling. It is reasonable to assume absence of spin-flip scattering during the tunneling process itself so that the spin of the electron is conserved during tunneling. If one further neglects any spin-dependence in the transmission through the barrier and focuses solely on the electronic properties of the two electrodes, the tunneling current I is proportional to the density of states in both electrodes. For parallel orientation, the majority/minority electrons of the first electrode tunnel into the majority/minority states in the second electrode respectively, as sketched in Fig. 1(a). Using Fermi's golden rule, for each spin channel the tunneling probability is proportional to the density N of tip (t) and sample (s) states at the Fermi edge. The tunneling current I is proportional to the sum of both channels. For antiparallel orientation, electrons of the majority character in one electrode tunnel into states of the minority character in the other electrode and I is given by the sum of the mixed products (see Fig. 1(b)). These two currents are, in general, not identical leading to a variation of I with the magnetization direction of the electrodes. Slonczewski treated the problem more rigorously (Slonczewski 1989) and calculated the dependence of I on the angle θ between the magnetization of the two electrodes. With the spin polarization $P = (N_{\uparrow} - N_{\perp})/(N_{\uparrow} +$ N_{\perp}), the current is given by

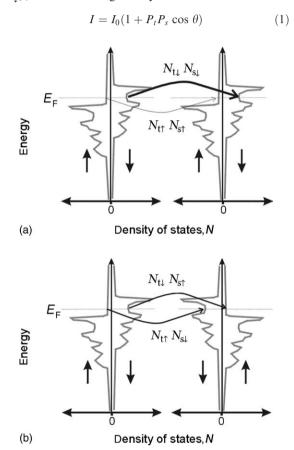


Figure 1 Tunneling between two ferromagnetic electrodes that show a spin-split density of state N. \uparrow/\downarrow denote majority/minority states. In (a) and (b), the magnetization of the two electrodes is parallel and antiparallel, respectively. The partial currents for tunneling from the left to the right electrode are indicated.

In Sp-STM, the TMR effect is used to obtain spin information via changes of the tunneling current. This way, the same high lateral resolution for topography and spin can be obtained.

2. Imaging Modes of Sp-STM

There are four principal imaging modes proposed by Pierce *et al.* (1988). The modes differ in the way the spin contrast is obtained and how it is separated from the topographic contrast of STM. All modes have been experimentally realized and are discussed below.

2.1 Optically Pumped Semiconductor Tips

An elegant way to spin-polarize the tip is to use GaAs tips and to photo-excite carriers into the conduction band using circularly polarized light (Pierce and Meier 1976). In the second step, the excited carriers tunnel from the conduction band of the tip into the sample. The spin-polarization of the electrons can be selected by the helicity of the light. By modulating the helicity, modulations in the tunneling current are induced due to spin-dependent tunneling. The modulations can be detected with a lock-in amplifier to separate spin information from topographic information (Suzuki et al. 1997). The optical modulation technique, however, suffers from a rather low contrast due to a minute net-polarization of the GaAs and an unintended magneto-optical contrast of low lateral resolution due to light-sample interaction. Very few studies on domain patterns in ferromagnetic films have been published using this technique.

2.2 Constant Current Mode

The constant current imaging mode is the simplest mode. In this mode, a spin-polarized tip is scanned over a surface in the conventional constant current mode of STM. Wiesendanger et al. (1990) were the first to use this imaging mode. They reported on spinpolarized vacuum tunneling between a ferromagnetic CrO₂ tip and the (100) surface of layer-wise antiferromagnetic b.c.c. Cr. Using a non-magnetic tip, topographic line-scans revealed atomic steps of the expected step height of 0.14 nm. When, however, the experiment was repeated with ferromagnetic tips, alternating step heights of 0.16 nm and 0.12 nm were observed. This was attributed to the TMR effect between the tip and the Cr surface. When the spin-polarizations of the tip and the Cr are parallel, the tunneling current is enhanced (see Eqn. (1)). In the constant current mode of the STM, the tip is retracted by a small amount (0.02 nm). Due to the antiferromagnetic order of Cr(001) (Blügel et al. 1989), the spin-polarization of the atoms is opposite on the adjacent atomic terrace of Cr. The TMR effect leads to a reduction of the current and the STM tip approaches.

This mechanism results in alternating step heights seen with a spin-polarized tip. It does, however, not allow a direct observation of the sample spin but only mixed topographic and spin information are obtained.

This mode is suited to study antiferromagnets or ferrimagnets. On the atomic scale, the topographic and spin information can be unfolded by studying the corrugation with nonmagnetic and spin-polarized tips. The additional corrugation due to the spin is of the order of several pm. In ferromagnets, the magnetic domains often extend over micron range. STM is at the moment not capable to resolve an additional corrugation of pm on these length scales such that the mode is not appropriate for these samples. Further, small variations in the density of states, for example, by contaminations, statistical variation of the composition in alloys, leads to a corrugation that is hard to distinguish from spin-induced corrugations.

2.3 Spectroscopic Mode

A mode that, under certain circumstances, allows to separate spin information from topographic information is spin-polarized scanning tunneling spectroscopy. It was initially suggested by Pierce (1988) and Stroscio et al. (1995) and was first realized by Bode et al. (1998). In general, if a finite bias is applied, all states between the two Fermi levels of tip and sample are involved in tunneling. The imaging mode uses the fact that the spin-polarization of the sample density of states is a function of energy. As a consequence, the size of the TMR depends on the bias voltage. The STM is operated in the constant current mode, that is, effective changes of the tunneling current due to the TMR are regulated away by the feedback mechanism as discussed above. At each lateral position, the bias voltage is changed while keeping the tip-sample separation fixed. In case the spin-polarization does not depend on energy, the recorded I(V) spectra for parallel and antiparallel magnetic orientation are identical. If, however, the spin-polarization varies, the I(V) curves also vary. Let us assume without loss of generality that the spin-polarization rises with energy. As a consequence, the TMR rises with bias, that is the I(V) curves for parallel magnetization rise steeper than those for antiparallel. Spin information can be separated by evaluating the spectra.

Figure 2(a) shows the topography of an Fe nanocluster grown on W(100) using an Fe-coated tip. To obtain spin contrast, the peak height of the differential conductance related to the minority surface state of Fe at 200 mV was used, as suggested by Stroscio et al. (1995). Figure 2(b) shows the lateral variations of the peak height which reflects the magnetic closure domains in the Fe nanocluster (Yamasaki et al. 2003). Figure 2(c) shows clearly the difference of the

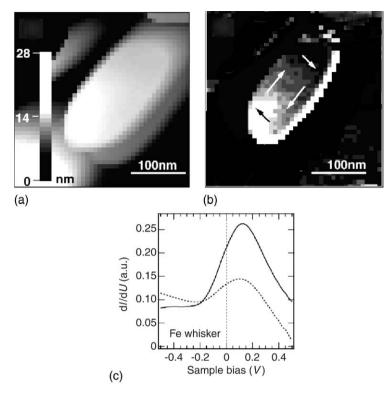


Figure 2
(a) Constant current image of an Fe nano-cluster on W(100) and (b) local height of the peak of the minority surface state of Fe in the dI/dU as function of position revealing the magnetic configuration. (c) Spin-dependent dI/dU spectra of the surface state of Fe(100). Data were recorded with Fe-coated W tips.

 $\mathrm{d}I/\mathrm{d}U$ spectra for parallel and antiparallel orientations of tip and sample magnetization.

The magnetic signal on samples of a homogeneous density of state can be separated from the topographic information, the prerequisite being the study of the differential conductivity with an unpolarized tip. On samples with inhomogeneous electronic structure caused, for example, by thickness variations or steps, often a strong nonmagnetic contrast covers up the magnetic contrast. As a spurious effect, the topographic images contain spin information similar to the constant current mode. Samples with a strong variation in the density of states and spin-polarization with bias are most suited for this mode. These are surfaces with surface states or thin films with quantized states. The contrast mode may be used with bulk and coated ferromagnetic or antiferromagnetic tips. This allows to minimize the influence of the magnetic stray field and operation in high magnetic fields. The application on systems with an inhomogeneous or unknown electronic structure has been used to image the magnetism of simple metallic ferromagnets and antiferromagnets and has shown to be somewhat problematic.

2.4 Differential Magnetic Mode

In the differential magnetic mode, the tip magnetization is modulated to strictly separate the spin information from the topography. Bulk ferromagnetic STM electrodes are used. First attempts using Ni-tips were of limited success (Johnson and Clarke 1990). but later, this mode was realized to obtain magnetic contrast (Wulfhekel and Kirschner 1999). The basic concept of this mode is related to Eqn. (1). The magnetization of a magnetically bistable tip is periodically reversed which is equivalent to changing the sign of the spin-polarization of the tip apex. In the experiment, the magnetization of the tip is reversed by an alternating current through a small coil that is fixed to the tip. The frequency of the alternating current is above the cut-off frequency of the feedback loop of the STM. Thus, the feedback loop only detects the averaged tunneling current I for the two spin-polarizations (positive and negative) of the tip. The spindependent part cancels out so that the topographic image contains no magnetic information. With a phase-sensitive lock-in amplifier, the alternating part of the tunneling current ΔI is detected which contains

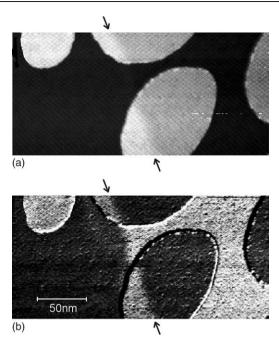


Figure 3
Sp-STM image of (a) the topography and (b) the corresponding spin signal of 11.9 ML Mn on Fe(001). A buried Fe step-edge is running almost vertically through the center of the images, indicated by arrows.

all the spin information. This way, the topographic and spin information are separated.

Figure 3(a) shows the topography of an Mn film on Fe(001). Due to the different lattice constants of Mn and Fe, a buried Fe substrate step-edge shines through the Mn film forming a step of subatomic height. It runs almost vertically through the center of the image indicated by arrows. Figure 3(b) shows the spin signal recorded simultaneously. The layerwise antiferromagnetic order of the Mn results in an alternating contrast. At the position of the buried Fe step, a 180° domain wall is induced in the Mn (Schlickum *et al.* 2004).

The differential magnetic mode allows the strict separation of topographic and spin information and can even be applied to electronically inhomogeneous samples. It has been used with bulk ferromagnetic tips for technical reasons. As the tip magnetization has to be switched periodically, thin film tips with high coercivities are less suited. A limitation is that the application of a large magnetic field affects the tip-switching so that only studies in limited applied fields are possible.

3. Tip Preparation

For spin-polarized STM, at least the last atom at the tip apex has to be spin-polarized, stable in size and

orientation. Two different approaches have been used in the past to reach these conditions. The whole tip may consist of a material with a certain spin-polarization or a nonmagnetic tip is coated with spinpolarized material. Most frequently, non-magnetic W tips were coated in situ with a thin film of either a ferromagnet or an antiferromagnet. The best results have been reported when the W tips were flashed to high temperatures before coating (Bode and Wiesendanger 2004). This cleans the tip apex from the natural oxide and leads to reproducible but rather blunt tips. When depositing, for example, an Fe layer of 10 ML on the tip followed by gentle annealing, the tip shows a magnetization in the sample plane and one in-plane component of the magnetization can be imaged (Bode et al. 1998). When the tip is coated with Gd, the tips show sensitivity for the out-of-plane component of the spin-polarization (Pietzsch et al. 2000). Both Fe- and Gd- coated tips produce a small magnetic stray field at the tip apex that, in some cases, may influence the magnetic object under investigation (Kubetzka et al. 2002). Alternatively, antiferromagnetic Cr (Kubetzka et al. 2002) or Mn (Yamada et al. 2003) coatings have been used. While the orientation of the spin-polarization of the tip is more difficult to control, the tips are mostly free of stray fields. Coated tips are most suitable for the constant current and the spectroscopic imaging mode.

Bulk tips are suitable for the constant current, the spectroscopic, and the differential magnetic imaging mode. The direction of magnetization in bulk magnetic tips is determined by the shape of the tip. To image the out-of-plane component of the spin-polarization, sharp tips have produced best results (Ding et al. 2002). For imaging one in-plane component of the spin-polarization, ring-shaped STM electrodes can be used. In these ring electrodes, the magnetization direction lies tangential to the outer perimeter of the ring. Thus, at the bottom of the ring where the tunneling occurs, the magnetization lies in the plane of the sample surface. Because the magnetic flux in an ideal ring is closed, the magnetic stray field is zero. By choosing the plane in which the ring is oriented, the magnetization direction of the ring is defined and thus the direction of the sensitivity. Although the rings used as STM electrodes are not sharp, a lateral resolution below 1 nm could be achieved (Schlickum et al. 2003, 2004).

For the differential magnetic mode, tips were electrochemically etched from CoFeSiB. The chosen material offers extremely low coercivities in the range of $50\,\mu\text{T}$ and negligible magnetostriction. This ensures virtually vanishing vibrations due to magnetostriction (Wulfhekel *et al.* 2002).

4. Conclusion

Sp-STM is a relatively new but matured technique offering a fresh view into magnetism on the nanometer

scale. It combines ultimate lateral resolution with a direct measurement of the spin-polarization. It is suited to image ferromagnetic as well as antiferromagnetic surfaces and allows the study of the spin-resolved density of states. With this, it combines classical magnetism and modern quantum mechanical solid state physics. It has had considerable impact on the study of magnetic nanostructures and will help to further develop that field.

See also: Atomic Force Microscopy; Magnetic Force Microscopy; Magnetoresistance: Magnetic and Nonmagnetic Intermetallics; Scanning Tunneling Microscopy; Surface Evaluation by Atomic Force Microscopy; Thin-film Magnetism: PEEM Studies.

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