Chapter 103 Spin-Polarized Scanning Tunneling Microscopy

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Keywords Spin-polarized tunneling current · Spin polarization · Magnetism

103.1 Principle

Spin-polarized scanning tunneling microscopy (SP-STM) is a powerful tool to visualize spin-polarization vectors of sample surface atoms. Variety of unique new magnetic properties have been discovered and studied, such as non-collinear magnetic exchange coupling between monolayer films, magnetoresistance of single atoms, single molecules, and nanomagnets, as well as skyrmion or magnon in magnetic films [1–5].

SP-STM measurements can be performed with an ultrahigh-vacuum (UHV)-STM setup combined with a scanning tunneling spectroscopy (STS) technique, while SP-STM requires a spin-polarized tip. Figure 103.1 shows the principle, in which local density of states (LDOS) of a tip and a sample is spin-polarized near the Fermi energy (E_F). Now only 3 spin-up electrons exist for the tip [spin polarization (P) = (3 - 0)/(3 + 0) = 100%], while 3 spin-up and 1 spin-down electrons for the sample [P = (3 - 1)/(3 + 1) = 50%]. In (a), tip and sample spin-polarization vectors are parallel. Then, tunneling probability can be described as $3 \times 3 + 0 \times 1 = 9$, where no spin-flip during tunneling is assumed. Next, the sample spin-polarization vector is reversed (antiparallel configuration, see Fig. 103.1b). In this case, tunneling probability is $3 \times 1 + 0 \times 3 = 3$. SP-STM detects this difference in current due to different spin-polarization vector configurations between the tip and the sample. This mechanism is comparable to tunnel magnetoresistance.

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Fig. 103.1 Spin-polarized electron tunneling process between the spin-polarized tip and the sample in the case of **a** parallel and **b** antiparallel spin-polarization (magnetization) configurations. [1-5]

103.2 Features

- Spin-polarization vectors of sample surfaces can be detected.
- Magnetoresistance can be detected at the atomic scale.
- Spin-polarized STM tips are required.
- Combined with IETS or magnetic coils, magnon or hysteresis loop can be measured.
- SP-STM has been performed in UHV at ~ 10 mK-300 K.

103.3 Instrumentation

SP-STM system is based on a UHV-STM setup. All experiments have been performed in UHV to prevent contamination adsorption on the tip and the sample surfaces. Single impurity atoms break local symmetry, and therefore, spin polarization can be quenched. To detect spin-polarized current, it is necessary to decrease current noise as much as possible (typical current noise: $I_{noise} < 1$ pA). Figure 103.2 shows an example of SP-STM measurement system, where a layerwise antiferromagnetically coupled Mn(001) film is scanned by a spin-polarized tip.

The most important (and difficult) point to make success SP-STM measurements is how to obtain a stable spin-polarization vector at the tip apex. Recent trends are uses of Fe-coated W tips, Mn-coated W tips, Cr-coated W tips, and bulk-Cr tips [1–6]. The Fe-coated tips have been used since they have the highest spin polarization of ~40%, while recently most of the SP-STM researchers use the antiferromagnetic tips because of no stray fields ($P \sim 10\%$) [2].



Fig. 103.2 Experimental setup of spin-polarized STM measurements. *Spheres* and *black* and *white arrows* denote atoms and spin-polarization vectors, respectively. *Blue* and *red arrows* denote spin-up and spin-down electrons, respectively. SP-STM measurements can be performed in UHV at 10 mK–300 K. [2–5]

SP tips	Thickness	Direction	Spin polarization	Refs.
Fe-coated W	2–10 nm	In-plane	~40%	[1–5]
Mn-coated W	3–6 nm	In-plane	~9%	[2]
Cr-coated W	>10 nm	In-plane	~10%	[2]
Cr-coated W	2–10 nm	Out-of-plane	~10%	[2]
Bulk-Cr	-	Out-of-plane	-	[6]

Table 103.1 Spin-polarization vectors of spin-polarized tips

These tips were prepared in a UHV preparation chamber before setting in STM. First, a W tip chemically etched in air was introduced into the preparation chamber through a load lock chamber and cleaned (Ar^+ sputter with a subsequent anneal to 2200 K). Next, magnetic films were deposited at 300 K on the cleaned W tip. Proper annealing of 500–600 K could increase crystalline quality of the film [2].

On the other hand, the bulk tip requires a different manner. A chemically etched bulk-Cr tip was cleaned inside the UHV-STM setup by applying voltage pulses (≤ 10 V) between the tip and the sample to remove oxide layers at the apex [6].

Depending on materials and thickness, the tip spin-polarization directions can be controlled by different magnetic anisotropies. Table 103.1 shows experimentally obtained tip spin-polarization vectors of SP-STM tips. Using these clean spin-polarized tips combined with STS, the spectroscopy d*I*/d*V* map shows spin contrast.

103.4 Applications

103.4.1 Magnetic Imaging

Experimentally obtained SP-STM results of Mn(001) films grown on Fe(001) are shown in Fig. 103.3 [2–5]. Seventh to tenth monolayers (ML) are exposed on the



Fig. 103.3 Spin-polarized STM results on bct-Mn(001) films with an Fe-coated W tip in UHV at 300 K. Topographic image $(100 \times 100 \text{ nm}^2)$ and spin-polarization dI/dV map at +0.2 V were obtained simultaneously. [2–5]

surface as shown in the STM topographic image $(100 \times 100 \text{ nm}^2)$. Since Mn(001) films higher than fourth layer have the same LDOS, non-spin-polarized W tips show no contrast; however, spin-polarized Fe-coated W tips with an in-plane sensitivity (see Table 103.1) show spin contrast in the dI/dV map at +0.2 eV (Fig. 103.3). Alternating spin contrast between the monolayers means that Mn atoms in the monolayer couple ferromagnetically, while they couple antiferromagnetically between the layers. Thus, we could obtain spin structures of nanomaterial by means of SP-STM.

103.4.2 Single-Molecule Junction

Using the STM tip, we can manipulate single atoms or single molecules. Left panels as shown in Fig. 103.4 show examples, where letters "N" and "S" were written with 17 Fe atoms, and "smile" was drawn with 11 CO single molecules. Using this STM manipulation technique, we can measure conductance through the target (see Fig. 103.4). Here, we measure I-z curve instead of dI/dV. By measuring the tunneling current (*I*), the tip is approached to the target molecule. When the tip contacts with the molecule, the measuring current becomes constant, forming a single-molecule junction. Thus, we can measure conductance through the molecule.

Using the spin-polarized tips, we can measure magnetoresistance (MR) through a single molecule (see right panels as shown in Fig. 103.4). Single phthalocyanine molecules were deposited on the Mn(001) films as shown in Fig. 103.3. First, the Fe-coated W tip was gently contacted to the molecule adsorbed on the ninth monolayer, in which spin polarization is parallel to the tip spin polarization and measured conductance "I". Next, the same tip was contacted to the molecule adsorbed on the eighth monolayer, in which spin polarization is antiparallel to the tip spin polarization and measured conductance "II". Thus, MR through a single



Fig. 103.4 STM manipulations of single atoms and molecules in UHV at 5 K. Magnetoresistance measurements through a single molecule by means of SP-STM [3]

phthalocyanine molecule was obtained by MR = (I–II)/II = -50% [3], which suggests that such a 1-nm-size π -conjugated organic molecule can be useful as a new nanomaterial for near-future spintronic devices.

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