Imaging Atomic-Scale Spin Structures

A Novel SPM Method Resolves Spin Structures on Insulator Surfaces

A fundamental understanding of magnetic and spin-dependent phenomena requires the determination of spin structures and spin excitations down to the atomic scale. The direct visualization of atomic-scale spin structures [1-4] has first been accomplished for magnetic metals by combining the atomic resolution capability of Scanning Tunnelling Microscopy (STM) [5, 6] with spin sensitivity, based on vacuum tunnelling of spin-polarized electrons [7]. The resulting technique, Spin-Polarized Scanning Tunnelling Microscopy (SP-STM), nowadays provides unprecedented insight into collinear and non-collinear spin structures at surfaces of magnetic nanostructures and has already led to the discovery of new types of magnetic order at the nanoscale (fig. 1 and [8]). More recently, the detection of spin-dependent exchange and correlation forces has allowed a first direct real-space observation of spin structures at surfaces of antiferromagnetic insulators [9]. This new type of scanning probe microscopy, called Magnetic Exchange Force Microscopy (MExFM), provides a powerful new tool to investigate different types of spin-spin interactions based on direct-, super-, or RKKY-type exchange down to the atomic level.

Spin-Polarized Scanning Tunnelling Microscopy and Spectroscopy

The technique of SP-STM is based on vacuum tunnelling of spin-polarized electrons as can be observed in STM-type tunnel junctions involving a magnetic tip and a magnetic sample. In this case, the tunnelling current does not only depend on the tip-surface separation and the applied bias voltage between tip and sample, but also on the spin polarization of the electronic states near the Fermi level of both electrodes as well as on the relative orientation of the local magnetization of the sample and the magnetic moment at the tip apex. Early experiments based on planar tunnel junctions involving magnetic electrodes have been performed by Jullière in the mid-seventies [10]. The invention of the STM by G. Binnig and H.
Rohrer [5] allowed the replacement of the ill-defined tunnel barrier of planar tunnel junctions by a well defined vacuum barrier and the replacement of one of the planar electrodes by a freely movable and positionable probe tip. Magnetic tips have first been employed in scanning probe microscopy to perform spatially resolved measurements of magnetic dipole forces [11]. The corresponding technique, Magnetic Force Microscopy (MFM), has become a routine method for magnetic domain imaging [12]. However, the spatial resolution of MFM is limited to 10 - 20 nm due to the long-range nature of the magnetic dipole forces [6]. In order to improve the spatial resolution of magnetic-sensitive scanning probe microscopy down to the atomic scale, two different approaches have been proposed at the end of the eighties: either to measure spin-dependent tunnelling currents making use of the strong exponential dependence of the tunnelling current on the tip-surface distance in order to combine atomic- with spin-resolution, or to measure spin-dependent exchange and correlation forces at very small distances between a magnetic probe tip and a magnetic sample based on the atomic force microscopy (AFM) technique [13].

The first successful observation of vacuum tunnelling of spin-polarized electrons in an STM experiment was made in 1990 [7], using a highly spin-polarized CrO$_2$ thin film probe tip and an antiferromagnetic Cr(001) sample in order to exclude a disturbing influence of superimposed magnetic dipole interactions. In these early SP-STM experiments, the constant-current mode of operation was employed where spin-polarized tunnelling current effects showed up as apparent height differences in constant-current STM contours. This mode of operation has turned out to be useful for atomic-scale SP-STM studies of surface spin configurations, as has been shown by resolving the spin structure at surfaces of ferrimagnetic oxides [1], two-dimensional antiferromagnetic metal layers (fig. 2 and [2 - 4]) and antiferromagnetic nitrides [14]. For nanoscale magnetic domain imaging the spectroscopic mode of SP-STM operation, introduced in 1998 [15], was found to be
superior compared to the constant-current mode, particular with respect to a clear separation between topographic, electronic, and magnetic contrast effects [16]. In the spectroscopic mode of SP-STM, the spin-resolved differential tunnelling conductance $dI/dU$ is measured with spatial resolution. The bias voltage applied between the magnetic tip and the magnetic sample is chosen in such a way that tunnelling into or out of a highly spin-polarized electronic state leads to a high spin contrast image. A nice example is the observation of the magnetic domain structure of rare-earth metal films, such as Dy(0001) epitaxially grown on W(110) single crystal substrates (fig. 3 and [17]).

**Magnetic Exchange Force Microscopy**

While the SP-STM method has provided unprecedented insight into atomic spin configurations at magnetic surfaces, its application is limited to electrically conducting samples such as magnetic metal films or magnetic semiconductors. In order to reveal atomic spin structures at surfaces of insulators and to open up the exciting possibility of studying spin ordering effects with atomic resolution while going through a metal-insulator transition, we have developed the technique of Magnetic Exchange Force Microscopy (MExFM). This technique is based on the detection of short-range spin-dependent exchange and correlation forces at very small tip-sample separations (a few Angstroms), in contrast to MFM where the magnetic dipole forces are probed with a ferromagnetic probe tip at a typical tip-to-surface distance of 10 - 20 nm. An important starting point for achieving the challenging goal of atomic-resolution spin mapping on surfaces of insulators has been the development of Non-Contact Atomic Force Microscopy (NC-AFM) with true atomic resolution [18]. Nowadays, NC-AFM allows atomically resolved studies of any material system [19], even in the case of curved surface topographies [20]. MExFM combines the possibilities of NC-AFM and atomic-scale spin resolution by making use of an atomically sharp probe tip with a very well defined spin state at its apex. Based on the knowledge gained during the development of SP-STM in preparing such tips, we have recently succeeded in resolving the surface spin structure of the antiferromagnetic insulator NiO(001) [9]. Figure 4a shows an atomic-resolution topographic NC-AFM image revealing chemical contrast between the oxygen atoms (bright sites) and the Ni atoms (dark sites) which originates from a different total charge density above O- and Ni-sites. No contrast is observed between magnetically inequivalent Ni sites, i.e. Ni atoms with a different orientation of their magnetic moments. By approaching the out-of-plane magnetized Fe-coated tip closer to the surface atoms, the spin-dependent exchange interaction between the rather localized Ni d-states of the sample and the Fe d-states of the tip leads to a different force or force gradient above Ni atoms with a different
orientation of their magnetic moments. As a result, a superperiodicity corresponding to an antiferromagnetically ordered state of the NiO(001) surface is observed in the MExFM image (fig. 4b). Notice that the apparent height difference between the magnetically inequivalent Ni-sites, corresponding to the different magnitude of the spin-dependent quantum-mechanical forces felt by the tip above the different Ni atoms, only amounts to 1.5 pm as can be deduced from the line section in figure 4b. To resolve such tiny signals, the AFM instrument has to be operated at low temperatures in order to reduce the thermal excitations of the AFM cantilever (force sensor).

To summarize, it has become possible to measure spin-dependent quantum mechanical exchange forces between a single atom at a tip apex and individual surface atoms, leading to a new type of scanning probe microscopy technique which can now resolve atomic spin structures at surfaces of any type of material system, thereby extending the range of applications of SP-STM considerably.

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