CHAPTER 1

Introduction

The semi-empirical Bethe–Slater curve, known since 1930 (figure 2.3), shows that, when the separation between atomic moments in a solid is varied by changing the element and hence the lattice constant, the exchange interaction changes from antiferromagnetic to ferromagnetic or vice versa. To directly access and measure this distance dependence of the exchange interaction strength between two magnetic atoms of the same element is not possible, because the separations between magnetic atoms in a solid of a given material are fixed by the crystal structure. On surfaces the situation changes and surface imaging techniques can be applied to address this theoretical hypothesis.

Scanning probe methods such as spin-polarized scanning tunnelling microscopy (SP-STM) [1–4], spin-flip spectroscopy [5, 6], and the recently developed atomic force microscopy (AFM) [7, 8] based magnetic exchange force microscopy (MExFM) [9,10], allow one to study magnetic ordering on surfaces with atomic scale precision. SP-STM and MExFM allow for direct characterization of even more complex magnetic structures with atomic-scale resolution. These techniques help to gain a fundamental understanding of spin-related magnetic phenomena within the field of condensed matter research.

In the past, scanning tunneling spectroscopy (STS) has been employed to measure the oscillatory distance dependent nature of the Ruderman-Kittel-Kasuya-Yosida (RKKY)-type magnetic exchange interaction between pairs of individual Co adatoms on Cu(111) by evaluating the Kondo resonance using a non-magnetic tip [11], and on Pt(111) by analyzing magnetization curves using a magnetic tip [12]. In both cases the magnetic coupling was mediated via conduction electrons of the non-magnetic substrate.

While the application of SP-STM and SP-STS, which combine the atomic-resolution capability of STM with spin sensitivity, is limited to probe conducting systems, that is, metallic and semiconducting magnetic nanostructures, MExFM opens up the possibility to extend spin characterization with at least equal precision towards insulators. Proposed by R. Wiesendanger *et al.* [13] in 1991, the basis of MExFM is to combine the atomic resolution capability of

AFM with the magnetic sensitivity of magnetic force microscopy (MFM) [14], but down to single spin resolution. Applying MExFM, the exchange interaction between a magnetic probe and the single magnetic moments of surfaces is visualized in real-space at length scales that are not accessible by other magnetic sensitive techniques, such as MFM [15] and magnetic resonance force microscopy (MRFM) [16]. Hence, MExFM promises to be a powerful tool to investigate many different types of exchange interactions, such as direct exchange or the prominent RKKY-interaction, either mediated by a substrate, between single molecules, or even single atoms.

The feasibility of MExFM to map spin structures with atomic resolution on insulating surfaces was demonstrated on antiferromagnetic NiO(001) [9, 17]. The key to success, in contrast to previous attempts to perform MExFM [18–22], was the application of an external magnetic field. The field was used to align the magnetic moment of the foremost probe tip apex atoms to enhance the interaction strength between the tip and sample moments. Implementation of the magnetic field paved the way for direct measurement of the magnetic exchange coupling between tip and sample atoms and for the visualization of the rowwise antiferromagnetic contrast between neighbouring rows of Ni atoms.

This work expands upon the first successful application of MExFM towards more systems and more capability. The sample system studied within this work is the first and second atomic layer of iron on tungsten with surface orientation (001). The focus, however, is put on the first atomic iron layer. This layer has been subject to theoretical considerations using density functional theory (DFT) [23], and it has already been studied earlier using SP-STM [24]. It has been found that, due to hybridization with the tungsten substrate, the iron monolayer becomes antiferromagnetic with out-of-plane anisotropy, that is, monolayer Fe atoms arrange in a checkerboard pattern with the moment of every second iron atom pointing either up or down, respectively.

The electronic and magnetic structure of iron is very different to that of the insulating NiO. Namely, the itinerant *d*-electron spins of the iron ML are delocalized, while, in NiO, the *d*-electrons are localized at the Ni atoms and couple via superexchange. Therefore, the study of Fe/W(001) with MExFM and the comparison to NiO(001) allows new insights into the contrast mechanism in MExFM. Since no further systems have been investigated successfully with MExFM, and since there is a lack of experiments on insulators which can be compared with SP-STM studies [1, 4, 12, 25–27], it is vital to understand the relevant contrast mechanisms in detail to clarify whether MExFM can be developed into a versatile tool for the analysis and manipulation of atomic scale magnetic structures.

Up to now, MExFM was only utilized in imaging mode. However, its spectroscopic mode, that is, measuring the interaction between probe tip and sample as a function of tip-sample distance, should allow to directly measure the distance dependence of the exchange interaction between single magnetic mo-

ments and a magnetic tip across a vacuum gap. By performing such measurements, as shown here, it is now possible to quantify the magnitude of the exchange interaction for the first time. Hence, this work presents the first successful application of the spectroscopic mode of MExFM, i.e. magnetic exchange force spectroscopy (MExFS), where, in contrast to previous STM-based experiments [11, 12], the magnetic exchange interaction is not mediated by a substrate.

After an introduction into the field of atomic magnetism in chapter 2, chapter 3 introduces the basic principles of AFM, where a super-sharp tip probe, mounted at the end of a vibrating cantilever, scans a surface. Within a certain distance between tip and surface, tip-sample interactions lead to a mechanical response of the cantilever vibration. Measuring this response allows for the analysis of the various interaction forces, such as chemical and magnetic exchange force, with a sensitivity of a few pN. A detailed description of the experimental setup and the preparation of Fe/W(001) are found in chapter 4.1. Chapter 5 discusses different properties of the used probes and the sample system, studied with KPFM and MFM.

After characterization of both, probe and sample, the c(2×2) surface magnetic unit cell of the iron monolayer is obtained by MExFM utilizing iron and chromium coated probes, presented in chapter 6. By comparison with theory, the origin of the obtained image contrast of the magnetic moments oriented perpendicular to the surface is revealed. Due to the difference in electronic structure, the corrugation amplitude as obtained on Fe/W(001) is essentially larger than it was observed in the first MExFM experiments performed in 2007 on NiO(001) [17]. Furthermore, chromium coated probes turn out to be much better suited than iron coated probes at small tip-sample separation for stable imaging with atomic and spin resolution. In contrast to the preliminary experiments on NiO(001), first successful MExFM experiments without external magnetic field are performed using chromium coated probes.

Chapter 7 presents the first direct measurement of the magnetic exchange interaction across a vacuum gap by combining magnetic exchange force spectroscopy (MExFS) and three-dimensional force field spectroscopy (3D-FFS). In MExFS, the spin sensitive probe is approached perpendicular to the sample surface and the total interaction potential between tip and sample is measured. Subtracting two curves obtained on oppositely oriented atomic magnetic moments on the surface yields the magnetic exchange interaction between both moments. All non-magnetic interactions are elegantly eliminated using this method, since all surface iron atoms are chemically identical. Comparison with DFT calculations reveals that theory is able to reproduce the measured magnetic exchange energy qualitatively and quantitatively using pyramidal shaped tips. However, some experimental results are not reproducable. They indicate that the used tips are not stable but undergo a strong modification during approach and retraction within single cantilever oscillation cycles. This assumption is supported by a strong variation of the energy dissipated while

keeping the oscillation amplitude of the cantilever constant. A survey on dissipation is presented in chapter 8 and leads to a classification of the used probe tips as either stable or unstable.

Chapter 8 further presents methods for the *in situ* preparation of magnetic sensitive tips. It is shown that, although coated with magnetic material, the tips used in this work do not show spin sensitivity from the beginning. Instead, spontaneously or deliberately induced modifications of the tip apex are needed to eventually perform successful MExFM and MExFS experiments. The findings of the last chapter are of further interest concerning the manipulation of single atoms in MExFM for realizing future atomic spin logic devices [28].