Chapter 1

Introduction

The investigation of magnetic thin films has become an important subject of research during the past two decades.

One of the striking discoveries in magnetism was the observation of hysteresis loops shifted away from the zero-field axis observed for ferromagnetic Co particles having an antiferromagnetic coating [MB56]. This phenomenon, called exchange bias (EB), also occurs in thin films, where it has important potential technological applications. Exchange bias is induced after field-cooling of a ferro-/antiferromagnetic bilayer below the Néel temperature of the antiferromagnet; the strength of the induced exchange bias and the coercivity usually increases with decreasing temperature. Generally, exchange bias is considered to be an interface effect; however, in recent studies on different EB trilayers, it was suggested that the coupling was transferred across a non-magnetic 'spacer', e.g. Cu, Ag or Au [GAC97] indicating a long range coupling mechanism. Nevertheless, there remains some controversy as to whether the exchange bias effect originates from 'pinholes' in the spacer or whether it is in fact a long range coupling persisting through the non-magnetic layer [NKS+00].

In this work different EB systems of the type Co/CoO and Co/Au/CoO and related systems have been investigated.

Therefore the structural properties of the samples had to be known. The samples were prepared in a controlled manner using Molecular Beam Epitaxy. A controlled in-situ oxidation method was applied to prepare ultrathin antiferromagnetic CoO layers [GR00a]. In order to analyze the structure and chemical composition of the samples, different in-situ (AES, LEED, STM) and ex-situ (X-ray diffraction, TEM) characterization methods have been employed.

Knowing the structural properties of the samples, various magnetic properties have been investigated using different appropriate measurement techniques, such as SQUID (Superconducting Quantum Interference Device) magnetometry, Polarized Neutron Reflectometry (PNR) and Low Temperature Nuclear Orientation (LTNO). SQUID magnetometry was used in order to determine the characteristic magnetic properties related to the EB effect, such as the EB shift, the coercive fields etc., before the EB samples were subjected to the technically demanding measurement techniques PNR and LTNO.

A main goal in this work was the study of magnetization reversal processes in a Co/CoO multilayer, which represents an example of an EB system consisting of individual granular Co and ultrathin granular CoO layers exhibiting a strong exchange anisotropy.

The second focus in this thesis was to explore the applicability of the hyperfine field technique LTNO for the investigation of thin magnetic layer systems. This method promises to be very well suited to study the EB effect in Co/Au/CoO EB-trilayers, since it is sensitive at the same time to the magnetic layers and to the induced nuclear polarization in the non-magnetic Au spacer adjacent to a magnetic material.

Study of magnetization reversal processes via Polarized Neutron Reflectometry

Magnetization reversal processes in EB systems have been a subject of many studies. In the EB state, one finds very good conditions to observe correlated phenomena such as asymmetric reversal processes on opposite sides of the hysteresis loop, as has been found in other EB systems [FYL+00]. Magnetization reversal processes are also important for magnetic storage technology. For the applicability of magnetic materials in magnetic memory devices it is of great importance to optimize the speed of magnetization reversal processes. In the case of Magnetic Random Access Memory (MRAM) devices e.g. reversal by fast coherent rotation is desired since reversal by domain wall motion is far too slow [GvdBH+02]. Therefore, it is crucial to study and to control the mechanisms which determine the reversal processes in different materials.

The present study on a [Co/CoO/Au] multilayer via Polarized Neutron Reflectometry in particular focuses on the magnetization reversal processes in EB systems consisting of individual granular Co/CoO bilayers. This type of granular EB system is characterized by only one prominent direction in the EB state, which is the direction of induced EB.

The PNR method is well suited to study magnetization reversal processes. In this technique, a polarized neutron beam hits a magnetic sample at low angles Θ_i (with respect to the sample plane). After interacting with the magnetic moments in the sample the neutrons are scattered at an angle Θ_f. Measuring the specular ($\Theta_i = \Theta_f$) reflected non spin-flip (NSF) intensities of up-neutrons (R^{++}) and down-neutrons (R^{--}), PNR can be used as a classical magnetometer. The first and second superscripts (+ -) denote the polarization of the incoming and scattered neutron beam. Additionally, by measuring the spin flip (SF) intensities R^{+-} and R^{-+}, the unique property of neutrons can be exploited, that their spin is flipped if the magnetization in the sample has an in-plane component perpendicular to the spin axis. In this work, PNR measurements are presented which treat the magnetization reversal in a [Co/CoO/Au] EB-multilayer at room temperature, i.e. above T_N of CoO and at 10 K when the sample is in the EB state.

In addition, first off-specular ($\Theta_i \neq \Theta_f$) PNR measurements on the same multilayer will be presented. Off-specular scattering arises from structural and magnetic inhomogeneities. Therefore this type of experiments potentially provides information about magnetic domains.

Studies by SQUID magnetometry

In the present work different Co/Au/CoO trilayers with varying Au spacer thicknesses have been investigated via SQUID magnetometry. SQUID magnetometry is sensitive only to the magnetic layers. Usually, information on the dependence of the EB coupling strength on the spacer thickness is obtained by studying changes of the magnetic properties of the FM material upon varying the spacer thickness. The corresponding simple Co/CoO bilayer system consisting of a granular Co and a very thin granular CoO layer is characterized by hysteresis loops of rectangular shape and an extraordinarily high exchange bias anisotropy [GR00b]. This system is particularly well suited to investigate the EB effect across non-magnetic spacers since it provides sensitivity even for samples containing relatively thick Au spacers.

Investigation of nuclear magnetic polarization in thin multilayer systems by Low Temperature Nuclear Orientation

It is of great interest to study how magnetic moments are induced in or transferred across a non-magnetic spacer layer. Therefore other experimental methods, which are sensitive to non-magnetic materials, have to be employed. Hyperfine field techniques, such as Low Temperature Nuclear Orientation (LTNO), can measure the nuclear spin polarization (direction and magnetic hyperfine field strength) of both the magnetic (e.g. Co) and non-magnetic spacer layers (e.g. Au) simultaneously. Using the LTNO technique, the average alignment of the nuclear magnetic moments and the magnitude of the magnetic hyperfine field are derived by measuring the γ-radiation emitted by radioactive probe atoms in the samples previously irradiated with thermal neutrons. Alignment of the radioactive magnetic probe nuclei is achieved at temperatures close to absolute zero. Under this condition the γ-rays are emitted anisotropically, and this anisotropy can be detected.

In contrast to most conventional magnetization measurements, LTNO determines the average magnitude of the nuclear alignment $<I_z^2>$ with respect to the applied magnetic field by observing the anisotropic γ-ray emission from radioactive probe nuclei [SP86]. Furthermore, LTNO is potentially sensitive to whether (i) the magnetization has rotated away from the applied field axis or (ii) it has broken up into domains aligned along the field axis.

Previous hyperfine field determinations have shown that strong spin perturbations (hyperfine fields $\geq$ 3 T) in an ultrathin non-magnetic spacer are to be expected only in the layer adjacent to the interface with a magnetic material. This has been demonstrated experimentally in earlier studies using perturbed angular correlation (PAC) studies on Fe/Ag bilayers with ^{111}In probes [RDF$^+$97], as well as with Mössbauer spectroscopy performed on Au/Ni and Au/Fe multilayers [KNES94]. Therefore the LTNO method is mainly sensitive to the interfacial Au since appreciable polarizations of the non-magnetic spacer are restricted to the interfaces adjacent to a magnetic material.

First measurements on Co/Au/CoO EB trilayers by means of LTNO are presented in this work. The EB effect was studied from the polarization of the nuclear moments in the ferromagnetic Co layer for different Co/Au/CoO trilayers with increasing Au layer thicknesses. At the same time the experiments reveal how the exchange bias effect is reflected in the induced polarization of the nuclear Au moments at the interface adjacent to the magnetic layers.

Additional information about the Co/Au and CoO/Au interfaces is obtained by comparing the results to LTNO measurements on related magnetic systems, such as Co/Au/Co and CoO/Au/CoO trilayers. In order to explore the use of LTNO for investigating the magnetization reversal mechanisms in EB systems, the Co/Au/CoO trilayers with large EB effects are particularly well suited. The extremely thin CoO films (2 nm) consist of small grains [GR00b, GR00a], and should have only one easy axis, induced by the applied magnetic field. In a preliminary investigation, the magnetization reversal processes in Co/Au/CoO were examined by means of LTNO, observing the behavior of both the polarization of the nuclear Co moments and the interfacial nuclear Au moments during hysteresis measurements (when the sample is in the EB state).

The present thesis is organized as follows: In chapter two a general description of the exchange bias effect will be given by summarizing the most important features occurring in exchange bias systems.

Chapter three deals with the the preparation and structural analysis of the MBE-grown samples. A short description of the ultra high vacuum (UHV) chamber and the applied characterization methods will be given. Thereafter follows a presentation of the experimental results revealing the structural properties of the investigated layer systems.

In the fourth chapter SQUID magnetometry measurements on the magnetic properties of Co/Au/CoO- and Co/CoO-layer systems are treated. After a brief description of the SQUID technique the results on Co/Au/CoO trilayers with different Au spacer thicknesses are presented. The dependence of the exchange bias effect upon the Au spacer thickness is discussed by interpreting the magnetization curves of various Co/Au/CoO systems with different Au layer thicknesses. Furthermore SQUID measurements on a Co/CoO bilayer and on a $[Co/CoO/Au]_{20}$ multilayer are presented. The magnetic properties related to the exchange bias effect are deduced from the magnetization curves. Finally the results for the two systems will be compared and discussed.

Chapter five deals with the magnetization reversal processes in an EB-multilayer when it is in the EB state. The Polarized Neutron Reflectometry (PNR) technique is used for this study. After a short introduction to the scattering theory the experimental set-up of the V6 reflectometer at the HMI is treated. Afterwards a presentation of the results obtained from specular scattering experiments is given. First results obtained from off specular scattering experiments are shown as well. The latter provide additional information on magnetic domains. Finally the results will be discussed and compared to earlier studies in that area.

Chapter six describes measurements on Co/Au/CoO EB trilayers and related systems using the Low Temperature Nuclear Orientation (LTNO) technique. The chapter begins with a short description of the contributions to the magnetic hyperfine fields and the alignment of the nuclear magnetic moments along the hyperfine field axis at temperatures close to absolute zero. The γ-radiation, which is emitted anisotropically by radioactive probe nuclei if they are oriented, is treated as well. Thereafter follows a description of the cooling procedure with a ^{3}He-^{4}He dilution refrigerator used to achieve mK temperatures. First LTNO results on Co/Au/CoO EB trilayers with different Au spacer thicknesses are presented with respect to the applicability of the LTNO method for studying thin magnetic EB films containing a non-magnetic spacer. The results are discussed with a focus on how the EB effect is reflected in the interfacial Au during a hysteresis measurement when the EB trilayers are in the EB state. Additionally, results on related trilayer systems, such as Co/Au/Co and CoO/Au/CoO, are presented and discussed.

The experimental results from the three main experimental techniques employed in this work are summarized in chapter seven. The characteristic features of the different measurements are discussed in the framework of the experimental data.