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Proceeding Paper

Magneto-Optical Investigation of Surface Magnetization in Comparison with Bulk Magnetization †

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Abstract: Exchange-biased specimens were produced by molecular beam epitaxy (MBE) of ferromagnetic (FM) Co-on-CoO substrates after the substrates had been irradiated by heavy ions to induce defects in the antiferromagnet (AFM). Measurements were obtained at different temperatures for different sample orientations with respect to the external magnetic field. While the EB was relatively small, measurements of the bulk magnetization at low temperatures revealed unusually shaped hysteresis loops. The surface magnetization, however, showed simple, nearly rectangular hysteresis loops. This study focuses on the advantage of complementary information on surface and bulk magnetization from optical and non-optical measurement methods.

Keywords: exchange bias; heavy ion irradiation; magneto-optical Kerr effect; SQUID

1. Introduction

The exchange bias (EB) is a unidirectional anisotropy in ferro-/antiferromagnetic (FM/AFM) systems, occurring after field cooling through the Néel temperature of the AFM [1,2]. Primarily, it leads to a horizontal shift in the magnetic hysteresis loop, but it can also lead to vertical shifts and pronounced asymmetries in the hysteresis loop [3]. After being discovered in Co/CoO core–shell nanoparticles, it has since come to be investigated in diverse material systems [4].

Co/CoO thin-film systems are among the most widely studied ones. In this material system, the crystal orientation of the layers strongly influences the EB, coercivity, and shape of the loop [5–7]. The samples investigated here were produced by molecular beam epitaxy (MBE) growth of 8 nm Co on CoO(100) substrates after the substrates had been irradiated by heavy ions (xenon or uranium) to induce defects in the AFM [8].

It is well known that defects at the FM/AFM interface and in the AFM bulk can strongly influence the EB [9–11]. Increasing the defect density of the AFM, e.g., by ion irradiation, has thus been tested in several studies. Bombardment with light ions, such as He, influences the FM/AFM interface and can thus increase the EB [12,13], while heavy ions penetrate deeper into the material. Lisha et al. found a fluence of 10^{12} Ag ions/cm^2 to lead to maximum EB in zinc ferrite/FeNiMoB bilayers [14]. Demeter et al., on the other hand, showed that oxygen ion implantation in FCC cubic Co films created an EB due to Co_xO_y formation [15].

The investigation of these EB systems was typically performed by techniques such as superconducting quantum interference device (SQUID) [9,12] or magneto-optical Kerr effect (MOKE) [16], sometimes combined with simulations. Only a few papers have compared different techniques [17], and in most cases there were no significant differences between the results of these measurements.

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Here, we compare measurements of the bulk magnetization by SQUID with surface magnetization measurements obtained by MOKE. Due to the high penetration depth of the heavy ions, we expect different findings for surface and bulk magnetization, including the possibility that besides the MBE-grown thin ferromagnetic layer on top of the sample, there may also be ferromagnetic material in the ion penetration channels, leading to different geometries of the FM/AFM interface. Indeed, our results show simple, nearly rectangular surface hysteresis loops, while SQUID measurements of the bulk magnetization at low temperatures reveal unusual shapes in the hysteresis loops.

2. Materials and Methods

The samples were prepared by high-energy ion irradiation of CoO(100) substrates of 0.5 mm thickness (lateral dimensions 10 mm · 5 mm) with uranium or xenon ions, using rates of 10^9 ions/cm^2 (samples U2a, Xe2a) or 10^{10}/cm^2 (samples U3a, Xe3a), leading to defects of up to 10–100 μm in depth [8]. Afterwards, 8 nm Co(100) was grown by molecular beam epitaxy (MBE), followed by a 5 nm Cu cap to avoid oxidation. It should be mentioned that the large ion implantation depth resulted in holes in the subsequently grown ferromagnetic layer and cap, so that the FM layer was also partly disturbed. A sample containing as-purchased CoO(100) substrate served as the reference (sample CoO). The crystal orientations were verified by reflection high-energy electron diffraction (RHEED), as depicted exemplarily for sample U2a (Figure 1). These images show that both the substrate and the Co layer are still epitaxially grown along large parts of the sample, in spite of the holes caused by ion irradiation.

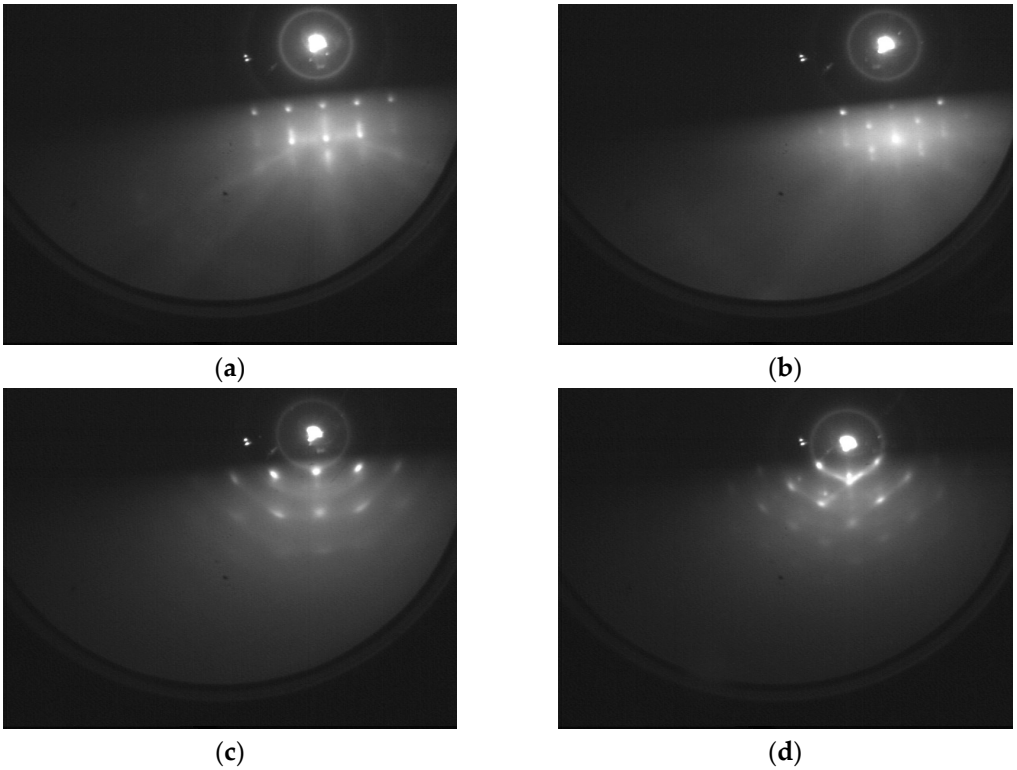


Figure 1. Reflection high-energy electron diffraction (RHEED) images of U3a: (**a**) CoO substrate along [010] and (**b**) [011] direction; Co layer along (**c**) [010] and (**d**) [011] directions.

The samples were investigated by a custom-built MOKE setup with a diode bridge technique [3] at room temperature, measuring surface magnetization, as well as by a magnetic property measurement system MPMS 3 magnetometer (Quantum Design) in VSM (vibrating sample magnetometer) mode, measuring the overall (bulk) magnetization

of the sample. The samples were cooled down from 350 K in an external magnetic field of 1 T prior to the temperature-dependent hysteresis loop measurements.

3. Results and Discussion

Figure 2 shows exemplary MOKE measurements of the longitudinal and transverse magnetization components at room temperature, measured on the U2a (Figure 2a) and Xe3a (Figure 2b) samples. While both longitudinal hysteresis loops show similar coercive fields and a similar shape, the transverse magnetization curves differ significantly. For sample U2a, there is no transverse signal visible, indicating that magnetization reversal may happen via domain wall processes.

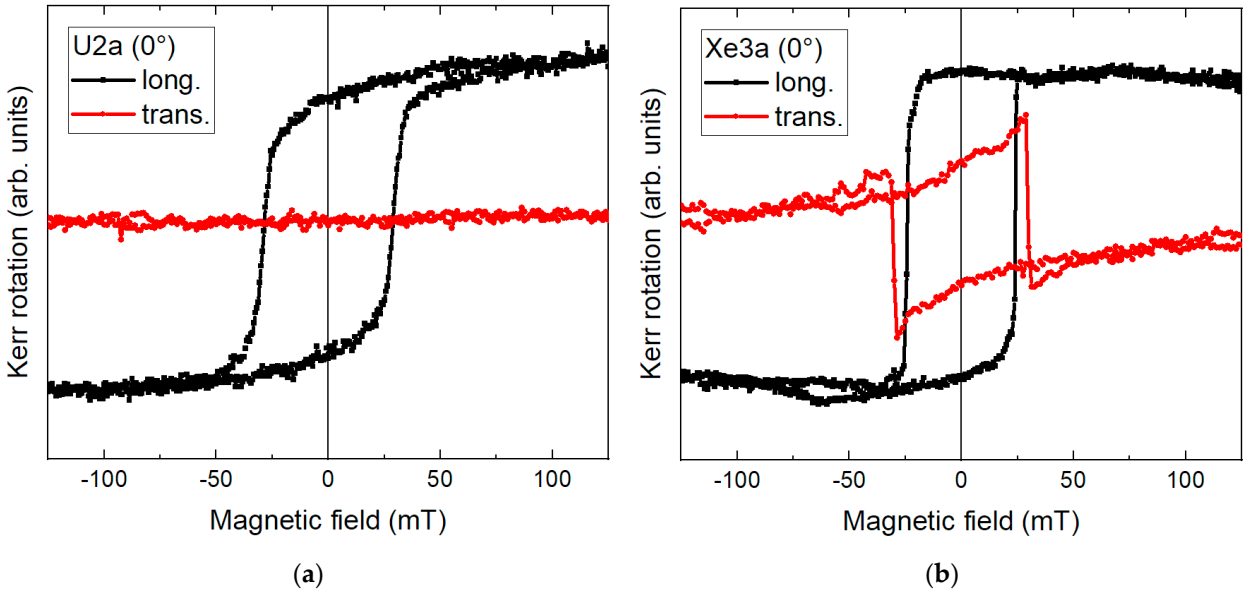


Figure 2. Magneto-optical Kerr effect (MOKE) measurements at room temperature: (**a**) sample U2a; (**b**) sample Xe3a.

For sample Xe2a, however, there is a strong transverse signal, showing coherent magnetization rotation as the magnetization reversal process. Apparently, the higher ion irradiation dose, resulting in a stronger dilution of the AFM, has changed the magnetization reversal, as has been described in the literature on previous occasions [9,10,12].

Figure 3 depicts MPMS measurements obtained at different temperatures, comparing room temperature (i.e., a temperature above the Néel temperature of the AFM, where no EB is expected) with a measurement at 10 K, i.e., far below the Néel temperature, where an EB is possible.

For the sample with lower dilution of the AFM, Xe2a (Figure 3a), very little EB is observed at low temperature, and there is no vertical shift, which is often found in EB systems. The deviation of the saturation at low temperature can be attributed to the strong underground signal from the AFM substrate which has to be subtracted and which increases the measurement error for larger external magnetic fields.

On the other hand, the sample with stronger AFM dilution, U3a (Figure 3b), shows not only a clear horizontal shift in the hysteresis loop at low temperature, i.e., an EB, but also a strong vertical shift in the loop due to the pinned magnetic moments [18,19]. Unexpectedly, this vertical shift also occurs at room temperature, i.e., above the blocking temperature of the AFM. This effect will be investigated in more detail in future experiments.

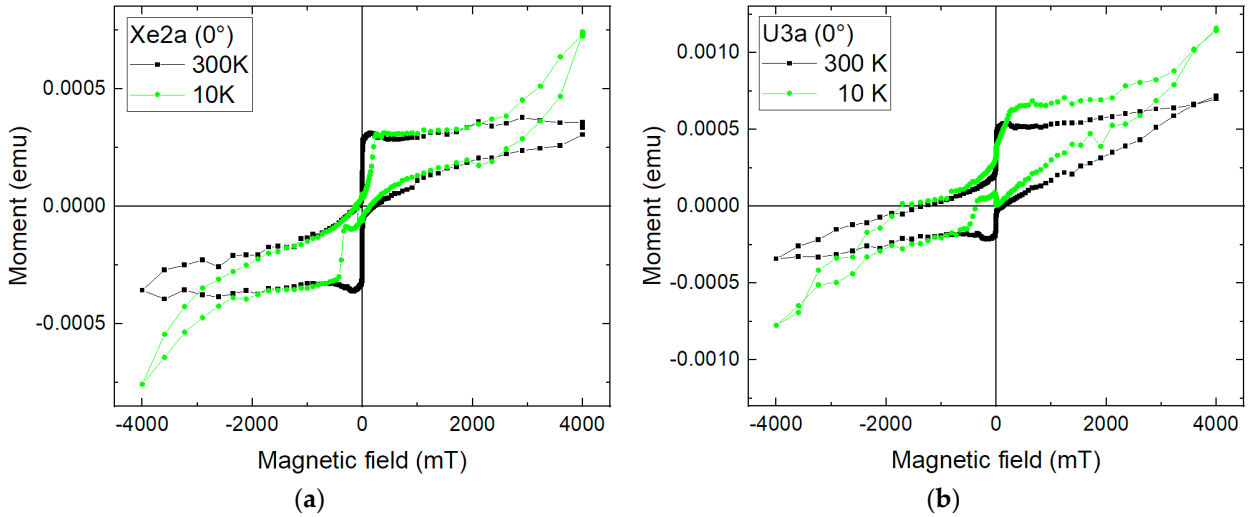


Figure 3. MPMS measurements at different temperatures: (**a**) sample Xe2a; (**b**) sample U3a.

It should be mentioned, however, that a horizontal shift in the hysteresis loop can also be measured without an exchange bias. On the one hand, the strong linear background of the thick antiferromagnetic substrate, which has to be subtracted to make the ferromagnetic hysteresis loops visible, may cause a gain error proportional to the magnetic field. On the other hand, the hysteresis loops may not be completely saturated, as the open ends indicate, meaning that a minor loop is measured [20,21]. Finally, it should be mentioned that Co/CoO systems in (100) orientation are known to show a change in the 90° coupling between FM and AFM in a temperature range of around 280 K to 320 K [4], while the Néel temperature of CoO is around 280–310 K, as reported in different studies [22]. Both effects may result in an asymmetric magnetization reversal, as found previously in Co/CoO thin film systems in (100) orientation [23]. To investigate the reliability of the apparent EB, measurements obtained with more precise equipment and potentially larger external magnetic fields are necessary.

More generally, these strong differences between bulk and surface magnetization can be attributed to ferromagnetic material inside the bulk, which is located along the irradiation channels, leading to differently shaped ferromagnetic domains and FM/AFM interfaces, as compared to the flat FM thin film on top of the AFM bulk. Such unusual FM/AFM interface shapes with thin FM-coated tubes inside an AFM bulk may be regarded as a system between point-like FM implantation [15] and exchange-biased nanotubes [24,25].

While samples irradiated with different ions and different irradiation doses are compared in greater detail in this paper, a preliminary study by SQUID (field range $\pm$ 5 T) showed a clear dependence of the exchange bias at low temperatures on the irradiation dose, while the heavy ions did not significantly influence the EB (Figure 4). In addition, the blocking temperature was the largest for the sample CoO8 with unmodified CoO substrate. It should also be mentioned that the smaller irradiation doses (U2a, Xe2a) even reduced the exchange bias at a low temperature.

In addition, it should be mentioned that the surface magnetization at room temperature, measured by MOKE, and the bulk magnetization at the same temperature, measured by MPMS, differ strongly, showing that both techniques can be complementarily used to achieve a comprehensive understanding of magnetization reversal processes throughout the whole magnetic system.

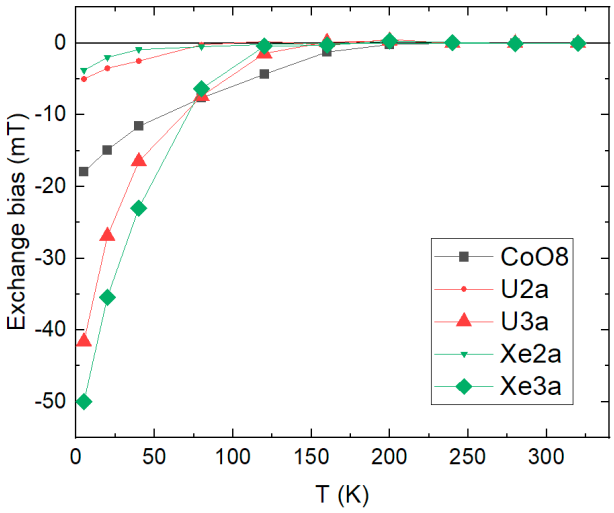


Figure 4. Preliminary SQUID measurements of the temperature-dependent exchange bias of the samples under investigation.

4. Conclusions and Outlook

Heavy ion irradiation of an AFM substrate was shown to change the magnetization reversal processes in exchange-biased CoO/Co samples. The samples with higher ion irradiation doses show a strong vertical shift even above the blocking temperature of the AFM which can be attributed to pinned AFM moments.

In addition, surface and bulk magnetization reversal processes differ strongly, as identified by MOKE and MPMS, respectively. Both techniques can be combined to fully understand magnetization reversal throughout the whole magnetic system.

More research is necessary to understand the influence of heavy ion irradiation on the magnetic characteristics of AFM substrates.

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