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Letter to the Editor

Anomalous magneto-optical behavior of uniaxial Co/CoO bilayer films

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ABSTRACT

We study the magneto-optical properties of thin epitaxially grown Co/CoO bilayer films with in-plane uniaxial anisotropy by means of Magneto-Optical Kerr Effect (MOKE) measurements. The magneto-optical response obtained from these samples exhibits a surprising and atypical behavior, in that the polarizer setting, at which signal inversion occurs, is a function of the sample orientation. This behavior even occurs for identical orientations of the magnetization with respect to the plane-of-incidence. Using Generalized Magneto-Optical Ellipsometry, we show that the origin of this behavior is the presence of optical anisotropy, which we attribute to the strained epitaxial CoO overlayer in our samples.

1. Introduction

Magneto-optical (MO) measurements, such as Magneto-Optical Kerr Effect (MOKE) spectroscopy and ellipsometry, have become important characterization techniques in magnetism [1]. Magneto-Optics has been also utilized to develop well-established technologies like MO recording [2], optical isolators [3,4], as well as novel applications within a wide range of different fields. Only the most recent ones alone cover topics from biosensing [5–7], magnetoplasmonics [8], nanomagnetism [9,10], ultrafast magnetization dynamics [11–13], magnetization reversal processes [14–16], to the anomalous behavior arising from the exchange bias effect [17–20], for instance.

In the vast majority of MO studies and applications, one makes the assumption that the MO effect is described by only one complex MO coupling constant Q, which leads to a modification of an otherwise isotropic dielectric tensor [21]. This comes from the fact that optical and especially MO anisotropies are usually not observed in metallic systems and thus not considered in the analysis of MO signals [22]. However, while this symmetry assumption is correct for cubic materials, it is generally not justified for materials with lower types of symmetry, such as uniaxial hcp Co, for instance. One has to be aware that non-cubic materials may exhibit optical or MO anisotropies that could induce a relevant modification of the MO response [23]. This is precisely what we find in this work and we show that the origin of this behavior is the presence of optical anisotropy, which we attribute to the strained epitaxial CoO overlayer in our samples. In addition, we demonstrate that Generalized Magneto-Optical Ellipsometry (GME) can be applied to unravel the relative orientation of the ordinary and extraordinary optical axes that describe the anisotropic material, and so, we prove it to be an efficient tool for studying material structures that are simultaneously magneto-optically active and optically anisotropic.

2. Results and discussion

The samples employed in this study consist of thin Co/CoO bilayer films fabricated by means of sputter deposition onto HF acid-etched Si (110) substrates. To achieve a good epitaxy as well as a suitable crystalline order, we followed a specific growth sequence consisting of a 75 nm Ag/50 nm Cr/30 nm Co multilayer structure. Due to this specific sequence, the hexagonal closed packed (hcp) Co films exhibit an in-plane orientation of the crystalline *c*-axis [24], which represents the easy axis of magnetization. After the sputter deposition, the sample was exposed to O_2 in order to induce a partial oxidation of the topmost Co-layers and form a CoO overlayer. This process yielded epitaxial CoO layers of approximately 3 nm thickness, which was measured via X-ray diffractometry and electron microscopy [25].

The MO characterization of our samples was carried out by means of a crossed polarizer MOKE-setup, which is well documented in the scientific literature and very frequently used [26,27]. This technique utilizes only two linear polarizers and a photo-detector for its optical setup. The first polarizer, which is placed in front of a laser whose light is aimed onto the sample under investigation, is set to p-polarization ($\varphi_1 = 0^\circ$), whereas the second polarizer in the reflected light beam path can be freely oriented (angle φ_2) to vary the signal level and the signal-to-noise ratio. The MO response is then obtained by measuring the light intensity via a photo-detector behind the second polarizer as a

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function of an applied magnetic field, which is oriented in the longitudinal configuration, i.e. in the plane-of-incidence and the sample plane. Typically, one finds optimal conditions for φ_2 as 1–2° away from s-polarization, depending on the total light intensity and the photo-detector and amplifier characteristics [28]. Usually, to obtain the most reliable MO measurement, several hysteresis loop measurements are done for different φ_2 orientations until the signal-to-noise ratio is optimized. Examples of this procedure are depicted in Fig. 1, where we plot two sets of normalized hysteresis loops for two different orientations of the same sample (denoted by the sample orientation angles Δ_1 and Δ_2). For the first one, the angle Δ between the easy axis of magnetization and the applied field direction is $\Delta_1 = -22.5°$ (see insets on the top of Fig. 1). The set of hysteresis loops corresponding to Δ_1 shows a minimum of the reflected light intensity in the immediate vicinity of $\varphi_2 = 90°$, which is the crossed polarizer condition and results in the fact that the signal-to-noise ratio becomes visibly worse. As can be seen from Fig. 1, this is also the configuration for which the hysteresis loop signal inverts due to the fact that $\varphi_2 - 90°$ acts as a bias angle for the MO signal detection, leading to a sign dependent summation of this very bias angle with the Kerr rotation angle [28].

For a second experiment, the sample orientation was changed to $\Delta_2 = +22.5°$, so that the magnetic easy axis has the same distance to the plane of incidence as in the first set but with opposite direction.[1] Given that all relevant measurement conditions are the same, one should expect to find not only very similar loop types, but also the compensation point at the same φ_2 orientation, which is obviously not the case. The smallest light intensity and lowest signal-to-noise ratio loop occurs at a shifted angular position, $\varphi_2 = 91°$. This result represents an anomalous MO behavior, inconsistent with conventional descriptions of the MOKE effect and associated data analysis schemes [26–28]. To find the origin of this anomalous behavior, it is necessary to employ a technique that provides more complete information on the optical and MO response. Thus, we have utilized GME, a reflection based technique that allows for a complete optical and MO characterization [29,30]. The experimental setup is fundamentally the same as the aforementioned one, but instead of using a fixed orientation for the first polarizer, this technique obtains multiple hysteresis loops as a function of both polarizer orientations (φ_1, φ_2). From these data, the light intensity change under magnetic state inversion ($\delta I/I$) is calculated for any particular value of the magnetic field, resulting in a series of $\delta I/I(\varphi_1, \varphi_2)$ datasets that can be conveniently plotted as maps. Given that analytical descriptions of these types of measurements have been derived, the methodology allows for least-squares (LSQ) fits of experimental data to determine the reflection matrix, optical and MO materials constants, as well as perform vector magnetometry [30,31].

Fig. 2 shows a series $\delta I/I(\varphi_1, \varphi_2)$ datasets in color map representation that were measured on the Co/CoO bilayer films. The angular step-width for both polarizers was 0.5° in our experiments and the color-scale is the same for all the maps. All datasets here were obtained for largest field value ($H = 1.7$ kOe) in our experiment, i.e. near saturation for the Co-film. The difference between the different datasets is the sample plane orientation angle Δ, which was varied in steps of 30°. The insets in Fig. 2 show the relative angle of the magnetic easy axis of the sample with respect to the magnetic field direction. Essentially, all maps show a very similar structure composed of two lobes, which are opposite in sign and aligned along the diagonal where the polarizers keep a relative angular orientation of 90°, irrespective of their absolute orientation in reference to the plane of incidence. As guide to the eye, we have also highlighted the inversion symmetry point of these structures as a blue dot as well as its angular evolution with a dashed curve. The sample rotation reveals the anomalous MO behavior of Fig. 1 as a shift of this symmetry point in terms of its φ_2 location. The maximum change is obtained for $\Delta \approx 60°$. Fig. 2 also reveals that the symmetry point shift exhibits a 180° periodicity for Δ. The location of the symmetry point also shifts in φ_1, with identical periodicity but smaller amplitude making this effect less visible.

The experimentally observed behavior cannot be ascribed to alignment inaccuracies of our experimental setup, since a small tilt of the rotation axis out of the plane of incidence should cause a 360° variation instead of the observed 180° periodicity. Furthermore, we have determined the maximum shift of the polarizer crossing point due to such misalignment to be less than $\pm 0.1°$. It is thus clear that the angular dependence of the $\delta I/I$ datasets shown in Fig. 2 must be related to the properties of the sample itself and, whatever the origin of this effect may be, it is uniaxial

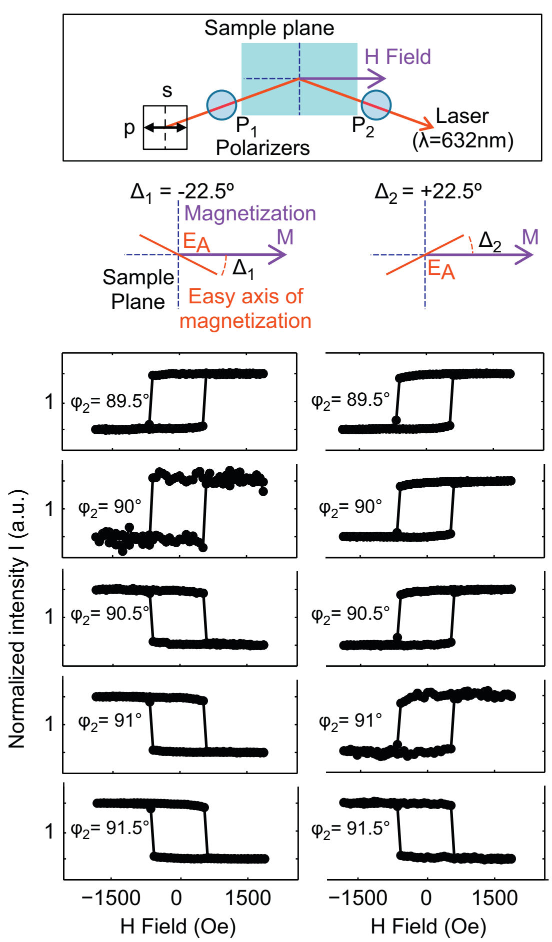

Fig. 1. (Top) Scheme of the Generalized Magneto-Optical Ellipsometry experimental setup. (Bottom) Normalized experimental hysteresis loops obtained as a function of the analyzer φ_2 angle: (left column) for orientation $\Delta_1 = -22.5°$; (right column) for orientation $\Delta_2 = +22.5°$. The polarizer orientations are common to both sets (the first polarizer is fixed to $\varphi_1 = 0°$ in all cases). The schematics on top of the data sets define the geometry, specifically the orientations of the easy magnetic axis with respect to the plane of incidence.

[1] Since the magnetization reversal process and thus the hysteresis loop shape depends on the relative angle between the easy axis of magnetization and the magnetic field direction, hysteresis loops generally differ for different sample orientations. Therefore, we chose two orientations here that produce very similar loop shapes to avoid distractions from the main point of Fig. 1. Only the transverse component of the magnetization is different for the two sample orientations here, a component to which our MOKE-setup is nearly insensitive in the configuration that we used.