Combining high temperature sample preparation and in-situ magnetic fields in XPEEM

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ABSTRACT

We present a custom-made sample holder system for use in Elmitec Low Energy and PhotoEmission Electron Microscopes. It consists of two different sample holder bodies: one with a filament for high temperature measurements (up to more than 1500 K) and the other with integrated electromagnets for the in-situ application of in-plane/out-of-plane small magnetic fields. The sample is placed on a platelet which can be transferred between the two holders. This opens up new possibilities for the preparation of samples at high temperatures and investigation of their behavior under applied magnetic fields without leaving the ultra high vacuum system.

1 Introduction

The study of static and dynamic properties of magnetic domains in ferromagnetic nanostructures has recently attracted a lot of attention beyond its fundamental interest due to the emergence of technological applications based, for example, on domain walls (DWs) and their dynamics in memory\textsuperscript{1–5} and logic devices\textsuperscript{6–9}.

Consequently their behaviour under applied magnetic field\textsuperscript{10–13} or spin-polarized electric currents\textsuperscript{14,15} as a driving force has been the subject of extensive investigations. For most spintronic applications, thin films or lower dimensionality structures are required. Thus there is a strong motivation to understand and control the growth of ferromagnetic films and nanostructures and probe the modification of their magnetic properties by applied magnetic fields.

The X-ray PhotoEmission Electron Microscopy (XPEEM) experimental station at the CIRCE beamline of ALBA Synchrotron\textsuperscript{16} possesses capabilities for the in situ observation of growth processes with high spatial and temporal resolution as well as for the chemical and magnetic characterization of surfaces, thin films and nanostructures. Optimization of the growth parameters and thus preparation of high quality thin films is therefore possible in the same microscope where their structural, chemical and magnetic study is carried out by a variety of spectromicroscopic techniques based on low-energy backscattered as well as photoemitted electrons excited by the beamline x-rays (LEEM and XPEEM, respectively). At ALBA, a suite of state-of-the-art sample holders and electronics provides a functional sample environment for applying electric and small magnetic fields, electrical signals (current pulses)\textsuperscript{17}, and time resolved measurements with sub -100 ps time resolution\textsuperscript{18,19}. The newly developed sample holder system enables the modification of the magnetic structure of the samples by applying magnetic fields in different directions despite the high temperatures involved in their preparation and the necessity to avoid air exposure.

In this article, we describe a custom-made exchangeable sample holder system comprising of two sample holder bodies. The first one, based on a modification of the standard Elmitec sample holder, is equipped with a filament for electron-bombardment that can be heated to 1500 K, and the second one is customizable with integrated electromagnets for in situ applications of small magnetic fields either in-plane or out-of-plane. An adapter for hosting sample platelets is mounted on both bases in order to provide a fast and convenient way to transfer the sample between them without leaving the ultra-high vacuum system. First experiments with the new sample holder system are presented in order to illustrate its capabilities: microstructures of ferrimagnetic Fe\textsubscript{3}O\textsubscript{4} were grown on a Ru(0001) substrate\textsuperscript{20,21} by high-temperature oxygen-assisted MBE and the switching of the individual magnetite microstructures under applied field was subsequently imaged at a resolution of tens of nm by means of XMCD-PEEM.

2 Design

The exchangeable sample holder system is designed to be compatible with the Elmitec microscopes widely used at synchrotrons. Drawings and pictures of the different parts are shown in Figure 1. There are two different sample holder bodies: one for high
temperature (Figure 1a-b) and one for magnetic field applications (Figure 1c-d). The former includes an electrically insulated thoriated tungsten filament for electron bombardment heating up to more than 1500 K and two electric contacts for a W/Re thermocouple. On the top there is an adapter for flag-type sample holder platelets of the same dimension as those of Omicron or Specs (Figure 1e), which can be inserted in UHV by means of a wobble stick. The sample itself is pressed between the platelet and a top plate with a central hole of variable diameter. The platelets are held together by short sink-head screws and are held in place by W springs which ensure stability irrespective of holder orientations and strong temperature variations.

Figure 1. 3D drawings (a) and picture (b) of the high temperature sample holder base with the electrically insulated filament in the center. 3D drawings (c) and picture (d) of the electromagnet sample holder base with an uniaxial in-plane electromagnet. (e) Platelet transferred between both holder types.

The second sample holder, shown in Figure 1c-d, features an integrated electromagnet for a uniaxial, in-plane magnetic field. It consists of a monolithic Permendur yoke and a coil wound from Kapton-insulated 0.14 mm diameter copper wire. With $2 \times 350$ windings and 3 mm gap it is possible to reach 50 mT/A. Although the available power supply provides $\pm 1$ A, the maximum field which can be applied during an experiment is limited by different factors such as the pressure increase due to wire heating and image distortion due to electron deflection. In practice we typically apply a maximum of 0.3 A for extended periods (i.e., imaging) and higher currents (up to 1 A) in the form of short pulses for switching or setting a certain magnetic state. The magnetic field produced in this assembly is measured with a Hall probe (Projekt Elektronik) with an active area $2 \text{ mm} \times 1 \text{ mm} \times 1 \text{ mm}$ as is shown in Figure 2. In the main panel, the measured magnetic field at the sample surface position is plotted (0.5 mm above the yoke plane). The field direction can be switched by 180 degrees, changing the current sense. In the upper left inset in Figure 2 the measured field B as a function of the in-plane position along the two principal axes is shown. Along the gap (X direction), the center represents the minimum of the field, while in the lateral (Y direction) the maximum field is at the center. In the vertical direction (lower right inset (Z axis)), the maximum of the field is at the yoke gap tips (0 mm), while at the sample position it is slightly reduced (typically 0.5 mm above the yoke).

The transfer between different sample holder bases is performed keeping the base on the standard main transfer bar and removing/inserting the sample platelet with a commercial magnetic wobble stick at 90 degrees to the transfer bar.

The complete assembly was tested in the ALBA XPEEM, operating at the most demanding conditions in terms of vacuum (sample at -20 kV). Both sample holder bodies were outgassed in UHV ($10^{-9}$ mbar) prior to the experiments, and the XMCD images with applied magnetic field were acquired with up to 0.25 A applied current. We enunciate that this sample holder system has been designed to be modular and highly customizable, e.g., for other electromagnet geometries. It is also compatible with the vacuum suitcase and could thus be extended to exchange samples with other systems such as the ALBA NAPP22, Boreas beamline23 or lab-STM. The vacuum suitcase is a custom-made miniature UHV chamber with a battery-powered iongetter pump, ending in a CF 40 gate valve. It includes a parking stack for several flag style platelets on a linear manipulator. It is mounted vertically on the PEEM experimental station and accessed by the same wobble stick used to transfer platelets between sample holder bases.

Representative data obtained are described in detail in the next section.
Figure 2. Measured magnetic field produced by the in-plane sample holder in the sample surface position (0.5 mm above the yoke plane) as a function of current. Left upper inset: field measured for 0.1 A when displacing the Hall probe along different in-plane directions (shown in the right panel). Right lower inset: measured field as a function of Hall probe position (for 0.1 A) as a function of height.

3 Experimental results

3.1 Growth of ultrathin magnetite films on Ru(0001)

Figure 3. (a) LEEM image at 5 eV electron energy and (b) microspot LEED pattern at 45 eV of the clean Ru(0001) substrate. (c) LEEM image of magnetite crystals grown on Ru with insets showing µ-LEED patterns of (d) a micrometric wide island and (e) the surrounding wetting layer.
The new sample holder system was tested while investigating the high-temperature growth of thin magnetite films on a Ru(0001) single crystal surface and the imaging of their domain landscape under different in-plane magnetic fields. The Ru substrate was sputtered with Ar$^+$ ions at 1.5 keV prior to the growth in order to remove any residual previous films. It was then annealed to 1200 K in 10$^{-6}$ mbar molecular oxygen and flashed to 1500 K in vacuum in order to remove the carbon diffusing from both the bulk and the surface oxide, respectively. Since the thermocouple is attached to the body of the cartridge, the real temperature of the sample cannot be extracted from TC measurements for short-time flashes. Thus the temperature was calibrated using a pyrometer placed in front of the sample surface in the preparation chamber. Figure 3a is a typical LEEM image of the clean Ru substrate with large flat terraces separated by monatomic steps (darker curved lines). The corresponding low energy electron diffraction (LEED) pattern is shown in Figure 3b. Magnetite was grown by oxygen-assisted high temperature MBE, as described elsewhere\textsuperscript{20}. As already reported, initially small islands start to nucleate and grow on Ru(0001). When they coalesce, the entire surface is covered by a so-called wetting layer with FeO composition, on top of which the growth proceeds in the form of 3-dimensional islands of magnetite (Fe$_3$O$_4$). The growth process in total took one hour during which the sample was kept at 1150 K. Figure 3c is a LEEM image of a couple of magnetite islands grown by this method at 1150 K on Ru(0001). The very different electron reflectivity of the islands and wetting layer in the LEEM image reveal the different nature of both structures. The crystallographic structure of the wetting layer and the island were confirmed by $\mu$-spot LEED measurements, as shown in Figure 3d-e. The wetting layer presents a 1x1 pattern with characteristic satellite spots arising from the in-plane mismatch between the substrate and the wetting layer. The resulting in-plane lattice spacing is 0.32 nm, which is in agreement with the value reported for FeO on Ru(0001)\textsuperscript{24}. In contrast, the large triangular islands present additional spots in 2x2 position characteristic of the Fe$_3$O$_4$(111) oxide surface, related to the larger unit cell of the spinel structure. The island’s oxygen lattice spacing obtained by LEED is similar to the wetting layer, 0.31 nm. The additional closely spaced satellite spots from the islands are assigned to a surface reconstruction of magnetite, the bi-phase reconstruction for which a recent model has been proposed\textsuperscript{25}. Thus we confirm that the film grown consists of magnetite crystals surrounded by a FeO wetting layer, as is the case for growth in the same conditions using the standard Elmitec high temperature sample holder.

3.2 Evolution of magnetic domains in magnetite microstructures

In the next step, the Fe$_3$O$_4$/Ru sample was transferred to the electromagnet holder without leaving the ultra high vacuum system.

![Figure 4. XAS (top) and XMCD spectra (bottom) at the Fe L-edge from the island shown in Figure 3c](image)

In order to gain insight into the chemical nature of the islands, X-ray Absorption Spectra (XAS) were measured at the Fe L$_{1,2}$ edges. Figure 4 shows the Fe XAS and X-ray Magnetic Circular Dichroism (XMCD) spectra from the island shown in Figure 3c. The Fe L$_3$-edge XMCD spectrum has a three-peak structure arising from the different iron cations (Fe$^{2+}$ and Fe$^{3+}$) located in octahedral and tetrahedral sites within the spinel structures, typical for magnetite\textsuperscript{26}. Next, the magnetic domains were mapped by means of (XMCD) PEEM images at the Fe L$_3$ absorption edge. The magnetic images were obtained by subtracting XAS images for opposite light polarizations at a photon energy that enhances the XMCD.
contrast (Fe $L_3$ absorption edge). Figure 5a shows the magnetic domain distribution of the magnetite microstructures following a demagnetization cycle performed by the built-in electromagnet (applying a decaying alternating magnetic field to a sample). A clear magnetic contrast is observed in the magnetite island (red and blue areas indicate different orientations of the magnetization with respect to the x-ray beam incidence direction, i.e., different ferrimagnetic domains) while the wetting layer does not show any magnetic contrast at room temperature.

Finally the magnetic state of a single magnetite island was changed by applying short magnetic field pulses. The evolution of the magnetic domains under applied field is shown in Figure 5b,c. It can be seen that even with a small field (10 mT) the distribution of the domains is changed. By increasing the value of $B$, the domains tend to align in the direction along the applied field. In order to check the reversibility of this process, the maximum possible field was applied in the opposite direction (Figure 5d). The magnetization inside the island shown in Figure 5d is almost fully reversed, with some residual opposite contrast around the island edges possibly nucleated when returning to zero field in order to measure in remanence. The domains have a nearly one to one inverse correspondence with those of Figure 5c when the magnetic field with the same value was applied in the opposite direction, suggesting that some of the domain walls are pinned to defects on the in-situ grown Fe$_3$O$_4$.

![Image of magnetic domains](image.png)

**Figure 5.** Magnetic domains of an Fe$_3$O$_4$ islands after different applied pulses of in-plane magnetic field, according to the XMCD contrast at the Fe $L_3$-absorption edge: (a) after the demagnetization cycle. After a short pulses of magnetic field of (b) -10 mT, (c) -52 mT and (d) 52 mT. Red and blue correspond to the two different antiparallel in-plane magnetization directions, white lines indicate the presence of a domain wall.

### 4 Summary

We have presented a new custom made exchangeable sample holder system for LEEM/PEEM consisting of a sample holder body for high-temperature measurements and another one with an integrated mini-electromagnet for the in-situ application of a magnetic field. We have demonstrated that it is possible to clean a Ru(0001) surface and perform growth at high-temperature by molecular beam epitaxy. Thin magnetite films on Ru(0001) were grown and characterized in-situ. Furthermore, we have shown experimental data confirming that by applying an in-plane small magnetic field, it is possible to change the magnetic state of ferrimagnetic thin films of Fe$_3$O$_4$ at tens of nm spatial resolution. The results suggest that XPEEM together with the presented sample holder opens up new possibilities for the study of magnetic properties of surfaces, thin films and nanostructures requiring high temperature preparation and/or growth and cannot be exposed to air.

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### References