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Fast elemental mapping and magnetic imaging with high lateral resolution using a novel photoemission microscope

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Abstract

Using tunable soft X-ray synchrotron radiation and a new-generation photoemission electron microscope with integral sample stage and microarea selector, elemental images and local XANES spectra have been measured. Given the present conditions (PM3 at BESSY), the lateral resolution was in the range of 130 nm with the potential of considerable improvement with high-brilliance sources (a base resolution of 25 nm was obtained in threshold photoemission). Measurements at the oxygen K-edge demonstrate that differences in the local chemical environment of the emitter atom are clearly revealed and can thus be used as a fingerprint technique for its chemical state and geometrical surroundings. By exploiting the magnetic circular dichroism effect it was possible to view magnetic domains and domain walls. © 1998 Elsevier Science B.V.

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1. Introduction

Owing to the development of modern electron storage rings as dedicated light sources, tunable synchrotron radiation has matured into a very powerful tool for investigating the electronic and geometrical structure of solids and surfaces. In particular, X-ray absorption spectroscopy (XAS) is now widely used in this field because it has two important advantages. First, electronic transitions from core levels of atoms at the surface are element-specific and can thus be used in a chemical 'fingerprint' technique. In this way elemental distributions can be easily visualized [1–3]. Second, and no less important, the X-ray absorption near edge structure (XANES) and the extended X-ray absorption fine structure (EXAFS) techniques can probe the spatial atomic arrangement (i.e. bond distances and angles) around the selected emitter atom [2,4,5]. These techniques have thus become widely used in many branches of chemistry, physics and the life sciences.

For the investigation of heterogenous samples, spatial resolution is mandatory in order to select and

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define the region of interest. The most natural way to obtain a small probe spot is, of course, to focus the exciting photon beam down to the desired spot size. Indeed, laterally resolved experiments have made use of this technique. However, in cases where a two-dimensional structure is to be investigated, the 'imaging' of this structure requires scanning of either the photon beam or the sample, both being comparably slow. Stöhr [2] and Tonner et al. [6] combined the parallel imaging technique of photoelectron emission microscopy (PEEM) with XAS using synchrotron radiation. Thus X-ray absorption microspectroscopy and element-specific imaging became possible on a lateral resolution scale of about 0.5 μm. It is the purpose of the present paper to present new measurements using this technique with an improved lateral resolution of around 130 nm. Special interest was focused on a detailed analysis of the XANES features measured in the microspectroscopy mode and on high-resolution magnetic imaging.

2. The photoemission electron microscope

We report measurements with a new-type photoemission microscope (FOCUS IS-PEEM), employing the beamline SX-700-3 (PM3) at BESSY yielding circularly polarized radiation in the energy range 50–1000 eV. The IS-PEEM is a three-lens electrostatic photoemission microscope. It comprises an electrostatic tetrode objective lens, a piezomotor-driven contrast aperture (variable in five different sizes between 1200 and 30 μm) located in the back focal plane, an octopole stigmator/deflector for maximum resolution, a continuously variable iris aperture located in the first intermediate image (for background reduction in the high-resolution mode as well as area selection for microanalysis), and two projective lenses. The imaged lateral electron distribution is intensified by a multichannel plate and made visible by a fluorescent screen (YAG crystal). Fig. 1 shows a schematic view of the principal arrangement.

The diameter of the contrast aperture determines both the resolution of the instrument (through the spherical and chromatic aberration coefficients) and also the intensity of the image (through its cross-section). In order to reach the ultimate resolution, the diameter of the contrast aperture must be chosen to be as small as possible. Additionally, the microscope must be operated with a stigmator which corrects non-spherical aberrations of the optics. The IS-PEEM uses an electrostatic octopole stigmator located in the back focal plane of the objective lens. The octopole arrangement ensures that astigmatism can be corrected in any rotational orientation, independently in the x/y orientation as well as in the $xy/-xy$ orientation. In addition, the octopole can be used as an x/y deflector in order to shift the field of view by about 1 mm without moving the sample. This can be useful for fine adjustment of the sample position as well as for optimization of the focusing of the field of view (compensation of small misalignments of the sample). With the SX-700-3 beamline, we obtained a lateral resolution of 130 nm at $h\nu = 540$ eV. Using a mercury lamp ($h\nu = 4.9$ eV), even a 25 nm lateral resolution was achieved (Fig. 2), since there is no contribution of the chromatic aberration due to the small energy width. More details of the experiment can be found in Ref. [2].

3. Elemental mapping and XANES microspectroscopy

By using soft X-rays in the energy range 50–1000 eV, characteristic core-levels become accessible and permit element- specific imaging by subtracting images, obtained at the photon energy of a characteristic absorption edge and at an energy some electron-volts below this. Because the IS-PEEM is further equipped with a continuously variable iris aperture, it acts as an efficient sample microspot selector, because both the image of the sample surface and the image of the hexagonal shape of the iris in the intermediate plane are simultaneously focused onto the screen. This 'microspot selector' allows the user to select well-defined sample areas down to about 1 μm in diameter for microspectroscopy purposes. A straightforward application of this operational mode is, for example, X-ray absorption spectroscopy (microXAS) by varying the excitation energy and recording the image intensity of a microspot with a CCD camera.

A result illustrating these techniques is shown in Fig. 3. The sample was a thin magnetite film

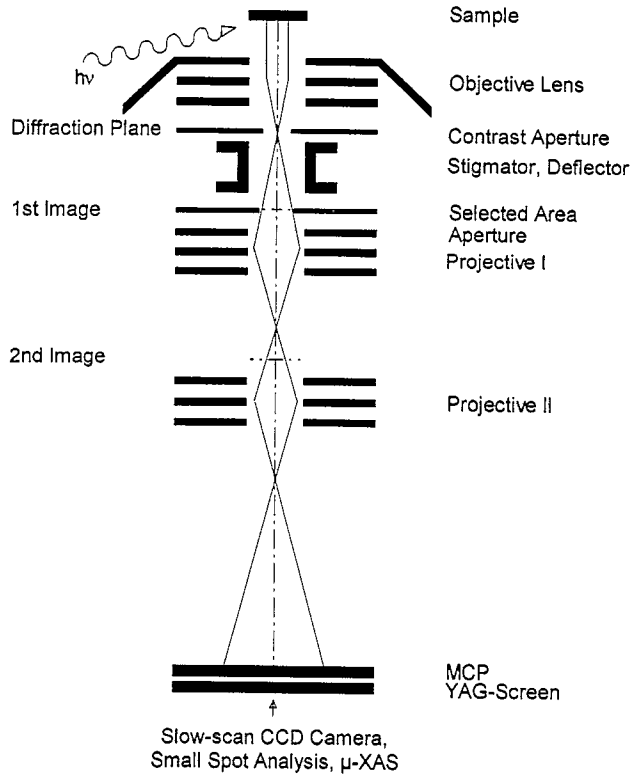

Fig. 1. Schematic view of the FOCUS IS-PEEM.

(Fe_3O_4) grown on the (100) face of a MgO single crystal platelet. The film was grown by molecular beam epitaxy (MBE) by electron beam evaporation from an Fe target at the Philips Research Laboratories in Eindhoven. During the growth, a substrate temperature of 225°C was used and the oxygen pressure was maintained at 2.8×10^{-5} mbar. The film was smooth over most of the surface area, with only a few small defects and scratches, one of which has been analyzed.

The lateral elemental distribution of Fe and O is shown in Fig. 3a and Fig. 3b. These images were obtained by subtracting from the images at 707 eV (Fe L_3-edge) and 543 eV (O K-edge) those recorded at photon energies just below these edges. Inside the scratch, the signal due to iron is diminished, but a high signal due to oxygen appears. This suggests that the Fe_3O_4 coverage in this region is thinner and the enhanced oxygen signal comes mainly from the MgO substrate.

The differences in the oxygen atom's chemical environment can be seen by close inspection of the fine structure in the near-edge absorption spectra. According to the well-known spectra of the O K-edge in MgO [7], at position D, where the scratch is so deep that the bare MgO surface appears, the near-edge structure shows the characteristics of oxygen chemically bonded to magnesium. At C and B, the magnetite layer becomes thicker and the features that are typical for MgO are significantly weaker. Finally, at position A the shape of the O K-edge shows that the magnetite layer is almost undamaged. Note the prepeak at 532 eV (arrow) from magnetite which is not present in spectra for D. This peak, which appears as a doublet (t_{2g} and e_g symmetry bands) at higher resolution due to crystal field splitting plus hybridization, can be attributed to oxygen 2p orbitals hybridized with the 3d metals of the transition metals. The broader region 10 eV above shows the mixing of the oxygen p states with the metal 4s and 4p orbitals. Further results for transition metal O K-edge XANES structures can be found in Ref. [8].

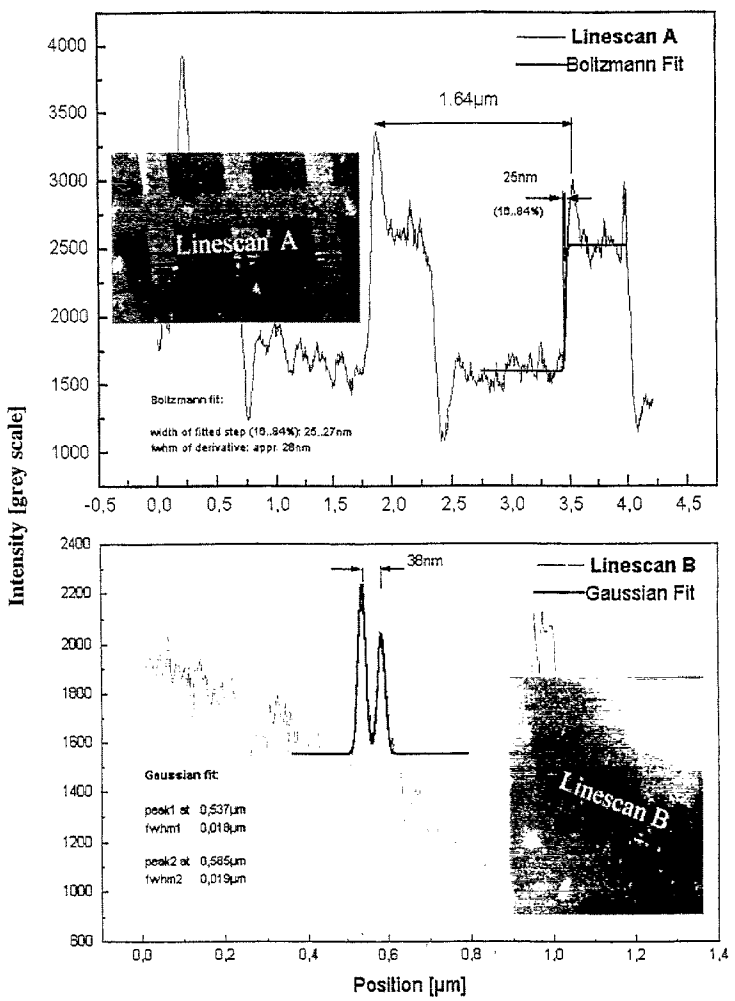

Fig. 2. Determination of the base resolution in threshold photoemission ($h\nu = 4.9$ eV). Upper: line scan across a microstructure of 20 nm gold (bright bars) on 10 nm chromium reveals an edge width of 25 nm (16% to 84% values). Lower: line scan across a pair of point defects at a distance of 38 nm reveals a half-width below 20 nm of each structure only.

This example illustrates the advantages of photoemission microscopes used for recording local XAS and XANES spectra. In addition to the possibility of identifying the chemical elements present on the sample surface, analysis of the near-edge structure reveals the lateral variation of the chemical environment, too.

4. Imaging of magnetic microstructures

In order to characterize the performance of the microscope for magnetic imaging, we investigated several microstructured samples using circularly polarized synchrotron radiation. The first sample was a 30 nm thick Permalloy film ($Fe_{19}Ni_{91}$), which had been deposited through a mesh onto a silicon wafer. This procedure resulted in a regular array of squares with a period of 25 μm (Fig. 4a). The second sample was a Co/Pt multilayer, also grown on silicon. This multilayer was patterned by ion etching with the same periodicity (Fig. 4b). Although being of similar shape, the structures show completely different magnetic domains. The images were recorded using the magnetic circular dichroism (MCD) at the Fe and Co

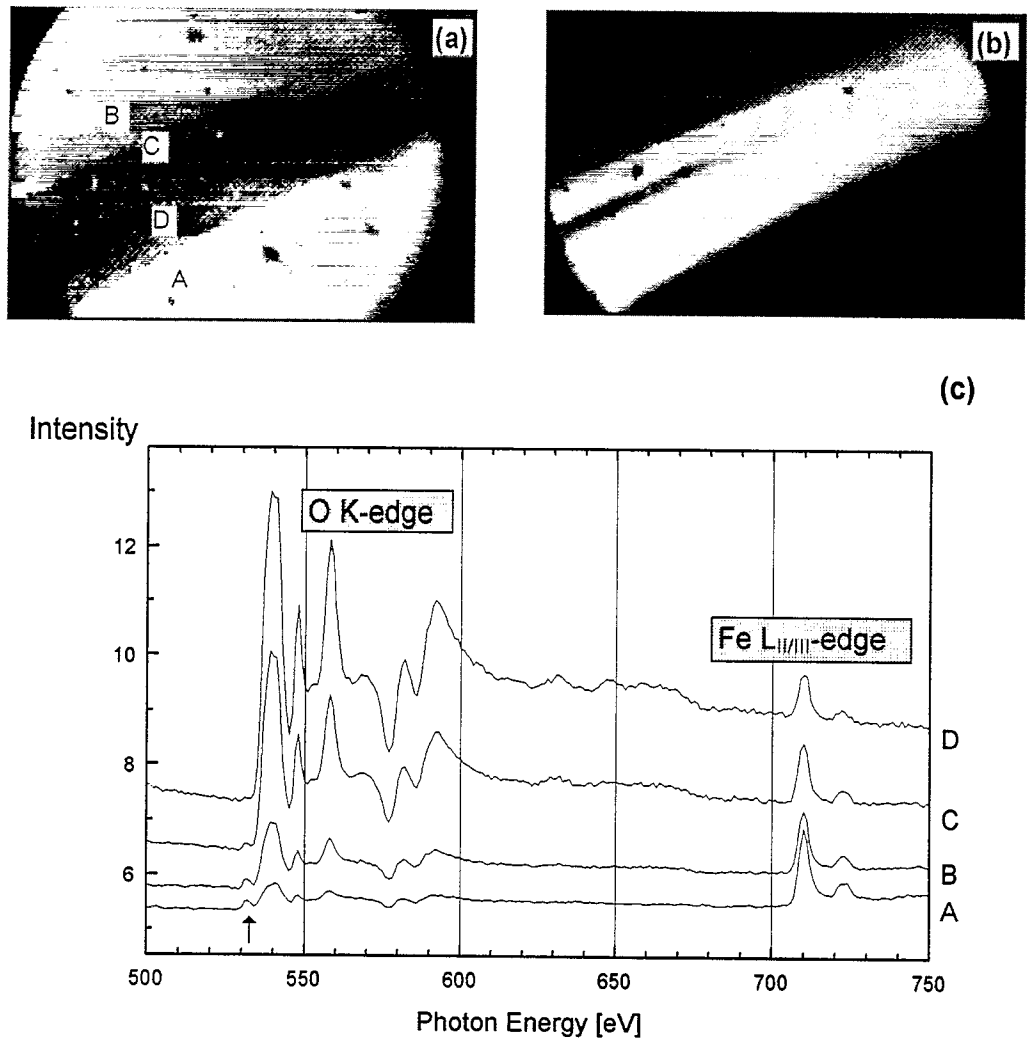

Fig. 3. Elemental imaging of (a) iron and (b) oxygen, and (c) X-ray absorption microspectroscopic analysis of a scratch in a magnetite layer on MgO. The spectra have been normalized to equal photon flux.

L_3/L_2-edges. Because the MCD signal reverses its sign when going from the L_3-edge to the L_2-edge, subtraction of images obtained at the corresponding wavelengths eliminates non-magnetic (chemical, topographical) contrasts and leaves only the magnetic domain pattern.

In the Permalloy pattern, we find an arrangement of four triangular domains, two of which are magnetized parallel and anti-parallel to the direction of the incoming photon beam $h\nu$. The other two domains are oriented perpendicular to this direction. In all cases, the magnetization vector lies within the surface plane. A reconstruction of the spatial orientation of $\boldsymbol{M}$ in one of the squares is indicated by the arrows, and shows a typical flux closure structure. This domain structure can be understood by the fact that Permalloy is a magnetically very soft material, i.e. with a small intrinsic magnetic anisotropy. For the given square, the tendency of the system to minimize the stray magnetic field shape yields the structure observed in the experiment. This ideal case may be easily destroyed by defects, as can be seen in the square in the center of the image. It shows a more complicated domain pattern, which is pinned to a structural defect on the

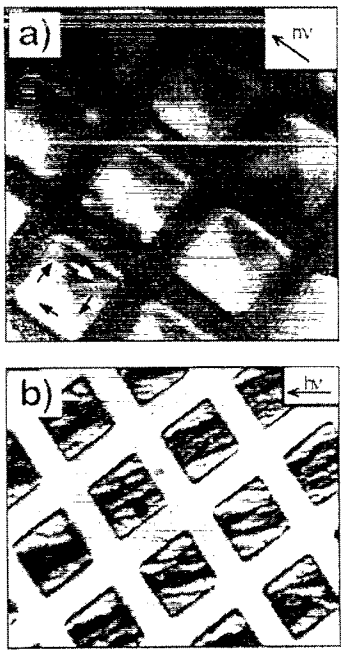

Fig. 4. Magnetic domains in microstructured samples (25 μm periodicity) of (a) Permalloy and (b) a Co–Pt multilayer (b) grown on silicon substrates. The images have been recorded using the MCD effect at the Fe and Co $L_{2,3}$-edges.

substrate. It should be noted that the same information about the magnetic microstructure is obtained when exciting at photon energies corresponding to the Ni $L_{2,3}$-edges.

The Co–Pt multilayer is known to have a very strong intrinsic magnetic anisotropy which is due to spin–orbit coupling (magnetocrystalline anisotropy). The filigree feather-like domain pattern within each square therefore reflects locally changing easy directions of the magnetization vector. A further difference observed from the Permalloy concerns the number of gray levels which reflect the possible spatial orientations of $\boldsymbol{M}$. A statistical analysis of the abundance of the various gray levels in the Co/Pt squares shows an almost continuous distribution, whereas one finds three distinct maxima for the Permalloy. This suggests that the magnetization vector in the Co–Pt system has a higher number of possible spatial orientations. This is compatible with the film being polycrystalline. In this case the easy axes of the magnetization—as determined by the magnetocrystalline anisotropy—change with the spatial orientation of the crystallite more or less randomly. The result is a rather complex variation of $\boldsymbol{M}$ along the sample surface, which is sometimes called a magnetization 'ripple' [9].

5. Summary and outlook

The examples presented in this paper show that the photoemission microscope has become a versatile tool for the detailed analysis of surfaces. A wide range of chemical information, starting from an elemental map to bond distances, bond angles or the geometrical reconstruction of adsorbates (if a systematic comparison of the absorption spectra with theoretical calculations is carried out), can be studied. The present lateral resolution is about 25 nm in threshold photoemission (work function contrast), 130 nm in the element-specific imaging mode, and 500 nm in the microspectroscopy mode (local XAS, XANES).

Using circularly polarized photon beams, magnetic circular or linear dichroism can be exploited in order to image magnetic microstructures with high lateral resolution ('domain imaging'), even chemically resolved. The photoemission electron microscope has been used for μ-ESCA studies, too [10].

On the basis of the present results, we estimated the future performance at third-generation synchrotron sources. It can be expected that time-resolved measurements with image sequences at 1 Hz and local XANES spectroscopy for the study of chemical reactions with 100 nm resolution can be carried out routinely. For high-resolution applications such as the imaging of magnetic domain walls, future experiments on magnetic systems may even be carried out in real-time, thus allowing the study of, for example, magnetization processes and magnetic relaxation phenomena.

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