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Superlattices and Microstructures 37 (2005) 99–106
Superlattices and Microstructures
www.elsevier.com/locate/superlattices

X-ray resonant magnetic scattering by Fe/Cr superlattices

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Received 3 June 2004; received in revised form 21 July 2004; accepted 21 July 2004

Available online 15 September 2004

Abstract

We have studied the structural and magnetic properties of an antiferromagnetically (AF) coupled Fe/Cr superlattice by means of soft x-ray resonant magnetic scattering. Strong and purely magnetic Bragg peaks are observed at the half-order positions in reciprocal space parallel to the [001] growth direction and in between the structural Bragg reflections from the superlattice periodicity. The magnetic hysteresis loops measured at the first-order and at the half-order Bragg peaks clearly demonstrate the strong AF coupling of the Fe/Cr multilayer. Transverse scans and off-specular reflectivity measurements confirm an AF domain structure of the superlattice in remanence with large perpendicular correlation. In addition, the transverse scan of the half-order Bragg peak exhibits a Lorentzian line shape at zero field, which diminishes in higher fields, indicative of a remanent multidomain state approaching a single-domain state towards saturation.

PACS: 75.25.+z; 07.85.Qe; 75.70.Cn; 61.10.Kw

Keywords: X-ray resonant magnetic scattering; Antiferromagnetically coupled superlattice

Magnetic heterostructures consisting of two or more ferromagnetic (F) layers separated by non-magnetic or antiferromagnetic (AF) spacer layers have received much attention due to their importance in fundamental science and technology. It has been shown that,

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doi:10.1016/j.spmi.2004.07.004

depending on the thickness of the spacer layer, ferromagnetic layers may be coupled parallel (ferromagnetically) or antiparallel (antiferromagnetically) via the interlayer exchange interaction [1–3]. The antiferromagnetic alignment of magnetic layers leads to a giant magnetoresistance effect (GMR) [4], which is used in magneto-electronic devices, such as GMR reading heads, spin valves and tunnel junctions [5]. The performance of these devices crucially depends on the chemical and magnetic structure, and especially on the quality of the AF/F interface.

It is well known that x-ray magnetic scattering provides direct information on the magnetic structure of materials [6]. Hannon et al. [7] have shown that the terms correcting the atomic scattering factor are sensitive to the magnetization of the sample and during the last decade a growing number of experiments have been carried out, using atom and shell tunability of synchrotron radiation, for the investigation of magnetic properties. In order to study x-ray resonant magnetic scattering (XRMS) of 3d transition metals, the *L* absorption edges (electronic transitions from 2p levels toward unoccupied band states) must be utilized; these are located in the soft x-ray range. Soft x-ray resonant magnetic scattering using either circularly [8–11] or linearly [12–14] polarized x-rays has proven to be a highly useful technique for the study of magnetic properties of buried layers or interfaces and depth-dependent magnetic properties. In the case of magnetic multilayers, the periodicity of the magnetization amplitude leads to a magnetic contribution at the position of low-angle Bragg peaks for ferromagnetically ordered multilayers [9,15,16] and at the half-order positions for antiferromagnetically ordered multilayers [9,17,18]. Moreover, a varying external magnetic field can be applied during XRMS measurements, such that the magnetization reversal at corresponding *L* absorption edges can be followed, i.e. element-selective hysteresis loops can be measured [19,20].

Up to now most XRMS studies have been carried out on Co/Cu multilayers, while data on Fe/Cr multilayers are scarce. This is in contrast to the literature available on Fe/Cr superlattices using polarized neutron reflectivity methods [21–24]. Partly, this is due to the fact that the demands for structural and magnetic quality as well as for the saturation fields required and available in XRMS experiments are better met for Co/Cu than for Fe/Cr systems. As we have now higher magnetic fields available than were used before in XRMS measurements, we would like to focus our attention on the magnetic and structural properties of a strongly AF coupled Fe/Cr superlattice. Furthermore, there is an uneasy discrepancy in the literature concerning the half-order AF reflection, which can usually be observed with neutron scattering but is often not seen in magnetic x-ray scans. In the present study we show the proper magnetic hysteresis of the half-order AF peak, which should exhibit a shape resembling the field dependence of the GMR effect. In addition, transverse scans across the half-order AF peak reveals the in-plane AF domain correlation and spin disorder at the interface as a function of field.

We have grown an Fe/Cr(001) superlattice by molecular beam epitaxy using thermal effusion cells for Fe and Cr with rates of 0.015 Å/s and 0.013 Å/s, respectively. The superlattice was grown on a MgO(001) substrate with a 240 Å thick Cr buffer layer deposited at 450 °C and at a rate of 0.15 Å/s. The buffer layer was then annealed for 30 min at the same temperature. A 25 Å thick Cr capping layer provided protection against oxidation of the Fe/Cr(001) superlattice. The base pressure of the chamber was 4×10^{-11} mbar before starting the deposition. The Cr layer thickness of 8 Å corresponds to the first maximum in the AF interlayer exchange coupling [1,2]. The number of repeats and the Fe layer thickness between 10 Å and 15 Å were chosen to not exceed the total penetration depth of the soft x-rays, which is about 500 Å at an energy just below the Fe L_3 absorption edge, where we performed most of our experiments. Using x-ray reflectometry and Bragg diffraction with hard x-rays ($\lambda = 1.542$ Å) the final layer sequence of the superlattice has been determined: MgO/Cr(250 Å)/[Fe(11.4 Å)/Cr(8 Å)] $\times$ 20/Cr(25 Å). The root mean square roughness of all interfaces is about 3 Å. The epitaxial relationship between the superlattice and substrate was determined as MgO(001){110} ∥ Fe(001){100}, which is the usually observed 45° in-plane relationship between bcc Fe and fcc MgO. SQUID magnetometry indicated a strong AF coupling with a saturation field of $H_S \approx 30$ kOe.

The XRMS experiments were carried out at the bending magnet beamline PM3 and at the undulator beamlines UE56/1 and UE56/2 of the Berlin storage ring for synchrotron radiation (BESSY). The beamline optics allows one to vary the energies in the range from 20 to 1900 eV with the possibility of tuning the polarization from linear to fully circular. Since for this energy range special vacuum conditions are required, a UHV chamber (ALICE) containing a two-circle diffractometer [25] was used for the scattering experiments to be described below. The sample environment of this diffractometer allows one to apply magnetic fields in the range ±2.7 kOe and to control the temperature in the range from 30 to 400 K. The incident photons were either about 90% (PM3) or 95% (UE56) circularly polarized with positive helicity or 100% linearly polarized with π polarization. The magnetic field was applied in the scattering plane and parallel to the sample surface. In the case of circularly polarized light this polarization corresponds to a longitudinal magneto-optic Kerr effect-type geometry [26]. The in-plane hard axis of the sample magnetization was oriented parallel to the scattering plane.

We start our discussion with experiments performed with circularly polarized light. By tuning the incident energy to just below the Fe L_3 edge, we observe strong magnetic Bragg peaks at the $1/2$ and $3/2$ positions in units of the reciprocal lattice vector of the first-order structural peak associated with the superlattice periodicity (Fig. 1). The fact that we can observe three orders of Bragg reflections as well as Kiessig interference fringes up to 85° clearly reflects the very high structural quality of the sample. Furthermore, from the presence of a half-order peak in the off-specular reflectivity curve (open symbols) we can infer two additional properties: first, the individual ferromagnetic layers are decomposed into magnetic domains; and second, the ferromagnetic domains are strictly antiferromagnetically correlated from top to bottom.

We have investigated the field dependence of the magnetic contribution to the first-order Bragg peak, which is sensitive to the total magnetization of the sample, and the half-order peak, which is sensitive to the antiferromagnetic coupling. Fig. 2 shows the hysteresis loop measured at the position of the first-order Bragg peak. The data (open and closed symbols) are a bit noisy, because the magnetic contribution to the total structure factor is very small compared to the non-magnetic charge contribution. However, the shape of the hysteresis loop is similar to the one measured by SQUID magnetometry as shown by a solid line in Fig. 2. In order to improve the magnetic/charge intensity ratio it is beneficial to determine the magnetic hysteresis at the half-order magnetic peak. Fig. 3 reproduces the half-order peak intensity (specular and off-specular), measured in the remanent state and at the magnetic field ($H = 2.7$ kOe) applied in the scattering plane parallel to the

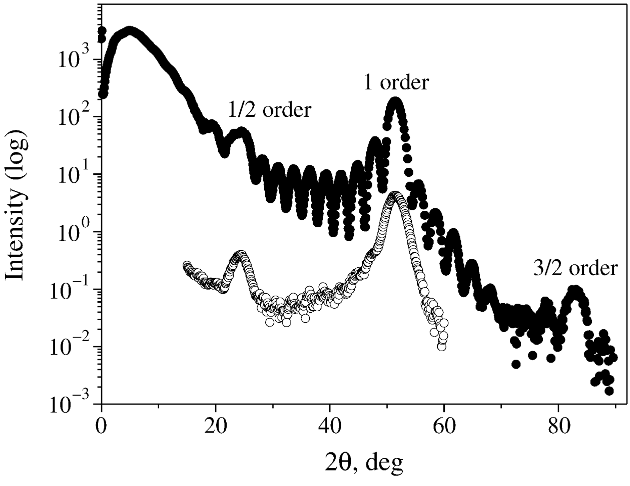

Fig. 1. Specular (closed symbols) and longitudinal diffuse scans (0.4° offset) (open symbols) taken at the Fe L_3 edge with circularly polarized radiation for an AF coupled superlattice with 20 repeats of [Cr(8 Å)/Fe(11.4 Å)] deposited on a Cr(240 Å)/MgO(001) buffer–substrate system.

sample surface and in two opposite directions. The intensity drop of the half-order peak at $2\theta = 24°$ is clearly seen for the specular as well as for the off-specular reflectivity curves. Since the saturation field is higher than the maximum field available, a small AF contribution remains in the intensity for the reflectivity curves measured at $H = \pm 2.7$ kOe. The magnetic hysteresis measured at the half-order AF peak is reproduced in Fig. 4. This hysteresis loop has a maximum at the field corresponding to zero magnetization of the multilayer (the coercive field H_c) and decreases with increasing magnetic field. Since the value of H_c is about 50 Oe only, this effect is not clearly seen for circularly polarized data, but it is more pronounced for linearly (π) polarized radiation (see the inset in Fig. 4). As circularly polarized x-rays are sensitive to the in-plane sample magnetization in the scattering plane, there is an additional magnetic contribution to the total scattering amplitude. This explains the difference between the intensities for positive and negative fields. The hysteresis loop measured with linearly (π) polarized x-rays exhibits the same background level for positive and negative field, since this polarization is not sensitive to ferromagnetic components of the magnetization (see the inset in Fig. 4).

It is well known that for magneto-electronic device applications the structural and magnetic quality of the AF/F interface plays a crucial role. The dependence of the diffuse scattering on the applied magnetic field provides information on the domain structure in the film plane and on the spin disorder at the interfaces [27–29]. Hase et al. have shown that the diffuse scattering around the half-order magnetic peak in AF coupled magnetic superlattices is of pure magnetic origin [30]. In order to avoid the additional magnetic contribution of the background, we have carried out these measurements with linearly polarized x-rays. Fig. 5 shows transverse scans (rocking curves) through the half-order peak measured in the remanent state (solid curve) and in the field of $H = +2.7$ kOe (dotted curve). The rocking curves measured for positive and negative field values are exactly

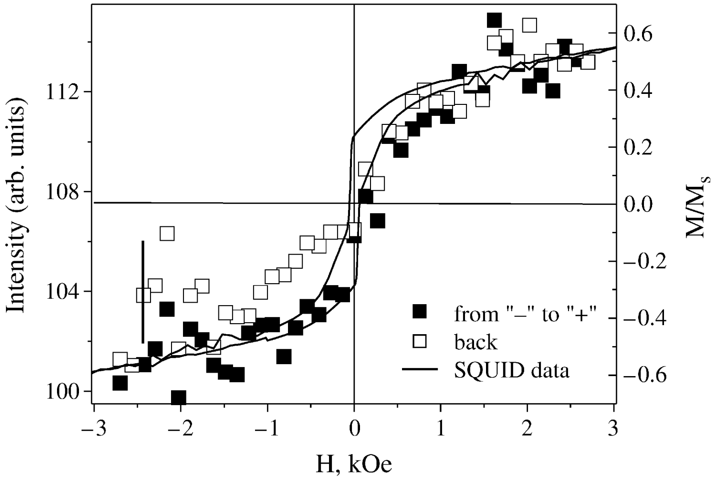

Fig. 2. The hysteresis loop measured at the position of the first-order Bragg peak with circularly polarized radiation (open and closed symbols). The line shows the part of the hysteresis loop measured by SQUID magnetometry (right y-axis), where M_S is the saturation magnetization at $H_S = 30$ kOe. The bar indicates the magnitude of the typical error.

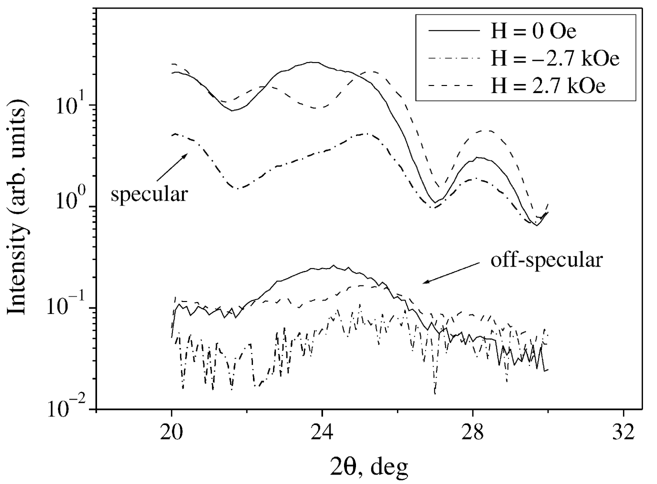

Fig. 3. The half-order peak intensity (specular and off-specular) in the remanent state and at the magnetic field ($H = \pm 2.7$ kOe) applied in the scattering plane along the sample surface (circularly polarized radiation).

identical. Therefore only one of them is shown in Fig. 5. All curves exhibit a Lorentzian line shape for the sharp component (see the inset in Fig. 5) and a broad diffuse shoulder. The width of the Lorentzian component drops by a factor of two from remanence to the maximum field value of $H = 2.7$ kOe. Similarly, the intensity of the diffuse shoulder also drops by a factor of two over the same field range. All changes of the rocking curves are completely reversible. Such a change of the intensity can only be observed at the

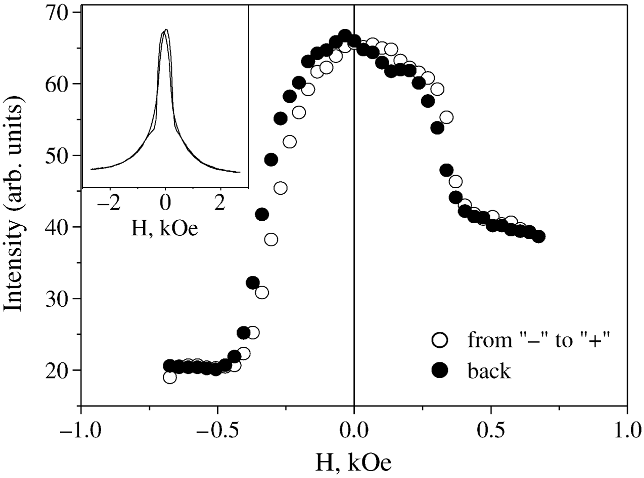

Fig. 4. The hysteresis loop measured at the position of the half-order magnetic peak ($2\theta = 24°$) measured with circularly and (π) linearly (the inset) polarized radiation.

half-order peak; further away, a field dependence can no longer be detected. Furthermore, if we had reached saturation, the centre peak would vanish and the broad shoulder may reflect residual interface spin disorder. Thus, we believe that the shape and intensity of the central peak are due to the magnetic domain structure, while the broad component results from misalignments of the magnetic spins at the interfaces between the Fe and Cr layers. We calculated correlation lengths and roughnesses according to the Born approximation and have found that the structural roughnesses are equal to the one measured with the hard x-rays ($\sigma_s = 3$ Å); the magnetic roughnesses in the remanent state are $\sigma_m \approx 0.65\sigma_s = 2$ Å with a decrease to $\sigma_m = 1.4$ Å at $H = 2.7$ kOe. The in-plane correlation lengths are 1300 ± 100 Å and 400 ± 50 Å for structural and magnetic roughnesses, respectively. The in-plane correlation lengths are independent of the magnetic field application.

The Lorentzian line shape of the rocking curves has already been reported for Fe/Cr multilayers from measurements by synchrotron Mössbauer and polarized neutron reflectometry [24] and for Co/Cu multilayers from measurements by neutron reflectometry [31]. The Lorentzian shape was explained by a multidomain state of the magnetic superlattice in the remanent state, which is superimposed on a standard Gaussian curve corresponding to the instrumental resolution. From an analysis in terms of Lorentzian line shape, we estimate the average size of the AF domains to be about 5 μm for the remanent state. This value is in a good agreement with the data presented in [32], where Kerr microscopy images of AF coupled Fe/Cr/Fe trilayers revealed that the magnetic domains have an average size on the order of a few μm. On applying a magnetic field $H = 2.7$ kOe, we have observed that the domains enlarge to ∼10 μm. On applying a higher magnetic field, the Fe/Cr superlattice approaches a single-domain state and there is a concomitant decrease of the Lorentzian contribution to the line shape, until in saturation only the Gaussian component remains.

In conclusion, we have shown that with XRMS it is possible to study in detail the magnetic properties of antiferromagnetically coupled Fe/Cr superlattices. Using the

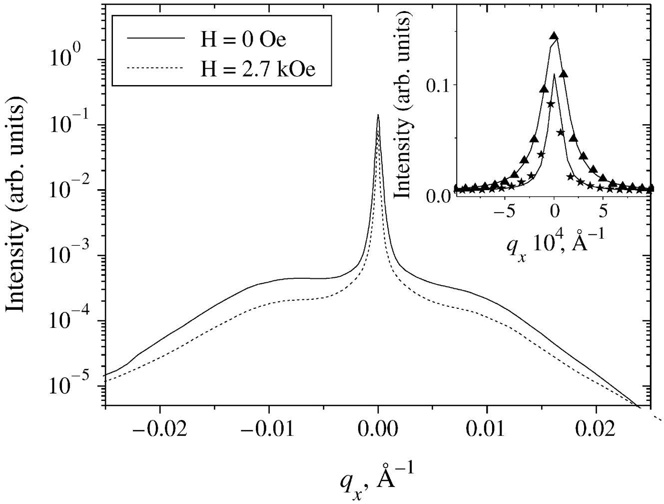

Fig. 5. Transverse scans through the half-order peak measured with linearly polarized radiation in the remanent state (solid curve) and at the applied magnetic field $H = +2.7$ kOe (dotted curve). Inset: the sharp component of the rocking curves. Symbols represent the experimental data (triangles: $H = 0$ Oe; asterisks: $H = 2.7$ kOe), while curves are corresponding fits with a Lorentzian line shape.

resonance condition close to the Fe L_3 edge, strong and purely magnetic Bragg peaks are observed at the half-order and three-half-order Bragg peaks in units of the reciprocal lattice vector of the superlattice periodicity. The half-order peaks reflect the antiparallel orientation of the Fe magnetization vectors of the adjacent layers in remanence. The hysteresis loop measured at this position demonstrates the AF coupling of the Fe/Cr superlattice. Transverse scans with linearly polarized light across the half-order peak exhibit a sharp peak with a Lorentzian line profile superimposed on a flat shoulder of diffuse intensity. With increasing field the width of the central Lorentzian peak and the intensity of the diffuse intensity both drop by roughly a factor of two. From the Lorentzian line shape of the central half-order peak we infer a multidomain state in remanence, which diminishes with increasing field. At the same time the broad diffuse shoulder may reflect spin disorder at the Fe/Cr interface, which decreases with increasing field.

Acknowledgements

We would like to thank J. Podschwadek for technical assistance during sample preparation, T. Kachel, B. Zada and W. Mahler (BESSY) for their help with the beamline operation. This work was supported by the German Federal Ministry of Education and Research (BMBF) under Contract No 03ZA6BC2.

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