Noncollinear Magnetic Hyperfine Fields in the Ag Spacers of Fe/Ag Multilayers

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Nearly perpendicular magnetic hyperfine fields have been observed for the first time in the Ag "spacers" of Fe/Ag multilayers using low temperature nuclear orientation of $^{110}Ag^m$ at 6 mK. At the same time, vibrating sample magnetometry measurements at temperatures down to 4 K have shown the magnetic anisotropy of the Fe to be in plane. The direction of the Ag hyperfine field is thus noncollinear (nearly orthogonal) to the Fe anisotropy. These results are compared with full potential linearized augmented plane wave calculations using the WIEN97 code.

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The magnetic properties of thin two-dimensional magnetic multilayers can be drastically different from the bulk material. By adjusting the thickness of the layers involved, the number of bilayers, and the roughness of the interface, it is possible to engineer properties that have great technological and scientific significance. In Fe/Ag multilayers both ferromagnetic and antiferromagnetic, as well as biquadratic (noncollinear), coupling have been observed [1]. The biquadratic coupling was found to be dependent on the roughness of the Fe/Ag interface [2] as theoretically predicted by [3].

The Fe/Ag interface was first theoretically studied using *ab initio* full potential linearized augmented plane wave (FLAPW) calculations [4]. This study predicts a small Ag magnetic moment of $(0.05-0.08)\mu_B$ and a Ag hyperfine field of -58.7 T in the first layer adjacent to the Fe. Recently, the magnetic hyperfine field of ^{111}Cd at various sites in an epitaxial Fe/Ag interface was measured using perturbed angular correlations (PAC). These experiments demonstrated that large magnetic hyperfine fields of the Cd are restricted to the first layer of the Ag at the interface [5].

In this work, we have measured for the first time the magnetic hyperfine fields in the Ag "spacers" of Fe/Ag multilayers, using the radioactive probe $^{110}Ag^m$ in low temperature nuclear orientation (LTNO) experiments, and compare these to the magnetic anisotropy of the Fe layers, obtained from vibrating sample magnetometry (VSM) measurements. Most conventional experimental magnetic techniques, including ferromagnetic resonance (FMR), magneto-optical Kerr effect [2,6], scanning electron microscopy with polarized analysis [1], Mössbauer spectroscopy [7], and photothermally modulated FMR (PM-FMR) [8], are sensitive only to the Fe layers. Thus, measurements of magnetic hyperfine fields in the Ag layers of Fe/Ag multilayers using LTNO can provide unique additional information on this system. Further, the LTNO technique allows one to deduce the average alignment and magnitude of the Ag hyperfine fields at the interface from the directional distribution of the γ rays from an isotope of the same element as the multilayer spacer, in this case $^{110}Ag^m$, so that no other foreign probes are needed. Finally, these results are supplemented by new *ab initio* FLAPW calculations [9], updating the 1984 results of [4] and providing further insight into the Fe/Ag interface.

The aim of these investigations was to determine the alignment of the Ag hyperfine field at the Fe/Ag interface. For multilayers with a magnetic shape anisotropy that is stronger than the surface anisotropy, the in-plane magnetized state is more stable than the perpendicular state; a stronger surface anisotropy on the other hand means a more stable perpendicular magnetized state. The Fe layer thickness d was chosen so as to have a surface anisotropy ($2K_s/d$ with K_s the surface anisotropy constant for an Fe/Ag interface) that is smaller than the shape anisotropy ($\mu_0 M_s^2/2 = 1.86 \times 10^6$ J/m^3 with M_s the saturation magnetization for Fe). Thus, the Fe layers all had thicknesses larger than the critical d value of five monolayers (ML) (using $K_s = 0.64$ mJ/m^2 [10]), at which it is expected that the easy axis lies in the plane of the multilayer.

The samples used were prepared with a Riber MBE (molecular beam epitaxy) system in ultrahigh vacuum (UHV) of 2×10^{-10} mbar during growth. On a substrate of MgO(001), a 100 Å seed layer of Fe(001) was first epitaxially grown at 175 °C. After cooling to 10 °C, the atomic layers were deposited and a final capping layer of 500 Å Fe was added for protection. The multilayers $[\mathrm{Ag}(x\ \mathrm{ML})/\mathrm{Fe}(y\ \mathrm{ML})]_{20}$ fabricated in this manner had (x, y) values of (2,10), (3,9), (3,18), and (5,10). The bcc lattice of the Fe (1 ML = 1.433 Å) closely matches the lattice of fcc Ag (1 ML = 2.043 Å) after a rotation by 45°, with a resulting in-plane lattice mismatch of only 0.8%. Thus, it is possible to grow high-quality superlattices and interfaces, provided there are not too many steps which tend to disturb crystalline growth within the layers.

The multilayers were characterized with x-ray diffraction (XRD) at both low and high angles using CuK_α

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radiation, ($\lambda = 1.5418$ Å). Low angle XRD is sensitive to large length scales and therefore indicates the interlayer structure, while high angle XRD is sensitive to the crystal structure quality within the layers. All samples except the (2,10) had well-defined Bragg peaks in both low and high angle XRD spectra. The high angle spectrum was analyzed using the x-ray analysis program SUPREX [11] and the average roughness was found to be less than 0.7 Å for both the Fe and Ag layers. In addition, so-called "rocking" spectra were obtained on a low and a high angle peak. The width of these peaks measures the parallelism of, respectively, the bilayers and the atomic layers. The observed full width half maximum of 0.12° and 1.5° for the low and high angles, respectively, indicates that the layers were very closely parallel.

In order to perform LTNO, the samples were activated for 10 days with thermal neutrons at a flux of $\phi = 10^{13} n/(\mathrm{cm}^2\,\mathrm{s})$ at the BER-II research reactor of the Hahn-Meitner-Institute in Berlin. During this activation at a temperature of 80 °C, ^{109}Ag is transformed into $^{110}\mathrm{Ag}^m$ which has a half-life of 249.7 days. X-ray spectra taken before and after irradiation showed that this 10 day irradiation caused only minimal damage.

The activated samples were soldered at a temperature of 30 °C onto the cold finger of a ^{3}He/^{4}He dilution refrigerator using a Ga-In eutectic and subsequently cooled to temperatures around 6 mK. At these temperatures the $^{110}\mathrm{Ag}^m$ nuclei are oriented, causing the γ rays from their decay to be emitted anisotropically. The directional distribution of these γ rays can be expressed as [12]

$$W(\theta) = 1 + \sum_{k=2,4} B_k U_k A_k Q_k P_k(\cos\theta)\,, \qquad (1)$$

where θ is the angle between the direction of the detector and the magnetic hyperfine field and $B_k(T, B_{\mathrm{hf}})$ are nuclear orientation parameters which are functions of both temperature T and magnetic hyperfine field B_{hf}. Further, U_k and A_k are known nuclear decay scheme parameters, Q_k are solid angle corrections, and P_k are the Legendre polynomials. The directional distribution of the 1384 keV γ rays was measured with two Ge γ detectors, one in the direction of the external magnetic field (detector 1) and the other perpendicular to it (detector 2).

The multilayers were mounted on the cold finger so that the external magnetic field was applied in the sample plane, along the (100) direction of the Ag. Because of the 45° rotation of the bcc Fe lattice relative to the fcc Ag, this corresponds to the (110) axis in Fe. Thus, detector 1 had its axis in the plane of the multilayer while detector 2 had its axis perpendicular to the multilayer surface. The directional distribution of the 1384 keV γ rays was measured for all samples as a function of the external magnetic field. For this γ ray, the A_k parameters in Eq. (1) are positive, so that $W(0°) > 1$ and $W(90°) < 1$. The data for the (3,9) sample are shown in Fig. 1. It is clear that detector 1 is a near 90° detector and detector 2 is a near 0° detector. Thus,

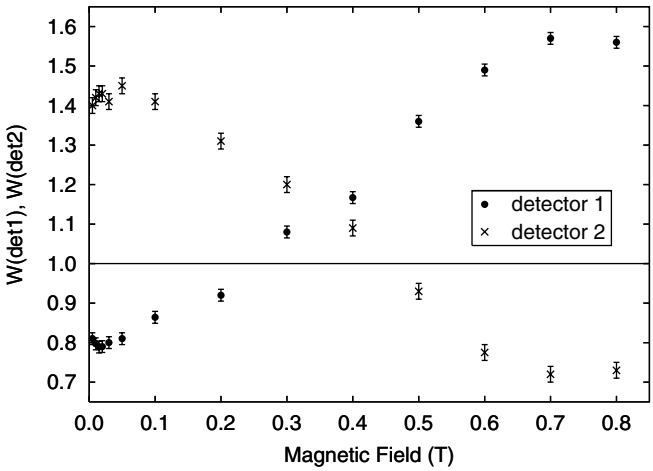

FIG. 1. The 1384 keV γ-ray anisotropy as a function of applied magnetic field for the [Ag(3 ML)/Fe(9 ML)] multilayer. The applied field was in the plane of the multilayer.

for low applied external fields, the Ag hyperfine fields point out of the plane of the multilayer. These out-of-plane Ag hyperfine fields were observed for all multilayers with well-defined Bragg diffraction peaks in the x-ray spectra [i.e., (3,9), (3,18), and (5,10)]. No out-of-plane Ag fields were observed for the (2,10) sample, but this had poor multilayer structure as seen from the x-ray diffraction. The maximum angle between the hyperfine field and the plane of the multilayers in small applied fields was found to be between 70° and 85° for the different multilayer systems. As the in-plane applied magnetic field is increased, the average direction of the hyperfine field rotates from out of plane to in plane, as can be seen from the change in anisotropy with magnetic field.

The (3,9) sample was resoldered onto the cold finger and cooled to $T \sim 6$ mK with the external magnetic field this time perpendicular to the multilayer. Detector 1, placed along the applied field direction, was now perpendicular to the multilayer and detector 2 was in plane, along the (110) axis of the Fe layers. The observed anisotropies are shown in Fig. 2 and it is clear that the Ag hyperfine fields point out of the plane of the multilayers. It is remarkable that over a wide range of applied fields, in which the Fe magnetism is expected to rotate from roughly in plane to out of plane, there is practically no change in the alignment of the Ag hyperfine fields: only a modest increase of 0.08 is observed in $W(\theta)$ in going from low to high magnetic fields.

Next, we compare the LTNO response with the VSM measurements at 4 K in both the in-plane and perpendicular orientations. The data for the (3,9) sample are shown in Fig. 3. The in-plane Fe magnetization saturates at 0.7 T for this (3,9) system. This is similar to the saturation field of the Ag hyperfine fields from the in-plane LTNO at 6 mK (Fig. 1). This correspondence of the saturation fields is also found for the other multilayer systems with good multilayer structure. There are two possible explanations for

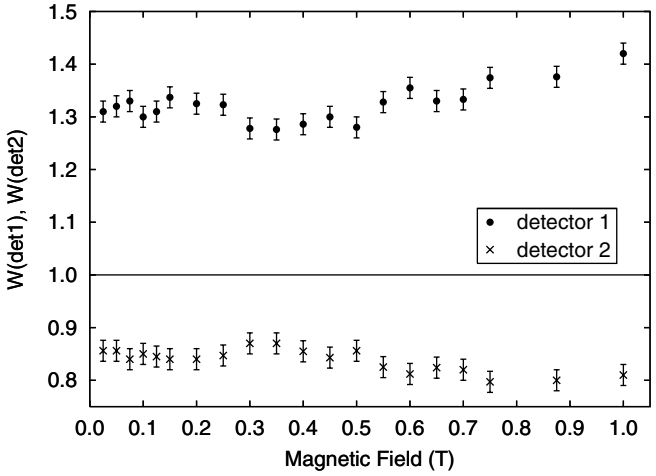

FIG. 2. The 1384 keV γ-ray anisotropy as a function of applied magnetic field for the [Ag(3 ML)/Fe(9 ML)] multilayer. The applied field was perpendicular to the multilayer.

this high saturation field: a coupling between the Fe layers or an out-of-plane Fe magnetization. Although PM-FMR measurements indicate that the Fe layers should couple ferromagnetically for 3 ML of Ag [8], it was shown in [3] that 1 ML steps at the interface could produce strong fluctuations in the coupling strength, thereby inducing a potentially large biquadratic coupling term. Recently, two alternative reasons have been proposed to explain how the magnetization can be canted out of the plane even when the ratio of surface to shape anisotropy is less than 1. First, the out-of-plane magnetization of Co/Au multilayers was attributed to an out-of-plane domain structure which minimized the shape anisotropy energy [13]. Second, Mössbauer measurements showed that Fe moments can emerge out of the plane for thin (4 ML) Ag layers in Fe/Ag multilayers grown on a Ag(001) surface with no capping layer [14]. The origin of this effect was attributed to atomic steps and the out-of-plane angle was found to be less than 45°. The perpendicular VSM magnetization curves show two saturation plateaus for all systems with good multilayer structure, in the case of the (3,9) system one at 1.2 T and the other at 2.19 T. The latter is the saturation field for a thin slab of Fe with only magnetic shape anisotropy. Assuming the second saturation plateau is associated with the 500 Å capping and 100 Å base Fe layers, the critical point at 1.2 T represents the saturation of the Fe in the thin multilayers, which have shape anisotropy as well as surface anisotropy. Finally, the total magnetic anisotropy energy obtained from a comparison of the in-plane and perpendicular orientations is given by $\int_0^{M_s}(H_{\parallel} - H_{\perp})\,dM$ with M and M_s the magnetization and saturation magnetization, and $H_{\parallel}$ and $H_{\perp}$ the magnetic field applied, respectively, in plane and perpendicular to the plane. For all multilayer systems this anisotropy energy was found to be negative. The negative value indicates that the magnetization in the Fe layers is more stable in the in-plane orientation than for the perpendicular state.

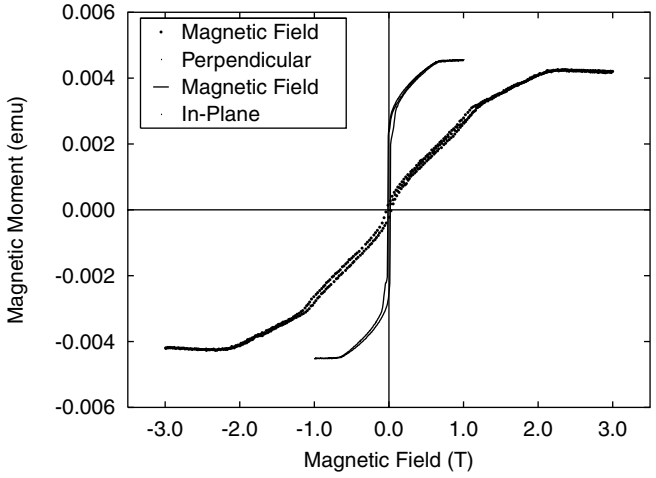

FIG. 3. VSM response for magnetic field in both in-plane and perpendicular orientations to the [Ag(3 ML)/Fe(9 ML)] multilayer, at 4 K.

The direction of the Ag hyperfine fields in near zero external magnetic field was found to be between 70° and 85° from the plane for the different multilayer systems. The magnetic anisotropy of the Fe layers from VSM measurements was determined to be in plane. Therefore in zero field the easy axis of the Ag surface sites and the anisotropy of the Fe layers are almost orthogonal.

This strange phenomenon has not been observed before in an Fe/Ag multilayer system nor is it theoretically expected. In a naive picture the Ag ions should follow the alignment of the Fe magnetization, as is expected by the fact that the saturation fields for the Ag hyperfine fields from LTNO correspond with the saturation fields for the Fe magnetization from VSM. However, this picture neglects the large amount of d hybridization at the interface as predicted by [4]. The magnetic hyperfine field measurements with the ^{111}Cd probe atoms on a Fe/Ag bilayer, using ^{111}In PAC, showed alignment parallel to the Fe moments, in plane [5]. Since bulk Ag and Cd both behave as sp metals, we could expect to see similar behavior at the interface of the Fe/Ag multilayers. However, this neglects an important difference between the Ag and Cd ions, i.e., the fact that the d band in Ag is much closer to the Fermi surface than in Cd. Thus the behavior of Ag atoms at the interface, particularly in regard to d hybridization, may be rather different from that of the Cd probe atoms. In addition, atomic steps and/or diffusion might play a role in causing magnetic frustration, thereby inducing noncollinear magnetic moments at the interface. Finally, one cannot rule out the possibility that magnetic coupling between Fe layers might play a role at the interface. Theoretical predictions of the coupling in noble metal spacers depend only on the Fermi surface of the bulk noble metal; interface states are not included [15]. In all these theories, the coupling between Fe layers oscillates between ferromagnetic and antiferromagnetic, and a collinear spin density wave (SDW) is assumed.

TABLE I. Hyperfine fields and electronic magnetic moments for a [Fe 4 ML/Ag 4 ML] multilayer using WIEN97.

	Ag(1)	Ag(2)	Fe(3)	Fe(4)
Hyperfine field (T)	−0.78	−31.84	−29.43	−33.05
Magnetic moment (μ_B)	0.001	+0.037	+2.683	+2.403

Theories which describe biquadratic coupling, induced by, e.g., monatomic steps, do not mention the topology of the SDW [3].

To complement the measurements, we did WIEN97 calculations for several "perfect" roughness-free, theoretical multilayers [Fe(x ML)/Ag(y ML)] with (x, y) values of (2,4), (4,4), (6,4), (4,2), and (4,6). While the calculation method is similar to that of [4], a number of important improvements are implemented in WIEN97, including (i) the possibility of increasing the number of points in k space while maintaining a reasonable computational effort, (ii) the ability to relax atomic positions to minimize the total energy, and (iii) improved local spin density approximations [9]. However WIEN97 does not give relevant results when parts of the unit cell have different magnetization directions. Therefore, we can only compare the calculations to the high-field data where the Ag and Fe magnetizations are parallel. The results presented here are on a [Fe(4 ML)/Ag(4 ML)] multilayer but the calculated hyperfine fields and electronic magnetic moments are found to be almost the same for the different systems. The unit cell that is used here has four pairs of equivalent atoms that have been moved from their bulk positions to minimize the total energy. The result of this calculation is summarized in Table I. Atoms Ag(1) and Fe(4) represent the center atoms within the Ag and Fe layers, respectively, while atoms Ag(2)/Fe(3) represent the interface sites.

While the results are in qualitative agreement with [4], some definite advances have been made. First, the hyperfine field and the moment in the second Ag layer away from the surface, i.e., Ag(1) were calculated for the first time. This layer turns out to have a very small magnetic moment and hyperfine field compared to the Ag at the interface. These calculations therefore indicate, in agreement with ^{111}In PAC [5] measurements, that large spin-polarized perturbations are restricted to the interface layer of Ag. It is then possible to estimate the average hyperfine field at the interface using LTNO, assuming that the Ag layers 1 ML away from the interface feel no hyperfine field and do not contribute to the γ-ray anisotropy. In the (3,9) sample, with a final in-plane saturation γ-ray anisotropy of 1.552(15) at 6 mK, a hyperfine field of |21.4(8)| T is derived for the Ag surface atoms. However, LTNO is sensitive to all Ag atoms in all surface sites, including those near lattice steps and defects, so that this value represents a lower limit for the Ag hyperfine field expected on a perfect surface. It is to be noted that within the experiment accuracy the data are not sensitive to the sign of the hyperfine field. Second, the hyperfine field at the interface was originally calculated to be −58.7 T [4], which is much larger than the value for an isolated Ag impurity in an Fe host, i.e., −45.42(2) T [16]. Our new calculated estimate using WIEN97 is −31.84 T, which is lower than the value for the Ag impurity and closer to the lower limit of |21.4(8)| T set by our LTNO measurement.

In summary, we have shown that in good quality Fe/Ag multilayers, the Ag hyperfine fields are pointing out of the plane, nearly perpendicular to the multilayer, while the magnetic anisotropy for the Fe layers lies in the plane. We have also improved on an earlier FLAPW calculation, using WIEN97, and have obtained an improved prediction for the Ag hyperfine field of −31.84 T at the interface with Fe for a perfect multilayer. This compares rather well with our LTNO measured value of |21.4(8)| T which represents a lower limit for the hyperfine field.

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