Magnetic Multilayers I Andreas Moser, Chairman

# Magnetism and structure of Fe/Cu multilayers studied by low-temperature conversion electron Mössbauer spectroscopy

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Magnetic properties, structure, and morphology of a (Cu 39 Å/Fe 7 Å)×36 multilayer prepared by dc magnetron sputtering were studied mostly by low-temperature conversion electron Mössbauer spectroscopy (LT-CEMS) down to 90 K. LT-CEMS measurements in external magnetic field of 0.1 T rule out superparamagnetism of Fe islands. The multilayer exhibits strong perpendicular anisotropy below its Curie temperature ($T_C = 185$ K) and the linear temperature dependence of the magnetization suggests two-dimensional ferromagnetism.  [S0021-8979(99)45408-8]

## I. INTRODUCTION

Superlattices formed from successive deposition of Fe and Cu layers provide a convenient system for studying complex phenomena such as fcc Fe magnetism, two-dimensional, surface/interface magnetism, and quantum tunneling of magnetization. Magnetic properties of Fe/Cu multilayers prepared by ion beam sputtering, magnetron sputtering, thermal evaporation, and electron beam evaporation with Fe and Cu layer thicknesses ($t_{Fe}$ and $t_{Cu}$, respectively) in the range from 5 Å [2.5 monolayers (ML)] to 50 Å have been studied extensively.[1–7] Superparamagnetic (SP) relaxation of magnetization of iron islands at room temperature (RT) was suggested for superlattices with Fe layer thicknesses less than 9 Å.[1–5,7] In our previous work[7] for a sample with $t_{Fe} = 7$ Å, the presence of a broad maximum in the temperature dependence of the ac susceptibility, the absence at RT of saturation in the magnetoresistance (MR) curve, the lack of oscillations in the dependence of MR on $t_{Cu}$, and the presence in the Mössbauer spectrum at RT of only paramagnetic components, all pointed to superparamagnetism of iron granules at RT. Here, we investigate the temperature dependence of magnetization measured on local and bulk scales for a single sample which allows us to establish a relationship among its magnetic behavior, structure, and morphology.

## II. EXPERIMENTAL METHODS

Fe/Cu multilayers were prepared by dc magnetron sputtering of 36 bilayers with individual layer thicknesses $t_{Fe} = 7$ Å and $t_{Cu} = 39$ Å, onto SiO substrates at ambient temperature. An Fe target of natural isotopic mixture was used. Before each deposition the vacuum reached $2\times10^{-7}$ Torr. Deposition was performed at an Ar pressure of 11 mTorr starting with a Cu layer and finishing with a 15 Å Cu capping layer to reduce oxidation. The deposition rates of Cu and Fe were about 2 and 1 Å/s, respectively. Determination of deposition rates, thickness calibration, and details of the structural characterization by low-angle x-ray diffraction (XRD) are described in a previous paper.[7] High-angle XRD showed that the Cu layers are fcc Cu(111) oriented along the multilayer normal. To study local properties we employed $^{57}$Fe low-temperature conversion electron Mössbauer spectroscopy (LT-CEMS) with the use of specially designed and an optimized gas-flow proportional counter. The counter gas was a He+4% $CH_4$ mixture at a flow rate of about 0.2 cm$^3$/s. $A \sim 1$ GBq $^{57}$CoRh source was used and the area of the beam spot was about 3 cm$^2$. Measurements of the initial suscepti-

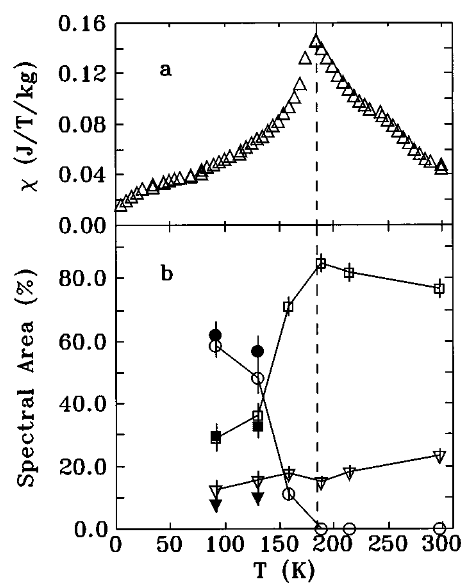


FIG. 1. Temperature dependence of the initial susceptibility (a), and of the relative spectral areas of different contributions to the CEM spectra (b): (squares) paramagnetic interior and alloyed interfaces, (triangles) sharp paramagnetic interface, (circles) ferromagnetic distribution of hyperfine fields, (filled symbols) measured in an external magnetic field of 0.1 T.

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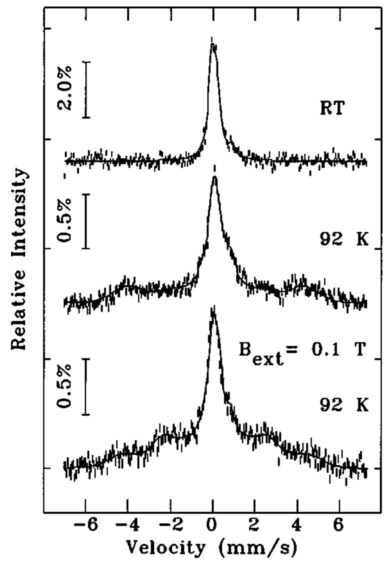


FIG. 2. Conversion electron Mössbauer spectra for the (Cu 39 Å/Fe 7 Å) $\times 36$ multilayer at room temperature and 92 K. Note the appearance of the broad distribution of hyperfine magnetic fields at 92 K. Also note the absence of any increase of hyperfine splitting in external magnetic field of 0.1 T applied in the plane of the multilayer.

bility and in-plane magnetization were performed using an extraction magnetometer. All measurements reported here were made on the same multilayer sample.

## III. RESULTS AND DISCUSSION

The initial susceptibility exhibits a clear cusp at 185 K shown in Fig. 1(a). A broad maximum in the susceptibility curve has been attributed to a distribution of the blocking temperatures of SP relaxation of ferromagnetic (FM) iron particles.[3,4] For systems with SP relaxation, the magnetization at low temperature follows a $T^{\alpha}$ law with $\alpha$ between 1.5 and 3.0.[8] The magnetization of our sample shows a linear dependence, i.e., $\alpha = 1.0$ which, in principle, could be attributed to interacting SP particles.[9] The hyperfine magnetic field at the $^{57}$Fe nucleus is approximately proportional to the local moment of the iron atom but will average to zero if relaxation is faster than the lifetime of the excited nuclear state. An external magnetic field should slow down the rate of fluctuations in the direction of the moment and thus lead to an increase in the hyperfine field. Mössbauer spectra of Cu 39 Å/Fe 7 Å multilayer at RT and 92 K are shown in Fig. 2. A paramagnetic (PM) component is present in the central part of each spectra. On cooling, a broad magnetic contribution appears below 190 K indicating a distribution of ferromagnetically split components. The onset of magnetic order is marked by the steep rise in the spectral area of the FM distribution in Fig. 1(b), and exactly matches the position of the cusp in the initial susceptibility shown in Fig. 1(a). Mössbauer spectra in an external magnetic field of 0.1 T applied in the sample plane were taken at 130 and 92 K and the spectrum at 92 K is also shown in Fig. 2. It is apparent, even without fitting, that the field does not increase the width of the Zeeman splitting present at low temperatures. The applied field merely leads to a redistribution of the intensity from the wings of the broad contribution to its middle part, suggesting a change in the relative intensity of the individual lines of each magnetically split subspectrum and thus we rule out SP behavior.

Having eliminated the possibility of SP relaxation, it is natural to ascribe the linear dependence of the bulk magnetization at low temperature to a manifestation of two-dimensional ferromagnetism. Linear temperature dependence is generally explained by suppression of spin-wave excitations normal to the plane of a thin FM film.[10,11] The slope of the temperature dependence of the magnetization (normalized to its value at 5 K) obtained from bulk measurements for the Cu 39 Å/Fe 7 Å multilayer is $(13.5\pm 0.4) \times 10^{-4}\,\mathrm{K}^{-1}$, which is close to that for a 2 Å Fe(110) layer on Ag(111)$\sim 11\times 10^{-4}\,\mathrm{K}^{-1}$,[11] but is 4–5 times higher than for (Ag 20 ML/Fe 3 ML)$\times 8$ multilayer prepared by molecular beam epitaxy $\sim 3\times 10^{-4}\,\mathrm{K}^{-1}$.[10] The larger slope obtained for the present multilayer may be attributed to a difference in structure and morphology which may change the spin-wave spectrum, exchange coupling in the interface zones, and interface magnetic anisotropy.

The large total width of the paramagnetic component in the RT Mössbauer spectra [full width half maximum (FWHM) $\simeq$0.65 mm/s compared with 0.28 mm/s for an $\alpha$-Fe calibration foil], the absence of any sharp features, and the small shoulder at the positive velocity side suggest several contributions. Although Fe and Cu have negligible equilibrium solubility, for thin films and multilayers the Fe/Cu interface may extend over 1–4 ML.[1,12] The expansion of the Fe layers with decreasing $t_{\mathrm{Fe}}$ approaches that of bulk Cu for $t_{\mathrm{Fe}} \simeq 7$ ML,[12,13] and provides an additional complication. Variations in composition and lattice distortion will lead to a distribution of isomer shifts and quadrupole splittings of subspectra in both the PM and FM states and also to a distribution of hyperfine fields ($B_{\mathrm{hf}}$) in the FM state.[1,14,15] A highly strained and alloyed interface can give rise to the small shoulder at the positive velocity side. However, we cannot exclude the possibility that this feature is due to Fe oxidation. Previously, oxide-like components were observed for thin Fe film,[16] and for Fe/Cu multilayers.[5] All of these effects, coupled with scatter in $t_{\mathrm{Fe}}$, lead to a smooth spectral shape suggesting a continuous distribution of isomer shifts and quadrupole splittings.

At present we will tentatively model the central peak and the shoulder by two doublets. The first doublet reflects mainly distorted interior layers together with more outer alloyed layers, while the second doublet represents the highly strained and alloyed Fe/Cu interface. For spectra exhibiting a magnetic splitting, a Gaussian distribution of hyperfine magnetic fields was introduced. For the spectrum at 92 K we had to introduce two Gaussian distributions with independent centers and widths. The fits show that the parameters of the doublets do not depend significantly on temperature. Relative spectral areas of the contributions are shown in Fig. 1(b). As expected, the area of the interface doublet is essentially constant. At 92 K the average hyperfine field $\langle B_{\mathrm{hf}}\rangle = 20\pm 3$ T, the most probable fields in the distribution are $12\pm 3$ and $26\pm 1$ T. The larger value is attributed to ferromagnetic Fe in a high-spin state with a tetragonally distorted fcc, i.e., fct structure, while the smaller value reflects the effect of alloying at the interface.[15]

The relative intensities of lines 2 and 5 to the innermost lines of the Zeeman sextet, $R_M$, follows a $4\sin^2(\Theta)/[1+\cos^2(\Theta)]$ dependence on the angle $\Theta$ between the direction of incident $\gamma$ beam and the direction of $B_{hf}$. At 92 K and in the absence of an external magnetic field $\langle\Theta\rangle=19°\pm19°$. Thus, on cooling, the fct Fe layers exhibit FM ordering with the $T_C=185$ K which is much smaller than reported previously for Fe(100) layers of comparable thickness on Cu(100)[15] and with almost complete perpendicular magnetic anisotropy. Application of a 0.1 T field in the sample plane does not change any of the parameters within error, except for $R_M$ which is clearly seen in the redistribution of intensity from the wings of the distribution to its middle part and results in $\langle\Theta\rangle=66°\pm11°$. This anisotropy is associated with the Fe/Cu interface and is clear evidence that the interfacial mixing and roughness occur in the zone much smaller than $t_{Fe}=7$ Å and is strong enough to prevent SP fluctiations and to lead to the cusp in the susceptibility measured in magnetic field with the small amplitude of 0.5 mT applied in the sample plane shown in Fig. 1(a).

## IV. CONCLUSIONS

LT-CEMS has been used to study the magnetism and structure of a (Cu 39 Å/Fe 7 Å)$\times$36 multilayer with natural content of $^{57}$Fe isotope prepared by dc magnetron sputtering. The Fe layers are in a complex structural state ranging from the alloyed interface to the inner Fe zone, with strong tetragonal distortion present across the whole layer thickness. On cooling, ferromagnetic order develops associated with the high-spin state of fct structure. The steep rise in the contribution of the ferromagnetic component to the Mössbauer spectrum matches the clear cusp in the temperature dependence of the initial susceptibility. Spontaneous magnetization at 92 K is directed along the multilayer normal. Application of a weak external magnetic field in the multilayer plane does not result in any increase of the hyperfine field and thus excludes interpretations based on SP relaxation. Magnetization shows the linear dependence on temperature characteristic of two-dimensional ferromagnets, and its large slope correlates with the weakening of the exchange interactions in the alloyed interface and the discontinuous structure of the Fe layers reflected in the very low $T_C=185$ K.


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