Spin wave excitations in Fe/Cu multilayers as a function of its parameters

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Correlation of low-temperature magnetic properties with morphology of (Cu(111)/Fe)×36/Cu multilayers prepared by dc-magnetron sputtering with $t_{Cu}=38.5$ Å, 21.0 Å, and $t_{Fe}=7.3$ Å, 8.5 Å, respectively, was investigated. The primary methods used were ^{57}Fe low-temperature conversion electron Mössbauer spectroscopy (LT–CEMS) from RT down to 50 K and measurements of magnetization from RT down to 5 K. The Fe layers were found to be discontinuous at $t_{Fe}=7.3$ Å, but continuous at $t_{Fe}=8.5$ Å. Transition to the continuity of the Fe layers enhances both in-plane shape and perpendicular surface magnetic anisotropies. This is reflected in the change of the behavior of the temperature dependencies of spontaneous magnetization σ_s measured with external field oriented parallel and perpendicular to the multilayer plane. © *2000 American Institute of Physics.* [S0021-8979(00)86408-7]

I. INTRODUCTION

Transition metal superlattices are attractive materials for information storage technologies. The tailoring of specific magnetic properties, such as the orientation and strength of the magnetic anisotropy, may be achieved by choosing appropriate materials and varying the thicknesses of constituent layers. Interfaces, size and relaxation effects (the latter being unfavorable for most applications) often control the magnetic behavior of such artificially structured substances and their influence strongly depends on crystalline structure and morphology. Fe layers in a (Cu(111)/Fe)×36/Cu multilayer prepared by dc-magnetron sputtering with $t_{Cu}=38.5\pm0.3$ Å and $t_{Fe}=7.3\times0.3$ Å were found to consist of iron islands[1] that were ferromagnetic as a result of a tetragonal lattice distortion.[2] The perpendicular magnetic anisotropy was strong enough to prevent superparamagnetic fluctuations of the magnetization and they exhibited a linear temperature dependence of spontaneous magnetization at low temperatures.[2] Here, we present experimental results on changes in the magnetic properties of Cu(111)/Fe multilayers as the Fe layers change from an island to continuous morphology. This shows the role of magnetic anisotropy and interfacial effects on spin-wave excitations.

II. EXPERIMENTAL METHODS

The multilayer film consisting of 36 bilayers with $t_{Fe}=8.5\pm1.0$ Å and much thicker Cu layers of $t_{Cu}=21.0\pm1.0$ Å (to minimize interlayer exchange coupling) was prepared by dc-magnetron sputtering onto Si substrates. The deposition sequence and all other conditions were as used previously.[1,2] The starting layer as well as the oxidation protective capping was Cu. Deposition onto a Si substrate kept at ambient temperature, was performed at an Ar pressure of 10 mTorr with rates of about 2 Å/s and 1 Å/s for Cu and Fe, respectively. The Cu/Fe interface roughness $\sigma_{Cu/Fe}$ (Fe on top of Cu) was found to be ~6 Å (as in the earlier $t_{Fe}=7.3$ Å multilayer), but that of the Fe/Cu interface $\sigma_{Fe/Cu}$ (Cu on top of Fe) was somewhat larger (~15 Å compared with 11 Å for the $t_{Fe}=7.3$ Å multilayer). The interface roughnesses and thicknesses calibration were obtained from low-angle x-ray reflectivity data.[1] As before[1,2] high-angle x-ray diffraction showed that the multilayer grows coherently with the Cu(111) direction normal to the film surface. Atomic scale properties were obtained using ^{57}Fe low-temperature conversion electron Mössbauer spectroscopy (LT-CEMS). A gas-flow He+4% CH_4 proportional counter was used for RT −90 K, while a newly designed and optimized He-filled proportional counter covered the range down to 50 K. Both counters provided efficient long-term operation with our natural Fe samples, and had an illuminated spot ~20 mm in diameter. Bulk magnetic properties were obtained by extraction magnetometer.

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III. RESULTS AND DISCUSSION

The initial susceptibility measured in the plane of the multilayer (excitation field=0.5 mT) exhibits a sharp shoulder at 225 K, slightly higher than the cusp observed for the earlier (Cu 38.5 Å/Fe 7.3 Å)×36 multilayer [Fig. 1(a) of Ref. 2]. This behavior cannot result from a broader distribution of Curie temperatures in the present (Cu 21.0 Å/Fe 8.5 Å)×36 multilayer because no hyperfine magnetic field is detected at temperatures above the shoulder, as illustrated by the RT Mössbauer spectrum shown in Fig. 2. The initial susceptibility measured along the multilayer normal shows no features at all. This is direct evidence for the absence of superparamagnetic relaxation.

Mössbauer spectra of the present multilayer measured at 93 K, i.e., below T_c (also shown in Fig. 2), are close to low-temperature spectra observed previously from similarly nanostructured Cu/Fe multilayers[3] which indicated perpendicular magnetic surface anisotropy. A satisfactory fit to the spectra is only possible by applying the same structural model used for the earlier 7.3 Å sample.[2] Distorted interior regions in paramagnetic (PM) state were modeled by a quad-

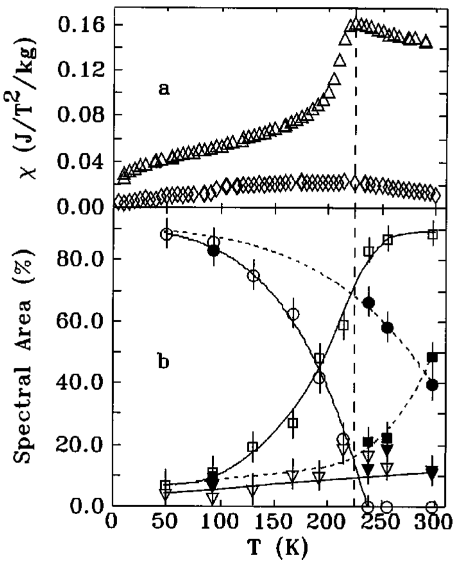

FIG. 1. Temperature dependence of the initial susceptibility (a): triangles, measured in the plane of the multilayer; diamonds, measured along its normal, and of the relative spectral areas of different contributions to the Mössbauer spectra (b): squares, paramagnetic interior and alloyed interfaces; triangles, highly strained and alloyed paramagnetic interface; circles, ferromagnetic distribution of hyperfine fields. Filled symbols, measured in an external magnetic field of 0.1 T.

rupole doublet. Highly strained and alloyed interfaces which were PM over the whole temperature range investigated, were included as a quadrupole doublet with a larger splitting. Ferromagnetic interior regions were fitted as a single Gaussian distribution at temperatures down to 130 K and by two Gaussian distributions with independent centers, widths and

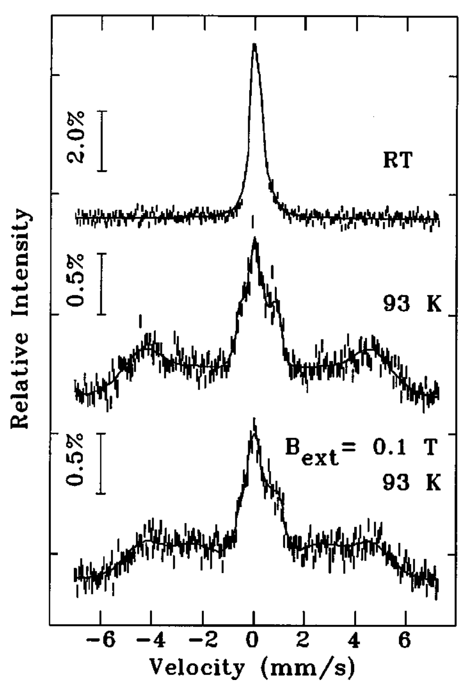

FIG. 2. Conversion electron Mössbauer spectra for the (Cu 21.0 Å/Fe 8.5 Å)×36 multilayer at room temperature and 93 K. Note the appearance of the broad distribution of hyperfine magnetic fields at 93 K. Also note the absence of any increase of hyperfine splitting in external magnetic field of 0.1 T applied in the plane of the multilayer.

amplitudes at lower temperatures. The lower field component corresponds to an alloyed interface, while the higher field one is due to tetragonally distorted fcc, (i.e., fct) inner regions.[4] As before, the appearance of a FM contribution on cooling occurs at the expense of the central PM component and starts at a temperature which matches the position of the sharp feature in the in-plane initial susceptibility as shown in Fig. 1(b).

In the same way as previously the absence of superparamagnetic relaxation is confirmed by taking Mössbauer spectrum at 93 K with external magnetic field of 0.1 T applied in the multilayer plane—no increase in the hyperfine field associated with slowing down of the rate of fluctuations in the direction of the local magnetic moment can be inferred. This spectrum is shown in the bottom of Fig. 2. The average hyperfine field, $\langle B_{\mathrm{hf}}\rangle = 21\pm 2$ T, and the most probable field ($B_{\mathrm{inn}} = 28\pm 1$ T) of the fct inner regions are distinctly smaller than the expected saturation value of about 33 T, and remain unchanged. The freezing of the moments would also lead to a widening of the hyperfine field distribution which is not the case—standard deviation $\sigma_{B_{\mathrm{hf}}} = 9\pm 1$ T also remains unchanged. The values of $\langle B_{\mathrm{hf}}\rangle$ and B_{inn} are practically the same as for the earlier 7.3 Å sample. This points to the unchanged structure of the inner regions of Fe layers. At the same time the interfaces in the present 8.5 Å sample are less alloyed as can be inferred from a slightly higher most probable field for the alloyed interfaces $B_{\mathrm{int}} = 17\pm 3$ T (compared with 12 ± 3 T for the earlier 7.3 Å sample). Less alloying of the interfaces is expected to lead to more continuous layers and stronger perpendicular surface anisotropy.

An external field of 0.1 T applied in the sample plane leads to essentially no redistribution of spectral intensity, indicating that the magnetic moments of the sample do not rotate towards the field, in contrast with the case of the earlier 7.3 Å sample. For the present 8.5 Å sample, the angle between the average moment direction and the γ beam ($\langle\Theta\rangle$) is the same without and with the applied field, being $27\pm 7°$ and $28\pm 13°$, respectively. By contrast, for the earlier 7.3 Å film, $\langle\Theta\rangle$ changes from $19\pm 19°$ to $66\pm 11°$ in the same field.[2] This shows that the present 8.5 Å sample possesses much stronger perpendicular anisotropy.

The slightly larger initial tilting of the average moment suggests a larger role of the in-plane shape anisotropy especially for the moments in a thicker alloyed interfaces where exchange is weaker. This points to a more continuous morphology of the Fe layers. The in-field data also support this conclusion. A moderate external field of 0.1 T increases T_c by 100 K. The shoulderlike shape of susceptibility curve may be explained then by the action of in-plane shape anisotropy associated with the 2D character of the FM layers induced by external field. The change in dimensionality is also confirmed by bulk magnetization data. Temperature dependence of hysteresis loops measured with in-plane applied field is very different for the present multilayer. For the earlier 7.3 Å sample, temperature dependence of the coercivity resembles the behavior for single domain particles, collapsing rapidly on warming from 5 to 90 K to a very small value (~5 mT). While for the present 8.5 Å sample the coercivity remains constant at ~15 mT from 25 K to 150 K. It de-

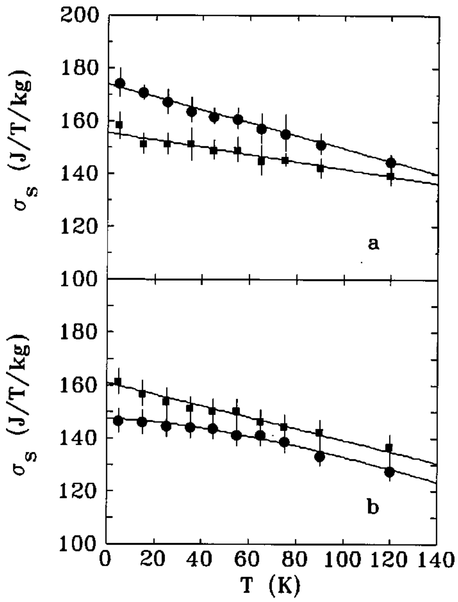

FIG. 3. Temperature dependence of spontaneous magnetization measured in external field of 4 T for the (Cu 38.5 Å/Fe 7.3 Å)×36 multilayer (a), and the (Cu 21.0 Å/Fe 8.5 Å)×36 multilayer (b): circles, external field oriented parallel to the multilayer plane; squares, external field oriented perpendicular to the multilayer plane.

creases linearly to zero only at higher temperatures. Such behavior implies multidomain state and hence continuous 2D character of the Fe layers.

For the earlier 7.3 Å sample the temperature dependence of spontaneous magnetization at low temperatures measured with external field oriented parallel to the multilayer plane $\sigma_s^{\parallel}(T)$ was found to be linear, with a fitted slope of $A_{\parallel} = (139.7 \pm 3.6) \times 10^{-5}\ \mathrm{K}^{-1}$.[2] Both the linear behavior and the value of the slope being consistent with the data on thin epitaxial Fe films having island structure.[5] The slope is inversely proportional to the number of atomic layers if the anisotropy associated with the spin wave gap energy is small.[6] For the slope of spontaneous magnetization measured with the external field oriented perpendicular to the multilayer plane ($\sigma_s^{\perp}(T)$) which is shown in Fig. 3(a) we get a much smaller value $A_{\perp} = (88.6 \pm 8.6) \times 10^{-5}\ \mathrm{K}^{-1}$. This may be explained by an increase in the effective thickness of Fe islands in the case when the external field acts along (approximately) the anisotropy direction, i.e., for the out-of-plane orientation of magnetization considering that the exchange interactions are weaker in the rough and alloyed interfaces.

The strong enhancement of the in-plane shape anisotropy upon transition to continuous Fe layers is reflected in the change of $\sigma_s^{\parallel}(T)$ from linear for the 7.3 Å sample to Bloch-like ($T^{3/2}$) for the 8.5 Å sample [as shown in Fig. 3(b)] supporting earlier observations that the transition to a quasilinear dependence correlates with a loss of the continuous structure.[7] For the 8.5 Å sample a fitted spin-wave parameter $B_{\parallel} = (9.9 \pm 0.5) \times 10^{-5}\ \mathrm{K}^{-3/2}$ is even larger than the $B = 6 \times 10^{-5}\ \mathrm{K}^{-3/2}$ found for interface magnetization of similar multilayers.[8] While $\sigma_s^{\perp}(T)$ clearly shows linear behavior with $A_{\perp} = (135.0 \pm 9.8) \times 10^{-5}\ \mathrm{K}^{-1}$ which is much larger than the corresponding value for the 7.3 Å sample. The large values of $B_{\parallel}$ and $A_{\perp}$ can be understood if to consider larger thickness of the alloyed interface regions in the present multilayer. The contribution of the hyperfine fields from alloyed interfaces in the Mössbauer spectrum at 93 K for the 8.5 Å sample is higher (70%) than for the 7.3 Å sample (50%) in agreement with the increased interface roughness as follows from low-angle x-ray reflectivity data. A weaker pinning of spins in a thicker alloyed interface will result in a weaker spin-wave stiffness constant and hence, in a larger net spin-wave parameters $B_{\parallel}$ and $A_{\perp}$.

For the 7.3 Å sample, $\sigma_s^{\perp}(T)$ may be modeled as well, though with larger error, by a $T^{3/2}$ law, while $\sigma_s^{\parallel}(T)$ cannot be modeled by this law satisfactory. This shows that in this sample, the perpendicular surface anisotropy is stronger than the in-plane shape anisotropy if the latter is present at all. A fitted $B_{\perp} = (7.6 \pm 0.9) \times 10^{-5}\ \mathrm{K}^{-3/2}$ is of the same order as $B_{\parallel}$ for the 8.5 Å sample in which in-plane shape anisotropy is clearly present but is weaker than perpendicular surface anisotropy. The less alloyed and more flat interfaces of the 8.5 Å sample lead to a stronger perpendicular surface anisotropy than for the 7.3 Å sample, a conclusion that is confirmed by the in-field Mössbauer measurements. Thus, $\sigma_s^{\perp}(T)$ dependence of the 8.5 Å sample is expected to exhibit appreciable curvature. In contrast to expectations, it cannot be fit by $T^{3/2}$ law. This shows the important role of the weakening of pinning of spins in the alloyed interfaces and of the in-plane shape anisotropy for excitations of spin waves in 2D ferromagnetic films with both normal and in-plane orientation of magnetization.

IV. CONCLUSIONS

Our results show that the curvature in the temperature dependence of spontaneous magnetization σ_s of Fe/Cu multilayers is associated with strong magnetic anisotropy either in-plane shape or perpendicular surface anisotropy. The transition of $\sigma_s^{\parallel}$ from Bloch-like ($T^{3/2}$) to T^1 dependence correlates with the change to discontinuous morphology of Fe layers. The shape of $\sigma_s^{\perp}$ depends on the interplay between perpendicular surface anisotropy and the weakening of pinning of spins in the alloyed interface region together with in-plane shape anisotropy.

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