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NMR and magnetization studies of Co/Cu superlattices

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Co NMR and magnetization have been measured in a series of Co/Cu superlattices grown by molecular-beam epitaxy, with Co thicknesses varying from 5 to 100 Å. The Co structure is mainly fcc for thicknesses of up to 65 Å. For thicker Co layers the structure becomes hcp. For thin layers it is shown that the hyperfine field corresponding to fcc regions is nearly isotropic, whereas that arising from faulted (hcp) regions is anisotropic, with the symmetry axis parallel to the film normal. The anisotropic part of this field has the same sign and order of magnitude as in bulk hcp Co. For the sample with 5-Å Co layers the dispersion of the magnetocrystalline anisotropy over the sample can be ascribed to Co-layer thicknesses which vary from 2 to 3 monolayers (4 to 6 Å) throughout the sample.

I. INTRODUCTION

In a previous paper[1] we have shown that the observation of a NMR signal originating at the interfaces in superlattices with thin layers proves to be useful in establishing the presence of atomically abrupt interfaces. We report in this paper on the investigation of structures with different Co-layer thicknesses. Both NMR and magnetization data are used in order to correlate the microscopic and macroscopic properties of the Co/Cu superlattices.

The samples were prepared in a Vacuum Generators VG-80 molecular-beam-epitaxy system on annealed GaAs(110) substrates. The Cu thickness was maintained at 23±3 Å, and the total superlattice thickness is 1500 Å. The Co-layer thickness ranged from 5 to 100 Å. Detailed x-ray studies[2] have shown fcc (111) growth for layer thicknesses of up to 65 Å. A single Co layer of thickness 1000 Å on GaAs(110) was also used for comparison.

The NMR measurements were carried out at 2 K using a variable-frequency spin-echo apparatus. The samples were mounted in a Dewar tail around which was fitted an exciting coil, such that the rf field was parallel to the film plane. A dc magnetic field up to 12 kOe can be applied perpendicular to the coil axis. The surface area of the film ranged from 0.3 to 0.8 cm^2. The magnetization measurements were performed at 4.2 K using a superconducting quantum interference device (SQUID) magnetometer with a dc field of up to 30 kOe.

II. EXPERIMENTAL RESULTS AND INTERPRETATION

The Co spin-echo spectra were first measured in zero external field (Fig. 1). For thin layers the peak near 215 MHz arises from regions having the fcc structure, whereas the high-frequency wing arises from stacking faults,[3] which produce local hcp symmetry. The Co spectra undergo a striking change when the Co thickness increases to 100 Å (Fig. 1), with a peak near 222 MHz. This value is rather low as compared to that observed in a Au/hcp Co/Au sandwich at 228 MHz.[4] It may be recalled that, for bulk hcp Co, the resonance frequencies are $F_{\parallel}=220$ MHz and $F_{\perp}=228$ MHz when the magnetic moment is parallel and perpendicular to the c axis, respectively.[5] The anisotropy in the hyperfine field originates mostly from the orbital contribution, giving rise to an anisotropic g factor. In order to obtain further insight on the structure of these films, we studied the effects of a dc magnetic field on the resonance frequencies, bearing in mind that the hyperfine field arising from the Fermi contact term is antiparallel to the magnetic moment. For

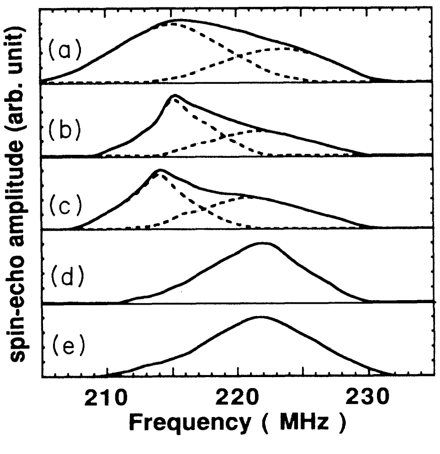

FIG. 1. Co spin-echo spectra in zero applied field at 2 K in Co-Cu superlattices with Co-layer thicknesses (in Å) of (a) 20, (b) 40, (c) 65, and (d) 100. (e) Spectrum for a single Co layer of 1000 Å.

^{59}Co nuclei the observed shift ΔF (MHz) in the resonance frequency is practically equal to the shift ΔH (kOe) in the resonance field, viz., $\Delta F/\Delta H = 1.0054$ MHz/kOe.

We first consider the case of a dc field parallel to the film plane. For the Co-Cu superlattice with a Co-layer thickness of 40 Å, the whole spectrum shifts to lower frequencies at a rate close to -1 MHz/kOe (Fig. 2), as expected for Co nuclei. This shift without deformation indicates that the hyperfine field arising from the hcp-like regions is isotropic within the plane, showing the coincidence of the c axis and the film normal. For Co thickness of 100 and 1000 Å, the resonance frequencies decrease more slowly (Fig. 3). This behavior is typical of an unsaturated magnetization and/or an anisotropic hyperfine field. Actually, the magnetization reaches the saturation in a dc field (parallel to the film) of about 10 kOe for both samples (Fig. 4). As the dc field H is much smaller than the hyperfine field H_n, only its projection onto H_n is effective for the shift in the resonance frequency. Since the hyperfine field is only slightly anisotropic, it remains practically antiparallel to the magnetic moment whatever the orientation of the latter. Thus the corresponding change in the effective field (resonance field) is given by $-H\cos\gamma$, where γ is the angle between H and the magnetic moment. The average value of $\cos\gamma$ was taken from the magnetization curve in Fig. 4. The $H\cos\gamma$ value thus obtained, which is significant only near the saturation field, is clearly greater than the observed shift (Fig. 3). The difference ΔH_n is then ascribed to the angular variation of H_n. This increase of H_n with H would reflect the rotation of the magnetic moment toward predominantly a direction perpendicular to the c axis. Indeed, for a uniaxial crystal the hyperfine field is given by the relation

$$H_n(\theta) = (H_{\parallel}^2\cos^2\theta + H_{\perp}^2\sin^2\theta)^{1/2} , \tag{1}$$

where θ is the angle between the magnetization and c axis. The $H_{\parallel}$ and $H_{\perp}$ values in the Au/Co/Au sandwich are about 215 and 226 kOe, respectively.[4] If the c axis in the samples with 100 and 1000 Å Co thicknesses is tilted from the film normal, the magnetization will tend to align along the projection of the c axis onto the plane because of the uniaxial anisotropy of hcp cobalt. Thus, in zero external field, the magnetization makes an angle $\theta < 90°$ with the c axis. This angle can be estimated to be $\theta = 50°$ from the maximum change $\Delta H_n \approx 4$ kOe in Fig. 3, assuming that the difference $H_{\parallel} - H_{\perp}$ is the same as for the Au/Co/Au sandwich.

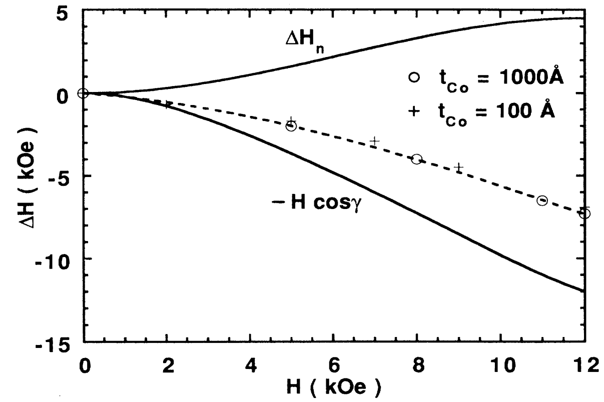

FIG. 3. Shift in the resonance field as a function of dc magnetic field parallel to the film plane for the samples with Co-layer thicknesses of 100 Å (circles) and 1000 Å (crosses). $-H\cos\gamma$ represents the expected shift for isotropic hyperfine field and ΔH_n the variation of the anisotropic part (see text).

When the field is applied perpendicular to the film, the demagnetizing field also contributes to the shift in the resonance frequency. For the 40-Å sample with Co(111) growth, the uniaxial magnetic anisotropy is expected to arise from the interfaces and hcp-like (faulted) regions. Owing to the strong exchange coupling within each layer, this anisotropy can be described by an effective constant

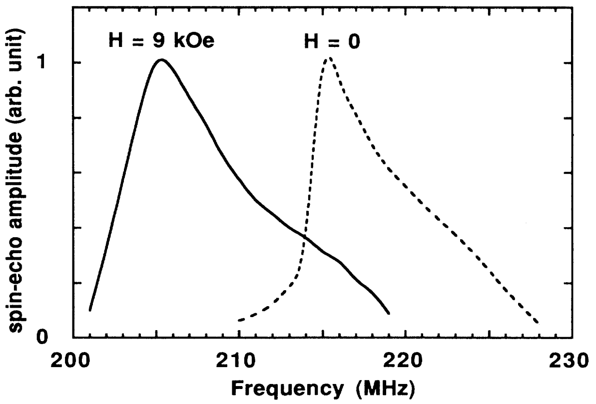

FIG. 2. Co spin-echo spectrum at 2 K in Co-Cu superlattice with a Co-layer thickness of 40 Å with a 9-kOe field parallel to the film plane. The dotted line represents the spectrum in zero external field, normalized at the same height.

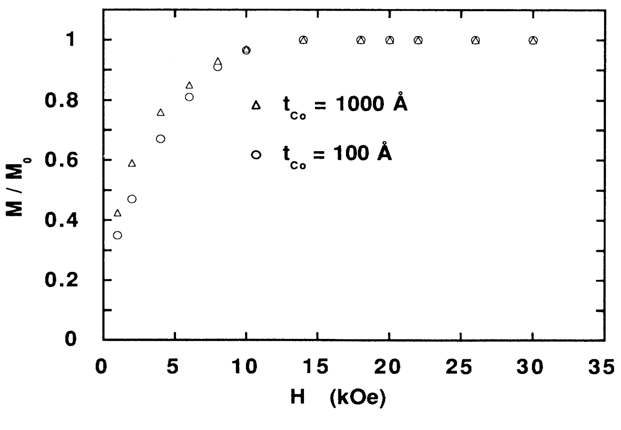

FIG. 4. Reduced magnetization as a function of the magnetic field parallel to the film plane for the samples with Co-layer thicknesses of 100 Å (circles) and 1000 Å (triangles).

anisotropy K_u. In NMR and SQUID experiments, the film normal can be oriented, with respect to the dc field, within a few degrees. We assume that no magnetic anisotropy is present in the film plane so that the dc magnetic field H, the magnetization M, and the film normal or c axis are coplanar. If α and θ are the angles, from the c axis, of H and M, respectively, the energy density is given by the expression

$$E=-HM\cos(\theta-\alpha)+K_u\sin^2\theta+2\pi M^2\cos^2\theta \ , \tag{2}$$

where the first term represents the Zeeman energy, the second the magnetocrystalline anisotropy, and the third the magnetostatic energy. The equilibrium position of the magnetization is given by the relation

$$(4\pi M-H_A)\sin\theta\cos\theta-H\sin(\theta-\alpha)=0 \ , \tag{3}$$

where $H_A=2K_u/M$ is the anisotropy field. This equation can be solved numerically for θ. We can now calculate the projections of the magnetic and demagnetizing field onto H_n, named ΔH, which gives rise to the shift in the resonance frequency. ΔH is given by the expression

$$\Delta H=4\pi M\cos^2\theta-H\cos(\theta-\alpha) \ . \tag{4}$$

The saturation field in perpendicular orientation is defined as $H_S=4\pi M-H_A$. For positive K_u the magnetization is more easily rotated to the c axis and ΔH is expected to increase up to approximately H_S, and then decrease as $\cos\theta$ is close to unity. For the 40-Å Co-layer sample, the spectrum peak indeed begins to shift to higher frequencies (Fig. 5), but unlike the case of parallel orientation, the line shape becomes more and more symmetric, showing differing behavior of fcc and hcp regions, represented by the dotted and dashed lines, respectively. The relative intensity of these two lines is taken as that observed in zero external field. As the dc magnetic field is increased, the fcc line clearly moves to higher frequencies, whereas the hcp spectrum remains practically unshifted. The fcc hyperfine field is expected to be isotropic, whereas the hcp one is necessarily anisotropic. The estimated shift of the fcc line is shown in Fig. 6. The magnetization of the 40-Å sample versus dc field in parallel and perpendicular orientations is presented in Fig. 7. The saturation magnetization was measured to be 1300 emu/cm^3, which agrees with the bulk value to within the uncertainty in the Co thickness. The solid curve for the perpendicular orientation represents the calculated value of $\cos(\theta-\alpha)$, using Eq. (3), with $H_S=10$ kOe and $\alpha=2°$. Next, the shift in the resonance field was calculated from Eq. (4), taking the demagnetizing field $4\pi M$ to be 16 kOe. The qualitative agreement for $\alpha=2°$ between the calculated and observed values shows that the hyperfine field in fcc regions is quasi-isotropic. This angle corresponds to the sample alignment error in the experimental setup.

For the hcp-like regions with a slightly anisotropic hyperfine field Eq. (1) can be rewritten as

$$H_n(\theta)=H_{\text{iso}}+H_{\text{an}}(3\cos^2\theta-1)/2 \ , \tag{5}$$

where H_{iso} and H_{an} constitute the isotropic and anisotropic parts, respectively. The $F_\perp$ value is about 222 MHz from the spectrum in zero external field [Fig. 1(b)]. By choosing $F_\parallel=217$ MHz, one obtains a variation of H_n which nearly cancels the shift ΔH due to the dc and demagnetizing fields (Fig. 6, dashed line). The resonance frequency corresponding to H_{iso} is then deduced to be 220.3 MHz, as compared to the bulk value of 225.3 MHz. The smaller H_{iso} value in the superlattice sample partially arises from an expansion of the Co lattice, as already observed for the fcc regions.[1] Nevertheless, the H_{an} values are of the same sign and order of magnitude, showing that the local symmetry of the layers with stacking faults is similar to that of bulk hcp Co. This similarity is consistent with the positive sign of K_u in Co-Cu superlat-

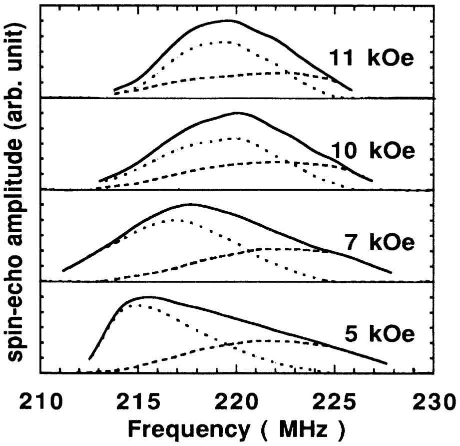

FIG. 5. Co spin-echo spectra at 2 K in Co-Cu superlattice with Co-layer thickness of 40 Å, for different values of magnetic field perpendicular to the film plane. The dotted and dashed lines represent the contributions of the fcc and hcp regions, respectively.

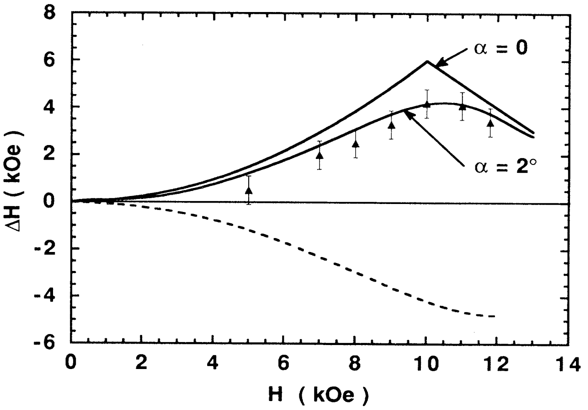

FIG. 6. Shift of the resonance field in the 40-Å sample as a function of the magnetic field perpendicular to the film plane. The solid lines represent the theoretical variations with deviation angles α of 0° and 2°. The dashed line represents the variation of the anisotropic hyperfine field corresponding to the hcp regions.

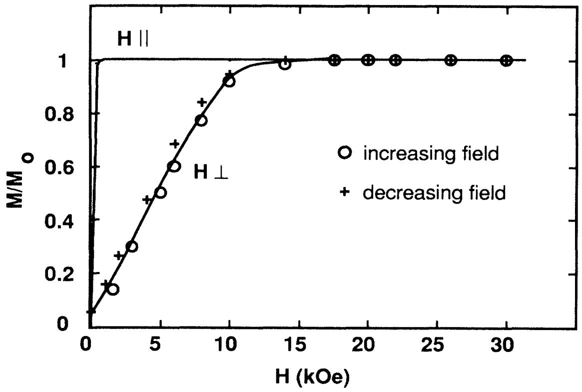

FIG. 7. Reduced magnetization as a function of the dc magnetic field, parallel and perpendicular to the film plane, for the 40-Å sample. The solid line for the perpendicular orientation represents the calculated curve with $H_S=10$ kOe and $\alpha=2°$ (see text).

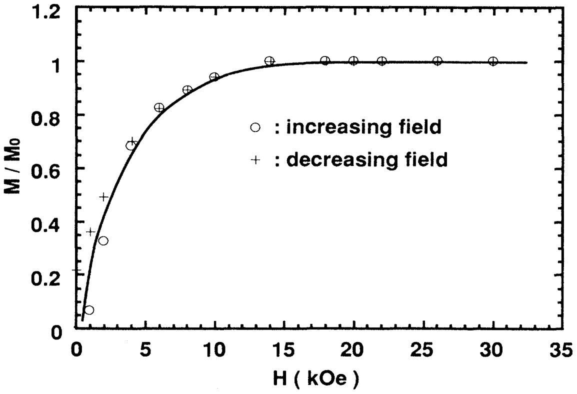

FIG. 8. Reduced magnetization as a function of the magnetic field parallel to the film plane (hard direction) for the Co-Cu superlattice with Co-layer thickness of 5 Å. The calculated solid line is the sum of six contributions with H_S values equal to 2, 4, 6, 8, 10, and 12 kOe, respectively.

tices, corresponding to that of the first anisotropy constant K_1 of hcp Co.

No NMR signal was observed in the range 150–230 MHz for the sample with 5-Å Co layers, which has a perpendicular easy axis of magnetization.[6] The absence of a nuclear signal reflects probably a very short nuclear transverse relaxation time in a quasi-two-dimensional ferromagnet layer.[7] The saturation magnetization was found to be 1427 emu/cm^3, which varies from the bulk by a value within the uncertainty in the Co thickness. The effective anisotropy field H_A can be deduced from the parallel saturation field $H_{S\parallel}=H_A-4\pi M$. However, unlike the case of the 40-Å sample (Fig. 7), the field dependence of the magnetization is far from linear (Fig. 8), revealing a significant distribution in $H_{S\parallel}$ values. The magnetization curve can be approximately fitted with a uniform distribution of $H_{S\parallel}$ values between 2 and 12 kOe. It is deduced that H_A would vary from about 18 to 28 kOe throughout the sample. Such a large distribution is not surprising, owing to the very small thickness of the Co layers. Indeed, the 5-Å nominal thickness can be considered as the average value of local thicknesses varying between 4 and 6 Å, i.e., containing either two or three monolayers, with the same probability. Assuming that the magnetic anisotropy arises mainly from the interfaces, H_A would vary as the inverse of the thickness, in agreement with the extreme H_A values of 18 and 28 kOe. If two regions with different thicknesses belong to the same layer and are exchange coupled, the effective H_A value will be intermediate.

III. CONCLUSION

In conclusion, by using the NMR and magnetization data, we have determined the local structure of Co in Co-Cu superlattices. For thin Co layers the hyperfine field corresponding to (111) fcc stacking is nearly isotropic, showing that the local environment has cubic symmetry. In hcp stacking regions the anisotropic part of the hyperfine field was found to have the same sign and order of magnitude as in bulk hcp Co, reflecting similar local symmetry. For the sample with a Co-layer thickness of 5 Å, a broad distribution of the perpendicular anisotropy was observed. This dispersion was related to spatial fluctuations of the Co thickness from two to three monolayers over the sample, with predominantly interface anisotropy.

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