# Interfacial Anisotropy in Magnetic Superlattices

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We have observed large discrepancies between the dc magnetization and ferromagnetic resonance (FMR) magnetic anisotropy in Mo-Ni superlattices. We show that higher-order anisotropy is present in both measurements and develop an analysis by which the first-order and second-order energies can be treated independently. The low-temperature results show a systematic divergence of the first-order FMR and dc anisotropy as the Ni-layer thickness decreases. It is shown that the source of the divergence is an enhanced surface anisotropy measured by FMR. Possible sources of this enhancement are discussed.

The properties of magnetic compositionally modulated structures are of considerable interest because of the insight they provide into the fundamental nature of magnetism. Of particular interest is the nature of the anisotropy in these materials because no clear-cut theoretical understanding has emerged of its origin as yet. Potential applications are also affected by these anisotropies. For example, high-density information storage in magnetic films requires the storage media to have anisotropic magnetic properties, and as the data transfer rates become increasingly higher the frequency dependence of the anisotropy will become more important. Artificial superlattices are ideal systems for the study of dimensional behavior in thin layers by conventional methods since sufficient material is present so that measurements can be performed with relative ease. The magnetic anisotropy of superlattices gives important information about the state of the material in the film and as we shall show, particularly the state of the interface.

This investigation examines the magnetic anisotropy as a function of Ni-layer thickness ($d$) in Mo-Ni superlattices. We show, for the first time, that in superlattices the anisotropy changes smoothly from planar to perpendicular as $d$ increases. In addition, we observe a discrepancy between the ferromagnetic resonance (FMR) and dc magnetization anisotropies and include higher-order anisotropy in the analysis to show that the first-order anisotropy discrepancy increases with decreasing Ni-layer thickness. We demonstrate that the difference is due largely to an enhanced surface anisotropy sensed by FMR.

Samples used in this investigation have been prepared and characterized by a variety of techniques and the results have been published elsewhere.[1-4] Briefly, the samples were prepared on temperature-controlled mica substrates by a magnetron sputtering technique. The struc-

ture of the films was determined by use of low- and high-angle x-ray diffraction and electron microscopy techniques. The dc magnetization was measured with a SQUID magnetometer in the temperature range 5–300 K and in fields up to 10 kG. Standard 9.2-GHz FMR measurements were made at room temperature and $T = 30$ K (to avoid a $T = 4$ K sample holder signal) on samples ranging in Ni-layer thickness from 9 to 5000 Å, each having a total thickness of approximately 1 $\mu$m. The temperature control was provided in a helium flow-through cryostat. The data were digitized and analyzed with the use of a computer. Magnetic field accuracy is $\pm 10$ G.

The anisotropy field is extracted from the FMR spectra according to the expressions

$$hf = g\mu_B H_r, \tag{1}$$

$$H_r = [H_\parallel (H_\parallel + D)]^{1/2}, \tag{2}$$

$$H_r = H_\perp - D, \tag{3}$$

where $h$ is Planck's constant, $f$ is the microwave frequency, $H_r$ is the internal resonance field of the Ni, $H_\parallel$ and $H_\perp$ are the applied fields parallel and perpendicular to the film surface, respectively, $D = 4\pi M + H_a$ is the total uniaxial anisotropy field, $M$ is the saturation magnetization obtained from dc magnetization measurements,[2,4] and $H_a$ is any uniaxial anisotropy in excess of the shape anisotropy. Note that $H_a > 0$ implies planar and $H_a < 0$ implies perpendicular anisotropy. Because of the rather high planar anisotropy energies present in these samples and because of the discrepancy between FMR and dc magnetization results to be discussed later, the magnetization is not aligned with the applied field in the perpendicular direction at 9.2-GHz resonance; therefore, although perpendicular measurements were made, the an-

isotropy analysis is based solely upon parallel measurements assuming $g = 2.21$.

Figure 1 shows the low-temperature anisotropy versus Ni thickness for $d_{Ni} = d_{Mo}$ samples obtained from FMR (solid squares) and dc magnetization (open squares). Similar results are found at room temperature. Several features are to be noted: (1) The anisotropy changes systematically from high values in thin Ni layers to lower values in thicker layers. This trend is contrary to expectation if strain is the sole source of $H_a$.[5] (2) The anisotropy changes from planar uniaxial $(H_a > 0)$ to perpendicular $(H_a < 0)$ at approximately 100-Å Ni thickness. (This change of the anisotropy has been previously observed in single-crystal NiFe films,[6] but to our knowledge it is the first observation of such behavior in superlattices.) (3) Very large planar anisotropy is present in thin Ni layers at low temperature. The resonance spectra for these thinner samples $(H_a > 10 \text{ kG})$ occur at very low fields and are progressively more truncated (although not appreciably distorted) at $H$(applied) $= 0$ as $H_a$ increases. (4) Systematic differences exist between anisotropies extracted from FMR and dc measurements. Particularly, there is no transition from parallel to perpendicular $H_a$ observed by dc measurements. A similar discrepancy between FMR and dc magnetization measurements was also reported earlier in Cu-Ni superlattices; however, a physical understanding of its origin was never presented.[7]

Since the Ni layers are textured, with the [111] direction perpendicular to the plane of the film, but polycrystalline in the plane, the anisotropy is expected to be uniaxial. This has been verified by torque measurements.[8]

Let us now examine the discrepancy between the low-temperature FMR and dc magnetization extracted anisotropies. The inset in Fig. 2 shows magnetization data obtained with the field perpendicular ($z$ direction) to the

Mo(25 Å)-Ni(25 Å) sample surface at $T = 5$ K. It is quite apparent from the curvature of the data that higher-order energies are present in the uniaxial anisotropy. The magnetization energy can be written as

$$E = (2\pi M^2 + K_1)\sin^2\phi + K_2 \sin^4\phi - MH\sin\phi, \quad (4)$$

where $\phi$ is the angle between the magnetization and the plane of the film. The first term is comprised of magnetostatic demagnetization and any additional first-order anisotropy energy, the second term represents second-order anisotropy energy, and the last term is the potential energy of $M$ in the external field. The condition of equilibrium in an applied field is given by $\partial E / \partial \phi = 0$, or

$$M_z/M = H/[H_K^{(1)} + H_K^{(2)} \sin^2\phi], \quad (5)$$

where $M$ is the saturation magnetization, $M_z = M\sin\phi$, $H_K^{(1)} = 4\pi M + 2K_1/M$, and $H_K^{(2)} = 4K_2/M$. This treatment neglects domain-wall interactions, most likely a valid assumption in the samples measured by dc techniques, because of the thickness ($\lesssim 100$ Å) and total anisotropy energies ($< 0.1$ J/cm$^3$)[9] and to the small amount of perpendicular hysteresis observed.[2] It follows from (5) that the Kittel resonance equations (1)–(3) are modified by higher-order uniaxial anisotropy as follows:

$$H_{a,\parallel} = 2K_1/M = H_a^{(1)},$$

$$H_{a,\perp} = 2(K_1 + 2K_2)/M. \quad (6)$$

Therefore, the parallel ($\phi = 0$) resonance will not be sensitive to the higher-order anisotropy and remains unaltered from Eq. (2), but the perpendicular resonance should reflect the higher orders in anisotropy. The $H_a$ values presented have been based upon parallel FMR measurements and therefore should be compared with the first-order anisotropies measured by dc magne-
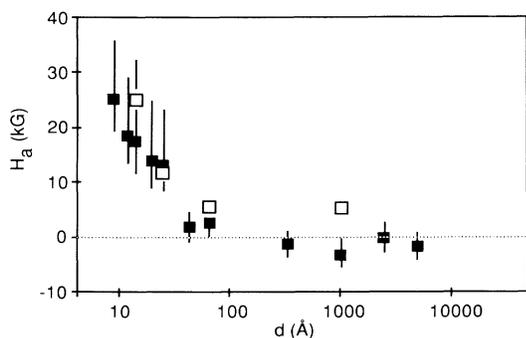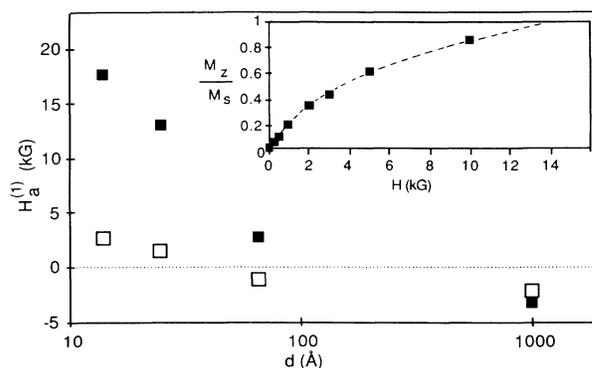


FIG. 1. Total low-temperature anisotropy, less demagnetization, vs Ni-layer thickness. Solid squares are FMR and open squares are dc magnetization results. FMR error bars are extracted from the uncertainty in the parallel resonance position. The dotted line marks the transition from planar to perpendicular $H_a$.



FIG. 2. Low-temperature, fist-order anisotropy vs Ni-layer thickness for FMR (solid squares) and dc magnetization (open squares). The dotted line marks the transition from planar to perpendicular $H_a$. Inset: dc data taken on the Mo(25 Å)-Ni(25 Å) sample, compared to the higher-order anisotropy model (dashed line).

tometry. The physical origin for the difference between the two types of measurements is clear. The FMR slightly tips the moment out of the parallel direction, whereas dc magnetization requires the moment to tip 90°; therefore, FMR in the parallel direction only senses the first-order anisotropy, in contrast to dc magnetization which senses all higher-order anisotropies. The first-order dc anisotropy can be extracted by fitting the experimental dc magnetization measurements to Eq. (5). An example of the fit is shown in the inset of Fig. 2 and the results extracted are as follows: For samples with $d_{Ni} = 14$, 25, 65, and 1000 Å the values of $2K_1/M$ are 2.7, 1.3, $-1.1$, and $-2.2$ kG and the values of $H_k^{(2)}$ are 22.1, 10.3, 6.5, and 7.4 kG, respectively. From this analysis we note the following: (1) The dc data fit very well the higher-order anisotropy analysis. This analysis allows an accurate extrapolation to $M_z/M = 1$, thus modifying the previously reported[2] results for $H_k$. (2) The first-order dc results show, similar to FMR, a transition from parallel to perpendicular anisotropy. (3) The second-order anisotropy energy is considerably larger than the first-order energy as deduced by dc techniques. According to the above theory the *perpendicular* FMR resonance should be sensitive to this second-order effect. However, at the frequency of the present experiment (9.2 GHz) the moment is not saturated (as shown by the dc magnetization) and so the simple resonance conditions do not apply. Further, theoretical work to understand the resonance conditions for $M$ not saturated along the perpendicular $H$ and experimental measurements at several frequencies are underway to address these points. (4) The magnetization is clearly saturated in the parallel FMR measurements; therefore, the difficulties discussed in point (3) above do not apply and $H_a^{(1)} = H_k^{(1)} - 4\pi M$ extracted from parallel FMR and dc measurements should be compared. Figure 2 shows good agreement in the thickest Ni-layer sample and a progressively greater discrepancy in the thinner layer samples. We considered the possibility that relaxation effects[10] were shifting our resonance peaks to produce this enhanced anisotropy. According to our calculations these effects would introduce in the worst case a 15% shift in our reported $H_a$ values; moreover, they would produce *even greater discrepancy* between FMR and dc results.

In order to understand the cause for the FMR-dc differences it is instructive to examine the sources of the first-order anisotropy. It is commonly accepted that the anisotropy (less demagnetization) can be expressed[11] in terms of surface and volume contributions according to

$$H_a = [2K_v + 4K_s(1/d_{Ni})]/M, \tag{7}$$

where $K_v$ and $K_s$ are the volume and surface anisotropy energies, respectively. The extra factor of 2 in the surface expression arises from the fact that each Ni layer has two (assumed identical) surfaces. It is known that the magnetization in these samples is dependent upon

Ni-Layer thickness.[2] In addition, in the spirit of the temperature dependence of crystal-field anisotropy (see, for example, Callen and Callen[12]) it will be assumed that the anisotropy energies are proportional to $M^c$ where $c$ is a constant to be determined. Upon correcting $H_a$ for magnetization dependence, Eq. (7) is fitted to

$$H_a/M^{(c-1)} = [a + b(1/d)], \tag{8}$$

where $a$, $b$, and $c$ are extracted, given the experimentally measured $D$, $d$, and $M$. The fitting routine involved a least-squares fit of the parameters $a$ and $b$, choosing the value of $c$ which provided the greatest correlation coefficient. The fit of the low-temperature measurements to the model for the samples that satisfy the $T \ll T_c$ condition is shown in Fig. 3, and the results of the analysis are as follows: for FMR (dc), $c = 1.44 \pm 0.1$ (1.38 $\pm 0.1$), $2K_v/M = -4.0 \pm 0.8$ ($-2.4 \pm 0.2$)kG, and $K_s = 0.54 \pm 0.08$ (0.11 $\pm 0.01$) erg/cm². General features to be noted are (1) the FMR and dc exponents on $M$ are in agreement, (2) the dc and FMR *volume* anisotropies are both perpendicular and are comparable in magnitude, and (3) there is a factor of 5 difference in the *surface* anisotropy energies.

This analysis leads one to conclude that the discrepancy between the low-temperature FMR and dc, first-order anisotropies lies primarily in an enhanced surface anisotropy sensed (and perhaps induced) by FMR. The origin of this anisotropy is not easy to ascertain; however, two possible mechanisms could be operational. They are as follows.

(1) Surface Currents: Although Ni and Mo are good metals, they have different electrical conductivities, and therefore interfacial surface currents will be induced by the microwave magnetic field. Such surface currents would likely alter the state of the interface, perhaps enhancing the surface anisotropy. Vittoria[13] has developed a detailed formalism which calculates multilayer
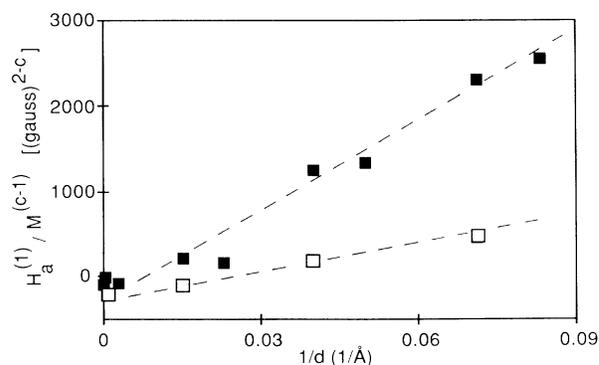
FIG. 3. Results of linear least-square fits (dashed line) of the anisotropy field to Eq. (8) for dc magnetization (open squares) and FMR (solid squares) low-temperature measurements.

interface impedances under FMR conditions and which may provide further insight into this large FMR anisotropy.

(2) Surface roughness: An intriguing model has been proposed recently by Liu,[14] which treats interface surface roughness between dissimilar materials with fractal geometry. By assuming fractal irregularities, one might be able to calculate equivalent ac impedances having well-defined frequency dependences for the interfaces. These impedances would presumably modify Vittoria's approach.

FMR measurements at various frequencies are essential in determining whether these or other mechanisms give rise to the anisotropy frequency dependence.

In conclusion, we have observed large discrepancies between dc and FMR first-order magnetic anisotropy energies in Mo-Ni sublattices. We have shown that higher-order anisotropy terms are important in both techniques and we have presented a method that allows the first- and second-order energies to be treated independently. Results show a systematic divergence of the first-order FMR and dc anisotropy as the Ni-layer thickness decreases. Its origin is an enhanced surface anisotropy sensed by FMR. It is clear from this analysis that future attempts to compare FMR and dc anisotropy energies need to take the higher-order terms into account. It is also apparent that the frequency dependence of magnetic multilayer anisotropy is a rich and largely unexplored area and warrants further investigation.

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