Observation of Thickness Dependence of Magnetic Surface Anisotropy in Ultrathin Amorphous Films

R. J. Hicken, G. T. Rado, Gang Xiao,[a] and C. L. Chien
Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218
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Ferromagnetic resonance (FMR) and SQUID magnetometry measurements have been made on multilayers of amorphous $Fe_{70}B_{30}$/Ag. The dependence of the magnetic surface anisotropy constant K_s on the magnetic layer thickness $2L$ has been determined in the range $1.6\ \text{Å} < 2L < 90\ \text{Å}$ using more than twenty samples. It is found that K_s is constant for $2L > 16.5$ Å, but decreases monotonically towards zero as $2L$ decreases from 16.5 Å towards zero. The FMR results can be well described by a theory developed for ultrathin amorphous ferromagnetic layers.

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There is currently much interest in artificially layered materials consisting of magnetic and nonmagnetic layers which have thicknesses of a few tens of Å or less. As the thickness $2L$ of the magnetic layer is decreased, the physical properties of the multilayer as a whole are increasingly affected by those of its interfaces. Of primary importance in the description of the magnetic properties of ultrathin films is the magnetic surface anisotropy energy density E_s first introduced by Néel.[1] Its uniaxial form for an amorphous material is given by $E_s = -K_s\cos^2\theta$, where θ is the angle between the magnetization and the film normal while K_s is the magnetic surface anisotropy constant.[2] In our notation the magnetic surface anisotropy term E_s favors an out-of-plane magnetization for $K_s > 0$. The value of K_s can be most conveniently determined in a flat-film geometry where the film thickness is varied. This approach has been used in several recent investigations. In most cases in which thin magnetic films were used, whether single layers or multilayers, K_s appears to be independent of thickness, the value depending upon whether the specific magnetic medium was Co,[3-5] Fe,[5,6] Ni,[7,8] or an amorphous ferromagnet.[9] In ultrathin films of Fe on Ag some variation of K_s has been noted among four samples.[10] In this Letter, we present the first systematic and conclusive study of a thickness dependence of K_s. As discussed below, amorphous $Fe_{70}B_{30}$/Ag is particularly suitable for such an investigation. More than twenty samples were used, the $Fe_{70}B_{30}$ thickness $2L$ spanning the range from 1.6 to 90 Å. We demonstrate that K_s is constant for thicker films ($2L > 16.5$ Å) but becomes strongly thickness dependent in the ultrathin limit ($2L < 16.5$ Å). The ferromagnetic resonance (FMR) results at two frequencies in both perpendicular and parallel applied fields are in good agreement with each other and can be excellently accounted for by the FMR theory of Zhang and Rado,[9] which was specifically developed for ultrathin amorphous ferromagnetic films. dc magnetometry measurements also agree very well with the FMR results.

The use of amorphous magnetic materials in multilayered structures is particularly attractive for a number of reasons. Amorphous films have no macroscopic in-plane anisotropies and may be combined with a variety of spacer materials and substrates, since they are not subject to the conditions required for epitaxial growth. The amorphous film is likewise not subject to the defects that such constraints may produce in a crystalline film[11] and which may cloud the magnetic properties of the system. The spacer layers of Ag were chosen because of their immiscibility with Fe. Characterization studies of our samples are described more fully in Ref. 12 but we shall restate the main findings here because of their relevance to the present work. Large-angle x-ray diffraction confirmed that the $Fe_{70}B_{30}$ layers were indeed amorphous while the Ag layers were crystalline. Small-angle x-ray diffraction revealed superlattice peaks appearing at scattering vector magnitudes given by integer multiples of $2\pi/d$, where d is the bilayer thickness. Cross-sectional electron microscopy revealed flat continuous layers of $Fe_{70}B_{30}$ and Ag with no sign of island structure for a sample in which $2L = 10$ Å. Resolution of the layer structure in the thinnest of our samples is of course very difficult and beyond the capabilities of the instruments available. However, even these films have a continuous and island-free layer structure because there is no hint of superparamagnetism from either SQUID measurements[12] or the FMR measurements that will be described below. We note that the frequencies used in the FMR measurements differ from those used in the SQUID measurements by 10 orders of magnitude. The saturation magnetization at $T = 8$ K was measured for a number of values of $2L$, the smallest of which was 3 Å, and was found to be independent of $2L$. This implies that the magnetization at the surface of the $Fe_{70}B_{30}$ layer is unaffected by the interface and hence that there is no interdiffusion between the $Fe_{70}B_{30}$ and Ag layers. This is not entirely unexpected since Fe and Ag have negligible mutual solubilities. Finally, the temperature dependence of the magnetization $M(T)$ for various layer thicknesses has also been determined.[12] These favorable

conditions facilitate the exploration of magnetic surface anisotropy in ultrathin films.

In order to explore the $2L$ dependence of K_s we have made FMR measurements at room temperature on $Fe_{70}B_{30}$/Ag multilayers grown on Kapton substrates. The use of multilayers rather than single layers was essential for the observation of a strong FMR signal. The ratio of the Ag thickness to the $Fe_{70}B_{30}$ thickness $2L$ was maintained at 3:1 throughout.[13] The multilayer samples contain from 32 to 726 bilayers. Details of fabrication and of the $2L$ dependence of the magnetization as determined by SQUID magnetometry measurements were given previously.[12] More than twenty samples were used to cover the range of $1.6\ \text{Å} < 2L < 90\ \text{Å}$. The sample used for FMR typically had a lateral dimension of about 1 mm to assure uniform film thickness across the sample. The FMR field was measured with the static applied field both parallel and perpendicular to the plane of the sample at frequencies of 9.52 and 23.9 GHz. Some of the perpendicular resonance fields could not be measured since they exceeded the 14 kOe available from our magnet.

Some representative FMR signals are shown in Fig. 1. The signals are clean despite the presence of ultrathin layers with $2L = 3$ Å. Thus they attest to the quality of samples consisting of several hundred layers and to the fact that the data reveal the behavior of a single layer. In the perpendicular configuration, the relevant FMR equations[9] are

$$\omega/\gamma = H - 4\pi M - 2Ak^2/M\,, \tag{1}$$

$$Ak\tanh(kL) = -K_s\,, \tag{2}$$

where H is the applied magnetic field, k is the wave number of the microwave magnetic field, ω is the circular frequency, and, for $Fe_{70}B_{30}$, $A = 1.38\times10^{-6}$ erg/cm and $\gamma = 18.38$ MHz/Oe. Since the resonance field can be measured, K_s can be determined from Eqs. (1) and (2). In the parallel configuration, the corresponding relations[9] are equally well known but more complicated in form and will not be reproduced here. The FMR equations[9] mentioned above were obtained from the equation of motion of the magnetization in which the surface anisotropy was introduced via the general exchange boundary conditions. These and other aspects of the theoretical background of the experimental observations (such as the nature of the k values and the associated excitations) are presented elsewhere.[9] The magnetization M was determined independently by SQUID magnetometry. While $M(0\ \text{K})$ is independent of $2L$, $M(300\ \text{K})$ does depend on $2L$. It is found that $M(300\ \text{K})$ can be adequately described by $M(300\ \text{K}) = [1054 - 1194(1/2L)]$ emu/cm^3 for $2L > 6.8$ Å and $M(300\ \text{K}) = [942 - 434(1/2L)]$ emu/cm^3 for $2L < 6.8$ Å.

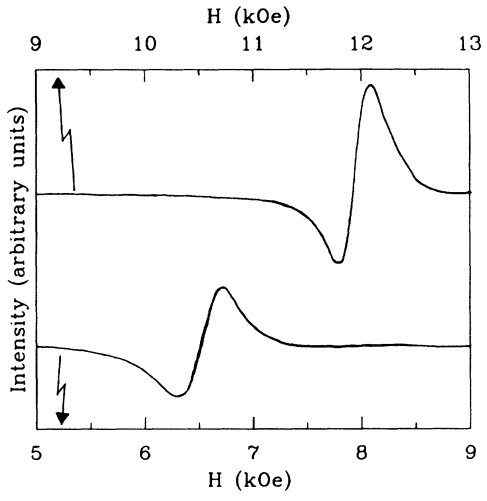

FIG. 1. FMR traces at 23.9 GHz for a 3.0-Å sample. The first derivative of the absorption is plotted against the applied static magnetic field H. The upper and lower traces are for the perpendicular and parallel configurations, respectively.

The resonance fields at 9.52 and 23.9 GHz with both perpendicular and parallel applied fields are shown in Fig. 2. The values of K_s calculated from each FMR measurement have been averaged and plotted in Fig. 3 for each thickness. To demonstrate the self-consistency of the results, the four theory curves (shown in Fig. 2) have been generated from Eqs. (1) and (2) and others using the single set of K_s values shown in Fig. 3. From Fig. 3 it can be seen that K_s has a constant value of 0.34 erg/cm^2 for $2L > 16.5$ Å but then decreases monotonically towards zero as $2L$ is decreased from 16.5 Å, conclusively demonstrating that in the ultrathin limit K_s decreases with film thickness.

We have also determined K_s for $2L = 4.1$, 6.7, 10.2, and 16.7 Å by extracting the "knee field" H_{knee} from the SQUID-determined perpendicular magnetization curves at $T = 300$ K. In the perpendicular configuration of the magnetization measurement, H_{knee} is defined as the field at which M is just saturated. With the inclusion of the magnetic surface anisotropy, $H_{\text{knee}} = 4\pi M - (4K_s/M) \times (1/2L)$, from which K_s can be determined. The values of K_s obtained from the SQUID data are also plotted in Fig. 3 and are seen to be in very good agreement with those determined from the four sets of FMR measurements.

It must be emphasized that the decrease in the value of K_s for $2L < 16.5$ Å is certainly not an artifact resulting from the $2L$ dependence of $M(300\ \text{K})$. If a constant value of M equal to 1054 emu/cm^3 is assumed for all values of $2L$, then K_s again decreases towards zero for $2L < 16.5$ Å but now the value of K_s for $2L > 16.5$ Å is found to be slightly higher, being about 0.40 erg/cm^2. The excellent agreement between five independent measurements of K_s, four at microwave frequencies and one essentially static, strongly supports the validity of the model[9] used, and rules out the possibility of the film

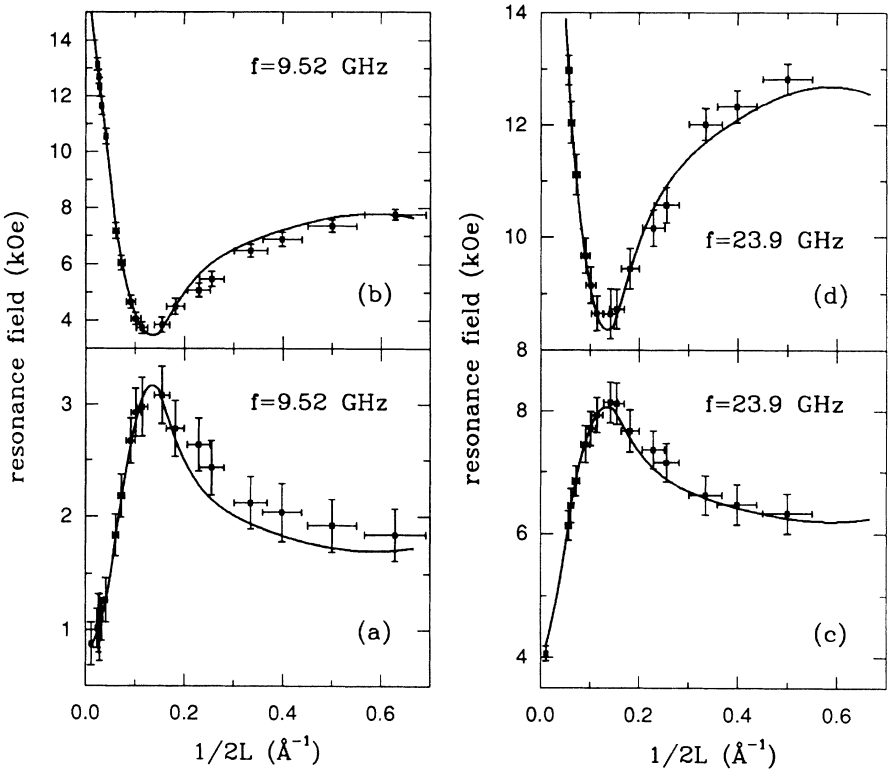

FIG. 2. Dependence of the magnetic-resonance field on the reciprocal of the magnetic layer thickness $2L$. The applied magnetic field is parallel to the film plane in (a) and (c), and perpendicular to the film plane in (b) and (d). The four curves were calculated on the basis of Ref. 9 by using the method described in the text.

breaking up into an island structure as an explanation for the decrease of K_s with decreasing $2L$. Furthermore, for an island structure the profound difference between the perpendicular and parallel FMR configurations would be strongly diminished. As shown in the picture reproduced in Ref. 12, cross-sectional transmission electron microscopy revealed the layer structure to be very good for $2L = 10$ Å; that is, well below the value of $2L$ at which K_s begins to decrease. We wish to stress that the experimental data on K_s shown in Fig. 3 differ from a proportionality to $2L$ by an amount exceeding experimental error. Even if K_s were exactly proportional to $2L$, it would be unreasonable to describe the experimental data by introducing a constant (i.e., thickness-independent) effective volume anisotropy K_v of some unknown origin for $2L < 16.5$ Å while retaining a constant surface anisotropy K_s for $2L > 16.5$ Å. This is because it is highly improbable that the values of K_v and K_s, which are independent, satisfy the equality $K_v = K_s/2L$ for $2L = 16.5$ Å, as demanded by the continuity of the experimental data.

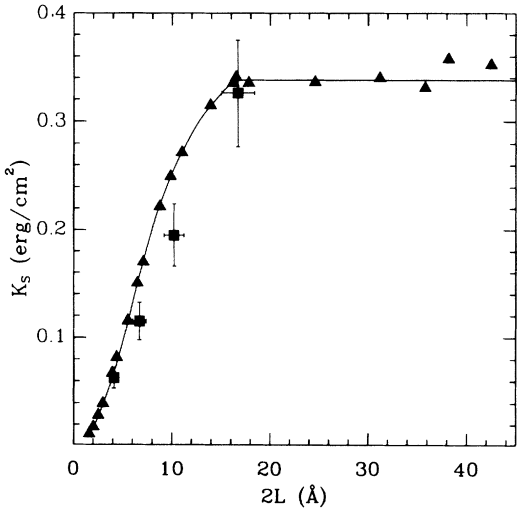

FIG. 3. Dependence of the magnetic surface anisotropy constant K_s on the magnetic layer thickness $2L$. Data points obtained by FMR and SQUID measurements are denoted by triangles and squares, respectively. The solid line is an interpolation of the data points.

The main result of this paper is that K_s is constant for $2L > 16.5$ Å but decreases monotonically and continuously towards zero as $2L$ decreases from 16.5 Å towards zero. Although several mechanisms have been suggested that may reduce[14] the value of K_s, it is by no means clear that one or more of these mechanisms would account for a monotonic decrease of K_s with decreasing $2L$ in an amorphous material. We suggest that the fundamental interactions contributing to the surface anisotropy energy in our samples have a range up to half of 16.5 Å. It seems conceivable that such relatively long-range interactions perhaps arise from the single-ion mechanism

described (after an appropriate averaging over the atomic sites) by the quadratic axial spin Hamiltonian term DS_z^2, where the z axis is now perpendicular to the film. This term involves, in our case, the combined action of the electrostatic field at the magnetic surface ions of the amorphous ferromagnet and the spin-orbit coupling. We note that in a ferrite a D term has already been shown to produce[15] a surface anisotropy energy in agreement with experiment. In the present case, of course, it would be essential to include in the calculations of D the electrostatic field arising from ions located farther than nearest neighbors.

The success of our model[9] in fitting the experimental data reaffirms the utility of the concept of a surface anisotropy energy. Moreover, we have demonstrated conclusively that the value of this energy is thickness dependent in ultrathin films and therefore cannot simply be inferred from experimental or theoretical investigations of the surface of a bulk solid.

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[a]Present address: Department of Physics, Brown University, Providence, RI 02912.

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