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SATURATION MAGNETIZATION AND PERPENDICULAR ANISOTROPY OF Fe/GaAs(110) EPITAXIAL FILMS STUDIED BY THE EXTRAORDINARY HALL EFFECT

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The magnetic field dependence of the extraordinary Hall effect (EHE) has been used to determine the sum of the perpendicular magnetic anisotropy and the saturation magnetization of thin (5–20 nm) Fe films. The films were grown on (110) surfaces of GaAs by molecular beam epitaxy. The free surface of the films was not protected thereby allowing an iron oxide layer to form upon removal from the MBE apparatus. The relation between the film thickness and the sum of the perpendicular anisotropy energy and the saturation magnetization was compared to that determined in a previous study which relied on ferromagnetic resonance (FMR) to measure the same quantity. The FMR study measured both oxide covered films and also films with a protective overcoating of Al to prevent oxidation. It is found that the data from the EHE are not in agreement with the FMR data taken on the iron oxide covered films but instead are in agreement with the FMR data of the protected or nonoxidized films. In addition a determination of the surface anisotropy energy can be made by subtracting the magnetization data measured on overcoated films from the sum determined by the EHE analysis. In this case there is no indication of a large surface anisotropy energy making the perpendicular direction an easy axis.

1. Introduction

The successful adaptation of molecular beam epitaxy (MBE) techniques for the manufacture of high quality single crystal magnetic films with thicknesses as small as a monolayer provides model systems to be used in fundamental magnetics research. In spite of the ideal nature of these epitaxed films, a number of the usual magnetic measurement techniques, e.g. SQUID and vibrating sample magnetometry, applied to these materials are complicated by the magnetic response of the substrate materials. Although the substrates possess smaller magnetic signals per unit of volume, the substrate constitutes nearly the entire volume of the film/substrate combination. One way to overcome the difficulty of the separation of the substrate contribution is to use a measurement which is insensitive to the magnetic signal from the substrate portion of the sample. One measurement of this type is ferromagnetic resonance (FMR) which measures only that portion of the sample which contains the magnetic ions. Electrical transport is another selective measurement and, although frequently overlooked, can be used to determine many of the same quantities usually determined by more traditional magnetic measurements.

In this paper we report the results of a study of the extraordinary Hall effect (EHE) [1] in epitaxial iron films and compare the magnetic information determined by the transport study with that previously obtained by FMR [2–4] on similar samples. As will be discussed in the theory section, the magnitude of the applied magnetic field necessary to saturate the magnetization perpendicular to the plane of a film can be determined in an EHE measurement. The magnitude of this saturation field is a linear combination of the saturation magnetization and the perpendicular anisotropy energy divided by the saturation magnetization of the magnetic film [5], i.e. $4\pi M_s + K_s/M_s$ where M_s is the saturation magnetization and K_s is the perpendicular or surface anisotropy energy. This same quantity can also be determined by FMR. For both the EHE study and the FMR study the films had thicknesses in the range from 5 to 20 nm. In the case of the EHE samples, the free iron surface was allowed to form an iron oxide surface layer when removed from the MBE system. For the FMR study two sets of samples were investigated; one with an iron oxide surface as in the EHE study and a second set which had a nonmagnetic overlayer of Al which prohibited the formation of the iron oxide.

In the final section of the paper, the discussion section, we report that the numerical values we obtain for the above sum determined by the EHE are consistent only with FMR data on films coated with the Al overlayer used to inhibit any oxidation of the iron. Although at first result may be surprising we attribute the result to the FMR in the oxide coated films as measuring a coupled resonance spectrum of the metallic and oxide iron ions whereas the FMR on the overcoated films and the transport measurement measure only metallic iron. Also in this final section we utilize previous magnetization measurements [6] made on the overcoated or protected films and the sum discussed above to make an estimate of an upper limit for the surface anisotropy energy in the oxide coated films.

2. Sample description

The epitaxial Fe films are deposited on polished, chemically etched and UHV thermally annealed GaAs(110) substrates using a PHI model 400 molecular beam epitaxy (MBE) system. Vacuum zone refined Fe is deposited at a rate of 0.3–0.4 nm/min with a background pressure during growth of about 5×10^{-8} Torr. Of particular importance to the magnetics results presented here is the substrate and Fe films are characterized using in situ reflection high energy electron difraction (RHEED) and Auger electron spectroscopy (AES). The RHEED studies suggest initial island growth below about 2.5 nm with the islands in registry with the GaAs substrate. Above about 2.5 nm the islands merge and epitaxial growth occurs. No evidence of Fe induced surface reconstruction or interface reaction is found. In contrast, recent sychrotron radiation photoemission studies [7] of the Fe/GaAs(110) system suggest that a reacted phase with both Ga and As intermixing with the Fe overlayer and significant surface segregation of As occurs. It is not clear whether a difference in substrate preparation (thermal anneal vs. cleaved), substrate temperature or some other factor is responsible for this discrepancy. Therefore, although RHEED indicates good epitaxial Fe films are produced, the possibility of significant Ga and As intermixing near the interface should not be neglected. In fact in the discussion section we will return to the question of intermixing and its effects on the surface anisotropy energies arising from the GaAs/Fe interface and the iron magnetic moment. Other details of the MBE growth process have been reported elsewhere [8].

Although the ratio of the lattice constants of GaAs and Fe is very nearly 2:1 enabling good epitaxial growth, the lattice match is not perfect and the Fe lattice is compressed 1.4% relative to bulk Fe at the Fe/GaAs interface. RHEED data indicates that this strain relaxes at a distance from the interface not greater than about 20 nm. The film thickness is determined by monitoring the Fe flux using a quadrupole mass analyzer calibrated by measuring thicker films with X-ray fluorescence. The nominal thickness range studied is 5–20 nm or roughly 25–100 monolayers. The actual metallic Fe thickness probed in the transport measurements is approximately 1–1.5 nm less [9] due to the formation of an oxide layer upon removal from the UHV system. In what follows, the effective thickness of the metallic Fe below the oxide will be assumed to be 1.5 nm less than the deposited film thickness. This oxide layer self passivates, inhibiting further oxidation, and is remarkably stable. For example, no significant resistivity changes are found after prolonged exposure to the ambient atmosphere and repeated thermal cycling. As stated in the introduction, part of the FMR study used samples coated with overlayers of Al to prevent the formation of the oxide layer. The transport properties of these samples was not measured due to the parallel conduction path in the Al overlayer.

The Fe/GaAs(110) films are prepared for transport measurements by producing Hall bar patterns using standard photolithography techniques. The Hall bars are oriented along the [001], [110] and [111] directions contained in the (110) plane and are configured to allow both magnetoresistance and Hall effect measurements along these high symmetry directions. After lithographic preparation the transport properties of the films are measured at ambient temperature, 77 and 4.2 K. The results presented here consist only of those measurements of the EHE taken at 4.2 K. The other transport results will be presented elsewhere.

3. Theory

For the iron films used in this study, one needs only to consider two contributions to the free energy [10,11] to describe the behavior of the perpendicular component of the magnetization in magnetic fields applied normal to the film plane. The first is the shape or demagnetization energy which is due to dipole–dipole interactions and the second is the surface or interface anisotropy energy arising from the reduced symetry of the free surface or interface of a magnetic film [12,13]. Although in general, one should also consider the usual cubic anisotropy energies, for the iron films studied here the demagnetization energy alone is sufficiently large so that they can be neglected. This energy argument cannot be made for the surface anisotropy in view of recent theoretical calculations by Gay and Richter [14] and we will therefore not neglect it.

The first of the two thin film energies, the demagnetization energy F_D, is given by

$$F_D = AtN_z M_z^2/2, \qquad (1)$$

where A is the surface area, t is the film thickness, M_z is the component of magnetization perpendicular to the film plane and $N_z = 4\pi$ is the demagnetization factor appropriate for a thin film. The other magnetic contribution to the free energy to be considered here is due to the reduced local symmetry at the surface or interface (F_S). This energy was first introduced by Néel [12,13] and is given to first order by

$$F_S = AK_s\left(M_z^2/M_s^2\right), \qquad (2)$$

where K_s is a phenomenological coefficient known as the surface anisotropy constant which is, of course, an average of contributions from both surfaces and M_s is the saturation magnetization value.

Using the above two energies and the energy of a magnetized film in an external field, the total free energy in an external magnetic field H_z can therefore be written as

$$F = -AtH_z M_z + 2\pi AtM_z^2 + AK_s\left(M_z^2/M_s^2\right). \qquad (3)$$

In equilibrium, M_z is easily found to depend on the applied field as

$$M_z = \left(4\pi + 2K_s/M_s^2 t\right)^{-1} H_z \qquad (4)$$

so that in this simple model the component of the magnetization perpendicular to the plane of the thin film is predicted to depend linearly on the applied field up to the point were the magnetization saturates out of the plane of the film. If the saturation field H_S is defined as the field at which $M_z = M_s$ then

$$H_s = 4\pi M_s + (2K_s/M_s)/t \qquad (5)$$

and so a linear relation between H_s and $1/t$ is predicted. From the intercept and slope of an H_s vs. $1/t$ plot one can extract the saturation magnetization and the surface anisotropy constant. Although the preceding analysis is extremely simple and depends only on the fact that the surface to volume ratio is $1/t$, it is interesting that precisely the same $1/t$ dependence is found for the ferromagnetic resonance (FMR) field using a more sophisticated approach [15]. It is important to note in the above analysis of the H_s vs. $1/t$ plot it is implicitly assumed that the saturation magnetization is constant and homogeneous throughout the film and not affected by the presence of the interface. The importance of this last comment will be manifest in the last section where a plot of H_s versus thickness is complicated by a thickness dependence of the saturation magnetization M_s.

4. Experimental details and analysis

The Hall voltage measurements were carried out using a standard ac resistance bridge with a constant excitation current of 20 μA. The three contact method [16] was used with a potentiometer to zero the measured voltage in the absence of an applied field. In a number of the measurements a magnetoresistive component was detected across the Hall contacts and was attributed to small inhomogeneities in the samples. This magnetoresistive component was subtracted from the Hall signal by reversing the applied field and using the fact that the Hall effect is odd and the magnetoresistance is even with respect to the applied field.

The saturation field H_s defined in the preceding section may be experimentally determined by measuring the hall voltage as a function of applied field [17]. The relation between H_s and the Hall voltage is related to the fact that in a ferromagnetic material, the Hall resistivity is given by

$$\rho_H = R_0 B_z + 4\pi R_s M_z, \qquad (6)$$

where R_0 and R_s are the ordinary and extraordinary Hall coefficients, respectively. This expression is simply the linear combination of the ordinary Hall effect and the extraordinary Hall effect. From the analysis of the previous section (in particular eq. (4)) one would expect ρ_H to increase linearly with applied field and show an abrupt change of slope at the field $H = H_s$. Above H_s the magnetic field dependence would be entirely due to the ordinary Hall effect since the magnetization would be saturated.

The experimentally observed Hall resistance as a function of applied magnetic field is somewhat different from this ideal behavior when the perpendicular component of the magnetization is close to the saturation magnetization of the films. An example of this behavior is shown in fig. 1 which is a plot of the Hall resistance as a function of applied field for a 6 nm thick Fe/GaAs(110) film. The lack of a linear dependence of ρ_H with applied fields in this field region (near H_s) may indicate either higher order anisotropy energy terms are required in eq. (3) or the applied field is not aligned with the sample normal (the misalignment is estimated to be less than 1°). Because of this lack of linearity H_s is operationally defined as the field value where the intersection of the two lines given by a linear fit to the low and high field regions of the curve occurs.

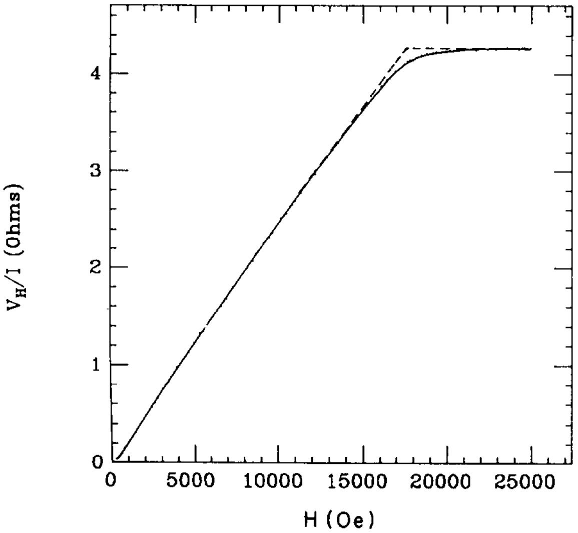

Fig. 1. The measured Hall voltage as a function of magnetic field is shown. The intercept of the linear fits to the low field and high field data is defined as H_s. Shown is data taken with the current in the ⟨110⟩ of a film with nominal thickness of 7.5 nm.

In addition to obtaining a value for H_s, data of the type exhibited in fig. 1 may be used to determined two other film properties of importance. One is the ordinary Hall coefficient which is determined by the slope of Hall resistance after the magnetization has been saturated and the second is the extraordinary Hall effect coefficient R_s, which is determined from the low magnetic field slope. The ordinary Hall coefficient provides information pertaining to possible alterations of the electron energy bands due to both growth associated strains and possible alloying effects at the

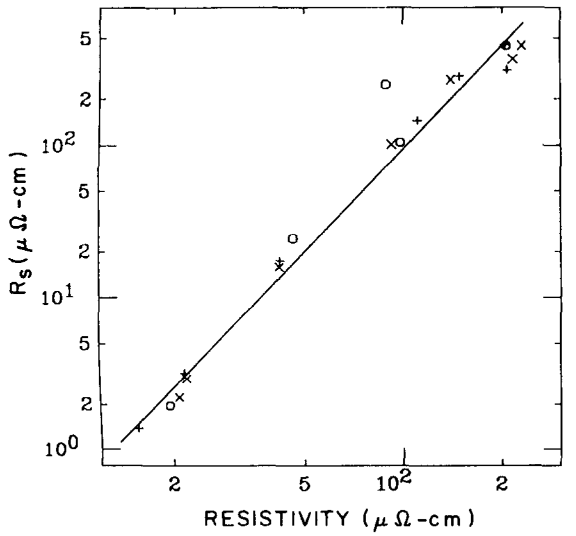

Fig. 2. A log–log plot of the saturation magnitude of the EHE or R_s versus the resistivity of the films at 4.2 K. The straight line fitted to these data determines the scattering mechanism responsible for the EHE in the iron films as described in the test. The crystallographic dicrection of the current is in the ⟨100⟩, ⟨110⟩ and the ⟨111⟩ direction for the data marked with the symbol +, × and o, respectively.

Fe/GaAs interface [18]. The extraordinary Hall coefficient is useful in determining the mechanism responsible for this asymetric magnetic scattering. As shown in fig. 2 the proportionality constant of the EHE, R_s, is found to depend quadratically on the resistivity of the films. This quadratic dependence is consistent with the side jump scattering mechanism proposed by Berger [19] as being the origin of the EHE in iron.

5. Discussion

In discussing the results of this study there are two major areas of interest on which we will focus; one is a comparison of the results of this study with data from an earlier FMR study and the second involves the question of surface anisotropy at the GaAs/Fe interface in these films. The data relevant to both areas is the thickness dependence of H_s. In fig. 3 is plotted the saturation field or H_s, determined in the manner described in the previous section, versus the thickness of the films. Included in this figure are two sets of data [20] from an earlier FMR study [3] which determines $4\pi M_s + (2K_s/M_s)/t$ (this quantity is the same as the H_s defined in eq. (5)) by fitting the angular dependence of the FMR field. The difference between the two sets of films used for accumulating the FMR data is that one set of films had the free surface protected by an Al overlayer whereas the second set was allowed to form an iron oxide on the surface. It can be seen that the EHE data are consistent with the Al protected FMR samples and not the oxidized samples which is surprising since the EHE samples have oxidized surfaces.

Although in detail we cannot state why this correlation exists we will present two different scenarios either of which could explain the data. The first is that if there were higher order anisotropy energies present in the oxidized films which alter the FMR spectrum then the simple model used to explain the FMR data would not provide the correct value for H_s. This origin, which has has observed in metallic superlattices [21], would indicate an anisotropy energy arising from the iron/iron oxide interface which is not present

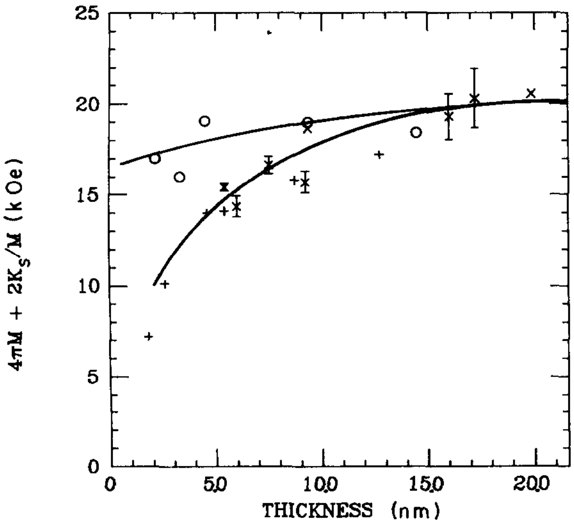

Fig. 3. Plot of $H_s = 4\pi M_s = 2K_s/M$ extracted from the EHE study compared to the results from FMR measurements from reference [3]. ×'s are the EHE data, +'s are the FMR data taken on the Al coated films, and o's are the FMR data taken on the oxide coated films. The lines are guides to the eye.

at the iron/aluminum interface. The other scenario stems in part from the fact that the difference between the EHE and FMR measurements of H_s is the EHE measurement is a transport measurement and therefore is only sensitive to the metallic portion of the epitaxial film. The FMR measurements on the other hand are a measure of the spin dynamics and if coupled strongly enough can be affected by either (both) the iron spins in the surface oxide or (and) the spins in the interfacial GaAsFe region [22]. This coupling might cause the FMR data to be an average over both the metallic and any other species of iron compound present. Naturally since the iron oxide layers are expected to be constant thickness [2,8] i.e. independent of the initial thickness of the epitaxial iron film, then this average of the FMR properties would be less representative of the pure iron for the thinnest films.

We now turn to the second area we would like to discuss; the possibility of large surface anisotropy energies at the GaAs/Fe interface. The rationale for considering this possibility stems from the calculation of Gay and Richter [14] which predicted a surface anisotropy energy for Fe grown on the (100) surface of Ag would be of sufficient magnitude that the easy axis of thin iron films would be in the direction perpendicular to the plane of the films. This remarkable prediction was subsequently confirmed experimentally [23]. For the case of iron films grown on the (110) surface of GaAs such as studied here the calculations are more difficult [24] and therefore a prediction of the magnitude of the surface anisotropy is not possible.

Although the calculations have proven difficult for iron films grown on the (110) surface of GaAs, experimentally the surface anisotropy energy may be probed by either EHE or FMR. As shown in fig. 3, the linear combination of the saturation magnetization and the anisotropy energy perpendicular to the plane of the film are a function of the thickness of the films. As shown in eq. (5) if there is a surface anisotropy present at the interface then one would expect there to be a thickness dependence to this sum. However, without an independent determination of the saturation magnetization one doesn't know if the thickness dependence exhibited in fig. 3 is an effect of the surface anisotropy energy or if there is a thickness dependence to the saturation magnetization. In ref. [6] SQUID magnetometry was used to measure the saturation magnetization of epitaxial Fe films grown on the (110) surface of GaAs. In fig. 4 the results of both the magnetization and the H_s determined by the EHE have been plotted as a function of the film thicknesses. As can been seen from this figure the values of H_s are consistent with a negligible surface anisotropy energy. This value for the surface anisotropy should be contrasted with the anisotropy energy of films grown on the (100) surface of GaAs where the surface anisotropy energy is sufficient to overcome the demagnetization energy and make the direction perpendicular to the plane the easy axis.

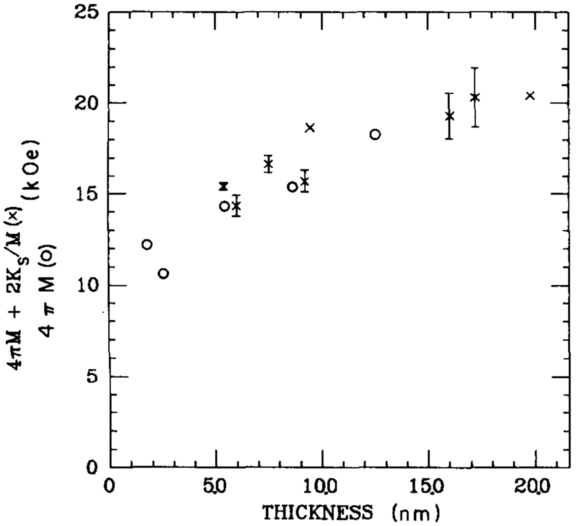

Fig. 4. Plot of H_s determined by the EHE (×) and the saturation magnetization (○) versus the thickness of the films. As can be seen in this figure, the value of H_s is consistent with the surface anisotropy being negligible compared to the demagnetization energy.

In conclusion we would like to note that in the case of the differences between the FMR and the EHE of the oxidized films there is not as yet sufficient information to determine the mechanism leading to the origin of this effect but we have presented two possibilities. The importance of understanding this phenomenon lies in the potential devices applications which rely an exchange coupling at interfaces. In the case of the perpendicular anisotropy energy, we have shown that these films, which were grown on the (110) surface of GaAs, exhibit no evidence of an unusually large anisotropy energy. We should caution however that the effect as predicted by Gay and Richter [14] should be extremely sensitive to the growth surface of the GaAs and any chemical processes which occur.

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