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Sputter Grown Fe and Fe/Cr Multilayers With Fourfold Magnetic Anisotropy on GaAs

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Thin films of Fe have been epitaxially sputtered on GaAs substrates with native oxide removal prior to the deposition carried out by an Ar ion milling. Films grown at substrate temperatures above 100 °C show well-defined fourfold anisotropies. The onset of epitaxial growth is accompanied by an increase in the surface roughness with growth occurring in a distinct island-like pattern. The Fe layers show significantly reduced moments, which decrease with increasing temperature. Antiferromagnetic coupling between Fe layers with Cr spacers was measured in a multilayer with a Cr thickness of 2.7 nm, around the second antiferromagnetic peak. The magnetic properties of the films are discussed in the context of multilayer storage applications.

Index Terms—Magnetic anisotropy, magnetic hysteresis, magnetic multilayers, spintronics.

I. INTRODUCTION

The growth of Fe on GaAs has been widely studied as a model system of ultrathin film magnetism, including the onset of ferromagnetism [1], [2], changes between uniaxial and cubic anisotropy with thickness [2], [3], domain wall motion [4], [5], and Ruderman–Kittel–Kasuya–Yosida (RKKY) coupling [6], [7]. It is also a system used for studying spin injection into semiconductors, where the relatively large spin polarization of Fe films has been combined with GaAs-based devices for both spin light emitting diode [8], [9] and lateral spin injection and detection experiments [10]. Another technologically relevant area using the thin magnetic films is data storage. For such applications, not only single layers but also multilayer structures with controllable interlayer coupling between the layers are of interest. Recently, it has been proposed to use RKKY coupled stacks to create 3-D data storage devices, where the information is propagated out-of-plane [11]–[14]. The growth of layers with well-defined uniaxial [13], [14] or fourfold anisotropies [15] and RKKY coupling is required for such multilayer devices.

Epitaxial Fe has been widely grown by molecular beam epitaxy (MBE) on GaAs surfaces prepared by high temperature oxide removal and annealing [1], [3], [8]. Growth by sputtering is a more commercially relevant technique but it is not certain that the same well-defined magnetic properties can be achieved. Previously, the epitaxial growth of Fe by sputtering has been obtained on oxide substrates, such as sapphire, using metal underlayers grown at high temperature before Fe growth at lower temperature [16]–[18]. In this way, controlled multilayer stacks demonstrating surface spin-flop and soliton states have been made. Whilst this method allows the growth of controlled multilayers, spintronic applications may also require a high-quality interface with GaAs [8]. Fe has been sputtered epitaxially on GaAs using either high temperatures [19], [20] or very low sputtering rates [21].

Fe grown on GaAs is known to intermix at relatively low temperatures (around 150 °C) although post-growth annealing at these temperatures can improve the quality of the interface [22]. Previous growths of Fe on GaAs using sputtering have been able to show both epitaxy and low ferromagnetic resonance linewidths [23], [24].

In this paper, the growth of single Fe layers as well as Fe/Cr multilayers grown epitaxially by sputtering on GaAs is studied. We look at optimizing the growth of single layers in terms of the anisotropies of the layers, particularly, how close they are to an ideal fourfold anisotropy. We also study multilayers in terms of the achievable RKKY coupling. For multilayer data storage, it is particularly important to be able to reproduce the anisotropy and coupling on a layer-by-layer basis.

II. SINGLE FE LAYERS

Samples were grown by magnetron sputtering in a system with a base pressure of $3 \times 10^{-8}$ mbar using an Ar gas pressure of $8 \times 10^{-3}$ mbar. Fe and Cr films were deposited at a power of 50 W, with the deposition rates of around 0.06 nm/s. No in situ characterization techniques are available. Semi-insulating GaAs(100) wafers were used as substrates. Untreated GaAs wafers have a native oxide present at the surface. Fe layers grown directly onto the GaAs with the native oxide still present show a polycrystalline behavior even at elevated growth temperatures. The oxide can be removed by chemical means, for example, etching in HCl. However, the native oxide will rapidly reform, requiring this procedure to be carried out immediately before the introduction of the wafer into the vacuum system. It is, therefore, preferable to use an in situ technique to remove the oxide. Here, the native oxide was removed by an Ar ion milling [19]. The milling system is located in the same chamber as the sputtering magnetrons, which allows for a very small delay between the milling and the start of the Fe growth, so that little oxide will reform on the surface even at the higher base pressures used here in comparison with MBE systems. This higher base pressure is also why annealing was not carried out prior to growth. A grided dc ion milling system with an acceleration...
TABLE I

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thickness (nm)</th>
<th>Ion milling time (s)</th>
<th>Growth temperature (°C)</th>
<th>Growth pressure (mbar)</th>
<th>Ms (emu/cc)</th>
<th>rms roughness (nm)</th>
</tr>
</thead>
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<td>12</td>
<td>15</td>
<td>20</td>
<td>1.5 × 10^-7</td>
<td>790</td>
<td>0.3</td>
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<tr>
<td>B</td>
<td>12</td>
<td>15</td>
<td>50</td>
<td>2.3 × 10^-7</td>
<td>710</td>
<td>0.7</td>
</tr>
<tr>
<td>C</td>
<td>12</td>
<td>15</td>
<td>100</td>
<td>2.6 × 10^-7</td>
<td>690</td>
<td>1.2</td>
</tr>
<tr>
<td>D</td>
<td>12</td>
<td>15</td>
<td>150</td>
<td>3.4 × 10^-7</td>
<td>520</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Fig. 1. Normalized angular remanent moment plots as a function of in-plane angle for 12 nm thick Fe samples grown at (a) 20 °C, (b) 50 °C, (c) 100 °C, and (d) 150 °C. (b) also shows the crystallographic directions of the GaAs substrate.

The Fe layers were then capped with Cr. Structural and magnetic properties of the films are listed in Table I. The magnetic properties of the films were characterized for different milling parameters were kept constant for all growths. The magnetic properties of layers were characterized for different milling times and it was found that 15 s, which corresponds to around 3 nm of milling, was optimal for Fe growth and is used for all the growths presented here. This would correspond to the thickness range of native oxide found on GaAs surfaces exposed to air for several years, which has been found to be between 2 and 3 nm [25]. For shorter milling times the layers do not show fourfold anisotropy (5 s) or have reduced anisotropy (10 s), whilst at longer times (20 s) a reduced anisotropy is seen (not shown here). This behavior is likely due to not fully removing the oxide for shorter milling times and due to increased roughness on the surface caused by the milling for longer times. A series of four 12 nm thick Fe layers were grown on GaAs substrates at different temperatures, which were ion milled for 15 s immediately prior to growth. The Fe layers were then capped with Cr. Structural and magnetic properties of the films are listed in Table I. The growth pressure refers to the background pressure of the chamber with the substrate at growth temperature immediately prior to deposition.

A. Magnetic Properties

Fig. 1 shows the angular remanent moment measurements of the four films. The maxima give the positions of the easy axes whilst minima define the hard axes. It is apparent that there is a change in the anisotropy of the films with increasing growth temperature. For all the measurements here, the [110] direction of the GaAs is aligned along 0°. For the 20 °C and 50 °C films, the anisotropy is a mix of uniaxial and fourfold terms. The uniaxial term easy axis appears to be at around 30°, between the [110] and [100] axes of the GaAs. For MBE grown Fe films on (001) GaAs two regimes of anisotropy with thickness are observed [3]. For films a few monolayers thick the anisotropy is dominated by an interfacial uniaxial term, which has an easy axis along the [110] direction [3]. For thicker films, the effect of this interface term is reduced and the films are dominated by the fourfold anisotropy term expected from the crystallography. The films studied here are 12 nm thick, which is beyond the thickness, where a uniaxial term is expected to dominate, assuming its origin at the GaAs interface.

At 100 °C and 150 °C, the samples acquire a dominant fourfold anisotropy. However, an asymmetry still remains between the two hard axes along the [110] (0°) and [110] (90°) directions. Vibrating sample magnetometer (VSM) hysteresis loops are shown in Fig. 2, for the [110], [110], and [100] directions of sample D (see Table I). The saturation along the [110] direction occurs at around 300 Oe, whilst that for the [110] direction occurs at around 500 Oe. For bulk Fe, the fourfold anisotropy gives a saturation field of 550 Oe. These results suggest the presence of a bulk uniaxial term in the Fe layers, with an easy axis along the [110] direction (0°). Such a term has been found before and shown to be along the [110] direction [26]. It is possible that this is also seen in the 20 °C and 50 °C data, which could be explained by combining a uniaxial term along [110] and a fourfold easy axis along the [100] direction, so causing the apparent intermediate easy direction found experimentally. The existence of the uniaxial term has been ascribed to defects in the bulk of the film, which may be more likely in sputter growth than in typical MBE films [27]. However, the low (<10 Oe) coercivity along the easy axes of the films [see Fig. 2(c)] suggests that defects are not impeding domain wall motion through the films, as has been suggested as a sign of a high density of impurities in the bulk [27].

The saturation magnetization of the films was also extracted from VSM measurements, and is listed in Table I. It is notable how reduced these values are from the expected 1760 emu/cm³ found in bulk as well as MBE grown films [2]. The saturation magnetization also decreases with the growth temperature. From MBE studies, it is known that growth at
raised temperatures may lead to reduced magnetization due to the intermixing of the Fe with the GaAs [28]. This may explain why the saturation magnetization decreases at higher temperatures, but this is on top of the significantly reduced $M_s$ even at room temperature. This may be due to roughness, defects, and inclusions in the film as well as possibly oxidation due to the high background pressure of the chamber combined with the relatively slow rate of growth. Using the $M_s$ values obtained, the effective anisotropy constants can be extracted using $K_1 = M_s H_{sat}/2$. Using the data in Table I and Fig. 2, the fourfold anisotropy terms along the $[1\bar{1}0]$ and $[110]$ directions are $0.8 \times 10^5$ erg/cm$^3$ and $1.3 \times 10^5$ erg/cm$^3$, respectively, compared with an expected $4.8 \times 10^5$ erg/cm$^3$ for bulk anisotropy [2]. Most simply the difference between the two hard axis anisotropies could be explained by the presence of a uniaxial term with an easy axis along the $[1\bar{1}0]$ direction. This would give a fourfold-term of $0.8 \times 10^5$ erg/cm$^3$ and a uniaxial term of $0.5 \times 10^5$ erg/cm$^3$ in this sample.

B. Surface Properties

Contact atomic force microscopy (AFM) on the single layer Fe samples was carried out, with images obtained for samples A and C as shown in Fig. 3(a) and (c), respectively. Single line scans for these samples are shown for sample A in Fig. 3(b) and for sample C in Fig. 3(d). For the samples grown at room temperature [Fig. 3(a)], a smooth surface is found with rms roughness around 0.3 nm, which is the same as that found for a GaAs substrate after 15 s of ion milling. As the temperature is increased, the roughness increases and the epitaxial samples have a distinctly textured surface with increased roughness as shown for sample C in Fig. 3(c). The surface appears to consist of circular columns several nanometers high, with no apparent tendency for growth along any of the underlying GaAs crystallographic directions. The same circular features are visible in the non-epitaxial sample, although much less prominently, which suggests that the texture is not related to the magnetic anisotropy of the films.

III. MULTILAYER SAMPLES

The Fe/Cr system has large RKKY coupling with oscillations over two different periods caused by the antiferromagnetism of the Cr [7]. For sputtered Fe/Cr multilayers the long-wavelength oscillation has been reported as a function of temperature [29].

Multilayer Fe/Cr superlattices were grown on an Fe (12 nm) / Cr (15 nm) buffer layer. The initial Fe buffer layer ensures the epitaxy of the growth, however, this layer may intermix with the GaAs, so a further Cr buffer is used before the multilayer growth. Shown in Fig. 4 are the hard [Fig. 4(a)] and easy [Fig. 4(b)] axis loops of a [Fe (1.5 nm)/Cr (2.7 nm)]$_9$/Fe(1.5 nm)/Cr (4.5 nm) multilayer. The loops show a slow saturation of the magnetization up to around 6000 Oe, in contrast to the behavior shown in Fig. 2. This is due to the extra field required to overcome the antiferromagnetic coupling between layers. The thickness of Cr used is around that expected for the second antiferromagnetic peak in Cr(001) layers [29]. The data also include the moment from the 12 nm Fe underlayer, which is shown in the low field data in the insets of Fig. 4. Here, the distinction between the easy and hard axis data, which mainly reflects the magnetization of the 12 nm Fe buffer, is more apparent, as seen in the higher remanence and more coercive switching in the easy axis direction.

Shown in Fig. 4(c) is the remanent magnetization after saturation as a function of angle, which shows a remanant
Fig. 4. Normalized VSM loops of an Fe/Cr multilayer sample. (a) [1\(\overline{1}\)0] GaAs direction. (b) [100] GaAs direction. Insets: low field behavior. (c) Remanent magnetization of the sample.

moment of about half the saturation moment with a clear fourfold anisotropy. This is probably due entirely to the Fe underlayer. Assuming the Fe underlayer has the same saturation magnetization as the single layer measured previously, the saturation magnetization of the 10 \(\times\) 1.5 nm Fe multilayer can be calculated as 700 emu/cm\(^3\), which is similar to that found in the single layer samples. From this an antiferromagnetic coupling strength of around \(-0.15\) erg/cm\(^2\) can be obtained. This is slightly larger than has been obtained previously [30], but is dependent on the assumptions made concerning the saturation magnetization of the layers. A large coupling would also be consistent with the lack of anisotropy, as shown in Fig. 4(c). However, multilayers with thicknesses around the first antiferromagnetic peak at 1 nm in Cr did not show any RKKY coupling, and neither did those around the third antiferromagnetic peak at 4.5 nm, both giving results consistent with strongly ferromagnetically coupled layers. This may be due to the roughness of the samples, leading to discontinuous layers for the thin Cr thicknesses. The sample that showed coupling had a large rms roughness of 4 nm and the other multilayer growths showed roughnesses between 2 and 4 nm.

IV. DISCUSSION

The possibility of using sputtered Fe on GaAs for spintronic applications has previously been demonstrated, at least at low temperature [21], [31]. Notably, these examples use ultrahigh vacuum sputtering, which allows low sputtering rates to be used without oxidizing the interface. The high temperatures, and consequently high background pressures, required to create epitaxial samples here may mitigate against the use of this devices in spintronic applications due to the likelihood of increased interfacial intermixing and oxidation. The surface preparation process here also may not favor spintronic applications compared with \textit{in situ} transfer techniques, which can produce sharp clean interfaces [8], [9], [32]. The strongly reduced \(M_s\) also suggests that the spin polarization of such devices would be significantly reduced.

For use in magnetic multilayers, a reduction in \(M_s\) is not problematic as long as it is fairly consistent across the multilayer. It was found that this was the case for the multilayers studied, where the magnetization of the ten times repeats was consistent with that of a single layer. More problematically, it appears that the roughness and column-like growth may lead to a limited range of available coupling strength. The sample shown in Fig. 4 is the only one, grown in the same conditions whilst varying the Cr thickness that showed RKKY coupling. Previously, oscillatory RKKY coupling in sputtered multilayers has been found in epitaxial [33] and non-epitaxial layers [29]. The roughness in the multilayers studied here may be due to the high temperatures used to establish the epitaxial growth of Fe on GaAs. For proposed multilevel memory applications, where each layer has reproducible properties [11]–[15] the use of high temperature buffer layers to provide a smooth metallic surface for the Fe/Cr epitaxy may be preferential, which can be achieved by using, for instance, MgO substrates [18], [33].

V. CONCLUSION

A series of single Fe and multilayer Fe/Cr films have been grown by magnetron sputtering on GaAs substrates prepared using an argon ion milling. Epitaxial growth of the Fe is achieved for temperatures above 100 °C, where a clear fourfold magnetization is found. Epitaxial layers show significant roughness and all layers show a reduction in saturation magnetization. An Fe/Cr multilayer sample showed significant antiferromagnetic coupling, where the Cr
thickness was around the second antiferromagnetic peak. However, despite the promising magnetic properties of the samples, the roughness of the layers most likely precludes the use of this materials system for in multilevel memory applications.

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REFERENCES