Development of flexible $Ni_{80}Fe_{20}$ magnetic nano-thin films

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Flexible magnetic $Ni_{80}Fe_{20}$ thin films with excellent adhesion, mechanical and magnetic properties have been fabricated using magnetron plasma deposition. We demonstrate that flexible $Ni_{80}Fe_{20}$ thin films maintain their non-flexible magnetic properties when the films are over 60 nm thick. However, when their thickness is reduced, the flexible thin films display significant increase in their magnetic coercive field compared to identical films coated on a solid Silicon substrate. For a 15 nm flexible $Ni_{80}Fe_{20}$ film coated onto 110µm Polyvinylidene fluoride polymer substrate, we achieved a remarkable 355% increase in the magnetic coercive field relative to the same film deposited onto a Si substrate. Experimental evidence, backed by micro-magnetic modelling, indicates that the increase in the coercive fields is related to the larger roughness texture of the flexible substrates. This effect essentially transforms soft $Ni_{80}Fe_{20}$ permalloy thin films into medium / hard magnetic films allowing not only mechanical flexibility of the structure, but also fine tuning of their magnetic properties.

Keywords: plasma sputtering, flexible thin films, NiFe, magnetic hysteresis, substrate roughness effects

1. Introduction

The ability to fabricate flexible nano-thin films is of great interest because of the increased demand for flexible technologies, a recent paradigm shift in high-tech and consumer electronics. Just as the transition from digital to quantum, from electronics to spintronics, from micro-electronics to nano-electronics would enable new and emergent technologies, the transitions from solid thin films to flexible thin films are already making significant technological and commercial impact by enabling the emergence of flexible photovoltaics [1], flexible electronics [2], flexible smart textiles [3] and flexible displays [4]. Flexible thin films are typically achieved by depositing a given material onto a flexible substrate. These are mostly done via a chemical vapour deposition (CVD) process where the coating ingredients are mostly organic materials and chemicals. Inorganic materials such as metals, functional metals, semiconductors, oxides and ceramics can be deposited via DC/RF plasma sputtering onto flexible substrates, but it is unclear how the flexible substrate affects their quality and properties relative to the same thin films coated onto traditional solid substrates like glass or Silicon wafers. This is a growing field of research and development and very interesting studies have already been published reporting the deposition of flexible structures for energy harvesting [5], flexile ferroelectric random access memories [6], flexible electroluminescent devices [7], flexible organic ferroelectrics [8], flexible synthetic anti-ferromagnets and nano-wires [9] and flexible solar cells [10].

In this paper we describe the deposition and experimental studies of flexible polycrystalline $Ni_{80}Fe_{20}$ permalloy thin films, together with a comparison study of their magnetic properties in rigid and flexible forms. For simplicity, instead of $Ni_{80}Fe_{20}$ we will use the contracted NiFe notation throughout the paper. This is demonstrated for NiFe thin solid and flexible film samples via magnetic properties investigation. We demonstrate the successful production of flexible magnetic thin films with excellent adhesion. Remarkably, the films maintain their structure, integrity and physical properties at any curvature bending applied to the flexible samples. We also show that the flexible substrates have identical physical properties to the films coated onto solid substrates, except that when the thickness of the film is reduced, the flexible substrates promote the growth of NiFe thin films of increased coercive field. We report experimental evidence that flexible NiFe films display an increase in the coercive field relative to solid NiFe thin films of identical thickness, with similar trends observed for both flexible substrates. Remarkably, the 15 nm NiFe flexible thin film on Polyvinylidene fluoride (PVDF) showed coercive fields of over 3 times larger than the identical film coated on a Si rigid substrate.

2. Experiments

Thin film samples have been deposited using a LabLine plasma sputtering machine from Kurt J. Lesker [11]. The vacuum chamber has been pumped to a base pressure of 5×10^{-7} Torr. The plasma sputtering system is equipped with four indexed substrate holders that can be independently rotated into the deposition position above one of the 5 magnetron Torus 2 sputtering targets. Each substrate position had 3 substrates mounted on it, namely Si, Kapton and PVDF. The Si is the solid substrate while the Kapton and PVDF are flexible substrates. The thickness of the Kapton substrate is 50 microns and PVDF is 110 microns. Moreover, the PVDF is also a functional polymer with piezo-electric properties. Before the deposition process commenced, each substrate was exposed to RF plasma for cleaning / etching for about 2 minutes. Each substrate plasma cleaning process had identical conditions, with RF power fixed at 60W and 10 mTorr Argon process pressure.

The structure of the deposited samples was Substrate/Cr (5 nm)/NiFe (t nm), where the substrate is Si (110 wafer), Kapton 50 microns thick flexible substrate or PVDF 110 microns thick piezo-electric polymer (see fig. 1). The thickness of the NiFe samples was t = 15 nm, 60 nm and 100 nm, respectively.

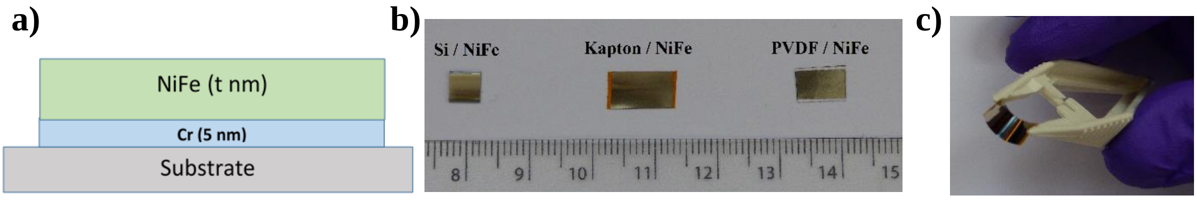

Figure 1. a) Schematic of the layered structure of the samples. Substrate = Si, Kapton and PVDF; b) Pictures of the solid (Si) and flexible (Kapton, PVDF) thin films in stress free state; c) Picture of Kapton / NiFe flexible thin film when flexed.

The 5 nm Cr film is a seed layer introduced to increase the adhesion to the substrate. Both Cr and NiFe layers were deposited using 3 mTorr Argon process pressure, which corresponded to an 11.2 sccm gas flow at the Mass Flow Controller (MFC). The magnetron DC power for Cr deposition was 60 W resulting in a deposition rate of 1.8 Å/sec. Various NiFe layer thicknesses were deposited at constant 100W DC magnetron power corresponding to 3.7 Å/sec deposition rate. The substrate temperature was monitored continuously during the deposition process and kept constant at around 22 – 24 ^{0}C.

Magnetic properties of the NiFe layers were tested using a room temperature Magneto-Optic Kerr Effect (MOKE) system in longitudinal geometry. The MOKE is capable of testing samples under maximum 5000 Oe applied magnetic field, with a field resolution of +/- 2 Oe. Since NiFe is a soft magnetic thin film, fields of maximum 600 Oe were sufficient to saturate the samples in our experiments. The MOKE system uses a HeNe 633 nm continuous wavelength (CW) red laser, which has an intensity stability of +/-5%. We reduced the intensity fluctuation of the laser by using a noise eater electro-optical modulator, to achieve laser stability of +/- 0.5%. The CW laser has been further modulated using a mechanical 10 blades beam chopper at 377 Hz frequency. A large area photo-detector with built in preamplifier has been used to sense the MOKE signal. The photo-detector signal was fed into a Stamford Research SR830 lock-in amplifier that used the reference signal of the mechanical beam chopper to detect the modulated MOKE signal. Using 100 ms time constant, we achieved a signal fluctuation of less than 0.1%, which allowed us to collect magnetic hysteresis loops in single scans without any averaging. Once optically aligned, a single MOKE magnetic hysteresis loop measurement took just 60 seconds.

Average roughness of each substrate has been determined using a Park NX10 Atomic Force Microscope. Z-height scans were performed over a 5 x 5 µm area using non-contact topography scanning mode. Park Systems PPP-NCHR cantilevers with resonances of 281.4 and 276.5 Hz were used. The scan rate was 0.1 to 0.15 Hz and the cantilever amplitude varied between 16.7 and 20.9 nm.

3. Results

Figure 2 shows the thickness effect on the magnetic hysteresis of NiFe thin films with thickness 15 nm, 60 nm and 100 nm coated onto three different substrates. The data indicates that thinner magnetic films display more pronounced changes of the magnetic properties, in particular the magnetic coercive fields. In the case of films that are 100 nm or thicker, the thickness effect vanishes and no changes are detected regardless of the substrate used and flexible / non-flexible structure of the film. Coercive fields of 100 nm NiFe films were 57 Oe for Si substrate, 59 Oe for Kapton and 72 Oe for PVDF. For 15 nm thin films, the coercive field increases by as much as 47% for solid thin films coated onto Si substrate from 57 Oe corresponding to 100 nm thin film to 84 Oe for 15 nm film on Si. The coercive field of 15 nm NiFe films coated onto Kapton substrate increased to 127 Oe from 59 Oe corresponding to 100 nm NiFe film on Kapton. This is an increase by 115% for flexible 15 nm thin films coated onto a Kapton substrate. The most remarkable thickness effect was observed for NiFe films coated onto PVDF substrates, where the coercive field increased from 72 Oe for 100 nm NiFe to 299 Oe for 15 nm NiFe thin films. This represents an increase by 315% for the 15 nm NiFe film relative to 100 nm coated onto PVDF. The data is summarized in figure 3, showing that the effect is consistent for all three different substrates and all three thicknesses.

However, the effect appears not only to scale with the thickness, but it is more enhanced for the flexible thin films, as seen in figure 2.a-c and figure 3. Initially this was linked to the possible interfacial stress induced in flexible films, but a closer inspection revealed that in fact the roughness of the substrates could play a more decisive role. The roughness was determined via surface topography scans using Atomic Force Microscopy imaging of the bare substrates as shown in Figures 2.d-f. Silicon substrate has an average roughness of R_a^{Si} = 1.81 nm, flexible Kapton has R_a^{Kapton} = 3.08 nm, and PVDF R_a^{PVDF} = 8.73 nm. The substrate roughness effect is clearly emphasised in figure 4 where the magnetic coercive field for each NiFe thickness and type of substrate is plotted as a function of the measured substrate roughness. Interestingly, it appears that coercive field scales linearly with the substrate roughness. The thinner the film, the larger the linear slope, indicating that larger roughness promotes the coercive field enhancement. In the case of 100 nm thin films the substrate roughness effect is minimal and no major changes are observed with the linear relationship displaying a very low slope. As the thickness decreases, the roughness effect is more pronounced and the slope of the linear trends increases. The magnetic coercive field of 15 nm NiFe on PVDF is larger by 255% than the coercive field of the same film coated onto Si, and coercive field of 15 nm NiFe on Kapton 51% larger than the same film coated on Si.

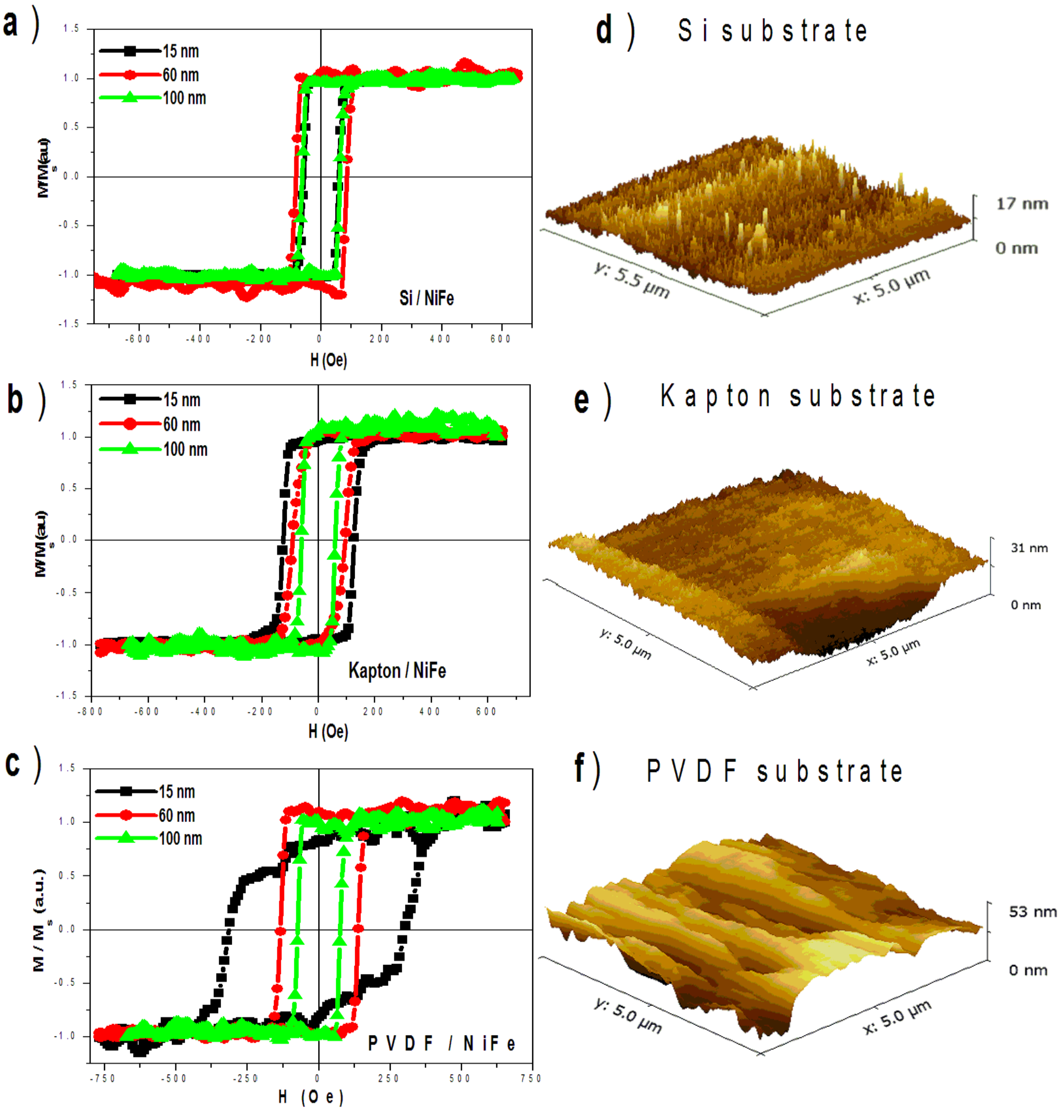

Figure 2. a) – c) Magnetic hysteresis loops for 15nm, 60nm and 100nm NiFe thin films. a) NiFe on Si substrate; b) NiFe on Kapton substrate; c) NiFe on PVDF substrate; d) – f) AFM surface topography. d) Si substrate; e) Kapton substrate; f) PVDF substrate.

In order to further support the conclusions of this study, micromagnetics simulations have been performed to analyse the variation of the magnetic coercivity with the measured surface roughness. An effective field roughness model previously introduced [12] was used to include the roughness profiles obtained from AFM scans into the computations.

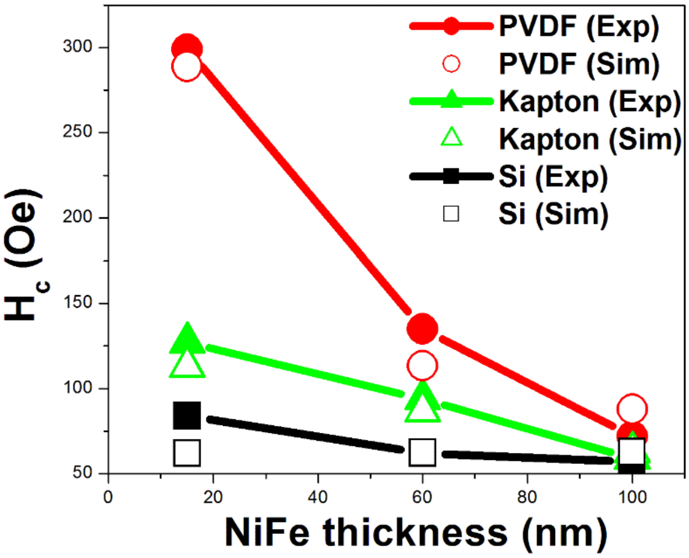

Figure 3. Magnetic coercive field versus thickness of NiFe thin film layer grown onto Si, Kapton and PVDF substrates. The solid symbols are the experimental values, whilst the open symbols are results from micromagnetics simulations.

As previously described [13], the Landau-Lifshitz-Bloch equation together with a thermal activation field was used at room temperature to simulate magnetic hysteresis loops and to extract the coercivity values, shown in Figure 3. Due to the thermal activation field a distribution in the coercivity values is obtained with standard deviation of ~20 Oe. The surface roughness profiles introduce a configurational anisotropy with effective roughness energy density proportional to the roughness depth and approximately inversely proportional to the ferromagnetic layer thickness. The roughness energy density within the simulated ferromagnetic body V is given by:

$$\varepsilon(\mathbf{r}_0) = \frac{\mu_0 M_S^2}{2} \sum_{\mathbf{r}\in V} \left(N_{xx}(\mathbf{r}-\mathbf{r}_0) m_x^2 + N_{yy}(\mathbf{r}-\mathbf{r}_0) m_y^2 + N_{zz}(\mathbf{r}-\mathbf{r}_0) m_z^2 \right) G(\mathbf{r},\mathbf{r}_0) \quad (\mathbf{r}_0 \in V) \quad (1)$$

Here M_S is the saturation magnetization, m_x, m_y, m_z are the magnetization direction cosines, N is the demagnetizing tensor with the diagonal components entering in the equation, and G is a function that depends on the particular roughness profile [12,13]. The actual roughness profiles from the AFM scans, see Figure 2, were used to calculate the roughness energy density. This appears as an energy perturbation on a smooth rectangular structure, from which effective roughness fields are obtained in the LLB equation using the relation $\mu_0 \mathbf{H} = -\partial\varepsilon / \partial\mathbf{M}$. For the

PVDF roughness texture, and to a lesser extent for Kapton due to the reduced roughness depth, the roughness energy density has a hard axis perpendicular to the sample plane, i.e. ε (averaged over the body V) has a maximum along the m_z direction. This introduces an additional energy barrier which must be overcome during the switching process, thus the external magnetic field required to switch the magnetization increases. Note that for a random surface roughness profile the energy roughness density has an easy axis perpendicular to the plane, thus the real roughness profiles obtained from AFM scans are crucial in reproducing the experimentally observed coercivity changes. The computed coercivity values are in very good agreement to the experimental values, as shown in Figure 3.

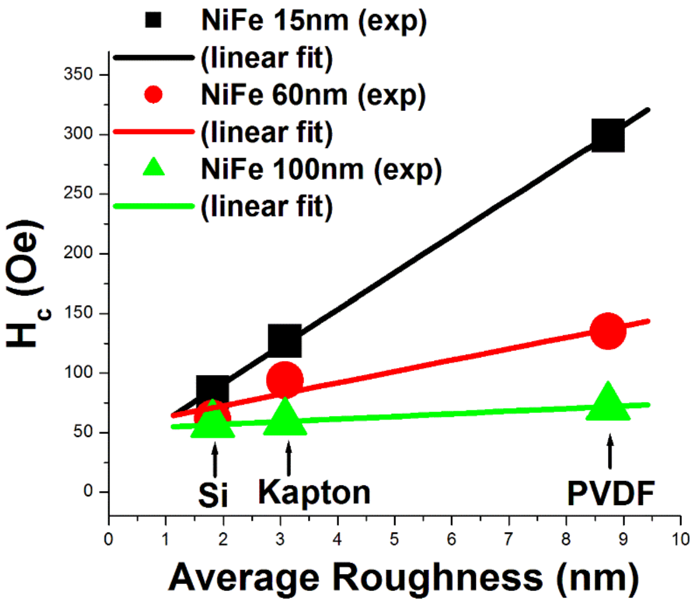

Figure 4. Magnetic coercive field of 15 nm, 60 nm and 100 nm NiFe thin films linear scaling with the average roughness of the substrates (Si, Kapton and PVDF).

Larger coercivity values are obtained for the thinner films, since the roughness energy density takes on the largest values. As the film thickness is increased, since the average roughness energy density is inversely proportional to the film thickness, the differences in coercivity between the different substrates decrease. For the thickest 100 nm films the roughness energy density is negligible, resulting in similar coercivity values for the different samples. These results confirm the roughness depth and texture as the origin of the observed change in the magnetic coercive fields of NiFe. Moreover, it is well known that the grain size of polycrystalline thin films has a dramatic influence on the film properties, including magnetic coercive field [14]. This effect is indeed enhanced when the grain size becomes comparable to the film thickness. However, previous studies on similar samples (not flexible), where the grain size was controlled via the deposition process, indicated that the coercive field increases with the grain size [15]. Our results show the exact opposite, that thinner films (i.e. smaller grain sizes) have larger coercive fields, reinforcing the conclusion that the roughness is the dominant driving force in this case, rather than the grain size.

4. Conclusions

NiFe thin films of various thickness have been deposited via magnetron plasma sputtering onto solid and flexible substrates. The flexible thin films displayed excellent adhesion and mechanical robustness. In addition we determined that magnetic properties are indeed preserved when flexible samples are fabricated, indicating that inorganic flexible thin films could be successfully fabricated via magnetron plasma sputtering. For lower thicknesses typically below 60 nm, the surface roughness effects kicked in resulting in a significant increase in the magnetic coercive field of 15 nm NiFe thin films. A linear relationship between substrate roughness and the value of the magnetic coercive field has been obtained for all three sets of sample thickness. Micro-magnetics simulations confirmed the significant roughness effect on the magnetic coercive fields of thinner films. Our results offer a platform for customization of the properties of magnetic thin films together with the additional features afforded by their mechanical flexibility and we hope this will stimulate further theoretical and experimental investigations into fabrication and studies of functional flexible thin films. While it is impossible to cover all the relevant literature, it is important to mention that other studies of NiFe thin films deposited onto flexible substrates have been recently reported, but unlike our studies, their emphasis was on the improvement of anisotropic magneto-resistance of flexible NiFe thin films [16,17]. It is also interesting to mention that our PVDF / NiFe samples are in fact free-standing, unclamped, bi-layer composite multiferroics consisting of piezo-ferroelectric active substrate / layer and ferromagnetic NiFe layer. These are also very attractive for further studies due to the fact that the samples are flexible multiferroic and the substrate clamping is totally removed making them suitable for magnetism control via electric fields [18].

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