Surface and Bulk Magnetization Behavior of Isolated Ferromagnetic NiFe Nanowires

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Abstract—The surface and bulk magnetization behavior of template released isolated ferromagnetic Ni$_{60}$Fe$_{40}$ nanowires of relatively thick diameters (~200 nm), deposited from a dilute suspension onto pre-patterned insulating chips have been investigated experimentally, using a highly sensitive Magneto-Optical Kerr Effect (MOKE) magnetometry and Magneto-Resistance (MR) measurements, respectively. The MR data were consistent with the theoretical predictions of the anisotropic magneto-resistance (AMR) effect. The MR measurements, in all the angles of investigations, showed large features and a series of nonmonotonic “continuous small features” in the resistance profiles. The extracted switching fields from these features and from MOKE loops were compared with each other and with the switching fields reported in the literature that adopted the same analytical techniques on the similar compositions and dimensions of nanowires. A large difference between MOKE and MR measurements was noticed. The disparate results between MOKE and MR results is attributed to the variance in the micro-magnetic structure of the surface and the bulk of such ferromagnetic nanowires. This result was ascertained using micro-magnetic simulations on an individual: cylindrical and rectangular cross sections NiFe nanowires, with the same diameter/thickness of the experimental wires, using the Object Oriented Micro-magnetic Framework (OOMMF) package where the simulated loops showed different switching events, indicating that such wires have different magnetic states in the reversal process and the micro-magnetic spin structures during switching behavior was complicated. These results further supported the difference between surface and bulk magnetization behavior in these nanowires. This work suggests that a combination of MOKE and MR measurements is required to fully understand the magnetization behavior of such relatively thick isolated cylindrical ferromagnetic nanowires.

Keywords—MOKE magnetometry, MR measurements, OOMMF package, micro-magnetic simulations, ferromagnetic nanowires, surface magnetic properties.

I. INTRODUCTION

The experimental and theoretical researches on the magnetization behavior and reversal process of ferromagnetic nanowires have intensified over the last few decades, due to their promise applications in magnetic storage media, logic circuits, sensors and spintronics technology, as well as; their importance for exploring the fundamental media, logic circuits, sensors and spintronics technology, as decades, due to their promise applications in magnetic storage...
surfaces of these structures, which leads to the scattering of the reflected light into different directions, may have donated to the valuable absence of measurements on individual cylindrical nanowires using this approach. During this effort, few papers have been published deliberating the magnetization behavior of cylindrical ferromagnetic nanowires using this technique [8]-[10].

MR is also a powerful technique implemented by many research groups to acquire information on the magnetic state of a range of ferromagnetic; rectangular nanowires, cylindrical nanowires embedded in their templates and isolated cylindrical nanowires. This technique, however, describes the change in the MR ratio as a function of the angles between the magnetization and the direction of the current flow, where the resistance is maximum and minimum when they are parallel and perpendicular to each other, respectively. This dependence is related to the anisotropic behavior of the electrons via a mechanism, known as; spin-orbit coupling. In this technique, the ferromagnetic nanowire is usually saturated by a high magnetic field along a certain orientation; as a result, the magnetization reversal occurs at a critical field value, due to the occurrence of instability in the magnetization state. Using this approach, one can get information about the bulk magnetization of the magnetic structures. Very limited works have been published on single cylindrical ferromagnetic nanowires using this method, due to the relative small sizes of such nanowires and the difficulties arising from electrically connecting these wires to the electrical contact pads and the external circuitry [12].

Today's computer systems and their large storage media allow use of OOMMF software, to analyze the magnetization behavior of nanowires and nanotubes. This approach is based on numerically solving the Landau–Lifshitz–Gilbert equation, which has the ability to simulate hysteresis loops and to visualize the magnetic state configurations at any point during a field cycle [11]. Therefore, researchers tried to use this software to examine; isolated rectangular cross-sections ferromagnetic nanowires, focusing on the smallest thicknesses that are likely to show single domain states and simpler magnetic behavior [11].

Much less work has focused upon thick cylindrical nanowires that are likely to display more complex domain structures compared to thinner one [11]. Thus, the aim of the work presented here is to investigate the magnetization behavior of template released ferromagnetic NiFe nanowires of relatively thick diameters (~200 nm), using a combination of highly sensitive MOKE magnetometry and MR measurements, to probe the surface and bulk magnetisation behaviour of such wires, respectively. The results of this work were discussed in accordance to the other experimental researches and with the detailed theoretical micro-magnetic simulations, using the OOMMF-package. Indeed, as it will be seen in the results and discussion sections, the contrast between magneto-optical properties and electrical transport measurements suggest that a combination of techniques is required to understand deeply the magnetization behavior of relatively thick nanowires. In accordance with our knowledge, this is the first article that combines both MOKE and MR measurements on the same isolated template released ferromagnetic nanowires deposited from a dilute suspension onto pre-patterned oxidized silicon substrates.

II. EXPERIMENTAL DETAILS

In this work, two dimensional arrays of ferromagnetic NiFe nanowires with a nominal composition of Ni$_{60}$Fe$_{40}$ were prepared, using the DC electro-deposition technique. WDS analysis was used to confirm the composition of these wires. The deposition process was carried out in a conventional three electrodes electrochemical cell (Metro-Ohm). The nanowires were synthesized within the pores of commercially available ~60 µm thick alumina template (Anodisc Whatman, Imm.) with a nominal pore diameter of ~200 nm after coating one side with ~100 nm gold layer, to serve as a working electrode. The electrochemical cell contains an electrolyte solution of ~1.3 M of NiSO$_4$ and ~0.151 M of FeSO$_4$ along with ~0.8 M of H$_3$BO$_3$. The pH of the solution was maintained at approximately 3.7. The electro-deposition was carried out under a constant voltage of approximately ~0.85 V, following a linear voltmetry sweep results. The length of the wires was fixed to ~20 µm and controlled via the electro-deposition time and current density.

The template was dissolved and entirely removed using ~2 M of NaOH solutions for a time of ~48 hours at room temperature, then the nanowires were washed repeatedly very well with distilled water and isopropanol alcohol (IPA). The beaker was placed in an ultrasonic bath for approximately 10 minutes, to release the nanowires from their base layer. This process will create suspended nanowires in the dilute suspension with shorter lengths. The suspended nanowires were then transferred onto gold pre-marked silicon substrates, by applying ~5 µl drops of the suspension, and then letting it dry at room temperature. To overcome the random orientation of the deposited wires, electromagnetic association was used during this process.

For MOKE and MR measurements, gold patterns (markers) of several microns in size and electrical contact gold pads was pre-fabricated onto clean SiO$_2$/Si chips, prior to the dispersion of the nanowires on them. These patterns were performed using a conventional electron beam lithography and lift-off techniques. Identifying isolated wires was then performed using high resolution scanning electron microscope. These images were used as a guide during measurements and electrical contact connections. After the dispersion of nanowires, gold electrodes were created, to electrically connect the selected wires with the contact pads, using electron beam lithography and lift-off techniques, as well. All these patterns (markers, electrical contacts and electrodes) were fabricated, as investigated in the following discussion.

Two layers of poly-methyl-methacrylate (PMMA) resist with a thickness of ~70 nm each were deposited on the top of a clean oxidized silicon substrate, using the spin coating method. The resist is chemically sensitive to the electron beam radiation and will serve to define the required patterns. In this
work, a high-resolution field emission scanning electron microscopy column on an FEI-Helios Nanolab dual focused ion (FIB)/scanning electron microscope (SEM) system with an electron beam energy of ~30 keV and a current of ~21 pA was used to expose the required patterns in the PMMA resist according to the images obtained in the previous step of SEM. During this work, care has to be taken not to expose the resist in the desired areas during all imaging and lithography processes. All the chips were then developed using 3:1 IPA: MIBK (Methyl-Isobutyl-Ketone) to dissolve the exposed area and leaving behind the unexposed areas in place. Thermal evaporation was then used, to deposit a combination of Cr and Au with thicknesses of ~3 nm and ~100 nm, respectively, for contact patterns, whilst ~3 nm and ~35 nm, respectively for the electrodes. The deposition of Cr is to enhance adhesion of the gold layer to the substrate. Finally, the remaining unexposed PMMA resist was removed using acetone and IPA solutions. At this stage, only the evaporated gold patterns in the regions that exposed to electron beam will remain. Finally, the wire bonding machine was used to electrically connect the contact pads with the chip carrier, using gold wires.

The hysteresis loops of an individual and a bundle of closely packed three NiFe nanowires were obtained using a highly sensitive MOKE magnetometry in the longitudinal configuration at room temperature. The MOKE setup is well suited for measuring the Kerr signal as a function of applied magnetic field, which is directly proportional to the magnetization of the sample. The laser light used in this magnetometry has an elliptical spot size of approximately 5-6 µm, and a wavelength of ~658 nm. An electromagnet with a maximum field of ±450 Oe was used with a lock-in amplifier to switch the magnetic hysteresis of the samples at a frequency of ~21 Hz.

The magneto-transport behavior of the same bundle of closely packed three NiFe nanowires was investigated using MR setup. The sample holder in this setup was cantilevered to measure the resistance, by applying the magnetic field in a plane parallel (longitudinal) and perpendicular (transverse) to the current flow. The current direction, of approximately 0.1 mA. This low current was chosen to minimize the thermal heating of nanowires, and not to damage them. All the measurements employed a magnetic field that was gradually increased or decreased at a steady rate of ~0.5 Oe/s. This was repeated for two full sweeps, in which the magnetic field was applied between +1 kOe and -1 kOe.

OOMMF package was used here to study the magnetic properties of an individual: cylindrical and rectangular Ni$_8$Fe$_{19}$ nanowires of ~200 nm diameter/thickness (width) with a length of 1 µm. This length was selected with consideration to the available computer memory space and the computation time. The simulated structures are relatively short, but the aspect ratio of these wires exceeds 5 and, therefore, there is no detrimental properties expected in terms of self-demagnetizing effect. Within this package, NiFe wires were created by defining a micro-magnetic input file (MIF). This file contains all the information required to solve the problem. Calculations were then, performed by dividing these nanowires into 3D arrays of cubic cells, with dimension of 5×5×5 nm$^3$. This size was chosen to allow the exchange interaction to be correctly modeled and to rationally approximate the shape. The magneto-crystalline anisotropy, $K_u$, was chosen to be zero, because Ni$_8$Fe$_{19}$ has zero magneto-crystalline anisotropy. The gyromagnetic ratio, $\gamma$, was 17.59 MHz/Oe. The value of saturation magnetization, $M_s$, was 8.6×10$^5$ A/m and the value of the exchange stiffness parameter, $A$, was used to be 0.5, to speed up the calculations and decrease the simulation time. Using smaller values of the Gilbert damping parameter is, however preferable, but the simulation will take longer time and a large amount of memory. The shape anisotropy was calculated using the demagnetizing field of the system through the magneto-static energy calculations. In complete simulations, the magnetic field was decreased from saturation, in steps of 5 to 10 Oe through zero to the opposite sign. All micro-magnetic simulations were conducted at 0 K, because OOMMF is limited to zero temperature calculations.

III. RESULTS

Typical examples of normalized hysteresis loops obtained from MOKE measurements of (a) an individual and (b) a bundle of template released NiFe nanowires are shown in Fig. 1. These measurements were utilized by applying the magnetic field nominally parallel to the nanowires long axis. The associated scanning electron micrographs showing the interrogated areas and the histogram of switching field distributions obtained from repeating measurements at different locations on these wires are presented in the insets of Fig. 1.

Details on the switching field distributions and the effect of increasing the number of nanowires in the closely packed bundle, on the switching field behavior can be found in other works [7].

Clearly, the loops show high squareness ratio ~1, indicating a characteristic magnetic easy axis along the wire long axis. This result is quite different from the results reported in the literature for 2D arrays of nanowires (see, for example [1]-[5]), which was measured using other characterization techniques. This was attributed to the strong magneto-static interaction between the nearest neighbor nanowires. This result, may also reflect the surface magnetic state compared to the nanowires bulk, since the magnitude of the detected MOKE signal is limited by the depth that laser light penetrates into it, owing to the absorption of laser light in the medium [6].

Fig. 2 shows typical results obtained from MR measurements of the same bundle of closely packed nanowires discussed above, by applying the magnetic field at longitudinal and transverse directions to the current flow. SEM micrograph showing this bundle of wires after electrically connected to the pad contact patterns is presented as inset in Fig. 2 (a). The MR measurements of different other angles are shown in other research [12]. Although, the applied magnetic field was between +1 kOe and -1 kOe, it is plotted in
Fig. 2, between +500 Oe and -500 Oe, because the resistance in this field region was saturated. These measurements were conducted by sweeping the magnetic field in both directions (red and black curves in electronic version). The arrows shown in the figure indicate the direction of the field change. From these profiles, it is interesting to note the good quality of signal to noise ratio. For both: longitudinal (Fig. 2 (a)) and transverse (Fig. 2 (b)) measurements, three different sections were identified. At high magnetic field strengths, between 250 and 500 Oe, the resistance displays saturation behavior, indicating that the magnetic moments within the nanowires are oriented parallel (with high resistance) or perpendicular (with low resistance) to the field applied. At moderate magnetic field strengths, between 100 and 250 Oe, the measurements show a series of nonmonotonic "continuous small features" in the resistance. These features are magnified and plotted as insets in Fig. 2, and they were noticed in all angles of measurements. Understanding these features is very important, as it will be seen in the discussion section. When the magnetic field was less than ±50 Oe, the resistance falls rapidly followed by positive jumps in the longitudinal measurements and jumped up followed by a drop in the transverse measurements.

The small and large changes in the resistance are symmetric about the zero field when reversing the field direction, using different colors of the curves (red in electronic version).

The resistance at magnetic saturation as a function of their long axis angle with respect to the applied field is plotted in Fig. 3. Clearly, a reduction in the saturated resistance is seen with increasing the nanowires’ long axis angle with respect to the field applied. Comparing the resistance at high magnetic fields (i.e. at saturation), for all angles investigated here, with the theoretical calculations, using the equations of AMR available in the literature (see, for example [8]-[10]), a good agreement was obtained, as demonstrated by the continuous line shown in Fig. 3. This is the high and low resistance relative orientation of the current and magnetization in AMR. Further information on the electro and magneto transport properties in such wires can be found elsewhere [12].

Fig. 4 shows examples of normalized hysteresis loops obtained from simulations of an individual (a) cylindrical and (b) rectangular cross sections NiFe nanowires, when the magnetic field was applied nominally parallel to the
nanowires’ long axis. These wires have the same diameter or thickness of the nanowires, experimentally investigated above.

The simulations were carried out by applying a large magnetic field along the long axis of the nanowires directed at a slight tilt (~1°) with respect to the nanowires’ long axis. This tilting has been used in the OOMMF software to break the symmetry of the magnetic structure and hence, to avoid numerical artifacts in the simulations, and yield stable meaningful results. Clearly, the loop shapes are quite complicated showing different switching events, indicating that there are at least two magnetic states in the reversal of such nanowire.

In order to understand the detailed magnetization behavior in such isolated ferromagnetic NiFe nanowires, the micromagnetic moment distributions are presented in Figs. 5 & 6, for cylindrical and planar nanowires, respectively, during magnetic remanence and both switching events. In the case of cylindrical wires, the micro-magnetic spin structures along the length of the nanowires are presented as an axial slice through the center of the nanowire. For the planar nanowires, the micromagnetic structures are shown along the length of the nanowires at the top, and as an axial slice through the center of the nanowire. Also, for both geometries, cross-sectional micro-magnetic structures are shown for each end of the nanowires, and for a slice through the center of the nanowire.

IV. DISCUSSION

The average switching field obtained from repeating MOKE measurements on an individual NiFe nanowire was found to be ~165 Oe (see, Fig. 1 (a) and its inset) whilst, the average switching field obtained from repeating measurements on the bundle of closely packed nanowires was found to be ~134 Oe (see, Fig. 1 (b) and its inset). Lupu et al. [6] used only MOKE technique to probe the magnetic properties of a range of isolated Ni80Fe20 nanowires with diameters ranged from ~35 to ~300 nm.

Fig. 3 Resistance at magnetic saturation for the isolated bundle of closely packed three NiFe nanowires as a function of their angle with respect to the field applied. The line is the theoretical curve based on anisotropy resistance, see text for details [12]

Fig. 4 Normalized hysteresis loops obtained from simulations of an individual: (a) Cylindrical and (b) Planar Ni81Fe19 nanowires with 200 nm diameter/thickness (width) using OOMMF code, when the field was nominally applied parallel to the nanowires long axis [11]

Fig. 5 Snapshots of magnetic moment distributions along the center of the nanowires long axis and normal cross-section of the left end, middle, and right end of the cylindrical Ni81Fe19 nanowire with 200 nm diameter at different field strengths; (a) Magnetic remanence, and during (b-c) first & second switching events. The color variation represents the x-z angle of the spin [11]
The estimated switching field of ~300 nm diameter from their data was found to be around 100 Oe. As reported elsewhere, increasing the Fe content in the NiFe alloys, decreases the switching field [3], whilst increasing the diameter, decreases the switching field. Thus, comparing this value with the value reported here, a reasonable agreement is clearly obtained, since the diameter and the composition of nanowires investigated here were different from Lupu’s nanowires [6].

For the MR measurements of the bundle of closely packed three wires, the switching fields obtained from the fields at which the large features occur in both longitudinal and transverse profiles were found to be ~16 Oe (see, Figs. 2(a) & (b)). Rheem et al. [9], [10] studied the magnetic behavior of an individual: Ni_{85}Fe_{15} and Ni_{80}Co_{20} nanowires, with 200 nm diameters and more than 5 µm lengths, using only MR measurements. The estimated switching fields from their data were found to be ~15 Oe and ~28 Oe, respectively. Comparing these values with the MR switching values presented here, again a good agreement is obtained. Comparing the field values obtained from the large features in the MR measurements (~16 Oe) with the switching field obtained from MOKE measurements (~165 Oe for an individual and ~134 Oe for a bundle of wires), a large difference is clearly seen, whilst, the fields at which the small features occur in the MR profiles were found to be around 100-240 Oe (see, Fig. 2 and its insets). Comparing these values with the switching field obtained from MOKE measurements, a comparable agreement is detected. This agreement suggests that the small features may correspond to the magnetization reversal at the surface of such nanowires, since the magnitude of the detected MOKE signal is limited by the depth that laser light penetrates into the nanowire.

Now, in order to get a full understanding of the magnetization behavior of these relatively thick nanowires, the switching fields obtained from micro-magnetic simulation need to be compared with the switching fields of their counterpart nanowires obtained experimentally from; MOKE and MR measurements.

As mentioned earlier in the results section, two different switching behaviors were noticed in the loops obtained from the simulations of cylindrical and rectangular cross-section nanowires (see Figs. 4(a) & (b)). These two switching events indicate that there are at least two magnetization features in the reversal of such relatively thick nanowires. The extracted switching fields from these loops are approximately 31 Oe and 263 Oe. Comparing these values, with the experimental results, reasonable agreement is observed, although, the simulation values are higher than the experimental results. It has been reported elsewhere [1], [3] that the switching fields of arrays of Ni, Fe and Co nanowires, decreases with increasing the temperature; from liquid helium to room temperature. Thus, the difference between the experimental and simulation switching fields may be reasonably attributed to the temperature difference between the room temperature experimental measurements and zero temperature micro-magnetic simulations. The nanowire compositional variations may have also influence on the switching field difference. This is due to the difference in the saturation magnetization and hence, the demagnetizing field. Moreover, the simulated nanowires are free from impurities, defects and structural variations that were noted in the real nanowires. These imperfections can distort the spin configuration structure and give different values of the switching fields. Thus, the results obtained here, from both; MOKE and MR measurements, may reflect the surface magnetization behavior compared to the bulk magnetization measurements, since MOKE setup is a sensitive technique to probe only the surface magnetization to a depth of the order of the skin-depth of the nanowires [6]. In contrast, MR measurements are sensitive to probe the whole bulk of the nanowires, but only when there are changes in the angular orientation of the magnetic moments.

It has been reported elsewhere that the surface magnetization of plane shaped iron single crystal thin film and amorphous micro-wires behaves differently from the bulk magnetization [13]. Therefore, the two different behaviors...
obtained here strongly suggest that the surface magnetization behaves differently from the bulk magnetization, of such relatively thick nanowires. To confirm this result, the magnetic moment distributions during magnetic remanence and first & second switching events are presented in Figs. 5 and 6 for both the cylindrical and planar cross sections nanowires, respectively. As expected, the magnetic moments at saturation magnetization were aligned parallel to the applied field with the existence of vortex states at both ends of the wire (snapshots are not shown here). At magnetic remanence, the nanowire shows multi-domain structures, and the magnetic moments are in a vortex state along the length of the wire, as it is clear in Figs. 5 (a) and 6 (a). This behavior indicates that, the shape anisotropy is less significant with such relatively thick nanowires. For the rectangular cross section nanowires, the magnetization structure along the wire axis is also quite different from the surface to the center (see, Fig. 6 (a)), suggesting that the surface sensitive magnetization measurements such as; MOKE magnetometry, would not fully capture the magnetization reversal behavior of such relatively thick nanowires. This finding explains the disparate observations for MR and MOKE for the other researchers and the combined MR and MOKE study presented here. During the first (Figs. 5 (b) and 6 (b)) and second (Figs. 5 (c) and 6 (c)) switching events, very complicated domain structures are clearly noticed, which further support the difference in the magnetic moment behavior between surface and bulk. After the second switching event, the magnetic moments change their orientation to the opposite direction and forming saturation magnetization with the existence of vortex states at both ends of the wires (snapshots are shown in [11]).

V. CONCLUSIONS

The magnetization behavior of template released isolated ferromagnetic NiFe nanowires with diameters of ~200 nm was investigated experimentally, using a combination of highly sensitive MOKE magnetometry and MR measurements. The results were then, compared with each other and with the theoretical micro-magnetic simulations using the OOMMF package.

The MR measurements showed large and continuous small features in the MR profiles. The extracted switching fields from these features were compared with the switching field obtained from MOKE measurements. The field values obtained from the small features were found to be in a reasonable agreement with the MOKE switching fields, whilst the fields extracted from the large features were found to disagree with MOKE switching fields. This work was combining both MOKE and MR measurements on the same ferromagnetic nanowires and their values were interpreted and compared with the values presented in other researches that adopted the same investigative techniques on similar composition and dimension of ferromagnetic nanowires. Agreements were obtained from these comparisons suggesting that the magnetic state at the surface of ferromagnetic nanowires behaves differently from their bulk.

The switching fields obtained from micro-magnetic loops of similar nanowires were compared with their experimental counterpart values. There was a reasonable agreement, which further supports the idea that the surface magnetic moments within relatively thick nanowires reverse differently from their bulk. The micro-magnetic moment distributions at different field strengths showed complicated domain structures.

Both micro-magnetic simulations and AMR measurements are sensitive to the orientation of the whole magnetic moments within the bulk of the wires, whilst the MOKE technique is sensitive to the surface magnetization, the micro-magnetic simulations may support that the magnetization at the surface of relatively thick nanowires behaves differently from their bulk.

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