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Magnetic properties of planar nanowire arrays of Co fabricated on oxidized step-bunched silicon templates

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Abstract
Planar nanowire (NW) arrays of Co grown on oxidized step-bunched Si(111) templates exhibit room temperature ferromagnetic behaviour for wire widths down to 25 nm. Temperature and thickness dependent magnetization studies on these polycrystalline NW arrays show that the magnetic anisotropy of the NW array is dominated by shape anisotropy, which keeps the magnetization in-plane with easy axis parallel to the wires. This shape related uniaxial anisotropy is preserved even at low temperatures (10 K). Thickness dependent studies reveal that the magnetization reversal is governed by the curling mode reversal for thick wires whereas thinner wires exhibit a more complex behaviour which is related to thermal effects and size distribution of the crystal grains that constitute the NWs.

Online supplementary data available from stacks.iop.org/Nano/23/235702/mmedia

(Some figures may appear in colour only in the online journal)

1. Introduction
Surface supported one-dimensional nanostructures such as nanowires (NWs), nanotubes and nanoribbons are currently the subject of intense interest due to their potential applications in spin-electronic devices and interesting physics perspectives [1–3]. Formation of such structures could be either process-directed or self-assembly based. The latter approach offers a greater throughput and prospects of synthesizing structures well below the limits of current lithography capabilities [4, 5]. Magnetic nanostructures of varying lateral width ranging from single atom to several hundred nm are reported on a variety of templates [4–17]. Reasonable control over the size distribution of magnetic NWs has been demonstrated in carefully prepared porous anodized alumina templates, where NW growth occurs perpendicular to the template surface [6–10]. Though there are several reports on growth and magnetic property investigations on planar NW arrays prepared using self-assembly methods such as step flow growth and step decoration [11–17], observation of ferromagnetism at room temperature (RT) is scarce [18]. This is mainly related to the effect of dimensionality and small thickness of the structures leading to dominance of superparamagnetism (SPM) in these planar structures [11–17]. These self-assembly based approaches [5, 9–13, 17] only work for certain selective combinations of NW and substrate materials which restricts their scope for future applications. In order to overcome SPM in self-assembled magnetic nanostructures one needs to enhance the energy barrier opposing spontaneous magnetization flipping, which scales with both the volume and edge atom anisotropy contributions [11, 14, 19]. Keeping this in mind, alternative approaches such as reactive deposition epitaxy [20, 21] and shallow angle deposition [22–24] methods have been employed to grow...
thick planar nanostructures which exhibit ferromagnetism at room temperatures.

Understanding magnetization reversal processes in these laterally engineered NW structures is of utmost importance considering their application potential and strong dependence of the reversal processes on the geometry (width, length, thickness, inter-wire separation etc.), associated length scales and material characteristics [25]. Only a few studies have focused on magnetization reversal in laterally engineered structures defined lithographically [26, 27]. To elucidate on the magnetization reversal mechanism in NW arrays with planar geometries, we report here a detailed study of magnetic properties of planar NW arrays consisting of Co-NWs with varying thickness deposited on oxidized vicinal Si(111) templates. We show that the Co-NW arrays are ferromagnetic at room temperature for wire width down to 25 nm with magnetic easy axis along the wire length. The magnetization reversal process in these planar NW arrays is found to be thickness dependent.

2. Experimental details

Planar NW arrays of Co were fabricated on oxidized step-bunched vicinal Si templates using a shallow angle deposition technique named ATLAS (atomic terrace low angle shadowing) [23, 24]. The growth was carried out at RT by depositing Co (0.02 Å s\(^{-1}\)) onto the oxidized templates at a small angle (3°) using a multi-pocket e-beam evaporator (Telemark, USA) in a UHV chamber with a base pressure of better than 5 × 10\(^{-10}\) Torr. The deposition flux was directed towards the ascending step direction. A schematic diagram illustrating the deposition geometry is shown in figure 1(a). Details of the ATLAS method are given elsewhere [23, 24]. The templates used in the present study were highly regular with 110 nm periodicity (sum of step-bunch width and terrace width) step-bunched surfaces of n-type doped vicinal Si(111) (miscut of 4° along the \((11\bar{2})\) and resistivity 1–10 Ω cm). Step-bunched surfaces were prepared by performing a dc-current annealing [28] under ultrahigh vacuum (UHV) conditions. These templates were oxidized using a standard thermal oxidation procedure carried out at 830 °C for a duration of 15 h producing a 110 nm thick amorphous surface oxide layer.

We grew two sets of NW array samples; for topographical investigations we prepared Co-NW arrays without cap layer, whereas for magnetization and TEM studies the Co-NW arrays were capped with a 5 nm MgO layer. The MgO cap layer of 5 nm thickness covering the whole surface was deposited in situ under normal deposition conditions. The templates used for preparing the two sets of NW arrays were produced under identical annealing conditions. The average step–terrace periodicity obtained on these templates was reproducible to a large extent (up to 80%), as determined from the statistics obtained from large area atomic force microscopy images taken at several locations on the samples. The morphology of the templates and NW arrays (without MgO capping layer) was characterized using an atomic force microscope (AFM, Solver Pro, NT-MDT) operated in semi-contact mode and a scanning electron microscope.
Figure 2. Cross-sectional TEM image of a 5 nm thick Co-NW array on a 110 nm average periodicity oxidized Si template. The inset shows a magnified view of the cross-sectional image depicting Co-regions that constitute the NWs. (b) HRTEM image showing the polycrystalline nature of the Co-NWs. (c) HRTEM image of a Co-NW crystallite with hcp-Co structure viewed along the [121] zone axis and (d) its corresponding fast Fourier transform.

(SEM, Ziess Ultra). The crystal structure of the NWs and its interfaces with substrate and cap layer was determined through a high-resolution transmission electron microscope (HRTEM, FEI Tecnai F30) operated at 300 kV. Sample cross-sections for HRTEM observation were prepared using a focused ion beam (FIB) on a Helios Nanolab microscope. Sample preparation for HRTEM required deposition of additional Au and Pt layers, which were deposited in the FIB chamber using an e-beam evaporator. The magnetic properties of the NW arrays were examined using a vibrating sample magnetometer (quantum design physical property measurements system) with a sensitivity of $5 \times 10^{-7}$ emu. The diamagnetic contribution from the substrate was removed from the measured data by subtracting a $M-H$ loop of the bare substrate.

3. Results and discussions

An AFM phase image of a step-bunched vicinal Si(111) template showing a regular array of step-bunches and terraces with an average periodicity of 110 nm (terrace width 85 nm and step-bunch width of 25 nm) is shown in figure 1(b). A representative SEM image of an uncapped Co-NW array with wire thickness of 3 nm is shown in figure 1(c). The deposition of NW arrays is realized by driving the Co flux towards the ascending step direction. It leads to the formation of nano-stripes covering the whole width of the step-bunched facet. One notices that the periodic arrays of Co are quite regular with the majority of the wires remaining straight up to 1 µm in length giving a large aspect (length/width) ratio. The NW thickness as determined from the height profiles of the AFM images taken from the same samples was 3.0 nm. It is possible to tune the wire width by varying the deposition angle or the template periodicity as reported in a previous communication on planar NW arrays of Fe [24]. One noticeable feature is the island-type morphology of the NWs, more clearly visible in the inset of figure 1(c). In the initial stages of growth, discontinuous chains of aligned Co islands form on the terraces. With increasing thickness, the density of the islands increases, eventually leading to the formation of a NW of coalesced islands. Figure 2 shows a cross-sectional TEM image of a Co-NW array grown with the deposition flux directed towards the descending step direction (NW thickness 5 nm, MgO cap layer 5 nm, template periodicity 110 nm). The width of the NWs varies between 50 and 70 nm in this particular sample, which is related to the statistical fluctuations in the periodicity of the self-assembled template. The inset of figure 2(a) shows a magnified view of the cross-section illustrating the presence of Co-regions that constitute the Co-NWs. The majority of the Co-NWs appear to be polycrystalline (see figure 2(b)), as expected for NWs formed of coalesced islands. In some cases the structure of single crystallites can be determined, and it is found to be mostly hcp-Co. An HRTEM image of a hcp-Co crystallite viewed along the [121] zone axis and its corresponding fast Fourier transform (FFT) are shown in figures 2(c) and (d). In a few areas we also observed fcc-Co and fcc-CoO structures, where oxidation is likely due to the proximity of the MgO cap layer. Further evidence of the presence of fcc-Co and fcc-CoO phases in the sample and its determination using TEM analysis is provided in the supplementary information (please see supporting information and figures S1 and S2 available at stacks.iop.org/Nano/23/235702/mmedia).

Magnetization studies on Co-NW array samples of 25 nm wire width and having different wire thickness of Co were performed in the 300–10 K temperature range. We refer to them as sample 1, 2, 3 and 4 hereafter and they correspond to 3, 4.5, 6.1 and 9 nm thickness, respectively. All of these samples were deposited on 110 nm periodicity oxidized templates and capped with a 5 nm MgO layer. Figure 3 shows the magnetization hysteresis ($M-H$) loops of all the Co-NW arrays measured at 300 K with an in-plane applied
Figure 3. Magnetization hysteresis loops measured at 300 K with an in-plane magnetic field directed either along \((H_\parallel)\) or across \((H_\perp)\) the length of the NWs for (a) sample 1, (b) sample 2, (c) sample 3 and (d) sample 4.

Table 1. Thickness \((t)\) of NWs, coercivity \((H_C)\), remnant magnetization \((M_R)\), switching field \((H_{sw})\) and width of switching field \((\delta H_{sw})\) measured at 300 K for all the samples with magnetic field applied in-plane along \((H_\parallel)\) and across \((H_\perp)\) of the NWs.

<table>
<thead>
<tr>
<th>Sample</th>
<th>(t) (nm)</th>
<th>(H_C) (Oe)</th>
<th>(M_R) (%)</th>
<th>(H_{sw}) (Oe)</th>
<th>(\delta H_{sw}) (Oe)</th>
<th>(H_C) (Oe)</th>
<th>(M_R) (%)</th>
<th>(H_{sw}) (Oe)</th>
<th>(\delta H_{sw}) (Oe)</th>
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<tr>
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<td>21</td>
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<td>400</td>
<td>90</td>
<td>11.5</td>
<td>85</td>
<td>640</td>
</tr>
<tr>
<td>2</td>
<td>4.5</td>
<td>450</td>
<td>54</td>
<td>450</td>
<td>600</td>
<td>110</td>
<td>18</td>
<td>105</td>
<td>1300</td>
</tr>
<tr>
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<td>6.1</td>
<td>920</td>
<td>82</td>
<td>910</td>
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<td>220</td>
<td>15</td>
<td>220</td>
<td>1400</td>
</tr>
<tr>
<td>4</td>
<td>9</td>
<td>530</td>
<td>53</td>
<td>525</td>
<td>590</td>
<td>260</td>
<td>22</td>
<td>260</td>
<td>1620</td>
</tr>
</tbody>
</table>

field directed either along \((H_\parallel)\) or across \((H_\perp)\) the wires (step-edges). From the magnetization data we find that the value of coercivity, \(H_C\), is thickness dependent. Values of \(H_C\) \((H_C\perp)\) are 110 (90), 450 (110), 920 (220) and 530 (260) Oe for samples 1, 2, 3 and 4 respectively. Values of various parameters determined from the \(M-H\) loops of all the samples are summarized in table 1. We observed that the \(H_C\) is strongly dependent on thickness (figure 4) of the NWs. In the thickness range \(3 \text{ nm} \leq t \leq 6 \text{ nm}\), there is an increase in the \(H_C\) as the thickness is increased. However, for \(t \geq 6 \text{ nm}\), we observe that the \(H_C\) decreases. As expected for wires with large shape anisotropy, the \(M-H\) loops are square for the field applied parallel to the wires \((H_\parallel)\), whereas for \(H_\perp\) the loops are sheared. Another noticeable feature is that the \(H_C\) is strongly enhanced for all the samples as compared to the measured \(H_C\) \((\sim 10 \text{ Oe})\) of a continuous Co-film grown at normal incidence on a flat oxidized Si substrate. All the samples exhibit an easier approach towards magnetic saturation for \(H_\parallel\) compared to \(H_\perp\), indicating that the magnetic easy axis is along the length of the wires, which suggests that the effective magnetic anisotropy \((K_{eff})\) is dominated by the shape anisotropy owing to the large aspect ratio of the wires. We further consider the effect of other contributions to \(K_{eff}\). The contribution from magnetocrystalline anisotropy is expected to be quite small owing to the polycrystalline nature of the wires. For Co the shape anisotropy energy density, \(K_s = \pi\). \(M_s^2 = 6.0 \times 10^6 \text{ erg cm}^{-3}\), is about an order of magnitude greater than the magnetocrystalline contribution for fcc Co \((K_1 = 6.1 \times 10^5 \text{ erg cm}^{-3}\)) and comparable to that for hcp Co \((K_1 = 5 \times 10^6 \text{ erg cm}^{-3}\)). In order to understand the observed differences in \(H_C\) on the applied field direction and thickness of the wires, we need to consider the influence of long range dipolar interactions on the magnetic anisotropy. Magnetostatic interactions favour an antiparallel distribution of magnetization in the neighbouring NWs and depend on the direction of field, length \((L)\) to width \((d)\) ratio and inter-wire separation \(D\) [29, 30]. For the case of large aspect ratio NWs \((d/L \ll 1)\) which are initially magnetized in the same direction, the increase of magnetostatic interactions results in the magnetization reversal of some NWs \((N)\). Assuming that the reversal of an individual NW produces a decrease of magnetostatic energy \(E_v\), it equals the magnetic anisotropy
where $K_1$ is the NWs. The thickness dependence of $H_C$ for both in-plane applied field directions. Figure 5 illustrates the temperature dependence of $H_C$ for all the samples measured as a function of $H_{||}$ and $H_{\perp}$. It is clear that $H_C$ increases with decreasing temperature for all samples. The increase in $H_C$ is more pronounced for the thinner samples, suggesting that the role of thermal fluctuations is important in determining the magnetization behaviour of these NW arrays. For sample 3, $H_C$ increases from 920 to 1575 Oe when cooled from 300 to 10 K, whereas for sample 1 its magnitude changes from 110 to 950 Oe. Similar changes in magnitude of $H_C$ were noticed for the case of $H_{\perp}$ when cooled from 300 to 10 K. Only a marginal increase ($\sim$5–12%) in the magnitude of the saturation magnetization was observed with a decrease in temperature from 300 to 10 K for all the samples, suggesting that the Curie temperature of the NWs is well-above 300 K. The uniaxial anisotropy related to the shape anisotropy of the wires is preserved at low temperatures (down to 10 K).

The results above suggest that the changes of $H_C$ with temperature for the Co-NW arrays is therefore only minimally related to the changes in $M_s$. Now we consider the influence of thermal activation on the magnetization reversal process in these nanostructures. Taking into account the thermal effects, the field dependence of the energy barrier within the Neel and Brown formalism can be written as:

$$E(H) = E_0(1 - H/H_0)^\alpha$$  \hspace{1cm} (3)$$

where $E_0$ is the energy barrier at zero field and $H_0$ is the switching field at zero temperature. The value of the exponent $\alpha$ depends on the size and distribution of the anisotropy axes relative to the applied field. For the curling mode and coherent rotation reversal mode its values are 3/2 and 2 respectively. Equation (3) does not deal with the temperature dependency of $E(H)$. If we take into consideration the thermal effects and the case where magnetic anisotropy is dominated by the shape anisotropy contribution, variation of $H_C$ with temperature can be modelled as [4, 33]

$$H_C(T) = H_C(0) \frac{M_S(T)}{M_S(0)} \left[1 - \left(\frac{25k_BT M_S}{E_0 M_S^2(T)}\right)^{\frac{1}{2}}\right]$$  \hspace{1cm} (4)$$

where $H_C(0)$ is the coercivity at 0 K, $E_0$ is the energy barrier for reversal related to the shape anisotropy, $M_s(T)$

Figure 4. Thickness dependence of $H_C$ of the Co-NW arrays measured at 300 K with an in-plane field applied along the length of the NWs.
Figure 5. Variation of the $H_C$ as a function of temperature measured with the in-plane field applied along ($H_{||}$) and across ($H_{\perp}$) the wires for all the four samples. Thickness labelling is the same as in figure 3. Also shown in the figure are the fitted curves using equation (4) with $\alpha = 0.66$ for all the samples.

Table 2. Parameters derived from the fitting of $H_C$ versus $T$ dependence using equation (4).

<table>
<thead>
<tr>
<th>Sample</th>
<th>$H_C(0)$ (Oe)</th>
<th>$E_0$ (eV)</th>
<th>$V^*$ (nm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>977</td>
<td>1.15</td>
<td>1300</td>
</tr>
<tr>
<td>2</td>
<td>1305</td>
<td>1.9</td>
<td>1670</td>
</tr>
<tr>
<td>3</td>
<td>1582</td>
<td>1.02</td>
<td>730</td>
</tr>
<tr>
<td>4</td>
<td>1278</td>
<td>2.14</td>
<td>1895</td>
</tr>
</tbody>
</table>

and $M_s(0)$ are the saturation magnetization at $T$ and 0 K, respectively. Analysis of the temperature dependences of $H_{C||}$ observed in our experiments reveals that its magnitude increases sharply for the thinner sample (sample 1) as opposed to thicker samples (figure 5). The magnitude of $H_{C\perp}$ is also found to exhibit similar features. The dependency of $H_C$ on temperature has been fitted for all three samples using equation (4) and is shown along with the experimental data in figure 5. The curves are fitted with a value of $\alpha = 3/2$ corresponding to curling mode magnetization reversal. The choice of this value for the exponent $\alpha$ is due to the fact that the wire widths of the arrays are above the critical width predicted for coherent magnetization reversal for Co [4] which is estimated to be 15 nm ($d_c = 3.68\sqrt{A/(\pi M_s^2)}$). In fitting the experimental data we neglect the variation in saturation magnetization as it remains nearly constant in the temperature range studied (5–12% increase with temperature decreasing to 10 K). We notice that the experimental curves fit better with an increase in the thickness of the NWs suggesting that the reversal in thick NW arrays takes place via curling mode reversal. Greater disagreement of experimental fit of $H_C$ data with equation (4) for the thinner sample (sample 1), suggests the need to consider other sources of anisotropy, as the shape anisotropy contribution alone is not sufficient to explain our data particularly for small thickness NW arrays. While a qualitative agreement is achieved for thicker NW arrays, the situation for thinner wires (sample 1) suggests that the wires act as a system of weakly interacting particles which undergo a complex magnetization reversal procedure. This can be understood from the fact that the Co-NWs are formed by coalescence of small sized islands (average size $\sim$5–10 nm).

This island size is comparable to the critical diameter for magnetization reversal via coherent rotation. The individual islands switch their magnetization via coherent rotation but the presence of magnetostatic and exchange interactions among the crystal grains constituting the wires complicates the situation leading to a stronger deviation in value of the exponent $\alpha$ from the curling mode value. The values of $H_C(0)$ and $E_0$ along with the activation volume ($V^*$) determined from the fitting of experimental data with equation (4) are summarized in table 2. The effective barrier $E_0$ can be converted approximately to an effective volume involved in the magnetization reversal [4] using $E_0 = H_C(0)M_s(0)V^*$. We find that the value of $V^*$ ($\sim 10^3$ nm$^3$) is much smaller than the NW volume, suggesting that the magnetization does...
not reverse coherently in the whole NW and the reversal is localized within $V^*$. The presence of polycrystalline grains and surface related structural imperfections are known to produce magnetic localization of the reversal modes [34].

4. Conclusions

In summary, from the temperature and thickness dependent magnetization studies on ATLAS grown planar NW arrays of Co, we find that they exhibit ferromagnetic behaviour at RT with shape anisotropy being the dominant contribution to the effective magnetic anisotropy throughout the temperature range studied. Analysis of $T$ dependent magnetization data based on an analytical model that takes into account the effect of thermal activation and magnetostatic interactions among the wire suggest that the magnetization reversal is governed by the curling mode reversal for thick wires. Whereas for smaller thickness, the polycrystalline nature and surface related structural imperfections of the NWs lead to a magnetic localization of the reversal modes and are the main factors in governing the reversal behaviour.

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