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The effect of fluence on the magnetic properties of superparamagnetic iron-nickel nanoparticles in SiO_2 made by dual Ni and Fe low energy ion implantation

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Superparamagnetic $Ni_{1-y}Fe_y$ nanoparticles were made in a SiO_2 film by 10 keV ion beam implantation of Ni followed by Fe with a Ni fluence of 4×10^{16} at.cm^{-2} and a Fe fluence fraction of 0.47. Nearly all of the moments magnetically ordered, which was not reported for an implanted film made with a Fe fluence fraction of 0.56 and half the Ni fluence. The temperature dependence of the saturation moment is remarkably similar for low and high Ni fluences where there is also the presence of very thin spin-disordered shells. The higher Ni fluence leads to a significant enhancement of the susceptibility by a factor of 9 when compared with the lower fluence sample. This enhancement is likely to be due to a larger magnetically ordered volume fraction.

1. Introduction

Ion beam implantation is proving to be a very useful method to create magnetic materials with new magnetic and transport properties [1–9]. For example, low energy Fe implantation into SiO_2 followed by electron beam annealing was reported to result in near surface Fe nanoclusters and a very high magnetoresistance that has potential applications in high magnetic field sensing [2]. A number of other studies have also reported the formation of magnetic nanoparticles after single magnetic ion implantation into SiO_2 that includes Fe [3–5], Co [6,7] and Ni [8,9] nanoparticles. Dual implantation using one nonmagnetic ion has been reported in a few studies. For example, Co and Pt implantation into SiO_2 resulted in an enhanced coercivity and magnetic remanence [7] and Fe and Al implantation into SiO_2 led to Fe nanoparticles with the size and space group depending on the implantation order [4]. It would be very interesting to try dual implantation of magnetic ions and in particular Fe and Ni. This could lead to the formation of $Ni_{1-y}Fe_y$ nanoparticles where the bulk compound is known to have a high permeability, low coercivity, and large anisotropic magnetoresistance [10,11]. $Ni_{1-y}Fe_y$ also has a degree of electronic spin polarization and in powders this is known to lead to a spin-tunnelling magnetoresistance [11,12].

We have previously reported the formation of magnetic nanoparticles by dual Fe and Ni implantation with Fe fluence fractions of 0.18 [13] and 0.56 [1] where the Ni fluences were kept constant. These Fe fluence fractions were selected because bulk $Ni_{1-y}Fe_y$ with $y = 0.18$ is known to have the lowest magnetocrystalline anisotropy [10] and y close to 0.56 is where the saturation moment is the highest [14]. It was found that the nanoparticles were superparamagnetic but the saturation moment was significantly less than that in the bulk due to the formation of a spin-glass and some magnetic ions not magnetically ordering. It would be interesting to increase the Ni and Fe fluences and see if this enhances the magnetic properties while still being superparamagnetic at room temperature.

In this paper, we report the results from Rutherford backscattered spectrometry (RBS) and magnetic measurements on SiO_2 films that were implanted with Fe and Ni at low energies where the Ni fluence is twice that reported in the earlier study [1]. We show below that this leads to the formation of superparamagnetic nanoparticles, the enhancement of the saturation moment per implanted ion, and a significantly larger susceptibility.

2. Experimental details

$Ni_{1-y}Fe_y$ nanoparticles were made by implanting $^{58}Ni^+$ and $^{56}Fe^+$ into a 500 nm thick SiO_2 film that was on a 0.3 mm thick

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crystalline Si substrate. The implantation was done at 10 keV using the low energy metal implanter at GNS Science [15] at 10^{-7} mbar and with a low current of less than 2 μA. $^{58}Ni^{+}$ was implanted first with a fluence of 4.0×10^{16} at.cm^{-2} and this was followed by $^{56}Fe^{+}$ was a fluence of 3.5×10^{16} at.cm^{-2}. The Fe fluence fraction, y, was 0.47. The $^{58}Ni^{+}$ fluence was twice that used in a previous study where y was slightly higher and it was 0.56 [1]. The implanted concentrations were modelled as a function of depth using Dynamic TRansport of Ions into Matter (DTRIM) simulations [16]. RBS measurements were made using a 2 MeV He^{+} beam at GNS Science with a current of 20 nA. The fluences were obtained by fitting the RBS data using RUMP software [17]. A magnetic measurements property system from Quantum Design was used to measure the magnetic properties. The samples were measured with the applied magnetic field parallel to the surface of the samples.

3. Results and discussion

The results from DTRIM simulations are shown in Fig. 1(a) for the high fluence sample with $y = 0.47$. The Ni and Fe peaks are at ~13 nm and the implanted ions extend ~38 nm into the SiO_2 film. Also shown in Fig. 1(a) is the Fe fraction where it can be seen that it is 44% near the surface and increases to 47% at the peak depth after which it continues to further increase. For comparison we also show the simulated Ni and Fe concentrations from a lower fluence sample that had a fluence fraction of 0.56 [1]. The peak Ni and Fe concentrations are at approximately the same depth. However, the Fe fraction decreases with increasing depth where it is 59% near the surface and 55% at the peak concentration.

Fig. 2 shows the RBS data for the high fluence sample plotted against energy. The peak at ~1.5 MeV can be attributed to Ni and Fe in the SiO_2 layer. The RBS spectral resolution was not high enough to see distinct Ni and Fe peaks. Si in the SiO_2 layer has an edge at ~1.14 MeV and another edge at ~0.74 MeV is from Si at the Si/SiO_2 interface. Oxygen in the SiO_2 layer leads to an edge at ~0.74 MeV and the oxygen peak can be seen down to ~0.52 MeV. The Ni/Fe fluence was measured by fitting the RBS data using RUMP software. The fitted total fluence were 7.4×10^{16} at. cm^{2}. It is the same as the total implanted fluence within the experimental uncertainty (±5%).

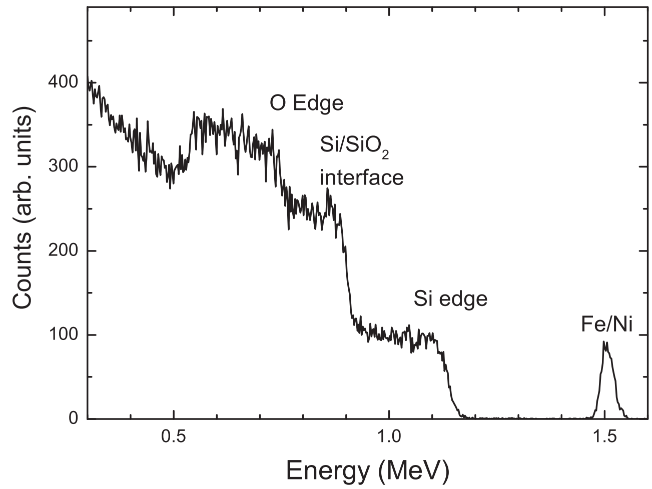

Fig. 2. Rutherford backscattering data for the high fluence sample with $y = 0.47$.

The moment per implanted ion, m_{ion}, is plotted in Fig. 3 for the high fluence sample against the applied magnetic field, B, at 5 K and 300 K. It was obtained from the measured moment, m, using $m_{ion} = [m - \chi_{Si} \times V \times B/\mu_0]/[F \times A]$, where A is the sample area, μ_0 is the permittivity of free space, F is the total Ni and Fe fluence, and χ_{Si}, is the silicon substrate susceptibility (-3.26×10^{-6}). m_{ion} increases with increasing B and then saturates above ~0.5 T where the 300 K saturation moment per implanted ion, $m_{ion,s}$, is only slightly lower that the value at 5 K. This is indicative of ferromagnetic order with a Curie temperature far above 300 K. $m_{ion,s}$ is 1.51 μ_B at 5 K, where μ_B is the Bohr magneton, and it is only slightly less than that found in bulk $Ni_{1-y}Fe_y$ with a similar x where $m_{ion,s} = 1.65$ μ_B [14]. This indicates that the ferromagnetic order arises from $Ni_{1-x}Fe_x$ nanoparticles that have formed after ion implantation. $Ni_{1-y}Fe_y$ nanoparticles also formed after ion implantation with a lower fluence and $y = 0.56$ [1]. However, in that case $m_{ion,s}$ was 0.56 μ_B and it was far less than the bulk value of 1.8 μ_B. The lower $m_{ion,s}$ was due to some Fe and Ni moments not magnetically ordering and there could also possibly be some antiferromagnetic phases. Thus, we find that doubling the Ni fluence has dramatically enhanced the magnetic order with nearly all of the Ni and Fe moments being ferromagnetically ordered.

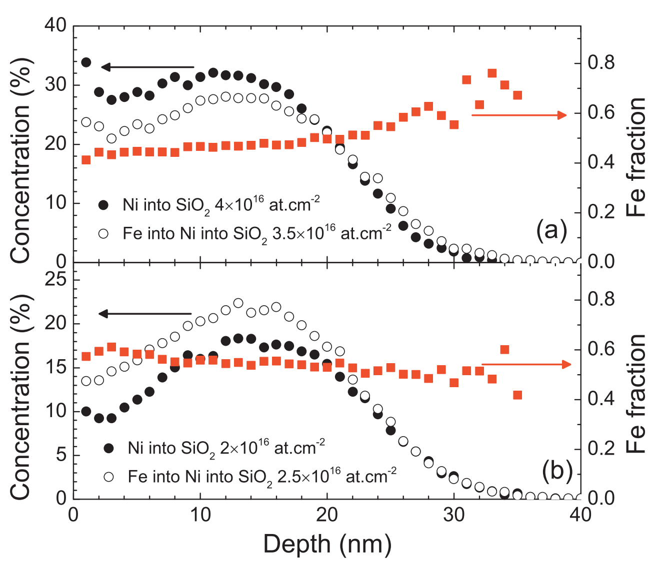

Fig. 1. DTRIM simulations for 10 keV implantation of Ni into SiO_2 followed by implantation of Fe with Ni and Fe fluences of (a) 4×10^{16} at.cm^{-2} (filled circles) and 3.5×10^{16} at.cm^{-2} (open circles) and (b) 2×10^{16} at.cm^{-2} (filled circles) and 2.5×10^{16} at.cm^{-2} (open circles), respectively [1]. Also show in the resulting Fe fraction as a function of depth (filled squares).

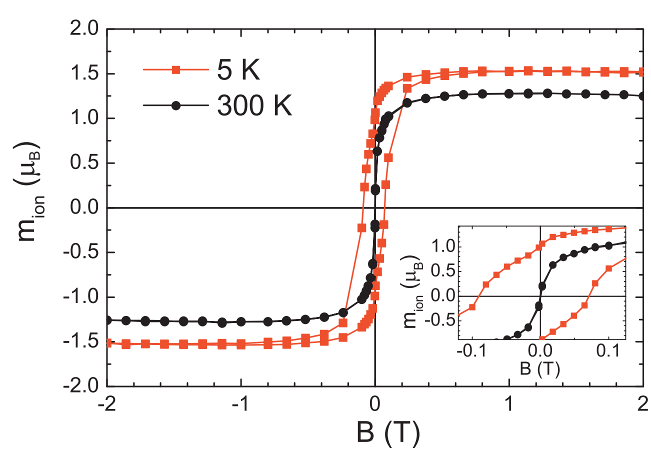

Fig. 3. The moment per implanted ion against the applied magnetic field, B, for the high fluence sample with $y = 0.47$ at 5 K (squares) and 300 K (circles). The inset shows the same data over a smaller magnetic field range.

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