Magnetic relaxation in nano-particle stacks

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Abstract. The remanent magnetisation and magnetic relaxation of Ni-Cu/Cu superlattice nanowires have been investigated. Arrays of superlattice nanowires were prepared by template deposition through polycarbonate nanoporous membranes using a single electrolyte bath. The thicknesses of nickel-rich layers ($t_{Ni}$) and copper layers ($t_{Cu}$) were independently controlled by monitoring the current during deposition. A study of the remanent magnetisation at 5K for $t_{Ni} = 30Å$ and a range of values of $t_{Cu}$ reveals the existence of inter-layer demagnetising interactions within each array. However the demagnetising interaction strength appears to reach a minimum level, believed to be due to intra-layer interactions caused by island formation within nickel-rich layers. Magnetic relaxation measurements on the same arrays after removal of a saturating (5T) field at various temperatures show $M$ to decrease linearly with $\ln(t)$. The data were analyzed using the $T\ln(t/\tau_0)$ scaling technique, revealing the effective energy barrier distribution of the arrays to be constructed of two components, possibly due to non- (or weakly-) interacting particles and strongly-interacting particles respectively. The weakly-interacting component is observed to decrease with decreasing $t_{Cu}$ and is believed to be caused by large individual nickel islands (corresponding to inter-layer interactions), while the strongly-interacting component is believed to be due to fragmented nickel islands (corresponding to intra-layer interactions).

1. Introduction
The advantages of electrodeposition as a method of preparing magnetic thin films are well-known, and include the low capital cost of the equipment required, and the selectivity, in the sense that deposition on a patterned substrate takes place only where there is a conducting path to the external circuit. The latter point is important when filling high aspect-ratio features in an insulating template with conducting base: where evaporation would lead to deposition on the insulator, possibly covering the features, electrodeposition only fills the features. Although the advantages of electrodeposition have long been appreciated for technological applications, they apply equally well to the fabrication of samples for so-called ‘pure’ research.

In this paper we report the results of investigations into the interactions and thermal relaxation of electrodeposited Ni-Cu/Cu multilayered nanowires. By electrodepositing metals within the pores of nanoporous nuclear track-etched membranes it is possible to prepare wires with lengths of several µm and diameters down to a few 10’s of nm. By switching the deposition potential between values at which different metals are electrodeposited it is possible to modulate the composition along the length of the wires with sub-nm precision [1]. If ferromagnetic layers alternate with non-magnetic layers the result is a multilayered nanowire that forms a stack of nanomagnets. These form a well-defined and easily controlled 1-D model system for studying magnetic interactions, but very little work has been

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done on this topic, despite the considerable attention paid to the magnetotransport properties of multilayered nanowires. In this paper we report how magnetic interactions between Ni-rich regions in the nanowires affect their bulk magnetic properties, in particular the magnetic remanence and the magnetic relaxation when a large field is first applied and then removed.

2. Experimental

Ni-Cu/Cu multilayered nanowires were deposited in commercially available polycarbonate membranes, 6 µm thick with a density of 6×10⁸/cm² track-etched pores, 80nm in diameter. A 200nm layer of gold was evaporated onto one side of the membrane to act as the working electrode. The electrolyte contained 2.3 M NiSO₄ and 0.05 M CuSO₄. Pure Cu was deposited at -0.4 V relative to a saturated calomel reference electrode, while a Ni-rich alloy was deposited at -1.9 V.

Magnetic data were obtained using a Quantum Design SQUID magnetometer. The magnetising remanence \( I_R(H) \) was measured by applying a series of increasing fields \( H \) to an initially demagnetised sample, and in each case determining the remanent magnetisation after removing the field. The demagnetising remanence \( I_D(H) \) was measured by applying a series of increasing fields \(-H\) to an initially saturated sample, again in each case determining the remanent magnetisation after removing the field. Clearly, \( I_R(0) = 0 \), \( I_R(0) = I_S \), \( I_R(\infty) = I_S \) and \( I_D(\infty) = -I_S \), where \( I_S \) is the saturation remanence.

More generally, for a system of non-interacting, single-domain particles possessing uniaxial anisotropy it is easy to show that

\[
I_D(H) / I_S = 1 - 2I_R(H) / I_S
\]

This is known as the Wohlfarth relationship [2], and deviations caused by interactions may easily be seen from a plot of \( I_D/I_S \) against \( I_R/I_S \), a so-called Henkel plot [3]. The thermal relaxation of the magnetisation at temperature \( T \) was measured by applying a field of 5 T to the sample, removing it and quenching the superconducting magnet to eliminate the residual field. The magnetisation was then measured as a function of time \( t \), the decay of which should follow the relationship

\[
M(t) = \int_0^{\infty} \exp\left[-(t / \tau_0)\exp(-E/kT)\right] f(E) dE
\]

where \( k \) is the Boltzmann constant and \( f(E) \) the energy barrier distribution [4].

3. Results and Discussion

Figure 1 shows magnetising and demagnetising remanence data measured for a series of multilayered nanowires consisting of alternating layers of Ni-Cu alloy with nominal thickness \( t_Ni \) = 30 Å and pure Cu with thickness \( t_Cu \) ranging from 50 to 300 Å. The data is presented in the form of Henkel plots, whereby \( I_D/I_S \) is plotted against \( I_R/I_S \) for the corresponding field \( H \), with the Wohlfarth relationship (eq.1) indicated by a straight line.

Note that in all cases the experimental data points lie beneath the Wohlfarth relationship line. This suggests that the remanence is influenced by demagnetising interactions. One obvious candidate would be the magnetostatic dipolar interactions between successive Ni-rich layers, which will tend to orient the layers antiferromagnetically, and are therefore demagnetising. Consistent with this, the deviation from the Wohlfarth relationship decreases with increasing \( t_Cu \), i.e. increasing layer separation. However, the deviation does not decrease between \( t_{Cu} = 200 \) Å and \( t_{Cu} = 300 \) Å, suggesting that there are intra-layer in addition to inter-layer magnetic interactions. These would be expected if rather than forming a single disc with diameter equal to the nanowire diameter, the Ni-Cu layers break up into islands separated by Cu or Cu-rich regions, as has been suggested by Bakonyi and co-workers [5].
Figure 1. Henkel plots measured at $T = 5$ K for a series of Ni-Cu/Cu multilayered nanowire arrays with $t_{\text{Ni}} = 30$ Å and the values of $t_{\text{Cu}}$ indicated on the figure. In all cases the data lies below the straight line.

Figure 2 shows magnetic relaxation data for an array of Ni-Cu/Cu multilayered nanowires with $t_{\text{Ni}} = 30$ Å and $t_{\text{Cu}} = 100$ Å measured at different temperatures $T$. At each $T$, $M$ decreases linearly with $\log t$. For a given $T$ we are only able to measure $M$ over a limited range of $t$, and the higher $T$, the greater the proportion of $M$ that has already relaxed before the magnetometer is able to start measuring. This is why $M$ corresponding to the lowest value of $t$ measured in the experiment decreases with increasing $T$. Similarly, the lower $T$, the greater the proportion of $M$ that relaxes after the end of the experiment. Thus by changing $T$, we change the part of the relaxation process that falls within our window of study.

Figure 3. Normalised magnetic relaxation data for an array of Ni-Cu/Cu multilayered nanowires with $t_{\text{Ni}} = 30$ Å and $t_{\text{Cu}} = 100$ Å measured at different temperatures $T$ indicated in the figure, plotted as a function of $T \ln(t/\tau_0)$. This may be seen by plotting all the data on a single curve, as a function of a variable that depends on both $T$ and $t$, as is done in Figure 3, known as the master plot. The reason for choosing $T \ln(t/\tau_0)$, where $\tau_0$ is a constant, as the independent variable in Figure 3 is that if $M$ is a single-valued function of this variable, then its gradient may be used to estimate the distribution of effective energy barriers in the system [4]. From Figure 3, it may be seen that although the data comes close to lying on a single curve with a physically reasonable value of $\tau_0$ ($10^{12}$ s), measurements at different $T$ do not quite overlap. The derivative of the master plot for multilayered nanowires with $t_{\text{Ni}} = 30$ Å and $t_{\text{Cu}} = 200$ Å is shown in Figure 4, revealing that the energy barrier distribution is constructed of two components. The first is an exponential decay, which can be related to strongly-interacting systems [6], the second is a log-normal distribution, normally associated with non- or weakly-interacting systems. Consistent with
this the log-normal component is observed to be suppressed as $t_{Cu}$ is decreased, increasing the inter-layer interaction strength (plots not shown). The weakly-interacting component is thus believed to be caused by large individual nickel islands, while the strongly-interacting component is believed to be due to fragmented nickel islands which give rise to the intra-layer interactions.

![Figure 4. Differential of the master plot for nanowires with $t_{Ni} = 30$ Å and $t_{Cu} = 200$ Å, revealing an energy barrier distribution which is constructed of two components](image)

![Figure 5. Simulated master plot for nanowires with $t_{Ni} = 30$ Å and $t_{Cu} = 100$ Å, showing no break down of scaling.](image)

A possible cause of the observed break down of scaling at high $T$ is an assumption integral to the scaling technique, by which the exponential term in equation (2) is approximated as a step function. The width of the step broadens as $T$ increases, and could cause the scaling to break down. In order to verify this, the experimentally derived $f(E)$ from the derivative of Figure 3 was used to simulate data using equation (2) over the same temperature range. This data was then scaled, and is shown in Figure 5. No break down of scaling was observed, indicating that the step function approximation holds true for the temperature range studied. The true cause of the break down of scaling is currently under investigation.

4. Conclusions.
Cu-Ni/Ni multilayered nanowires have been shown to exhibit both inter- and intra-layer demagnetising interactions, caused by the possible fragmentation of Ni layers into islands. The thermal magnetic relaxation of the wires over a wide range of temperatures has been shown to scale close to a single curve, with more than 50% of the nanowires’ magnetisation being thermally activated. The energy barrier distribution derived from these master plots has been shown to be constructed of two components, possibly related to weakly- and strongly-interacting particles. A break down of scaling is observed, the cause of which is under investigation.

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