Magnetotransport effects of ultrathin Ni$_{80}$Fe$_{20}$ films probed \textit{in situ}

S Krzyk, A Schmidsfeld, M Kläui$^1$ and U Rüdiger

Fachbereich Physik, Universität Konstanz, Universitätsstr. 10, D-78457 Konstanz, Germany
E-mail: mathias.klaeui@uni-konstanz.de

\textit{New Journal of Physics} 12 (2010) 013001 (15pp)
Received 7 October 2009
Published 11 January 2010
Online at \url{http://www.njp.org/}
doi:10.1088/1367-2630/12/1/013001

Abstract. We investigated the magnetoresistance of Permalloy (Ni$_{80}$Fe$_{20}$) films with thicknesses ranging from a single monolayer to 12 nm, grown on Al$_2$O$_3$, MgO and SiO$_2$ substrates. Growth and transport measurements were carried out at 80 K in UHV. Applying in-plane magnetic vector fields up to 100 mT, the magnetotransport properties were ascertained during growth. With increasing thickness the films exhibited a gradual transition from tunnelling magnetoresistance to anisotropic magnetoresistance. This corresponds to the evolution of the film structure from separated small islands to a network of interconnected grains, as well as the film’s transition from superparamagnetic to ferromagnetic behaviour. Using an analysis based on a theoretical model of island growth, we found that the observed evolution of the magnetoresistance in the tunnelling regime originated from changes in the island size distribution during growth. Depending on the substrate material, significant differences in the magnetoresistance response in the transition regime between tunnelling magnetoresistance and anisotropic magnetoresistance were found. We attributed this to an increasingly pronounced island growth, and to a slower percolation process of Permalloy when comparing growth on SiO$_2$, MgO and Al$_2$O$_3$ substrates. The different growth characteristics resulted in a markedly earlier onset of both tunnelling magnetoresistance and anisotropic magnetoresistance for SiO$_2$. For Al$_2$O$_3$ in particular the growth mode results in a structure of the film containing two different contributions to ferromagnetism, which lead to two distinct coercive fields in the high thickness regime.

$^1$ Author to whom any correspondence should be addressed.
1. Introduction

Spin-dependent transport phenomena have become a focus of research recently, with many different magnetoresistive (MR) effects being investigated. These effects are interesting from the point of view of fundamental physics as well as for possible applications in sensors and storage and logic devices. While a variety of novel effects have been discovered in nanoscale systems, such as giant magnetoresistance (GMR) \[1, 2\] and tunnelling magnetoresistance (TMR) \[3, 4\], anisotropic magnetoresistance (AMR) \[5\], which is well known, can be tailored for specific purposes in ultrathin films \[6\]. These effects have already entered into use in industrial devices such as hard drive read heads (GMR and TMR) and angular and positional sensors (AMR).

Discontinuous films of ferromagnetic metals on insulating substrates can exhibit both AMR and TMR effects \[7\]. When increasing the thickness of the film it will start to coalesce at some point, a process that is accompanied by changes in its electrical and magnetic properties, and accordingly changes of the MR can be expected. This radical change in transport behaviour from tunnelling to diffusive will also entail a radical change in prevailing magnetoresistive effects.

The first MR effect that will contribute to magnetotransport is tunnelling magnetoresistance, which occurs in a ferromagnet-insulator-ferromagnet (FM/I/FM) junction when the relative orientation of the magnetization of the FM contacts changes due to an external magnetic field \[3\]. This field tends to align the magnetizations in parallel, thus enhancing the probability of spin-dependent tunnelling and lowering the resistance during application of the field. Such a situation arises if separated islands are grown on an insulating substrate. The probable conduction mechanism for islands with a size of tens of nanometres and separations below 10 nm is thermally activated tunnelling, either substrate assisted or through vacuum. The tunnelling conductivity of these processes increases exponentially with decreasing island distance \[8\]. A film of separated islands consists of a large number of FM/I/FM junctions that form a complex conduction network. It has been shown that the magnetoresistance of such a network of ferromagnetic grains can be approximated by the magnetoresistance of a linear chain of contacts and even a single FM/I/FM junction \[9\]. Therefore, the behaviour of the film can be understood if we consider in the following the growth of a few neighbouring islands.

The magnetic behaviour of a discontinuous film depends on the interplay between exchange coupling giving rise to ferromagnetism and thermal excitation leading to superparamagnetism, with a strong dependence on the size of the magnetic islands. Using a value of $10^3$ Jm$^{-3}$ for the magnetocrystalline anisotropy $K$ of bulk Ni$_{80}$Fe$_{20}$ \[10\] and 80 K for
the film temperature \( T \), we get an upper limit for the radius \( r \) of a spherical superparamagnetic grain of [11]

\[
r \approx \left( \frac{6k_B T}{K} \right)^{1/3} \approx 20 \text{ nm}
\]

where \( k_B \) is the Boltzmann constant. If this value is larger than the typical island size in a film prior to percolation, we can assume that the film will be superparamagnetic. It has been shown that the magnetoresistance of a discontinuous film in this superparamagnetic regime can be modelled by assuming just two different grain sizes [12]. The magnetization for a single grain size as a function of the external field \( B \) is given by

\[
M(B) = M_{\text{sat}} \left( \coth \left( \frac{\mu B}{k_B T} \right) - \frac{k_B T}{\mu B} \right)
\]

with \( M_{\text{sat}} = N \cdot \mu \), where \( M_{\text{sat}} \) is the saturation magnetization of the film, \( \mu \) the magnetic moment of each grain, \( N \) the number of grains and \( T \) the temperature. Assuming two discrete grain sizes in the Permalloy film, corresponding to magnetic moments \( \mu_S \) and \( \mu_L \) for the smaller and larger grain size respectively, the total magnetization is then given by

\[
M(B) = M_{\text{sat}}^S \left( \coth \left( \frac{\mu_S B}{k_B T} \right) - \frac{k_B T}{\mu_S B} \right) + M_{\text{sat}}^L \left( \coth \left( \frac{\mu_L B}{k_B T} \right) - \frac{k_B T}{\mu_L B} \right)
\]

where the \( M_{\text{sat}}^i \) depend on the volume fractions \( \phi_i \) (\( i = S, L \)) of the respective grain sizes by

\[
M_{\text{sat}}^i = N_i \cdot \mu_i = \phi_i \cdot M_{\text{sat}}.
\]

The magnetoresistance can be calculated by assuming conduction only between neighbouring islands of similar size [13]. Considering that the tunnelling probability depends on the relative orientation of the neighbouring grains, we get a dependence on the square of the total magnetization and a total magnetoresistance of

\[
\Delta R/R = -A_S \left( \coth \left( \frac{\mu_S B}{k_B T} \right) - \frac{k_B T}{\mu_S B} \right)^2 - A_L \left( \coth \left( \frac{\mu_L B}{k_B T} \right) - \frac{k_B T}{\mu_L B} \right)^2
\]

with fractional magnetoresistance amplitudes at saturation of \( A_i \propto (\phi_i)^2 \). Accordingly, the ratio \( A_L/A_S \propto (\phi_L/\phi_S)^2 \) can be used to deduce changes in the relative volume fractions of smaller and larger grain size.

After growing above the superparamagnetic threshold, the formation of a classic ferromagnetic phase is expected, but there are several processes that can lead to a deviation from the magnetic properties of a bulk ferromagnet. As neighbouring islands approach each other, they can couple ferromagnetically even if they are electrically separated (superferromagnetism), via dipolar interaction [14], tunnelling exchange coupling [15] or surface atom exchange bridges [16]. Furthermore, higher-order multipolar interactions have been shown to be a possible mechanism for a changing coercivity in superferromagnetically coupled dense grain magnetic materials [17]. When two roughly circular islands grow together, they form a larger particle with a considerably higher aspect ratio. This gives rise to a non-negligible shape anisotropy and increases the coercivity significantly in comparison to the value of \( \lesssim 1 \text{ mT} \) for a homogeneous \( \text{Ni}_{80}\text{Fe}_{20} \) thin film [18, 19]. Due to the statistical distribution of island sizes and separations, these magnetic phases will not occur exclusively in a certain thickness range, but rather show a gradual transition with increasing thickness.

As the film thickness grows, bulk MR effects set in, with anisotropic magnetoresistance (AMR) being the most important. AMR in transition metals such as Fe and Ni is believed
to be a consequence of changes in the scattering probabilities of conduction electrons due to spin-orbit interaction that depends on the relative orientation of the local magnetization and the current [20]. Phenomenologically, the AMR is characterized by a \( \cos^2 \phi \) dependence of the resistance on the angle \( \phi \) between magnetization and current direction. For a discontinuous film it is necessary to consider that AMR only occurs intra-island, and so the observed resistance change is smaller than the intrinsic AMR effect within the islands. For a total resistance dominated by tunnelling and interface scattering, as is the case before a considerable coalescence takes place, the AMR contribution is thus expected to be negligible, but it will increase in importance with increasing film coalescence.

Previously, experiments have been carried out that have focused either on the low thickness regime where TMR dominates or on continuous films with a dominant AMR contribution [7, 12], [21]–[23], without studying the transition regime in detail. Results include measurement of the TMR in a granular Ni\(_{80}\)Fe\(_{20}\)/Al\(_2\)O\(_3\) film for various Permalloy concentrations [22], of the TMR in Ni on SiO\(_2\) intermittently grown up to the percolation threshold [23] and of the temperature dependence of TMR and AMR in Ni films grown on GaAs at a fixed film thickness [7]. From these measurements it could be established that the TMR in granular films diminishes when approaching the percolation threshold, and that coexistence of TMR and AMR in granular magnetic films is a possibility. But, so far no study has been made available that investigates the particularly exciting thickness regime where percolation occurs, and the development of TMR as well as AMR associated with the change in the transport regime from tunnelling to diffusive. Only with this information will a detailed understanding of the changes of the resistance and magnetic properties during this transition be obtained.

In this paper we use a unique combination of in-plane magnetic vector fields and in situ transport measurements during deposition via thermal evaporation under ultra-high vacuum (UHV) conditions to reveal the transition of the magnetoresistance response from TMR to AMR. This measurement procedure allows us to determine the TMR and AMR for an identical film area as a function of Ni\(_{80}\)Fe\(_{20}\) thickness without externally influencing its mechanical, electrical, thermal or chemical properties, as would be inevitable when using conventional set-ups involving sample transfer, either in situ or extra situm. From the measured magnetic and electric information we deduce the correlation between structural growth and magnetic properties. For growth on different substrates, we show that magnetotransport measurements are a useful tool to identify growth conditions without the need for complicated structural in situ scanning probe techniques.

2. Experimental

The samples used in this work consist of Permalloy (Ni\(_{80}\)Fe\(_{20}\)) deposited on different substrates via thermal evaporation from a rod of 6 mm diameter under UHV conditions. Multiple samples with Al\(_2\)O\(_3\), MgO and SiO\(_2\) substrates were investigated. The measurements carried out show a qualitatively similar behaviour for samples of the same substrate material. The chamber pressure during deposition was \( 5 \times 10^{-10} \) mbar. Prior to insertion into UHV, the samples were cleaned with acetone and isopropanol and Au contacts were defined on the surface in a rectangular pattern with 2 mm separation. The samples were degassed for 16 hours at 350 K and then cooled down to 77 K with LN\(_2\). Permalloy was evaporated at a constant rate, varying for different samples between 0.7 and 5 nm h\(^{-1}\), while the film thickness was monitored using a quartz microbalance. The sample temperature remained below 80 K during evaporation.
Figure 1. Sketch of measurement layout. $R_\perp$ and $R_\parallel$ indicate the resistance measurement configuration for measurements with the current direction perpendicular and parallel to the magnetic field, respectively.

All thickness values mentioned in this work refer to the nominal thickness as indicated by the quartz microbalance, which corresponds to the average thickness in the surface area between two Au contacts. Sample resistance was measured with a commercial multimeter (Keithley 6430) in a two-terminal set-up, covering the range from $20\,\Omega$ to $100\,\Omega$ without changes in the measurement set-up. The measurement layout is shown in figure 1. The in situ vector magnet allows for the application of in-plane fields up to $100\,\mu\text{T}$ at the sample position in an arbitrary orientation. Two measurement modes were used, either field ramps from $-100\,\mu\text{T}$ to $+100\,\mu\text{T}$ and back at a constant field angle (field sweep), or rotation of the field with a constant amplitude of $20\,\mu\text{T}$ (angle sweep). We define the value of the magnetoresistance as $\text{MR} = (R(100\,\mu\text{T}) - R(0\,\mu\text{T}))/R(0\,\mu\text{T})$. For each measurement step, several sweeps were carried out and averaged for drift and noise reduction. Magnetoresistance measurements were carried out at fixed film thicknesses by interrupting the deposition via a mechanical shutter and acquiring field-sweep and angle-sweep curves for several different contact pairs. After deposition of additional material, the measurements were repeated with increasing thickness up to a final thickness between 3.5 and 12 nm. Analysis of the sample topography was carried out extra situm by atomic force microscopy (AFM) at room temperature.

3. Results and Discussion

3.1. Resistance and AFM measurements

First we look at the resistance as a function of deposited Permalloy thickness (figure 2(a)) for MgO, Al$_2$O$_3$ and SiO$_2$ substrates. For all substrates the resistance initially drops exponentially with increasing film thickness, but the decrease in resistance slows significantly above some 3 nm for MgO (empty red circles) and Al$_2$O$_3$ (empty green squares) and above 1.5 nm for SiO$_2$ (empty black triangles). In comparison to MgO and Al$_2$O$_3$, the resistance at a given film thickness is orders of a magnitude lower for SiO$_2$, and the difference decreases with increasing film thickness.

The exponential decrease at low thickness is explained by the reduction of island separation with continuing deposition. As the film develops interconnects between islands and eventually
Figure 2. (a) Film resistance as a function of nominal film thickness for MgO (red empty circles), Al₂O₃ (green empty squares) and SiO₂ (black empty triangles) samples. (b-d) The magnetoresistance (solid symbols) is shown together with the island size distribution ratio as calculated from the MR loops ($A_L/A_S$) (empty symbols) versus nominal film thickness for MgO (b), Al₂O₃ (c) and SiO₂ (d) substrates. Arrows indicate the thicknesses corresponding to the curves shown in figure 4(a)–(d). The resistance measurements were carried out with the current direction perpendicular to the magnetic field. Magnetoresistance values below 2.0 nm for (d) were deduced by combining the results of parallel measurements with angle sweeps. The lines are guides to the eye.

becomes continuous, the resistance curve flattens off with higher film thickness. The conduction in the percolating regime is characterized by a combination of bulk-like behaviour, where the resistance is caused by scattering at the lattice, impurities and defects [24], and scattering at interfaces typical for thin films [25]. These effects limit the conductivity compared to the ideal ohmic resistor. Therefore, the resistance drops more slowly than the $1/d$ ratio expected for purely ohmic behaviour. The markedly lower resistance for SiO₂ can be explained by a lower average distance and height for the islands of the SiO₂ sample, which leads to percolation at significantly lower nominal film thickness. For higher thicknesses, all substrate types approach the continuous film state, resulting in a reduction of the conductance differences for a given film thickness. For low film thickness we find that the resistance decreases with increasing temperature, whereas for higher thicknesses it increases with increasing temperature. This is
in agreement with a changeover from conduction due to thermally activated tunnelling to a metallic behaviour as thicker films are deposited.

After the end of the Permalloy deposition, extra situm tapping-mode AFM images of the samples were acquired at room temperature. Accordingly, the surface morphology is expected to be significantly altered compared to conditions during growth at low temperature. Therefore the AFM measurements are used to qualitatively cross check the conclusions drawn from the transport measurement regarding surface structure. For a thickness of several nanometres, the films consist of islands of roughly circular shape with a typical diameter of 50 nm (figure 3(a)). Although single islands are clearly visible, partial overlap is evident from the height profile (figure 3(b)). The profile also shows an average island height of 4 nm for a film of 6 nm nominal thickness, suggesting a coalescent film. This indicates an island growth mode on our samples, which leads to the development of separated grains for the first few nanometres of film thickness. With additional deposition of material, these grains increase in volume, corresponding to a decrease in the average gap between grains, which eventually leads to inter-island connections and the formation of a continuous film.

3.2. Magnetoresistance

Next we investigate magnetotransport. The magnetoresistance curves of the Permalloy thin films, as well as the total magnetoresistance, show distinctive and non-monotonous changes with increasing film thickness. Figures 2(b)–(d) show the magnetoresistance amplitudes measured on an MgO ((b), solid red circles), an Al₂O₃ ((c), solid green squares) and an SiO₂ substrate ((d), solid black triangles) as a function of film thickness. The magnetoresistance initially increases with increasing film thickness and reaches a maximum at about 2.3 nm for MgO (figure 2(b)) and 2.8 nm for Al₂O₃ (figure 2(c)), corresponding to the thickness where the resistance curve begins to deviate from the exponential drop (see figure (a)). With a further increase in film thickness, magnetoresistance decreases. The general trend of magnetoresistance with increasing film thickness is similar for the MgO and Al₂O₃ sample. The MgO sample reaches a higher magnetoresistance, however, and the maximum is at a lower film thickness than for the Al₂O₃.
sample. This is attributed to the dependence of the growth process on substrate material and the statistical nature of the island growth process, leading to a different distribution of the island sizes and separations for both samples and accordingly leading to differences in the ratio of inter- and intra-island resistance for a given film thickness. In comparison to MgO and Al$_2$O$_3$, magnetoresistance as a function of film thickness for SiO$_2$ (figure 2(d)) develops markedly differently. Below 0.8 nm the magnetoresistance is below the noise floor, which is very high for the low thickness regime, as discussed later. Between 0.8 and 1.6 nm the magnetoresistance increases slightly to 0.05%, and then drops sharply to 0.01% above 1.6 nm, and increases again above 2.2 nm film thickness. The changes in magnetoresistance and the different behaviour for SiO$_2$ compared to, for instance, MgO with increasing thickness can be explained by looking at the resistance as a function of field strength (field-sweep measurements) for different film thicknesses. The shape of the MR curve for several thicknesses of the MgO and the SiO$_2$ sample from figure 2(b) and (d) respectively, are shown in figure 4, and these thicknesses are denoted by arrows in figure 2(b) and (d). Note that the curves in the low thickness range for Al$_2$O$_3$ and MgO have a similar shape, although Al$_2$O$_3$ exhibits more complex behaviour with increasing thickness, which will be discussed later (figure 7). At the lowest Py thickness (1.8 nm for MgO, 0.8 nm for SiO$_2$), negative MR is clearly visible (figure 4(a)). The MgO curve (black line) has a smooth maximum at zero field and drops with a slow decrease of the slope at higher fields. In contrast, the SiO$_2$ curve (blue line) shows a triangular shape typical for TMR in multilayers or granular films, with saturation reached for 100 mT. For the maximum MR (2.4 nm for MgO, 1.3 nm for SiO$_2$), the MR curve has narrowed for MgO, showing an increasing slope with decreasing field and a sharp maximum at zero field (figure 4(b)). The curve for SiO$_2$ has an almost identical shape, only deviating at higher fields and again reaching saturation for 100 mT, which is also the case for the next two thicknesses. After some further 50% of increases in thickness (to 3.6 nm for MgO, 1.7 nm for SiO$_2$), the MR has dropped by more than one order of magnitude; the curve shows an almost linear decrease with increasing field and a slightly higher slope for low fields (figure 4(c)). At 4.0 nm (2.3 nm for SiO$_2$) the MR has increased again; the curve is similar to figure 4(c), but with a significantly higher peak around 0 mT (figure 4(d)).

The changes of the magnetoresistance with increasing film thickness for MgO can be explained by the growth of superparamagnetic islands as follows. At low thickness, the film consists of small islands below the superparamagnetic threshold. When applying an external magnetic field, thermal fluctuations lead to deviations of the magnetization orientation of the islands from the parallel alignment to the field. This causes a broadening of the resistance peak at zero field and a slow increase of the film magnetization with increasing field (figure 4(a)). As the average island size increases, the influence of thermal fluctuations of the magnetization diminishes. This results in an increase of the tunnelling magnetoresistance and a more pronounced resistance peak at zero field, as can be seen by comparing figure 4(a) to (b). Concurrent to this process is the onset of percolation above a certain thickness, reducing the contribution of tunnelling to the total resistance and therefore reducing TMR. This leads to a decrease in the total MR in figure 4(c). When the coalescence of the film has sufficiently increased, bulk-like conduction begins to contribute significantly, corresponding to the onset of AMR. If the measurement current is orientated perpendicularly to the magnetic field, the AMR leads to a reduction of the resistance with increasing field, which is indicated by the enhancement of the peak close to zero field in figure 4(d). The different behaviour for SiO$_2$ in the low thickness regime can be explained by larger islands compared to MgO, leading to a lower influence of thermal excitations on the magnetization, which is more stable due
Figure 4. Field sweep MR curves of Permalloy on MgO (black circles) and SiO$_2$ (blue circles) for different film thicknesses as indicated in figure 2(b) and (d). Comparable states of percolation for MgO and SiO$_2$ respectively are shown adjacent to one other: (a) 1.8 nm MgO, 0.8 nm SiO$_2$; (b) 2.4 nm MgO, 1.3 nm SiO$_2$. Note that (c) 3.6 nm MgO, 1.7 nm SiO$_2$ and (d) 4.0 nm MgO, 2.3 nm SiO$_2$. The dotted black line in (b) shows the curve from (a) for comparison. For the MgO samples, the red lines in (a), (b) and (c) show a fit of the experimental data using the model of two different superparamagnetic island sizes [4]. The black and blue lines are guides to the eye. Note that we do not attain saturation of our samples for all measurements, as can be seen by the significant slope of the curves at the maximum fields for MgO.

to the larger volume. For lower temperatures the curves for the MgO substrate would have a comparable shape (as shown for the Ni/SiO$_2$ system at 4 K in [23]). The fast increase of the magnetoresistance above 2.3 nm for SiO$_2$ is due to a rapidly increasing AMR above the percolation threshold.

This interpretation of the magnetotransport data can be supported by theoretical calculations based on equation (4). This model allows us to deduce the changes in the size distribution of the islands with increasing film thickness from the shape of the MR curves.
as shown in figure 4 (red solid lines). Using equation (4) with $A_S$, $A_L$, $\mu_S$ and $\mu_L$ as free parameters, the field sweep MR curves can be fitted very well in the TMR regime, as exemplified by figure 4(a)–(c). Although the physical size distribution of the islands can be assumed to be continuous, the MR behaviour can be described satisfactorily by two discrete island sizes. For low thicknesses, only one pair of parameters is required, suggesting that all islands contributing to the MR are of similar size. For higher film thicknesses, the curves deviate from the model describing the situation of superparamagnetic islands, which becomes less and less valid.

The values for the magnetic moments $\mu_L$ and $\mu_S$ are a measure of the respective volumes of the grains. The saturation magnetoresistances $A_L$ and $A_S$ are proportional to the square of the respective volume fraction $\phi_i$ and also to the magnetic moments $\mu_i$. For the MgO and Al$_2$O$_3$ substrates, the parameters $\mu_L$ and $\mu_S$ become larger with increasing film thickness, indicating an overall increase in size for all islands. This development stops on reaching the TMR/AMR transition regime and then reverts slightly on further increase of the film thickness. For the SiO$_2$ substrate, the changes for the $\mu_i$ are less pronounced.

The behaviour of the saturation magnetoresistances $A_L$ and $A_S$ is sensitive to changes in average size as well as to the quantity of the large and the small island species. By using two constant values for $\mu_L$ and $\mu_S$ in equation (4) we obtain a good fit for the TMR regime. This eliminates the $\mu_i$ as variables and allows us to use the ratio $A_L/A_S \propto (\phi_L/\phi_S)^2$ as a single parameter to describe the development of the average size of the superparamagnetic islands by changes in the relative numbers of two discrete island sizes with good agreement to the experiment. For MgO the best-fit values for the island magnetic moments are between 1500$\mu_B$ and 70 000$\mu_B$, and the fixed values used were $\mu_S = 4700\mu_B$ and $\mu_L = 47 000\mu_B$. For Al$_2$O$_3$ the best-fit values for the island magnetic moments are between 5000$\mu_B$ and 90 000$\mu_B$, and the fixed values used were $\mu_S = 7000\mu_B$ and $\mu_L = 70 000\mu_B$. The best-fit values for the SiO$_2$ substrate are between 10 000$\mu_B$ and 120 000$\mu_B$, and the fixed values are $\mu_S = 14 000\mu_B$ and $\mu_L = 95 000\mu_B$. While the values of the magnetic moments for the MgO and Al$_2$O$_3$ substrates show similar behaviour, the larger and less diffuse values for SiO$_2$ indicate a larger island size with a narrower size distribution, leading to a lower percolation threshold.

The changes in $A_L/A_S$ with increasing film thickness for the different sample types can be seen in figure 2(b)–(d) (empty symbols). For MgO and Al$_2$O$_3$, $A_L/A_S$ mirrors the changes in the total MR (solid symbols), with a small shift to higher thicknesses. This similar behaviour is at first surprising given the fact that while $A_L$ and $A_S$ are both correlated with the total MR (see equation (4)), the ratio $A_L/A_S$, which describes the relative contribution of the large and small island types to the overall resistance and can therefore be used as a measure of the average size of the superparamagnetic islands, is not necessarily directly related to the MR.

To understand how the changes of $A_L/A_S$ affect the overall magnetoresistance, one needs to take into account the fact that the films incorporate Permalloy islands ranging in size from small nonmagnetic grains and superferromagnetic islands to larger ferromagnetic regions. Only the intermediate-sized superferromagnetic islands contribute to the TMR, and accordingly only these affect the shape of the TMR loops and are thus modelled. As we do not reach saturation at 100 mT and larger islands reach saturation at lower fields, an increase in average island size translates into an increase in the TMR amplitude, assuming a constant value of the saturation MR. This is reflected in the parallel rise of the MR and $A_L/A_S$ for low film thicknesses in 2(b) and (c). When the films reach the percolation threshold, the largest islands tend to merge to form large ferromagnetic continuous areas, which are outside the model; hence, effectively, this reduces the number of large superparamagnetic islands. This effect
counteracts the overall increase of the size of each island due to deposition, leading to a peak of the ratio $A_L/A_S$ shortly after the onset of percolation. Additionally, the area fraction of the superparamagnetic islands diminishes, so transport to an increasing extent takes place in the ferromagnetic part of the film, thereby reducing the TMR amplitude. As a result, the overall MR starts to decrease before $A_L/A_S$ reaches the maximum. With continued percolation, the contribution of superparamagnetic islands to the overall conductivity steadily decreases, which, in addition to the decreasing $A_L/A_S$, leads to a fast drop of the MR. The SiO$_2$ sample exhibits a slightly different behaviour compared to MgO and Al$_2$O$_3$. For low thicknesses, the average island size rises slowly in parallel with the MR to 1.5 nm film thickness. When the MR drops sharply with further increases in thickness, $A_L/A_S$ also starts to decline, although less abruptly. We note that the separate values of $A_L$ and $A_S$ follow the sharp drop of the MR from 1.5 to 1.7 nm. This can be understood by considering that the transition to a continuous film occurs at a lower thickness and in a somewhat narrow thickness range compared to those for MgO and Al$_2$O$_3$, corresponding to a larger and more uniform island size on SiO$_2$. Therefore the changes in the size distribution ratio $A_L/A_S$ are more closely correlated to the development of the MR for the SiO$_2$ sample. So we see that our theoretical analysis confirms our interpretation of the MR evolution as a function of film thickness; the rise of the MR in the low thickness regime can be attributed to a steady increase in average island size, resulting in an increase in average magnetization for a given magnetic field strength and an increase of the TMR. With the onset of percolation, the average island size decreases and the superparamagnetic fraction of the film is reduced, leading to a reduction of the overall MR, up to the thickness where AMR starts to contribute significantly to the MR.

As can be inferred from the shape of the MR curves in figure 4(d), the contributions of AMR and TMR appear superimposed in the field-sweep measurements. The magnetoresistance in the TMR thickness regime was found to be isotropic, as expected for a random distribution of island sizes and distances. Under the assumption that this is also the case for higher film thicknesses, angle sweep measurements can be used to separate the contributions of the angle-dependent AMR and the isotropic TMR. The development of the AMR for Permalloy grown on MgO, Al$_2$O$_3$ and SiO$_2$ substrates is shown in figures 5(a)–(c) for a SiO$_2$ sample, figures 5(d)–(f) for a MgO sample and figures 5(g)–(i) for Al$_2$O$_3$. The onset of the AMR occurs at the lowest thickness for SiO$_2$, followed by MgO and Al$_2$O$_3$. The AMR exhibits a clear $\cos^2\phi$ oscillation of the resistance for all samples, and increases with increasing thickness. A notable feature is the significantly higher noise at the onset of AMR when compared to larger thicknesses for all substrates. This can be clearly seen when comparing the angle-sweep curves at the onset of AMR, and after a slight increase in thickness (figure 5(a) versus (b)) for SiO$_2$, figure 5(d) versus (e) for MgO and figure 5(g) versus (h) for Al$_2$O$_3$). Although the signal amplitudes differ by less than a factor of 2, the noise is considerably higher for lower thicknesses. This is consistent over all measurements, making it impossible to detect AMR below a relative value of some $10^{-4}$. Sousa et al have attributed this behaviour to Barkhausen jumps of domain walls in the percolating film [26]; this effect could also be due to an increased sensitivity of the conductivity to random fluctuations of the atomic structure of the Permalloy film in the percolating regime.

The increase of the AMR as a function of Permalloy film thickness is shown in figure 6 for an Al$_2$O$_3$ substrate (empty green squares), for an SiO$_2$ substrate (empty black triangles) and for an MgO substrate (empty red circles). After its onset (for instance, for Al$_2$O$_3$, between 3 and 4 nm film thickness), the AMR increases roughly linearly with increasing thickness. This is in good agreement with measurements in continuous multilayer films, where a linear...
Figure 5. Angle sweep MR curves of Permalloy grown on SiO$_2$, MgO and Al$_2$O$_3$ substrates for various film thicknesses up to 7.5 nm: (a) SiO$_2$, 2.0 nm; (b) SiO$_2$, 2.3 nm; (c) SiO$_2$, 4.0 nm; (d) MgO, 3.1 nm; (e) MgO, 3.7 nm; (f) MgO, 5.0 nm; (g) Al$_2$O$_3$, 3.6 nm; (h) Al$_2$O$_3$, 4.4 nm and (i) Al$_2$O$_3$, 7.5 nm. Field angles of 90$^\circ$, 170$^\circ$ and 45$^\circ$ correspond to the parallel orientation of the magnetic field and the current direction for the SiO$_2$, MgO and Al$_2$O$_3$ substrates, respectively.

increase of the AMR up to 10 nm Permalloy thickness was found [27]. In comparison to the magnetoresistance values extracted from field-sweep measurements, which are also shown in figure 6 (solid symbols), the onset of the AMR coincides with the drop of the TMR, which is followed by a transition regime up to about 7 nm thickness for the Al$_2$O$_3$ sample, where both AMR and TMR occur. Above 7 nm, only AMR contributes to the total magnetoresistance; therefore AMR and MR rise in proportion. The MR effect for the SiO$_2$ sample shows an analogous behaviour, but at significantly lower thicknesses. Results for the MgO samples lie in between those for Al$_2$O$_3$ and SiO$_2$.

For Al$_2$O$_3$ the transition between TMR and AMR is even more directly visible in the field-sweep measurements, as shown in figure 7(a)–(d). At 4.3 nm thickness, only a TMR curve is observable, with peaks (dark blue arrows) at $\pm 2$ mT (figure 7(a), inset). As the thickness is increased, AMR peaks (denoted by light red arrows in figure 7(b)) appear superimposed on the TMR curve. The TMR peaks are visible at a higher field of $\pm 4$ mT. With further increases in film thickness the AMR peaks become more pronounced, staying at low fields close to zero.
Figure 6. Magnetoresistance measured by field sweeps with the current direction perpendicular to the magnetic field (solid green squares for Al$_2$O$_3$, solid red circles for MgO and solid black triangles for SiO$_2$) and AMR as extracted from angle sweep measurements (empty green squares for Al$_2$O$_3$, empty red circles for MgO and solid black triangles for SiO$_2$) for Permalloy grown on Al$_2$O$_3$, MgO and SiO$_2$ substrates. The lines are guides to the eye.

By contrast, the TMR peak heights are reduced and they move to even higher absolute fields ($\pm 5$ mT in figure 7(c)). At 6.9 nm, the TMR is barely visible and the peaks are at about $\pm 6$ mT, while the AMR peaks at low fields dominate (figure 7(d)). For the MgO and SiO$_2$ samples on the other hand, no differences in the coercive fields detected by TMR and AMR were observed, while the value of the coercive field remains below 1 mT (see figures 4(b)–(d)).

The observation of two different coercive fields for the TMR and the AMR effect for the Al$_2$O$_3$ samples (figure 7) indicates the presence of two different contributions to the ferromagnetism in the TMR/AMR transition regime. These can be identified as the bulk ferromagnetism of continuous film areas that gives rise to the AMR and which thus shows a low coercivity, and either superferromagnetism or multi-island ferromagnetism that give rise to an enhanced coercivity of the small islands that contribute to the TMR. As these processes are very sensitive to island size, shape and separation, this implies that differences in the distribution of island size and separation exist for Al$_2$O$_3$ when compared to MgO and SiO$_2$ substrates, and have a clear influence on the magnetoresistive behaviour only in the percolating regime. A possible explanation for this surprising behaviour exhibited by the Al$_2$O$_3$ substrates is indicated by the higher film thickness where percolation occurs and the broader thickness range of the transition regime from TMR to AMR for Al$_2$O$_3$ substrates in comparison to the other substrate types. This means that the morphological transition from separated islands to a continuous film is quite gradual for Al$_2$O$_3$ substrates, with a broad intermediate regime where separate islands with inter-island tunnelling transport exist, and at the same time continuous film areas are present where diffusive transport with AMR occurs. In this transition regime both conduction mechanisms can contribute to a similar extent to the resistance of the film.
Figure 7. Field sweep MR curves of Permalloy on Al$_2$O$_3$ for film thicknesses between 4 and 7 nm: (a) 4.3 nm, (b) 5.2 nm, (c) 6.4 nm and (d) 6.9 nm. The insets show the low field regime of the field sweeps. The multiple peaks of the MR curves are indicated by arrows above the peaks and lines on the field axis (dark blue for TMR at large fields and light red for AMR at small fields). The positions of the TMR peaks move to higher field with increasing thickness, from 2 mT at 4.3 nm (a) to 6 mT at 6.9 nm (d). The resistance measurements were carried out with the current direction perpendicular to the magnetic field.

3.3. Conclusion

In conclusion, we have determined the magnetotransport properties of ultrathin Permalloy (Ni$_{80}$Fe$_{20}$) films on insulating substrates. The films exhibit a change from the TMR regime to the AMR regime with increasing thickness, with a transition zone where both AMR and TMR are present. The MR loops in the TMR regime can be fitted using a theoretical model and remarkable agreement between the MR amplitude and the island size distribution is found. The transition to AMR corresponds to the percolation process with continuing material deposition and the transition from superparamagnetic behaviour to ferromagnetism. The film thickness dependence of the resistance, the TMR and the AMR is qualitatively similar for Al$_2$O$_3$, MgO and SiO$_2$ substrates for the thickness regimes where the MR response is clearly dominated by either TMR or AMR. SiO$_2$ exhibits a markedly earlier onset of both tunnelling magnetoresistance and anisotropic magnetoresistance, due to an onset of percolation at lower thickness. In the transition zone from the TMR to the AMR regime we find markedly different behaviour for the different substrate materials. SiO$_2$ shows a very abrupt reduction in TMR within a thickness increase of 0.2 nm, whereas for Al$_2$O$_3$ and MgO the transition is broader.
For Al₂O₃ in particular the transition zone covers a larger thickness range (3 nm), and this is accompanied by the TMR and AMR effect revealing different coercivities. This in turn points to the slower percolation process and a large thickness range where tunnelling between islands and intra-island transport contribute significantly to the conduction for Permalloy films grown on Al₂O₃ in comparison to MgO and SiO₂. So even though polycrystalline growth occurs on all substrates, the transition zone exhibits reproducibly very different behaviour for the different substrate materials, further highlighting the importance of the substrate even if no epitaxy is used. For additional investigation, measurements at different deposition temperatures are conceivable. Our results clearly demonstrate that in situ transport measurements are a suitable tool for in-depth investigation of growth conditions of ultrathin magnetic films.

**Acknowledgments**

The authors acknowledge support by the DFG (SFB 513 and 767), the Landesstiftung Baden-Württemberg and the ERC (ERC-200)-Stg 208162-MASPI

**References**

[1] Baibich M N et al 1988 Phys. Rev. Lett. 61 2472
[2] Binasch G, Grünberg P, Saurenberg F and Zinn W 1989 Phys. Rev. B 39 4828
[3] Julliere M 1975 Phys. Lett. A 54 225
[4] Moodera J S, Kinder L S, Wong T M and Meservey R 1995 Phys. Rev. Lett. 74 3273
[5] Thomson W 1857 Proc. Royal Soc. 5 546
[6] Ding L et al 2009 Appl. Phys. Lett. 94 162506
[7] Gürttler C M, Xu Y B and Bland J A C 2001 J. Magn. Magn. Mater. 226 655
[8] Neugebauer C A and Webb M B 1962 J. Appl. Phys. 33 74
[9] Vilchik H, Frydman A and Berkovits R 2006 Phys. Status Solidi c 3 288
[10] Yin L F et al 2006 Phys. Rev. Lett. 97 067203
[11] O’Handley R 2000 Modern Magnetic Materials (New York: John Wiley)
[12] Honda S, Okada T, Nawate M and Tokumoto M 1997 Phys. Rev. B 56 14566
[13] Helman J S and Abeles B 1976 Phys. Rev. Lett. 37 1429
[14] Beleggia M, Zhu Y, Tandon S and de Greaf M 2005 Appl. Phys. Lett. 87 202504
[15] Bakuzis A F and Morais P C 2005 J. Magn. Magn. Mater. 285 145
[16] Hansen M F, Koch C B and Mørup S 2000 Phys. Rev. B 62 1124
[17] Vedmedenko E Y, Mikuszein N, Oepen H P and Wiesendanger R 2005 Phys. Rev. Lett. 95 207202
[18] Michelini F et al 2002 J. Appl. Phys. 92 7337
[19] Cowburn R P 2003 J. Appl. Phys. 93 9310
[20] Potter R I 1974 Phys. Rev. B 10 4626
[21] Gittleman J I, Abeles B and Bozowski S 1974 Phys. Rev. B 9 3891
[22] Brućas R et al 2007 J. Appl. Phys. 101 073907
[23] Frydman A, Kirk T L and Dynes R C 2000 Solid State Commun. 114 481
[24] Ziman J M 1962 Electrons and Phonons (Oxford: Oxford University Press) p 486
[25] Sondheimer E H 1952 Advan. Phys. 11
[26] Sousa J B et al 2004 J. Appl. Phys. 96 3861
[27] Thanh N T et al 2007 J. Appl. Phys. 101 053702

New Journal of Physics 12 (2010) 013001 (http://www.njp.org/)