Magnetoresistance of Ag/Fe/Ag and Cr/Fe/Cr trilayers

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Magnetoresistance arising from a nanoscale magnetic structure was first discovered in antiferromagnetically coupled multilayers prepared by molecular beam epitaxy (MBE) [1,2]. It is generally referred to as giant magnetoresistance (GMR) due to the large resistance change as compared to the anisotropic magnetoresistance (AMR) of bulk ferromagnetic materials. The large effect gives great potential for technological applications, however, as concerning the underlying physics the absence of anisotropy in case of the GMR effect [1] is more important. This means that $[R(H) - R(0)]/R(0)$ is negative irrespective of the direction of the measuring current and the applied magnetic field when both lie in the sample plane. Since the first reports on GMR it turned out to be a more complex phenomenon, as it was observed in polycrystalline multilayers [3], granular materials [4,5], magnetic domain walls [6] or supersaturated alloys [7], as well. The criteria for observing GMR can be put more generally: the characteristic length scales of the magnetic inhomogenities should be in the order of the electronic mean free path. However, the respective role of different bulk and interface scattering processes [8] is not yet clear even in the case of multilayers. Many recent experimental [9,10] and theoretical [8,11] works addressed this question. Serious efforts were made to compare measurements on multilayer samples after altering the impurity content and the interface quality by different methods [9], however, analysis of the structural changes and its impact on the different scattering processes is usually not straightforward. Trilayers containing two magnetic layers separated by a non-magnetic spacer were also studied [2] in order to investigate the GMR arising from antiferromagnetic interlayer coupling.

In this study we present a different approach and show that the magnetoresistance arising from chemical mixing at the interface can be studied by measurements on trilayers with a sequence of non-magnetic/magnetic/non-magnetic (N/M/N) layers. The magnetoresistance of N/M/N trilayers has been scarcely investigated [12] in the past. The magnetoresistance of our vacuum evaporated N/M/N trilayers has GMR characteristics, which is attributed to the formation of a granular interface structure. The observation of a granular interface magnetoresistance raises the question as to how this term is related to the magnetoresistance observed in multilayers. Our study demonstrates that the granular interface contribution is dominant in Fe/Ag multilayers. By investigating Cr/Fe/Cr trilayers it is also shown that the granular interface magnetoresistance is not restricted to immiscible elements.

The trilayer and multilayer samples were prepared by vacuum evaporation with two electron guns in a base pressure of 10^{-7} Pa. The deposition was made with a rate between 0.1 and 0.2 nm/s on Si single crystal substrate at room temperature. The layer thickness was controlled by a

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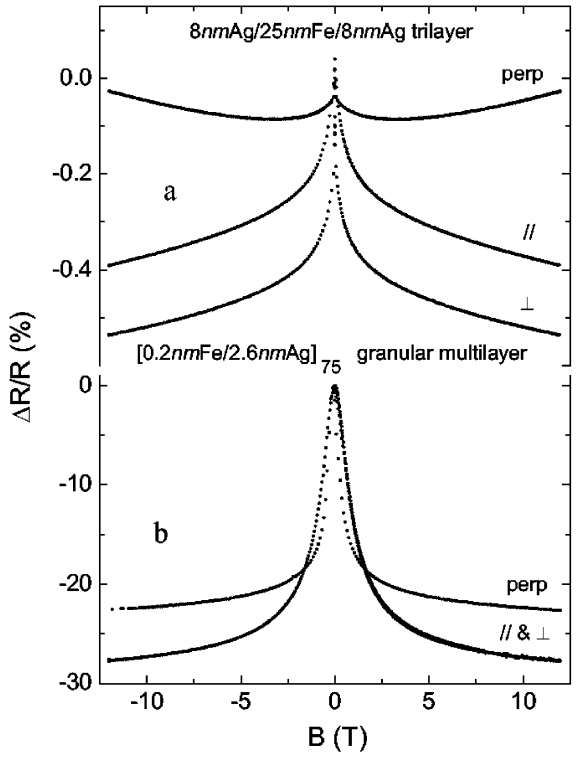

Fig. 1. Magnetoresistance measured at $T = 4.2$ K with magnetic field aligned parallel ($\|$), transversal ($\perp$) and perpendicular (perp) to the measuring current for a 8 nm Ag/25 nm Fe/8 nm Ag trilayer (a) and a granular sample prepared as a discontinuous multilayer of [0.2 nm Fe + 2.6 nm Ag]$_{75}$ nominal sequence (b).

quartzoscillator during sample deposition. The magnetoresistance was measured by four contact method on 2 mm thick and 10 mm long samples with current in the plane geometry. The magnetic field was applied in three geometries: (i) in the sample plane parallel to the current (ii) in the sample plane perpendicular to the current and (iii) perpendicular to the sample plane. Magnetoresistance measured in the earlier geometries are usually called parallel ($R_{\|}$), transversal ($R_{\perp}$) and perpendicular (R_{perp}), respectively. The layer thickness range of the trilayers (8 nm for Ag and Cr and 25 nm for Fe) was small enough that the interface magnetoresistance was not shunted by the resistance of the layers, however it was thick enough that the Fe layer shows normal ferromagnetic behavior with parameters (saturation magnetization, Curie temperature, demagnetization field) similar to bulk layers. The multilayered samples also exhibited ferromagnetic magnetization curves with in-plane anisotropy. Superparamagnetic behavior could only be observed in multilayers with Fe layer thickness smaller than 0.7 nm [14].

Magnetoresistance curve of the as deposited 8 nm Ag/25 nm Fe/8 nm Ag trilayer measured in parallel, transversal and perpendicular geometries up to $B = 12$ T magnetic field at $T = 4.2$ K is shown in Fig. 1(a). $R_{\|}$ and $R_{\perp}$ show similar magnetic field dependence in the high field region, but the two curves are shifted relative to each other because of a small AMR below 0.2 T. The lower value of $R_{\perp}$ is consistent with thin film measurements on Fe [13] indicating a cross-over in the sign of the parallel and the transversal magnetoresistance around this layer thickness. However, the high field behavior is rather unusual. The equal decrease of the parallel and the transversal magnetoresistance and the absence of saturation up to 12 T magnetic field have not yet been observed on a single ferromagnetic Fe layer. On the other hand the cusp like shape of the magnetoresistance curves and the extremely high saturation field is typical of granular systems [4,5].

Magnetoresistance of a Fe–Ag sample which has a [0.2 nm Fe + 2.6 nm Ag]$_{75}$ nominal multilayer structure is shown in Fig. 1(b). The thin Fe layers are not continuous in this sample and this specimen shows characteristics of a granular system, e.g. it is superparamagnetic with a blocking temperature around 40 K [14]. The magnetic field dependence of $R_{\|}$ and $R_{\perp}$ also shows the characteristic features observed on granular samples prepared by co-deposition of the constituents [15]. A distinct feature of our granular sample prepared by sequential deposition is the anisotropy observed when the magnetic field is perpendicular to the sample plane. In co-deposited granular materials [16] there are only minor differences between $R_{\|}$, $R_{\perp}$ and R_{perp}. The layered growth seems to strongly affect the sample morphology and probably the shape of the granules. Note that R_{perp} of the trilayer sample also shows distinct behavior (see Fig. 1(a)).

In granular materials the GMR phenomenon is attributed to spin dependent scattering on single domain ferromagnetic particles and is shown to scale with the reduced magnetization [4] as:

$$\frac{R(H) - R(0)}{R(0)} = -A\left(\frac{M}{M_s}\right)^2, \tag{1}$$

where M is the global magnetization of the sample and M_s is the saturation magnetization. The prefactor A depends on the number and the size of the single domain particles. According to the classical theory of superparamagnetism the reduced magnetization can be described by the Langevin function, therefore

$$\frac{R(H) - R(0)}{R(0)} = -AL^2(mH/kT), \tag{2}$$

where $L(x) = \text{cth}(x) - 1/x$ and m is the magnetic moment of the superparamagnetic particles. Eq. (2) was extended in order to be applicable for the trilayer and the multilayers, as well:

$$\frac{R(H) - R(0)}{R(0)} = -A_1L^2(mH/kT) - A_2H^2 + A_3 : \tag{3}$$

To account for possible scattering on single Fe impurities in the non-magnetic matrix a term proportional to H^2 was

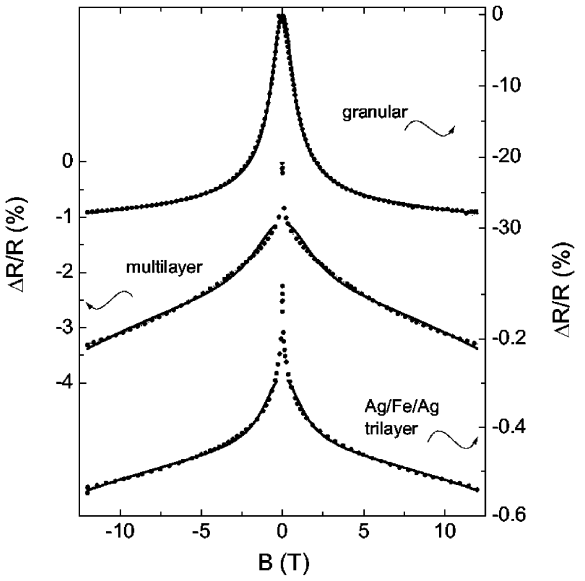

Fig. 2. Magnetoresistance curves measured with transversal magnetic field alignment at 4.2 K and fitted according to Eq. (3) in the $|B| > 0.25$ T magnetic field range. Upper panel is for a granular sample prepared as a discontinuous multilayer of [0.2 nm Fe + 2.6 nm Ag]$_{75}$ nominal sequence. Middle panel is for a multilayer of [1.4 nm Fe + 2 nm Ag]$_{60}$ nominal sequence with continuous ferromagnetic layers. Lower panel is for a 8 nm Ag/25 nm Fe/8 nm Ag trilayer. Since the measured points and the fitted curves mostly overlap on this scale the measured points are rarefied for clarity.

included [17]. The constant A_3 describes the shift in the high field magnetoresistance due to the AMR contribution of ferromagnetic particles. All the experimental curves could be satisfactorily fitted with Eq. (3) in the $B >$ 0.25 T range.

Fig. 2 shows the fit results for three representative samples of different morphology. The first one is a granular sample, the same as in Fig. 1(b), prepared by sequential deposition. The second one is a multilayer with continuous ferromagnetic layers of [1.4 nm Fe + 2 nm Ag]$_{60}$ nominal sequence. For this thickness range a RKKY-type magnetic coupling is expected between the Fe layers [18]. The last one is a trilayer sample, the same as in Fig. 1(a), containing one magnetic layer. For the earlier three samples $m = 19$, 7, and $11\mu_B$ cluster moments are obtained, respectively. The magnetic moment of Fe atoms belonging to different size Fe clusters in the Ag matrix [19] is about $3\mu_B$. According to the earlier analysis the magnetoresistance is determined by clusters containing a few (3–7) Fe atoms. We found that for all the samples the Langevin term is the dominant one ($A_1 = 5.3 \times 10^{-1}$, 1.3×10^{-1} and 4.4×10^{-2}, respectively). The AMR shift is zero for the granular sample, however it is non-negligible for the multilayer and the trilayer ($A_3 = -0.01$ and -0.003, respectively) due to the continuous ferromagnetic layers. The term proportional to H^2 is

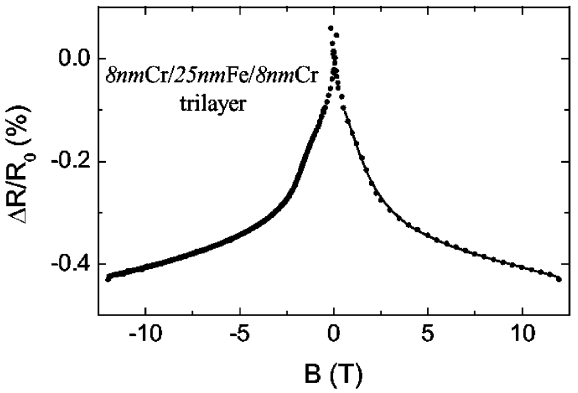

Fig. 3. Transversal magnetoresistance of a 8 nm Cr/25 nm Fe/8 nm Cr trilayer measured at 4.2 K. The fitted curve corresponds to Eq. (3) in the $|B| > 0.25$ T range. For $B > 0$ T the measured points are rarefied for clarity.

relatively small in the granular sample ($A_2 = 5 \times 10^{-5} T^2$), however it is non-negligible as compared to A_1 for the multilayer and the trilayer ($A_2 = 6 \times 10^{-5} T^2$ and $5 \times 10^{-6} T^2$, respectively). The earlier A_2 amplitudes can be associated with Fe impurities in the non-magnetic matrix in the order of a few hundred ppm [17]. Application of a second Langevin function instead of the term proportional to H^2 results in an equally good fit of the high field magnetoresistance, however it provides unphysically small (below $1\mu_B$) cluster moments.

According to the earlier analysis the unusual high field magnetoresistance of the trilayer sample is attributed to mixing of the Fe and Ag atoms at the interface and the formation of a granular-like interface alloy. Fe and Ag are immiscible at equilibrium but the substantially smaller surface free energy of silver makes an Ag covered surface energetically favorable. It has been shown that this acts as a driving force for Ag diffusion through ultra thin Fe layers, either during sample deposition [20] on substrates at or above room temperature or during a heat treatment [21] at low temperature (200–300 °C). However, if interface mixing can occur in case of immiscible elements it is even more likely for constituents with a limited solubility and the question can be put forward, if an interface magnetoresistance is to be observed, as well. To answer this question the Fe–Cr system was studied. The phase diagram of the Fe–Cr system shows solubility above 1094 K in the entire concentration range and at room temperature the solubility limit is a few at.% on each side.

Magnetoresistance measured on the 8 nm Cr/25 nm Fe/8 nm Cr trilayer is shown in Fig. 3. The granular type magnetoresistance can also be observed and could also be fitted according to Eq. (3) ($m = 9\mu_B$, $A_2 = 3 \times 10^{-6} T^2$ and $A_3 = -0.001$) as shown in Fig. 3. Non-equilibrium alloying at the interface can also be an adequate explanation, since similar behavior was observed in supersaturated bulk Fe–Cr alloys [7] and in co-sputtered alloy films [22].

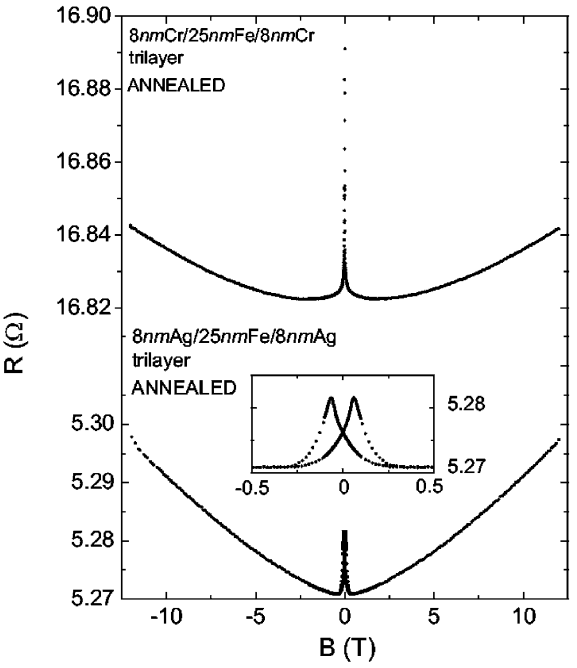

Fig. 4. Magnetoresistance measured $T = 4.2$ K on the 8 nm Cr/25 nm Fe/8 nm Cr and the 8 nm Ag/25 nm Fe/8 nm Ag trilayers after a heat treatment in vacuum at 500 °C. The insert shows the low field behavior for the 8 nm Ag/25 nm Fe/8 nm Ag trilayer.

In case of the relatively large layer thickness of our Ag/Fe/Ag trilayer a demixing of the interface alloy can be achieved by annealing without basically destroying the layered geometry. Magnetoresistance of the sample heat treated in vacuum at 500 °C for 1 h is shown in Fig. 4. The unusual high field magnetoresistance cannot be observed after the heat treatment but the resistance increases as $[R(H) - R(0)]/R(0) \propto H^{1.5}$ in accordance with former results on Fe [23]. The equality of $R_{\parallel}$ and $R_{\perp}$ has already made evident that the negative and non-saturating high field magnetoresistance is not a thin film effect. The disappearance of the high field anomaly after the heat treatment further supports the idea that it arises from the granular nature of the interface in the as-deposited sample. As it can be seen in Fig. 4 the same heat treatment removes the high field anomaly of the Cr/Fe/Cr trilayer, as well. This is in accordance with the results on bulk supersaturated alloys [7] where the recovery of the usual field dependence of the magnetoresistance was attributed to the precipitation of large Fe clusters.

On the basis of the interface magnetoresistance observed in trilayers it is tempting to estimate the magnitude of this effect in multilayers. If the mean free path of the electrons is less than the layer thickness, a parallel resistor network can approximate the interface magnetoresistance of a multilayer. Supposing that the interface conductivity is negligible in zero field but it gives a significant contribution at the highest field applied, one obtains a value around 7.7 and 5.4 kOhm for the high field resistance of a single interface in the trilayer and the $[1.4\text{ nm Fe} + 2\text{ nm Ag}]_{60}$ multilayer, respectively. The order of magnitude agreement in this simplified model and the good fit of the field dependence according to Eq. (3) suggest that the magnetoresistance of Fe–Ag multilayers mainly arise from the interface mixing. Rutherford backscattering spectrometry could also detect interface roughness [24] and will be used to characterize the chemical mixing in a more detailed study.

In conclusion the non-equilibrium structure of the interface has been demonstrated to be the source of a granular-type magnetoresistance behavior by studying Ag/Fe/Ag and Cr/Fe/Cr trilayers with a thick and continuous ferromagnetic layer. The granular interface magnetoresistance is explained by the formation of small magnetic clusters and impurities through chemical mixing at the interface during sample deposition. We believe that the investigation of N/M/N trilayers can largely contribute to a better understanding of the GMR phenomena in multilayers by elucidating the role of non-periodic structural features in the spin dependent scattering. We have shown that the granular interface magnetoresistance is dominant in polycrystalline Fe/Ag multilayers by comparing samples with different morphology. The similar shape of the magnetoresistance curves for the N/M/N trilayer, the multilayer and the granular samples indicates the possibility of a common explanation. We suggest that very small clusters and single impurities play a decisive role in case of the Fe–Ag system. The observed magnetoresistance in the Cr/Fe/Cr trilayer hints to a non-negligible granular interface contribution in Fe/Cr multilayers, as well. To establish how the magnetoresistance of N/M/N trilayers depends on the layer thickness and on the different parameters of the deposition technique needs further studies.

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