Giant Magnetoresistance at the Interface of Iron Thin Films

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Ag/Fe/Ag and Cr/Fe/Cr trilayers with a single 25 nm thick ferromagnetic layer exhibit giant magnetoresistance (GMR) type behavior. The resistance decreases for parallel and transversal magnetic field alignments with a Langevin-type magnetic field dependence up to \( B = 12 \) T. The phenomenon is explained by a granular interface structure. Results on Fe/Ag multilayers are also interpreted in terms of a granular interface magnetoresistance.

75.70.Pa, 75.70.Cn

Magnetoresistance arising from a nanoscale magnetic structure was first discovered in antiferromagnetically coupled multilayers prepared by molecular beam epitaxy (MBE)\(^ 1\). 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\(^ 2\) is more important. This means that \( [R(H) - R(0)]/R(0) \) is negative irrespectively 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 inhomogeneities 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 case of multilayers. Many recent experimental\(^ 9,10\) and theoretical\(^ 11,12\) works addressed this question, but the role of the interface was examined only in conjunction with the multilayer structure.

In this letter we show that by studying the magnetoresistance of trilayers containing only one magnetic layer it is possible to separate the contribution of a single interface. The interface magnetoresistance has GMR characteristics and is attributed to a granular interface structure. The existence of a granular interface magnetoresistance raises the question how this term is related to the magnetoresistance arising from interlayer coupling in case of 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 on Si single crystal substrate at room temperature by vacuum evaporation in a base pressure of \( 10^{-7} \) Pa. The layer thickness was controlled by a quartz oscillator 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 above geometries are usually called parallel (\( R_\parallel \)), transversal (\( R_\perp \)) and perpendicular (\( R_{\text{perp}} \)), 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 magnetic properties (saturation magnetization, Curie temperature, demagnetization field) similar to bulk layers.

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. 1a. \( R_\parallel \) 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\(^ 12\) indicating a crossover in the signe 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
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. 1b. 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 [13]. 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 [14]. 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 [15] there are only minor differences between \( R_{||} \) and \( R_{\perp} \) and \( R_{\text{perp}} \). The layered growth seems to strongly affect the sample morphology and probably the shape of the granules. Note that \( R_{\text{perp}} \) of the trilayer sample also shows distinct behavior (see Fig. 1a).

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 [3,4] as:

\[
\frac{R(H) - R(0)}{R(0)} = -A \left( \frac{M}{M_s} \right)^2,
\]

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

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

where \( L(x) = \text{cth}(x) - 1/x \) and \( m \) is the magnetic moment of the superparamagnetic particles. Equation (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 nonmagnetic matrix a term proportional to \( H^2 \) was included [16]. 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. (2) in the \( B > 0.25 \) T range.

Figure 2 shows the fit results for three representative samples of different morphology. The first one is a granular sample, the same as in Fig. 1b, 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 [17]. The last one is a trilayer sample, the same as in Fig. 1a, containing one magnetic layer.

**FIG. 1.** Magnetoresistance measured at \( T = 4.2 \) K with magnetic field aligned parallel (||), transversal (\( \perp \)) and perpendicular (\( \text{perp} \)) 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).

**FIG. 2.** Magnetoresistance curves measured with transversal magnetic field alignment at 4.2 K and fitted according to Eq. (2) 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.
For the above three samples $m = 19, 7, \text{ and } 11 \mu_B$ cluster moments are obtained, respectively. The magnetic moment of Fe atoms belonging to different size Fe clusters in the Ag matrix is about $3 \mu_B$. According to the above 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 ($A_1 = 5.3 \times 10^{-1}, 1.3 \times 10^{-1}$ and $4.4 \times 10^{-2}$, respectively). The AMR shift is zero for the granular sample, however it is non-negligible as compared to the multilayer and the trilayer ($A_3 = -0.01$ and 0.003, respectively) due to the continuos ferromagnetic layers. The quadratic term is relatively small in the granular sample ($A_2 = 5 \times 10^{-5}$), however it is non-negligible as compared to $A_1$ for the multilayer and the trilayer ($A_2 = 6 \times 10^{-5}$ and $5 \times 10^{-6}$, respectively). The above $A_2$ amplitudes can be associated with Fe impurities in the nonmagnetic matrix in the order of a few hundred ppm.

According to the above 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 favourable. It has been shown that this acts as a driving force for Ag diffusion through ultra thin Fe layers, either during sample deposition on substrates at or above room temperature or during a heat treatment at low temperature ($200-300^\circ\text{C}$). 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 8nm Cr/25nm Fe/8nm Cr trilayer is shown in Fig. 3.

The granular type magnetoresistance can also be observed and could also be fitted according to Eq. 4 ($m = 9 \mu_B$, $A_2 = 3 \times 10^{-6}$ 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 and in co-sputtered alloy films.

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$^\circ\text{C}$ for one hour 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. The equality of $R_\|\text{ and } R_\bot$ 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 where the recovery of the usual field dependence of the magnetoresistance was attributed to the precipitation of large Fe clusters.

The measured points are rarefied for clarity.
tude 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 kOhm and 5.4 kOhm for the high field resistance a single interface in the trilayer and the $[1.4nm \text{Fe} + 2nm \text{Ag}]_{60}$ 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.

In conclusion it is demonstrated for the first time that the non-equilibrium structure of the interface in magnetic multilayers is the source of a granular-type GMR behavior, which gives a contribution to the magnetoresistance independently from the nature of coupling between the layers. Separation of this interface magnetoresistance was possible by studying a single magnetic layer sandwiched between nonmagnetic layers. To establish how the interface structure depends on the layer thickness and on the different parameters of the deposition technique need further studies. However, the granular interface magnetoresistance was shown to be dominant in polycrystalline Fe/Ag multilayers. The observed magnitude of this effect in Cr/Fe/Cr trilayer indicates that the granular interface magnetoresistance can be non-negligible in Fe/Cr multilayers, as well.

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