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Influence of Grain Size on the Transport Properties of $\text{Ni}_{80}\text{Fe}_{20}$ and Cu Thin Films

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Abstract—The conductivity of sputtered Ta/Cu/Ta and Ta/ $\text{Ni}_{80}\text{Fe}_{20}$ /Ta thin films was measured for a film thickness ranging from 20 to 1500 Å. The measured data were analyzed using a semi-classical model for the electron transport, that includes grain boundary scattering. It was found that in these films grain boundaries are an important source of electron scattering, in $\text{Ni}_{80}\text{Fe}_{20}$ leading to an effective spin dependence of the scattering which is considerably smaller than the intrinsic spin dependence of the scattering.

I. INTRODUCTION

In order to increase the understanding of the magneto-transport properties of exchange biased $\text{Ni}_{80}\text{Fe}_{20}$ /Cu/ $\text{Ni}_{80}\text{Fe}_{20}$ /Fe₅₀Mn₅₀ spin valve multilayers, which are commonly grown on a Ta buffer layer, the transport properties of polycrystalline Ta/Cu/Ta and Ta/ $\text{Ni}_{80}\text{Fe}_{20}$ /Ta thin films have been studied. Mayadas *et al.* [1] and many other authors [2] have demonstrated already that two size effects have to be taken into account to explain the thickness dependence of the resistivity in polycrystalline films, i.e. film boundary and grain boundary scattering. Resistivity data are generally analyzed in terms of the Fuchs-Sondheimer theory [3,4], combined with a model for grain boundary scattering proposed by Mayadas and Shatzkes [5]. In this paper, using a similar approach, we will show that the average grain size in Cu and $\text{Ni}_{80}\text{Fe}_{20}$ thin films, grown on a Ta buffer layer, is significantly smaller than the intrinsic mean free path of the electrons in Cu and $\text{Ni}_{80}\text{Fe}_{20}$. As the grain size is found to be considerably smaller than the film thickness, grain boundaries are an important source of electron scattering, even up to large film thickness. In $\text{Ni}_{80}\text{Fe}_{20}$ films this leads to a considerable reduction of the effective spin dependence of the scattering.

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II. EXPERIMENTAL METHOD

The films under investigation had the following structure:
30 Å Ta/ t_x X/30 Å Ta, where X = Cu, $\text{Ni}_{80}\text{Fe}_{20}$
and $t_{\text{Cu}} = 50\text{--}1500$ Å, $t_{\text{NiFe}} = 20\text{--}1230$ Å.

The films were prepared by dc magnetron sputtering with a base pressure of typically 10^{-8} Torr, on 4×12 mm² Si(100) substrates precleaned by a 2% HF dip. The argon pressure during sputtering was 5.0 mTorr, the deposition rates were about 2 Å/s. The Ta buffer layer was used to improve the (111) texture of the films. The full width at half maximum of the x-ray diffraction rocking curve, measured on a film of 400 Å, is 3.7° and 5.2° for $\text{Ni}_{80}\text{Fe}_{20}$ and Cu, respectively. The grain size in these polycrystalline films was determined by plan view transmission electron microscopy (TEM) and grazing incidence X-ray diffraction.

The sheet conductance for varying film thickness (Fig. 1), measured in a four probe setup, was analyzed using a semi-classical model similar to the Fuchs-Sondheimer theory, which involves the solution of the Boltzman transport equation in linear response for a free electron gas in the relaxation time approximation. In order to treat the anisotropic magnetoresistance effect in $\text{Ni}_{80}\text{Fe}_{20}$, the mean free path of the electron was taken to depend on the angle between its velocity vector and the magnetization vector [6]. In this model diffusive electron scattering at the film boundaries is assumed. Grain boundary scattering was taken into account in the following way: the mean free path λ , which is the only adjustable model parameter, is given by

$$\frac{1}{\lambda} = \frac{1}{\lambda_i} + \frac{1}{\lambda_{gr}} \quad (1)$$

where λ_i and λ_{gr} are the mean free paths for intrinsic scattering and grain boundary-scattering, respectively; λ_i and therefore also λ are spin dependent in $\text{Ni}_{80}\text{Fe}_{20}$, λ_{gr} is assumed to be spin independent. Eq. (1) is valid when λ_i is considerably larger than the grain size.

Fig. 1: Sheet conductance versus film thickness at 4.2 K for Cu (squares) and $\text{Ni}_{80}\text{Fe}_{20}$ (circles) thin films. The open and closed circles represent different sample series.

This is a similar but somewhat more general approach, as compared to the Mayadas-Shatzkes model [5], which describes grain boundary scattering in terms of specular reflection on a Gaussian distributed set of randomly spaced planes perpendicular to the current direction.

III. RESULTS AND DISCUSSION

In the Cu films, with film thickness t , $\lambda_{gr}(t)$ was determined from modelling the sheet conductance measured at 4.2 K, neglecting at this temperature the intrinsic scattering ($\lambda_i \rightarrow \infty$) and taking an intermixed layer of thickness t_{int} at the Ta/Cu interfaces into account. λ_{gr} as a function of film thickness is shown in Fig. 2. In order to determine a consistent value of t_{int} , $\lambda_{gr}(t)$ was implemented in the modelling of the room temperature conductivities of the same films, using Eq. (1), which yields a finite value of λ_i for each film. This procedure was repeated while varying t_{int} until λ_i , which is a bulk parameter, was found to be independent of film thickness. For an intermixed layer thickness of 10 Å, λ_i was found to be about 330 Å. This is reasonably consistent with the value of 395 Å, as derived from the room temperature resistivity of Cu within the free electron model [7], which confirms the applicability of our model. The λ in the intermixed layer was determined by analyzing the sheet conductance of a nominally 30 Å Ta/ t_{int} Cu/30 Å Ta film, assuming that this corresponds to $(30 \text{ Å} - \frac{1}{2}t_{int}) \text{ Ta}/2t_{int} \text{ Ta-Cu}/(30 \text{ Å} - \frac{1}{2}t_{int}) \text{ Ta}$ due to intermixing. For $t_{int} = 10 \text{ Å}$, a λ for Ta-Cu of 10 Å at 4.2 K and 2 Å at room temperature was found. In the Ta layers a λ of 3 Å at 4.2 K and 11 Å at room temperature was used, determined from a separate series of Ta single films with varying film thickness. The negative temperature coefficient

Fig. 2: λ_{gr} versus film thickness at 4.2 K for Cu thin films. The inset shows the ratio of λ_{gr} to the measured average grain size D versus film thickness. The lines are guides to the eye.

of the resistivity of Ta is characteristic for β -Ta. It must be remarked that in the model calculations, presented in this paper, the Fermi velocity of Cu is applied to all the metals.

In addition, the average grain size D was measured by plan view TEM and grazing incidence X-ray diffraction. With both characterization techniques the presence of a columnar grain structure was confirmed. The thickness dependence of the grain size is in accordance with the expected type of film growth [8]. The ratio $\lambda_{gr}(t)/D$, which is shown in the inset of Fig. 2, was found to be essentially constant over the entire thickness range. This confirms that the electron scattering described by λ_{gr} is related to the grain structure of the films.

For $\text{Ni}_{80}\text{Fe}_{20}$, Eq. (1) was applied to each spin direction separately. Using $(\lambda_i' + \lambda_i'') = 361 \text{ Å}$, as derived from the low temperature resistivity of $\text{Ni}_{80}\text{Fe}_{20}$ within the free electron model [9], $\lambda_{gr}(t)$ was determined from the low temperature sheet conductance for various values of $\alpha \equiv \lambda_i'/\lambda_i''$, the intrinsic spin dependence of the scattering. The results for $\alpha = 4$, a typical value according to [12,13], are shown in Fig. 3. For the thickness range shown, $\lambda_{gr}(t)$ is smaller than $(\lambda_i' + \lambda_i'')$, clearly pointing out that grain boundary scattering cannot be neglected. As $\lambda_{gr}(t)$ proved to be only very weakly dependent on interfacial intermixing, this is neglected in the analysis of the $\text{Ni}_{80}\text{Fe}_{20}$ films. We would like to emphasize that in our analysis λ_{gr} and α are dependent parameters; additional experiments would be required to determine their separate values.

Plan view TEM measurements show the development of voids between the grains for $\text{Ni}_{80}\text{Fe}_{20}$ films thicker than about 200 Å, indicating a different film morphology as compared to Cu films [8]. This will seriously influence the conductivity, as the presence of voids between the grains

Fig. 3: λ_{gr} versus film thickness at 4.2 K for $\text{Ni}_{80}\text{Fe}_{20}$ films, assuming $\alpha = 4$ and $(\lambda_i' + \lambda_i'') = 361$ Å. The open and closed symbols represent different sample series. The inset shows the ratio of λ_{gr} to the measured average grain size D .

strongly reduces the transmission probability of electrons at the grain boundaries. The inset in Fig. 3 shows that the ratio $\lambda_{gr}(t)/D$ is essentially constant over a large thickness range. The average grain size D is estimated from TEM measurements. The relatively low value of $\lambda_{gr}(t)/D$, as compared to the Cu films, indicates a high electron scattering probability at the grain boundaries, which is consistent with the presence of voids.

As mainly the mean free path of the spin up electrons, which have the longest intrinsic mean free path, is affected by grain boundary scattering, this will lead to a considerably lower effective spin dependence of the scattering $\alpha_{eff}(t) \equiv \lambda^+/\lambda^- = \alpha \cdot [\lambda_i^+ + \lambda_{gr}(t)]/[\lambda_i^+ + \lambda_{gr}(t)]$, as compared to the intrinsic spin dependence of the scattering α . This is clearly illustrated in Fig. 4, which shows $\alpha_{eff}(t)$ for $\alpha = 4, 10$, and 20 , using $(\lambda_i^+ + \lambda_i'') = 361$ Å and the values of $\lambda_{gr}(t)$, as discussed above. In particular for high values of α , α_{eff} is much smaller than α .

For comparison, values of 2.7 [10] and 11 [11] are reported for the α of $\text{Ni}_{80}\text{Fe}_{20}$ (at 4.2 K), derived from modelling the current-in-plane magnetoresistance in sputtered $\text{Ni}_{80}\text{Fe}_{20}/\text{Cu}$ multilayers. In the first case, contrary to the second, no grain boundary scattering was taken into account, resulting in a lower spin dependence of the scattering, i.e. α_{eff} . From measurements of the current-perpendicular-to-plane magnetoresistance, α values between 3 and 5 were derived [12,13]. In the CPP case a more accurate analysis of the different contributions to the total conductivity is possible as the current is forced to cross the interfaces.

In conclusion, for $\text{Ni}_{80}\text{Fe}_{20}$ and Cu films, grown on a Ta buffer layer, it was found that grain boundaries are an important source of electron scattering. In $\text{Ni}_{80}\text{Fe}_{20}$ films

Fig.4: Effective spin dependence of the scattering α_{eff} for $\alpha = 4$ (squares), 10 (triangles), and 20 (circles). The open and closed symbols represent different sample series.

(spin independent) grain boundary scattering leads to a considerable reduction of the effective spin dependence of the scattering. In $\text{Ni}_{80}\text{Fe}_{20}$ based spin valve multilayers, which are commonly grown on a Ta buffer layer, grain boundary scattering will reduce the giant magnetoresistance.

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