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Temperature dependence of the resistivity and tunneling magnetoresistance of sputtered FeHf(Si)O cermet films

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We have studied the tunneling resistivity and magnetoresistance of reactive sputter deposited FeHfO and FeHfSiO thin granular films. Maximum magnetoresistance ratios at room temperature of 2% and 3.2% were observed for films with compositions of Fe47Hf10O43 and Fe40Hf6Si6O48, respectively. The magnetoresistance shows a decrease with temperature, which cannot be explained by spin-dependent tunneling only. We propose that spin-flip scattering in the amorphous FeHf(Si)O matrix causes this decrease as function of temperature. A two current model for the tunnel magnetoresistance, taking into account spin-flip scattering, is presented which can describe the observed temperature dependence of the magnetoresistance. © 1998 American Institute of Physics.

I. INTRODUCTION

Recently there has been a great deal of interest in the magnetoresistance (MR) effect in materials which combine ferromagnetic metals and insulators. The observed MR effect in these materials is often denoted as tunnel magnetoresistance (TMR) or junction magnetoresistance (JMR) because it is ascribed to the spin-dependent tunneling of electrons between two ferromagnetic materials across an insulating barrier.

This TMR effect can be found in layered structures in which a current flows from one ferromagnetic layer (e.g., Co, CoFe) across an insulating layer (often Al2O3) to another ferromagnetic layer in a so-called tunnel junction. The preparation of these junctions is, however, rather difficult as it requires lithographic techniques or shadow evaporation with the help of masks. TMR can also be observed in a different class of materials, the so-called cermet films. These are composed of metallic magnetic grains embedded in an insulating matrix, in which the probability for electrons to tunnel from one grain to another depends on the relative orientation of the magnetic moments of the grains. Several granular systems have been studied, for example CoSiO2, FeSiO2, CoAl2O3, FeHfO, and FeSiO2. Among these materials there is particular interest in FeHfO not only for its magnetoresistance but also for its soft magnetic properties.

In this article we report on the observed TMR in reactive sputter deposited FeHfO and FeHfSiO thin films. We will focus on the temperature dependence of the TMR and resistivity which has not been studied in great detail so far and this yields additional insight into the mechanisms for spin-dependent conductivity. The unusual temperature dependence of the TMR of our films is in contrast with earlier studies of, for example, CoAl2O3 (Ref. 8) and FeSiO2 (Ref. 5) and cannot be explained with spin-dependent tunneling only. We propose a model for the temperature dependence of the magnetoresistance in which we have included spin-flip scattering.

II. EXPERIMENT

All films were prepared by rf diode sputtering on a Perkin–Elmer 2400 machine, with a base pressure of about 4×10−7 mTorr, at a sputter pressure of 3–4 mTorr from a Fe83Hf17 target. The composition of the films was varied by changing the partial O2 pressure of the Ar/O2 flow. For the sputtering of the FeHfSiO films 4% of the target was covered with Si pellets. The films are sputtered on glass and the thickness of the FeHfO and FeHfSiO films is 2.1 and 0.7 μm, respectively. The composition of the materials was determined with electron probe microanalysis (EPMA). Details on the microstructure of these films will be published in a separate article. Resistivity and TMR measurements were done in a standard four point contact geometry with current and field in the plane of the films.

III. RESULTS AND DISCUSSION

Figure 1 shows the MR ratio as a function of the applied field at room temperature of a Fe47Hf10O43 film. The MR is measured with current either perpendicular or parallel with respect to the applied field. As can be seen, there is almost no...
difference between the two geometries, which shows the absence of a significant contribution of an anisotropic MR effect. The MR curves of this film are typical for all of our FeHfO and FeHfSiO cermet materials.

We have prepared a series of FeHfO and FeHfSiO films in which we have varied the partial O\textsubscript{2} pressure during the sputter process. This allows us to fabricate films with different Fe concentrations. Figure 2 shows the Fe concentration dependence of the TMR and the resistivity at room temperature. The resistivity is extremely large and increases strongly with decreasing Fe concentration. This can be understood as follows. For a large Fe concentration a metallic conductance arises because part of the Fe grains are connected. For lower Fe content the percolation concentration will be approached\textsuperscript{14–16} and the grains become electrically isolated by an insulating amorphous FeHf\textsubscript{2}Si\textsubscript{12}O\textsubscript{42} phase resulting in tunnel-type conductivity with orders of magnitude larger resistivities as compared to metallic iron ($\rho_{Fe} = 10 \mu\Omega \text{ cm}$).

Note the difference in scale between the resistivity of FeHfO and the FeHfSiO films. For Fe concentrations below 45\% for FeHfO and below 40\% for FeHfSiO the layers become fully oxidized and insulating. In both cases the TMR at room temperature peaks near the percolation limit where a small barrier between the grains can be expected. The TMR for the FeHfSiO films has a maximum of about 3.2\% for the composition Fe\textsubscript{40}Hf\textsubscript{2}Si\textsubscript{12}O\textsubscript{42}. For the FeHfO films the maximum TMR of approximately 2\% appears at higher Fe concentration for the film with composition Fe\textsubscript{45}Hf\textsubscript{10}O\textsubscript{43}.

Next we present the temperature dependence of the resistivity and the TMR ratio of the FeHfO and FeHfSiO films. We will focus on the films around maximum TMR because they are believed to have an optimal grain size and separation between the grains with respect to the observation of spin-dependent tunneling. X-ray diffraction with Cu K\textsubscript{α} radiation for these films, presented in Fig. 3, display clear Fe peaks from the grains, a large amorphous background, and a peak around 30°, which possibly results from a combination of HfO\textsubscript{2}, FeO\textsubscript{3}, and SiO\textsubscript{2} crystalline phases.

The temperature dependence of the resistivity of several FeHfO and FeHfSiO films, around maximum TMR, is presented in Fig. 4, plotted as $\log \rho$ vs $T^{-1/2}$. This proportionality should represent a thermally activated tunneling current flowing from grain to grain through an insulating matrix as calculated by Sheng et al.\textsuperscript{14–16} viz. $\rho$ = $\rho_0 \exp \left(2 \alpha(1/\kappa B T)\right)$, which represents a thermally activated tunneling current flowing from grain to grain through an insulating matrix.

FIG. 1. MR ratio as a function of the applied field at room temperature for Fe\textsubscript{47}Hf\textsubscript{10}O\textsubscript{43} with current perpendicular (a) and parallel (b) with respect to the applied field. The MR ratio is defined as $[\rho - \rho(B = 1.3 \text{ T})]/\rho(B = 1.3 \text{ T}) \times 100\%$.

FIG. 2. (a) TMR and (b) resistivity $\rho$ at room temperature as a function of the Fe concentration for FeHfO and FeHfSiO. The solid lines are guides for the eye only.

FIG. 3. X-ray diffraction θ-2θ scan with Cu K\textsubscript{α} radiation for (a) Fe\textsubscript{45}Hf\textsubscript{10}O\textsubscript{43} and (b) Fe\textsubscript{40}Hf\textsubscript{2}Si\textsubscript{12}O\textsubscript{42}. The patterns display clear Fe peaks from the grains, a large amorphous background, and a peak around 30°, which possibly results from a combination of HfO\textsubscript{2}, FeO\textsubscript{3}, and SiO\textsubscript{2} crystalline phases.

FIG. 4. Temperature dependence of the resistivity, log $\rho$ vs $T^{-1/2}$, for several (a) FeHfO and (b) FeHfSiO films with compositions around maximum TMR. The solid lines are fits to the experimental data according to $\rho = \rho_0 \exp \left(2 \alpha(1/\kappa B T)\right)$, which represents a thermally activated tunneling current flowing from grain to grain through an insulating matrix.
\[ \text{for FeHfSiO are rather good} \]
\[ \text{Fe}_{40} \text{Hf}_6 \text{Si}_6 \text{O}_{48} \]
\[ \text{barrier height}, \]"s
\[ \text{data for Fe}_{47} \text{Hf}_{10} \text{O}_{43}. \]
\[ \text{small values of} \]"s
\[ \text{compared to, for example, CoAl}_2 \text{O}_3 \]
\[ \text{is independent of temperature, more or less consistent with} \]
\[ \text{down, respectively. Within this model the magnetoresistance} \]
\[ \text{cable to our films since a leak conductivity leads to zero} \]
\[ \text{grains. As we will show later on, this seems not to be appli-} \]
\[ \text{to a leak conductivity which flows through interconnected} \]
\[ \text{ berry deviation was observed for FeSiO}_2 \text{ and this was attributed} \]
\[ \text{ature dependent TMR may still be observed in these materi-} \]
\[ \text{magnetically clusters in an insulating matrix is equal to} \]
\[ \text{andr deviation was observed for FeSiO}_2 \text{ and this was attributed} \]
\[ \text{tation in the ferromagnetic material. Here,} \]
\[ \text{therefore a deviation from this} \]
\[ \text{C} \text{ of temperature of} \text{Fe}_{47} \text{Hf}_{10} \text{O}_{43}. \]
\[ \text{ratio of} \text{Fe}_{47} \text{Hf}_{10} \text{O}_{43} \text{ increases strongly} \]
\[ \text{this suggests that the individual grains are only poorly sepa-} \]
\[ \text{in the ferromagnetic material. Here,} \]
\[ \text{deviation between calculation and experimentally observed} \]
\[ \text{Figure 5. Temperature dependence of the TMR for} \]
\[ \text{Fe}_{47} \text{Hf}_{10} \text{O}_{43} \text{ and} \]
\[ \text{Fe}_{39} \text{Hf}_7 \text{Si}_5 \text{O}_{49} \text{ which increases only slightly. Inoue} \]
\[ \text{First of all, a reduction of the polarization} \]
\[ \text{temperature dependent TMR may still be observed in these materi-} \]
\[ \text{magnetically clusters in an insulating matrix is equal to} \]
\[ \text{P} \text{ at higher} \]
\[ \text{therefore a deviation from this} \]
\[ \text{m} \text{ is the electron mass,} \]
\[ \text{and the effective barrier} \]
\[ \text{and} \]
\[ \text{activation energy} \]
\[ \text{C} \text{ } \text{is the electron mass,} \]
\[ \text{a condition} \]
\[ \text{the variable} \]
\[ \text{dependence of the TMR is observed. Moreover, Fig. 7 shows} \]
\[ \text{large spread in grain sizes, with a blocking temperature} \]
\[ \text{between 200 and 300 K. Superparamagnetic behavior is lim-} \]
\[ \text{width saturated at 1.3 T, the maximum field applied in the} \]
\[ \text{or} \]
\[ \text{transport measurements, which ensures a good parallel align-} \]
ment of the grains at all temperatures. On the other hand the remanent magnetization is close to zero which guarantees a high degree of antiparallel alignment of the grains at zero field. It should be noted that the saturation magnetization decreases slightly with increasing temperature which may indicate a small fraction of paramagnetic grains.

It is clear from the foregoing analysis that there is no dramatic change in magnetization behavior which can account for the large decrease of TMR at higher temperatures, and we believe another mechanism plays a role, causing the temperature dependence of the magnetoresistance. We propose that the strong decrease of TMR at higher temperatures is due to spin-flip scattering which is caused, for example, by magnetic impurities or iron-rich phases in the matrix. Recently, the effect of barrier impurities in ferromagnetic tunnel junctions was investigated and it was shown that these impurities can severely reduce the TMR as a result of spin-flip scattering.19 The impact of spin-flip scattering will be discussed the temperature dependent resistance in ferromagnetic materials diluted with transition metal ions and used the resistor scheme as illustrated in Fig. 8 to account for the resistivity including spin-flip scattering. The resistance of such a circuit is given by

\[ \rho = \frac{\rho_+ \rho_- + \rho_{||} \rho_{\perp}}{\rho_+ \rho_- + 4 \rho_{||}}, \]

where \( \rho_+ \) and \( \rho_- \) denote the resistivities of the up and down electrons, respectively, and where \( \rho_{||} \) is the spin-mixing resistivity. We apply this model directly to our granular system. The resistivities \( \rho_+ \) and \( \rho_- \) now consist of the sum of the part of the grains with magnetization “up” and magnetization “down,” respectively:

\[ \rho_+ = \frac{N}{M} \rho_{||} \left( 1 - \frac{N}{M} \right) \rho_{\perp}, \]

\[ \rho_- = \frac{N}{M} \rho_{||} \left( 1 - \frac{N}{M} \right) \rho_{\perp}. \]

Here \( M \) is the total number of grains and \( N \) is the number of grains with magnetization up, \( \rho_{||} \) and \( \rho_{\perp} \) are the resistivities for the majority and minority spin electrons with respect to the local magnetization, respectively. The resistivity of Eq. (2) can now be expressed in terms of the relative magnetization \( m = (2N-M)/M \), as

\[ \rho(m) = \frac{1}{4} (\rho_+ + \rho_-) - \frac{1}{4} m^2 \frac{(\rho_+ - \rho_-)^2}{\rho_+ + \rho_- + 4 \rho_{||}}. \]

Equation (4) can be transformed to a similar equation for the resistivity as derived by Inoue et al.17 with \( \rho_{||}, \rho_\perp \approx \exp(2 \sqrt{C/k_B T}), \)

\[ \rho(m) = \rho_0 (1 - P^2 m^2 F) \exp(2 \sqrt{C/k_B T}), \]

with \( \rho_0 \) constant and \( F \) representing the spin-flip scattering term given by

\[ F = \frac{1}{1 + \frac{\rho_{||}}{\rho_0} \exp(-2 \sqrt{C/k_B T})}, \]

and \( P \) the polarization of the ferromagnetic material [in this model given by \( (\rho_{||} - \rho_\perp)/(\rho_1 + \rho_\perp) \)]. We use Eqs. (5) and (6) to describe our data of the TMR ratio, which is defined as

\[ \text{TMR} = 100\% \cdot \frac{\rho(m=0) - \rho(m=1)}{\rho(m=1)}. \]

The spin-mixing rate at finite temperature has been modeled by Fert et al.21,22 and evaluated as \( \rho_{||} = \rho_{||}^* T^n \) \( (n = 2 \) in case of electron-magnon scattering). The solid lines in Fig. 5 show that this model can describe the temperature dependence of the TMR ratio very well with \( P = 0.26, \rho_{||} = 138 \mu \Omega \text{cm K}^{-n}, \) with \( n = 1.3 \) for FeHfO, and \( P = 0.18, \rho_{||} = 38 \mu \Omega \text{cm K}^{-n}, \) with \( n = 1.3 \) for FeHfSiO \( (\rho_0 \) and \( C \) are the same as for the resistivity measurements). The magnitude of \( \rho_{||}^* \) and \( n \) are determined by the details of the spin-flip scattering mechanism, of which we don’t know the exact origin. The spin polarization \( P \) is for both systems lower than the polarization of iron \( P_{Fe} = 0.4 \) as reported by Meservey et al.,23 determined from Al/Al₂O₃/Fe junctions at low temperatures. We should not be surprised by this, since our calculations are inspired by the models of Julliere1 and Inoue et al.,17 in which the TMR is determined solely by the spin polarization of the ferromagnetic material. However, it is theoretically argued24 that the barrier material and the interface matching between barrier and magnetic material may determine the TMR effect as well, although no conclusive experimental data are available yet to verify this. Additionally, we have to realize that our granular films are far from an ideal system of pure Fe grains in an isolating Hf(Si)O₂ matrix and therefore negative effects on the magnitude of the polarization can be expected from, for example, intermixing of Fe and Hf and oxidation of Fe. Further experimental study is necessary to determine the exact composition of the grains and the matrix and this may also reveal why spin mixing is much more prominent in FeHfO than in FeHfSiO.

IV. CONCLUSIONS

In summary we have measured the TMR and the resistivity of reactive sputtered FeHfO and FeHfSiO cermet films. Maximum magnetoresistance ratios of 2% and 3.2% at room temperature are observed for films with the composition Fe₄7Hf₁₀O₄₃ and Fe₄₀Hf₆Si₆O₄₈, respectively. The resistivity and magnetoresistance show an unusual temperature dependence which cannot be explained by spin-dependent tunneling only. We propose that spin-flip scattering in the amorphous FeHf(Si)O matrix causes a decrease of the TMR as a function of temperature.
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