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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 Fe$_{47}$Hf$_{10}$O$_{43}$ and Fe$_{40}$Hf$_{6}$Si$_{6}$O$_{48}$, 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 Al$_2$O$_3$) 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 CoSiO$_2$, FeSiO$_2$, Co$_3$Al$_2$O$_4$, and FeHfO. 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, CoAl$_2$O$_3$ (Ref. 8) and FeSiO$_2$ (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 Fe$_{83}$Hf$_{17}$ target. The composition of the films was varied by changing the partial O$_2$ pressure of the Ar/O$_2$ 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 Fe$_{47}$Hf$_{10}$O$_{43}$ 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 O2 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 is approached and the grains become electrically isolated by an insulating amorphous FeHf~SiO 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 Fe40Hf6Si6O48. For the FeHfO films the maximum TMR of approximately 2% appears at higher Fe concentration for the film with composition Fe47Hf10O43.

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 Kα 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 HfO2, Fe2O3, and SiO2 crystalline phases.

![FIG. 1. MR ratio as a function of the applied field at room temperature for Fe47Hf10O43 with current perpendicular (a) and parallel (b) with respect to the applied field. The MR ratio is defined as \(\frac{[\rho - \rho(B = 1.3 \, \text{T})]/\rho(B = 1.3 \, \text{T}) \times 100\%}{\rho(B = 1.3 \, \text{T})}\).](image1)

![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.](image2)

![FIG. 3. X-ray diffraction θ-2θ scan with Cu Kα radiation for (a) Fe47Hf10O43 and (b) Fe40Hf6Si6O48. 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 HfO2, Fe2O3, and SiO2 crystalline phases.](image3)

![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 \(\rho \approx \exp \left(2.5/(k_BT)\right)\), which represents a thermally activated tunneling current flowing from grain to grain through an insulating matrix as calculated by Sheng et al.\(^{14-16}\) viz. \(\rho\).](image4)
for FeHfSiO are rather good

\[ \text{Fe}_{40} \text{Hf}_{6} \text{Si}_{6} \text{O}_{48} \]

barrier height, data for \( \text{Fe}_{47} \text{Hf}_{10} \text{O}_{43} \).

Small values of compared to, for example, \( \text{CoAl}_{2} \text{O}_{3} \)

the TMR ratio for \( \text{Fe}_{39} \text{Hf}_{7} \text{Si}_{5} \text{O}_{49} \), but in contrast with the

is independent of temperature, more or less consistent with

down, respectively. Within this model the magnetoresistance

through the interconnected grains. We also have to realize

to a leak conductivity which flows through interconnected

gains. As we will show later on, this seems not to be applica-
to our films since a leak conductivity leads to zero

grains. As we will show later on, this seems not to be appli-
table to a leak conductivity which flows through interconnected

gains. As we will show later on, this seems not to be applica-
to our films since a leak conductivity leads to zero

TMR at low temperature when all of the current is shunted

through the interconnected grains. We also have to realize

that \( \rho \propto \exp 2.1(C/k_B T) \) was derived for \( C > k_B T \), a condition

not satisfied in our films and therefore a deviation from this

proportionality may be expected at higher temperatures. The

magnitude \( C \) is to a great extent determined by the average

separation between the grains \( s \) and the effective barrier

height \( \phi \). Since the values of \( C \) obtained from our data are on the

order of \( 10^{-3} \) eV, which is one to two orders smaller as compared to, for example, \( \text{CoAl}_{2} \text{O}_{3} \) (Ref. 7) and \( \text{NiSiO}_{2} \), this suggests that the individual grains are only poorly separ-

ated by the amorphous matrix.

In Fig. 5 the temperature dependence of the TMR ratio is

presented. The TMR ratio of \( \text{Fe}_{39} \text{Hf}_{10} \text{O}_{43} \) increases strongly

with decreasing temperature from about 2% at room tem-

terature to about 7% at 4.2 K, in contrast to the TMR ratio of

\( \text{Fe}_{39} \text{Hf}_{10} \text{Si}_{5} \text{O}_{49} \) which increases only slightly. Inoue et al.\(^{17}\)
have shown that the magnetoresistance for ferromagnetic

metallic clusters in an insulating matrix is equal to \( P^2 \)

for small values of \( P \), with \( P = (N_{\uparrow} \cdot N_{\downarrow})/(N_{\uparrow} + N_{\downarrow}) \) the spin

polarization in the ferromagnetic material. Here, \( N_{\uparrow, \downarrow} \) is the
density of states at the Fermi level with spin-up and spin-
down, respectively. Within this model the magnetoresistance

is independent of temperature, more or less consistent with

the TMR ratio for \( \text{Fe}_{39} \text{Hf}_{10} \text{Si}_{5} \text{O}_{49} \), but in contrast with the
data for \( \text{Fe}_{39} \text{Hf}_{10} \text{O}_{43} \).

There are several possible explanations why a tempera-
ture dependent TMR may still be observed in these materi-

als. First of all, a reduction of the polarization \( P \) at higher

temperatures leads to a decrease of the TMR.\(^{18}\) However, we

expect this to be a small effect because of the high Curie
temperature of Fe. Moreover an almost temperature indepen-
dent TMR was already observed for \( \text{FeSiO}_{2} \).\(^{5}\) Second, the temperature dependence of the TMR might be related to a

change in magnetization behavior of the grains at low tem-

perature. It is known that a superparamagnetic behavior of

the magnetic grains in these material leads to a \( 1/T^2 \) depen-
dence of the TMR.\(^{17}\) Therefore we have measured the field-
cooled and zero-field-cooled magnetic moment of

\( \text{Fe}_{39} \text{Hf}_{10} \text{O}_{43} \) as a function of temperature as shown in Fig. 6.

A wide peak in the zero-field-cooled measurement indicates

a large spread in grain sizes, with a blocking temperature

between 200 and 300 K. Superparamagnetic behavior is lim-

ted to temperatures above the blocking temperature, which

is well above the regime where the strongest temperature

dependence of the TMR is observed. Moreover, Fig. 7 shows

four magnetization loops measured at \( T = 10, 75, 150, \) and

300 K of \( \text{Fe}_{37} \text{Hf}_{10} \text{O}_{43} \) with the applied field in the plane of

the layers. For all temperatures the magnetization loops are

similar in shape and seem to behave like a ferromagnetic

layer, with out of plane loops (not shown) that saturate at

\( H = M_s \). For the interpretation of the magnetoresistance data

it is important to mention that the magnetization in plane is

well saturated at 1.3 T, the maximum field applied in the

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 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 analyzed via a simple model calculation. Fert \(\text{et al.}\)\(^{(20,21)}\) described 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

\[
\rho = \frac{\rho_+ \rho_- + \rho_{\uparrow\downarrow} (\rho_+ + \rho_-)}{\rho_+ + \rho_- + 4 \rho_{\uparrow\downarrow}},
\]

where \(\rho_+\) and \(\rho_-\) denote the resistivities of the up and down electrons, respectively, and where \(\rho_{\uparrow\downarrow}\) 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:

\[
\begin{align*}
\rho_+ & = \frac{N}{M} \rho_1 + \left(1 - \frac{N}{M}\right) \rho_1, \\
\rho_- & = \frac{N}{M} \rho_1 + \left(1 - \frac{N}{M}\right) \rho_1.
\end{align*}
\]

Here \(M\) is the total number of grains and \(N\) is the number of grains with magnetization up, \(\rho_1\) and \(\rho_1\) 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

\[
\rho(m) = \frac{1}{4} (\rho_+ \rho_-) - \frac{1}{4} m^2 (\rho_1 - \rho_1)^2 (\rho_1 + \rho_1 + 4 \rho_{\uparrow\downarrow}).
\]

Equation (4) can be transformed to a similar equation for the resistivity as derived by Inoue \(\text{et al.}\)\(^{(17)}\) with \(\rho_1\), \(\rho_1\) \(\propto \exp(2(C/k_B T))\), yielding:

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

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

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

and \(P\) the polarization of the ferromagnetic material [in this model given by \((\rho_1 - \rho_1)/(\rho_1 + \rho_1))\]. 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 \(\text{et al.}\)\(^{(21,22)}\) and evaluated as \(\rho_{\uparrow\downarrow} = \rho_{\uparrow\downarrow}^* T^n\) (\(n = 2\) 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 \(P = 0.26, \rho_{\uparrow\downarrow}^* = 138 \mu \Omega \text{ cm K}^{-n}\), with \(n = 1.3\) for FeHfO, and \(P = 0.18, \rho_{\uparrow\downarrow}^* = 38 \mu \Omega \text{ cm K}^{-n}\), with \(n = 1.3\) for FeHfSiO (\(\rho_0\) and \(C\) are the same as for the resistivity measurements). The magnitude of \(\rho_{\uparrow\downarrow}^*\) and \(n\) are determined by the details of the spin-flip scattering mechanism, of which we can’t know the exact origin. The spin polarization \(P\) is for both systems lower than the polarization of iron \(P_F = 0.4\) as reported by Meservey \(\text{et al.}\),\(^{(23)}\) determined from AlAl\(_2\)O\(_3\)/Fe alloys at low temperatures. We should not be surprised by this, since our calculations are inspired by the models of Julliere\(^{(1)}\) and Inoue \(\text{et al.}\)\(^{(17)}\) in which the TMR is determined solely by the spin polarization of the ferromagnetic material. However, it is theoretically argued\(^{(24)}\) 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)O\(_2\) 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 Fe\(_47\)Hf\(_{10}\)O\(_{43}\) and Fe\(_{40}\)Hf\(_{40}\)Si\(_6\)O\(_{48}\), 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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