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On the frequency dependence of the magnetic permeability of FeHfO thin films

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The frequency dependence of the magnetic permeability as well as of the electrical impedance have been investigated for soft-magnetic granular FeHfO thin films. The impedance measurements indicate that capacitive effects resulting from the inhomogeneous structure of the layers are of no importance for the roll off of the permeability of the present films. The frequency behavior at various FeHfO thicknesses shows that eddy current effects start to play a role above thicknesses of 10 μm. Below this thickness ferromagnetic resonance dominates the roll off. © *1998 American Institute of Physics.* [S0021-8979(98)02324-X]

I. INTRODUCTION

Currently a considerable amount of attention is directed at the investigation of soft-magnetic oxidic/metallic granular thin films. Examples are CoAlO, CoSiO,[1] FeHfO,[2] and other Fe–M–O systems in which M is mostly a transition metal from the group IVa or Va or a rare earth element, see, e.g., Refs. 3 and 4. The origin for this interest lies in the remarkable combination of a high magnetic permeability and a relatively high electrical resistivity. The resistivities of the oxide based systems are commonly one to two orders of magnitude larger than of, for instance, the well-known soft-magnetic nitride based Fe–M–N systems.

The reason for this difference in resistivity is believed to originate in specific differences in the microstructure. The Fe–M–N are accepted to exhibit a microstructure consisting of nanometer-sized Fe crystallites with a M–N phase on the triple points. Here, the Fe crystallites are in contact with each other (no nonmagnetic grain boundary) enabling the direct exchange interaction to average out the large α-Fe magnetocrystalline anisotropy thus yielding the soft-magnetic properties.[5] This direct contact is inevitably accompanied by electrically percolating paths through the sample resulting in relatively low electrical resistivities. On the other hand, for the Fe–M–O systems the microstructure is believed to consist of again nanometer-sized α-Fe crystallites now fully embedded in an amorphous high resistive Fe–M–O phase. Percolation between the Fe crystallites is no longer possible and the resistivity is dominated by the amorphous Fe–M–O phase. This phase is accepted to be ferromagnetic so that it forms a medium transmitting direct exchange interaction between the Fe crystallites necessary to average out the crystalline anisotropy and to yield high permeabilities.

The combined high resistivity and high permeability is particularly suitable for high-frequency devices such as inductors and magnetic heads because for these applications it is preferable that the magnetic permeability μ should remain constant as a function of the frequency up to as large as possible frequencies *f*. In literature a few studies of the frequency dependence of the magnetic permeability on oxidic Fe–M–O systems have been published. In a number of articles a problem is mentioned that one is not able to properly describe the frequency dependent behavior using the well-known loss mechanisms related to eddy currents and ferromagnetic resonance and it is suggested that the problem might originate in capacitive effects.[6] Indeed at high frequencies dielectric currents may start to run between Fe crystallites across the highly resistive Fe–M–O phase, i.e., the corresponding capacitors effectively lower the resistivity at high frequencies and consequently give rise to enhanced eddy current losses. If this mechanism were correct it should also be visible as a capacitive-like characteristic in the electrical impedance of the films, see also Refs. 7 and 8.

Earlier Huijbregtse *et al.*[9] reported on the frequency dependence of the magnetic permeability of a thick FeHfO film and interpreted the behavior in terms of combined eddy current shielding and ferromagnetic resonance (FMR) effects. In the present article we present a more detailed analysis including the thickness dependence. We also report on the frequency dependence of the electrical impedance of FeHfO films. It will be shown that displacement currents are of no importance for the roll off of the magnetic permeability for the present films.

II. EXPERIMENT

All films were prepared by radio frequency (rf) diode sputtering on a Perkin Elmer 2400 machine at a pressure of 3–4 mTorr corresponding to an Ar flow of 30 sccm. The oxygen has been applied with a 9 sccm flow of a gas mixture consisting of 95% Ar and 5% O_2. The samples have been annealed at 400 °C with rapid thermal processing in a magnetic field of about 600 Oe. For the typical dependence of the magnetic and electrical properties on the deposition conditions and information on the microstructure we refer to Ref. 9. The conditions used to prepare the present films result in films with a magnetic permeability of about 1500, a coercive

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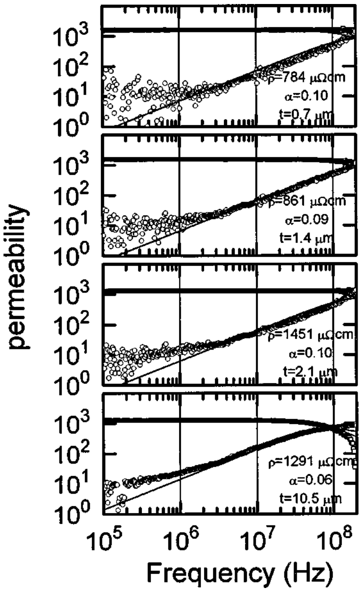

FIG. 1. Frequency dependence of the magnetic permeability for FeHfO films of different thicknesses as indicated. The solid lines are model calculations taking eddy current effects as well as ferromagnetic resonance effects into account. The used damping parameters α and (measured) resistivities are also indicated.

field of about 0.5 Oe, a uniaxial magnetic anisotropy constant close to 300 J/m^3, and a magnetostriction constant of about $+5\times10^{-6}$.

III. RESULTS AND DISCUSSION

To unravel the various mechanisms that play a role in the frequency dependent behavior of the magnetic permeability a study of the dependence on the thickness of the soft-magnetic film is important. This is because the two main mechanisms causing the permeability to roll off viz. eddy current shielding and ferromagnetic resonance, have different thickness dependencies.

In the case of the FMR mechanism, the frequency at which the magnetic permeability starts to roll off does not depend on thickness since it is only determined by material dependent properties viz. the damping factor α, the gyromagnetic ratio γ, and the uniaxial anisotropy constant K_u. On the contrary, in the case of eddy current shielding, the roll-off frequency is strongly thickness dependent.

Figure 1 displays the real (μ') and imaginary (μ'') parts of the complex relative magnetic permeability $\mu=\mu'-i\mu''$ as measured as a function of the frequency for several FeHfO thin films with thicknesses ranging from 0.7 to 10.5 μm. It is noted that the initial permeability is fairly independent of film thickness. What is seen from Fig. 1 is that the frequency at which the real and imaginary parts of magnetic permeability cross each other is almost independent of thickness. If the eddy current mechanism would have been the dominating mechanism then one would expect, for instance, that films having thicknesses differing by a factor of 10 would differ in cross frequency by a factor of 100. This is because the cross frequency varies with thickness t as:

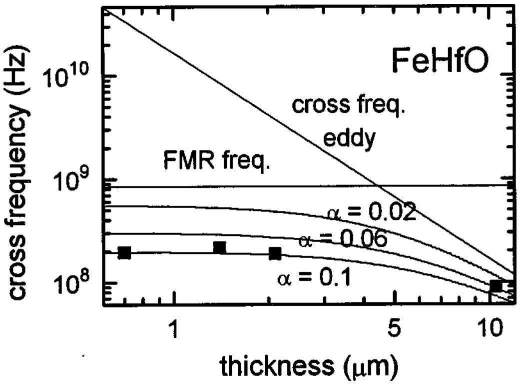

FIG. 2. Cross frequency as a function of the film thickness. The experimental values are represented by the solid squares. The calculated FMR frequency (horizontal line), the eddy current behavior (descending line), and the combined FMR–eddy current behavior for several values of the damping parameter α are plotted (curved lines).

$$f_{\mathrm{cross}}=\frac{\pi\rho}{\mu_0\mu_r t^2}=\frac{2\pi\rho K_u}{B_s^2 t^2}, \tag{1}$$

an equation that is easily derived from the well-known eddy current formula (see Ref. 10) by setting $\mu'=\mu''$. Here, ρ is the resistivity and B_s is the saturation induction. Evidently, this dependence does not apply to the experiment and other mechanisms dominate the behavior in this thickness regime. This becomes even more clear from Fig. 2 where we have plotted the cross frequency versus the film thickness on a double logaritmic scale. Here, the data points derived from the measurements shown in Fig. 1 are represented by the solid squares. The straight solid line that is decreasing with a slope of -2 has been calculated using Eq. (1). The meaning of the other lines will be explained later on. At low thicknesses Eq. (1) predicts cross frequencies that are much higher than observed. Now, as mentioned already in the introduction, Eq. (1) is derived for a thin film having uniform properties which is not the case for the present granular films. Capacitive coupling may result in larger eddy currents than one might expect on the basis of the direct current (dc) resistivity. To infer if these capacitive effects play a role we have theoretically estimated at which frequency these effects would occur and moreover measured the frequency dependence of the electrical impedance of our FeHfO films. Figure 3 displays a typical experimental result. It is seen that the real part of the impedance $Re(Z)$ is independent of frequency up to frequencies above those where the roll off occurs. If capacitive effects play a role then one would expect $Re(Z)$ to decrease with frequency near f_{cross}. Clearly Fig. 3 shows that this is not the case. In Appendix A it is shown that capacitive effects are indeed expected not to occur in this frequency regime but at much higher frequencies. It is therefore concluded that capacitive effects do not determine the roll off for the present films.

The obvious mechanism that will largely account for the roll off is ferromagnetic resonance. We now return to Fig. 2. The horizontal line at 0.84 GHz indicates the ferromagnetic resonance frequency calculated from

$$\omega/\gamma=\sqrt{2\mu_0 K_u}. \tag{2}$$

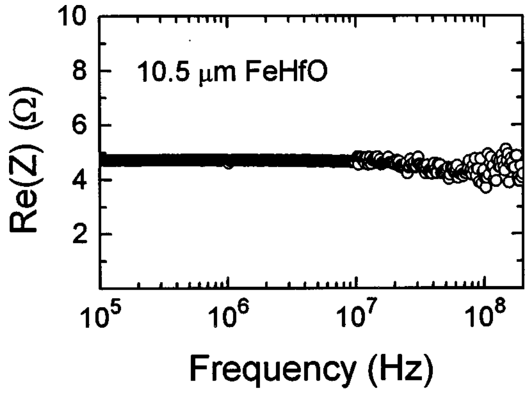

FIG. 3. Frequency dependence of the real part of the complex impedance of a 10.5-μm-thick FeHfO film.

Clearly this frequency is above the experimentally observed cross frequencies and a certain linewidth is necessary to account for the roll off. We took this into account as is done commonly in literature, i.e., we solved the Landau–Lifshitz (LL) equation including a damping term—the magnitude of which is determined by the parameter α. Since there are several different forms of the damping term and thus several definitions of α we have listed the LL equation that we used in Appendix A. To calculate the combined effect of FMR and eddy current shielding, we have substituted the solution for the complex magnetic permeability from the LL equation in the well-known eddy current expression—a procedure that is commonly followed, see, e.g., Refs. 11, 12, and 13. In doing so we have numerically searched for the cross frequency. The results as a function of the magnetic layer thickness are presented in Fig. 2 as the curved lines. The values for the damping parameters α used in the calculations are indicated in Fig. 2 also. All other parameters are fixed: γ=185 GHz/T, ρ=1000 $\mu\Omega$ cm, B_s=1.1 T and K_u close to 300 J/m^3 fitting μ' at low frequencies. It appears that the data can be fitted rather well using a damping parameter α between 0.06 and 0.1—values that are not unreasonable.

We have attempted to obtain a value for α from an independent ferromagnetic resonance experiment. At a fixed frequency of the microwaves of 9.34 GHz, the field directed along the film plane, was swept through the resonance. The result for the field derivative of the absorbed power for a 0.7 μm FeHfO film is shown in Fig. 4 (open circles). The solid line in this figure represents a calculation, again based on the LL equation taking eddy currents into account in the same manner as was done for the magnetic permeability. To obtain a good fit we needed to use a damping parameter of α=0.02. Clearly this value is inconsistent with the value needed to describe the permeability data. The origin for this discrepancy is unclear. It is possible that it is a result of the approximation that one makes in decoupling the LL equation from the Maxwell equations: FMR and eddy current shielding are, however, coupled phenomena. For example, in separately solving the LL equation it is assumed that the spins precess uniformly. This is clearly not correct since the spins at the surface experience an rf field that is different from that in the interior of the film: The rf field has a finite penetration depth. The spins will process differently depending on their position with respect to the film surface and an exchange term should be included in the LL equation. The coupled problems should be solved simultaneously, as has been done in Ref. 14 for an infinite thickness film. The magnitude of the error that one makes by using the above approximation will likely depend on frequency and may consequently give rise to different α values for the FMR experiment (gigahertz range) and the permeability experiment (megahertz range). The modeling error may also be the origin of the earlier mentioned problems in literature in describing permeability measurements, see introduction and Ref. 6.

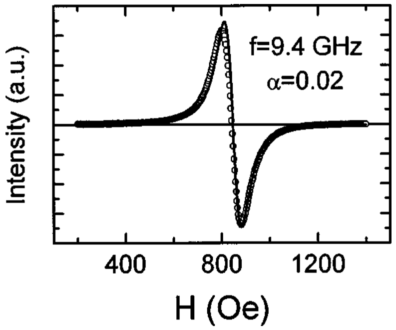

FIG. 4. Derivative with respect to the magnetic field of the absorbed power as a function of this field for a 0.7 μm FeHfO film. The magnetic field was directed along the film plane.

In conclusion, frequency dependent permeability measurements have been performed for highly resistive FeHfO films. The combined thickness and frequency dependence can be understood reasonably well with the commonly used models. The roll off of the permeability for thicknesses below 10 μm is dominated by the FMR phenomenon whereas above 10 μm eddy current effects start to play a significant role. A discrepancy has been found between the damping parameters α determined from the permeability measurements and the FMR experiment. Capacitive effects between Fe crystallites do not play a role in the behavior of the magnetic permeability with frequency.

APPENDIX A: MODELING

The definition of the damping parameter α that we have used in the present article corresponds to the Landau–Lifshitz equation in the following form:

$$\frac{1}{\gamma}\frac{\partial\vec{M}}{\partial t}=\mu_0\vec{M}\times\vec{H}_{\mathrm{int}}-\frac{\alpha}{M_s}\frac{1}{\gamma}\left(\vec{M}\times\frac{\partial\vec{M}}{\partial t}\right). \tag{A1}$$

Here, H_{int} represents the sum of the demagnetizing field and the anisotropy field.

To calculate the effect of displacement eddy currents on the magnetic permeability we introduce an effective complex resistivity ρ_{eff} that accounts for the effect of capacitive coupling between Fe crystallites. This resistivity may then be substituted in the common eddy current formula.

An expression for ρ_{eff} can be derived by modeling the granular system as a series of leak capacitors of thickness d connecting the Fe crystallites of size t. We write ρ_{eff} as:

$$\rho_{\mathrm{eff}}=\frac{\rho_{\mathrm{Fe}}t+\rho_{\mathrm{am}}d}{t+d}, \tag{A2}$$

with ρ_{Fe} the specific resistivity of an Fe crystallite and with ρ_{am} the specific complex resistivity of the amorphous phase which, in turn, is represented by a resisivity ρ_a and in parallel with this an ideal capacitor with specific impedance $-i/\epsilon\omega$, i.e., ρ_{am} is given by:

$$\rho_{\mathrm{am}}=\frac{\rho_a}{1+i\omega\epsilon\rho_a}. \tag{A3}$$

We may now substitute Eq. (A2) using Eq. (A3) into the common eddy current formula for the resistivity. After calculation (not shown) it appears that the behavior for the magnetic permeability in our frequency range is not altered. Here, we have used $\rho_a=1000\ \mu\Omega$ cm, $d=t=5$ nm, and $\epsilon=25\epsilon_0$. This relative permittivity value $\epsilon_r=25$, is an overestimation based on the relative permittivity of $\mathrm{HfO_2}$. The fact that the behavior is unaltered is completely understandable since the characteristic frequency $f=1/(2\pi\epsilon\rho_a)$ where the impedance starts to deviate from the dc value and can be estimated to be of the order of 10^{14} Hz, i.e., much higher than typical cross frequencies observed for the magnetic permeability. Our conclusion is therefore that capacitive coupling between Fe crystallites is not a relevant loss mechanism for the roll off of the magnetic permeability.

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