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Magnetic properties of nanocrystalline materials for high frequency applications

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Chapter 7

Summary

Ultrasoft ferromagnetic materials can be incorporated in high frequency devices due to their distinctive properties. A control of the structure at the nanoscale range is the key element in obtaining the right parameters for such material. The objective of this thesis is to contribute to the understanding of the correlation between the structure and magnetic properties of new nanostructured materials as potential candidates for high frequency applications. In the first chapter, the principal requirements which have to be considered for the synthesis of a magnetic material used in high frequency applications are introduced together with a justification from the experimental point of view.

The samples investigated in this study were obtained by two different methods: nitriding treatments of cold rolled alloys and reactive sputtering. A detailed description of the experimental methods is given in Chapter 2. Here special attention is given to the measurement technique of permeability spectra. The operating bandwidth of the spectrometer extends up to 6GHz. This was high enough to measure complete resonance lines of our iron based nanocrystalline films.

A characteristic of our materials is that most of them have nitrogen as constituent element. The use of nitrogen was motivated by the following reasons: i) control of the various phases of iron nitrides, ii) grain size dependence on nitrogen concentration in case of reactive sputtered samples, iii) possibility to reduce the magnetostriction constant and iv) possibility to induce a magnetic anisotropy. In Chapter 3 a study of the effect of low temperature nitriding on the magnetic properties of $\text{Fe}_{93}\text{Ni}_4\text{Cr}_3$ and $\text{Fe}_{94}\text{Ni}_4\text{Ti}_2$ cold-rolled alloys is presented. Low temperature nitriding in the α region leads to formation of *fcc* CrN and TiN precipitates uniformly distributed within the grains. This process is accompanied by an increase of the interstitial nitrogen concentration which in case of the Ti containing alloy is about 3 at.%. From the structural point of view, these precipitates can act as nucleation centers for Fe nitride phases formed in subsequent steps and can also restrict the grain growth when formed at the grain boundaries. The role of interstitial nitrogen dissolved in the α phase is demonstrated by annealing a (Fe-Ti-Ni) sample in a H_2 atmosphere at the same temperature as used for nitriding. We

suggest that the lower coercive field measured in nitrated samples has to do with a decrease of the magnetostriction constant, which changes with nitrogen concentration.

Nitriding in the γ' region yields another important result concerning the magnetic properties of our samples. We show that a combination of α - γ' phases can lead to a soft magnetic behavior. The most probable mechanism is the reduction of the total free energy of the system when averaging over regions containing both phases. This is possible since the signs of the magnetic anisotropy constants of the α and γ' phases are opposite.

Although both nitriding techniques mentioned above yield positive results related to the coercive fields, still the structural aspect have to be improved in order to obtain a material suitable for high frequency applications. The reasons are the following: i) the samples have grains too large as compared with the magnetic correlation length L_m and ii) the initial texture of the as rolled samples is not removed. Our approach in solving these problems was to perform $\alpha \leftrightarrow \epsilon$ transformations. In this respect, the best results were obtained for the (Fe-Cr-Ni) alloy. An almost complete reduction of the texture was obtained after 12 cycles, accompanied by reduction of the grain size to 100-200nm, still larger than L_m . Further phase transformations did not improve the structure or magnetic response. A secondary structural effect was also identified as a distribution of microstress. In consequence we conclude that in these systems soft magnetic properties cannot be obtained via solid state phase transformations.

In Chapter 4 the investigation of the magnetization dynamics in nanocrystalline soft magnetic materials is presented. The samples were produced by reactive sputtering on a SiO_2 substrate. The parameters which have been systematically varied in this study were the sample composition (different Zr or N concentrations), the grain size and the magnitude of the induced anisotropy. The highest resonance frequency in zero external field was 2.1 GHz for a sample with 14at.% N and a saturation magnetization $4\pi M_S = 16.9\text{kG}$. These values make this material suited for high frequency applications.

All permeability spectra could be analyzed using the Landau-Lifshitz interpretation. Furthermore, the measurements of permeability spectra in constant external fields H_{DC} up to 70Oe reveal the following new findings: i) the ferromagnetic resonance frequency is shifted towards higher frequency values than expected on basis of measured anisotropy field H_K and ii) the phenomenological damping parameter has a pronounced nonlinear dependence on the external field. These experimental observations were supported by ferromagnetic resonance measurements at 9.4GHz which yield saturation magnetization values.

The anisotropy fields were calculated from the permeability measurements performed at the same external field, subtracting the resonance values specific for $H_{RF}||HA$ and $H_{RF}||EA$. We conclude that this method is more appropriate for such magnetic systems than DC hysteresis loops or field-dependent ferromagnetic resonance. Combining the resonance values in both orientations of the sample at the

same H_{DC} we obtain the equivalent field H_{Shift} responsible for the FMR line shift. In the regime in which the spins precess coherently this field is independent of H_{DC} or saturation magnetization and increases with decreasing grain size. Discussing this effect we arrive at the conclusion that H_{Shift} originates from the variations of the magnitude of the magnetization, associated with atomic density fluctuations in the nanocrystalline structure. These variations lead to local demagnetizing fields acting on a length scale comparable to the grain size. Since the local demagnetizing fields are averaged-out in the direction of H_{DC} (Ox axis), only the local in-plane demagnetizing fields along the Oz axis contribute to the effect observed by us. Assuming a sinusoidal variation of the magnetization and exchange stiffness, we estimate a variation of the magnitude of the magnetization of about 3%. The effect of spin wave excitations is also discussed; we argue that they do not provide an explanation of the behavior of the linewidth as a function of the external field.

The influence of the substrate roughness on the magnetization dynamics of soft nanocrystalline thin films is presented in chapter 5. For this purpose we have chosen $(Fe_{99}Zr_1)_xN_{1-x}$ as material of study with a saturation magnetization $4\pi Ms \cong 15.5 \text{ kG}$ and an induced anisotropy field $H_k \cong 20 \text{ Oe}$. The substrates used in this study were: a) Si covered with Si-oxide (like the one used in Chapter 4), b) Si covered with a thin organic layer and c) Si covered with a Cu layer. The morphology of the samples was analyzed using atomic force microscopy. The highest root mean square roughness amplitude $w=5.1 \text{ nm}$ was obtained for the sample deposited on the Cu covered substrate. Computing the height-difference correlation function in the fast scan direction we were able to calculate a lateral correlation length ξ of 78.2 nm for this sample. The demagnetizing factors were calculated assuming an isotropic and translation invariant random roughness in two directions. A different morphology has the sample deposited on polymer, where the root mean square does not yet saturate over the scan sizes of 6000 nm . This sample is characterized by hillocks with a lateral base size $L \approx 400 \text{ nm}$ and an average height $\Delta \approx 5 \text{ nm}$. The demagnetizing fields for this sample were estimated based on the dimensions of these hillocks. For the sample deposited on Si-oxide, the surface roughness does not contribute to the magnetic properties.

The magnetic structure in the remanent state was single-domain as shown from the hysteresis loops. This is because the spins do not follow all imperfections due to the strength of the exchange coupling and are aligned by the induced uniaxial anisotropy obtained during sputtering in a magnetic field. The permeability spectra of these samples show a clear influence of the substrate morphology. While for the sample deposited on Si-oxide the resonance line-width was $\Delta\omega \cong 520 \text{ MHz}$, the resonance lines of the other samples were much broader, with $\Delta\omega > 1 \text{ GHz}$. In the same time the average resonance frequency is characterized by a positive shift which increases for the samples with a rougher morphology. The spectra were analyzed by using two different approaches: i) we consider that the local demagnetizing field follows a simple periodic position-dependence (a reasonably good fit of the spectra

was obtained) and ii) transforming the spectra $\mu=\mu(f)$ into $\mu=\mu(H_{eff})$ via the dispersion relationship. A good agreement between these interpretations was obtained.

In the discussion part of this chapter we refer to the line asymmetry, the line position and the homogenous linewidth as observed via the damping parameter. We show that the line shape and the line position can be understood from the rough profile of the film interfaces. The values obtained for the halfwidth of the local field distribution compare well with the average demagnetizing field calculated from the AFM measurements. Like we did in chapter 4 we attribute the line shift to atomic density fluctuations in the fine grained material, which increase when the interfaces become rougher. For the same sample we have some evidence that the homogenous damping parameter α is much larger in the permeability measurements ($\nu_{FMR}\approx 2\text{GHz}$) than in the X-band measurement ($\nu_{FMR}\approx 9.4\text{GHz}$). In the same time α is increasing with the sample roughness. Our point of view regarding the last effect is that probably the two magnon process can be responsible.

The static and dynamic magnetic properties of Fe-Zr-N nanocrystalline films with perpendicular anisotropy are given in Chapter 6. Here we study a set of samples in which a transition from stripe domains to in-plane orientation of the spins is observed. Once again the nitrogen is the element which correlates most with the magnetic behavior. While the sample with a nitrogen concentration of almost 8at.% shows a stripe periodicity of 220nm, the stripe domain pattern disappears for a nitrogen concentration of approximately 4at.%. This behavior corresponds to an increase of the saturation magnetization from 18.3kG to 19.6kG and a decrease of the perpendicular anisotropy constant from $4.3 \cdot 10^5 \text{erg/cm}^3$ to an insignificant value. The magnetization distribution in the cross-section of the films was determined assuming a model with two types of magnetic domains: i) bulk magnetic domains characterized by an orientation of the magnetization out of the sample plane and ii) closure domains with an orientation of the magnetization in the sample plane. Quantitative estimations were done combining Mössbauer spectroscopy to probe the entire sample and magnetic force microscopy to image the sample surface.

Torque measurements combined with DC hysteresis loops were used in order to determine the magnetization reversal mechanism of these samples. The in-plane magnetization reversal is a two-step process. First the spins turn out of the sample plane. When a critical value of the out of plane angle is reached, a 180° in-plane rotation of the stripe pattern takes place over a very small field interval. If the external field makes an angle close to 90° with respect to the sample plane, the reversal mechanism is mostly determined by the shape of the sample. The influence of the perpendicular anisotropy can be observed when this external field has relatively small values as compared with the saturation magnetization and is associated by a rotational hysteresis behavior in the torque measurements.

The permeability spectra of the samples were measured for the magnetic structure at the remanence state. In this way we have shown that these materials are

also potential candidates for high frequency applications. The FMR frequency of the sample with the highest perpendicular anisotropy is higher than 2GHz and the linewidth is about 500MHz. It is clear that a control of the magnetization dynamics can be achieved by controlling the perpendicular anisotropy and the thickness of the samples.