Magnetic properties of nanocrystalline materials for high frequency applications

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Chapter 1

Introduction

1.1. Magnetic materials for high frequency applications.

The ongoing demand for miniaturization in electronic devices used in telecommunication and recording industry has stimulated the increase of research in areas like materials development, device analysis, design and microlithography.

Magnetic materials are used in a large variety of devices like passive circuit elements (see examples in Fig. 1.1), sensors, reading-writing heads (example in Fig. 1.2), information storage media, etc. The synthesis and understanding of their properties are essential requirements for their integration in a certain industrial fabrication process. One class of magnetic materials which is of great interest in recent years is that of ultra-soft magnetic materials [1]. These materials can be used to extend the operation frequency range of various passive circuit elements into the GHz regime.

It is well known that when a highly permeable soft magnetic material is placed near a conductor carrying an electrical current, the inductance of the conductor will increase. In the ideal case, if the conductor is enclosed in an infinite magnetic medium, the inductance is increased by a factor of μ_r, the relative permeability of the medium. Additionally, the quality factor Q of such a structure increases if the magnetic losses are small. This means that if a highly permeable material is incorporated into an inductor without producing extra losses, a substantially higher inductance and Q value can be obtained without increasing the size of the device. Alternatively, for the same inductance a much smaller area is needed. Moreover, the cross-talk between inductors on the same chip would be reduced because the magnetic flux is confined within the magnetic core. These issues are particularly relevant for electronic circuits used in mobile communication systems where miniaturization and integration of the inductors used in LC (inductor-capacitor) filters and voltage controlled oscillators are of great interest.

The most commonly used design for making planar inductors is the spiral (Fig. 1.1a). This is mainly because its inductance is proportional to the number of turns in the spiral. In practice, typical RF (radio frequency) spiral inductors have only

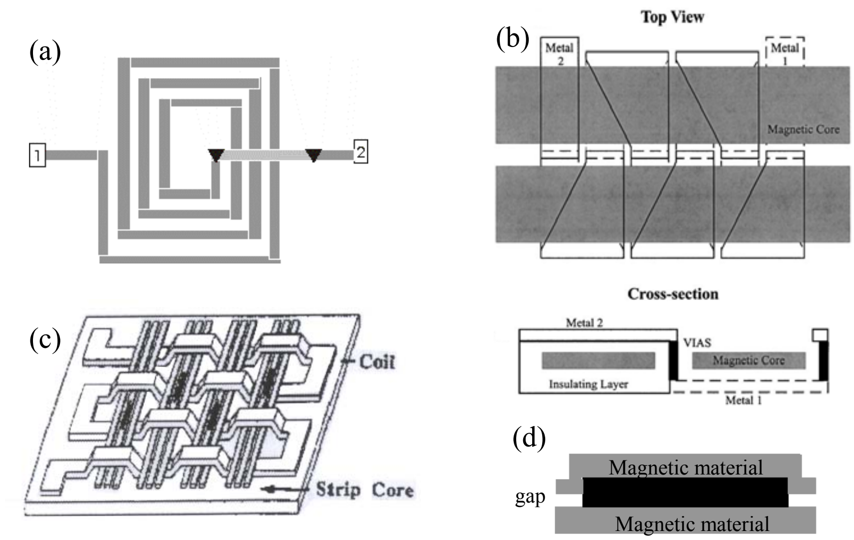

Fig. 1.1 Examples of high frequency inductors. a) spiral inductor, b) top view and cross-section of a planar inductor with two magnetic cores, c) thin film cloth-structured inductor with magnetic strip core array, d) geometry of the electromagnetic model for a magnetic sandwich inductor with variable edge gaps

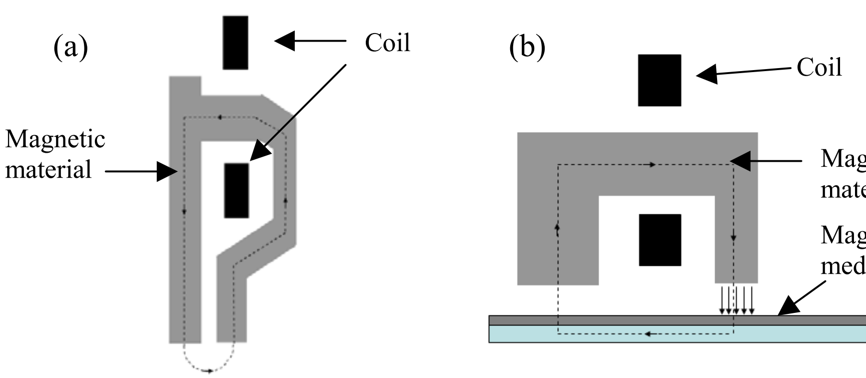

Fig. 1.2. Schematic drawings of a (a) longitudinal writing head and (b) perpendicular writing head and a magnetic medium with a soft underlayer.

a few turns, and their inductance is only a factor of 2 or less larger than for a straight conductor of the same length, width and thickness as the conductor of the spiral. This is equivalent with the fact that the major contribution to the inductance of the spiral comes from the self inductance of the conductor and that the spiral is basically a compact way of placing a long conductor on the substrate. On the other hand sandwiching a spiral between two magnetic layers can lead to an inductance enhancement as high as a factor of μ_r for very thick films [2]. Because the RF field from the spiral coil is not unidirectional, the biasing of the magnetic films to keep them in a single domain state is not an easy task. In order to solve this issue a new design was proposed having the magnetic material in a stripe-like shape and the conductor oriented perpendicular to it. Recently, many publications were devoted to the optimization of these designs [3, 4]. As pointed out by Korenivski and Dover [5], understanding of the physics describing a "magnetically enhanced" stripe inductor is the straightforward way of approaching the circuit integration. For an example of such a magnetic sandwich structure see Fig. 1.1d.

As we already mentioned, the soft magnetic materials integrated in high frequency devices must be characterized by a well controlled magnetic domain pattern. In the absence of an external magnetic field these domains have their magnetization oriented perpendicular to the RF field. In this way a relatively frequency-independent magnetic permeability is obtained because the change in magnetization is caused by a rotation process and not by domain wall movements [6]. The most important mechanisms which contribute to the total losses in such material are the *eddy currents* and the *ferromagnetic resonance*. Eddy currents are produced in conductors by the time varying magnetic field. On their turn, the eddy currents produce a magnetic field which is opposite in orientation to the external field, thus preventing the field to enter in the conductor. Ferromagnetic resonance (FMR) is a phenomenon in which the magnetic moments precess coherently around the effective field. It normally occurs in the GHz frequency range, although for some highly permeable materials this phenomenon can be observed in the MHz range.

The requirements which must be satisfied by a material used as magnetic medium in high frequency inductors are:

- *High saturation magnetization* M_S to ensure a large value of permeability $\mu_r \cong \frac{4\pi M_S}{H_k}$
- *Controllable uniaxial anisotropy* field H_k=10÷50Oe in order to bring the FMR frequency $f_{FMR} \cong \gamma\sqrt{4\pi M_S H_k}$ in the GHz range.
- *The FMR line-width must be small*. In a real system, with dissipation, the FMR line can be broadened by processes like spin lattice relaxation, nonuniform magnetization modes (spin waves), eddy currents.

- *Sufficiently high resistivity* to guarantee that the intensity of the RF field does not change as a function of depth. In sandwich stripe inductors no insulation is required if the conductivity of the nonmagnetic layer is ~10^2 larger than the conductivity of the soft magnetic material. This eliminates the unwanted capacitance.
- *Low magnetostriction* is required because in the fabrication process a stress-induced anisotropy may appear.
- *Process compatibility.* There are several requirements, for instance the magnetic material should be compatible with the chosen substrate. In addition, the temperature of processing steps after deposition of the magnetic layer, should not exceed the maximum temperature at which the magnetic material is stable.

For applications in the kHz to MHz range the best choice of material is the ferrite magnetic medium, e.g. NiZn ferrite. This is mainly because of its high resistivity. The upper limit of such material in absence of a bias magnetic field is determined by the ferromagnetic resonance frequency, which for these materials was calculated by Snoek [6]. The Snoek limit [6] is the line $\mu=f(\omega)$, calculated in the absence of an external field, above which the permeability can not have values, as long as a cubic magnetocrystalline anisotropy is present. Furthermore the sintering of ferrite films requires a high temperature which makes these materials incompatible with the rest of the elements of a high frequency integrated circuit. Soft metal alloys films with a well defined in-plane anisotropy may overcome this limitation. Historically the first alloy investigated for the purpose of use at high frequencies was permalloy ($Ni_{75}Fe_{25}$) [7, 8, 9]. This alloy has a resonance frequency below 1GHz. Almost similar values for the permeability and resonance were measured for CoNbZr films [10] which are studied because of their very low values of magnetostriction. Values of the ferromagnetic resonance above 1GHz were obtained with a FeCoB alloy. Recently nanocrystalline Fe-based oxides and nitrides were investigated as components of microinductors. The examples presented in Table 1.1 show the relationship between the composition of the alloys and the magnetic properties of interest. Remarkable are the results obtained in Ref. 21 which demonstrate that it is possible to achieve a value of H_k as high as 160Oe in boron-based amorphous alloys. Other interesting results are obtained in Ref. 22 where the maximum resonant frequency is obtained if the soft magnetic material is placed between two layers of permalloy with a much smaller thickness. The explanation proposed by the authors is based on the coupling of those spins situated at the interface between the Fe-Co-N alloy and the permalloy. This is yet to be verified.

The most important property which makes the materials presented in Table 1.1 suitable for high frequency applications is their crystalline structure (nanocrystalline or amorphous). In this way the effective anisotropy contribution of

Table 1.1 Magnetic properties of materials suited for ultra high frequency applications. The alloy composition is given in at%.

Material	H_k (Oe)	$4\pi M_S$ (kG)	μ_r (relative units)	f_r (GHz)	Ref.
Nitrides: Fe-M-N with M=Ta, Cr, Ti, Zr					
$FeCr_{4.6}Ta_{0.2}N_{7.4}$	90	20.2	200	3.7	11
$FeTa_{3.3}N_{1.6}$	4.5	21.5	1880	0.8	12
FeAlN	10	20	2000	1.2	13
FeZrN	5	18	4000	0.8	14
$FeTi_{(8-10)}N_{(10.5-14)}$	10	24	3200	1.3	15
$FeSi_{10}Al_5N_2$	14	11	800	1.1	16
Oxides: Fe-M-O with M=Al, Si					
$FeAl_{3.5}O_8$	4	18.2	4600	0.7	17
$FeHf_{17}O_{28}$	6	10.5	1600	0.7	18
$FeSm_{3.4}O_{13.2}$	5	12.6	2600	0.6	19
Other materials					
$FeCo_{18}B_{15}$	22	17.5	740	1.7	20
$FeCo_{17}B_{16}Si_1$	160	18	110	4.2	21
$(Fe_{0.7}Co_{0.3})_{95}N_5$	20	24.5	1200	1.9	22
Materials used in this thesis					
$FeZr_2N_{17}$	26	17	680	1.8	23
$(Fe_{0.7}Co_{0.3})_{83}Ta_2N_{15}$	48	17	340	2.5	24

the randomly oriented grains can be reduced by the exchange interactions. When the grain size becomes smaller than the magnetic correlation length, the magnetization will not follow the orientation of the local easy axis but will be forced to align parallel by the exchange interaction. The local anisotropies are averaged out over a large number of grains [25]. Supplementary, a well defined uniaxial anisotropy has to be induced in the sample plane, see Fig. 1.3a. For iron-based alloys, the magnetocrystalline anisotropy can be reduced in some cases up to three orders of magnitude if the grain size is 10-15nm, ensuring ultra soft magnetic properties. This was shown by Herzer [26] for materials produced by rapid solidification and subsequently annealed above crystallization temperature, see coercivity curves in Fig. 1.3.b.

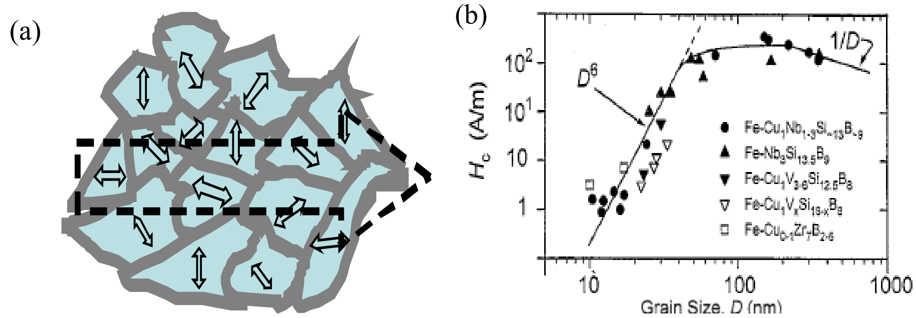

Fig. 1.3. a) Schematic representation of a nanocrystalline structure; left-right arrows indicate the orientation of the local easy axis and the dashed arrow indicates the average orientation of the magnetization. b) coercivity H_C versus average grain size [26]

We also point out that Herzer's materials were almost free of magnetostriction in order to minimize magneto-elastic anisotropy.

Going back to Table 1.1, we can see that while most of the materials have a large saturation magnetization due to their high Fe content, only those with a large induced anisotropy field have also a high resonance frequency. The advantage of those alloys which contain interstitial nitrogen or carbon is that we can induce magnetic anisotropy in a certain direction if the deposition is performed in a magnetic field [27]. Other methods of inducing anisotropy are: oblique deposition [28] and patterning the substrate [29].

The resistivity of these alloys is related to the concentration and nature of the non-metallic constituent. While in case of N-containing alloys the resistivity can go up to 100μΩcm, in those magnetic materials containing oxygen the resistivity may reach even 1mΩcm [18]. A solution for the resistivity problem is a laminar structure in which thin magnetic layers are separated by dielectric films. The reason is that the electromagnetic energy is stored mainly in the dielectric layers. Such structure is also very convenient for notch filters [30, 31]. The crystalline state may also influence the resistivity since the number of scattering centers increases with the surface of the grain boundaries. Annealing at temperatures in the interval 300-500°C leads to grain coarsening and consequently the disappearance of soft magnetic properties. Although this is not important for high frequency circuits, in case of applications like hard disk recording heads the thermal stability is a fundamental issue.

1.2. This thesis

The aim 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. Since our approach was an experimental one, a substantial effort was made in the area of synthesis and characterization techniques. Details of the various experimental methods used in this work are given in Chapter 2.

In Chapter 3 the results concerning the structure and magnetic properties of nitrided $Fe_{93}Ni_4Cr_3$ and $Fe_{94}Ni_4Ti_2$ cold-rolled alloys are presented. The idea was to investigate the possibility of obtaining an ultra-soft magnetic material via phase transformations in the Fe-N system. First, nitriding in alpha phase is discussed considering the influence of the CrN or TiN precipitates. The formation of these precipitates enables us to control the nitrogen concentration and consequently the magnetic anisotropy. We have also studied the role of an additional magnetic phase in obtaining soft magnetic behavior. In such biphasic systems containing a mixture of α and γ' phases, the coercive field was reduced even more. The next step was to create a nanocrystalline material by repeated phase transformations. Because of the low temperatures needed, nitriding/ reducing was an attractive method. The advantage over similar conventional methods was the effect of grain coarsening. Although a significant grain refinement can be obtained using this method, it turns out that such process does not lead to the desired soft magnetic properties. We note in passing that it may be interesting to investigate the mechanical properties of these fine-grained alloys.

One of the most successful methods for synthesis of soft magnetic materials is reactive sputtering. We have applied this method in order to study the Fe-Zr-N system. In addition some samples having Co and Ta as alloying elements were also fabricated, see example in Chapter 2. A fine tuning of structural and magnetic properties was obtained by varying the substrate temperature. In Chapter 4 the study of magnetization dynamics of soft nanocrystalline Fe-Zr-N films with random magnetocrystalline anisotropy and induced uniaxial anisotropy is presented. The parameters which were systematically modified were the composition, the grain size and the induced anisotropy. We show that the magnetization dynamics is strongly influenced by the structural parameters. A positive shift of the resonant frequency with respect to the theoretical prediction for a perfectly homogenous and smooth layer was present in all samples. The extra field needed in order to have agreement between experiment and calculations does not depend on the saturation magnetization and increases significantly when the grain size decreases from 10 to 2 nm. After discussing a number of effects we conclude that the frequency shift is due to the slight difference in magnitude of the magnetization in the interior of the grains and the grain boundaries.

Another factor which can influence the high frequency permeability response is the sample roughness. This is shown in Chapter 5 where the magnetization dynamics of samples deposited on substrates with different roughness is presented. The analysis is performed for samples having the same composition. The samples deposited on rough substrates (polymer or Cu) have a much large resonance linewidth than the one deposited on Si-oxide. Using atomic force microscopy (AFM) we were able to characterize the topography of the samples and to calculate the demagnetizing factors. The analysis of the permeability spectra was done introducing a distribution of the local demagnetizing fields associated with the sample topography. It turns out that the high frequency behavior can be understood from the rough profile of the film interfaces.

In Chapter 6 nanocrystalline films with perpendicular anisotropy are investigated. By changing the power of sputtering, a transition from stripe domain magnetic structure to in-plane orientation of magnetization was obtained. This effect is related to the nitrogen content of the samples. Mössbauer spectroscopy measurements in combination with magnetic force microscopy were performed in order to determine the magnetization distribution of the samples. This method was introduced by us for the first time. In addition torque measurements were used for understanding the magnetization reversal mechanism. The permeability spectra demonstrate the possibility to use such material for applications in the GHz range.

The summary of this thesis is given in Chapter 7.

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