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STRUCTURE CONSTANT AND GRAIN SIZE DETERMINATION BY FERROMAGNETIC RESONANCE IN THIN MAGNETIC FILMS

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The paper shows that the structure constant and the average crystal grain size of anisotropic nanocrystalline magnetic film can be determined by analyzing the shape of the microwave absorption peak in sweeping the external magnetic field along the hard magnetization axis. In the theory of magnetization ripple, the surface energy density of the local magnetic anisotropy is connected with the structure constant, which can be used to determine the quality of nanocrystalline films. The effectiveness of the structure constant measurements is demonstrated on a 300-nm-thick nanocrystalline Co–P *film. Spectral data on the microwave absorption are collected in the ~1 mm^2 region of the film using a scanning ferromagnetic resonance spectrometer. The structure constant obtained from the spectral analysis allows detecting the average grain size of the magnetic film, which is in good agreement with transmission electron microscopy observations.*

Keywords: nanocrystallites, thin magnetic films, random magnetic anisotropy, magnetization ripple, structure constant, ferromagnetic resonance, microwave frequencies.

INTRODUCTION

The development of the synthesis technologies of nanocrystalline thin magnetic films is primarily connected with their unique magnetic properties, that set them apart from solid ferromagnetic materials. In particular, nanocrystalline thin magnetic films demonstrate high-frequency magnetic susceptibility and low losses on the microwave frequencies due to their specific microstructure [1]. This makes them suited for the use in high frequency sensors of weak magnetic fields and signal processing devices [2–5].

Nanocrystalline thin magnetic films are mainly characterized by a small grain size as compared to the effective radius of the exchange and magnetodipole interactions. The magnetic coupling between the grains leads therefore to averaging a random magnetic anisotropy and its partial suppression in individual grains. However, averaging the local magnetic anisotropy within the effective exchange-interaction radius is not usually complete. In this case, the magnetization vector deviates from a mean direction. This results in a peculiar magnetic structure, which is called magnetization ripple [6, 7]. The magnetic properties of such films are described by the theory of magnetization ripple. The theoretical conclusions are in good agreement with the numerical simulations [8, 9] and experimental data [10].

Unlike the Stoner–Wohlfarth model [11] widely used for the description of the magnetic properties of a uniaxial ferromagnetic single domain particle (thin film in the limiting case), the theory of magnetization ripple creates a link between the nanocrystalline film structure and its magnetic properties. This theory introduces a new

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mateial parameter, namely the structure constant, which describes the thickness-averaged surface energy density of the local magnetic anisotropy [7]. The structure constant S of the magnetization ripple should be considered as a quality factor of the magnetic film. For example, $S = 0$ means that the magnetization ripple is absent in the film, and its behavior can be described by the Stoner–Wohlfarth model. But this condition is never satisfied in real nanocrystalline magnetic films, despite the fact that the structure constant can be an arbitrary small value.

The theory of magnetization ripple also leads to a simple and rather effective method of deriving the structure constant from the field dependent transverse magnetic susceptibility of thin films [12]. This work presents a novel method for calculating the structure constant from the microwave absorption spectrum of nanocrystalline thin films. This method is based on a specificity of the high-frequency susceptibility discovered in our early research [13] in sweeping the external magnetic field along the hard magnetization axis.

1. TRANSVERSE MAGNETIC SUSCEPTIBILITY IN NANOCRYSTALLINE MAGNETIC FILMS

The term of transverse magnetic susceptibility was first introduced by Aharony *et al*. [14]. They calculated a tensor of quasi-static magnetic susceptibility of a uniaxial single domain particle using the Stoner–Wohlfarth model [11]. The measurement technique of the transverse magnetic susceptibility illustrated in Fig. 1, includes recording the deviation $\Delta\boldsymbol{M}$ of the magnetic moment $\boldsymbol{M}$ of the thin film exposed to a small variable magnetic field $\boldsymbol{h}$ normal to the constant sweeping field $\boldsymbol{H}$. The external magnetic field sweeping either along the hard magnetization axis (HMA) or easy magnetization axis (EMA) is usually used for anisotropic magnetic films. The frequency of the variable magnetic field is commonly selected in the kilohertz range.

The transverse magnetic susceptibility for the homogeneously magnetized thin magnetic film with in-plane uniaxial magnetic anisotropy, is identified in the linear approximation as

$$\chi_t = \lim_{h\sim\to 0} \frac{\Delta M}{h} = \frac{M_\mathrm{s}}{H_\mathrm{eff}}, \tag{1}$$

where M_s is the saturation magnetization; H_eff is the effective magnetic field, which includes the constant external field H and the magnetic anisotropy field H_a. In Fig. 1, the effective internal magnetic field is calculated as $H_\mathrm{eff} = H - H_\mathrm{a}$. In practice, the experimental dependence $\chi_t(H)$ differs from theoretical calculations (Eq. (1)). This difference can be explained by the magnetization ripple [12, 15], which occurs in real magnetic films owing to inhomogeneity of different nature.

Based on the theory of magnetization ripple, Hoffmann [16] derived the equation for the transverse magnetic susceptibility of nanocrystalline thin films. He showed that the total effect from the real film structure on the magnetic susceptibility was determined by the structure constant

$$S = \frac{DK_s\sigma_1}{\sqrt{n}}, \tag{2}$$

which is the most important quality parameter of the magnetic film. The structure constant S is the thickness-averaged surface energy density of the local magnetic anisotropy. It is determined by the average grain size D; the magnetic anisotropy constant K_s allowing for the effect of local crystallographic anisotropy and elastic stresses; the constant σ_1 describing the root-mean-square deviation of EMA grains; the thickness-averaged grain number $n = d/D$, where d is the film thickness.

Doyle and Finnegan [17] derived the general equation for the local anisotropy $K_s\sigma_1$ in polycrystalline films with regard to the contribution of both crystallographic and magnetoelastic energies. In the general case of isotropic deformation of the polycrystalline films, the average local anisotropy $K_s\sigma_1$ was calculated as

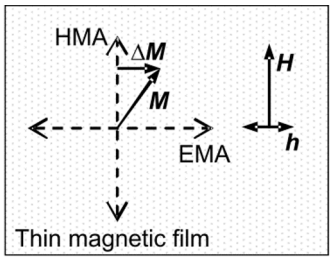

Fig. 1. Directions of external magnetic fields in measuring transverse magnetic susceptibility.

$$K_s\sigma_1 = 2\left(\frac{2}{105}\right)^{1/2}\left[\left(K_1+\frac{3}{8}\lambda_s\sigma_i\right)^2+\frac{7}{16}\left(\frac{3}{2}\lambda_s\sigma_i\right)^2\right]^{1/2}, \tag{3}$$

where K_1 is the magnetocrystalline anisotropy constant of individual particles, λ_s is the magnetostriction constant of the film, σ_i is the structure-dependent parameter describing the internal in-plane isotropic stress in the film.

Within the frame of the theory of magnetization ripple, Harte [6] and Hoffmann [7] showed that fluctuations of the direction of magnetization in thin films was accompanied by internal demagnetizing fields. The mean value of the effective demagnetizing field H_d of the magnetization ripple was calculated by Hoffmann [16].

The effective demagnetizing field H_d at the film magnetization along the hard magnetization axis is obtained from

$$H_d = \frac{1}{2\pi 2^{1/4}}\frac{S^2\sqrt{d}}{A^{5/4}M_s^{1/4}}\left(H-H_a\right)^{-1/4}.$$

Here A is the exchange copuling constant. Formally, on the right-hand side of Eq. (4), it would be necessary to write down the field $H - H_a + H_d$ instead of the effective field $H - H_a$. In this case, the effective demagnetizing field H_d would be a finite quantity at $H = H_a$. However, the approximation (4) is well founded within the fields $H > H_a$, that are used to determine the structure constant.

With regard to the additional internal field H_d, the effective field H_{eff} included in Eq. (1) for the transverse magnetic susceptibility, can be calculated as $H_{eff} = H - H_a + H_d$. And with a glance to Eq. (4), we obtain

$$\chi_t = \frac{M_s}{(H-H_a)+B(H-H_a)^{-1/4}}. \tag{5}$$

Equation (5) includes the parameter B of the demagnetizing field

$$B = \frac{1}{2\pi 2^{1/4}}\frac{S^2\sqrt{d}}{A^{5/4}M_s^{1/4}}, \tag{6}$$

that can be used as an adjustable parameter in approximating the experimental curve $\chi_t(H)$ by the theoretical one (Eq. (5)). Equation (6) allows us to derive the structure constant S from the experimental value of the demagnetizing field parameter B.

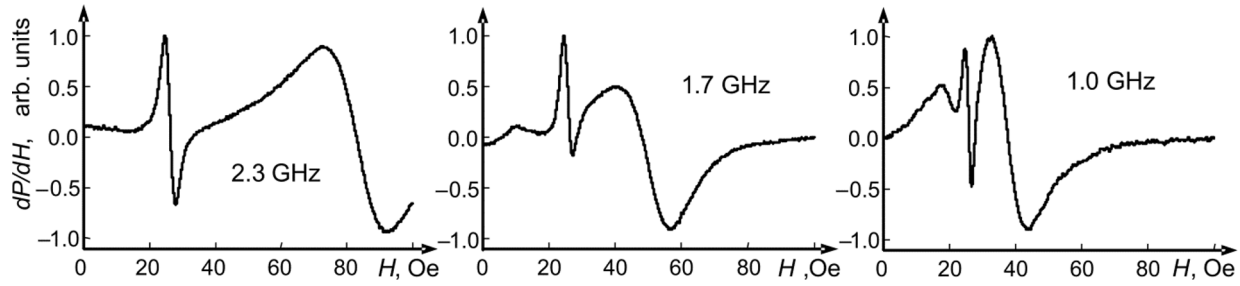

Fig. 2. FMR spectra of the center of 300-nm-thick Co–Ni–P film. *dP*/*dH* is the differential absorption spectrum of the local region.

2. HIGH-FREQUENCY SUSCEPTIBILITY OF THIN FERROMAGNETIC FILMS WITH UNIAXIAL MAGNETIC ANISOTROPY

It is known that in sweeping the planar magnetic field along the hard magnetization axis, one or two resonance peaks are observed (depending on the pumping frequency) in the ferromagnetic resonance (FMR) spectrum of the uniaxial single domain magnetic films [18]. The resonance fields of the peaks can be obtained from

$$\omega = \gamma\sqrt{\left(4\pi M_{\mathrm{s}} + H_{\mathrm{a}}\right)\left(H_{\mathrm{a}}^2 - H^2\right)/H_{\mathrm{a}}} \quad \text{for } H \le H_{\mathrm{a}},$$
$$\omega = \gamma\sqrt{\left(4\pi M_{\mathrm{s}} + H\right)\left(H - H_{\mathrm{a}}\right)} \qquad \text{for } H \ge H_{\mathrm{a}}, \tag{7}$$

where $\omega = 2\pi f$ is the frequency of variable magnetic field, γ is the gyromagnetic ratio.

In our early research [13], in addition to homogeneous FMR, we detected a narrow susceptibility peak in the field equaling the anisotropy field H_{a} in the microwave absorption spectrum of local regions of Co–Ni–P films in sweeping the constant magnetic field H along the hard magnetization axis. The FMR spectroscopic technique provided measurements within an ~1 mm^2 area determined by a diameter of the measuring hole of the microstrip resonator. When the constant magnetic field H shifted by merely 1 degree, the narrow peak disappeared. The peak width was by an order of magnitude smaller than that of homogeneous FMR peaks, the peak position being not dependent on the pumping frequency. This is depicted in Fig. 2.

In Fig. 3, the solid line indicates the dependence between the FMR resonance field and frequency calculated from Eqs. (7), and dots indicate the experimental data obtained in the HMA-oriented magnetic field. Black dots in this figure match the position of the new peak observed in the FMR spectrum. These dots form a vertical (dashed) line, which cuts the 28 Oe magnetic anisotropy H_{a} in the investigated film region. It is important to note that the peak amplitude rapidly lowers with decreasing pumping frequency below 1 GHz as a result of the peak suppression by the homogeneous FMR peaks approaching to each other. And this peak disappears already at 0.2 GHz. The peak amplitude decreases monotonely with the pumping frequency increase over 2.6 GHz. This can be explained by the skin effect.

In [13], we showed that the new peak detected in the FMR spectrum resulted from the transverse (quasi-static) magnetic susceptibility of the magnetic film and can therefore be used to measure the structure constant.

3. STRUCTURE CONSTANT DETERMINATION FROM THE MICROWAVE ABSORPTION SPECTRUM

High-frequency magnetic susceptibility χ can be calculated from the Landau–Lifshitz–Gilbert equation [18]. In the linear approximation, we have

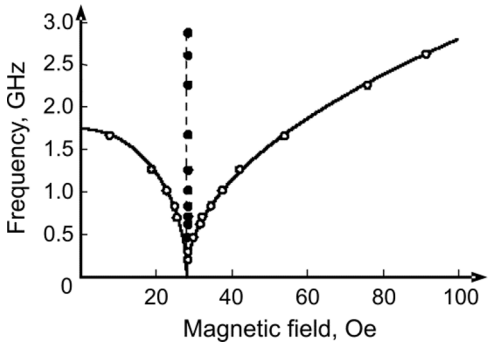

Fig. 3. Dependence between the FMR resonance field and frequency at magnetic field orthogonal to easy magnetization axis. Black dots indicate positions of the new peak in the FMR spectrum.

$$\chi = \frac{\gamma M_s(\Omega_z + i\alpha\omega)}{\omega_0^2 - \omega^2 + i\alpha\omega(\Omega_\theta + \Omega_z)}, \tag{8}$$

where the homogeneous FMR frequency is $\omega_0 = \sqrt{\Omega_z \Omega_\theta}$, $\alpha = \gamma\Delta H/(2\omega)$ is the damping parameter, ΔH is the FMR linewidth at ω frequency. In sweeping the external magnetic field along the hard magnetization axis, as illustrated in Fig. 1, Ω_z and Ω_θ values can be found from

$$\begin{aligned} &\Omega_z = \gamma(4\pi M_s + H_a); \quad \Omega_\theta = \gamma(H_a^2 - H^2)/H_a \quad \text{for } H \le H_a, \\ &\Omega_z = \gamma(4\pi M_s + H); \quad \Omega_\theta = \gamma(H - H_a) \qquad \text{for } H \ge H_a. \end{aligned} \tag{9}$$

Dividing the magnetic susceptibility χ into the real and imaginary parts $\chi = \chi' - i\chi''$, we obtain the relation for the imaginary part:

$$\chi'' = \gamma M_s \frac{\alpha\omega(\Omega_z^2 + \omega^2)}{(\omega_0^2 - \omega^2)^2 + \alpha^2\omega^2(\Omega_\theta + \Omega_z)^2}. \tag{10}$$

As is known, the energy absorption P by the magnetic film is determined by the imaginary part of the magnetic susceptibility χ'' [19]:

$$\begin{aligned} &P = \omega h_\sim^2 V \chi'' H^2 / H_a^2 \quad \text{for } H \le H_a, \\ &P = \omega h_\sim^2 V \chi'' \qquad \text{for } H \ge H_a. \end{aligned} \tag{11}$$

Figure 4*a* shows the example of the experimental differential absorption spectrum dP/dH obtained in the local region of the 300-nm-thick Co–P thin film. This spectrum was recorded by the scanning FMR spectrometer [20] at a frequency of 2274 MHz. Chemical solution deposition at 96–97°C [21] onto a 10×10 mm glass substrate was used to produce the Co–P thin film. The in-plane uniaxial magnetic anisotropy H_a was indiced by the homogeneous magnetic field (H = 3 kOe) applied to the substrate during the film deposition. The microstructure of the Co–P thin film was

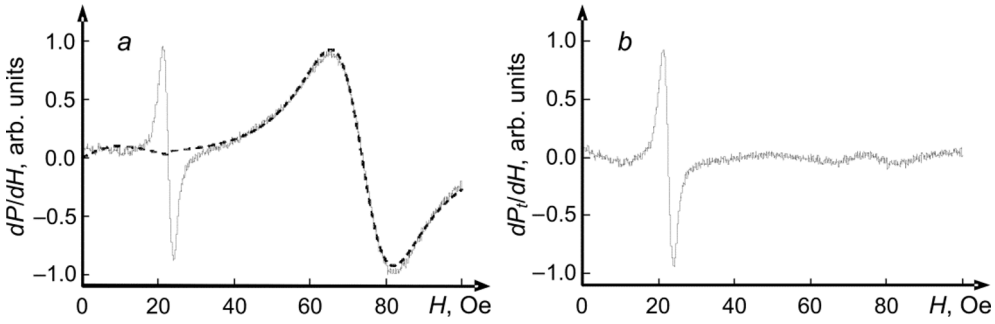

Fig. 4. Differential absorption spectrum dP/dH in the local region of the Co–P thin film (*a*). Solid and dashed lines indicate experimental and theoretical (Eqns (10), (11)) results, respectively. Extracted from the total spectrum, the differential absorption spectrum dP_t/dH is determined by the transverse (quasi-static) magnetic susceptibility (*b*).

investigated on a PREM 200 transmission electron microscope (TEM). According to TEM observations, the grain size of the Co–P thin film ranged between 2 and 6 nm.

In [22, 23] we developed a measurement technique for the determination of the magnetic parameters based on the angular dependence of the resonance field. This technique was used to obtain the main parameters of the local film region, namely: 1017.4 G saturation magnetization, 22.56 Oe uniaxial magnetic anisotropy, and 0.176 damping parameter. Based on these data and Eqns (10) and (11), the theoretical dP/dH dependence presented in Fig. 4*a* (dashed line) was calculated.

According to Fig. 4*a*, theoretical calculations are in good agreement with the experimental data on the homogeneous FMR. At the same time, the difference observed in the region $H \sim H_a$, is attributed to the transverse quasi-static magnetic susceptibility in the microwave absorption spectrum.

From the experimentally obtained differential absorption spectrum dP/dH, it is possible to extract the dP_t/dH part responsible for the transverse quasi-static magnetic susceptibility, as a difference between the theoretical and experimental curves presented in Fig. 4*b*. It is worth noting that this dependence does not result from the direct measurements of the transverse quasi-static magnetic susceptibility. Hence, the dP_t/dH dependence contains errors owing to a discrepancy between the real film and the model of a uniform, coherent rotation of magnetization. In order to minimize these errors, the spectrum overlap of the homogeneous FMR and the transverse quasi-static magnetic susceptibility must be reduced. For this purpose, it is advisable to use the pumping frequency of $f >> f_0$, where $f_0 = (\gamma/2\pi)[H_a(H_a + 4\pi M_s)]^{1/2}$ is the natural ferromagnetic resonance frequency.

In Fig. 5*a*, white circles indicate the experimental dependence between the transverse quasi-static magnetic susceptibility and the applied external magnetic field obtained by the integration of the dP_t/dH curve. The ferromagnetic resonance spectrometer records the signal of the differential magnetic susceptibility dP/dH of the thin film with accuracy up to the constant proportionality factor K. In this case, the integrated curve $P_t(H)$ is connected with $\chi_t(H)$ function as $P_t(H) = K\chi_t(H)$, where χ_t is the microwave absorption spectrum. A certain calibration is therefore required to determine the magnetization ripple parameters using the experimental curve.

According to Eq. (5),

$$P_t = K\chi_t = Ka\frac{1}{h + bh^{-1/4}}, \tag{12}$$

$$a = \frac{M_s}{H_a}, \quad b = \frac{B}{H_a^{5/4}}, \quad h = (H - H_a)/H_a,$$

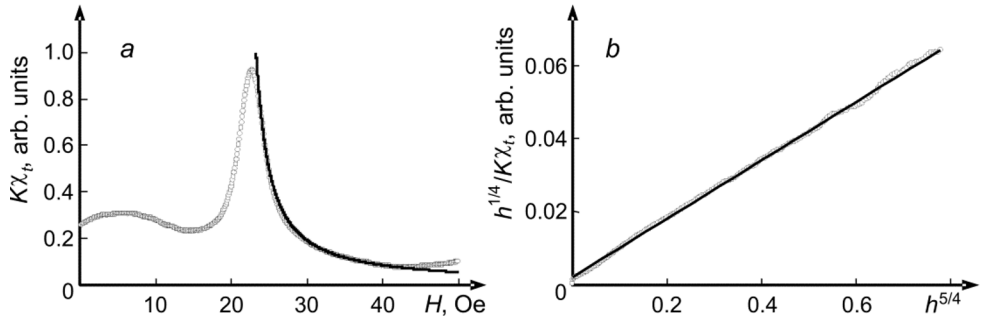

Fig. 5. Experimental field dependent transverse magnetic susceptibility $K\chi_t$ (white circles) of thin films and its theoretical approximation (solid line) in accord with Eq. (5) (*a*); experimental dependence between $h^{1/4}/K\chi_t$ and $h^{5/4}$ (white circles) and its linear approximation (solid line) (*b*).

or

$$\frac{1}{P_t} = \frac{1}{K\chi_t} = a'h + b'h^{-1/4}, \tag{13}$$

where $a' = 1/Ka$, $b' = b/Ka$.

Multiplying Eq. (13) by $h^{1/4}$, we obtain

$$h^{1/4} / K\chi_t = a'h^{5/4} + b'. \tag{14}$$

In this case, the experimental curve of the transverse quasi-static magnetic susceptibility must be linear in $x = h^{5/4}$ and $y = h^{1/4}/K\chi_t$ coordinates.

White circles in Fig. 5*b* indicate the dependence between $h^{1/4}/K\chi_t$ and $h^{5/4}$. The approximation of this dependence determines the constants a' and b' from Eq. (14), which help to derive the demagnetizing field parameter $B = bH_a^{5/4} = b'H_a^{5/4}/a'$ attributable to the magnetization ripple. The theoretical dependence $K\chi_t(H)$ given in Fig. 5*a* is in good agreement with the experimental data.

Using the tabulated data of the exchange coupling constant $A = 3\cdot10^{-6}$ erg/cm and the magnetocrystalline anisotropy constant $K_1 = 5\cdot10^6$ erg/cm^3 of cobalt [24] disregarding the internal isotropic stresses, we obtained the structure constant $S = 0.0269$. And using Eq. (2), we found the average grain size of 2.25 nm of the Co–P thin film. Note that the obtained grain size was in good agreement with the measurement results of TEM investigations [21].

CONCLUSIONS

In this study, a new technique was proposed for the determination of the structure constant. The latter was introduced within the frame of the theory of magnetization ripple to describe the influence of the real structure of nanocrystalline thin films on their magnetic properties. In contrast to the traditional method of measuring the structure constant based on the field dependent transverse magnetic susceptibility usually measured in the kilohertz range, the proposed technique suggested the use of experimentally collected FMR spectra in sweeping the magnetic field along the hard magnetization axis. The proposed technique was based on the new narrow peak of the magnetic susceptibility detected in the microwave absorption spectrum in the uniaxial magnetic anisotropy field, independently of the pumping frequency. It is interesting that the similar effect was recently discovered by Zhuravlev *et al*. [25] in $BaFe_{12}O_{19}$ hexaferrite synthesized by the sol-gel process.

Importantly, the proposed technique allowed measuring the structure constant only in the local regions of the thin film, rather than its integrated value of the bulk sample as is generally the case with the traditional technique. The local regions were determined by a diameter of the measuring hole of the microstrip resonator of a FMR spectrometer [26]. The use of the novel technique eliminated the inhomogeneity effect from the uniaxial magnetic anisotropy on the structure constant measurements, which was well pronounced at the magnetic film edges [27, 28].

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