Physics of the Solid State, Vol. 43, No. 6, 2001, pp. 1108–1111. Translated from Fizika Tverdogo Tela, Vol. 43, No. 6, 2001, pp. 1072–1075.

MAGNETISM AND FERROELECTRICITY

Spin-Wave Spectroscopy Study of Spatial Magnetization Fluctuations in Metastable Nanocrystalline Films of Fe-Based Alloys

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Received September 6, 2000; in final form, November 17, 2000

Abstract—The spectrum of standing spin waves is investigated in nanocrystalline Fe films prepared by the pulsed plasma-spraying method. The dispersion relation of these waves is determined in the wave-vector range $(0.2–3.2) \times 10^6$ cm^{-1} and is found to be affected by spatial magnetization fluctuations 100 Å in size. These fluctuations are supported as being due to the inhomogeneous distribution of C atoms in the atomic structure of nanocrystalline Fe films. © *2001 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

The objective of this paper is to investigate magnetic inhomogeneities of metastable nanocrystalline films of Fe-based alloys prepared by the pulsed plasma-spraying (PPS) method. This method allows one to produce ferromagnetic supersaturated Fe(C) solid solutions in the form of films with local fcc (hcp) structure [1, 2]. Previously, Mössbauer spectra [^{57}Co(Cr) source] at room temperature were taken for these samples doped with the ^{57}Fe isotope [3]. The spectral lines of an as-prepared film are very broad, which is characteristic of iron in the ultradisperse or amorphous state. After annealing at T = 470 K for an hour, the spectral lines become narrower and can be reasonably approximated by two Zeeman sextets with hyperfine fields H_1 = 211 kOe and H_2 = 189 kOe. Therefore, there are two different environments of Fe atoms in the material under study; that is, the Fe films are magnetically inhomogeneous. It is not easy to investigate inhomogeneous ferromagnets, in particular, to determine which parameter fluctuates [the exchange constant $\alpha(r)$, the saturation magnetization $M(r)$, the anisotropy constant $\beta(r)$, etc.] and to measure the correlation length r^* of the fluctuating parameter. Information on magnetic inhomogenety types and on their space scale can be derived from spin-wave spectra.

Theory predicts (and numerous experiments reveal) that the spectrum of spin waves in ferromagnetic films consists of several separate dispersion curves $\omega(k_n, \chi)$. The dependence of the frequency ω on the wave vector χ for waves propagating in the plane of a film can be examined experimentally by Brillouin scattering. In the case of χ = 0, the spectrum characterizes standing (across the film thickness) spin waves, which can be examined experimentally by the spin-wave resonance (SWR) method. The resonance frequencies for these spin waves are determined both by the average values of the magnetic parameters of the ferromagnetic film at hand and by the fluctuations of these parameters.

At the present time, two types of dispersion curves $\omega(k)$ (associated with two types of magnetic inhomogeneities) have been predicted theoretically and discovered experimentally in thin ferromagnetic films by the SWR method. Inhomogeneities of the first type are those characterized by an isotropic and homogeneous distribution of a fluctuating parameter such as $\alpha(r)$; the $\omega(k)$ curve in this case has a break (change in slope) "due to exchange." Inhomogeneities of the second type are fluctuations of $M(r)$, and the corresponding $\omega(k)$ curve has a break "due to magnetization." The dispersion relation for spin waves in such inhomogeneous systems has the form [4, 5]

$$\omega(k) = \omega_0 + \langle \alpha M \rangle g k^2 (1 - \gamma_i^2 J_i(k)), \qquad (1)$$

where ω_0/g is the internal field of the ferromagnet; i = α, M; and $\gamma_i^2 = (\Delta_i/i)^2$ is the strength of the fluctuating parameter i. The functions $J_\alpha(k)$ and $J_m(k)$ differ markedly near a characteristic wave vector $k^* = 1/r_i$, which is determined by the correlation length of magnetic inhomogeneities r_i; specifically, in the vicinity of k^*, the function J_α increases sharply (from 1/3 to 5/4), whereas the function J_m sharply decreases (from 1/2 to 0), and only in the region of $k = 2k^*$ does J_m start to increase (from 0 to 5/4). The difference in the behavior of $J_\alpha(k)$ and $J_m(k)$ in the region of $k \sim k^*$ allows one to experimentally reveal the dominant fluctuating parameter of the spin system of an inhomogeneous ferromag-

1063-7834/01/4306-1108$21.00

netic alloy (α or M) and to determine the correlation length of this parameter. This experimental identification can be made by spin-wave spectroscopy. Many recent SWR studies have revealed that the spin-wave dispersion relations in ferromagnetic films of amorphous alloys and inhomogeneous supersaturated solid solutions are associated with both α and M fluctuations. It was found that fluctuations of α and M are due to chemical inhomogeneities; these inhomogeneities give rise to spatial fluctuations of the exchange parameter α in inhomogeneous alloys of the transition metal–metalloid type (CoP [6], FeB [7]) and to fluctuations of the magnitude of the magnetization M in transition metal–transition metal alloys (CoZr [8], FeZr [9]).

2. SAMPLES AND EXPERIMENTAL TECHNIQUE

Nanocrystalline films of $Fe_{80}C_{20}$ and $Fe_{74}C_{20}B_6$ alloys are prepared by the PPS method in vacuum with a residual gas pressure of $P_0 = 5.5 \times 10^{-6}$ mm Hg. The substrates are made of glass. The films ranged in thickness from 300 to 3000 Å. The electronic and chemical structures of the films were determined from photoelectron and Auger spectra measured with a photoelectron spectrometer (with Mg anode) at the Institute of Semiconductor Physics (Siberian Division, Russian Academy of Sciences). The SWR spectra of nanocrystalline Fe films were examined with a standard x-band spectrometer (f = 9.2 GHz) at room temperature. These SWR spectra are presented in [2], where the boundary conditions for the magnetization of these films are also discussed. Here, we merely point out that, in the films under study, antisymmetric boundary conditions are realized for which the relation between the mode index of the SWR spectrum and the wave vector **k** of a standing spin wave has the form $k_n = \pi n/d$ (with n = 1, 2, 3, … and d being the film thickness) and the resonance fields of the SWR peaks are given by

$$H_n = \omega/\gamma + 4\pi M - \eta^{\text{eff}} k_n^2. \qquad (2)$$

Using Eq. (2), the effective exchange stiffness η^{eff} ($\eta = \alpha M$) is calculated numerically from the formula

$$\eta^{\text{eff}}(k) = (d/\pi)^2 (H_1 - H_2)/(n^2 - 1). \qquad (3)$$

A correlation is made between the $\eta^{\text{eff}}(k)$ dependence thus found and the theoretical dependence $n^{\text{eff}} = \langle\eta\rangle(1 - \gamma_i^2 J_i(k))$ (see Eq. (1) and [4, 5]) with the aim of determining the dominant fluctuating magnetic parameter i (α or M) and the correlation length r*.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Let us first discuss the experimental SWR spectra of $Fe_{74}C_{20}B_6$ films. Figure 1 shows the experimental dependence of $\delta H_{1,n} = H_1 - H_n$ on n^2 for two films of different thickness, d_1 = 1100 Å and d_2 = 960 Å. It

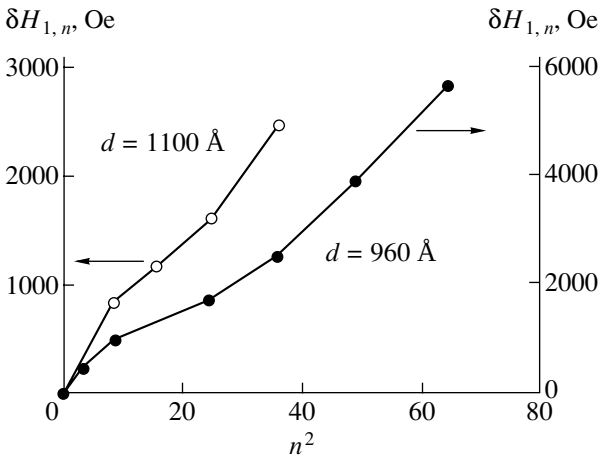

Fig. 1. Difference in the resonance fields of SWR peaks $\delta H_{1,n} = H_1 - H_n$ as a function of n^2 for two $Fe_{74}C_{20}B_6$ alloy films differing in thickness (n is the peak index).

can be seen from Fig. 1 that the $\delta H_{1,n}(n^2) \sim \omega(k^2)$ dispersion curve has a break in the long-wavelength range of the SWR spectrum: the effective exchange stiffness η^{eff} sharply decreases ($\eta_1 > \eta_2$). Therefore, according to the classification introduced in [4], this break is due to exchange. By measuring the coordinate of the break point n_i, one can determine the critical wave vector ($k = \pi n/d$); in this case, we have $k_\alpha = 1.1 \times 10^6$ cm^{-1} for both films. As the wave vector **k** increases further, one more change in the effective exchange stiffness η occurs: this time, η sharply increases ($\eta_2 < \eta_3$) and, therefore, the break is due to magnetization (in the classification of [4]). For the $Fe_{74}C_{20}B_6$ alloy at hand, the corresponding wave vector is $k_m = 1.85 \times 10^6$ cm^{-1}. As indicated above, the $\omega(k)$ dispersion curve affected by fluctuations of M and having a break due to magnetization must have two characteristic features at the wave vectors k_m and $2k_m$. In the case in question, the experimental $\delta H_{1,n}(n^2)$ curve corresponds to the portion of the theoretical $\omega(k^2)$ dispersion curve in the range of $k < 2k_m$, because the boundary wave vector k_b, corresponding to the extreme peak of the SWR spectrum, is $k_b \sim 2.2 \times 10^6$ cm^{-1} and, therefore, $k_b/k_m < 2$.

The observation of breaks of two types (those being due to exchange and to magnetization) in the dispersion curves of films of the $Fe_{74}C_{20}B_6$ alloy under study is a surprising experimental finding. The point is that this alloy belongs to the class of transition metal–metalloid alloys, the dispersion curves of which were observed earlier to show only breaks due to exchange. For instance, in films of alloys CoP [6], FeB [7], etc., the experimental $\delta H_{1,n}(n^2)$ curves obtained by the SWR method are affected only by fluctuations of the exchange constant α. For this reason, we attribute the break due to magnetization observed in the metastable

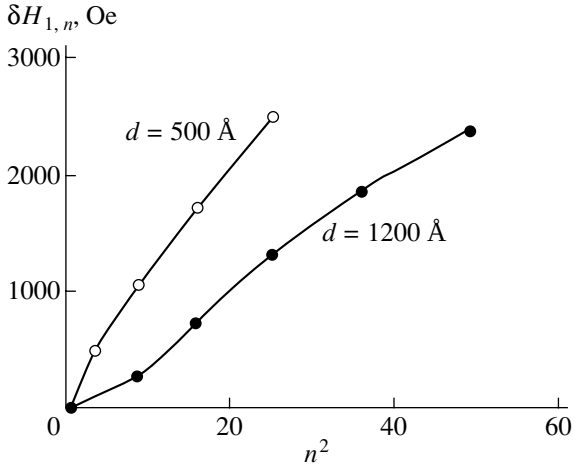

Fig. 2. Difference in the resonance fields of SWR peaks $\delta H_{1,n} = H_1 - H_n$ as a function of n^2 for two $Fe_{80}C_{20}$ alloy films differing in thickness.

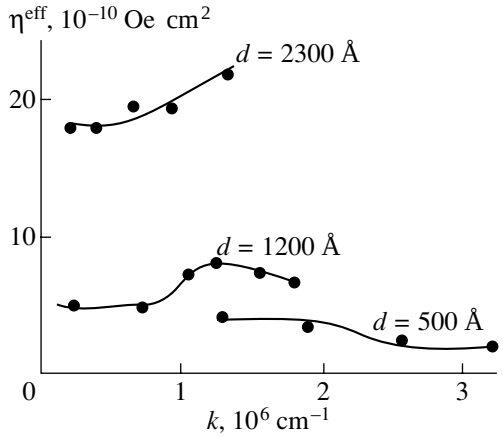

Fig. 3. Effective exchange stiffness η^{eff} as a function of wave vector **k** for $Fe_{80}C_{20}$ alloy films differing in thickness.

$Fe_{74}C_{20}B_6$ alloy to the presence of atoms of another metalloid, carbon, in the alloy. Further investigation provided support for this assumption.

Figure 2 shows the dependences of the resonance fields $\delta H_{1,n}$ on the spin-wave mode index squared n^2 for SWR spectra of two Fe(C) films differing in thickness (d_1 = 1200 Å, d_2 = 500 Å). It can be seen that the $\delta H_{1,n}(n^2) \sim \omega(k^2)$ dispersion curve of the d_1 = 1200 Å thick film has two characteristic features (breaks) at wave vectors $k' = 0.99 \times 10^6$ cm^{-1} and $k'' = 1.6 \times 10^6$ cm^{-1}. The exchange stiffness sharply increases near the characteristic wave vector k' ($\eta_1 < \eta_2$) and sharply decreases near the wave vector k'' ($\eta_2 > \eta_3$). This behavior of the $\delta H_{1,n}(n^2)$ curve suggests that the dispersion relation of spin waves in the d_1 = 1200 Å thick film of Fe(C) is affected by fluctuations of the magnetization M. This conclusion is also supported by the fact that the numerical values of the characteristic wave vectors k' and k'' approximately satisfy the theoretically predicted relation $k'' = 2k'$ (see [4]). The value of the wave vector $k' = k_m$ is determined by the scale of spatial inhomogeneities of the magnetization M in the Fe(C) alloy at hand, $k_m = 1/r_m$, where r_m is the correlation length of fluctuations of M. For films of the Fe(C) alloy, r_m is estimated to be 100 Å. It should be noted that the $\delta H_{1,n}(n^2)$ curve for the thinner (500 Å thick) film exhibits a break which is seemingly due to exchange, but in actual fact, this curve is affected by fluctuations of magnetization in the inhomogeneous Fe(C) alloy. From Fig. 2, it is seen that this $\delta H_{1,n}(n^2)$ curve has a feature near the wave vector $k' = 1.57 \times 10^6$ cm^{-1}; namely, the exchange stiffness sharply decreases in the vicinity of this point ($\eta_1 > \eta_2$). However, the range of observable wave vectors $k_n = \pi n/d$ in the SWR spectrum of the d = 500 Å thick film is such that the wave vector k_m, which characterizes the scale of fluctuations of the magnetization M, satisfies the inequalities $k_1 < k_m < k_2$. Indeed, we have $k_1 = 0.63 \times 10^6$ cm^{-1}, $k_2 = 1.26 \times 10^6$ cm^{-1}, and $k_m = 1 \times 10^6$ cm^{-1}. Therefore, the feature observed in the $\delta H_{1,n}(n^2)$ curve is basically determined by the feature of the $J_m(k)$ function in the vicinity of $2k_m$, which is also supported by the fact that the characteristic wave vector $k' = 1.57 \times 10^6$ cm^{-1} is close to the wave vector k'' at which a break is observed in the dispersion curve for the d_1 = 1200 Å thick film. We note that a similar situation was observed [4] to occur with amorphous $Co_{93}Zr_5P_2$ alloy films differing in thickness. The entire $\omega(k^2)$ dispersion curve affected by fluctuations of M and exhibiting breaks due to magnetization was first observed experimentally for those transition metal–metal alloy films.

Thus, for films of the metastable Fe(C) alloy, which is an interstitial solid solution, the experimental $\delta H_{1,n}(n^2)$ dependence agrees well with the theoretically predicted $\omega(k^2)$ dependence affected by magnetization fluctuations [4, 5]; in particular, the experimental curve exhibits breaks (changes in slope) due to magnetization. We note that the $\omega(k^2)$ curve for this Fe(C) alloy differs in character from those for analogous alloys, substitutional solid solutions, of the transition metal–metalloid system (FeB, CoP, etc.). In those alloys, the dispersion curves exhibited only breaks due to exchange.

Experimental $\delta H_{1,n}(n^2)$ dependences also allow one to calculate some other characteristics of magnetic inhomogeneities, such as the strength of fluctuations γ_m^2 and the average $\langle\eta\rangle$. The effective exchange stiffness $\eta^{eff}(k)$ calculated from the resonance fields $H_n(n^2)$ by Eq. (2) is shown in Fig. 3 for free films differing in thickness (d_1 = 500 Å, d_2 = 1200 Å, d_3 = 2300 Å). The character of the $\eta^{eff}(k)$ dependence suggests that only fluctuations of M are significant in the Fe(C) alloy. The wave vector defined by the relation $r_m = 1/k_m$ is the

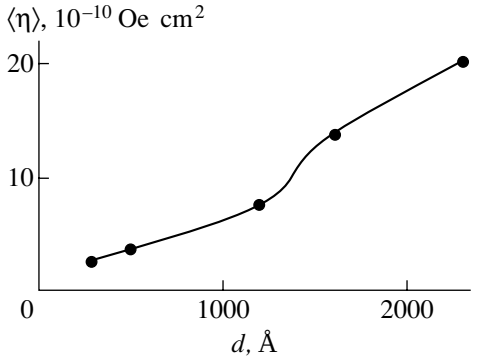

Fig. 4. Average exchange stiffness ⟨η⟩ as a function of the film thickness for an $Fe_{80}C_{20}$ alloy.

same ($k_m = 10^6$ cm^{-1}) for all films of the alloy under study, irrespective of their thickness. From theoretical expressions for the function $J_m(k)$ in Eq. (1) (see [4, 5]), it follows that the effective exchange stiffness $\eta^{\text{eff}}(k)$ measured in the wave-vector range $k_m < k < 2k_m$ is the average exchange stiffness ⟨η⟩ of the film under study. Figure 4 shows ⟨η⟩ calculated in this range of wave vectors **k** as a function of *d* for films of the Fe(C) alloy. From Fig. 4, it is seen that for the films prepared by the method indicated in Section 2, the average ⟨η⟩ depends on the film thickness *d*, which was not observed earlier [6–9]. The reason for this dependence is the following. The kinetics of solidification of an Fe(C) condensate depends on the cooling rate of the condensate, which, in turn, depends on its thickness. Therefore, the phase composition of the Fe films under study (fcc-Fe(C), hcp-Fe(C), cementite Fe_3C [1, 2]) varies with film thickness. This conclusion was supported by the findings of [10], where, by means of Mössbauer spectroscopy based on conversion electrons, it was shown that the phase composition of an Fe(C) film varies across the film. Given the average ⟨η⟩, the strength of magnetization fluctuations can be calculated from Eq. (1); the result is $\gamma_m^2 = 0.6$. It should be noted that, for Fe(C) films, γ_m^2 is independent of the film thickness. Thus, from experimental $\delta H_{1,n}(n^2)$ dependences, we calculated the correlation length r_m and made estimates of the average exchange stiffness ⟨η⟩ and the strength γ_m^2 of fluctuations of *M* in films of metastable Fe(C) alloys. It is found that in films prepared by the PPS method, γ_m^2 and r_m are independent of the thickness of the film, while ⟨η⟩ is thickness dependent. The latter property distinguishes these films from some other films, e.g., from those of CoP [6], in which all three quantities do not depend on the film thickness.

4. CONCLUSION

Thus, in this paper, the dispersion relation of spin waves in films of a nanocrystalline Fe(C) alloy is measured by the SWR method and found to be affected by spatial magnetization fluctuations approximately 100 Å in size. Fluctuations of magnetization *M* are likely to be due to the inhomogeneous distribution of C atoms in the atomic structure of nanocrystalline Fe films. The dispersion relation of the type indicated above distinguishes Fe(C) alloys, which are interstitial solid solutions, from analogous alloys of the transition metal (Fe, Co)–metalloid (B, P, Si) system, which are substitutional solid solutions and in which fluctuations of the exchange constant dominate.

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Translated by Yu. Epifanov