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Spin-Wave Resonance in One-Dimensional Magnonic Crystals by an Example of Multilayer Co–P Films

R. S. Iskhakov^a, S. V. Stolyar^b, L. A. Chekanova^a, and I. G. Vazhenina^{a, *}

^a Kirensky Institute of Physics, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia ^b Federal Research Center "Krasnoyarsk Science Center of the Siberian Branch of the Russian Academy of Sciences," Krasnoyarsk, 660036 Russia

*e-mail: irina-vazhenina@mail.ru

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Abstract—Layered magnetic films of Co–P alloy, which are sets of laminates with certain atomic and chemical structures, have been investigated by spin-wave resonance. Coatings of two types were synthesized: a magnonic crystal (amorphous Co(P)/fcc Co(P))_N and gradient Co(P) films. They exhibit two different modifications of the spin-wave resonance (SWR) spectrum. In the first case, the SWR spectrum is described as $H_n(n) \sim n^2$ with a modification caused by the formation of the first stop band of the magnonic crystal. In the second case, the dependence of resonance fields H_n of the spin-wave modes on mode number *n* has the form of $H_n(n) \sim n^{2/3}$. Thermal annealing leads to crystallization (amorphous alloy \rightarrow fcc) and causes transformation of the layered films to the films of single-phase Co–P alloys, which is accompanied by a change in the shapes of the SWR spectra and occurrence of a "exchange kink" spectral modification.

Keywords: spin-wave resonance, surface-anisotropy constant, exchange-interaction constant, amorphous and nanocrystalline alloys

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1. INTRODUCTION

Spectra of waves in periodic structures have a band character with forbidden and allowed energies. In each allowed energy band, the dispersion relation for spin waves is described by a quadratic dependence in the form $\omega \sim k^2$. It is known that the dispersion relation of spin waves in a ferromagnet is $\omega \sim k^2$; therefore, the dependences of the positions of the resonance fields of standing exchange spin waves H_n on mode number *n* in the spin-wave resonance (SWR) spectra of ferromagnetic films are discrete: $H_n \sim n^2$ [1–3]. A particular solution of the magnetization equation for ferromagnetic films is a harmonic function [4]. If the experimental range of wave vectors in magnonic crystals covers several allowed energy bands, the edges of these bands can be observed in the recorded SWR spectra as a doublet of the mode with wave vector k_{critic} [5, 6].

During the first decades after the discovery of the SWR phenomenon in thin magnetic films [1, 3], the related studies mainly addressed homogeneous thin films (100-400 nm) with quadratic dependence $H_n(n)$, because one could determine in a relatively simple way the exchange hardness of spin waves by measuring dependence $H_n \sim n^2$. However, at the same time, nonquadratic dependences $H_n(n)$ were observed

experimentally for ferromagnetic films, and a number of theoretical studies [7-11] were devoted to interpretation of the experimental results of [11–16]. Two mechanisms leading to deviations from a quadratic law can be distinguished. The first one yields deviations from the dependence $H_n \sim n^2$ in a narrow wave range around the characteristic wave vector k_c and is related to random distributions of magnetic parameters (exchange and magnetization) in the film bulk. In the case of dominance of some magnetic parameter, one could determine (based on the form of the modification of the experimental SWR spectrum) not only the type of this parameter but also some statistical characteristics (average magnetic parameter, its dispersion, and correlation radius $r_C \approx 1/k_C$ [9–11]. The second mechanism, which modifies the SWR spectra in the entire wave range, is due to the dominance of the magnetic-parameter distribution described by some functional dependence along the film thickness over random fluctuations. Portis [7] suggested a parabolic change in the magnetization along the film thickness and explained the linear dependence $H_n(n)$ for FeNi films, which was observed experimentally by Kooi [12]. Schlömann [8] interpreted the experiments by Nisenoff and Terhune [13] on Permalloy films proceeding from a linear change in the effective field



Fig. 1. (a) Concentration dependences of the exchange constant and saturation magnetization in Co–P alloys [37] and (b) geometry of the experiment.

along the film thickness, which yielded the theoretical dependence of the resonance fields on the mode number in the form $H_n \sim n^{2/3}$ corresponding to the experimental one.

In the recent years, a lot of experimental studies [17–20] were devoted to the mechanisms causing the dependences in the form $H_n \sim n$ and $H_n \sim n^{2/3}$ in the SWR spectra. A distinctive feature of those studies is the use of single-layer films, in which the spatial distribution of the magnetic parameters along the film thickness is hard to be monitored; therefore, different results were often obtained for identical systems [21, 22].

Therefore, specific features of microwave spectra are often studied using a laminate (multilaver) film, which makes it possible to form desired atomic, chemical, and magnetic structures of a ferromagnetic film. As successful examples of this approach, one can refer to works [5, 6, 23-28], in which the spectrum of spin waves in a magnonic crystal and an effective medium was studied. It was found that the SWR method makes it possible to identify changes in not only magnetic but also structural parameters of a ferromagnetic system [29–34]. The latter possibility is due to the fact that the fundamental parameters of a spin system (such as exchange-interaction constant A, effective magnetization $M_{\rm eff}$, and anisotropy constant K), which in turn depend on the atomic and chemical structures of the material, can be determined by the SWR method.

The purpose of this study was to analyze (by an example of multilayer Co(P) films) compositionally modulated structures with a specified distribution of transformation of the structure from nanocrystalline to amorphous, which made it possible to obtain films of two types: a one-dimensional magnonic crystal and a gradient structure. The implemented modulations

allowed us to record two types of modifications of the exchange spin wave spectrum in microwave spectra by the SWR method: the first modification is due to the first stop band of the magnonic crystal, whereas the second one is characterized by the distribution of resonance fields in the form $H_n \sim n^{2/3}$.

2. EXPERIMENTAL

The layered (laminate) films of Co-P alloys under study were synthesized by chemical vapor deposition from solutions of salts of the corresponding metals. The influence of phosphorus content on both the atomic structure of Co-P alloys and the magnetic characteristics of Co-P alloy films caused by the atomic and chemical structures therein was investigated previously in many studies [32, 33, 35, 36]. The results of studying single-layer films suggest the influence of P content in Co–P alloy on the change in the alloy phase state (hcp and fcc atomic lattices and amorphous structure). These structural transformations affect the main fundamental magnetic characteristics (exchange, magnetization, and anisotropy) and are most pronounced in the concentration dependence of the exchange-interaction constant in the range of 0–10 at % P (Fig. 1a).

We synthesized two sets of layered films with different concentration profiles along the film thickness. Samples from the first set are laminate structures with alternating layers of amorphous (90 at % Co and 10 at % P) and nanocrystalline (93 at % Co and 7 at % P) Co–P alloy, which made it possible to form a periodic step profile of a change in the phosphorus concentration along the sample thickness (Fig. 2a). Thus, multilayer $[Co_{90}^{am}P_{10}(d_1)/Co_{93}^{crystal}P_7(d_2)]_7$ films are structures, all magnetic parameters of which are peri-



Fig. 2. (a) Step profile at variation in the phosphorus content and (b) phosphorus distribution along the gradient-film thickness.

odically modulated (i.e., magnonic crystals). Therefore, their spectrum should have a band character with allowed and forbidden energies for spin waves. The edges of the Brillouin zones are determined by the wave vectors $k_{\text{critic}} = \pi (d_1 + d_2)^{-1}$. The ranges of magnitudes of these vectors for the first and second allowed bands were brought into correspondence with the ranges of magnitudes of the measured wave vectors. Having chosen thicknesses of individual layers $(d_1 = d_2 = 15 \text{ nm}, \text{ where } d_1 + d_2 \text{ is the one-dimensional}$ modulation period) and number of pairs (N = 7) in the synthesized samples, we could form the first and second Brillouin zones with the edge at the 7th mode, so that the value of the forbidden gap in the field coordinates is described by a doublet of SWR modes at n = 7.

The second set is also comprised of layered films of the same alloy; however, the phosphorus concentration profile along the coating thickness was chosen so that to make the concentration function (at % P) linear. In this case, a transition from the amorphous to nanocrystalline state (or vice versa) (Fig. 2b) divides the thickness of the Co-P alloy film into three regions: amorphous-nano fcc-amorphous (or nano fcc-amorphous-nano fcc). The thickness of individual layers in the gradient films was varied from 30 to 35 nm and the number of the layers was 9. Note that this chemical structure can be considered as a scaled boundary between the amorphous and crystalline layers (Fig. 2a), which makes it possible to study the effects occurring at the interface between the layers (d_1 and d_2).

The room-temperature ferromagnetic resonance (FMR) and SWR spectra of the films were recorded on a standard EPA-2M spectrometer with a pump frequency of 9.2 GHz. The dynamic characteristics were measured under external magnetic fields oriented parallel ($\theta_H = 90^\circ$) and normally ($\theta_H = 0^\circ$) with respect to the surface (Fig. 1b). The microwave-absorption curves with a complex shape (Fig. 3) were decom-

posed into components using the differentiated Lorenz function, which was chosen taking into account the absence of electrical-component contribution (due to cavity design and sample sizes).

3. RESULTS AND DISCUSSION

It is known that, under certain boundary conditions [1, 38], standing exchange spin waves can be excited in a thin homogeneous ferromagnetic film under constant (**H**) and uniform alternating (\mathbf{h}_{\sim}) magnetic fields. When the external magnetic field **H** is applied perpendicular to the film plane ($\mathbf{H} \perp \mathbf{h}_{\sim}$), the positions of the resonance fields H_n are described by the expression

$$H_n = \frac{\omega}{\gamma} + 4\pi M_{\rm eff} - \frac{2A}{M_S}k^2, \qquad (1)$$

where $\gamma = 1.758 \times 10^7$ Hz/Oe is the gyromagnetic ratio, ω is the fixed microwave-field frequency, M_{eff} is the effective magnetization, A is the exchange-interaction constant determined by the expression $A = 2JS^2/a$ and related to the exchange hardness η ($\eta = 2A/M_s$), and k is the wave vector ($k = n\pi/d$, where n is the mode number and d is the film thickness). The dependence of H_n on n^2 can be plotted based on the experimental SWR curve by identifying n according to the rules described in detail in [2, 39]. The possibility of using expression (1) for describing standing spin waves in multilayer films was substantiated in [6, 31, 40–44].

The positions of the resonance fields of standing exchange spin waves (in an inhomogeneous medium with fluctuating magnetic parameters) in nanocrystalline and amorphous films are described by the expression

$$H_n = \frac{\omega}{\gamma} + 4\pi M_{\text{eff}} - \langle \eta \rangle k^2 (1 - \gamma_i^2 J_i(k)), \qquad (2)$$



Fig. 3. SWR spectra (a, b) before and (c, d) after annealing for the (a, c) layered $[Co_{90}^{am}P_{10}(15 \text{ nm})/Co_{93}^{crystal}P_7(15 \text{ nm})]_7$ film and (b, d) gradient film $[Co_X P_Y]_N$.

where the function $J_i(k)$ describes the influence of fluctuations, which manifest themselves in deviations (kinks) of the dependence $H_n(n^2)$ from a straight line at the characteristic wave vector $k^*=1//r_C$ [9–11], and γ_i^2 is the mean square fluctuation of the spin parameter. To analyze the results of this study, exchange fluctuations ("exchange" kink) should be selected among possible modifications of the dispersion relation, which are described by (2); the presence of these fluctuations can be identified based on the decrease in the slope of the dependence $H_n(n^2)$. For example, the SWR spectra recorded for reference single-layer Co– P films (both nano fcc and amorphous ones) were described by the nonstandard Kittel's ratio $H_n \sim n^2$ with the exchange kink in the vicinity of $k \sim k_c$.

If the effective field in the magnetic structure of the film changes linearly [8], the analytical expression relating the positions of resonance fields H_n and the mode number is as follows:

$$H_n = H_0 - \left(\frac{2A}{M}\right)^{1/3} \left[\frac{3\pi}{2}\left(n + \frac{1}{4}\right)\right]^{2/3} \left(\frac{H_{\text{grad}}}{d}\right)^{2/3}, \quad (3)$$

where H_0 is the effective field of uniform magnetization precession and H_{grad} is the parameter characterizing the magnetic-field gradient along the sample thickness.

Random (natural) and artificial inhomogeneities in thin films manifest themselves especially clearly as characteristic features in the dependence of exchange hardness $\eta^{\text{eff}}(n)$, which is described by the following fluctuat

$$\eta^{\rm eff} = \frac{H_1 - H_j}{n_j^2 - 1} \left(\frac{d}{\pi}\right)^2.$$
 (4)

For implementation of the Brillouin quasi-zone, a specific feature of $\eta^{\text{eff}}(n)$ is a decrease in the η value for the waves with $k \le k_{\text{critic}}$, a step in the η value at $k = k_{\text{critic}}$, and a further decrease in η to the mean exchange hardness $\langle \eta \rangle$ for the waves with $k \ge k_{\text{critic}}$. The magnitude of the critical wave vector is described by the relation

expression:

$$k_{\rm critic} = \frac{n\pi}{N(d_1 + d_2)},\tag{5}$$

in which the number of bilayers N equals to the mode number n.

Exchange fluctuations are characterized by a decrease in the $\eta^{\text{eff}}(n)$ value in the vicinity of k_c^* [9–11, 45, 46].

Figures 3a and 3c show the experimental spectra of layered $[Co_{90}^{am}P_{10}(15 \text{ nm})/Co_{93}^{crystal}P_7(15 \text{ nm})]_7$ films with the above-described specific features: a doublet at the 7th mode (energy gap between the first and second Brillouin zones) is implemented before annealing and "exchange" kink after the annealing, which crystallizes the amorphous alloy. The structures of all the recorded spectra have a number of specific features. First, the presence of surface modes (S1 and S2 in Fig. 3) indicates antisymmetric pinning of surface spins, when the hard axis of surface anisotropy is normal to the film surface (the surface-anisotropy constant is $K_S < 0$ [47]. The provided boundary conditions lead to implementation of even modes in the spectra, which are much weaker than the neighboring odd modes. The specified synthesis conditions (the number of layers and their thicknesses) make it possible to observe the formation of a doublet of the seventh mode, which corresponds in the field coordinates to the edges of the band gap in the spin-wave spectrum. The modes with numbers *n* equal to 1, 3, 4, 5, and 7 characterize the first Brillouin guasi-zone, while the modes with numbers 7, 9, and 11 are related to the second quasi-zone. It should be noted that expression (1) describes the positions of the resonance fields within each band in dependence of the mode number (Fig. 4a). Band gap ΔH_B in the field coordinates is equal to the difference between the resonance fields of the seventh modes (~ 500 Oe). The manifestation of the band-gap edges in the SWR spectrum for structures made of Co–P alloy was also observed in [5]; however, the adjacent layers had a nanocrystalline structure (hcp Co/fcc Co). The band gap in this magnonic crystal was measured to be ~200 Oe. The data obtained may indicate that the difference in the ΔH_B values can be caused by not only different types of the

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fluctuating parameter (see [5]) but also different atomic structures of the alternating layers.

The identification of the spin-wave mode numbers shown in Fig. 3a and expression (4) allowed us to determine effective spin-wave hardness η^{eff} ; its dependence on the mode number is given in the inset in Fig. 4a. The presence of two individual branches in the dependence $\eta^{\text{eff}}(n)$ (Fig. 4a) with the edge at the 7th mode is in good agreement with the previously described specific features and the k_{critic} value determined from (5).

Thus, the SWR spectrum in the form of a step concentration function was recorded for a one-dimensional periodic magnonic crystal formed as a multilayer film with the structure modulation of the "nanocrystalline ferromagnet/amorphous ferromagnet" type; its processing made it possible to determine the modification of the spin-wave spectrum caused by the formation of the first and second Brillouin zones of the magnonic crystal.

Note that the interface between layers is not atomically sharp (the interface thickness is 1-2 nm) because of technological reasons (chemical vapor deposition). However, based on the SWR-spectrum modifications, one can distinguish the contributions from the layer periodicity and the interface by choosing an appropriate relation between the modulation period and the recorded wave vectors.

The interface-induced effects were investigated on a gradient film with a linear profile of change in the phosphorus concentration along the film thickness. The experimental SWR spectrum of the gradient film is shown in Fig. 3b. Dependence of the position of the resonance fields on the mode number coincide with high accuracy with the dependence $(H_1 - H_n) \sim n^{2/3}$ (Fig. 4b).

Crystallization (amorphous state \rightarrow fcc) annealing of both the periodic magnonic crystal and the gradient film induced the formation of a macroscopically homogeneous film of Co–P alloy with nanoscale fluctuations. Thus, one-dimensional periodic (aperiodic) modulation of a composite structure was replaced by isotropic fluctuations of the magnetic parameter.

The experimental SWR spectra after thermal annealing are shown in Figs. 3c and 3d. The dependences of the position of the resonance fields on the squared mode number (Figs. 4c and 4d) demonstrate the SWR-spectrum modification caused by the dominance of the exchange-interaction constant in random fluctuations of the magnetic parameters of the system [11, 15, 16, 46]. The form of the dependence $\eta^{\text{eff}}(n)$ for each sample after annealing (Figs. 4c, 4d, insets) also confirms the dominant role of the nonuniformity of the exchange parameter. The values of effective magnetization M_{eff} , exchange-interaction constant A, and surface-anisotropy constant K_S were calculated at the



Fig. 4. Dependences of the position of resonance field H_i on mode number *n* (a, b) before and (c, d) after annealing for the (a, c) layered film and (b, d) gradient film $[Co_X P_Y]_N$. The dependences of the effective exchange hardness on the mode number are shown in the insets.

quadratic portions of the dependence $H_n(n^2)$ using (1), and the exchange inhomogeneity scale (correlation radius r_c) was determined based on the wave-vector magnitude k^* at the kink point (see Table 1).

4. CONCLUSIONS

Modifications of the spectrum of standing spin exchange waves caused by both the formation of the first and second Brillouin zones (magnonic Co–P crystal at equal critical wave vector k_c and wave vector k_B of the Brillouin-zone edge) and the formed linear profile of the internal effective field ($H_n \sim n^{2/3}$) were found experimentally in multilayer films of the nanocrystalline ferromagnet/amorphous ferromagnet type (fcc Co(P)/amorphous Co(P)) and layered films with a gradient chemical structure by the SWR method. It was also established that the crystallization (amorphous alloy \rightarrow fcc Co(P)) thermal annealing of layered Co-P films induces transformation to the structure of disordered fcc solid solution with almost isotropic inhomogeneous spin system, which manifests itself in the corresponding modifications of the dispersion relation of exchange spin waves. The results obtained suggest that the differences in the structural types of the adjacent layers (fcc-hcp or fcc-amorphous state) affect significantly the magnetic parameters of the system (energy gap between the first and second Brillouin zones in the field coordinates ΔH_{R} before annealing and correlation radius r_c after annealing).

| Table | 1. | Magnetic | parameters | of the | films | before | and | after | annealing |
|-------|----|----------|------------|--------|-------|--------|-----|-------|-----------|
|-------|----|----------|------------|--------|-------|--------|-----|-------|-----------|

| | $M_{\rm eff},{ m G}$ | $A \times 10^{-6}$, erg/cm | $ K_{S1} $, erg/cm ² | $ K_{S2} $, erg/cm ² | <i>r</i> _{<i>C</i>} , nm | | | | | | |
|-----------------------------------------------------------------------|----------------------|-----------------------------|----------------------------------|----------------------------------|-----------------------------------|--|--|--|--|--|--|
| Before annealing | | | | | | | | | | | |
| [Co ^{am} (15 nm)/Co ^{crystal} (15 nm)] ₇ | 1366 | ~0.95 | ~0.67 | ~0.32 | | | | | | | |
| After annealing | | | | | | | | | | | |
| [Co ^{am} (15 nm)/Co ^{crystal} (15 nm)] ₇ | 1372 | ~0.65 | ~0.39 | ~0.17 | ~26 | | | | | | |
| Gradient film $[Co_X P_Y]_N$ | 1274 | ~0.378 | ~0.22 | ~0.16 | ~22 | | | | | | |

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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