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FMR Line Splitting in Fe/Cr Multilayers

L.P. AKINSHINA, A.V. KOBELEV[1]), A.A. KOSHTA, M.A. MILYAEV, V.I. MININ, and A.A. ROMANYUKHA

Institute for Metal Physics, Russian Academy of Sciences, Urals Division, 620219 Yekaterinburg, Russia

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Measurements of the magnetization curves and ferromagnetic resonance (FMR) were made on a series of samples of multilayers $[Fe(13\ Å)/Cr(t_{Cr})]_{12}$, with $7\ Å < t_{Cr} < 40\ Å$, deposited by the method of molecular-beam epitaxy on a sapphire substrate with the Fe buffer layer of 40 Å thickness. The effective demagnetizing field $4\pi M_s - H'_u$ was estimated for all samples from the angular dependence of an FMR resonant field and linewidth for the uniform mode at the transverse excitation, where M_s is the saturation magnetization of the Fe layer, H'_u is the uniaxial anisotropy field. An additional FMR line was observed for samples with small spacer thickness ($t_{Cr} < 16$ Å). Its intensity was equal to that of a main line under the condition of $t_{Cr} \approx t_{Fe} \approx 12$ Å, decreasing down to zero both at reduction, and at the increase of t_{Cr}. The FMR lineshape in a field parallel to the surface was simulated by a superposition of two curves, possessing in view of the magnetization curve approximately Lorentzian shape with different effective field values H'_u. The possible reason of the features observed can be the formation of regions with different effective anisotropy field values in a sample plane during the deposition. For all samples investigated at longitudinal excitation, an FMR signal which corresponds to an optical mode was not observed.

1. Introduction

Multilayer metal structures are intensively investigated in connection with the phenomenon of giant magnetoconductivity. This causes the interest in the magnetic properties and structure of multilayers. In superlattices consisting of magnetic (Fe) and non-magnetic (Cr) layers of metals, in the absence of an external field, ferromagnetic, antiferromagnetic or non-collinear magnetic ordering of magnetic layers has been found [1, 2]. The magnetic ordering depends on the thickness of the non-magnetic spacer, because, as had been shown by various experiments, the exchange parameter oscillates as a function of spatial coordinate in these systems.

On the other hand, the investigations of a thin structure of the multilayers revealed their columnar crystal morphology, that is connected coherently with the structure of the substrate. This structure changes with the increase of Cr layer thickness [3]. The interlayer boundary also has a complex structure, due to the formation of various magnetic phases [4, 5].

The type of magnetic order in these systems can be determined by static magnetic measurements of the magnetization curve in a field parallel to the surface, and in some cases it gives the quantitative information on interlayer exchange parameters. Ferromagnetic resonance (FMR) is more sensitive to the details of a magnetic structure.

[1]) Corresponding author: e-mail: Kobelev@ifm.ural.ru

However, this method does not determine the magnetic parameters directly. Moreover, the interpretation of the shapes of the FMR spectra is a separate task which requires the respective model. The research on the same multilayers by other methods is also useful.

For the study of antiferromagnetic and non-collinear magnetic structure in multilayers the excitation of an optical mode at longitudinal pumping [7] has special significance. The observation of an optical mode may confirm the existence of two magnetic sublattices and may help to define the parameters of exchange coupling. Some particular peculiarities of an optical mode excitation in the model of biquadratic exchange were predicted in [8], and this should be verified experimentally.

The purpose of the present work is to consider the mentioned problems by studying a series of Fe/Cr multilayers with the variable thickness of a Cr layer. We have worked out an approach described further to obtain information about the magnetic structure of the multilayers from the FMR and magnetization curve measurements. The results of structural research [3] and Mössbauer effect [4] for the same samples were invoked to interpret the FMR data.

2. Samples

The method of molecular-beam epitaxy was used to obtain the Fe/Cr multilayers. The single crystal sapphire Al_2O_3 substrate with orientation $[21\bar{1}0]$ was used. The substrate, however, does not play the main role in the process of epitaxial growth of the multilayer. The buffer layer of Fe with a thickness of about 40 Å was put on the substrate. Such a thickness was chosen to provide the transition from the hexagonal crystal structure of the substrate surface to the cubic structure of the multilayer Fe/Cr film, which has orientation $\{100\}$. We have used the Fe buffer layer for the reason that it grows better than, for instance, the Cr one, although the latter grows smoother and does not add to the magnetic parameters of the sample. Later we have realized that the Cr buffers have some advantages, and the results of the studies on these samples, being not completed as yet, have to be published elsewhere.

The multilayer Fe/Cr structure was deposited on the buffer layer, with the number of layers hold constant (both 12 layers of Fe and Cr). The thickness of the Fe layer is kept constant (≈13 Å), whereas the thickness of Cr layers changes from 7.7 up to 40 Å. These values of the spacer thickness were chosen to ensure magnetic ordering over the

Table 1
List of the samples investigated

sample	buffer layer (Å)	t_{Fe} (Å)	t_{Cr} (Å)
1	42	14.6	7.7
2	40	13.0	9.0
3	42	12.3	11.1
4	36	11.2	14.5
5	40	12.2	18.8
6	36	10.8	22.0
7	42	12.9	29.6
8	40	11.1	35.1

whole range starting with ferromagnetic order at small $t_{Cr} < 7$ Å, through non-collinear, and to antiferromagnetic order at $t_{Cr} > 30$ Å. At the last stage, the multilayer structure was covered by an additional layer of Cr for stabilization. The nominal values of layer thickness were estimated from the time and speed of deposition. The optimal deposition parameters were defined from the preliminary deposition of thick layers of Cr and Fe. The values of thickness of the layers t_{Cr} and t_{Fe} were determined by X-ray diffraction and micro-structural X-ray analyses of the samples (Table 1).

3. Experimental

The FMR spectra were recorded at room temperature with an ERS-231 spectrometer (produced in the former GDR) operating in the X-band. For the computer treatment of spectral files this spectrometer was connected with an IBM-PC. The experimental parameters of the spectra recording were the following: 100 kHz modulation frequency, 10^{-4} T modulation amplitude, 5 T field sweep. The magnetic system allowed to vary the magnetic field strength in the range from 0.01 up to 1.0 T.

The cylindrical microwave cavity TE_{102} has been used, which allows to carry out measurements of the FMR signal at the transverse excitation (direction of the static magnetic field being perpendicular to the magnetic vector of the microwave field). The angular dependences of FMR signal parameters were measured by a rotation of the electromagnet relative to a fixed microwave cavity (Fig. 1a). The rotation of a magnetic field over an angle θ from 0 to 360°, with respect to the sample plane, was executed with an accuracy of 0.1°.

The rectangular cavity TE_{102} has been used to detect an optical mode, in which the type of FMR excitation varies from longitudinal to transverse by the rotation of the magnet, at fixed position of the sample in the cavity (Fig. 1b).

The magnetization curves and hysteresis loops were measured for all investigated samples by a standard Faraday method on a vibrating sample magnetometer in a range of field 0 kOe $< H <$ 18 kOe. The samples of size 5×5 mm^2 were oriented by their surface parallel to the magnetic field direction. The normalized magnetization curves

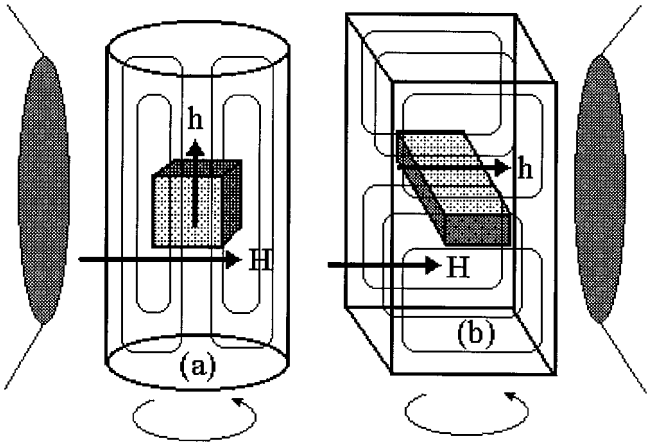

Fig. 1. Microwave cavities used for the FMR investigations at a) transverse and b) longtitudinal excitation

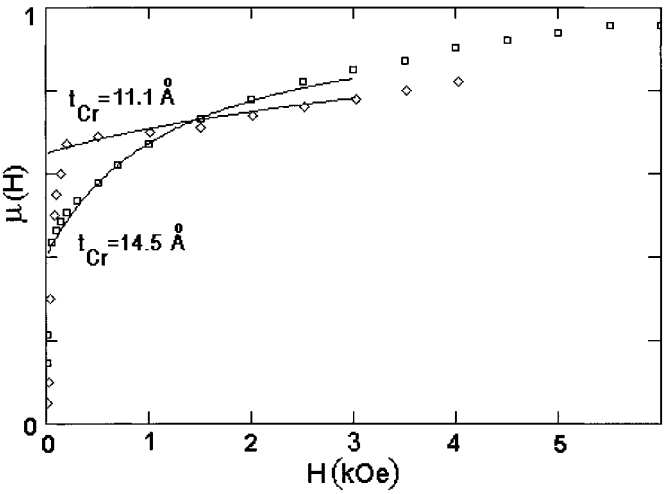

Fig. 2. Relative magnetization curves $\mu(H)$ for the sample with spacer thickness t_{Cr} = 11.1 Å (◇) and 14.5 Å (□). Solid lines are approximations (eq. (5) with m_0 = 0.65, H_b = 5 kOe and m_0 = 0.4, H_b = 1.2 kOe, respectively)

$\mu(H)$ were obtained (Fig. 2) using high fields, approaching approximately saturation magnetization.

4. Assumptions

According to our experimental conditions, we assume that only magnetically uniform modes are excited, and that the sample can be treated as an infinitely-layered one. In general, it is sufficient to consider an infinite antiferromagnet with two sublattices composed of Fe layers with different magnetization directions [8]. The Cr layer is treated as a non-magnetic spacer. Non-collinear ordering can be described by a combination of bilinear and biquadratic interlayer exchange interaction. The acoustic mode in this model, contrary to an optical one, does practically not depend on the interlayer exchange. Therefore, we assume further that the processing of the angular dependence of the FMR field and the linewidth can be done in the usual way [16, 18]. A possible error may be caused in this case by the factor $\mu(H)$, so that estimated values of the effective anisotropy field received from angular dependences are rather uncertain.

5. Modeling

The calculations of the resonant field and linewidth as functions of the angle θ were made in the model, taking into account the difference in the directions of magnetization and magnetic field vectors. In the samples investigated the values of demagnetizing fields exceed the fields of anisotropy, so it is possible to speak in general about an "easy plane" type of anisotropy. In the absence of an external static field the magnetization vector lies in the sample plane. At an increase of the field, directed normally to the surface, magnetization leaves the plane. At the turn of the external field off a sample plane up to the normal the directions of magnetization and external field vectors do not coincide. This effect results in a slower increase of the resonant field H_r when θ

increases from zero ($\mathbf{H_0}$ is in the film plane) up to 90°. The size of this backlog is proportional to an effective anisotropy constant and, consequently, by fitting of the angular dependence $H_r(\theta)$ to experimental data it is possible to evaluate it. From the standard equation describing uniform FMR [16] we have

$$(\omega/\gamma)^2 = [H_r(\theta)\cos(\theta - \theta_m) - H_u \sin^2\theta]\,[H_r(\theta)\cos(\theta - \theta_m) + H_u \cos 2\theta], \quad (1)$$

where θ is the angle of the magnetic field vector, θ_m is the angle of the magnetization vector relative to a surface normal, $H_u = 4\pi M_s - H'_u$ where H'_u is the effective field of uniaxial anisotropy, M_s the saturation magnetization of an iron layer and γ the gyromagnetic ratio. The equation relating θ and θ_m is [17]

$$2H_r(\theta)\sin(\theta_m - \theta) = -H_u \sin 2\theta_m . \quad (2)$$

The parameters γ and H_u were estimated by fitting of experimental data with theoretical dependences following from (1) and (2). Good agreement was achieved generally. The values of γ were found close to the free electron value.

The angular dependence of FMR linewidth $dH(\theta)$ was described by the expression [18]

$$dH(\theta) = H_r(\omega(1 + \alpha)) - H_r(\omega(1 - \alpha)), \quad (3)$$

where $H_r(\omega)$ denotes the FMR resonance field as a function of frequency, α is the Gilbert relaxation parameter, ranking as the fitting parameter and related with the FMR linewidth by the expression

$$dH(0) = 2\alpha\omega/\gamma .$$

The modeling of the FMR lineshape was carried out for the case of a static field, lying in the sample plane. It was assumed, that two values of anisotropy fields H_{u1} and H_{u2} are present. Thus, a signal is excited with a lineshape, characteristic of a multilayer (transverse acoustic mode), which is a superposition of signals from two subsystems with the weight factors k_1 and k_2,

$$P(H) = k_1 P(H, H_{u1}) + k_2 P(H, H_{u2}), \quad (4)$$

$$P(H, H_u) = (\partial/\partial H)\,\mathrm{Im}\,\chi_a(H, H_u),$$

$$\mathrm{Im}\,\chi_a(H, H_u) = \frac{[H - H_u\mu(H)]^2 + \mu^2(H)\,(\omega/\gamma)^2\,(1 + \alpha^2)}{\{[H(H - H_u\mu(H)] - (\omega/\gamma)^2\,(1 + \alpha^2)\}^2 + \alpha^2(\omega/\gamma)^2 A^2(H, H_u)},$$

$$A(H, H_u) = H[\mu(H) + 1/\mu(H)] - H_u .$$

Expression (4) differs from the expression valid in the ordinary ferromagnet by the function $\mu(H)$, describing the relative magnetization of a multilayer structure, formed by alternating layers of magnetic and non-magnetic metals. Expression (4) is adequate for the joint movement of the total moment of sublattices. This formula is sufficient by general and it is suitable for the description of a similar mode in a magnet with arbitrary magnetic structure, characterized by the magnetization $\mu(H)$. As a convenient trial function, approximating the experimentally received magnetization, the function depen-

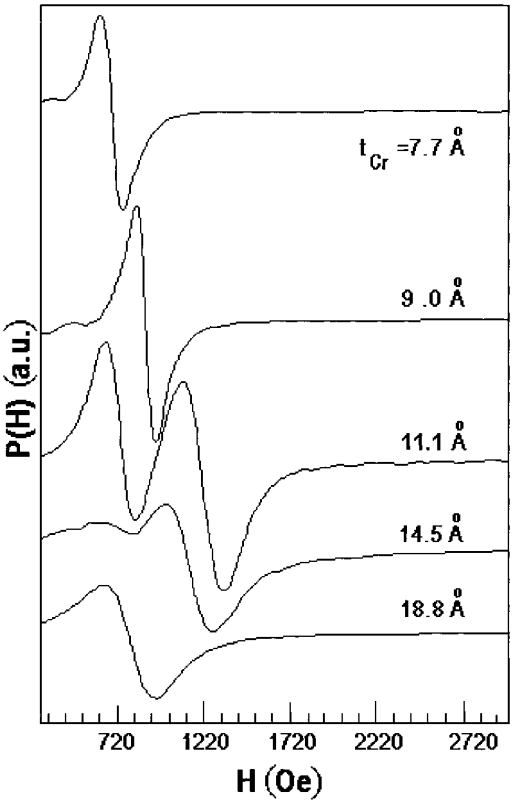

Fig. 3. Evolution of the FMR spectra at the transverse excitation in the static field parallel to the film surface for a series of samples with varying spacer thickness t_{Cr}

dent on the two following parameters m_0 and H_b has been used:

$$\mu(H) = \frac{m_0 + H/H_b}{1 + H/H_b} . \qquad (5)$$

The parameter H_b can be treated as the saturation field, and m_0 is the zero-field relative magnetization.

6. Results

None of the investigated samples has an FMR signal at parallel excitation. However, at transition to mixed excitation by the turn of the magnetic field relative to the sample, when the transverse component of the high-frequency field being present, the resonance signal was observed.

At completely transverse excitation all samples have an intense and well resolved single, or a splitted FMR line shape. The evolution of the FMR spectrum for the samples with increasing spacer thickness t_{Cr} is presented in Fig. 3. At small spacer

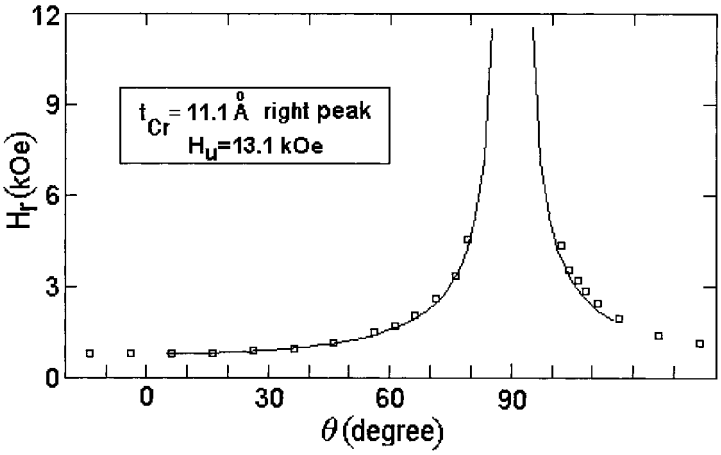

Fig. 4. Angular dependence of the resonance field $H_r(\theta)$ for the sample with t_{Cr} = 11.1 Å (right peak). The curve is obtained according to (1) and (2) with the value of the demagnetizing field H_u = 13.1 kOe. The upper part of the curve is omitted

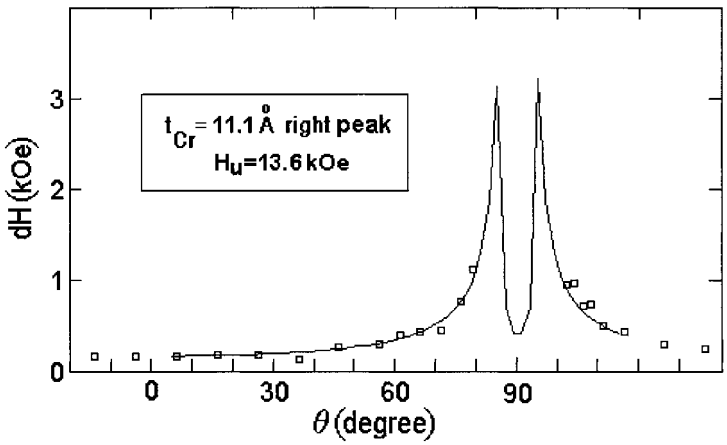

Fig. 5. Angular dependence of the linewidth $dH(\theta)$ for the sample with $t_{Cr} = 11.1$ Å (right peak). The curve is obtained according to (3) with the value of the demagnetizing field $H_u = 13.6$ kOe and $dH_0 = 390$ Oe

thickness ($t_{Cr} < t_{Fe}$) a narrow single line was registered. The additional FMR line arises at the left wing of the main one. The intensity of the additional line grows, becomes equal to the intensity of the main line at $t_{Cr} \approx t_{Fe} \approx 12$ Å, and then decreases again. For the samples with $t_{Cr} > 16$ Å the FMR line becomes a broad single line with asymmetric shape. The results of the determination of the effective demagnetizing field (uniaxial anisotropy H_u) from the angular dependence of H_r (Fig. 4) and dH (Fig. 5) of the main line are presented in Table 2. The tendency of growth of H_u (in the assumption of constant M_s) was demonstrated for increasing t_{Cr} up to $t_{Cr} = 16$ Å (Fig. 6), when the stabilization of the effective demagnetizing field (H_u) occurred. In the same region (Fig. 7) the linewidth remarkably grows, remaining constant at large t_{Cr}.

The determination of the effective demagnetizing field was done once again independently by the modeling of the lineshape in a field parallel to the multilayer plane for those samples, where a splitted FMR signal has been observed. Thus, the fitting param-

Table 2
Results of modeling

modeling of the angular dependence of H_r and dH						modeling of the FMR line shape			
sample	$H_u(H_r)$ (kOe)	$H_u(dH)$ (kOe)	dH (Oe)	H_{u1} (kOe)	H_{u2} (kOe)	k_1/k_2	m_0	H_b (kOe)	dH (Oe)
1	15.4	15.8	280	28.0	15.0	0.04	0.8	0.19	202
2	11.2	11.2	240	26.0	13.0	0.04	0.75	4.5	182
3	13.1	13.6	390	19.5	10.7	1	0.65	5.0	270
4	9.0	9.0	450	20.5	11.5	0.4	0.4	1.2	404
5	15.0	13.0	650		16.2		0.55	0.4	471
6	14.0	14.0	650		17.5		0.75	0.8	471
7	13.0	13.0	550		16.5		0.1	0.13	471
8	12.0	12.0	700		16.5		0.6	0.6	679

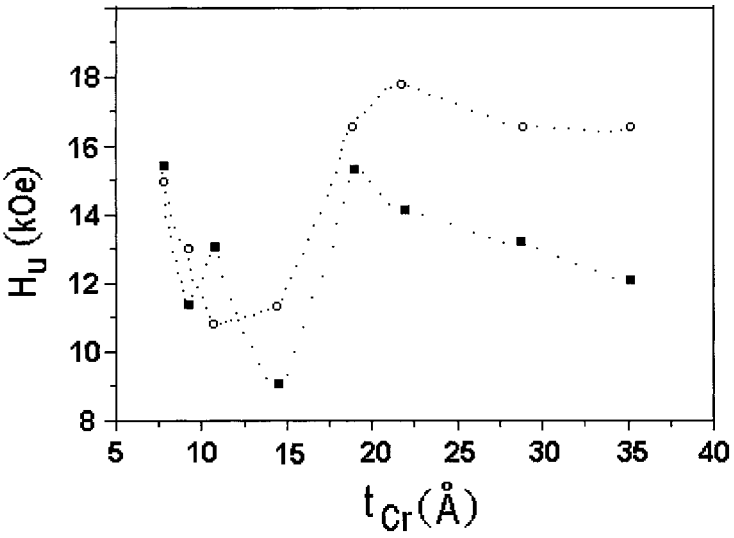

Fig. 6. Dependence of the effective demagnetizing field on t_{Cr} for the series of the samples. ○ obtained by processing of angular dependence of H_r and dH data, ■ obtained by modeling of the FMR lineshape in the field parallel to the surface. The dotted lines may guide the eyes

eters m_0 and H_b for these samples are essentially different (see Table 2). The curve representing the formula (4) in view of the chosen approximation for $\mu(H)$, for four samples with t_{Cr} = 7.7 to 14.5 Å agrees rather well with the experimental data (see, for example, Fig. 8). The values of the anisotropy fields H_{u1} and H_{u2} (Table 2) differ for

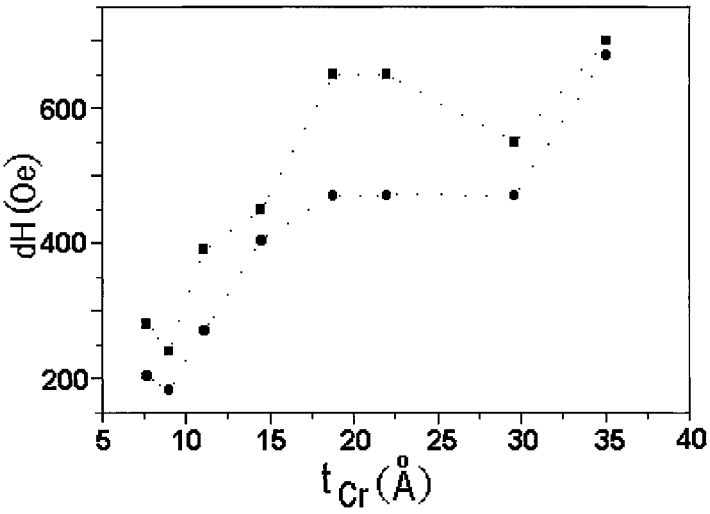

Fig. 7. Dependence of the FMR line width on t_{Cr} for the series of samples. ■ obtained by processing of angular dependence of H_r and dH data, ● obtained by modeling of the FMR lineshape in the field parallel to the surface. The dotted line may guide the eyes

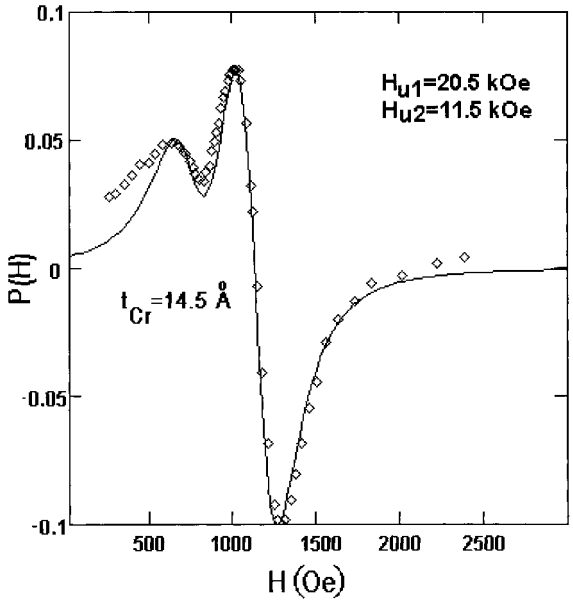

Fig. 8. FMR spectra in the field parallel to the surface for the sample with $t_{Cr} = 14.5$ Å (◇). The curve is obtained by modeling according to (4) with the parameter values $dH_0 = 404$ Oe, $H_{u1} = 20.5$ kOe and $H_{u2} = 11.5$ kOe

each sample by about a factor of two. The value of H_{u2} approaches the value of H_u, obtained by the simulation of the angular dependences of H_r and dH. The dependence of the intensity of the peaks ratio k_1/k_2 on the spacer thickness t_{Cr} is depicted in Fig. 9. For the sample with $t_{Cr} = 11.1$ Å the intensities of both peaks become equal.

7. Discussion

One of the results received is the silence of an optical mode in the studied samples at the experimental conditions used. Different reasons for this should be considered. Estimates of the parameters of interlayer exchange interaction extracted from the values of

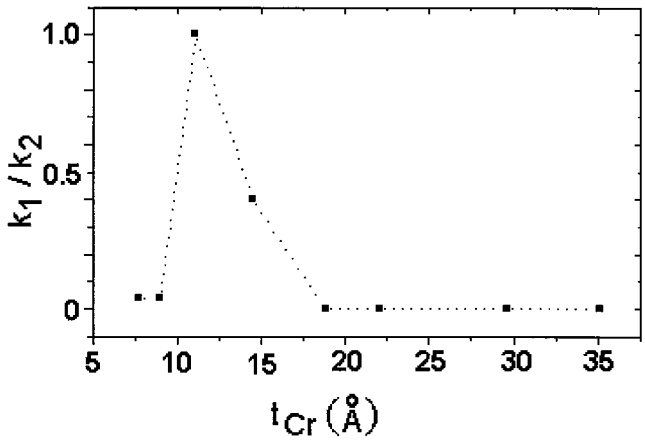

Fig. 9. Relative intensity of the low-field and high-field peaks of the FMR signal for the series of samples with different t_{Cr}

m_0 and H_s of the magnetization curves are rough because of the contribution of the relatively thick buffer layer (≈1/3 of the whole volume of the iron). Nevertheless, the calculations based on the theory of biquadratic exchange [8], in the assumption of an ideal structure of Fe/Cr layers, have shown that the values of characteristic resonant fields received for an optical mode are different for the investigated samples. These fields (≈30 kOe) for a part of the samples are inaccessible in our experimental conditions, but for others optical mode should be observed at longitudinal excitation.

The absence of an FMR signal at longitudinal excitation for all samples can also mean that the uniform model used for the estimation of the resonance fields [8] is not adequate. Instead, a more complicated, non-uniform magnetic structure of a multilayer may exist. According to the results of Mössbauer effect investigations of the same samples [4], mass transfer of Fe and Cr atoms on the boundaries between the layers partially washes out the interface. The latter process can lead to the formation of various solid solutions with different magnetic properties [4]. Therefore, the excitation of the uniform FMR mode corresponding to a simple two-sublattice magnetic structure can hardly be expected.

Judging from the magnetization curves of the investigated samples at the spacer thickness ≈6 to 7 Å, the saturation fields are not high (some hundreds of Oe), and the shape of the curve is close to the observed one on a thin single layer of iron [9 to 13]. For comparison, the single iron layers of thicknesses 30, 60 and 300 Å were investigated. For all of them a sharp saturation effect in the fields of 10 to 1000 Oe was observed. Processing of the FMR spectra having single narrow lines gave the values of $4\pi M_s - H'_u$ close to the value for bulk iron. Thus, the samples with small spacer thickness are magnetically weakly coupled and, in their magnetic properties, they are similar to thin single Fe layers.

For the samples with thicker spacer t_{Cr} (starting from the sample 2), there is a section of the magnetization curve with almost linear change of magnetization. For these samples the m_0 values (see Table 2) are still close to unity. At the thickness of 10 to 12 Å (samples 3, 4) the saturation field H_s increased up to 12 kOe. At further increase of the spacer thickness up to 20 Å the reduction of H_s, together with a more sharp magnetization curve, is observed again. Then H_s grows a little at $t_{Cr} \approx 40$ Å (sample 8). Hence, in the interval of spacer thickness 12 to 40 Å the change of the magnetic structure, connected with the growth of the parameters of interlayer exchange occurs. Thus, the tendency to the formation of antiferromagnetic or non-collinear magnetic order may be assumed.

The influence of the buffer layer on the FMR signal may be evaluated from the comparison with the results of FMR measurements on single Fe layers of similar thicknesses (30 and 60 Å). It appears that the buffer layer contributes a narrow single line to the main FMR line, with small effect on its position and intensity. The role of the magnetic interaction of the Fe buffer layer with the principal body of the Fe/Cr multilayer is minimal due to the short range of the interaction involving the nearest Fe layer separated by a Cr spacer. The above conclusion is also supported by the evolution of the FMR spectra found (Fig. 3) at increasing spacer thickness. The additional signal revealed for some of the samples studied is not connected with the *buffer* layer because all samples have the same buffer.

Generally, the additional line found in the FMR spectrum at the transverse excitation (Fig. 3) can be attributed to the surface mode [6] or a spin wave harmonic [14]. How-

ever, such interpretation contradicts to the observed spectra evolution with the sample rotation in a magnetic field. The more realistic reason for the observation of the above mentioned line is the existence of an additional phase, having different magnetic anisotropy. A similar phase spreading through the whole depth of the multilayer has been already observed by electron microscope [3]. Different values of the anisotropy field can be caused, for example, by magnetostriction effects. The latter assumption is in agreement with the results of FMR measurements on single iron layers and Fe/Cr multilayers [15]. The characteristic FMR lineshape with a bent part in the center was observed for the sample $(Fe/Cr)_{30}$. The modeling of this lineshape gave two values of an effective field, i.e. 17.6 and 28 kOe, respectively. The relative volume of participating phases (main and additional) may be related to the relative intensity of the peaks observed (Fig. 9).

8. Conclusions

The performed investigations have shown the possibilities of FMR to study the magnetic structure of the multilayers in combination with the magnetization and crystalline structure data. The approach, developed by computer simulation of the angular dependences of the resonance field and linewidth together with FMR lineshape modelling, allows to get reliable values of magnetic anisotropy. The most interesting result obtained is the evolution of the FMR spectrum in dependence on the spacer thickness.

The splitting of the FMR spectrum observed in Fe/Cr multilayers can be explained by inhomogeneities of its structure in the form of their columnar texture [3]. This leads to the existence of two phases with different values of anisotropy fields, giving two contributions to the total FMR signal. Evaluations of the effective demagnetizing fields of these phases show that their values differ by a factor of two. The value of one of them, attributed to a low-field peak, coincides with that of bulk iron. This means, in the assumption of constant M_s, that in the latter phase the anisotropy field is close to zero.

The FMR measurements have specified the range of Cr spacer thickness, smaller or equal to the thickness of Fe layer (<15 Å), where magnetic inhomogeneities of a multilayer are essential. Such a feature has not been detected for larger spacer thickness. This conclusion is supported by the observation of the complex structure of the interfaces, stipulated by alloying in the same samples at small spacer thickness [4]. A possible explanation of the FMR spectra evolution in dependence of the spacer thickness may be associated with an effect of percolation between the iron layers. Thus, one could argue that at small spacer thickness values (<15 Å) there are no permanent interlayer boundaries, whereas they exist at thicker spacers. Supposed this interpretation would be correct, FMR-lineshape analysis may provide a good tool to check the quality of the samples prepared.

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