SPIN-WAVE RESONANCE IN MANGANITES

P. ALESHKEVYCH, M. BARAN, V. DYAKONOV,
R. SZYMCZAK, H. SZYMCZAK

Institute of Physics, Polish Academy of Sciences
(32/46, Al. Lotnikow, Warsaw PL-02-668, Poland; e-mail: szymh@ifpan.edu.pl; wydz3@pan.pl)

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A review of experimental studies of magnon excitations in manganites by microwave technique is presented. The main result obtained is the observation of the spin-wave resonance (SWR) consisting of a series of well-resolved standing spin-wave modes. The surface spin-wave modes have been observed in manganites for the first time. The surface modes data are consistent with the surface-inhomogeneity model, in which the surface-anisotropy field acts on the surface spin. At low temperatures for small wave vectors $k \to 0$, the dispersion relation has a quadratic shape similar to that observed in Heisenberg ferromagnets.

1. Introduction

During the last years, the physical properties of doped manganites with the general formula $La_{1-x}D_xMnO_3$, where D is a divalent ion, have been extensively studied both from a fundamental point of view and for their potential applications ([1] and references therein). At a fundamental level, the new physics associated with such systems arises from a strong correlation between the structural, transport, and magnetic properties of these materials.

The character of spin ordering at the transition to the ferromagnetic metallic state was shown to influence the mobility of charge carriers and, correspondingly, the electronic transport. Therefore, studying the magnetic properties of manganites, it is important to investigate the spin dynamics as it is intimately connected with a charge transfer between Mn^{3+} and Mn^{4+} ions through the so-called "double-exchange" interaction [2].

There are three main experimental methods in studying the spin dynamics: an inelastic neutron scattering (INS), Brillouin scattering, and magnetic resonance. The inelastic neutron scattering is a powerful technique to study the spin dynamics in large samples; however, it experiences some difficulties when thin films are necessary to study. The Brillouin scattering technique could be used for studying the thin films but, at the present time, there are no reports, which use this technique in the thin films of manganites. The microwave technique is a very valuable method for the study of magnon excitations in thin films. The microwave field may excite spin waves in ferromagnets. The important problem concerning the microwave technique is the achievement of film's specific boundary conditions necessary for the spin-wave resonance (SWR) appearance. The next important problem is the proper choice of an appropriate theoretical model for the data interpretation. The existing models concerning SWR can be divided into two main types: the volume inhomogeneity (VI) model and the surface inhomogeneities (SI) one. The essential difference between those models relies on a formulation of the boundary conditions in a film leading to the pinning of surface spins. The VI model assumes that the volume magnetization is maximal in the middle of a film and, decreasing towards the surfaces, provides the pinning of surface spins [3]. In the SI model [4—7], a volume magnetization is assumed to be homogeneous, and a surface anisotropy is responsible for the pinning of surface spins. A special interest in SWR consists in the fact that additional resonance lines corresponding to surface spin waves can be observed [7—10]. The existence of acoustic surface spin waves in a ferromagnet implies that its ground state is not uniform.

The SWR in manganite thin films was observed, for the first time, in $La_{0.67}Ba_{0.33}MnO_3$ [11] and next in $La_{0.75}Sr_{0.11}Ca_{0.14}MnO_3$ [12]. In both cases the authors have used the ideal Kittel's model to describe their results. Moreover, for $La_{0.67}Ba_{0.33}MnO_3$ [11], the temperature dependence of the spin-wave stiffness coefficient was evaluated and found to be in an agreement with the spin-wave theory. The SWR was also observed in $La_{0.7}Mn_{1.3}O_{3-\delta}$ films, where the microwave technique was successfully used to excite spin waves [13—15].

In this work, the magnetic resonance technique, as a highly precise and effective method, is used for studying the magnetic dynamics in magnetoresistive films, as well as for the determination of both the effective nearest-neighbour Heisenberg interaction and spin wave stiffness coefficient, and other fundamental characteristics of the spin system.

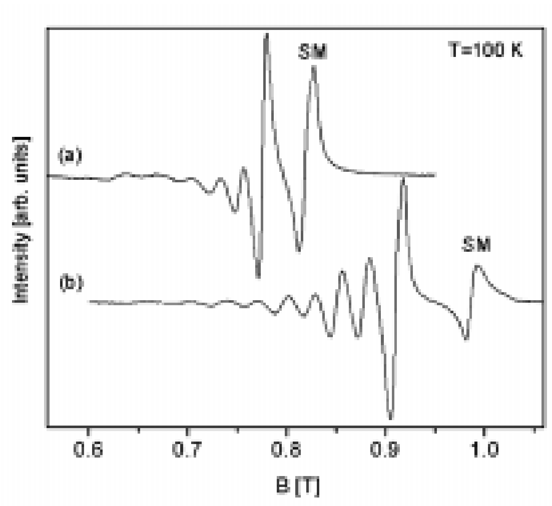

Fig. 1. Resonance spectra for (a) the as-deposited sample and (b) for the annealed one, recorded in the perpendicular orientation at T=100 K

2. Samples and Experimental Procedure

The samples studied are epitaxial $La_{0.7}Mn_{1.3}O_3$ films. The preparation technology of both films and ceramic pellets used as targets has been previously described in detail [16,17]. In brief, the films were deposited onto [001] oriented single-crystalline $LaSrAlO_4$ substrates using DC-magnetron sputtering. The temperature of the substrate surface was 600 °C. After the film was deposited to the suitable thickness, it was cooled to room temperature at a rate of 5 K/min. In order to achieve a homogeneous film, next it was annealed at 600 °C for additional 30 min in the oxygen flow. The thickness of films was estimated to be about 3500 Å.

The chemical composition of the films studied, as well as that of the ceramic targets used for the film preparation, has been discussed previously [18,19]. The ionic structure of

$$(La^{3+}_{0.66}V^{(c)}_{0.05}Mn^{2+}_{0.29})_A(Mn^{3+}_{0.65}Mn^{4+}_{0.29}V^{(c)}_{0.06})_B O^{2-}_{2.84}V^{(a)}_{0.16} \quad (1)$$

obtained for ceramics was conserved for $La_{0.7}Mn_{1.3}O_3$ films with an uncertainty of about 10%.

The measurements of SWR spectra were performed using an X-band reflection spectrometer operating at a fixed frequency (about 9.235 GHz) in conjunction with a variable temperature flowing gas cryostat. The 100 kHz field modulation and the phase sensitive detection techniques were used, so that the detected signal corresponded to the field derivative of the absorbed microwave energy.

3. Experimental Results

The SWR spectrum at the out-of-plane magnetic field direction orientation reveals a complex structure depending on temperature. The greatest number of resonance peaks was found in a spectrum measured when the static magnetic field is applied perpendicularly to the surface of a film. The profile of the spectrum strongly changes with a deviation from the perpendicular configuration ($\boldsymbol{H}_\perp$). Some examples of resonance spectra for two samples at 100 K and in the configuration $\boldsymbol{H}_\perp$ are shown in Fig. 1 for (a) the as-deposited sample and for (b) the annealed one.

The influence of the pinning conditions at the surfaces of a film, which are responsible for the excitation of standing spin waves, was described in detail within the SI model by Puszkarski ([8] and references therein). Assuming the SI model, it was found that the spectra contain one surface mode, whereas all other peaks correspond to non-uniform bulk modes [8]. The surface peaks are labeled as SM in Fig. 1. Within the SI model, the existence of only one surface (acoustic) wave at the perpendicular orientation is possible in the situation where the surface anisotropy energy of the film has different values at both surfaces. This situation can be expected in these films, since one of the surfaces is in direct contact to the substrate, while the other surface can be treated as completely free. The appearance of only one surface wave testifies to that the surface anisotropy $E_S > 0$ on one of the surfaces (on the free surface, as will be shown below), while the spins are pinned on the other surface (interface) (i.e. $E_S < 0$). In other words, the asymmetric boundary conditions are realized in the given film. Therefore, this film should be characterized by two different surface parameters: A_f and A_S for the free surface and the interface, respectively. As for volume spin waves, this asymmetry in the boundary conditions makes it possible to excite asymmetric spin waves in the spectrum in addition to symmetric modes.

The results described above are related to wave excitations in the low-temperature region, where ferromagnets are characterized by a high degree of spin ordering. In this analysis, the magnon—magnon interactions were neglected. But the SWR spectra change with increase in temperature. This means that the surface anisotropy changes with temperature similarly to the volume magnetic anisotropy. Fig. 2 shows the integral intensity of spin wave modes vs mode numbers for the spectra recorded in the perpendicular configuration at three different temperatures (T = 10,

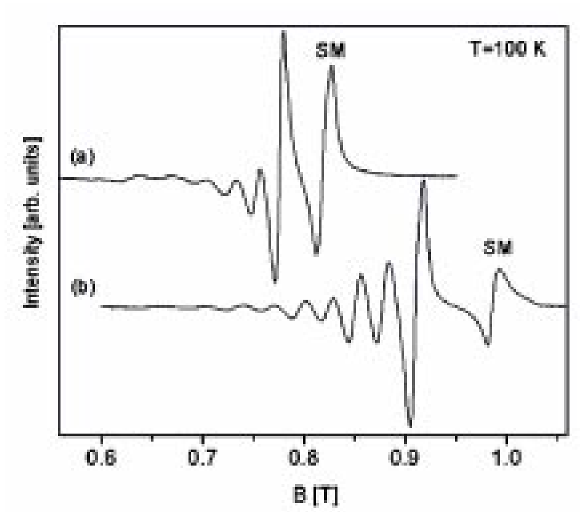

Fig. 2. Resonance peak intensity vs mode number for three temperatures

90 and 130 K). At 10 K, the intensity of even and odd peaks is highly nonmonotonic because of the large asymmetry of the surface anisotropies. With the temperature increase, the surface anisotropies decrease and, consequently, the asymmetry decreases resulting in a more monotonous intensity dependence.

A very unusual transformation of the SWR spectrum appears when the temperature becomes higher than 120 K. In this case each of the resonance lines begins to split into two lines. This splitting grows with increase in temperature. A typical example of such a spectrum recorded at 165 K is shown in Fig. 3,*a*. The inset in Fig. 3 shows the splitting between adjacent resonance lines within the doublet vs T. This splitting monotonously grows up to the Curie temperature T_C=205 K. This "doublet structure" of the SWR spectrum can be comprehensively explained within the framework of the SI model [20]. The values of the wave vectors (which determine the positions of resonant peaks) can be found from the relation

$$\mathrm{tg}(Lk) = \frac{(A_f A_S - 1)\sin(ka)}{(A_f A_S + 1)\cos(ka) - (A_f + A_S)}, \quad (2)$$

where k is the wave vector, L is the thickness of the film, and a is the lattice constant. Only two parameters (A_f, A_S) define a set of wave vectors — the roots of Eq. (2). Upon changing one of these parameters, Eq. (2) should give another set of wave vectors not conterminous with the previous one. This means that the doublet structure of the SWR spectrum can be explained if the film is characterized by three different surface parameters (it

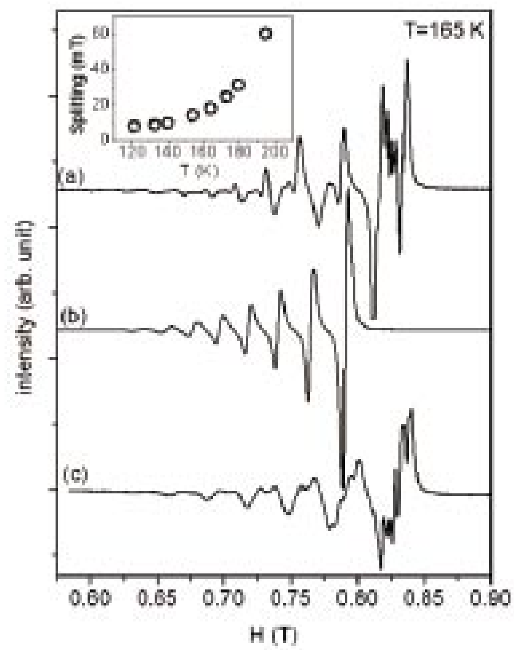

Fig. 3. Illustration of consecutive changes of the SWR spectrum as a result of the sample annealing and with time. All three spectra are recorded in the perpendicular orientation at T=165 K. Spectrum (*a*) has been recorded within long time after a post-preparation annealing, spectrum (*b*) — immediately after the second annealing in the oxygen atmosphere, spectrum (*c*) — three months later than spectrum (*b*). Inset shows the distance between adjacent resonance lines within doublet vs T for the sample after a post-preparation annealing

requires that one of surfaces should be characterized by two surface parameters). A description of the surface by two parameters is possible if magnetic inhomogeneities are created on this surface with a periodic topology [21]. It means that the doublet structure of the resonance lines testifies to that two magnetic sublattices are realized on one of the surfaces. In the $La_{0.7}Mn_{1.3}O_{3-\delta}$ manganites (as well as in any other manganites), the existence of two types of manganese ions (Mn^{3+} and Mn^{4+}) can be a natural source of that magnetic inhomogeneity. And these ions should be aligned periodically, forming two magnetic sublattices (or stripes). It is important to note, that even in the presence of two types of ions but without a periodic structure, such a surface would be characterized only by one average value of the surface parameter. Such periodic stripe structure was observed in manganites $La_{1-x}Ca_xMnO_3$ ($x > 0.5$) using electron microscopy technique [22,23]. It should be mentioned that the manganites studied in these papers are charge-ordered antiferromagnets in contrast to our samples, which are ferromagnetically ordered. Nevertheless, several theoretical and experimental investigations have indicated the existence of charge-ordered phase also in ferromagnets [24—26].

It is known that in manganites with excess oxygen, for example, in ($LaMnO_{3+\delta}$) [27, 28] or in $La_{0.7}Mn_{1.3}O_{3-\delta}$ [18] presented in this study, the appearance of Mn ions in mixed valence states is a result of the charge compensation caused by the enhanced defect structure. In general, in this crystal structure, there exist vacancies in both the cation and anion sublattices. Moreover, those studies have showed that the defect chemistry can be described with a cluster model, where cation and anion vacancies may not only be randomly distributed but also form more complex defects - mesoscopic cluster-type inhomogeneities [18]. In addition to the manganites, ordering of the oxygen vacancies is also known to exist in related compounds such as cuprates, nickelates [29], and cobaltites [30]. The probable ordering of oxygen vacancies in $La_{0.7}Mn_{1.3}O_{3-\delta}$ is in favor of the suggested stripe formation by Mn ions with different valences. Any type of oxygen vacancies cannot directly affect the resonance lines. However, oxygen vacancies that assist the electronic ordering of Mn ions can also assist the charge ordering.

It is well known [31, 32] that annealing in reducing or oxidizing atmospheres can be used to control the Mn^{3+}/Mn^{4+} ratio. This points to a simple way to confirm the assumption of stripe formation by Mn^{3+} and Mn^{4+} ions on the free surface. Since the film annealing in an oxygen atmosphere can change the Mn^{3+}/Mn^{4+} ratio, a second annealing was carried out under the same conditions as the first post-preparation annealing. As was expected, the repeated annealing has not changed the magnetic properties in the film volume, since the value of the magnetic moment has remained the same as it was before, but this operation has strongly modified the free surface properties. As an example, the SWR spectrum recorded just after the additional annealing is labeled as (*b*) in Fig. 3. As one can see, spectrum (*b*) does not show any traces of splitting. It is interesting to note, that three months after the additional annealing, the SWR spectrum again shows the splitting. This spectrum is labeled as (*c*) in Fig. 3. For convenience of comparison all three spectra in Fig. 3 are taken out at the same temperature (165 K) and orientation ($H_\perp$).

At annealing, the ratio of Mn^{4+}/Mn^{3+}changes resulting in the disappearance of the splitting in the spectrum. After the elapse of months, the reverse process takes place, probably accompanied with the release of O_2 ions, and the splitting in the SWR spectrum is restored. Thus, the stripe formation of charge-ordered manganese ions seems to be the preferable state for $La_{0.7}Mn_{1.3}O_{3-\delta}$. It is a matter to assume, of course, that each subsequent annealing will causes the same changes in the film. The application of this technique for the observation of stripes was possible because of the specific influence of the periodic boundary conditions on SWR spectra in the periodically inhomogeneous materials.

4. Energy of Spin Waves

It is known from the theory of spin waves [33], that the energy of spin waves in ferromagnets can be described as $E = Dk^2$ for $k \to 0$, where D is the stiffness coefficient of spin waves. The multipeak resonance spectrum allows one to check the dispersion relation, since a position of each resonance peak is proportional to the energy of an appropriate spin wave according to the well-known Kittel's resonance condition [4]

$$\frac{\omega}{\gamma} = H_{\rm res} - 4\pi M + H_a + Dk^2, \tag{3}$$

where $H_{\rm res}$ is the resonance field, ω is the resonance frequency, M is the saturation magnetization, H_a is the anisotropy field, and γ is the spectroscopic splitting factor. As follows from Eq. (3), the $H_{\rm res}$ as a function of k^2 should be linear. The dependence $H_{\rm res}(k^2)$ for a spectrum recorded at three different temperatures in the perpendicular orientation is shown in the inset of Fig. 4. As can be remarked, for the longest-wave modes (three first modes), there is a deviation from a simple linear dependence. It is related to the fact that these modes (and especially surface ones) are more sensitive to an inhomogeneity of the magnetic structure at a surface layer. Therefore, to determine D, it is necessary to take into account only the linear part of the dispersion dependence. In this case, D is the average bulk exchange constant. The temperature dependence of D is shown in Fig.4. From these data, a value of $D(0)$=156 meV·Å^2 was found. This value agrees within the experimental error with that determined from the magnetization saturation temperature dependence [15].

For spin waves with a small k, the experimental $D(T)$ data can be compared with the Dyson formalism of two-spin-wave interactions in a Heisenberg ferromagnet [34] which predicts that the dynamical interaction

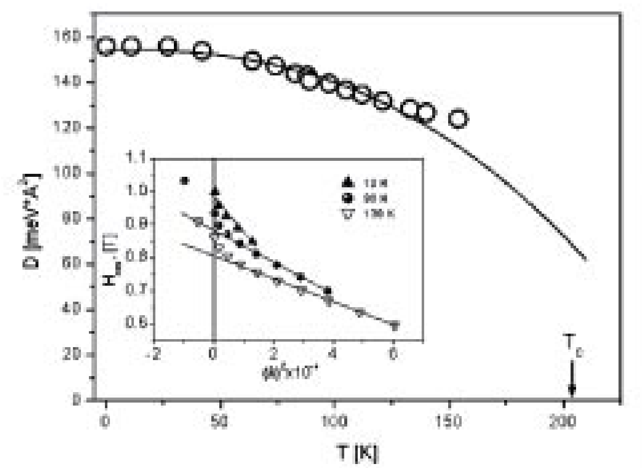

Fig. 4. Spin-wave stiffness coefficient D vs T for the annealed sample. The solid curve presents fitting of Eq. (4) to experimental points. The arrow points to the Curie temperature position. The inset shows the resonance field H_{res} versus the square of the spin wave vector k^2 at T = 10, 90, and 130 K. Note, that the values of k are normalized to the lattice constant and therefore are dimensionless

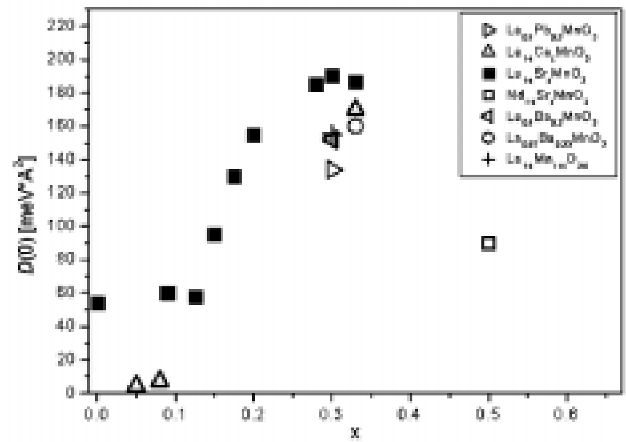

Fig. 5. Spin-wave stiffness coefficient $D(0,x)$ as a function of the doping level x evaluated from the data of both inelastic neutron scattering and microwave technique

between spin waves gives a $T^{5/2}$ behavior:

$$D(T) = D(0)\left\{1 - \frac{\upsilon_0 \bar{l}^2 \pi}{S}\zeta(5/2)\left[\frac{k_{\mathrm{B}}T}{4\pi D(0)}\right]^{5/2} + ...\right\}, \quad (4)$$

where υ_0 is the volume of a unit cell determined by nearest neighbors, S is the average value of the manganese spin, and $\zeta(5/2)$ is the Riemann integral. $\bar{l}^2$ is defined by $\bar{l}^2 = S/3D\left\{\sum l^{n+2}J(\vec{l}\,)\right\}$, which gives information on the range of the exchange interaction. The solid curves in Fig. 4 present fittings of Eq. (4) to the experimental data being in good agreement with them for temperatures up to 165 K, which is not far from the Curie point, T_{C}=205 K. The fitted values of $\bar{l}^2$ give $\sqrt{\bar{l}^2} = 5.4a_0$, where a_0 is the distance between the nearest neighbors (a_0=3.907 Å), and indicate that the exchange interaction extends significantly beyond nearest neighbors. For T > 165 K, the resonance peaks become unresolved not allowing the D evaluation. However, it should be noted that the experimental $D(T)$ dependence deviates from theoretical curve for T > 150 K, when the SWR spectrum begin to show splitting. It is known for other manganites [11, 35] that, at temperatures close to T_{C}, their $D(T)$ dependences have rather power-law behavior, and appearing to collapse as $T \to T_{\mathrm{C}}$. In $La_{0.7}Mn_{1.3}O_{3-\delta}$, the deviation in the opposite direction can be explained by a decrease in $\bar{l}^2$, which means that itinerant e_g electrons become localized, and this is consistent with the charge ordering and the stripe formation of Mn ions.

The spin-wave stiffness coefficient at $T \to 0$ K, $D(0)$, is related to the intrinsic property of magnets, and it seems to be of some interest to compare these values for different manganites known from the literature. Fig. 5 shows $D(0)$ as a function of the doping level x evaluated in different manganites using both inelastic neutron scattering [35—42] and microwave techniques [11, 15]. The $D(0)$ values obtained using SWR data are in good agreement with those measured in other ferromagnetic manganites. There are no compound that was studied by two techniques simultaneously, however the $D(0)$ values are very similar both for $La_{0.7}Ba_{0.3}O_3$ studied by INS technique and $La_{0.67}Ba_{0.33}O_3$ studied by SWR.

5. Conclusions

The analysis of the magnetoexchange-branch dispersion relation in $La_{0.7}Mn_{1.3}O_3$ films has shown that the internal fields in these films are very homogeneous and that the spin-wave modes depend on the surface pinning conditions rather than variations in the internal field.

For small wave vectors at low temperatures, the dispersion relation is quadratic as that in Heisenberg ferromagnets. In the case of thin films, INS experiments can be successfully replaced by microwave technique for studying the spin dynamics. In addition to bulk magnetic properties, the spin wave resonance spectrum allows one to study the surface effects like the excitation of surface spin waves or the stripe formation, as was demonstrated by the example of $La_{0.7}Mn_{1.3}O_{3-\delta}$ films.

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СПІН-ХВИЛЬОВИЙ РЕЗОНАНС У МАНГАНІТАХ

П. Алєшкевич, М. Баран, В. Дьяконов, Р. Шимчак, Г. Шимчак

Р е з ю м е

Подано огляд досліджень магнонних збуджень у манганітах методами мікрохвильової техніки. Основний результат — спостереження спін-хвильового резонансу, що являє собою послідовність добре розділених стоячих спін-хвильових мод. Поверхневі спін-хвильові моди у манганітах спостережено вперше. Дані з поверхневих мод узгоджуються з поверхнево-неоднорідною моделлю, в якій поле поверхневої анізотропії діє на поверхневий спін. При низьких температурах для малих хвильових векторів дисперсійне співвідношення є квадратичним подібно тому, що спостерігається у гейзенбергівських феромагнетиках.

СПИН-ВОЛНОВОЙ РЕЗОНАНС В МАНГАНИТАХ

П. Алешкевич, М. Баран, В. Дьяконов, Р. Шимчак, Г. Шимчак

Р е з ю м е

Представлен обзор исследований магнонных возбуждений в манганитах методами микроволновой техники. Основной ре-

зультат — наблюдение спин-волнового резонанса, представляющего собой последовательность хорошо разрешенных стоячих спин-волновых мод. Поверхностные спин-волновые моды в манганитах наблюдались впервые. Данные по поверхностным модам согласуются с поверхностно-неоднородной моделью, в которой поле поверхностной анизотропии действует на поверхностный спин. При низких температурах для малых волновых векторов дисперсионное соотношение является квадратичным подобно тому, что наблюдается в гейзенберговских ферромагнетиках.