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A remarkable transformation of magnetic resonance spectra as a result of a mutual influence of coexisting para- and ferromagnetic phases

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Abstract

In this work, we carry out the analysis of the resonance absorption of electromagnetic radiation for the system in which para- and ferromagnetic phases coexist over a wide temperature region. It is found that taking account of the mutual influence of coexisting phases gives rise to the appearance of substantial changes in the curves of resonance absorption and values of resonance fields, as well as to making the geometry of a phase distribution dependent on an external magnetic field. Near the temperature boundaries of the phase coexistence region, the expressions for description of the curves of the dispersive absorption of electromagnetic radiation are obtained and the rules of the behaviour of the resonance fields for each of the phases are specified. As follows from the calculations, the resonance field for the paramagnetic phase becomes dependent on the shape of the sample, the saturation magnetization and the fraction of ferromagnetic phase. It is shown that the character of magnetic resonance spectra and the features of their temperature change agree well with the experimental data, obtained by various groups of researchers on the single crystalline and polycrystalline samples of doped perovskite manganites.

1. Introduction

In recent years, the doped perovskite manganites $R_{1-x}A_xMnO_3$, where R is a rare earth and A is an alkaline or alkaline earth element, have attracted a great deal of research interest due to a number of unusual physical properties including the colossal magnetoresistance effect (CMR) [1]. The essence of the CMR consists in a very large (several orders in magnitude) decrease in electric resistance under the action of magnetic field. Primarily due to this effect the doped perovskite manganites are considered as promising materials for a new generation of magnetic sensors and readout elements in magnetic storage devices.

The results of numerous theoretical and experimental investigations have shown that strong tendencies toward phase separation play an essential role in the given class of materials

and often an inhomogeneous state, characterized by a coexistence of different electronic and magnetic phases, becomes more favourable than a homogeneous one [1–7]. To clarify the physical picture within the region of magnetic phase coexistence, the method of magnetic resonance can be used as a highly informative tool. It is well known that even a minor inhomogeneity of magnetic materials manifests itself in noticeable transformations of magnetic absorption spectra. To date, a number of research groups (see, for example [2–15]) have studied the spectra of magnetic resonance, but an unambiguous picture is mainly obtained for magnetically homogeneous regions. The results that embrace the temperature regions near the points of magnetic phase transitions considerably differ from each other and are often interpreted from different points of view.

It is revealed in papers [3, 8–11] that in the vicinity of a paramagnetic (PM) to ferromagnetic (FM) transition the spectra of electron spin resonance (ESR) for a number of $R_{1-x}A_xMnO_3$ compounds consist of two well resolved lines. What is more, in all the cases, the resonance field of one of the lines decreases with the temperature lowering, whereas that of the other increases. It follows from the analysis of the experimental results that such behaviour is characteristic of both the single crystalline and polycrystalline samples. The authors identify one of the lines of the resonance absorption as belonging to the ferromagnetic phase. As concerns the other line, the researchers tried to ascribe it to PM [3, 8, 10, 16], spin glass [9] or antiferromagnetic [11] phases, but its nature still remains unclear.

It should be noted that the fact that an ESR spectrum consists of two lines does not necessarily imply the coexistence of two different magnetic phases. Under some specific conditions, such a picture can also be observed in single phase ferromagnetic samples [17–22]. Let us analyse such cases in more detail and examine if they are applicable to the explanation of the above phenomena.

It is known that a number of parameters, namely the presence of crystallographic anisotropy, the shape and orientation of a sample, strain, degree of inhomogeneity, porosity, and others, exert a strong influence on the character of the ESR spectra. In most cases, the presence of these factors leads to a change in the width of the resonance curve, a shift of its maximum, and a distortion of its shape, but not to the appearance of two lines (a review of the influence of such factors can be found in [19]). The splitting of the ESR spectrum is most likely in the systems which are characterized by these kinds of *nonuniform* angular or size distribution of the local effective magnetic fields H_{eff}, which lead to the appearance of two or more preferred directions of the orientation of local magnetic moments (for example, textured polycrystals, ensembles of particles with specific shape or size distributions, and others). Theoretical calculations show that this effect can also be observed in polycrystalline ferromagnets or in an ensemble of single domain anisotropic particles with the *uniform* distribution of the effective local magnetic fields (in particular, anisotropy fields), but only under the condition that the magnitude of H_{eff} exceeds a certain threshold value [19–22].

The analysis shows that such approaches are inapplicable to the explanation of the phenomena of the ESR spectrum splitting observed in single crystalline and polycrystalline samples of doped manganites. It is known that the crystals of these materials are weakly anisotropic (see, for example [23]) and this especially concerns the region of temperatures near the para- to ferromagnetic transition. Therefore, the realization of the latter scenario seems to be unlikely. At the same time, there are no reasons to suggest that at the heart of the observed effects lies the nonuniformity of the distribution of crystallites in shape or direction (the former scenario), since the splitting and unusual behaviour of resonance fields have been observed in the samples of various microstructures and textures: in spherical [8] and rectangular [3] single crystals, in poly- and nanocrystalline powders [10, 16], and in films [9, 11].

A complex investigation of the properties of a spherical single crystal of $La_{0.7}Ca_{0.3}MnO_3$ was performed in work [8]. Based on the results of the comparative analysis of the temperature behaviour of magnetic and electric properties, on the one hand, and two lines observed in the ESR spectra, on the other hand, the authors came to a conclusion that the lines of the resonance absorption correspond to the PM and FM phases coexisting over a wide temperature range. At the same time, they made a suggestion that the presence of the FM phase gives rise to a change in the resonance field of the PM phase and vice versa. However, no quantitative estimation of this effect was made.

In the present work, we analyse the behaviour of the system in which the PM and FM phases coexist over a wide temperature range and show that taking into account the mutual influence of the phases leads to remarkable changes in the behaviour of resonance fields and curves of dispersive absorption, and that the geometry of the phase distribution becomes governed by an external magnetic field.

2. Thermodynamic aspect of the problem

A PM to FM transition is usually considered to be a second-order (continuous) phase transition (PT) [24]. A distinguishing characteristic of such kinds of PTs is that at a transition point a thermodynamic potential of a system, as well as its first derivatives with thermodynamic variables, change continuously, whereas a second derivative exhibits a discontinuity. The continuity of the first derivatives at a PT point means that such transitions have no latent heat [24]. In the case of magnetic materials, the expressions for the second derivatives of the corresponding thermodynamic potentials include, in particular, a heat capacity and magnetic susceptibility. In the case where a PM to FM transition belongs to the continuous PT, the magnetization, which characterizes the first derivative of the thermodynamic potential with a magnetic field, increases continuously as the temperature is lowered through the transition point, whereas the magnetic susceptibility changes anomalously [24, 25].

The first-order PTs are those that involve a latent heat [24]. The first-order transitions are associated with a mixed-phase regime in which some parts of the system have completed the transition and others have not; this means that both the phases coexist within a certain temperature region. At a fast increase or decrease in temperature, a temperature hysteresis is observed, which is often used for the identification of the first-order PTs [24–26].

Magnetic systems can exhibit magnetic transitions that are thermodynamically of first order when the magnetic order parameter (magnetization) is strongly coupled to the lattice deformations [25–28]. The necessary criterion which determines if a magnetic transition occurs via the first-order PT was obtained in papers [29–31]. This criterion is often satisfied in CMR manganites [25–28, 32–35]. For these materials, the experimental evidence for the occurrence of PM–FM transitions as first-order PTs is contained in papers [25–28, 32, 33]. A satisfactory description of the thermodynamics of these and similar systems is achieved in papers [25, 34–37]. So, Novak *et al* [25] analysed the temperature behaviour of magnetization using the mean field theory, which besides the exchange interaction also includes the dependence, characterized by a coupling parameter η, of this interaction on the interatomic distances. A satisfactory agreement of the theoretical calculations with the experimental data is found. The critical parameter to determine the order of the transition, introduced by the authors, was shown to be proportional to the coupling parameter η.

As was noted above, when the PM to FM transition occurs via the first-order PT, both the phases can coexist over a wide temperature region, being at the same time in an equilibrium state. It is this situation that we consider below and for which the magnetization dynamics is studied. While analysing the behaviour of such a system, we assume that the strong coupling

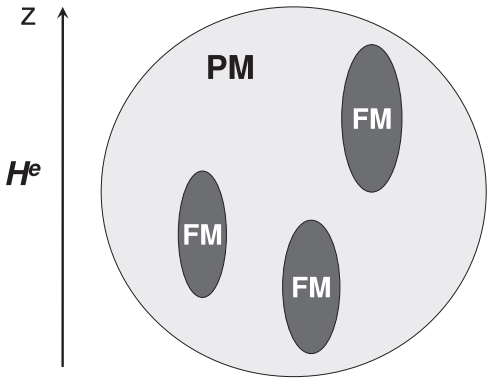

Figure 1. A graphic illustration of a spherical PM particle containing FM clusters. $\mathbf{H}^{\mathrm{e}}$ is the external magnetic field.

of the magnetic order parameter to the lattice deformations, which makes the PM to FM transition belong to the first-order PT, influences for the most part the temperature behaviour of magnetization, whereas its effect can be neglected while considering the situation at a fixed temperature.

3. Splitting of the resonance fields in a region of coexistence of PM and FM phases

Consider a spherical particle at a temperature T, where T is high enough that the whole volume of the particle is in the paramagnetic state. As the temperature decreases below a certain value T_{c1}, the clusters of the FM phase are nucleated in the particle volume (figure 1). The nucleation mechanisms at the first-order PTs were analysed in detail in [24]. The most favourable regions for the nucleation are those which contain various kinds of composition or structure imperfections (dislocations, intergrain boundaries, and others). In especially perfect crystals, a fluctuation mechanism is predominant [24].

Denote the total volume of the FM and PM phases as V_{FM} and V_{PM}, respectively. It is noteworthy that $V_{\mathrm{FM}} + V_{\mathrm{PM}} = V$, where V is the particle volume. Assume that a fraction of the FM phase increases with a further decrease in temperature and the whole volume of the particle becomes ferromagnetic as the temperature reaches a value T_{c2}. Carry out theoretical analysis of the splitting of magnetic resonance fields for two limiting cases where $T \to T_{\mathrm{c1}}^{-}(V_{\mathrm{FM}} \ll V_{\mathrm{PM}})$ and $T \to T_{\mathrm{c2}}^{+}(V_{\mathrm{FM}} \gg V_{\mathrm{PM}})$.

3.1. Splitting of the resonance fields in the vicinity of the temperature of the FM phase nucleation

Assume that the temperature T of the particle meets a condition $0 < (T_{\mathrm{c1}} - T)/T_{\mathrm{c1}} \ll 1$. Under this condition, the FM clusters are formed in the particle volume, but the fraction of the FM phase is very small ($V_{\mathrm{FM}} \ll V_{\mathrm{PM}}$). Consider a case where the magnetization of the FM clusters is aligned parallel to the external magnetic field whose direction coincides with the Oz axis (see figure 1). The reason for making this assumption will be discussed below.

To determine the magnetic field at a point of the location of a certain FM cluster, let us neglect the local contributions depending on the nearest neighbours' arrangement and take into account only magnetostatic field depending on shape and average magnetization of the particle:

$$\mathbf{H} = \mathbf{H}^{\mathrm{e}} - N_z\langle\mathbf{M}\rangle, \tag{1}$$

where $\mathbf{H}^{\mathrm{e}}$ is the external magnetic field, N_z the demagnetizing factor along the z axis, whose value equals $4\pi/3$ for a spherical particle, and $\langle\mathbf{M}\rangle$ is the average magnetization of the particle.

For a particle of a spherical shape, the direction of the internal magnetic field coincides with that of the external one, which sufficiently simplifies the further consideration and allows us to concentrate on the essence of the phenomenon of the resonance fields splitting. Later on, some of the expressions will be generalized to the case where a particle has a shape of an ellipsoid of rotation around the z axis ($N_x = N_y \equiv N_\perp$; $N_z + 2N_\perp = 4\pi$).

Consider an isolated FM cluster. Assume for simplicity that it has the shape of an ellipsoid of rotation elongated along the Oz axis, parallel to the field lines of the permanent magnetic field. Denote the demagnetization factors of the ellipsoid along and perpendicular to the Oz axis as $n_\parallel$ and $n_\perp$, respectively ($n_\parallel < n_\perp$).

Focus on the case where apart from the permanent magnetic field H_z^{e}, a weak alternating field $\mathbf{h}_\perp^{\mathrm{e}}$, aligned perpendicular to the Oz axis, acts on the system. The latter gives rise to small transverse perturbations of the cluster magnetization. Accounting for the fact that in the ferromagnetic state, the absolute value of the magnetization keeps constant ($\mathbf{M}^2 = \mathrm{const}$), represent the magnetization vector of the cluster in the form

$$\mathbf{M} = \begin{pmatrix} m_x \\ m_y \\ M_0 - \mathbf{m}_\perp^2/2M_0 \end{pmatrix} \qquad \mathbf{m}_\perp = m_x\mathbf{e}_x + m_y\mathbf{e}_y, \tag{2}$$

where M_0 is the saturation magnetization and $\mathbf{m}_\perp$ is the small transverse perturbation of the magnetization of the FM cluster.

While determining the energy of the cluster, let us take into account the field $\mathbf{H}$ acting on it (see (1)), the cluster's own magnetostatic field, and surface energy. Then, assuming that in the ground state the cluster is magnetized along the Oz axis and accounting for (2), the cluster's energy can be written as

$$\begin{aligned} &W = W_1 + W_2; \\ &W_1 = v\cdot(\tfrac{1}{2}n_\parallel M_0^2 - M_0\cdot(H_z^{\mathrm{e}} - N_z\langle M_z\rangle) - \sigma(T)s/v), \\ &W_2 = v\cdot(\tfrac{1}{2}(H_{\mathrm{FM}}/M_0)\cdot\mathbf{m}_\perp^2 - \mathbf{m}_\perp\cdot(\mathbf{h}_\perp^{\mathrm{e}} - N_\perp\langle\mathbf{M}_\perp\rangle)), \\ &H_{\mathrm{FM}} = (n_\perp - n_\parallel)\cdot M_0 + H_z^{\mathrm{e}} - N_z\cdot\langle M_z\rangle \end{aligned} \tag{3}$$

where $\mathbf{h}_\perp^{\mathrm{e}} = h_x^{\mathrm{e}}\mathbf{e}_x + h_y^{\mathrm{e}}\mathbf{e}_y$ is a small in magnitude alternating magnetic field aligned perpendicular to the Oz axis, v is the cluster volume, $\sigma(T)$ is the density of the surface energy, and s/v is the ratio of the surface area of the cluster to its volume.

The first term in expression (3), W_1, determines the magnetic energy of the cluster in the ground state, whereas the second one, W_2, describes the small magnetostatic and Zeeman perturbations caused by the action of the alternating magnetic field. The analysis of the expression for W_1 makes it possible to draw some conclusions about the shape of ferromagnetic clusters. As the volume of the cluster is fixed, the surface energy achieves a minimum when the cluster takes a spherical shape, but the minimization of the magnetostatic energy term, $n_\parallel M_0^2 v/2$, requires elongation of the cluster along the magnetic field lines. Therefore, to achieve a compromise, it is favourable for the clusters to take a shape which is close to the ellipsoid of rotation with a long axis parallel to the direction of the external magnetic field. For this reason, we can conclude that $n_\parallel < 4\pi/3$.

The cluster magnetization, being kept as a single whole due to the strong intracluster exchange interaction, oscillates around the equilibrium position. The dynamics of the cluster magnetization is described by the Landau–Lifshitz equations, which, with regard for the

relaxation term in the Hilbert form, read

$$\begin{aligned}(H_{\mathrm{FM}} + \mathrm{i}\alpha_{\mathrm{FM}} H_\omega) m_x - \mathrm{i} H_\omega m_y &= M_0 (h_x^{\mathrm{e}} - N_\perp \langle M_x \rangle), \\ \mathrm{i} H_\omega m_x + (H_{\mathrm{FM}} + \mathrm{i}\alpha_{\mathrm{FM}} H_\omega) m_y &= M_0 (h_y^{\mathrm{e}} - N_\perp \langle M_y \rangle),\end{aligned} \tag{4}$$

where $H_\omega = \omega/\gamma$ ($\gamma = 2\mu_0/\hbar$), μ_0 is the Bohr magneton, and α_{FM} the dissipative constant of the FM phase.

To simplify the calculations, let us introduce the variables $m^\pm = m_x \pm \mathrm{i} m_y$. Then, the solutions of equations (4) can be expressed as

$$\begin{aligned} m^\pm &= \chi^\pm \cdot (h^{\mathrm{e}^\pm} - N_\perp \langle M^\pm \rangle); \\ \chi^\pm &= \frac{M_0/H_\omega}{(H_{\mathrm{FM}}/H_\omega - 1 \pm \mathrm{i}\alpha_{\mathrm{FM}})}, \\ h^{\mathrm{e}^\pm} &= h_x^{\mathrm{e}} \pm \mathrm{i} h_y^{\mathrm{e}}, \qquad \langle M^\pm \rangle = \langle M_x \rangle \pm \mathrm{i} \langle M_y \rangle, \\ N_\perp &= N_x = N_y. \end{aligned} \tag{5}$$

The fact that the average magnetization of the particle, $\langle M^\pm \rangle$, depends itself on the oscillations of the magnetic moments of individual clusters complicates the analysis. However, in the vicinity of the point of the FM phase nucleation, $T_{\mathrm{c}1}$, where $|\langle M^\pm \rangle| \ll |H^{\mathrm{e}}|$, the second term in expression for $m^\pm$ (see (5)) can be neglected and the expression can be reduced to $m^\pm = \chi^\pm \cdot h^{\mathrm{e}^\pm}$.

In sections 4 and 5, expression (5) will be used to calculate the dispersive absorption curves. At this stage, let us only calculate the resonance conditions for the oscillations of the cluster magnetic moment. As follows from (5), in the limit $\alpha_{\mathrm{FM}} \to 0$, the denominator in the expression for $\chi^\pm$ becomes zero when the equality

$$H_\omega = H_{\mathrm{FM}} = (n_\perp - n_\parallel) M_0 + H_z^{\mathrm{e}} - N_z \langle M_z \rangle \tag{6}$$

is fulfilled. This allows us to find the value of the external field H_z^{e} at which $\chi^\pm$ goes to infinity. With regard for the facts that the separate clusters differ from each other in a shape and the particle magnetization is small in the vicinity of $T_{\mathrm{c}1}$, the expression for H_z^{e} can be rewritten as

$$H_z^{\mathrm{e}} = H_\omega - \left(\overline{n_\perp - n_\parallel}\right) M_0. \tag{7}$$

The overbar means the average over the ensemble of clusters contained in the particle. Actually, the relation (7) determines the value of external field at which the resonance for the FM phase occurs in the vicinity of $T_{\mathrm{c}1}$. It is noteworthy that the value of the resonance field for the FM phase is determined not by the particle shape as is usually suggested at the analysis of experimental results [8–10, 16], but by the shape of the FM clusters.

Calculate the resonance field for the PM phase. According to [19], the resonance conditions for an arbitrary chosen PM region occur when $H_\omega = \omega/\gamma$ becomes equal to the value of magnetic field acting on this region. In the case under consideration, the average field which acts on the PM region is not equal to the external magnetic field, but to the effective field which is determined by expression (1). Thus, the condition for the paramagnetic resonance to occur reads

$$H_\omega = H_z^{\mathrm{e}} - N_z \langle M_z \rangle. \tag{8}$$

Taking into account that the magnetization of the particle is mainly determined by the amount of FM phase, the value of the resonance field for the PM phase can be written in the form

$$H_z^{\mathrm{e}} = H_\omega + N_z M_0(T) \cdot \frac{V_{\mathrm{FM}}(T)}{V}, \tag{9}$$

where V is the particle volume and $V_{\mathrm{FM}}(T)/V$ is the fraction of the FM phase at a given temperature. It should be noted that the resonance field of the PM phase becomes dependent

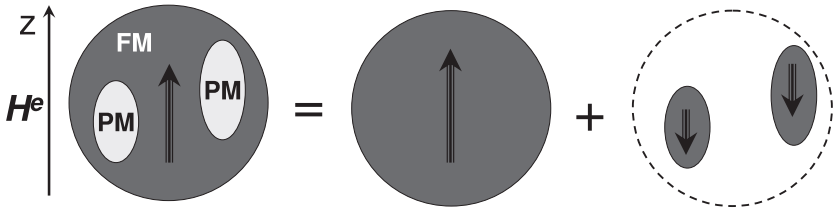

Figure 2. A graphic illustration of the application of a superposition principle to a FM particle with PM inclusions.

on the particle shape, the saturation magnetization of ferromagnetic clusters, and the fraction of FM phase at a given temperature.

3.2. Splitting of the resonance fields in the vicinity of T_{c2}

For temperatures slightly higher than T_{c2} ($0 < (T - T_{c2})/T_{c2} \ll 1$), the inequality $V_{FM} \gg V_{PM}$ is valid. This situation is reverse to that considered above. Assume that the whole volume of the particle is in the ferromagnetic state except for the small clusters of PM phase.

To analyse the effects of a shape and effective field of PM clusters, calculate the energy of the spherical FM particle containing the inclusions of PM phase. Let us proceed from a superposition principle, i.e. consider the PM clusters as the fictitious FM fragments of the same shape, which are magnetized opposite to the magnetic field direction and put into the uniformly magnetized particle (figure 2).

The energy of the system can be written as

$$E = E_0 + \sum_a v_a\{M_0 \cdot (H_z^e - N_z\langle M_z\rangle) + \tfrac{1}{2} \cdot n_\parallel^a M_0^2 + \sigma(T) \cdot s_a/v_a\}, \qquad (10)$$

where E_0 is the energy of the uniformly magnetized particle, $n_\parallel^a$ the demagnetizing factor, taken in a magnetization direction, of a fictitious cluster, and v_a is the volume of the cluster number a. The last term in expression (10) accounts for the surface energy of the clusters. It includes the density of the surface energy $\sigma(T)$ and the ratio of the surface area of the cluster to its volume, s_a/v_a. As in the case considered above, it is favourable for the clusters to take the shape which is close to the ellipsoid of rotation with a long axis parallel to the direction of the external magnetic field. Thus, we can conclude again that $n_\parallel^a < 4\pi/3$.

The magnetic field inside the paramagnetic cluster number a is equal to

$$H^a = H_z^e - N_z\langle M_z\rangle + n_\parallel^a M_0. \qquad (11)$$

Accounting for the dependence $\langle M_z\rangle = M_0(1 - V_{PM}/V)$ and neglecting the latter term (since $V_{PM} \ll V_{FM} < V$ in the vicinity of T_{c2}), we find that *the resonance field for the PM phase* tends to a limit

$$H_z^e = H_\omega + \overline{(N_z - n_\parallel^a)}M_0 \qquad (12)$$

in the vicinity of $T_{c2}(T \to T_{c2}^+)$. At the same time, it is noteworthy that the contribution of the PM phase to the dispersive absorption curve tends to zero.

Finally, it should be noted that the resonance field of the FM phase approaches a value which is characteristic of the ferromagnetic particle of the spherical (in the given case) shape, i.e. H_ω. In a more general case, for the particle in the form of an ellipsoid of rotation around the z axis, *the resonance field for the FM phase* tends to a limit

$$H_z^e = H_\omega + (N_z - N_\perp)M_0 \qquad (13)$$

as $T \to T_{c2}^+$.

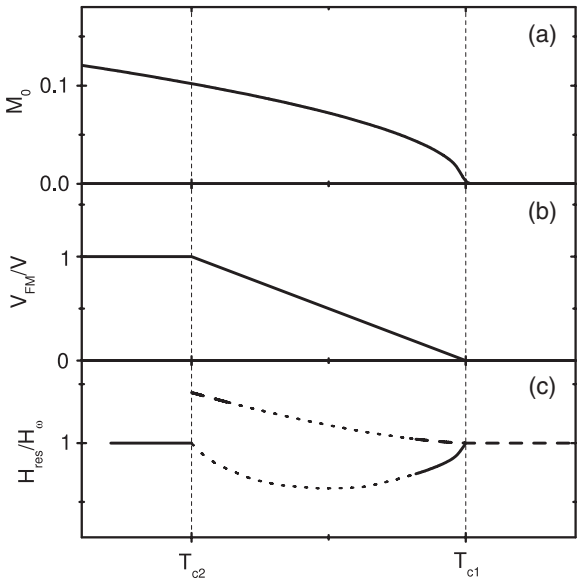

Figure 3. Temperature dependences of (a) the saturation magnetization of the FM phase; (b) the fraction of the FM phase; (c) the resonance fields for the PM (dashed line) and FM (solid line) phases for the spherical particle. The parts of curves for which the approximations used for the calculations of resonance fields are invalid are shown by a dotted line.

3.3. A qualitative analysis of the splitting of resonance fields in the system

For a qualitative analysis of the temperature dependence of resonance fields, let us utilize the Weiss theory [38], according to which $M_0(T) \approx M_0(0)\{3(T_0 - T)/T_0\}^{1/2}$ (figure 3(a)). Consider the case where the clusters take the form of an infinitely elongated cylinder. Then, $(\overline{n_\perp - n_\parallel})$ in expression (7) is equal to 2π and $\overline{N_z - n_\parallel^a}$ in (12) is reduced to N_z. Carry out the calculations for the case where the fraction of FM phase linearly decreases as the temperature increases from T_{c2} to T_{c1} (figure 3(b)).

Figure 3(c) shows the results of the calculations performed for the case of the spherical particle ($N_z = 4\pi/3$) for the following values of the theory parameters: $4\pi M_0(0)/H_\omega = 1$ and $(T_{c1} - T_{c2})/T_{c1} = 0.5$. For the temperature range from T_{c2} to T_{c1}, the approaches that we used to calculate the resonance fields are valid only for the temperatures for which the conditions $(T_{c1} - T)/T_{c1} \ll 1$ and $(T - T_{c2})/T_{c2} \ll 1$ are fulfilled (dashed and solid lines in figure 3(c)). The dotted lines show one of the possible options and are obtained by means of joining the corresponding points near T_{c2} and T_{c1} by a smooth curve.

As is seen from the figure, for the case where the temperature is high enough ($T > T_{c1}$) and the whole particle is in the PM state, the resonance field depends only on the frequency of the exciting electromagnetic field and does not depend on T. Below T_{c1}, when the FM clusters are nucleated and grow, both the PM and FM phases make contributions to the energy dissipation. As the temperature is lowered, the resonance field for the former phase increases, whereas that for the latter decreases (for the temperatures where $(T_{c1} - T)/T_{c1} \ll 1$). At the same time, the intensity of the PM signal gets lower, in contrast to that of the FM one, which becomes higher. As T approaches T_{c2}, the PM resonance field tends to the value $H_\omega + 4\pi M_0(T_{c2})/3$, but the intensity of the PM phase is lowered to zero. The resonance field of the FM phase approaches the value which is characteristic of the ferromagnetic particle of the spherical (in the given case) shape, i.e. H_ω.

As follows from the analysis of experimental data (see, for example, [8, 10, 11, 39]), the character of the temperature dependence of the resonance fields below and above T_{c1} agrees well with what is obtained by calculations in the present work. However, as concerns the data for the temperatures far below T_{c1}, either reliable results are absent or the reliability of the available data is questionable. One of the possible reasons for this may be the low intensity of the PM signal in the low-temperature region and the appearance of ambiguity when one tries to decompose the integral signal into corresponding constituents. So, in paper [8], the detailed analysis of the magnetic resonance curves for the spherical single crystal of $La_{0.7}Ca_{0.3}MnO_3$ is carried out only for the region where the intensities of the FM and PM signals are comparable (from 270 to 210 K), although the authors note that both the signals coexist down to 160 K.

In addition, it should be noted that the experimental results obtained in work [16] are very close to the data shown in figure 3(c), including the non-monotonic dependence of the resonance field of FM phase on temperature. However, the authors do not make any comments on this behaviour.

4. Oscillations of the particle magnetization in the range of the phase coexistence

Under the resonance conditions, the magnetic field inside the particle can strongly differ from the external field due to the magneto-dipole contributions enhanced by the resonance and this can occur even if the fraction of the FM phase is small.

As follows from expressions (5), the oscillations of the magnetic moment of an individual cluster depend on the motion of the average magnetization of the particle. To describe the motion of the average magnetization, carry out a summation of the values of the magnetic moments, determined in (5), of individual clusters and divide the sum by the particle volume. To simplify the calculations, choose the Ox axis in the direction of the alternating external field. At the same time, put $h_y^{\mathrm{e}} = 0$. Then, the results obtained read

$$\begin{aligned} \langle M^{\pm} \rangle &= \chi_{\mathrm{FM}}^{\pm} \cdot h^{\mathrm{i}^{\pm}}, \\ h^{\mathrm{i}^{\pm}} &= \frac{h^{\mathrm{e}^{\pm}}}{1 + N_{\perp} \chi_{\mathrm{FM}}^{\pm}}, \end{aligned} \tag{14}$$

where $\chi_{\mathrm{FM}}^{\pm} = V^{-1} \sum_a v_a \chi_a^{\pm}$. Here, the index a, used for the summation, denotes the cluster number, h_x^{i} is the average magnetic field inside the particle, and $h^{\mathrm{e}^{\pm}} = h_x^{\mathrm{e}} = h_0 \cos \omega t$ is the external alternating magnetic field.

At the same time, the relation (5) which describes the oscillations of a separate cluster takes the form

$$m_a^{\pm} = \chi_a^{\pm} \cdot h^{\mathrm{i}^{\pm}}. \tag{15}$$

It follows from (14) and (15) that the character of the motion of magnetization is governed by $\chi_{\mathrm{FM}}^{\pm}$. It is noteworthy that the susceptibility $\chi_a^{\pm}$ is determined by the shape of the clusters and does not depend on their sizes. For this reason, the mean value of the cluster volume and the average susceptibility are statistically independent.

Accounting for this, rewrite $\chi_{\mathrm{FM}}^{\pm}$ in the form

$$\chi_{\mathrm{FM}}^{\pm} = f \cdot \overline{\chi_a^{\pm}} = (f \cdot M_0 / H_\omega) \cdot 1 \overline{/ (H_{\mathrm{FM}}^a / H_\omega - 1 \pm \mathrm{i}\alpha_{\mathrm{FM}})} \tag{16}$$

where the overbar denotes the averaging over the ensemble of FM clusters. $f = \overline{v_{\mathrm{a}}} N / V$ is the fraction of the FM phase in the particle, $\overline{v_{\mathrm{a}}}$ is the average volume of the FM cluster, and N is the number of the clusters in the particle. To calculate the components of $\chi_{\mathrm{FM}}^{\pm}$, assume that for

the ensemble of FM clusters the distribution of the fields can be characterized by a mean field $\bar{H}_{\mathrm{FM}}$ and root mean square (RMS) deviation from the mean field, ΔH_{FM}. Then we can write

$$\begin{aligned} &\bar{H}_{\mathrm{FM}} = H_z^{\mathrm{e}} - N_z M_0 f + \overline{(n_\perp - n_\parallel)} M_0, \\ &\Delta H_{\mathrm{FM}} = \sigma M_0, \\ &\sigma = \sqrt{\overline{((n_\perp - n_\parallel) - (n_\perp - n_\parallel)^2)}}. \end{aligned} \tag{17}$$

The overbars in (17) mean the average over the ensemble of FM clusters.

Consider the case where $\bar{H}_{\mathrm{FM}} \gg \Delta H_{\mathrm{FM}}$ and the distribution of the internal fields of the FM clusters constituting the ensemble is the Gaussian one:

$$\rho_{\mathrm{FM}}(H_{\mathrm{FM}}^a) = \frac{\exp(-(H_{\mathrm{FM}}^a - \bar{H}_{\mathrm{FM}})^2/\Delta H_{\mathrm{FM}}^2)}{\Delta H_{\mathrm{FM}}\sqrt{2\pi}}. \tag{18}$$

After performing the averaging in (16) with the use of the distribution (18) we obtain the expression for the coefficients of $\chi_{\mathrm{FM}}^{\pm}$:

$$\begin{aligned} &\chi_{\mathrm{FM}}^{\pm} = \chi_{\mathrm{FM}}' \pm \mathrm{i}\chi_{\mathrm{FM}}''; \\ &\chi_{\mathrm{FM}}'' = \sqrt{\frac{\pi}{2}} \cdot \frac{f M_0}{\Delta H_{\mathrm{FM}}} \cdot \exp\left\{-(H_\omega - \bar{H}_{\mathrm{FM}})^2/\Delta H_{\mathrm{FM}}^2\right\}, \\ &\chi_{\mathrm{FM}}' = \sqrt{\frac{2}{\pi}} \frac{f M_0}{\Delta H_{\mathrm{FM}}} \cdot \exp\left\{-(H_\omega - \bar{H}_{\mathrm{FM}})^2/\Delta H_{\mathrm{FM}}^2\right\} \cdot \int_0^\infty \frac{\mathrm{d}x}{x} \mathrm{e}^{-x^2} \mathrm{sh}(2x(\bar{H}_{\mathrm{FM}} - H_\omega)/\Delta H_{\mathrm{FM}}). \end{aligned} \tag{19}$$

5. Dispersive dependence of the absorption of electromagnetic radiation for the coexisting PM and FM phases

Usually, what researchers observe in experiments is the curves of the dispersive absorption of electromagnetic radiation. It is obvious that in the case under consideration both the FM and PM phases give the corresponding contributions to the energy dissipation. We take these contributions into account additively.

First, we calculate the dissipative contribution originating from the FM phase. To do this, let us use the dissipation function in the Hilbert form [40]:

$$I_{\mathrm{FM}} = \frac{\alpha_{\mathrm{FM}}}{2\gamma M_0} \sum_a \nu_a \overline{\dot{m}_a^+ \dot{m}_a^-}, \tag{20}$$

where I_{FM} is the intensity of the energy losses in the FM phase and the index a refers to the cluster number a. Here, the overbar means average in time.

With regard for the expression for $m_a^{\pm}$ (see (14) and (15)), I_{FM} can be transformed to the form

$$I_{\mathrm{FM}} = \frac{\pi}{4} \frac{\gamma H_\omega M_0 |h_0|^2}{(1 + N_\perp \chi_{\mathrm{FM}}')^2 + (N_\perp \chi_{\mathrm{FM}}'')^2} \sum_a \nu_a \left\{ \frac{1}{\pi} \frac{\alpha_{\mathrm{FM}} H_\omega}{(H_{\mathrm{FM}}^a - H_\omega)^2 + (\alpha_{\mathrm{FM}} H_\omega)^2} \right\}. \tag{21}$$

In the limit $\alpha_{\mathrm{FM}} \to 0$, the expression in braces can be replaced by the Dirac delta function $\delta(H_{\mathrm{FM}}^a - H_\omega)$. Then, taking into account that the distribution of the internal fields of FM clusters, H_{FM}^a, is described by (18), we can write the dispersive dependence of the energy losses for the FM phase in the final form:

$$I_{\mathrm{FM}} = V \cdot \frac{\gamma H_\omega \cdot |h_0|^2}{4} \cdot \frac{\chi_{\mathrm{FM}}''}{(1 + N_\perp \chi_{\mathrm{FM}}')^2 + (N_\perp \chi_{\mathrm{FM}}'')^2}, \tag{22}$$

where the real and imaginary components of χ_{FM} are given by (19).

To calculate the dissipative contribution originating from the PM phase, let us use the known expression [40]

$$I_{\mathrm{PM}} = -\frac{\omega}{2} \int\limits_{V-V_{\mathrm{FM}}} \mathrm{d}v \cdot \chi''_{\mathrm{PM}} \cdot h^{i^+} h^{i^-}, \tag{23}$$

where χ''_{PM} is the imaginary component of the tensor of paramagnetic susceptibility and $h^{i^\pm}$ are the components of the effective magnetic field inside the particle.

With regard for (14), the expression (23) can be rewritten as

$$I_{\mathrm{PM}} = -\frac{\omega}{4} \cdot V \cdot \overline{\chi''_{\mathrm{PM}}} \cdot \frac{|h_0|^2}{(1 + N_\perp \chi'_{\mathrm{FM}})^2 + (N_\perp \chi''_{\mathrm{FM}})^2}, \tag{24}$$

where $\overline{\chi''_{\mathrm{PM}}} = V^{-1} \int\limits_{V-V_{\mathrm{FM}}} \mathrm{d}v \cdot \chi''_{\mathrm{PM}}$ is the average value of the imaginary component of paramagnetic susceptibility of the particle.

To calculate $\overline{\chi''_{\mathrm{PM}}}$, let us proceed from the results obtained by Bloch [41]. According to them, the local value of the imaginary component of susceptibility is determined by the relation

$$\frac{\chi''_{\mathrm{PM}}}{\chi_0} = \frac{H_{\mathrm{PM}}}{H_\omega} \cdot \frac{1/\omega\tau_2}{(H_{\mathrm{PM}}/H_\omega - 1)^2 + (1/\omega\tau_2)^2 + (h^{i^+} h^{i^-} \tau_1 / H_\omega^2 \tau_2)}, \tag{25}$$

where τ_1, τ_2 are the constants of spin–spin and spin–lattice relaxation, respectively, and χ_0 is the static paramagnetic susceptibility.

It should be noted that for the typical range of frequencies ($\omega \sim 10^{10}$ s^{-1}) and fields ($h_0 \sim 0.5$ Oe) used in ESR measurements, and for the typical values of the relaxation constants ($\tau_1 \sim 10^{-6}$ s and $\tau_2 \sim 10^{-9}$ s), the condition $(h^{i^+} h^{i^-} \tau_1 / H_\omega^2 \tau_2) \ll (1/\omega\tau_2)^2 \ll 1$ is fulfilled. Thus, the above expression can be considerably simplified and transformed to the form

$$\chi''_{\mathrm{PM}} \approx \chi_0 \pi H_\omega \delta(H_{\mathrm{PM}} - H_\omega), \tag{26}$$

where $\delta(H_{\mathrm{PM}} - H_\omega)$ is the Dirac delta function.

Since the value of magnetic field is changed in the vicinity of the average value $\bar{H}_{\mathrm{PM}} = H_z - N_z M_0 f$, carry out the averaging of χ''_{PM} in (24) for the case where the distribution of H_{PM} is the Gaussian one:

$$\rho_{\mathrm{PM}}(H_{\mathrm{PM}}) = \frac{\exp(-(H_{\mathrm{PM}} - \bar{H}_{\mathrm{PM}})^2 / \Delta H_{\mathrm{PM}}^2)}{\Delta H_{\mathrm{PM}} \sqrt{2\pi}}, \tag{27}$$

where ΔH_{PM} is the RMS variance of the field in the volume occupied by the PM phase.

To proceed with the calculations of $\overline{\chi''_{\mathrm{PM}}}$, let us use the natural assumption that the average over the volume is equal to the average over the distribution (27). Then

$$\overline{\chi''_{\mathrm{PM}}} = \frac{1}{V} \int\limits_{V-V_{\mathrm{FM}}} \mathrm{d}v \cdot \chi''_{\mathrm{PM}} = (1 - f) \int_{-\infty}^{+\infty} \mathrm{d}H_{\mathrm{PM}} \cdot \chi''_{\mathrm{PM}} \cdot \rho_{\mathrm{PM}}(H_{\mathrm{PM}}). \tag{28}$$

Integration in (28) with regard for (26) and (27) results in

$$\overline{\chi''_{\mathrm{PM}}} = \chi_0 (1 - f) \sqrt{\frac{\pi}{2}} \cdot \frac{H_\omega}{\Delta H_{\mathrm{PM}}} \cdot \exp\left\{-(H_\omega - \bar{H}_{\mathrm{PM}})^2 / \Delta H_{\mathrm{PM}}^2\right\}. \tag{29}$$

In the final form, with regard for the contributions originating from the FM and PM phases, the expression for the dissipative losses can be represented as

$$\begin{aligned} I &= I_0 \cdot \frac{\chi''_{\mathrm{FM}} + \overline{\chi''_{\mathrm{PM}}}}{(1 + N_\perp \chi'_{\mathrm{FM}})^2 + (N_\perp \chi''_{\mathrm{FM}})^2}, \\ I_0 &= V \cdot \frac{\gamma H_\omega h_0^2}{4}. \end{aligned} \tag{30}$$

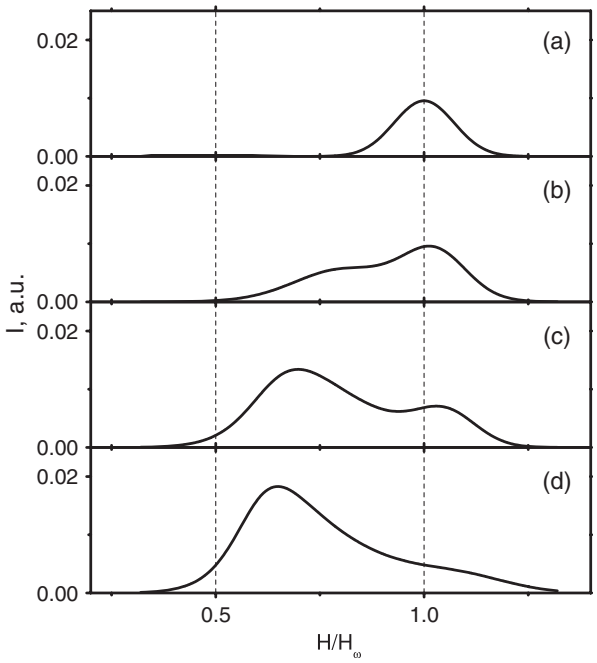

Figure 4. The curves of the dispersive absorption for the spherical particle, calculated for various temperatures. The value of the parameter $(T_{c1} - T)/T_{c1}$ is equal to 0 (a); 0.045 (b); 0.01 (c); 0.15 (d). The other parameters used for the calculations are $\Delta H_{FM}/H_\omega = 0.2$, $\Delta H_{PM}/H_\omega = 0.1$, and $\chi_0 = 7 \times 10^{-3}$.

Figures 4(a)–(d) show the curves of the dispersive absorption calculated for the spherical particle for a series of temperatures in the vicinity of T_{c1}. The calculations were performed for the same values of the parameters as were used in section 3.3. The variables that are functions of temperature are the magnetization $M_0(T)$ and the fraction of FM phase $f(T)$ (see figure 3). All the other parameters were set to not depend on temperature. It is noteworthy that account of the temperature dependence of χ_0 will not change the picture qualitatively but only lead to the renormalization of the temperature scale.

As follows from the calculations, for the high temperature region ($T/T_{c1} \geqslant 1$), the absorption spectrum consists of a single line centred at H_ω (see figure 4(a)). As the temperature is lowered below T_{c1}, the contribution which is associated with the nucleation and growth of the FM clusters and characterized by a resonance field smaller than H_ω becomes noticeable (see figure 4(b)). Further decrease in the temperature leads to the increase in the FM signal intensity with a simultaneous decrease in its resonance field value. The inverse situation is characteristic of the PM signal (see figure 4(b)–(d)). The shape of the resulting curve strongly depends on the saturation magnetization and fraction of the FM phase, magnetic susceptibility of the PM phase, the values of ΔH_{FM} and ΔH_{PM}, as well as on the character of the temperature changes of these parameters. The further decrease in the temperature gives rise to the complete domination of the FM phase and to the approach of the parameters of the resonance curve to those characteristic of the uniformly magnetized FM particle whose shape is the governing factor determining the location of the ferromagnetic resonance line.

Finally, let us discus the limits of applicability of the approach developed. The main idea of this approach is to replace all interactions at any point of the particle with an average (effective)

interaction (see (1)). Thus, the whole formalism is valid provided that the basic concepts of the mean field approximation are true [19, 38]. Comparison of the results calculated in this paper with those obtained experimentally shows that both the character of the dependences and features of their temperature change (see figures 3 and 4) well agree with the experimental data obtained by various research groups on the single- and polycrystalline samples of the doped manganites [9, 11, 16, 39]. Further extensive work is needed to specify the factors influencing the parameters of resonance curves within the region of the PM and FM phase coexistence in order to deepen the comprehension of the physics of this class of materials and show ways for the goal-oriented modification of their properties.

6. Conclusions

In the present work, the magnetization dynamics is considered for the system in which the PM and FM phases coexist over a wide temperature range and are in an equilibrium state. It is shown that in the cases where one of the phases prevails the energy of the system achieves a minimum when the clusters of the second phase take the shape of the regions elongated along the direction of the external magnetic field. Near the boundaries of the phase coexistence region, the expressions for the resonance fields of the PM and FM phases are obtained and the dispersive dependence of the energy absorption of electromagnetic radiation is calculated. The resonance field for the PM phase is shown to increase with the temperature lowering, while the resonance field for the FM phase first decreases and then approaches a value characteristic of the uniformly magnetized ferromagnetic sample. It is shown that the scenario described agrees well with the experimental data obtained on various kinds of samples of doped perovskite manganites.

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References

[1] Dagotto E, Hotta T and Moreo A 2001 *Phys. Rep.* **344** 1
[2] Rivadulla F, Frita-Alvite M, Lopez-Quintela M A, Hueso L E, Miguens D R, Sande P and Rivas J 2002 *J. Appl. Phys.* **91** 785
[3] Volkov N V, Petrakovskii G A, Vasil'yev V N and Sablina K A 2002 *Phys. Solid State* **44** 1350
[4] Winkler E, Causa M T, Ramos C A and De Biasi E 2004 *Physica* B **354** 51
[5] Ding T, Zheng W, Zang J, Tian H, Zheng B, Wang X, Yu S and Wang Y 2005 *J. Magn. Magn. Mater.* **293** 782
[6] Tovstolytkin A, Pogorily A, Vovk A, Podyalovskii D, Lezhnenko I and Matviyenko A 2004 *J. Magn. Magn. Mater.* **272–276** 1839
[7] Tovstolytkin A I, Pogorily A N, Lezhnenko I V, Matviyenko A I, Podyalovskii D I and Kravchik V P 2003 *Phys. Solid State* **45** 1857
[8] Joh K W, Lee C H, Lee C E, Hur N H and Ri H-C 2003 *J. Phys.: Condens. Matter* **15** 4161
[9] Aswal D K, Singh A, Kadam R M, Bhide M K, Page A G, Bhattacharya S, Gupta S K, Yakhmi J V and Sahni V C 2005 *Mater. Lett.* **59** 728
[10] Shames A I, Auslender M, Rozenberg E, Gorodetsky G, Sominski E, Gedanken A and Mukovskii Ya M 2006 *J. Magn. Magn. Mater.* **300** 12
[11] Singh A, Chowdhury P, Padma N, Aswal D K, Kadam R M, Babu Y, Jayanth Kumar M L, Viswanadham C S, Goswami G L, Gupta S K and Yakhmi J V 2006 *Solid State Commun.* **137** 456
[12] Shengelaya A, Zhao G-M, Keller H and Müller K A 1996 *Phys. Rev. Lett.* **26** 5296
[13] Huber D L 1998 *J. Appl. Phys.* **83** 6949

[14] Huber D L, Laura-Ccahuana D, Tovar M and Causa M T 2007 *J. Magn. Magn. Mater.* **310** e604
[15] Zhao G-M, Keller H, Hofer J, Shengelaya A and Müller K A 1997 *Solid State Commun.* **104** 57
[16] Phan T L, Tho N D, Phand M H, Ha N D, Chau N and Yu S C 2006 *Physica* B **371** 317
[17] Sparks M, Loudon R and Kittel C 1961 *Phys. Rev.* **122** 791
[18] Shlömann E 1969 *Phys. Rev.* **182** 632
[19] Gurevich A G and Melkov G A 1996 *Magnetization Oscillations and Waves* (Boca Raton, FL: CRC Press)
[20] Raikher Yu L and Stepanov V I 1994 *Phys. Rev.* B **50** 6250
[21] Raikher Yu L and Stepanov V I 1995 *J. Magn. Magn. Mater.* **149** 34
[22] Marin C N 2006 *J. Magn. Magn. Mater.* **300** 397
[23] Haghiri-Gosnet A-M and Renard J-P 2003 *J. Phys. D: Appl. Phys.* **36** R127
[24] Landau L D and Lifshitz E M 1994 *Statistical Physics* Part 1 (*Course of Theoretical Physics* vol 5) 3rd edn (Oxford and New York: Pergamon)
[25] Novak P, Marysko M, Savosta M M and Ulyanov A N 1999 *Phys. Rev.* B **60** 6655
[26] Hwang H Y, Cheong S-W, Radaelli P G, Marezio M and Batlogg B 1995 *Phys. Rev. Lett.* **75** 914
[27] Amaral V S, Araujo J P, Pogorelov Yu G, Tavares P B, Sousa J B and Vieira J M 2002 *J. Magn. Magn. Mater.* **242–245** 655
[28] Archibald W, Zhou J-S and Goodenough J B 1996 *Phys. Rev.* B **53** 14445
[29] Rodbell A S and Bean C P 1962 *J. Appl. Phys.* **33** 1037
[30] Bean C P and Rodbell D S 1962 *Phys. Rev.* **126** 104
[31] Banerjee B K 1964 *Phys. Lett.* **12** 16
[32] Dörr K 2006 *J. Phys. D: Appl. Phys.* **39** R125
[33] Gor'kov L P 1998 *Phys.—Usp.* **41** 589
[34] Jaime M, Lin P, Chun S H, Salamon M B, Dorsey P and Rubinstein M 1999 *Phys. Rev.* B **60** 1028
[35] Alonso J L, Fernandez L A, Guinea F, Laliena V and Martin-Mayor V 2001 *Phys. Rev.* B **63** 054411
[36] Lesnik A G 1985 *Metallofizika* **7** 3
[37] Lesnik A G 1987 *Metallofizika* **9** 83
[38] Chikazumi S 1997 *Physics of Ferromagnetism* 2nd edn (Oxford: Oxford University Press)
[39] Tovstolytkin A I, Pogorily A N, Dzhezherya Yu I, Podyalovskii D I and Lysenko V A 2006 *Proc. 20th Int. Conf. On New Magnetic Materials of Microelectronics (Moscow, June 2006)* (Moscow: Physical Faculty of Moscow State University) p 654 (in Russian)
[40] Landau L D, Lifshitz E M and Pitaevskii L P 1984 *Electrodynamics of Continues Media* 2nd edn (Oxford: Pergamon)
[41] Bloch F 1946 *Phys. Rev.* **70** 460