

## FMR linewidth and resonance frequency shift caused by double exchange in doped lanthanum manganites

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A mechanism is proposed for FMR broadening and resonance frequency shift in a ferromagnetic conducting sample (hopping conductivity) of a doped lanthanum manganite due to the double exchange (DE) under FMR conditions (i.e. by the dynamic double exchange, DDE). It has been shown that the mechanism defines the resonance frequency shift and the FMR damping coefficient  $\omega''$  (the band width). This shift and broadening mechanism is effective when the variable field frequency  $\omega$  is of the order of the mobile electron relaxation rate  $\tau^{-1}$ . The inequality  $\tau^{-1} > \omega''$  should be met for the mechanism validity. The expressions obtained for  $\omega\tau < 1$  agree qualitatively with the observed dependence of the FMR band width on the exciting variable field frequency as well as (under certain conditions) with experimental data on the temperature dependence of the FMR spectrum. In our opinion, this research may favor the obtaining of quantitative data on the DE constants.

Предложен механизм сдвига резонансной частоты и уширения ФМР в ферромагнитном проводящем образце (прыжковая проводимость) допированного лантанового манганита, обусловленный двойным обменом (ДО) в условиях ФМР (динамическим двойным обменом, ДДО). Показано, что этот механизм обуславливает сдвиг резонансной частоты и коэффициент затухания  $\omega''$  (ширина линии) ФМР. Предложенный механизм сдвига и уширения, обусловленный ДДО, является эффективным, когда частота переменного поля  $\omega$  имеет порядок скорости релаксации подвижных электронов  $\tau^{-1}$ . Для справедливости предложенного механизма необходимо выполнение неравенства  $\tau^{-1} > \omega''$ . Полученные выражения при  $\omega\tau < 1$  качественно согласуются с наблюдаемой зависимостью ширины линии ФМР от частоты возбуждающего его поля, а также, при определенных условиях, с экспериментальными данными по температурной зависимости спектра ФМР. По нашему мнению, данное исследование может способствовать получению количественной информации о константах ДО.

The lanthanum manganites (LM) with the perovskite structure doped with divalent metal ions are under an intensive study [1-3]. An information of great importance on the materials is provided by studying the internal magnetic fields, especially in the region of the phase transitions in LM samples with the divalent ion concentration near 0.3, where the strongest colossal magnetoresistance effect is observed. The temperature dependence of EPR spectrum (the resonance frequency and the band width) in a wide temperature range is investigated in [4-9]. In particular, the works [4, 5] are worth to be noted, where the EPR spectrum recording in  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  with  $x=0.1, 0.2, 0.25$  and 0.33 is complemented by the independent measurement of the longitudinal electron relaxation time using the modulation technique. The following experimental results have been obtained in

[4-9] for LM ferromagnetic metallic (FMM) phase. The measured band width has been found [7, 8] to increase at the increasing microwave frequency exciting ferromagnetic resonance (FMR) ([7], sample  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ ). The authors [7] believe that the strong dependence of the band width on the microwave frequency is due to the magnetization inhomogeneity caused, in particular, by the demagnetization effects. At  $T < T_C$ , the following effects are observed as the temperature decreases: the resonance field shift toward lower fields being observed at the approaching to  $T_C$  from above becomes sharper in the FMM phase as compared to the paramagnetic one [8]. In ceramic and polished samples, the band broadening observed at the approaching to  $T_C$  from above becomes sharper [4, 5, 7]; as to the single crystals, either the same behavior takes place [9], or the narrowing of the paramagnetic region is replaced by a plateau.

As is known, the double exchange (DE) model is the base one for the interpretation of the doped LM physical properties in the FMM phase [9, 11]. The DE microscopic model in LM began to be elaborated still at the first flare of interest to these compounds half-century ago [12]. The modern status of this direction is reflected in the reviews [2, 3] and references therein. Those reviews are concentrated mainly on the double exchange manifestations in LM transport properties (conductivity, magnetoresistance). In contrast, we shall be interested in the electron spin dynamics under magnetic resonance conditions, i.e. under the action of a microwave (mw) field on the samples. The above-mentioned dependence of FMR width on mw frequency can testify that the spin dynamics in LM under the resonance conditions is a specific phenomenon.

In contrast to the approach of [2, 12], where the change of the longitudinal (along the spontaneous magnetization) component of the total magnetization during the double exchange was taken into account, we shall also consider the non-zero stationary values of the transverse magnetization components, i.e. the phased spin precession. Therefore, unlike the works [2, 12], the DE influence on the transverse magnetization component motion will be investigated in this paper, too. The macroscopic interaction of magnetization components caused by the double exchange at the presence of ferromagnetic resonance (FMR) of  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions, which will be introduced by us in what follows, will be referred to as the dynamic double exchange (DDE). In this model, the DE is the reason for FMR frequency shift and band broadening.

Note that, in contrast to the FMM phase, there is a strong non-uniaxial magnetocrystalline anisotropy in the ferromagnetic insulating (FMI) phase, which is characteristic for the orbitally ordered FMI systems [10]. In the FMI phase, the magnetocrystalline anisotropy contributes substantially to the shift and asymmetrical broadening of the FMR band [9]; however, we shall not consider such a case. The magnetic state of  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  with  $x = 0.22$  at temperatures exceeding 100 K is estimated in [9] as the homogeneous and metallic; similar samples will form the object of our investigation. The elaborated DDE model is used to calculate the FMR spectrum (resonance frequency and band width) in the LM ferromagnetic conducting phase. The obtained results agree qualitatively with the experiments.

At  $x < 0.5$ , there is a fraction  $x$  of holes ( $\text{Mn}^{4+}$  ions) with respect to the total number of  $\text{Mn}$  ions in doped manganites. However, to describe the effect the such doping on the sample dynamic magnetic properties, it is more convenient to speak about the extra (mobile) electrons ( $e_g$ ) in  $\text{Mn}^{4+} + \text{Mn}^{4+}$  pairs. In the well-known work [12], the double exchange problem is reduced to the microscopic interaction within a single pair of  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions. In [12] model, the mobile electron energy includes the Hund exchange with a constant  $J$  (diagonal terms of the Hamiltonian in the matrix form) and hopping energy with transfer integral  $b$  (non-diagonal terms). When the condition  $J \gg b$  is met (what is most adequate to the DE situation [2, 11]), the secular equation solution corresponding to the lower Hund level has the form  $E = -JS \pm b \cos(\theta/2)$ , where  $\theta$  is the angle between the core spins. Thus, this level is split into two levels symmetrically situated with respect to  $-JS$ . The *electron level* split value proportional to the transfer integral is used below in the expression for the probability of the total spin to have the definite value of its z-component in the macroscopic problem of the *sample* static susceptibility calculation. At this point, the initial (non-split) position of the

electron level (-JS) does not characterize DE, since in the  $J \gg b$  approximation, the Hund exchange does not depend on the mutual position of core spins [2].

Similar to [12], we shall consider also one pair of  $\text{Mn}^{3+}$ - $\text{Mn}^{4+}$  ions with the  $\mathbf{S}_1$  and  $\mathbf{S}_2$  spins, where the difference in the indices is due to the mobile electron presence in one of the ions, the spin of that electron being parallel to the core spin. The state corresponding to the electron presence in ion 1 is energetically degenerate with the state of its presence in ion 2. In the quantum-mechanical sense, the electron hopping between the ions is the tunnel transition from one potential well to the another having the same depth. As is known [13], such transitions result in the symmetric double split of each of the initial levels. Let this result be described classically, in the language of coupled oscillators, what is necessary for the further description of the magneto-resonance processes. The inter-oscillator coupling is characterized by the transfer integral value (without the explicit consideration of the hopping process). The total core spin direction will be taken as the Z axis. Then, it can be supposed approximately that, as in [12],  $\cos(\theta/2) \approx (S_{1z} + S_{2z})/2S$ . Using the result of [12], the hopping can be believed to cause "molecular field"  $B_z = -(b/g\mu_B)\cos(\theta/2)$  in the pair. As a result of this field action, the pair spin-system possesses the internal energy  $U_{DE}^{micr} = -(b/2S)(S_{1z} + S_{2z})^2$ . In contrast to [12], where the problem of the LM spin-system static equilibrium susceptibility calculation was studied, the problem of its behavior under FMR conditions is considered by us. Hence, the main attention will be paid to the tilting of the total spin from the Z axis at the electron hopping. That is why, using the total spin square saving condition, the  $U_{DE}^{micr}$  energy can be presented in the form

$$U_{DE}^{micr} = (b/2S) \left\{ (S_{1x} + S_{2x})^2 + (S_{1y} + S_{2y})^2 \right\} \quad (1)$$

Similar to [2, 12], at  $J \gg b$  the Hund exchange will not contribute to the FMR spectrum. Therefore, instead of it, we shall take the Zeeman energy in the external constant magnetic field  $B_0$  as the ground state of the coupled oscillators:  $g\mu_B B_0 (S_{1z} + S_{2z})$ .

The equations of the pair spin precession under the action of the Hamiltonian  $g\mu_B B_0 (S_{1z} + S_{2z}) + U_{DE}^{micr}$  in the linear case (when the operators  $S_{1z}$  and  $S_{2z}$  are considered as the c-numbers and  $S_{1z} = S_{2z} \equiv S_z$ ) have the form

$$\begin{aligned} \dot{S}_1^+ &= (i/\hbar)(g\mu_B B_0 - bS_z/S)S_1^+ - i(bS_z/\hbar S)S_2^+ \\ \dot{S}_2^+ &= (i/\hbar)(g\mu_B B_0 - bS_z/S)S_2^+ - i(bS_z/\hbar S)S_1^+ \end{aligned} \quad (2)$$

Here we have used the operators  $S_{1,2}^+ \equiv S_{1,2x} + iS_{1,2y}$  instead of  $S_{1,2x}$  and  $S_{1,2y}$  because in such a way it is possible to work with the two coupled first order equations instead of second order ones. It is seen that the Eqs. (2) are the motion equations of two coupled oscillators with the equal partial frequencies  $\omega_p = g\mu_B B_0 - bS_z/S \approx g\mu_B B_0$  and the coupling coefficients  $\alpha = (bS_z/\hbar S)$  [14]. The normal frequencies of these oscillators are equal to

$$\omega_{\pm} = g\mu_B B_0 / \hbar \pm bS_z / \hbar S. \quad (3)$$

Here  $S_z/S \approx \cos(\theta/2)$ . Note that the total transverse projection precesses at the frequency  $\omega_-$ .

Thus, the interaction (1) results in the appearance of two precession frequencies of the pair spin-system shifted with respect to their partial frequency by the values equal to  $\pm(b/\hbar)\cos(\theta/2)$ . Here, the situation is similar to the inversion split of energy levels of ammonia molecule [15]. Therein, the nitrogen atom nucleus can be found at the both sides of the plane defined by hydrogen atoms and make exactly identical oscillations about two steady positions (at one side and another). However, these oscillations are not independent: the nitrogen nucleus can transit from one side of the hydrogen plane to another due to the tunnel effect. Thus, the ammonia molecule is also similar to two coupled oscillators with the equal partial frequencies and the normal frequencies, one of which

is lower than the partial one, and the other is higher. Therefore, each of its energy levels appears to be split into two sublevels (just this split is referred to as the inversion one). For the mobile electron hopping in  $\text{Mn}^{3+} + \text{Mn}^{4+}$  ion pair, a similar split takes place.

Till now, we had to do with one such pair. If a set of pairs is considered, their precession motions, being combined, will not result in a non-zero transverse component of the sample magnetization because of their dephasing. However, under an external variable field with the frequency close to the precession frequency, i.e. in FMR experiment, the precessions of separate pairs can be phased, providing the non-zero transverse components  $M_x$  and  $M_y$  necessary for the stationary macroscopic manifestation of the hopping effect on the precession characteristics. Due to the phasing of the pair precession, the macroscopic values of  $M_x$  and  $M_y$  can be introduced at this stage without the pair explicit summation. We suppose that the sample is a small sphere magnetized up to the saturation at the set temperature, and we shall limit ourselves by consideration of the magnetization homogeneous oscillations.

The macroscopic magnetic energy density corresponding to (1) under FMR conditions is believed to have the form

$$U_{DE}^{macr} = b_0 \frac{M_x^2 + M_y^2}{2}. \quad (4)$$

Here  $M_x$  и  $M_y$  correspond to the total spin of the  $\text{Mn}^{3+} + \text{Mn}^{4+}$  ions (in microscopic languages, to the total projections  $S_{1x,y} + S_{2x,y}$ ). We consider that, similar to the Heisenberg exchange of the nearest magnetic ions [16], the interaction constant is equal to

$$b_0 = \frac{zb}{(g\mu_B)^2 n_m}. \quad (5)$$

Here  $z$  is the number of the nearest ions, between which the mobile electron has the non-zero transfer integral,  $n_m$  is the total concentration of magnetic ions. The totale magnetic energy density of a ferromagnetic conducting LM sample subjected to a constant field  $B_0 \parallel Z$ , where  $Z \parallel \mathbb{D}_0$  and to a variable field  $B_1(t) \parallel X$  has the form:

$$H = -M_z B_0 + U_{DE}^{macr} - 2B_1 M_x \cos \omega t. \quad (6)$$

To investigate the evolution of magnetization components in a magnetically ordered sample, we use the Landau equation [17]

$$\frac{d\mathbf{M}}{dt} = -\gamma [\mathbf{M} \times \mathbf{B}] \quad (7)$$

where  $\gamma = g\mu_B/\hbar > 0$  is the giromagnetic ratio considered in the frame of continual approach as a phenomenological parameter [17]. The effective field  $\mathbf{B}$  acting on the spin-system, according to Landau [17], is calculated as

$$\mathbf{B} = -\frac{\partial H}{\partial \mathbf{M}}$$

The equations of motion for the transverse magnetization components following from the Landau equation at the magnetic energy density  $-M_z B_0 + U_{DE}^{macr}$  have the form

$$\begin{aligned} \dot{M}_x &= -\gamma B_0 M_y - \gamma b_0 M_y M_z \\ \dot{M}_y &= \gamma B_0 M_x + \gamma b_0 M_x M_z \end{aligned} \quad (8)$$

(the longitudinal magnetization component does not change under the action of  $-M_z B_0 + U_{DE}^{macr}$ , it

can be changed only under the action of the relaxation processes, as considered below).

The split of the each core level in a sample described above means the existence possibility of the appropriate additive to the longitudinal magnetization  $m_z$  directed against the Z axis. In other words, the mobile electron hopping between the cores cause a potential possibility of the fixed deviation of the total core magnetization from the equilibrium direction. This possibility can be realized under the action of a mw field which tilts the magnetization by such an angle that the longitudinal magnetization becomes equal to  $M_z = M_0 - m_z$ , where  $m_z$  is the magnetization connected with different populations of the doubly split levels. At thermodynamic equilibrium, the populations are defined by the Boltzmann distribution; then  $m_z = m_{z\infty}$ . If the equilibrium disturbance takes place, then  $m_z$  relaxes to  $m_{z\infty}$  with a large relaxation rate  $\tau^{-1}$  characteristic for mobile electrons according to the equation

$$\dot{m}_z = -\frac{m_z - m_{z\infty}}{\tau}$$

From the equations (8) it is seen that the core energy is decreased by the value

$$\Delta\varepsilon = g\mu_B b_0 m_z \quad (9)$$

because of the electron hopping at the presence of mw field.

Since the energy is fast dissipated from the mobile electrons to the lattice, the magnetization will easily absorb the mw field energy. From the condition of the total magnetic moment conservation in an ideal (loss-free) ferromagnet at the tilting under a mw field [18], we have  $m_z \approx (M_x^2 + M_y^2)/2M_0$ , and  $\Delta\varepsilon$  takes the form

$$\Delta\varepsilon = g\mu_B b_0 \frac{M_x^2 + M_y^2}{2M_0}. \quad (10)$$

Due to the relation of  $m_z$  with  $M_x$  and  $M_y$ , the  $m_z$  value is modulated by mw field. The modulated value  $m_z$  develops under the relaxation what, in turn, is reflected back to the precession, i.e. on FMR.

If the electron relaxation is indefinitely fast,  $m_z$  will be always equal to its equilibrium value  $m_{z\infty}$ . If it has the finite rate, then  $m_z$ , on the one hand, is modulated by the variable field ( $m_z = \tilde{m}_z \exp(i\omega t)$  with the complex amplitude  $\tilde{m}_z$  because of the retardation of the field following), and on the other hand, tends at the electron relaxation rate to the instantaneous equilibrium value, which is modulated, too:  $m_{z\infty} \exp(i\omega t)$ . This process occurs according to the equation:

$$\frac{d}{dt}(\tilde{m}_z \exp(i\omega t)) = -\frac{\tilde{m}_z \exp(i\omega t) - m_{z\infty} \exp(i\omega t)}{\tau}. \quad (11)$$

Thus, we obtain the following picture of the magnetization motion: the magnetization deviates from the Z axis under the variable field action, and the relaxation of the mobile carriers, which energy is dissipated to the lattice, tends to bring the magnetization to the equilibrium position. This process is reflected by the negative additive  $-m_z$  to  $M_0$ , which also has its equilibrium value. It follows from the above that, besides the precession, also the magnetization nutation takes place. The combination of the nutation motion in two directions (from the Z axis and towards it) results finally in the modulation of  $m_z$  and  $m_{z\infty}$ .

For a stationary process ( $d\tilde{m}_z/dt = 0$ ), the solution of Eq. (11) has the form:

$$\tilde{m}_z = \frac{m_{z\infty}}{1 + i\omega t}. \quad (12)$$

Let a term related to the action of the rotating component  $B_1 \exp(i\omega t)$  of the variable field be

added to the equations (8). Let make use of the complex amplitude method for the magnetization transverse components –  $M_{x,y} = \widetilde{m}_{x,y} \exp(i\omega t)$ . Also, the value  $m_z$  be expanded in series with respect to the magnetization transverse components to find its influence on the precession:

$$m_z = \left\{ \left( \frac{\partial m_{z\infty}}{\partial M_x} \right)_0 \tilde{m}_x + \left( \frac{\partial m_{z\infty}}{\partial M_y} \right)_0 \tilde{m}_y \right\} \frac{\exp(i\omega t)}{1+i\omega\tau} \quad (13)$$

Then we obtain from (8) the following precession equations:

$$\begin{aligned} i\frac{\omega}{\gamma} \tilde{m}_x + \left[ B_0 - \frac{M_0/g\mu_B}{1+i\omega\tau} \left( \frac{\partial \Delta\varepsilon}{\partial M_y} \right)_0 \left( \frac{\partial m_{z\infty}}{\partial M_y} \right)_0 \right] \tilde{m}_y &= 0 \\ \left[ B_0 - \frac{M_0/g\mu_B}{1+i\omega\tau} \left( \frac{\partial \Delta\varepsilon}{\partial M_x} \right)_0 \left( \frac{\partial m_{z\infty}}{\partial M_x} \right)_0 \right] \tilde{m}_x - i\frac{\omega}{\gamma} \tilde{m}_y &= M_0 B_1 \end{aligned} \quad (14)$$

In (13, 14) the values of the derivatives are taken at the equilibrium direction of the sample total magnetization at the presence of an mw field (this direction is tilted with respect to Z axis), what is denoted by the index “0”. The instantaneous equilibrium value of  $m_z$  corresponding to the split  $\Delta\varepsilon_\infty = g\mu_B b_0 m_{z\infty}$ , is equal to the solution of the transcendental equation

$$m_{z\infty} = \frac{g\mu_B n_e}{2} \tanh \frac{g\mu_B b_0 m_{z\infty}}{2k_B T}.$$

Here  $n_e = xn_m$  is the concentration of the mobile holes (in this text are referred to as the “mobile electrons”).

Let these equations to be considered first without the term  $M_0 B_1$ , i.e. formally as the free precession equations. Then the complex frequency of this precession  $\omega = \omega' + i\omega''$  contains the FMR resonance frequency  $\omega'$  and the delay factor  $\omega''$  (the band width) of the spin-system of a ferromagnetic conducting sample caused by the DDE. For the corresponding field values appearing in an experiment with the field passage we have from the Eqs. (14)

$$\frac{\omega''}{\gamma} = \frac{M_0 n_e}{8\Delta\varepsilon_\infty} \left\{ \left( \frac{\partial \Delta\varepsilon}{\partial M_x} \right)_0^2 + \left( \frac{\partial \Delta\varepsilon}{\partial M_y} \right)_0^2 \right\} C(T) \frac{\omega' \tau}{1 + \omega'^2 \tau^2} \quad (15)$$

$$\frac{\omega'}{\gamma} = B_0 - \frac{M_0 n_e}{8\Delta\varepsilon_\infty} \left\{ \left( \frac{\partial \Delta\varepsilon}{\partial M_x} \right)_0^2 + \left( \frac{\partial \Delta\varepsilon}{\partial M_y} \right)_0^2 \right\} \frac{C(T)}{1 + \omega'^2 \tau^2} \quad (16)$$

where the notation is introduced

$$C(T) \equiv \frac{\Delta\varepsilon_\infty}{k_B T} \operatorname{sech}^2 \left( \frac{\Delta\varepsilon_\infty}{2k_B T} \right).$$

Let the angles  $\theta$  and  $\Phi$  be introduced by the ratios

$$M_x = M_0 \sin \Delta\theta \approx M_0 \Delta\theta; \quad M_y = M_0 \sin \Delta\Phi \approx M_0 \Delta\Phi$$

( $\theta$  is the polar angle and  $\Phi = \varphi \sin \theta$ , where  $\varphi$  is the azimuth angle of the sample magnetization in the spherical co-ordinate system). Note that

$$\frac{\partial}{\partial M_x} = \frac{1}{M_0} \frac{\partial}{\partial \theta}; \quad \frac{\partial}{\partial M_y} = \frac{1}{M_0} \frac{\partial}{\partial \Phi}.$$

Let these relations be substituted into (15), (16) and extract from  $B_0 - \frac{\omega'}{\omega}$  its dynamic part (i.e. the part, which tends to 0 at  $\omega\tau \rightarrow 0$ ). Then it is easy to see the complete analogy between the expressions (15), (16) of this work with the expressions (9.6.29) and (9.6.30) of [17], where the form of  $\Delta\varepsilon$  is not detailed. For our  $\Delta\varepsilon$  (see (10)), the resonance field and FMR band width values (the expressions (15), (16)) will have the following form:

$$\frac{\omega''}{\gamma} = \frac{1}{4} n_e g \mu_B b_0 C(T) \frac{\omega' \tau}{1 + \omega'^2 \tau^2} \quad (17)$$

$$\frac{\omega'}{\gamma} = B_0 - \frac{1}{4} n_e g \mu_B b_0 \frac{C(T)}{1 + \omega'^2 \tau^2} \quad (18)$$

It is of interest that (17) differs considerably from the usual expression for the spin resonance band width due to internal local magnetic field fluctuations on the spins under consideration. The expression (17) is also proportional to the square of the internal field, but of the regular one. The irreversibility is entered here not by the averaging of the internal field square, but by the fast relaxation of the mobile carriers (for the validity of the offered mechanism, the inequality  $\tau^{-1} \gg \omega''$  is to be met). Note that this inequality is fulfilled when  $\tau$  decreases at the temperature decreasing from 300 up to 60 K from  $1.6 \cdot 10^{-11}$  s up to  $10^{-13}$  [19], and the homogeneous band width of about  $3 \cdot 10^{-3}$  T [4, 5].

The experimental study of the electron resonance broadening in a wide temperature range (both higher and lower than  $T_c$ ) was carried out in numerous works [4-9]. First of all, it is to note the non-trivial fact found out in [7, 8], that the FMR width in the region near to  $T_c$  depends on the frequency of the variable field exciting the resonance, namely, it is increased parallel with the frequency. Such dependence follows from the formula (17), obtained in this work (where in the r.h.s. we take  $\omega' = \omega$  for simplicity) at  $\omega\tau < 1$ . Further, the temperature dependence of the EPR width observed in [4-9] was characterized by a diminution at the temperature decrease from 300 K, had a minimum at  $T = T_{\min}$  somewhat above  $T_c$  ( $T_{\min} \sim (1.1 - 1.5)T_c$ ), while below it, either a broadening was revealed (ceramics and polycrystalline samples), or the narrowing observable in the paramagnetic region was replaced by a plateau. The reasons for the temperature behavior at  $T > T_c$  are discussed in detail in [4, 5]. The subject of our research is the behavior at  $T < T_c$  being considered using the proposed DDE model. It is described by the expression (17) which gives the FMR homogeneous width. Its temperature behavior is caused by the temperature behavior of the functions  $C(T)$  and  $f(\omega, T) \equiv \omega\tau / (1 + \omega^2\tau^2)$ . The latter function has a maximum at  $\omega\tau = 1$ , i.e. at the mobile electron relaxation rate equal to the resonant frequency of the localized spins. The tendency to the electron resonance line broadening with the temperature decrease below  $T_{\min}$  can be provided by (i) the growth of  $C(T)$  with the lowering  $T$ , occurring at  $(k_B T / \Delta\varepsilon) > 0.65$  [17]; or (ii) increase of  $f(\omega, T)$  at

$$-\frac{\tau}{T} (1 + \omega^2 \tau^2) + \frac{\partial \tau}{\partial T} (1 - \omega^2 \tau^2) < 0. \quad (19)$$

The observed frequency dependence is favored by the condition  $\omega\tau < 1$ . If the condition is met, the inequality (19) is automatically fulfilled at  $\partial\tau/\partial T < 0$ . For this relation, the usual activation law (with the activation energy independent on  $T$ ) of the  $\tau$  temperature dependence matches:  $\tau = \tau_\infty \exp(E_a/k_B T)$ ; ( $E_a = \text{const}$ ). If  $\partial\tau/\partial T > 0$ , what takes place, for example, at tempera-

ture dependence  $\tau = \tau_0 \exp(aT)$ ;  $(E_a = ak_B T^2)$  used successfully in [19] in the interpretation of NMR experiments on  $^{55}\text{Mn}$  in FMM phase of LM, then the condition

$$aT < \frac{1 + \omega^2 \tau^2}{1 - \omega^2 \tau^2} \quad (20)$$

is favorable for the band broadening with the temperature lowering.

If  $\omega\tau > 1$ , the  $\tau$  temperature dependence from [19] *a priori* matches, however, the assumption  $E_a = \text{const}$  requires the validity of the inequality

$$\frac{\text{const}}{k_B T} < \frac{\omega^2 \tau^2 + 1}{\omega^2 \tau^2 - 1} \quad (21)$$

At  $\omega\tau \sim 1$  (when the proposed relaxation mechanism is most effective) the fulfillment of the both inequalities (20) and (21) seems to be real. Certainly, the  $\omega''$  increase with  $T$  decrease can be interpreted also as a result of  $C(T)$  growth at the decrease of  $f(\omega, T)$ , or by  $f(\omega, T)$  growth at the decrease of  $C(T)$ . However, the absence of maxima in the observed  $\omega''(T)$  dependence suggests that, most likely, the combination of (i) and (ii) is real.

The resonance field (i.e. the constant field value at which the absorption signal  $\chi''$  maximum is observed) decreases at the temperature lowering [4, 5, 7, 8]. Such a dependence follows from (18) at  $\frac{(1 + \omega^2 \tau^2)}{T} + \frac{\partial \tau}{\partial T} 2\omega^2 \tau > 0$ , which is automatically satisfied at  $\partial \tau / \partial T > 0$ , i.e. the activation law from [19] matches.

As to the quantitative comparison with the experimental results on FMR band width and shift in non-single-crystal samples, it cannot to be strictly carried out at this stage, since the measured band width in such samples contains a considerable inhomogeneous contribution caused by the disorder of demagnetizing fields, related to the microinhomogeneities of the sample surface and the pores between the crystallites [8].

Summarizing, the following mechanism is proposed for FMR broadening and resonance frequency shift in a ferromagnetic conducting sample (hopping conductivity) of a doped lanthanum manganite, caused by the double exchange (DE) under FMR conditions (the dynamic DE, or DDE). The mobile electron hopping within a pair of  $\text{Mn}^{4+}+\text{Mn}^{4+}$  ions results in a tilting of the pair total spin from its equilibrium direction, i.e. to an increase of its nonzero transverse components. The precession of the tilted total spin in internal local fields occurs out of phase in different pairs. However, the precession is phased under the action of a transverse external variable field, thus resulting in the nonzero transverse magnetization component. On the other hand, the magnetization longitudinal component deviation from its equilibrium value, tends to zero with the relaxation rate  $\tau^{-1}$  exceeding that of the core magnetization components due to the good electron-lattice contact (for the validity of the offered mechanism, the inequality  $\tau^{-1} \gg \omega''$  is necessary). The joint action of variable field and electron relaxation results in the magnetization longitudinal component modulation with the variable field frequency, what can influence magnetization precession, i.e. FMR. The corresponding macroscopic DDE spin Hamiltonian is proposed. It is shown that for one ion pair it provides the level split similar to that in [12]. The magnetization motion under the action of this Hamiltonian is investigated. It is shown that this mechanism causes the resonance frequency shift and the FMR decay (band width). This shift and broadening mechanism is effective, when the variable field frequency  $\omega$  is of the order of  $\tau^{-1}$ . The obtained expressions at  $\omega\tau < 1$  agree qualitatively with the observed dependence of FMR band width on the variable field frequency [7, 8], as well as (at the certain assumptions) with the experimental data on temperature dependence of FMR spectrum [4-9]. This research is believed to promote the obtaining of the quantitative information on DE constants.

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## **Ширина лінії та зсув частоти ФМР, обумовлені подвійним обміном у допованих лантанових мanganітах**

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Запропоновано механізм зсуву резонансної частоти та розширення ФМР у феромагнітному провідному зразку (стрибкова провідність) допованого лантанового манганіту, обумовлений подвійним обміном (ПО) в умовах ФМР (динамічним подвійним обміном, ДПО). Показано, що цей механізм обумовлює зсув резонансної частоти та коефіцієнт згасання  $\omega''$  (ширину лінії) ФМР. Запропонований нами механізм зсуву та розширення, обумовлений ДПО, є ефективним, коли частота змінного поля  $\omega$  має порядок швидкості релаксації рухливих електронів  $\tau^{-1}$ . Для справедливості цього механізму необхідним є виконання нерівності  $\tau^{-1} >> \omega''$ . Одержані вирази при  $\omega$  якісно узгоджуються зі спостережуваною залежністю ширини лінії ФМР від частоти змінного поля, що його збуджує, а також, за певних умов, з експериментальними даними з температурної залежності спектру ФМР. Вважаємо, що це дослідження може сприяти отриманню кількісної інформації про константи ПО.