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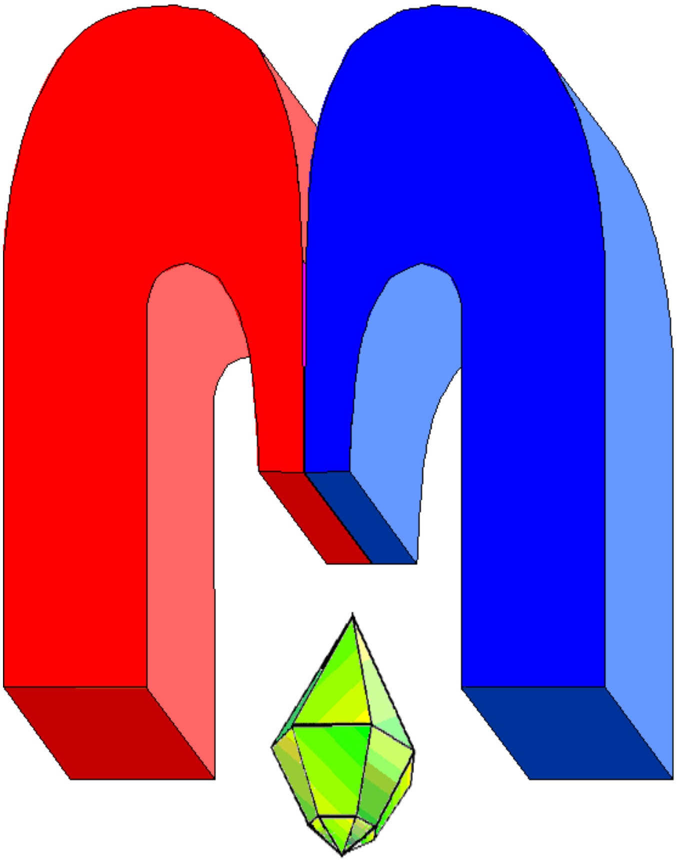

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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Angular and temperature dependencies of EPR linewidth and Gorter relaxation rates in concentrated paramagnets: Application to $La_{0.9}Sr_{0.1}MnO_3$ and $La_{0.875}Sr_{0.125}MnO_3$

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The angular and temperature dependencies of the electron paramagnetic resonance (EPR) linewidth in the weak constant field and of the relaxation rates measurable by the Gorter type experiments in zero constant magnetic field (Gorter RRs) are analytically investigated in magnetically concentrated paramagnets with the dominating exchange interaction. The consideration is restricted to the experiments, where the EPR linewidth is both anisotropic and linear over the temperature. It is suggested that under such conditions the EPR broadening is caused by the spin-lattice relaxation of the anisotropic interaction via the one-phonon mechanism. The analytical results are brought to the form suitable for the extraction of the Dzyaloshinsky-Moriya and the crystal field interaction constants from the experiment. It is shown that the EPR linewidth at the constant field direction along any crystal axis is equal to the half sum of the zero-field RRs for the two other crystal axes. The obtained results are successfully used for the interpretation of the EPR experimental results in the $La_{0.9}Sr_{0.1}MnO_3$ and $La_{0.875}Sr_{0.125}MnO_3$ single crystals in the Jahn-Teller strongly distorted phase in the definite temperature interval. The angular dependencies of the Gorter RRs and the EPR linewidth are presented graphically at the constant field continuous rotations in the three crystallographic planes in $La_{0.9}Sr_{0.1}MnO_3$.

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1. Introduction

The lightly doped manganites $La_{1-x}Sr_xMnO_3$ show in the paramagnetic phase intense exchange-narrowed electron paramagnetic resonance (EPR) signals with a large variation of the EPR linewidth as a function of the temperature. Moreover, the single crystals exhibit a pronounced anisotropy of the linewidth in the Jahn-Teller strongly distorted $O^{'}/I,PM$ phase and a less anisotropy in weakly distorted $O/I,PM$ phase (see the $x-T$ phase diagram in Refs. 1, 2).

The systematic weak field EPR investigations on $La_{1-x}Sr_xMnO_3$ eight single crystals with the Sr dopings $0 \le x \le 0.2$ were carried out in [2]. The EPR linewidth angular dependence for $x = 0.05$ and temperature dependencies for all samples were presented there. The angular dependence was interpreted as caused by the Dzyaloshinsky-Moriya (DM) antisymmetric interaction along the antiferromagnetically coupled b axis (we use here the axis notations according to the papers [3]). Further, in the detailed paper [3], which were the refinement of the paper [2] for $La_{0.95}Sr_{0.05}MnO_3$ sample, it was shown that one has to take into account also the DM interaction $\mathcal{H}_{DM}$ within the ferromagnetically coupled ac plane. Besides, another source of EPR broadening, namely, the interaction with the crystal field (CF) $\mathcal{H}_{CF}$ was taken in the Ref. 3 into account. It should be mentioned that the linewidth temperature dependence in [3] was sufficiently weak (of the form $\left(T_L-\theta_{CW}\right)/T_L$, where T_L is the lattice temperature, and θ_{CW} is the compound Curie-Weiss temperature). Therefore, the pure spin origin of the EPR broadening was suggested there, without taking the spin-lattice relaxation (SLR) into account. Since the strong exchange narrowing takes place in these concentrated paramagnets, the EPR linewidth was determined by the second moment conditioned by the anisotropic interactions $\mathcal{H}_{an} = \mathcal{H}_{DM} + \mathcal{H}_{CF}$ divided by the exchange frequency ω_{ex} [4]. Differing from the weak temperature dependence in [3], the linear temperature dependence $\Delta H^{EPR}(T_L) = \Delta H^{EPR}(\theta_{CW}) + b(T_L - \theta_{CW})$ of the anisotropic linewidth was observed in some samples in [2] within the definite temperature range. For instance, there is a temperature interval for samples with $x = 0.1$ and $x = 0.125$, where the EPR linewidth is both anisotropic and linear. The linear increase of the linewidth was ascribed in [2] to the one-phonon SLR, as it was suggested earlier in Refs. 5 and 6 for the same range of the doping. However, the one-phonon SLR of a Zeeman subsystem of magnetic ions should have T_L directly proportional dependence and should not be anisotropic. Besides, in [5] the EPR linewidth values for the three crystals ($x = 0.1$, $x = 0.2$, $x = 0.3$) were essentially the same at the frequencies $\nu = 9$ GHz and $\nu = 36$ GHz. If the one-phonon SLR of a Zeeman subsystem would be the reason of the EPR broadening, there would be ν^2 proportional dependence of its rate. It should be mentioned here that recently we suggested the weak field EPR broadening model conditioned by the one-phonon mechanism of the SLR most of all of the $\mathcal{H}_{an}$ freedom degrees rather than these of the Zeeman one for the concentrated paramagnets with the dominating exchange interaction [7]. It was demonstrated there that the corresponding SLR rate and, consequently, the EPR linewidth are proportional to the anisotropic second moments conditioned by $\mathcal{H}_{an}$ rather than proportional to ν^2. On the other hand, just the anisotropic second moments are able to cause the angular dependence of the EPR linewidth observed in [2, 3]. So, to our mind, the one-phonon SLR mechanism of the EPR broadening proposed in [7], in which the linewidth is anisotropic and does not depend on ν, should be appropriate both for the interpretation of the experiments of Refs. 5, 6 and experiments on the single crystals $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ in the definite temperature intervals in Ref. 2.

The same mechanism should be valid also for the relaxation rates (RR) measurable in the Gorter type experiments on $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ in the zero constant field ("zero-field" RR in terms of Refs. 8, 9). It was shown in [7] that the weak field EPR linewidth and the Gorter RRs are connected with one another by the simple linear relation. The particular case of this relation was revealed in Ref. 10 (see also the references therein) in the experiments on another magnetically concentrated crystal $\mathrm{CuCl_2 \cdot 2H_2O}$: the EPR half width on the half height at the constant field direction along any crystal axis is equal to the half sum of the zero-field RR measured by means of the Gorter method for the two other crystal axes (further, "Gorter RRs"). There was also an approach of Ref. 11 (see also [12] and [13]), where the different RRs were ascribed to the magnetization components along the different axes transverse to a constant field in the laboratory frame of reference in the case of the arbitrary anisotropic interaction.

Possessing the analytical temperature and angular dependencies of the EPR linewidth and the zero-field RRs, both including DM and CF parameters, it is possible to duplicate by the Gorter type experiments the obtaining of the interaction constants from EPR. We would like especially attract the attention of the experimentalists to such a possibility. In the given paper, we suppose to refine the results of our paper [7] and to apply them to the temperature and angular dependence interpretation of the EPR linewidth in [2] and to the description of these dependencies for the "zero-field" Gorter RRs in the $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ compounds within the definite temperature range.

2. The statement of the problem

We consider here the spin-system of the single crystals of the magnetically concentrated compounds $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ interacting with the lattice in the weak constant field (the condition of the EPR constant field weakness will be presented in Appendix 1).

The Hamiltonian of the problem is as follows:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{sph}, \qquad \mathcal{H}_0 = \mathcal{H}_{ex}^{is} + \mathcal{H}_{an} + \mathcal{H}_Z + \mathcal{H}_{ph}, \tag{1}$$

where the isotropic Heisenberg superexchange between Mn ions via the oxygen ions $\mathcal{H}_{ex}^{is} = -J\sum_{i<j} \mathbf{S}_i \cdot \mathbf{S}_j$ is the dominant interaction (J is the superexchange constant), and the EPR anisotropy is conditioned by the antisymmetric Dzyaloshinsky-Moriya (DM) interaction $\mathcal{H}_{DM}$ and the interaction with the crystal field (CF) $\mathcal{H}_{CF}$ (their sum is denoted by $\mathcal{H}_{an}$). So, we'd like to bring the explicit expressions of the DM interaction $\mathcal{H}_{DM}$ between i and j spins and the one-spin interaction $\mathcal{H}_{CF}$:

$$\mathcal{H}_{DM}(i,j) = \mathbf{d}_{ij}\left[\mathbf{S}_i \times \mathbf{S}_j\right], \qquad \mathcal{H}_{CF}(j) = \sum_{m,n} \tilde{D}_{mn}^{(j)} S_j^m S_j^n,$$

where $\mathbf{d}_{ij}$ is the Dzyaloshinsky-Moriya vector for the pair of i, j spins $\mathbf{S}_i$, $\mathbf{S}_j$; $\tilde{D}_{mn}^{(j)}$ are the tensor components of the CF interaction constant; $m, n = X, Y, Z$. The axes X, Y, Z of the laboratory frame of reference (LFR) are determined in the EPR experiments by the external magnetic field along the Z axis and the varying field along the X axis. $\mathcal{H}_{ph}$ is the phonon Hamiltonian; we assume that the phonons are at the lattice temperature. As in the case of EPR in the sited experimental papers, here the Zeeman interaction of Mn ions $\mathcal{H}_Z$ is supposed to be the smallest one. A model Hamiltonian is taken for the spin-lattice interaction

$$\mathcal{H}_{sph} = \sum_{n,\alpha',\beta'} e_{\alpha'\beta'} \mathcal{G}_{\alpha'\beta'}^{n} S^n = \sum_{\alpha',\beta'} e_{\alpha'\beta'} \left\{ \mathcal{G}_{\alpha'\beta'}^{X} S^X + \mathcal{G}_{\alpha'\beta'}^{Y} S^Y + \mathcal{G}_{\alpha'\beta'}^{Z} S^Z \right\}, \tag{2}$$

where $e_{\alpha'\beta'}$ is the component of the crystal deformation tensor, $\mathcal{G}_{\alpha'\beta'}^{n}$ is the component of the spin-phonon bond tensor.

We suppose that in the weak constant field it is the spin subsystem $\mathcal{H}_{an} + \mathcal{H}_Z \approx \mathcal{H}_{an}$, which exchange energy with the lattice. The interaction $\mathcal{H}_{ex}^{is} >> \mathcal{H}_{an}$ provides the fast modulation of the local magnetic fields, therefore $\mathcal{H}_{ex}^{is}$ cannot participate in the spin-lattice relaxation, as a separate reservoir (see Ref. 14). Since the stated problem is essentially many-particle, we shall proceed using the thermodynamical model. At that, we suppose that due to the strong spin-spin interactions the subsystem $\mathcal{H}_{an} + \mathcal{H}_Z \approx \mathcal{H}_{an}$ reaches the internal equilibrium faster than SLR can occur. Therefore, the Zubarev method [15] of the nonequilibrium statistical operator was applied by us leading to the following formula for the SLR rate (see Appendix 1 for details)

$$1/T_{sph} = \left(\frac{\lambda}{\Delta}\right)^2 \frac{k_B}{2\pi^2\hbar^2\rho_{cr}\bar{v}^5} \frac{\omega_m^3\tau_c}{3\omega_{an}^2} \sum_n \left(\mathcal{G}_{\alpha\beta}^n\right)^2 \left(M_2^n\right)_{an}(\infty)\left(T_L - \theta_{CW}\right), \tag{3}$$

where $n = X, Y, Z$; ω_m is the Debye frequency; ρ_{cr} is the crystal density; $\tau_c = \hbar / J$ is the exchange correlation time; $\bar{v}$ is the average sound velocity; $\hbar\omega_{an}$ is the mean-square quantum of $\mathcal{H}_{an}$; λ is the spin-orbit coupling parameter, and Δ is the distance of the excited orbit level from the ground one; $\left(M_2^n\right)_{an}(\infty)$ is the second moment calculated with the n-th component of the magnetization of the exchange non-coupled spins – in our case, effectively, at the infinitely large temperature. The expressions for the second moments $\left(M_2^X\right)_{an}(\infty)$, $\left(M_2^Y\right)_{an}(\infty)$ conditioned by the interactions $\mathcal{H}_{DM}$ and $\mathcal{H}_{CF}$, which enter the Eq. (3), with their angular dependencies in the crystallographic frame of reference (CFR) are brought in the Section 4 (some raw results were presented in Ref. 16).

3. Gorter RRs, EPR linewidth, and their connection

Let us consider the Gorter type experiment with the low-frequency field directed along the definite axis. Suppose that the Gorter low-frequency field is directed along the X or Y axis of our LFR. Then S^X or S^Y will be the only non-zero variable spin component in Eq. (2), correspondingly. Just S^X or S^Y will determine then the form of $\mathcal{H}_{sph}$: in the first case $\mathcal{H}_{sph}=\sum_{\alpha',\beta'} e_{\alpha',\beta'}\mathcal{G}^X_{\alpha'\beta'}S^X$, while in the second case $\mathcal{H}_{sph}=\sum_{\alpha',\beta'} e_{\alpha',\beta'}\mathcal{G}^Y_{\alpha'\beta'}S^Y$.

Therefore, the following expressions for the relaxation rates measurable in the corresponding experiments of the Gorter type (Gorter RRs)

$$1/T_{X,Y}=\left(\frac{\lambda}{\Delta}\right)^2\frac{\mathcal{G}^2}{2\pi^2\hbar^2\rho_{cr}}\frac{\omega_m^3\tau_c}{3\omega_{an}^2}\sum_{\alpha',\beta'}\left(\mathcal{G}^{X,Y}_{\alpha'\beta'}\right)^2\left(M_2^{X,Y}\right)_{an}(\infty)k_B\left(T_L-\theta_{CW}\right), \tag{4}$$

are following from Eq. (3). It was shown in Ref. 7 with the help of the macroscopic consideration that the T_X^{-1}, T_Y^{-1} relaxation rates are at the same time the relaxation rates of the X and Y components of the macroscopic magnetization.

On the other hand, at the EPR experiment the both S^X and S^Y are the non-zero variable spin components. Then $\mathcal{H}_{sph}=\sum_{\alpha',\beta'} e_{\alpha',\beta'}\left\{\mathcal{G}^X_{\alpha'\beta'}S^X+\mathcal{G}^X_{\alpha'\beta'}S^X\right\}$ and the EPR relaxational broadening $\Delta\omega_Z^{EPR}=(1/2)T_{sph}^{-1}$ [17], corresponding to the SLR rate T_{sph}^{-1}, is equal to

$$\Delta\omega_Z^{EPR}=\left(\frac{\lambda}{\Delta}\right)^2\frac{\mathcal{G}^2}{2\pi^2\hbar^2\rho_{cr}\bar{v}^5}\frac{\omega_m^3\tau_c}{3\omega_{an}^2}\frac{1}{2}\sum_{\alpha',\beta'}\left\{\left(\mathcal{G}^X_{\alpha'\beta'}\right)^2\left(M_2^X\right)_{an}(\infty)+\left(\mathcal{G}^Y_{\alpha'\beta'}\right)^2\left(M_2^Y\right)_{an}(\infty)\right\}k_B\left(T_L-\theta_{CW}\right). \tag{5}$$

Comparing (5) with (4), we get the known from the literature [11, 12] relation (further,"Gorter-EPR" relation) between the EPR linewidth and the Gorter RRs in the LFR

$$\Delta\omega_Z^{EPR}=(1/2)\left(T_X^{-1}+T_Y^{-1}\right), \tag{6}$$

which is confirmed also by the macroscopic consideration (see Ref. 7). It should be mentioned that the Gorter-EPR relation is valid at the arbitrary anisotropic interactions.

We would like to note that the EPR linewidth and Gorter RR angular dependencies as well as their inter-relation expressed by the Eq. (6) were obtained by us earlier for the KCuF$_3$ compound [13]; however, only the spin-spin interactions determine there the EPR broadening.

At the constant field arbitrary direction relatively to the CFR, there is the one-to-one correspondence between the angles, which $\mathbf{H}_0$ makes with the axes of CFR, and the angles, which the Gorter field $\mathbf{H}_1$ measuring T_X^{-1}, and the Gorter field $\mathbf{H}_1$ measuring T_Y^{-1}, make with the CFR axes (see Fig. 1 and Table 1).

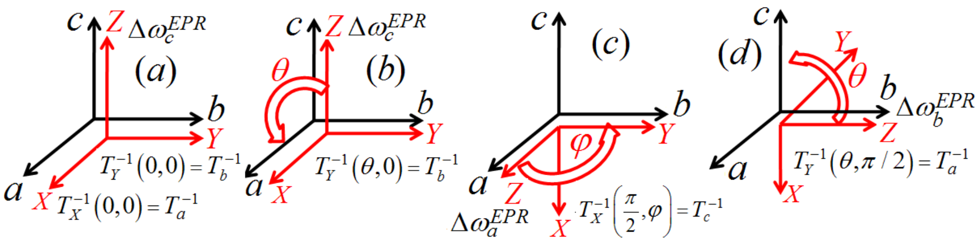

Figure 1. Connection between the rotation of the LFR relatively to the CFR and the obtaining T_a^{-1}, T_b^{-1}, T_c^{-1} from T_X^{-1}, T_Y^{-1} (see Appendix 2 for comments).

Table 1. Coordinates of the Gorter field vector $\mathbf{H}_1(\theta,\varphi)$ which is able to measure relaxation rates of X ($T_X^{-1}(\theta,\varphi)$) and Y ($T_Y^{-1}(\theta,\varphi)$) components of magnetization drawn in Fig. 1 (see Appendix 2 for comments).

Position of LFR respectively to CFR	Coordinates of the Gorter field $\mathbf{H}_1(\theta,\varphi)$ recording $T_X^{-1}(\theta,\varphi)$	Coordinates of the Gorter field $\mathbf{H}_1(\theta,\varphi)$ recording $T_Y^{-1}(\theta,\varphi)$
Fig. 1(a)	$\mathbf{H}_1(0,0)=(H_1,0,0)$ $\Rightarrow T_X^{-1}(0,0)=T_a^{-1}$	$\mathbf{H}_1(0,0)=(0,H_1,0)$ $\Rightarrow T_Y^{-1}(0,0)=T_b^{-1}$
Fig. 1(b)	$\mathbf{H}_1(\theta,0)=(H_1\cos\theta,0,-H_1\sin\theta)$ $\mathbf{H}_1(\pi/2,0)=(0,0,-H_1)$ $\Rightarrow T_X^{-1}(\pi/2,0)=T_c^{-1}$	$\mathbf{H}_1(\theta,0)=(0,H_1,0)$ $\Rightarrow T_Y^{-1}(\theta,0)=T_b^{-1}$
Fig. 1(c)	$\mathbf{H}_1(\pi/2,\varphi)=(0,0,-H_1)$ $\Rightarrow T_X^{-1}(\pi/2,\varphi)=T_c^{-1}$	$\mathbf{H}_1(\pi/2,\varphi)=(-H_1\sin\varphi,H_1\cos\varphi,0)$ $\mathbf{H}_1(\pi/2,\pi/2)=(-H_1,0,0)$ $\Rightarrow T_Y^{-1}(\pi/2,\pi/2)=T_a^{-1}$
Fig. 1(d)	$\mathbf{H}_1(\theta,\pi/2)=(0,H_1\cos\theta,-H_1\sin\theta)$ $\mathbf{H}_1(0,\pi/2)=(0,H_1,0)$ $\Rightarrow T_X^{-1}(0,\pi/2)=T_b^{-1}$	$\mathbf{H}_1(\theta,\pi/2)=(-H_1,0,0)$ $\Rightarrow T_Y^{-1}(\theta,\pi/2)=T_a^{-1}$

The relation (6) coincides with the corresponding relations of Refs. 8-10 in the particular cases when the LFR axes *X*, *Y*, *Z* coincide with the CFR axes *a*, *b*, *c* in the arbitrary order. It should be mentioned that a simple method of T_a^{-1}, T_b^{-1}, T_c^{-1} obtaining from EPR follows from (6): if the EPR linewidths are measured at the constant field directed in turn along the three crystallographic axes, then the solution of the three "Gorter-EPR" relations with respect to T_a^{-1}, T_b^{-1}, T_c^{-1} will give their values. The relaxation rates T_a^{-1}, T_b^{-1}, T_c^{-1}, which enter such relations, are nothing but the "zero-field" relaxation rates measured in the zero constant fields in the Gorter type experiments known from the literature [8-10]. We would like to present the method of their value calculation using the obtained by us T_X^{-1}, T_Y^{-1} expressions for the cases of the constant magnetic field rotations within the three crystallographic planes (the LFR rotations relatively to the CFR). These rotations can be depicted in the following form:

It is seen from Fig. 1 that in the case of the arbitrary anisotropic interactions, the analytical expressions for T_a^{-1}, T_b^{-1}, T_c^{-1} can be obtained from these for T_X^{-1}, T_Y^{-1} (Eqs. (4)) after the transition to CFR at the following rotation angles:

$$\begin{aligned} &T_b^{-1}=T_Y^{-1}(\theta,\varphi=0)\text{, at } \mathbf{H}_0 \text{ rotation in the plane } ac;\\ &T_c^{-1}=T_X^{-1}(\theta=\pi/2)\text{, at } \mathbf{H}_0 \text{ rotation in the plane } ab;\\ &T_a^{-1}=T_Y^{-1}(\theta,\varphi=\pi/2)\text{, at } \mathbf{H}_0 \text{ rotation in the plane } bc. \end{aligned} \tag{7}$$

Here, the following angular coordinates are used: the polar angle θ of the LFR *Z* axis ($\mathbf{H}_0$ direction) with respect to the crystallographic axis *c*, and the corresponding azimuthal angle φ which is counted from the *a* axis.

Having in mind the comparison with the experimental data, we take into account in (5) the Debye frequency value $\omega_m^3=6\pi^2 c\bar{v}^3$, where c is the magnetic ion concentration, the ratio $c/\rho_r=N_A/M$, where N_A is Avogadro number, M is the molar mass of the compound under consideration. Then the EPR linewidth in kOe can be written in the following form:

$$\Delta H_Z^{EPR}\left(kOe\right) = C_{sph}\frac{10\hbar}{g\mu_B}\sum_{\alpha'\beta'}\left\{\left(\mathcal{G}_{\alpha'\beta'}^{X}\right)^2\left(M_2^X\right)_{an}\left(\infty\right)+\left(\mathcal{G}_{\alpha'\beta'}^{Y}\right)^2\left(M_2^Y\right)_{an}\left(\infty\right)\right\}\tau_c\left(T_L-\theta_{CW}\right), \tag{8}$$

where $C_{sph}\equiv\left(\dfrac{\lambda}{\Delta}\right)^2\dfrac{1}{\hbar^2\overline{v}^2\omega_{an}^2}\dfrac{N_A}{M}k_B$.

4. Calculation of the second moments and the mean-square frequencies

Firstly, we would like to bring some comments to the calculation of the second moments in the compounds under consideration. The investigations of Deisenhofer et al [3] show that the contributions of the DM and CF interactions enter the full moments additively: $\left(M_2^n\right)_{an}=\left(M_2^n\right)_{DM}+\left(M_2^n\right)_{CF}$. Similar to Ref. 3, we shall present the second moments $\left(M_2^n\right)_{an}$ and the value ω_{an}^2, as the expressions averaged over the r magnetically nonequivalent ions of the magnetic structure $\left(M_2^n\right)_{an}=\left(1/r\right)\sum_{j=1}^{r}\left(M_{2an}^n\right)_j$, $\omega_{an}^2=\left(1/r\right)\sum_{j=1}^{r}\left(\omega_{an}^2\right)_j$. Here the values with the *j* subscript are calculated for the one *j*-s position ion. At that the DM interaction with the q nearest *i*-neighbors of the *j*-s ion contribute to $\left(M_{2DM}^n\right)_j$ and $\left(\omega_{DM}^2\right)_j$ in the form of the sums over $i = 1\ldots q$. For the crystals of the distorted perovskite structure, – the materials under our consideration, $r=4$; $q=6$. It should be noted that the summation over *i* is not required for the one-spin CF interaction.

The partial second moments of DM interaction in the LFR are equal to

$$M_{2DM}^X=\frac{4a}{3\hbar^2 r}\sum_{i,j}\left\{\left(d_{ij}^Y\right)^2+\left(d_{ij}^Z\right)^2\right\},\qquad M_{2DM}^Y=\frac{4a}{3\hbar^2 r}\sum_{i,j}\left\{\left(d_{ij}^X\right)^2+\left(d_{ij}^Z\right)^2\right\}, \tag{9}$$

where $a=S\left(S+1\right)$ and the summation way is as described above. The following should be mentioned in connection with the a value. As it is demonstrated in [2], the EPR signal in the doped lanthanum manganites is in accordance with the behavior expected within an intermediate octahedral ligand field. Such field splits the five d orbitals of Mn ions into a lower triplet t_{2g} and an upper doublet e_g. The dominant Hund's coupling causes the parallel spin alignment of the four d electrons of the Mn^{3+} spins and leads to the so-called high-spin state $S = 2$. Then, if the surrounding is ideally octahedral, the Zeeman effect is rather complicated and the observation of an EPR signal is rather unlikely. On the other hand, the Jahn-Teller distortion splits the orbital doublet into two singlets, where the ground state is a spin quintuplet with the usual Zeeman effect. The three d electrons of Mn^{4+} occupy the triplet t_{2g} with the parallel spin alignment $S = 3/2$. The ground state is an orbital singlet with a spin quadruplet. So, within the Jahn-Teller strongly distorted O$'$ phase the EPR signal is due to the both Mn^{3+} and Mn^{4+} spins. However, experiment shows the non-zero contribution of the Mn^{3+} ions even after the transition O$'$→O. This fact witnesses that the weak orthorhombic distortion in O phase is still enough strong to retain the EPR signal of Mn^{3+} ions. Therefore, we assume that for the these phases of the lanthanum manganites with the x doping the value $S(S+1)$ is the linear superposition of the both Mn^{3+} and Mn^{4+} spin contributions: $S\left(S+1\right)\big|_x=6\left(1-x\right)+3.75x$. At that, we do not take into account that according to [1] the effective moments are considerably higher than the spin-only values. The authors of [1] believe that this increase is due to the formation of the superparamagnetic clusters (the so-called "giant" spins).

For the obtaining the angular dependencies of the different second moments, the transition from the LFR to the CFR is needed.

Accomplishing the transition to the CFR, we take into account the known from Ref. 3 fact that all sums over the cross terms $d_{ij}^{\alpha}d_{ij}^{\beta}$ with $\alpha \neq \beta$, where $\alpha, \beta = a, b, c,$ vanish. Then the second moments (9) take the form:

$$M_{2DM}^{X} = \frac{4a}{3\hbar^2 r}\sum_{i,j}\left\{\left(d_{ij}^{a}\right)^2\left(\sin^2\varphi + \sin^2\theta\cos^2\varphi\right) + \left(d_{ij}^{b}\right)^2\left(\cos^2\varphi + \sin^2\theta\sin^2\varphi\right) + \left(d_{ij}^{c}\right)^2\cos^2\theta\right\}, \tag{10}$$

$$M_{2DM}^{Y} = \frac{4a}{3\hbar^2 r}\sum_{i,j}\left\{\left(d_{ij}^{a}\right)^2\cos^2\varphi + \left(d_{ij}^{b}\right)^2\sin^2\varphi + \left(d_{ij}^{c}\right)^2\right\}, \tag{11}$$

where $d_{ij}^{a}, d_{ij}^{b}, d_{ij}^{c}$ are the DM vector $\mathbf{d}_{ij}$ components along the CFR axes. At the calculation of the sums $\sum_{i=1}^{6}\left(d_{ij}^{a,b,c}\right)^2$ for the compounds under our consideration, we use the results of the Ref. 3. It is shown there that these sums are equal for all of *j* and can be presented as the functions of only the two parameters d_1, d_2.

The values of M_{2CF}^{n} in the LFR have the form

$$\begin{aligned} M_{2CF}^{X} = \frac{4a-3}{20\hbar^2 r}\sum_{j=1}^{r}\Big\{&\left(\tilde{D}_{XX}^{(j)} + \tilde{D}_{YY}^{(j)} - 2\tilde{D}_{ZZ}^{(j)}\right)^2 + \left(\tilde{D}_{XX}^{(j)} - \tilde{D}_{YY}^{(j)}\right)^2 \\ &-2\left(\tilde{D}_{XX}^{(j)} + \tilde{D}_{YY}^{(j)} - 2\tilde{D}_{ZZ}^{(j)}\right)\left(\tilde{D}_{XX}^{(j)} - \tilde{D}_{YY}^{(j)}\right) + 4\left(\tilde{D}_{XY}^{(j)2} + \tilde{D}_{XZ}^{(j)2} + 4\tilde{D}_{YZ}^{(j)2}\right)\Big\}, \end{aligned} \tag{12}$$

$$\begin{aligned} M_{2CF}^{Y} = \frac{4a-3}{20\hbar^2 r}\sum_{j=1}^{r}\Big\{&\left(\tilde{D}_{XX}^{(j)} + \tilde{D}_{YY}^{(j)} - 2\tilde{D}_{ZZ}^{(j)}\right)^2 + \left(\tilde{D}_{XX}^{(j)} - \tilde{D}_{YY}^{(j)}\right)^2 \\ &+2\left(\tilde{D}_{XX}^{(j)} + \tilde{D}_{YY}^{(j)} - 2\tilde{D}_{ZZ}^{(j)}\right)\left(\tilde{D}_{XX}^{(j)} - \tilde{D}_{YY}^{(j)}\right) + 4\left(\tilde{D}_{XY}^{(j)2} + \tilde{D}_{YZ}^{(j)2} + 4\tilde{D}_{XZ}^{(j)2}\right)\Big\}. \end{aligned} \tag{13}$$

Here $\tilde{D}_{mn}^{(j)}$ are the components of the CF interaction tensor in the LFR, which are linearly connected [3] with those in the CFR $D_{\alpha\beta}^{(j)}$. The values of M_{2CF}^{n} in the CFR will be written only for the $\mathbf{H}_0$ rotation in the *ab* plane of the crystal because of their inconvenience. They have the following form

$$M_{2CF}^{X}\left(ab\ \text{plane}\right) = \frac{4a-3}{5\hbar^2 r}\sum_{j=1}^{r}\left[\left(D_{aa}^{(j)} - D_{bb}^{(j)}\right)^2 + 4\left(D_{ab}^{(j)}\right)^2 + \left(D_{ac}^{(j)}\right)^2 + \left(D_{bc}^{(j)}\right)^2\right], \tag{14}$$

$$\begin{aligned} M_{2CF}^{Y}\left(ab\ \text{plane}\right) = \frac{4a-3}{10\hbar^2 r}\sum_{j=1}^{r}\Big\{&0.5\left(2D_{cc}^{(j)} - D_{aa}^{(j)} - D_{bb}^{(j)}\right)^2 + 0.5\left[\left(D_{aa}^{(j)} - D_{bb}^{(j)}\right)^2 + 4\left(D_{ab}^{(j)}\right)^2\right] \\ &+5\left(\left(D_{ac}^{(j)}\right)^2 + \left(D_{bc}^{(j)}\right)^2\right) + \left[\left(2D_{cc}^{(j)} - D_{aa}^{(j)} - D_{bb}^{(j)}\right)\left(D_{bb}^{(j)} - D_{aa}^{(j)}\right) + 3\left(\left(D_{ac}^{(j)}\right)^2 - \left(D_{bc}^{(j)}\right)^2\right)\right]\cos 2\varphi\Big\}. \end{aligned} \tag{15}$$

Note that it is possible to obtain the expressions for $M_{2CF}^{X,Y}$ in the *ac* and *bc* planes from (14, 15), if the replacements $X \leftrightarrow Y$ are made in the l.h.s. and replacements $\varphi \to \theta$; *a*, *b*, *c* → *c*, *a*, *b* and *a*, *b*, *c* → *c*, *b*, *a*, – in the r.h.s. of the above expressions, respectively.

The values of $\left(M_2^{+-}\right)_{an}$ in the CFR, which are necessary for the EPR linewidth angular dependence obtaining, can be easily found from the known $\left(M_2^{n}\right)_{an}$.

Also, the straightforward calculation gives the following expression for the DM mean quantum squared

$$\left(\hbar\omega_{DM}\right)^2 = \frac{4a}{3r}\sum_{i,j}\left\{\left(d_{ij}^{X}\right)^2 + \left(d_{ij}^{Y}\right)^2 + \left(d_{ij}^{Z}\right)^2\right\}. \tag{16}$$

The expression of $\left(\hbar\omega_{DM}\right)^2$ contains the square of the DM vector length; hence, it is angle independent in the CFR and has the following form there:

$$\left(\hbar\omega_{DM}\right)^2 = \frac{4a}{3r}\sum_{i,j}\left\{\left(d_{ij}^{a}\right)^2 + \left(d_{ij}^{b}\right)^2 + \left(d_{ij}^{c}\right)^2\right\}. \tag{17}$$

As to the CF interaction mean quantum squared, it is equal in the LFR to

$$\begin{aligned}\left(\hbar\omega_{CF}\right)^2 = \frac{1}{20r}\sum_{j=1}^{r}\Big\{&(4a-3)\left[\left(\tilde{D}_{XX}^{(j)} - \tilde{D}_{YY}^{(j)}\right)^2 + 4\tilde{D}_{XY}^{(j)2} + 2\left(\tilde{D}_{XZ}^{(j)2} + \tilde{D}_{YZ}^{(j)2}\right)\right] \\ &+(8a-1)\left(\tilde{D}_{XX}^{(j)} + \tilde{D}_{YY}^{(j)}\right)^2 + 4(2a+1)\left(\tilde{D}_{XX}^{(j)} + \tilde{D}_{YY}^{(j)}\right)\tilde{D}_{ZZ}^{(j)} + 4(3a-1)\tilde{D}_{ZZ}^{(j)2}\Big\}.\end{aligned} \tag{18}$$

After the transition to the CFR for the $\mathbf{H}_0$ rotation in the *ab* plane (18) takes the form

$$\begin{aligned}\left(\hbar\omega_{CF}\right)^2(ab\ \text{plane}) = \frac{1}{20r}\sum_{j=1}^{r}\Big\{&(4a-3)\Big[(3/4)\left[\left(D_{bb}^{(j)} - D_{aa}^{(j)}\right)^2 + 4D_{ab}^{(j)2}\right] \\ &+(1/4)\left[\left(D_{bb}^{(j)} - D_{aa}^{(j)}\right)^2 - 4D_{ab}^{(j)2}\right]\cos 4\varphi + 3\left(D_{bc}^{(j)2} + D_{ac}^{(j)2}\right) + \left(D_{bc}^{(j)2} - D_{ac}^{(j)2}\right)\cos 2\varphi\Big] \\ &+(8a-1)\left(D_{bb}^{(j)} + D_{aa}^{(j)}\right)^2 + 4(2a+1)\left(D_{bb}^{(j)} + D_{aa}^{(j)}\right)D_{cc}^{(j)} + 4(3a-1)D_{cc}^{(j)2}\Big\}.\end{aligned} \tag{19}$$

We have made sure with the help of the computer calculations that the terms of $\left(\hbar\omega_{CF}\right)^2$ containing angular dependence practically does not influence the value of $\left(\hbar\omega_{CF}\right)^2$ (their contribution is less than 3%), so, in (19) they can be neglected, and for the numerical calculations we have used Eq. (19) without them.

We would like to make a definite note regarding the spin-phonon bond tensor components $\mathcal{G}_{\alpha'\beta'}^{n}$. They can be considered as the crystal electric field potential arising from the crystal lattice distortions [18, p. 241; 19, p. 619]. Further the rough evaluation $\left|\mathcal{G}_{\alpha'\beta'}^{n}\right| \approx 3e^2\overline{r}^2/4\pi\varepsilon_0 R^3 \equiv \left|\mathcal{G}\right|$ from [18, p. 241] will be used for $\mathcal{G}_{\alpha'\beta'}^{n}$, where $\overline{r}^2$ is the mean distance of the d-electrons from the nucleus squared; $R = \left\{(8/3)\left(R_l^{-3} + R_m^{-3} + R_s^{-3}\right)\right\}^{-1/3}$ is the nearest ion O^{2-} average distance from the Mn ion; R_l, R_m, R_s are the long, the middle and the short octahedron axes; ε_0 is the electric constant. So, we suppose that the approximate equality of $\mathcal{G}_{\alpha'\beta'}^{X}$, $\mathcal{G}_{\alpha'\beta'}^{Y}$, $\mathcal{G}_{\alpha'\beta'}^{Z}$ parameters takes place, their mutual value being denoted further, as $\mathcal{G}$.

Knowing the above-calculated second moments, the Gorter RRs T_a^{-1}, T_b^{-1}, T_c^{-1} (4) can be written in the following explicit form:

$$\begin{aligned}T_a^{-1} &= C_{sph}\mathcal{G}^2\left(\tau_c\right)_{eff}\left(T_L - \theta_{CW}\right)\left\{\frac{4a-3}{5\hbar^2}\left[\left(D_{cc} - D_{bb}\right)^2 + 4D_{bc}^2 + D_{ac}^2 + D_{ab}^2\right] + \frac{4a}{3\hbar^2}\left[\left(d_{ij}^{b}\right)^2 + \left(d_{ij}^{c}\right)^2\right]\right\}, \\ T_b^{-1} &= C_{sph}\mathcal{G}^2\left(\tau_c\right)_{eff}\left(T_L - \theta_{CW}\right)\left\{\frac{4a-3}{5\hbar^2}\left[\left(D_{cc} - D_{aa}\right)^2 + 4D_{ac}^2 + D_{cb}^2 + D_{ab}^2\right] + \frac{4a}{3\hbar^2}\left[\left(d_{ij}^{a}\right)^2 + \left(d_{ij}^{c}\right)^2\right]\right\}, \\ T_c^{-1} &= C_{sph}\mathcal{G}^2\left(\tau_c\right)_{eff}\left(T_L - \theta_{CW}\right)\left\{\frac{4a-3}{5\hbar^2}\left[\left(D_{aa} - D_{bb}\right)^2 + 4D_{ab}^2 + D_{ac}^2 + D_{bc}^2\right] + \frac{4a}{3\hbar^2}\left[\left(d_{ij}^{a}\right)^2 + \left(d_{ij}^{b}\right)^2\right]\right\},\end{aligned} \tag{20}$$

where we write $\left(\tau_c\right)_{eff}$ instead of τ_c for the case when the narrowing of the EPR linewidth is caused not only by the isotropic exchange (see below).

5. Comparison with the experiment

Comparison of the analytical angular and temperature dependencies of the EPR linewidth obtained in the refined model of the EPR spin-phonon broadening with the experimental data for the compounds $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ will be carried out in the temperature interval $\theta_{CW} < T_L < T_{O'O}$. Here θ_{CW} is the Curie-Weiss temperature of the O' phase transition to the ferromagnetic phase; $T_{O'O}$ is the temperature of the structural transition from the O' phase to the O phase. The temperature interval reflects the requirement of the strong Jahn-Teller distortions leading to the DM interaction ($T_L < T_{O'O}$), and the condition $\theta_{CW} < T_L$ is necessary for the validity of the Eq. (A11) (see Appendix 1). As is shown below, the EPR linewidth angular and temperature dependencies following from this model agree with the experimental data. So, to our mind, the angular and temperature dependencies of the Gorter RRs presented are also adequate for the pointed compounds.

Firstly, we would like to make some general notes regarding the result (8). The Eq. (8) is anisotropic and linear over the temperature, what is in agreement with the experimental results of Ref. 2 for the samples $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ in the mentioned temperature interval. At that, Eq. (8) does not depend on the generator frequency. This fact is in accordance with the observation in Ref. 4: the measured EPR linewidths in the similar compounds were essentially the same at the generator frequencies $\nu = 9$ GHz and $\nu = 36$ GHz. Note, that if it would be the Zeeman subsystem which relaxed to the lattice, then its relaxation rate and, correspondingly, the EPR linewidth determined by it would be proportional to ν^2. On the other hand, the EPR linewidth (8) is proportional to $\left(M_2^{+-}\right)_{an}$. Hence, our suggestion that the EPR linewidth is determined in the given case rather by the relaxation to the lattice of the $\mathcal{H}_{an}$ interaction, and not of the Zeeman one, leads to the result agreeing with the experiment.

It is shown in Ref. 3, bearing on the experiment, that in a material, where the both DM and CF interactions present, their contributions to EPR linewidth are summed up additively. Our formulae confirm this fact. Really, since the joint heat capacity of these interactions is proportional to $\omega_{an}^2 = \omega_{DM}^2 + \omega_{CF}^2$ and the equality $\left(M_2^n\right)_{an} = \left(M_2^n\right)_{DM} + \left(M_2^n\right)_{CF}$ takes place, the relation $\Delta\omega_{an}^{EPR} = \Delta\omega_{DM}^{EPR} + \Delta\omega_{CF}^{EPR}$ is also seen from our Eq. (8). Obviously, the same is valid for the Gorter RRs, too. Besides, it is demonstrated by the authors of Ref. 3 that the angular dependence of $\Delta\omega_{an}^{EPR}$ is proportional to that of $\left(M_2^{+-}\right)_{an}$. Such regularity also follows from our Eq. (8) in the case, when approximate equality of $\mathcal{G}_{\alpha'\beta'}^X$, $\mathcal{G}_{\alpha'\beta'}^Y$, $\mathcal{G}_{\alpha'\beta'}^Z$ parameters takes place, since ω_{an}^2 is practically angle independent, and the identity $\left(M_2^X\right)_{an}(\infty) + \left(M_2^Y\right)_{an}(\infty) \equiv 2\left(M_2^{+-}\right)_{an}(\infty)$ is valid.

Let now begin with the comparison of the EPR linewidth temperature dependence (8) with the experimental dependencies in the samples with $x = 0.1,\ 0.125$ from the paper [2], where it is of the linear form: $\Delta H_Z^{EPR} = a + bT_L$. Following the Ref. 5, we rewrite this dependence in a somewhat different form:

$$\Delta H_Z^{EPR}\left(T_L\right) = \Delta H_Z^{EPR}\left(\theta_{CW}\right) + b\left(T_L - \theta_{CW}\right), \tag{21}$$

where $\Delta H_Z^{EPR}\left(\theta_{CW}\right) = a + b\theta_{CW}$. Note that the contribution to the EPR linewidth directly proportional to the temperature was observed in Ref. 17 in the A-site ordered compound $\mathrm{YBaMn_2O_6}$ with the mixed valence of the Mn ions ($\mathrm{Mn^{3+}}$ and $\mathrm{Mn^{4+}}$). This contribution was successfully interpreted there, as the result of the Korringa relaxation of the collective e_g electrons. For the quantitative agreement with the experiment it was assumed that this relaxation channel was the bottleneck of the relaxation to phonons of the united system of e_g electrons and Mn^{4+} spins. The presence of the highly mobile collective e_g electrons on the Mn^{4+} background was the necessary condition for such a relaxation. This condition was fulfilled in the A-site ordered compound $YBaMn_2O_6$. Differing from that situation, our consideration is restricted to the region $O^{'}/I,PM$ of the $x-T$ phase diagram of the Sr doped lanthanum manganites [1, 2]. Here the conductivity of the isolator type (see, for instance Ref. 3) takes place by means of the thermally activated hopping of the e_g electrons between Mn^{3+} and Mn^{4} sites [2]. Therefore, the Korringa relaxation cannot be realized here.

The experimental linewidth temperature dependence in the samples $La_{0.9}Sr_{0.1}MnO_3$ and $La_{0.875}Sr_{0.125}MnO_3$ of Fig. 3 from the Ref. 2 can be presented in kOe in the form:

$$\begin{aligned}\Delta H(T)\big|_{x=0.1} &= 0.2+0.0041(T_L-156),\\ \Delta H(T)\big|_{x=0.125} &= 0.09+0.0043(T_L-190).\end{aligned} \tag{22}$$

In (22) the values of the Curie-Weiss temperature of the O' phase $\theta_{CW}(x=0.1)=156\,\text{K}$ and $\theta_{CW}(x=0.125)=190\,\text{K}$ are substituted, which are known from Ref. 1. From the comparison of (22) with the Eq. (8) obtained by us it is seen at once that the Eq. (8) corresponds to the second term of the experimental temperature dependencies (21), (22), while the first term of (21), (22) is not present in the Eq. (8). The origin of $\Delta H_Z^{EPR}(\theta_{CW})\neq 0$ in Ref. 5 was connected with the spin-spin broadening. However, such contribution calculated by the Kubo-Tomita formula [4] under the conditions of the strong exchange narrowing, contains the second moment, which in the concentrated paramagnets should depend on the temperature according to (A11) (see Appendix 1). Following the papers [20], and [21], we suggest that the non-zero temperature independent contribution $\Delta H_Z^{EPR}(\theta_{CW})$ to the EPR linewidth in $La_{0.9}Sr_{0.1}MnO_3$ and $La_{0.875}Sr_{0.125}MnO_3$ single crystals is conditioned by the inhomogeneous broadening, which is known to be temperature independent. Naturally, the same inhomogeneous contribution should be added also to the Gorter RRs (4), so, the correlation (6) remains valid. The large inhomogeneous broadening $\Delta^{*}=\Delta H_Z^{EPR}(\theta_{CW})$ (~200 Oe) can follow from the noted in Ref. 2 twin structure of the investigated crystals.

Finally, the suggested by us EPR linewidth temperature dependence is of the form

$$\Delta H_Z^{EPR}(T_L)=\Delta^{*}+b(T_L-\theta_{CW}), \tag{23}$$

where the value b is equal to

$$b=C_{sph}\mathcal{G}^2\frac{10\hbar}{g\mu_B}(\tau_c)_{eff}\left(M_2^{+-}\right)_{an}(\infty).$$

Here the value $(\tau_c)_{eff}$ replaces the value $\tau_c=\hbar/J$, its appearance will be explained below.

We would like to compare the temperature dependence (23) with the experimental ones for the samples with $x=0.1;\,0.125$ from Ref. 2 also quantitatively. This comparison faces difficulties because the DM and CF interactions parameters, which are necessary for the calculation of $\left(M_2^{+-}\right)_{an}$ and ω_{an}^2 (see Section 4), are not known to us for the compounds with $x=0.1;\,0.125.$ However, the DM and CF interactions parameters were extracted from the experiment in Ref. 3 for $x=0.05.$ We see the way out in the fact that b contains the ratio of the values $\left(M_2^{+-}\right)_{an}$ and ω_{an}^2, each of which is some linear combination of these parameters squared. Therefore we suppose that

$\left.\frac{\left(M_2^{+-}\right)_{an}}{\omega_{an}^2}\right|_{x=0.1}, \left.\frac{\left(M_2^{+-}\right)_{an}}{\omega_{an}^2}\right|_{x=0.125} \approx \left.\frac{\left(M_2^{+-}\right)_{an}}{\omega_{an}^2}\right|_{x=0.05}$. Not allowing for the latter assumption, b can be written in the more general form:

$$b=\frac{10\hbar}{g\mu_B}\left(\frac{\lambda}{\Delta}\right)^2 k_B \frac{\mathcal{G}^2\left(\tau_c\right)_{eff}}{\hbar^2\overline{v}^2}\frac{N_A}{M}\left.\frac{\left(M_2^{+-}\right)_{an}(\infty)}{\omega_{an}^2}\right|_{x=0.05}\left\{\left.\frac{\left(M_2^{+-}\right)_{an}(\infty)}{\omega_{an}^2}\right|_{x=0.1,x=0.125} \Big/ \left.\frac{\left(M_2^{+-}\right)_{an}(\infty)}{\omega_{an}^2}\right|_{x=0.05}\right\}. \quad (24)$$

Though it seems to us that the ratio in the parenthesis is of the unity order, further, to be on the right side, one should take into account that the plots of Fig. 2 are really built in the units

$$\left\{\left.\frac{\left(M_2^{+-}\right)_{an}(\infty)}{\omega_{an}^2}\right|_{x=0.1,x=0.125} \Big/ \left.\frac{\left(M_2^{+-}\right)_{an}(\infty)}{\omega_{an}^2}\right|_{x=0.05}\right\}.$$

Note that in (24) we have replaced the correlation time $\tau_c=\hbar/J$ conditioned by the usual isotropic exchange for $\left(\tau_c\right)_{eff}$, which is determined by us by the fit of the Eq. (23) to the experimental temperature dependence plots from Ref. 2. The point is that the correlation time calculated with the help of the expression $\tau_c=2S\left(S+1\right)\hbar/k_B\theta_{CW}$ leads to the value of the constant b, which is an order of magnitude larger than the experimental one. Since as a result of the fit it turned out that $(\tau_c)_{eff}\approx\tau_c/30$ for the both dopings $x=0.1$ and $x=0.125$, the angular dependence plots of the Fig. 2 are built with this value of $\left(\tau_c\right)_{eff}$. The values lT_X^{-1}, lT_Y^{-1}, where $l=10\hbar/g\mu_B$, are plotted in kOe for the relaxation rates of the X and Y components of Mn ion magnetization, correspondingly, there.

We explain the effective decrease of the correlation time in the samples under consideration analogously to the Ref. 5. Namely, we suppose that this decrease reflects the additional motional narrowing of the EPR linewidth due to e_g electron hopping between Mn^{3+} and Mn^{4+} sites. The motional narrowing was absent in $La_{0.95}Sr_{0.05}MnO_3$, but, to our mind, plays role for the increased e_g electron hopping intensity in the samples under consideration with the increased doping.

We have obtained the following curves of the angular dependencies of the transverse magnet-

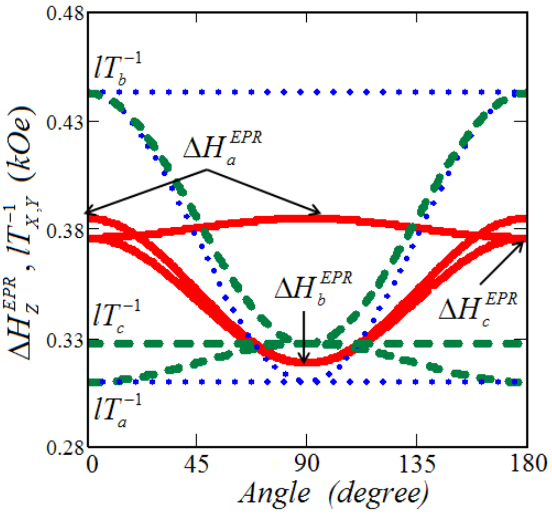

Figure 2. Dashed green and dotted blue curves describethe angular dependencies of the relaxation rates lT_X^{-1}, lT_Y^{-1} in kOe of the X and Y components of Mn ion magnetization, correspondingly, at the constant magnetic field rotation relatively to the crystallographic axes a, b, c of the sample within its three crystallographic planes, $l=10\hbar/g\mu_B$. Solid red curve describes the angular dependence of the EPR linewidth ΔH^{EPR} in kOe. The plot is built for $La_{0.9}Sr_{0.1}MnO_3$ at $T_L=200$ K.

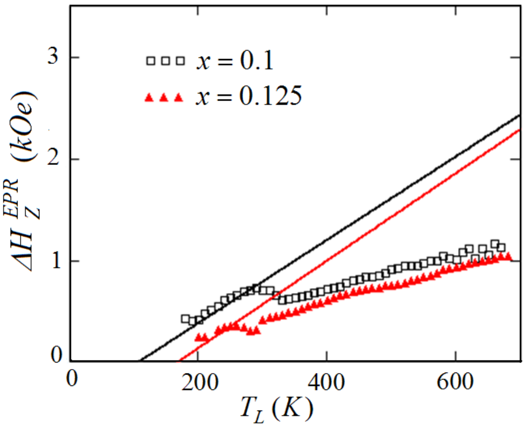

Figure 3. Temperature dependencies of the EPR linewidth in $La_{0.9}Sr_{0.1}MnO_3$ and $La_{0.875}Sr_{0.125}MnO_3$. The squares and triangles depict the experimental data from Ref. 2. The fit according to Eq. (23) of the given paper is indicated by a solid line.

ization relaxation rates and that of the EPR linewidth in kOe in $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ compound (see Fig. 2; here, as in Ref. 3, the *b* axis is perpendicular to the ferromagnetically ordered *ac* planes).

The comparison of the obtained temperature dependencies with the experimental ones are depicted on the Fig. 3. It is seen that there is a good agreement with the experiment in the definite temperature interval.

Though the roughness of the used model spin-phonon Hamiltonian and the corresponding value evaluations, together with the presence of the fitting parameter, do not enable us to make unambiguous quantitative conclusions, the suggested model agreement with the experimental temperature dependence seems to be reasonable.

6. Conclusion

Summarizing, the following results are obtained for the magnetically concentrated paramagnets under conditions of the dominating exchange interaction and for the weak constant magnetic field EPR broadening caused by the one-phonon spin-lattice relaxation. The main result is that the analytical angular and temperature dependencies of the EPR linewidth obtained agree with the experimental data of the Ref. 2 for the $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ single crystals. So, one may hope that the angular and temperature dependencies of the zero-field Gorter relaxation rates obtained are also adequate. Further, as it was shown in our recent paper [7], in such materials the EPR half width on the half height at a weak constant field $\mathbf{H}_0$ along *Z* axis can be presented as the half sum of the relaxation rates T_X^{-1}, T_Y^{-1} of the magnetization components along *X* and *Y* axes measurable in the Gorter type experiments. These relaxation rates can be measured in the experiments of the Gorter type in the zero constant field with the definite (pointed out in the Table 1) directions of the low-frequency field relatively to the CFR. The angular dependencies of the half sum of T_X^{-1}, T_Y^{-1} coincide with the experimentally observed EPR linewidth angular dependencies, thus demonstrating the continuous “Gorter-EPR relation”. This continuous “Gorter-EPR relation” coincides with the discrete ones, which were confirmed experimentally, in the particular cases of the axes *X*, *Y*, *Z* (LFR) coincidence with the axes *a*, *b*, *c* (CFR). Our investigation combines the continuous and the discrete approaches, differing both from the discrete consideration of the Refs. 8-10, restricted to the CFR, and the continuous consideration of Refs. 11, 12 restricted to the LFR.

The refined model of Ref. 7 is used for the interpretation of the experimental results of Ref. 2 for the single crystals $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ and $\mathrm{La_{0.875}Sr_{0.125}MnO_3}$ in the temperature interval, where the EPR linewidth is simultaneously anisotropic and linearly dependent on the temperature. At that, the anisotropic interaction consisting of the DM and CF interactions effectively relaxes to the lattice. It is supposed that all the relaxation rates contain the temperature independent contributions from the inhomogeneous broadening. At such a supposition, the temperature dependence of the EPR linewidth qualitatively agrees with the experiments of Ref. 2. The angular dependence of the EPR linewidth qualitatively agreeing with the experiment of Ref. 2 is presented analytically and for the single crystal $\mathrm{La_{0.9}Sr_{0.1}MnO_3}$ graphically. For the quantitative agreement of the obtained results with the experiments of Ref. 2, it was necessary to suppose that the correlation time of the interaction narrowing the EPR line is approximately an order of magnitude smaller than the one given only by the isotropic exchange. We suppose that the correlation time decreasing is caused by the additional motional effect due to the e_g-electron hopping between the $\mathrm{Mn^{3+}}$ and $\mathrm{Mn^{4+}}$ sites.

The obtained microscopic analytical expressions for the zero-field relaxation rates measurable in Gorter type experiments and for the EPR linewidth in a weak constant field connect the experimentally obtainable macroscopic values of the relaxation rates with the spin structure. Since all these quantities are expressed via the constants of the interactions causing magnetic anisotropy, duplication of the getting of information on these constants in the new technological materials becomes possible.

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Appendix 1. Calculation of the one-phonon spin-lattice relaxation rate

At the calculation of the spin-lattice relaxation rate, we follow Eq. (15, 29) in Ref. 22:

$$1/T_{sph} = (1/2)\left\langle \mathcal{H}_{an}^2 \right\rangle^{-1} \int\limits_{-\infty}^{\infty} dt \left\langle K_{an} K_{an}(t) \right\rangle, \tag{A1}$$

where

$$K_{an} = (i\hbar)^{-1}\left[\mathcal{H}_{an}, \mathcal{H}_{sph}\right]; \quad \left\langle \mathcal{H}_{an}^2 \right\rangle \equiv (\hbar\omega_{an})^2 \left\langle \left(S^z\right)^2 \right\rangle; \tag{A2}$$

the triangular brackets mean the equilibrium thermodynamic averaging; $\hbar\omega_{an}$ is the mean-square quantum of $\mathcal{H}_{an}$. Further,

$$K_{an}(t) = \exp\left((i/\hbar)\left(\mathcal{H}_{an} + \mathcal{H}_{ph}\right)t\right) K_{an} \exp\left(-(i/\hbar)\left(\mathcal{H}_{an} + \mathcal{H}_{ph}\right)t\right). \tag{A3}$$

It should be mentioned that the presence of the strong exchange interaction $\mathcal{H}_{ex}^{is}$ will be taken into account later so, as it was done in Ref. 7.

The following inequalities are fulfilled in the experiments under consideration

$$H_0 << J / g\mu_B\mu_0, \sqrt{H_{an}^2}, \tag{A4}$$

$$k_B T_L > \gamma^{\max} \equiv \left\{2S(S+1)n_0 J_{nn}^2\right\}^{1/2}, \tag{A5}$$

where $\mathbf{H}_0$ is the external constant magnetic field in EPR; $\sqrt{H_{an}^2} = (g\mu_B\mu_0)^{-1}\sqrt{\mathrm{Sp}\left(\mathcal{H}_{an}^2\right)/\mathrm{Sp}\left(S^{z2}\right)}$ is the mean-square local field of $\mathcal{H}_{an}$; T_L is the lattice temperature; n_0 is the number of the nearest spin neighbors and $J_{nn} = J$ is the exchange integral for them; for the doping x one has $S(S+1)\big|_x = 6(1-x) + 3.75x$; k_B and μ_0 are the Boltzmann and magnetic constants, μ_B is the Bohr magneton.

The weak field condition (A4) provides the strong exchange narrowing of the EPR line and guarantees that it is the anisotropic interaction $\mathcal{H}_{an}$, which relaxes to the lattice during EPR rather than the Zeeman interaction. The inequality (A5), according to Huber [23], provides the efficiency of the one-phonon spin-lattice mechanism of the EPR broadening and the EPR linewidth linear temperature dependence even at sufficiently high temperatures.

Basing on the arguments brought in the Introduction, we suggest further that the relaxational EPR broadening takes place in the single crystal lanthanum manganites with the Sr doping $x = 0.1$ and $x = 0.125$ in the definite temperature interval. Spin transitions under the influence of the lattice vibrations, shortening the lifetime of the magnetic ions on the spin levels, serve as the broadening cause of these levels, and consequently, of the EPR line in the compounds under consideration, the corresponding linewidth being [14] $\Delta\omega_Z^{EPR} = (1/2)T_{sph}^{-1}$. We suggest also that the SLR is of the one-phonon type in $La_{0.9}Sr_{0.1}MnO_3$ and $La_{0.875}Sr_{0.125}MnO_3$ and is realized by the crystal electric field modulation at the vibrations of the O^{2-} ions nearest to the magnetic ions (O^{2-} ions are situated in the vertices of the octahedrons surrounding Mn ions). Since our quantitative calculations will have the evaluation character, we proceed from the model spin-lattice Hamiltonian (2). There $n = X, Y, Z$ are the LFR axes; α', β' are the O^{2-} ion coordinates in the local Cartesian frame of reference with the origin in the magnetic ion and with the axes along the O^{2-} octahedron axes; $\mathcal{G}^{X,Y,Z}_{\alpha'\beta'}$ are the components of the spin-phonon bond tensors;

$$e_{\alpha',\beta'} = \frac{i}{2}\left(\frac{\hbar}{4\pi M}\right)^{1/2} \sum_{\mathbf{k}_p} \frac{\sqrt{2\pi \mathbf{k}_p}}{\sqrt{\omega_{\mathbf{k}_p}}} \left(a^{+}_{\mathbf{k}_p} \exp\left(i\mathbf{k}_p\mathbf{R}\right) - a_{\mathbf{k}_p} \exp\left(-i\mathbf{k}_p\mathbf{R}\right)\right)\left(\lambda_{\alpha'} f_{\beta'} + \lambda_{\beta'} f_{\alpha'}\right) \tag{A6}$$

is the crystal deformation tensor component; M_{cr} is the crystal mass; $\omega_{\mathbf{k}_p}$ and $v_{\mathbf{k}_p}$ are correspondingly the frequency and the velocity of a *p*-s branch phonon possessing wave vector $\mathbf{k}_p$; $a^{+}_{\mathbf{k}_p}$ and $a_{\mathbf{k}_p}$ are the birth and annihilation operators of such a phonon; λ_α are the directing cosines of the phonon polarization vector; **f** is the wave vector ort.

Further, the fact is used that in the case of the one-phonon SLR Hamiltonian (2) K_{an} can be presented in the form

$$K_{an} = -\sum_{\alpha',\beta'} e_{\alpha'\beta'} \sum_{n=x,y,z} \mathcal{G}^{n}_{\alpha'\beta'} K^{n}_{an}, \quad \text{where } K^{n}_{an} = \left(1/i\hbar\right)\left[S^{n}, \mathcal{H}_{an}\right]. \tag{A7}$$

It follows from (A3) and (A7) that

$$\left\langle K_{an} K_{an}(t)\right\rangle = \sum_{\alpha',\beta'} \left(\mathcal{G}^{n}_{\alpha'\beta'}\right)^{2} \left\langle e_{\alpha'\beta'} e_{\alpha'\beta'}(t)\right\rangle \left\langle K^{n}_{an} \tilde{K}^{n}_{an}(t)\right\rangle, \tag{A8}$$

where $\tilde{K}^{n}_{an}(t) = \exp\left(\frac{i}{\hbar}\mathcal{H}_{an} t\right) \tilde{K}^{n}_{an} \exp\left(-\frac{i}{\hbar}\mathcal{H}_{an} t\right)$.

It should be mentioned that the SLR rate in Ref. 7 was obtained in the high temperature approximation over all the energies, including the phonon energy. If one does not do the latter approximation and passes to the continual limit in the phonon spectrum, then the spin-phonon relaxation rate in the long wave approximation in frames of the Debye model has the following form

$$\begin{gathered}\frac{1}{T_{sph}} = \frac{1}{16\pi^{3}\rho_{cr}\hbar\omega^{2}_{an}} \sum_{j} v^{-5}_{kp} \sum_{n} \left\langle \left(K^{n}_{an}\right)^{2}\right\rangle / \left\langle \left(S^{n}\right)^{2}\right\rangle \left[\sum_{\alpha',\beta'} \left(\mathcal{G}^{n}_{\alpha'\beta'}\right)^{2} \int d\Omega \frac{1}{2}\left(\lambda_{\alpha'} f_{\beta'} + \lambda_{\beta'} f_{\alpha'}\right)\right]^{2} \\ \cdot\int_{-\infty}^{\infty} d\omega K^{n}_{an}(\omega) \int_{0}^{\omega_m} d\omega' \omega'^{3} \coth\frac{\hbar\omega'}{2k_B T_L} \frac{\omega_{ex}}{\left(\omega' - \omega\right)^{2} + \omega^{2}_{ex}}.\end{gathered} \tag{A9}$$

Here ω_m is the Debye frequency; ρ_{cr} is the crystal density. Since $K^{n}_{an}(\omega)$ is the Fourier-transform of the normalized correlation function $\left\langle K^{n}_{an} \tilde{K}^{n}_{an}(t)\right\rangle / \left\langle \left(K^{n}_{an}\right)^{2}\right\rangle$, one has $\int_{-\infty}^{\infty} d\omega\, K^{n}_{an}(\omega) = 1$. Note that the influence of the strong exchange interaction with the exchange frequency $\omega_{ex} = J/\hbar$, which is suggested to be larger than all other frequencies in the problem under consideration is already taken into account in the equation (A9) in the same way, as it was done in Ref. 7.

Further, in the considered model the following equation is valid:

$$\left\langle \left(K^{n}_{an}\right)^{2}\right\rangle / \left\langle \left(S^{n}\right)^{2}\right\rangle_{T_L} = -\left\langle \left[S^{n}, \mathcal{H}_{an}\right]^{2}\right\rangle \hbar^{-2} \left\langle \left(S^{n}\right)^{2}\right\rangle^{-1}_{T_L} = \left(M^{n}_{2}\right)_{an}\left(T_L\right). \tag{A10}$$

Following the result of Ref. 24 and Ref. 25 at the calculation of $\left\langle \left(S^n\right)^2 \right\rangle_{T_L} = (1/2)\left\langle S^+S^- \right\rangle_{T_L}$ in the second moment denominator we use the relation $\left\langle S^+S^- \right\rangle_{T_L} / \left\langle S^+S^- \right\rangle_{\infty} = \chi(T_L)/\chi_0(T_L)$, where $\chi_0(T_L)$ is the single ion static susceptibility, and $\chi(T_L)$ corresponds to the behavior of the exchange coupled Mn system. Similar to Ref. 24, we assume that the single ion susceptibility $\chi_0(T_L)$ obeys the Curie law $\chi_0(T_L) = C/T_L$, while $\chi(T_L)$ can be approximated by the Curie-Weiss behavior $\chi(T_L) = C/(T_L - \theta_{CW})$, where C is the Curie constant and θ_{CW} is the Curie-Weiss temperature. Then the following relation is valid at $T_L > \theta_{CW}$:

$$\left(M_2^n\right)_{an}(T_L) = \frac{T_L - \theta_{CW}}{T_L}\left(M_2^n\right)_{an}(\infty). \tag{A11}$$

Note that the Eq. (A11) was used in Ref. 7 at $T_L \to \infty$.

Further, evaluating the term $\left[\sum_{\alpha',\beta'}\left(\mathcal{G}^n_{\alpha'\beta'}\right)^2 \int d\Omega \frac{1}{2}\left(\lambda_{\alpha'} f_{\beta'} + \lambda_{\beta'} f_{\alpha'}\right)\right]^2$ in (A9), we make use of the corresponding value $4\pi\Phi_{mm'}$, which is brought in [18], p. 231. However, we shall use it in the simplified form – assuming that the longitudinal and the transverse velocities of the acoustic waves are equal to one another $v_l = v_t = \bar{v}$. We also suppose that the approximate equality of $\mathcal{G}^X_{\alpha'\beta'}$, $\mathcal{G}^Y_{\alpha'\beta'}$, $\mathcal{G}^Z_{\alpha'\beta'}$ parameters takes place, their mutual value being denoted further, as $\mathcal{G}$. At that, $\left[\sum_{\alpha',\beta'}\left(\mathcal{G}^n_{\alpha'\beta'}\right)^2 \int d\Omega \frac{1}{2}\left(\lambda_{\alpha'} f_{\beta'} + \lambda_{\beta'} f_{\alpha'}\right)\right]^2 = 4\pi\mathcal{G}^2/3$. Note also that if one passes to the variable $x = \omega'/\omega_{ex}$, which was introduced in Ref. 23, in the integral over $d\omega'$, then this integral takes the form

$$\omega_{ex}^3 \int_0^{\omega_m/\omega_{ex}} dx \cdot x^3 \coth\frac{xJ}{2k_BT_L}\frac{1}{\left(x - \omega/\omega_{ex}\right)^2 + 1}. \tag{A12}$$

The expression (A12) is analogous to the function

$$f\left(k_BT_L/\gamma^{\max}\right) = \int_0^\infty dx \cdot x^3 \coth\frac{x\gamma^{\max}}{2k_BT_L}\exp\left(-\frac{x^2}{2}\right), \tag{A13}$$

from Ref. 23, in spite of the fact that the different model of the exchange interaction influence on the spin-lattice relaxation rate is used there (note that from (A5) $\gamma^{\max} \propto J$). Because of the difficulties to take the integral (A12) exactly analytically, we approximate (A12) as $\frac{2k_BT_L\omega_{ex}^2}{\hbar}\int_0^{\omega_m/\omega_{ex}} dx \cdot x^2 = \frac{2k_BT_L}{\hbar\omega_{ex}}\frac{\omega_m^3}{3}$. At that, we use the fact that $\omega_{ex} \equiv \tau_c^{-1}$ is the largest frequency in the problem under consideration and that according to Ref. 23, at $k_BT_L > \gamma^{\max}$ the expression (A13) is directly proportional to the temperature. Further, the coefficient $(\lambda/\Delta)^2$ should be added to T_{sph}^{-1}, where λ is the spin-orbit coupling parameter, and Δ is the distance of the excited orbit level from the main one [14]. This is the consequence of the well-known fact that for the case of iron group ions (the case under our consideration) the crystal field reduces an orbital magnetic moment λ/Δ times [14].

Taking into account all the above mentioned and using the definition of the exchange correlation time $\tau_c = \hbar/J$, we obtain the expression (3) for the one-phonon spin-lattice relaxation rate in the main text.

Appendix 2. Comments to Fig. 1

Fig. 1(a). On this figure one of the three versions of the classical "axial" implementation of the Eq. (6) (see also [10]) is presented: EPR is observed at $\mathbf{H}_0 \parallel Z \parallel c$, while the Gorter RR measurements are observed at the low-frequency field along the a ($X \parallel a$, $T_X^{-1} = T_a^{-1}$) and b ($Y \parallel b$, $T_Y^{-1} = T_b^{-1}$) axes. The other two "axial" implementations are as follows: $\mathbf{H}_0 \parallel Z \parallel a$, $X \parallel -c$ ($T_X^{-1} = T_c^{-1}$), $Y \parallel b$ ($T_Y^{-1} = T_b^{-1}$) and $\mathbf{H}_0 \parallel Z \parallel b$, $X \parallel -c$ ($T_X^{-1} = T_c^{-1}$), $Y \parallel -a$ ($T_Y^{-1} = T_a^{-1}$). The values of T_a^{-1}, T_b^{-1} and T_c^{-1} can be calculated with the help of the expressions (20), and the EPR linewidth – by the formula $\Delta\omega_i^{EPR} = (1/2)\left(T_j^{-1} + T_k^{-1}\right)$, where $i, j, k = a, b, c$.

Fig. 1(b). The $\mathbf{H}_0$ rotation from the c axis to the a axis (*ac* plane) is shown by the arrow. At that, the X axis of the LFR turns also in the *ac* plane (from a to $-c$), and it is possible to measure $T_X^{-1}(\theta, \varphi = 0)$ for any value of θ (at $\varphi = 0$) from the value T_a^{-1} up to the value T_c^{-1} (rotation axis Y stays immovably, and $T_Y^{-1}(\theta, \varphi = 0) = T_b^{-1}$).

Fig. 1(c). $\mathbf{H}_0$ turns from the a axis to the b axis (*ab* plane). Here the Y axis turns in the *ab* plane (from b to $-a$), and it is possible to measure $T_Y^{-1}(\theta = \pi/2, \varphi)$ between the values T_b^{-1} and T_a^{-1} (rotation axis X stays immovably along the $-c$ axis, and $T_X^{-1}(\theta = \pi/2, \varphi) = T_c^{-1}$).

Fig. 1(d). Finally, the $\mathbf{H}_0$ returns to the c axis, X axis turns from $-c$ to b, and in the *bc* plane the value of $T_X^{-1}(\theta, \varphi = \pi/2)$ can be measured within the limits T_c^{-1}, T_b^{-1}), while $T_y^{-1}(\theta, \varphi = \pi/2) = T_a^{-1}$ (Y axis does not turn).

It should be noted that the relaxation rates for any θ and φ values can be calculated by the formulae (4) with the taking into account the expressions (10, 11, 14, 15) for $\left(M_2^{X,Y}\right)_{an}$ and the additional expressions obtained from (14, 15) with the help of the replacements specified nearby.

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