Magnetic structure and antiferromagnetic resonance spectrum in manganites: the effect of orbital structure

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The paper presents an investigation concerning temperature dependence of magnetic structure and antiferromagnetic resonance frequencies. Taking into account the crystal and orbital structures of pure lanthanum manganite, it is shown that the orbital structure plays the crucial role in forming of the magnetic structure. The $H-T$ phase diagrams for the magnetic structure are drawn.

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1. Introduction

The interest in pure rare-earth orthorhombic manganites both doped by alkaline-earth manganite compounds is quickened, because a lot of unusual effects studied in experiments and theoretically predicted. We turn our attention to the interrelation between crystal, charge, orbital and magnetic subsystems of pure rare-earth manganite.

The present work is aimed to theoretical study of magnetic resonance spectra of manganite. The external magnetic field dependence of the magnetic structure and AFMR frequencies shown the presence of the spin-flop transition [1]. Because of a strong effect of the orbital structure upon the magnetic structure, the field behavior of the magnetic subsystem is more complicated than the one of an usual antiferromagnet. The temperature dependences of the magnetic structure and AFMR frequencies show critical behavior [1–3]. The experimentally determined Neel temperature is 138 K [1–3]. At this temperature, the magnetization and the AFMR frequencies go to zero.

For a description of the external magnetic field and temperature effects upon the magnetic structure and AFMR frequencies, we use the recently developed model [4] which includes orbitally-dependent interactions: superexchange interaction and single-ion anisotropy. The temperature dependence is described in the mean-field approximation. The field dependence and a value of the spin-flop transition field are in an excellent agreement with experiment [1]. Taking into account the Neel temperature overestimation due to approximation, temperature dependences are also in a good agreement with experiment [1–3] and explain the splitting of AFMR frequencies without an external magnetic field and the disappearance of this splitting near the Neel temperature [3].

Using the described calculations, the phase diagrams $H-T$ in the main field directions (a, b, c)are drawn, which shows antiferromagnetic, ferromagnetic and paramagnetic regions of the pure manganite's magnetic structure.

2. Crystal and orbital structures of $LaMnO_3$

The crystal structure of manganite (Fig. 1) is the distorted perovskite structure (*Pnma* space group). The distortions are of two kinds: the rotational distortions of oxygen octahedra and the lanthanum ion shifts like in orthoferrite crystals and the Jahn–Teller distortions of oxygen octahedra of *e*-type [5]. The strong electron-lattice interaction causes the presence of *e*-type distortions due to the degenerate

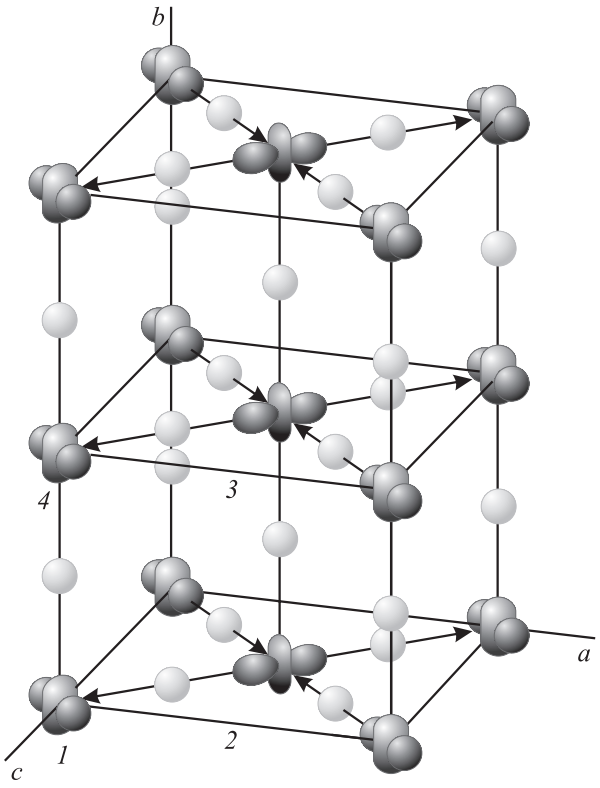

Figure 1. Crystal and orbital structures. Lanthanum ions are omitted. Manganese ions are drawn as *e*-orbitals. Arrows show *e*-type distortions of oxygen octahedral. Numbers enumerate orbital and magnetic sublattices.

trivalent manganese-ion ground state and helps the orbital structures to be established.

The wave function of the ground state of each manganese ion in the distorted perovskite crystal is [4]

$$\psi_n = \sin(\phi_n/2)\varphi_{n\theta} + \cos(\phi_n/2)\varphi_{n\varepsilon}, \qquad (1)$$

where $\varphi_{n\theta}$ and $\varphi_{n\varepsilon}$ are the eigenfunctions of the degenerated ^5E term. The values of ϕ_n could be derived from experimental data using parameters of lattice distortions [5] within the framework of strong electron-lattice coupling. The orbital structure is described by a correlation of orbital angles

$$\phi_1 = \phi_2 = -\phi_3 = -\phi_4 = \phi, \qquad (2)$$

where ϕ characterize the orbital state of the Mn^{3+} sublattice according to Eq. (2).

3. Orbital-dependent magnetic interactions

In order to predict the magnetic properties, we use following spin-Hamiltonian

$$\widehat{H} = \sum_{n>m} J_{n,m}(\mathbf{S}_n \cdot \mathbf{S}_m) + \widehat{H}_n^{\text{anis}} + g\mu_B \sum_n (\mathbf{H} \cdot \mathbf{S}_n), \qquad (3)$$

where

$$J_b = J_0 \frac{\cos^2 \varphi_b}{r_b^{10}} [1 + 2\alpha \cos\phi + \beta \cos^2 \phi], \qquad (4)$$

$$J_{ac} = J_0 \frac{\cos^2 \varphi_{ac}}{r_{ac}^{10}} [1 - \alpha \cos\phi + \beta(\cos^2 \phi - 3/4)], \qquad (5)$$

the parameters of dependencies are [4] $J_0 = 1.69 \cdot 10^4\,\text{K} \cdot \text{Å}^{10}$, $\alpha = 1.0$, $\beta = 4.5$, and the experimental structural parameters are $\varphi_b = 155.1°$, $\varphi_{ac} = 154.8°$ (Mn–O–Mn bond angles), $r_b = 1.97$ Å, $r_{ac} = 2.05$ Å (mean Mn–O distances), $\phi = 107.8°$ (angle of orbital mixing, without ambient pressure [5]). The single-ion anisotropy of manganese ions in octahedral oxygen environment is given by

$$\widehat{H}_n^{\text{anis}} = D_n S_{nz\,l}^2 + E_n(S_{nx\,l}^2 - S_{ny\,l}^2),$$
$$D_n = 3P\cos\phi, \quad E_n = \sqrt{3}P\sin\phi, \qquad (6)$$

where $P = -1.15$ K [4], n enumerates manganese ions, index l denotes local axes of octahedra nearly corresponding to cubic axes. In order to take into account the rotational distortions, we have to transform the Hamiltonian (6) into the $Pnma$ axes.

The temperature dependence cound be taken into account within the framework of the molecular field approximation

$$\langle S_i \rangle = S_i B_{S_i}\left(\frac{g\mu_B \langle H_i^{\text{eff}} \rangle S_i}{kT}\right), \qquad (7)$$

where $B_{S_i}(x)$ is the Brillouin function and

$$\mathbf{H}_i^{\text{eff}} = \sum_j J_{ij}\mathbf{S}_j + \partial H_i^{\text{anis}}/\partial \mathbf{S}_i + g\mu_B \mathbf{H} \qquad (8)$$

is the effective magnetic field, acting on i-th sublattice.

4. Results and discussion

These parameters describe a magnetic structure of A-type. The single-ion anisotropy (or the Dzyaloshinsky–Moria interaction, as in paper [3]) estabilishes the magnetic structure of $A_xF_yG_z$ type (in $Pnma$ notation) [4]. The molecular field theory gives overestimated critical temperatures for magnetic transitions. For $LaMnO_3$, we obtained $T_N = 216$ K while experimentally $T_N \approx 140$ K is observed [2,3]. But qualitatively our results are in a good agreement with experiments (see Figs 2, 3). Temperature dependence of total magnetization is represented in Fig. 3.

Little discrepancy for the AFMR frequencies temperature dependence is reasoned by an inaccuracy of measurements near the edge of the receptiveness band at low temperatures. As for Fig. 3, divergence between theoretical and experimental [6] data (which almost coincide even quantitatively) is also caused by a twinning of the $LaMnO_3$ crystal and magnetic domains (which could be treated by field cooling of the sample [7]). Magnetic domains are the reason of presence negative magnetization at low temperature and low field in plane (see Fig. 3).

In order to find out and explain different magnetic structures of pure lanthanum manganite, magnetic phase diagrams field-temperature ($H-T$) in different directions

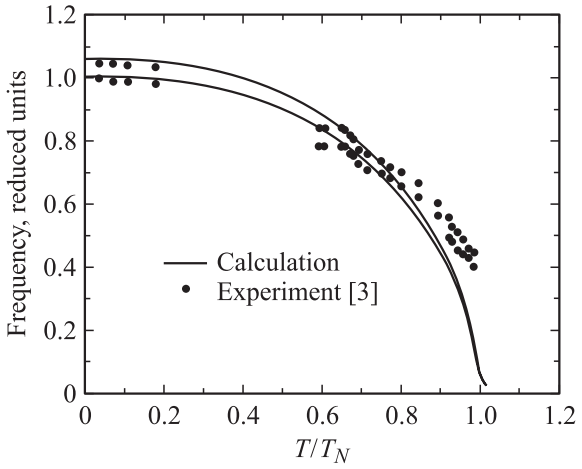

Figure 2. Temperature dependence of AFMR frequencies.

of magnetic field were obtained (Fig. 4). As it is known, pure lanthanum manganite is a canted antiferromagnet. Its magnetic structure, therefore, is characterized by a mixture of basic magnetic structures (A, C, G — AFM structures and F — FM structure). With increasing temperature, the antiferromagnetic A-component decreases, but the ferromagnetic F-component remains almost constant. That is why the model predicts an A-antiferromagnetic to F-ferromagnetic transition (AFM A to FM). While temperature of this transition is close to that of the ferromagnetic–paramagnetic transition (FM to PM), one can observe a direct switch from the AFM A to PM phase (for fields up to 45 kOe for H perpendicular to the easy axis and up to 190 kOe for H parallel to the easy axis), and the gap between AFM A–FM and FM–PM (FM gap on the diagrams) transition temperatures widens with raising of the magnetic field. This effect is stimulated by strong damping of the A-component by magnetic field.

Due to a spin-flop transition in the external magnetic field directed along the easy axis x near $H = 190$ kOe [1,4], there is slight step on the corresponding diagram. The point is that there are two structures, which are close energetically

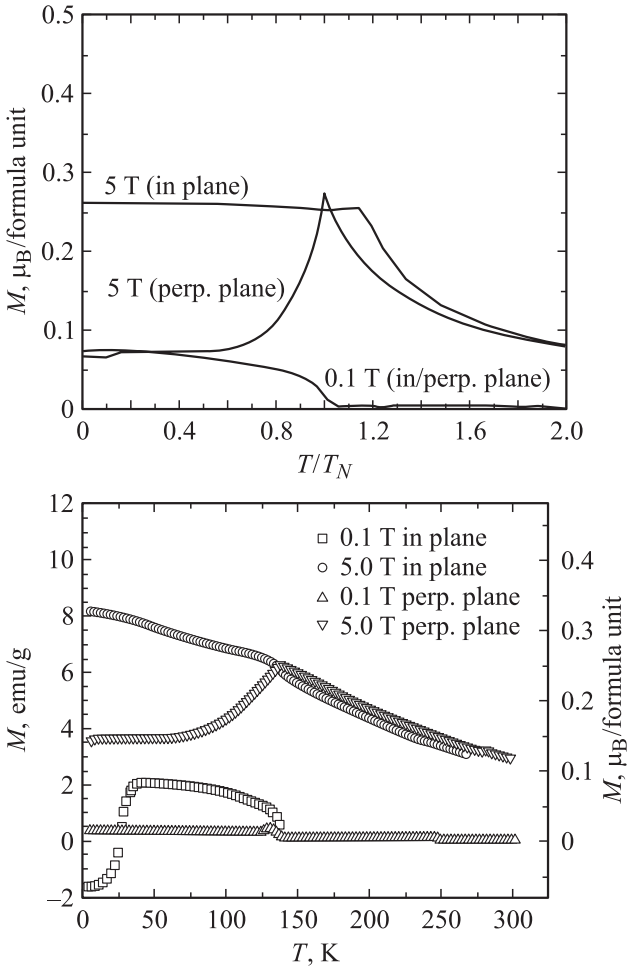

Figure 3. Temperature dependence of total magnetization. Experimental data are taken from paper [6].

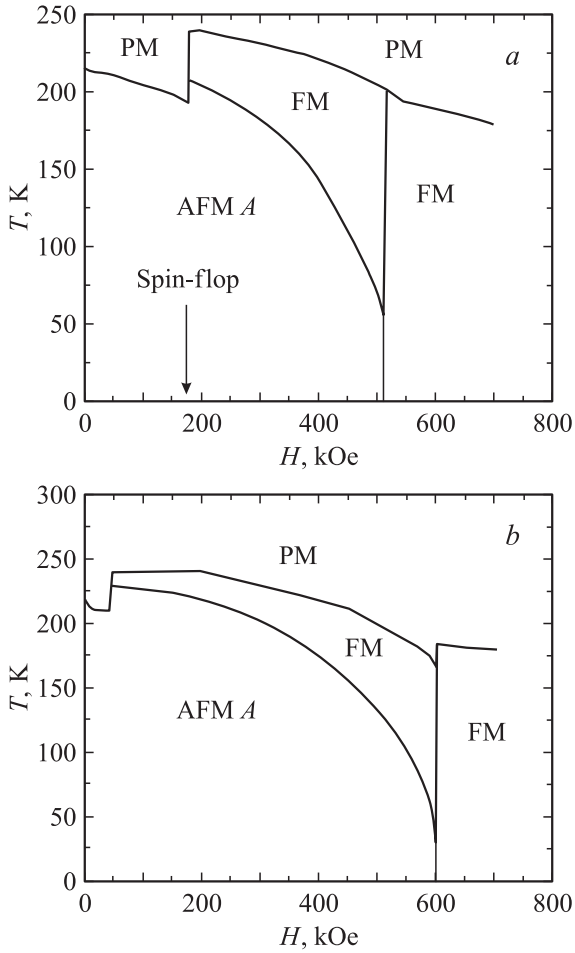

Figure 4. Phase diagram $H-T$. a — $H \parallel x$; b — $H \parallel y$. „AFM A“ is the A-type antiferromagnet.

($A_xF_yG_z$ and $G_xC_yF_z$), and the energy minimum switch takes place in the spin-flop field, when the magnetic structure is changed rapidly.

5. Conclusion

The theoretical investigation of temperature dependences of magnetic properties of $LaMnO_3$ was performed within the framework of spin-Hamiltonian including three parts: isotropic exchange, single-ion anisotropy and Zeeman interaction. Mean field approximation was used for obtaining macroscopic parameters.

Experimental temperature dependencies for total magnetizations in low (0.1 T) and high (5 T) magnetic fields were reproduced. Phase diagrams $H-T$ are obtained and qualitatively discussed. A rich phase diagram is originated by non trivial character of magnetic interactions caused by the orbital structure in the compound. Variation of different magnetic parameters was performed to find out their influence on the behavior of the system in the magnetic field. It was found that single-ion anisotropy plays a crucial role in the reaction of the magnetic system on the external

magnetic field along the easy axis of magnetization (where a spin-flop transition is observed). Thus, this factor does not seem to be of such importance as antiferromagnetic exchange is, if we investigate the perpendicular direction (hard direction) of the external magnetic field. Thereby, it is evident that $LaMnO_3$ possesses a non-trivial strongly anisotropic magnetic structure caused by its orbital and crystal structures.

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