Antiferromagnetic-Resonance Spectrum in Charge-Ordered $R_{0.5}Ca_{0.5}MnO_3$ Manganites ($R = La, Pr, Tb$): The Effect of Orbital and Charge Structures

L. É. Gonchar* and A. E. Nikiforov
Ural State University, Yekaterinburg, 620083 Russia
*e-mail: lyudmila.gonchar@usu.ru
Received August 21, 2002

Abstract—A theoretical study is made into the effect of the crystal, orbital, and charge structures on the magnetic structure and spin-wave spectra and on the antiferromagnetic resonance (AFMR) for $R_{0.5}Ca_{0.5}MnO_3$ crystals of monoclinic structure. The model assumes fixed crystal, charge, and orbital structures and enables one to determine the orbitally dependent exchange interaction and single-ion anisotropy for $R = La, Pr, Tb$. A 16-sublattice weakly noncollinear magnetic CE-structure without a ferromagnetic component is obtained. The behavior of magnetic structure in an external magnetic field is simulated, and the values of fields of spin-flop-transition for different Rs are obtained. The law of spin-wave dispersion and the field dependence of the antiferromagnetic-resonance spectrum are calculated.

1. INTRODUCTION

Attention given to manganites at present is not confined to the well-known effect of colossal magnetoresistance and is directed at a large number of unusual properties. The dielectric phases of manganites likewise are becoming the subject of thorough theoretical and experimental studies. Certain difficulties involved in studying these compounds are associated with the description of all crystal subsystems, namely, the crystal lattice and the charge, orbital and spin subsystems, with regard for their correlation. This paper deals with the magnetic properties of dielectric $R_{0.5}Ca_{0.5}MnO_3$ manganites ($R = La, Pr, Tb$) of monoclinic structure. The objective of our study is to demonstrate the effect of the crystal, charge, and orbital structures on magnetic ordering and spectra of magnons.

The investigation of the correlation between the orbital and magnetic structures of manganites was begun quite some time ago. Starting with the studies by Goodenough [1] and Wollan and Koehler [2], the presence of such a correlation was determined qualitatively. The quantitative characteristics of interaction of all four subsystems are still being discussed at present. Several models exist, which are used for the description of manganites. The effect of lattice distortions on the magnetic structure is recognized today by most researchers [3]; however, the inclusion of Jahn–Teller lattice distortions, which play the main part in the formation of unusual properties of manganites, is done differently by different authors. In the charge-ordered phases of manganites, the ordering of localized charge carriers formed due to nonisovalent doping is assumed.

Two models are largely used at present for explaining the correlation between the crystal, orbital, and magnetic structures in manganites. The first of these models, referred to as the Kugel’–Khomskii model [4], implies the formation of the orbital structure owing to the orbitally dependent exchange interaction eventually causing a crystal lattice distortion. Various modifications of this model are used in numerous studies [5–9]. This model is used quite frequently for prediction and investigation of orbital excitations (orbitons) [6, 8–10]. However, the temperature of decomposition of the orbitally ordered phase is much higher than the Neél temperature [11, 12], which proves the assumption that the orbital structure is formed as a result of stronger-than-exchange interactions. The second model, referred to as the Kanamori model [13], presumes the presence of the cooperative Jahn–Teller effect. This model likewise presumes the presence of the orbitally dependent exchange interaction; however, the main part is played by the electron-vibrational interaction. The development of the Kanamori model for manganites is described in [14–17].

The description of superexchange interaction in many-electron systems is a fairly complicated problem. In some compounds, the signs of exchange parameters may be defined by the Goodenough–Kanamori rules [1]. These rules, however, do not define the values of exchange integrals. The exchange parameters were originally calculated [5] for pure manganite; however, they turned out to be overestimated compared to experiment [18, 19]. The general approach to the description of pure and doped manganites is based on the Hubbard model with regard for Hund interaction, Coulomb interaction (in-site and intersite), and Jahn–Teller inter-
action in different models. As a rule, the double-exchange contribution is added to the Hamiltonian [6, 9, 15, 16, 20–22] in order to describe additional charge carriers emerging when the rare-earth sublattice is doped with alkali-earth ions. This approach is characterized by a simplified model of the orbital structure \((d_{3z^2-r^2}, d_{x^2-y^2})\)-orbitals, with the \(x\), \(y\), and \(z\) axes assumed to be dependent on the position of manganese ion. In some studies, the \(t_2_g\) shell is disregarded. The magnetic structure (including pure and charge-ordered manganites) is described in the double-exchange model [5, 7, 16]. Some models [21, 23] are incapable of describing the experimentally observed [2, 24–26] magnetic structure of charge-ordered manganites of the \(CE\) type. Therefore, it is not always possible to describe the experimental data and more exact calculations need to be performed. The large number of experimental studies into the crystal, charge, orbital, and magnetic structures makes it possible to apply the semiempirical approach to the description of the mechanism of superexchange interaction in dielectric manganites, which was developed in previous papers [17, 27].

The magnetic structure of half-doped charge-ordered manganites was studied in sufficient detail for different compositions of the rare-earth/alkali-earth sublattice [2, 24–26, 28–30]. These compounds are characterized by the presence or absence of charge ordering, and this feature is of main importance from the standpoint of formation of some or other magnetic structure. In the charge-ordered phase, the \(Mn^{3+}\) and \(Mn^{4+}\) ions are present in equal amounts and are ordered in space, the orbital structure is characteristic of the sublattice of \(Mn^{3+}\) ions, the magnetic structure corresponds to the \(CE\) type, and the easy magnetic axis is directed mainly along the \(c\) axis (in \(Pnma\) notation) [2, 24–26, 28, 29, 31]. A fairly accurate symmetry classification was performed for pure manganites [32]; however, there exists the problem of performing a classification of the \(CE\)-structure proper of the charge-ordered phase and experimental determination of noncollinear components [33]. The charge, orbital, and \(CE\)-magnetic structures may be destroyed by temperature or magnetic field [3].

We used the model of the orbital and magnetic structures [17, 27, 34], based on fixed crystal and charge structures, strong electron-vibrational interaction, and orbitally dependent superexchange. The transition to the charge-ordered phase leads to localization of holes on manganese ions with the formation of an ordered structure. In compounds of half-doped orthomanganites, the \(Mn^{3+}/Mn^{4+}\) ions alternate in the \(ac\) plane and do not alternate along the \(b\) axis (\(C\) type of charge structure). In so doing, the \(Mn^{3+}\) ions are Jahn–Teller ions and the \(Mn^{4+}\) ions do not exhibit a degeneracy of the ground state in an ideal octahedral coordination. In view of these features, it is possible to use our model [17] to quantitatively estimate the parameters of superexchange interaction and Neél temperature in dielectric manganites. In this study, we calculated the characteristic features of a multisublattice magnetic structure of monoclinic charge-ordered manganites, the spin-wave spectra, and the field dependence of antiferromagnetic resonance under conditions of the external magnetic field directed along the easy magnetic axis. The suggested model is semiempirical; however, it enables one to obtain the magnetic structure and spectra of magnons proceeding from the crystal structure. Our results may be used for interpreting experimental data in case an exact magnetic structure is required.

2. CRYSTAL AND ORBITAL STRUCTURES

At low temperatures, the \(R_{0.5}Ca_{0.5}MnO_3\) crystals being treated exhibit a distorted perovskite structure of symmetry \(P2_1/m\) and charge ordering [2, 24–26]. This group is a \(Pnma\) subgroup which describes the space symmetry of a crystal of pure manganite. Because of the presence of manganese ions in different charge states, which are arranged in a staggered order in the basal plane (see Figs. 1 and 2), additional distortions arise in the crystal. The sublattice of manganese ions is divided into three positions (see Table 1). The ions in positions \(a\) and \(b\) exhibit \(e\)-type distortions of oxygen coordination, and the ions in position \(d\) do not. The orthorhombic distortion of an ideal perovskite lattice [35] varies in accordance with low-symmetry distortion due to the charge nonequivalence of the ions.

1. An \(R\)-type distortion is a rotation about the pseudocubic [110]$_p$ axis with cell doubling on all three axes \((k_{11})_p\tau_0(C,C,0)\) in the notation of Kovalev [36] or \((\varphi\varphi\varphi)\) in the notation of [37]). Three values of the angle \(\varphi\), namely, \(\varphi_1\), \(\varphi_2\), and \(\varphi_3\), are distinguished in the monoclinic structure, which correspond to this distortion in sign and are different in magnitude.

2. An \(M\)-type distortion is a rotation of oxygen octahedrons about the [001]$_p$ axis with cell doubling on two axes \((k_{11})_p\tau_1(00C_2)\) in the notation of [36] or \((000\psi)\) in the notation of [37]). Three values of the angle \(\psi\), namely, \(\psi_1\), \(\psi_2\), and \(\psi_3\), are distinguished in the monoclinic structure, which correspond to this distortion in sign and are different in magnitude.

3. A \(Q\)-type distortion describes the deformation of \(e\)-type oxygen octahedrons (see Fig. 2) with cell doubling on two axes \((k_{11})_p\tau_1(00C_2)\) in the notation of [36] and with the choice of ray \([1/2 1/2 0]_p\). In the monoclinic structure, this distortion is characteristic only of the coordination of trivalent manganese ions. The doubling occurs on a single [110]$_p$ axis.

4. The coordination of tetravalent manganese is characterized by an \(a\)-type distortion (uniform compression) because of the smaller radius of the \(Mn^{4+}\) ion compared to \(Mn^{3+}\).