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2. Non-collinear magnetism in distorted perovskite compounds

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5. Abstract

Using results of the band structure calculations in the local-spin-density approximation we demonstrate how the crystal distortions affect the magnetic structure of orthorhombically distorted perovskites leading to a non-collinear spin arrangement. Our results suggest that the non-collinearity of the spin magnetic moments, being generally small in LaMO\textsubscript{3} series with M=Cr-Fe, is large in SrRuO\textsubscript{3}.

6. \textit{keywords}: Heisenberg exchange, Dzyaloshinskii-Moriya exchange, perovskite transition metal oxides, band structure calculations

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Perovskite transition-metal oxides are known to be the most striking example of materials where magnetic, transport and structural properties are strongly coupled. The reciprocal influence of the spin and lattice degrees of freedom can be due to the spin-orbit interaction (SOI) or the orbital ordering effects [1]. Both mechanisms can be responsible for the non-collinear magnetic arrangement through the antisymmetric Dzyaloshinskii-Moriya exchange interaction [2] or through strong dependence of the interatomic exchange on the orbital ordering [1] resulting in pronounced non-Heisenberg behavior [3]. We consider the first possibility for several orthorhombically distorted perovskites with $D_{16}^h$ structure where the non-collinear magnetism is allowed by symmetry [4]. However, the magnitude of the effect itself depends on the relative strength of several magnetic interactions.

We use the LMTO Green’s function technique in the real space and perturbative approach both for small deviations of the spin magnetization near the scalar-relativistic equilibrium and SOI. Then, the total energy change can be expressed analytically as

$$\delta E = E_H + E_{DM} + E_{MAE}.$$  

The first term $E_H \simeq -1/2 \sum_{ij} J_{ij} e_i e_j$ (where $e_i$ is the direction of the spin magnetization at the site $i$) describes the isotropic Heisenberg exchange interaction and appears in the second order with respect to nonuniform rotations of spins [5]. The magnetocrystalline anisotropy energy (MAE) firstly appears in the second order with respect to the SOI. The antisymmetric coupling $E_{DM} \simeq \sum_{i>j} d_{ij} [e_i \times e_j]$ corresponds to the mixed type perturbation with respect to spin rotations and SOI [6].

$J_{ij}$ and $d_{ij}$ parameters for La$M$O$_3$ series with $M=$Cr-Fe and SrRuO$_3$ are shown in Tables 1 and 2. General tendencies of the nearest neighbor interactions $J_{ij}$ can be understood by using simple tight-binding arguments given in [7]. (i) $J_{ij} < 0$ at the half of the band filling: $t_{2g}$-type exchange interaction in LaCrO$_3$ (formal atomic configuration of Cr is $t_{2g}^3 e_g^0$), both $t_{2g}$ and $e_g$ interactions in LaFeO$_3$ ($t_{2g}^3 e_g^2$). (ii) $J_{ij} > 0$ at the beginning and at the end of the band filling: $e_g$ exchange interaction dominating in LaMnO$_3$ ($t_{2g}^3 e_g^1$) [6]. (iii) $J_{ij} \sim 0$ around $1/3$ and $2/3$ of the band filling: SrRuO$_3$ case ($t_{2g}^4 e_g^0$). On the other hand, $d_{ij}$ parameters being proportional to the SOI are generally larger in SrRuO$_3$. The structural factor defined by rotations of the $MO_6$ octahedra relative to each other is of the same
order of magnitude for all compounds considered here. Thus, for LaMO₃ compounds the Heisenberg exchange interaction is clearly the strongest, whereas for SrRuO₃ it is considerably smaller and comparable with the antisymmetric exchange.

It is particularly interesting in LaMnO₃ that the interlayer exchange coupling $J_{1B} = \sum_{j \in B} J_{1j}$ with $j$ running over Mn atoms in the plane $B$ in Fig.1 crucially depends on the Jahn-Teller distortion (JTD) and varies between ferro- (FM) and antiferromagnetic (AFM) [6]. For the pure compound, the AFM interlayer coupling stabilized by JTD is large enough ($J_{1B} \approx -1.4$ mRy [6]) to overcome the antisymmetric interactions and the magnetic spin structure is nearly collinear. Weak FM canting due to the $d_{12}$ interaction and estimated as $\sin^{-1} |\alpha_c/J_{1B}|$ is less than $2^\circ$ (see [6] for details). However, JTD is suppressed rapidly with the hole doping, directly affecting the interlayer coupling constant $J_{1B}$. At certain concentration of the holes one expects $|J_{1B}| \sim |\alpha_c|$ and large non-collinearity. This tendency qualitatively explains the appearance of the spin-canted AFM phase accompanying the AFM-to-FM transition in the low-doped manganites [8].

In conclusion, the non-collinear spin structure, imposed by general symmetry rules, is suppressed in LaMO₃ oxides by the strong isotropic exchange interaction. The latter is reduced in SrRuO₃ suggesting essentially non-collinear magnetic arrangement.

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Table 1: Parameters of the isotropic exchange interaction $J_{ij}$ (in mRy). Atomic positions 1, 2 and 3 are shown in Fig.1.

| compound  | $J_{12}$  | $J_{13}$  |
|-----------|-----------|-----------|
| LaCrO$_3$ | -1.321    | -1.372    |
| LaMnO$_3$ | 0.225     | 0.668     |
| LaFeO$_3$ | -2.559    | -3.119    |
| SrRuO$_3$ | 0.306     | -0.101    |

Table 2: Parameters of the antisymmetric exchange interaction $d_{ij}$ for two $M$–O–$M$ bonds (in mRy). The symmetry of the nearest neighbor interactions is shown in Fig.1.

| compound  | $d_{12}$:(-$\alpha_c$,-$\beta_c$, 0) | $d_{13}$:($\alpha_{ab}$,-$\beta_{ab}$,$\gamma_{ab}$) |
|-----------|--------------------------------------|-------------------------------------------------|
| LaCrO$_3$ | (-0.005,-0.044, 0)                   | (0.029,-0.028, 0.035)                            |
| LaMnO$_3$ | (-0.032,-0.052, 0)                   | (0.032,-0.024, 0.039)                            |
| LaFeO$_3$ | (-0.019,-0.125, 0)                   | (0.075,-0.059, 0.086)                            |
| SrRuO$_3$ | (-0.138,-0.286, 0)                   | (-0.062,-0.127, 0.198)                           |
Figure 1: Parameters of the antisymmetric exchange interaction associated with different $M-O-M$ bonds in $D_{2h}^{16}$ structure (black and white spheres are $M$ and $O$ respectively). $\alpha$, $\beta$ and $\gamma$ are the components of $d_{ij}$ vectors along orthorhombic $a$, $b$ and $c$ axes for inplane ($ab$) and interplane ($c$) interactions.