Journal of Magnetism and Magnetic Materials 377 (2015) 121–125

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Effects of magnetic correlation on the electric properties in multiferroic materials

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ARTICLE INFO

Article history:
Received 16 May 2014
Received in revised form
10 October 2014
Accepted 17 October 2014
Available online 22 October 2014

Keywords:
Magnetic correlation
Multiferroic materials
Green's function
Magnetic field control of polarization

ABSTRACT

The effects of magnetic correlation on the electric properties in the multiferroic materials are studied, where the phase transition temperature of the magnetic subsystem T_m is lower than that of the electric subsystem T_e. A Heisenberg-type Hamiltonian and a transverse Ising model are employed to describe the ferromagnetic and ferroelectric subsystems, respectively. We find that the magnetic correlation can influence the electric properties above the T_m, and magnetic transverse and longitudinal correlations have opposite functions. In the curves of temperature dependence of polarization, kinks appear at T_m which is dominated by the sharp change of decreasing rate of the magnetic correlation. The kinks can be eliminated by an external magnetic field. The magnetic transverse and longitudinal correlations play contrary roles on the manipulation of polarization by the external magnetic field.

1. Introduction

Since the promising technological potentials in storage devices and sensors and other fields, multiferroic materials, in which ferroelectric (FE) and ferromagnetic (FM) or antiferromagnetic (AFM) orderings coexist, have been the subject of intensive theoretical and experimental studies [1–3]. Because of the interaction between the two kinds of orders in such materials, i.e. the magnetoelectric coupling, the magnetic properties could be changed by the ferroelectric ordering or an external electric field, and the electric properties can also be changed by the fluctuation of spin ordering or an external magnetic field. The effect of magnetoelectric coupling is particularly strong when temperature is close to or just below magnetic phase transition temperature and may result in a change of electric properties [4–6]. In recent studies, it has been found that pair correlation between magnetic lattice sites, hereafter referred to as magnetic correlation for brevity, played an important role on the electric properties [7–11], and there have been theoretical works devoted to the investigations of this effect [11–16].

In a multiferroic material, the FE and FM (AFM) subsystems have their own order–disorder transition temperatures, the values of which are denoted as T_e and T_m, respectively. In the previous microscopic studies, it was generally believed that the magnetoelectric coupling would disappear above the lower one of the T_e and T_m [17,18]. For example, if T_m is lower than T_e, then the magnetic correlation (MC) above the T_m was considered to be zero [19]. In a pure magnetic system, the MC in fact contains two parts: magnetic transverse correlation (MTC) and magnetic longitudinal correlation (MLC). Both of them are nonzero at the phase-transition temperature [20]. Therefore, whether the MC will be zero or not above the transition point should be carefully studied. This is necessary in the investigation of the effect of MC on polarization. In this paper, the effects of the MTC and MLC on polarization are studied in detail. Our investigation is performed in a multiferroic material with T_e is larger than T_m. We find that the growth of coupling constant can raise the polarization and reduce the T_e, which demonstrates that MC can influence the electric properties above the T_m. Such conclusion is different from the previous studies [8]. Moreover, by evaluating the effects of MTC and MLC on T_e, we find that T_e is increased by MTC while reduced by the MLC.

In Section 2 the Hamiltonian is presented. For magnetic subsystem, a Heisenberg-type Hamiltonian is employed by which both MTC and MLC can be computed correctly. For the electric subsystem, the transverse Ising model (TIM) is employed [21,22], since it is suitable for describing a large class of FEs. We use the double-time Green's function method which enables us to treat both subsystems satisfactorily [20,23]. In Section 3, the numerical results of the temperature dependence of magnetization, MTC, MLC, and MC are presented, and their effects on the electric properties are studied. Finally, a summary is presented in Section 4.

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http://dx.doi.org/10.1016/j.jmmm.2014.10.072
0304-8853/

2. The model Hamiltonian and formalism

2.1. The model Hamiltonian

In present discussion, we assume that the studied multiferroic compound is in a perovskite like structure, and the magnetic and electric orderings are originated from different units. Therefore, the multiferroic crystal contains two subsystems: the magnetic and electric subsystems. There is a coupling interaction between them. The total Hamiltonian can be presented as [24–27]

$$H = H_m + H_e + H_{me}. \tag{1}$$

H_m and H_e denote the Hamiltonians of the magnetic and electric subsystems, respectively. H_{me} describes the coupling interaction.

The magnetic subsystem is described by a Heisenberg model. Its Hamiltonian reads [7,8]

$$H_m = -J_1\sum_{[i,j]} \mathrm{S}_i\cdot\mathrm{S}_j - J_2\sum_{\langle i,j\rangle} \mathrm{S}_i\cdot\mathrm{S}_j - B_z\sum_i S_i^z. \tag{2}$$

$\boldsymbol{S}_i = \left(S_i^x, S_i^y, S_i^z\right)$ is the spin operator at site i. Here, J_1 and J_2 are the exchange integral parameters between the nearest and next-nearest magnetic lattice sites. The denotations $[i, j]$ and $\langle i, j\rangle$ mean that the nearest and next-nearest neighbors are involved, respectively. $B_z = g\mu_B H_z$, where g is the Landé factor and μ_B is the Bohr magneton, and H_z is the external magnetic field along the z direction. Here, we use the B_z in calculation instead of H_z. In this paper, we set Boltzmann constant k_B=1. By modifying parameters J_1 and J_2 properly, the magnetic subsystem may show a FM or AFM configuration.

The electric subsystem is modeled by the TIM, which can be written as [21–23]

$$H_e = -\Omega\sum_i P_i^x - J_e\sum_{[i,j]} P_i^z\cdot P_j^z - E_z\sum_i \mu P_i^z. \tag{3}$$

P_i^x and P_i^z are the spin-1/2 operators of the pseudo-spins which obey the commutation relation of convenient spin operators. The pseudo-spin P_i^z represents the two positions of ferroelectric unit at the lattice point i with a barrier between them. P_i^x is responsible for the tunneling process between two positions, and Ω is the tunneling frequency. J_e denotes the interaction between adjacent electric lattice sites. E_z represents the external electric field along the z direction, and μ is the effective dipolar moment of each pseudo-spin. In this subsystem, the mean electric polarization is proportional to the statistical average of P_i^z.

For current discussion, the coupling term H_{me} is written as [24–27]

$$H_{me} = -g\sum_{[ij]}\sum_{[kl]} \mathrm{S}_i\cdot\mathrm{S}_j P_k^z P_l^z = -g\sum_{[ij]}\sum_{[kl]}\left(S_i^+S_j^- + S_i^zS_j^z\right)P_k^zP_l^z. \tag{4}$$

Here, g is the intensity of the magnetoelectric coupling, and $S_i^\pm = S_i^x \pm iS_i^y$. Such coupling means that the magnetic and electric subsystems have independent mechanism. The MTC and MLC are defined as the statistical averages of the terms $\sum_{[i,j]}S_i^+S_j^-$ and $\sum_{[i,j]}S_i^zS_j^z$, respectively. They are written as $C_T = \sum_{[i,j]}\left\langle S_i^+S_j^-\right\rangle$ and $C_L = \sum_{[i,j]}\left\langle S_i^zS_j^z\right\rangle$. The sum of them is the MC defined as $C_M = C_T + C_L$. The coupling Hamiltonian Eq. (4) indicates that the MC can influence the polarization directly, and the magnetization is also affected by the correlation between the pseudo-spins.

2.2. The formalism

We use the double-time Green's function method [20]. It is applicable to the whole temperature range, and easy to study various systems. Especially, the MTC and MLC can be evaluated by means of this method in a satisfactory way. For the magnetic subsystem, the operators $A = \left(S_m^+, S_e^+\right)^{\mathrm{T}}$, $B = \left(e^{uS_m^z}S_m^-\ \ e^{uS_e^z}S_e^-\right)$ are chosen to construct Green's functions as follows [28,29]:

$$G_{ij}(t, t') = \langle\langle A_i(t); B_j(t')\rangle\rangle. \tag{5}$$

The H_m and H_{me} can merge to be an effective magnetic Hamiltonian:

$$\begin{aligned} H_M &= H_m + H_{me} \\ &= -\left(1 + \frac{g}{J_1}\sum_{[kl]} P_k^zP_l^z\right)J_1\sum_{[i,j]} \mathrm{S}_i\cdot\mathrm{S}_j - J_2\sum_{\langle i,j\rangle} \mathrm{S}_i\cdot\mathrm{S}_j - B_z\sum_i S_i^z. \end{aligned} \tag{6}$$

Roughly speaking, the first term on the right hand side of Eq. (6) can be regarded as a simple Heisenberg exchange form $-J_1'\sum_{[i,j]}\mathrm{S}_i\cdot\mathrm{S}_j$, where the effective exchange integral parameter is

$$J_1' = \left(1 + \frac{g}{J_1}\sum_{[kl]}\langle P_k^zP_l^z\rangle\right)J_1. \tag{7}$$

Green's function method is employed following the standard routine [30–32]. Here we do not intend to present the tedious formalism. The well-known spectral theorem and its derivative with respect to time t help us to calculate various thermodynamic quantities [20].

On the other hand, if H_e and H_{me} are merged, we can define an effective electric Hamiltonian as follows:

$$\begin{aligned} H_E &= H_e + H_{me} \\ &= -\Omega_i\sum_i P_i^x - \left(1 + \frac{g}{J_e}\sum_{[i,j]}\boldsymbol{S}_i\cdot\boldsymbol{S}_j\right)J_e\sum_{[i,j]} P_i^z\cdot P_j^z - E_z\sum_i \mu P_i^z. \end{aligned} \tag{8}$$

And also an effective coupling constant between nearest electric lattice sites is defined as follows:

$$J_e' = \left(1 + \frac{g}{J_e}\sum_{[i,j]}\left\langle\boldsymbol{S}_i\cdot\boldsymbol{S}_j\right\rangle\right)J_e. \tag{9}$$

We use the creation and annihilation operators of Fermion type $a_{i+}(t)$ and $a_{j+}^+(t)$ to construct Green's functions of electric subsystem as follows [23]:

$$G_{i,j}'(t, t') = \langle\langle a_{i+}(t); a_{j+}^+(t)\rangle\rangle. \tag{10}$$

Following the treatment in Ref [23], the relative polarization is obtained by the simple decoupling approximation:

$$\langle P_i^z\rangle = \frac{J_e'\langle P_j^z\rangle + E\mu}{\sqrt{\left(J_e'\langle P_j^z\rangle + E\mu\right)^2 + \Omega_i^2}}\tanh\left(\frac{\sqrt{\left(J_e'\langle P_j^z\rangle + E\mu\right)^2 + \Omega_i^2}}{2k_BT}\right). \tag{11}$$

Obviously, the expression coincides completely with the usual MFA.

3. Results and discussions

We assume that the magnetic subsystem is a FM one. The parameters are chosen as $S = 2$, $J_1 = 20$, $J_2 = 2$, $J_e = 720$,and $\Omega = 50$ so that T_m is lower than T_e. The coupling parameter g is set to various values. Parameters used here do not contrapose a given multiferroic compound, but they are representative for a number of multiferroic compounds for a qualitative study.

First of all, we investigate the effect of magnetoelectric coupling on the magnetization. Fig. 1 plots the magetizaiton versus temperature curves at three coupling values. It is shown that T_m increases with the growing coupling constant g, which is in accordance with the previous theoretical predications [9,33]. In the