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The inherent magnetoelectric properties in the three-dimensional ferroelectromagnetic system

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Abstract

Ferroelectromagnet is a special compound which shows the coexistence of the spontaneous ferroelectricity and (anti)ferromagnetism in the same phase. The inherent magnetoelectric (ME) coupling effect is the most attractive property for the ferroelectromagnetic compound. In this paper, we thoroughly research into the influence of the inherent ME coupling on the properties of the three-dimensional ferroelectromagnetic system, including the magnetization, magnetic susceptibility, polarization and dielectric susceptibility. Calculation reveals that the magnetic and dielectric properties both display some anomalous change for the influence of the ME effect. For dielectric properties, an anomaly in both spontaneous polarization and dielectric susceptibility is observed, indicating the onset of the magnetic order. For magnetic properties, an excursion in Néel temperature T_N is observed, indicating the influence of the ferroelectric order. In addition, the ME-coupling dependence of the spin wave excitations of the three-dimensional ferroelectromagnetic system is also considered.

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Keywords: Ferroelectromagnet; Magnetoelectric; Mean-field theory

1. Introduction

The earliest discovery of the ferroelectromagnet (FEM) can be traced back to the late 1950s and the early 1960s by the former soviet union scientists Smolenskii and co-workers [1–4]. In the meantime, Bertaut et al. found the hexagonal rare-earth manganites having the overall formula $RMnO_3$, where R = Y, Ho, Er, Tm, Yb, Lu or Sc [5], are FEMs with antiferromagnetic and weak ferroelectric properties [3–5]. Ever since,

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much work has been done on the investigation of the magnetoelectric (ME) effect. In theoretical aspect, in 1959 Landau and Lifshitz first pointed out the possible existence in magnetically ordered crystals of an equilibrium electric polarization proportional to the magnetic-field intensity and of an equilibrium magnetization proportional to the electric-field intensity. Moreover, they predicted there was an allowed term in the free energy of the form $\alpha_{i,j}H_iE_j$ based on the group theory, where E and H represent external electric and magnetic field, $\alpha_{i,j}$ is the element of a tensor showing the correlation between H and E [6]. Then Rado proposed the two-ion model and explained the temperature dependence of the ME effect [7], followed by the ameliorated approaches of Hornreich and Shtrikman [8], Yatom and Englman [9], and Gehring [10]. These theories, however, are concerned only with the coupling term caused by the external field as perturbation to the system, thus it is only applicable to the system which has weak coupling.

As far as FEM is concerned, although experimental evidence has shown that the ME energy, which comes from the coexistence of the spontaneous ferroelectric and antiferromagnetic order, can be comparable with the magnitudes of the intrinsic spin and ferroelectric energies [11,12], it seems that this kind of ME energy does not trigger off research interest. One of the most important reasons is that, for many of the ferroelectromagnetic compounds examined, the ferroelectric transition temperature T_E is much higher than the antiferromagnetic transition temperature T_{N}. Another reason is that it is difficult to find a suitable theory to describe the coupling. Besides, devising experiments to probe the nature of the coupling is likewise difficult. Until recently, Z.J. Huang et al. have detected an inversed S-shaped anomaly in both dielectric constant and loss tangent in bulk yttrium $YMnO_3$ [13]. Furthermore, in single crystal $RMnO_3$ ($R = Y, Yb, Lu$), the dielectric and magnetic anomalies were also observed [14]. The interest in the ME effect is again ignited. In order to describe the intrinsic coupling between the electric and magnetic subsystem in FEM, Gao et al. [15,16] utilized Monte Carlo simulations on the base of the Ising-DIFFOUR model to investigate the phase transition in a one- and two-dimensional ferroelectromagnetic lattice. They thought the coupling interaction between the ferroelectric and magnetic subsystems should be described as $-\varepsilon_{ij}gu_k^2s_is_j$, where g is the coupling coefficient, u_k is the electric displacement for spontaneous polarization, and s_i is the Ising spin operator.

In this paper, we will investigate the intrinsic ME coupling in the three-dimensional ferroelectromagnetic lattice based on the Heisenberg model. In our previous work, we have studied the ME properties related with the external electric (or magnetic) fields in the three-dimensional A-type antiferromagnet [17]. Now, we further presume that the A-type antiferromagnet is simultaneously a ferroelectric, i.e. ferroelectromagnet. For such a FEM, the intrinsic ME coupling is dominant compared with the external-field-induced coupling. Our work emphasizes on researching into the contribution of the intrinsic ME coupling. To our knowledge, A-type antiferromagnet shows a clear anisotropy in magnetic structure. Below T_{N}, the spins are ferromagnetically coupled in the basal plane and antiferromagnetically coupled in the normal direction of the plane. This anisotropic magnetic structure may also result in the anisotropy of the ME coupling form. Therefore, we use $g_{\parallel}u_k^2(\mathbf{S}_{ai}\cdot\mathbf{S}_{aj})$, $g_{\parallel}u_k^2(\mathbf{S}_{bi}\cdot\mathbf{S}_{bj})$ and $g_{\perp}u_k^2(\mathbf{S}_{ai}\cdot\mathbf{S}_{bj})$ to describe the intraplane and interplane coupling, respectively. In formula, a and b represent the nearest planes. $\mathbf{S}_{ai}$ denotes the Heisenberg spin operator at site i in basal plane a, $g_{\parallel}$ and $g_{\perp}$ are the intraplane and interplane ME-coupling coefficient, respectively.

Our paper is organized as follows: in Section 2.1, we give an analysis of the ferroelectromagnetic system by partitioning it into ferroelectric and magnetic subsystem. Soft-mode theory and mean-field theory are utilized to deal with the ferroelectric and magnetic subsystems. For ferroelectric subsystem, an anomaly in dielectric susceptibility and polarization is observed around T_{N} indicating the onset of the magnetic order, which is qualitatively with the results obtained from the experiments [13,14]. For magnetic subsystem, an excursion in Néel temperature T_{N} is observed, indicating the influence of the ferroelectric order. In Section 2.2, we use spin wave theory to investigate the influence of the ME coupling on the excited spectrum of the spin wave. The advantage of the spin wave theory lies in that the correlation of the spins is considered instead of a mean field, therefore, it enables us to gain insight into the ME coupling on the spin wave