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Magnetodielectric effects in the ferroelectric ferromagnet BiMnO₃

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Abstract

The changes in the dielectric constant of perovskite BiMnO₃ at about T_C , which are induced by the magnetic ordering as well as by the application of magnetic fields, indicate that coupling exists between the magnetism and the dielectric properties. The soft-mode theory based on the single-well potential model and the molecular-field approximation to the Heisenberg model are successfully applied to the electric and magnetic subsystems coexisting in the perovskite-type ferroelectromagnet BiMnO₃, respectively. By adding an appropriate coupling term, which is related to a combination of electrical polarization and spin correlation, we investigate the inherent coupling between the ferroelectric and ferromagnetic orders, and find that the application of magnetic fields induces a fairly large suppression of χ_p around T_C , and the field-induced change in the magnetocapacitance effect becomes maximal at T_C , which are in good agreement with experimental data in ferroelectromagnetic BiMnO₃.

1. Introduction

The behaviour of systems with strongly coupled magnetic and electronic degrees of freedom has drawn significant interest in recent years due to both challenges for many-body theory as well as new phenomena for potential applications as transducers, actuators, and sensors [1–3]. One manifestation of such coupling in the itinerant limit is the interplay between ferromagnetism and charge order in the colossal magnetoresistance manganites [4, 5]. The effects of strong coupling are rather subtle in the limit of localized charge, and are manifested chiefly through a magnetocapacitive response, which can take several different forms [6–8]. After the first experimental realization of magnetoelectric coupling in Cr₂O₃ [9], similar effects have been

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observed in many other materials including Gd_2CuO_4 [10], YMnO_3 [11], EuTiO_3 [12] and BiMnO_3 [13]. The present strong interest in coupled magnetic dielectric properties is further motivating the search for so-called multiferroic materials in which two or all three ferroic orders (ferroelectrics, ferromagnetics, and ferroelastics) coexist in the same phase [14, 15].

Ferroelectromagnets, as a kind of multiferroic materials, in which (anti)ferroelectric and (anti)ferromagnetic ordering coexist spontaneously at low temperature, have also been the object of intensive theoretical and experimental studies from the 1960s to 1970s [16, 17]. It has been reported that the yttrium and rare-earth manganites AMnO_3 crystallize in two structural phase [18, 19]: a hexagonal phase for A with a small ionic radius; and an orthorhombic phase for A possessing a greater ionic radius. It turns out that the structure is primarily determined by the size of the A-site ions. In these compounds, magnetic ordering occurs in both hexagonal and orthorhombic manganite due to 180° and 90° superexchange in Mn–O–Mn bonds and strong Jahn–Teller distortion. Ferroelectric ordering occurs only in hexagonal systems which belong to the noncentrosymmetric symmetries (the $P6_3cm$ space group). A good example is yttrium manganite YMnO_3 or LuMnO_3 , in which each Mn^{3+} ion with $s = 2$ is surrounded by three in-plane and two apical oxygen ions, and thus MnO_5 blocks are formed. In these MnO_5 , the displacement of Mn ions along the c -axis, which is subject to a trigonal crystal field, causes a ferroelectric polarization. In the orthorhombic phase such as LaMnO_3 , however, spontaneous polarization does not exist, due to the centrosymmetric structure.

Recently, Atou, Moreira and their co-workers performed band-structure determinations on a perovskite BiMnO_3 by electron diffraction and neutron powder diffraction [20–22], and thought that although the ionic radii of trivalent lanthanum in LaMnO_3 and trivalent bismuth in BiMnO_3 are close to each other (Bi^{3+} : 1.24 Å, La^{3+} : 1.22 Å), their structures are different to a great extent. LaMnO_3 has an orthorhombic GdFeO_3 -type structure, while BiMnO_3 is related to a heavily distorted perovskite structure where the bismuth cation is shifted from the centre of the hexagon of the oxide anions due to the polarized lone pair $6s^2$ of the Bi^{3+} , resulting in a lowering of the symmetry of the whole structure (space group $C2$) below 750–770 K. All these indicate that the system has an off-centre distortion responsible for the polar behaviour.

Therefore, compared with the perovskite LaMnO_3 , BiMnO_3 is a ferroelectromagnet, which shows the coexistence and coupling between the two orderings. For the magnetic structure, data analysis reveals a collinear ferromagnetic structure with the spin direction along [010] and a magnetic moment of $3.2 \mu\text{B}$, which is slightly smaller than that of the fully aligned spin value of $4 \mu\text{B}$ for Mn^{3+} . This is because the ferromagnetism of BiMnO_3 stems directly from orbital ordering, and among the six unique Mn–O–Mn superexchange pathways, four are ferromagnetic and two antiferromagnetic [21].

In fact, due to the inherent coupling between the ferroelectric and ferromagnetic orders in the multiferroic BiMnO_3 , Kimura *et al* [13] reported that not only does the magnetic ordering and the application of magnetic fields suppress the dielectric constants, but a fairly large negative magnetocapacitance has also been observed in the vicinity of the ferromagnetic transition temperature T_C . As to these experimental results, although they have made a phenomenological interpretation, a profound understanding of the magnetoelectric coupling mechanism in BiMnO_3 is still an important and necessary issue.

Liu and Gao *et al* utilized Monte Carlo simulations on the basis of the Ising–Diffour model to investigate the phase transition in the two-dimensional ferroelectromagnetic lattice where the spin moment and polarization interact [23, 24]. For BiMnO_3 , however, things are different. First, the Heisenberg model must be applied instead of the Ising model, since the electronic configuration of Mn^{3+} ion in BiMnO_3 is $t_{2g}^3 e_g^1$ (spin quantum number $s = 2$) as in the famous orbital-ordered manganite LaMnO_3 . Second, based on the available experimental data for the incipient ferroelectric EuTiO_3 and the hexagonal ferroelectromagnetic YMnO_3 ,

Katsufuji *et al* [12] found that the changes in dielectric and magnetic properties of these materials are dominated by the pair correlation of the nearest-neighbour Mn ion spins, $\vec{s}_i \cdot \vec{s}_j$. So they proposed that the dielectric constant and spin correlation $\langle \vec{s}_i \cdot \vec{s}_j \rangle$ are related as $\varepsilon = \varepsilon_0(1 + \alpha \langle \vec{s}_i \cdot \vec{s}_j \rangle)$, which differs from the coupling term previously suggested in [23] and [24]. In our earlier work [25], we proposed a possible form for general magnetoelectric coupling, which is related to a combination of electrical polarization and spin correlation, to study the thermodynamic properties of the system with ferroelectric and antiferromagnetic coupling. It was found that an additional electrical polarization is induced by the effect of the magnetic moment, and this correspondingly leads to a reduction in the dielectric susceptibility at low temperature, which is consistent with the experimental results [11].

Thus, it is reasonable to add a term in the Hamiltonian of the form $g \sum_l \sum_{(i,j)} q_l^2 \vec{s}_i \cdot \vec{s}_j$, which describes the coupling interaction between the electrical and magnetic subsystems, in order to study the changes in the dielectric constant induced by the magnetic ordering as well as by the application of magnetic fields which were observed in BiMnO₃. On such an assumption, we obtain an analytical relation between the polarization susceptibility and the spin correlation as well as between the dielectric constants and the spin correlation, which is in good agreement with the experimental results. Our results show that the polarization and polarization susceptibility are related to the spin correlation $\langle \vec{s}_i \cdot \vec{s}_j \rangle$. We also discuss the behaviour of the magnetocapacitance at different temperatures and the polarization susceptibility under different magnetic fields.

In our present article, we first deal with the electrical subsystem, including electrical-magnetic coupling, using the soft-mode theory under the mean-field approximation. Second, to further investigate the temperature and magnetic-field dependence of the polarization susceptibility and magnetocapacitance, we study the magnetic spin correlation, which influences the behaviour of the polarization. Finally, we give our numerical results and a detailed discussion.

2. Model and analysis

Considering the difficulty in dealing with the magnetic structure of BiMnO₃ and noticing that the highly distorted perovskite structure may be transformed at a high temperature and pressure to the orthorhombic perovskite-type [13], we simulated this complicated ferromagnet with a partial antiferromagnetic order by a full ferromagnet with orthorhombic perovskite structure, to discuss the magnetodielectric effect. Thus, we consider the Hamiltonian for a three-dimensional cubic system with periodic boundary conditions. Two parameters, q_l and s_i are introduced here to represent the electrical displacement at the Bi site and the Heisenberg spin at the Mn site, respectively. In addition, the coupling interaction between the electrical and magnetic subsystems is taken into account. Following Janssen's model [26] the Hamiltonian for this coupling system can be presented as follows:

$$\mathcal{H} = \mathcal{H}^m + \mathcal{H}^e + \mathcal{H}^{me} \quad (1)$$

where \mathcal{H}^m denotes the Hamiltonian for the magnetic subsystem, \mathcal{H}^e is the Hamiltonian for the electrical subsystem and \mathcal{H}^{me} the coupling interaction between the two subsystems. Here, the soft-mode theory under the mean-field approximation is applied to solve \mathcal{H}^e . In treating \mathcal{H}^e , one assumes that the Mn and O ions are fixed and only Bi ions vibrate like a harmonic oscillator with small additional anharmonic part. That is to say q_l at each site is subjected to a single-well potential. Thus \mathcal{H}^e in the presence of the electric field can be written as follows:

$$\mathcal{H}^e = \sum_l \left[\frac{p_l^2}{2m} + \frac{1}{2} m \omega_0^2 q_l^2 + \frac{1}{4} \gamma q_l^4 \right] - \frac{1}{2} \sum_l \sum_{l'} v_{ll'} q_l q_{l'} - \sum_l E q_l \quad (2)$$

where p_l is the momentum at site l , and ω_0 is the inherent frequency. The second term is the harmonic part and the third term the anharmonic one. $\frac{1}{2} \sum_l \sum_{l'} v_{ll'} q_l q_{l'}$ denotes the two-body interaction potential and the last term the electric field effect. Here we use the simplest form to describe the anharmonic motion at the Bi site.

The interaction between Mn^{3+} spins originates from superexchange (mainly ferromagnetic) through the O 2p state. \mathcal{H}^m with $s = 2$ can be written as

$$\mathcal{H}^m = \sum_{\langle i,j \rangle} J_1 \vec{s}_i \cdot \vec{s}_j + \sum_{[i,j]} J_2 \vec{s}_i \cdot \vec{s}_j - \sum_i \vec{h} \cdot \vec{s}_i \quad (3)$$

where we take into account exchange interactions up to the second nearest neighbours. J_1 and J_2 represent the ferromagnetic coupling between the nearest neighbours and the next-nearest neighbours, respectively. \vec{s}_i is the Heisenberg spin at the Mn site i , and h the external magnetic field parallel to the z axis. $\langle i, j \rangle$ and $[i, j]$ denote the single summation over the nearest neighbours and the next nearest neighbours, respectively.

As pointed out in the previous section and [25], we consider the coupling term between the magnetic and electric subsystems as

$$\mathcal{H}^{me} = -g \sum_l \sum_{\langle i,j \rangle} q_l^2 \vec{s}_i \cdot \vec{s}_j \quad (4)$$

where g is the coupling coefficient. Here, only the quadratic term of q_l is taken because of the time-reversal and space-reversal symmetries. l represents the lattice site of the Bi ion, and i and j denote distinct nearest-neighbour Mn ions to a given Bi ion.

First, we deal with the electrical subsystem, considering electrical–magnetic coupling. Under the mean-field approximation, the single-ion Hamiltonian can be written as

$$\mathcal{H}_l^E = \mathcal{H}_l^e + \mathcal{H}_l^{me} = \frac{p_l^2}{2m} + \left(\frac{1}{2} m \omega_0^2 - g \sum_{\langle i,j \rangle} \langle \vec{s}_i \cdot \vec{s}_j \rangle \right) q_l^2 + \frac{1}{4} \gamma q_l^4 - \sum_{l'} v_{ll'} \langle q_{l'} \rangle q_l - E q_l. \quad (5)$$

To facilitate further discussion, we define

$$\frac{1}{2} m \omega_0^2(g) = \frac{1}{2} m \omega_0^2 - g \sum_{\langle i,j \rangle} \langle \vec{s}_i \cdot \vec{s}_j \rangle. \quad (6)$$

The magnetic effect on the electrical subsystem is embodied by $\omega_0(g)$, thus,

$$\mathcal{H}_l^E = \frac{p_l^2}{2m} + \frac{1}{2} m \omega_0^2(g) q_l^2 + \frac{1}{4} \gamma q_l^4 - \sum_{l'} v_{ll'} \langle q_{l'} \rangle q_l - E q_l. \quad (7)$$

According to the soft-mode theory based on the mean-field approximation, we can then obtain

$$\begin{aligned} (m \omega_0^2(g) + 3\gamma \sigma_l + \gamma \langle q_l \rangle^2) \langle q_l \rangle &= \sum_{l'} v_{ll'} \langle q_{l'} \rangle + E \\ (m \omega_0^2(g) + 3\gamma (\sigma_l + \langle q_l \rangle^2)) \sigma_l &= k_B T \end{aligned} \quad (8)$$

where k_B and T are Boltzmann's constant and thermal temperature, and σ_l is the fluctuation of the Bi ion displacement, which is defined by $\sigma_l = \langle q_l - \langle q_l \rangle \rangle^2$. Likewise, the frequency of the soft-mode can be derived:

$$m \omega_{\vec{q}}^2(g) = m \omega_0^2(g) + 3\gamma (\sigma_l + \langle q_l \rangle^2) - v_{\vec{q}} \quad (9)$$

where

$$v_{\vec{q}} = \sum_{l'} v_{ll'} \exp[-i\vec{q} \cdot (\vec{R}_l - \vec{R}_{l'})] \quad (10)$$

is the interaction potential at the wavevector \vec{q} . For ferroelectric BiMnO₃, the soft-phonon mode is in the centre of the Brillouin zone ($\vec{q} = 0$). Therefore, beginning with the single-mode theory under the mean-field approximation, the frequency of the soft-phonon mode can be written as

$$m\omega_{\vec{q}=0}^2(g) = m\omega_{\vec{q}=0}^2(0) (1 - \alpha \langle \vec{s}_i \cdot \vec{s}_j \rangle) \quad (11)$$

where

$$\alpha = \frac{2z_2g}{m\omega_{\vec{q}=0}^2(0)} = \frac{2z_2g}{m\omega_0^2 + 3\gamma(\sigma_l + \langle q_l \rangle^2) - \nu_{\vec{q}=0}}. \quad (12)$$

$\omega_{\vec{q}=0}(0)$ is the frequency of the soft-phonon mode as the coupling interaction is not considered, and it is approximately temperature independent at very low temperature. $z_2 = 12$ is the number of the spin correlations that will directly affect the motion of Bi in one unit. The static polarization susceptibility, which embodies the dielectric constant, is given by

$$\chi_p = \frac{1}{m\omega_{\vec{q}=0}^2(0)} (1 + \alpha \langle \vec{s}_i \cdot \vec{s}_j \rangle) = \chi_p(0) (1 + \alpha \langle \vec{s}_i \cdot \vec{s}_j \rangle) \quad (13)$$

where $\chi_p(0)$ is the static susceptibility in the absence of spin correlation. Thus, the relative dielectric constant ε can be derived from equation (13) as follows:

$$\varepsilon = \varepsilon(0) (1 + \beta \langle \vec{s}_i \cdot \vec{s}_j \rangle) \quad (14)$$

where

$$\varepsilon(0) = 1 + \chi_p(0) \quad (15)$$

$$\beta = \frac{\alpha \chi_p(0)}{1 + \chi_p(0)}. \quad (16)$$

From the above equation, we can conclude that our theoretical result of ε shows a good agreement with the dielectric properties fitted from the experimental data [12]. It is shown that the coupling between the polarization and spin correlations arises from the coupling of magnetic fluctuations to the soft-phonon frequencies. That is, the spin correlations perturb the optical phonon frequencies, which in turn shift the dielectric constant. To investigate how the dielectric constant varies under different conditions of magnetic field, we take into account the spin correlation in the coupling system. Then, the Hamiltonian for the magnetic subsystem can be written:

$$\mathcal{H}^M = \mathcal{H}^m + \mathcal{H}^{\text{mc}} = \sum_{\langle i,j \rangle} \left(J_1 - g \sum_l \langle q_l^2 \rangle \right) \vec{s}_i \cdot \vec{s}_j + \sum_{\langle i,j \rangle} J_2 \vec{s}_i \cdot \vec{s}_j - \sum_i \vec{h} \cdot \vec{s}_i. \quad (17)$$

Using a standard molecular-field approach, we rewrite the Hamiltonian for spin at site i :

$$\mathcal{H}_i^M = -H_i^x s_i^x - H_i^z s_i^z \quad (18)$$

where H_i^v ($v = x, z$) is the equivalent magnetic field imposed on spin at site i . x, z denote the spin components.

$$\begin{aligned} H_i^x &= -z_1 (J_1 - z_3 g \langle q_l^2 \rangle) \langle s_i^x \rangle - z_2 J_2 \langle s_i^x \rangle \\ H_i^z &= h - z_1 (J_1 - z_3 g \langle q_l^2 \rangle) \langle s_i^z \rangle - z_2 J_2 \langle s_i^z \rangle \end{aligned} \quad (19)$$

where $z_1 = 6$, $z_2 = 12$ are the coordinate numbers of the nearest and the next-nearest neighbours of Mn³⁺, and $z_3 = 4$ is the number of Bi³⁺ that will directly affect the spin configuration at each pair of correlation. According to statistical physics, the average magnetic

moment of different directions can be derived by using the following solutions in the case of $s = 2$:

$$\langle s_i^v \rangle = -\frac{1}{Z_i} \sum_{k=1}^5 \frac{\partial \lambda_i^{(k)}}{\partial H_i^v} \exp(-\beta \lambda_i^{(k)}); \quad Z_i = \sum_{k=1}^5 \exp(-\beta \lambda_i^{(k)}) \quad (20)$$

where $\beta = 1/k_B T$, and $\lambda_i^{(k)}$ are the eigenvalues of the matrix

$$\begin{pmatrix} -2H_i^z & -H_i^x & 0 & 0 & 0 \\ -H_i^x & -H_i^z & -\frac{\sqrt{6}}{2}H_i^x & 0 & 0 \\ 0 & -\frac{\sqrt{6}}{2}H_i^x & 0 & -\frac{\sqrt{6}}{2}H_i^x & 0 \\ 0 & 0 & -\frac{\sqrt{6}}{2}H_i^x & H_i^z & -H_i^x \\ 0 & 0 & 0 & -H_i^x & 2H_i^z \end{pmatrix}.$$

The eigenvalues are readily obtained. Then, the self-consistent equation (20) reduces to

$$\langle s_i^x \rangle = \frac{H_i^x}{H_i} \frac{4 \sinh 2\beta H_i + 2 \sinh \beta H_i}{2 \cosh 2\beta H_i + 2 \cosh \beta H_i + 1} \quad (21)$$

$$\langle s_i^z \rangle = \frac{H_i^z}{H_i^x} \langle s_i^x \rangle \quad (22)$$

where $H_i = \sqrt{(H_i^x)^2 + (H_i^z)^2}$. Within the framework of the mean-field approximation, on condition that only two spin components are considered, the spin correlation between the nearest neighbours can be decoupled as

$$\langle \vec{s}_i \cdot \vec{s}_j \rangle = \langle s_i^x \rangle \langle s_j^x \rangle + \langle s_i^z \rangle \langle s_j^z \rangle. \quad (23)$$

3. Results and discussions

In order to investigate the magnetodielectric effect in multiferroic perovskite BiMnO₃, the thermodynamic average value of physical quantities via temperature $k_B T$ and via magnetic field h can be obtained numerically by the above self-consistent equations (8), (21) and (22), respectively. In the light of references [23, 24] and in the case of real multiferroic perovskite BiMnO₃, which displays a lower magnetic ordering $T_C = 105$ K and a rather high ferroelectric $T_E = 750$ K, the parameters used in our calculation are $J_1 = -0.25k_B$ K, $J_2 = -0.1k_B$ K which produce a magnetic phase transition temperature $T_C = 10$ K for a cubic perovskite ferromagnetic system, $m\omega^2 = 15$, $\gamma = 55$, $\nu = 20$ which produce a phase temperature $T_E = 80$ K for a ferroelectric interaction, and $g = 0.05$ for the magnetodielectric coupling interaction. Although the T_E and T_C given by these parameters are different from those in real material BiMnO₃, the ratio of T_C and T_E is approximately equal to that in BiMnO₃; thus we use these parameters to study analogously the magnetodielectric effect in this ideal multiferroic single-crystal BiMnO₃.

The isothermal magnetization and magnetocapacitance curves at various temperatures are displayed in figures 1(a) and (b), respectively. Here, we give the situations for the temperatures $T = 6, 8, 10, 12$ and 14 K. As seen in figure 1(a), the saturated moment is near that expected for the full Mn moment at the lowest temperature (6 K), and gradually decreases with increasing temperature. In multiferroic perovskite BiMnO₃, because of a collinear ferromagnetic coupling structure with the spin direction along [010], the ground state magnetization should be zero in the z axis direction in the absence of magnetic field. However, if a magnetic field perpendicular to the ground state spin direction is applied to such ferromagnetically ordered Heisenberg spins, they are immediately flipped parallel to the magnetic field and then are gradually increased

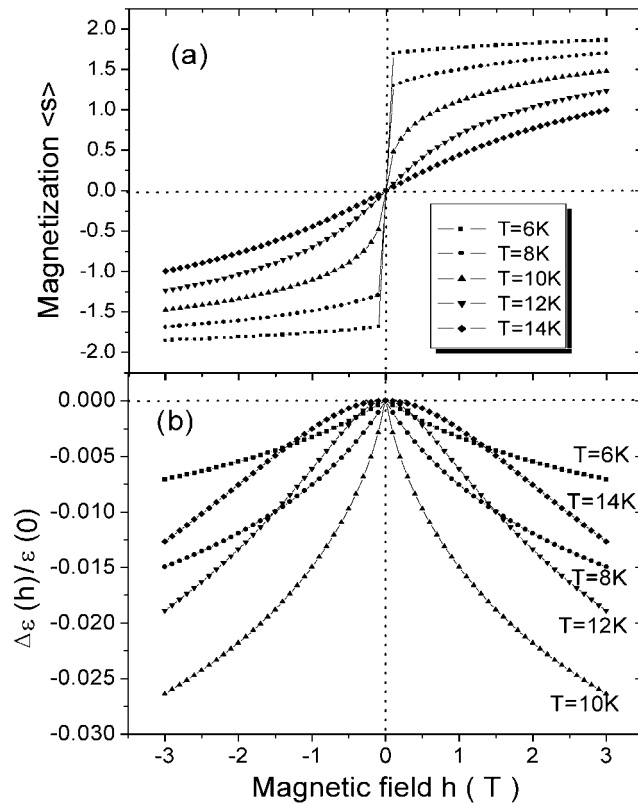


Figure 1. Isothermal (a) magnetization and (b) field-induced change in the dielectric constant as a function of a magnetic field at various temperatures.

to saturation with increasing magnetic field, as shown in figure 1(a). At the same time, we can also find that the higher the temperature is, the more slowly the spins parallel to the magnetic field are flipped. This is analogous to the situation of Heisenberg antiferromagnet. If the magnetic field is applied to an antiferromagnetically ordered Heisenberg spins, they are immediately flipped perpendicular to the magnetic field and then are gradually changed into a ferromagnetic arrangement with increasing magnetic field.

To further clarify the character of the magnetocapacitance effect, we define $\Delta\varepsilon(h)/\varepsilon(0) = [\varepsilon(h) - \varepsilon(0)]/\varepsilon(0)$. From figure 1(b), we can find that a fairly large negative magnetocapacitance effect is observed in the vicinity of the ferromagnetic transition temperature $T_C = 10$ K. For the real material BiMnO₃, experimental data show that the magnitude of the magnetocapacitance reaches a maximum $-\Delta\varepsilon/\varepsilon(0) = 0.6\%$ at 100 K, which is the critical temperature of BiMnO₃. So our numerical calculation is qualitatively consistent with the results of experiments [13] carried out on multiferroic BiMnO₃. By comparing figure 1(a) with (b), it is easy to find that the magnetocapacitance is negligibly small at temperatures sufficiently below T_C , although the magnetization has reached ≥ 0.8 of its saturation value by a small magnetic field. This suggests that the magnetic domain rotation due to the external magnetic field least affects the dielectric constant at low temperature. With increasing temperature, the magnitude of the magnetocapacitance becomes larger, and reaches its maximum around $T_C = 10$ K. On further increasing the temperature above T_C ,

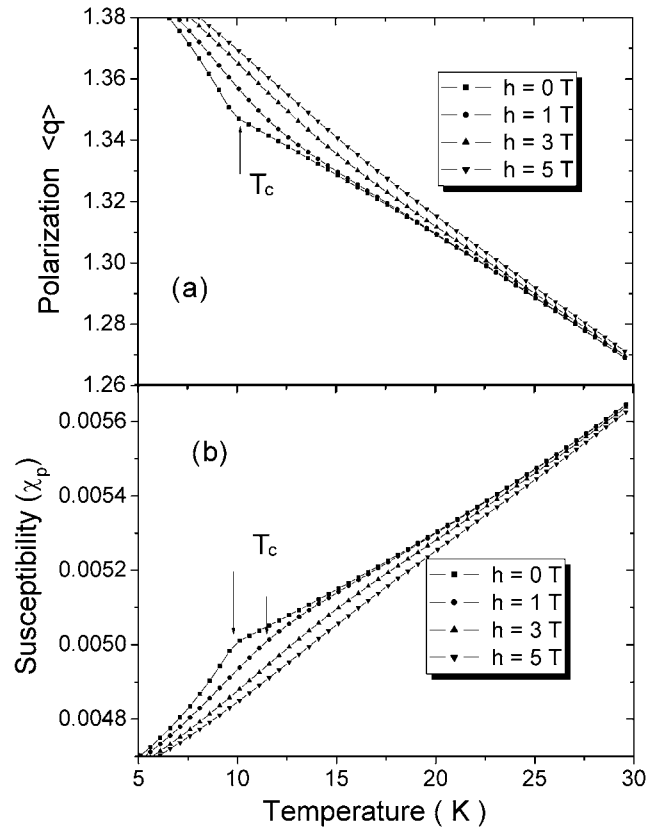


Figure 2. (a) Polarization and (b) polarization susceptibility as a function of temperature under different conditions of magnetic field.

the magnetocapacitance again decreases. As for the one that will be also seen from the curve of $\chi_p - k_B T$, it should be ascribed to the magnetodielectric coupling term $g \sum_i \sum_{\langle i,j \rangle} q_i^2 \vec{s}_i \cdot \vec{s}_j$ in the thermodynamic potential. Furthermore, by fitting $-\Delta\varepsilon/\varepsilon(0)$ to the square of $\langle s_i \rangle$, it is not difficult to find that there exists such a simple relation: $-\Delta\varepsilon/\varepsilon(0) \propto \langle s_i \rangle^2$, which is in a agreement of the result of Ginzburg–Landau theory [16]. This is because [13, 16] the magnetocapacitance effect can also be phenomenologically interpreted in terms of a simple Ginzburg–Landau theory for the second-order phase transition of ferroelectromagnets.

The polarization $\langle q \rangle$ and polarization susceptibility χ_p as a function of temperature are shown in figures 2(a) and (b), in which the different magnetic fields are given as 0, 1, 3 and 5 T. It can be seen that the polarization $\langle q \rangle$ decreases with the increase in temperature. This is because increasing the temperature leads to a disorder in the electrical polarization order. The different magnetic fields, nevertheless, change not only the ferromagnetic order phase transition temperature T_C but also the value of the ferroelectric polarization $\langle q \rangle$ at a certain temperature around T_C . Since the magnetic-order parameter of a ferromagnet can be controlled by the application of a relatively low magnetic field near T_C , correspondingly, the single-well potential parameter is also modified due to the magnetodielectric coupling term: $g \langle q_i^2 \rangle \sum_{\langle i,j \rangle} \vec{s}_i \cdot \vec{s}_j$, i.e., the harmonic part parameter becomes $(1/2)m\omega_0^2 - g \sum_{\langle i,j \rangle} \vec{s}_i \cdot \vec{s}_j$. In particular, we find that this enhancement of polarization $\langle q \rangle$ is very obviously around T_C because of the increase of the magnetic field.

At the same time, from figure 2(b), it is easy to find that remarkable features in the $\chi_p - k_B T$ curves are anomalies in the vicinity of T_C , which is also consistent with the data of Kimura *et al* [13]. At zero magnetic field, χ_p decreases steeply below T_C ; this suggests that the magnetic ordering suppresses the dielectric constant. Due to the application of magnetic fields, a fairly large suppression of relative dielectric constant is induced around T_C ; that is to say, the application of magnetic fields suppresses the anomaly in dielectric constant at T_C , which is consistent with figure 1(b) where we have drawn a conclusion that the magnetic-field-induced change in the magnetocapacitance effect becomes maximal at T_C . In addition to the occurrence in ferroelectric ferromagnet BiMnO₃, of course, a similar cusp of the $\chi_p - k_B T$ curves correlated with the magnetic ordering can be observed in some ferroelectric antiferromagnets such as hexagonal rare-earth manganite YMnO₃ [11, 27].

4. Summary

In summary, we have examined the relation between magnetocapacitance and magnetization of a ferroelectromagnet BiMnO₃ with orbital ordering and magnetic field by an approach which combines the soft-mode theory for ferroelectric order and the mean-field approximation for ferromagnetic order. We have also found that a fairly large negative magnetocapacitance effect occurs around the ferromagnetic transition temperature T_C . At the same time, the application of magnetic fields also induces a rather large suppression of the relative dielectric constant in the vicinity of T_C . The results obtained are qualitatively consistent with previous theoretical and experimental measurements.

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