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Magnetic field control of ferroelectric polarization and magnetization of $LiCu_2O_2$ compound*

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A spin model of $LiCu_2O_2$ compound with ground state of ellipsoidal helical structure is adopted. Taking into account the interchain coupling and exchange anisotropy, we investigate the magnetoelectric properties in a rotating magnetic field and perform the Monte Carlo simulation on a two-dimensional lattice. A prominent anisotropic response is observed in both the magnetization curve and the polarization curve, qualitatively coinciding with the behaviors that are detected in the experiment. In addition, the influences of the magnetic field with various magnitudes on polarization are also explored and analyzed in detail. As the magnetic field increases, a much smoother polarization of angle dependence is exhibited, indicating the strong correlation between the magnetic and ferroelectric orders.

Keywords: multiferrocity, magnetoelectricity, spiral order, Monte Carlo simulation

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1. Introduction

Multiferroics, which possess magnetic and ferroelectric orderings in the same material, yield abundant physical mechanisms and broad application prospects. In recent years, the magnetism-driven ferroelectricity discovered in frustrated multiferroic materials has attracted considerable attention.[1–3] In these compounds, spiral spin states are usually found and give rise to surprising physical properties, such as flop, reversal and rotation of the electric polarization in an external magnetic field.[4–7] This brings us a hopeful prospect of controlling charges by applied magnetic fields, thereby constructing new forms of multifunctional devices.

The $LiCu_2O_2$ multiferroic compound is a prototype of one-dimensional (1D) $S = 1/2$ spiral magnet.[8–15] It has an orthorhombic crystal structure with identical numbers of magnetic Cu^{2+} and nonmagnetic Cu^{1+} ions. The magnetic Cu^{2+} ions are located at the center of edge-sharing CuO_4 plaquettes and form a zigzag-like spin-chain structure along the b axis. The Cu–O–Cu angle is nearly 94°.[16–19] As a result, according to the Kanamori–Goodenough rule, the nearest-neighbor ferromagnetic (FM) coupling J_2 and the next-nearest-neighbor antiferromagnetic (AFM) coupling J_4 are expected along the chain direction b axis. Such competing interactions will lead to frustration and an incommensurate helimagnetic state in this quantum spin system. Spontaneous electric polarization has been detected when the helical order sets in, and shows sensitive response to the applied field.

In order to reveal the origin of the polarization, many experiments have been carried out to study the magnetic structures of $LiCu_2O_2$. A previous neutron scattering study has indicated the helical magnetic order in the ab plane.[16] However, it is in discrepancy with the spin-current model or the inverse Dzyaloshinskii–Moriya interaction since spontaneous electric polarization emerges along the c axis. Later, Park *et al.*, according to the polarized neutron scattering experiments, proposed the bc-spiral plane,[11] which is partially supported by Seki *et al.*'s results, but the quantitative calculation of the intensity of polarized neutron reflections shows a prominent discrepancy.[20] Recently, by combining studies of neutron diffraction and Li-NMR results, Sato *et al.* proposed an ellipsoidal helical spin structure with the helical axis tilted by about 45° with respect to the a or b axis within the ab plane.[21,22] This new noncollinear spin configuration has received strong evidence support from the very recent experiments performed by Li *et al.*[23] They implemented the measurements of the dielectric constant and spontaneous polarization in nearly untwinned $LiCu_2O_2$ single crystals. The highly anisotropic responses observed in different magnetic fields are perfectly explained by using this 45°-tilt spin model.

Although a large number of experiments on $LiCu_2O_2$ have been done,[24–28] theoretical studies on its thermodynamic properties or field-dependent behaviors are still rare and unsatisfied, owing to inadequate information about ground states. The ellipsoidal helical spin structure suggested by Sato *et al.* provides us with the new clues to unveiling the underlying mechanism in this multiferroic compound. In this paper, concentrating on magnetoelectric coupling effect, we investigate the magnetization and polarization under a rotation of the

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magnetic field by Monte Carlo method. The results demonstrate the strong coupling between the ferroelectric and magnetic orders in this spiral magnet, and in some respects are qualitatively in good agreement with experimental results.

2. Model and simulation

In $LiCu_2O_2$, there are two linear Cu^{2+} chains, which propagate along the b axis and form a zigzag ladder-like structure (Fig. 1). The Hamiltonian of the system is written as

$$\begin{aligned} H = & \sum_{i,j}[J_1(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)_{XXZ}+J_2(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)_{XXZ} \\ & +J_4(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)_{XXZ}+J_\perp(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)_{XXZ}] \\ & -\sum_i D_i(\boldsymbol{S}_i\cdot\boldsymbol{e}_i)^2-\boldsymbol{h}\cdot\boldsymbol{S}_i-\boldsymbol{h}_{\rm e}\cdot\boldsymbol{P}_i, \end{aligned} \tag{1}$$

where $(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)_{XXZ}=S_i^xS_j^x+S_i^yS_j^y+\Delta S_i^zS_j^z$ with unit vectors of the classical spin components being S_i^x, S_i^y, and S_i^z. $\Delta\leq 1$ is an exchange anisotropy and theoretically expected to stabilize a vector chiral order.[26] Actually, in the edge-sharing spin–chain compounds substantial exchange anisotropies have been detected.[29] In order to yield results that are much close to experimental results, the value of $\Delta=0.7$ is adopted. Considering the 45°-tilt spin model proposed by Sato *et al.* (Fig. 2), a planar anisotropy in the [110] direction is added into the Hamiltonian. D_i represents the magnitude of the magnetic anisotropy. In Eq. (1), $\boldsymbol{e}_i$ is a unit vector and represents the direction of magnetic anisotropy. For the sake of effectively pinning the spiral axis at the diagonal of ab plane, a large negative $D=-5$ is made. In addition, this planar anisotropy will lead to a strong spin coupling along the c axis, which is found to be essential for inducing electric polarization.[30] Here $\boldsymbol{h}$ is an external magnetic field including an extra factor $g\mu_{\rm B}$, and $\boldsymbol{h}_{\rm e}$ is an externally applied electric field along the c axis. The average magnetization is evaluated as

$$m=\frac{M}{N}=\frac{1}{N}\sum_i S_i, \tag{2}$$

where N is the total number of spin sites and M is the total magnetization.

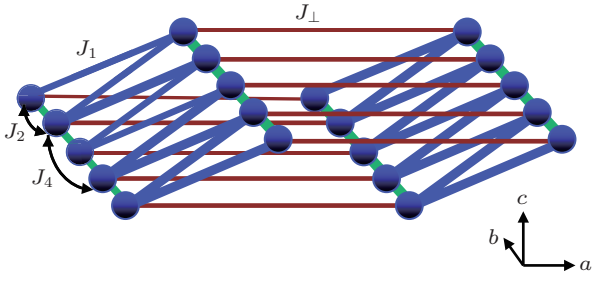


Fig. 1. (color online) Schematic plot of exchange interactions between magnetic Cu^{2+} ions in $LiCu_2O_2$.

According to the spin current model, or equivalently, the inverse Dzyaloshinskii–Moriya interaction, the polarization $\boldsymbol{P}_i$ induced by two neighboring spins ($\boldsymbol{S}_i$ and $\boldsymbol{S}_j$) is formulated as

$$\boldsymbol{P}_i=A\boldsymbol{e}_{ij}\times(\boldsymbol{S}_i\times\boldsymbol{S}_i), \tag{3}$$

where $\boldsymbol{e}_{ij}$ denotes the vector connecting the two sites of $\boldsymbol{S}_i$ and $\boldsymbol{S}_j$, namely in the direction of magnetic modulation vector along the b axis; A is a proportional constant determined by the spin-exchange and the spin–orbit interactions as well as the possible spin–lattice coupling term. It is assumed to be unity here. Thereby the third item in Hamiltonian Eq. (1) corresponds to electric energy.

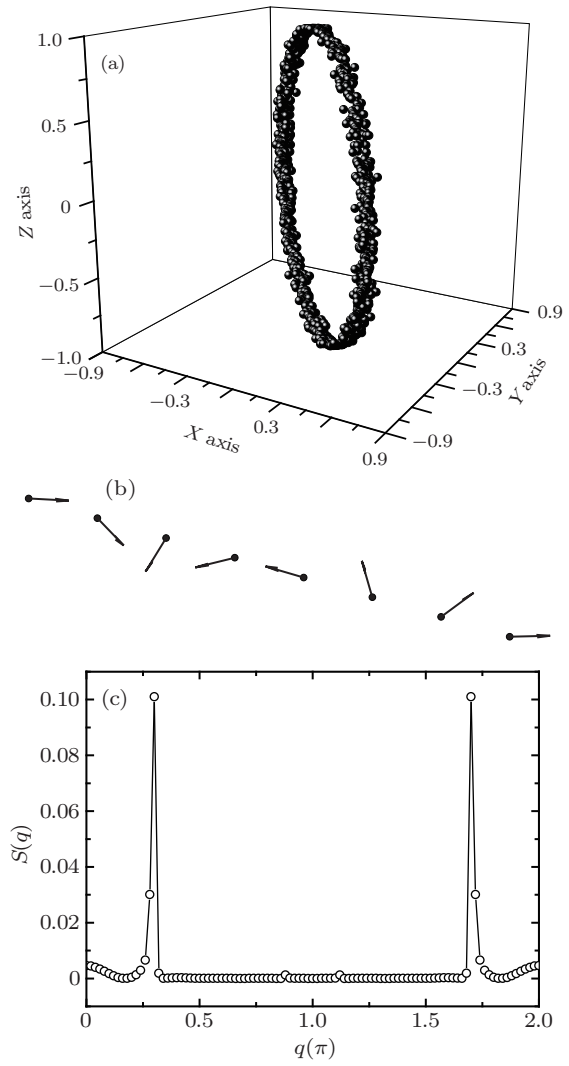


Fig. 2. (a) Snapshot for the coordinates of spins in all zigzag chains after the system has been polarized by a small electric field, (b) spin configurations for partial spins in a single chain, and (c) wave vector dependence of the spin structure factor.

The low-dimensional $LiCu_2O_2$ compound contains multifold spin interactions as shown in Fig. 1. It is well accepted that J_2 is ferromagnetic and J_4 is antiferromagnetic. The estimated ratio $|J_4/J_2|$ approximately varies from 0.5 to 0.65, which has been experimentally verified.[16,28] However, the relative magnitudes of interchain interactions J_1 and $J_\perp$ remain controversial. Based on inelastic neutron scattering experiments and spin wave theory, Masuda *et al.*[17,18] extracted

the relevant exchange constants presented in this spin system. Their data suggest a strong AF 'rung' interaction J_1 and a weak interchain coupling $J_\perp$, in contrary to the conclusion obtained by Drechsler *et al*.[13] In this paper, we adopt the suggestion of Masuda *et al*. Our previous study showed that this enhanced antiferromagnetic coupling J_1 would cause a drastic competition with the antiferromagnetic bond J_4 and lead to the modification of the modulated period of the incommensurate spiral order, and more importantly, the thermodynamic behaviors under this condition were in qualitatively good agreement with experimental results.[31] Therefore, according to the exchange parameters from neutron scattering and our previous investigation on this model, the parameters are as follows: $J_1 = 3.4$, $J_2 = -6.0$, $J_4 = 3.0$, and $J_\perp = 0.9$. One can note that the ratio $|J_4/J_2| = 0.5$ here is also in the range provided by the experiment.

Since $LiCu_2O_2$ compound possesses a weak $J_\perp$ interaction and the spiral order is formed along the chain, the number of the zigzag chains is fixed at $L' = 10$. For the length of a zigzag chain, we have carefully examined the size effects and find that $L = 100$ is large enough to eliminate it. Hence the lattice size in the present simulation is 100×10. It is assumed that a, b, and c axes are respectively, along the directions of [100], [010], and [001]. The standard Monte Carlo simulation is employed and performed on this $L \times L'$ lattice with periodic boundary conditions. The spin is updated according to the Metropolis algorithm. For each temperature T, the initial 5×10^4 Monte Carlo steps (MCS) are discarded for equilibration, and then the results are obtained by averaging 1.5×10^4 data. Each datum is collected at every 10 MCS.

The final results are obtained by averaging twenty independent data sets obtained by selecting different seeds for random number generation.

3. Results and discussion

Like the measurement process in experiment, the system is initially polarized by an electric field $\boldsymbol{h}_e = 0.5$ along the c axis, which is implemented under the condition of the zero magnetic field cooling (ZFC). When the system is polarized down to a very low temperature $T = 0.01$, a single magnetoelectric domain will be produced with magnetic modulation vector along the chain direction. Based on the cubic symmetry of magnetic structure, the spiral axes are along [110], $[\bar{1}\bar{1}0]$, $[1\bar{1}0]$, or $[\bar{1}10]$ with equal probability. The choice depends on the choice of parameters. In the present simulation, we have considered a large negative magnetic anisotropy $D = -5$ in the diagonal direction of ab plane, therefore the ground state consisting of the incommensurate spiral structure with its spiral axis along the [110] direction can be expected. As shown in Fig. 2(a), if all the sites in the chains are moved to one point, the spin vectors would almost lie on the secondary diagonal plane of the cube and form an ellipsoidal shape, which well reproduces the ground state configuration described in Refs. [21] and [22]. In Fig. 2(b), the snapshot of the spin configuration in a single chain also demonstrates a vivid picture of the low-temperature spiral magnetic order.

In order to analyze the spiral spin state accurately, the spin structure factor is evaluated along the spin chain via a Fourier transform of the spin correlation function written as[32]

$$S(q) = \sum_{i,r} \cos(q \cdot r) \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+r} \rangle , \tag{4}$$

where q is the wave vector and r is the distance between two nearest-neighbor correlated spins. The spin structure factor, which can be measured by a diffuse neutron-scattering experiment, provides the direct information about the spin ordering at finite temperature through the position of its maximum. In Fig. 2(c), one can see two sharp characteristic peaks appearing at $q = 0.3\pi$ and 1.7π that is its equivalent position, which confirms a good spiral spin order along the b axis as shown in Figs. 2(a) and 2(b). Furthermore, the non-zero value at $q = 0$ indicates the weak ferromagnetism induced by the strong 'rung' coupling J_1.

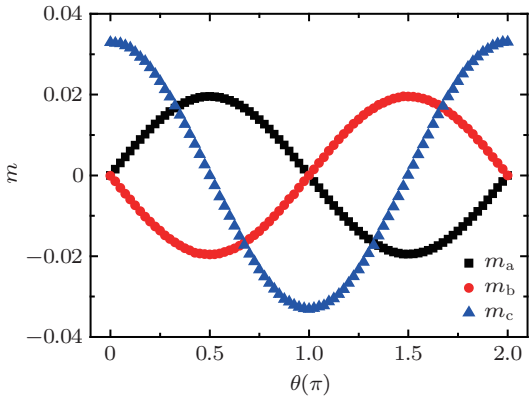


Fig. 3. (color online) Angle-dependent magnetization under $|\boldsymbol{h}| = 1$.

After the polarization procedure, $\boldsymbol{h}_e$ is removed and the low temperature calculations of the macroscopic physical properties in a rotating magnetic field $\boldsymbol{h}$ are executed, where $\boldsymbol{h}$ rotates in a clockwise direction from the c axis and in the plane perpendicular to the spiral axis. In $LiCu_2O_2$, the anisotropic magnetic phase transition has been observed in the magnetic susceptibility for magnetic field $\boldsymbol{h} \parallel ab$ and $\boldsymbol{h} \parallel c$, respectively. In order to study the anisotropic behavior and compare with the experimental results, we explore the angle dependence of magnetization for $\boldsymbol{h}$ rotating in the plane where the spiral spin order lies. It is worth mentioning that it yields the identical results whether $\boldsymbol{h}$ rotates in a clockwise direction or in a counterclockwise direction. As shown in Fig. 3, all the three components of magnetization follow $\boldsymbol{h}$ all the time, presenting a well-defined harmonic wave-form. Despite an exchange

anisotropy ($\Delta = 0.7$) in the Hamiltonian (1), the amplitude of m_c curve still exceeds those of m_a and m_b curves. This can be used to account for the strong planar anisotropy in the [110] direction, which ensures the dominated coupling between spins along the c axis. Furthermore, m_a and m_b curves show almost no distinction except for the sign of the amplitude, which is consistent with recent report on untwinned crystals.[15]

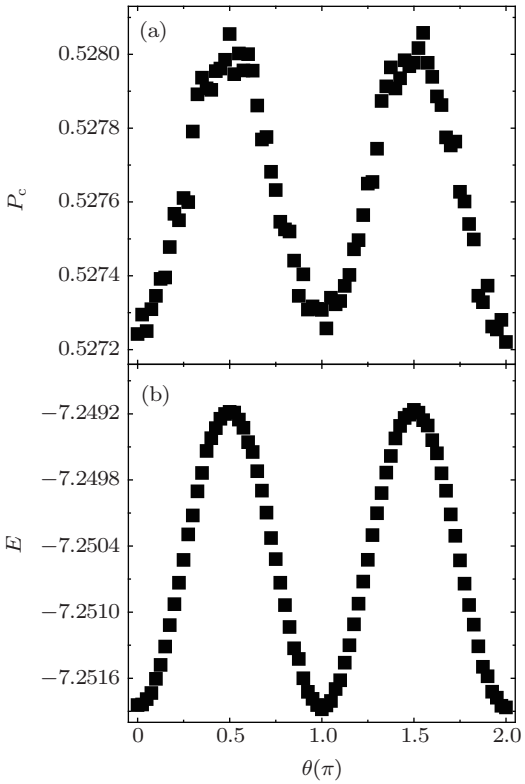


Fig. 4. Angle-dependent (a) polarization P_c and (b) energy E under $|\boldsymbol{h}| = 1$.

Owing to the magnetic origin of ferroelectricity, the ferroelectric behaviors in these materials are highly sensitive to the magnetic field. Figure 4(a) displays the angle dependence of the electric polarization P_c under a rotation of $\boldsymbol{h}$. According to the spin current theory, the polarization in this model should occur along the c axis. Our simulation results also confirm this deduction, which is in agreement with the experimental report.[11] As seen in this figure, a hump structure appears in the polarization curve. At $\theta = 0$ or π where $\boldsymbol{h}$ is aligned parallel or antiparallel to the c axis, P_c presents a narrow valley peak, reaching its minimum value. At $\theta = \pi/2$ or $3\pi/2$, namely $\boldsymbol{h}$ is along the secondary diagonal of ab plane, P_c arrives at its maximum, exhibiting a round peak. The variation tendency of P_c in a rating magnetic field can be understood in the following way. From Eq. (3), it is known that the magnitude of the polarization is proportional to $S_i S_j \sin\varphi$ and reaches its maximum at $\varphi = \pi/2$, where φ denotes the pitch angle between nearest-neighbor spin along the chain. Thus, the results here indicate that the pitch angle increases with $\boldsymbol{h}$ sweeping from zero to $\pi/2$ and decreases with $\boldsymbol{h}$ sweeping from $\pi/2$ to π. When $\boldsymbol{h}$ sweeps from π to 2π, the behaviors depicted above are repeated again. The repeated behavior observed in P_c also demonstrates the fact that the exotic magnetic structure of $LiCu_2O_2$ at low temperature is symmetric with respect to c and diagonal axes as argued in Ref. [23]. In addition, from the weak P_c, it can be conjectured that the pitch angle still maintains much less than $\pi/2$ in the whole process. Moreover, the variation in amplitude of P_c is also very small because of a weak magnetic field applied. The angle-dependent energy E as shown in Fig. 4(b) also verifies the analysis above. It presents the analogy picture with that of P_c. The minimum of E appears at the maximum of pitch angle due to the dominated contribution from the ferromagnetic exchange energy between the nearest-neighbor spins in chain.

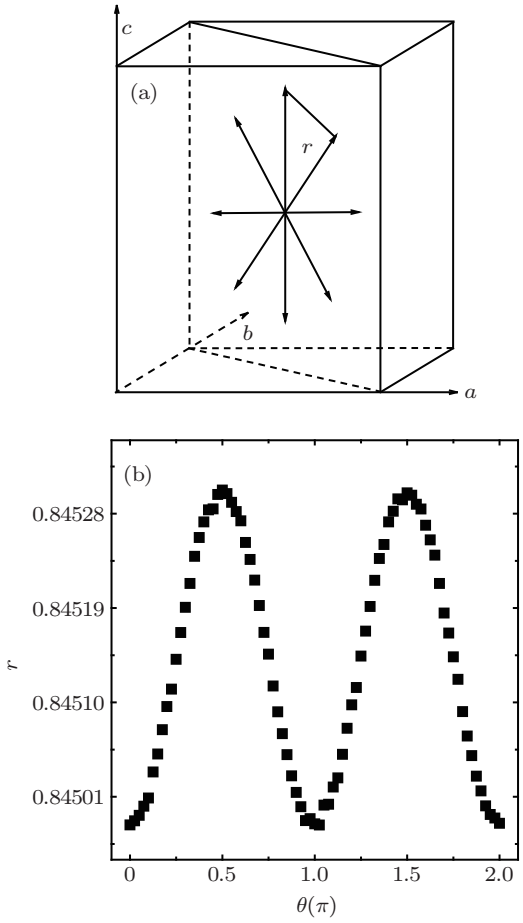


Fig. 5. (a) Sketch map of the spin structure in ground state, where the distance between the nearest-neighbor spin vectors along the chain is denoted as r and (b) angle-dependent r under $|\boldsymbol{h}| = 1$.

In order to obtain a thorough comprehension of the magnetoelectric coupling and microscopic spin structure, the distance r between two nearest-neighbor spin vectors is investigated. The sketch map in Fig. 5(a) illustrates the meaning of the physical quantity r. It reflects the magnitude of the angle included between two adjacent spins along the chain, which is closely related to the value of the polarization in the system.

According to the initial value of r in Fig. 5(b), we can obtain a rough estimation of the pitch angle with $\boldsymbol{h}$ applied along the c axis. Its value is around $\pi/4$. It can also be found that when $\boldsymbol{h}$ is far from the c axis, the pitch angle increases. However, when $\boldsymbol{h}$ is near the c axis, the variation of the pitch angle in an opposite way. Furthermore, the difference between the maximum and minimum pitch angles is no more than 0.1°. These behaviors indicate the small pitch angle of the spin vector in ground state, and consequently lead to a low-temperature weak electric polarization as shown in Fig. 4(a).

In addition to the anisotropic behaviors, the intriguing field responses are also detected in $LiCu_2O_2$. Since it is a complex magnetic-order-driven multiferroic compound, exploring the influences of various magnetic fields on the magnetoelectric effect will be an effective way to understand the underlying mechanism of magnetism inducing ferroelectricity. Figure 6 shows the variation tendencies of the magnetization and polarization under different magnetic fields. As expected, the magnetization is strengthened with the increase of $\boldsymbol{h}$ as shown in Figs. 6(a) and 6(b). One can see that the increment of m_c exceeds that of m_a due to the strong spin coupling of z component, which is also reflected in the P_c curve. In Fig. 6(c), the polarization P_c exhibits large-amplititude variations under a big value of $\boldsymbol{h}$. And it is greatly smoothed compared with the profile in Fig. 4(a), demonstrating the intimate correlation between the magnetism and ferroelectricity. It reveals that strong magnetic field will be helpful to stabilize the polarization in a frustrated system. However, as $\boldsymbol{h}$ increases, P_c is suppressed for $\boldsymbol{h}$ parallel to the c axis or the secondary diagonal of ab plane (see $q = 0$ and $\pi/2$), which is contrary to the experimental results.[11] With regards to this discrepancy, the reasons can be attributed to the following points. Based on the calculation of distance r in Fig. 5(b), it is known that the pitch angle in ground state is much less than $\pi/2$. When $\boldsymbol{h}$ is parallel to c axis or secondary diagonal of ab plane, the spin orientation will tend to the direction of the magnetic field. And this tendency will be augmented by the enhancement of $\boldsymbol{h}$. As a direct consequence of this variation, the pitch angle decreases, and eventually leads to a decrease of P_c, which is proportional to the sine of pitch angle.

In fact, if the ratio of $|J_4/J_2|$ is adjusted to be much greater than 1, the pitch angle larger than $\pi/2$ can be expected in ground state, thereby the variation trends of polarization under different magnetic fields will be similar to experimental observations. But it violates the fact that the real value of the pitch angle in $LiCu_2O_2$ is 62.6°.[11] This contradiction manifests the significant effect of quantum fluctuation on the classical incommensurate magnetism, although three-dimensional (3D) interactions in real quasi-1D materials tend to suppress the quantum fluctuations. It also implies that the discrepancy between our simulation results and experimental results are mainly ascribed to the neglect of the quantum effects. Therefore, a deep investigation into the competition between the quantum commensurate and classical incommensurate correlations is very important and meaningful for understanding the multiferroic nature. In the future work, we will concentrate on this aspect.

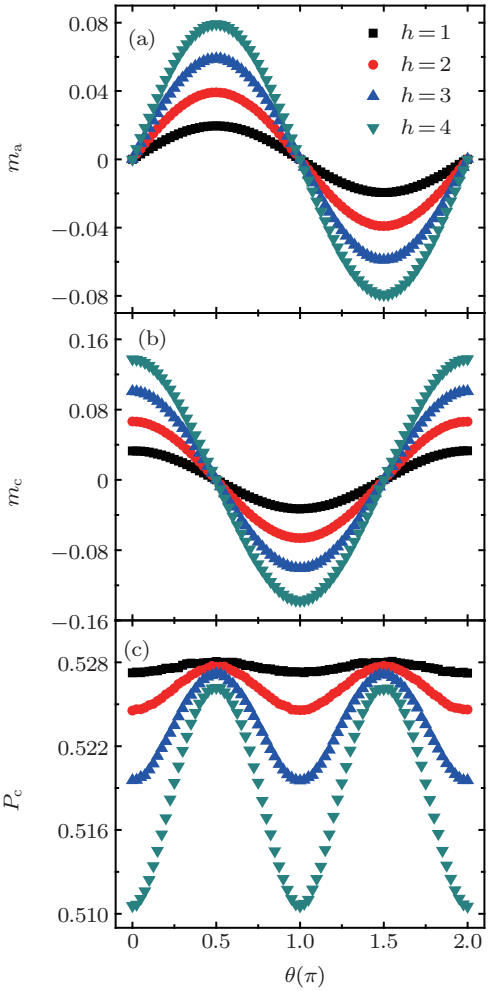


Fig. 6. (color online) Angle-dependent (a) magnetization m_a, (b) magnetization m_c, and (c) polarization P_c under different magnitudes of $\boldsymbol{h}$.

4. Summary

In summary, the ellipsoidal spiral structure in ground state is produced by performing Monte Carlo simulation. By computing the spin structure factor, the low-temperature spiral order of the system is confirmed. Focusing on magnetoelectric phenomenon, we investigate the variations of magnetic and ferroelectric behaviors under rotating $\boldsymbol{h}$. The anisotropic responses to the magnetization and polarization indicate the strong coupling between spins along the c axis, which qualitatively accords with the experimental results. In addition, we also find that the strong magnetic field can stabilize the ferroelectricity in frustrated multiferroic compounds, demonstrating the intrinsic connection between magnetism and ferroelectricity in this class of materials. Last but not least, the

responses to the polarization under different magnetic fields, which are contrary to the experimental observations, are discussed. The analysis implies the non-neglectable effects of the quantum fluctuations in $LiCu_2O_2$ compound.

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