Dynamical magnetoelectric effects induced by the Dzyaloshinskii-Moriya interaction in multiferroics

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Abstract – We study the dynamical interplay between ferroelectricity and magnetism in a multiferroic with a helical magnetic order. We show that the dynamical exchange-striction induces a biquadratic interaction between the spins and transverse phonons resulting in quantum fluctuations of the spontaneous ferroelectric polarization \( P \) in the ferroelectric phase. The hybridization between the spin wave and the fluctuation of the electric polarization leads to low-lying transverse phonon modes. Those are perpendicular to \( P \) and to the helical spins at small wave vector but then turn parallel to \( P \) at a wave vector close to the magnetic modulation vector. For helical magnetic structure, the spin chirality which determines the direction of \( P \), also possesses a long-range order. Due to the dynamical Dzyaloshinskii-Moriya interaction, the spin chirality is strongly coupled to the spin fluctuation which implies an on-site inversion of the spin chirality in the ordered spin-(1/2) system and results in a finite scattering intensity of polarized neutrons from a cycloidal helimagnet.

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Introduction. – Multiferroic compounds in which the electric and the magnetic order coexist are in the focus of current research. Of particular interest is the possibility of controlling the direction of spontaneous electric polarization by a magnetic field [1]. Beside this technological relevance of such a strong interplay between the magnetic and electric order parameters, it is also fundamentally interesting to understand how such a coupling comes about and what is the microscopic mechanism behind the magnetoelectric (ME) coupling in multiferroics.

Among the family of multiferroics, two different types of manganites, RMnO\(_3\) (R = Gd, Tb, Dy) [2] and RMn\(_2\)O\(_5\) (R = rare earth, Tb, Y, Bi) [3] play a special role as they exhibit different microscopic ME coupling mechanisms. In the perovskite multiferroic RMnO\(_3\), it is shown experimentally that the onset of helical magnetic order induces spontaneous ferroelectric (FE) polarization, which can be well described by the so-called spin-current model [4]. In these compounds, the spin-orbit coupling within the \( d(p)\)-orbitals of magnetic (oxygen) ions produces an electric polarization of the form [5], \( P \propto S_i \times S_j \). Non-collinearity of the spins \( S_j \) (at sites \( j \)) is strictly required in the spin-current model. In the main FE phase of RMn\(_2\)O\(_5\) the electric dipole moments are directed along the \( b \)-axis, the spins however are almost collinear in the \( ab \)-plane which indicates that a spin-orbit–driven mechanism cannot be the primary source for FE in RMn\(_2\)O\(_5\). As an alternative explanation, the (super)exchange-striction [6,7] is believed to be the origin of ferroelectricity in RMn\(_2\)O\(_5\), \( P \propto S_i \cdot S_j \). On the other hand, inspecting carefully the dynamical properties of the multiferroics, we find both, the antisymmetric Dzyaloshinskii-Moriya (DM) interaction and the symmetric magnetostriction play an essential role and need to be taken into account.

Based on the spin-current model, the dynamical properties of DM interaction were studied in refs. [8–10]. A collective ME excited mode, so-called electromagnon, was theoretically predicted. Moreover, it was found [8] that this new low-lying mode is perpendicular to both the spontaneous polarization and the helical wave vector. It corresponds to a rotation of the spin plane with respect to the axis of the helical wave vector, the rotation frequency is \( \sqrt{SJD} \), where \( S \) is the spin value, \( J \) is the exchange coupling, and \( D \) is the magnetic anisotropy. Electromagnons have been detected in RMnO\(_3\) [11] and Eu\(_{0.75}\)Y\(_{0.25}\)MnO\(_3\) [12], seemingly consistent with the theoretical analysis. However, a detailed study of the terahertz spectrum of Eu\(_{1-x}\)Y\(_x\)MnO\(_3\) [13] revealed that

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infrared-absorption along the spontaneous polarization direction is also observable, which is not explained by theory.

In this paper we show that the dynamical exchange striction intrinsically generates bi-quadratic coupling between the spin and the transverse acoustic (TA) phonons, \( \sim (\mathbf{u}_i^j - \mathbf{u}_j^i) \langle S_i \cdot S_j \rangle \), where \( \mathbf{u}_i^j \) is a transverse displacement at site \( j \). This dynamical coupling does not contribute any additional static electric polarization but induces the fluctuation of the electric dipole moment due to the low-frequency excitation modes of the TA phonons. One thus has a mode mixing behavior and the polarization correlation function follows the soft magnetic behavior of the system parallel to the uniform electric polarization \( \mathbf{P} \). Moreover, in \( S = 1/2 \) multiferroics, the spin fluctuation is accompanied with an inversion of the onsite electric dipole moment according to the spin-current fluctuation is accompanied with an inversion of the spin-current model. The hybridization between phonons and spins results also in a large quantum fluctuation of the spin chirality which allows for a finite differential scattering intensity of polarized neutrons from a cycloidal magnet LiCu2O2 [14].

\[ H = H_s + H_{DM} + H_p, \]
\[ H_s = \sum_{(ij)_{nn}} J_i(r_i - r_j) S_i \cdot S_j + \sum_{(lm)_{nnn}} J_2(r_l - r_m) S_l \cdot S_m, \]
\[ H_{DM} = -\lambda \sum_i \mathbf{u}_i \cdot [\hat{e}_z \times (S_i \times S_{i+1})], \]
\[ H_p = \frac{k}{2} \sum_i \mathbf{u}_i^2 + \frac{1}{2M} \sum_i \mathbf{p}_i^2, \]

where the notation \((ij)_{nn}\) indicates nearest-neighboring (nn) \( i \) and \( j \), and \((lm)_{nnn}\) corresponds the next-nearest-neighboring (nnn) \( l \) and \( m \). The competition between the nn ferromagnetic interaction (\( J_1 < 0 \)) and the nnn antiferromagnetic interaction (\( J_2 > 0 \)) leads to magnetic frustration and to a spiral spin ordering with the wave vector \( Q = -J_1/4J_2 \) [15]. \( H_p \) is an optical phonon model. The spin-phonon interaction \( H_{DM} \) originates from a spin-orbital (DM) coupling and breaks the inversion symmetry along the chain. Minimizing the energy yields the condition of the atomic displacement and the local spin-configuration

\[ \mathbf{u}_i = \frac{\lambda}{k} \hat{e}_z \times (S_i \times S_{i+1}). \]

Particularly, if the \( xx \) helical spins along the chain, \( i.e. S_i = S(\sin Q, 0, \cos Q) \), eq. (2) leads to a macroscopic uniform lattice displacements along the \( x \) direction

\[ \mathbf{u}_i^x = -\frac{\omega_0^x}{k} \sin Q \hat{e}_z. \]

In the helical spin-ordering phase \( \mathbf{u}_x \) cannot be softened through the hybridization between the TO phonons and the magnons because \( k/M \gg J_S \). The spontaneous FE polarization \( \mathbf{P}_S \) is frozen at \(-\mathbf{u}_0^x\). The fluctuation \( \delta P_x \) can therefore be neglected [8]. However, accounting for the superexchange striction, the TA phonon mode emerges. As is well known, TA phonons possess a low-frequency mode at long wavelength, \( \omega^2_{TA}(q) \propto q^2 \), which gives rise to the fluctuation of the FE polarization. Such polarization fluctuations are hybridized with the spin bosons and soften thus the transverse phonon behavior.

Existing experimental data suggests that the exchange energy \( J \) falls off as a power law with the separation of the magnetic ions

\[ J_{1,2}(r_i - r_j) = J_{1,2}[R_i^z + \mathbf{u}_i] - [R_j^z + \mathbf{u}_j] |^{\gamma_{1,2}}, \]

where \( \gamma \) is in the range 6–14 [16]. \( R_i^z \) is the bare value of the position of the atom at site \( i \), and \( [R_i^z - R_j^z] \) determines the lattice constant \( a \) (set here to 1). \( \mathbf{u}_i \) is the displacements of site \( i \). Generally, \( |\mathbf{u}_i| \) is small and does not destroy the lattice structure, \((|\mathbf{u}_i|/a \sim 10^{-3})\). We inspect the dominant term for \( J \) in the following two case.

\[ \sum_{i,j} \mathbf{e}_i \cdot (\mathbf{u}_i^z - \mathbf{u}_j^z) \]

Longitudinal phonons. When the atoms are displaced along the chain one finds

\[ J_{1,2}(r_i - r_j) \approx J_{1,2} \left[ 1 - \gamma_{1,2} \mathbf{e}_z \cdot (\mathbf{u}_i^z - \mathbf{u}_j^z) \right], \]

with \( \mathbf{e}_z \) being the unit vector connecting two sites \( i \) and \( j \). A trilinear coupling between the phonon and spin is induced. One can easily check that \( (\mathbf{u}_i^z) = 0 \) because of the local rotational symmetry of the spin-spin correlation \( \langle S_i \cdot S_j \rangle \) in the helically ordered phase. Since \( \mathbf{u}_i^z \) is not involved in the DM interaction we do not consider it in the following discussion.

Transverse phonons. For atomic displacements perpendicular to the chain, \( \mathbf{u}_i^t \cdot \mathbf{e}_z = 0 \) we find, for \( J_{1,2}(r_i - r_j) \),

\[ J_{1,2}(r_i - r_j) \approx J_{1,2} \left[ 1 - \frac{\gamma_{1,2}}{2} (\mathbf{u}_i^t - \mathbf{u}_j^t)^2 \right], \]

which gives a TA phonon mode coupled to the spins with the bi-quadratic interaction \(-\gamma_{1,2} J_{1,2} (\mathbf{u}_i^t - \mathbf{u}_j^t)^2\) \( \langle S_i \cdot S_j \rangle \). The dynamical exchange striction will harden the frequency of the transverse phonon mode as

\[ \omega_0^2 = \omega_0^2(1 + F_t), \]

where \( \omega_0^2 = k/M \) and \( F_t = -\gamma_1 J_1 \langle S_i \cdot S_j \rangle - \gamma_2 J_2 \langle S_i \cdot S_m \rangle \)/\( k \). As the temperature decreases below the transition temperature for magnetic order \( T_N \) the spin-spin

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correlation function $\langle S_i S_j \rangle$ increases and so does the phonon frequency. Lowering further the temperature to the FE transition temperature $T_{FE}$, an additional frequency hardening occurs due to the dynamical DM interaction [8,10]. The complete scenario is thus that the phonon frequency hardens at two onsets at $T_N$ and $T_{FE}$, a conclusion consistent with the experimental observation for Eu$_{75}$Y$_{25}$MnO$_3$ [12]. Assuming $k \sim 1$ eV/Å$^2$ and $JS^2 \sim 10$ meV [8] the frequency hardening can be estimated to be $\delta \omega/\omega_0 \approx 1\%$, which is in good agreement with the experimental data [12]. Phenomenologically, the exchange restriction suggests that in a Ginzburg-Landau (GL) theory for the coupling between the spin $S$ and the transverse electric dipole $P_z$ terms of the form $-\alpha S^2 P_z^2$ appear. As a consequence, $P_z$ and $S$ condense at the same temperature due to the strong spin-lattice coupling $\alpha \sim J$ and the two transition temperatures merge, a conclusion which is in line with the experimental observations in YMnO$_3$ [17]. There the electric-dipole moment $\Delta P_z$, which is along the $z$-direction, obeys the same temperature dependence as the magnetic moment that is aligned in the $ab$-plane with $120^\circ$ structure below 80 K.

**Electromagnon.** We split the atomic displacements into two parts: i) the statical part $\mathbf{u}_i = (u_{0x}, 0, 0)$ driven by the DM interaction, and ii) the dynamical part $\delta \mathbf{u}_i = (-\delta u_{Q x}^i, \delta u_{Q y}^i, 0)$ induced by the exchange striction. As the softness of the system is due to the magnetic part, we concentrate at first on the spin excitations. For the $zx$ helical spins, it is convenient to rotate the spins locally (at each site) along its classical direction ($\tilde{S}_i^x$):

$$S_i^x = \tilde{S}_i^x \cos iQ + \tilde{S}_i^z \sin iQ,$$

$$S_i^y = \tilde{S}_i^y,$$

$$S_i^z = -\tilde{S}_i^z \sin iQ + \tilde{S}_i^x \cos iQ.$$  

Disregarding the high-order terms of the interplay between the spins and the dynamical part of lattice displacements, i.e. using the standard linear-spin-wave approximation, we have

$$H = E_0 + \sum_q A(q)\tilde{S}_q^- S_i^+ + B(q)(\tilde{S}_q^- S_i^- + \tilde{S}_i^+ S_q^+),$$  

where $E_0 = N[J_1 S^2 \cos Q + J_2 S^2 \cos 2Q - \frac{k}{2}|u_{0x}|^2]$, and

$$A(q) = -J_1[\cos Q + \frac{1}{2}(1 + \cos Q) \cos q]$$

$$-J_2[\cos 2Q + \frac{1}{2}(1 + \cos 2Q) \cos 2q]$$

$$+ \frac{\lambda^2 S^2 \sin^2 Q}{2k}(2 - \cos q),$$

$$B(q) = \frac{J_1}{4}(1 - \cos Q) \cos q + \frac{J_2}{4}(1 - \cos 2Q) \cos 2q$$

$$- \frac{\lambda^2 S^2 \sin^2 Q}{4k} \cos q.$$  

$H$ can be easily diagonalized by a Bogoliubov transformation.

The energy dispersion of the spin excitation reads

$$\omega_s(q) = \left[A(q)^2 - (2B(q))^2\right]^{1/2}.$$  

The effective spin anisotropy introduced by the spin-phonon (DM) interaction results in an energy gap of the spin-wave spectrum for non-collinear spin ordering, i.e. $\omega_s(q = Q) \neq 0$, if $Q \neq 0$ or $\pi$. One can see in the further discussion that the spin fluctuations with the wave vector $q \approx Q$ are important in connection with the magnetic softening of the transverse phonons.

Now we turn our attention to the dynamical spin-phonon interaction. Retaining terms up to the second order in the quantum fluctuation, the spin-current model delivers the following coupling terms:

$$\tilde{H}_{DM} = -\lambda S \cos Q \sum_i \delta \mathbf{u}_i^x (\tilde{S}_{i+1}^x - \tilde{S}_i^x)$$

$$-\lambda S \sum_i \delta \mathbf{u}_i^y \cos Q \cos Q_{i+1} - \tilde{S}_{i+1}^y \cos Q_i)\cos Q_i + 1)$$

$$-\lambda S \sum_q \delta \mathbf{u}_q^y \cos Q (e^{iQ} - e^{i(q \pm Q)})/2,$$

where $\delta \mathbf{u}_q^y$ is hybridized with the spin at $q \pm Q$, but $\delta \mathbf{u}_q^x$ is coupled to $S^2$ at $q$. As expected, $\delta \mathbf{u}_q^y$ has the same long-wavelength behavior as the magnons. No static displacement exists along the $x$-direction, i.e. $\delta \mathbf{u}_q^x = 0$. On the other hand, a uniform lattice deformation along the $y$-direction ($\delta \mathbf{u}_q^y \neq 0$) may occur due to the hybridization between the electric polarization and the spin ordering [8]. After some algebra, we find for the polarization correlation functions

$$\langle \langle \delta \mathbf{u}_q^y | \delta \mathbf{u}_{q'}^y \rangle \rangle = \frac{\omega^2 - \omega_q^2}{M[\omega^2 - \omega_q^2 + \omega_{q'}^2 + \omega_{q'+\pm Q}^2]},$$

$$\langle \langle \delta \mathbf{u}_q^y | \delta \mathbf{u}_{q'}^y \rangle \rangle = \frac{1}{M[\omega^2 - \omega_q^2 + \lambda^2 S^2 \cos^2 Q / 2k \sum_{q'=q\pm Q} G_s(q')]}.$$
and $\omega_{TA}(Q) \neq 0$ both the symmetric and antisymmetric magnetoelastic interaction respond to the fluctuations of the polarization. Especially, in the direction parallel to the FE polarization $P_z$, there is a low frequency range around $\omega_c \equiv \omega_4(Q)$, where $\omega_c$ couples resonantly to light even if $D = 0$. For finite $D$, assuming $D \gg \hbar^2 S^2/k$ we observe nearly the same low-frequency behavior of the polarization correlation functions $\omega_c \approx \sqrt{JSD} \approx \omega_4$. These conclusions are also qualitatively consistent with experiment observations (fig. 8 in ref. [13]).

Spin-flip. – Recently, LiCu$_2$O$_2$ ($S = 1/2$) has been found to be ferroelectric in the bc-spiral state at low temperatures [14]. In contrast to large spin multiferroics, in spin-(1/2) magnet the spin fluctuations may spontaneously reverse the local spin. According the spin-current model eq. (2) the direction of the on-site electric-dipole moment can also be completely reversed by the spin fluctuations. Large quantum fluctuations of the FE polarization $\delta P = -2\mu_0 \bar{c}$ is induced by the hybridization between the phonon and the spin.

Defining the vector of spin chirality as the average of the outer product of two adjacent spins $\hat{c}_i = (s_i \times s_{i+1})/(|s_i \times s_{i+1}|)$, in the RMnO$_3$-type multiferroics the direction of electric polarization is determined by the spin chirality. Reversing the direction of electric polarization does also reverse $\hat{c}_i$. Clearly, the spin chirality $\hat{c}_i$ has only two eigenvalues, +1 and −1, and possesses long-range ferromagnetic order in the FE phase. Thus, $\hat{c}_i$ can be simply treated as the Pauli operator. According to the dynamical exchange striction eq. (5) the interaction term involving the spin chirality has the structure $\sim -J_z(\hat{c}) \cdot \hat{c}_{i+1}$. The $x$-component of the spin chirality operator $\hat{c}_i^x$ acts as a direction reversal operator which can be traced back to the quantum fluctuation of the FE polarization. Considering the dynamical DM interaction eq. (14), the coupling term between spin and spin chirality in the spin-(1/2) multiferroics is given by $\sum_i \hat{c}_i^x (\hat{c}_{i+1}^x - \hat{c}_i^x) = \sum_i \hat{c}_i^x (\hat{c}_{i+1}^x - \hat{c}_i^x)$, which indicates that when the spin at site $i$ is flipped, $\hat{s}_i \rightarrow -\hat{s}_i$, the directions of spin chirality $\hat{c}_i$ and $\hat{c}_{i-1}$ are also reversed, an observation consistent with the spin-current model and with the definition of spin chirality.

For the one-dimensional spin-(1/2) chain, the quantum model predicts a gapped spin-liquid state in the range of the frustration exchange parameters in LiCu$_2$O$_2$. The very existence of the magnetic helix state suggests that the quantum fluctuations is significantly suppressed and the spins tend to recover a semiclassical behavior. In the ground state of the spin system all spins point along their corresponding classical directions as in NaCu$_2$O$_2$, where a $J_1 - J_2$ spin model provides a good description of the helix state [15]. So the spin interaction can be simply given as $-J_z(\hat{c}) \hat{s}_i \cdot \hat{s}_j$, where $Q$ is taken as the pitch angle along the chain.

Based on the above arguments an effective model that describes the interplay between the helical spin and spin chirality has the form

$$H_{sc} = -\sum_{i,j} (J_s \hat{s}_i \cdot \hat{s}_j + J_c \hat{c}_i \cdot \hat{c}_j) - \gamma \sum_i \hat{c}_i^x (\hat{c}_{i+1}^x - \hat{c}_i^x).$$

(15)

The Hilbert space can be considered as the tensor product space $|i\rangle \rightarrow |s_i\rangle \otimes |\hat{c}_i\rangle$. The ground state of $H_{sc}$ possesses the ferromagnetic order both for $\hat{s}$ and $\hat{c}$, $\hat{c}_i = |\hat{c}_i\rangle \otimes |\hat{c}_i\rangle$. Now let us consider the effect of the quantum fluctuation. If the spin at site $i$ is flipped, we have $|s_i\rangle = \hat{c}_i^x |s_i\rangle \otimes |\hat{c}_i\rangle$, which is the ground state with the spin at site $i$ being flipped. Noting that $\hat{c}_i^x |s_i\rangle = |s_i\rangle \otimes |\hat{c}_i\rangle$, $|\hat{c}_i\rangle \otimes |\hat{c}_i\rangle$, $|\hat{c}_i^2\rangle |s_i\rangle = |s_i\rangle$, we apply $H_{sc}$ to the state $|s_i\rangle |\hat{c}_i\rangle$ we find

$$H_{sc} |s_i\rangle |\hat{c}_i\rangle = \left[ E_0(q) + E_{sp} \begin{bmatrix} \cos \theta & \sin \theta \\ \sin \theta & -\cos \theta \end{bmatrix} \right] |s_i\rangle |\hat{c}_i\rangle,$$

where the state $|s_i\rangle |\hat{c}_i\rangle$ is essentially a flipped spin (spin chirality) delocalized across all the lattice. $E_0 = -NJ_s s^2 - NJ_c c^2 + |J_s(q) + J_c(q)|^2/2$, $J_s(q) = 2\pi J_s(1 - \cos q)$, $J_c(q) = 2\pi J_c(1 - \cos q)$, and $E_{sp} = \sqrt{[J_s(q) - J_c(q)]^2/2 + \gamma(1 - \cos q)}$, $\gamma(1 - \cos q)$, $\cos \theta = (J_s(p) - J_c(p))/E_{sp}$, and $\sin \theta = \gamma(p)/E_{sp}$. Applying the rotation $|\hat{c}_i\rangle = \begin{pmatrix} \cos \theta/2 & \sin \theta/2 \\ -\sin \theta/2 & \cos \theta/2 \end{pmatrix} |\hat{c}_i\rangle$, the Hamiltonian is brought in the diagonal form

$$H_{sc} = \sum_q (E_0(q) + E_{sp}) |s_q\rangle + \sum_q (E_0(q) - E_{sp}) |\hat{c}_q\rangle.$$

Due to the spin-phonon coupling the spin and spin chirality excitations are mixed. Two separated channels are identified: the spin channel $|s_q\rangle$ and the phonon channel $|\hat{c}_q\rangle$. In each channel, we have

$$\langle \hat{s} \rangle + \langle \hat{c} \rangle = 1.$$  

(16)

Generally, the expected value of $\hat{c}$ is less then one due hybridization with spin excitations. A non-unitary $\hat{c}$ is the origin for a finite-scattering intensity of polarized neutrons. For the cycloidal helimagnet we have [18]

$$\langle \hat{c} \rangle = I_{on} - I_{off} \over I_{on} + I_{off},$$  

(17)

where $I_{off}(I_{off})$ is the reflection intensity of polarized neutrons parallel (antiparallel) to the scattering vector. On the basis of the experimental data for LiCu$_2$O$_2$ [14] we infer $\langle \hat{c} \rangle \approx 0.3$. On the other hand, the magnitude of the ordered moment per magnetic copper site is 0.56 $\mu_B$ [15]. Together with the typical $g$-factor for Cu$^{2+}$ in a square-planar geometry ($g \approx 2$) from eq. (16) we conclude that $\langle \hat{c} \rangle = 0.44$, which is consistent with the previous estimated value.

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Summarizing, both the (symmetric) exchange striction and (antisymmetric) DM interaction affect dynamically the magnetoelastic coupling in multiferroics. At a small wave vector, the DM interaction determines the low-frequency behavior of the phonons. For a wave vector close to that of the magnetically modulated structure, the exchange striction induces fluctuations in the FE polarization, and additional low-lying mode parallel to the FE polarization emerges. For spin-\(\frac{1}{2}\) multiferroics, the effect of the quantum fluctuation is particularly large. The local polarization can be completely reversed by the spin fluctuation, and so does the direction of the on-site spin chirality. These findings are in line with experimental observations.

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