FERROMAGNETIC-FERROELECTRIC COMPOSITE 1D NANOSTRUCTURE IN THE PURSUIT OF MAGNON-PHONON INTERACTION

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ABSTRACT

Nanocomposites of linear chain of ferroelectric-ferromagnetic crystal structure is considered. It is analyzed theoretically in the motion equation method on the pursuit of magnonic excitations, lattice vibration excitations and their interactions leading to a new collective mode of excitations, the electromagnons. In this particular work, it is observed that the magnetizations and polarizations are tunable in a given temperature ranges for some specific values of the coupling order parameter.

KEY WORDS

Multiferroics, Electromagnons, ferroelectromagnets

1 Introduction

An ever-increasing interest of multi-functional materials both for research purposes and technological aspirations coiled the focus towards multiferroics. The theoretical backgrounds and experimental endeavors helped the field to envisage fast, huge memory and multifunctional devices in the electronics industry. Prominently, in the spintronics world where the spin and the charge degrees of freedom play independent roles so as to manipulate spin and electronic properties separately, these features of the composite multiferroics can be considered as an exclusive means to point out the coexistence of coupled order parameters¹,². The existence of simultaneous order parameters in some naturally occurring compounds is very weak to be used in wide ranges of applications and also very rare due to several constraints. The comparatively few number of single phase (naturally found) multiferroics put sever limitations to the researchers not to proceed further on the same track. However, the possibility of the coupled order parameters in synthesized compounds of different strengths have triggered the spirit onto composite ferroelectric-ferrromagnetic multiferroics providing magnetically ordered phases³ and electrically active states in addition to the elastic responses⁴,⁵. Studies focus on low dimensional thin film composites of magnetically ordered phases and ferroelectrically ordered phases in different configurations⁶,⁷,⁸. Most often, thin film hetrostructures and laminates on various substrates have been analyzed. Composite materials of both ferromagnetic and ferroelectric states have been examined experimentally and showed appreciable couplings of the two phases at room temperatures⁹,¹⁰.
Nanofibers have also shown enhanced magnetoelastic responses. Such composites have rich phase diagrams not only because they have the properties of their parent compounds but also coupled interactions between the magnetic and electric orders lead to additional functionalities such as the four state logic devices. The general principle underlying the quantum mechanical description of the interacting particles with the surrounding environment is on the pursuit of small perturbation that leads to elementary excitations. In the composite systems of ferroelectric-ferromagnetic nanostructured materials, coupling is inevitable so long as there are magnetostriction and electrostriction effects that can be shared from one component to the other and, consequently, modify the magnetizations and polarizations of the system. These responses are suggested to happen through strain mediation which can be strictly linkup with the phonon theoretical formalism on one hand and to the magnonic formalism on the other hand. To be specific, we consider a multiferroic composite of nanoparticles of linear chain consisting of both ferroelectric and ferromagnetic phases along the x-axis. The paper is organized in sections, the theoretical model helps to cast the specific Hamiltonian to be analyzed. The results and discussions include the brief outcomes to be compared with recent experimental findings. Finally, a short summary condenses the whole findings.

2 Theoretical model

The remarkable topic of research in magnetically induced multiferroicity is the appearance of magnon-phonon excitations in which an electric field induced spin excitations and magnetic field induced electric polarizations collectively called, "electromagnons" are immediate consequences of the interaction. In different literatures, magnetic properties and lattice vibrations have been considered separately and various model Hamiltonians were used. In this particular paper theoretical model Hamiltonian of composites of ferroelectric-ferromagnetic material is proposed. We consider the elementary quasi-particle excitations of composites of the two phases that can be described by making use of the motion equation method. To deal with this, we tried to model the crystal as a regular array of atoms in a particular direction, the positive x-axis. Each atom is expected to have a total spin while lattice vibrations is a natural outcome of every crystal. Hence, it is common to write the effective Hamiltonian in component formalism

\[ H = H_m + H_{ph} + H_{me} \]  

Here, \( H_m \) is the magnetic excitation energy component, \( H_{ph} \) is the energy associated with the lattice vibrations and, \( H_{me} \) corresponds to their interaction.

Collective magnetic excitation modes

In this linear chain of atoms the magnetic excitation modes are obtained for composite of ferroelectric-ferromagnetic crystal assuming the superexchange interaction between single domain magnetic structures on the pursuit of Heisenberg model. The effective magnetic and electric fields are also considered to be finite while the easy axis of magnetization is taken to be along the z-axis. However the polarization direction depends on the direction of the effective electric field. Assuming that the spin-electric field coupling has the most important contributions of the spin system to the total Hamiltonian arising from the interaction of spins with the external or intrinsic field (Zeeman interaction), the exchange and anisotropic interactions among spins we develop the Hamiltonian. The following Hamiltonian describes the effective magnetic interaction of spin frustrated nanostructural chains of atoms whose electrons are localized and
their ions are free to oscillate about equilibrium positions.

\[ H_m = \sum_i J_{ij} S_i \cdot S_j - g\mu_B H \sum_i S_{iz} - \mu E \sum_i S_{iz} + D \sum_i S_{iz}^2 \]

where \( S_i (S_j) \) is spin operator at sites \( i(j) \) and \( J_{ij} \) is the exchange coupling. \( \mu_B \) and \( D \) are the Bohr magneton and magnetic anisotropic while \( g \) is the gyromagnetic ratio. Whereas, \( H \) and \( E \) magnetic and electric fields and \( \mu \) describes the coupling strength between the spins and electric field. Thus, the interaction Hamiltonian through subsequent transformations and mathematical operations followed by suitable approximations to the bilinear order of spin operators become,

\[ H_m = \sum_k \hat{n}_k \omega_k \]

where

\[ \omega_k = -2JS(\gamma_k - 1) - 2DS + g\mu_B H + \mu E \]

For \( |k, \delta| \ll 1 \), long wavelength limit and cubic symmetry

\[ Z(1 - \gamma_k) \approx 1/2 \sum_\delta (k, \delta)^2 \approx 1/2 (k, a)^2 \]

So that, the excitation frequency of quasi-particles is

\[ \omega_k = JSa^2 k^2 - 2DS + g\mu_B H + \mu E \]

Defining \( \beta = JSa^2 \) and \( \gamma = -2DS + g\mu_B H + E \), the dispersion function of magnetic excitation is written as

\[ \omega_k = \beta k^2 + \gamma \]

**The lattice vibration excitation modes**

Electron clouds, despite the fact that they are bounded strongly in insulating ferroelectric-ferromagnetic multiferroic crystals, can polarize the nearby lattices. Therefore, it is very fundamental to deal with the magnetic excitations and lattice vibrations hybridized in a single crystal system as predicted that the Ferroelectric polarization direction is not merely on the spin configurations but also depends on ionic displacements from their centrosymmetric positions. We suggest that the enter-play of the electrostrictions and magnetostrictions arising from inhomogeneous crystal fields and external fields cooperatively can enhance the lattice vibration excitations which can be well described by harmonic oscillations to obtain the Hamiltonian. Lattice vibrations play very crucial roles in the properties of crystals due to the fact that it occurs at finite temperature in very quantized quasi-particle states and hence often necessary to study lattice distribution functions to understand crystal dynamics as well. We assume that in such composites of insulating materials, the lattice vibration is particularly the ion density oscillation in a relatively static electron cloud backgrounds, i.e the ion plasma oscillation where the driving force is the long range coulomb interaction. The predominating interaction is the electron cloud with the lattice ions and gives rise to induced polarization. This is based on the Jellium
oscillation theory that treats the ion density dynamics approximating the electronic contributions in a lesser extent. We begin with a simple one dimensional quantum mechanical model consisting of linear chain of atoms of finite length $L$, having $N$ ions of different masses $m_1$ and $m_2$ each interacting with the nearest neighbors. These ions execute simple harmonic oscillations (back and forth) about their equilibrium positions through a linear force constant, $c$. The equilibrium displacements of the masses $m_1$ and $m_2$ are $u_j$ and $u_i$ respectively. In a linear approximation, the restoring forces of the masses being expressed as

$$m_2 \frac{d^2 u_i}{dt^2} = c(u_j - u_i) + c(u_j - u_i)$$

Whereas,

$$m_1 \frac{d^2 v_j}{dt^2} = c(u_{i+1} - v_j) + c(u_i - v_j)$$

The nontrivial solutions for equations making use of the lattice displacement vectors of different amplitudes are proportional to $e^{-i\omega t}$, $v_j = v e^{i\omega t}$, lattice spacing $a$, and to the nearest neighbor interaction leads to eigen frequencies,

$$\omega^2_\pm = c\left\{ \frac{1}{m_1} + \frac{1}{m_2} \right\} \pm \sqrt{\left( \frac{1}{m_1} + \frac{1}{m_2} \right)^2 - \frac{2}{m_1 m_2} (1 - \cos qa)}$$

Here the optical mode frequency corresponds to

$$\omega^2_+ = c\left\{ \frac{1}{m_1} + \frac{1}{m_2} \right\} \pm \sqrt{\left( \frac{1}{m_1} + \frac{1}{m_2} \right)^2 - \frac{2}{m_1 m_2} (1 - \cos qa)}$$

With our usual assumptions of nearest-neighbor interactions and harmonic forces, the effective Hamiltonian of the linear chain in quantum viewpoint described as a sum of potential and kinetic energy terms can be expressed equivalently with the electrostatic energy at the specified optical frequency. Thus, the total energy on the long range coulomb interaction is

$$H_e = \sum_j p_i \cdot E_{loc}$$

The local dipole moment is $p_i = \alpha_i E_{loc}$ Where $\alpha_i$ is the polarizability of the $i^{th}$ ion and for uniform polarization simply $\alpha$, and $E_{loc}$ is the local electric field. Thus,

$$H_e = \sum_j p_i \frac{p_i}{\alpha}$$

The dipole moment is also defined in terms of the Born charge and the effective lattice displacement
The ionic operator can be transformed in to wave coordinates as 

\[ u_i = N^{-\frac{1}{2}} \sum_q Q_q e^{iq \cdot x_i} \]

whose Fourier transform is 

\[ Q_q = N^{-\frac{1}{2}} \sum_i u_i e^{-iq \cdot x_i} \]

Once again the displacement vector can be represented in terms of the phonon creation and annihilation operators

\[ u_i = \sum_q \left( \frac{\hbar}{2M\omega_q} \right)^{\frac{1}{2}} (c_q e^{iq \cdot x_i} + c_q^\dagger e^{-iq \cdot x_i}) \]

The energy expectation value tends to be 

\[ H_e = \sum_q \frac{e^{q^2}}{\alpha} \frac{\hbar}{M\omega_q} (c_q^\dagger c_q + \frac{1}{2}) \]

**The interaction component of the Hamiltonian**

Let’s consider a ferroelectromagnet crystal of ferroelectric and ferromagnetic components of a continuous linear chain of composite fine particles, perhaps, in single domain configurations and elastically isotropic with reduced mass. The elastic deformation associated with electrostriction and magnetostriction of such crystal can be expressed in terms of the vector operator \( u = x' - x_0 \) along the chain direction\(^{15}\), while \( x' \) is the position after deformation and \( x_0 \) is the equilibrium position of the atoms at a particular site. The coupling occurs, in fact, while the magnetic orders break inversion symmetry. The ferroelectric crystals are non-centrosymmetric in character, i.e. the spontaneous polarization arises from the John Tellor distortion and magnetostrictions\(^{16}\). On the other hand, the magnetization in magnetic insulators is due to localized electrons tightly bound to the lattice ions. Though, the electronic clouds tends to be bound strongly it can polarize the nearby lattices\(^{17}\) and hence there exists an interaction of the dipole moments (deformed lattices induce dipole moments) with intrinsic fields through exchanges. Therefore, it helps to switch magnetic properties making use of electric fields and vice versa\(^{18,19,20}\).

The interaction Hamiltonian of the spin system and and lattice vibration is represented by

\[ H_{me} = -\lambda \sum_{i \neq j} (u_i \cdot r_j) \cdot (s_i \cdot s_{i+j}) \]

Where \( \lambda \) is the spin-phonon coupling parameter, \( r_i \) position vector along chain axis, and \( s_i \) the spin operator. Using the spin operators transformed into magnon operators and the lattice displacement vectors expressed in terms of phonon annihilation and creation operators,
Where, \( \sigma \) stands for the lattice vibration polarization; \( \sigma = x, y, z \). At large coordination number

\[ z \gamma_{q-1} \approx z \gamma_q \]

and on symmetric considerations while the operators are maintained to the bilinear orders, at \( q = \pm k \), the interaction energy is

\[ H'_{me} = -\frac{\lambda Na}{2s^2} \sum_{qz} \left( \frac{h}{M \omega_{qz}} \right)^{\frac{1}{2}} \gamma_q \{ c_{qz} b_q^\dagger + c_{qz}^\dagger b_q \} \]

Thus, the effective Hamiltonian by virtue of equations 3, 11 and 13 becomes

\[ H = \sum_k \omega_m b_k^\dagger b_k + \sum_{q \sigma} \omega_{q \sigma} c_{q \sigma}^\dagger c_{q \sigma} - \sum_{qz} \lambda' \{ c_{qz} b_q^\dagger + c_{qz}^\dagger b_q \} \]

Where \( \lambda' = \frac{\lambda Na}{2s^2} \left( \frac{h}{M \omega_{qz}} \right)^{\frac{1}{2}} z \gamma_q \)

3 Results and discussion

The Hamiltonian is diagonalized by the motion equation method

\[ i \frac{db_k}{dt} = [b_k, H] = \omega_m b_k - \lambda' c_{qz} \]

\[ i \frac{db_k^\dagger}{dt} = [b_k^\dagger, H] = -\omega_m b_k^\dagger + \lambda' c_{qz}^\dagger \]

we denote, \( \omega_m \) the free magnon frequency, \( \omega_{qz} = \omega_p \) the \( z \)-direction lattice vibration frequencies, respectively.

\[ i \frac{dc_{qz}}{dt} = [c_{qz}, H] = \omega_{qz} c_{qz} - \lambda' b_k \]

\[ i \frac{dc_{qz}^\dagger}{dt} = [c_{qz}^\dagger, H] = -\omega_{qz} c_{qz}^\dagger + \lambda' b_k^\dagger \]

In the stationary states all the operators have exponential variations proportional to \( e^{-i\omega t} \) as a solution and \( \omega' \) being the excitation frequency.

\[ (\omega' - \omega_m) b_k + \lambda' c_{qz} = 0 \]

\[ \lambda' b_k + (\omega' - \omega_p) c_{qz} = 0 \]

These linear homogeneous equations have solutions if the determinant vanishes and leads to an excitation frequency of
\( \omega' = 1/2 \left[ \omega_m + \omega_p \pm \sqrt{(\omega_m - \omega_p)^2 + 4\lambda'^2} \right] \)

This is the dispersion of the excitation of hybridized crystal systems. In recent literatures, the excitation of both magnonic and phononic hybrid is the electromagnon. This novel excitation is a quantized quasi-particle described by the aforementioned frequency ranges. At the crossover region, in which the dispersions coincide for values of \( \omega_m = \omega_p \) at some particular values of \( q = k \), the above equation reduces to

\( \omega' = 1/2 \left[ \omega_m + \omega_p \pm 2\lambda' \right] \)

It indicates that if the interaction exists, the two dispersions differ significantly to the order of the coupling order parameter \( \lambda \). Whereas, if it tends to zero, the excitation energy approaches to either the pure magnon or pure phonon components. We can also rewrite it either

\( \omega' = \omega_m \pm \lambda' \quad \text{or} \quad \omega' = \omega_p \pm \lambda' \)

Approximation of \( \lambda' \) on the symmetric argument

\[ z|\gamma_q = z \cdot z^{-1} \sum_{\delta} e^{i q \cdot \delta} \approx z - \frac{1}{2} \sum_{\delta} (q, \delta)^2 \approx z - \frac{1}{2} (q, a)^2 \]

Where \( \delta = a \). Finally,

\[ \lambda' \approx \frac{\lambda N a}{2} \left( \frac{\hbar}{M \omega_p} \right)^{\frac{1}{2}} \left\{ z - \frac{1}{2} (q, a)^2 \right\} \]

Using the notion \( q = k \) and \( \omega_m = \beta k^2 + \gamma \) at the limiting optical frequency value of \( \omega_p \), the excitation frequency is

\[ \omega' = (\beta - \beta') k^2 + \gamma + \gamma' \]

Where \( \beta' = 1/4 \lambda N a s^3 / 2 (\sqrt{L M \omega_p}) \) and \( \gamma' = 1/2 \lambda N a s^3 / 2 (\sqrt{L M \omega_p}) \) such that

\[ \beta' / \gamma' = a^2 / 2z \]

In the same way,

\[ \omega' = (\beta + \beta') k^2 + \gamma - \gamma' \]

The magnetization characters of the array

An essential part of this work is to obtain the magnetization. The saturation magnetization per unit length is calculated making use of the excitation energy.

\[ M_s(T) = \mu_B S_z = \mu_B \sum_i (S_{iz}) \]

The magnetization of the hybrid described in terms of bosonic quasi-particle occupation number density at the new excitation frequency is
Where the occupation number density at the excitation frequency is obtained as

\[ \langle n_k(\omega') \rangle = \frac{1}{e^{\eta \omega'} - 1}; \quad \eta = \frac{1}{k_B T} \]

However, in one dimensional crystal, \( V^* = 2\pi/L \)

\[ M_s(T) = M_s(0) - g \mu_B N \int \frac{dk}{2\pi} \frac{1}{e^{\frac{\eta \omega'}{2}} - 1} \]

The term \( \omega' \) is a function of magnetic and electric fields in addition to the wave vector.

At low temperature, setting \( \omega' \rightarrow +\infty \) and about \( \eta \rightarrow +\infty \), the magnetization

\[ M_s(T) = M_s(0) - g \mu_B N \frac{e^{-\eta(\gamma + \gamma')}}{2\pi} \sqrt{\eta(\beta - \beta')} \text{erf}(\sqrt{\eta(\beta - \beta')}k) \]

Where \( \beta = J\alpha^z \). If the argument \( k \) goes to infinity, this function will be expressed as the Gaussian integral of zero mean and half variance. That is, \( \text{erf}(ax) = \pm 1 \) for \( ax \rightarrow \pm \infty \).

\[ M_s(T) = M_s(0) - g \mu_B \frac{N}{4\sqrt{\beta - \beta'}} e^{-\eta(\gamma + \gamma')} \sqrt{\frac{\pi}{\eta(\beta - \beta')}} \]

Figure 1: Magnetization versus temperature plot. It indicates the magnetic characteristic of the linear chain of atoms in the low temperature limits for some values of tunable parameter \( \alpha' = \frac{\gamma + \gamma'}{k_B} \). The range of values given arbitrarily as, 0, 1, 10 and ensures how the magnetization monotonically drops with the temperature in comparison with experimental observations 21,22,23.
The electric polarization of the chain

In this section, we look at the polarization of the crystal structure considered so far. We discuss the lattice distortions reasonably accounted for the electrical phenomenon. Ferroelectrics are viable for structural distortions, consequently so do the ferromagnetics due to magnetostrictions and induce dipole moments are expected to be seen. Thus, it is assumed that there exists an induced charge density fluctuation $\rho(x)$ in every small local dilation of the lattice. We denote the displacement $x$ to represent the fluctuation. The general expression for induced polarization is different from zero, i.e.

$$\int x \rho(x) dx = P$$

The density fluctuation operator is $\rho(x) = \sum_q \rho_q e^{iq.x}$ where the Fourier transform

$$\rho_q = \sum_i \rho(x) e^{-iq.x}$$

$$P = \sum_q \rho_q e^{iq.x} \frac{1}{q^2} - i \sum_q \rho_q e^{iq.x} x \frac{1}{q}$$

For discrete dipoles of density fluctuation, we can also write,

$$\rho(x) = \sum_j \delta(x-x_j)$$

So that

$$\rho_q = \int \sum_j \delta(x-x_j) e^{iq.x} dx = \sum_j e^{iq.x}$$

The polarization becomes

$$P = \sum_q \frac{1}{q^2} \sum_j e^{iq.(x-x_j)} - i \sum_q \frac{1}{q} \sum_j e^{iq.(x-x_j)} x$$

At $x=x_j$, if we define the average occupation number density at a particular site $j$, the polarization becomes

$$P = \sum_q \frac{n_q}{q^2} - i \sum_q \frac{n_q}{q} x_j$$

24

Integrating over $q$ space

$$P = \frac{1}{2\pi} \int dq \frac{1}{q^2} e^{\eta q} - \frac{1}{q} e^{\eta q} - i \sum_j x \frac{1}{2\pi} \int dq \frac{1}{q} e^{\eta q} - \frac{1}{q} e^{\eta q}$$

$$P = \frac{1}{2\pi} e^{-\eta(\gamma+\gamma')} \left\{ \int dq q^{-2} e^{-\eta(\gamma-\gamma')} q - i x \int dq q^{-1} e^{-\eta(\gamma-\gamma')} q \right\}$$

26

To evaluate these integrals, the limits of integrals are chosen to run to infinity. Meanwhile, using
the standard integrals and the lattice spacing $x_j = a$, we obtain the expression

$$P = \frac{360}{\pi} e^{-\eta(\gamma+\gamma')} \eta(\beta-\beta') \sqrt{\frac{\pi}{(\beta-\beta')}} + i \frac{a}{4\pi} e^{-\eta(\gamma+\gamma')}$$

This equation shows that polarization is the sum of real and imaginary terms. The real part is proportional to the inverse square root law of the temperature, $T^{-1/2}$ apart from the exponential dependency. The general characteristics of the polarization dependency on the fields and temperature is indicated briefly in figure 2 considering an approximate value of $P= \frac{360 \sqrt{\beta-\beta'}}{\pi} \approx 100$, and at some values of $\gamma+\gamma' = 1$, and $\gamma+\gamma' = 10$. The real part is significantly variable and tunable with the parameters at low temperatures, while the imaginary part saturates to one even if the fields tend to vanish.

Figure 2: On the left panel of the figure we observe enhanced polarization both at low temperatures and fields. However, it tends to saturate to some lower value at high field values. On the right panel the imaginary plot indicates how the polarization approaches to a specific value regardless of the magnitude of the fields.

Figure 3: The figure herein shows how the susceptibility varies with the electric field keeping the other parameters constant at some specific low temperatures.
In conclusion, we consider a linear chain of composites of ferroelectric-ferromagnetic crystal structure where magnon-phonon interaction is responsible for the flexibility of the magnetic and electric properties of the crystal. This is, of course, due to the magnetostriction and electrostriction behaviors of the linear chain cooperatively inducing the new classes of collective excitation modes the "electromagnons" that can be observed at new excitation energies parametrized by the coupling order parameter. The hybrid novel excitation is a quantized quasi-particle described by the frequency ranges at the crossover region, in which the dispersions coincide for values of \( \omega_m = \omega_p \) at some particular values of \( q = k \). \( \omega'_m = \omega_m + \lambda' \) or \( \omega'_p = \omega_p + \lambda' \). It indicates that if the interaction exists, the two dispersions differ significantly to the order of the coupling order parameter \( \lambda' \). Whereas, if it tends to zero, the excitation energy approaches either the pure magnon or pure phonon modes.

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