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Electric field induced and enhanced magnetic properties of multiferroic material ($BiFeO_3$)

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ABSTRACT: In this research work, enhancement and induction of magnetization and other related properties of ferro-electromagnet is reported. The ascertained enhancement and induction is supported by the Hamiltonian composed of itinerant electrons, localized electrons, and their mutual interactions. Using double time temperature dependent Green's function formalism and equation of motion, the study exhibited the effect of external electric field that can induce and enhance the magnetization and other related parameters of the material. The expressions for spin wave energy, magnetization, magnetic susceptibility, and heat capacity of the system have been obtained and influenced by the action of an external electric field. These enhanced and induced functional responses of this material will present an opportunity to create and implement thin film devices that actively couple the magnetic and ferroelectric order parameters. The results obtained in this work are in a good agreement with the previous theoretical and experimental findings.

Keywords/phrases: Multiferroic, Green's function, Magnetization; Order parameter, Magneto-electric coupling, Spin wave energy

INTRODUCTION

Multiferroic materials are an exceptional class of solid state compounds, in which at least two order states such as magnetic, electric or piezo-elastic phases coexist (Liu Yu, et al., 2015). Thus, well known materials such as ferromagnetic materials that show strong magnetic properties, as well as ferroelectrics are possibly a special class of electro-elastic coupled multiferroics. The interaction between the two types of orders, called magneto-electric coupling introduces possible multi-functional responses to an external electric and magnetic fields (Liang and Huai, 2015). Multiferroics are considered now as promising materials for magnetic storage, sensors or spintronics devices and microwave technique as they provide a novel approach to the magnetic field or electric field responses (Melvin, 2015). In these materials ferromagnetism and ferroelectric ordering coexist no matter how their course of existence is different from material to material (Liu Yu, et al., 2015; Smolenskii, *et. al., 1960*).

These materials have a unique character when subjected to an applied external magnetic and electric fields, the electric polarization, magnetization, and other related properties are totally modified from the former one. The magneto-electric effect in multiferroics can therefore be electrically or magnetically induced; and it is mathematically described by the magneto-electric coupling coefficient. The electrically induced magneto-electric effect (α_E) is defined as the change in magnetization (M) of sample due to the action of an electric field (E). That is,

$$\alpha_E = \frac{\partial M}{\partial E} \qquad (1)$$

Similarly, the magnetically induced magneto-electric effect (α_M) is the change in the electric polarization (P) of the sample due to the application of magnetic field (H). i.e.,

$$\alpha_H = \varepsilon_0 \varepsilon_r \frac{\partial E}{\partial H} \qquad (2)$$

where ε_o and ε_r are the electric permittivity of the vacuum and relative permittivity of the medium, respectively.

There are two possible types of magneto-electric coupling (Nan, et al., 2008), known as direct and indirect. The direct magneto-electric coupling refers to multiferroics in which the direct application of electric or magnetic field results in changes of magnetization or electric polarization of the system, respectively. This effect is characteristic to a special class of multiferroic materials called

"single-phase" multiferroics. In the case of indirect magneto-electric effect, the application of magnetic or electric field does not affect directly the electric polarization or magnetization, respectively. Instead, they are modified indirectly via a strain mediated magneto-electric coupling. Being one of the very few room temperature multiferroics, $BiFeO_3$ is arguably the most important multiferroic. Ferroelectric order originates from the lone electron pair of Bi, while magnetic ordering is due to the unpaired Fe spins (Ohno, et. al., 2000; Sosnowska and Steichele, 1982).

Theoretical Model of the Hamiltonian

In this work, we considered spin Hamiltonian for ferro-electromagnetic thin film order to demonstrate magnetic and other properties of the system under study, we need to formulate the Hamiltonian consisting of an external electric field and magnetic field, which is written as in equation (3) (Liang and Huai, 2015; Shegaw and Singh, 2012).

$$H = \sum_{i,j}^{N} J_{ij} S_i S_j - g\mu_B H \sum_{i}^{N} (S - n_i) + D \sum_{i}^{N} (S^2 - n_i) - g\mu_E H \sum_{i}^{N} (S - n_i) + \lambda \sum_{i}^{N} S_i^z . \sigma \quad (3)$$

In equation (3) the first term represent the Hamiltonian for spin interaction in a localized state, where S_i and S_j are spin operators at sites *i* and *j*. J_{ij} is the exchange coupling that depends on the relative positions of neighboring spins and is defined by $J_{ij} = J(R_i - R_j)$. The constant *g* is the gyro-magnetic ratio and the second term describes the effective Zeeman field with an external deriving field while in this model it is chosen to be parallel to the +z-axis to line up all the spins in the same direction. The third term represents Hamiltonian for total spin operator. The fourth term include an electric field *E* interacts with the spins exist in the material. The constant μ_E describes the coupling strength between the spins and the radiation field. The terms μ_B and *D* are the Bohr magneton and magnetic anisotropic (or spin wave stiffness) constants, respectively; and the last term describes the interaction between localized magnetic moment and the itinerant electrons, where λ is the corresponding exchange constant.

Transforming the spin ladder operator problems into many-body interaction systems, Bosonic creation and annihilation operators expressed as (Shegaw and Singh, 2012).

$$S_i^+ = \sqrt{2S}\sqrt{(1 - \frac{a_i^+ a_i}{2S})}a_i \quad (4)$$

and

$$S_i^- = \sqrt{2S}a_i^+\sqrt{(1 - \frac{a_i^+ a_i}{2S})} \quad (5)$$

where a_i and a_i^+ are annihilation and creation spin deviation operators, respectively, which satisfy the commutation relation $[a_i,\ a_i^+] = 1$.

It is likely transform these Bosonic operators to magnon variable operators to a_i^+ and a_i in a lower temperature limit, that can be presented as

$$a_k = \frac{1}{\sqrt{N}} \sum_i^N e^{ik.\omega x_i} a_i \quad (6)$$

and

$$a^+_k = \frac{1}{\sqrt{N}} \sum_i^N e^{-ik.\omega x_i} a_i^+ \quad (7)$$

where the inverse transformation is rewrite as,

$$a_i = \frac{1}{\sqrt{N}} \sum_i^N e^{-ik.\omega x_i} a_k \quad (8)$$

$$a^+_i = \frac{1}{\sqrt{N}} \sum_i^N e^{ik.\omega x_i} a_k^+ \quad (9)$$

Spin density conduction electron or itinerant electrons for ferroelectricity material before and after interaction process is expressed by,

$$\sigma_{k,q}^z = C_{k\uparrow}^+ C_{k\uparrow} - C_{(k+q)\downarrow}^+ \quad (10)$$

Organizing equation (4-10) and substituting in to equation (3) and consider system is handling at low temperature phenomena where the interaction is predominated by bilinear spin variables, neglecting higher order interaction, the effective Hamiltonian of equation (3) is reduced in to,

$$H = (\beta k^2 - 2DS + g\mu_B H + \mu_E E) \sum_i^N a_i^+ a_i + JS \sum_i^N a_{(k-q)\downarrow}^+ a_{(k-q)\downarrow} + \frac{JS}{N} \sum_i^N a_{k\uparrow}^+ a_{k\uparrow} \left(C_{q\downarrow}^+ C_{q\downarrow} - C_{(k-q)\downarrow}^+ C_{(k-q)\downarrow} \right) \quad (11)$$

or,

$$H = \omega_o \sum_i^N n_k \omega_k \quad (12)$$

where

$$\omega_o = (-2NS^2J - Ng\mu_B H + N\mu_E E + 2NS\lambda - Nm(g\mu_B H - \mu_E E)) \quad (13)$$

Here, m is spin carrier polarization, expressed as,

$$m = \frac{1}{N}\left(\sum_{k,q}^{N}\langle C_{k\uparrow}^{+}C_{k\uparrow}\rangle - \langle C_{(k+q)\downarrow}^{+}C_{(k+q)\downarrow}\rangle\right) \quad (14)$$

In order to calculate multiferroic order parameter, first it is better to write the equation of motion, using Green's function formalism which is given by

$$G_a(t,t') = \ll A(t); B(t) \gg_a \quad (15)$$

After doing some mathematical steps and certain approximations, the equation of motion for the Fourier transform of GF is provided by

$$\omega G(\omega)_\omega = \frac{1}{2\pi} < [A;B] > + \ll [A;H] \gg_\omega \quad (16)$$

where A and B are the Heisenberg operators which might be represented by creation and annihilation magnon operators as

$$\omega \ll a_{k\uparrow}; a_{k\downarrow}^{+} \gg = \frac{1}{2\pi} < \left[a_{k\uparrow}; a_{k\downarrow}^{+}\right] > + \ll [a_{k\uparrow}; H]; a_{k\downarrow}^{+} \gg_\omega \quad (17)$$

Substituting equation (11) into equation (17) and then evaluate commutation relation, from which we obtain,

$$\omega \ll a_{k\uparrow}; a_{k\downarrow}^{+} \gg = \frac{1}{2\pi} + \left(JS(ka)^2 - 2DS + g\mu_B H + \mu_E E \ll a_{k\uparrow}; a_{k\downarrow}^{+} \gg\right) - \frac{\lambda}{N}\left(\ll a_{k\uparrow} C_{k\uparrow}^{+} C_{k\uparrow}; a_{k\downarrow}^{+} \gg - \ll a_{k\uparrow} C_{(k+q)\downarrow}^{+} C_{(k+q)\downarrow}; a_{k\downarrow}^{+} \gg\right) \quad (18)$$

Employing decoupling procedure on the higher order Green's function, we obtain the following simplified equation of motion:

$$\omega \ll a_{k\uparrow}; a_{k\downarrow}^{+} \gg = \frac{1}{2\pi} + \left(JS(ka)^2 - 2DS + g\mu_B H + \mu_E E\right) \ll a_{k\uparrow}; a_{k\downarrow}^{+} \gg - m\lambda \ll a_{k\uparrow}; a_{k\downarrow}^{+} \gg] \quad (19)$$

From equation (19), we get the Green's function as:

$$G(\omega)_k = \frac{1}{2\pi}\left(\frac{1}{\omega - \beta k^2 + 2DS - \mu_B H - \mu_E E - m\lambda}\right) (20)$$

From equation (20) we can find the pole's of the Green's function, which is written as,

$$\omega_k = \beta k^2 + \gamma \quad (21)$$

Equation (21) describes the spin wave energy of multiferroic material as a function of wave vector k, where the constant beta and gamma terms are defined as,

$$\beta = JSa^2$$
$$\gamma = 2DS - \mu_B H - \mu_E E - m\lambda \quad (22)$$

From equation (22) the gamma term represents the coupling interaction between ferromagnetism and ferroelectricity subsystems.

RESULTS AND DISCUSSION

Spin wave energy

An applied external electric field is induced and enhanced spin wave energy of multiferroic material, which results to high mobility of the magnetic moment in the given domain (Shegaw and Singh, 2012). The material parameters that are used for the analysis of spin wave energy are the following: J =1/6 *meV*, S = 3/2, g = 25, a = 5.639 Å, and m = 0.845. Based on equation (21) spin wave energy versus wave vector for γ = 0, 5, and 10 are plotted as shown figure here below.

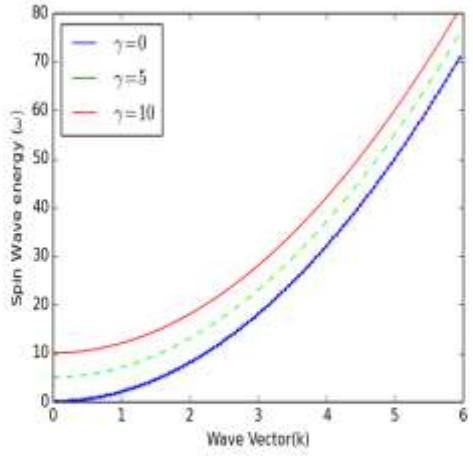

Figure 1. Calculated spin wave energy for multiferroic material as a function of wave vector k subjected to an external electric field.

As cab be seen from Figure 1, spin wave energy is altered by an external electric field and exchange coupling term. It is found that, spin wave energy increases with the increase of coupling term or electric field parameter as anticipated. We also found that as gamma term increases by a factor of five, spin wave energy gap increases with an equal gap as observed in Figure 1.

Magnetization

One important feature of multiferroic materials is the mutual control between the magnetization and polarization; that is, the magnetization (polarization) could be controlled by an external electric (magnetic) field. Microscopic theoretical studies have hardly been seen. Our present model enables us to study the properties of magnetization by the action of an external electric field. In most of magnetic materials, magnetic properties of these materials are initiated by the action of an external magnetic field, but the best things that we can see in the present work is magnetic properties of some selected materials like multiferroic that can be possibly controlled by the application of an external electric field. Manipulating magnetization of magnetic devices using an external electric field will lead to new devices that will be energy efficient, fast and compact as compared to those devices tuned by electromagnets (Sosnowska and Steichele, 1982). Magnetic excitation's possibly occurred at some temperature T and can be possibly calculated by the correlation function as,

$$< a_{k\uparrow}^{+}(t')a_{k\uparrow}(t) >= \lim_{\epsilon\to 0}\int_{-\infty}^{\infty}\frac{e^{-\omega(t'-t)}}{e^{\beta\omega}-1}\left(G(\omega+\epsilon)-G(\omega-\epsilon)\right)d\omega \qquad (23)$$

The equal time ($t = t'$) correlation function gives the number of magnon and is obtained as

$$< n_k >= \frac{1}{e^{\beta\omega}-1} \qquad (24)$$

Magnetization is the order parameter that is used to study ferromagnetism or anti-ferromagnetism of a system under study and it is given by,

$$M(T) = M(0) - m(T) \qquad (25)$$

$$M(0) = g\mu_B NS \qquad (26)$$

$$m(T) = g\mu_B \sum_{k=1}^{N}\frac{1}{e^{\beta\omega}-1} \qquad (27)$$

The magnetization M(T) can be expressed using the Bloch law as

$$M(T) = g\mu_B(NS - \sum < n_k >) \qquad (28)$$

where N is the number of magnetic atoms and S is magnetic spin of the system.
Using equations (22) and (27) into equation (28), we have

$$M(T) - M(0) = g\mu_B(NS - \sum_{k}\frac{1}{e^{\frac{(\beta' k^2 + zDS - \mu_B H - \mu_E E - m\lambda)}{K_B T}} - 1} \qquad (29)$$

For small value of k, the summation in equation (29) can be changed in to the integral form over the whole value of k space, and it becomes

$$M(T) = g\mu_B NS\left[1 - \frac{1}{NS}\int_{-\infty}^{\infty}\frac{d^2k}{e^{\frac{(\beta' k^2 + zDS - \mu_B H - \mu_E E - m\lambda)}{K_B T}} - 1}\right] \qquad (30)$$

Then, computing the magnetization gives,

$$M(T) = g\mu_B NS - \frac{g\mu_B}{\beta'(2\pi)^2}K_B T\, e^{\frac{-\gamma}{K_B T}} \qquad (31)$$

Rearranging terms, we obtained

$$\frac{M(T)}{M(0)} = 1 - A * e^{\frac{-\gamma}{K_B T}} \qquad (32)$$

where equation (32) is mathematical expression for reduced magnetization as function temperature for various values of an external electric field and A is constant term.

Based on equation (32), we have plotted the reduced magnetization as a function of temperature at different values of external electric field parameters, shown in Figure 2. This order parameter tends to increase towards the transition temperature as the electric field parameter increase. The electric field increases the coupling interaction between localized spin and the itinerant holes spins and produces spin polarized carriers which in turn creates molecular field which is highly responsible to the higher magnetization of the system (Ohno, *et. al., 2000)*.The following material parameters are used for the analysis. J = 1/6 meV, a = 5.77 Å , m = 0.85, S = 3/2, A = 0.999, $\mathbf{k_B}$ = 1.5.

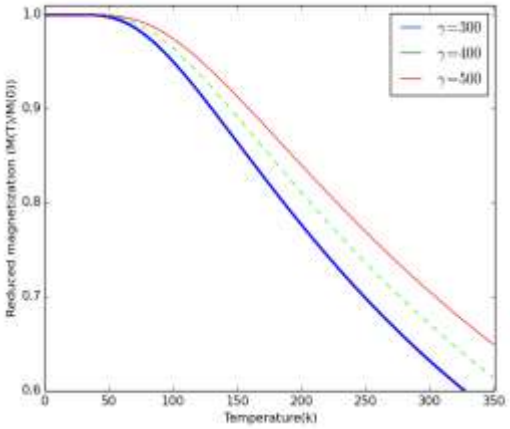

Figure 2. Calculated reduced magnetization as a function of temperature subjected to an applied electric field.

Referring equation (31), it is also possible to plot the graph of magnetization versus external electric field for various magnetic fields as depicted in Figure 3 below. The following material parameters are used for the analysis. A = 0.7, k_B = 1 and T = 6 K.

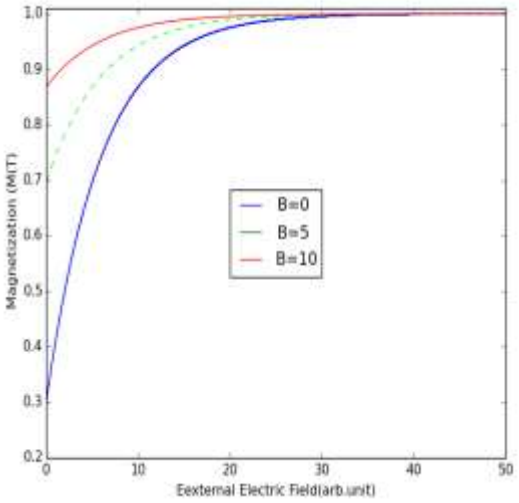

Figure 3. Calculated magnetization as a function of electric field for multiferroic material ($BiFeO_3$) subjected to various magnetic fields.

Magnetization versus external electric field subjected to various magnetic fields is shown in Figure 3, from which we see that, the magnetization of the system increases until it reaches saturation magnetization, as the external magnetic field increase, which enhances the critical temperature (*Tc*) of system.

Magnetic susceptibility

The susceptibility indicates whether a material is attracted into or repelled out of a magnetic field, which in turn has implications for practical applications. Ferromagnetic, ferrimagnetic, or antiferromagnetic materials have positive susceptibility and possess permanent magnetization even without an external magnetic field.

The magnetic susceptibility is given by

$$X = \frac{\partial M}{\partial H} \; H \to 0$$

where M is magnetization of multiferroic material which is written in equation (31), using equation (31) in equation (33), and doing certain mathematical steps we obtain reduced magnetic susceptibility as

$$X_m = CTe^{\frac{\sigma}{K_B T}} \qquad (34)$$

where C′ and σ are constants given by

$$C = \frac{g\mu^2{}_B K_B T}{\beta'(2\pi)^2}$$

and

$$\sigma = -2DS + \mu_E E + m\lambda$$

To plot equation (34), the following material parameters are used for the analysis. J = 50 meV, a = 5.77 Å , m = 0.85, S = 3/2, C =3, $\mathbf{k_B}$=2.

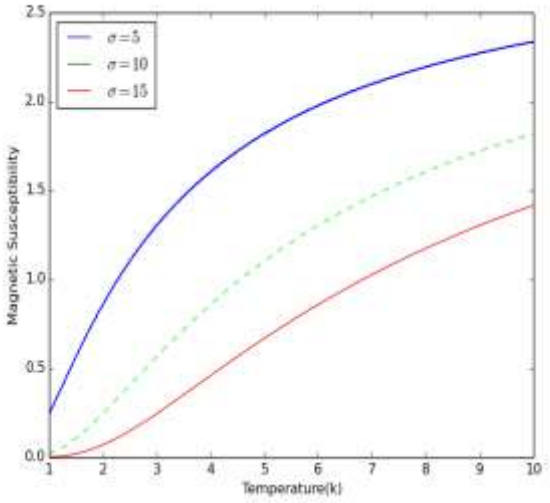

Figure 4. Magnetic susceptibility as a function of temperature for various values of an external electric field.

As can be seen from Figure 4, magnetic susceptibility of the system varies with applied external electric fields. From this figure, we have seen that magnetic susceptibility of system is controlled by an external electric field which is an astonishing event.

Heat capacity

To obtain the heat capacity system under study, first we have to find an expression for internal energy which is given by

$$U = \sum_{K} n_k \omega_k \tag{35}$$

where $< n_k >$ is the mean number of spin wave with a wave vector k at a temperature T given by

$$< n_k >= \frac{1}{e^{\frac{\omega}{k_B T}} - 1} \tag{36}$$

where ω is spin wave energy for multiferroic material which is written in equation (21). Then, substituting equations (21) and (36) into equation (35), we get that

$$U = \sum_k \frac{(\beta' k^2 + \gamma)}{e^{\frac{\beta' k^2 + \bar{E}}{k_B T}} - 1} \tag{37}$$

Changing the sum of equation (37) into integration form, then using standard integration techniques and certain approximations, we found that the total energy for multiferroic materials becomes the following expression

$$U \sim \frac{1}{4\pi^2}(k_B T + \gamma) k_B T e^{\frac{\gamma}{k_B T}} \tag{38}$$

Magnon obey Bose statistics and allow us for the calculation of the low temperature thermal properties of magnetic materials. Heat capacity is defined as the amount of heat that a unit mass of a material must gain or lose to change its temperature by a given amount. Mathematically heat capacity is defined as

$$C = \frac{\partial U(T)}{\partial T} \tag{39}$$

Hence, using equation (38) into equation (39) and manipulating certain mathematical steps we get reduced form of heat capacity as provided in equation (40). The following material parameters are used for the analysis. β' = 1/12, a = 6 Å , m = 0.85, S = 3/2, $\mathbf{k_B}$ = 1.

$$C = \frac{k_B}{4\beta' \pi^2}\left(2k_B T + \frac{\gamma^2}{k_B} + \gamma\right) e^{\frac{-\gamma}{k_B T}}$$

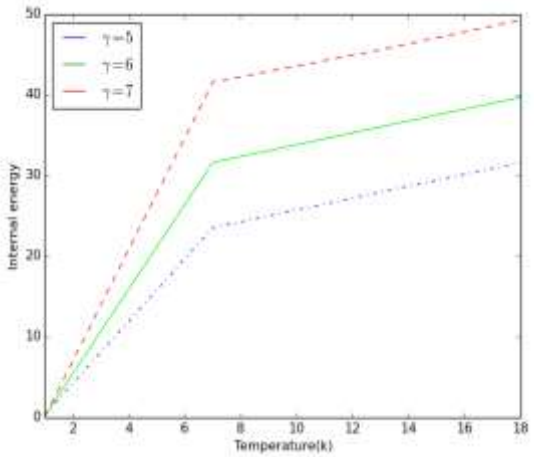

Figure 5. Heat capacity of multiferroic material as a function of temperature subjected to various values of an external eclectic field.

From Figure 5, the heat capacity varies as external eclectic field is varied; it makes the graph change in fixed time interval. From this graph, we can study the actual situation of system occurring in the presence of temperature and an external electric and magnetic fields.

CONCLUSIONS

In this article, we have shown that the magnetization of multiferroic material ($BiFeO_3$) is induced and enhanced by imposing an external electric field into the system. Thus, the magnetization is induced and enhanced with increasing the external electric field. This could be explained by the fact that an external electric field would accumulate large enough spin polarized carriers which affect the orientation of the magnetic Fe-ions and induce ferromagnetism. In addition, the critical temperature, heat capacity, and the spin wave energy of the material increase with an increase of the external electric field. These

electric field dependent magnetic properties of the materials will lead to the control of the magnetism by an electric field which is essential for spintronics applications. Our theoretical results agree very well with others theoretical and experimental findings (Ohno, *et. al., 2000).*

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