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A phenomenological Landau theory for electromagnons in cubic spinel multiferroic $CoCr_2O_4$

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Abstract

Non-anisotropic free energy is considered which under minimization yields two magnetic phases: a conical spin density wave and a low temperature conical cycloid. Using equations of motion, the excitation spectrum is studied. Knowing the nature of these excitations, the dielectric function as well as the fluctuation specific heat is computed and compared with the experimental spectrum. Due to the electromagnon going soft, the dielectric function (imaginary part) as well as the specific heat capacity show peaks at the temperature where ferroelectricity appears in the system.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

There has been a renewal of interest recently in multiferroics, i.e. a class of materials where long range ferroelectric as well as magnetic order coexist together, due to their potential technological applications, such as in multi-state memory devices, magnetically/electrically switchable optical devices, etc. Most of the multiferroics discovered recently show the coexistence of a spatially modulated magnetic order and a uniform polarization, which is induced by the broken inversion symmetry due to the modulated magnetism [1–7]. Since such a polarization is inherently of magnetic origin, an unusual magnetoelectric (ME) effect appears which shows an ability to tune polarization by the application of a magnetic field. Among these exciting classes of multiferroics, the cubic spinel oxide $CoCr_2O_4$ is most unusual in the sense that it not only displays uniform polarization and spatially modulated magnetism but also uniform magnetization in the conical cycloid state [8, 9]. The most appealing phenomenon of this material is that one can tune polarization by the uniform component of magnetization by application of a magnetic field of nearly 0.5 T. This question has been addressed by Zhang *et al* [10, 11] in a phenomenological Landau theory where they considered a spin rotational invariant free energy.

The crystal structure of $CoCr_2O_4$ is a cubic normal spinel, in which magnetic Co^{2+} ions occupy the A (8a) sites and Cr^{3+} ions the B (16d) sites [12, 13] of AB_2O_4. It enters into a ferrimagnetic state at $T = T_c = 93$ K, but in our formulation we consider only the ferromagnetic (FM) component of the magnetization. With further lowering the temperature, the compound undergoes a transition to conical spin states, i.e. a uniform plus transverse spiral spin state, with an incommensurate propagation vector of $(q\ q\ 0)$ $(q \approx 0.63)$ at 26 K. The helical modulation of the magnetization is in a plane transverse to the uniform component. Such a state can be described by an order parameter,

$$\vec{M} = m_1\hat{e_1}\cos(\vec{q}\cdot\vec{r}) + m_2\hat{e_2}\sin(\vec{q}\cdot\vec{r}) + m_3\hat{e_3}, \qquad (1)$$

where $\{\hat{e_i}\}$ form an orthonormal triad. When the pitch vector $\vec{q}$ is normal to the rotational components, the rotating components form a 'conventional helix'. When $\vec{q}$ lies in the plane of the rotating components then, for $\vec{m}_3 = 0$, it is an 'ordinary cycloid' otherwise it is called for $\vec{m}_3 \neq 0$ a conical cycloid state. The latter magnetic modulation has been observed in $CoCr_2O_4$ at low temperature $T < 26$ K. In the low temperature phase (below 26 K), the polar order appears and since it is coupled to spatially modulated magnetism, it would be interesting to understand the associated collective modes in the system. In this paper, we precisely address this issue in the framework of the phenomenological Ginzburg–Landau approach. We develop this in a rotationally invariant way and show that multiferroicity occurs in this formulation even without easy-plane spin and easy-plane lattice anisotropies. This is in accordance with the material $CoCr_2O_4$ where the cubic symmetry forbids easy-plane spin and easy-plane lattice

anisotropies. Using the equation of motion and Fourier decomposition, we study the excitation spectrum, dispersion of the modes, and dielectric susceptibility as well as the fluctuation induced specific heat in this system.

2. Theoretical framework

Much effort has been devoted to understanding the origin of magnetically induced ferroelectricity [14] and it is still under debate. The microscopic models used to address multiferroicity are based on the theory of spin current [15], spin–orbit coupling [16], etc, whereas the symmetry based phenomenological approach deals with constructing a free energy which takes into account the coupling between ferroelectric and magnetic orders [17]. This is mostly done by introducing a term in the free energy of type $(\vec{P}\cdot(\vec{M}\times\vec{\nabla}\times\vec{M}))$ to lowest order. This yields a nonzero polarization $\vec{P}$ which can be expressed as $P \propto M_1 M_2(\hat{e_3}\times\vec{q})$. Thus $\vec{P}$ becomes normal to both $\vec{q}$ and $\hat{e_3}$. Since in the cycloid state $\vec{q}\perp\hat{e_3}$, $\vec{P}$ becomes nonzero. This was precisely done by Zhang *et al* [10, 11] where they considered a spin rotational invariant free energy. They predicted that a second-order phase transition from ferromagnet (FM) to conical cycloid (CC) state occurred through an intervening conical longitudinal or transverse spin density wave state, even though the direct FM–CC transition was first order. We, in what follows, consider only the conical spin density wave (CSDW)–CC transition where the CC phase is characterized by a polarization along the b-axis. Thus, based on the symmetry, we consider a Landau free energy as

$$F = \int f\,\mathrm{d}^3 r \tag{2}$$

where

$$f = \frac{a}{2}\vec{M}^2 + \frac{u}{4}\vec{M}^4 + \frac{\gamma}{2}(\vec{\nabla}\cdot\vec{M})^2 + \frac{\gamma_1}{2}(\vec{\nabla}\times\vec{M})^2 + \frac{\alpha}{2}(\vec{\nabla}^2\vec{M})^2 + \frac{b}{2}\vec{P}^2 - \nu\vec{P}\cdot(\vec{M}\times\vec{\nabla}\times\vec{M}). \tag{3}$$

In constructing the free energy, the details of the unit cell for $CoCr_2O_4$ have not been taken into account. Here, the parameters u, b, and γ_1 are positive [11]. The parameters $a = a_0(T - T_0)$, $\gamma = \gamma_0(T - T_N)$, and $\alpha = \alpha_0(T - T_N)$ have been introduced to obtain the required magnetic phases in the system. Here $\gamma < 0$ and $\alpha > 0$ below T_N (α_0: − ve) and $T_0 > T_N$ (T_0 is some reference temperature below which M_3 becomes nonzero). The parameter ν represents the coupling between the ferroelectricity and the magnetization. It should be noted here that the above free energy is considered in order to explain the observed low temperature properties of the cubic spinel $CoCr_2O_4$. Since this material lacks spin/lattice anisotropies, we artificially employ coupling such as γ, γ_1, etc in the free energy to obtain the required phases. This can be justified by writing the term $|\nabla M|^2 = (\nabla\cdot M)^2 + |\nabla\times M|^2 +$ an unimportant surface term. The equivalence of γ and γ_1 will make the free energy rotationally invariant. We introduce a small difference between γ and γ_1 due to the smallness of spin–orbit coupling. The effects of competing magnetic interactions which are present in the multiferroic systems are responsible for the modulation of M which are embodied in γ and γ_1. The specific temperature dependence gives rise to the required magnetic phases in the formulation. Considering the equilibrium magnetic states in $CoCr_2O_4$ to be $M_{x0} = M_1\cos kx$, $M_{y0} = M_2\sin kx$, and $M_{z0} = M_3$, the above free energy is minimized with respect to the order parameters M_1, M_2, M_3, P_y, and the modulation vector k. Thus, one finds the CSDW (I) and the CC(II) phases which are given as follows:

$$\text{I:}\begin{cases} M_1 = 0; \qquad M_2 = 0; \qquad P = 0; \\ M_3^2 = \dfrac{-a}{u} \end{cases} \tag{4}$$

$$\text{II:}\begin{cases} M_2 = 0; \qquad P = 0; \\ M_3^2 = \dfrac{-1}{u}(a - 2\gamma k^2 + 2\alpha k^4) \\ M_1^2 = \dfrac{-4}{u}(\gamma k^2 + \alpha k^4) = \dfrac{\gamma_0^2}{u\alpha_0}(T - T_N) \\ k^2 = \dfrac{-3\gamma}{7\alpha} \end{cases} \tag{5}$$

$$\text{III:}\begin{cases} M_3^2 = \dfrac{-1}{u}(a - 2\gamma k^2 - 2\alpha k^4) \\ M_1^2 = \dfrac{-1}{2u}[(7\gamma k^2 - \gamma_1 k^2 + 6\alpha k^4) + \dfrac{2k^2\nu^2}{ub}(5\gamma_1 k^2 + \gamma k^2 + 6\alpha k^4)] \\ M_2^2 = \dfrac{-3}{2u}[(\gamma k^2 + \gamma_1 k^2 + 2\alpha k^4) - \dfrac{2k^2\nu^2}{ub}(3\gamma k^2 - \gamma_1 k^2 + 2\alpha k^4)] \\ \quad = \dfrac{9\gamma_0^2}{48u\alpha_0}\left(1 + \dfrac{30k^2\nu^2}{ub}\right)(T - T_l) \\ P_y = \nu k M_1 M_2/b, \end{cases} \tag{6}$$

where $T_l = T_N - \frac{7\gamma_1}{\gamma_0}(1 - \frac{32k^2\nu^2}{ub})$. In obtaining such a phase diagram, we assumed the magnetoelectric coupling to be weak, that is $\frac{\nu^2 k^2}{ub} \ll 1$. The transition temperature T_N and T_l are related as, $T_l = T_N - \frac{7\gamma_1}{\gamma_0}(1 - \frac{32k^2\nu^2}{ub})$ such that $T_l < T_N$. Thus, the above minimization scheme yields a phase diagram where on lowering T, one obtains the CSDW phase below T_N and on further lowering (below T_l), it enters into the conical cycloidal magnetic phase. The ferroelectricity of magnetic origin appears below T_l.

Since the system under consideration is ME coupled, the excitations in such a system will be coupled magnetic and lattice modes. This has been analyzed using the equations of motion method. In order to do this, we consider the fluctuations in the magnetic vector ($\vec{m} = \vec{M} - \vec{M}_0$) and the electric polarization ($\vec{p} = \vec{P} - \vec{P}_0$) with respect to their equilibrium values. Further, these excitations are coupled to the y-component of the electromagnetic field (E_y), propagating along the x-direction ((E, $P \parallel b$) and (k, $q \parallel a$)) (see figure 1). This is taken care of by Maxwell's equations since its electric field component E_y excites polarization along the y-direction. Thus, Maxwell's equations and the equations of motion both, in the linear approximation, give rise to a coupled set of equations for p_y and m_y which are written as,

$$\lambda\ddot{p_y} + \delta_1\dot{p_y} + \frac{\delta F}{\delta p_y} = 0 \qquad \mu\ddot{m_y} + \delta_2\dot{m_y} + \frac{\delta F}{\delta m_y} = 0, \tag{7}$$

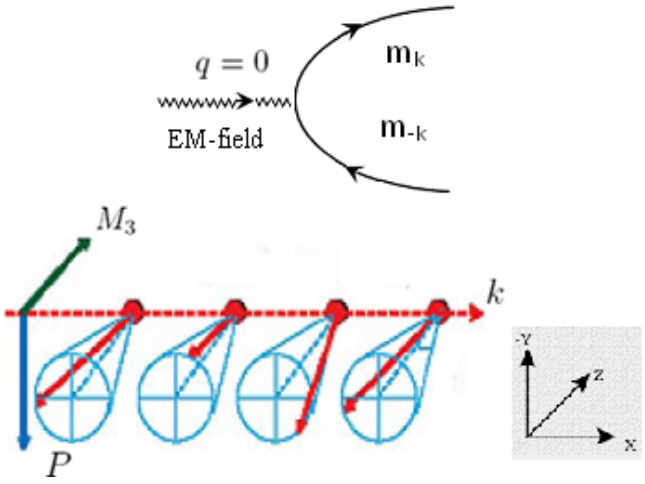

Figure 1. Light interaction with electromagnons and spin ordering in the cycloidal phase in $CoCr_2O_4$.

where μ and λ are the density parameters which characterize the kinetic energy of the system. In the limit of no damping ($\delta_1 = 0 = \delta_2$), these equations can explicitly be written as

$$\begin{aligned}&\lambda\ddot{p_y} + bp_y - E_y - \nu(M_{x0}\partial_x m_y - m_y\partial_x M_{x0}) = 0\\&\mu\ddot{m_y} + (a + uM_{x0}^2 + uM_{z0}^2 - \gamma_1\partial_x^2 + \alpha\partial_x^4)m_y\\&\quad + \nu(2p_y\partial_x M_{x0} + M_{x0}\partial_x p_y) = 0.\end{aligned} \tag{8}$$

In order to obtain the modes of excitations, these equations are solved in Fourier space and are taken to be $m_y(x,t) = \sum_l m_l \exp[\mathrm{i}\{(lk+q)x - \omega t\}]$ and $E_y(p_y)(x,t) = E_0(p_0)\exp[\mathrm{i}(qx - \omega t)]$. Here, q is the wavevector associated with the electromagnetic field propagation and is assumed to be much smaller than that of the wavevector of the magnetic order parameter which has sinusoidal variation ($q \ll k$). In such an approximation, the higher harmonics in l except the first one can be neglected. Moreover, using Maxwell's equation, E_0 can be eliminated from the above equation by using the relation, $4\pi p_0 = E_0(n^2 - 1)$, $n(= qc/\omega)$ being the refractive index of the material. Thus, the above equation can be reduced to a coupled equation in terms of the electric and magnetic eigenvectors p_0, m_1, and m_{-1} as

$$\begin{pmatrix} B & -\mathrm{i}D_k & -\mathrm{i}D_{-k} \\ \mathrm{i}D_{-k} & -R & L_+ \\ \mathrm{i}D_k & L_- & -R \end{pmatrix}\begin{pmatrix} p_0 \\ m_{-1} \\ m_1 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \tag{9}$$

where, $B = \lambda\omega^2 - b + 4\pi/(n^2-1)$, $L_\pm = \mu\omega^2 - a - \gamma_1(q \pm k)^2 - \alpha(q\pm k)^4 - uM_1^2/2 - uM_3^2$, $D_k = \nu M_1(k - q/2)$, and $R = uM_1^2/4$. As, already mentioned above, $q \ll k$, we take $q = 0$ (optic modes only), which simplifies the above parameters as $D_{-k} = -D_k$, $L_+ = L_-$ and $m_{-1} = -m_1$. Thus, the above equation simplifies to a 2×2 matrix equation in terms of p_0 and $m(m = m_1 = -m_{-1})$ whose solution is obtained by solving the determinant. These yield the energies/frequencies of new $q = 0$ coupled modes of excitations called *electromagnons* which are given as

$$\omega_{1,2}^2 = \frac{1}{2}\left[(\omega_p^2 + \omega_0^2) \mp \left((\omega_p^2 - \omega_0^2)^2 + \frac{8k^2\nu^2M_1^2}{\lambda\mu}\right)^{\frac{1}{2}}\right],$$

and the corresponding longitudinal modes given as

$$\Omega_{1,2}^2 = \frac{1}{2}\left[(\Omega_p^2 + \omega_0^2) \mp \left((\Omega_p^2 - \omega_0^2)^2 + \frac{8k^2\nu^2M_1^2}{\lambda\mu}\right)^{\frac{1}{2}}\right],$$

where ω_0 and ω_p are, respectively, the magnetic and dipole frequencies defined as $\omega_0^2 = \frac{69\gamma_0^2}{49\mu\alpha_0}[T - T_l + \frac{224\gamma_1k^2\nu^2}{\gamma_0 ub}] - 72\gamma_0\gamma_1/7\alpha_0$, $\omega_p^2 = b/\lambda$, and $\Omega_p^2 = (b + 4\pi)/\lambda$. It should be noted here that the magnetic frequency ω_0 depends on temperature and it takes the lowest value at $T = T_l$.

It is clear from the expression of electromagnon energies ω_1 and ω_2 that they are independent of the ferromagnetic component of magnetization. This is a surprising result and is in accordance with the work of Zhang *et al* [10]. This looks similar to that of electromagnons in $RMnO_3$ materials even though the origins of temperature dependence are different. Such a phenomenon can be understood from the consideration of the free energy. Since the free energy does not contain a spin/lattice anisotropic term, M_3 is canceled and disappears from the expression of the electromagnon energy, and hence is independent of M_3. However, we believe that, in the presence of lattice/spin anisotropies, $\omega_{1,2}$ will depend on the ferromagnetic component of the magnetization.

3. Dielectric function and fluctuation specific heat capacity

The frequency and temperature dependent dielectric function ($\epsilon(\omega, T)$) (which is the square of the refractive index n) in such a formulation is calculated as

$$\epsilon(\omega, T) = \frac{(\omega^2 - \Omega_1^2)(\omega^2 - \Omega_2^2)}{(\omega^2 - \omega_1^2)(\omega^2 - \omega_2^2)}. \tag{10}$$

This result looks similar to the polariton problem in conventional ferroelectrics where the longitudinal and the transverse optic mode frequencies appear as zeroes and poles, respectively, in the frequency dependent dielectric function. One of the transverse optic modes softens toward the phase transition and is condensed according to the Lyddane–Sachs–Teller (LST) relation [18] where the static dielectric function diverges. This conventional viewpoint is not relevant in the case of multiferroics since the lattice displacement is not essential to the electronic polarization. It is the electromagnon mode, which is the outcome of the ME coupling, that goes soft at $T = T_l$. This further causes divergence in the imaginary part of the dynamic dielectric function, which becomes the origin of ferroelectricity in these systems.

Considering the long wavelength fluctuations in the order parameter and using the standard phenomenology of Gaussian theory [19], we computed the fluctuation specific heat capacity in our model, which turns out to be

$$\Delta C_P^{\mathrm{flu}} = \frac{-K_B T}{\pi^2}[I_1 - I_2], \tag{11}$$

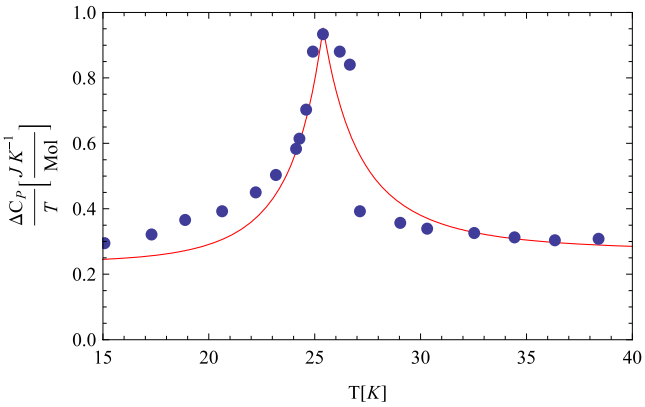

Figure 2. Temperature variation of change in specific heat $\Delta C_P/T$ versus T (data are taken from [8]).

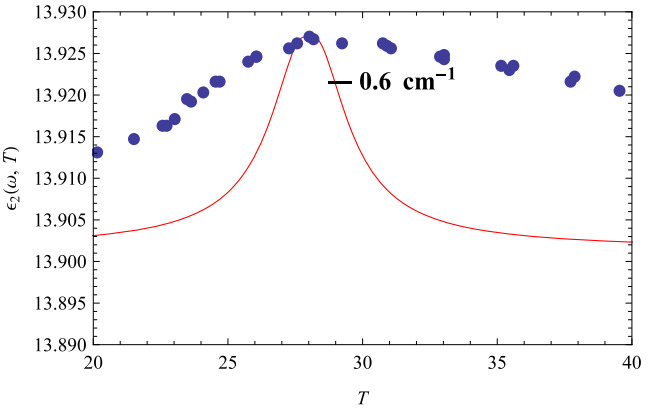

Figure 3. Temperature variation of the imaginary part of the dielectric function at $\omega = 0.6$ cm^{-1} (in arbitrary units) (data are taken from [8]).

where

$$I_1 = 2\int_0^{q_{\max}} \{(\gamma_0 q^2 + \alpha_0 q^4) q^2\, \mathrm{d}q\} \times \left\{a + \gamma_1 q^2 + (\gamma_0 q^2 + \alpha_0 q^4)(T - T_l) + \frac{3}{2}\left(\gamma_1 q^2 + \frac{\gamma_1 \alpha_0}{\gamma_0} q^4\right)\left(1 + \frac{2k^2 v^2}{ub}\right)\right\}^{-1} \tag{12}$$

and

$$I_2 = T\int_0^{q_{\max}} \{(\gamma_0 q^2 + \alpha_0 q^4)^2 q^2\, \mathrm{d}q\} \times \left\{a + \gamma_1 q^2 + (\gamma_0 q^2 + \alpha_0 q^4)(T - T_l) + \frac{3}{2}\left(\gamma_1 q^2 + \frac{\gamma_1 \alpha_0}{\gamma_0} q^4\right)\left(1 + \frac{2k^2 v^2}{ub}\right)\right\}^{-2}. \tag{13}$$

Here we have neglected the higher order terms in M such as $M^4 \ldots$, etc. The above integrals are evaluated near $T = T_l$ and the computed $\Delta C_P^{\mathrm{flc}}$ is compared with the experimental observations.

4. Results and discussion

In the present paper for $CoCr_2O_4$ which has ferrimagnetic order along with antiferromagnetic spiral order, we start with a magnetic order parameter $\vec{M} = m_1\hat{e_1}\cos(\vec{k}\cdot\vec{r}) + m_2\hat{e_2}\sin(\vec{k}\cdot\vec{r}) + m_3\hat{e_3}$ and an antisymmetric magnetoelectric coupling and conclude that the contribution of m_3 to the dielectric constant and electromagnons is negligible. This is in accordance with the magnetocapacitive measurements on $CoCr_2O_4$ where the dielectric constant is found to be strongly coupled to the spiral magnetic order parameter but insensitive to the ferrimagnetic component [20]. Thus for the study of dielectric constant and electromagnons $CoCr_2O_4$ is analogous to $TbMnO_3$ which has antiferromagnetic spiral ordering with no ferromagnetic component. A brief introduction to low frequency electromagnons in $TbMnO_3$ is given below.

A Raman scattering experiment on $TbMnO_3$ reveals (i) two peaks (at 30 and 60 cm^{-1}) with electric field $E \parallel a$ and (ii) one peak (30 cm^{-1}) with $E \parallel c$ in the cycloidal phase [21]. The (ii) case can be associated with the antisymmetric part of the magnetoelectric effect and can be well described by the Dzyaloshinskii–Moriya (DM) interaction. Here the selection rule is $q = 0$, $E \parallel c$, $p \parallel c$. The (i) case, however, is associated with both the symmetric (leading role) and antisymmetric magnetoelectric coupling [22]. The 60 cm^{-1} peak is assigned to be the 'zone-edge magnon' mode. The origin of the 30 cm^{-1} peak is more tricky. It might be assigned to the zone-center magnon mode. A recent model based on cross-coupling between magnetostriction and spin–orbit interaction can explain both the peaks at 30 and 60 cm^{-1} [23]. In this model the 30 cm^{-1} is not connected to the zone-center magnon mode but corresponds to an excitation combining the zone-edge magnon wavevector and twice the cycloid wavevector.

Keeping the data of $TbMnO_3$ in mind, we begin with the idea that there must be an electromagnon mode emerging from the rotation of the spiral plane. Since there is a lack of experiments on optical studies of $CoCr_2O_4$ we take experimental input from the temperature dependence of the imaginary part of the dielectric function and specific heat [8] and find a peak in the frequency dependence of the imaginary part of the dielectric function, the selection rule for which is $q = 0$; $E \parallel b$; $P \parallel b$. This mode is the 'zone-center magnon mode' and is comparable to the 30 cm^{-1} peak with $E \parallel c$. More peaks with different selection rules can be observed if one employs the symmetric magnetoelectric effect in the theory [24–26].

In the computation of the fluctuation specific heat the following parameters ($a/\mu = 1.0 \times 10^{-5}$, $\frac{\gamma_0\gamma_1}{\alpha_0\mu} = 250$ cm^{-2}, $\frac{\gamma_0^2}{\alpha_0\mu} = 66$ cm^{-2} K^{-1}, $\frac{k^2v^2}{ub} = 0.124$) are used such that it fits the data [8] very well (figure 2). In addition to this, the magnetic and lattice frequencies ($\omega_p^2 = 6400$ cm^{-2}, $\Omega_p^2 = 8000$ cm^{-2}, $\omega_0^2(T = 20\ \mathrm{K}) = 531$ cm^{-2}) are used to compute the imaginary part of the temperature dependent dielectric function which shows a broad peak at $T = T_l$ (figure 3). In computing this we use a width parameter $\eta = 11.0$ cm^{-1} in the dielectric function by letting $\omega \to \omega + i\eta$. The variation of the dielectric function with respect to ω at $T = 20$ K also has been shown in figure 4.

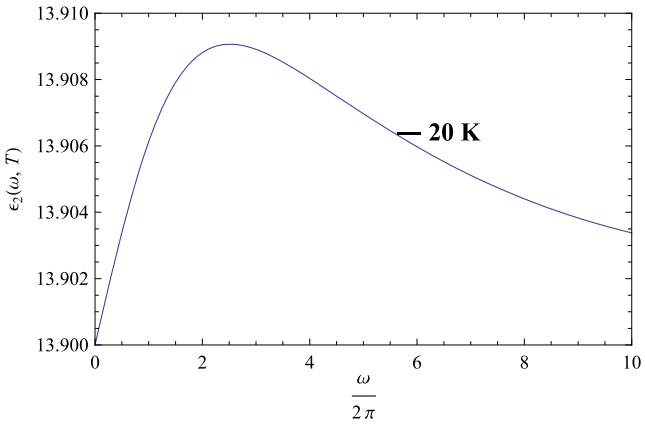

Figure 4. Variation of the imaginary part of the dielectric function (in arbitrary units) with frequency at temperature $T = 20$ K.

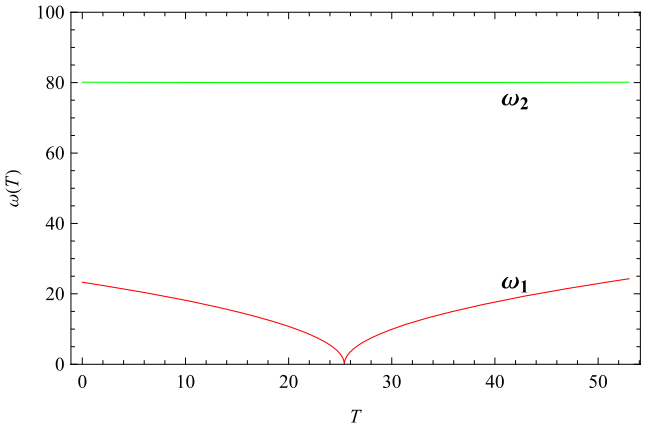

Figure 5. Variation of electromagnon frequencies ω_1 and ω_2 with temperature showing the soft mode.

A general discussion on the computed electromagnons is in order. The electromagnon modes ω_1 and ω_2 obtained in this formulation are nothing but composites of magnetic and lattice modes. The role of ME coupling is to lower the frequency of the magnetic modes (ω_0) to ω_1 whereas it pushes up the lattice mode (ω_P) to ω_2. The interaction between the electromagnon modes and that of electromagnetic radiation becomes very strong near the frequencies ω_1 and ω_2. The nature of the ω_1 mode is mostly dominated by the magnetic one so that it becomes soft at $T = T_l$ and gives rise to multiferroicity whereas the ω_2 mode is dominated by the lattice mode, which is almost temperature independent. Moreover, ω_2 is always larger than ω_1. The softening of the ω_1 mode as well as the dispersion of both the modes discussed above are shown in figures 5 and 6. The dispersion of the electromagnon here is an artifact of coupling to light. Thus, most of the low energy optical excitation occurs near ω_1. It is obvious that the low energy physics in these systems will be dominated by ω_1-excitations.

In conclusion, we briefly summarize the main results of the paper. We considered free energy which lacks spin and lattice anisotropies, minimized it and obtained the required magnetic phases, that is a conical spin density wave and a low temperature conical cycloid state. A detailed version of it was given in recent papers by Zhang *et al* [10, 11]. Using the equations of motion, the nature of the excitation spectrum (electromagnons) was studied. It was shown that one of the electromagnons goes soft at $T = T_l$ and is responsible for multiferroicity. The temperature dependent dielectric function as well as the fluctuation specific heat capacity was computed and compared with the experimentally observed spectrum.

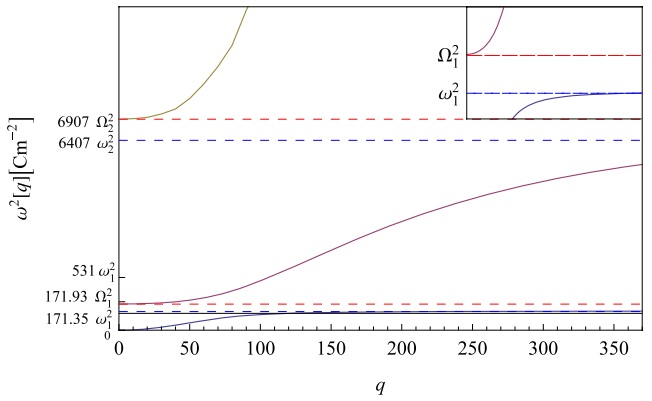

Figure 6. The electromagnon dispersion curve.

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