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Dynamic magnetoelectric couplings in ferroelectric ferromagnets

Gérard A. Maugin
Laboratoire de Mécanique Théorique Associé au Centre National de la Recherche Scientifique, Université Pierre-et-Marie-Curie, Tour 66, 4 Place Jussieu, 75230 Paris, Cedex 05, France
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A simple dynamical phenomenological theory is deduced from a variational principle with a view to studying coupled dynamical effects in rigid ferroelectric ferromagnets of the easy-axis type. In the absence of dissipation the analytical study of coupled bulk modes of wave propagation allows one to exhibit a resonance type of coupling accompanied by a repulsion of dispersion branches. The latter is of the order of magnitude of the magnetoelectric coupling constant and disappears when the bias static polarization and magnetization are either aligned or at right angles to one another. When coupled ferroelectric and ferromagnetic relaxations are taken into account, then an exchange of relaxation takes place in the neighborhood of the critical coupling wave number and the repulsion of real dispersion branches is slightly increased while the resonance effect is smoothed out, but still vanishes for an alignment of the bias fields. In all cases the nonlinear ferroelectric behavior modifies the magnetic anisotropy constant and also renders anisotropic the high-frequency electric susceptibility.

I. INTRODUCTION

Certain materials, especially compounds, may possess simultaneously ferroelectric and ferromagnetic properties.[1,2] Among those can be mentioned ferroelectric magnet solutions of $BaTiO_3Sr_{0.3}La_{0.7}MnO_3$ with 75–100 mole % concentrations for $BaTiO_3$. The ferromagnetic ordering favors the phenomenon of collective magnetic-spin oscillations, spin waves or magnons,[3] while the ferroelectric property favors high-frequency vibrations in the P (electric polarization) system. The energies and wave numbers of those two types of coherent oscillations may match and the problem arises as to the form taken by the interaction of those high-frequency vibrations. The coupling will be especially interesting in those ferroelectric ferromagnets which possess a sufficiently large magnetoelectric coupling constant. Recently, some authors, among those Bar'yakhtar and Chuppis,[4] have studied this dynamical magnetoelectric coupling with methods similar to those of quantum statistical mechanics (Holstein-Primakoff transformation, cf. Ref. 5). In the present paper we first construct a rather simple phenomenological model of rigid ferroelectric ferromagnets with the use of a classical variational principle, which follows along the same line as variational principles developed for deformable magnetically saturated media.[6] The inertia associated with the electric polarization density, the nonlinear ferroelectric behavior, the magnetoelectric couplings, and both ferromagnetic and ferroelectric relaxations are taken into account. The ferromagnet is of the easy-axis type and ferromagnetic relaxation is of the type proposed by Gilbert in the fifties (and not of the Landau-Lifshitz type, even though both formulations become equivalent for small ferromagnetic damping, cf. Ref. 7). The remainder of the paper is devoted to the study of the propagation of plane time-harmonic perturbations, first in the absence of relaxation, and next with this dissipative effect taken into account. To that purpose a static, spatially uniform, background solution is first constructed (i) where static magnetic and electric susceptibilities can be defined in terms of the initial magnetization and polarization fields and of the various phenomenological coefficients of the theory, and (ii) on which can be superimposed time-varying, spatially nonuniform perturbations. The nonlinear ferroelectric behavior renders anisotropic the high-frequency electric susceptibility. Magnetoelectric couplings, while bringing interesting dynamical interactions (see below), have also for effect to alter the value of both magnetic anisotropy constant and electric susceptibility. Magnetostatic and electrostatic Maxwell equations are used because of the special attention paid to the long-wavelength approximation (phenomenological theory). This leads to the neglect of the interactions between electromagnetic waves and spin waves on the one hand, and between electromagnetic waves and oscillations in the P system on the other hand. This in turn implies that so-called polaritons (Ref. 8, coupled electromagnetic soft optic modes) are reduced here to simple oscillations in the P system which are driven by the electric field and ultimately couple with magnons via the magnetoelectric effect.

The following results emerge from the analysis. For a propagation along the easy axis of magnetization, the initial polarization field being set along this axis and the initial magnetization field being set at an angle $\theta(0 < \theta < \frac{1}{2}\pi)$ with this axis, it is shown in the absence of relaxation that: (i) Transverse polaritons travel independently of the other modes with a typical parabolic dispersion relation whose zero wave-number value is determined by a combination of the optical value of the electric susceptibility and the initial values of the polarization and magnetization fields via the nonlinear ferroelectric behavior and the magnetoelectric coupling; and (ii) longitudinal polaritons present a *resonance*-type of coupling with magnons at a certain critical wave number (which defines a so-called crossover region). The latter coupling is not without recalling the magnetoacoustic resonance effect which shows up in the study of magnon-phonon couplings in elastic ferromagnets (Chap. 4 in Ref. 3; see also Ref. 9) or the resonance effect of the acousto-soft optical type in elastic ferroelectrics.[10] The dispersion branches thus placed in evidence present a *repulsion* of which the magnitude is of the order of the magnetoelectric coupling constant and which vanishes if the initial magnetization is set either parallel to, or at an angle $\frac{1}{2}\pi$ of the initial polarization. The fully phenomenological approach thus given yields results which are quite comparable to those obtained by Bar'yakhtar and Chuppis.[4] These authors have coined the term *"segnetomagnons"* for those bonded ferroelectric-ferromagnetic oscillations. In the case where both ferroelectric and ferromagnetic relaxations are taken into account, it is approximately shown that (i) the crossover region is slightly translated as compared to that of the nondissipative case, and (ii) where the mixed magnon-polariton dispersion branches behave like one of the uncoupled branches, the damping of the coupled branches practically is that of the corresponding uncoupled branch while in the crossover region both branches suffer equal damping which equally results from ferroelectric and ferromagnetic dampings. This equal contribution of both dampings to coupled branches is of the same nature as the phenomenon observed for magnon-phonon couplings in presence of viscosity and spin-lattice relaxation.[11] The ferromagnetic damping is wave-number dependent. The expression obtained for the repulsion of branches in the dissipative case shows that this repulsion is slightly increased as compared with the nondissipative case, but it does not vanish for vanishing magnetoelectric constant or for an alignment of the initial magnetization with the initial polarization as a result of mixed damping. Like in the magnon-phonon case, the resonance coupling placed in evidence in ferroelectric ferromagnets allows us to envisage an *energy-conversion* process in spatially disuniformly magnetized or polarized bodies via magnetoelectric couplings.

II. BASIC EQUATIONS

In order to arrive at the field equations which govern rigid ferroelectric ferromagnets we consider the following variational principle:

$$\delta \int_{\mathcal{B}} \mathcal{L}\, d^3x = \delta\mathcal{H} + \delta\mathcal{R} \ . \tag{2.1}$$

Here $\mathcal{L}$ is a volume Lagrangian density, $\delta\mathcal{H}$ stands for a Hertzian nonholonomic variation which allows us to account for the dynamics of magnetic spins, and $\delta\mathcal{R}$ is the dissipative contribution which is derived from a Rayleigh dissipation potential. $\vec{M}$ and $\vec{P}$ are the volume density of magnetic and electric dipoles, respectively, and $d\vec{M}/dt$ and $d\vec{P}/dt$ are the corresponding time rates. $P=|\vec{P}|=(\vec{P}\cdot\vec{P})^{1/2}$ and $M=|\vec{M}|=(\vec{M}\cdot\vec{M})^{1/2}$ will denote the magnitude of the vectors $\vec{P}$ and $\vec{M}$. Let θ_C and θ_E denote the ferromagnetic and ferroelectric phase-transition temperatures. The range of temperatures θ considered herein after is such that θ is much below both θ_C and θ_E. This in turn implies low levels of energy and a magnetization which has practically reached its saturation value. That is,

$$M[\theta << (\theta_C, \theta_E)] \simeq M_S = \text{const} \ .$$

Therefore, as is also the case in deformable magnetically saturated bodies,[6] $|\vec{M}| = \text{const}$ implies that the variation $\delta\vec{M}$ necessarily is of the form

$$\delta\vec{M} = \delta\vec{\omega} \times \vec{M} \ , \tag{2.2}$$

where $\delta\vec{\omega}$ is an infinitesimal angular vectorial variation.

For the sake of example we consider a ferromagnet of easy axis pointing in the direction of unit vector $\vec{d}$ and we write the Lagrangian $\mathcal{L}$ as

$$\begin{aligned}\mathcal{L} = {} & \tfrac{1}{2}\alpha(\nabla\vec{M})^2 - \tfrac{1}{2}\beta(\vec{M}\cdot\vec{d})^2 - \vec{M}\cdot\vec{H} + \tfrac{1}{2}\chi_\infty^{-1}\vec{P}^2 \\ & + \tfrac{1}{4}\zeta P^4 + \tfrac{1}{2}\lambda(\nabla\vec{P})^2 \\ & + \tfrac{1}{2}d_E\left(\frac{d\vec{P}}{dt}\right)^2 - \vec{P}\cdot\vec{E} - \tfrac{1}{2}\xi P^2(\vec{M}\cdot\vec{d})^2 \ ,\end{aligned} \tag{2.3}$$

where $\vec{E}$ and $\vec{H}$ are the Maxwellian electric and magnetic fields, α is the ferromagnetic exchange constant, $(\nabla\vec{M})^2 = M_{i,j}M_{i,j}$ in rectangular Cartesian components, β is the magnetic anisotropy constant (uniaxial ferromagnets), χ_∞ is the optical value of the electric susceptibility, ζ is the fourth-order electric constant (which measures the nonlinearity of the ferroelectric behavior), λ might be referred to as the ferroelectric "exchange" constant (since it accounts for the ferroelectric ordering), d_E is the polarization inertia, and ξ is the magnetoelectric coupling constant.

If γ is the gyromagnetic ratio of the material, then

$$\delta\mathcal{H} = \int_{\mathcal{B}} \gamma^{-1}\frac{d\vec{M}}{dt}\cdot\delta\vec{\omega}\, d^3x \ . \tag{2.4}$$

This contribution reflects the gyroscopic nature of the magnetic spin. Finally, R being a Rayleigh dissipation potential describing relaxation effects,

$$\delta\mathcal{R}=\int\left[\frac{\partial R}{\partial(d\vec{M}/\partial t)}\cdot\delta\vec{M}+\frac{\partial R}{\partial(d\vec{P}/dt)}\cdot\delta\vec{P}\right]d^3x \quad .(2.5)$$

Consistently with (2.3) and for the sake of example, we consider the following potential R, which respects time reversal:

$$R=\frac{1}{2}\left[\delta_E\left(\frac{d\vec{P}}{dt}\right)^2-\left(\frac{\delta_S}{\omega_M}\right)\left(\frac{d\vec{M}}{dt}\cdot\vec{d}\right)^2\right] , \quad (2.6)$$

where

$$\omega_M=\gamma M_0 \ . \quad (2.7)$$

$M_0=M_S$ is a background reference magnetization. δ_S and δ_E are two coefficients which account for ferromagnetic relaxation (of the Gilbert type)[7] and ferroelectric relaxation, respectively.

With vanishing variations $\delta\vec{\omega}$ and $\delta\vec{P}$ at the regular boundary of the material body—which occupies the region $\mathcal{B}$ of Euclidean physical space—and accounting for (2.1) through (2.6), we find that $\vec{M}$ and $\vec{P}$ are governed, within the bulk of the material, by the equations

$$\frac{d\vec{M}}{dt}=\vec{\Omega}\times\vec{M},\quad \vec{\Omega}=-\gamma\vec{H}^{\rm eff},$$
$$\vec{H}^{\rm eff}=-\left[\frac{\delta\mathcal{L}}{\delta\vec{M}}+\frac{\partial R}{\partial(d\vec{M}/dt)}\right] , \quad (2.8)$$

and

$$\frac{\delta\mathcal{L}}{\delta\vec{P}}+\frac{\partial R}{\partial(d\vec{P}/dt)}=\vec{0} , \quad (2.9)$$

where Euler-lagrange variational derivatives are defined by, e.g.,

$$\frac{\delta\mathcal{L}}{\delta\vec{P}}=\frac{d}{dt}\left[\frac{\partial\mathcal{L}}{\partial(d\vec{P}/dt)}\right]+\left[\frac{\partial\mathcal{L}}{\partial\vec{P}}-\vec{\nabla}\cdot\left[\frac{\partial\mathcal{L}}{\partial\nabla\vec{P}}\right]\right] , \quad (2.10)$$

a similar definition holding true for $\delta\mathcal{L}/\delta\vec{M}$. Thus

$$\frac{d\vec{M}}{dt}=\gamma\vec{M}\times\left[\vec{H}+(\beta+\xi P^2)(\vec{M}\cdot\vec{d})\vec{d}+(\delta_S/\omega_M)\left(\frac{d\vec{M}}{dt}\cdot\vec{d}\right)\vec{d}+\alpha\nabla^2\vec{M}\right] \quad (2.11)$$

and

$$d_E\frac{d^2\vec{P}}{dt^2}+\delta_E\frac{d\vec{P}}{dt}-\vec{E}+[\chi_\infty^{-1}+(\tfrac{1}{2}\zeta)P^2-\xi(\vec{M}\cdot\vec{d})^2]\vec{P}-\lambda\nabla^2\vec{P}=\vec{0} \ . \quad (2.12)$$

Demagnetizing and depolarizing effects have been discarded.

Since we shall only consider the long-wavelength approximation, (2.11) and (2.12) are supplemented with Maxwell's magnetostatic and electrostatic equations in insulators:

$$\vec{\nabla}\cdot(\vec{H}+\vec{M})=0,\quad \vec{\nabla}\times\vec{H}=\vec{0} ,$$
$$\vec{\nabla}\cdot(\vec{E}+\vec{P})=0,\quad \vec{\nabla}\times\vec{E}=\vec{0} \ . \quad (2.13)$$

III. BACKGROUND SOLUTION

In order to study small perturbations superimposed on the fields $\vec{M}$ and $\vec{P}$, we assume that there exists a unique background, static, spatially homogeneous solution S_0, with fields $\vec{H}_0, \vec{E}_0, \vec{P}_0$, and $M_0=M_S$, such that Eqs. (2.11) through (2.14) are identically satisfied. At S_0 (2.11) and (2.12) yield

$$\vec{M}_0\times[\vec{H}_0+(\beta+\xi P_0^2)(\vec{M}_0\cdot\vec{d})\vec{d}]=\vec{0} \quad (3.1)$$

and

$$\vec{E}_0=[\chi_\infty^{-1}+(\tfrac{1}{2}\zeta P_0^2-\xi(\vec{M}_0\cdot\vec{d})^2]\vec{P}_0 \ . \quad (3.2)$$

Equation (3.1) means that there exists a multiplier χ_{m0}, whose dimension is that of a magnetic susceptibility and which is such that

$$\vec{M}_0=\chi_{m0}[\vec{H}_0+(\beta+\xi P_0^2)(\vec{M}_0\cdot\vec{d})\vec{d}] \ . \quad (3.3)$$

We generally assume that $\vec{M}_0$ makes an angle $0<\theta<\frac{1}{2}\pi$ with $\vec{d}$ and $\vec{H}_0$ makes an angle $0<\theta<\psi<\frac{1}{2}\pi$ and $\vec{d}$. It first follows from (3.3) that

$$\vec{M}_0\times\vec{d}=\chi_{mo}\vec{H}_0\times\vec{d} \quad (3.4)$$

and

$$\vec{M}_0\cdot(\vec{H}_0\times\vec{d})=0 \ . \quad (3.5)$$

Thus $\vec{d}$, $\vec{H}_0$, and $\vec{M}_0$ are coplanar. Upon multiplying scalarly both sides of (3.3) by $\vec{M}_0$, we obtain

$$\chi_{mo}^{-1}=(H_0/M_0)\cos(\theta-\psi)+(\beta+\xi P_0^2)\cos^2\theta \ . \quad (3.6)$$

According to (3.2), $\vec{E}_0$ and $\vec{P}_0$ are aligned and we can define a *static* electric susceptibility χ_{po} by

$$\chi_{po}^{-1}=\chi_\infty^{-1}+(\tfrac{1}{2}\zeta)P_0^2-\xi(\vec{M}_0\cdot\vec{d})^2=|\vec{E}_0|/|\vec{P}_0| \ . \quad (3.7)$$

The orientations of $\vec{P}_0$ and $\vec{M}_0$ *a priori* are not correlated.

IV. EQUATIONS FOR PERTURBATIONS

Now let $\vec{m}(\vec{x},t)$, $\vec{p}(\vec{x},t)$, $\vec{h}(\vec{x},t)$, and $\vec{e}(\vec{x},t)$ be time-varying, spatially nonuniform perturbations superimposed on the solution S_0 and such that

$$
\begin{aligned}
|\vec{m}(x,t)| &= |\vec{M}(\vec{x},t)-\vec{M}_0| \ll M_0, \\
|\vec{p}(\vec{x},t)| &= |\vec{P}(\vec{x},t)-\vec{P}_0| \ll P_0 , \\
|\vec{h}(\vec{x},t)| &= |\vec{H}(\vec{x},t)-\vec{H}_0| \ll H_0, \\
|\vec{e}(\vec{x},t)| &= |\vec{E}(\vec{x},t)-\vec{E}_0| \ll E_0 .
\end{aligned}
\tag{4.1}
$$

From (2.13) and (2.14) the standard linearization procedure yields

$$\vec{\nabla}\cdot\vec{h} = -\vec{\nabla}\cdot\vec{m}, \quad \vec{\nabla}\times\vec{h}=\vec{0} \tag{4.2}$$

and

$$\vec{\nabla}\cdot\vec{e} = -\vec{\nabla}\cdot\vec{p}, \quad \vec{\nabla}\times\vec{e}=0 . \tag{4.3}$$

From (2.11) and (2.12) we obtain

$$
\begin{aligned}
\frac{d\vec{m}}{dt} = \gamma\vec{M}_0\times\{\vec{h}&+(\beta+\xi P_0^2)(\vec{m}\cdot\vec{d})\vec{d}-\chi_{mo}^{-1}\vec{m} \\
&+\alpha\nabla^2\vec{m}+(\delta_S/\omega_M)[(d\vec{m}/dt)\cdot\vec{d}]\vec{d} \\
&+2\xi(\vec{P}_0\cdot\vec{p})(\vec{M}_0\cdot\vec{d})\vec{d}\}
\end{aligned}
\tag{4.4}
$$

and

$$
\begin{aligned}
d_E\frac{d^2\vec{p}}{dt^2}+\delta_E\frac{d\vec{p}}{dt}-\vec{e}+\chi_{po}^{-1}\vec{p}+\zeta(\vec{P}_0\cdot\vec{p})\vec{P}_0& \\
-\lambda\nabla^2\vec{p}-2\xi(\vec{M}_0\cdot\vec{d})(\vec{m}\cdot\vec{d})\vec{P}_0&=\vec{0}
\end{aligned}
\tag{4.5}
$$

on account of (3.6) and (3.7). From the saturation condition we have

$$\vec{M}_0\cdot\vec{m}=0 , \tag{4.6}$$

while from (4.4)

$$\vec{M}_0\cdot\frac{d\vec{m}}{dt}=0 . \tag{4.7}$$

V. DYNAMICAL COUPLINGS IN ABSENCE OF RELAXATION

With $\delta_S=\delta_E=0$ and $\vec{M}_0\cdot\vec{d}=M_0\cos\theta$, Eqs. (4.4) and (4.5) reduce to

$$
\begin{aligned}
\frac{d\vec{m}}{dt} = \gamma\vec{M}_0\times[\vec{h}&+(\beta+\xi P_0^2)(\vec{m}\cdot\vec{d})\vec{d}-\chi_{mo}^{-1}\vec{m} \\
&+\alpha\nabla^2\vec{m}+2\xi(\vec{P}_0\cdot\vec{p})M_0\vec{d}\cos\theta]
\end{aligned}
\tag{5.1}
$$

and

$$
\begin{aligned}
d_E\frac{d^2\vec{p}}{dt^2}-\vec{e}+\chi_{po}^{-1}\vec{p}+\zeta(\vec{P}_0\cdot\vec{p})\vec{P}_0-\lambda\nabla^2\vec{p}& \\
-2\xi M_0\cos\theta(\vec{m}\cdot\vec{d})\vec{P}_0&=\vec{0} .
\end{aligned}
\tag{5.2}
$$

In Eqs. (4.2), (4.3), (5.1), and (5.2) we try plane time-harmonic solutions of the type

$$(\vec{m},\vec{p},\vec{h},\vec{e})=(\hat{m},\hat{p},\hat{h},\hat{e})\exp[i(\omega t-\vec{k}\cdot\vec{r})] , \tag{5.3}$$

where ω is a real circular frequency and $\vec{k}$ is the wave vector. $\hat{m},\hat{p}$, etc., are the amplitudes. We set $k=|\vec{k}|$. For the sake of simplicity we consider a propagation along the direction $\vec{d}$, i.e., $\vec{k}=k\vec{d}$, and we assume that S_0 is such that $\vec{P}_0$ is set along $\vec{d}$, i.e., $\vec{P}_0=P_0\vec{d}$. An orthonormal basis $\{\vec{e}_x,\vec{e}_y,\vec{e}_z\}$ is selected in such a way that the z axis lies along $\vec{d}$, the x axis lies along $\vec{M}_0\times\vec{d}$ or $\vec{H}_0\times\vec{d}$, and the z axis completes the triad. Therefore,

$$
\begin{aligned}
&\vec{k}=(0,0,k), \quad \vec{P}_0=(0,0,P_0) , \\
&\vec{M}_0=(0,M_0\sin\theta,M_0\cos\theta) , \\
&\vec{H}_0=(0,H_0\sin\psi,H_0\cos\psi) , \\
&\hat{m}\cdot\vec{d}=\hat{m}_z, \quad \vec{P}_0\cdot\hat{p}=\hat{p}_zP_0 .
\end{aligned}
\tag{5.4}
$$

We shall set

$$\omega_S(k)=\omega_M(\alpha k^2+\chi_{mo}^{-1}) , \tag{5.5}$$

$$
\begin{aligned}
\bar{\omega}_S(k) &= \omega_M[\alpha k^2+(H_0/M_0)\cos(\theta-\psi) \\
&\qquad -(\beta+\xi P_0^2)\sin^2\theta+1] \\
&=\omega_M[\alpha k^2+\chi_{mo}^{-1}+1-(\beta+\xi P_0^2)] ,
\end{aligned}
\tag{5.6}
$$

and

$$\omega_E^2(k)=d_E^{-1}\{\lambda k^2+[\chi_\infty^{-1}+1+(3\zeta/2)P_0^2-\xi M_0^2\cos^2\theta]\} , \tag{5.7}$$

$$\bar{\omega}_E^2(k)=d_E^{-1}\{\lambda k^2+[\chi_\infty^{-1}+(\tfrac{1}{2}\zeta)P_0^2-\xi M_0^2\cos^2\theta]\} . \tag{5.8}$$

These formulas define the fundamental magnon and polariton frequency spectra, respectively.

We call $\hat{p}_\perp$ that vectorial component of $\hat{p}$ which lies in the plane spanned by $\vec{e}_x$ and $\vec{e}_y$. Noting that Maxwell's equations (4.2) and (4.3) classically yield

$$\hat{h}=-(\vec{k}\cdot\hat{m})\vec{k}/k^2, \quad \hat{e}=-(\vec{k}\cdot\hat{p})\vec{k}/k^2 , \tag{5.9}$$

we can write the equations satisfied by the amplitudes $\hat{m}$ and $\hat{p}$ as follows. First for $\hat{p}_\perp$,

$$\hat{p}_\perp[\bar{\omega}_E^2(k)-\omega^2(k)]=0 . \tag{5.10}$$

Next for $\hat{m}$ and $\hat{p}_z$, for which we have the matrix system

$$
\begin{pmatrix}
i\omega & -\omega_S\cos\theta & \bar{\omega}_S\sin\theta & \omega_M\xi P_0M_0\sin2\theta \\
\omega_S\cos\theta & +i\omega & 0 & 0 \\
\omega_S\sin\theta & 0 & -i\omega & 0 \\
0 & 0 & -2\xi d_E^{-1}P_0M_0\cos\theta & (\omega^2-\omega_E^2)
\end{pmatrix}
\begin{pmatrix}\hat{m}_x\\ \hat{m}_y\\ \hat{m}_z\\ \hat{p}_z\end{pmatrix}=(0) , \tag{5.11}
$$

the compatibility condition for solving nontrivially this linear system for the amplitudes is obtained by the vanishing of the determinant of the matrix factor. That is, after some rearrangement and for $\omega \neq 0$:

$$\mathfrak{D}_I(\omega,k,\xi) = (\omega^2-\omega_E^2)(\omega^2-\tilde{\omega}_S^2) - \xi^2 d_E^{-1}\omega_M\omega_S P_0^2 M_0^2 \sin^2 2\theta = 0 , \quad (5.12)$$

where we have set

$$\tilde{\omega}_S^2(k) = \omega_S(k)[\omega_S(k)\cos^2\theta + \bar{\omega}_S(k)\sin^2\theta] . \quad (5.13)$$

If the transverse component $\hat{p}_\perp$ is not zero, then (5.10) implies that *transverse polaritons* propagate with a frequency spectrum given by

$$\omega^2(k) = \bar{\omega}_E^2(k) . \quad (5.14)$$

Whenever either the magnetoelectric coupling constant ξ vanishes or the angle θ is vanishingly small or in the neighborhood of $\frac{1}{2}\pi$, the last contribution in the left-hand side of (5.12) can be discarded; the system (5.11) uncouples for the vector amplitude $\hat{m}$ and the scalar amplitude $\hat{p}_z$. The compatibility condition for the first system yields the dispersion relation for *pure magnons* (coherent magnetic spin oscillations) as

$$\mathfrak{D}_S(\omega,k) = \omega^2 - \tilde{\omega}_S^2(k) = 0 \quad (5.15)$$

for $\hat{m} \neq \vec{0}$, while for $\hat{p}_z \neq 0$, we obtain for *pure longitudinal* polaritons,

$$\omega^2 = \omega_E^2(k) . \quad (5.16)$$

The general systems (5.11) and (5.12), however, indicate a dynamical coupling between magnons and longitudinal polaritons if θ differs from zero and $\frac{1}{2}\pi$ and if ξ is sufficiently large (of course, d_E is supposed to be finite). Let $\omega_I^2(k)$ and $\omega_{II}^2(k)$ be the two solutions of the biquadratic equation (5.12). We have

$$\omega_{I,II}^2(k) = \tfrac{1}{2}\{[\omega_E^2(k)+\tilde{\omega}_S^2(k)] \mp [(\omega_E^2+\tilde{\omega}_S^2)^2 - 4(\omega_E^2\tilde{\omega}_S^2 - \xi^2\mu^2)]^{1/2}\} , \quad (5.17)$$

where we have set

$$\mu^2(k,\theta) \equiv d_E^{-1}P_0^2M_0^2\,\omega_M\omega_S(k)\sin^2 2\theta . \quad (5.18)$$

Equation (5.17) can also be written in the form

$$\omega_{I,II}^2(k) = \tfrac{1}{2}\{(\omega_E^2+\tilde{\omega}_S^2) \mp [(\omega_E^2-\bar{\omega}_S^2)^2 + 4\xi^2\mu^2]^{1/2}\} . \quad (5.19)$$

As k goes to zero we have the following typical behavior:

$$\omega_I^2(k) \simeq \tilde{\omega}_S^2(k) \simeq \omega_S^2(0) \quad (5.20)$$

and

$$\omega_{II}^2(k) \simeq \omega_E^2(k) \simeq \omega_E^2(0) , \quad (5.21)$$

while for large k's we have the asymptotic behavior

$$\omega_I^2(k) \simeq \omega_E^2(k), \quad \omega_{II}^2(k) \simeq \tilde{\omega}_S^2(k) . \quad (5.22)$$

There therefore exists a so-called *crossover region* of the two dispersion branches ($\omega > 0$, $k > 0$) for an intermediate value of k. This crossover region is defined by the critical value $k^*(\theta)$ given by the intersection point of (5.15) and (5.16) at which the energy spectra of pure magnons and pure longitudinal polaritons match, i.e.,

$$\omega_E^2(k) - \bar{\omega}_S^2(k) = 0 \Rightarrow k = k^*(\theta) . \quad (5.23)$$

The *repulsion* of the two branches $\omega_I(k)$ and $\omega_{II}(k)$ which occurs at $k^*(\theta)$ is evaluated as follows:

$$\Delta\omega(k^*) = \omega_{II} - \omega_I = (\omega_{II}^2 - \omega_I^2)/(\omega_{II}+\omega_I) . \quad (5.24)$$

That is, from (5.19)

$$\Delta\omega(k^*) \simeq 2\xi\mu(k^*(\theta),\theta)/[\omega_{II}(k^*)+\omega_I(k^*)] \simeq \xi\mu(k^*(\theta),\theta)/\tilde{\omega}_S(k^*) . \quad (5.25)$$

As $\tilde{\omega}_S(k)$ is very flat (small k) we have

$$\tilde{\omega}_S(k^*) \simeq \omega_S(k^*) \simeq \omega_S(0) \simeq \omega_M , \quad (5.26)$$

so that finally at the first order in ξ

$$\Delta\omega(k^*) = \xi d_E^{-1}P_0M_0\sin 2\theta . \quad (5.27)$$

Thus for θ not zero and $\frac{1}{2}\pi$ and finite d_E the repulsion of branches at k^* behaves like ξ. For finite d_E and prescribed ξ, as θ goes to zero $\Delta\omega(k^*)$ behaves like θ since, then

$$\Delta\omega(k^*) = \xi d_E^{-1}O(\theta) . \quad (5.28)$$

The repulsion phenomenon disappears whenever $\vec{M}_0$ is aligned with $\vec{P}_0$, i.e., $\Delta\omega(k^*(0)) = 0$ or when $\theta = \frac{1}{2}\pi$. In the first case we have

$$\tilde{\omega}_S^2(k) = \omega_S^2(k) = \omega_M^2[\alpha k^2 + (\beta + \xi P_0^2) + (H_0/M_0)\cos\psi]^2 . \quad (5.29)$$

Obviously, $\Delta\omega$ diverges as d_E goes to zero. Note finally that the repulsion phenomenon is accompanied, in the absence of dissipation, by a *resonance* for the amplitudes. Indeed, on eliminating $\hat{m}_y$ and $\hat{m}_z$ from the matrix system (5.11), we find that

$$|\hat{m}_x/\hat{p}_z| = \xi\omega(k)\omega_M P_0M_0\sin 2\theta[\omega^2(k) - \tilde{\omega}_S^2(k)]^{-1} . \quad (5.30)$$

This blows up when $\omega^2(k)$ approaches $\omega_E^2(k)$ in the neighborhood of k^* (with $\theta \neq 0$ and $\frac{1}{2}\pi$, ξ finite as well as d_E).

In conclusion we have sketeched out a fully phenomenological approach which is equivalent to the quantum statistical approach of Bar'yarkhtar and Chuppis.[4] Our general equations (4.4) and (4.5), however, allow us to carry on the study when coupled relaxation effects are taken into account.

VI. DYNAMICAL COUPLINGS IN PRESENCE OF RELAXATION

Now we consider complex ω's with

$$\omega=\Omega+i\Gamma,\quad \Omega=\mathrm{Re}(\omega),\quad \Gamma=\tau^{-1}=\mathrm{Im}(\omega)\ , \tag{6.1}$$

where Ω is the *real* circular frequency and τ is the relaxation time. Both are real functions of the *real* wave number k. We set

$$\Gamma_E=\tfrac{1}{2}(\delta_E/d_E)\ , \tag{6.2}$$

$$\Gamma_S(k,\theta)=\tfrac{1}{2}\delta_S\omega_S(k)\sin^2\theta\ , \tag{6.3}$$

$$\Omega_E^2(k)=\omega_E^2(k)+\Gamma^2(k)-2\Gamma_E\Gamma(k)\ , \tag{6.4}$$

$$\overline{\Omega}_E^2(k)=\overline{\omega}_E^2(k)+\Gamma^2(k)-2\Gamma_E\Gamma(k)\ , \tag{6.5}$$

and

$$\tilde{\Omega}_S^2(k)=\tilde{\omega}_S^2(k)+\Gamma^2(k)-2\Gamma_S(k)\Gamma(k)\ . \tag{6.6}$$

Equations (6.4) through (6.6) assume beforehand that $\Gamma(k)$ has been determined, which is not the case. It is immediately shown that (5.10) is replaced by

$$\hat{p}_\perp[(\Omega^2-\overline{\Omega}_E^2)+2i\Omega(\Gamma-\Gamma_E)]=0\ , \tag{6.7}$$

while the corresponding z component reads

$$\hat{p}_z[(\Omega^2-\Omega_E^2)+2i\Omega(\Gamma-\Gamma_E)]-2d_E^{-1}\xi P_0M_0(\cos\theta)\hat{m}_z=0\ . \tag{6.8}$$

For pure transverse polaritons ($\hat{p}_\perp\neq\vec{0}$), Eq. (6.7) yields

$$\Gamma=\Gamma_E,\quad \Omega^2=\overline{\Omega}_E^2=\overline{\omega}_E^2-\Gamma_E^2\ . \tag{6.9}$$

For coupled magnons and longitudinal polaritons, in place of (5.12) we obtain

$$\mathfrak{D}_1'(\Omega,k;\xi)=[(\Omega^2-\Omega_E^2)+2i\Omega(\Gamma-\Gamma_E)][(\Omega^2-\tilde{\Omega}_S^2)+2i\Omega(\Gamma-\Gamma_S)]-d_E^{-1}\xi^2P_0^2M_0^2\,\omega_M\omega_S\sin^2 2\theta=0\ . \tag{6.10}$$

On separating the real and imaginary parts of this equation we obtain a biquadratic equation determining the real frequencies $\Omega(k)$, once $\Gamma(k)$ is known, in the form

$$(\Omega^2-\Omega_E^2)(\Omega^2-\tilde{\Omega}_S^2)-4\Omega^2(\Gamma-\Gamma_E)(\Gamma-\Gamma_S)-\xi^2\mu^2=0\ , \tag{6.11}$$

where μ^2 is defined as in (5.18), and the equation determining $\Gamma(k)$ as

$$\Gamma(k)=\frac{\Gamma_E(\Omega^2-\tilde{\Omega}_S^2)+\Gamma_S(\Omega^2-\Omega_E^2)}{2\Omega^2-(\tilde{\Omega}_S^2+\Omega_E^2)} \tag{6.12}$$

if $\Omega(k)$ is known.

The solutions of (6.11) are shown to be

$$\Omega_{\mathrm{I,II}}^2(k)=\tfrac{1}{2}(\{\Omega_E^2+\tilde{\Omega}_S^2+4(\Gamma-\Gamma_E)(\Gamma-\Gamma_S)\}\mp\{(\Omega_E^2-\tilde{\Omega}_S^2)^2+4[\xi^2\mu^2+4(\Gamma-\Gamma_E)^2(\Gamma-\Gamma_S)^2+2(\Omega_E^2+\tilde{\Omega}_S^2)(\Gamma-\Gamma_E)(\Gamma-\Gamma_S)]\}^{1/2})\ . \tag{6.13}$$

Note that if $\Omega\simeq\Omega_E$ (6.12) yields

$$\Gamma\simeq\Gamma_E\ , \tag{6.14}$$

whereas if $\Omega\simeq\tilde{\Omega}_S$, then from the same equation we have

$$\Gamma\simeq\Gamma_S(k)\ . \tag{6.15}$$

Therefore,

$$\Omega_E^2(k)\simeq\omega_E^2(k)-\Gamma_E^2 \tag{6.16}$$

and

$$\tilde{\Omega}_S^2(k)\simeq\tilde{\omega}_S^2(k)-\Gamma_S^2(k)\ . \tag{6.17}$$

This means that in the presence of relaxation the crossover region is defined by

$$\omega_E^2-\Gamma_E^2=\tilde{\omega}_S^2(k)-\Gamma_S^2(k)\Rightarrow k=k^{**}(\theta)\ . \tag{6.18}$$

Because of the smallness of the Γ's involved the critical wave number k^{**} cannot differ very much from that corresponding to the nondissipative case [k^*; cf. Eq. (5.23)].

If $\Omega\simeq\tilde{\Omega}_S$ in the neighborhood of k^{**}, then (6.12) yields

$$\Gamma(k)\simeq\tfrac{1}{2}[\Gamma_E+\Gamma_S(k)]\ . \tag{6.19}$$

This means that both branches $\Omega_{\mathrm{I}}(k)$ and $\Omega_{\mathrm{II}}(k)$ in the crossover region have equal damping which *equally* results from ferroelectric and ferromagnetic relaxations. At $k=k^{**}$ we have thus

$$\Omega^2_{\mathrm{I,II}} = \tfrac{1}{2}[(\Omega_E^2+\tilde{\Omega}_S^2)-(\Gamma_E-\Gamma_S)^2] \mp [\xi^2\mu^2+\tfrac{1}{4}(\Gamma_E-\Gamma_S)^4+4\tilde{\Omega}_S^2(\Gamma_E-\Gamma_S)^2]^{1/2} \; . \tag{6.20}$$

The repulsion of branches at k^{**} is given by

$$\Delta\Omega(k^{**}) = \left.\frac{\Omega_{\mathrm{II}}^2-\Omega_{\mathrm{I}}^2}{\Omega_{\mathrm{II}}+\Omega_{\mathrm{I}}}\right|_{k^{**}} \simeq \left.\frac{\Omega_{\mathrm{II}}^2-\Omega_{\mathrm{I}}^2}{2\tilde{\Omega}_S}\right|_{k^{**}} \; . \tag{6.21}$$

That is

$$\begin{aligned}\Delta\Omega(k^{**}) &\simeq \tilde{\Omega}_S^{-1}[\xi^2\mu^2+\tfrac{1}{4}(\Gamma_E-\Gamma_S)^4 - 4\tilde{\Omega}_S^2(\Gamma_E-\Gamma_S)^2]^{1/2} \\ &= \left[\frac{\xi^2\mu^2+(\Gamma_E-\Gamma_S)^4/4}{\omega_M^2}+4(\Gamma_E-\Gamma_S)^2\right]^{1/2}\end{aligned} \tag{6.22}$$

or, on account of the smallness of the Γ's,

$$\Delta\Omega(k^{**}) \simeq (\xi\mu/\omega_M)+\frac{1}{2}\left[\frac{(\Gamma_E-\Gamma_S)^4}{\omega_M^2}+4(\Gamma_E-\Gamma_S)^2\right] \; . \tag{6.23}$$

As compared to the nondissipative case (and for $\theta\in[0,\frac{1}{2}\pi]$, ξ and d_E finite) the value of the repulsion is slightly increased. The important point, however, is that a slight repulsion occurs even when θ equals 0 or $\frac{1}{2}\pi$.

Proceeding as in Sec. V we find that the resonance condition (5.29) is now replaced by

$$\left|\frac{\hat{m}_x}{\hat{p}_z}\right| = \left|\frac{\xi(\Omega^2-\Gamma^2)^{1/2}\omega_M P_0 M_0 \sin 2\theta}{[(\Omega^2-\Omega_S^2)-4\Omega^2(\Gamma-\Gamma_S^2)]^{1/2}}\right| \; . \tag{6.24}$$

In the crossover region, on account of (6.19), we find that this reduces to the finite value

$$\left|\frac{\hat{m}_x}{\hat{p}_z}\right| \simeq \xi\omega_M \frac{P_0 M_0 \sin 2\theta}{|\Gamma_S(k^{**})-\Gamma_E|} \tag{6.25}$$

if $\Gamma_S(k^{**})$ does not match with Γ_E.

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