Surface and volume modes of polarization waves in ferroelectric films

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We investigate the characteristic modes of polarization waves in ferroelectric films. This is motivated by the recent surge of interest in the excitations of the ferroelectric order inspired by the duality between electric dipoles in ferroelectrics and magnetic dipoles in ferromagnets that has led to the introduction of the area of ferronics by analogy to magnonics. We report that a ferroelectric film supports surface and volume modes of polarization waves, analogous to the surface and volume magnetostatic spin-wave modes in a ferromagnetic film. However, while in ferromagnets each type of mode has only one (positive) frequency band, in a ferroelectric film the surface and volume modes have two frequency bands each. We present the dependence of the frequencies on the wave vector for both modes for the parameters of the classic ferroelectric $LiNbO_3$, with polarization either parallel or perpendicular to the film. The frequencies lie in the low terahertz band that is experimentally accessible.

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I. INTRODUCTION

In recent years there has been a growing interest in studies of polarization waves in ferroelectric materials, whose quanta have been called ferrons [1–13]. Predicted properties, such as polarization transport by thermal gradients or ferronic Seebeck effect [2,6], analogous to the spin Seebeck effect [14–21], have yet to be unequivocally observed. Among the motivations for the recent surge in interest in polarization waves are the analogies with the well-studied spin waves, or magnons, in ferromagnets. While in ferromagnets a spin wave consists of magnetic dipoles precessing about an equilibrium direction [20] with a phase that varies in space, in ferroelectrics a polarization wave consists of oscillating electric dipoles with a finite wave vector [2,3].

In ferromagnets, the most important interaction between neighboring spins arises from the exchange energy. In most parts of the Brillouin zone, this interaction dominates the frequency versus wave number relation of the spin waves, called a dispersion relation [20]. However, for very small wave numbers k, neighboring spins are essentially parallel to each other, so that the exchange interaction has no influence on the frequency. In this case, for spin excitations with wavelength $\lambda = 2\pi/k$ comparable to the sample dimensions, the frequencies and mode configurations are determined by the long-range dipolar interactions between the magnetic moments subject to boundary conditions at the sample surfaces. These so-called magnetostatic waves obey $\nabla \times \vec{H} = 0$, where $\vec{H}$ is the associated magnetic stray field and are governed by the Walker equation for the magnetic potential [22]. They have been extensively studied in samples of various shapes, both theoretically and experimentally, since their frequencies lie in convenient microwave ranges [20,22–39].

The fact that magnetism and ferroelectricity break, respectively, time-reversal and space-inversion symmetry [40–43], leads to analogies as well as differences of dynamic phenomena in the two areas [2,9,13]. In magnets (ferroelectrics) magnetic (electric) dipoles order below some critical temperature in the ordered phase spin (polarization) waves [2,3, 6–13]. However, while in magnets there are no controversies regarding the nature of the excitations, in the case of ferroelectrics the situation is not so clear cut because in most ferroelectrics the excitation of the electric dipoles necessarily involves lattice vibrations.

Here we present the results of an investigation of polarization waves in ferroelectric films. We show that a ferroelectric film supports surface and volume modes, analogous to the surface and volume magnetostatic spin-wave modes in ferromagnetic films. In ferromagnets each type of mode has only one frequency band. However, in a ferroelectric film, polarized either parallel or perpendicular to the plane, the surface and volumes modes have two frequency bands each. Calculations of the dispersion relations for both modes for the parameters of the classic ferroelectric $LiNbO_3$ show frequencies in the low terahertz range that await experimental observation.

After a general discussion of the electrodynamics of excited ferroelectric films in Sec. II, we discuss films with in-plane equilibrium polarization in Secs. III (surface waves) and IV (bulk waves) as well as those with perpendicular polarization in Sec. V, followed by an assessment of the results in Sec. VI.

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II. THE ELECTRIC POTENTIAL IN FERROELECTRICS

We wish to study polarization waves with wave number k in the range $\omega/c \ll k \ll 1/a$ (c is the speed of light and a is the lattice parameter), which is small enough to make the boundary conditions in the film important but is larger than the photon wave number, such that we can disregard the ac magnetic field and polariton effects [3,13]. The electric field $\vec{E}$ and the displacement field $\vec{D}$ obey Maxwell's equations in the electrostatic limit, viz.,

$$\nabla \cdot \vec{D} = 0, \quad \nabla \times \vec{E} = 0. \tag{1}$$

In order to solve Eqs. (1), we need a constitutive relation between the field $\vec{E}$ and the polarization $\vec{P}$. Considering a cubic ferroelectric material polarized in the y direction, the linearized relations between the components of the polarization vector and the electric field read [7]

$$(1/\Omega_p)^2 \frac{\partial^2 P_x}{\partial t^2} + K_\perp P_x = \varepsilon_0 E_x, \tag{2a}$$

$$(1/\Omega_p)^2 \frac{\partial^2 P_y}{\partial t^2} + K_\parallel P_y = \varepsilon_0 E_y, \tag{2b}$$

$$(1/\Omega_p)^2 \frac{\partial^2 P_z}{\partial t^2} + K_\perp P_z = \varepsilon_0 E_z, \tag{2c}$$

where Ω_p is the ionic plasma frequency,

$$K_\perp = \varepsilon_0 \lambda \quad \text{and} \quad K_\parallel = \varepsilon_0(\alpha + 3\beta P_0) \tag{3}$$

parameterize the stiffness of the transverse and longitudinal fluctuations, ε_0 is the vacuum permittivity, λ, α, and β are parameters of the Landau free energy, and P_0 is the polarization of the ferroelectric material [7]. With harmonic electric field and polarization $\vec{E}, \vec{P} = \mathrm{Re}[\vec{E}(\vec{r}), \vec{P}(\vec{r})\, e^{-i\omega t}]$, we can write Eqs. (2a)–(2c) in the matrix form

$$\vec{P} = \begin{bmatrix} 1/c_1 & 0 & 0 \\ 0 & 1/c_2 & 0 \\ 0 & 0 & 1/c_1 \end{bmatrix} \varepsilon_0 \vec{E}, \tag{4a}$$

where

$$c_1 = -(\omega/\Omega_p)^2 + K_\perp, \quad c_2 = -(\omega/\Omega_p)^2 + K_\parallel. \tag{4b}$$

Since $\vec{D} = \varepsilon_0 \vec{E} + \vec{P}$,

$$\vec{D} = \begin{bmatrix} (1 + 1/c_1) & 0 & 0 \\ 0 & (1 + 1/c_2) & 0 \\ 0 & 0 & (1 + 1/c_1) \end{bmatrix} \varepsilon_0 \vec{E}. \tag{5}$$

One important difference with magnets is the absence of the off-diagonal imaginary elements in the tensor relation between $\vec{B}$ and $\vec{H}$. The latter is a direct consequence of the time-reversal symmetry breaking that gives rise to the chiral properties of magnetization waves. Introduce the electric potential ψ defined as

$$\vec{E} = -\nabla \psi. \tag{6}$$

According to Eqs. (1), (5), and (6), the electric potential in a ferroelectric material with polarization in the y direction obeys the wave equation

$$(1 + 1/c_1)\left(\frac{\partial^2 \psi}{\partial x^2} + \frac{\partial^2 \psi}{\partial z^2}\right) + (1 + 1/c_2)\frac{\partial^2 \psi}{\partial y^2} = 0\,. \tag{7}$$

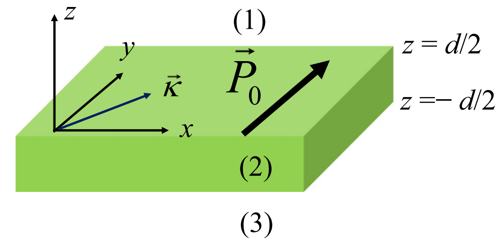

FIG. 1. Illustration of a ferroelectric film with polarization in the plane.

Interestingly, this equation is very similar to the Walker equation for the magnetic potential in the magnetostatic regime [22]:

$$(1 + \chi)\left(\frac{\partial^2 \psi}{\partial x^2} + \frac{\partial^2 \psi}{\partial z^2}\right) + \frac{\partial^2 \psi}{\partial y^2} = 0\,, \tag{8}$$

where χ is a parameter that depends on the frequency and the magnetization of the material. In regions outside the ferroelectric (or ferromagnetic) sample, the potential obeys the Laplace equation

$$\nabla^2 \psi = \frac{\partial^2 \psi}{\partial x^2} + \frac{\partial^2 \psi}{\partial y^2} + \frac{\partial^2 \psi}{\partial z^2} = 0. \tag{9}$$

III. SOLUTIONS FOR A FERROELECTRIC FILM POLARIZED IN THE PLANE

Consider a ferroelectric film of thickness d with polarization vector $\vec{P}_0$ in the (in-plane) y direction, as shown in Fig. 1.

The electric potential is expressed by different functions in the three regions of space, inside and outside the film: $\psi_1(\vec{r})$ in region (1), $z > d/2$; $\psi_2(\vec{r})$ in region (2), $-d/2 < z < d/2$; and $\psi_3(\vec{r})$ in region (3), $z < -d/2$. In all regions Eqs. (7) and (9) can be solved by the method of separation of variables,

$$\psi_\lambda\,(x, y, z) = X_\lambda(x)\, Y_\lambda(y)\, Z_\lambda(z), \tag{10}$$

where each function depends on only one variable. We consider propagating waves in the film plane, with wave vector $\vec{\kappa} = \hat{x}\, k_x + \hat{y}\, k_y$. Thus, in order to satisfy the tangential boundary conditions at the film surfaces, in all regions

$$X_\lambda(x)\, Y_\lambda(y) = e^{ik_x x} e^{ik_y y}. \tag{11}$$

The solutions for the function $Z_\lambda(z)$ are $e^{\pm k_z z}$ in regions (1) and (3) with signs chosen to vanish for $z \to \pm\infty$, while in region (2) we consider harmonic solutions. Hence

$$\psi_1(\vec{r}) \;=\; C e^{-k_z^{(e)} z} e^{ik_x x} e^{ik_y y}, \tag{12a}$$

$$\psi_2\,(\vec{r}) = [A\,\sin(k_z z) + B\,\cos(k_z z)]\, e^{ik_x x} e^{ik_y y}, \tag{12b}$$

$$\psi_3(\vec{r}) \;=\; D e^{k_z^{(e)} z} e^{ik_x x} e^{ik_y y}, \tag{12c}$$

where $k_z^{(e)}$ and k_z denote, respectively, the wave numbers in the exponents in $Z_\lambda(z)$ outside and inside the film. The four coefficients in Eq. (12) are determined by the boundary conditions at the surfaces, viz., continuity of the tangential component of $\vec{E}$ and continuity of the normal component of $\vec{D}$. The first condition implies continuity of the electric potential, i.e., $\psi_1 = \psi_2$ at $z = d/2$ and $\psi_2 = \psi_3$ at $z = -d/2$. The second conditions read $D_z = -(1 + 1/c_1)\varepsilon_0 \partial\psi/\partial z$ inside, and

$D_z = -\varepsilon_0 \partial\psi/\partial z$ outside the film. In matrix form

$$\begin{bmatrix} \sin(k_z d/2) & \cos(k_z d/2) & -e^{-k_z^{(e)} d/2} & 0 \\ -\sin(k_z d/2) & \cos(k_z d/2) & 0 & e^{-k_z^{(e)} d/2} \\ (1+1/c_1)\,k_z \cos(k_z d/2) & -(1+1/c_1)\,k_z \sin(k_z d/2) & k_z^{(e)} e^{-k_z^{(e)} d/2} & 0 \\ (1+1/c_1)\,k_z \cos(k_z d/2) & (1+1/c_1)\,k_z \sin(k_z d/2) & 0 & -k_z^{(e)} e^{-k_z^{(e)} d/2} \end{bmatrix} \begin{pmatrix} A \\ B \\ C \\ D \end{pmatrix} = 0. \tag{13}$$

We need two additional relations that eliminate $k_z^{(e)}$ and k_z. When substituting either (12a) or (12c) in the Laplace equation (9),

$$k_x^2 + k_y^2 - k_z^{(e)2} = 0, \tag{14}$$

while substituting (12b) in Eq. (7) gives

$$c_1 c_2 k^2 + c_2\left(k_x^2 + k_z^2\right) + c_1 k_y^2 = 0, \tag{15}$$

where $k^2 = k_x^2 + k_y^2 + k_z^2$. This relation is the same as Eq. (7) of Ref. [7], obtained with a Green's function approach to calculate the electric field. Expanding the determinant of the matrix (13), using the relation $k_z^{(e)} = \kappa$ obtained from Eq. (14), the condition $\det = 0$ gives

$$2\kappa(c_1+1)c_1 k_z \cos(k_z d) + \left[c_1^2\kappa^2 - (c_1+1)^2 k_z^2\right]\sin(k_z d) = 0, \tag{16}$$

which agrees with Eq. (8) of Ref. [7]. The solutions of Eqs. (15) and (16) are the dispersion relations for polarization waves in a ferroelectric film with arbitrary thickness d. For a complex wavenumber $k_z = \eta_1 + i\eta_2$ and using

$$\sin(k_z d) = \sin(\eta_1 d)\cosh(\eta_2 d) + i\,\cos(\eta_1 d)\sinh(\eta_2 d), \tag{17a}$$

$$\cos(k_z d) = \cos(\eta_1 d)\cosh(\eta_2 d) - i\,\sin(\eta_1 d)\,\sinh(\eta_2 d), \tag{17b}$$

Eq. (16) leads to two equations, for the real and imaginary parts, viz.,

$$\begin{aligned}&\left[\left(\eta_2^2-\eta_1^2\right)(c_1+1)^2 + c_1^2\kappa^2\right]\sin(\eta_1 d)\,\cosh(\eta_2 d) + 2\eta_1\eta_2(c_1+1)^2\cos(\eta_1 d)\,\sinh(\eta_2 d)\\ &\quad + 2\kappa(c_1+1)c_1\eta_1\cos(\eta_1 d)\cosh(\eta_2 d) + 2\kappa(c_1+1)c_1\eta_2\sin(\eta_1 d)\sinh(\eta_2 d) = 0,\end{aligned} \tag{18a}$$

$$\begin{aligned}&\left[\left(\eta_2^2-\eta_1^2\right)(c_1+1)^2 + c_1^2\kappa^2\right]\cos(\eta_1 d)\sinh(\eta_2 d) - 2\eta_1\eta_2(c_1+1)^2\sin(\eta_1 d)\cosh(\eta_2 d)\\ &\quad - 2\kappa(c_1+1)c_1\eta_1\sin(\eta_1 d)\,\sinh(\eta_2 d) + 2\kappa(c_1+1)c_1\eta_2\cos(\eta_1 d)\cosh(\eta_2 d) = 0.\end{aligned} \tag{18b}$$

Dividing both equations by $\cosh(\eta_2 d)$ and rearranging terms we obtain two transcendental characteristic equations:

$$\begin{aligned}&\left[\left(\eta_2^2-\eta_1^2\right)(c_1+1)^2 + c_1^2\kappa^2 + 2\kappa(c_1+1)c_1\eta_2\tan(\eta_2 d)\right]\sin(\eta_1 d)\\ &\quad + \left[2\kappa(c_1+1)c_1\eta_1 + 2\eta_1\eta_2(c_1+1)^2\tanh(\eta_2 d)\right]\cos(\eta_1 d) = 0,\end{aligned} \tag{19a}$$

$$\begin{aligned}&\left[\left(\eta_2^2-\eta_1^2\right)(c_1+1)^2 + c_1^2\kappa^2\right]\tanh(\eta_2 d) + 2\kappa(c_1+1)c_1\eta_2\right]\cos(\eta_1 d)\\ &\quad - \left[2\eta_1\eta_2(c_1+1)^2 + 2\kappa(c_1+1)c_1\eta_1\tanh(\eta_2 d)\right]\sin(\eta_1 d) = 0.\end{aligned} \tag{19b}$$

Equations (15) and (19) give the frequency of polarization waves for arbitrary wave vectors in a ferroelectric film with any thickness.

IV. DISPERSION RELATIONS FOR SURFACE AND VOLUME MODES

Ferroelectrics support waves of either surface or volume nature [44]. In this section we derive the dispersion relations for both modes in a ferroelectric film polarized in the plane and calculate them for the parameters of the classic ferroelectric $LiNbO_3$.

A. Surface waves

The dispersion relations for surface waves follow from Eq. (19b) by setting $\eta_1 = 0$. In this case, k_z is imaginary and the modes are evanescent with exponentially decaying amplitudes as a function of distance from one of the surfaces, i.e., surface modes. For $\eta_1 = 0$, Eq. (19b) can be written as

$$2c_1(c_1+1)\eta_2\kappa + \left[(c_1+1)^2\eta_2^2 + c_1^2\kappa^2\right]\tanh(\eta_2 d) = 0, \tag{20}$$

where

$$\kappa = \left(k_x^2 + k_y^2\right)^{1/2}, \tag{21}$$

and the coefficients c_1 and c_2 as defined by Eq. (4b) are functions of frequency. On the other hand, Eq. (15) reads in this case as

$$\eta_2 = \pm\left[k_x^2 + \frac{c_1(c_2+1)}{c_2(c_1+1)}k_y^2\right]^{1/2}. \tag{22}$$

Equations (20)–(22) can be solved numerically for the dispersion relations of the surface waves. When propagating along the polarization in the ferroelectric film, the surface modes

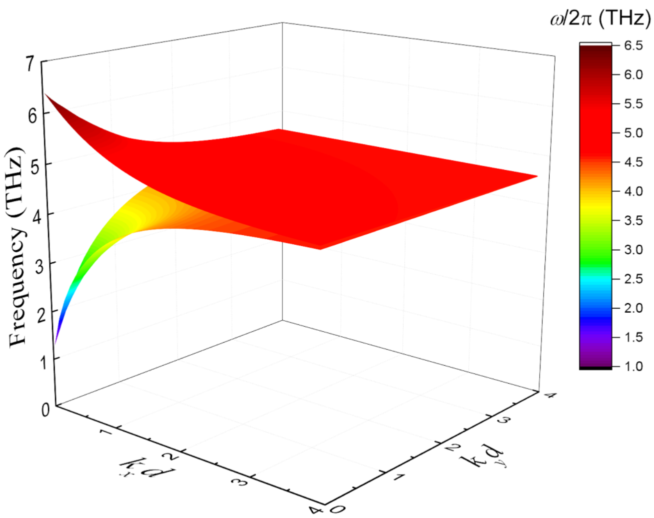

FIG. 2. Dispersion relations of surface mode polarization waves in a ferroelectric $LiNbO_3$ film polarized in the y direction, with wave vectors in the film plane.

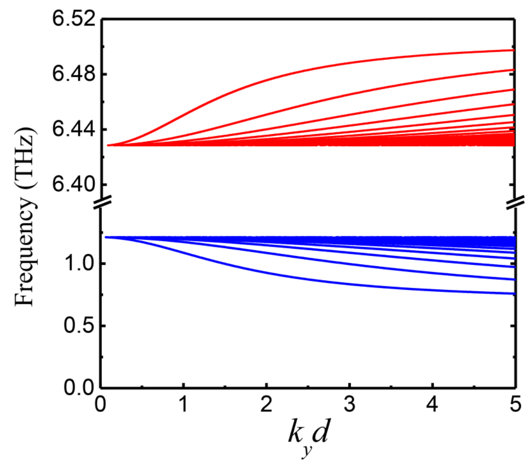

FIG. 3. Dispersion relations of volume polarization waves in a ferroelectric $LiNbO_3$ film propagating in the y direction along the equilibrium polarization.

have two branches. For $k_x \to 0$, $k_y \to 0$, the higher (upper) and lower frequencies are

$$\omega_{0l} = (K_\parallel)^{1/2}\Omega_p,$$
$$\omega_{0u} = (K_\perp + 1)^{1/2}\Omega_p. \quad (23)$$

For $k_y \to 0$ and $k_x d \gg 1$, or $k_x \to 0$ and $k_y d \gg 1$, the two frequencies converge to

$$\omega_\infty = \left(\frac{K_\perp + K_\parallel + 1}{2}\right)^{1/2}\Omega_p. \quad (24)$$

Let us apply the results obtained to $LiNbO_3$, a classic insulating ferroelectric at room temperature. Its primitive cell contains two formula units that allow 27 degrees of freedom to be assigned to optical-phonon modes, that have been identified by Raman and neutron inelastic scattering, and calculated in detail with lattice dynamics [45–47]. Considering the longitudinal and transverse optical phonons with frequencies $\Omega_{LO} = 9.7$ THz and $\Omega_{TO} = 7.3$ THz, and using the expression for the plasma frequency $\Omega_p^2 = \Omega_{LO}^2 - \Omega_{TO}^2$ [48,49], we obtain $\Omega_p = 6.39$ THz. As in Ref. [7], using the parameters for the Landau free energy from Ref. [50], $\alpha = -2.012\,\mathrm{J\,m/C^2}$, $\beta = 3.608\,\mathrm{J\,m^5/C^4}$, $\lambda = 1.345 \times 10^9\,\mathrm{J\,m/C^2}$, and the polarization at room temperature $P_0 = 0.746\,\mathrm{C/m^2}$, with Eq. (3) we find $K_\perp = 0.012$ and $K_\parallel = 0.036$. We solve Eqs. (20)–(22) for these parameters numerically.

Figure 2 shows a plot of the frequency dispersion relations of the surface waves in a $LiNbO_3$ film polarized in the plane as a function of the in-plane components of the wave vector times film thickness. There are two bands of surface modes: those at lower frequencies are forward-moving waves with positive group velocities, while those at higher frequencies are backward-moving waves with negative group velocities. Magnetostatic waves in ferromagnetic films magnetized in the plane have only one band of purely forward-moving surface waves that are chiral, i.e., when the in-plane wave vectors are normal to the magnetization they can propagate on a given surface in only one direction. Here, the polarization waves may always propagate in opposite directions in the plane.

The limiting values of the frequencies for small and large wave vectors, Eqs. (23) and (24), become for $LiNbO_3$ $\omega_{0l} = 1.212$ THz, $\omega_{0u} = 6.428$ THz, and $\omega_\infty = 4.626$ THz, in quite good agreement with the values in Fig. 2.

B. Volume waves

The volume modes in a ferroelectric film are characterized by a real k_z inside the film. Using $\eta_2 = 0$ and setting $k_z = \eta_1$ in Eq. (19a), we obtain a transcendental equation

$$2\kappa k_z c_1(c_1 + 1) + \left[c_1^2\kappa^2 - k_z^2(c_1 + 1)^2\right]\tan(k_z d) = 0, \quad (25)$$

where $\kappa^2 = k_x^2 + k_y^2$. Note that the periodicity of the $\tan(k_z d)$ function leads to multiple solutions that represent the ladder of standing waves in the z direction. The full dispersion relations of the volume modes are obtained by solving this equation together with Eq. (15), that can be written as

$$c_2(c_1 + 1)k_x^2 + c_1(c_2 + 1)k_y^2 + c_1 c_2 k_z^2 = 0. \quad (26)$$

Equations (25) and (26) can be solved numerically by fixing a pair of values for $k_x d$, $k_y d$ and finding the multiple roots of Eq. (25) that correspond to different values of k_z, i.e., perpendicular standing-wave modes. In contrast to the surface modes, the bulk waves propagating normal to the polarization vector, $k_y = 0$, do not depend on k_x because the electric fields generated by the dynamic bound charges are perpendicular to the polarization. The dispersion relations of waves propagating along the polarization vector, $k_x = 0$, are shown in Fig. 3. They have two manifolds, one for backward-moving waves at lower frequencies and one for forward-moving ones at higher frequencies. The dispersion relations for the surface waves in Fig. 2 lie in the gap of the volume waves, which is again markedly different from the volume magnetostatic modes in a ferromagnetic film, with a single manifold of backward-moving waves, similar to the lower branch in Fig. 3.

The frequency gap of the volume modes follows from Eqs. (25) and (26) by setting $k_x = k_y = 0$. The solutions are $c_1 + 1 = 0$ or $c_2 = 0$, which with Eqs. (4) give

$$\omega_{0l} = \sqrt{K_\parallel}\,\Omega_p,$$
$$\omega_{0u} = \sqrt{1 + K_\perp}\,\Omega_p. \tag{27}$$

Using the parameter values for $LiNbO_3$ we obtain $\omega_{0l} = 1.212$ THz and $\omega_{0u} = 6.428$ THz. The asymptotic frequencies for $k_x = 0$ and $k_y \to \infty$ of Eqs. (25) and (26) are

$$\omega_{\infty l} = \sqrt{K_\perp}\,\Omega_p,$$
$$\omega_{\infty u} = \sqrt{1 + K_\parallel}\,\Omega_p. \tag{28}$$

For $LiNbO_3$ we obtain $\omega_{0l} = 1.212$ THz, $\omega_{0u} = 6.428$ THz, $\omega_{\infty l} = 0.70$, and $\omega_{\infty u} = 6.504$ THz.

V. POLARIZATION WAVES IN PERPENDICULARLY POLARIZED FILMS

In Secs. III and IV we focus on films polarized in the plane. Analogously to the magnetostatic waves in ferromagnetic films [20,22,28,32,37], volume and surface polarization waves can be supported by ferroelectric films with polarization vector in any direction. Let us consider now a ferroelectric film with polarization vector $\vec{P}_0$ perpendicular to the plane, pointing along the z direction in Fig. 1. The properties of the polarization waves can be calculated as before, but with an equation for the electric potential that differs from Eq. (7). The matrix relation for the ac components of the electric field and the displacement vector now reads

$$\vec{D} = \begin{bmatrix} (1+1/c_1) & 0 & 0 \\ 0 & (1+1/c_1) & 0 \\ 0 & 0 & (1+1/c_2) \end{bmatrix} \varepsilon_0 \vec{E}, \tag{29}$$

and an electric potential that is governed by

$$(1+1/c_1)\left(\frac{\partial^2\psi}{\partial x^2} + \frac{\partial^2\psi}{\partial y^2}\right) + (1+1/c_2)\frac{\partial^2\psi}{\partial z^2} = 0. \tag{30}$$

As in Sec. III, the coefficients of the solutions in the film and the vacuum are determined by the electric boundary conditions. The secular equation is similar to Eq. (13) and using the relation $k_z^{(e)} = \kappa$ obtained from the Laplace equation (9), gives

$$2\,\kappa k_z\,(c_2+1)c_2\cos(k_z z) + \left[c_2^2\kappa^2 - (c_2+1)^2 k_z^2\right]\sin(k_z z) = 0. \tag{31}$$

Replacing c_2 by c_1 would lead to Eq. (16) for in-plane polarization. Since k_z is complex, we can use Eqs. (19a) and (19b) by exchanging c_2 and c_1. Substituting the expression for the potential inside the film into Eq. (30) leads to

$$c_2(c_1+1)\kappa^2 + c_1(c_2+1)\,k_z^2 = 0. \tag{32}$$

The dispersion relations for surface waves are obtained by demanding evanescence with $k_z = i\eta_2$. Equation (31) then leads to Eq. (20) but with $c_2 \leftrightarrow c_1$

$$2c_2(c_2+1)\eta_2\kappa + \left[(c_2+1)^2\eta_2^2 + c_2^2\kappa^2\right]\tanh(\eta_2\,d) = 0, \tag{33}$$

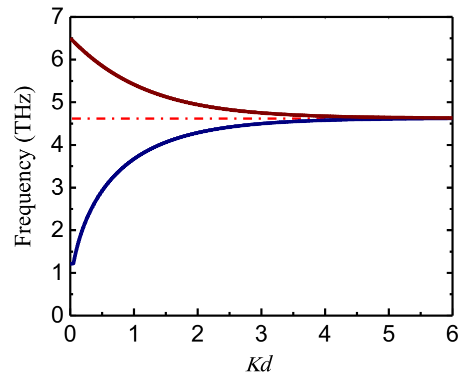

FIG. 4. Dispersion relations of surface polarization waves propagating in the film plane in a ferroelectric $LiNbO_3$ film with perpendicular polarization.

while Eq. (32) gives

$$\eta_2 = \left[\frac{c_2(c_1+1)}{c_1(c_2+1)}\right]^{1/2}\kappa. \tag{34}$$

Substitution of η_2 given by (34) in Eq. (33) leads to

$$2c_1c_2(c_1+1)(c_2+1) + [c_1c_2 + (c_1+1)(c_2+1)]\tanh(\eta_2\,d) = 0, \tag{35}$$

with c_1 and c_2 from Eq. (4b). Numerical solutions of Eqs. (34) and (35) for the parameters of $LiNbO_3$ used in the previous section gives the dispersion relations for surface waves in Fig. 4. Reflecting the axial symmetry, the dispersion relation is isotropic, i.e., it depends only on $|\vec{\kappa}|$. Again, and in contrast to the magnetostatic surface waves in a ferromagnetic film magnetized in the perpendicular direction, we find two branches with opposite radial group velocities.

From Eqs. (34) and (35) with $\kappa \to 0$ we find the frequencies of the lower and upper modes,

$$\omega_{0l} = (K_\parallel)^{1/2}\Omega_p,$$
$$\omega_{0u} = (K_\perp + 1)^{1/2}\Omega_p, \tag{36}$$

which are the same as those for films polarized in the plane. For $\kappa d \gg 1$, the two frequencies converge to a value identical to that in films with in-plane polarization,

$$\omega_\infty = \left(\frac{K_\perp + K_\parallel + 1}{2}\right)^{1/2}\Omega_p. \tag{37}$$

For $LiNbO_3$ the frequencies are $\omega_{0l} = 1.212$ THz, $\omega_{0u} = 6.428$ THz, and $\omega_\infty = 4.626$ THz.

Similarly, the dispersion relation of volume modes in a perpendicularly polarized ferroelectric film follows from Eq. (25) with c_1 and c_2 exchanged:

$$2\kappa k_z c_2(c_2+1) + \left[c_2^2\kappa^2 - k_z^2(c_2+1)^2\right]\tan(k_z\,d) = 0, \tag{38}$$

and from Eq. (32)

$$k_z = \left[-\frac{c_2(c_1+1)}{c_1(c_2+1)}\right]^{1/2}\kappa. \tag{39}$$

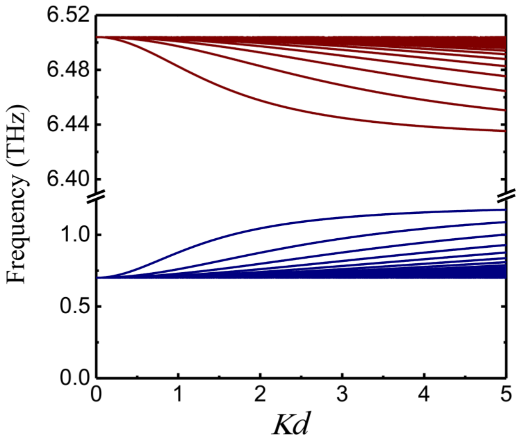

FIG. 5. Dispersion relations of volume polarization waves in a ferroelectric $LiNbO_3$ film polarized perpendicularly to the plane, propagating in the film plane.

Since for the frequencies of interest, $c_1 < 0$ and $c_2 > 0$, k_z is real. Equations (38) and (39) can be solved numerically by finding the multiple roots of (38) for a given value of κd, corresponding to different values of k_z. The solutions for $LiNbO_3$ are shown in Fig. 5. We observe again two manifolds, for backward- and forward-moving waves. However, group velocities are inverted since the forward-moving waves now form the lower frequency band. The dispersion relations for the surface waves, shown in Fig. 4, again lie in the gap of the volume waves.

For $\kappa = 0$ Eqs. (38) and (39) reduce to

$$\omega_{0l} = (K_\perp)^{1/2}\Omega_p,$$
$$\omega_{0u} = (K_\parallel + 1)^{1/2}\Omega_p, \tag{40}$$

while for $\kappa d \to \infty$,

$$\omega_{\infty l} = (K_\parallel)^{1/2}\Omega_p,$$
$$\omega_{\infty u} = (K_\perp + 1)^{1/2}\Omega_p. \tag{41}$$

For $LiNbO_3$ $\omega_{0l} = 0.70$ THz, $\omega_{0u} = 6.504$ THz, $\omega_{\infty l} = 1.212$ THz, and $\omega_{\infty u} = 6.428$ THz.

VI. CONCLUSIONS

We obtained the frequency dispersion relations for surface and volume polarization waves in ferroelectric films and compared them with the surface and volume magnetostatic spin-wave modes in ferromagnetic films. The equation of motion for the electric potential turns out to be similar to the well-known Walker equation for magnetostatic waves. The numerical results for ferroelectric $LiNbO_3$ films, polarized in-plane or perpendicular to the plane, lie in the experimentally accessible low terahertz range. While in ferromagnets surface and bulk modes emerge in single frequency bands, they always come in two bands for each type in ferroelectrics. We attribute this to the transverse and longitudinal character of the polarization waves while magnetostatic waves are purely transverse. In the near future, we intend to address the amplitudes of the polarization waves and their ferron character, i.e., dc and ac electric polarization, and the associated polarization transport and optical properties, also in the limit of two-dimensional ferroelectrics.

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