Eigenfrequencies and Eigenvectors of Polaritons with Application to $LiNbO_3$

I. Long Optical Phonons

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The properties of polaritons in uniaxial crystals with an arbitrary number of atoms in the unit cell are discussed in the harmonic approximation. For $LiNbO_3$, which seems to become a very important substance for applications in tunable Raman lasers, the complete set of eigenfrequencies and eigenvectors (normal coordinates, electric field, electric polarisation) is calculated numerically.

In the limiting case $k \to \infty$ (long optical phonons), which is treated in detail in this first paper, an example for each possible type of extraordinary phonon branches is discussed, namely a $(T \to L)$- a $(L \to T)$-, a $(T \to T)$- and a $(L \to L)$-branch of each type. The results are comparable with experimental data only for the frequencies of the 45°-phonons, measured by Barker and Loudon. The agreement is very satisfactory.

Introduction

Although the theory of long-wavelength optical lattice vibrations and polaritons has been developed by several authors [1–4] in recent years, there are only a few publications [5–8], which deal with calculations for the dispersion relation of polaritons in more complicated crystals. To the authors knowledge up to now there are no calculations of the eigenvectors in the whole range of wave vector $\boldsymbol{k}$ of polaritons, i. e. the dependence of normal coordinates, electric field and electric polarisation both on modulus and direction of $\boldsymbol{k}$.

In this paper the theory of polaritons for crystals of the orthorhombic system and higher symmetry is re-discussed in the harmonic approximation for the purpose of numerical calculations. For the application we have chosen $LiNbO_3$, which belongs to the hexagonal crystal class and therefore is uniaxial. This substance seems to become a very important one for experiments of stimulated Raman-effect and for tunable Raman lasers [9–11]. The knowledge of normal coordinates and electric field is necessary for calculations of Raman scattering cross-sections and intensities of Raman lines of polaritons. This calculation shall be given elsewhere.

In this first part we only treat the limiting case $k \to \infty$ $(n^2 \to \infty)$ of polaritons, i. e. the infrared active long optical phonons. Polaritons of ranges of $k \lesssim 10^4\ \mathrm{cm}^{-1}$ shall be treated in the second part. Polaritons in the former range take part in backward (stimulated) or right angle Raman scattering experiments, whereas those of the latter range take part in forward or near-forward (stimulated) Raman scattering experiments.

Theory

It has been shown [4], that the properties of polaritons including IR-active long optical phonons are determined by the following set of equations:

$$-\omega^2 \boldsymbol{Q} = B^{11}\boldsymbol{Q} + B^{12}\boldsymbol{E}\,, \qquad (1\,\mathrm{a})$$

$$\boldsymbol{P} = (B^{12})^{+}\boldsymbol{Q} + B^{22}\boldsymbol{E}\,, \qquad (1\,\mathrm{b})$$

$$\boldsymbol{E} = \frac{4\pi}{n^2 - 1}\,(\boldsymbol{I} - n^2\,\boldsymbol{s}\,\boldsymbol{s})\,\boldsymbol{P} \qquad (1\,\mathrm{c})$$

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with the following notations: ω: phonon frequency, $\boldsymbol{E}$: electric field, $\boldsymbol{P}$: electric polarisation, I: unit tensor, $\boldsymbol{Q}$: vector of the quasi-normal coordinates Q with dimension r where r is the number of IR-active lattice vibrations *, $n = c\,k/\omega$: refractive index, c: velocity of light in vacuum, $\boldsymbol{k} = k\,\boldsymbol{s}$: wave vector, B^{11}: $(r \times r)$-matrix, B^{12}: $(r \times 3)$-matrix, B^{22}: (3×3)-matrix, $(B^{12})^+$: transposed matrix of B^{12}.

The Eqs. (1 a) and (1 b) follow from the existence of an energy density of the crystal, as it has been shown in more detail by BORN and HUANG [1] and COCHRAN and COWLEY [3]. Equation (1 c) results directly from Maxwell's equations for insulators.

The Eqs. (1) in this form are valid for all crystal classes. In the special case of orthorhombic crystals and higher symmetry the elements of B^{11}, B^{12}, B^{22} are determined in the following way (see Ref. [4]):

a) B^{11} is a diagonal matrix, the diagonal elements $B^{11}_{i\alpha}$ are given from the dispersion frequencies ω_{ai} by

$$B^{11}_{\alpha i} = -(\omega_{ai})^2 \quad (i = 1, \ldots, r). \tag{2}$$

b) B^{22} is a diagonal matrix, the diagonal elements B^{22}_{α} are determined by

$$B^{22}_{\alpha} = (\varepsilon^{\infty}_{\alpha} - 1)/4\pi \qquad (\alpha = 1, 2, 3), \tag{3}$$

where $\varepsilon^{\infty}_{\alpha}$ is the high frequency dielectric constant in α-direction.

c) The elements of B^{12} are connected with the dynamic dielectric constant ε_{α} as follows:

$$\varepsilon_{\alpha} = -\sum_i 4\pi \frac{(B^{12}_{i\alpha})^2}{B^{11}_{\alpha i} + \omega^2} + 4\pi B^{22}_{\alpha} + 1. \tag{4}$$

As KUROSAWA first has shown [12], it is useful to factorize Eq. (4), yielding

$$\varepsilon_{\alpha} = \varepsilon^{\infty}_{\alpha} \frac{\prod_j \left((\omega^1_{\alpha j})^2 - \omega^2\right)}{\prod_j (\omega^2_{\alpha j} - \omega^2)}. \tag{5}$$

Herein the $\omega^1_{\alpha j}$ are the frequencies of exactly longitudinal waves along the principal directions of the crystal.

Combining (4) with (5) and substituting $B^{11}_{\alpha i}$ and B^{22}_{α} by (2) and (3), respectively, one finds

$$\varepsilon^{\infty}_{\alpha} \prod_j \left((\omega^1_{\alpha j})^2 - \omega^2\right) = \varepsilon^{\infty}_{\alpha} \prod_j (\omega^2_{\alpha j} - \omega^2) + \sum_i 4\pi (B^{12}_{i\alpha})^2 \prod_{j \neq i} (\omega^2_{\alpha j} - \omega^2). \tag{6}$$

For the special value ω_{ak} for ω the first term in (6) on the right vanishes and also all terms in the sum, except the term with $i = k$. Therefore the elements $B^{12}_{k\alpha}$ are given by

$$(B^{12}_{k\alpha})^2 = \frac{\varepsilon^{\infty}_{\alpha}}{4\pi} \frac{\prod_j \left((\omega^1_{\alpha j})^2 - \omega^2_{\alpha k}\right)}{\prod_{j \neq k} (\omega^2_{\alpha j} - \omega^2_{\alpha k})}. \tag{7}$$

In this way the coefficients of the system (1) are completely determined, if the values $\varepsilon^{\infty}_{\alpha}$, ω_{aj}, $\omega^1_{\alpha j}$ are known from experimental data.

To solve the Eqs. (1) we combine the vectors $\boldsymbol{Q}$, $\boldsymbol{E}$, $\boldsymbol{P}$ to a vector $\boldsymbol{X} = (\boldsymbol{Q}, \boldsymbol{E}, \boldsymbol{P})$ of dimension $r + 6$. With this notation the system (1) takes the simple form of $A\,\boldsymbol{X} = \boldsymbol{O}$, by blocking up Eqs. (1) to a $(r + 6 \times r + 6)$-matrix A:

$$A\,\boldsymbol{X} = \begin{pmatrix} B^{11} + \omega^2 I & B^{12} & 0 \\ (B^{12})^+ & B^{22} & -I \\ 0 & -I & T \end{pmatrix} \begin{pmatrix} \boldsymbol{Q} \\ \boldsymbol{E} \\ \boldsymbol{P} \end{pmatrix} = \boldsymbol{O}, \tag{8}$$

where 0 is the zero-matrix and

$$T \equiv \frac{4\pi}{n^2 - 1} (I - n^2\,\boldsymbol{s}\,\boldsymbol{s}). \tag{8 a}$$

The condition for a non-trivial solution of (8) is

$$\det A = 0. \tag{9}$$

Because of the simple form of the submatrices the determinant can be calculated directly. As it has been shown [4], the condition (9) leads to a generalized Fresnel equation for the wave normals:

$$\frac{{s_1}^2}{\frac{1}{n^2} - \frac{1}{\varepsilon_1}} + \frac{{s_2}^2}{\frac{1}{n^2} - \frac{1}{\varepsilon_2}} + \frac{{s_3}^2}{\frac{1}{n^2} - \frac{1}{\varepsilon_3}} = 0, \tag{10}$$

where $\boldsymbol{s} = \boldsymbol{k}/k = (s_1, s_2, s_3)$ is the wave normal vector.

For application to uniaxial crystals such as $LiNbO_3$ the Eqs. (1) can be specialized. Every vector in (1) can be split into a component perpendicular (ordinary component, index 0) and a component parallel (extraordinary component, index e0) to the plane built up by $\boldsymbol{s}$ and the optic axis:

$$\boldsymbol{Q} = (\boldsymbol{Q}_0, \boldsymbol{Q}_{e0}), \qquad \boldsymbol{E} = (\boldsymbol{E}_0, \boldsymbol{E}_{e0}), \quad \boldsymbol{P} = (\boldsymbol{P}_0, \boldsymbol{P}_{e0}).$$

In analogy every matrix B^{ik} can be split into two matrices B^{ik}_0 and B^{ik}_{e0}.

* In this paper we call the coordinates Q quasi-normal coordinates: They are not normal coordinates in the usual sense, because they diagonalize not the total energy density but only its first term containing only the displacement vectors but not the electric field $\boldsymbol{E}$. These coordinates become normal coordinates in the usual sense only for waves with vanishing $\boldsymbol{E}$.

[12] T. KUROSAWA, J. Phys. Soc. Japan **16**, 1298 [1961].

Because of the assumption $\boldsymbol{s}\cdot\boldsymbol{P}_0=0$ for the ordinary part $\boldsymbol{P}_0$ of the polarisation $\boldsymbol{P}$ the second term in (1 c) vanishes and one gets:

$$-\omega^2\,\mathbf{Q}_0 = B_0{}^{11}\,\mathbf{Q}_0 + B_0{}^{12}\,\boldsymbol{E}_0\,, \qquad (1\,a')$$

$$\boldsymbol{P}_0 = (B_0{}^{12})^+\,\mathbf{Q}_0 + B_0{}^{22}\,\boldsymbol{E}_0\,, \qquad (1\,b')$$

$$\boldsymbol{E}_0 = \frac{4\pi}{n^2-1}\,\boldsymbol{P}_0\,. \qquad (1\,c')$$

Because Eqs. (1′) do not contain the vector $\boldsymbol{s}$ any longer, the solutions $\omega=\omega_\perp$, E_0, P_0 are independent of the orientation of $\boldsymbol{k}$. By definition every vector in (1′) is perpendicular to the plane built up by $\boldsymbol{s}$ and the optic axis, therefore ordinary polaritons are exactly transverse.

In the case of extraordinary polaritons one obtains:

$$-\omega^2\,\mathbf{Q}_{e0} = B_{e0}^{11}\,\mathbf{Q}_{e0} + B_{e0}^{12}\,\boldsymbol{E}_{e0}\,, \qquad (1\,a'')$$

$$\boldsymbol{P}_{e0} = (B_{e0}^{12})^+\,\mathbf{Q}_{e0} + B_{e0}^{22}\,\boldsymbol{E}_{e0}\,, \qquad (1\,b'')$$

$$\boldsymbol{E}_{e0} = \frac{4\pi}{n^2-1}\,(\boldsymbol{P}_{e0} - n^2\,\boldsymbol{s}\,\boldsymbol{s}\cdot\boldsymbol{P}_{e0}), \qquad (1\,c'')$$

$[\boldsymbol{s}=(s_\perp,\ s_\parallel)$ with $s_\perp=\sqrt{s_1^2+s_2^2}$, $s_\parallel=s_3]$.

The extraordinary polaritons in general have mixed longitudinal-transverse character. Because (1″) contains the vector $\boldsymbol{s}$, the solutions ω, $\boldsymbol{E}_{e0}$, $\boldsymbol{P}_{e0}$ are dependent on the orientation of $\boldsymbol{k}$. Every vector in (1″) by definition lies in the plane built up by $\boldsymbol{s}$ and the optic axis.

In the special case of $\boldsymbol{s}$ being parallel to the optic axis, ordinary and transverse extraordinary polaritons degenerate into the type $E(T)$. In addition, there are longitudinal extraordinary polaritons, e. g. of type $A(L)$.

The equations for the ordinary and extraordinary frequencies now can be derived directly from (1′) and (1″):

If the system (1′) is written in the form $A_0\,\mathbf{X}_0=\boldsymbol{O}$ analogously to (8) with *

$$(B_0^{11})_{jj} = -\omega_{\perp j}^2\,; \qquad B_0^{22} = (\varepsilon_\perp^\infty - 1)/4\pi;$$

$$((B_0^{12})_j)^2 = \frac{4\pi\prod_n\left((\omega_{\perp n}^1)^2-\omega_{\perp j}^2\right)}{\varepsilon_\perp^\infty\prod_{n\neq j}(\omega_{\perp n}^2-\omega_{\perp j}^2)}$$

it follows from $\det A_0=0$:

$$\varepsilon_\perp^\infty\,\omega^2\prod_j\left((\omega_{\perp j}^1)^2-\omega^2\right) - c^2\,k^2\prod_j(\omega_{\perp j}^2-\omega^2)=0\,. \qquad (11)$$

This equation which is equivalent to $n^2=\varepsilon_\perp(\omega)$ determines the frequencies of ordinary polaritons. Because (11 a) contains only the modulus of $\boldsymbol{k}$, these frequencies depend only on the modulus but not on the orientation of the wave vector.

Analogously one can find the frequencies of extraordinary polaritons via $A_{e0}\,\mathbf{X}_{e0}=\boldsymbol{O}$ and $\det A_{e0}=0$. With elements of the matrices *

$$(B_{e0}^{11})_{jj}^{(1)} = -\omega_{\perp j}^2\,, \quad (B_{e0}^{11})_{kk}^{(2)} = -\omega_{\parallel k}^2\,; \quad (B_{e0}^{22})_1 = \frac{\varepsilon_\perp^\infty-1}{4\pi}\,, \quad (B_{e0}^{22})_2 = \frac{\varepsilon_\parallel^\infty-1}{4\pi}\,;$$

$$((B_{e0}^{12})_{j1})^2 = \frac{\varepsilon_\perp^\infty}{4\pi}\,\frac{\prod_n\left((\omega_{\perp n}^1)^2-\omega_{\perp j}^2\right)}{\prod_{n\neq j}(\omega_{\perp n}^2-\omega_{\perp j}^2)}\,, \qquad ((B_{e0}^{12})_{k2})^2 = \frac{\varepsilon_\parallel^\infty}{4\pi}\,\frac{\prod_m\left((\omega_{\parallel m}^1)^2-\omega_{\parallel k}^2\right)}{\prod_{m\neq k}(\omega_{\parallel m}^2)^2-\omega_{\parallel k}^2)}$$

it follows:

$$\varepsilon_\perp^\infty\prod_j\left((\omega_{\perp j}^1)^2-\omega^2\right)\left[\varepsilon_\parallel^\infty\,\omega^2\prod_k\left((\omega_{\parallel k}^1)^2-\omega^2\right) - c^2\,k^2\prod_k(\omega_{\parallel k}^2-\omega^2)\right]s_\perp^2$$

$$+\,\varepsilon_\parallel^\infty\prod_k\left((\omega_{\parallel k}^1)^2-\omega^2\right)\left[\varepsilon_\perp^\infty\,\omega^2\prod_j\left((\omega_{\perp j}^1)^2-\omega^2\right) - c^2\,k^2\prod_j(\omega_{\perp j}^2-\omega^2)\right]s_\parallel^2 = 0\,. \qquad (11\,b)$$

This is equivalent to

$$n^2 = \varepsilon_\perp\,\varepsilon_\parallel/(\varepsilon_\perp\,s_\perp^2 + \varepsilon_\parallel\,s_\parallel^2)\,.$$

Because (11 b) contains k and the vector $\boldsymbol{s}$, these frequencies depend both on modulus and orientation of $\boldsymbol{k}$.

As it has been shown [13], one also can derive (11 a), (11 b) directly from (10) with

$$s_\perp^2 = s_1^2 + s_2^2\,, \qquad s_\parallel^2 = s_3^2\,; \quad \varepsilon_\perp=\varepsilon_1=\varepsilon_2\,, \qquad \varepsilon_\parallel=\varepsilon_3\,.$$

Substituting the eigenvalue of (11 a) or (11 b) into the system $A\,\mathbf{X}=\boldsymbol{O}$ we now can calculate for any uniaxial crystal the complete set of vectors $\mathbf{Q}$, $\boldsymbol{E}$, $\boldsymbol{P}$ over the whole range of k and ϑ, where ϑ is the angle between the wave vector and the optic axis.

[13] L. MERTEN, Phys. Stat. Sol. **30**, 449 [1968].

* Notice the notation of indices now being different from that in Eqs. (1) – (10):

$$(B_0^{11})_{jj} \equiv (B_{e0}^{11})_{jj}^{(1)} = B_{1j}^{11}\,, \quad (B_{e0}^{11})_{kk}^{(2)} \equiv B_{3k}^{11}\,; \quad (B_0^{12})_j \equiv (B_{e0}^{12})_{jj} \equiv B_{j2}^{12} = B_{j2}^{12},$$

$$(B_{e0}^{12})_{k2} \equiv B_{k3}^{12}\,; \quad B_0^{22} \equiv (B_{e0}^{22})_1 \equiv B_1^{22} = B_2^{22}\,, \quad (B_{e0}^{22})_2 \equiv B_3^{22}.$$

Eigenfrequencies ω and Eigenvectors Q, E, P of Long Optical Phonons in $LiNbO_3$

As an example for applications of polariton theory given in the preceeding section we have chosen $LiNbO_3$, for this substance seems to become very important for devices in nonlinear optics, in particular for tunable Raman lasers.

The values for numerical calculation $\varepsilon_{\parallel}^{\infty}$, $\varepsilon_{\perp}^{\infty}$, $\omega_{\parallel}$, $\omega_{\parallel}^{1}$, $\omega_{\perp}$, $\omega_{\perp}^{1}$, which have been measured by several authors, are collected in Table 1. We used the frequencies given by KAMINOW and JOHNSTON JR.[14] which seems to be complete according to group theory.

In Table 2 we have collected the non-zero elements of the matrices B^{ik} calculated from (2), (3), (7).

In this section we regard only the limiting case $k \to \infty$. The other ranges of the wave vector $\boldsymbol{k}$ ($k \lesssim 10^4\,\mathrm{cm}^{-1}$) will be treated numerically in part II. In this limiting case, according to (1 c) or (1 c′) and (1 c″), the electric field of the ordinary phonons exactly vanishes whereas the electric field of the extraordinary phonons is always longitudinal. In the following we need to discuss only the extra-

Table 1. Frequencies $\omega_{\parallel}$, $\omega_{\parallel}^{1}$, $\omega_{\perp}$, $\omega_{\perp}^{1}$ and dielectric constants of $LiNbO_3$.

	ω [cm^{-1}]				
	Raman[14]	Raman[16]	Raman[17]	IR[17]	IR[18]
$\omega_{\parallel}$	253	255	252	248	187
	275	277	276	274	248
	334	334	333	307	305
	637	632	634	628	605
$\omega_{\parallel}^{1}$	273	—	—	273	200
	331	—	—	306	298
	428	—	—	423	412
	874	—	873	869	882
$\omega_{\perp}$	92	—	—	—	—
	152	152	152	152	147
	238	239	238	236	234
	262	266	264	265	258
	322	321	321	322	317
	368	369	367	363	354
	436	430	434	431	421
	582	580	579	586	557
	630	—	—	—	657
$\omega_{\perp}^{1}$	95[+]	—	—	—	—
	198	—	—	198	194
	243	—	—	238	244
	298	—	299	296	290
	345	—	333	342	345
	428	—	—	418	408
	448	—	—	450	444
	621	—	—	—	626
	881	883	880	878	901

$\varepsilon_{\perp}^{\infty}=4.2$; $\varepsilon_{\parallel}^{\infty}=4.5$ (see [19]). [+] Corrected value from Ref. [15].

Table 2. Non-zero elements of B^{11}, B^{12}, B^{22} for $LiNbO_3$ [+].

index[++] (i,k)	B_{ik}^{11} ($10^{26}\,\mathrm{sec}^{-2}$)	index[++] (i,α)	$B_{i\alpha}^{12}$ ($10^{12}\,\mathrm{sec}^{-1}$)
(1,1), (10,10)	— 3.007	(1,1), (10,2)	8.96
(2,2), (11,11)	— 8.209	(2,1), (11,2)	35.7
(3,3), (12,12)	— 20.13	(3,1), (12,2)	15.5
(4,4), (13,13)	— 24.39	(4,1), (13,2)	29.8
(5,5), (14,14)	— 36.84	(5,1), (14,2)	22.0
(6,6), (15,15)	— 48.12	(6,1), (15,2)	25.5
(7,7), (16,16)	— 67.54	(7,1), (16,2)	6.21
(8,8), (17,17)	—120.4	(8,1), (17,2)	50.9
(9,9), (18,18)	—141.0	(9,1), (18,2)	24.7
(19,19)	— 22.74	(19,3)	50.1
(20,20)	— 26.87	(20,3)	15.4
(21,21)	— 39.64	(21,3)	9.03
(22,22)	—144.2	(22,3)	52.9

index (α,β)	$B_{\alpha\beta}^{22}$
(1,1), (2,2)	0.278
(3,3)	0.255

[+] It may be noticed that the coefficients B_{ik}^{11} and $B_{i\alpha}^{12}$ are calculated from Table I with frequencies in units of sec^{-1} instead of cm^{-1}.

[++] The indices used in this table are indices of matrix elements referring to the matrices of Eq. (1) and are not to be confused with indices chosen in Eqs. (2) — (7).

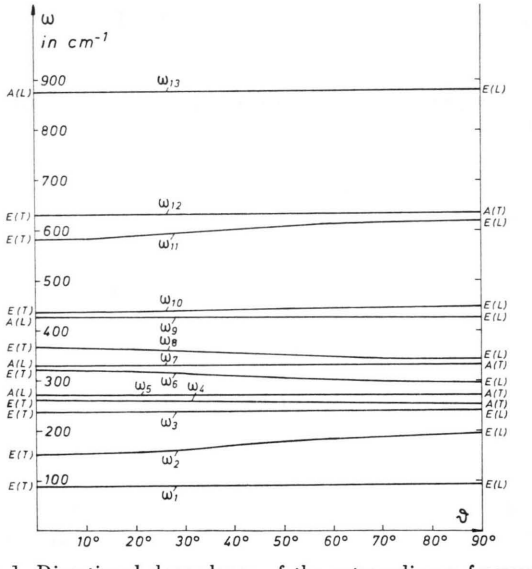

Fig. 1. Directional dependence of the extraordinary frequencies in $LiNbO_3$.

[14] I. P. KAMINOW and W. D. JOHNSTON, JR., Phys. Rev. **160**, 519 [1967].
[15] W. D. JOHNSTON, JR., Phys. Rev. **B 1**, 3494 [1970].
[16] R. F. SCHAUFELE and M. J. WEBER, Phys. Rev. **152**, 705 [1966].
[17] A. S. BARKER, JR., and R. LOUDON, Phys. Rev. **158**, 443 [1967].
[18] J. D. AXE and D. F. O. KANE, Appl. Phys. Letters **9**, 58 [1966].
[19] G. D. BOYD, R. C. MILLER, K. NASSAU, W. L. BOND, and A. SAVAGE, Appl. Phys. Letters **5**, 234 [1964].

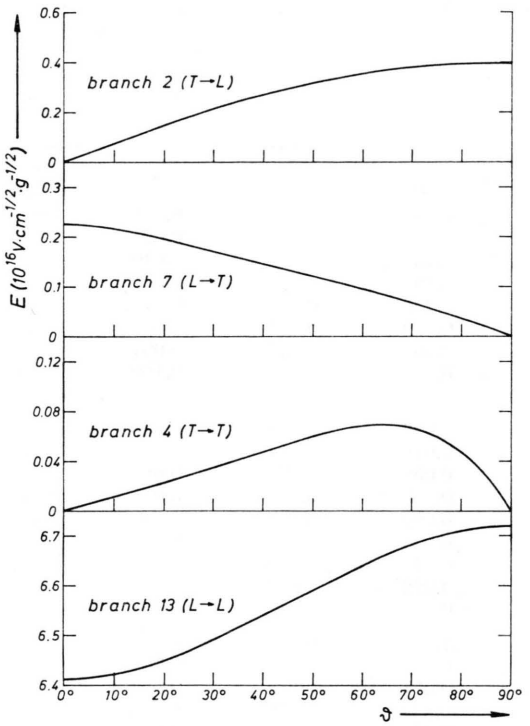

Fig. 2. Directional dependence of the electric field E of extraordinary branches in $LiNbO_3$.

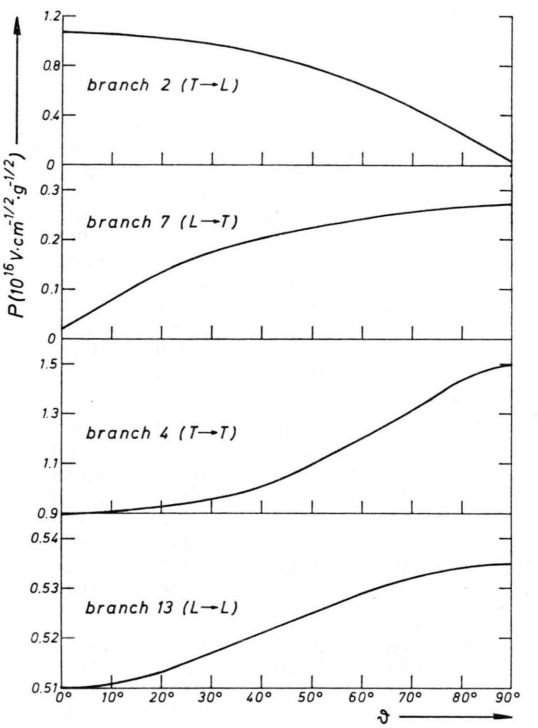

Fig. 3. Directional dependence of the polarization P of extraordinary branches in $LiNbO_3$.

Table 3. 45°-extraordinary optical phonons in $LiNbO_3$.

branch	$\omega_{e0}(45°)$ [cm^{-1}] theoretical due to Eq. (12b)	experimental due to BARKER Jr. and LOUDON[17]
1	94.05	—
2	175.4	177
3	239.0	—
4	259.6	—
5	273.7	—
6	311.5	308
7	332.7	—
8	352.8	353
9	428.0	—
10	444.0	447
11	605.3	610
12	631.4	—
13	877.4	878

ordinary phonons in detail, because the ordinary ones are essentially orientation independent and agree with the extraordinary phonons in the case of $\vartheta = 0$.

The directional dependence of frequencies of all extraordinary phonon branches of $LiNbO_3$ is represented in Fig. 1. As expected, the phonon branches have very similar properties to those of α-quartz [7, 8]. Besides the phonons of the principal directions only the 45°-phonons have been measured by BARKER and LOUDON [17]. They are collected together with the calculated values in Table 3. The agreement between experimental and theoretical values is satisfactory and proves that the input frequencies $\omega_{\parallel}$, $\omega^1_{\parallel}$, $\omega_{\perp}$, $\omega^1_{\perp}$ at least for the branches 2, 6, 8, 10, 11, 13 are correctly combined.

In Table 4 we represent an example for each possible type of vibration in $LiNbO_3$ **, which now will be discussed separately:

1. $(T \to L)$-branch (branch 2)

This vibration is exactly transverse for $\vartheta = 0$. Because the coupling of the quasi-normal coordinates is caused by the electric field which vanishes from (1 c) for $k \to \infty$, the phonon for $\vartheta = 0°$ is determined only by one single coordinate ($\overline{Q}_{\perp 2}$ corresponding to $\omega_{\perp 2}$). If the angle increases, there is a coupling of all other coordinates due to the electric field, but $\overline{Q}_{\perp 2}$ still remains the strongest quasi-normal coordinate over the whole range of ϑ. For

** Readers who are interested in the complete set of data also of the other branches may request them from the authors.

Table 4. Eigenfrequencies ω, quasi-normal coordinates $\overline{Q}_{\perp j} = \frac{Q_{\perp j}}{\partial}$ $(j=1,\ldots,9)$ and $\overline{Q}_{\parallel k} = \frac{Q_{\parallel k}}{Q}$ $(k=1,\ldots,4)$,

	ϑ	ω [cm^{-1}]	$\overline{Q}_{\perp 1}$	$\overline{Q}_{\perp 2}$	$\overline{Q}_{\perp 3}$	$\overline{Q}_{\perp 4}$	$\overline{Q}_{\perp 5}$	$\overline{Q}_{\perp 6}$	$\overline{Q}_{\perp 7}$
branch 2	0°	152.0	0	1.0	0	0	0	0	0
($T \rightarrow L$)	10°	153.5	0.007	0.996	− 0.006	− 0.008	− 0.003	− 0.003	− 0.0005
	20°	157.6	0.026	0.983	− 0.023	− 0.033	− 0.013	− 0.011	− 0.002
	30°	163.9	0.049	0.963	− 0.053	− 0.072	− 0.029	− 0.024	− 0.004
	40°	171.4	0.071	0.937	− 0.094	− 0.125	− 0.049	− 0.040	− 0.006
	50°	179.3	0.087	0.909	− 0.145	− 0.188	− 0.071	− 0.057	− 0.009
	60°	186.6	0.098	0.882	− 0.206	− 0.256	− 0.093	− 0.074	− 0.012
	70°	192.6	0.105	0.858	− 0.267	− 0.319	− 0.111	− 0.088	− 0.014
	80°	196.6	0.109	0.842	− 0.315	− 0.365	− 0.124	− 0.097	− 0.015
	90°	198.0	0.110	0.836	− 0.334	− 0.382	− 0.129	− 0.100	− 0.016
branch 7	0°	331.0	0	0	0	0	0	0	0
($L \rightarrow T$)	10°	331.1	0.003	0.015	0.010	0.026	0.131	− 0.035	− 0.003
	20°	331.5	0.006	0.026	0.018	0.046	0.224	− 0.063	− 0.005
	30°	332.0	0.007	0.033	0.023	0.058	0.272	− 0.082	− 0.006
	40°	332.5	0.008	0.036	0.025	0.063	0.284	− 0.091	− 0.007
	50°	333.0	0.008	0.036	0.025	0.062	0.268	− 0.091	− 0.007
	60°	333.4	0.007	0.031	0.022	0.055	0.229	− 0.082	− 0.006
	70°	333.7	0.005	0.024	0.017	0.041	0.169	− 0.063	− 0.005
	80°	333.9	0.003	0.013	0.009	0.022	0.090	− 0.034	− 0.002
	90°	334.0	0	0	0	0	0	0	0
branch 4	0°	262.0	0	0	0	− 1.0	0	0	0
($T \rightarrow T$)	10°	261.9	0.0003	0.001	0.002	− 0.993	− 0.001	− 0.0007	− 0.0001
	20°	261.6	0.001	0.006	0.010	− 0.971	− 0.005	− 0.003	− 0.0004
	30°	261.0	0.002	0.013	0.022	− 0.934	− 0.010	− 0.006	− 0.0008
	40°	260.1	0.004	0.023	0.040	− 0.878	− 0.018	− 0.011	− 0.001
	50°	258.9	0.007	0.035	0.064	− 0.797	− 0.026	− 0.016	− 0.002
	60°	257.4	0.008	0.046	0.089	− 0.685	− 0.032	− 0.020	− 0.003
	70°	255.5	0.009	0.050	0.106	− 0.527	− 0.034	− 0.022	− 0.003
	80°	253.8	0.007	0.037	0.086	− 0.303	− 0.024	− 0.015	− 0.002
	90°	253.0	0	0	0	0	0	0	0
branch 13	0°	874.0	0	0	0	0	0	0	0
($L \rightarrow L$)	10°	874.2	0.012	0.050	0.023	0.045	0.035	0.042	0.011
	20°	874.8	0.024	0.099	0.045	0.089	0.069	0.084	0.022
	30°	875.7	0.036	0.146	0.066	0.130	0.101	0.123	0.033
	40°	876.8	0.046	0.189	0.086	0.168	0.130	0.159	0.042
	50°	878.0	0.056	0.226	0.102	0.201	0.156	0.190	0.051
	60°	879.2	0.063	0.257	0.116	0.229	0.177	0.216	0.058
	70°	880.1	0.069	0.279	0.127	0.249	0.193	0.235	0.063
	80°	880.8	0.072	0.294	0.133	0.262	0.203	0.247	0.066
	90°	881.0	0.074	0.298	0.135	0.266	0.206	0.251	0.067

Table 5. ($T \rightarrow L$)- and ($L \rightarrow T$)-transition of branch 2 and branch 7 in $LiNbO_3$ near the principal directions.

branch 2		*branch* 2	
ϑ	φ	ϑ	φ
80°	82.8°	87°	67.2°
81°	82.0°	87.5°	63.2°
82°	81.0°	88°	57.8°
83°	79.8°	88.5°	49.9°
84°	78.1°	89°	38.4°
85°	75.8°	89.5°	21.6°
86°	72.5°	90°	0°

branch 7		*branch* 7	
ϑ	φ	ϑ	φ
0°	0°	4°	60.7°
0.5°	12.6°	5°	65.8°
1°	24.0°	6°	69.4°
1.5°	33.8°	7°	72.2°
2°	41.7°	8°	74.2°
2.5°	48.1°	9°	75.9°
3°	53.2°	10°	77.2°

electric field E, polarisation P (relative values) and angle φ between $\boldsymbol{K}$ and $\boldsymbol{P}$ of extraordinary branches in $LiNbO_3$.

$\overline{Q}_{\perp 8}$	$\overline{Q}_{\perp 0}$	$\overline{Q}_{\parallel 1}$	$\overline{Q}_{\parallel 2}$	$\overline{Q}_{\parallel 3}$	$\overline{Q}_{\parallel 4}$	E/Q (10^{16} V · cm$^{-1/2}$ · g$^{-1/2}$)	P/Q (10^{16} V · cm$^{-1/2}$ · g$^{-1/2}$)	φ
0	0	0	0	0	0	0	1.07	90.0°
− 0.002	− 0.0008	− 0.088	− 0.021	− 0.007	− 0.010	0.076	1.06	89.7°
− 0.008	− 0.003	− 0.169	− 0.040	− 0.014	− 0.018	0.150	1.03	89.3°
− 0.017	− 0.007	− 0.237	− 0.056	− 0.019	− 0.024	0.216	0.978	89.0°
− 0.027	− 0.011	− 0.285	− 0.066	− 0.022	− 0.028	0.274	0.900	88.6°
− 0.038	− 0.016	− 0.304	− 0.068	− 0.022	− 0.027	0.321	0.794	88.2°
− 0.049	− 0.020	− 0.288	− 0.063	− 0.020	− 0.024	0.357	0.653	87.5°
− 0.057	− 0.023	− 0.229	− 0.049	− 0.015	− 0.018	0.382	0.472	86.3°
− 0.062	− 0.025	− 0.128	− 0.027	− 0.008	− 0.009	0.397	0.251	82.8°
− 0.064	− 0.026	0	0	0	0	0.402	0.032	0.0°
0	0	0.235	0.097	− 0.966	− 0.038	0.227	0.018	0.0°
− 0.008	− 0.003	0.222	0.091	− 0.960	− 0.036	0.218	0.079	77.2°
− 0.014	− 0.005	0.190	0.078	− 0.948	− 0.031	0.197	0.136	83.4°
− 0.018	− 0.007	0.152	0.062	− 0.942	− 0.025	0.172	0.176	85.5°
− 0.020	− 0.008	0.114	0.046	− 0.943	− 0.019	0.147	0.203	86.7°
− 0.019	− 0.008	0.079	0.032	− 0.952	− 0.013	0.122	0.225	87.5°
− 0.017	− 0.007	0.048	0.019	− 0.966	− 0.008	0.096	0.242	88.2°
− 0.013	− 0.005	0.023	0.009	− 0.982	− 0.004	0.067	0.257	88.8°
− 0.007	− 0.003	0.006	0.002	− 0.995	− 0.001	0.035	0.267	89.4°
0	0	0	0	− 1.000	0	0	0.271	90.0°
0	0	0	0	0	0	0	0.895	90.0°
− 0.0004	− 0.0001	0.116	− 0.023	− 0.002	− 0.002	0.011	0.903	89.9°
− 0.001	− 0.0006	0.233	− 0.044	− 0.004	− 0.003	0.023	0.925	89.9°
− 0.003	− 0.001	0.351	− 0.059	− 0.006	− 0.004	0.035	0.964	89.8°
− 0.005	− 0.002	0.472	− 0.066	− 0.007	− 0.005	0.048	1.02	89.8°
− 0.008	− 0.003	0.595	− 0.064	− 0.007	− 0.006	0.060	1.10	89.7°
− 0.010	− 0.004	0.719	− 0.052	− 0.006	− 0.005	0.068	1.20	89.7°
− 0.011	− 0.004	0.840	− 0.032	− 0.004	− 0.003	0.067	1.31	89.8°
− 0.008	− 0.003	0.948	− 0.010	− 0.001	− 0.001	0.047	1.44	89.9°
0	0	1.000	0	0	0	0	1.50	90.0°
0	0	0.431	0.135	0.083	0.888	6.410	0.510	0.0°
0.125	0.070	0.425	0.133	0.082	0.875	6.420	0.511	1.1°
0.247	0.139	0.406	0.127	0.078	0.836	6.450	0.513	2.1°
0.362	0.203	0.376	0.117	0.073	0.772	6.490	0.517	2.8°
0.467	0.262	0.334	0.104	0.065	0.685	6.540	0.521	3.2°
0.558	0.313	0.282	0.088	0.054	0.576	6.590	0.525	3.2°
0.633	0.355	0.220	0.069	0.042	0.449	6.640	0.529	2.9°
0.688	0.385	0.151	0.047	0.029	0.307	6.680	0.532	2.1°
0.722	0.404	0.077	0.024	0.015	0.156	6.710	0.534	1.1°
0.733	0.411	0	0	0	0	6.720	0.535	0.0°

Remark to Table 4: Splitting the solutions $\boldsymbol{X} = (\boldsymbol{Q}, \boldsymbol{E}, \boldsymbol{P})$ of Eq. (8) into itc component $\boldsymbol{X} = (Q_\perp, E_\perp, P_\perp)$ perpendicular and $\boldsymbol{X}_\parallel = (\boldsymbol{Q}_\parallel, \boldsymbol{E}_\parallel, \boldsymbol{P}_\parallel)$ parallel to the optic axis it follows from symmetry of uniaxial crystals, that $\boldsymbol{X}' = (-\boldsymbol{X}_\perp, \boldsymbol{X}_\parallel)$, $\boldsymbol{X}'' = (\boldsymbol{X}_\perp, -\boldsymbol{X}_\parallel)$, $\boldsymbol{X}''' = (-\boldsymbol{X}_\perp - \boldsymbol{X}_\parallel)$ must be solutions of (8) too, i. e. the sign of both vectors $\boldsymbol{X}_\parallel$ and $\boldsymbol{X}_\perp$ is arbitrary. In our calculations the signs are chosen in such a way that the wave vector $\boldsymbol{s}$ has no negative components and the vector $\boldsymbol{E}$ is antiparallel to $\boldsymbol{s}$ for $k \to \infty$.

the angle of $\vartheta = 90°$ the vibration becomes exactly longitudinal. Because the electric field vector lies in the basic plane together with $\boldsymbol{k}$, all the quasi-normal coordinates $\overline{Q}_{\perp j}$ $(j = 1, \ldots, 9)$ couple, whereas all $\overline{Q}_{\parallel k}$ $(k = 1, \ldots, 4)$ must vanish. It ist interesting that the polarisation $\boldsymbol{P}$ turns into the exactly longitudinal character only in the immediate vicinity of the principal direction $\vartheta = 90°$ (cf. Tab. 5) in contrast to the electric field, which is exactly longitudinal over the whole range of ϑ as mentioned.

2. *(L → T)-branch (branch 7)*

This branch essentially has the same properties as branch 2 but with opposite direction of ϑ. Therefore this case must not be discussed in detail. The behavior of $\boldsymbol{P}$ near the principal direction $\vartheta = 0°$ is completely analogous to that of the $(T \to L)$-branch (cf. Table 5).

3. $(T \rightarrow T)$-branch (branch 4)

This vibration is exactly transverse for both principal directions and therefore characterized by only one quasi-normal coordinate ($\overline{Q}_{\perp 4}$ respectively $\overline{Q}_{\parallel 1}$). Going from $\vartheta = 0^\circ$ to $\vartheta = 90^\circ$ these two coordinates interchange their predominance. In the intermediate range $0^\circ < \vartheta < 90^\circ$ there is a weak coupling of all other coordinates to the predominant coordinates $\overline{Q}_{\perp 4}$ and $\overline{Q}_{\parallel 1}$. The polarisation $\boldsymbol{P}$ is nearly transverse over the whole range of ϑ, the electric field is very weak compared to the other branches because it is exactly longitudinal.

4. $(L \rightarrow L)$-branch (branch 13)

Because this vibration is exactly longitudinal for both principal directions and the electric field therefore being relatively strong over the whole range of ϑ, there is a strong coupling of the quasi-normal coordinates for all directions. Only for $\vartheta = 0^\circ$ and $\vartheta = 90^\circ$ all $\overline{Q}_{\parallel k}$ and $\overline{Q}_{\perp j}$, respectively, vanish for reasons of symmetry. The polarisation $\boldsymbol{P}$ is nearly longitudinal for all angles ϑ.

All numerical calculations were carried out on the IBM computer 360/50 of the Universität Münster.

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A Simple Algebraic Calculation of the Resolvent in the V Θ-Sector of the Lee Model

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It is shown that the resolvent in the $V\,\Theta$-sector of the Lee model may be expressed by simple algebraic operations in terms of known matrix elements between states of the V-sector.

1. Introduction

It is known that the stationary states in the $V\,\Theta$-sector of the LEE model [1] may be obtained from the solution of the KÄLLÉN-PAULI integral equation [2], which has been determined in closed form by several authors [3–8] using the theory of analytic functions. On the other hand, FIVEL [9] has shown that it is also possible to solve the eigenvalue problem in the $V\,\Theta$-sector in a completely algebraic way. His method consists in applying a unitary transformation to the HAMILTONIAN such that a separable potential problem arises which may be reduced to a system of linear equations. The calculations required are, however, rather complicated, especially as regards the comparison of the result with that of the aforementioned authors.

In the present paper we wish to show that by extremely simple algebraic operations the resolvent in the $V\,\Theta$-sector may be expressed in terms of known matrix elements from the V-sector. In fact, the problem reduces to solving a linear integral equation with a degenerate kernel which is equivalent to one linear equation for one unknown constant. In comparing our result with the standard form of the solution we have to use some relations between analytic functions, which are however rather trivial.

We need not distinguish explicitly between the cases of a stable or an unstable V-particle. In order

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