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PHONON DISPERSION RELATION OF BERYLLIUM OXIDE

G. L. Ostheller, R. E. Schmunk and R. J. Kearney

IDAHO NUCLEAR CORPORATION
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PHONON DISPERSION RELATION OF BERYLLIUM OXIDE

by

G. L. Ostheller*, R. E. Schmunk and R. J. Kearney†

ABSTRACT

The phonon dispersion relation measurements for BeO have been extended by neutron inelastic scattering, using the MTR triple axis spectrometer. New data include a transverse acoustic branch and an optical branch with wave propagation in the $[01\bar{1}0]$ direction and in addition the transverse acoustic, longitudinal acoustic and a transverse optic branch, all with wave propagation in the $[0001]$ direction. The two optical branches observed are degenerate at the center of the Brillouin zone with a frequency $\omega = 6.21 \pm 0.11 \times 10^{13}$ rad sec⁻¹. This frequency is in fair agreement with the optical data of Loh which gives $\omega = 6.41 \times 10^{13}$ rad sec⁻¹ for the same mode. From the neutron scattering data of this experiment and the optical data of Loh it appears that the low lying optical branches reported previously by Brugger et al. are not single phonon data but are the result of multiphonon processes. Comparison is made between the measured branches of the dispersion relation and the results of preliminary lattice dynamics model calculations by Young and by Solbrig.

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PHONON DISPERSION RELATION OF BERYLLIUM OXIDE

I. INTRODUCTION

Beryllium oxide is sometimes classified as a metal oxide ceramic. It has a very large coefficient of thermal conductivity and is a poor electrical conductor. In the crystal state, it has the wurtzite structure and a preferential growth along the c axis¹. Investigations of the properties of BeO have been hampered by the relative unavailability of large single crystals. Austerman et al.² were among the first to investigate the growth of BeO crystals in detail and have developed recipes suitable for single crystal growth. The crystals have been grown from both the liquid and vapor state with the physical shape being quite sensitive to growth temperature. The crystals tend to reject impurities and appear nearly perfect, that is, with no visible flaws.

The advent of the nuclear age in the last twenty years has stimulated great interest in the properties of beryllium oxide. It is somewhat rare and its toxic character has been one of the reasons preventing its use in commercial manufacturing. In spite of these difficulties, beryllium oxide is rapidly becoming a major reactor moderator and reflector. These uses have stimulated great interest in the physical properties of beryllium oxide.

A knowledge of the lattice vibrational spectra of crystalline material is of fundamental importance to a basic understanding of its physical properties. Measurements of the dispersion relations in BeO are of particular interest to nuclear engineers since the

experimental data are necessary for the development of an accurate lattice dynamics model. Once the model has been developed it can be used for calculating inelastic neutron scattering cross sections and a scattering kernel, which are needed for reactor design considerations.

Several groups have made preliminary calculations to obtain the dispersion relation for BeO. J. A. Young³ has followed the general formulation for ionic crystals given by Born and Huang⁴ in which the lattice potential is expressed as the sum of repulsive and Coulomb contributions. Solbrig⁵ has approached the problem using general quadratic valence force fields and applying symmetry coordinates to factor the secular equation. The latter approach was used quite successfully in the case of diamond⁶.

The frequency dispersion relations can be deduced by measuring the inelastic scattering of x-rays or neutrons. The experimental x-ray work is extremely difficult and gives rather poor resolution of the phonon branches. However, excellent resolution is obtained when low energy thermal neutrons are used as a probe. Low energy thermal neutrons, those with energy of less than one tenth electron volt, have momenta and energies of the same order of magnitude as the normal modes in a crystal, and the deBroglie wavelength $\lambda = h/p$ of the neutron, is of the same order of magnitude as the crystal lattice spacings. Thermal neutrons readily interact with the normal modes of vibration in a solid, and the experimental techniques for determining the energy and momentum transfer in the scattering process are well established.

Parts of the phonon dispersion relation for BeO were measured by Brugger et. al⁷ using the MTR velocity selector and time-of-flight techniques⁸. Their measurements were confined to phonons propagating

in the hexagonal plane and primarily to the $[01\bar{1}0]$ symmetry direction in that plane. For the $[01\bar{1}0]$ direction their data included two of the acoustic branches and six optical branches.

In the present work, scattering measurements were confined to phonons propagating in the crystallographic plane defined by the $[0001]$ and $[01\bar{1}0]$ directions. The MTR three axis spectrometer, operating in the "constant Q" mode, was used for these measurements. The third acoustical branch as well as an optical branch were mapped for the $[01\bar{1}0]$ direction, and in addition the acoustical branches and an optical branch were mapped for the $[0001]$ direction. The two observed optical branches have an intercept at $\vec{q} = 0$ of 40.9 ± 0.75 meV ($\omega = 6.21 \pm 0.11 \times 10^{13}$ rad sec⁻¹) which is compared to the value of 42.2 meV (340 cm⁻¹) obtained from recent optical measurements by Loh⁹. General aspects of the lattice dynamics of BeO are discussed in the text; comparison is made between the present data, and 1) the data of Brugger et. al., 2) optical and elastic constant data, and 3) the preliminary lattice dynamics calculations of Young and Solbrig. The present data indicate that the low lying branches observed by Brugger et. al. are not branches of the dispersion relations, but probably arise from multiphonon processes. Finally, estimates are made of the as yet unmeasured phonon branches for the $[0001]$ and $[01\bar{1}0]$ directions.

II. THEORY

A. Lattice Dynamics

The subject of lattice dynamics in the harmonic approximation has been treated by a number of authors^{4,10} and will not be discussed in detail here. In this approximation the potential energy function is expanded to second order in the displacements of the particles from

their equilibrium positions. When this expansion is introduced into the Hamiltonian the resulting equations of motion yield a non-trivial solution for the displacement vectors when the determinant of the coefficients of the displacement vectors vanish, i.e. for the secular equation:

$$|\omega^2(\vec{q}) \delta_{\alpha\beta} \delta_{\kappa\kappa'} - D_{\alpha\beta}(\vec{q}, \kappa\kappa')| = 0.$$

The solutions (roots) ω^2 of the secular equation are the eigenvalues of the dynamical matrix $D_{\alpha\beta}(\vec{q}, \kappa\kappa')$ which is a function of the phonon wave vector $\vec{q}$. Subscripts α, β refer to Cartesian coordinates x, y, z , while indices κ, κ' range over the different atoms in the unit cell. The matrix $D_{\alpha\beta}(\vec{q}, \kappa\kappa')$ in the secular equation is $3n \times 3n$, where n equals the number of atoms in the primitive unit cell. In general, solution of the secular equation yields $3n$ values of ω^2 for a given value of the phonon wave vector. The relationship $\omega^2 = \omega_j^2(\vec{q})$ is referred to as the dispersion relation where the index $j = 1, \dots, 3n$ indicates a particular branch of the relation.

Of the $3n$ branches, 3 are acoustical for which $\omega \rightarrow 0$ as $\vec{q} \rightarrow 0$ while the other $3n-3$ branches are optical branches for which the ω_j tend to non-zero values as $\vec{q} \rightarrow 0$. In cases where phonon propagation is along crystallographic directions of high symmetry, the atomic displacements $\vec{U}(\kappa)$ for a particular mode can be constrained by symmetry to well defined directions, i.e. the modes are either pure longitudinal or transverse. However, for $\vec{q}$ in directions of lower symmetry the phonon polarizations are often less well defined.

Beryllium oxide has the wurtzite type structure which is shown in Figure 1. This structure is described in terms of a hexagonal lattice

Figure 1 - a) Perspective drawing of the beryllium oxide structure with beryllium atoms denoted by solid points and oxygen atoms as open points. b) Separation of the structure into nearest neighbors, second nearest neighbors, etc.

with a basis having Be atoms at $(0, 0, 0)$ and $(1/3, 2/3, 1/2)$ and oxygen atoms at $(0, 0, \mu)$ and $(1/3, 2/3, 1/2 + \mu)$. The lattice constants for BeO are taken to be $a_1 = a_2 = a = 2.698 \text{ \AA}$, $a_3 = c = 4.377 \text{ \AA}$ and $\mu = 0.378$, and the c/a ratio is very close to ideal at $1.6223^{11,12,13,14}$. With 4 atoms in the hexagonal primitive cell there are 12 branches in the dispersion relation.

We are concerned primarily with phonon propagation in the two symmetry directions $[0001]$ and $[01\bar{1}0]$ in BeO. Along $[0001]$ there are 8 distinct branches for the dispersion relation, 4 longitudinal and 4 transverse (1 acoustical and 3 optical branches for each polarization) with each of the transverse branches being doubly degenerate. Along $[01\bar{1}0]$ the 12 branches are divided into two groups: first, $8 \Sigma_1$ branches which have their polarizations in the plane of the $[01\bar{1}0]$ and $[0001]$ directions, and second, $4 \Sigma_2$ branches which have their polarization perpendicular to both $[01\bar{1}0]$ and $[0001]$ directions. Of the $8 \Sigma_1$ branches 2 are acoustical, one predominantly transverse and one predominantly longitudinal and the other 6 are optical whereas for the $4 \Sigma_2$ branches one is acoustical and the other 3 are optical. The Σ_2 branches have a well defined (transverse) polarization while the Σ_1 branches generally do not, being dependent upon the details of the interatomic force field in the BeO crystal.

Several lattice dynamics calculations have been made in the harmonic approximation in which varying assumptions have been made regarding the nature and range of the interatomic forces. These assumptions are reflected in the details of the matrix $D_{\alpha\beta}(\vec{q}_k)$ in the secular equation. Calculations which have been applied directly to BeO were made by Young³ and by Solbrig⁵. In Young's work the potential was expressed as the sum

of a long range Coulomb contribution and a short range repulsive contribution, the latter term including interactions with only nearest neighbors. He used the value for the cohesive energy and the maximum phonon energy observed by Sinclair¹⁵ in evaluating the two disposable parameters in the model. Solbrig has approached the problem using methods developed for calculating molecular dynamics which involve general quadratic valence force fields and the use of symmetry coordinates to factor the secular equation. His force constants are all short range in character relating to specific bond stretching and angle bending constants. The work of both Young and Solbrig should be considered as preliminary due to the lack of sufficient experimental data to evaluate parameters in the models. This point is accentuated further in the work of Sullivan¹⁶ who has attempted to apply the shell model to wurtzite structure crystals. Even limiting short range interactions to nearest neighbors Sullivan was unable to make a unique determination of the constants in his model.

The most recent and complete lattice dynamics calculation for a wurtzite structure material is that of Nusimovici and Birman¹⁷ on CdS. These authors expressed the potential function in terms of Coulomb and short range contributions. The latter contributions are expressed in terms of bond stretching, bond angle and cross coupling constants and include interactions out to third nearest neighbors. Nusimovici and Birman were able to evaluate the constants in their model from infrared and Raman data available on CdS. Of interest are the frequency shifts of the Γ_1 and Γ_5 modes which have different frequencies when the point $\Gamma(\vec{q} = 0)$ is approached from different directions such as $[0001]$ and $[01\bar{1}0]$. These frequency shifts are caused by the long range

electric field associated with the infrared active optical modes and are discussed in detail by Loudon¹⁸.

B. Neutron Scattering

Coherent inelastic scattering of thermal neutrons from a solid is restricted by the interference conditions

$$\Delta E = E - E_0 = \hbar \omega_j(\vec{q})$$

and

$$\vec{Q} \equiv \vec{k}_0 - \vec{k} = 2\pi\vec{\tau} = \vec{q}$$

where E_0 , $\vec{k}_0$ and E , $\vec{k}$ are respectively the neutron energy and wave vector before and after scattering. The quantity $\vec{\tau}$ is a translation vector of the reciprocal lattice, and ω and $\vec{q}$ are the phonon frequency and wave vector which were defined previously. From a knowledge of the incident and scattered neutron energies, the angle of scattering and the orientation of the sample in the scattering experiment it is possible to obtain pairs of values for ω and $\vec{q}$ in the scattering from a single crystal specimen. By varying the experimental parameters and by making a series of measurements it is possible to map out the dispersion relation $\omega_j(\vec{q})$.

Brockhouse and Iyengar, in their work on germanium¹⁹, presented an equation for the one phonon scattering cross section for crystals having more than one atoms per unit cell. This cross section is directly proportional to a quantity $g_j^2(\vec{q}, \vec{\tau})$ which has come to be known as the dynamic structure factor, a function of the phonon wave vector $\vec{q}$ and reciprocal lattice translation vector $\vec{\tau}$. The importance of the structure factor lies in the guidance it can give the experimentalist in mapping

out a particular j^{th} branch of the dispersion relation, i.e. in structures having more than one atom per primitive unit cell the periodic range of the factor $g_j^2(\vec{q}, \vec{\tau})$ is larger than the Brillouin zone and it is valuable to know around which reciprocal lattice point the factor will have an optimum value for a given branch of the dispersion relation. According to Chen²⁰ the structure factor is given by

$$g_j^2(\vec{q}, \vec{\tau}) = \left| \sum_{\kappa} \frac{b_{\kappa} [\vec{Q} \cdot \vec{e}(j, \kappa, \vec{q})]}{\sqrt{M_{\kappa} v_j}} e^{-w(\kappa)} e^{i\vec{Q} \cdot \vec{R}_{\kappa}} \right|^2$$

where the summation is over all κ atoms in the primitive unit cell, M_{κ} and b_{κ} are the mass and bound atom scattering lengths of the κ^{th} atom, v_j is the frequency of the j^{th} mode, $\vec{e}(j, \kappa, \vec{q})$ is a component of the normalized polarization vector belonging to the κ^{th} atom, $\vec{R}_{\kappa}$ is the position vector of the κ^{th} atom in the cell relative to the cell origin and $e^{-w(\kappa)}$ is the Debye-Waller factor. Evaluation of the structure factor requires the application of a lattice dynamics model to evaluate the $\vec{e}(j, \kappa, \vec{q})$. Solbrig has adopted the valence force model to evaluate the structure factor for BeO which has been used as a guide in the experiments²¹. His results are reported in terms of 1) the polarization factor $|\vec{Q} \cdot \hat{\beta}_j|^2$, 2) a modified structure factor γ_j^2 , and 3) the product of the first two quantities. In the evaluation of the structure factor Solbrig set the Debye-Waller factor equal to unity. The dynamic structure factor and the x-ray structure factor for BeO are periodic in the hexagonal plane but not along the c axis due to the odd spacing factor (μ) between interpenetrating lattices in the structure.

III. EXPERIMENTAL METHOD

Experimental measurements were made with the BeO single crystals using a triple axis spectrometer installed at the HT-1 south beam port²² of the MTR. In Figure 2 we have displayed a schematic outline of the experiment together with an artist's sketch of the spectrometer. The spectrometer consists of two separate units: 1) a shielded monochromator facility which was used to obtain a diffracted monoenergetic beam for the experiment, and 2) a sample platform and energy analyzer unit which were mounted on a large diameter rotary table. Because of the lack of coupling between the two units all measurements were made using the same incident neutron energy of 54.2 meV. A beryllium single crystal with a mosaic spread of 12 min. (on the (0002) planes) served as monochromator giving a beam intensity at the sample position of 5×10^6 neutrons/cm²sec and the (111) planes of an aluminum crystal were used for the analyzer. The sample orientation angle ψ , scattering angle ϕ , analyzer crystal angle θ_A , and analyzer arm angle $2\theta_A$ were controlled by Slo-Syn stepping motors through worm gears which permitted angle changes in increments of 0.01° for ψ , ϕ , and $2\theta_A$ and in increments of 0.005° for θ_A . Beam collimators with an angular divergence of 30 min. were positioned between monochromator and sample and between sample and analyzer crystal.

All data were obtained with the spectrometer operated in the constant Q²³ mode using a preset monitor scaler to control the duration of the count for the detector on the analyzer arm for each spectrometer setting. The constant Q method requires changing the sample, scattering and analyzer angles for each energy that is observed. Angle settings for the constant Q runs were generated by a computer program. At the

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Figure 2 - Schematic diagram and pictorial view of the three axis spectrometer.

start of a run the spectrometer angles were set manually and then advanced automatically from one setting to the next after each counting period, using a drum programmer to introduce angle correction instructions to control circuits for stepping the angle drive motors. In Figure 3 we have displayed data peaks for four data runs, each pertaining to a different branch of the BeO dispersion relation as indicated above the data. Considerations which affect the choice of "Q" values to be used in a given run include 1) the polarization factor $(\vec{Q} \cdot \hat{\beta}_j)$ requires that the branch to be observed have at least a component of its polarization $\hat{\beta}_j$ in the plane of the observed scattering and preferably $\hat{\beta}_j$ should be parallel to $\vec{Q}$, 2) the cross section is proportioned to Q^2 through the polarization factor, 3) the modified structure factor γ_j^2 should have an optimum value, and 4) any focusing effects^{24,25} available should be used to advantage.

In the experiment, the single crystals used were two beryllium oxide crystals on loan from Austerman², each of which was less than one cubic centimeter in volume. The crystals were individually mounted on adjustable aluminum disks and adjusted so that the reciprocal lattice plane defined by the [0001] and [01 $\bar{1}$ 0] directions was parallel to the scattering (horizontal) plane. The two crystals were aligned by observing elastic (Bragg) scattering from (0006), (03 $\bar{3}$ 0) and (02 $\bar{2}$ 3) crystallographic planes. The collimator between sample and detector was masked so that scattering was observed from one crystal at a time and then by comparing the two and adjusting one crystal to bring the elastic peaks into coincidence. With the sample crystals mounted as described above it was possible to investigate phonon branches having $\vec{q}$ parallel to the [0001] direction and branches having $\vec{q}$ parallel to [01 $\bar{1}$ 0] which have

Figure 3 - Data plots obtained from runs with the spectrometer operated in the constant Q mode. Identification of the branch and wave vector magnitude pertaining to each data plot is made above the data.

their polarization in the plane of these two symmetry directions. This means that for the $[01\bar{1}0]$ direction we could observe Σ_1 modes but not Σ_2 modes, the latter having their polarization perpendicular to the scattering plane.

In applying the diffraction technique to these measurements it is necessary to guard against the possibility of accepting false peaks. A major contributor to this problem in applying the diffraction technique arises from the fact that order contamination is present in the monoenergetic beam incident on the sample and in the neutrons diffracted by the analyzer crystal. In certain cases where the elastic scattering from the sample is strong or even highly directional one may observe resonances for conditions where $N_1^2 E_O = N_2^2 E_F$ where N_1 and N_2 are integers and E_O and E_F are the first order energies for monochromator and analyzer respectively. One might erroneously interpret such a resonance as having a neutron energy change $E_F - E_O$ where in fact it should be zero. Multiple scattering effects such as Bragg-inelastic or inelastic-Bragg double scattering from the sample may also lead to misunderstanding of the data.

IV. RESULTS AND DISCUSSION

Six branches of the BeO dispersion relation were measured in the present experiment. Five of these represent new information while one branch, LA $[01\bar{1}0]$, duplicates a previous measurement by Brugger et al. Table I lists the observed data giving the phonon wave vector $|\vec{q}|$, phonon frequency ω as well as the energy transfer ΔE in the scattering process, peak strength and width information and the region in κ -space where the phonon was observed. The latter information can be understood recalling that the scattering was confined to the plane containing the orthogonal crystallographic directions $[01\bar{1}0]$ and $[0001]$. Taking

these directions to be coincident with x-z axes respectively we interpret $R(01\bar{1}5)$ as away from the reciprocal lattice point $(01\bar{1}5)$ in the direction of increasing x (to the right). $L(01\bar{1}5)$ would be away from the same lattice point in the direction of decreasing x (to the left), while $A(01\bar{1}5)$ and $B(01\bar{1}5)$ would be in directions of increasing z (above) and decreasing z (below) respectively from the same point. The full width at half maximum (fwhm) figure given in the table should not be construed as the accuracy of the measurements because in most cases the resonances gave well defined peaks for which the centroids of the peaks could be determined to much higher accuracy. Figure 4 displays the dispersion relation plot of the data listed in the table. Data shown for the $[01\bar{1}0]$ direction are all Σ_1 modes.

Figure 5 displays the data of Brugger et al. for comparison with the present data. Data for the $LA[01\bar{1}0]$ branch, the only measurement repeated in the present work, are in good agreement with the previous data. Considerable effort was made to observe low lying optical branches in the range 10-20 meV which were reported in the previous experiment. It was impossible to get a good measurement at $\vec{q} = 0$ due to a strong background of elastic scattering. This Bragg scattering was present at lattice points for which first order Bragg scattering should be absent and was traced in every case to a double Bragg scattering from two different sets of planes which simulated the forbidden scattering. Away from the lattice points phonons were not observed, and the existence of these low lying branches is questioned. This position is supported by recent optical data which will be discussed in the following paragraphs.

Figure 4 - Dispersion relation plot of BeO phonon branches mapped in the present experiment. All branches represent new data except for the longitudinal acoustic (LA) branch for the $[01\bar{1}0]$ direction.

Figure 5 - BeO dispersion relation data obtained previously by Brugger et al⁷.

TABLE I

Listing of observed phonon data obtained by neutron scattering giving the type of mode, phonon wave vector $|\vec{q}|$, neutron energy transfer ΔE and phonon frequency ω , area of observation (r&p) in k space, peak width and signal to noise ratio.

Type	$\frac{q}{q_{\max}}$	ΔE (meV)	$ \vec{q} $ ($\text{\AA}^{-1}$)	ω (10^{13} rad. sec $^{-1}$)	r&p	Peak Background	E(meV) fwhm
TA(Σ_1) [01 $\bar{1}$ 0]	.2	12.9	.2689	1.96	R(01 $\bar{1}$ 5)	50	3
	.4	24.7	.5378	3.75	L(01 $\bar{1}$ 5)	31.3	3
	.6	36.3	.8067	5.51	L(01 $\bar{1}$ 5)	21	3
	.8	47.2	1.0757	7.17	L(01 $\bar{1}$ 5)	11	3
	.9	51.7	1.2101	7.85	L(01 $\bar{1}$ 5)	5	3
	1.0	55.5	1.3446	8.43	L(01 $\bar{1}$ 5)	3	3.5
LA(Σ_1) [01 $\bar{1}$ 0]	.2	22.5	.2689	3.42	R(03 $\bar{3}$ 0)	3	6.0
	.4	41.7	.5378	6.34	R(03 $\bar{3}$ 0)	4	8.2
	.6	59.0	.8067	8.96	R(03 $\bar{3}$ 0)	5	6
	.8	70.25	1.0757	10.67	R(03 $\bar{3}$ 0)	3	2.5
	1.0	78.0	1.3446	11.85	R(03 $\bar{3}$ 0)	2	6.8
LO(Σ_1) [01 $\bar{1}$ 0]	.1	44.25	.1344	6.72	R(03 $\bar{3}$ 1)	5	6.3
	.2	48.2	.2689	7.32	R(03 $\bar{3}$ 1)	5	5
	.4	60.2	.5378	9.15	R(03 $\bar{3}$ 1)	5	4.1
	.6	73.5	.8067	11.17	R(03 $\bar{3}$ 1)	3	3.8
	.8	81.38	1.075	12.36	R(03 $\bar{3}$ 1)	2	3.8
	.9	81.38	1.2101	12.36	R(03 $\bar{3}$ 1)	1.5	3.8
	1.0	79	1.3446	12.0	R(03 $\bar{3}$ 1)	1.2	6
TA(Δ_5) [0001]	.2	6.75	.1435	1.03	A(03 $\bar{3}$ 0)	3	4
	.4	13.2	.2871	2.01	A(03 $\bar{3}$ 0)	48	3
	.6	19.88	.4306	3.02	A(03 $\bar{3}$ 0)	86	3.4
	.8	25.55	.5742	3.88	A(03 $\bar{3}$ 0)	60	2.7
	.9	28.2	.6459	4.28	A(03 $\bar{3}$ 0)	62	2.3
	1.0	30.15	.7177	4.58	A(03 $\bar{3}$ 0)	77	2.4
TO(Δ_6) [0001]	0	40.9	0	6.21	B(03 $\bar{3}$ 1)	5	6.8
	.2	40.1	.1435	6.09	B(03 $\bar{3}$ 1)	6	3.8
	.4	39.0	.2871	5.92	B(03 $\bar{3}$ 1)	7	3
	.5	38.1	.3588	5.79	B(03 $\bar{3}$ 1)	11	4.2
	.6	37.3	.4306	5.67	B(03 $\bar{3}$ 1)	14	3.3
	.7	35.7	.5024	5.42	B(03 $\bar{3}$ 1)	10	2.7
	.8	34.0	.5742	5.17	B(03 $\bar{3}$ 1)	30	2.6
	.9	32.0	.6459	4.86	B(03 $\bar{3}$ 1)	30	2.3
	1.0	30.15	.7177	4.58	B(03 $\bar{3}$ 1)	30	2.3
LA(Δ_1) [0001]	.3	17.35	.4034	2.64	B(01 $\bar{1}$ 3)	17	5
	.4	22.5	.5378	3.42	B(01 $\bar{1}$ 5)	2.5	6
	.5	29.15	.6723	4.43	B(01 $\bar{1}$ 3)	4	3.8
	.6	34.9	.8067	5.30	B(01 $\bar{1}$ 3)	2	3.8
	.8	43.3	1.0757	6.58	A(01 $\bar{1}$ 5)	2	9.8
	.9	48	1.2101	7.29	B(01 $\bar{1}$ 5)	1.2	5.3
	1.0	51	1.3446	7.75	A(01 $\bar{1}$ 5)	2	9.8

Other experimental data to be compared with present data include Loh's optical data and the elastic constants of Bente²⁶ and of Cline et al.²⁷. Loh reports observing an Σ_2 mode at 340 cm^{-1} (42.2 meV) which is 3.2% higher than our optical intercept at $40.9 \pm .74 \text{ meV}$. Although Table I indicates that the observed neutron scattering resonance at 40.9 meV was broad enough to overlap the value reported by Loh, the centroid of the observed neutron resonance was definitely lower in value than the 42.2 meV reported by Loh. Loh also reports observing another Σ_2 mode at $684 \pm 9.5 \text{ cm}^{-1}$ (84.8 meV) which coincides with the optical branches (both Σ_1 and Σ_2 types) reported previously by Brugger et al. Our acoustical branches appear to be in agreement with both sets of elastic constant data although we have not attempted to take data at $q/q_{\text{max}} < 0.2$.

Figures 6 and 7 give respectively the results of preliminary calculations by Young and Solbrig. Neither of these calculations give very good agreement with the data, nor do they give the correct slopes for the acoustic branches. Solbrig has re-evaluated the constants in his model giving much improved results, however further refinements in the model are still in order²⁸. Structure factor calculations based on Solbrig's model have been helpful in conducting the experiments, whether the frequencies obtained from the model are accurate or not.

In addition to the optical modes of Loh which we cited in a previous paragraph he has also reported other infrared and Raman active modes as: E_1 transverse at 725 cm^{-1} (89.9 meV), E_1 longitudinal at 1095 cm^{-1} (135.8 meV) and A_1 longitudinal at 1085 cm^{-1} (134.5 meV). With these additional mode frequencies and their symmetry types we have projected what the full dispersion relation looks like for the two

Figure 6 - Dispersion relation of BeO predicted from the calculation of J. A. Young³.

Figure 7 - Dispersion relation of BeO predicted by the preliminary general valence force model calculations of Solbrig⁵.

symmetry directions $[01\bar{1}0]$ and $[0001]$ in Figure 8. Branches which have been mapped by slow neutron inelastic scattering in either the present or previous experiments are displayed as solid lines. All other branches are shown as dotted lines. These unmapped branches should not be taken seriously except those intercepts at $\vec{q} = 0$ which are indicated by solid points having been established from either neutron scattering data or Loh's optical data. There remain, then, two optical intercepts at $\vec{q} = 0$ to be determined along with the mapping of 6 branches in the Σ direction and 5 branches in the Δ direction.

The rejection of the low lying optical branches previously reported by Brugger et al. is due to the fact that they have not been observed in this experiment or the optical work of Loh and all four Σ_2 branches required by symmetry¹⁶ can be accounted for without including these branches.

Two Σ_2 branches can be seen clearly in Figure 8 as having been mapped by neutron scattering (the transverse acoustic branch and a flat optical branch at approximately 85 meV), Loh has determined the intercept at $\vec{q} = 0$ for a second Σ_2 optical branch as 89.9 meV, and a third Σ_2 optical branch is required to be degenerate with the observed Σ_1 optical branch at the center of the Brillouin zone at 40.9 meV. It would seem that the low lying modes previously reported were not due to single phonon scattering but to some higher order process. While similar arguments cannot be advanced at this time to apply to the low lying Σ_1 optical branches previously reported, their existence is doubted also.

The two Σ_1 optical branches in Figure 8 which are shown at $\vec{q} = 0$ with values of 78 meV and 98 meV are unknown and merely estimated. Note

Figure 8 - Beryllium oxide dispersion relation. The solid curves represent branches which have been mapped by neutron scattering techniques while the dashed curves represent branches which have not yet been mapped. The solid points plotted at $\vec{q} = 0$ indicate intercept frequencies which have been determined by either neutron scattering or optical measurements.

that the Σ_1 optical branch, which has a value of 134.5 meV at $\vec{q} = 0$ based on Loh's data, is not degenerate with the Δ branch which Loh reports as 135.8 meV at the center of the zone.

V. CONCLUSIONS

The present data completes the acoustical branches of the dispersion relation for both $[01\bar{1}0]$ and $[0001]$ directions. These data are in good agreement with ultrasonic data which give the initial slopes of the acoustical branches. New optical branches which were measured and which intercept at $\vec{q} = 0$ at 40.9 meV are in fair agreement with recently reported infrared and Raman data. A search was made for low lying optical branches of the type reported by Brugger et al. with negative results. This fact together with the optical branches which have been measured and the infrared and Raman data of Loh indicate that the previously reported low lying optical branches are not the result of single phonon scattering. The present data help to clarify the situation for the BeO dispersion relation and with the aid of Loh's data an estimate has been made of the full dispersion relation for the $[01\bar{1}0]$ and $[0001]$ directions.

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