Extension of the Dispersion-Relation Measurements of Beryllium*

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The phonon dispersion-relation data for beryllium have been extended for waves propagating in the [01$\bar{1}$0] and [11$\bar{2}$0] directions by recent neutron scattering measurements. Measurements were confined to phonons which have their polarization vectors aligned parallel to the hexagonal plane. Branches for the [01$\bar{1}$0] direction of wave propagation designated TO(||) and TA(||) have intercept frequencies at the zone boundary of 1.72 and 1.21, respectively, in units of 10^{13} sec^{-1}. Comparison is made between these data and lattice-dynamical models of Gupta and Dayal, Iyengar *et al.*, and DeWames *et al.* The model of DeWames *et al.* gives the best agreement with the data but further improvements are desired. The Rosenstock sum Rule which has been applied to the dispersion-relation data for the symmetry directions indicates the presence of considerable nonelectrostatic or trace-variable forces. These forces are presumed to arise from the ion-core repulsion and the kinetic energy of the conduction electrons in beryllium.

I. INTRODUCTION

RENEWED interest in the dispersion relation of beryllium arose because of recent comparisons of the general features of the dispersion relation with data for magnesium and zinc[1–4] and because of recent calculation of lattice-dynamics models.[5–9] The dispersion relation of beryllium is of interest because of the information it conveys to solid-state physicists about lattice dynamics and because application of the data permits one to calculate the specific heat, neutron moderation in beryllium,[10] and other physical properties from first principles. Given a lattice-dynamical model which accurately describes the dispersion-relation data, one should be able to apply the model to the calculation of neutron scattering from polycrystalline beryllium.[10]

For these reasons the dispersion relation of beryllium has been extended by making further neutron-inelastic-scattering measurements at the Materials Testing Reactor (MTR). In this work the determination of the $\omega_j(\mathbf{q})$ relation was completed for wave propagation in the [01$\bar{1}$0] direction by measurements on the transverse optical and acoustical branches which have their polarization parallel to the basal plane. In addition, a limited amount of data was obtained for wave propagation in the [11$\bar{2}$0] direction. These dispersion-relation data, combined with the previous data,[4] have been compared with several lattice dynamical models. These models include (1) the model of Gupta and Dayal[5] (GD) which includes the extended Slutsky and Garland[4] simplified central-force model with an added contribution from the electron gas, (2) the tensor-force model (TF) of Begbie and Born[11,12] extended to include interactions out to fourth-nearest neighbors,[6] and (3) the modified axially symmetric model (MAS) of DeWames, Lehman, and Wolfram.[7] Reference to these models in the rest of the paper will be in terms of the letters given in parentheses, i.e., (GD), (TF), and (MAS), respectively. Of these models the last gives the best fit to the neutron-scattering data, although some improvement in the fit is still desired.

The Rosenstock sum rule[13] has been applied to the

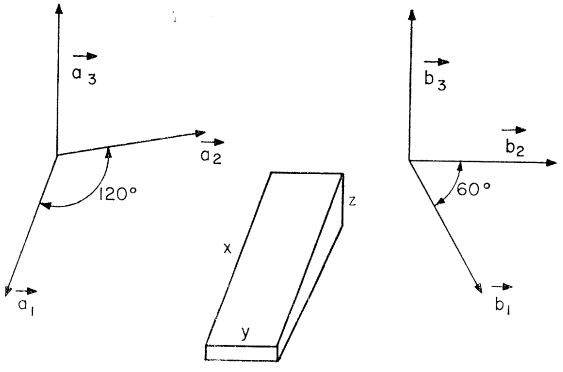

FIG. 1. Diagram showing the orientation of the direct and reciprocal lattice basis translation vectors relative to the crystal sample block.

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[1] P. K. Iyengar, G. Venkataraman, P. R. Vijayaraghavan, and A. P. Roy, in *Proceedings of the Bombay Symposium on Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1965), Vol. 1, p. 153.
[2] E. Maliszewski, Phys. Letters **1**, 338 (1962).
[3] G. Borgonovi, G. Caglioti, and J. J. Antal, Phys. Rev. **132**, 683 (1963).
[4] R. E. Schmunk, R. M. Brugger, P. D. Randolph, and K. A. Strong, Phys. Rev. **128**, 562 (1962).
[5] R. P. Gupta and B. Dayal, Phys. Status Solidi **8**, 115 (1965).
[6] P. K. Iyengar, G. Venkataraman, P. R. Vijayaraghavan, and A. P. Roy, J. Phys. Chem. Solids, Suppl. I, 223 (1965).
[7] R. E. DeWames, T. Wolfram, and G. W. Lehman, Phys. Rev. **138**, A717 (1965).
[8] M. F. Collins, Proc. Phys. Soc. (London) **80**, 362 (1962).
[9] A. Czachor, in *Proceedings of the Bombay Symposium on Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1965), Vol. 1, p. 181.
[10] J. A. Young and J. U. Koppel, Phys. Letters **16**, 235 (1965); J. A. Young, General Atomic Report GA-4638, 1963 (unpublished).
[11] G. H. Begbie and M. Born, Proc. Roy. Soc. (London) **A188**, 179 (1946).
[12] G. H. Begbie, Proc. Roy. Soc. (London) **A188**, 189 (1946).
[13] H. B. Rosenstock, Phys. Rev. **129**, 1959 (1963); H. B. Rosenstock, J. Phys. Chem. Solids, Suppl. I, 205 (1965); H. B. Rosenstock and G. Blanken, Bull. Am. Phys. Soc. **11**, 249 (1966).

dispersion-relation data, the results indicating the presence of trace-variable or nonelectrostatic forces. From theoretical calculations that have been made for the elastic constants of beryllium,[14] it is expected that these nonelectrostatic forces arise from the kinetic energy of the conduction electrons and the ion-core repulsion in the solid. In view of the important role that the conduction electrons have in the binding and stiffness of divalent hexagonal-close-packed metals, it appears that a more fundamental approach, along the lines of Toya's[15] work on sodium, is needed to describe the lattice dynamics of these metals.

II. EXPERIMENTAL PROCEDURE

The phased-chopper velocity selector has been used for the present measurements, employing basically the same techniques that were applied previously.[4,16,17] Improvements in the velocity selector have been achieved through an increase of the chopper speed to 12 000 rpm and an increase by a factor of 30 in the neutron beam flux at the sample position. The higher chopper speed is reflected in the improved time resolution of the neutron burst which has been reduced to 1.6% for 0.025-eV neutrons. All scattering angles were defined by single BF_3 detectors, 1-in. diameter×4-in. active length, placed approximately 2.45 m from the sample. The 15 scattering angles were arranged with one group of five detectors (separately connected) centered at a scattering angle of 90°, a second such group of five detectors centered at 105°, and the third group of five detectors centered at 120°. The scattering surface method[17] has been used to obtain all new data which are presented here. A channel width of 5 μsec was used in obtaining all time-of-flight data.

The same crystal has been used in this experiment as was used previously, and the sample geometry relative to the crystallographic axes is indicated in Fig. 1. In order to study the transverse acoustical and optical branches, having $\mathbf{q}$ in the $[01\bar{1}0]$ direction and with the phonon polarization vector $\boldsymbol{\varepsilon}$ parallel to the basal plane, it was necessary to orient the sample so that the lattice vector $\mathbf{a}_3$ was perpendicular to the scattering plane of the experiment. Changes in crystal orientation were made by rotation of the crystal about the $\mathbf{a}_3$ or c axis. In this way all observed scattering events were confined to the reciprocal lattice plane containing $\mathbf{b}_1$ and $\mathbf{b}_2$. The large sample dimensions in the plane of scattering (7 in.×1.72 in.) placed rather severe restrictions on the variation of sample orientation because of the need for a well-defined neutron flight path in order to make the time-of-flight measurements.

[14] B. T. Bernstein, J. Appl. Phys. **33**, 142 (1962).
[15] T. Toya, J. Res. Inst. Catalysis, Hokkaido Univ. **6**, 183 (1958).
[16] R. M. Brugger and J. E. Evans, Nucl. Instr. Methods **12**, 75 (1961).
[17] R. E. Schmunk and R. M. Brugger, Nucl. Instr. Methods **12**, 365 (1961).

III. LATTICE DYNAMICS

The general features of the lattice dynamics of hexagonal-close-packed crystals have been reviewed elsewhere,[1] and we cite here only those features which are pertinent to the present measurements. With two atoms per primitive unit cell there are three acoustical branches and three optical branches in the dispersion relation. For wave propagation along the $[01\bar{1}0]$ direction the phonon polarization for longitudinal waves is parallel to $[01\bar{1}0]$ and for transverse waves the polarization is either perpendicular $T(\perp)$ or parallel $T(\parallel)$ to the basal plane. These polarizations for the $[01\bar{1}0]$ direction of propagation are all pure, whereas for wave propagation along $[11\bar{2}0]$ only the transverse branches with polarization perpendicular to the basal plane are pure while the other four branches have mixed polarization.[18] Our interest here is focused primarily on the study of TO($\parallel$) and TA($\parallel$) for wave propagation in the $[01\bar{1}0]$ direction. A study of these branches is most easily achieved by neutron-scattering measurements in the plane of reciprocal lattice vectors $\mathbf{b}_1$ and $\mathbf{b}_2$ or the direct lattice vectors $\mathbf{a}_1$ and $\mathbf{a}_2$.

The structure factor $g_j^2(\mathbf{q})$ [19] has been evaluated previously for the TO($\parallel$) and TA($\parallel$) branches by Borogonovi *et al.*,[3] and also by Iyengar *et al.*,[1] and their results have been used as a guide in selecting a region of reciprocal space in which to perform the measurements. This approach is justified considering the close similarity in the structure factor of Be, Mg, and Zn for the other branches, even where the calculations were based on different lattice models.

Two of the lattice dynamical models, namely GD and MAS, which are compared with the data in this paper are both central-force models. In the MAS model interactions between atoms are expressed in terms of bond stretching and bond bending force constants whereas in the GD model the bond-bending constants are set equal to zero. This simplification, which was introduced initially by Slutsky and Garland[20] in their third-neighbor model, reduces the number of force constants for interactions with a given neighbor from 2 to 1. The modification which Gupta and Dayal introduced to the simplified central-force model is an electron-gas term which contributes to the dynamical matrix as well as the relations between the force constants and elastic constants. The GD model includes interactions out to fifth-nearest neighbors giving a total of six constants including the electron gas constant, whereas the MAS model includes interactions out to sixth-nearest neighbors giving a total of 13

[18] P. K. Iyengar, G. Venkataraman, K. R. Rao, P. R. Vijayaraghavan, and A. P. Roy, in *Proceedings of the Chalk River Symposium on Inelastic Scattering of Neutron in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), Vol. 2, p. 99.
[19] B. N. Brockhouse and P. K. Iyengar, Phys. Rev. **111**, 747 (1958).
[20] L. J. Slutsky and C. W. Garland, J. Chem. Phys. **26**, 787 (1957).

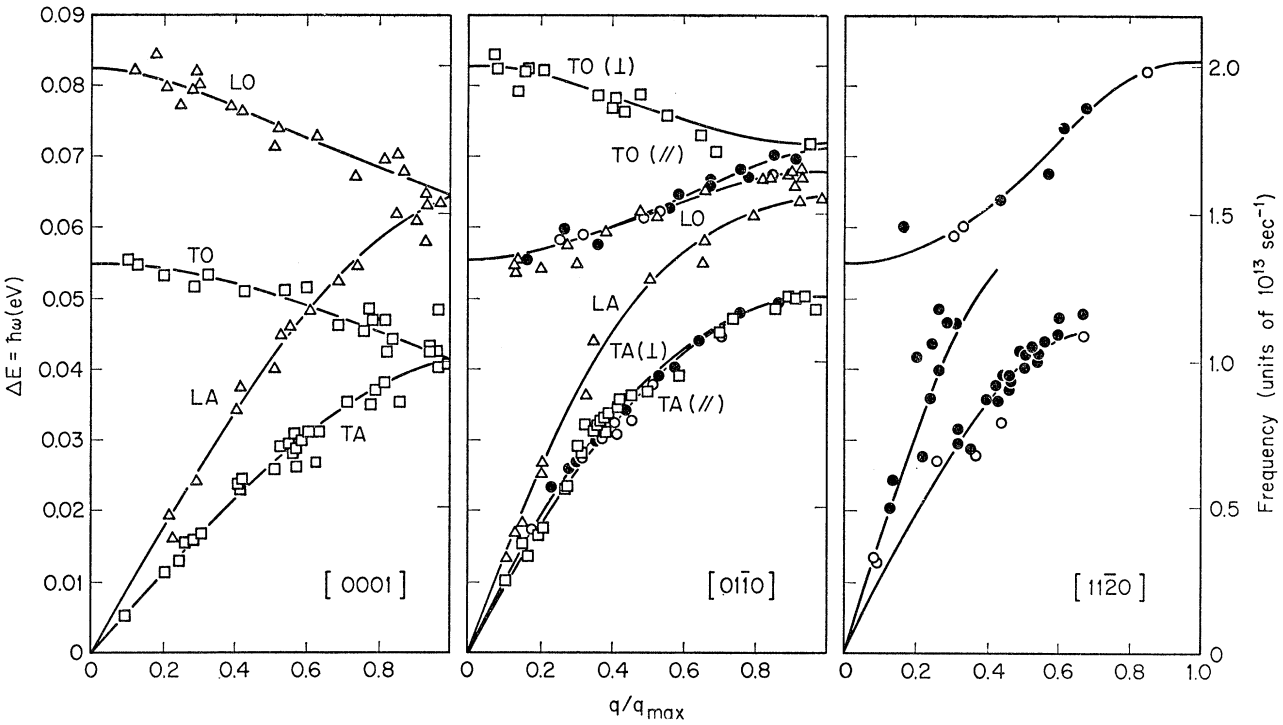

FIG. 2. Beryllium dispersion-relation data for the three crystallographic directions [0001], [01$\bar{1}$0], and [11$\bar{2}$0]. The data obtained recently are plotted as circles, open circles for neutron energy loss events and closed circles for neutron energy gain events. The smooth curves are the author's interpretation of the dispersion-relation data. Phonon polarizations labeled (||) and (⊥) for the [01$\bar{1}$0] direction are parallel and perpendicular respectively to the basal plane. Phonons for the [11$\bar{2}$0] direction all have their polarization parallel to the basal plane.

constants, one of these being an anisotropy factor. In contrast to the central-force models, the TF model of Begbie and Born,[11,12] extended by Iyengar to include interactions out to fourth-nearest neighbors, has a total of 14 force constants. However, only 11 of these constants appear in the dynamical equations for the symmetry directions.

In all three models the force constants have been forced to satisfy the equations relating them to the elastic constants of beryllium[21] i.e., conditions of elastic consistency. The extended Slutsky and Garland model, per se, has not been included because of its failure in this respect. In addition to the elastic-constant data, certain of the neutron-scattering data have also been used to evaluate the force constants. At special points in the Brillouin zone, i.e., at the center of the zone and at the zone boundaries in the [0001] and [01$\bar{1}$0] directions, the eigenvalue ($m\omega^2$) of each branch of the dispersion relation is related to a particular linear combination of the force constants in each of the models. Thus, from the three special points in the Brillouin zone we obtain ten equations relating the force constants and neutron-scattering data. Added to the five equations relating force constants and elastic constants we have a total of 15 equations which can be applied in evaluating the force constants. Force constants for the TF model have been evaluated by the author while the constants in the GD and MAS models were evaluated previously.[4,7]

IV. RESULTS AND DISCUSSION

In Fig. 2 the dispersion-relation data for beryllium are displayed for phonon propagation in the crystallographic directions [0001], [01$\bar{1}$0], and [11$\bar{2}$0]. The data which were obtained recently include the TO(||) and TA(||) branches for the [01$\bar{1}$0] direction and the data for the [11$\bar{2}$0] direction. These new data are plotted as circles, solid points for neutron energy gain and open points for neutron energy loss. Those data which have been obtained for the [11$\bar{2}$0] direction pertain to phonons which have their polarization in the basal plane, modes which are neither purely transverse nor longitudinal. All data which are displayed for the [11$\bar{2}$0] direction were obtained in the process of mapping the TO(||) and TA(||) branches for the [01$\bar{1}$0] direction. The curves displayed in Fig. 2 are not representative of a model but are the author's interpretation of the dispersion-relation data. The spread in the data points is considered to give a reasonable estimate of the errors in the dispersion-relation data, and is due primarily to the quality of the crystal.

[21] J. F. Smith and C. L. Arbogast, J. Appl. Phys. **31**, 99 (1960).

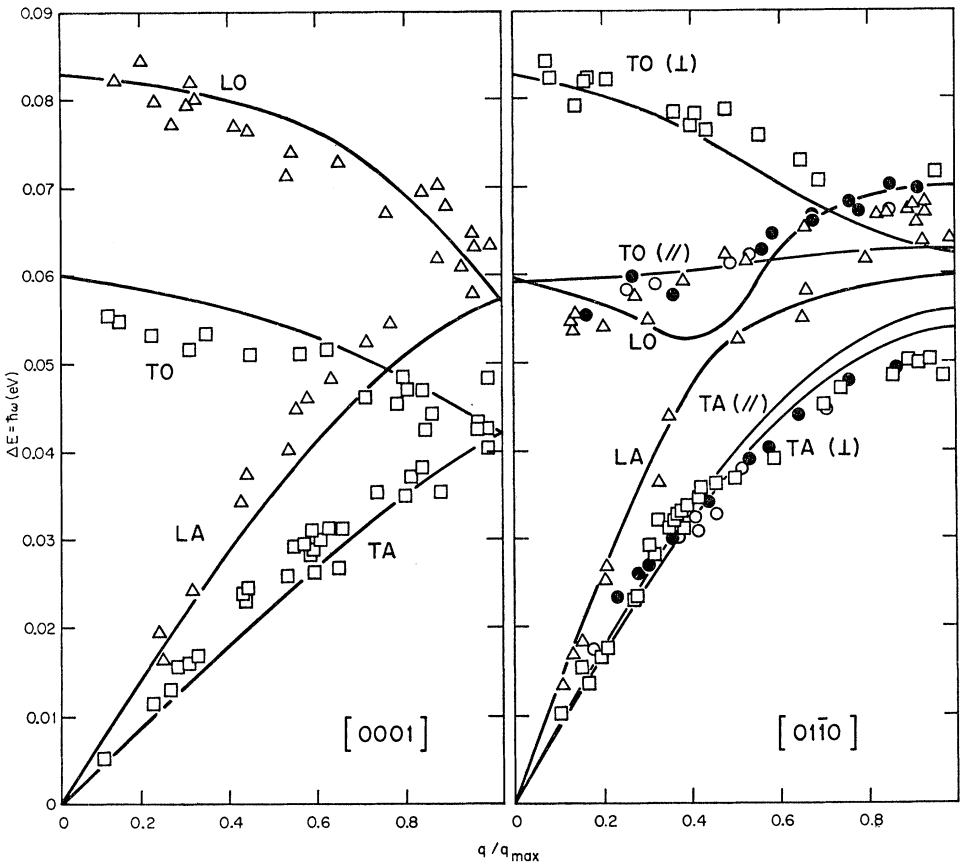

FIG. 3. Comparison of the lattice dynamical model of Gupta and Dayal, which is represented by the curves, and the experimental data.

In a previous experiment[22] in which the same equipment and techniques were used a much smoother dispersion relation was obtained. Although elastic constant data predict initial slopes that differ by 10% for TA(‖) and TA(⊥) in the [01$\bar{1}$0] direction these branches appear to be degenerate, or nearly so, in the dispersive region. This near degeneracy is not predicted from the symmetry of the structure, but is a result of the force field which is peculiar to beryllium.

In Figs. 3, 4, and 5 comparisons are given between the neutron-scattering data and the GD, TF, and MAS models, respectively, where the predictions of the models are displayed as solid lines. The electron-gas term which Gupta and Dayal added to the extended Slutsky and Garland model contributes terms to both the elastic constant-force constant equations and to the dynamical matrix in such a way that the model is elastically consistent in the long-wavelength limit. In adjusting the magnitude of the electron-gas contribution to their model, Gupta and Dayal used a value of the elastic constant C_{44} which is 25% lower than the most recent determination of the elastic constant[21,23] and makes the initial slope of the transverse acoustic (TA) branch in the [0001] direction 12% lower than the neutron-scattering data. The electron-gas term which Gupta and Dayal added to the simplified central-force model is partly responsible for the dip in the longitudinal optic (LO) branch in the [01$\bar{1}$0] direction and the low intercept for LO and longitudinal acoustic (LA) branches at the zone boundary for the [0001] direction. Use of the most recent value of the elastic constant C_{44} would have made the dip in the LO branch larger and also would have lowered the intercepts of the LO and LA [0001] branches even further and at the same time raised the lower optical branch at $\mathbf{q}=0$. Criticism of the electron-gas term used by Gupta and Dayal arises from the assumption that it is shear free, a position which is untenable for the hexagonal-close-packed divalent metals.[14,24] In addition, Krebs has pointed out that this model violates symmetry requirements.[25] No attempt has been made to adjust the force constants in the GD model in view of the shortcomings of the model.

For both the TF and MAS models an attempt was made to evaluate the force constants in the models by a least-squares solution of the set of equations relating force constants and experimental data on an IBM 7040

[22] R. E. Schmunk and W. R. Gavin, Phys. Rev. Letters **14**, 44 (1965).

[23] Previous values of the elastic constants were due to L. Gold, Phys. Rev. **77**, 390 (1950). In Gold's measurements the value of C_{44} was determined from ultrasonic measurements on polycrystalline beryllium.

[24] J. R. Reitz and C. S. Smith, Phys. Rev. **104**, 1253 (1956).

[25] K. Krebs, Phys. Rev. **138**, A143 (1965).

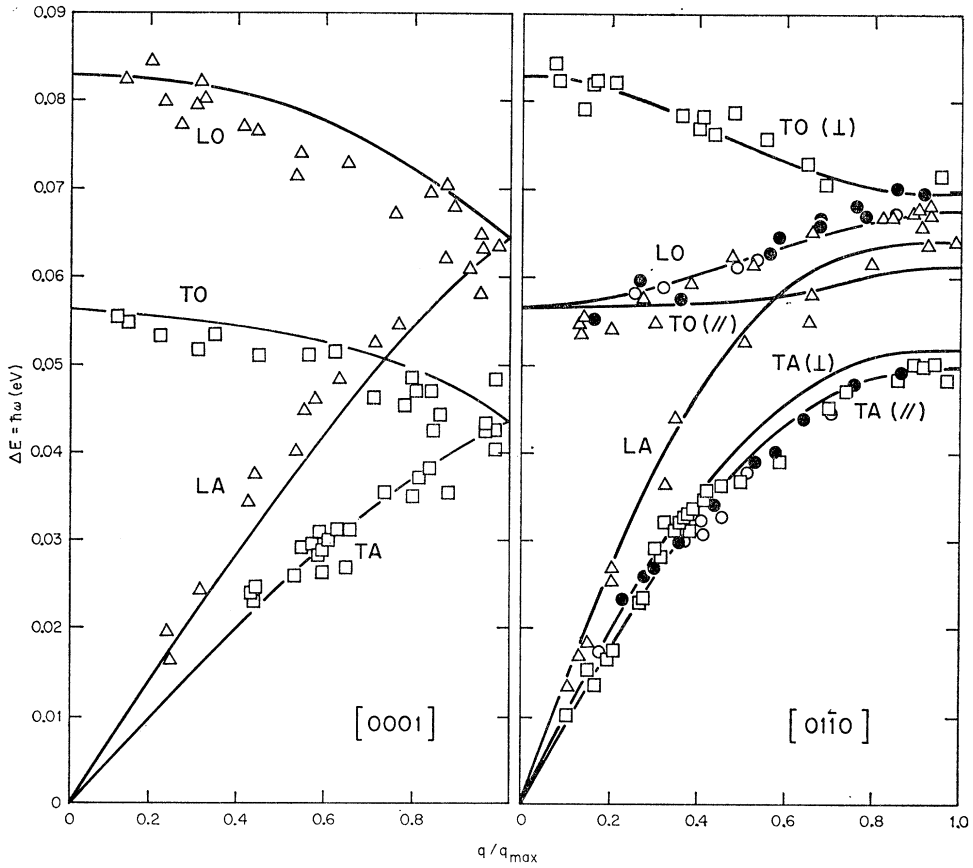

FIG. 4. Comparison of the tensor force lattice dynamical model of Iyengar *et al.*, which is represented by the curves, and the experimental data.

computer. With both models the full set of 15 equations referred to in the previous section was used. Results of these attempted calculations were negative, in part because of the input experimental data and the ill condition[26] of the coefficient matrices. As a result of these difficulties the force-constant values used in comparison of the models with the data are the results of a hand calculation for both the TF and MAS models, in the latter model by DeWames *et al.* in their published paper.[27]

The fourth-neighbor TF model is compared with the neutron-scattering data in Fig. 4. Because of the difficulties cited in the previous paragraph, only part of the neutron-scattering data were used in evaluating the force constants. Values assigned to the force constants in the notation of Iyengar *et al.*,[6] are as follows: α, 1.753; β, 0.120; γ, 0.636; λ, 0.318; μ, -0.225; ν, 1.557; ξ, 0.553; η, -0.473; ζ, 0.389; θ, 0.373; ϕ, 0.649; all in units of 10^4 dyn cm^{-1}. Difficulties with this model are particularly evident for the [01$\bar{1}$0] direction where the intercepts at the zone boundary are considerably in error. In the tensor force model the dispersion curves for the [01$\bar{1}$0] direction are described primarily in terms of interactions with first- and second-nearest neighbors with only minor contributions from third-neighbor forces and no contribution from fourth-nearest neighbors. Hence, the requirements placed on the force constants are too restrictive, and interactions with more distant neighbors are needed in order to relax these restrictions and obtain a better fit to the experimental data. In the modified axially symmetric model interactions with fifth- and sixth-nearest neighbors contribute significantly to the dispersion relation for the [01$\bar{1}$0], and the same would be expected for the tensor force model extended to include interactions with these more distant neighbors.

The best agreement between models and experiment is obtained for the MAS model, displayed in Fig. 5. For the symmetry direction [0001] and [01$\bar{1}$0] the model agrees with the data except for the transverse optic (TO)(||) and the TA(⊥) branches along [01$\bar{1}$0], the disagreement being primarily in the TO(||) branch. Consideration of the small amount of data which has been obtained for the [11$\bar{2}$0] direction indicates further disagreement with the MAS model; the optical branch plotted for this latter direction rises to a value about 20% higher than the model predicts. In spite of these differences and the difficulties encountered in attempting to adjust the force constants to agree with the recent data, the MAS model gives the best agreement with the neutron-scattering data and is the most satisfying from the standpoint of the physical basis for the model. Although it has not been demonstrated

[26] N. Mendelsohn, Am. Math. Monthly **63**, 285 (1956).
[27] R. E. DeWames (private communication).

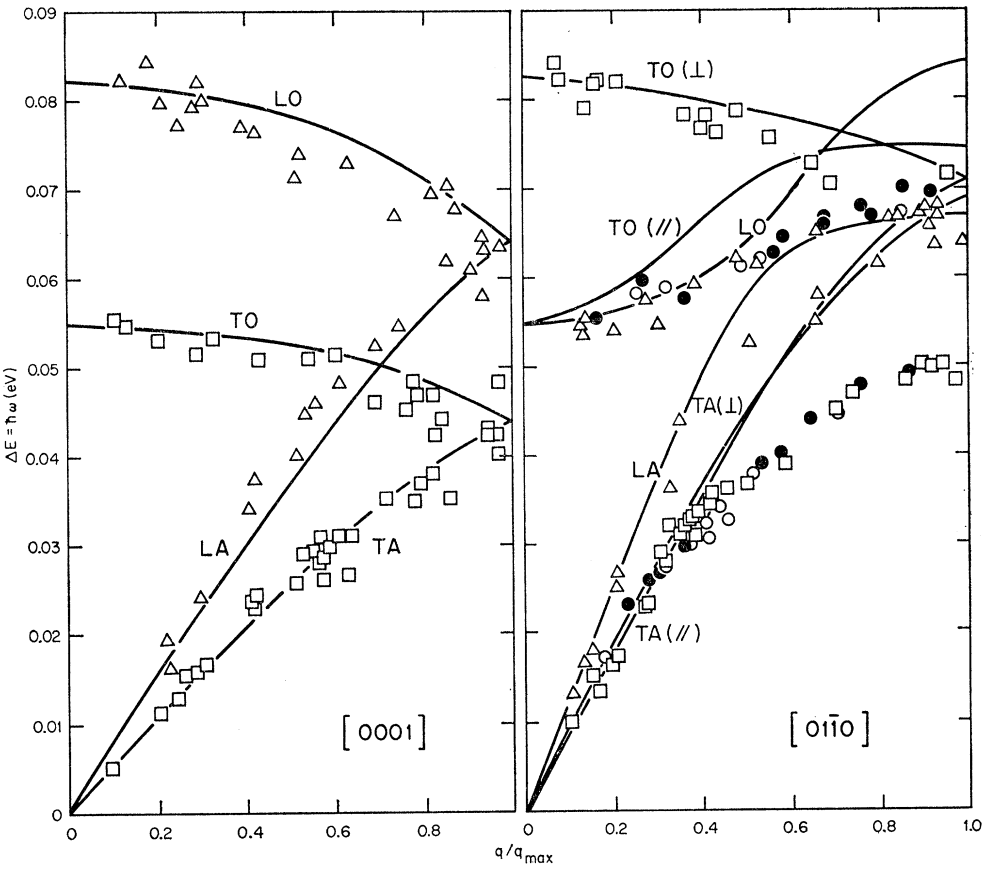

FIG. 5. Comparison of the central force lattice dynamical model of DeWames *et al.*, which is represented by the curves, and the experimental data.

here, the possibility exists that a better fit between the model and the data can be obtained.

Rosenstock[13] has described a sum rule that can be applied to the dispersion-relation data. The sum rule states that $\Sigma_i \omega_i^2(\mathbf{q}) = \text{constant}$, where ω_i is the frequency of the ith branch, and the sum extends over all branches for a given wave vector $\mathbf{q}$. Rosenstock has shown that the sum rule holds for cases where the interatomic forces can be classed as electrostatic or as interactions between unlike atoms. In monatomic crystals such as beryllium where the structure can be described in terms of atoms arranged on interpenetrating simple hexagonal lattices, atoms which are arranged on a given sublattice are denoted as "like" atoms and atoms on different sublattices as "unlike" atoms. Deviation of the sum from a constant value is interpreted as evidence for the existence of nonelectrostatic forces. The sum rule has been evaluated from the beryllium dispersion relation data for the [0001] and the [01$\bar{1}$0] directions. In Fig. 6 the sum rule data are plotted in the form $[\Sigma_i \omega_i^2(\mathbf{q}) - \Sigma_i \omega_i^2(0)]/\Sigma_i \omega_i^2(0)$ versus the normalized phonon wave vector. Comparing the results for the two directions it is apparent that the contributions from nonelectrostatic forces differ considerably for the two symmetry directions and in fact are quite strong for the [01$\bar{1}$0] direction. For comparison with the beryllium data the sum rule was computed from published dispersion relation data for Mg[1] and Zn[3] at the Brillouin-zone boundaries for the two symmetry directions cited above with the following results: at the [0001] boundary the sum rule gave 0.05 and 0.0 and at the [01$\bar{1}$0] boundary 1.37 and 2.67 for Mg and Zn, respectively.[28] The sum rule suggests, then, that electrostatic forces and/or forces between "unlike" atoms tend to dominate the dynamics of the structure for phonon propagation along the [0001] direction for Be, Mg, and Zn, and that for phonon propagation in the [01$\bar{1}$0] direction there are additional contributions

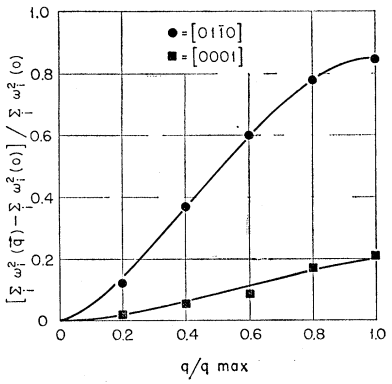

FIG. 6. The Rosenstock sum rule evaluated from the beryllium experimental data and plotted as a function of phonon wave vector for the [0001] and [01$\bar{1}$0] directions.

[28] In order to evaluate the sum rule for Mg at the zone boundary in the [01$\bar{1}$0] direction it was necessary to use Iyengar's quoted values for the force constants of the tensor force model as they have measured only 5 of the 6 branches for that direction.

from nonelectrostatic forces. From calculations of the elastic shear constants of beryllium[14] it is expected that the nonelectrostatic forces of interest are contributed by the kinetic energy of the conduction electrons and ion-core repulsive interactions. The relative importance assigned to these two contributions was found by Bernstein[14] to be sensitive to the energy-band model chosen for the calculations. Harrison[29] has attempted to calculate the physical properties of polyvalent metals from first principles and has obtained some information about the phonon spectrum in Zn, and for Al has obtained a dispersion relation. Further calculations of this type are needed to give an adequate description of the dispersion relations in polyvalent metals, and they are certainly desirable for giving a unified approach to various physical properties.

In calculating quantities such as frequency distributions, specific heat, and Debye-Waller factor for Be from the lattice dynamical models one would certainly expect to obtain the best results from the MAS model of DeWames *et al.*, in view of the better agreement between this model and the neutron-scattering data. Young and Koppel[10] have applied the extended Slutsky-Garland model to the calculation of coherent inelastic neutron scattering from polycrystalline Be, a case where there is justification for using a simpler model. This model fits the dispersion-relation data with maximum deviations of about 10%, although it does not satisfy the elasticity conditions in the long-wavelength limit. In view of the fact that an accuracy to within 10% is acceptable for the neutron-scattering kernel calculations, the simplified central-force model should suffice for this purpose and is expected to require less computation time than the more elaborate models.

[29] W. A. Harrison, Phys. Rev. **129**, 2512 (1963); **136**, A1107 (1964).

V. CONCLUSIONS

Comparison of the extended dispersion-relation data for Be with lattice dynamical models reveals continued discrepancies between the models and experimental data, although considerable improvement has been made in the modified axially symmetric model of DeWames *et al.* Application of Rosenstock's sum rule test to the dispersion-relation data demonstrates the presence of trace-variable or nonelectrostatic forces in the dynamics of the solid. From theoretical calculations of the elastic shear constants of beryllium it is expected that these nonelectrostatic forces are due to the kinetic energy of the conduction electrons and ion-core repulsive interactions.

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Influence of Impurities on the de Haas–van Alphen Effect

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The influence of impurities, or solute atoms, upon the de Haas–van Alphen effect associated with a metal having a Fermi surface of arbitrary shape is investigated. It is shown that the well-known result of Dingle for the decrease in amplitude of the oscillations in the magnetic susceptibility may be derived without recourse to the additional phenomenological assignment of a specific line shape to each Landau level. The relaxation parameter used by Dingle is shown to be *twice* the lifetime of a state at the Fermi energy. This is compared with the relaxation time for electrical conduction for a class of impurity potentials of variable range in real space. The derivation of the Dingle result presented here depends only upon certain continuity arguments related to the effect of impurities on the electronic band structure. These are investigated in detail for the free-electron model. Simultaneously, the method yields the change in period of the oscillations upon alloying. The conditions under which the rigid-band model is applicable to this problem are derived and a possible generalization is suggested to account for the changes in period which are observed when the solute and solvent have the same valency. The information which can be obtained from experiment concerning the change in amplitude and period is correlated with previous theoretical studies.

I. INTRODUCTION

THE qualitative aspects of the influence of impurities on the amplitude of the oscillatory component of the magnetic susceptibility of metals [de Haas–van Alphen (dHvA) effect] have been understood ever since the pioneering work of Dingle.[1] On the basis of an assumed Lorentzian line shape, characterized by a "relaxation time" τ, for every Landau level, he showed that each harmonic in the oscillatory magnetization was modified in magnitude by the multiplicative factor $\exp[-2\pi l/\omega_c\tau]$, where $\omega_c=eB/mc$ is the cyclotron frequency and l is the order of the harmonic. Moreover, Dingle's treatment, originally valid only for free electrons, has since been extended

[1] R. B. Dingle, Proc. Roy. Soc. (London) **A211**, 517 (1952).