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MACS J
A PROGRAM FOR COMPUTING CRYSTAL PHONON DISPERSION RELATIONS,
STRUCTURE FACTORS FOR NEUTRON INELASTIC SCATTERING,
AND JACOBIAN MATRICES

J. K. Boyter and A. W. Solbrig, Jr.

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U. S. ATOMIC ENERGY COMMISSION

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IDAHO NUCLEAR CORPORATION

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MACS J
(PPCo 40*0929)

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ABSTRACT

This report describes a FORTRAN IV program MACS J (PPCo 40*0929) which computes (a) the dynamical matrix for the harmonic oscillations of a crystal, (b) the eigenvalues and eigenvectors of this matrix, (c) the corresponding structure factors for coherent single-phonon scattering of neutrons, and (d) Jacobian matrices for use in adjusting force constants to fit calculated dispersion curves to observed dispersion curves. Appendixes contain discussion of fitting procedures and the linearity assumption as well as sample input for calculation of a Jacobian matrix.

Running time on an IBM 7044 (32K, batch processing) for the 7 problems required to calculate a Jacobian with 8 rows and 6 columns for BeO (4 atoms per primitive cell) is less than 5 minutes.

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MACS J

A PROGRAM FOR COMPUTING CRYSTAL PHONON DISPERSION RELATIONS, STRUCTURE FACTORS FOR NEUTRON INELASTIC SCATTERING, AND JACOBIAN MATRICES

I. GENERAL DESCRIPTION OF PROGRAM

The FORTRAN IV program MACS J (PPCo 40*0929) computes certain quantities which are of interest in the theory of harmonic oscillations of a crystal and in the theory of inelastic scattering of neutrons from a crystal. These quantities are (a) the dynamical matrix, (b) eigenvalues and eigenvectors of this matrix, (c) corresponding structure factors for coherent single-phonon scattering of neutrons, and (d) rows from Jacobian matrices for use in a program⁽¹⁾ which adjusts force constants to fit calculated dispersion curves to observed dispersion curves⁽²⁾. The eigenvalues lead directly to the dispersion relations for the crystal. The structure factors are useful in planning and interpreting certain neutron scattering experiments. The structure factors are also an important part of the calculation of cross sections for inelastic scattering of neutrons from crystalline or polycrystalline materials. The calculation of the dynamical matrix is especially adapted to the use of a potential in valence coordinates⁽²⁾, but the input allows use of a potential in Cartesian coordinates.

MACS J is a modification of MACS M⁽³⁾. We assume that the reader has available the description of MACS M contained in Ref. (3). The ways in which MACS J differs from MACS M are described in this section. More detailed discussion of the first item below, calculation of

Jacobian matrices, is given in Section II. Appendix A includes discussion of fitting procedures and the linearity assumption. Appendix B gives sample input for calculation of a Jacobian matrix.

a. The program permits calculation of rows from Jacobian matrices for use in the ADJUSTER program described in Ref. (1). These rows are not only printed but are also punched on cards which may be used directly as input to the ADJUSTER program. The Jacobian consists of an array of first partial derivatives of eigenvalues with respect to force constants. These first derivatives are approximated numerically with the aid of the "force constant changer" feature (Control Card 3 and Card Types 4, 5) of MACS M. The Jacobian calculation is controlled by a punch in Control Card 3, Column 20. This punch is the only change in input card format from MACS M.

b. Internal changes have been made which speed up and make more accurate both the calculation of the $\underline{D}$ matrix from the $\underline{D}_{0,l}$ matrices and the calculation of the $\underline{F}$ matrix from the $\underline{F}_{0,l}$ matrices.

c. Suppose a change in the $\underline{F}_{0,l}$ matrices is called for (Control Card 3 and Card Types 4, 5) in a given problem: (1) The initial force constants (X_i) and the changed force constants (Y_i) are printed. (2) The changed $\underline{F}_{0,l}$ are used in that problem. (3) Changed $\underline{F}_{0,l}$ are printed if called for by a 0 punch in columns 25 and/or 30 of Control Card 2. (4) If Control Card 1 has a 1 punch in columns 10 and 30, the changed $\underline{F}_{0,l}$ are stored on tape for later use as desired. (5) If Control Card 1 has a 0 punch in columns 10 and/or 30, then the initial $\underline{F}_{0,l}$ remain on tape. Notice that only (1), (3), and (4) differ from MACS M.

d. If a raw eigenvalue is negative, then the eigenvalue in wave numbers and the eigenvalue in electron volts are meaningless because they are proportional to the square root of the raw eigenvalue. The corresponding raw Jacobian element may be meaningful, but the Jacobian element in electron volts is meaningless. If the program computes a negative raw eigenvalue, then the number 7.7777E 07 is printed for the corresponding eigenvalue in wave numbers and for the corresponding eigenvalue in electron volts. This number is used in the calculation of the corresponding Jacobian element in electron volts, which is of course meaningless.

II. CALCULATION OF JACOBIAN MATRICES

1. Introduction

Our Jacobian matrix is defined as an array J of elements

$$J_{st} = - \frac{\partial v_s}{\partial f_t},$$

where the v_s are raw or eV eigenvalues and the f_t (not to be confused with the components f_1, f_2, f_3 of the wave vector $\underline{k}$) are the distinct nonzero force constants appearing in the matrices $F_{0,l}$. We have inserted a minus sign in the definition in order to achieve compatibility with Ref. (1). The program calculates J_{st} by use of the first difference approximation

$$\begin{aligned} J_{st} &= - \frac{\Delta v_s}{\Delta f_t} \\ &= - \frac{v_s(f_1, \dots, f_t + \Delta f_t, \dots, f_m) - v_s(f_1, \dots, f_t, \dots, f_m)}{\Delta f_t}. \end{aligned}$$

This approximation should give reasonably accurate results if there exist Δf_t such that (a) the remainder term (related to second derivatives) in the Taylor series for Δv_s is small and (b) the program can compute a reasonably accurate value for Δv_s by the subtraction indicated in the formula above. Satisfying (a) often requires small Δf_t , but satisfying (b) requires Δf_t not too small. (Clearly if Δf_t is "too small", then numerical calculation of Δv_s with a fixed-word-length computer by the indicated subtraction will lead to numerical nonsense.)¹¹

It is feasible to calculate rows of J for use in the ADJUSTER program by use of MACS M or by use of MACS J without invoking the Jacobian calculator feature of MACS J. The procedure would be as follows: (1) Compute by machine a set of v_s for the initial $f_1, f_2, \dots, f_m$. (2) Compute by machine a set of v_s with f_1 only changed to $f_1 + \Delta f_1$, a set of v_s with f_2 only changed to $f_2 + \Delta f_2$, etc. It is simple to use the "force constant changer" to generate the $F_{0,l}$ matrices containing the changed f 's. (3) Calculate by hand the J_{st} elements, recording the results directly on a form for keypunching. Each element requires only one subtraction and one division. (4) Punch by hand operation of a keypunch machine the cards for entry into the ADJUSTER program. (5) Check the punching of the cards by hand operation of a verifier machine or by visual inspection of the cards. If (as in our

* Long, Gravenor, and Woodger⁽⁴⁾ use a formula from perturbation theory for calculating the Δv_s . Their method seems to have some advantages, including almost complete elimination of the "crossover" error discussed in Section II.4 below.

current work on Be0) there are 25 frequencies and 6 force constants, then there are 150 J_{st} elements to handle. This is not a prohibitively difficult job, but it is tedious; furthermore, human errors, especially in steps (3), (4), and (5), can cause the ADJUSTER program to give misleading results. We considered it worthwhile to develop a program which allows the machine to perform steps (1)-(4) in a single run. We are aware that the input to the Jacobian calculator portion of MACS J involves duplication and could be shortened, but the program is still reasonably easy to use; and it seems to work correctly.

The program MACS J may punch rows of J corresponding to frequencies which are not being fitted. In this case, the user may manually select those cards corresponding to the frequencies being fitted. Certain punches (described in Section II.3 below) in columns 73-80 assist the user in this selection. Alternatively, the user may (a) leave some extra rows in the Jacobian deck for the ADJUSTER program and then (b) not fit the frequencies corresponding to these rows by putting zero weights in the corresponding positions in the diagonal weighting matrix W .

2. Procedure

Here we describe in general terms the input for and output from the Jacobian calculator portion of MACS J. In Appendix B, we present a specific example comprising part of the written card input for key-punching. Except for a punch (IJAC) in Control Card 3, Column 20, controlling the J calculation, the format of input cards for MACS J is identical to the format of input cards for MACS M. The IJAC = 1 if a JACobian column is to be calculated.

Suppose that we are given a set of ℓ vectors and $D_{0,\ell}$ matrices on cards, a wave vector $\underline{k}$ in terms of its components f_1, f_2, f_3 on cards, a matrix A on cards, and a set of $F_{0,\ell}$ matrices on cards. Suppose that the $F_{0,\ell}$ cards contain m distinct nonzero entries $f'_1, f'_2, \dots, f'_m$ (not to be confused with the components f_1, f_2, f_3 of the wave vector $\underline{k}$). Suppose further that we wish to evaluate a Jacobian matrix at a set of $F_{0,\ell}$ matrices which (a) have nonzero entries in positions identical to those of the nonzero entries in the $F_{0,\ell}$ matrices on cards and (b) have m distinct entries $f_1, f_2, \dots, f_m$ which may or may not have the same value as the $f'_1, f'_2, \dots, f'_m$ in the $F_{0,\ell}$ matrices on cards. In other words, the $F_{0,\ell}$ matrices on cards merely serve as a pattern giving the location of the distinct nonzero force constants. We wish to evaluate a Jacobian at a set of $F_{0,\ell}$ matrices with this same pattern of nonzero force constants but with perhaps different numerical values of force constants. To evaluate this Jacobian requires $m+1$ "passes" or problems. These passes should be performed consecutively; and for each pass the D matrix, the wave vector $\underline{k}$, and the A matrix should be the same. Let a superscript j on the $X_i^{(j)}$ and $Y_i^{(j)}$ of Card Types 4 and 5 indicate the "pass number".

Pass 0: Control Card 1 should be such that the following quantities are on tape at the close of this pass: (1) The $F_{0,\ell}$ to be used in the J calculation. These $F_{0,\ell}$ should contain m distinct nonzero force constants $f_1, f_2, \dots, f_m$. (2) The A matrix to be used in the J calculation. (3) The components f_1, f_2, f_3 (not to be confused with force constants) of the wave vector $\underline{k}$ to be used in the J calculation. Column 65 of Control Card 1 should contain a 1 punch.

Control Card 2 for printing or not printing various quantities, as well as the cards for computing structure factors and/or transformed eigenvectors, may be entered as desired by the user in accord with the description in Ref. (3) and in Section I above. The following quantities and always printed and labeled on Pass 0: X's, Y's, components of the wave vector κ (labeled "F"), eigenvalues, trace check, eigenvectors.

On Control Card 3 put NOCH = m, IJAC = 0 (Column 20). (The NOCH is a mnemonic or acronym for NO. of CHanges.)

On Cards Type 4 put $X_j^{(0)} = f'_j$,
and on Cards Type 5 put $Y_j^{(0)} = f_j$, $\left. \begin{array}{l} \\ \end{array} \right\} j = 1, 2, \dots, m.$

Pass j (j = 1, 2, \dots, m): Control Card 1 should contain 0's in columns 5, 10, 15, \dots, 40. Columns 41-70 should be identical to the corresponding columns from Pass 0.

Control Card 2 for printing or not printing various quantities, as well as the cards for computing structure factors and/or transformed eigenvectors, may be entered as desired by the user in accord with the description in Ref. (3) and in Section I above.

On Control Card 3 put NOCH = IJAC = 1. On Card Type 4 put $X_1^{(j)} = Y_j^{(0)}$, and on Card Type 5 put $Y_1^{(j)} = Y_j^{(0)} + \Delta f_j$. In the calculation of Δf_j for use as a divisor in the Jacobian element, the program subtracts the $Y_j^{(0)}$ entered in Pass 0 from $Y_1^{(j)}$. When IJAC = 1, the program does not compute a D matrix. Instead it uses the D matrix left on tape 2 by Pass 0.

The following quantities are always printed and labeled on Pass j (j = 1, 2, \dots, m): X's, Y's, components of the wave vector κ

(labeled "F"), eigenvalues, trace check, eigenvectors, J elements (raw and eV) computed in Pass j . On Pass m only, the check numbers

$$(\text{raw eigenvalue})_k + \sum_{j=1}^m J_{kj} Y_j^{(0)}$$

are always printed and labeled. Here the $(\text{raw eigenvalue})_k$ are from the output of Pass 0, and the $Y_j^{(0)}$ are from the input to Pass 0. In a "good" check, $|\text{check number}|_k \ll |\text{raw eigenvalue}|_k$ --except for acoustic modes at $k = 0$. (See Appendix A.3, 4 for further discussion on this check.) On Pass m only, the program always prints and labels the computed raw and eV J matrices. On Pass m only, the program always punches on cards the computed raw and eV J matrices. The printed and punched J matrices are in the WRIM format described on pages 11-12 of Ref. (3). The punched card output is discussed further in Section II.3 below.

If there is another problem for this machine run, this problem should have IJAC = 0 on Control Card 3. The other cards for the problem may be entered as desired by the user in accord with the description in Ref. 3 and in Section I above.

3. Punched Card Output

Each number on a card is in a given row of J ; e.g., $J_{i,1}$, $J_{i,2}$, . . . , $J_{i,6}$ may be on one card. If a card contains fewer than six Jacobian elements, the remaining entries on the card may be meaningless junk; however, this fact does not affect the correct use of the cards in the ADJUSTER program. The WRIM format punches in columns 73-77 (see pages 11-12 of Ref. (3)) may aid the user in selecting from the cards punched by MACS J those cards which he wishes to use in the

ADJUSTER input. The punched "RAW" or "EV" in columns 78-80 may also be of aid in identifying the cards desired for use in ADJUSTER. The ADJUSTER makes no use of columns 73-80 of the J cards.

4. "Crossover" Error

The kind of error which we call "crossover" error occurs because the program prints raw eigenvalues in order of decreasing magnitude; that is,

$$(\text{raw eigenvalue})_{1\geq} (\text{raw eigenvalue})_{2\geq} \cdots \geq (\text{raw eigenvalue})_{\text{last}}.$$

In case some eigenvalues from Pass 0 are close together, then these eigenvalues may interchange, or "cross", in the passes which compute Jacobian elements. If this happens, then some of the Jacobian elements are almost certainly in error because an incorrect pairing of eigenvalues has been assumed in the calculation of the Δv 's. This sort of error may be indicated by failure of the printed check on the raw Jacobian. If the crossover involves accidental degeneracy between inequivalent representations then examination of the eigenvectors, which are always printed with each pass, should show how to pair eigenvalues correctly. In case the symmetries of the printed eigenvectors are not readily apparent, the user may find it helpful to examine structure factors and/or transformed eigenvectors. The program is sufficiently flexible to allow structure factors and/or transformed eigenvectors to be calculated and printed in any problem, or pass.

If the crossover involves accidental degeneracy between

equivalent representations^{*}, determination of the correct pairing of eigenvectors may be difficult. If this sort of crossover error is anticipated or suspected in a certain part of the Jacobian, it might be worthwhile to calculate that part again with Δf_j replaced by $-\Delta f_j$. If this replacement causes abnormally large changes in some of the Jacobian elements, then these elements may be afflicted by crossover error.

Once the correct pairing is known, the corrected Δv 's and the corresponding J elements may be computed by hand. The J elements can be keypunched by hand into the proper locations in the J cards.

^{*} It is said (see Ref. (8) for example) that this sort of crossover cannot occur. However, our experience with BeO and the experience of Raubenheimer and Gilat⁽⁸⁾ with Zn show that crossover of this sort may occur with simplified sets of force constants. We found Heine's brief discussion⁽⁹⁾ of different types of degeneracy to be illuminating.

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APPENDIX A

FITTING PROCEDURES AND THE LINEARITY ASSUMPTION

1. Need for a Fitting Procedure

Sometimes it is possible to fit observed phonon dispersion curves by use of a harmonic potential function.⁽²⁾ If this is possible, then the force constants in the potential function are of interest for both theory and applications. The force constants afford an opportunity and a challenge for theoretical explanation on the basis of more fundamental physical concepts. The force constants may be used to predict those portions of the dispersion curves which have not been observed experimentally. The complete dispersion relations and the calculated normal modes may then be used to calculate the phonon distribution function, thermodynamic functions, neutron scattering cross sections, and perhaps other properties of the material.

2. Discussion of Fitting Procedures

It might be possible to select force constants to fit observed curves by a procedure which is largely human, or manual. The human worker can compare observed and calculated curves. He can examine the computed eigenvectors and the nature of the force constants involved, perhaps taking into account the results of group theoretical calculations^(5,6). Then he can attempt to adjust the force constants to improve fit between calculated and observed curves. By use of judgment and insight, he might obtain good fit after considering the results of repeated changes in force constants.

On the other hand, a highly automated procedure may be envisioned. One might formulate a quantitative criterion by which the goodness of fit is to be judged. Then one might set up some mathematical devices which should lead to a set of force constants which yield a better fit in terms of the quantitative criterion. By an iterative process, which might be programmed for a computer, a set of force constants which yield a value of the fit criterion smaller than some preassigned value might be obtained.

We have made efforts to develop both of the above methods.* With regard to the "highly automated" method, we agree with Long and other workers⁽⁴⁾: "Experience supports the view . . . that although in principle the whole sequence of refinement can be programmed automatically at this stage of our knowledge it is essential to reflect on the results of each stage of the calculation before proceeding with the next." With regard to the "manual" method, our work indicates that it is too demanding in terms of insight, patience, and time to be generally successful.

We have attempted to set up and apply a compromise method in which human judgment is aided by a force constant ADJUSTER program⁽¹⁾. This program predicts force constants on the basis of a linearity assumption to minimize a weighted least squares deviation. Human judgment seems to be essential in the assignments of calculated to observed points and in the decisions regarding the types of force constants that must be included to obtain a satisfactory fit.

*The work on a "highly automated" method was done by Mr. C. Noble and Dr. H. L. McMurtry.

3. Remarks on the Linearity Assumption

It is desired to predict at least approximately the changes in force constants which will minimize a weighted least squares deviation of calculated frequencies from observed frequencies. In order to do this conveniently, the assumption is made that the changes in calculated frequencies are linear functions of changes in force constants: In the Taylor series, terms involving second and higher derivatives are ignored. This linearity assumption is sometimes inaccurate. We shall not discuss the doctrine of least squares or its vagaries, but we shall present some remarks about the linearity assumption.

Let the column vector $\underline{f}$ have as elements the m distinct nonzero force constants f_t (not to be confused with the components f_1, f_2, f_3 of the wave vector $\underline{\kappa}$) in the $F_{0,\underline{\kappa}}$ matrices. Let the column vector $\underline{\nu}$ have as elements the n frequencies ν_s under consideration. These frequencies are a complicated function of $\underline{f}$, $\underline{\nu} = \underline{\nu}(\underline{f})$. Here the elements of $\underline{\nu}$ may all be quantities proportional to squared frequencies (raw eigenvalues), or they may all be quantities proportional to frequencies (wave number or eV eigenvalues). Let $\underline{J} = \underline{J}(\underline{f})$ denote an $n \times m$ Jacobian matrix with elements $J_{st} = -\frac{\partial \nu_s}{\partial f_t}$ evaluated at $\underline{f}$. The linearity assumption is

$$\underline{\nu}(\underline{f} + \Delta \underline{f}) - \underline{\nu}(\underline{f}) = -\underline{J}(\underline{f}) \Delta \underline{f},$$

or $\Delta \underline{\nu} = -\underline{J} \Delta \underline{f}$. Presumably this assumption is fairly accurate in the neighborhood of $\underline{f}$, or for small $\Delta \underline{f}$. Now in our harmonic approximation, the raw eigenvalues $\nu_s(\underline{f})$ are eigenvalues of a Hermitian matrix $\underline{M}(\underline{f})$, where the elements of $\underline{M}$ are linear homogeneous functions of the f_t . It is not difficult to see that the raw eigenvalues are linear

homogeneous functions of the force constants f_t ; that is, for any real number a ,

$$v_s(af_1, af_2, \dots, af_m) = a v_s(f_1, f_2, \dots, f_m), s = 1, 2, \dots, n,$$

or

$$v(af) = a v(f).^*$$

By Euler's theorem on homogeneous functions,

$$v_s(f_1, \dots, f_m) = \sum_{t=1}^m \frac{\partial v_s}{\partial f_t} f_t, s = 1, 2, \dots, n,$$

or

$$v(f) = -J(f) f;$$

therefore

$$a v(f) = -J(f) af,$$

or

$$v(af) = -J(f) af.$$

It follows that if $f + \Delta f = af$, or if $\Delta f = (a - 1)f$, then the linearity assumption is exact for the raw eigenvalues. We conclude that for raw eigenvalues the linearity assumption is fairly accurate in a small region about the point $\Delta f = 0$ and in a small region about the "straight line" in m -dimensional space defined by $\Delta f = (a - 1)f$.

*(1) This result does not imply that the v_s are linear functions of any one of the f_t . For example, the eigenvalues of the Hermitian matrix $\begin{pmatrix} a & c \\ c & b \end{pmatrix}$ (a and b real; a, b, c distinct and nonzero) are linear homogeneous functions of a, b , and c ; yet the eigenvalues are not linear functions of a . (2) It is interesting to notice that the raw eigenvalues are homogeneous functions of the masses of degree -1 .

If the raw eigenvalues are not negative, the the eV or wave number eigenvalues are proportional to square roots of raw eigenvalues. It follows that the eV eigenvalues are homogeneous functions of the force constants f_t of degree one half; that is, if $a > 0$, then for eV eigenvalues $\nu(af) = a^{\frac{1}{2}} \nu(f)$. By Euler's theorem on homogeneous functions,

$$\nu(f) = -2J(f) f \text{ for eV eigenvalues;}$$

hence,

$$\nu(af) = -J(f) 2a^{\frac{1}{2}} f.$$

If $\Delta f = (a - 1)f = \epsilon f$, then $\nu(f + \Delta f) = -J(f) 2(1 + \epsilon)^{\frac{1}{2}} f$ and $\nu(f + \Delta f) - \nu(f) = -J(f) [2(1 + \epsilon)^{\frac{1}{2}} - 2] f$. If furthermore $\epsilon \ll 1$, then $(1 + \epsilon)^{\frac{1}{2}} \approx 1 + \frac{\epsilon}{2}$; and $\Delta \nu \approx -J \Delta f$ for J and ν in eV form. This result is in accord with the expectation that the linearity assumption is fairly accurate for either the eV or the raw form near $\Delta f = 0$.

4. Conclusions, with Suggestions for Further Investigation

There appear to be advantages to using raw eigenvalues and raw Jacobians rather than eV eigenvalues and eV Jacobians in the ADJUSTER program: On the basis of the discussion just above, it seems plausible that the region of accuracy of the linear approximation is more extensive for the raw Jacobian. For acoustic branches of the dispersion relation near $k = 0$, negative raw eigenvalues may appear during the fitting procedure. Calculations using the raw Jacobian can handle such negative raw eigenvalues and attempt to make them positive without any change in procedure. In the eV form, existence of such eigenvalues implies an "improper" situation requiring special procedures.

The linear homogeneous property of the raw eigenvalues has at least two applications. (The fact that the ν eigenvalues are homogeneous functions of degree one-half might be applied similarly, but we discuss only the raw form explicitly below.) In MACS J, we use the relation $\nu(\underline{f}) + \underline{J}(\underline{f})\underline{f} = 0$ as a check (Section II.2) on the computation of the elements of $\underline{J}$. Failure of this check may indicate that an unsuitable $\Delta \underline{f}$ has been used in the calculation of $\underline{J}$ or that the $\underline{J}$ elements have been associated with the wrong elements in $\underline{f}$ or that "crossover" error (Section II.4) has occurred.

At the beginning of any fitting procedure, human judgment and experience are probably required to obtain an initial set of force constants. Clearly there are advantages to having a reasonably good initial set of force constants in insert into an iterative procedure involving Jacobian matrices. The relation $\nu(a\underline{f}) = a \nu(\underline{f})$ might be used in an attempt to improve the force constants before starting the iterative procedure. It is not difficult to find an a (real) to minimize the least squares deviation between calculated and observed frequencies without computing a Jacobian. Suppose that we have calculated $\nu(\underline{f})$ by use of MACS J and that we wish to find an a (real) to minimize the weighted least squares deviation from the observed ν_o .

$$\begin{aligned} S &= [\nu(a\underline{f}) - \nu_o]^T \underline{W} [\nu(a\underline{f}) - \nu_o] \\ &= [a \nu(\underline{f}) - \nu_o]^T \underline{W} [a \nu(\underline{f}) - \nu_o] \\ &= \sum_j W_{jj} [a \nu_j(\underline{f}) - \nu_{oj}]^2. \end{aligned}$$

Now

$$\begin{aligned}\frac{dS}{da} &= \sum_j W_{jj} [2a \dot{v}_j^2(f) - 2 \dot{v}_j(f) \dot{v}_{oj}] \\ &= 2[a \dot{v}^T(f)W \dot{v}(f) - \dot{v}^T(f)W \dot{v}_o];\end{aligned}$$

so $\frac{dS}{da} = 0$ is satisfied by

$$a = \frac{\dot{v}^T(f)W \dot{v}_o}{\dot{v}^T(f)W \dot{v}(f)}.$$

It could be interesting to investigate the usefulness of this preliminary step in simple cases resembling those of Long and Gravenor⁽⁷⁾. It could also be interesting to see if a similar step between iterations in the more complicated crystal problems would accelerate convergence. No doubt it is of more value to investigate the following questions for various physical materials: (1) Can one set of force constants be found such that the calculated and observed dispersion curves are in satisfactory agreement? (2) Can other sets of force constants which yield comparable agreement be found?

APPENDIX B

SAMPLE INPUT FOR A CALCULATION OF A JACOBIAN MATRIX

In this sample we show part of the input for calculation of a Jacobian matrix containing eight rows and six columns. Seven passes, or seven problems, are required. Since no structure factors or transformed eigenvectors are called for, there is no structure factor data or transformation matrix. The following additional input (not shown) is needed to run the problems:

```

      1
      D0,1 (32 x 12)
      .
      .
      5
      D0,5 (32 x 12)
      D0,0 (32 x 12)
      D0,-1 (32 x 12)
      .
      .
      D0,-5 (32 x 12)
      F0,1 (32 x 32)
      .
      .
      F0,5 (32 x 32)
      F0,0 (32 x 32)
      A (8 x 12)
  
```

The $F_{0,l}$ matrices read in from cards contain six distinct nonzero entries (or force constants), 0.03, 1.32, 0.05, 0.04, 0.004, and 0.002; but it is desired to evaluate the Jacobian with these constants

changed respectively to 0.2596, 1.4793, 0.124, 0.0659, 0.004, and -0.0228. The $\Delta f_j = 0.01$ for $j = 1, 2, \dots, 6$. This sample problem is taken from work on beryllium oxide (four atoms per primitive cell). Running time on an IBM 7044 (32K, batch processing) for this sample calculation was slightly less than five minutes.