

absorptions. The inability of the approach taken in this work to obtain a good fit of the theoretical transmittance curves to the experimental transmittance curves, besides predicting the frequencies of maximum absorption, is not unexpected. In all powdered materials the particles may be expected to vary in shape about some average, with some average shape factor g . Variations in shape cause line broadening which would be extremely difficult to take into account quantitatively, and the effort probably would not be justified. Size variations may result in the occurrence of absorptions of both the surface-polarization and bulk-crystal type in the same spectrum. We have semiquantitatively accounted for the effect of shape variation on linewidth by virtue of the fact that both scattering theory and the Fröhlich relation predict the presence of absorption at frequencies all the way from the bulk-

crystal TO frequencies to the LO frequencies for extreme variations in particle shapes.

The success of our effort to interpret absorption spectra of particulate materials in terms of size and shape makes abundantly clear the danger of reporting absorption spectra of powdered polar materials without taking into account the effects of particle size and shape. On the other hand, with care such spectra may be interpreted and then used in support of other data. In some cases powder spectra may provide supplementary information when it is not readily available by other means. For example, when sufficiently large single crystals are not available for low-frequency reflection studies, or in the event that one or more frequencies are too low to measure with available equipment, the polarization-shifted frequency may be measurable.

Scattering-Matrix Method in Lattice Dynamics*

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Localized vibrational modes of plane defects in crystals can be studied by using the scattering matrix originally introduced by Saxon and Hutner for a one-dimensional crystal model. The method is generalized and illustrated for a (001) defect plane in the rocksalt structure with general first-nearest-neighbor forces.

1. INTRODUCTION

THE scattering matrix provides a simple method for studying localized states due to impurities in crystal lattices. It was first used by Saxon and Hutner in the study of localized electronic-energy states in linear lattices¹ and applied by Fukuda to localized modes of vibration in a linear chain with isotopic defects.² Hori and Asahi applied the method to study the localized modes of vibration in monatomic and diatomic linear chains, with isotopic impurities at the free end.³ They have also studied surface modes of vibration in a diatomic linear chain and a two-dimensional lattice with isotopic impurities at one edge.³

It is the purpose of this paper to show that the scattering-matrix method is more useful than has been generally recognized so far. It is simpler than the Green's-function method, in that the complete eigenvalue problem of the unperturbed system need not be

solved beforehand. It will be shown that this method can specifically be applied to $(N-1)$ -dimensional "plane" defects in N -dimensional crystal lattices; however, this domain of applicability indicates at the same time the limitation of the scattering-matrix method.

The present paper describes the generalization of the scattering-matrix method to three-dimensional crystals in the harmonic approximation. An application to the study of the localized vibrational modes in a simple cubic diatomic lattice containing a plane of isotopic impurities is made. For the special case of equal masses, this study yields the same result as obtained by Lengeler and Ludwig⁴ by a different method. The correct equation for surface modes of vibration are obtained for a (100) surface of the rocksalt structure with general nearest-neighbor forces, first considered by Takeno.⁵

A subsequent paper will deal with the application of the scattering-matrix method to the study of surface modes in a semi-infinite ionic crystal.

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¹ D. S. Saxon and R. A. Hutner, *Philips Res. Rept.* **4**, 81 (1949).

² Y. Fukuda, *J. Phys. Soc. Japan* **17**, 766 (1962).

³ J. Hori and T. Asahi, *Progr. Theoret. Phys. (Kyoto)* **31**, 49 (1964).

⁴ W. Ludwig, in *Theory of Crystal Defects*, edited by B. Gruber (Academic Press Inc., New York, 1966).

⁵ In Ref. 2, dealing with the local vibrational modes of a one-dimensional crystal, there is an error in the definition of the S matrix. Although Fukuda uses the incorrect definition throughout his derivation, he arrives at the right condition for localized modes, namely $R_{22}=0$.

2. PRINCIPLE OF SCATTERING-MATRIX METHOD

In the following, a generalized formulation of the scattering-matrix method for two-dimensional plane defects in three-dimensional crystal lattices will be presented. Consider a three-dimensional lattice with a defect formed by one or several adjacent layers of impurities, or by a free surface, or both. The perturbation extends on either side along the normal of the defect plane up to a distance corresponding to the range of the interatomic forces. The slab consisting of this perturbed region forms the scattering unit. Instead of the conventional description, a larger unit cell will be chosen, with one base vector parallel to the slab normal, and its length equal to the slab thickness. This is labeled as the 3-direction. The other two base vectors are chosen so as to provide a primitive two-dimensional unit cell in the plane of the defect. Let s be the number of atoms in the new three-dimensional unit cell. The position vector of the atoms will be denoted by $\mathbf{X}(\mathbf{m}\mu) = \mathbf{X}(\mathbf{m}) + \mathbf{X}(\mu)$, where \mathbf{m} is the vector with integer components labeling the unit cells, and $\mu = 1, \dots, s$ is the index of the atoms within a cell. Periodic boundary conditions will be assumed in the two directions in the defect plane.

A plane wave propagating through all sublattices of the crystal will be scattered by the defect slab. The scattering matrix $S_{ij}^{AB}(\mu\nu)$ relates the amplitudes $\alpha_j^B(\nu)$ of incident waves in the ν th sublattice (on both sides of the defect) to the amplitudes $\beta_i^A(\mu)$ of waves scattered into the μ th sublattice ($i, j = 1, 2, 3$; $\mu, \nu = 1, 2, \dots, s$):

$$\begin{aligned}\beta_i^1(\mu) &= \sum_{j\nu} [S_{ij}^{11}(\mu\nu)\alpha_j^1(\nu) + S_{ij}^{12}(\mu\nu)\alpha_j^2(\nu)], \\ \beta_i^2(\mu) &= \sum_{j\nu} [S_{ij}^{21}(\mu\nu)\alpha_j^1(\nu) + S_{ij}^{22}(\mu\nu)\alpha_j^2(\nu)].\end{aligned}\quad (1)$$

The indices $A, B = 1, 2$ denote the two regions on either side of the slab. Figure 1 shows the relation between incident and scattered waves. Because of the periodic boundary conditions in the two directions in the defect plane, a plane wave propagating in a direction lying in the defect plane will not be scattered. Further, for a plane wave propagating in an arbitrary direction, the component of the wave vector in the defect plane will not be affected. As far as the component k_3 in the direction of the defect plane normal is concerned, the problem, therefore, reduces essentially to a one-dimen-

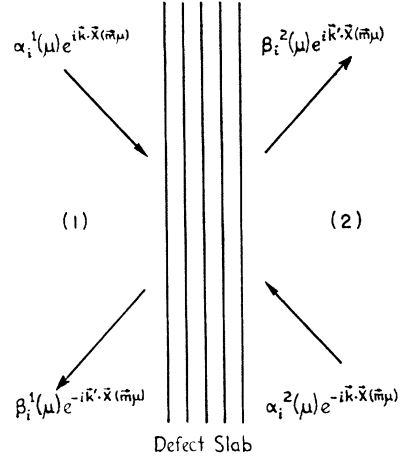


FIG. 1. Incident and scattered waves at a plane defect slab.

sional one. The directions of the wave vectors of all four kinds of waves present follow from the condition that the components parallel to the defect plane are continuous across the defect plane and are not affected by the scattering, that the normal components should represent standing waves, and that the scattering be elastic. This results in the "reflection law" shown in Fig. 1, the wave vectors satisfying the relations $k_{11} = k_{11}'$ and $k_{12} = -k_{12}'$. Although the scattering process involves only k_3 , the scattering matrix depends on all three components of \mathbf{k} , with the components k_1 and k_2 in the defect plane entering as parameters.

Localized modes are obtained from the condition that all elements of the scattering matrix be infinite¹⁻³ (resonance condition) as this corresponds to outgoing waves of finite amplitude without incoming waves. Further, the component k_3 of the wave vector in the direction of the plane normal must be imaginary, since otherwise the impurity would represent a source of outgoing waves, which is physically impossible.⁴

An even simpler condition for localized modes can be obtained in terms of a second matrix \mathbf{R} , relating the amplitudes of the two plane waves on one side to the two plane waves on the other side of the scattering unit¹:

$$\begin{aligned}\beta_i^2(\mu) &= \sum_{j\nu} [R_{ij}^{11}(\mu\nu)\alpha_j^1(\nu) + R_{ij}^{12}(\mu\nu)\beta_j^1(\nu)], \\ \alpha_i^2(\mu) &= \sum_{j\nu} [R_{ij}^{21}(\mu\nu)\alpha_j^1(\nu) + R_{ij}^{22}(\mu\nu)\beta_j^1(\nu)].\end{aligned}\quad (2)$$

It is easily shown that the two matrices are inter-related as follows:

$$\begin{pmatrix} \mathbf{S}^{11}(\mu\nu)\mathbf{S}^{12}(\mu\nu) \\ \mathbf{S}^{21}(\mu\nu)\mathbf{S}^{22}(\mu\nu) \end{pmatrix} = \begin{pmatrix} -\sum_{\lambda} [(\mathbf{R}^{22}(\lambda\mu))^{-1}\mathbf{R}^{21}(\lambda\nu)] & [\mathbf{R}^{22}(\nu\mu)]^{-1} \\ \mathbf{R}^{11}(\mu\nu) - \sum_{\lambda\sigma} \mathbf{R}^{12}(\mu\sigma)[\mathbf{R}^{22}(\lambda\sigma)]^{-1}\mathbf{R}^{21}(\lambda\nu) & \sum_{\sigma} \mathbf{R}^{12}(\mu\sigma)[\mathbf{R}^{22}(\nu\sigma)]^{-1} \end{pmatrix}.\quad (3)$$

Each of the four matrix elements \mathbf{S}^{AB} and \mathbf{R}^{AB} ($A, B = 1, 2$) is a square matrix of rank $3s$. Because of the factor $[\mathbf{R}^{22}(\lambda\mu)]^{-1}$ in Eq. (3), the condition $\mathbf{S} = \infty$ now reduces to the condition that for every i, j

$$\det(\mathbf{R}_{ij}^{22}(\mu\nu)) = 0.\quad (4)$$

TABLE I. Coupling parameters in the plane $m_3=0$ according to Lengeler and Ludwig.^a

$\Phi_{ij}(m_1, m_2, 0, \mu; m_1 \pm 1, m_2, 0, \nu)$	$\Phi_{ij}(m_1, m_2, 0, \mu; m_1, m_2 \pm 1, 0, \nu)$	$\Phi_{ij}(m_1, m_2, 0, \mu; m_1, m_2, \pm 1, \nu)$	$\Phi_{ij}(0, 0, 0, \mu; 0, 0, 0, \nu)$
(a) Impurity atoms in the plane $m_3=0$			
$-\alpha''$	0	0	$2(\alpha''+\beta''+\beta')$
0	$-\beta''$	0	0
0	0	$-\gamma''$	$2(\alpha''+\beta''+\beta')$
$-\beta''$	0	0	0
0	$-\alpha''$	0	0
0	0	$-\gamma''$	$2\alpha'+4\gamma''$
(b) Free surface at $m_3=0$			
$-\alpha$	0	$\mp\delta$	$2\alpha+3\beta$
0	$-\beta$	0	0
$\pm\delta$	0	$-\beta$	0
$-\beta$	0	0	0
0	$-\alpha$	$\mp\delta$	$2\alpha+3\beta$
0	0	$-\beta$	0
0	0	$-\alpha$	$\alpha+4\beta$

^a Reference 7.

This is the proper generalization of the condition given by Saxon and Hutner¹ for the electronic states of a one-dimensional crystal.⁵

While the **S** matrix is unitary, the **R** matrix has only the property that $\det(\mathbf{R})=1$.

As can be seen from Eq. (2), \mathbf{R}^{22} relates the amplitudes $\alpha_j^2(\mu)$ and $\beta_j^1(\nu)$, each of which has a spatial dependence of the form $e^{-ik \cdot x(m\mu)}$. The elements of \mathbf{R}^{22} are, therefore, obtained by substituting in the equations of motion displacements of the form

$$u_i(\mathbf{m}\mu) = \alpha_i(\mu)e^{i[\omega t - \mathbf{k} \cdot \mathbf{x}(m\mu)]} \quad (5)$$

and collecting coefficients of $\alpha_i(\mu)$. The determinant of these coefficients is the determinant required in Eq. (4). It should be remembered, however, that the component k_3 is purely imaginary for these modes, $k_3 = -iq_3$, where q_3 is an attenuation constant.

Since the displacements according to Eq. (5) must be eigenstates of the unperturbed lattice, they satisfy the equations of motion of the unperturbed crystal for imaginary values of k_3 . Thus, both the frequency ω and the attenuation constant q_3 can be obtained from the simultaneous solution of the condition (4), and the secular equation of the perfect crystal for imaginary value of k_3 . This is much simpler than the Green's-function method, and the solutions can be obtained graphically.

The simplicity of the scattering-matrix method arises from the translational symmetry within the defect plane, and it is, therefore, obvious that it is applicable

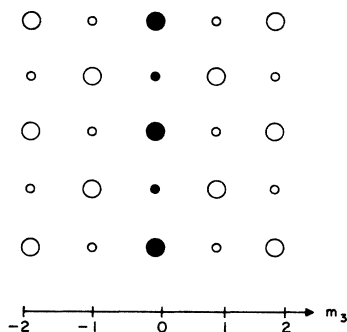


FIG. 2. (001) plane of impurities in the rocksalt structure.

to $(N-1)$ -dimensional plane defects in N -dimensional crystal lattices.

3. APPLICATIONS

Consider first the localized vibrational modes in a rocksalt-type crystal with general first nearest-neighbor forces, containing a (001) plane of impurity atoms. It will be shown how the correct equation for surface modes of vibration for the system first studied by Takeno⁶ can be obtained. It will also be shown that in the limit of equal masses the results obtained by Lengeler and Ludwig⁷ for localized modes are reproduced.

Let the plane of impurity atoms be at $m_3=0$ (Fig. 2). The site $(m_1m_2m_3)$ is occupied by the heavier mass M_1 or the lighter mass M_2 , depending on whether $(m_1+m_2+m_3)$ is even or odd, respectively. Let M_1' and M_2' denote the impurity masses. The nearest-neighbor coupling parameters are shown in Table I. The condition of rotational invariance⁷ requires that $\beta'=\beta$ and $2\delta=\beta$. The necessary modifications will be made before presenting the final results for the various examples under study.

First, we present the general equations which are applicable to a lattice containing extended defects, such as a plane of impurities.⁷ There is no translational invariance of the lattice in a direction perpendicular to the defect, although it is still present in a direction parallel to the plane of impurities. In two directions parallel to the plane, periodic boundary conditions are applied.

The time-dependent equations of motion are in the harmonic approximation

$$M_\mu \omega^2 u_i(\mathbf{m}\mu) = \sum_{\nu n j} \Phi_{ij}(\mathbf{m}\mu, \mathbf{n}\nu) u_j(\mathbf{n}\nu), \quad (6)$$

where $\Phi_{ij}(\mathbf{m}\mu, \mathbf{n}\nu)$ are the second-order coupling parameters. Because of the periodicity in the two directions parallel to the plane, one can write

$$u_i(\mathbf{m}\mu) = U_i(m_3\mu)e^{i\mathbf{k}\rho \cdot \mathbf{R}_\rho(\mathbf{m})}, \quad (7)$$

⁶ S. Takeno, Progr. Theoret. Phys. (Kyoto) **30**, 1 (1963).

⁷ B. Lengeler and W. Ludwig, Phys. Status Solidi **7**, 463 (1964).

where

$$\mathbf{R}_\rho(\mathbf{m}) = m_1 \mathbf{a}_1 + m_2 \mathbf{a}_2, \quad (8a)$$

$$\mathbf{k}_\rho = 2\pi(\mathbf{b}_1 k_1 + \mathbf{b}_2 k_2). \quad (8b)$$

\mathbf{b}_1 and \mathbf{b}_2 are the base vectors of the two-dimensional lattice reciprocal to \mathbf{a}_1 and \mathbf{a}_2 . Substituting (7) into the equations of motion gives

$$M_\mu \omega^2 U_i(m_3 \mu) = \sum_{n_3 \nu} \psi_{ij}^{k\rho}(m_3 \mu, n_3 \nu) U_j(n_3 \nu), \quad (9)$$

where

$$\psi_{ij}^{k\rho}(m_3 \mu, n_3 \nu) = \sum_{n_1 n_2} \Phi_{ij}(00m_3 \mu, 00n_3 \nu) e^{i\mathbf{k}_\rho \cdot \mathbf{R}_\rho(n)}. \quad (10)$$

The coefficients Ψ represent the force constants between sublattice planes⁸ in the presence of the wave of wave vector \mathbf{k}_ρ . Accordingly, the equations of motion for the atoms in the layers $m_3 = -1, 0, 1$ are

$$\begin{aligned} M_1 \omega^2 U_i(\bar{1}1) &= \sum_j [\psi_{ij}^{k\rho}(\bar{1}1, \bar{1}1) U_j(\bar{1}1) + \psi_{ij}^{k\rho}(\bar{1}1, \bar{1}2) U_j(\bar{1}2) \\ &\quad + \hat{\psi}_{ij}^{k\rho}(\bar{1}1, 02) U_j(02) + \psi_{ij}^{k\rho}(\bar{1}1, \bar{2}2) U_j(\bar{2}2)], \\ M_2 \omega^2 U_i(\bar{1}2) &= \sum_j [\psi_{ij}^{k\rho}(\bar{1}2, \bar{1}2) U_j(\bar{1}2) + \psi_{ij}^{k\rho}(\bar{1}2, \bar{1}1) U_j(\bar{1}1) \\ &\quad + \hat{\psi}_{ij}^{k\rho}(\bar{1}2, 01) U_j(01) + \psi_{ij}^{k\rho}(\bar{1}2, \bar{2}1) U_j(\bar{2}1)], \\ M_1' \omega^2 U_i(01) &= \sum_j [\hat{\psi}_{ij}^{k\rho}(01, 01) U_j(01) + \hat{\psi}_{ij}^{k\rho}(01, 02) U_j(02) \\ &\quad + \hat{\psi}_{ij}^{k\rho}(01, 12) U_j(12) + \hat{\psi}_{ij}^{k\rho}(01, \bar{1}2) U_j(\bar{1}2)], \\ M_2' \omega^2 U_i(02) &= \sum_j [\hat{\psi}_{ij}^{k\rho}(02, 02) U_j(02) + \hat{\psi}_{ij}^{k\rho}(02, 01) U_j(01) \\ &\quad + \hat{\psi}_{ij}^{k\rho}(02, 11) U_j(11) + \hat{\psi}_{ij}^{k\rho}(02, \bar{1}1) U_j(\bar{1}1)], \\ M_1 \omega^2 U_i(11) &= \sum_j [\psi_{ij}^{k\rho}(11, 11) U_j(11) + \psi_{ij}^{k\rho}(11, 12) U_j(12) \\ &\quad + \hat{\psi}_{ij}^{k\rho}(11, 02) U_j(02) + \psi_{ij}^{k\rho}(11, 22) U_j(22)], \\ M_2 \omega^2 U_i(12) &= \sum_j [\psi_{ij}^{k\rho}(12, 12) U_j(12) + \psi_{ij}^{k\rho}(12, 11) U_j(11) \\ &\quad + \hat{\psi}_{ij}^{k\rho}(12, 01) U_j(01) + \psi_{ij}^{k\rho}(12, 21) U_j(21)]. \end{aligned} \quad (11)$$

⁸ The two-dimensional Fourier transform based on Eq. (7) leading to the coefficients ψ in Eq. (10) has been used before by Feuchtwang and is the basis of his lattice theoretical treatment of semi-infinite crystals [T. E. Feuchtwang, Phys. Rev. **155**, 715 (1967); **155**, 731 (1967)].

The symbol $\hat{}$ indicates that the corresponding coefficients are different from those in the perfect lattice. Writing the solutions in the form

$$\begin{aligned} U_i(m_3 \mu) &= \alpha_i^1(\mu) e^{ik_3 m_3 a} + \beta_i^1(\mu) e^{-ik_3 m_3 a}, \quad m_3 \leq 0 \\ U_i(m_3 \mu) &= \beta_i^2(\mu) e^{ik_3 m_3 a} + \alpha_i^2(\mu) e^{-ik_3 m_3 a}, \quad m_3 \geq 0 \end{aligned} \quad (12)$$

the continuity condition at $m_3 = 0$ requires

$$\alpha_i^1(\mu) + \beta_i^1(\mu) = \alpha_i^2(\mu) + \beta_i^2(\mu), \quad \mu = 1, 2. \quad (13)$$

After substituting from (12) into the equations of motion (11) the condition $\det(\mathbf{R}_{ij}^{22}(\mu\nu)) = 0$ for localized modes becomes

$$\begin{vmatrix} A_{11} & A_{12} \\ A_{21} & A_{22} \end{vmatrix} = 0, \quad (14)$$

with

$$\begin{aligned} A_{11} &= (M_1 e^{i2k_3 a} - M_1') \omega^2 \delta_{ij} - \psi_{ij}^{k\rho}(\bar{1}1, \bar{1}1) e^{i2k_3 a} \\ &\quad + \hat{\psi}_{ij}^{k\rho}(01, 01), \\ A_{12} &= -\psi_{ij}^{k\rho}(\bar{1}1, \bar{1}2) e^{i2k_3 a} - \psi_{ij}^{k\rho}(\bar{1}2, \bar{2}1) e^{i3k_3 a} \\ &\quad + \hat{\psi}_{ij}^{k\rho}(02, 01) + \hat{\psi}_{ij}^{k\rho}(02, \bar{1}1) e^{ik_3 a}, \\ A_{21} &= -\psi_{ij}^{k\rho}(\bar{1}1, \bar{1}2) e^{i2k_3 a} - \psi_{ij}^{k\rho}(\bar{1}1, \bar{2}2) e^{i3k_3 a} \\ &\quad + \psi_{ij}^{k\rho}(01, 02) + \hat{\psi}_{ij}^{k\rho}(01, \bar{1}2) e^{ik_3 a}, \\ A_{22} &= (M_2 e^{i2k_3 a} - M_2') \omega^2 \delta_{ij} - \psi_{ij}^{k\rho}(\bar{1}2, \bar{1}2) e^{i2k_3 a} \\ &\quad + \hat{\psi}_{ij}^{k\rho}(02, 02). \end{aligned} \quad (15)$$

If the lattice is monatomic, condition (14) reduces to

$$(M e^{i2k_3 a} - M') \omega^2 \delta_{ij} - \psi_{ij}^{k\rho}(\bar{1}, \bar{1}) e^{i2k_3 a} - \psi_{ij}^{k\rho}(\bar{1}, \bar{2}) e^{i3k_3 a} + \hat{\psi}_{ij}^{k\rho}(0, 0) + \hat{\psi}_{ij}^{k\rho}(0, \bar{1}) e^{ik_3 a} = 0. \quad (16)$$

This can be shown to reproduce the results obtained by Lengeler and Ludwig in their analysis of localized modes in a simple cubic lattice with extended defects.⁷ For example, for $i = j = 1$, Eq. (16) is equivalent to

$$(M e^{i2k_3 a} - M') \omega^2 - \psi_{11}^{k\rho}(\bar{1}, \bar{1}) e^{i2k_3 a} - \psi_{11}^{k\rho}(\bar{1}, \bar{2}) e^{i3k_3 a} + \psi_{11}^{k\rho}(0, 0) + \psi_{11}^{k\rho}(0, \bar{1}) e^{ik_3 a} = 0. \quad (17)$$

The sum of the first three terms in Eq. (17) is equal to $\psi_{11}^{k\rho}(01) e^{ik_3 a} - M' \omega^2$. Hence (17) reduces to

$$[\psi_{11}^{k\rho}(0, 1) + \hat{\psi}_{11}^{k\rho}(0, \bar{1})] e^{ik_3 a} + \hat{\psi}_{11}^{k\rho}(0, 0) - M' \omega^2 = 0. \quad (18)$$

For the surface modes of this model, Eq. (18) is equivalent to

$$\begin{aligned} -M \omega^2 + (2\alpha + 3\beta) - 2\alpha \cos k_1 a \\ - 2\beta \cos k_2 a - \beta e^{ik_3 a} = 0. \end{aligned} \quad (19)$$

This is the same as Eq. (2.7) given by Lengeler and Ludwig.⁷ However, this is inconsistent with the condition of rotational invariance, as has been pointed out in Ref. 7. But Eq. (16) is quite correct, and will involve nondiagonal terms. The above digression had as its purpose only the reproduction of results formerly obtained by Lengeler and Ludwig.⁷ The condition for surface modes in a semi-infinite diatomic simple cubic crystal is obtained by setting $M_1' = M_1$, $M_2' = M_2$

and noting that the surface is free [$\hat{\psi}_{ij}^{k\rho}(01, \bar{1}2) = \hat{\psi}_{ij}^{k\rho}(02, \bar{1}1) = 0$]. The condition $\det(\mathbf{R}_{ij}{}^{22}(\nu\mu)) = 0$ can again be written in the form (14), but with

$$\begin{aligned} A_{11} &= M_1 \omega^2 (e^{i2k_3 a} - 1) \delta_{ij} - \psi_{ij}^{k\rho}(11, 11) e^{i2k_3 a} + \hat{\psi}_{ij}^{k\rho}(01, 01), \\ A_{12} &= -\psi_{ij}^{k\rho}(11, 12) e^{i2k_3 a} - \psi_{ij}^{k\rho}(12, 01) e^{i3k_3 a} + \hat{\psi}_{ij}^{k\rho}(02, 01), \\ A_{21} &= -\psi_{ij}^{k\rho}(11, 12) e^{i2k_3 a} - \psi_{ij}^{k\rho}(11, 02) e^{i3k_3 a} + \psi_{ij}^{k\rho}(01, 02), \\ A_{22} &= M_2 \omega^2 (e^{i2k_3 a} - 1) \delta_{ij} - \psi_{ij}^{k\rho}(12, 12) e^{i2k_3 a} + \hat{\psi}_{ij}^{k\rho}(02, 02). \end{aligned} \quad (20)$$

This will give the correct solution to the problem first considered by Takeno.⁶ As indicated earlier, a simultaneous solution of Eq. (14) [with the matrix elements given by Eq. (20)] and the secular equation of the infinite crystal,

$$\det \begin{vmatrix} \psi_{ij}^{k\rho}(11, 11) - M_1 \omega^2 \delta_{ij} & \psi_{ij}^{k\rho}(11, 12) + 2\psi_{ij}^{k\rho}(11, 02) \cosh q_3 a \\ \psi_{ij}^{k\rho}(12, 11) + 2\psi_{ij}^{k\rho}(12, 01) \cosh q_3 a & \psi_{ij}^{k\rho}(12, 12) - M_2 \omega^2 \delta_{ij} \end{vmatrix} = 0 \quad (21)$$

would yield both the frequency and the damping constant q_3 (defined by $k_3 = iq_3$) of the surface modes of vibration. The solution can be obtained graphically by plotting the two functions $\omega(k_3)$ defined by Eqs. (14) [in connection with Eq. (20)] and (21) along the imaginary k_3 axis.

The model of the rocksalt structure considered here corresponding to general first-nearest-neighbor forces is not applicable to real materials, since it follows that the relation $c_{12} = -c_{44}$ must hold for the elastic constants. As c_{44} must be positive in view of the stability criterion this implies that c_{12} is negative. No cubic material with this property is known, however.

For UC, the Cauchy relation $c_{12} = c_{44}$ is approximately fulfilled,^{9,10} suggesting that a model with central forces between first- and second-nearest-neighbor interaction might be adequate. For ZrC and TiC the Cauchy relation is not fulfilled¹⁰ and both c_{12} and c_{44} are positive, so that the simplest adequate model would consist of general first-nearest-neighbor and central second-nearest-neighbor interaction. For ionic crystals, such

as the alkali halides, the model becomes, of course, inadequate because of the long-range Coulomb forces. The application of the scattering-matrix method to ionic crystals demands a more elaborate treatment and will be made in a subsequent paper. The simple first-nearest-neighbor model considered here was chosen only to illustrate the simplicity of the scattering-matrix approach and to reproduce and correct the results of Lengeler and Ludwig⁷ and Takeno,⁶ respectively.

4. SUMMARY AND CONCLUSIONS

The scattering-matrix method has been generalized to extended plane defects in polyatomic three-dimensional crystals, and has been applied to the study of localized modes in the rocksalt structure with general nearest-neighbor forces containing a plane of impurities. The results agree with those obtained by Lengeler and Ludwig⁷ in the limit of equal masses for the atoms. In addition, the correct solution for the surface modes of vibration for a free (001) surface of the rocksalt structure is obtained. The approach based on the scattering matrix turns out to be much simpler than the Green's-function method, but is limited to the study of localized modes only.

⁹ L. J. Graham, H. Nadler, and R. Chang, *J. Appl. Phys.* **34**, 1572 (1963).

¹⁰ R. Chang and L. J. Graham, *J. Appl. Phys.* **37**, 3778 (1966).