Elastic Spectra of Two-Dimensional Disordered Lattices*

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The elastic vibrational spectra of perturbed square-lattice systems with nearest-neighbor central and noncentral interactions have been derived. The unperturbed system consists of masses m on the lattice points and interacting with force constants α, which determines the resistance to compression, and β, which determines the resistance to shear along the direction [10]. In one case, the perturbations are Nq randomly positioned isotopic impurities of mass m', where N is the number of lattice sites. It is shown that the elastic spectrum for this and all other isotopic impurity systems is completely determined by the average mass, $\bar{m}=(1-q)m+qm'$. In the other case, corresponding to certain order-disorder situations, the constants describing the interactions between $2Nq$ randomly positioned pairs of nearest neighbors are replaced by α' and β'. To first order in q, the resulting elastic modes are then completely determined by an average α,

$$\bar{\alpha}=\alpha\left[1+\frac{q(\alpha'-\alpha)}{\alpha+(\alpha'-\alpha)(2/\pi)\tan^{-1}(\alpha/\beta)^{1/2}}\right],$$

and a similar average, $\bar{\beta}$, which is obtained from the previous expression via the interchange of α and β. The appearance of β in the expression for $\bar{\alpha}$ implies that the virtual-crystal approximation fails. It is shown, however, that two different forms of the virtual-crystal approximation place bounds on the $\bar{\alpha}$ and $\bar{\beta}$, in accordance with a general theorem of Paul. A physical interpretation of the results is also presented.

I. INTRODUCTION

IN a recent paper,[1] (denoted, hereafter, as P-B) the authors obtained an expression for the vibrational frequency spectrum of a one-dimensional chain of atoms with the same mass and nearest-neighbor interactions. Randomly positioned springs were replaced by springs which had a different force constant, and the spectral distribution function was calculated to first order in the concentration of the impurity springs. The mathematical techniques used were a modification of those used by Langer[2] to solve the identical problem for the same unperturbed system in which the impurities were atoms with a different mass. While the motivation for the previous paper was an attempt to obtain more information about the effect of disordering on the vibrational spectrum of binary alloys such as CuZn and CoFe, in which the masses of the two constituents are almost identical, the one-dimensional nature of the problem limited the value of the solution. Nevertheless, the mathematical techniques developed are quite helpful, and will be used here.

In this paper, two related problems are treated. In both cases, the unperturbed lattice is a square-lattice array of atoms with same mass, and with nearest-neighbor central and noncentral forces. In one case, the two force constants describing the interaction between randomly positioned nearest-neighbor pairs of atoms are replaced by a different pair of force constants. In the other, randomly positioned atoms are replaced by atoms of a different mass.

For convenience, we call the problems the impurity-spring and impurity-mass problems. In both cases, the elastic constants are obtained as a function of concentration q, to first order in q.

The most striking feature of the results is that while the virtual-crystal approximation is found to be valid for the impurity-mass problem, it is valid only for special cases in the impurity-spring problem. The explanation and implications of this result are discussed.

II. THE UNPERTURBED CASE

In the unperturbed case, we are considering the vibrational spectrum of a square-lattice array of atoms of mass m. In order to maintain both stability and only nearest-neighbor interactions, these interactions must be noncentral. We label the atom at the origin by (0,0) and its four nearest neighbors by (1,0), (0,1), ($\bar{1}$,0), and (0,$\bar{1}$). The equations of motion of the lth atom are then

$$m\ddot{x}_{l,\alpha}+\sum_{l',\alpha'}C_{\alpha\alpha'}(l,l')x_{l',\alpha'}=0. \tag{1}$$

Here, the l' summation runs over the atom l and its four nearest neighbors, and the α' summation runs over the two orthogonal coordinates.

The force-constant matrices have the forms

$$C(1,0)=C(\bar{1},0)=-\begin{pmatrix}\alpha & 0\\ 0 & \beta\end{pmatrix},$$

$$C(0,1)=C(0,\bar{1})=-\begin{pmatrix}\beta & 0\\ 0 & \alpha\end{pmatrix}, \tag{2}$$

$$C(0,0)=2(\alpha+\beta)\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}.$$

For central force interactions, $\beta=0$ and the lattice

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[1] H.-C. Poon and A. Bienenstock (to be published).
[2] J. S. Langer, J. Math. Phys. 2, 584 (1961).

shows zero-frequency shear modes for propagation along certain principal directions.

Expressing the displacements in terms of normal modes

$$x_{l,\gamma}=\sum_{\mathbf{k},j} Q_{j,\gamma}(\mathbf{k})\,\exp i(\mathbf{k}\cdot\mathbf{R}_l-\omega_{j,\mathbf{k}}t)\,, \tag{3}$$

where $j(=1$ or $2)$ labels the two orthogonally polarized modes and γ labels a displacement component, we obtain the normal-mode solutions

$$\begin{aligned}\omega_{1,\mathbf{k}}{}^2&=\frac{4}{m}\left[\alpha\sin^2\frac{k_xa}{2}+\beta\sin^2\frac{k_ya}{2}\right],\\ \omega_{2,\mathbf{k}}{}^2&=\frac{4}{m}\left[\beta\sin^2\frac{k_xa}{2}+\alpha\sin^2\frac{k_ya}{2}\right],\end{aligned} \tag{4}$$

with eigenvectors

$$Q_1(\mathbf{k})=\begin{pmatrix}1\\0\end{pmatrix},\quad Q_2(\mathbf{k})=\begin{pmatrix}0\\1\end{pmatrix}. \tag{5}$$

Equation (5) implies that for any propagation vector $\mathbf{k}$ the two normal modes, 1 and 2, have polarization vectors along $\hat{x}$ and $\hat{y}$, respectively. Thus, one of the two labels, j and γ, of $Q_{j,\gamma}$ in Eq. (3) is redundant, and will not be used hereafter.

III. THE IMPURITY-SPRING PROBLEM

A. Formulation

In this problem, the force constants α and β, associated with $2N$ randomly positioned pairs of atoms, are replaced by force constants α' and β'. To simplify the equations of motion, the four nearest neighbors of an atom which are separated from it by vectors (1,0), $(\bar{1},0)$, (0,1), and $(0,\bar{1})$ are denoted by $(l+1)$, $(l+2)$, $(l+3)$, and $(l+4)$, respectively. The equations of motion can then be written in the general form

$$\begin{aligned}-\omega^2m\begin{pmatrix}x_{l,1}\\x_{l,2}\end{pmatrix}&+\begin{pmatrix}\alpha&0\\0&\beta\end{pmatrix}\begin{pmatrix}2x_{l,1}-x_{l+1,1}-x_{l+2,1}\\2x_{l,2}-x_{l+1,2}-x_{l+2,2}\end{pmatrix}\\&+\begin{pmatrix}\beta&0\\0&\alpha\end{pmatrix}\begin{pmatrix}2x_{l,1}-x_{l+3,1}-x_{l+4,1}\\2x_{l,2}-x_{l+3,2}-x_{l+4,2}\end{pmatrix}\\&=\begin{pmatrix}\alpha-\alpha_{l,l+1}&0\\0&\beta-\beta_{l,l+1}\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+1,1}\\x_{l,2}-x_{l+1,2}\end{pmatrix}\\&+\begin{pmatrix}\alpha-\alpha_{l,l+2}&0\\0&\beta-\beta_{l,l+2}\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+2,1}\\x_{l,2}-x_{l+2,2}\end{pmatrix}\\&+\begin{pmatrix}\beta-\beta_{l,l+3}&0\\0&\alpha-\alpha_{l,l+3}\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+3,1}\\x_{l,2}-x_{l+3,2}\end{pmatrix}\\&+\begin{pmatrix}\beta-\beta_{l,l+4}&0\\0&\alpha-\alpha_{l,l+4}\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+4,1}\\x_{l,2}-x_{l+4,2}\end{pmatrix}.\end{aligned} \tag{6}$$

Equation (6) can be rewritten in terms of the normal coordinates of the transformation

$$x_{l,\gamma}=\textstyle\sum_{\mathbf{k}} Q_\gamma(\mathbf{k})\,\exp i\mathbf{k}\cdot\mathbf{R}_l\,. \tag{7}$$

Since all the force-constant matrices are diagonal, the x and y components of the displacement are independent. Thus, equations of motion can be written simply for each direction, using Eqs. (6) and (7). In addition, the two directions are indistinguishable. Thus, it is sufficient to study one direction of polarization over a complete quadrant of the propagation plane. For the x components of the displacements, we have

$$\begin{aligned}-\omega^2mx_{l,1}&+\alpha(2x_{l,1}-x_{l+1,1}-x_{l+2,1})\\&\qquad+\beta(2x_{l,1}-x_{l+3,1}-x_{l+4,1})\\&=(\alpha-\alpha_{l,l+1})(x_{l,1}-x_{l+1,1})+(\alpha-\alpha_{l,l+2})(x_{l,1}-x_{l+2,1})\\&\quad+(\beta-\beta_{l,l+3})(x_{l,1}-x_{l+3,1})+(\beta-\beta_{l,l+4})(x_{l,1}-x_{l+4,1})\,.\end{aligned} \tag{8}$$

By substituting Eq. (7) into Eq. (8), and using Eq. (4) to simplify the resulting expression, one obtains

$$\begin{aligned}-m\omega^2\textstyle\sum_{\mathbf{k}} Q_1(\mathbf{k})\,\exp i\mathbf{k}\cdot\mathbf{R}_l&+m\textstyle\sum_{\mathbf{k}}\omega_{1,\mathbf{k}}{}^2Q_1(\mathbf{k})\,\exp i\mathbf{k}\cdot\mathbf{R}_l\\&=(\alpha-\alpha_{l,l+1})\textstyle\sum_{\mathbf{k}} Q_1(\mathbf{k})(1-\exp ik_xa)\,\exp i\mathbf{k}\cdot\mathbf{R}_l\\&\quad+(\alpha-\alpha_{l,l+2})\textstyle\sum_{\mathbf{k}} Q_1(\mathbf{k})[1-\exp(-ik_xa)]\,\exp i\mathbf{k}\cdot\mathbf{R}_l\\&\quad+(\beta-\beta_{l,l+3})\textstyle\sum_{\mathbf{k}} Q_1(\mathbf{k})(1-\exp ik_ya)\,\exp i\mathbf{k}\cdot\mathbf{R}_l\\&\quad+(\beta-\beta_{l,l+4})\textstyle\sum_{\mathbf{k}} Q_1(\mathbf{k})[1-\exp(-ik_ya)]\,\exp i\mathbf{k}\cdot\mathbf{R}_l\,.\end{aligned} \tag{9}$$

After multiplying both sides of Eq. (9) by

$$\exp(-i\mathbf{k}'\cdot\mathbf{R}_l)$$

and summing over l, one obtains

$$\begin{aligned}&mN(\omega_{1,\mathbf{k}'}{}^2-\omega^2)Q_1(\mathbf{k}')\\&\quad=\sum_{l,\mathbf{k}} Q_1(\mathbf{k})\,\exp i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_l\\&\qquad\times\{(\alpha-\alpha_{l,l+1})(1-\exp ik_xa)+(\alpha-\alpha_{l,l+2})\\&\qquad\times[1-\exp(-ik_xa)]+(\beta-\beta_{l,l+3})(1-\exp ik_ya)\\&\qquad\qquad+(\beta-\beta_{l,l+4})[1-\exp(-ik_ya)]\}\,.\end{aligned} \tag{10}$$

After interchanging $\mathbf{k}$ and $\mathbf{k}'$ and performing some trivial mathematical manipulations, Eq. (10) can be written in the form

$$(\omega_{1,\mathbf{k}}{}^2-\omega^2)Q_1(\mathbf{k})=-\textstyle\sum_{\mathbf{k}}\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}Q_1(\mathbf{k}')\,, \tag{11}$$

where

$$\begin{aligned}\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}=\frac{(-2i)}{Nm}\sum_l\{&\exp[-i(\mathbf{k}-\mathbf{k}')]\cdot\mathbf{R}_l\\&\times[(\alpha_{l,l+1}-\alpha)\sin(k_x'a/2)\exp(ik_x'a/2)\\&-(\alpha_{l,l+2}-\alpha)\sin(k_x'a/2)\exp(-ik_x'a/2)\\&+(\beta_{l,l+3}-\beta)\sin(k_y'a/2)\exp(ik_y'a/2)\\&-(\beta_{l,l+4}-\beta)\sin(k_y'a/2)\exp(-ik_y'a/2)]\}\,.\end{aligned} \tag{12}$$

The superscript (1) of $\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}$ is used to indicate that polarizations along $\hat{x}$ only are included.

Equation (11) is identical in form to Langer's[2] equation (4) and Eq. (4) of (P-B). The three problems differ only in the form of the $\Phi_{\mathbf{k},\mathbf{k}'}$. We shall follow the approach described in (P-B) because it is closer to the methods used here, although the basic technique was originally presented, and is more completely described, in Langer's paper.

For a particular configuration of springs, we can write

$$[D^{(1)}(\omega^2)]_{\mathbf{k},\mathbf{k}'}{}^{-1}=(\omega_{\mathbf{k}}{}^2-\omega^2)\delta_{\mathbf{k},\mathbf{k}'}+\Phi_{\mathbf{k},\mathbf{k}'} \tag{13}$$

such that Eq. (11) becomes

$$\sum_{\mathbf{k}'}[D^{(1)}(\omega^2)]_{\mathbf{k},\mathbf{k}'}{}^{-1}Q_1(\mathbf{k}')=0. \tag{14}$$

The spectral distribution function for this configuration, defined by

$$g(\omega)=\lim_{N\to\infty}\frac{2\omega}{N}\sum_n\delta(\Omega_n{}^2-\omega^2), \tag{15}$$

where the Ω_n are the exact eigenfrequencies for the particular configuration of springs, is given by[2]

$$g(\omega)=\frac{4\omega}{\pi}\lim_{\substack{N\to\infty\\ \epsilon\to 0}}\frac{1}{N}\operatorname{Im}\operatorname{Tr}D^{(1)}(\omega^2+i\epsilon). \tag{16}$$

In this expression, use of symmetry has made it unnecessary to include $D^{(2)}(\omega^2+i\epsilon)$.

To obtain the spectral distribution function for the random distribution of springs, $\bar{g}(\omega)$, we must average the distribution function over all configurations which are consistent with a fixed value of q. As shown by Langer, it is sufficient to take the configuration average, $\bar{D}^{(1)}(\omega^2+i\epsilon)$, of $D^{(1)}(\omega^2+i\epsilon)$. As in the one-dimensional problem, $\bar{D}^{(1)}(\omega^2+i\epsilon)$ is diagonal in $\mathbf{k}$, so that

$$\bar{g}(\omega)=\frac{4\omega}{\pi}\lim_{\substack{N\to\infty\\ \epsilon\to 0}}\frac{1}{N}\operatorname{Im}\sum_{\mathbf{k}}\bar{D}_{\mathbf{k}}{}^{(1)}(\omega^2+i\epsilon). \tag{17}$$

B. Configuration Averages

As in (P-B), $D_{\mathbf{k},\mathbf{k}'}{}^{(1)}(\omega^2)$ may be iterated to yield

$$D_{\mathbf{k},\mathbf{k}'}{}^{(1)}(\omega^2)=\frac{1}{\omega_{\mathbf{k}}{}^2-\omega^2}\Bigg\{\delta_{\mathbf{k},\mathbf{k}'}-\frac{\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}}{\omega_{\mathbf{k}'}{}^2-\omega^2}+\sum_{\mathbf{k}_1}\frac{\Phi_{\mathbf{k},\mathbf{k}_1}{}^{(1)}\Phi_{\mathbf{k}_1,\mathbf{k}'}{}^{(1)}}{(\omega_{\mathbf{k}_1}{}^2-\omega^2)(\omega_{\mathbf{k}'}{}^2-\omega^2)}+\cdots\Bigg\}. \tag{18}$$

The average over configurations is performed at this point. Since the $\omega_{\mathbf{k}}$ are properties of the unperturbed lattice, it is only necessary to obtain configurational averages of the products $\Phi_{\mathbf{k},\mathbf{k}_1}{}^{(1)}$, $\Phi_{\mathbf{k}_1,\mathbf{k}_2}{}^{(1)}$, $\Phi_{\mathbf{k}_2,\mathbf{k}_3}{}^{(1)},\cdots$, $\Phi_{\mathbf{k}_n,\mathbf{k}'}{}^{(1)}$. We proceed to obtain these averages. The first, $\langle\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}\rangle$ is easily obtained. We need

$$\langle\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}\rangle=\frac{(-2i)}{Nm}\Big\langle\sum_l\exp[-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_l]\{\sin(k_x'a/2)[(\alpha_{l,l+1}-\alpha)\exp(ik_x'a/2)-(\alpha_{l,l+2}-\alpha)\exp(-ik_x'a/2)]$$
$$+\sin(k_y'a/2)[(\beta_{l,l+3}-\beta)\exp(ik_y'a/2)-(\beta_{l,l+4}-\beta)\exp(-ik_y'a/2)]\}\Big\rangle. \tag{19}$$

This expression is evaluated by keeping l fixed while summing over configurations. No contribution to the terms linear in the α's is obtained from any configuration in which $\alpha_{l,l+1}$ or $\alpha_{l,l+2}$ is equal to α. The analogous situation holds for the terms linear in β. Denoting

$$\Delta\alpha=\alpha'-\alpha,$$

and

$$\Delta\beta=\beta'-\beta, \tag{20}$$

the configurational average is

$$\langle\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}\rangle=\frac{(-2i)}{Nm}\sum_l\exp i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_l\{q[2i\Delta\alpha\sin^2(k_x'a/2)+2i\Delta\beta\sin^2(k_y'a/2)]\}. \tag{21}$$

Performance of the summation over l yields

$$\langle\Phi_{\mathbf{k},\mathbf{k}'}{}^{(1)}\rangle=(4q/m)[\Delta\alpha\sin^2(k_x'a/2)+\Delta\beta\sin^2(k_y'a/2)]\delta_{\mathbf{k},\mathbf{k}'}. \tag{22}$$

Next, consider the second-order term,

$$\langle\Phi_{\mathbf{k},\mathbf{k}_1}{}^{(1)}\Phi_{\mathbf{k}_1,\mathbf{k}'}{}^{(1)}\rangle=\frac{(-2i)^2}{N^2m^2}\sum_{l_1,l_2}\exp i(\mathbf{k}_1-\mathbf{k})\cdot\mathbf{R}_{l_1}\exp i(\mathbf{k}'-\mathbf{k}_1)\cdot\mathbf{R}_{l_2}$$
$$\times\langle\{\sin(k_{1x}a/2)[(\alpha_{l_1,l_1+1}-\alpha)\exp(ik_{1x}a/2)-(\alpha_{l_1,l_1+2}-\alpha)\exp(-ik_{1x}a/2)]$$
$$+\sin(k_{1y}a/2)[(\beta_{l_1,l_1+3}-\beta)\exp(ik_{1y}a/2)-(\beta_{l_1,l_1+4}-\beta)\exp(-ik_{1y}a/2)]\}$$
$$\times\{\sin(k_x'a/2)[(\alpha_{l_2,l_2+1}-\alpha)\exp(ik_x'a/2)-(\alpha_{l_2,l_2+2}-\alpha)\exp(-ik_x'a/2)]$$
$$+\sin(k_y'a/2)[(\beta_{l_2,l_2+3}-\beta)\exp(ik_y'a/2)-(\beta_{l_2,l_2+4}-\beta)\exp(-ik_y'a/2)]\}\rangle. \tag{23}$$

As before, only terms in which the $\alpha_{i,j}$ and $\beta_{i,j}$ differ from α and β, respectively, contribute to the right-hand side of Eq. (23). In addition, as in (P-B), only the product terms in which the α_{i_1,j_1} and β_{i_1,j_1} represent the same springs as the α_{i_2,j_2} and β_{i_2,j_2}, respectively, contribute to first order in q. It should be noted that all the α terms correspond to springs which are parallel to the direction (1), while all β terms correspond to springs which are perpendicular to the direction (1). Thus, all product terms which involve both α's and β's must vanish, to first order in q, for a product of Φ's of any order. As a result of these considerations, we may write

$$\langle \Phi_{\mathbf{k},\mathbf{k}_1}{}^{(1)} \Phi_{\mathbf{k}_1,\mathbf{k}'}{}^{(1)} \rangle = \frac{(-2i)^2}{N^2 m^2} \sum_{l_1,l_2} \exp[i(\mathbf{k}_1-\mathbf{k})\cdot \mathbf{R}_{l_1}] \exp[i(\mathbf{k}'-\mathbf{k}_1)\cdot \mathbf{R}_{l_2}]$$
$$\times \{ \langle [(\alpha_{l_1,l_1+1}-\alpha)\exp(ik_{1x}a/2) - (\alpha_{l_1,l_1+2}-\alpha)\exp(-ik_{1x}a/2)] \sin(k_{1x}a/2)$$
$$\times [(\alpha_{l_2,l_2+1}-\alpha)\exp(ik_x'a/2) - (\alpha_{l_2,l_2+2}-\alpha)\exp(-ik_x'a/2)] \sin(k_x'a/2) \rangle$$
$$+ \langle [(\beta_{l_1,l_1+3}-\beta)\exp(ik_{1y}a/2) - (\beta_{l_1,l_1+4}-\beta)\exp(-ik_{1y}a/2)] \sin(k_{1y}a/2)$$
$$\times [(\beta_{l_2,l_2+3}-\beta)\exp(ik_y'a/2) - (\beta_{l_2,l_2+4}-\beta)\exp(-ik_y'a/2)] \sin(k_y'a/2) \rangle \}. \quad (24)$$

The important feature of Eq. (24) which makes this problem soluble is that the two-dimensional average of Eq. (23) has been split into two one-dimensional averages which are mathematically identical to those which have been evaluated in (P-B). This splitting occurs, for terms which are first order in q, for all order products of the Φ's. As a result, using Eq. (30) of (P-B), we obtain the general expression

$$\langle \Phi_{\mathbf{k},\mathbf{k}_1}{}^{(1)} \Phi_{\mathbf{k}_1,\mathbf{k}_2}{}^{(1)} \cdots \Phi_{\mathbf{k}_{n-1},\mathbf{k}_n}{}^{(1)} \rangle = \frac{q\delta_{\mathbf{k},\mathbf{k}_n}}{N^{n-1}} \left(\frac{4}{m}\right)^n \Big[(\Delta\alpha)^n \sin^2\left(\frac{k_{1x}a}{2}\right) \sin^2\left(\frac{k_{2x}a}{2}\right) \cdots \sin^2\left(\frac{k_{nx}a}{2}\right)$$
$$+ (\Delta\beta)^n \sin^2\left(\frac{k_{1y}a}{2}\right) \sin^2\left(\frac{k_{2y}a}{2}\right) \cdots \sin^2\left(\frac{k_{ny}a}{2}\right) \Big]. \quad (25)$$

C. Calculation of the Elastic Spectrum

The form of Eq. (25) indicates, as expected, that $\bar{D}^{(1)}$ is diagonal in $\mathbf{k}$. As a result, we may follow Langer and write the diagonal elements of $\bar{D}^{(1)}$ as $D_{\mathbf{k}}{}^{(1)}$ where

$$D_{\mathbf{k}}{}^{(1)}(\omega^2) = \frac{1}{\omega_{\mathbf{k}}{}^2 - \omega^2 + G_{\mathbf{k}}{}^{(1)}(\omega^2)}. \quad (26)$$

The configuration average of Eq. (18) may then be rewritten, using Eqs. (25) and (26) as

$$G_{\mathbf{k}}{}^{(1)} = q \sum_n \frac{(-1)^{n-1}(4)^n}{N^{n-1}m^n} \Big[(\Delta\alpha)^n \sin^2\left(\frac{k_x a}{2}\right) \Big(\sum_{\mathbf{k}_1} \frac{\sin^2(k_{1x}a/2)}{\omega_{1,\mathbf{k}}{}^2 - \omega^2} \Big)^{n-1} + (\Delta\beta)^n \sin^2\left(\frac{k_y a}{2}\right) \Big(\sum_{\mathbf{k}_1} \frac{\sin^2(k_{1y}a/2)}{\omega_{1,\mathbf{k}}{}^2 - \omega^2} \Big)^{n-1} \Big]. \quad (27)$$

Since there are no terms in which both $\Delta\alpha$ and $\Delta\beta$ appear, Eq. (27) is easily transformed into the expression

$$G_{\mathbf{k}}{}^{(1)} = \frac{[4q(\Delta\alpha)/m] \sin^2(k_x a/2)}{1 + [4(\Delta\alpha)/mN] \sum_{\mathbf{k}_1} [\sin^2(k_{1x}a/2)/(\omega_{1,\mathbf{k}_1}{}^2 - \omega^2)]} + \frac{[4q(\Delta\beta)/m] \sin^2(k_y a/2)}{1 + [4(\Delta\beta)/mN] \sum_{\mathbf{k}_1} [\sin^2(k_{1y}a/2)/(\omega_{1,\mathbf{k}_1}{}^2 - \omega^2)]}. \quad (28)$$

The entire problem is then reduced to an evaluation of the summations

$$I = \frac{1}{N} \sum_{\mathbf{k}} \frac{\sin^2(k_x a/2)}{\omega_{1,\mathbf{k}}{}^2 - \omega^2}, \quad (29)$$

and

$$J = \frac{1}{N} \sum_{\mathbf{k}} \frac{\sin^2(k_y a/2)}{\omega_{1,\mathbf{k}}{}^2 - \omega^2}. \quad (30)$$

Because of the symmetry associated with this lattice,

$$I(\alpha,\beta) = J(\beta,\alpha). \quad (31)$$

Equation (31) implies that one need evaluate Eq. (29) only. Transforming the right-hand side of Eq. (29) into an integral, we obtain

$$I=\lim_{\epsilon\to 0}\frac{a^2}{4\pi}\int_{-\pi/a}^{\pi/a}\int_{-\pi/a}^{\pi/a}\frac{\sin^2(k_xa/2)dk_xdk_y}{(4\alpha/m)\sin^2(k_xa/2)+(4\beta/m)\sin^2(k_ya/2)-\omega^2-i\epsilon}. \tag{32}$$

In Appendix A, it is shown that the value of this integral is given by

$$I=(m/2\pi\alpha)\tan^{-1}(\alpha/\beta)^{1/2}+(im^2\omega^2/32\alpha\beta)(\beta/\alpha)^{1/2} \tag{33}$$

under the conditions

$$m\omega^2/4\alpha\ll 1$$

and

$$m\omega^2/4\beta\ll 1. \tag{34}$$

These conditions limit our discussion to the elastic portion of the spectrum. It would be reasonably simple, but tedious, to obtain higher order terms in the ω^2 power series expansion of I. Since interesting physical results are obtained from the lowest order terms, we shall not continue the procedure, but shall continue with the determination of the elastic portion of the spectrum.

Substitution of Eq. (33) into Eq. (31) yields

$$J=(m/2\pi\beta)\tan^{-1}(\beta/\alpha)^{1/2}+(im^2\omega^2/32\alpha\beta)(\alpha/\beta)^{1/2}. \tag{35}$$

Substitution of Eqs. (31) and (33) into Eq. (28) yields

$$G_{\mathbf{k}}{}^{(1)}=\frac{(4q\Delta\alpha/m)\sin^2(k_xa/2)}{1+(2\Delta\alpha/\pi\alpha)\tan^{-1}(\alpha/\beta)^{1/2}+(im\omega^2\Delta\alpha/8\alpha\beta)(\beta/\alpha)^{1/2}}+\frac{(4q\Delta\beta/m)\sin^2(k_ya/2)}{1+(2\Delta\beta/\pi\beta)\tan^{-1}(\beta/\alpha)^{1/2}+(im\omega^2\Delta\beta/8\alpha\beta)(\alpha/\beta)^{1/2}}. \tag{36}$$

As in one dimension[1,2] we may associate the positions of the poles of the function $D_{\mathbf{k}}{}^{(1)}(\omega^2)$ with the modes of vibration of the lattice with the randomly distributed impurity springs. The frequencies, for each $\mathbf{k}$, and with polarization vector parallel to $\hat{x}$, are given by the real part of the roots of the equation

$$\omega_{\mathbf{k}}{}^2-\omega^2+G_{\mathbf{k}}{}^{(1)}(\omega^2)=0. \tag{37}$$

A complete solution of $G_{\mathbf{k}}{}^{(1)}(\omega^2)$ would lead to a number of roots, for fixed $\mathbf{k}$, of Eq. (37). The existence of these roots is a mirror of the fact that $\mathbf{k}$, for any one member of the ensemble, is not a good label. By restricting our calculation of $G_{\mathbf{k}}{}^{(1)}(\omega^2)$ to small ω^2, and keeping terms of lowest order only in the resulting small parameters, we have suppressed the multiplicity of these roots. The resulting expression for the elastic mode frequencies is given by

$$\omega_1{}^2=\frac{4\alpha}{m}\left[1+\frac{q(\Delta\alpha)}{\alpha+(2/\pi)(\Delta\alpha)\tan^{-1}(\alpha/\beta)^{1/2}}\right]\sin^2(k_xa/2)+\frac{4\beta}{m}\left[1+\frac{q(\Delta\beta)}{\beta+(2/\pi)(\Delta\beta)\tan^{-1}(\beta/\alpha)^{1/2}}\right]\sin^2(k_ya/2). \tag{38}$$

The solution of Eq. (37) also contains an imaginary part. Writing the solution as

$$\omega=\omega_{1,\mathbf{k}}+\Delta_{1,\mathbf{k}}-i\Gamma_{1,\mathbf{k}}, \tag{39}$$

the imaginary part,

$$\Gamma_{1,\mathbf{k}}=\frac{q\omega_{\mathbf{k}}}{4(\alpha\beta)^{1/2}}\left\{\frac{(\Delta\alpha)^2\sin^2(k_xa/2)}{\alpha[1+(2\Delta\alpha/\pi\alpha)\tan^{-1}(\alpha/\beta)^{1/2}]^2}+\frac{(\Delta\beta)^2\sin^2(k_ya/2)}{\beta[1+(2\Delta\beta/\pi\beta)\tan^{-1}(\beta/\alpha)^{1/2}]^2}\right\}, \tag{40}$$

corresponds[1] to the reciprocal of the lifetime of the mode. It should be noted that $\Gamma_{1,\mathbf{k}}$ is of one order higher in the small parameters of Eq. (34) than is $\Delta_{1,\mathbf{k}}$. Thus, although the low-frequency modes are shifted in frequency, they are quite well defined.

Finally, the density of states $\bar{g}(\omega)$, as indicated in Eq. (17) and calculated in Appendix B, is given by the expression

$$\bar{g}(\omega)=\omega m/\pi(\bar{\alpha}\bar{\beta})^{1/2}. \tag{41}$$

Here, $\bar{\alpha}$ and $\bar{\beta}$ are defined by Eqs. (57) and (58). These results will be discussed in Sec. V, after the analogous expressions have been obtained for the isotopic-impurity problem.

IV. THE ISOTOPIC-IMPURITY PROBLEM

A. Formulation

In this problem, the unperturbed lattice is again the square lattice discussed in Sec. II. In this case, the Nq atoms at randomly positioned sites are replaced by isotopes of mass m'. The mass of the isotope at site l is denoted by m_l. The equation of motion of the atom at this site is given by

$$-\omega^2\begin{pmatrix}x_{l,1}\\x_{l,2}\end{pmatrix}+\frac{1}{m}\begin{pmatrix}\alpha&0\\0&\beta\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+1,1}\\x_{l,2}-x_{l+1,2}\end{pmatrix}+\frac{1}{m}\begin{pmatrix}\alpha&0\\0&\beta\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+2,1}\\x_{l,2}-x_{l+2,2}\end{pmatrix}+\frac{1}{m}\begin{pmatrix}\beta&0\\0&\alpha\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+3,1}\\x_{l,2}-x_{l+3,2}\end{pmatrix}+\frac{1}{m}\begin{pmatrix}\beta&0\\0&\alpha\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+4,1}\\x_{l,2}-x_{l+4,1}\end{pmatrix}$$
$$=\begin{pmatrix}\frac{1}{m}-\frac{1}{m_l}\end{pmatrix}\left\{\begin{pmatrix}\alpha&0\\0&\beta\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+1,1}\\x_{l,2}-x_{l+1,2}\end{pmatrix}+\begin{pmatrix}\alpha&0\\0&\beta\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+2,1}\\x_{l,2}-x_{l+2,2}\end{pmatrix}+\begin{pmatrix}\beta&0\\0&\alpha\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+3,1}\\x_{l,2}-x_{l+3,2}\end{pmatrix}+\begin{pmatrix}\beta&0\\0&\alpha\end{pmatrix}\begin{pmatrix}x_{l,1}-x_{l+4,1}\\x_{l,2}-x_{l+4,2}\end{pmatrix}\right\}. \quad (42)$$

As in Sec. III, the x and y components of the motion are completely decoupled, and may be considered separately. As a result of symmetry, it is sufficient to consider the x-component motion for a complete quadrant of the space of the propagation vector $\mathbf{k}$. The transformation to the normal coordinates of the unperturbed system, Eq. (7), yields the equation of motion,

$$-\omega^2\sum_{\mathbf{k}}Q_1(\mathbf{k})\exp i\mathbf{k}\cdot\mathbf{R}_l+(\alpha/m)\sum_{\mathbf{k}}[2-\exp(ik_xa)-\exp(-ik_xa)]Q_1(k)\exp i\mathbf{k}\cdot\mathbf{R}_l$$
$$+(\beta/m)\sum_{\mathbf{k}}[2-\exp(ik_ya)-\exp(-ik_ya)]Q_1(\mathbf{k})\exp i\mathbf{k}\cdot\mathbf{R}_l$$
$$=\sum_{\mathbf{k}}Q_1(\mathbf{k})[(1/m)-(1/m_l)]\exp i\mathbf{k}\cdot\mathbf{R}_l\{\alpha[2-\exp ik_xa-\exp(-ik_xa)]+\beta[2-\exp ik_ya-\exp(-ik_ya)]\}. \quad (43)$$

Equation (43) is similar in form to Eq. (9). By mathematical manipulations which are similar to those which lead from Eq. (9) to Eq. (11), Eq. (43) is transformed into

$$(\omega_{1,\mathbf{k}}^2-\omega^2)Q_1(\mathbf{k})=-\sum_{\mathbf{k}'}Q_1(\mathbf{k}')\Phi_{\mathbf{k},\mathbf{k}'}, \quad (44)$$

where

$$\Phi_{\mathbf{k},\mathbf{k}'}=(\omega_{1,\mathbf{k}'}/N)\sum_l[(m/m_l)-1]\exp i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_l. \quad (45)$$

This result is identical to that obtained by Langer[2] for the one-dimensional problem. It is easily shown that the entire difference between the two problems is represented in the form of the function $G_{\mathbf{k}}^{(1)}(\omega^2)$ which, for this problem, is given by

$$G_{\mathbf{k}}^{(1)}(\omega^2)=\frac{q\lambda\omega_{1,\mathbf{k}}^2}{1+(\lambda/N)\sum_{\mathbf{k}'}[\omega_{1,\mathbf{k}'}^2/(\omega_{1,\mathbf{k}'}^2-\omega^2)]}, \quad (46)$$

where

$$\lambda=(m/m')-1. \quad (47)$$

The remainder of the problem consists of the evaluation of the sum

$$\sum_{\mathbf{k}'}\frac{\omega_{1,\mathbf{k}'}^2}{\omega_{1,\mathbf{k}'}^2-\omega^2}=\frac{4\alpha}{m}\sum_{\mathbf{k}'}\frac{\sin^2(k_x'a/2)}{\omega_{1,\mathbf{k}'}^2-\omega^2}$$
$$+\frac{4\beta}{m}\sum_{\mathbf{k}'}\frac{\sin^2(k_y'a/2)}{\omega_{1,\mathbf{k}'}^2-\omega^2}. \quad (48)$$

The two sums which appear on the right-hand side of Eq. (48) have been evaluated in Appendix A and have been used in Sec. III, for the case where Eq. (34) holds. Substituting Eqs. (33) and (35) into Eq. (48), and inserting the result into Eq. (46), one obtains the result

$$G_{\mathbf{k}}^{(1)}(\omega^2)=q\lambda\omega_{1,\mathbf{k}}^2\{1+\lambda[1+(i\omega^2m/(\alpha\beta)^{1/2})]\}^{-1}. \quad (49)$$

With this result, it is easily shown that the frequencies of propagation, in the elastic limit, are given by the expression

$$\omega^2=\omega_{1,\mathbf{k}}^2(1+q\kappa), \quad (50)$$

where

$$\kappa=1-(m'/m). \quad (51)$$

The inverse lifetimes are given by

$$\Gamma_{\mathbf{k}}^{(1)}=\frac{q\lambda^2m\omega_{1,\mathbf{k}}^3}{2(1+\lambda)^2(\alpha\beta)^{1/2}}. \quad (52)$$

Before leaving this problem, we should point out that another aspect of it has been treated by Mahanty, Maradudin, and Weiss.[3] They considered the local modes which arise when $m'<m$. Many of the mathematical difficulties, which these authors surmounted quite elegantly, have been avoided in this work because attention was restricted to the elastic spectrum.

V. DISCUSSION: THE VIRTUAL-CRYSTAL APPROXIMATION

One frequently finds statements in the literature that the so-called "virtual-crystal approximation" must hold for the elastic spectrum of a crystal containing a small number of impurities, or a small degree of disorder. In this approximation, the masses are all taken as the average mass, and the spring constants are all taken as the average spring constant. The justification given for this approximation is usually that the wavelengths of the modes are very long compared to the range of the local perturbations, so that these perturbations are just averaged. Other authors have carried the approximation further, and have concluded that the failure of the

[3] J. Mahanty, A. A. Maradudin, and G. H. Weiss, Progr. Theoret. Phys. (Kyoto) **24**, 648 (1960).

virtual-crystal approximation to explain the vibrational frequencies near the Brillouin zone surface, of dilute alloys, is evidence for the existence of short-range order in these alloys. In this section, we examine this approximation with the aid of the solutions obtained in the preceding sections.

The two one-dimensional solutions obtained previously[1,2] yield virtual-crystal-approximation type results to first order in q. Langer[1] finds that the elastic spectrum of a system with isotopic impurities is completely described by an average mass given by

$$\bar{m}=(1-q)m+qm'. \tag{53}$$

In (P-B), the authors find that it is the inverse of the spring constant which must be averaged. That is, taking γ as the nearest-neighbor spring constant of the unperturbed system, and γ' as the spring constant which replaces it at randomly positioned sites, the elastic spectrum is completely described by an effective spring constant $\bar{\gamma}$, which is given by the expression

$$1/\bar{\gamma}=[(1-q)/\gamma]+(q/\gamma'). \tag{54}$$

Henceforth, we shall speak of this equation as the virtual-crystal approximation for spring impurities.

For the isotopic substitution in two dimensions, the situation is quite similar. Equation (50) can be rewritten as

$$\omega^2=m\omega_{1,k}{}^2[m(1-q)+qm']^{-1} \tag{55}$$

to first order in q. Since $m\omega_{1,k}{}^2$ is independent of mass, this is the result expected from the virtual-crystal approximation.

There is good reason to believe that the approximation must hold for the elastic spectrum of a system of any dimensionality and any concentration where the only substitutions are isotopic and the system is homogeneous on a macroscopic scale. The reasoning goes as follows. The masses enter the dynamical equation through acceleration terms only. Thus, they do not contribute at all to the static elastic constants, which depend only on the interactions. The only way that the masses enter into the classical expressions for the elastic spectrum is through the density, since the elastic spectrum is, of course, entirely determined, for a homogeneous solid, by a knowledge of the elastic constants and the density. Thus, the only condition for the applicability of the virtual-crystal approximation to the elastic spectrum of an isotope substituted system is that it be homogeneous. That is, one must be able to divide the crystal into volume elements whose dimensions are small compared to the wavelength of the elastic modes, all of which have the same density associated with them. In this case, the virtual-crystal approximation is just a means of calculating that density.

The argument just presented leads one to suspect that it is in the case where the spring constants are changed that one expects a breakdown of the approximation. This is the case. For comparison, we may rewrite Eq. (54) as

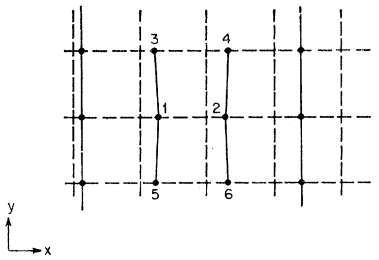

FIG. 1. Pictorial representation of the displacements of atoms in the vicinity of an impurity spring, while taking part in a (10) longitudinal mode. The dashed lines represent the equilibrium positions. The impurity spring, with $\alpha'>\alpha$, is between the atoms denoted 1 and 2. Shear along the lines 3-1-5 and 4-2-6 introduces the factor β into the expression for the effective α.

$$\bar{\gamma}=\gamma[1+q\Delta\gamma/(\gamma+\Delta\gamma)]. \tag{56}$$

Comparison of Eq. (38), for the perturbed system, with Eq. (4) for the unperturbed system, shows that one can define effective force constants $\bar{\alpha}$ and $\bar{\beta}$, by the relations

$$\bar{\alpha}=\alpha\left[1+\frac{q\Delta\alpha}{\alpha+\Delta\alpha(2/\pi)\tan^{-1}(\alpha/\beta)^{1/2}}\right] \tag{57}$$

and

$$\bar{\beta}=\beta\left[1+\frac{q\Delta\beta}{\beta+\Delta\beta(2/\pi)\tan^{-1}(\beta/\alpha)^{1/2}}\right]. \tag{58}$$

These results indicate immediately that a knowledge of q, α, and α' is not sufficient to define $\bar{\alpha}$. The relationship between α and β also enters into the expression for $\bar{\alpha}$. The same is true for $\bar{\beta}$.

The reasons for this dependence can be visualized easily. Consider Fig. 1, which portrays the displacements of atoms in the region of an impurity spring from equilibrium while participating in a longitudinal mode. Here, we have taken $\alpha'>\alpha$. The intersections of the dashed lines represent the equilibrium positions. The straight vertical lines represent the lines of constant phase associated with the mode of the unperturbed system. Atoms 1 and 2 are connected by a spring which is stronger, and which prevents them from separating by the distance associated with the unperturbed mode. As a result, there is a shear component in the motion of, say, atom 1 with respect to atoms 3 and 5, and the shear constant β enters into the frequency of a mode whose frequency is determined entirely by α in the unperturbed system. Some of the relationships of this picture to the mathematical solution, Eq. (57), are easily seen. We note, first of all, that this is a calculation to first order in q. Thus, we can neglect other "wrong" springs. As a result, the picture indicates that there is no shear motion of atom 1 with respect to atom 2. Thus, β' does not enter into the expression for $\bar{\alpha}$. Similarly, we would expect, from this picture, that the

virtual-crystal approximation would hold for $\bar{\alpha}$ if $\beta=0$. This is the case, since $(2/\pi)\tan^{-1}(\infty)=1$, making Eq. (57) identical to Eq. (56). Setting $\beta=0$ also has the effect, of course, of making the lattice unstable. Finally, we may look at the problem as the picture does, with a classical perturbation theory viewpoint, treating $(\Delta\alpha)$ as the perturbation on this mode. In first order, the energy shift is obtained by letting the system undergo its unperturbed motion. In this order, β should not appear. It is only in second order, when the motion has been changed to first order, that the effect of β should enter in. It is readily seen from Eq. (57) that β only enters into terms of second and higher order in $\Delta\alpha$.

Equations (57) and (58) do show that the two virtual-crystal approximations which may be used to obtain average spring constants form upper and lower bounds for the actual $\bar{\alpha}$ and $\bar{\beta}$, for the model discussed here. The $\tan^{-1}(\alpha/\beta)^{1/2}$ and $\tan^{-1}(\beta/\alpha)^{1/2}$ in these equations have upper and lower bounds of 0 and $\pi/2$. For $\tan^{-1}(\beta/\alpha)^{1/2}=0$, these equations become

$$1/\bar{\alpha}=[(1-q)/\alpha]+[q/\alpha'], \tag{59}$$

and

$$\bar{\beta}=(1-q)\beta+q\beta'. \tag{60}$$

For $\tan^{-1}(\beta/\alpha)^{1/2}=\pi/2$, of course, the situations are reversed. Equation (59) corresponds to the $1/\gamma$ averaging which was obtained in one dimension, while Eq. (60) is the straightforward averaging of the spring constants. Any intermediate values of $\tan^{-1}(\beta/\alpha)$ will yield results which lie between the two simple forms of averaging. Note that this discussion has been restricted to the cases where $\Delta\alpha/\alpha>-1$ and $\Delta\beta/\beta>-1$. These conditions must be satisfied if each atom has its motion centered about a position of stable equilibrium in the static crystal.

Paul[4] has obtained the identical upper and lower bounds for the elastic constants of isotropic multiphase materials. He shows that such bounds hold in general in the situation where "$\cdots$the constituents are distinct and capable of separation by purely mechanical means (e.g., not solid solutions)." In the language of lattice dynamics, Paul is restricting his work to situations in which the force constants between atoms belonging to the interiors of different constituents are zero. That is, the elastic constants associated with each of the constituents completely describe the interactions. This situation is quite similar to that discussed here. In the determination of the elastic constants, it is only the pairwise interactions, and not the masses, which are significant. Thus, since nearest-neighbor interactions only are considered, we may consider each impurity spring as an independent constituent, and the results are consistent with Paul's bounds. For some reason that we do not understand, the fact that the system is not isotropic does not seem to be important in this case.

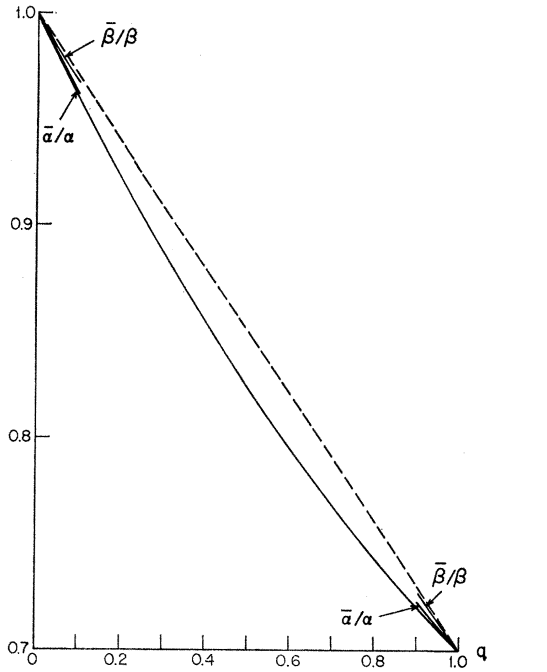

FIG. 2. Comparison of the effective force constants $\bar{\alpha}$ and $\bar{\beta}$, as a function of concentration q, with virtual crystal approximation predictions. Here, $\Delta\alpha/\alpha$ and $\Delta\beta/\beta$ have been taken as -0.3, while $\alpha/\beta=9$. The dashed line represents the linear averaging of the force constant, while the solid curve shows the averaging of the inverse of the force constant.

This analysis raises the possibility, however, that important and interesting effects may appear when longer range interactions appear.

Figure 2 shows the deviations of $\bar{\alpha}$ and $\bar{\beta}$, as given by Eqs. (57) and (58), from the virtual-crystal approximation. In these graphs, we have taken $\alpha/\beta=9$. This ratio was obtained from the elastic constants of β brass, as measured by McManus[5] by setting $\alpha/\beta=C_{11}/[(C_{11}-C_{12})/2]$. As expected, $\bar{\alpha}$ lies close to the curve defined by Eq. (59), while $\bar{\beta}$ is given, approximately, by Eq. (60). A choice of $\alpha/\beta\approx 1$, corresponding to a strongly covalent crystal, would lead to lines for $\bar{\alpha}$ and $\bar{\beta}$ which have slopes, at $q=0$ and 1, that are approximately equal to the average of the slopes associated with the two virtual-crystal approximations.

Since $\bar{\alpha}$ and $\bar{\beta}$ also determine, through Eq. (41), the spectral distribution function for small ω^2, the remarks about $\bar{\alpha}$ and $\bar{\beta}$ apply to it as well.

Finally, it is important to note that the random-spring calculation performed in this work applies specifically to the order-disorder problem. There exists another interesting related situation which remains to be treated. It is the case where impurity atoms, which are substituted into the two-dimensional lattice, have approximately the same mass as the solvent atoms, but change the spring constants. In this case, the four sets of spring

[4] B. Paul, Trans. A.I.M.E. **218**, 37 (1960).

[5] G. M. McManus, Phys. Rev. **129**, 2004 (1963).

constants associated with the interaction of the impurity atom with each of its neighbors must appear together in the averaging. Work is under way on this problem, and it is expected that the results will soon be presented. The results obtained here indicate that it is highly likely that the virtual-crystal are inadequate for the description of the modifications of the elastic spectrum of such a system. If this is the case, any attempt to assign observed breakdowns of the approximation to ordering or clustering, without corroborative diffraction evidence, would appear unjustified. This is particularly true when the modes are not a portion of the elastic spectrum, but are associated with the Brillouin zone surface.

ACKNOWLEDGMENTS

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The authors are indebted to U. F. Kocks and B. Budiansky, who pointed out the existence of the work of B. Paul (Ref. 4).

APPENDIX A: EVALUATION OF THE INTEGRAL OF EQ. (32)

The value of the integral

$$I=\lim_{\epsilon\to 0}\int_{-\pi/a}^{\pi/a}\int_{-\pi/a}^{\pi/a}\frac{\sin^2(k_xa/2)dk_xdk_y}{(4\alpha/m)\sin^2(k_xa/2)+(4\beta/m)\sin^2(k_ya/2)-\omega^2-i\epsilon} \tag{A1}$$

is required in the limits $\omega^2\ll 4\alpha/m$ and $\omega^2\ll 4\beta/m$. We begin by fixing k_y and evaluating the integral over k_x. Consider the case $\omega^2>(4\beta/m)\sin^2(k_ya/2)$. Let

$$\omega'^2=\omega^2-(4\beta/m)\sin^2(k_ya/2), \tag{A2}$$

and

$$I_1=\int_{-\pi/a}^{\pi/a}\frac{\sin^2(k_xa/2)dk_x}{(4\alpha/m)\sin^2(k_xa/2)-\omega'^2-i\epsilon}. \tag{A3}$$

Letting

$$Z=\exp ik_xa, \tag{A4}$$

$$\omega''^2=(m/\alpha)\omega'^2, \tag{A5}$$

and

$$\epsilon''=(m/\alpha)\epsilon, \tag{A6}$$

Eq. (A3) becomes

$$I_1=\frac{m}{4\alpha ai}\oint\frac{(Z-1)^2dZ}{Z[Z^2-(2-\omega''^2-i\epsilon'')Z+1]}, \tag{A7}$$

where the contour of integration is the unit circle. The denominator of the integrand has three roots,

$$Z_0=0, \tag{A8}$$

$$Z_1=(1-\tfrac{1}{2}\omega''^2-\tfrac{1}{2}i\epsilon'')+i[1-(1-\tfrac{1}{2}\omega''^2-\tfrac{1}{2}i\epsilon'')^2]^{1/2}, \tag{A9}$$

$$Z_2=(1-\tfrac{1}{2}\omega''^2-\tfrac{1}{2}i\epsilon'')-i[1-(1-\tfrac{1}{2}\omega''^2-\tfrac{1}{2}i\epsilon'')^2]^{1/2}. \tag{A10}$$

Since $\omega''^2<1$, Z_1 is inside and Z_2 is outside the unit circle. Thus, we obtain the solutions

$$I_1=\frac{m\pi}{2\alpha a}\left\{\frac{1}{Z_1Z_2}+\frac{(Z_1-1)^2}{Z_1(Z_1-Z_2)}\right\}. \tag{A11}$$

Evaluating Eq. (A11) in terms of the variables of Eq. (A1), we obtain

$$I_1=\frac{m\pi}{2\alpha a}\left\{1+\frac{i(\beta/\alpha)^{1/2}[(m\omega^2/4\beta)-\sin^2(k_ya/2)]^{1/2}}{[1-(m\omega^2/4\alpha)+(\beta/\alpha)\sin^2(k_ya/2)]^{1/2}}\right\}. \tag{A12}$$

In a similar manner, we consider the case $\omega^2<(4\beta/m)\sin^2(k_ya/2)$. Letting

$$\omega'^2=(4\beta/m)\sin^2(k_ya/2)-\omega^2, \tag{A13}$$

we obtain the integral

$$I_2=\int_{-\pi/a}^{\pi/a}\frac{\sin^2(k_xa/2)dk_x}{(4\alpha/m)\sin^2(k_xa/2)+\omega'^2}$$

$$=\frac{m\pi}{2\alpha a}\left\{1-\frac{(\beta/\alpha)^{1/2}[\sin^2(k_ya/2)-(\omega^2m/4\beta)]^{1/2}}{[1+(\beta/\alpha)\sin^2(k_ya/2)-(\omega^2m/4\alpha)]^{1/2}}\right\}. \tag{A14}$$

Substitution of Eqs. (A12) and (A14) into Eq. (A1) yields the expression

$$I=\frac{am}{8\pi\alpha}\int_{-\pi/a}^{\pi/a}dk_y+\frac{iam}{4\pi\alpha}\int_0^{(2/a)\sin^{-1}(m\omega^2/4\beta)^{1/2}}\frac{(\beta/\alpha)^{1/2}[(\omega^2m/4\beta)-\sin^2(k_ya/2)]^{1/2}dk_y}{[1-(m\omega^2/4\alpha)+(\beta/\alpha)\sin^2(k_ya/2)]^{1/2}}$$

$$-\frac{am}{4\pi\alpha}\int_{(2/a)\sin^{-1}(m\omega^2/4\beta)^{1/2}}^{\pi/a}\frac{(\beta/\alpha)^{1/2}[\sin^2(k_ya/2)-(\omega^2m/4\beta)]^{1/2}dk_y}{[1+(\beta/\alpha)\sin^2(k_ya/2)-(\omega^2m/4\alpha)]^{1/2}}. \tag{A15}$$

It should be noted at this time that the integrands are all real. Thus, the first and third terms of the right-hand side of Eq. (A15) contribute the real part of I, while the second term yields the imaginary part.

For convenience, we denote

$$I_3=(am/8\pi\alpha)\int_{-\pi/a}^{\pi/a}dk_y=m/4\alpha, \tag{A16}$$

$$I_4=\frac{iam}{4\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_0^{(2/a)\sin^{-1}(m\omega^2/4\beta)^{1/2}}\frac{[(m\omega^2/4\beta)-\sin^2(k_ya/2)]^{1/2}dk_y}{[1-(m\omega^2/4\alpha)+(\beta/\alpha)\sin^2(k_ya/2)]^{1/2}}, \tag{A17}$$

$$I_5=\frac{am}{4\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_{(2/a)\sin^{-1}(m\omega^2/4\beta)^{1/2}}^{\pi/a}\frac{[\sin^2(k_ya/2)-(m\omega^2/4\beta)]^{1/2}dk_y}{[1-(m\omega^2/4\alpha)+(\beta/\alpha)\sin^2(k_ya/2)]^{1/2}}. \tag{A18}$$

Let

$$t=\sin(k_ya/2),$$

$$\delta=(m\omega^2/4\beta)\ll1,$$

and

$$\delta'=(m\omega^2/4\alpha)\ll1. \tag{A19}$$

Then Eq. (A17) can be written as

$$I_4=\frac{ima^2}{8\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_0^{(\delta)^{1/2}}\frac{(\delta-t^2)^{1/2}dt}{[1-\delta'+(\beta/\alpha)t^2]^{1/2}[1-t^2]^{1/2}}. \tag{A20}$$

It is easily seen that $t\ll1$ over the range of integration. Thus, we may approximate the entire denominator of the integrand of Eq. (A20) by unity to obtain an expression which is good to only first order in δ and δ'. In this case, we may write

$$I_4\approx\frac{im}{2\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_0^{(\delta)^{1/2}}(\delta-t^2)^{1/2}dt$$

$$=\frac{im^2\omega^2}{32\alpha\beta}\left(\frac{\beta}{\alpha}\right)^{1/2}. \tag{A21}$$

Substitution of Eq. (A19) into Eq. (A18) yields the expression

$$I_5=\frac{m}{2\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_{(\delta)^{1/2}}^1\frac{(t^2-\delta)^{1/2}dt}{[1-t^2]^{1/2}[1-\delta'+(\beta/\alpha)t^2]^{1/2}}. \tag{A22}$$

In this case, the approximations are less straightforward. It is clear that δ' may be set equal to zero, since it is always very much less than the other terms contained in the radical in which it appears. Further simplification of the integrand is hindered by the fact that $\delta/t^2\approx1$ near the lower limit of integration. Nevertheless, we may write $[1-(\delta/t^2)]^{1/2}\approx1$ in the integrand for the reasons which follow. The factor $[1+(\beta/\alpha)t^2]^{1/2}$ of the denominator is a

relatively slowly varying function. As a result, the remainder of the integrand is sharply peaked near the upper limit of integration. In this region, it is quite reasonable to replace the radical of the numerator by unity. Hence, if I_5 contained the entire contribution to the real part of I, it would be sufficient to make this replacement. Unfortunately, however, a major portion of I_5 is cancelled by I_3. Thus it is necessary to justify this replacement a bit more carefully. We do this by calculating an upper and lower bound to the integral. It is then shown that the difference between the two is negligible compared to the real part of I. To compute these bounds, we note that the integrand is positive and real over the range of integration. In addition, over this range, $[1-(\delta/t^2)]\leqslant[1-(\delta/t^2)]^{1/2}\leqslant 1$. Thus, lower and upper bounds to the integral can be obtained by using the upper and lower bounds of the radical in the integrand. To obtain the values of the associated integrals, we need

$$\frac{m}{2\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_{(\delta)^{1/2}}^{1}\frac{t\,dt}{[1-t^2]^{1/2}[1+(\beta/\alpha)t^2]^{1/2}}=\frac{m}{4\alpha}-\frac{m}{2\pi\alpha}\tan^{-1}\left(\frac{\alpha}{\beta}\right)^{1/2}+O(\delta)\,, \tag{A23}$$

and

$$\frac{m\delta}{2\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\int_{(\delta)^{1/2}}^{1}\frac{dt}{t[1-t^2]^{1/2}[1+(\beta/\alpha)t^2]^{1/2}}$$
$$=\frac{m\delta}{4\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\ln\left\{\frac{[\alpha+(\beta-\alpha)\delta-\beta\delta^2]^{1/2}+\alpha}{\delta(\alpha)^{1/2}}+\frac{(\beta-\alpha)}{2\alpha}\right\}-\frac{m\delta}{4\pi\alpha}\left(\frac{\beta}{\alpha}\right)^{1/2}\ln\left[\frac{(\beta-\alpha)}{2\alpha}\right]. \tag{A24}$$

Equation (A24) shows clearly that the difference between the upper and lower bounds is of the order of $\delta\ln\delta$. This should be compared with the result for I, obtained by using Eq. (A23) for I_5, and substituting the results of Eqs. (A16) and (A21) into Eq. (A15). We get

$$I=\frac{m}{2\pi\alpha}\tan^{-1}\left(\frac{\alpha}{\beta}\right)^{1/2}+\frac{im^2\omega^2}{32\alpha\beta}\left(\frac{\beta}{\alpha}\right)^{1/2}. \tag{A23}$$

Since the real part of the right-hand side of Eq. (A23) is of order unity, we are justified in neglecting the difference between the upper and lower bounds of I_5.

APPENDIX B: EVALUATION OF THE SPECTRAL DISTRIBUTION FUNCTION FOR THE IMPURITY-SPRING PROBLEM

As indicated in Eq. (17), the spectral distribution function for this problem is given by

$$\bar{g}(\omega)=\frac{4\omega}{\pi}\lim_{\substack{N\to\infty\\ \epsilon\to 0}}\frac{1}{N}\,\mathrm{Im}\,\mathrm{Tr}D^{(1)}(\omega^2+i\epsilon)\,. \tag{B1}$$

Using Eq. (26) for $D^{(1)}$ and Eq. (36) for $G^{(1)}$, Eq. (B1) becomes, for small ω^2,

$$\bar{g}(\omega)=\lim_{\substack{N\to\infty\\ \epsilon\to 0}}(4\omega/N\pi)\sum_{\mathbf{k}}[(4\bar{\alpha}/m)\sin^2(k_xa/2)+(4\bar{\beta}/m)\sin^2(k_ya/2)-\omega^2-i\epsilon]^{-1}$$
$$=(a^2\omega/\pi^3)\lim_{\epsilon\to 0}\mathrm{Im}\int_{-\pi/a}^{\pi/a}\int_{-\pi/a}^{\pi/a}[(4\bar{\alpha}/m)\sin^2(k_xa/2)+(4\bar{\beta}/m)\sin^2(k_ya/2)-\omega^2-i\epsilon]^{-1}dk_xdk_y\,. \tag{B2}$$

Here, $\bar{\alpha}$ and $\bar{\beta}$ are defined by Eqs. (57) and (58), respectively.

Let $I(\omega^2)$ be the integral of Eq. (B2). It can be written as

$$I=2(I_1+I_2)\,, \tag{B3}$$

where

$$I_1=\int_0^{(2/a)\sin^{-1}(m\omega^2/4\beta)^{1/2}}dk_y\int_{-\pi/a}^{\pi/a}[(4\bar{\alpha}/m)\sin^2(k_xa/2)+(4\bar{\beta}/m)\sin^2(k_ya/2)-\omega^2-i\epsilon]^{-1}dk_x\,, \tag{B4}$$

and

$$I_2=\int_{(2/a)\sin^{-1}(m\omega^2/4\beta)^{1/2}}^{\pi/a}dk_y\int_{-\pi/a}^{\pi/a}[(4\bar{\alpha}/m)\sin^2(k_xa/2)+(4\bar{\beta}/m)\sin^2(k_ya/2)-\omega^2-i\epsilon]^{-1}dk_x\,. \tag{B5}$$

The real part of the denominator of the integrand of I_2 is always positive. As a result, the imaginary part of the integral is of first and higher order in ϵ, and vanishes when the ϵ limit of Eq. (B2) is taken.

Let

$$\xi^2=m\omega^2/4\bar{\beta}, \tag{B6}$$

$$\omega''^2=(m/\bar{\alpha})[\omega^2-(4\bar{\beta}/m)\sin^2(k_y a/2)], \tag{B7}$$

$$I_3=\frac{m}{\bar{\alpha}}\int_{-\pi/a}^{\pi/a}\frac{dk_x}{4\sin^2(k_x a/2)-\omega''^2-i\epsilon}. \tag{B8}$$

Then

$$I_1=\int_0^{(2a)\sin^{-1}\xi} I_3 dk. \tag{B9}$$

Substitution of Eq. (A4) into Eq. (B8) yields

$$I_3=\frac{im}{a\bar{\alpha}}\oint\frac{dZ}{Z^2-(2-\omega''^2-i\epsilon)Z+1}, \tag{B10}$$

where the contour is the unit circle.

The denominator of the integrand of Eq. (B10) is similar to that of Eq. (A7). It has roots, Z_1 and Z_2, given by Eqs. (A9) and (A10), respectively. As before, Z_1 lies inside and Z_2 lies outside the unit circle. Thus,

$$\begin{aligned}I_3&=(mi/a\bar{\alpha})[2\pi i/(Z_1-Z_2)]\\&=(2\pi i/a)\{[\omega^2-(4\bar{\beta}/m)\sin^2(k_y a/2)][(4\bar{\alpha}/m)+(4\bar{\beta}/m)\sin^2(k_y a/2)-\omega^2]\}^{-1/2}.\end{aligned} \tag{B11}$$

After some simple manipulations which use Eq. (B6), substitution of Eq. (B11) into Eq. (B9) yields

$$I_1=\frac{im\pi}{2a(\bar{\alpha}\bar{\beta})^{1/2}}\int_0^{(2/a)\sin^{-1}\xi}\frac{dk_y}{[\xi^2-\sin^2(k_y a/2)]^{1/2}[1-(m\omega^2/4\bar{\alpha})+(\bar{\beta}/\bar{\alpha})\sin^2(k_y a/2)]^{1/2}}. \tag{B12}$$

To lowest order in ω^2, we have

$$\begin{aligned}I_1&\approx\frac{im\pi}{2a(\bar{\alpha}\bar{\beta})^{1/2}}\int_0^{(2/a)\sin^{-1}\xi}\frac{dk_y}{[\xi^2-\sin^2(k_y a/2)]^{1/2}}\\&=\frac{im\pi^2}{2a^2(\bar{\alpha}\bar{\beta})^{1/2}}.\end{aligned} \tag{B13}$$

Substitution of Eqs. (B3) and (B13) into Eq. (B2) yields the density of states

$$\bar{g}(\omega)=\frac{\omega m}{2\pi(\bar{\alpha}\bar{\beta})^{1/2}}. \tag{B14}$$