APPENDIX B. THE CASE OF EQUAL MASSES AND RANDOM FORCE CONSTANTS

The equation of motion of the $(p-1)$th atom of a chain of random masses and equal spring constants is, from (1),

$$\begin{aligned}\frac{d^2u_{p-1}}{dt^2}&=\frac{\alpha}{m_{p-1}}[u_{p-2}-2u_{p-1}+u_p]\\&=\frac{\alpha}{m_{p-1}}[u_{p-2}-u_{p-1}]-\frac{\alpha}{m_{p-1}}[u_{p-1}-u_p]\\&=\frac{\alpha}{m_{p-1}}w_{p-1}-\frac{\alpha}{m_{p-1}}w_p,\end{aligned}\qquad\text{(B.1)}$$

where

$$w_p=u_{p-1}-u_p.$$

Similarly

$$\frac{d^2u_p}{dt^2}=\frac{\alpha}{m_p}w_p-\frac{\alpha}{m_p}w_{p+1}.\qquad\text{(B.2)}$$

Substracting we obtain

$$\frac{d^2w_p}{dt^2}=\frac{\alpha}{m_{p-1}}w_{p-1}-\left(\frac{\alpha}{m_{p-1}}+\frac{\alpha}{m_p}\right)w_p+\frac{\alpha}{m_p}w_{p+1},\qquad\text{(B.3)}$$

and this is of exactly the same form as (1) with constant m and random α.

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Vibration Frequency Spectra of Disordered Lattices. II. Spectra of Disordered One-Dimensional Lattices*

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By using a combination of the moment-trace method and a new method, the "delta-function" method, the vibrational frequency spectrum of a randomly disordered, two-component, isotopic, linear chain has been computed for a wide range of the concentrations of the two kinds of particles and of their mass ratios. In addition the particular case of a chain in which the mass of one of the isotopic constituents becomes infinite can be treated exactly, and the results of this analysis shed light on the form of the spectra for lattices with large but finite mass ratios for the two constituents. The spectra are characterized by the disappearance of the square-root singularity at the maximum frequency which is found in ordered one-dimensional lattices, and by the appearance of impurity bands, the nature of which is discussed. Finally, the zero-point energy of a randomly disordered lattice is calculated and compared with the zero-point energy of an ordered lattice and of the separated phases.

1. INTRODUCTION

In a previous paper[1] a method for obtaining the moments of the vibrational frequency spectrum of a disordered two-component linear chain was described, and explicit expressions for the even moments up to μ_{20} were given. In the present paper we apply these results to the construction of frequency spectra of disordered isotopic linear chains for a wide variety of concentrations of the two kinds of particles and of the ratios of their masses. In obtaining these spectra the moments are used in two different ways: In the first method, the spectrum is expanded in an infinite series of Legendre polynomials whose coefficients are linear combinations of the moments; in the second method, which is believed to be new, polynomial approximations to a Dirac delta-function are used to give the value of the spectrum at any point as a linear combination of the moments.

We begin by considering a particular one-dimensional disordered lattice problem which can be solved exactly, namely the case in which the mass of one atomic species becomes infinite, and obtain the distribution of normal mode frequencies for this case. We then obtain spectra for the finite-mass case by the two methods mentioned above. The zero-point energy of a randomly disordered linear chain is also calculated. In an Appendix the relation between asymptotic properties of the moments and the high- and low-frequency behavior of the spectrum is discussed.

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[1] Domb, Maradudin, Montroll, and Weiss, preceding paper [Phys. Rev. **115**, 18 (1959)].

2. SPECTRUM FOR A DISORDERED LATTICE WITH THE MASS OF ONE OF THE ATOMIC SPECIES INFINITE

In one dimension there is one disordered lattice problem that can be solved exactly: the case in which the mass of one species of atom is infinite. We believe that the details of this spectrum shed some light on the problem of the disordered chain with the mass of one of the atomic species very much larger than that of the second.

The essential simplification introduced by an infinite mass is that the linear chain is divided into "islands" of light masses bounded by "walls" of rigid atoms. In higher dimensions even the infinite-mass case is difficult, if not impossible, to solve analytically. Consider a chain of N atoms and let τ be the probability that a light atom occupies a given lattice site. Then the probability of having an "island" or run of n light atoms is, in the limit $N\rightarrow\infty$, $\tau^n(1-\tau)^2$. A run of length zero corresponds to a single infinite mass, and the probability of this is $(1-\tau)$. The normal mode frequencies of a chain of n particles of mass m enclosed between rigid walls are

$$\omega=\omega_L\left\{\left(1-\cos\frac{\pi s}{n+1}\right)\Big/2\right\}^{\frac{1}{2}}$$

$$=\omega_L\sin\frac{\pi s}{2(n+1)}. \quad s=1,\,2,\,\cdots,\,n. \qquad (1)$$

Thus every rational number in the interval (0,1) except for the end points generates a frequency given by Eq. (1). We now derive that weighting factor which is to be associated with a given rational number. Let us assume that $s/(n+1)$ is reduced to lowest terms. This ratio can be realized in many different ways since $s/(n+1)=2s/(2n+2)=\cdots$. Hence the probability associated with the frequency $\omega_L\sin[\pi s/(2n+2)]$ is

$$(1-\tau)^2\sum_{k=1}^{\infty}\tau^{kn+(k-1)}=\frac{(1-\tau)^2\tau^n}{1-\tau^{n+1}}. \qquad (2)$$

The relation

$$(1-\tau)^2\sum_{n=0}^{\infty}\frac{\varphi(n+1)\tau^n}{1-\tau^{n+1}}=1, \qquad (3)$$

where $\varphi(n)$ is the number of integers less than and prime to n, is known from the theory of Lambert series[2]; hence the probabilities given by Eq. (2) sum to unity, as they must. We therefore see that the frequency spectrum for a one-dimensional disordered lattice containing atoms with an infinite mass is given by a series of lines (delta functions) with varying weights located at a set of points in one-to-one correspondence with the rational numbers. There is also a delta-function at the origin of strength $(1-\tau)$ which corresponds to the zero frequencies due to the infinite masses. The spectrum differs essentially from a normal one-dimensional spectrum of a chain of N atoms. Although this also consists of N lines, we can divide these lines into groups in which the variation of their magnitudes is less than any given quantity ϵ, and as $N\rightarrow\infty$ the number of lines in each group becomes large and the spectrum tends to a continuous function. However, as $N\rightarrow\infty$ in our particular case the limiting function still exists only at points given by Eq. (1). Thus, near any one point of the spectrum there will be infinitely many others, but from Eq. (2) we see that the closer any other point comes to the first the larger is the difference in their magnitudes [since by Eq. (2), these magnitudes depend only on n and not on s].

Some idea of the distribution of frequencies in the spectrum can be obtained from Table I which lists the expected distribution of frequencies in $(0,\omega_L)$ in intervals of $\omega_L/10$ for $\tau=\frac{1}{2}$.

It is possible to calculate the behavior of the expected integrated frequency spectrum in a neighborhood of the maximum frequency. Let $N(\omega)$ be defined as $\int_\omega^{\omega_L}g(\omega)d\omega$, where $g(\omega)$ is the expected frequency spectrum. We shall study the behavior of $N(\omega)$ in the neighborhood of $\omega=\omega_L$. From Eq. (1) we see that we must study the fraction of frequencies in an interval specified by the requirement that $s/(n+1)$ be close to one. For simplicity we take n large and set $s=n$. Then the length of the interval to be considered in the frequency domain is

$$\omega_L-\omega=\omega_L\left(1-\sin\frac{\pi n}{2(n+1)}\right)=\omega_L\left(1-\cos\frac{\pi}{2(n+1)}\right)$$

$$\approx\frac{\omega_L\pi^2}{8(n+1)^2}. \qquad (4)$$

There will be frequencies in this interval which

TABLE I. Fraction of frequencies in the intervals $[(j-1)\omega_L/10,\ j\omega_L/10]$.

ω/ω_L	0–0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
Fraction	0.5019	0.0038	0.0120	0.0504	0.0760	0.0183	0.0052	0.1719	0.0902	0.0703

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[2] K. Knopp, *Theory and Application of Infinite Series* (Blackie and Son, Ltd., London and Glasgow, 1951), p. 448.

correspond to the rational numbers

$$\frac{n}{n+1},\ \frac{n+1}{n+2},\ \cdots,\ \frac{2n}{2n+1},\ \frac{2n+1}{2n+2},\ \cdots,$$
$$\frac{3n}{3n+1},\ \frac{3n+1}{3n+2},\ \cdots,\ \text{etc.}$$

The expected fraction of frequencies is therefore equal to

$$(1-\tau)^2\{\tau^n+\tau^{n+1}+\tau^{n+2}+\cdots$$
$$+\tau^{2n}+\tau^{2n+2}+\tau^{2n+4}+\cdots+\tau^{3n}+\tau^{3n+3}+\cdots+\cdots\}$$
$$=(1-\tau)^2\left[\frac{\tau^n}{1-\tau}+\frac{\tau^{2n}}{1-\tau^2}+\cdots\right]\approx(1-\tau)\tau^n \quad (5)$$

for large n. Hence the desired density is given by

$$N(\omega)\approx(1-\tau)\exp\left\{\frac{\pi}{[8(1-\omega/\omega_L)]^{\frac{1}{2}}}\ln\tau\right\}. \quad (6)$$

From this result one can obtain some information about the behavior of $N(\omega)$ for a disordered chain with a finite ratio of masses. We first note that the moments are a linear combination, with positive coefficients, of the $\nu_n[=\tau/m^n+(1-\tau)/M^n]$.[1] The moments of a disordered chain with masses m, M and probability τ are thus bounded below by the moments of a disordered chain with masses m, ∞ and probability τ. But the moments are also bounded above by the moments of a disordered chain with masses m, ∞, and probability τ', where $\tau<\tau'<1$. This can be seen from the inequality

$$\frac{\tau}{m^n}+\frac{1-\tau}{M^n}=\frac{\tau}{m^n}\left[1+\frac{1-\tau}{\tau}\left(\frac{m}{M}\right)^n\right]$$
$$\leqslant\frac{\tau+(1-\tau)(m/M)}{m^n}. \quad (7)$$

The last term on the right is an expression for ν_n for a disordered lattice with masses m, ∞, and a probability $\tau'=\tau+(1-\tau)(m/M)$. Let us now consider the function $N(\omega)$ for a disordered chain with masses m and M and probability τ and compare it with $N_\infty'(\omega)$ for a chain with masses m and ∞ and probability τ'. It is shown in the Appendix that it is impossible for $N(\omega)$ to be strictly greater than or equal to $N_\infty'(\omega)$ in any neighborhood of $\omega=\omega_L$. Similarly it cannot be strictly less than or equal to $N_\infty(\omega)$. Therefore it behaves qualitatively like $N_\infty'(\omega)$ about $\omega=\omega_L$.

3. DETERMINATION OF THE SPECTRUM FROM THE MOMENTS

If all the moments of a spectrum are known, it is clear that the characteristic function can be written down, and hence the spectrum derived exactly. If only a finite number of moments is known, we can nevertheless derive approximations to the spectrum which will improve steadily with the number of exactly determined moments. We shall be concerned with a chain in which any atom can have mass m or M $(M>m)$, these values occurring randomly with frequencies τ, $1-\tau$, respectively. Then ν_s, the sth moment of the M^{-1} distribution will be equal to

$$\nu_s=\frac{\tau}{m^s}+\frac{1-\tau}{M^s}=\frac{\tau}{m^s}(1+\beta\xi^s);$$
$$\beta=(1-\tau)/\tau,\quad \xi=m/M. \quad (8)$$

By Rayleigh's theorem[3] no frequency of the random chain can be greater than the highest frequency of a homogeneous chain of light atoms m. Since we consider a lattice with only masses chosen at random, the maximum frequency will be equal to that of a lattice containing light masses only with probability τ^N; hence the frequency spectrum is nonzero in at most a finite region $(0,\omega_L)$ where ω_L is the maximum frequency of a lattice with light atoms only.[4,5] The results of the last section suggest that the high-frequency end is approached by an exponential tail in the spectrum.

A considerable simplification in the determination of the spectrum arises when it is known that the spectrum exists only for a finite range of frequencies. The asymptotic values of the moments then determine the behavior of the spectrum at its upper end. Thus if $\mu_n\sim\omega_L{}^n f(n)$, where $\lim_{n\to\infty}[f(n)]^{1/n}=1$, then the upper limit of the spectrum is ω_L and $f(n)$ determines the form of the spectrum near $\omega=\omega_L$ in certain cases; for example, if $f(n)\sim 1/n^k$, then $g(\omega)\sim A(1-\omega/\omega_L)^{k-1}$. This result is derived in the Appendix, and it is shown how a detailed asymptotic expansion of μ_n for large n enables us to obtain a corresponding expansion of $g(\omega)$ near $\omega=\omega_L$.

In what follows, it will be more convenient to discuss the dimensionless distribution functions,

$$f(x)=\omega_L g(\omega_L x),\quad F(x)=\omega_L{}^2G(\omega_L{}^2x), \quad (9)$$

where $G(\omega^2)$ is the distribution function for the squares of the normal mode frequencies, and is related to $g(\omega)$ by $g(\omega)=2\omega G(\omega^2)$. The even moments of the frequency distribution function $g(\omega)$ are given in terms of these new functions by

$$u_{2k}=\mu_{2k}/\omega_L{}^{2k}=\int_0^1 x^{2k}f(x)dx$$
$$=\int_0^1 x^kF(x)dx. \quad (10)$$

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[3] Lord Rayleigh, *The Theory of Sound* (Dover Publications, New York, 1945), second edition, Chap. 4.

[4] G. H. Weiss and A. A. Maradudin, J. Phys. Chem. Solids **7**, 327 (1958).

[5] Maradudin, Mazur, Montroll, and Weiss, Revs. Modern Phys. **30**, 175 (1958).

We restrict ourselves to the consideration of even moments only since it is these which are obtained by the moment-trace method. In order to calculate the spectrum using these moments, the function $g(\omega)$ is extended symmetrically from $-\omega_L$ to 0; all odd moments over this extended range are then zero.

It is easy to transfer the origin to $x=1$ and calculate the transformed moments v_{2k}. We have

$$v_{2k}=\int_0^1 (1-x^2)^k f(x)dx$$

$$=\sum_{m=0}^{k}(-1)^m\binom{k}{m}u_{2m}. \tag{11}$$

As we shall see in Sec. 4, these transformed moments are intimately connected with the evaluation of the zero-point energy of a crystal lattice. Furthermore, we can use the above results to obtain the expansion of $f(x)$ near $x=0$.

To approximate to the spectrum over the whole range, two independent methods have been used. The first is to obtain the best type of polynomial approximation with the moments available, and this is achieved by the use of Legendre polynomials. The details of the method have been described previously,[5] and will therefore not be reproduced again here. Since even moments were available up to μ_{20}, Legendre polynomials up to the twentieth order were used. It is important in methods of this kind to maintain a sequence of approximations rather than to rely on one approximation of the highest order. By noticing the differences between successive approximations in the sequence it is possible to assess the reliability of the final estimate.

The second method, which to the best of our knowledge has not been used before, we have called the delta-function approximation. This method makes use of the formula

$$F(x)=\int_0^1 F(\xi)\delta(\xi-x)d\xi,\quad 0<x<1, \tag{12}$$

and the nth order approximation replaces $\delta(\xi-x)$ by a polynomial of order n, $\delta_n(\xi-x)$.

However, for the study of $f(x)$ about $x=0$ we need a one-sided delta-function, and we can use

$$\delta_n(x)=\frac{(2n+1)!}{2^{2n+1}(n!)^2}(1-x^2)^n=A_n(1-x^2)^n, \tag{13}$$

where the normalization constant has been chosen in such a way that

$$\int_{-1}^{1}\delta_n(x)dx=1. \tag{14}$$

For one-dimensional lattices the expansion of $f(x)$ for $x\sim 0$ is

$$f(x)=c_0+c_2x^2+c_4x^4+\cdots. \tag{15}$$

Using the delta-function method we can obtain values for the coefficients c_{2n} as follows. We have that

$$\int_0^1 \delta_n(x)f(x)dx$$

$$=A_n\int_0^1 f(x)(1-x^2)^n dx$$

$$=A_n\int_0^1 (1-x^2)^n[c_0+c_2x^2+c_4x^4+\cdots]dx, \tag{16}$$

i.e., that

$$2A_nv_{2n}=c_0+\frac{c_2}{(2n+3)}+\frac{3c_4}{(2n+3)(2n+5)}$$

$$+\frac{15c_6}{(2n+3)(2n+5)(2n+7)}+\cdots. \tag{17}$$

The coefficient c_0 has been determined exactly,[6] and we see that from the relation

$$(2n+3)[2A_nv_{2n}-c_0]$$

$$=c_2+\frac{3c_4}{(2n+5)}+\frac{15c_6}{(2n+5)(2n+7)}+\cdots \tag{18}$$

that the coefficients c_2, c_4, $\cdots$ can be obtained with reasonable accuracy provided that a large number of moments are available. An explicit expression for the c_{2n} can be derived in terms of limits and differences of limits of the quantities A_nv_{2n} as in Theorems 4 and 5 of the Appendix. If the functional form of $f(x)$ for $x\sim 1$ is known, similar methods using the u_{2n} can give us precise information about the spectrum in this region.

At an intermediate point of the spectrum we need a two-sided delta-function, and we have used

$$\delta(\xi-x)=A_n\left[1-\left(\frac{\xi-x}{1-x}\right)^2\right]^n,\quad x\leqslant\tfrac{1}{2},$$

$$=B_n\left[1-\left(\frac{\xi-x}{x}\right)^2\right]^n,\quad x\geqslant\tfrac{1}{2}, \tag{19}$$

where A_n and B_n are given by

$$A_n=\frac{1}{1-x}\frac{(2n+1)!}{2^{2n+1}(n!)^2},$$

$$B_n=\frac{1}{x}\frac{(2n+1)!}{2^{2n+1}(n!)^2}, \tag{20}$$

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[6] A. A. Maradudin and G. H. Weiss, J. Chem. Phys. **29**, 631 (1958).

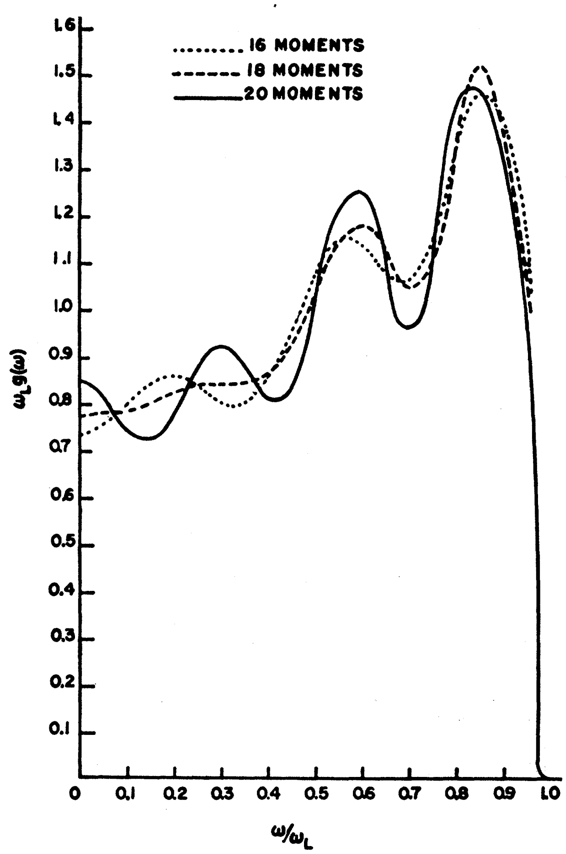

FIG. 1. This figure shows successive approximations to the frequency spectrum of a randomly disordered two-component linear chain in which the ratio of masses $M/m=2$, and the fraction of light masses $\tau=0.5$. These were obtained by the Legendre polynomial method using 16, 18, and 20 moments respectively.

respectively. This function overlaps the interval (0,1) but this does not affect our results, since there is no contribution from outside this interval. Clearly the nth approximation, $F_n(x)$, can be expressed in terms of moments of up to order $2n$ for an end point and of up to order $4n$ for an interior point of the spectrum. We can similarly obtain estimates of $F'(x)$ and $F''(x)$ at any point x by using $\delta'(\xi-x)$, $\delta''(\xi-x)$. The spectrum and its properties can then be calculated point by point, the accuracy obtainable depending on the number of moments available.

The two methods of approximation seem to be roughly complementary. The Legendre polynomial method provides an approximation which oscillates above and below the correct spectrum, and hence tends to exaggerate the maxima and minima and oscillations of this spectrum; spurious oscillations can usually be separated from genuine maxima and minima since they are not consistent in successive approximations (see for example Fig. 1). On the other hand, the delta-function method produces an averaged spectrum with a breadth of averaging which is determined by the order of the approximation, becoming finer as n increases; it therefore tends to smooth out the oscillations of the correct spectrum. Both methods (and more particularly the Legendre polynomial method) are highly sensitive to the correctness of the moments; several small errors in the initial derivation of the expression for the twentieth moment were detected by the obvious incorrectness of the corresponding approximations to the spectrum.

The delta-function method has the advantage of great simplicity, and ready application without excessive computational difficulty to moments of a high order; it provides approximations to a positive function which are always positive. The Legendre polynomial method is less satisfactory to use with moments of high order since a very high degree of accuracy is required in computing the coefficients of high order Legendre polynomials, and this leads to heavy computational work. In the present application we have used the delta function method largely at the ends of the spectrum, as the Legendre polynomials seem to provide a better estimate in the interior frequency range. If more moments were available it is likely that the delta-function method could have wider application; this might provide an alternative method for accurate calculations of spectra of homogeneous lattices, where moment calculations can be carried out quite easily.

4. ZERO-POINT ENERGY OF DISORDERED LATTICES

The zero-point energy of a crystal lattice is given by

$$E_0=\tfrac{1}{2}\hbar \textstyle\sum_j \omega_j, \tag{21}$$

where the summation is over all the normal mode frequencies of the lattice. Since $g(\omega)d\omega$ is defined as the *fraction* of normal modes with frequencies in the interval $(\omega, \omega+d\omega)$, we can replace the sum in Eq. (21) by an integral to obtain the zero-point energy per particle as

$$\begin{aligned} E_0/N &= \tfrac{1}{2}\hbar \int_0^{\omega_L} \omega g(\omega)d\omega \\ &= \tfrac{1}{2}\hbar\omega_L \int_0^1 x f(x)dx, \end{aligned} \tag{22}$$

where N is the number of particles in the lattice. Following the method used by Domb and Salter[7] for homogeneous lattices we rewrite Eq. (22) as

$$\begin{aligned} E_0/N &= \tfrac{1}{2}\hbar\omega_L \int_0^1 [1-(1-x^2)]^{\frac{1}{2}} f(x)dx \\ &= \tfrac{1}{2}\hbar\omega_L \sum_{n=0}^{\infty} (-1)^n \binom{\frac{1}{2}}{n} \int_0^1 f(x)(1-x^2)^n dx \\ &= \tfrac{1}{2}\hbar\omega_L \sum_{n=0}^{\infty} (-1)^n \binom{\frac{1}{2}}{n} v_{2n}. \end{aligned} \tag{23}$$

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[7] C. Domb and L. Salter, Phil. Mag. 43, 1083 (1952).

Since in the present case we have only the moments up to μ_{20} at our disposal, it is necessary to estimate the contribution to E_0/N from the sum

$$S=\sum_{n=11}^{\infty}(-1)^n\binom{\frac{1}{2}}{n}v_{2n}. \tag{24}$$

The method for obtaining this correction is quite general and can be applied to homogeneous as well as to disordered lattices.

We begin by obtaining the asymptotic form of v_{2n} for large n. Since we know that about $x=0$, $f(x)$ can be expanded as

$$f(x)=c_0+c_2x^2+c_4x^4+\cdots, \tag{25}$$

and since the v_{2n} are moments of the frequency distribution function about the origin, then with the aid of Lemma 1 of the Appendix we can assert that the large-n behavior of v_{2n} is given by

$$\begin{aligned}v_{2n}&\sim\int_0^1(1-x^2)^n(c_0+c_2x^2+c_4x^4+\cdots)dx\\&=\frac{c_0}{2}\frac{\Gamma(\frac{1}{2})\Gamma(n+1)}{\Gamma(n+\frac{3}{2})}+\frac{c_2}{2}\frac{\Gamma(\frac{3}{2})\Gamma(n+1)}{\Gamma(n+\frac{5}{2})}\\&\qquad+\frac{c_4}{2}\frac{\Gamma(\frac{5}{2})\Gamma(n+1)}{\Gamma(n+\frac{7}{2})}+\cdots.\end{aligned} \tag{26}$$

If this result is substituted into Eq. (24), we obtain

$$\begin{aligned}S=\frac{c_0\pi}{4}\sum_{n=11}^{\infty}\frac{(-1)^n}{\Gamma(\frac{3}{2}+n)\Gamma(\frac{3}{2}-n)}+\frac{c_2\pi}{8}\sum_{n=11}^{\infty}\frac{(-1)^n}{\Gamma(\frac{5}{2}+n)\Gamma(\frac{3}{2}-n)}\\+\frac{3c_4\pi}{16}\sum_{n=11}^{\infty}\frac{(-1)^n}{\Gamma(\frac{7}{2}+n)\Gamma(\frac{3}{2}-n)}+\cdots,\end{aligned} \tag{27}$$

where we have used the explicit expression for the binomial coefficient $\binom{\frac{1}{2}}{n}$. With the aid of the relation

$$\Gamma(z)\Gamma(1-z)=\pi/\sin\pi z,$$

the sums in Eq. (27) are transformed into

$$\begin{aligned}S=-\frac{c_0}{4}\sum_{n=11}^{\infty}\frac{1}{(n+\frac{1}{2})(n-\frac{1}{2})}-\frac{c_2}{8}\sum_{n=11}^{\infty}\frac{1}{(n+\frac{3}{2})(n+\frac{1}{2})(n-\frac{1}{2})}\\-\frac{3c_4}{16}\sum_{n=11}^{\infty}\frac{1}{(n+\frac{5}{2})(n+\frac{3}{2})(n+\frac{1}{2})(n-\frac{1}{2})}-\cdots.\end{aligned} \tag{28}$$

By breaking them up by partial fractions, we can evaluate the sums immediately with the result that

$$S=-\frac{c_0}{2\times21}-\frac{c_2}{4\times21\times23}-\frac{c_4}{2\times21\times23\times25}-\cdots. \tag{29}$$

Our expression for the zero-point energy per particle becomes finally

$$\begin{aligned}\frac{E_0}{N}=\frac{\hbar\omega_L}{2}\Bigg\{\sum_{n=0}^{10}(-1)^n\binom{\frac{1}{2}}{n}v_{2n}-0.0238095c_0\\-0.0005176c_2-0.0000414c_4-\cdots\Bigg\}.\end{aligned} \tag{30}$$

In applying this result to the calculation of the zero-point energy of the disordered lattice the exact values of c_0, c_2, c_4, which were obtained by the methods of Sec. 3, were used.

The zero-point energy per particle of the disordered lattice with the ratio M/m infinite cannot be evaluated in this manner since the correction terms become infinite. However, from our analysis of the frequency spectrum for this case, we know the value of every normal mode frequency and the probability of its occurrence, so that we may evaluate the zero-point energy by direct summation. The sum converges rapidly, and good accuracy can be achieved in this way.

In order to compare the zero-point energy per particle of the disordered lattice with the zero-point energy per particle of the ordered ($ABABAB\cdots$) lattice and the lattice in which the isotopic constituents have separated into two phases, the latter two energies were computed.

The zero-point energy per particle of an alternating diatomic chain with masses M and m has been obtained by Mazur, Montroll, and Potts[8] for a chain with fixed ends. In the present case, where we have assumed cyclic boundary conditions, their analysis leads to the following expression

$$\begin{aligned}\frac{E_0{}^{\text{ordered}}}{N}=\frac{\hbar\omega_L}{\pi}\left[\frac{1}{2}\left(1+\frac{m}{M}\right)\right]^{\frac{1}{2}}\frac{(M/m)^{\frac{1}{4}}+(m/M)^{\frac{1}{4}}}{[(M/m)^{\frac{1}{2}}+(m/M)^{\frac{1}{2}}]^{\frac{1}{2}}}\\\times E\left(\frac{\pi}{4},\frac{2}{(M/m)^{\frac{1}{4}}+(m/M)^{\frac{1}{4}}}\right),\end{aligned} \tag{31}$$

where $E(\varphi,k)$ is an incomplete elliptic integral of the second kind.

The zero-point energy for the separated phases is computed from the fact that the zero-point energy per particle of a monatomic linear chain with masses M and force constants α is $(\hbar/\pi)(4\alpha/M)^{\frac{1}{2}}$. The zero-point energy per particle for the separated phases is then given by

$$\frac{E_0{}^{\text{sep}}}{N}=\frac{\hbar}{\pi}\left[\tau\left(\frac{4\alpha}{m}\right)^{\frac{1}{2}}+(1-\tau)\left(\frac{4\alpha}{M}\right)^{\frac{1}{2}}\right]. \tag{32}$$

If we normalize this expression to give the zero-point energy in terms of the maximum frequency of the

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[8] Mazur, Montroll, and Potts, J. Wash. Acad. Sci. **46**, 2 (1956).

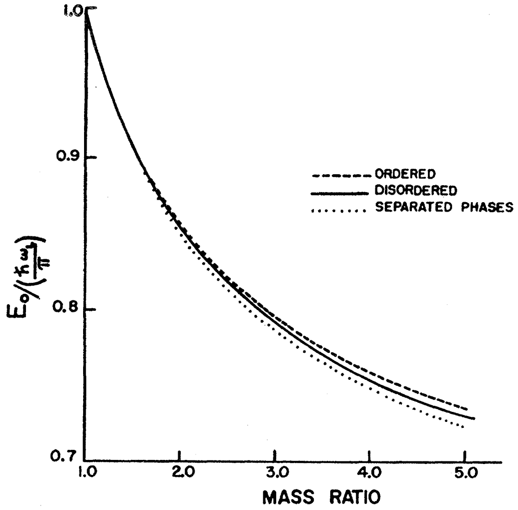

FIG. 2. The zero-point energy for an ordered ($ABAB\cdots$) diatomic linear chain, for a randomly disordered two-component chain with $\tau=0.5$, and for a diatomic chain in which the components have separated into two phases ($\cdots AAABBB\cdots$), plotted as a function of the mass ratio M/m.

disordered lattice, we obtain

$$\frac{E_0^{\text{sep}}}{N}=\frac{\hbar\omega_L}{\pi}\left[1-\tau+\tau\left(\frac{m}{M}\right)^{\frac{1}{2}}\right]. \qquad (33)$$

By using Eqs. (30), (31), and (33), the zero-point energies of the ordered and disordered lattices and the separated phases were calculated as a function of the mass ratio for $\tau=\frac{1}{2}$. The results are given in Fig. 2. For all mass ratios the ordered lattice has the highest zero-point energy, while the separated phases have the lowest zero-point energy. This result is in agreement with results obtained earlier and by different methods by several investigators.[4,5,9]

5. PROPERTIES OF THE SPECTRA

Even moments of the spectrum up to μ_{20} were evaluated from the expressions given in reference 1 on the IBM 704 computer at the National Bureau of Standards for $M/m=1.1$, 0.5, 2.0, 3.0, 5.0 and for $\tau=0.1$, 0.25, 0.50, 0.75, and 0.90. Corresponding approximations to the spectra using Legendre polynomials were also computed with 16, 18, and 20 moments. We found it necessary to do our calculations in double precision (i.e., keeping twenty decimal places), since in the Legendre polynomial calculations many of the terms arise as small differences of large numbers, thus necessitating increased accuracy. A typical set of estimates of the spectrum is shown in Fig. 1 for $\tau=0.5$, $M/m=2$. The advantage of maintaining the series of approximations referred to in Sec. 3 will be clearly seen. At long wavelengths the small oscillations occurring in successive approximations are not consistently maintained, and have the appearance of spurious oscillations introduced by the approximation. On the other hand, the major peaks are consistent in successive approximations, and we feel confident that they are not spurious.

The Legendre polynomial approximation thus seems to be rather unsatisfactory in the long-wave region of the spectrum. Fortunately an alternative and independent source of information is available for this region. Clearly the forces in any given region are independent of the masses, and hence the stress across any given area, and the corresponding stress tensor, remain the same as the masses change. Hence for a large region containing different masses the equations of motion are of the form[10]

$$\iiint \rho(x,y,z)\ddot{u}(x,y,z)dxdydz=\iint X_\nu dS, \qquad (34)$$

where $\rho(x,y,z)$ is the density of the body at the point (x,y,z), $u(x,y,z)$ is the displacement of the point (x,y,z), and X_ν is the x component of the traction across the bounding surface of this region. Now it is this macro-

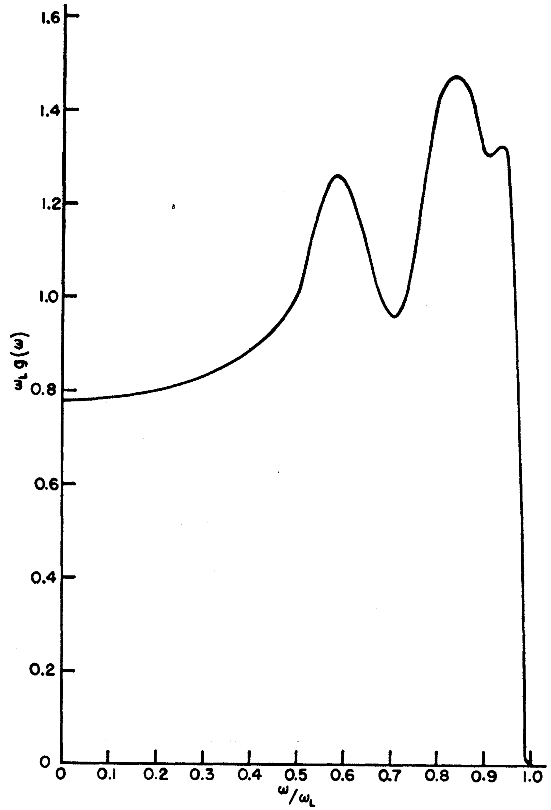

FIG. 3. The final estimate for the frequency spectrum of a randomly disordered two-component chain with $M/m=2$ and $\tau=0.5$.

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[9] Prigogine, Bingen, and Jeener, Physica **20**, 383, 516 (1954).

[10] A. E. H. Love, *A. Treatise on the Mathematical Theory of Elasticity* (Dover Publications, Inc., New York, 1944), p. 76.

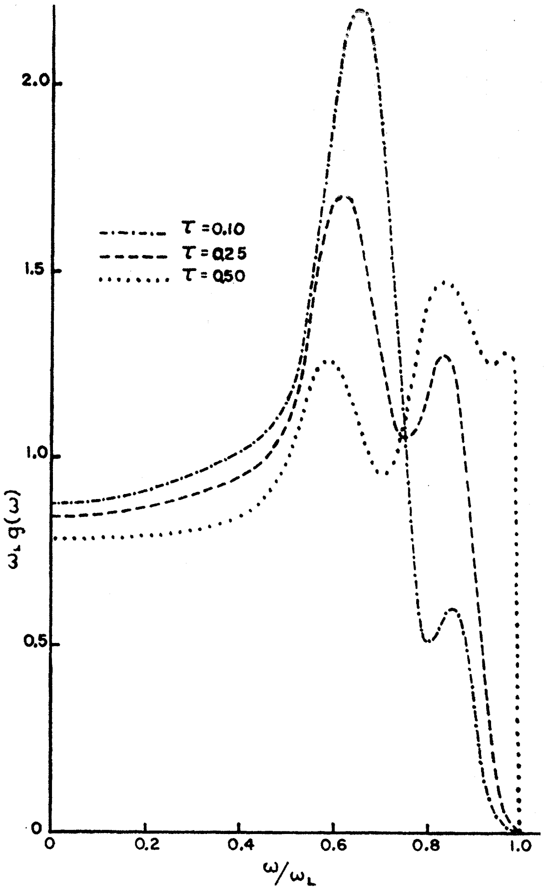

FIG. 4. The frequency spectra of randomly disordered two-component chains for the cases $M=2m$, and $\tau=0.1$, 0.25, and 0.50.

scopic equation which determines the elastic constants, and hence the long-wave behavior of the spectrum; and if the region concerned contains a large number of lattice points, we can neglect density fluctuations and replace $\rho(x,y,z)$ by the mean density. We should thus expect the long-wave behavior to correspond to that of a monatomic lattice of mean mass.

This result can be established independently for the linear chain by a perturbation method.[4] In fact, the latter method provides a long-wavelength expansion for the disordered lattice of the form

$$g(\omega)=C_0+C_2\omega^2+C_4\omega^4+\cdots, \tag{35}$$

where

$$\begin{aligned} C_0&=\frac{1}{\pi}\left(\frac{\bar{M}}{\gamma}\right)^{\frac{1}{2}},\\ C_2&=\frac{1}{8\pi}\left(\frac{\bar{M}}{\gamma}\right)^{\frac{3}{2}}\left[1-\beta_3\mu^3+\frac{15}{16}\beta_2^2\mu^4+O(\mu^5)\right],\\ C_4&=\frac{1}{32\pi}\left(\frac{\bar{M}}{\gamma}\right)^{\frac{5}{2}}\left[\frac{3}{4}-\frac{5}{2}\beta_3\mu^3-\frac{15}{32}\beta_2^2\mu^4+O(\mu^5)\right], \end{aligned} \tag{36}$$

with

$$\begin{gathered} \beta_2=\tau(1-\tau),\quad \beta_3=\tau(1-\tau)(2\tau-1),\quad \mu=\Delta M/\bar{M},\\ \Delta M=M-m,\quad \bar{M}=\tau m+(1-\tau)M. \end{gathered} \tag{37}$$

We have used this method to fill in the long-wavelength part of the spectrum. We also tried the delta-function method for this region of the spectrum and found that the resulting curve did not differ appreciably from that given by (35).

The curve in Fig. 3 represents our final estimate of the spectrum for $M/m=2$, $\tau=\frac{1}{2}$ and was obtained from the twentieth Legendre polynomial approximation at the upper end of the spectrum (which is terminated exponentially at the upper end in accordance with Sec. 2) and the methods of the previous paragraph at the lower end. There are indications (by comparison with corresponding Legendre polynomial approximations for the ordered $ABAB\cdots$ lattice) that the peaks in this spectrum do not represent infinite singularities.

In Figs. 4 and 5 we have plotted the frequency spectra for the case $M=2m$ as the concentration of particles of lighter mass m increases from 10% to 90%. For refer-

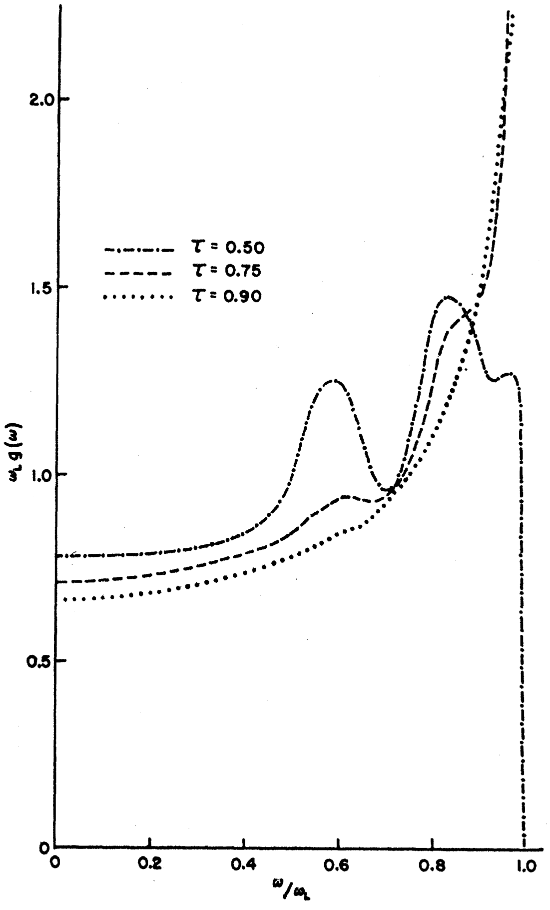

FIG. 5. The frequency spectra of randomly disordered two-component chains for the cases $M=2m$, and $\tau=0.5$, 0.75, 0.90.

ence, the spectrum corresponding to $\tau=0.50$ has been included in each figure. We note that for $\tau=0.10$ the spectrum does not differ qualitatively from the spectrum of a monatomic lattice containing only heavy particles in that it increases monotonically to a maximum at $\omega/\omega_L=0.65$ and then drops steeply. (The additional subsidiary maximum in this spectrum near the high-frequency end is believed to be associated with an impurity band and will be discussed below.) The corresponding monatomic lattice has a square root infinity at $\omega/\omega_L=0.7071$, and is zero beyond this point. As the concentration of light masses increases, the main peak decreases, while the small subsidiary peak increases until at $\tau=0.50$ it becomes the dominant peak. At the same time, a smaller peak begins to develop on the high-frequency side. As τ increases further, this new peak increases while the remaining two peaks decrease. At $\tau=0.90$ the spectrum is almost that of a monatomic lattice with light masses only. The spectra for both $\tau=0.75$ and $\tau=0.90$ eventually bend over and approach zero at $\omega/\omega_L=1$. It might be remarked that the spectrum of a lattice containing only 10% heavy atoms resembles the spectrum of the corresponding light atom monatomic lattice more closely than the spectrum of the lattice containing only 10% light masses resembles the spectrum of its corresponding heavy mass monatomic lattice. The reason for this is found in Rayleigh's theorem.[3] The addition of a small number of heavy masses to the light atom lattice will only depress the frequencies slightly, while the addition of a small number of light masses to a heavy mass lattice will shift the frequencies upward and can displace the uppermost frequencies into an impurity band.

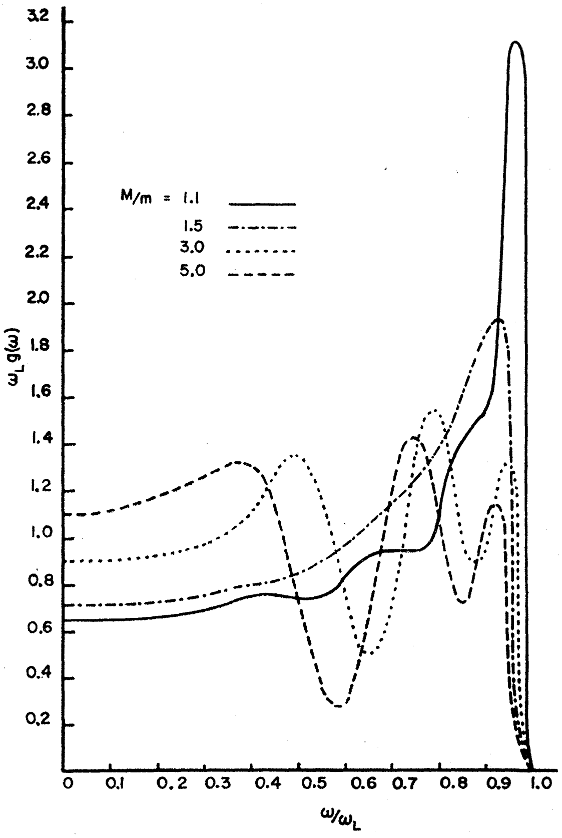

FIG. 6. The frequency spectra of randomly disordered two-component chains for the cases $\tau=0.5$, and $M/m=1.1$, 1.5, 3.0, 5.0.

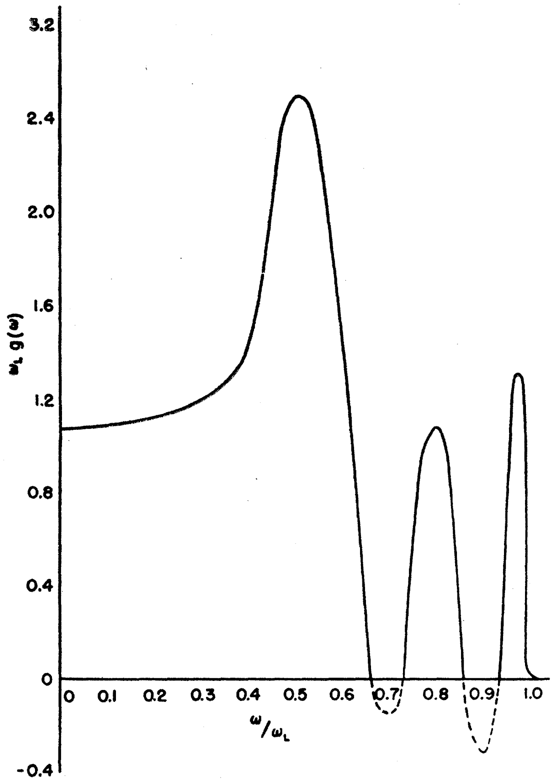

FIG. 7. The frequency spectrum for a randomly disordered two-component linear chain with $M/m=3$ and $\tau=0.1$. Note the two impurity bands.

In Fig. 6 we have plotted the frequency spectra for $\tau=0.50$ as the ratio M/m varies from 1.1 to 5. (The spectrum for the case $M/m=2$ is contained in Figs. 4 and 5.) It is seen that the change from a rather smooth spectrum containing no sharp peaks (except at $\omega\approx\omega_L$) to one containing several maxima takes place for values of the mass ratio approximately equal to two.

In Figs. 7 and 8, we have plotted the spectra for the cases $\tau=0.1$, $M/m=3$, and $\tau=0.1$, $M/m=5$, respectively. These represent the spectra of homogeneous lattices with a small impurity concentration. In each case the existence of two impurity bands is clearly suggested.

A quantitative explanation for the features of the spectra is very difficult to give at the present time. However, some qualitative remarks can be made which may aid in the interpretation of our results. In the case that the concentration of light atoms is small, as in the case of the spectra of Figs. 7 and 8, the main contribution to the frequency spectrum is associated with the spectrum of the monatomic lattice composed only of heavy masses. The maximum frequency for such a lattice is given by

$$\omega_{\max}/\omega_L=(m/M)^{\frac{1}{2}}, \tag{38}$$

which yields values of 0.577 and 0.447 in reasonable agreement with the values which mark the cutoff on the first band in Figs. 7 and 8.

The presence of the two impurity bands may perhaps be explained in the following way. From the work of Montroll and Potts[11] it is known that adding a single lighter-mass-isotope defect to an otherwise monatomic linear chain gives rise to a localized normal mode of vibration whose frequency lies above the band of allowed frequencies and is given by

$$\omega_{\mathrm{imp}}/\omega_L=(2-m/M)^{-\frac{1}{2}}. \tag{39}$$

The broadening of this frequency into an impurity band for small concentrations of defects has also been discussed,[5,12] but the approximation of the spectra by twentieth degree polynomials is undoubtedly too coarse to allow us to expect quantitative agreement between the observed and calculated impurity band widths. Substituting the values $(M/m)=3$ and $(M/m)=5$ into Eq. (39), we obtain values of 0.775 and 0.745, respectively. These values are in good quantitative agreement with the values of ω/ω_L at which the first impurity band occurs in the two cases illustrated in Figs. 7 and 8.

The second impurity band is believed to be associated with the replacement of a heavy atom by a light atom in an ordered portion of the chain. In the limit of very long chains we can expect to find "islands" where the atoms are arranged in an ordered array, with the ratio of the number of consecutive light atoms to the number of consecutive heavy atoms equal to the ratio $\tau/(1-\tau)$. Then, if in such an island one of the heavy atoms is replaced by a light atom, the normal mode associated with such a defect will have a frequency which will lie above the maximum frequency of the ordered lattice. The value of this frequency has been calculated only in the case of an alternating diatomic lattice $(ABAB\cdots)$, with the result that[5]

$$\frac{\omega_{\mathrm{imp}}}{\omega_L}=\left\{\frac{1}{4}+\frac{1}{2(2-m/M)}\left[1+\left(2-\frac{3m}{M}+\frac{5}{4}\frac{m^2}{M^2}\right)^{\frac{1}{2}}\right]\right\}^{\frac{1}{2}}. \tag{40}$$

If we substitute the values $M/m=3$ and 5 into Eq. (40), we obtain values of 0.933 and 0.929, respectively, in good quantitative agreement with the locations of the second impurity bands in Figs. 7 and 8. We also note the qualitative feature that the second impurity band in Fig. 7 occurs at a higher frequency than does the corresponding band in Fig. 8, in qualitative agreement with the calculated results. The agreement is probably fortuitous since with $\tau=0.10$ there is a greater probability for finding repeating units of nine heavy atoms and one light atom than for finding repeating units of one heavy atom and one light atom.

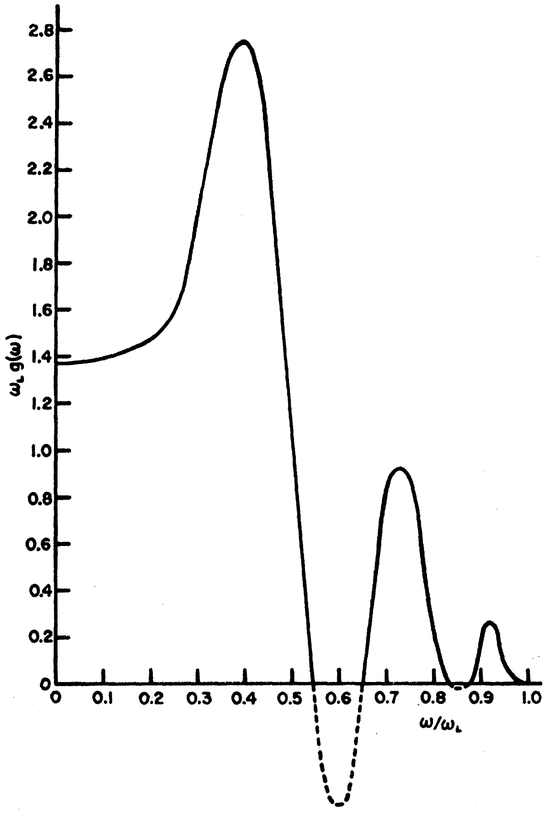

FIG. 8. The frequency spectrum for a randomly disordered two-component linear chain with $M/m=5$ and $\tau=0.1$. Note the two impurity bands.

In applying this analysis to the spectrum for the case $M/m=2$, $\tau=0.1$, which is illustrated in Fig. 4, we find that it predicts impurity bands at $\omega/\omega_L\approx0.816$ and $\omega/\omega_L\approx0.940$, respectively. However, only one such band is observed, at $\omega/\omega_L=0.87$. It is suggested that this band is associated with the first of the proposed mechanisms, i.e., it is the impurity band due to the presence of a small number of light isotope defects in an otherwise monatomic lattice. It is possible that a recalculation of this spectrum using higher moments than available at present would reveal a second impurity band at a higher frequency. At present we must assume that the mass difference is not sufficiently large for a second isolated band to appear.

For values of $\tau\approx0.5$ the analysis of the spectra is more difficult since peaks can now arise from more complicated sources, and a detailed discussion of Figs. 4, 5, and 6 will not be given.

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[11] E. W. Montroll and R. B. Potts, Phys. Rev. **100**, 525 (1955).

[12] Montroll, Maradudin, and Weiss, *Proceedings of the Stevens Institute Conference on the Many-Body Problem* (Interscience Publishers, Inc., New York, 1958).

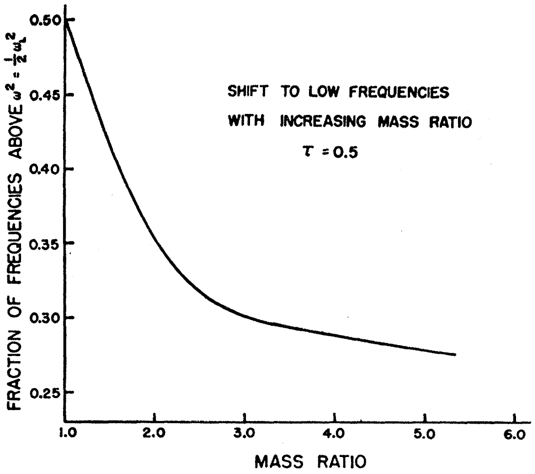

FIG. 9. This figure illustrates Rayleigh's theorem that the addition of heavy masses to a system of vibrating springs and masses lowers the normal mode frequencies of the system. The fraction of normal mode frequencies satisfying $\omega^2 > \frac{1}{2}\omega_L^2$ as a function of the ratio of the masses of the two constituents of a randomly disordered linear chain is plotted.

In this case, it would appear to be more convenient to pick the ordered diatomic lattice as our base lattice, and to regard the disordering as taking place about this configuration. It is well known[5] that the ordered diatomic lattice has three square-root infinities: at the top of the acoustic band, at the bottom of the optical band, and at the high-frequency end of the spectrum. For the disordered chain we might expect that the forbidden band will be filled in and the infinities reduced to finite peaks. The frequencies at which these infinities occur, which we denote by ω_1, ω_2, and ω_3, respectively, are given in the present case by

$$\frac{\omega_1}{\omega_L}=\left(\frac{m}{2M}\right)^{\frac{1}{2}},\quad \frac{\omega_2}{\omega_L}=\left(\frac{1}{2}\right)^{\frac{1}{2}},\quad \frac{\omega_3}{\omega_L}=\left(\frac{1}{2}+\frac{m}{2M}\right)^{\frac{1}{2}}. \tag{41}$$

In addition, one might expect in this case an impurity band at a frequency given by Eq. (40).

However, the results shown in Figs. 4, 5, and 6 do not agree well with this interpretation, except for the positions of the high-frequency maxima in the cases $\tau=0.5$, $M/m=2$, 3, 5 which agree qualitatively with the values predicted by Eq. (40). It may very well be that the two infinities which occur at frequencies ω_2 and ω_3 in the ordered diatomic lattice coalesce into a finite peak in the disordered lattice. For additional information on this and other points, we must await the calculation of higher moments.

Finally, Fig. 9 has been drawn to illustrate Rayleigh's theorem[3] that the increase of any mass of a vibrating system decreases the normal mode frequencies. We have plotted the fraction of normal modes of a disordered chain whose frequencies satisfy $\omega^2 > \frac{1}{2}\omega_L^2$ as a function of the mass ratio M/m. This fraction equals $\frac{1}{2}$ for the homogeneous chain, but decreases rapidly for $M/m>1$ until it approaches the value for $M/m=\infty$.

ACKNOWLEDGMENTS

We should like to thank Mr. Andreas Schopf of the National Bureau of Standards for his expert programming assistance, and Mr. John Wood for help with the numerical calculations. We would also like to acknowledge a helpful communication from Dr. R. Sherman Lehman.

APPENDIX. ASYMPTOTIC VALUES OF THE MOMENTS

In the remarks that follow we assume that $\omega_L=1$, with only trivial modifications being necessary to translate our results for general ω_L. It is almost immediately evident that the asymptotic behavior of the moments is intimately related to the behavior of the frequency spectrum in the neighborhood of the maximum frequency. This follows from the fact that $\lim_{n\to\infty}\omega^n=0$ for $|\omega|<1$ and $=1$ for $\omega=1$. It is therefore not surprising that under quite general conditions one can discuss the behavior of the frequency spectrum at its upper end in terms of the asymptotic form of the moments. We shall discuss the relation between the form of the frequency spectrum at the high-frequency end and the asymptotic form for the moments in this Appendix.

Let the frequency distribution function be denoted by $g(\omega)$ and let us define a truncated distribution function $g_\epsilon(\omega)$ for $\epsilon>0$ by

$$\begin{aligned} g_\epsilon(\omega)&=0 && \text{for}\quad 0\leqslant\omega\leqslant 1-\epsilon\\ &=g(\omega) && \text{for}\quad 1-\epsilon<\omega\leqslant 1. \end{aligned} \tag{A1}$$

Denote the kth moment of $g(\omega)$ by μ_k and the kth moment of $g_\epsilon(\omega)$ by $\mu_k(\epsilon)$. Fundamental to our later remarks is the following obvious lemma:

Lemma 1.—The difference between the real and truncated moments is bounded in absolute value by a function which is $O[(1-\epsilon)^n]$.

Proof: We have the estimate

$$\mu_k-\mu_k(\epsilon)=\int_0^{1-\epsilon}x^k g(x)dx\leqslant(1-\epsilon)^k\int_0^1 g(x)dx. \tag{A2}$$

With the aid of this lemma we can discuss the sense in which the asymptotic form of the moments is determined by the behavior of $g(\omega)$ in an arbitrarily small neighborhood of $\omega=1$.

Theorem 1. Let $f(\omega)\geqslant 0$ be such that

1. $\int_0^1 f(\omega)d\omega$ exists[13] and has moments

$$\zeta_k=\int_0^1\omega^k f(\omega)d\omega.$$

2. $g(\omega)=f(\omega)+\theta(\omega)$ where for any $\delta>0$ there exists an ϵ such that $|\theta(\omega)|<\delta$ when $|1-\omega|<\epsilon$.

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[13] The existence of $\int_0^1 f(\omega)d\omega$ is enough to insure the existence of ζ_k.

3. For any $\epsilon>0$ the truncated moments $\zeta_k(\epsilon)$ go to zero more slowly than $(1-\epsilon)^k$, i.e., $\lim_{k\to\infty}(1-\epsilon)^k/\zeta_k(\epsilon)=0$.

4. The difference function $\theta(\omega)$ satisfies $\lim_{\omega\to 1}|\theta(\omega)|/|f(\omega)|=0$. Then

$$\lim_{k\to\infty}(\mu_k/\zeta_k)=1.$$

Hypothesis 3 insures that $f(\omega)$ does not go to zero too quickly. It is clear that if $f(\omega)$ approaches zero too rapidly we would not be justified in neglecting contributions from the interval $(0, 1-\epsilon)$ as is done in the following proof.

Proof: We may write

$$\frac{\mu_k}{\zeta_k}=\frac{\mu_k-\mu_k(\epsilon)+\mu_k(\epsilon)}{\zeta_k-\zeta_k(\epsilon)+\zeta_k(\epsilon)}. \tag{A3}$$

By the proof of Lemma 1 we have the property $|\zeta_k-\zeta_k(\epsilon)|$, $|\mu_k-\mu_k(\epsilon)|\leqslant O[(1-\epsilon)^k]$; hence, using hypothesis 3 we find

$$\lim_{k\to\infty}\frac{\mu_k(\epsilon)}{\zeta_k(\epsilon)}=\frac{\mu_k}{\zeta_k}. \tag{A4}$$

We can also write

$$\frac{\mu_k(\epsilon)}{\zeta_k(\epsilon)}=1+\int_{1-\epsilon}^{1}\omega^k\theta(\omega)d\omega\bigg/\int_{1-\epsilon}^{1}\omega^k f(\omega)d\omega. \tag{A5}$$

By condition 4 the last ratio must tend to zero as ϵ approaches zero, thus proving the theorem.

If, as $\omega\to 1$, $g(\omega)\to A(1-\omega^2)^{-\alpha}$ sufficiently rapidly, where $\alpha\neq 1, 2, 3, \cdots$, then we may assert that $\mu_k\sim A\Gamma(1-\alpha)2^{-\alpha}k^{\alpha-1}$. If, as $\omega\to 1$, $g(\omega)\to A\ln(1-\omega)^{-1}$ sufficiently rapidly, then $\mu_k\sim A(\ln k/k)$.

The question now arises as to the conditions under which it is possible to reverse the property of Theorem 1, i.e., if it is known that $\mu_k\sim\zeta_k$ as $k\to\infty$ where the ζ_k are the moments of some function $f(\omega)$ can we assert that $g(\omega)\to f(\omega)$ as $\omega\to 1$? It is indeed true that under a wide set of conditions the asymptotic form of the moments determines the behavior of $g(\omega)$ in a neighborhood of $\omega=1$ as shown by Theorem 2.

Theorem 2. Let

$$\mu_k\sim\sum_{n=1}^{\infty}a_nk^{-s_n}\quad\text{for}\quad k\to\infty$$

where $0<s_1<s_2<s_3<\cdots$: If it is known that $g(\omega)$ has a monotonic behavior near $\omega=1$, then

$$g(\omega)\sim\sum_{n=1}^{\infty}a_n\frac{\ln^{s_n-1}\omega^{-1}}{\Gamma(s_n)} \tag{A6}$$

as $\omega\to 1$.

Proof: We may write

$$\mu_k=\int_0^1\omega^kg(\omega)d\omega=\int_0^\infty e^{-kt}g(e^{-t})e^{-t}dt$$
$$\sim\sum_{n=1}^{\infty}a_nk^{-s_n}\quad\text{for}\quad k\to\infty, \tag{A7}$$

or the kth moment can be written as the Laplace transform of the function $g(e^{-t})e^{-t}$. The hypotheses of this theorem allow us to use a Tauberian theorem[14] for the Laplace transform, to assert that

$$g(e^{-t})e^{-t}\sim\sum_{n=1}^{\infty}a_n\frac{t^{s_n-1}}{\Gamma(s_n)} \tag{A8}$$

near $t=0$ or

$$g(\omega)\sim\sum_{n=1}^{\infty}a_n\frac{\ln^{s_n-1}\omega^{-1}}{\Gamma(s_n)}. \tag{A9}$$

If the monotonicity properties of $g(\omega)$ are not known, then we may at least assert that

$$\int_\omega^1 g(\omega)d\omega\sim\sum_{n=1}^{\infty}a_n\frac{\ln^{s_n}\omega^{-1}}{\Gamma(s_n)}. \tag{A10}$$

Notice that when there is only a single term, say $\mu_k\sim Ak^{-\alpha}$, we may expand the logarithm in the vicinity of $\omega=1$ to find

$$g(\omega)\sim\frac{A(1-\omega)^{\alpha-1}}{\Gamma(\alpha)}+O[(1-\omega)^{\alpha-1}]. \tag{A11}$$

Another problem along these lines is: Knowing the form of $g(\omega)$ near $\omega=1$, how can we fit the relevant parameters from the asymptotic form of the moments? As an example of how this question might be answered, we have theorems 3 and 4.

Theorem 3: If $g(\omega)$ can be expanded around $\omega=1$ in a series of the form

$$g(\omega)=\sum_{n=0}^{\infty}a_n(1-\omega)^n,$$

then, in terms of the moments μ_k,

$$a_n=(-1)^n\lim_{k\to\infty}\frac{1}{(n!)^2}(k+2n+1)(k+2n+2)$$
$$\times\Delta\{(k+2n-1)(k+2n)$$
$$\times\Delta\{(k+2n-3)(k+2n-2)\Delta\cdots$$
$$\times\Delta\{(k+1)\mu_k\}\cdots\}, \tag{A12}$$

$$a_0=\lim_{k\to\infty}[(k+1)\mu_k],$$

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[14] G. Doetsch, *Theorie und Anwendung der Laplace Transfortioman* (Dover Publications, New York, 1945).

where

$$\Delta f_k = f_{k+1} - f_k.$$

Proof: The moments, μ_k are given by

$$\begin{aligned}\mu_k &= \sum_{n=0}^{\infty} a_n \int_0^1 \omega^k(1-\omega)^n d\omega \\ &= k!\sum_{n=0}^{\infty} a_n \frac{\Gamma(n+1)}{\Gamma(n+k+2)} \\ &= \sum_{n=0}^{\infty} \frac{n!a_n}{(k+1)(k+2)\cdots(k+n+1)},\end{aligned} \tag{A13}$$

or, equivalently,

$$(k+1)\mu_k = a_0 + \frac{a_1}{k+2} + \frac{2!a_2}{(k+2)(k+3)} + \cdots. \tag{A14}$$

Hence we have

$$a_0 = \lim\nolimits_{k\to\infty}(k+1)\mu_k. \tag{A15}$$

If we take first differences and use the formulas

$$\begin{aligned}&\Delta\ \text{constant} = 0, \\ &\Delta\frac{1}{(k+2)(k+3)\cdots(k+n+1)} \\ &\qquad = -\frac{n}{(k+2)(k+3)\cdots(k+n+2)},\end{aligned} \tag{A16}$$

we find

$$\begin{aligned}\Delta\{(k+1)\mu_k\} &= -\frac{a_1}{(k+2)(k+3)} \\ &\quad - \frac{2\cdot 2!a_2}{(k+2)(k+3)(k+4)} - \cdots,\end{aligned} \tag{A17}$$

or

$$a_1 = -\lim_{k\to\infty}(k+2)(k+3)\Delta\{(k+1)\mu_k\}. \tag{A18}$$

If we continue in the same fashion we find, in general, the expression given by Eq. (A12).

Theorem 4 is proved in a similar manner.

Theorem 4: If $g(\omega)$ can be expanded around $\omega=1$ in a series of the form

$$g(\omega) = \sum_{n=0}^{\infty} a_n(1-\omega)^{(n-1)/2},$$

then

$$a_0 = \lim_{k\to\infty} \frac{1}{\sqrt{\pi}} \frac{\Gamma(k+\frac{3}{2})}{\Gamma(k+1)}\mu_k, \tag{A19}$$

$$\begin{aligned}a_n = \frac{(-1)^n 2^{n-1}}{n!\Gamma[(n+1)/2]} \lim_{k\to\infty} &\frac{\Gamma(k+\frac{3}{2}(n+2))}{\Gamma(k+\frac{3}{2}(n+1))} \\ &\times\Delta\left\{\frac{\Gamma(k+\frac{3}{2}(n+1))}{\Gamma(k+\frac{3}{2}n)}\Delta\cdots\right. \\ &\left.\times\Delta\left\{\frac{\Gamma(k+\frac{3}{2})}{\Gamma(k+1)}\mu_k\right\}\cdots\right\}.\end{aligned} \tag{A20}$$

Since we are dealing with the limit of k tending to infinity we may, for all practical purposes, write $(k+m)(k+m+1)\sim k^2$ and $\Delta\to d/dk$. In this notation for the case of Theorem 3 we have

$$a_n \sim \frac{(-1)^n}{(n!)^2} \lim_{k\to\infty} k^2 S^{n-1}\left\{\frac{d}{dk}k\mu_k\right\}, \tag{A21}$$

where the operator S is defined as $(d/dk)k^2$. Similarly Eq. (A20) can be written

$$a_n \sim \frac{(-1)^n 2^{n-1}}{n!\Gamma[(n+1)/2]} \lim_{k\to\infty} k^{\frac{3}{2}} T^{n-1}\left\{\frac{d}{dk}k^{\frac{1}{2}}\mu_k\right\}, \tag{A22}$$

where the operator T is defined as $(d/dk)k^{\frac{3}{2}}$.

In Sec. 2 we have shown that the moments of the disordered chain with masses m and M and probability τ are less than or equal to those of a disordered chain with masses m and ∞ and probability τ'. Denoting the moments of $N_\infty(\omega)$ by μ_n and those of $N(\omega)$ by ν_n we have:

Theorem 6: Let $\mu_n \geqslant \nu_n$ with the inequality valid for $n>1$. Then $N_\infty(\omega)-N(\omega)$ cannot be strictly negative in any neighborhood of $\omega=1$.

Proof: Let $H(\omega)=N_\infty(\omega)-N(\omega)$. By hypothesis

$$\int_0^1 \omega^n H(\omega)d\omega \geqslant 0, \quad n=0,1,2,\cdots,$$

where we have re-expressed the moments by means of an integration by parts. Suppose $H(\omega)$ were strictly negative in a neighborhood $(1-\epsilon, 1)$ of 1. Then on some subset E of nonzero measure $m(E)$ which lies in $(1-\epsilon_1, 1)$ where $\epsilon_1<\epsilon$, we must have $H(\omega)<-\theta$ where $\theta>0$. We may therefore write

$$\int_0^{1-\epsilon} \omega^n H(\omega)d\omega + \int_E \omega^n H(\omega)d\omega + \int_{C(E)} \omega^n H(\omega)d\omega \geqslant 0,$$

where $C(E)$ denotes the complement of E with respect to $(1-\epsilon, 1)$. The first term is at most $O[(1-\epsilon)^n]$ and must be positive. The second term is negative and less than $-\theta(1-\epsilon_1)^n m(E)$, and the third term is negative. Consequently as $n\to\infty$ the second (or third term) dominates the sum; hence the sum cannot be positive as asserted. This proves the theorem.