Normal Modes of a Lattice of Oscillators with Many Resonances and Dipolar Coupling*

U. FANO
National Bureau of Standards, Washington, D. C.
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The normal modes of a lattice of coupled dipoles are studied as a model of the collective excitations of electrons in condensed materials. Two types of oscillations are found in which electrostatic coupling has a dominant influence. One of them is analogous to the oscillation of an electron plasma and has a high dipole moment. Other collective oscillations have a low net dipole moment, owing to destructive interference between out-of-phase components. These two types of oscillation occur in systems with a sufficiently high density of oscillator strength in space and in spectrum. A simple estimate indicates that most condensed materials fulfill this condition.

1. INTRODUCTION

MOST condensed materials are excited, by the passage of fast electrons, to "characteristic" energy levels. The valence electrons of distant atoms are understood to participate in these excitations collectively owing to the dominant influence of their Coulomb interaction.[1] The collective excitations have been treated theoretically, in the main, by means of the plasma model of metal electrons.[1,2] It has been pointed out[3] that the mechanism which yields collective excitations in a plasma should also operate in insulators, but it has been possible thus far to develop only sketchily a theory that would apply equally to all types of macroscopically homogeneous condensed materials.[4]

To complement the detailed results of the plasma model, which is somewhat unrealistic (especially for nonmetals), we consider in this paper another model, which is many ways complementary to the plasma model and which can also be worked out in detail. This model consists of a cubic lattice of identical electric dipole oscillators coupled by their electrostatic interaction. Whereas, in the plasma model, the Fermi gas of uncoupled electrons has a single band of excitation levels beginning at zero energy, we assume at each lattice site an unspecified number of isotropic oscillators with different proper frequencies ω_n.[5] No interaction is assumed among the oscillators at the same lattice site.[6]

Our objective will be to determine the normal modes of the whole lattice of coupled oscillators, i.e., its eigenfrequencies and eigenvectors. It is hoped the physico-mathematical properties of the special model considered in this paper will occur again in the eventual theory of the excitation of realistic types of condensed matter.

2. DESCRIPTION OF THE MODEL AND CALCULATION

Consider a cubic lattice of identical sites, 1, 2, $\cdots$, i, $\cdots$, with space coordinates $\mathbf{r}_i$. At each site there are a number of oscillators, each of which is identified by an index 1, 2 $\cdots$, n, $\cdots$. Each oscillator consists of a particle of mass m and charge $ef_n^{\frac{1}{2}}$, bound by an elastic force with force constant $k_n=m\omega_n^2$ to a particle of charge $-ef_n^{\frac{1}{2}}$ which is fixed at the lattice site. The number of sites in the lattice will be indicated by N, with the understanding that we always consider the limit $N\to\infty$. The coefficient f_n, which serves to express the charge of each oscillator in terms of the electron charge e, will be called the strength of the oscillator. We seek to determine the displacement $\mathbf{s}_{ni}$ of each oscillator particle from its equilibrium position when the whole lattice of dipoles oscillates in a normal mode.[7]

Because the lattice is invariant under translation by any lattice vector, the displacement $\mathbf{s}_{ni}$ must be, in a normal mode, a sinusoidal function of the lattice site position $\mathbf{r}_i$, characterized by a wave vector $\mathbf{k}$. We shall be interested in normal modes with a long wavelength, i.e., with a small $\mathbf{k}$. All oscillators will be assumed to be polarized in a single direction, indicated by a unit vector $\hat{p}$. (It will turn out that $\hat{p}$ is either parallel or perpendicular to $\mathbf{k}$, in a normal mode.) The displacement $\mathbf{s}_{ni}$ will then be represented by

$$\mathbf{s}_{ni}=a_n(t)\,\hat{p}\cos(\mathbf{k}\cdot\mathbf{r}_i+\varphi), \tag{1}$$

where φ is arbitrary and $a_n(t)$ has to be determined. The net dipole moment of the ith lattice site will then be

$$\mathbf{p}_i=\sum_n ef_n^{\frac{1}{2}}\mathbf{s}_{ni}=e\left[\sum_n f_n^{\frac{1}{2}}a_n(t)\right]\hat{p}\cos(\mathbf{k}\cdot\mathbf{r}_i+\varphi). \tag{2}$$

The kinetic energy of all the lattice oscillators is

$$K=\sum_{in}\tfrac{1}{2}m\dot{a}_n(t)^2\cos^2(\mathbf{k}\cdot\mathbf{r}_i+\varphi)=\tfrac{1}{4}Nm\sum_n\dot{a}_n^2, \tag{3}$$

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[1] See, e.g., L. Marton, L. B. Leder, and H. Mendlowitz, *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic Press, New York, 1955), Vol. 7, p. 183.

[2] See, e.g., D. Pines, *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1955), Vol. 1.

[3] N. F. Mott, Solvay Congress, Brussels, 1954 (unpublished) quoted in reference 2, p. 400.

[4] U. Fano, Phys. Rev. **103**, 1202 (1956).

[5] This model is quite analogous, in its main concept, to that of W. R. Heller and A. Marcus, Phys. Rev. 84, 809 (1951), but differs from it in its emphasis on the characteristics of many-frequency systems. In addition, Heller and Marcus overlooked that the polarization must obey the Eq. (13) of this paper.

[6] Such interactions may be regarded as having been removed by a previous reduction to normal modes.

[7] A classical treatment is sufficient for our purpose since the normal modes of coupled harmonic oscillators are determined by the same procedure in classical as in quantum mechanics.

and the potential energy of the elastic forces is

$$U=\sum_{in}\tfrac{1}{2}k_n a_n(t)^2\cos^2(\mathbf{k}\cdot\mathbf{r}_i+\varphi)=\tfrac{1}{4}Nm\sum_n\omega_n^2a_n^2. \quad (4)$$

The electrostatic interaction energy between all the dipoles at different sites is

$$V=\tfrac{1}{2}\sum_i\sum_{j\neq i}[\mathbf{p}_i\cdot\mathbf{p}_j-3\,\mathbf{p}_i\cdot\hat{r}_{ij}\,\mathbf{p}_j\cdot\hat{r}_{ij}]/r_{ij}^3$$
$$=\tfrac{1}{2}e^2[\sum_n f_n^{\frac{1}{2}}a_n(t)]^2\sum_i\sum_{j\neq i}\cos(\mathbf{k}\cdot\mathbf{r}_i+\varphi)$$
$$\times\cos(\mathbf{k}\cdot\mathbf{r}_j+\varphi)[1-3(\hat{p}\cdot\hat{r}_{ij})^2]/r_{ij}^3, \quad (5)$$

where $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$ and $\hat{r}_{ij}=\mathbf{r}_{ij}/r_{ij}$ is a unit vector. Consider that

$$\cos(\mathbf{k}\cdot\mathbf{r}_i+\varphi)\cos(\mathbf{k}\cdot\mathbf{r}_j+\varphi)$$
$$=\tfrac{1}{2}\{\cos[\mathbf{k}\cdot(\mathbf{r}_i+\mathbf{r}_j)+2\varphi]+\cos\mathbf{k}\cdot\mathbf{r}_{ij}\},$$

that the first term in these braces averages out upon summation over lattice sites, and that the residual $\sum_{j\neq i}$ has the same value for each lattice site i. Eq. (5) reduces, therefore, to

$$V=\tfrac{1}{8}Ne^2(\sum_n f_n^{\frac{1}{2}}a_n)^2\sum_{j\neq i}e^{i\mathbf{k}\cdot\mathbf{r}_{ij}}[1-3(\hat{p}\cdot\hat{r}_{ij})^2]/r_{ij}^3$$
$$+\text{compl. conj.} \quad (6)$$

The $\sum_{j\neq i}$ in (6) has been the subject of many studies, in recent years particularly by Cohen and Keffer[8] and also, for $\mathbf{k}\sim0$, in reference 5 and by Nijboer and De Wette.[9] Its limit for $\mathbf{k}=0$ depends on the angle between $\mathbf{k}$ and $\hat{p}$. We shall consider only the value of V for this limit, but reference 8 gives also data throughout the first Brillouin zone in $\mathbf{k}$ space. In the limit, Eq. (18) of reference 8 yields

$$V=N(\pi e^2/a^3)(\sum_n f_n^{\frac{1}{2}}a_n)^2[(\hat{p}\cdot\hat{k})^2-\tfrac{1}{3}], \quad (7)$$

where a indicates the cell edge in the cubic lattice and $\hat{k}=\mathbf{k}/k$. The average total energy of a lattice site is, then,

$$E=(K+U+V)/N=\tfrac{1}{4}m\sum_n\dot{a}_n^2+\tfrac{1}{4}m\sum_n\omega_n^2a_n^2$$
$$+\tfrac{1}{4}mC\omega_p^2(\sum_n f_n^{\frac{1}{2}}a_n)^2, \quad (8)$$

where

$$C=(\hat{p}\cdot\hat{k})^2-\tfrac{1}{3}, \quad (9)$$

and where

$$\omega_p^2=4\pi e^2/ma^3 \quad (10)$$

indicates the squared plasma frequency of a gas of a^{-3} particles of charge e and mass m per unit volume.

The amplitudes $a_n(t)$, the polarization $\hat{p}$, and the frequency Ω of each normal mode can be determined by requiring that the time integral of the Lagrangian $L=K-U-V$ be stationary with respect to variations of the $a_n(t)$ and of $\hat{p}$. (Ordinarily one would consider the variations of all Cartesian components of the vectors $a_n(t)\hat{p}$ as independent variables, but here the direction $\hat{p}$ is treated separately from $a_n(t)$ because it is common to all vectors $a_n(t)\hat{p}$ and because L depends on it only through the coefficient C.) Variation of the amplitudes $a_n(t)$ leads to the Euler-Lagrange equations

$$\frac{d}{dt}\frac{\partial L}{\partial\dot{a}_n}-\frac{\partial L}{\partial a_n}=0, \quad (11)$$

that is, to the system of coupled equations of motion

$$\ddot{a}_n=-\omega_n^2a_n-C\omega_p^2f_n^{\frac{1}{2}}\sum_m f_m^{\frac{1}{2}}a_m. \quad (12)$$

Variation of $\hat{p}$, subject to the condition $\hat{p}\cdot\hat{p}=1$, leads to the eigenvalue equation

$$\text{grad}_{\hat{p}}(C-\lambda\hat{p}\cdot\hat{p})=2(\hat{p}\cdot\hat{k})\hat{k}-2\lambda\hat{p}=0. \quad (13)$$

This equation has the eigenvalues $\lambda=1$ and 0 and eigenvectors $\hat{p}$ parallel or perpendicular to $\mathbf{k}$, corresponding to *longitudinal* (l) or *transverse* (t) normal modes, with

$$\hat{p}=\hat{p}_l=\hat{k}, \quad C=C_l=\tfrac{2}{3}, \quad (14a)$$

or

$$(\hat{p}\cdot\hat{k})=(\hat{p}_t\cdot\hat{k})=0, \quad C=C_t=-\tfrac{1}{3}. \quad (14b)$$

The solutions of (12) are of the form

$$a_n(t)=b[f_n^{\frac{1}{2}}/(\omega^2-\omega_n^2)]\cos(\omega t+\delta), \quad (15)$$

where b is a normalization constant and the frequency ω has to be determined by inserting (15) into (12). We obtain thus for ω the eigenvalue equation

$$1=C\omega_p^2\sum_n f_n/(\omega^2-\omega_n^2)=-C\alpha(\omega), \quad (16)$$

where

$$\alpha(\omega)=\omega_p^2\sum_n f_n/(\omega_n^2-\omega^2), \quad (17)$$

represents the polarizability of a unit volume of the oscillator lattice, computed without regard to the dipole interactions.

The eigenvalues of (16) will be indicated by Ω, and subscripts l_n or t_n will be added when necessary to specify the eigenvalue of the nth longitudinal or transverse mode. The normalization constant b may be expressed in terms of the mean energy per site E by substituting (15) into (8). This yields

$$E=\tfrac{1}{4}m\omega^2b^2\sum_n\frac{f_n}{(\omega^2-\omega_n^2)^2}=\tfrac{1}{4}mb^2\frac{\omega^2}{\omega_p^2}\frac{d\alpha}{d(\omega^2)}, \quad (18)$$

that is

$$b=\left(\frac{4E\omega_p^2}{m\omega^2d\alpha/d(\omega^2)}\right)^{\frac{1}{2}}. \quad (19)$$

Utilizing this result and the eigenvalue equation (16), we may express the dipole moment (2) of the ith lattice site, for a normal mode oscillation of frequency Ω_n and average site energy E, in the form

$$\mathbf{p}_i^{(n)}=e\left[\left(\frac{4E\omega_p^2}{m\omega^2d\alpha/d(\omega^2)}\right)^{\frac{1}{2}}\sum_{n'}\frac{f_{n'}}{\omega^2-\omega_{n'}^2}\right.$$
$$\left.\times\cos(\omega t+\delta)\right]_{\omega=\Omega_n}\hat{p}\cos(\mathbf{k}\cdot\mathbf{r}_i+\varphi)$$
$$=eF_n^{\frac{1}{2}}(4E/m\Omega_n^2)^{\frac{1}{2}}\hat{p}\cos(\mathbf{k}\cdot\mathbf{r}_i+\varphi)\cos(\Omega_nt+\delta), \quad (20)$$

[8] M. H. Cohen and F. Keffer, Phys. Rev. **99**, 1128 (1955). The factor ρ in Eq. (18) of this reference is due to a misprint and should be deleted.

[9] B. R. A. Nijboer and F. W. De Wette, Physica **24**, 422 (1958).

where

$$F_n=\left[C^2\frac{d\alpha}{d(\omega^2/\omega_p{}^2)}\right]^{-1}_{\omega=\Omega_n}$$
$$=\left[C^2\omega_p{}^4\sum_{n'}\frac{f_{n'}}{(\Omega_n{}^2-\omega_{n'}{}^2)^2}\right]^{-1}. \quad (21)$$

The same dipole moment would arise if there were at the ith site a single oscillating particle with mass m, charge $eF_n{}^{\frac{1}{2}}$, force constant $m\Omega_n{}^2$ and energy $2E\cos^2(\mathbf{k}\cdot\mathbf{r}_i+\delta)$. Notice that

$$\sum_n F_n=\sum_n f_n, \quad (22)$$

for longitudinal or transverse modes, as shown in Appendix A.

3. DISCUSSION

A macroscopic medium, whose electric properties are characterized by a dielectric constant $\epsilon(\omega)$, can be the seat of longitudinal or transverse polarization waves whose proper frequencies lie at the zeros[10] or at the poles of $\epsilon(\omega)$, respectively. Therefore, the roots of the eigenvalue equation (16), with $C=\frac{2}{3}$ or $-\frac{1}{3}$, identify, respectively, the zeros or poles of the dielectric constant of our lattice. This information, combined with the information that $\epsilon(\omega)$ is analytic and $\epsilon(\infty)=1$, shows that $\epsilon(\omega)$ is related to the lattice site polarizability $\alpha(\omega)a^3/4\pi$, defined by (17), by the Lorentz-Lorenz formula

$$\epsilon(\omega)=\frac{1+\frac{2}{3}\alpha(\omega)}{1-\frac{1}{3}\alpha(\omega)}, \quad \text{or} \quad \frac{\epsilon(\omega)-1}{\epsilon(\omega)+2}=\tfrac{1}{3}\alpha(\omega). \quad (23)$$

The Lorentz derivation of this formula treats macroscopically the mass of dielectric in a condenser with the exception of a small volume within which the interaction of microscopic dipoles is considered in detail. In the derivation by Nijboer and de Wette,[9] the microscopic treatment is extended to all dipoles in a mass of dielectric and the existence of boundaries of the medium influences only the geometrical procedure for summing over the dipole interactions. In this paper, as in an earlier one by the present author,[4] the macroscopic dielectric properties of an unbounded aggregate of atomic systems are defined in terms of its eigenstates of excitation, and the wave vectors of these eigenstates play a geometrical role somewhat analogous to that of boundary conditions.

Notice, however, that the theory of reference 4 treated the electrostatic interactions only approximately (in accordance with the "random phase approximation" of the plasma theory) and led to the Sellmeyer-Drude formula $\epsilon(\omega)=1+\alpha(\omega)$. Thereby it yielded eigenfrequencies of longitudinal or transverse oscillations which are roots of $\alpha=-1$ or $\alpha=\infty$ instead of $\alpha=-1/C=-\frac{3}{2}$ or 3. Apart from this difference, some of the following discussion is analogous to the discussion on pp. 1208–1209 of reference 4, and the remainder stresses essential features of the polarization waves that were overlooked in that earlier study.[11]

Since $\alpha(\omega)$ rises monotonically from $-\infty$ to ∞ in the interval between any two successive oscillator frequencies ω_n and ω_{n+1}, there lies in such an interval one eigenvalue of (16) for longitudinal waves ($C=\frac{2}{3}$) and one for transverse waves ($C=-\frac{1}{3}$). These two eigenvalues will be indicated by Ω_{ln} and $\Omega_{t\,n+1}$, respectively. In addition, there is one longitudinal eigenfrequency above the highest oscillator frequency, and one transverse eigenfrequency Ω_{t1} below the frequency ω_1, provided $\alpha(0)<3$. The eigenfrequencies Ω_{ln} or Ω_{tn} can thus be determined graphically, if necessary, after which they can be entered in (21), (20), and (15) to examine the characteristics of the normal modes of oscillation of the lattice.

Three different typical situations occur, depending on whether the value of $\alpha(\omega)=\sum_n \omega_p{}^2 f_n/(\omega_n{}^2-\omega^2)$ in the proximity of a particular eigenfrequency is determined primarily: (a) by a *single* term of the $\sum_n$, (b) by two terms (or groups of terms) with *opposite* signs, or (c) by one group of terms with the *same* sign.

(a) When the oscillators of frequency ω_n are perturbed only weakly by the electrostatic interaction, as it happens in gases, the polarizability $\alpha(\omega)$ varies sharply very near to ω_n but slowly for somewhat larger values of $\omega-\omega_n$. It is then represented approximately throughout a range of ω in the form $\alpha(\omega)=\langle\alpha(\omega)\rangle_n+\omega_p{}^2 f_n/(\omega_n{}^2-\omega^2)$, where the mean value $\langle\alpha(\omega)\rangle_n$ is often negligible. A necessary condition for the validity of this representation is

$$\omega_p{}^2 f_n/|\omega_n{}^2-\omega_{n\pm1}{}^2|\ll 1. \quad (24)$$

The eigenvalue equation (16) has now the approximate solutions

$$\Omega_{ln}{}^2\sim\omega_n{}^2+\frac{\frac{2}{3}\omega_p{}^2 f_n}{1+\frac{2}{3}\langle\alpha(\omega)\rangle_n}, \quad \Omega_{tn}{}^2\sim\omega_n{}^2-\frac{\frac{1}{3}\omega_p{}^2 f_n}{1-\frac{1}{3}\langle\alpha(\omega)\rangle_n}, \quad (25)$$

and one finds, from (15) and (21)

$$a_{n'}\ll a_n, \quad \text{for} \quad n'\neq n, \quad F_n\sim f_n/[1+C\langle\alpha(\omega)\rangle_n]^2. \quad (26)$$

(b) A tight electrostatic coupling between oscillators of different frequencies occurs under the condition opposite to (24), namely when

$$\omega_p{}^2 f_n/|\omega_n{}^2-\omega_{n\pm1}{}^2|\gg 1. \quad (27)$$

The polarizability $\alpha(\omega)$ varies then rapidly throughout the interval between successive oscillator frequencies

[10] See, e.g., J. Hubbard, Proc. Phys. Soc. (London) **A68**, 441 (1955).

[11] Each of the transverse normal modes considered in this paper is coupled to oscillations of the electromagnetic field with equal polarization and wave vector. This coupling is to be taken into account separately by the method of reference 4. No such further coupling is to be considered for the longitudinal modes, because the longitudinal components of the electromagnetic field are merely an alternative representation of the electrostatic interaction which has already been taken into account in the calculation of the normal modes.

ω_n and ω_{n+1}, and the eigenvalues of (16) lie rather in the middle of such an interval.[12] The normal mode is characterized here by the fact that two terms, or groups of terms, in the $\sum_{n'}$ of contributions to the dipole moment (20) have comparable magnitude and opposite sign. That is, the net dipole moment of the normal mode is reduced by the *destructive interference* between the contributions of oscillators that are out of phase because their own proper frequencies would be, respectively, somewhat larger and somewhat smaller than the eigenfrequency Ω_n of the normal mode. One can also see directly that the effective oscillator strength F_n of the normal mode must be small, when (27) holds, owing to the large value of the slope $d\alpha/d(\omega^2)$ in (21), that is, owing to inequalities

$$F_n=\left(C^2\omega_p{}^4\sum_{n'}\frac{f_{n'}}{(\Omega_n{}^2-\omega_{n'}{}^2)^2}\right)^{-1} \ll f_n\left(C^2\frac{\omega_p{}^4 f_n{}^2}{(\Omega_n{}^2-\omega_n{}^2)^2}\right)^{-1}\ll f_n. \quad (28)$$

(c) When a number of oscillators, say, from $n=r$ to $n=s$, have frequencies in a range where (27) holds, their contribution to the polarizability

$$\alpha^{(rs)}(\omega)=\sum_{n=r}^{s}\omega_p{}^2 f_n/(\omega_n{}^2-\omega^2),$$

remains $\gtrsim 1$ (in absolute value) for a considerable spectral interval above ω_s and below ω_r. This contribution resembles that of a single "effective" oscillator whose frequency ω_{eff} lies somewhere between ω_r and ω_s and whose oscillator strength $f_{\text{eff}}\sim\sum_{n=r}^{s} f_n$. For purpose of orientation we may set

$$\alpha^{(rs)}(\omega)\sim\omega_p{}^2 f_{\text{eff}}/(\omega_{\text{eff}}{}^2-\omega^2). \quad (29)$$

If (24) holds, instead of (27), for $\omega>\omega_s$ or $\omega<\omega_r$, we have in these spectral ranges $\alpha(\omega)\sim\alpha^{(rs)}(\omega)$, except in the immediate proximity of oscillator frequencies ω_n. We can then determine here the eigenfrequencies and normal modes by the approximation method of case (a) and find, in analogy with (25) and (26), normal modes characterized by

$$\Omega_{l\text{eff}}{}^2\sim\omega_{\text{eff}}{}^2+\tfrac{2}{3}\omega_p{}^2 f_{\text{eff}},\quad \Omega_{t\text{eff}}{}^2\sim\omega_{\text{eff}}{}^2-\tfrac{1}{3}\omega_p{}^2 f_{\text{eff}}, \quad (30)$$

$$F_{\text{eff}}\sim f_{\text{eff}},\quad a_{n'}\ll a_n \quad \text{for} \quad \begin{cases}(n'<r<n<s),\\ (n'>s>n>r).\end{cases} \quad (31)$$

The eigenfrequency $\Omega_{t\text{eff}}$ exists only if $\alpha^{(rs)}(0)<3$. On the other hand a normal mode of longitudinal oscillation of the type considered here—or, at least, something akin to it—is expected to occur whenever (27) holds in some spectral range, because this range would not extend to $\omega=\infty$ and would be followed by a range where (24) holds.

The polarization properties of the plasma model of metal electrons are equivalent to those of a lattice of dipoles for which (27) holds within a narrow band at $\omega_{\text{eff}}=0$ and f_{eff} represents the number of conduction electrons per crystal cell. The collective excitation of the plasma corresponds then to the longitudinal normal mode with frequency $\Omega_{l\text{eff}}$. Our study of the lattice of dipoles has shown that: (1) A plasma-like longitudinal excitation with many particles oscillating in phase occurs, as expected, under conditions much more general than those of the plasma model. (2) Normal modes of type (b) also occur, in which different oscillators are excited collectively as in the plasma-like mode, but with phase relations that yield a weak net polarization and therefore prevent ready excitation by the passage of fast charged particles.

The occurrence of collective excitations is thus seen to hinge on the condition (27). Therefore, it is interesting to consider the physical significance of this condition and the extent to which it should be expected to hold in realistic systems. Having in mind the definition (10) of $\omega_p{}^2$, the expression $\omega_p{}^2 f_n/|\omega_n{}^2-\omega_{n\pm1}{}^2|$ is seen to be a *density of oscillator strength* in space and in spectrum, namely, to be the density $f_n/a^3|\omega_n{}^2-\omega_{n\pm1}{}^2|$ expressed in the atomic unit $m/4\pi e^2=4.0\times10^{-10}$ cm^{-3} (radian/sec)$^{-2}$. (Notice that the frequency *squared* serves here as a coordinate for the spectral distribution of oscillators.) The relevancy of the spectral density is most apparent in the limiting case of a continuous spectrum of oscillators, which is treated in Appendix B.

The concept of spatial and spectral density of oscillator strength applies to any material system whose internal charges may be polarized. For any given material one may consider the density of atoms and the number of electrons in each atom for which the frequencies of dipole oscillation lie within a given spectral range. In particular, the valence electrons of any atom yield the highest spectral density because their levels of dipole excitation are concentrated mainly in a narrow energy range, say, up to 10 or 20 ev. For purpose of orientation, we may evaluate a mean spectral density of oscillator strength $\langle df/d(\omega^2)\rangle$ for an atom by allocating its number of valence electrons Z_{val} to a spectral band width equal to the ionization potential of the hydrogen atom. For a material of density ρ (in g/cm^3) and mean atomic weight A we find thus the mean spatial and spectral density of oscillator strength

$$\omega_p{}^2\langle df/d(\omega^2)\rangle\sim 4.5(\rho/A)Z_{\text{val}}. \quad (32)$$

The ratio ρ/A lies between $\frac{1}{5}$ and $\frac{1}{10}$ for most condensed materials, so that our density index is likely to be $\gtrsim 1$ on the average over a broad spectral range, and probably considerably larger than 1 in more limited ranges Therefore we are led to surmise that collective excitations of the types (b) and (c) described above occur quite generally in condensed matter.

The occurrence of collective excitations of types (b) and (c) shows a formal analogy to effects that have been recently emphasized in nuclear physics (giant

[12] A contrary surmise given at the end of p. 1208 of reference 4 was erroneous.

resonance,[13] ground states of heavy nuclei[14]) and in superconductivity. It may, therefore, be worthwhile to summarize its physico-mathematical origin. One considers initially a set of variables, the oscillation amplitudes a_n in our problem, which correspond to energy levels $\hbar\omega_n$ when a certain interaction is disregarded. This interaction is represented by a matrix whose elements have the form $D_{nm}=d_nd_m$; in our equation (12) the matrix represents a dipole-dipole interaction and each coefficient d_n is proportional to the dipole strength $f_n^{\frac{1}{2}}$ associated with one of the variables a_n. Owing to the interaction, the variables a_n and energy levels $\hbar\omega_n$ are replaced by the eigenvectors and eigenvalues of the matrix $\omega_n\delta_{nm}\pm D_{nm}$, where the $\pm$ sign corresponds to repulsive or attractive interaction. The eigenvalues are roots of $\sum_n d_n^2/(\omega-\omega_n)=\pm 1$, analogous to our Eq. (16), and the eigenvectors have components proportional to $d_n/(\omega-\omega_n)$. If the coupling coefficients d_n^2 are much larger than the spacings $\omega_{n+1}-\omega_n$ for a group of frequencies in a limited range $\omega_r\leq\omega_n\leq\omega_s$, one of the matrix eigenvalues "squirts" out of this range to a distance $\sim\pm\sum_{m=r}^{s} d_m^2$ and the corresponding eigenvector has components $\sim d_n/\sum_{m=r}^{s} d_m^2$ proportional to the d_n. This eigenvector represents a state of the system—our excitation (*c*)—in which the interaction finds maximum expression. All the other eigenvectors that result from the superposition of the variables a_n with $r\leq n\leq s$—excitations of type (b)—involve a destructive interference which neutralizes the energetic effect of the interaction so that the corresponding eigenvalues remain confined in the range (ω_r,ω_s). The qualitative features outlined here should presumably be noticeable provided only that the condition $D_{nm}=d_nd_m$ is verified approximately. Notice also that these features follow automatically when one assumes, for lack of better information, that the interaction matrix elements are all equal (since $D_{nm}\sim$const is equivalent to $D_{nm}=d_nd_m$ plus $d_n\sim$const). The transition from conditions of weak interaction, when each eigenvector nearly coincides with one of the a_n, to the condition of strong interaction may well occur rapidly as the critical ratio $d_n^2/(\omega_{n+1}-\omega_n)$ increases through the range ~ 1. Exploration of matrix properties to provide a specific answer to these questions seems worthwhile.

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[13] G. E. Brown and M. Bolsterli, Phys. Rev. Letters, 3, 472 (1959). Advance communication of this result is gratefully acknowledged.

[14] S. T. Belyaev, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 31, No. 11 (1959).

APPENDIX A

The sum rule $\sum_n F_n=\sum_n f_n$ follows from a comparison of two expansions of the function $[1+C\alpha(\omega)]^{-1}$ near $\omega=\infty$. The roots $\omega=\Omega_n$ of the eigenvalue equation (16) are the poles of this function. Near each of these poles, the function is represented approximately by

$$\{C[d\alpha/d(\omega^2)]_{\omega=\Omega_n}(\omega^2-\Omega_n^2)\}^{-1}=C\omega_p^2F_n/(\omega^2-\Omega_n^2).$$

Moreover $[1+C\alpha(\omega)]^{-1}$ approaches 1 at $\omega\sim\infty$ because α vanishes there. It can therefore be represented in terms of the residues at its poles by

$$[1+C\alpha(\omega)]^{-1}=1+C\omega_p^2\sum_n F_n/(\omega^2-\Omega_n^2). \quad (33)$$

The expansion of this representation near $\omega=\infty$ is

$$[1+C\alpha(\omega)]^{-1}\sim 1+C(\omega_p^2/\omega^2)\sum_n F_n. \quad (34)$$

On the other hand we have $\alpha(\omega)\sim-(\omega_p^2/\omega^2)\sum_n f_n$, near $\omega=\infty$, and therefore

$$[1+C\alpha(\omega)]^{-1}\sim 1+C(\omega_p^2/\omega^2)\sum_n f_n. \quad (35)$$

The equivalence of the expansions (34) and (35) requires that $\sum_n F_n=\sum_n f_n$.

APPENDIX B

We seek the limit of (21) when the spectrum of oscillator frequencies is continuous. The limit will be approached by considering first a discrete spectrum of frequencies $\omega_m=(\bar{\omega}^2+m\Delta^2)^{\frac{1}{2}}$, distributed about a mean frequency $\bar{\omega}$ with $-\infty<m<\infty$. To each of these frequencies corresponds an oscillator strength $\bar{f}$, with the understanding that $\bar{f}/\Delta^2$ will be indicated by $df/d(\omega^2)$ in the limit $\Delta^2\to 0$. Eq. (17) yields now the polarizability

$$\alpha(\omega)=\omega_p^2\sum_m\frac{\bar{f}}{\bar{\omega}^2+m\Delta^2-\omega^2}=\pi\omega_p^2\frac{\bar{f}}{\Delta^2}\operatorname{cotan}\pi\frac{\bar{\omega}^2-\omega^2}{\Delta^2}, \quad (36)$$

and

$$\frac{d\alpha}{d(\omega^2)}=\pi\omega_p^2\frac{\bar{f}}{\Delta^2}\frac{1}{\sin^2[\pi(\bar{\omega}^2-\omega^2)/\Delta]}\frac{\pi}{\Delta^2}$$

$$=\pi^2\omega_p^2\frac{\bar{f}}{\Delta^4}\left[1+\frac{\Delta^4}{\pi^2\omega_p^4\bar{f}^2}\alpha^2(\omega)\right]. \quad (37)$$

This expression may now be entered in (21), setting $\alpha=-1/C$ at $\omega=\Omega$ as required by the eigenvalue equation (16). We find

$$\bar{F}=\bar{f}/(1+\pi^2C^2\omega_p^4\bar{f}^2/\Delta^4), \quad (38)$$

and, in the limit,

$$\frac{dF}{d(\omega^2)}=\frac{df/d(\omega^2)}{1+[\pi C\omega_p^2df/d(\omega^2)]^2}. \quad (39)$$

In this limit we need no longer consider the inequality (28) between F_n and f_n since we have obtained the explicit dependence of the ratio F/f on the density of oscillator strength $\omega_p^2df/d(\omega^2)$.