COHERENCE PHENOMENA IN THE RADIATION OF IDENTICAL OSCILLATORS CONSTITUTING A CRYSTAL

Č. MUZIKAŘ*

Joint Institute for Nuclear Research

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A classical theory is given for the electromagnetic interaction of identical oscillators which form a crystal of finite dimensions. General formulas for the radiation field are derived. It is shown that in a crystal of large size there is in most directions a large broadening of the line (as compared with the line of an isolated oscillator) when there is spontaneous emission of radiation by one of the oscillators.

1. INTRODUCTION

THE effect of the interaction of an oscillator with other oscillators of the same kind surrounding it on the properties of the line omitted by the first oscillator has been studied by Dicke [1] and by Faĭn. [2] The case envisaged was that of microwave radiation from bunches of molecules, for which the distance between molecules is much smaller than the wavelength. In this case there is a large broadening of the line, proportional to the number of oscillators in the bunch.

The inverse case, in which the wavelength λ is less than the distance a between radiating systems, has been studied by Podgoretskii and Roĭzen [3]; they considered the cases of two oscillators (a diatomic molecule) and of a long linear chain. The phenomena in such systems are obviously connected with the emission of radiation by atomic nuclei arranged in a crystal, under the conditions in which the Mössbauer effect is observed. The presence of a linear chain of nuclei causes a shift and broadening of the line which depends on the direction of observation and is of the order $(\lambda/a)\gamma_{is}$, where γ_{is} is the line width for the isolated nucleus. Only in certain directions does the shift reach a large value.

When it is extended to the case of three-dimensional crystals, however, the method of Podgoretskii and Roĭzen [3] leads to complications which hinder the further development of the theory. Therefore it is expedient to choose a somewhat different way of solving this problem. In this paper we shall consider a simple cubic lattice whose sites are at the points $\mathbf{r}_s = a\mathbf{s}$, where a is the lattice constant and the components of the vector $\mathbf{s}$ are integers, and shall assume that all of the sites are occupied by identical dipoles, which we shall think of as composed of vibrating particles of charge e and mass μ.

If a dipole were isolated, then after excitation it would emit a line of frequency Ω and width $\gamma + \gamma_e = \gamma_{is}$, where $\gamma_e = (2e^2/3\mu c)\cdot(\Omega/c)^2$ is the partial width caused by the damping owing to radiation, and γ is the partial width associated with loss of energy from the oscillator through other channels. In what follows it is assumed that the time for passage of light through the crystal is much less than the times $1/\gamma_e$ and $1/\gamma$.

2. THE EQUATIONS OF MOTION

We shall denote the dipole moment at the point $\mathbf{r}_s$ and its Fourier transform by $\mathbf{p}_s(t)$ and $\mathbf{p}_s(\omega)$, respectively. At the point $\mathbf{r}$ the dipole $\mathbf{p}_s$ produces an electric field $\mathbf{E}(\mathbf{r}_s; \mathbf{r}, t)$, which we represent by a superposition of plane waves:

$$\mathbf{E}(\mathbf{r}_s; \mathbf{r}, t) = -\frac{1}{(2\pi)^4}\int d\mathbf{k}d\omega G(\mathbf{k}, \omega) \times \left\{(\mathbf{k}\,\mathbf{p}_s(\omega))\,\mathbf{k} - \left(\frac{\omega}{c}\right)^2 \mathbf{p}_s(\omega)\right\} e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}_s)-i\omega t}. \quad (1)$$

Here the retarded Green's function $G = G_0 + G_r$ is defined by the formulas

$$G_0 = 4\pi P\frac{1}{k^2 - (\omega/c)^2}, \qquad G_r = 4\pi^2 i\,\delta(k^2 - (\omega/c)^2)\,\mathrm{sign}\,\omega. \quad (2)$$

Let the origin of the reference system be inside the crystal. At a sufficiently distant point $\mathbf{R}_0$ the field from the entire crystal is a plane wave prop-

*Charles University, Prague, Czechoslovakia.

agated in the direction $\mathbf{n}_0 = \mathbf{R}_0/R_0$. In the dipole approximation the amplitude of this field is given by the formula

$$\mathbf{E}(\mathbf{R}_0, \omega) = -R_0^{-1}[\mathbf{k}_0 [\mathbf{k}_0 \mathbf{P}(\mathbf{k}_0, \omega)]] e^{i\mathbf{k}_0\mathbf{R}_0}, \quad (3)^*$$

where $\mathbf{k}_0 = \mathbf{n}_0\omega/c$ and

$$\mathbf{P}(\mathbf{k}_0, \omega) = \sum_s \mathbf{p}_s(\omega) e^{-i\mathbf{k}_0\mathbf{r}_s}. \quad (4)$$

The total radiated energy

$$W_{rad} = \int d\mathbf{n}_0 R_0^2 \int dt \mathbf{S}\mathbf{n}_0$$

($\mathbf{S}$ is the Poynting vector) takes the form

$$W_{rad} = \int d\mathbf{n}_0 \int_0^\infty d\omega I(\mathbf{n}_0, \omega). \quad (5)$$

In this formula $d\mathbf{n}_0$ is an element of solid angle and

$$I(\mathbf{n}_0, \omega) = \frac{c}{4\pi^2}\left(\frac{\omega}{c}\right)^4 \mathbf{P}_\perp \mathbf{P}_\perp^* \quad (6)$$

is the density in angle and frequency of the radiation from the crystal; $\mathbf{P}_\perp = \mathbf{P} - (\mathbf{P}\cdot\mathbf{n}_0)\mathbf{n}_0$.

To determine the quantity $\mathbf{P}_\perp$, which is analogous to the function F introduced in the paper of Podgoretskii and Roĭzen,[3] we use the equations of motion of the dipoles. Each dipole is acted on by the following forces: a) elastic and dissipative forces associated with the proper frequency Ω of the vibrations of the isolated dipole and the partial width γ; b) the retarded electric field of all the other dipoles; c) its own electric field, of which we are concerned with only the part associated with G_r in Eq. (1), since the part from G_0 leads to the infinite field mass of the dipole and it is assumed that this mass is already included in the total mass μ; d) the force $\mathbf{f}_s(t)$ by whose action the system is brought into a vibrating state. The nature of this force will be considered in more detail later. We neglect the magnetic interaction of the dipoles, since it brings in only relativistic corrections.

By means of Eq. (1) we get the following equations of motion of the oscillators:

$$\mu(\omega^2 + i\gamma\omega - \Omega^2)\mathbf{p}_s(\omega) = -e\mathbf{f}_s(\omega) + \frac{e^2}{(2\pi)^3}\int d\mathbf{k}\left[e^{i\mathbf{k}\mathbf{r}_s} G(\mathbf{k}, \omega)\left\{(\mathbf{k}\mathbf{P}(\mathbf{k}, \omega))\mathbf{k} - \left(\frac{\omega}{c}\right)^2 \mathbf{P}(\mathbf{k}, \omega)\right\} - G_0(\mathbf{k}, \omega)\left\{(\mathbf{k}\mathbf{p}_s(\omega))\mathbf{k} - \left(\frac{\omega}{c}\right)^2 \mathbf{p}_s(\omega)\right\}\right]. \quad (7)$$

Multiplying these equations by $\exp(-i\mathbf{k}_0\cdot\mathbf{r}_s)$ and summing over all s, we arrive at an integral equation for the determination of $\mathbf{P}$:

$$L(\omega)\mathbf{P}(\mathbf{k}_0, \omega) = -\mathbf{F}(\mathbf{k}_0, \omega) + \mathbf{J}_1 + \mathbf{J}_2; \quad (8)$$

$$\mathbf{J}_1 = \frac{e^2}{(2\pi)^3\mu}\int d\mathbf{k} G_r(\mathbf{k}, \omega) S(\mathbf{k} - \mathbf{k}_0)[\mathbf{k}[\mathbf{k}\mathbf{P}(\mathbf{k}, \omega)]], \quad (9)$$

$$\mathbf{J}_2 = \frac{e^2}{(2\pi)^3\mu}\int d\mathbf{k} G_0(\mathbf{k}, \omega)[S(\mathbf{k} - \mathbf{k}_0)\{[\mathbf{k}[\mathbf{k}\mathbf{P}(\mathbf{k}, \omega)]] + (k^2 - (\omega/c)^2)\mathbf{P}(\mathbf{k}, \omega)\} - \{\mathbf{k}[\mathbf{k}\mathbf{P}(\mathbf{k}_0, \omega)]] + (k^2 - (\omega/c)^2)\mathbf{P}(\mathbf{k}_0, \omega)\}], \quad (10)$$

$$L(\omega) = \omega^2 + i\gamma\omega - \Omega^2, \quad (11)$$

$$\mathbf{F}(\mathbf{k}_0, \omega) = \frac{e}{\mu}\sum_s \mathbf{f}_s(\omega) e^{-i\mathbf{k}_0\mathbf{r}_s}, \quad (12)$$

$$S(\mathbf{k} - \mathbf{k}_0) = \sum_s e^{i(\mathbf{k}-\mathbf{k}_0)\mathbf{r}_s}. \quad (13)$$

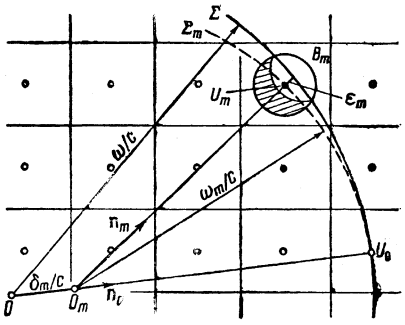
The structure factor S is a periodic function in the space of the reciprocal lattice, which reduces in our case to a cubic lattice with distance $2\pi/a$ between its nodes. At these nodes the function S has sharp maxima ($S = N_1N_2N_3$, where N_i is the number of oscillators in the direction of the i-th axis of the crystal). Since the integral of S over an elementary cell of the reciprocal lattice is $(2\pi/a)^3$, we can approximate the structure factor (13) in the following way: $S = N_1N_2N_3$ inside a parallelepiped whose edges are $2\pi/N_ia$ and whose center is at the node (hereafter we shall call this parallelepiped the nodal region), and $S = 0$ outside this region. Using the fact that the nodal region is small in comparison with the elementary cell of the reciprocal lattice and that $\mathbf{P}(\mathbf{k}, \omega)$ is a periodic function of the reciprocal lattice, we can replace $\mathbf{P}(\mathbf{k}, \omega)$ by $\mathbf{P}(\mathbf{k}_0, \omega)$ in Eqs. (9) and (10). Thus Eq. (8) is an equation for the vector $\mathbf{P} = \mathbf{P}(\mathbf{k}_0, \omega)$, which does not depend on the variable of integration $\mathbf{k}$. In what follows we shall confine ourselves to crystals of cubical shape ($N_i = N$).

3. DETERMINATION OF THE DIPOLE MOMENT P OF THE SYSTEM

If we substitute for G_r the expression (2), it can be seen from Eq. (9) that the integrand in the formula for $\mathbf{J}_1$ differs from zero only at the points where the sphere $k^2 - (\omega/c)^2 = 0$ (the sphere of propagation Σ) intersects the nodal regions of the reciprocal lattice. To determine them we can use a construction analogous to the well known Ewald construction[4]: For a given frequency ω and a given direction of observation $\mathbf{n}_0$ we lay off from an (arbitrary) node U_0 the vector $-\mathbf{k}_0 = -\omega\mathbf{n}_0/c$, and its end determines the center O of the sphere of propagation Σ (see diagram).

Thus for given ω and $\mathbf{n}_0$ we can determine the nodal regions that contribute to $\mathbf{J}_1$. Denoting these

*$[\mathbf{k}_0[\mathbf{k}_0\mathbf{P}]] = \mathbf{k}_0 \times [\mathbf{k}_0 \times \mathbf{P}]$.



regions by an index m, we get as the expression for $\mathbf{J}_1$

$$\mathbf{J}_1 = i\omega\gamma_e \frac{3}{8\pi} N^3 \sum_{m=0}^{M} \int_{(m)} d\mathbf{n}\,(\mathbf{n}(\mathbf{Pn}) - \mathbf{P}), \qquad (14)$$

where (m) is the solid angle subtended at the center O by the portion of the sphere Σ that intersects the m-th nodal region, and M is the number of regions intersected.

For simplicity let us replace the nodal region by a sphere of radius $\rho = (3/4\pi)^{1/3}(2\pi/Na)$. Because of the smallness of the nodal regions we can neglect the variation of the integrand in Eq. (14) and replace it by a constant value $(\mathbf{P}\cdot\mathbf{n}_m)\mathbf{n}_m - \mathbf{P}$, which is determined in the following way. In the diagram the point O is the center of the sphere of propagation Σ for the frequency ω and direction of observation $\mathbf{n}_0$. The point O_m is the center of the sphere of propagation Σ_m for this same direction of observation but for a frequency ω_m such that the sphere passes exactly through the node U_m. Thus for each direction $\mathbf{n}_0$ and each node U_m one determines a direction $\mathbf{n}_m$ and a frequency ω_m.*

Introducing the notation $\delta_m = \omega - \omega_m$, we get for the distance ϵ_m between the nodal point U_m and the sphere of propagation Σ the value

$$\varepsilon_m = (1 - \mathbf{n}_0\mathbf{n}_m)\,\delta_m/c. \qquad (15)$$

By means of these results the formula (14) can be represented with good accuracy in the form

$$\mathbf{J}_1 = i\omega\gamma_e \left(\frac{4\pi}{3}\right)^{1/3} \frac{3}{8\pi^2}\left(\frac{\lambda}{a}\right)^2 N \sum_m g\left(\frac{\varepsilon_m}{\rho}\right)((\mathbf{Pn}_m)\,\mathbf{n}_m - \mathbf{P}), \qquad (16)$$

where $g(x) = (3\pi/4)(1-x^2)$.

The calculation of $\mathbf{J}_2$ is more complicated. Let us divide the entire $\mathbf{k}$ space into cells B_ν. (Each such cell is essentially a first Brillouin zone.)

*One can obtain all of the points O_m by means of the following construction: we take the nodal point U_0 as the origin and construct the Brillouin zones.[5] The points O_m are the points of intersection of the boundary surfaces of these zones and the straight line that passes through the point O_0 in the direction $\mathbf{n}_0$.

Using Eq. (2) and the properties of the function S, we get

$$\int_{(B_\nu)} d\mathbf{k}\,(S(\mathbf{k}-\mathbf{k}_0) - 1)\,G_0(\mathbf{k},\omega)\left(\mathbf{k}^2 - \left(\frac{\omega}{c}\right)^2\right)$$

$$= 4\pi \int_{(B_\nu)} d\mathbf{k}\,(S(\mathbf{k}-\mathbf{k}_0) - 1) = 0. \qquad (17)$$

Then $\mathbf{J}_2$ takes the form

$$\mathbf{J}_2 = \frac{e^2}{2\pi^2\mu} \sum_\nu \mathrm{P} \int_{(B_\nu)} d\mathbf{k}\left(S(\mathbf{k}-\mathbf{k}_0) - 1\right)\frac{[\mathbf{k}\,[\mathbf{kP}]]}{k^2 - (\omega/c)^2}. \qquad (18)$$

In the calculation of this integral an important part is played by the relative positions of the cell B_ν and the sphere of propagation Σ. Three different cases are possible: 1) the sphere of propagation $k^2 - (\omega/c)^2 = 0$ does not intersect the cell B_ν; 2) it intersects the cell, but not the nodal region (see diagram, $|\epsilon_m| > \rho$); 3) it intersects both the cell and the nodal region ($|\epsilon_m| < \rho$). It can be shown that (owing to the large value of N) the main contribution to the expression (18) comes from the last case. Thus in the formula (18) we are to keep only the integrals

$$\mathrm{P}\int d\mathbf{k}\,[\mathbf{k}\,[\mathbf{kP}]]/(k^2 - (\omega/c)^2) \qquad (19)$$

over nodal regions intersected by the sphere Σ. In these integrals, owing to the antisymmetry of the function $k^2 - (\omega/c)^2$, the unshaded parts of the nodal regions (see diagram) make practically no contribution. Then on replacing the fraction in Eq. (19) by its value at the nodal point, we get as an approximate expression for the integral (19):

$$-\frac{\omega}{c}\left(\frac{2\pi}{Na}\right)^3 \frac{[\mathbf{n}_m[\mathbf{n}_m\mathbf{P}]]}{2\rho} f\left(\frac{\varepsilon_m}{\rho}\right), \quad f(x) = \frac{3}{2}\left(1 - \frac{x^2}{3}\right)\mathrm{sign}\,x. \qquad (20)$$

Using all of these results, we arrive at the following approximate expression for $\mathbf{J}_2$:

$$\mathbf{J}_2 = -i\omega\gamma_e \left(\frac{4\pi}{3}\right)^{1/3} \frac{3}{8\pi^2}\left(\frac{\lambda}{a}\right)^2 N \sum_m f\left(\frac{\varepsilon_m}{\rho}\right)((\mathbf{Pn}_m)\,\mathbf{n}_m - \mathbf{P}). \qquad (21)$$

The summation is taken over just the same nodal points as in Eq. (16). We note further that for m = 0 all of the spheres of propagation pass through the nodal point U_0, and therefore $f(\epsilon_0/\rho) = 0$, $g(\epsilon_0/\rho) = 3\pi/4$.

Introducing the notations

$$A = (2\pi)^{-1}(3/4\pi)^{2/3}(\lambda/a)^2 N\gamma_e, \qquad (22)$$

$$\Phi_m = \omega A\,(f(\varepsilon_m/\rho) - ig(\varepsilon_m/\rho)), \qquad (23)$$

we rewrite Eq. (8) for $\mathbf{P}$ in the form

$$\mathcal{L}\mathbf{P} = -\mathbf{F} - \sum_m \Phi_m((\mathbf{Pn}_m)\,\mathbf{n}_m - \mathbf{P}). \qquad (24)$$

Multiplying Eq. (24) by $\mathbf{n}_q$ ($q = 0, 1, 2, \ldots, M$), we get a system of linear equations

$$\Big(L - \sum_m \Phi_m\Big)\,\mathbf{P}\mathbf{n}_q + \sum_m \Phi_m\,(\mathbf{n}_q\mathbf{n}_m)\,(\mathbf{P}\mathbf{n}_m) = -\,\mathbf{F}\mathbf{n}_q, \quad (25)$$

from which we can determine the values of $\mathbf{P}\cdot\mathbf{n}_q$ and substitute them in the equation

$$\Big(L - \sum_m \Phi_m\Big)\,\mathbf{P}_\perp = -\,\mathbf{F}_\perp - \sum_m{}' \Phi_m\,(\mathbf{P}\mathbf{n}_m)\,\mathbf{n}_{m\perp} \quad (26)$$

for the determination of $\mathbf{P}_\perp$. Equation (26) follows simply from Eqs. (24) and (25). The prime on the summation sign means that the value $m = 0$ is excluded from the sum. The index $\perp$ denotes the component perpendicular to the direction $\mathbf{n}_0$.

Thus the investigation of the effect of the crystal lattice on the line shape of the emitted light reduces to the finding of the nodal regions intersected by the sphere of propagation Σ and the solution of the system of linear equations (25) and (26).

4. SPONTANEOUS EMISSION IN A CRYSTAL OF LARGE SIZE

As an example of the use of these formulas let us consider spontaneous emission. Suppose that for times $t < 0$ all of the oscillators are in the unexcited state, with the exception of the one located at the point $\mathbf{s} = 0$, which is in an excited state with the energy W_0 and begins to radiate at the time $t = 0$. We can obtain a classical model of such a system by assuming that $\mathbf{p}_s(t) = 0$ for $t < 0$ for all oscillators and that at the time $t = 0$ the oscillator at the point $\mathbf{s} = 0$ receives an instaneous impulse. To produce this we set $\mathbf{f}_s(t) = K\delta_{0s}\,\delta(t)$, where $K = (2\mu W_0)^{1/2}$. Then the vector $\mathbf{F}$ in the formulas (24) — (26) takes the form

$$\mathbf{F}_{sp} = (e/\mu)\,K. \quad (27)$$

The use of the retarded Green's function for the electric field of the interaction assures that the condition $\mathbf{p}_s(t) = 0$ is satisfied for $t < 0$. Then since for spontaneous emission the direction of the exciting force is not determinate, we must average $I_{sp}(\mathbf{n}_0, \omega)$ over all directions of the vector $\mathbf{K}$.

We assume that the crystal is sufficiently large. In this case the dimensions of the nodal regions in the reciprocal lattice space are very small and consequently it is most probable that the only intersection is that with the nodal region $m = 0$ (which intersects all the spheres). Thus for most directions of observation $\mathbf{n}_0$ and frequencies ω the sum in Eq. (26) reduces to just the term $m = 0$, and

$$\mathbf{P}_\perp = -\,\mathbf{F}_\perp\,(L - \Phi_0)^{-1}. \quad (28)$$

On using the formula (6) for the averaged density of spontaneously emitted radiation, we get

$$\bar{I}_{sp}(\mathbf{n}_0, \omega) = \frac{W_0}{2\pi^2}\left[1 + \left(\frac{\omega - \Omega}{\Gamma/2}\right)^2\right]^{-1}\frac{\gamma_e}{\Gamma^2}, \quad (29)$$

where W_0 is the excitation energy and Γ is the total line width, given by the formula

$$\Gamma = \gamma + \frac{3}{8}\left(\frac{3}{4\pi}\right)^{2/3}\left(\frac{\lambda}{a}\right)^2 N\gamma_e. \quad (30)$$

We obtain the conditions under which we can regard the crystal as large in the sense indicated above in the following way: if $\lambda \ll a$, then there are approximately $(4\pi/3)(\omega/c)^3(a/2\pi)^3$ nodal points inside the sphere of radius ω/c. The number of nodal regions intersected by the sphere is determined by the number of points inside a spherical shell with the radii $\omega/c \pm \rho$, and is consequently equal to $8\pi\,(3/4\pi)^{1/3}(a/\lambda)^2(1/N)$. For a large crystal this quantity must be much smaller than unity, and from this we have

$$N \gg 10\,(a/\lambda)^2. \quad (31)$$

If the nonelectromagnetic width γ is not very large in comparison with γ_e, then it can be seen from Eqs. (30) and (31) that $\Gamma \gg \gamma + \gamma_e$. Thus the radiation emitted spontaneously from an oscillator which is a constituent of a large crystal is in most directions a line which retains the Lorentz shape but is much broadened in comparison with the line emitted by an isolated source.

The treatment is complicated for the phenomena that occur for directions of propagation for which the sphere of propagation intersects more than one nodal region. The expression for $\mathbf{P}_\perp$ is now a fraction, with a denominator which consists of two factors; one is $L - \Sigma\Phi_m$, and the other is the determinant of the system (25). The properties of these factors determine the shape of the line. It can be seen at once that the presence of terms $f(\epsilon_m/\rho)$ leads to a shift of the line, and that the presence of the function $g(\epsilon_m/\rho)$ again leads to a broadening of this line. Since the determinant can be a higher-degree polynomial in the frequency ω, the line emitted may have a complicated multiplet structure depending on the direction of observation, and the line loses its Lorentz shape.

For very small crystals the sphere of propagation intersects many nodal regions and it can be shown that in this case the crystal has almost no effect on the line shape.

The scattering of radiation incident on a large crystal is treated in greater detail in [6]. Scattering occurs at the Bragg angle, and the width of the scattered line depends on the ratio of the width of the incident line to the width Γ given by Eq. (30), in accordance with the general theory of scattering. [7]

All of our results are derived for an ideal single crystal, in which identical oscillators are distributed in a strictly periodic crystal lattice. For direct comparison with experiment one must keep in mind the effect of possible violations of these conditions.

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[1] R. H. Dicke, Phys. Rev. **93**, 99 (1954).

[2] V. M. Faĭn, Usp. Fiz. Nauk **64**, 273 (1958).

[3] M. I. Podgoretskii and I. I. Roĭzen, JETP **39**, 1473 (1960), Soviet Phys. JETP **12**, 1023 (1961).

[4] R. W. James, The Optical Principles of the Diffraction of X-Rays, London, 1950.

[5] L. Brillouin and M. Parodi, Propagation des Ondes dans les Milieux Periodiques, Paris, 1956.

[6] Č. Muzikař, Joint Inst. Nuc. Res. Preprint P-709, Dubna, 1961.

[7] W. Heitler, The Quantum Theory of Radiation, Oxford, 1954.

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