

MOLECULAR SCATTERING IN A BOUNDED MEDIUM

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The possibility is considered of developing a theory of Rayleigh and Raman scattering of waves by bodies of finite dimensions (the dimensions being arbitrary with respect to the wavelength of the incident and scattered radiations) for the case when the electromagnetic properties of the body differ sharply from those of the medium in which the body is located. The classical results of the theory of molecular scattering of light are derived from the relations obtained under conditions when the refractive index unperturbed by fluctuations in the scattering body is identical with the refractive index of the surrounding medium.

INTRODUCTION

IF there are no fluctuations in a medium and the requirement of optical homogeneity is met (dimensions of the particles, the distance d between the particles, and their mean free paths small compared to the wavelength λ of the incident radiation), then, as Mandel'shtam first showed,^[1] such a medium cannot be turbid. Forward coherent scattering of the oscillators leads only to an index of refraction different from unity, and the same kind of scattering in the backward direction in the presence of a plane of separation between the initial medium and the medium perturbed by the presence of the particles evokes a field that can be calculated from the Fresnel equations. It is clear that in the case of a body bounded on all sides coherent scattering causes the field that is determined as a result of solving the ordinary diffraction problem for the given body.

In the same paper Mandel'shtam^[1] noted that in an optically inhomogeneous medium the addition of the intensities scattered by the individual particles (Rayleigh) is perfectly valid, and leads to an electromagnetic field scattered also in lateral directions, which is also called incoherent scattering. The difficulty uncovered by Mandel'shtam, namely that in air (an optically homogeneous medium) one nonetheless observes incoherent scattering obeying the Rayleigh relations, was subsequently reconciled in the investigations of Smoluchowski^[2] and Einstein.^[3] It was found that in the presence of fluctuations incoherent scattering takes place even under the condition $\lambda \gg d$, i.e., in practically all bodies independently of the relation between the wavelength and the distance between the particles.

For a body bounded on all sides this means that the total field scattered by this object consists of the ordinary diffraction field and an incoherently scattered one that is associated with the presence of fluctuations in the electromagnetic parameters. It is clear that the solution of the second problem (the determination of the intensity of molecular scattering) is closely linked with the solution of the first (diffraction), since the coherently scattered field, together with the incident field in vacuum, determines in final analysis the exciting electric field which, by interacting with the fluctuations, evokes the incoherently scattered waves. Of course, if

the scattering volume is only slightly different from the surrounding space with respect to its electromagnetic properties, then one should take as the exciting field the ordinary field, for example, of a plane wave in unbounded space filled by a medium with dielectric constant ϵ , and then all complications associated with the necessity of solving the diffraction problem disappear. It was just in this way, as far as we know (see^[4-7] and literature cited therein), that the various results of the classical theory of molecular scattering were obtained.

There is no doubt, however, that setting up and solving a rigorous boundary-value problem of electrodynamics, which permits one to take consistently into account all diffraction effects in the determination of both the exciting and the incoherently scattered fields, is of great interest. It has become of particular importance in recent years with the advent of powerful sources (masers and lasers) and the increase in sensitivity of detectors, which makes it possible experimentally to observe extremely fine diffraction effects during molecular scattering in a bounded medium.

1. STATEMENT OF THE PROBLEM. CHOICE OF METHOD OF SOLUTION

Consider some bounded body with sharp boundaries. Let it be required to determine the spatial distribution of the intensity of the molecular scattering of waves incident on this body from without. In general, besides volume scattering there is also diffusion scattering, associated with the roughness of the free surface of the body,^[2, 8] but with the assumption of a sharp boundary it is sufficient to consider only volume scattering.

In order that the theory of molecular scattering in a bounded medium be applicable to a wide class of bodies and interacting waves, the form of its general results should not depend, firstly, on the relation between the wavelengths and the dimensions of the body; in the second place, on the relation between the electromagnetic parameters of the body and the surrounding space; thirdly, on the type of incident wave (plane, spherical, with curved direction, etc.); and finally, on the shape of the scattering body. Of course, with such firm require-

ments, it is inevitable that the results will be of extremely general character, but they will possess the decisive advantage that the region of their concrete applications will be extraordinarily wide.

Levin and Rytov^[9-10] have constructed a theory of equilibrium thermal fluctuations of the electromagnetic field in bounded bodies using the electrodynamic reciprocity principle, the application of which insures that all the aforementioned requirements will be met. The generality and simplicity of this theory stimulated the choice of method of solution of the problem of molecular scattering in a bounded medium which is the subject of this paper. This method is also based on the reciprocity theorem for the field created by an auxiliary source (this is usually a point dipole located in the Fraunhofer diffraction zone) and the field scattered by the fluctuations in the body under consideration. The first step is to determine all those quantities that play the role of sources of the incoherently scattered field, i.e., the "scattering currents." For this it is necessary to write down the equations of the scattered field such that the sources will be explicitly distinguished.

2. EQUATIONS OF THE INCOHERENTLY SCATTERED FIELD

In the presence of electromagnetic fluctuations the total field \mathbf{E}_σ , \mathbf{H}_σ in the medium consists of the sum of fields: exciting (initial) \mathbf{E}_0 , \mathbf{H}_0 and incoherently scattered \mathbf{E} , \mathbf{H} . Obviously, in the determination of the initial field in the medium one should surely take into account the contribution from the coherent scattering. In a boundless medium this is attained at the macroscopic level automatically by the introduction of ε ; in a semi-bounded medium it is possible to make use of the Fresnel formulas in determining \mathbf{E}_0 and \mathbf{H}_0 , and in scattering by fully bounded bodies it is natural to define as the primary field that field which is the solution of the problem of diffraction in the given body in the ordinary, coherent sense.^[11] Only when the refractive indices inside and around the body are almost the same and the boundary problem can be solved in the Rayleigh-Gans-Born approximation is it sufficient to take the usual field of the incident wave as the internal field acting on the fluctuating oscillators.

In the above-mentioned determination of the initial field, the change in its amplitude due to the molecular scattering is not considered, since the cross section of this process is extremely small (if the fluctuations are not too great), and the length of the traversed wave path cannot be very large by virtue of the statement of the problem (dimensions of the body comparable to the wavelength). For the same reasons, in determining \mathbf{E} and \mathbf{H} it suffices to consider one-time scattering, i.e., to confine oneself at this stage to the solution of the problem in the Born approximation.^[6] Thus, in our statement of the problem both the exciting and the incoherently-scattered fields must be determined from the solution of the boundary-value problem for the body considered (this is where it differs from the classical theory of molecular scattering), but in formulating the differential equations of the scattered field use is made, as before, of the first (Born) approximation of the method of small perturbations.^[6] In addition, in spite of the presence of boundaries the fluctuations of the dielectric

constant are assumed to be homogeneous, which imposes definite limitations on the dimensions of the body (see Sec. 3).

The vectors of the total field \mathbf{E}_σ and \mathbf{H}_σ satisfy the usual equations of macroscopic electrodynamics

$$\text{rot } \mathbf{E}_\sigma = -\frac{1}{c} \frac{\partial \mathbf{H}_\sigma}{\partial t}, \quad \text{div } \mathbf{H}_\sigma = 0, \quad (1)$$

$$\text{rot } \mathbf{H}_\sigma = \frac{1}{c} \frac{\partial \mathbf{D}_\sigma}{\partial t}, \quad \text{div } \mathbf{D}_\sigma = 0,$$

where

$$\mathbf{E}_\sigma = \mathbf{E}_0 + \mathbf{E}, \quad \mathbf{H}_\sigma = \mathbf{H}_0 + \mathbf{H}, \quad \mathbf{D}_\sigma = \mathbf{D}_0 + \mathbf{D}, \quad \mathbf{D}_\sigma = \hat{\varepsilon}_\sigma \mathbf{E}_\sigma. \quad (2)$$

In the case of an isotropic homogeneous medium, to which we are here confined, the operator of the dielectric constant can also be represented as a sum of two parts:

$$\hat{\varepsilon}_\sigma = \hat{\varepsilon} + \delta \hat{\varepsilon}(t, \mathbf{r}), \quad (3)$$

where the dependence of $\delta \hat{\varepsilon}$ on time and coordinates reflects the space-time behavior of the fluctuations.

For the sake of simplicity, we shall henceforth take into account in (3) only isotropic fluctuations, since when anisotropic fluctuations^[12] are also present, the boundary problem being considered is solved in completely analogous fashion.

If in the investigation of the propagation of the incident and scattered waves one ignores spatial dispersion (allowance for which may, however, be necessary for some cases in plasmas^[13] and in crystal optics^[14]), then, omitting non-linear terms in (2), we obtain for the Fourier component with respect to time the following relation:

$$\mathbf{D}_\omega = \varepsilon(\omega) \mathbf{E}_\omega + \delta \varepsilon_{\Delta\omega}(\mathbf{r}) \mathbf{E}_{0\omega}. \quad (4)$$

The quantity $\Delta\omega = \omega - \omega_0$ has the meaning of the difference of the frequencies of the scattered and incident waves. Because of the neglect of spatial dispersion, $\delta \varepsilon_{\Delta\omega}(\mathbf{r})$ is not an integral operator. However, in calculating the correlation function for $\delta \varepsilon_{\Delta\omega}(\mathbf{r})$ the spatial dispersion should, generally speaking, be taken into account, if the problem is that of investigating the spectral constitution of the scattered radiation.

Substituting (4) into (1), we arrive at the standard equations for the Fourier components of the incoherently scattered field:

$$\text{rot } \mathbf{E}_\omega = \frac{i\omega}{c} \mathbf{H}_\omega, \quad (5)$$

$$\text{rot } \mathbf{H}_\omega = -\frac{i\omega}{c} \varepsilon(\omega) \mathbf{E}_\omega + \frac{4\pi}{c} \mathbf{J}_\omega, \quad (6)$$

where the role of the source evoking molecular scattering is played by the quantity

$$\mathbf{J}_\omega = -\frac{i\omega}{4\pi} \delta \varepsilon_{\Delta\omega} \mathbf{E}_{0\omega}. \quad (7)$$

From (5) and (6) we get the following inhomogeneous wave equation for \mathbf{E}_ω :

$$\Delta \mathbf{E}_\omega + \frac{\omega^2}{c^2} \varepsilon(\omega) \mathbf{E}_\omega = -\frac{\omega^2}{c^2} \left\{ \delta \varepsilon_{\Delta\omega} \mathbf{E}_{0\omega} + \frac{c^2}{\omega^2 \varepsilon(\omega)} \text{grad}(\mathbf{E}_{0\omega}, \text{grad } \delta \varepsilon_{\Delta\omega}) \right\}. \quad (8)$$

If we neglect the time rate of change of ε on the wave, then (8) goes over to the equation first obtained by Einstein.^[3]

The distribution of the incoherently scattered field over all space may now be found either by solving the

boundary problem for the inhomogeneous wave equation (8) with appropriate boundary conditions or by using the reciprocity theorem for the current of the auxiliary emitter and of the "scattering current" given by (7).

3. MOLECULAR SCATTERING CROSS SECTION

Since the main interest is usually the field of molecular scattering in the far zone, we place an auxiliary point dipole

$$\mathbf{p} = p_0 c^2 \omega^{-2} \delta(\mathbf{r}' - \mathbf{r}) e^{-i\omega t}$$

in the Fraunhofer diffraction zone (\mathbf{p}_0 is a unit vector in the direction of the dipole). By orienting the auxiliary dipole each time along the normals to the observation direction $\mathbf{n} = \mathbf{r}/r$, we find by the usual rules^[15] its field in the zone where the body is located:

$$\mathbf{E}'_{\omega}{}^{\text{inc}}(\mathbf{r}, \mathbf{r}') = p_0 \frac{e^{i\mathbf{k}\mathbf{r}}}{r} e^{-i\mathbf{k}\mathbf{r}'}, \quad (9)$$

where

$$\mathbf{k} = n\mathbf{k}, \quad k = \omega \sqrt{\bar{\epsilon}(\omega)} / c. \quad (10)$$

The dielectric constant of the surrounding space also may depend on frequency and equals $\bar{\epsilon}(\omega)$. From (9) we see that the auxiliary field \mathbf{E}'_{ω} inside the object being considered is the result of the diffraction of a plane wave of unit amplitude by the given body and is well known for the majority of simple configurations.

By applying the reciprocity theorem^[4] and considering the expression for the "scattering current" (7), we immediately obtain the desired relation for the electric field of molecular scattering in the Fraunhofer zone:

$$|\mathbf{E}_p(\mathbf{r})|^2 = \frac{(\omega/c)^4}{16\pi^2 r^2} \iint \Phi(\mathbf{r}') \Phi^*(\mathbf{r}'') \langle \delta\epsilon_{\Delta\omega}(\mathbf{r}') \delta\epsilon_{\Delta\omega}^*(\mathbf{r}'') \rangle d\mathbf{r}' d\mathbf{r}'', \quad (11)$$

where

$$\Phi(\mathbf{r}') = \mathbf{E}_{0\omega}(\mathbf{r}') \mathbf{E}'_{\omega}(\mathbf{r}') \quad (12)$$

and the integration is carried out over the region occupied by the fluctuating oscillators. Equation (11) allows one to calculate the field for two independent polarizations, which in the spherical system are naturally called the ϑ - and φ -polarizations depending on which of the two components (p_{ϑ} or p_{φ}) is zero.

As is seen from (11), for the determination of the quadratic characteristics of the molecular scattering field yielded by some completely bounded body it is necessary to solve first two diffraction problems: that of the field of the point source and that of the coherent field created inside the body by the incident wave. Since in diffraction theory the difficulty lies not in obtaining formal results but in discussing them (Sommerfeld), it can be said that formal solutions for molecular scattering in a bounded medium can be easily obtained from (11) in all simple particular cases (sphere, cylinder, etc.). On the other hand, for their analysis one requires all the asymptotic or approximate methods of diffraction theory (from the long-wave Rayleigh approximation to geometrical optics), depending on the relation between the wavelengths of the incident and scattered radiations and the dimensions of the body, as well as on the electromagnetic properties of the object itself. This analysis can result in the discovery of new features compared to molecular scattering in a boundless

medium (another directivity diagram, additional resonances in the spectrum of the scattered radiation due to the presence of boundaries, for example magnetostatic resonances in the scattering by a small ferrite sample or electroacoustical resonances in the scattering by small plasma bodies, etc.).

Of principal significance is the question of the correlation function $\langle \delta\epsilon_{\Delta\omega}(\mathbf{r}') \delta\epsilon_{\Delta\omega}^*(\mathbf{r}'') \rangle$, if fluctuations occur in bounded bodies. As far as we know, this question has never been specially investigated (and such an investigation is evidently a difficult problem, particularly in those cases where one has to do with a consistent calculation of spatial dispersion in a completely bounded body), in connection with which one has to use, at least in the first step, the correlation function for boundless space even in boundary value problems. In any case, it is obvious that the validity of this approach depends on the relation of the correlation radius and the characteristic dimensions of the system. Hence, in the absence of wave motion, when the fluctuations are correlated only within molecular distances,^[4] (i.e., on the macroscopic level the correlation radius equals zero), this approach is rigorously valid. We can be confident that it suffices also even when there is wave motion in those cases when the correlation radius is at least small compared to the dimensions of the system. Fortunately, this is almost always the case in experiments, as was pointed out in^[5, 16].

Considering these remarks and postulating that the fluctuation field is spatially homogeneous,^[6] we write

$$\langle \delta\epsilon_{\Delta\omega}(\mathbf{r}') \delta\epsilon_{\Delta\omega}^*(\mathbf{r}'') \rangle = \langle |\delta\epsilon|^2 \rangle_{\Delta\omega R}, \quad (13)$$

where

$$\mathbf{R} = \mathbf{r}' - \mathbf{r}''. \quad (14)$$

Introducing, as usual,^[6] a new variable \mathbf{R} and extending the limits of integration over \mathbf{R} to infinity (on account of the smallness of the correlation radius), we transform (11) to the form

$$|\mathbf{E}_{\omega p}(\mathbf{r})|^2 = \frac{(\omega/c)^4}{16\pi^2 r^2} \int_V d\mathbf{r}' \int_{-\infty}^{\infty} \Phi(\mathbf{r}') \Phi^*(\mathbf{r}' - \mathbf{R}) \langle |\delta\epsilon|^2 \rangle_{\Delta\omega R} d\mathbf{R}. \quad (15)$$

In considering Rayleigh scattering (without substantial frequency change) it is possible to ignore the space-time behavior of the fluctuations,^[3] and then

$$\langle |\delta\epsilon|^2 \rangle_{\Delta\omega R} = \langle \delta\epsilon^2 \rangle \delta(\Delta\omega) \delta(\mathbf{R}), \quad (16)$$

which makes possible the integration in (15) both over frequency and over \mathbf{R} . As a result, the molecular-scattered field will have the same frequency as the incident field, and

$$|\mathbf{E}_p(\mathbf{r})|^2 = \frac{(\omega_0/c)^4 \langle \delta\epsilon^2 \rangle}{16\pi^2 r^2} \int_V |\Phi(\mathbf{r}')|^2 d\mathbf{r}'. \quad (17)$$

However, if it is a question of investigating the spectral composition of the scattered radiation (e.g., Mandel'shtam-Brillouin scattering),^[16-18] then it is necessary to expand the correlation function in a Fourier integral

$$\langle |\delta\epsilon|^2 \rangle_{\Delta\omega R} = \int \langle \delta\epsilon^2 \rangle_{\Delta\omega \mathbf{k}} e^{i\mathbf{k}\mathbf{R}} d\mathbf{k}. \quad (18)$$

The Fourier components $\langle |\delta\epsilon|^2 \rangle_{\Delta\omega \mathbf{k}}$ are determined in the correlation theory developed by Rytov.^[16]

We present, finally, the differential cross sections of molecular scattering in an element of solid angle $d\Omega$,

which are obtained without difficulty with the aid of (15) and (17):

$$d\sigma_\omega = \sqrt{\frac{\varepsilon(\omega)}{\varepsilon(\omega_0)}} \frac{\omega^4}{16\pi^2 c^4} \int_V d\mathbf{r}' \int_{-\infty}^{\infty} \Phi(\mathbf{r}') \Phi^*(\mathbf{r}' - \mathbf{R}) \langle |\delta\varepsilon|^2 \rangle_{\Delta\omega\mathbf{R}} d\mathbf{R} d\Omega \quad (19)$$

or in the Einstein case

$$d\sigma = \frac{\omega^4}{16\pi^2 c^4} \langle \delta\varepsilon^2 \rangle \int_V |\Phi(\mathbf{r}')|^2 d\mathbf{r}'. \quad (20)$$

In calculations with Eqs. (19) and (20) it must be kept in mind that the amplitude of the incident wave outside the body is normalized to unity, and the total cross section is obtained as the sum of contributions from two independent polarizations.

4. BORN APPROXIMATION. TRANSITION TO THE EINSTEIN FORMULA

Equations (19) and (20) differ from the results of classical molecular scattering theory in the sense that they are obtained by rejecting the Born approximation in solving the boundary problem and hence are free of the shortcomings associated with this approximation. They permit determination of the molecular scattering cross section for any relations between the refractive indices outside and inside the body. Under the condition $\bar{\varepsilon} \approx \varepsilon$, which corresponds to the Rayleigh-Gans-Born approximation, these results should go over into the classical ones, since as the incident field one may now again take the field of a propagating plane wave in the given medium. Then

$$\Phi(\mathbf{r}') \Phi^*(\mathbf{r}' - \mathbf{R}) = \cos^2 \theta e^{-i\mathbf{q}\mathbf{R}}, \quad (21)$$

where

$$\mathbf{q} = \mathbf{k} - \mathbf{k}_0 \quad (22)$$

(\mathbf{k}_0 is the wave vector of the incident wave, $\cos \theta$ is the angle between the electric vectors of the incident and auxiliary waves). Since in (21) one should take into account the contribution of both ψ - and φ -polarizations, we get

$$\cos^2 \theta = \cos^2 \psi + \cos^2 \varphi. \quad (23)$$

If we choose the coordinate system so that the incident wave propagates along the z axis (the polar axis in spherical coordinates), then the role of the scattering angle (i.e., the angle between \mathbf{k}_0 and \mathbf{k}) will be played by the polar angle ψ . Then it is easy to show that

$$\cos \psi = \cos \theta \cos(\varphi - \varphi_0), \quad \cos \varphi = \sin(\varphi_0 - \varphi), \quad (24)$$

where φ_0 is the angle between the direction of the electric vector of the incident wave and the x axis. From this we conclude that

$$\cos^2 \theta = 1 - \sin^2 \psi \cos^2(\varphi - \varphi_0). \quad (25)$$

In the case of unpolarized incident radiation it is still necessary to average over all orientations of \mathbf{E}_0 (i.e., over the angle φ_0) and

$$\overline{\cos^2 \theta} = \frac{1}{2}(1 + \cos^2 \psi). \quad (26)$$

Substituting (21) with account taken of (26) into (19) and integrating over \mathbf{r}' (which gives simply the total volume V of the scattering body), we obtain

$$d\sigma_\omega = \sqrt{\frac{\varepsilon(\omega)}{\varepsilon(\omega_0)}} \frac{\omega^4}{32\pi^2 c^4} (1 + \cos^2 \psi) \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\mathbf{k} d\mathbf{R} e^{i(\mathbf{k}-\mathbf{q})\mathbf{R}} \langle |\delta\varepsilon|^2 \rangle_{\Delta\omega\mathbf{k}} d\Omega. \quad (27)$$

Since

$$\int_{-\infty}^{\infty} e^{i(\mathbf{k}-\mathbf{q})\mathbf{R}} d\mathbf{R} = (2\pi)^3 \delta(\mathbf{k} - \mathbf{q}), \quad (28)$$

we get finally after integration over \mathbf{k}

$$d\sigma_\omega = \frac{\pi\omega^4}{4c^4} \sqrt{\frac{\varepsilon(\omega)}{\varepsilon(\omega_0)}} (1 + \cos^2 \psi) \langle |\delta\varepsilon|^2 \rangle_{\Delta\omega\mathbf{q}} d\Omega \quad (29)$$

—a result well known from the theory of molecular scattering.

If further the space-time behavior of the fluctuations is unimportant, then (21) should be substituted into (20), and then

$$d\sigma = \frac{\omega^4 V}{32\pi^2 c^4} (1 + \cos^2 \psi) \langle \delta\varepsilon^2 \rangle d\Omega. \quad (30)$$

Integrating over all solid angles and dividing by V , we calculate the extinction coefficient

$$h = \frac{\omega^4}{6\pi c^4} \langle \delta\varepsilon^2 \rangle. \quad (31)$$

Substituting into this the known^[4] correlation function

$$\langle \delta\varepsilon^2 \rangle = \rho T \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \left(\frac{\partial \rho}{\partial T} \right)_T + \frac{T^2}{\rho c_v} \left(\frac{\partial \varepsilon}{\partial T} \right)_\rho^2, \quad (32)$$

we obtain the result given in Landau and Lifshitz.^[4] If temperature fluctuations are not taken into account ($\langle \delta T^2 \rangle = 0$), then

$$h = \frac{\omega^4}{6\pi c^4} T \rho \left(\frac{\partial \rho}{\partial p} \right)_T \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T^2, \quad (33)$$

which is the same as Einstein's formula.^[3]

In conclusion we remark that the approach developed in this paper to the theory of molecular scattering by bounded objects can be used also in problems of scattering of electromagnetic and acoustic waves by turbulent fluctuations in the atmosphere,^[19] scattering and transformation of waves by electromagnetic fluctuations in a plasma,^[20, 21] and in acoustical problems where the relation of the properties of the scattering body and the surrounding medium are such as to require a rigorous solution of the boundary problem. The greatest difficulties arise in those cases when calculation of spatial dispersion is found to be necessary, not only in determining the correlation function, but also in the diffraction stage of the solution of the corresponding problems.

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