

On the theory of light scattering in gases

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Contents

1. Introduction	75
2. Rayleigh scattering law	75
3. On the dispersion theory	76
References	79

Abstract. The history of development of the theory of light scattering and dispersion in gases is outlined, from the works of Rayleigh, Planck, and Mandelstam to those of Lorentz and Einstein. It is shown that of central concern in these studies was actually the problem of thermal fluctuations in a medium. A formula for the permittivity $\varepsilon(\omega)$ taking account of radiation friction forces is derived for the case of an isotropic medium.

1. Introduction

Early in the 20th century, i.e. approximately 100 years ago, there occurred a polemic concerning the problem of light scattering in gases. The starting point was the work of Lord Rayleigh [1] who described the scattering of light in the terrestrial atmosphere, provided an explanation for the blue color of the sky, and derived the Loschmidt number to the correct order of magnitude using the experimental data as the base.

This paper came under criticism by Mandelstam (1907), who also touched upon Planck's paper on dispersion theory in addition to Rayleigh's paper. A debate broke out between Mandelstam and Planck (1907–1908). Recalling it seems to be rather instructive.

Getting ahead of the story, it is pertinent to note that Lorentz also advanced his views on the problem (1910), even though he did not participate in the polemic. He performed calculations of the permittivity $\varepsilon(\omega)$ of gases, which led him to the results of Planck and Rayleigh (for more details, see Section 3 below).

Today, commenting upon the works of even the great physicists, which date back to a century ago, is easy and at the same time difficult. Easy, because many of the present-day truisms were then completely unknown. Difficult, because the central physical problems of that time are not topical today.

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Everything may appear to be trivially simple. In particular, this applies to the problem of light scattering and the dispersion theory.

When discussing the Mandelstam–Planck polemic I will endeavor to assume an unbiased attitude. I will note fallacies and inaccuracies, but in doing this I will not simplify the problems that faced the physicists a century ago. I will also try to show that the dispute between Mandelstam and Planck was actually concerned not with a particular problem of light scattering. The case in point was a controversy about whether a medium can be homogeneous despite the thermal molecular motion in the medium. Or whether a medium without fluctuations is possible, as we would put it today. But at that time the concept of fluctuations, their unavoidable and universal nature did not exist. The works of Smoluchowski and Einstein made their appearance later. Planck proved to be right in this dispute. Although he did not invoke the notion of fluctuations explicitly, the results for light scattering in gases he arrived at turned out to be the same as if he were doing all the calculations with due regard for fluctuations.

2. Rayleigh scattering law

The problem of light scattering in the terrestrial atmosphere was first considered by Rayleigh late in the 19th century [1]. He assumed that molecules scatter incoherently because they participate in thermal motion. This allows a summation to be made over the intensities of scattering by individual oscillators. It is customary to characterize the scattering intensity by an extinction coefficient $h[\text{cm}^{-1}]$ which describes the attenuation of intensity of a light beam along a path L in the absence of true absorption: $I(L) = I(0) \exp(-hL)$. Rayleigh obtained the result

$$h[\text{cm}^{-1}] = \frac{8\pi}{3} k^4 |\alpha|^2 N. \quad (1)$$

Here, $k = \omega/c$, α is the polarizability of an oscillator, and N is the number of the oscillators per cubic centimeter. The polarizability enters into the simplest formula for the permittivity ε of a medium:

$$\varepsilon = 1 + 4\pi N\alpha.$$

In gases, outside the absorption bands, the real part of $\alpha(\omega)$ as well as of $\varepsilon(\omega)$ far exceeds the imaginary part. This permits

formula (1) to be expressed in terms of $\varepsilon - 1 \cong 2(n - 1)$, where n is the index of refraction. Eventually the Rayleigh scattering law is commonly written as

$$h = \frac{2}{3\pi} \frac{\omega^4}{c^4} \frac{(n - 1)^2}{N}. \quad (2)$$

The central point of Rayleigh's paper, specifically, that the motion of oscillators results in the incoherence of their light scattering, was severely criticized by Mandelstam [2]. His arguments reduced to the following. The wavelength λ of visible light is of the order of 10^{-4} cm. At the same time, for $N \cong 10^{19} - 10^{18}$ cm $^{-3}$ (the upper terrestrial atmosphere) the average distance between the molecules is $\bar{R} \cong 10^{-6}$ cm. Therefore, a volume smaller than λ^3 contains very many molecules, viz. $\sim 10^6$. From here Mandelstam concluded that the light scattering by the particles which make up the medium is responsible for the light field of the same intensity and phase, irrespective of whether the particles are at rest or in motion. The last-mentioned statement is simultaneously formulated in more general terms: "an optically homogeneous medium cannot scatter light".

Undoubtedly the above is correct. But the statement that the motion is insignificant on condition that $\bar{R} \ll \lambda$ is not true. This is easy to verify. Induced oscillations of an oscillator in the field $\mathbf{E} \propto \exp[-i\omega t + \mathbf{kR}(t)]$ contain a factor $\exp[i\mathbf{kR}(t)]$, where $\mathbf{R}(t)$ is the oscillator coordinate. In a mean free time τ of a molecule, the phase $\mathbf{kR}(t)$ acquires an increment $\mathbf{k}\mathbf{v}\tau$, where \mathbf{v} is the oscillator velocity. But $v\tau = l$, where l is the mean free path. Hence one finds

$$\mathbf{k}\mathbf{v}\tau \cong \frac{2\pi}{\lambda} l,$$

and the question of coherence of induced oscillations of the oscillators in the medium should be solved by comparing λ not with \bar{R} but with the mean free path l .

But $l = 1/N\sigma$, where σ is the elastic cross section. For gases which make up the terrestrial atmosphere, σ is of the order of 3×10^{-16} cm 2 . For the optical spectral range, where $\lambda \cong 0.5 \times 10^{-4}$ cm, and altitudes $H \geq 15$ km, where $N \leq 4 \times 10^{18}$ cm $^{-3}$, we have $l \geq 5 \times 10^{-3}$ cm and $l/\lambda \geq 1$, so that random phase shifts $\mathbf{k}\mathbf{v}\tau$ exceed 2π . For lower layers of the atmosphere, viz. $H \sim 5 - 10$ km, these shifts are still large. For instance, for $H = 6$ km we have $2\pi l/\lambda = 1.75$. The above estimates, which rely on the model for terrestrial atmosphere of Ref. [3], show that Rayleigh was right when treating the light scattering by the molecules of the upper atmosphere as incoherent.

The summary of this part of the polemic is as follows:

(1) the coherence criterion is the ratio between λ and l rather than between λ and \bar{R} , as Mandelstam believed;

(2) assuming that the motion of molecules can be neglected, i.e. considering the light scattering in the medium where the molecules stay at rest, Mandelstam assumed that the condition $\bar{R} \ll \lambda$ also ensures the homogeneity of the medium.

Here are citations of Mandelstam's papers [2, 4]:

"The air should be treated as an optically homogeneous medium, for a cube λ^3 contains 5×10^6 molecules" (Ref. [2], p. 116).

"Gases under normal conditions (at atmospheric pressure) should be regarded as optically homogeneous bodies" (Ref. [4], p. 130).

Both of these statements rest on the $\bar{R} \ll \lambda$ criterion.

3. On the dispersion theory

What at all has the microscopic dispersion theory got to do with the question of light scattering?

Two approaches can be pursued to calculate the extinction coefficient h [cm $^{-1}$] for a rarefied gas. One can proceed from the calculations of the scattering intensity and the effective scattering cross section σ_{sc} by the particles of a medium or proceed from the definition of h in terms of the permittivity $\varepsilon(\omega)$ of the medium:

$$h = \sigma_{sc} N = \frac{8\pi}{3} k^4 |\alpha|^2 N, \quad (3)$$

or

$$h = \frac{\omega}{c} \text{Im } \varepsilon(\omega) \dots \quad (4)$$

The difference between these formulas is as follows: no additional constraints are imposed on α in formula (3). If α is complex, i.e. $\alpha = \alpha' + i\alpha''$, the inequality $\alpha' \gg \alpha''$ is always (in gases, outside of an absorption band) fulfilled. That is why in the calculation of σ_{sc} one can assume that α is a real quantity and obtain a (quantitatively) correct result, which was taken advantage of in formula (2).

Im ε is another matter. It is precisely the imaginary value of $\varepsilon(\omega)$ and the reasons underlying its imaginary nature that are the heart of the problem, no matter how small the Im ε quantity may be in comparison with Re ε .

In his works on dispersion theory [5], Planck showed that the attenuation of a transmitted light wave in the absence of dissipation is determined by its scattering, resulting in the imaginary value of the permittivity $\varepsilon(\omega)$, which is determined by the radiative damping constant $\gamma_{rad} = 2e^2\omega^2/3mc^3$. This damping results from the deceleration of oscillator oscillations caused by its intrinsic radiation field $\mathbf{E} = (2/3c^3)\ddot{\mathbf{d}}$, (see Ref. [6]), where \mathbf{d} is the oscillator dipole moment. In his calculations Planck adopted Rayleigh's postulate that the light scattering in the terrestrial atmosphere proceeds incoherently. It is precisely this result by Planck that was criticized by Mandelstam (Ref. [4], p. 125): "If the entire medium, including resonators, is regarded as optically homogeneous, no damping due to scattering should be expected in this case".

Therefore, starting from formula (4), one should perform calculations of $\varepsilon(\omega)$ taking into account the action on the medium oscillators exerted by their radiation fields. The intensity of scattering (or its absence) should be made consistent with the magnitude of Im $\varepsilon(\omega)$. This relationship follows from the comparison of formulas (3) and (4).

By way of example I will consider the light scattering by a harmonic oscillator. The induced oscillations are given by the equation for the oscillator dipole moment \mathbf{d} :

$$\ddot{\mathbf{d}} + \gamma\dot{\mathbf{d}} + \omega_0^2\mathbf{d} = \frac{e^2}{m} \{ \mathbf{E} \exp(-i\omega t) + \mathbf{E}^* \exp(i\omega t) \}, \quad \mathbf{d} = e\mathbf{x}. \quad (5)$$

Here, the term $\gamma\dot{\mathbf{d}}$ describes the possible damping of oscillator oscillations, the cause of damping in no way being defined concretely. It is easy to calculate the polarizability $\alpha(\omega)$, the total cross section σ_{tot} defined by the work done on the oscillator by the field, and the scattering cross section σ_{sc} :

$$\alpha = \frac{e^2}{m} \frac{1}{-\omega^2 + \omega_0^2 - i\omega\gamma}, \quad (6)$$

$$\sigma_{\text{tot}} = 4\pi k \text{Im } \alpha, \quad (7)$$

$$\sigma_{\text{sc}} = \frac{8\pi}{3} k^4 |\alpha|^2. \quad (8)$$

Here, $k = \omega/c$. Because in the general case $\sigma_{\text{tot}} = \sigma_{\text{sc}} + \sigma_{\text{dis}}$, where σ_{dis} is the dissipation cross section, there occurs an inequality $\sigma_{\text{tot}} \geq \sigma_{\text{sc}}$, so that

$$\text{Im } \alpha(\omega) \geq \frac{2}{3} k^3 |\alpha|^2, \quad (9)$$

which is referred to as the optical theorem. From expression (6) and inequality (9) it follows that

$$\gamma \geq \frac{2e^2\omega^2}{3mc^3}.$$

The right-hand side of this inequality is precisely the radiative damping constant (see Ref. [6]):

$$\gamma_{\text{rad}} = \frac{2e^2\omega^2}{3mc^3}. \quad (10)$$

If no dissipation occurs, γ in Eqn (5) becomes $\gamma = \gamma_{\text{rad}}$, i.e. the damping is determined by the light scattering alone. We can put it somewhat differently. If an oscillator does scatter light, the damping constant in Eqn (5) cannot be equal to zero: $\gamma \geq \gamma_{\text{rad}}$.

One can see that the definition of h by formula (4) is a very fine instrument. L I Mandelstam addressed himself to precisely this instrument, i.e. he decided to undertake the calculations of $\varepsilon(\omega)$ and in this way determine h . But he approached the realization of this task with the assumption that the condition $\bar{R} \ll \lambda$ ensures the optical homogeneity of the medium. We are reminded of the following statement from Ref. [2]: “The air should be treated as an optically homogeneous medium, for a cube λ^3 contains 5×10^6 molecules”. If so, it is assumed in advance that the calculations of $\varepsilon(\omega)$ in the absence of dissipation will yield $\text{Im } \varepsilon(\omega) = 0$ and $h = 0$.

All this took place in the days when an understanding of the role of fluctuations was completely missing; their universal character and principal role in several phenomena were not recognized. An absolutely correct idea that a homogeneous medium cannot scatter light ruled Mandelstam’s mind. In his dispute with Lord Rayleigh he tried to take advantage of the inequality $\bar{R} \ll \lambda$ to actually impose homogeneity on the medium. In Ref. [2], first of all he says that molecular motion is insignificant, that it should be neglected and the oscillators should be ‘brought to rest’, and mentions in passing that the medium therewith becomes homogeneous. In the subsequent discussion the assertion that the medium is homogeneous comes to be central. But $h = (\omega/c) \text{Im } \varepsilon$ should be equal to zero, since the medium is homogeneous.

By contrast, M Planck adopted, after Rayleigh, that independent oscillators scatter light (incoherently). Starting from the condition $h \neq 0$, he constructed the dispersion theory in such a way as to obtain $\text{Im } \varepsilon \neq 0$ and make damping, with $\gamma = \gamma_{\text{rad}}$ given by expression (10), responsible for this. He thereby introduced fluctuations implicitly and, what is more, correctly. Later on, when the concept of fluctuations was realized (Smoluchowski, Einstein), it became clear that the scattering in rarefied gases is determined by the fluctuations of density or the number of

particles, i.e. by the quantity $\overline{\Delta N^2}$. But for an ideal gas one has $\Delta N^2 = N$, i.e. the result arrived at is precisely the same as in the consideration of the light scattering by individual particles. In the Mandelstam–Planck discussion Planck was doomed to obtain the correct result. He supposedly sensed that the thermal molecular motion is bound to disturb the homogeneity.

Starting from the statement that the medium is homogeneous, Mandelstam was certain to show that $\text{Im } \varepsilon = 0$ and that the radiative damping in the equation for the oscillator of a medium is compensated for by the presence of neighbors.

I note in passing that the well-known Einstein formula for the light scattering by the fluctuations of density ρ and temperature T (1910):

$$h = \frac{\omega^4}{6\pi c^4} \left\{ \rho T \left(\frac{\partial \rho}{\partial P} \right)_T \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T^2 + \frac{T^2}{\rho C_V} \left(\frac{\partial \varepsilon}{\partial T} \right)_\rho^2 \right\} \quad (11)$$

is easily shown, if we go over to the variables P and S (the pressure and the entropy, which is always done, by the way, in the description of the spectral composition of the scattered light), to be strongly simplified for gases and to pass into the Rayleigh formula (2) (see Ref. [7])¹

$$h = \frac{2\omega^4}{3\pi c^4} \frac{(n-1)^2}{N}.$$

After the above-mentioned we need not outline in detail the works of Mandelstam and Planck on the dispersion theory (so firmly did the idea of medium homogeneity rule Mandelstam’s mind). I will only note the main points.

In his first paper on the dispersion theory [4] (a semiphenomenological work with elements of microscopic description) Mandelstam wrote down the Maxwell equations, where the induction vector is

$$\mathbf{D} = \mathbf{E} + 4\pi N \mathbf{d}.$$

For $\mathbf{d}(t)$, advantage is taken of the equation

$$\ddot{\mathbf{d}} + \omega_0^2 \mathbf{d} - \frac{e^2}{m} \frac{2}{3c^3} \ddot{\mathbf{d}} = \frac{e^2}{m} \mathbf{E}', \quad (12)$$

where \mathbf{E}' is the electric field which would have existed at the location of a resonator had we removed the corresponding resonator together with its field. Planck referred to \mathbf{E}' as ‘the driving force’ [4]. The term containing $\ddot{\mathbf{d}}$ is responsible for the damping due to radiative friction. Since the damping is weak, for harmonic oscillations at a frequency ω it is possible to make the change

$$-\frac{e^2}{m} \frac{2}{3c^3} \ddot{\mathbf{d}} \cong \frac{2}{3} \frac{e^2\omega^2}{mc^3} \mathbf{d} = \gamma_{\text{rad}} \mathbf{d}.$$

According to Planck, one sets

$$\mathbf{E}' = \mathbf{E} + \frac{4\pi}{3} N \mathbf{d}.$$

¹ Now we could say that the Rayleigh scattering law is the correct formula for h in gases (not only under the assumption of incoherent light scattering).

Mandelstam's calculations and the corresponding reasoning give [4]

$$\mathbf{E}' = \mathbf{E} + \frac{4\pi}{3} N\mathbf{d} - \frac{2}{3c^3} \ddot{\mathbf{d}}.$$

Therefore, the radiative friction is removed from Eqn (12). In response to Planck's criticism [8], pointing out that the radiation fields of the neighbors should also be included, Mandelstam took these into account in the subsequent papers [9, 10]. He carried out an extensive calculation of the radiation fields of the oscillators in the medium, but in the summation of the fields of the neighboring oscillators he made every effort to retain the homogeneity of the medium. In calculating the resultant sums, a large volume V is divided into cells, each of which contains strictly one particle.

As a result, Mandelstam obtained a complete compensation for the radiative friction forces: "The refractive index proves to be real. Therefore, no attenuation occurs due to scattering" [9, 10]. This is undoubtedly true for a homogeneous medium. It fluctuations are nonexistent — no light scattering occurs. But in fact, this is specially thrust upon by the calculations.

In 1910, H A Lorentz interfered in the polemic by publishing his paper "On the question of light scattering by molecules" [11]. It is pertinent to note that he made no mention of Mandelstam and Planck at all, as if their polemic was nonexistent. The paper bore a reference only to Rayleigh. But one can see from the text of the paper, very comprehensive and detailed, that the paper was a direct answer to the questions posed by Mandelstam. Lorentz gave a thorough derivation of the formulas which define the interaction of the oscillators in the medium via their radiation fields. The resultant sums over the oscillators of the medium surrounding a given oscillator were calculated in two ways — first assuming the oscillators of the medium to be regularly distributed in space, and next for an irregular distribution. In the former case, the result he obtained is that in the absence of dissipation the $\varepsilon(\omega)$ function is real and $\text{Im } \varepsilon = 0$. In the latter case, he arrived at the results of Rayleigh and Planck. Lorentz complemented the $\varepsilon(\omega)$ computations with direct calculations of the intensity of scattering by a medium with an irregular spatial distribution of oscillators. A perfect agreement was reached. The main result was formulated as follows: "Scattering can occur only when molecules are irregularly distributed, as they are in gases and liquids".

Given below is the main point of Lorentz's calculations of the $\varepsilon(\omega)$ function, namely, the method of summation of the oscillator radiation fields in the medium. I will take advantage of the approach and the terminology of present-day dispersion theory, which allows the presentation to be made more compactly, but in doing this I will closely follow Lorentz.

The radiation field of an oscillator at a distance \mathbf{R} is given by the expression (see Ref. [6])

$$\mathbf{E}(t) = \text{rot rot} \frac{\mathbf{d}(t - R/c)}{R}.$$

By going over to the Fourier components at a frequency ω and employing the general relation for the Fourier transform of a function $f(t')$ of a retarded argument $t' = t - a$:

$$\{f(t - a)\}_\omega = \{f(t)\}_\omega \exp(i\omega a),$$

we obtain

$$\mathbf{E}_\omega = \text{rot rot} \frac{\exp[i(\omega/c)R]}{R} \mathbf{d}_\omega.$$

That is why the equation of oscillator motion in the medium with the inclusion of the response of the intrinsic radiation field and the fields of all remaining oscillators, which is written for the Fourier component \mathbf{d}_ω^j of the dipole moment, assumes the form

$$\mathbf{d}_\omega^j \{ -\omega^2 - i\omega\gamma_{\text{rad}} + \omega_0^2 \} = \frac{e^2}{m} \sum_{j \neq i} \text{rot rot} \frac{\exp[i(\omega/c)|\mathbf{R}_i - \mathbf{R}_j|]}{|\mathbf{R}_i - \mathbf{R}_j|} \mathbf{d}_\omega^j + \frac{e^2}{m} \mathbf{E}_\omega. \quad (13)$$

Here, \mathbf{R}_i and \mathbf{R}_j are the oscillator coordinates; the rot operation in the sum over j acts on \mathbf{R}_j ; possible dissipation processes are neglected; the damping is determined by the radiative friction force alone, and \mathbf{E}_ω is the external field.

It is shown that the expression

$$\text{rot rot} \frac{\exp[i(\omega/c)|\mathbf{R}_i - \mathbf{R}_j|]}{|\mathbf{R}_i - \mathbf{R}_j|} \mathbf{d}_\omega^j$$

for $|\mathbf{R}_i - \mathbf{R}_j| \ll \lambda$ includes the effect of radiative friction. We expand the exponent

$$\frac{\exp[i(\omega/c)|\mathbf{R}_i - \mathbf{R}_j|]}{|\mathbf{R}_i - \mathbf{R}_j|}$$

in a series and retain only the first nonvanishing imaginary term in this expansion to obtain

$$\begin{aligned} \frac{e^2}{m} \text{rot rot} \frac{\exp[i(\omega/c)|\mathbf{R}_i - \mathbf{R}_j|]}{|\mathbf{R}_i - \mathbf{R}_j|} \mathbf{d}_\omega^j &\cong i \frac{e^2}{m} \frac{2}{3} \left(\frac{\omega}{c}\right)^3 \mathbf{d}_\omega^j \\ &= i\omega\gamma_{\text{rad}} \mathbf{d}_\omega^j. \end{aligned} \quad (14)$$

The calculations of $\varepsilon(\omega)$ with the aid of system (13) are performed as follows (see, for instance, Ref. [12]). We consider Eqn (13) without the external field \mathbf{E}_ω . We average the right-hand side of Eqn (13) over the coordinates \mathbf{R}_j assuming that every oscillator can find itself at any point in space with an equal probability, irrespective of the location of other oscillators, including the i th oscillator (this is precisely the ideal gas approximation invoked by Lorentz):

$$\begin{aligned} \sum_{j \neq i} &\rightarrow \lim_{V \rightarrow \infty} (NV - 1) \\ &\times \frac{1}{V} \int_{\delta} d\mathbf{R}_j \text{rot}_{\mathbf{R}_i} \text{rot}_{\mathbf{R}_j} \frac{\exp[i(\omega/c)|\mathbf{R}_i - \mathbf{R}_j|]}{|\mathbf{R}_i - \mathbf{R}_j|} \mathbf{d}_\omega(\mathbf{R}_j). \end{aligned} \quad (15)$$

The lower limit δ of integration signifies that the integration should be taken over the range $|\mathbf{R}_i - \mathbf{R}_j| > \delta \rightarrow 0$. We next take advantage of the relation (see Ref. [13])

$$\begin{aligned} &\int_{\delta \rightarrow 0} d\mathbf{R}' \text{rot}_{\mathbf{R}} \text{rot}_{\mathbf{R}'} \frac{\exp[i(\omega/c)|\mathbf{R} - \mathbf{R}'|]}{|\mathbf{R} - \mathbf{R}'|} \mathbf{d}_\omega(\mathbf{R}') \\ &= \int d\mathbf{R}' \text{rot}_{\mathbf{R}} \text{rot}_{\mathbf{R}'} \frac{\exp[i(\omega/c)|\mathbf{R} - \mathbf{R}'|]}{|\mathbf{R} - \mathbf{R}'|} \mathbf{d}_\omega(\mathbf{R}') - \frac{8\pi}{3} \mathbf{d}_\omega(\mathbf{R}) \end{aligned} \quad (16)$$

to obtain the integral equation for $\mathbf{d}_\omega(\mathbf{R})$:

$$\mathbf{d}_\omega(\mathbf{R}) = N\alpha(\omega) \int d\mathbf{R}' \text{rot}_{\mathbf{R}} \text{rot}_{\mathbf{R}} \frac{\exp[i(\omega/c)|\mathbf{R} - \mathbf{R}'|]}{|\mathbf{R} - \mathbf{R}'|} \times \mathbf{d}_\omega(\mathbf{R}') - \frac{8\pi}{3} N\alpha(\omega) \mathbf{d}_\omega(\mathbf{R}). \quad (17)$$

Here, $\alpha(\omega)$ is the polarizability of one isolated oscillator defined by expression (6); for $\gamma = \gamma_{\text{rad}}$, one obtains

$$\alpha(\omega) = \frac{e^2}{m} \frac{1}{-\omega^2 + \omega_0^2 - i\omega\gamma_{\text{rad}}}. \quad (18)$$

The solution of Eqn (17) will be sought in the form of plane waves, which can propagate through the medium of dipoles whose oscillations are given by the system of equations (13) or the integral equation (17). The wave vector of a plane wave in the medium is not \mathbf{k} , as in vacuum. The sought plane waves will be assumed to be of the form

$$\exp(-i\omega t + i\boldsymbol{\eta}\mathbf{R}),$$

where $\boldsymbol{\eta} \neq \mathbf{k}$ and $\eta = \sqrt{\varepsilon(\omega/c)}$. The dependences of the Fourier amplitudes $\mathbf{d}_\omega(\mathbf{R})$ and $\mathbf{d}_\omega(\mathbf{R}')$ respectively on \mathbf{R} and \mathbf{R}' are of the form

$$\mathbf{d}_\omega(\mathbf{R}) = \mathbf{d}_\omega \exp(i\boldsymbol{\eta}\mathbf{R}), \quad \mathbf{d}_\omega(\mathbf{R}') = \mathbf{d}_\omega \exp(i\boldsymbol{\eta}\mathbf{R}'),$$

where \mathbf{d}_ω is a constant independent of coordinates. The calculation of the integral in Eqn (17) can be found in Ref. [13]:

$$\int d\mathbf{R}' \left\{ \text{rot}_{\mathbf{R}} \text{rot}_{\mathbf{R}} \frac{\exp[i(\omega/c)|\mathbf{R} - \mathbf{R}'|]}{|\mathbf{R} - \mathbf{R}'|} \mathbf{d}_\omega(\mathbf{R}') \right\} \times \exp[i\boldsymbol{\eta}(\mathbf{R}' - \mathbf{R})] = \frac{4\pi\eta^2}{\eta^2 - k^2}, \quad k = \frac{\omega}{c}. \quad (19)$$

As a result, we obtain the dispersion equation which relates $\eta(\omega) = \sqrt{\varepsilon(\omega)}(\omega/c)$ and the frequency ω :

$$1 = N\alpha \frac{4\pi\eta^2}{\eta^2 - (\omega/c)^2} - \frac{8\pi}{3} N\alpha, \quad \eta^2 = \varepsilon\left(\frac{\omega}{c}\right)^2, \quad (20)$$

whence after simple identity transformations there follows the well-known Lorentz–Lorenz formula

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{4\pi}{3} N\alpha(\omega) = \frac{4\pi}{3} N \frac{e^2}{m} \frac{1}{-\omega^2 + \omega_0^2 - i\omega\gamma_{\text{rad}}}. \quad (21)$$

This formula was obtained by application of the technique of summation over j in Eqns (13), which was developed in Lorentz's work for an ideal gas [11].

As shown by Klimontovich and Fursov [14], for a medium wherein density fluctuations differ from the case of an ideal gas, the radiative damping in the expressions for $\alpha(\omega)$ and $\varepsilon(\omega)$ is determined by the constant

$$\gamma_{\text{rad}} \frac{\overline{\Delta N^2}}{N}. \quad (22)$$

Here, $\overline{\Delta N^2}$ and N are respectively the rms fluctuation and the average number of particles residing in a specific volume selected in the body. For an ideal gas, we have $\overline{\Delta N^2} = N$, which leads to formula (21). If the possibility of fluctuations is completely neglected by putting $\overline{\Delta N^2} = 0$, the radiative damping vanishes altogether.

Therefore, in a homogeneous medium of isotropic oscillators, the interaction of the oscillators through their radiation fields results in a complete compensation for radiative damping. A homogeneous medium does not scatter electromagnetic waves. By postulating the uniformity of oscillator distribution in a medium, Mandelstam obtained this result by direct calculation of $\varepsilon(\omega)$ [9, 10]. The occurrence of fluctuations is responsible for light scattering, the intensity of scattering by density fluctuations in an ideal gas proving to be equal to the sum of the intensities of scattering by each of the isolated oscillators, as was adopted in the works of Rayleigh and Planck. Indeed, in gases $|\varepsilon| \sim 1$ and formula (21) gives

$$\text{Im } \varepsilon(\omega) = 4\pi N |\alpha|^2 \frac{2}{3} \left(\frac{\omega}{c}\right)^3,$$

whence follows, in view of the general definition $h = (\omega/c) \text{Im } \varepsilon$, the Rayleigh scattering law (1) or (2).

In the calculation of the $\varepsilon(\omega)$ function in the foregoing, the motion of oscillators was disregarded (like in the papers by Mandelstam and Lorentz). The inclusion of motion is significant in the calculation of the light scattering spectrum, but does not influence the spectrum-integrated extinction coefficient h .

The works of Lorentz and Einstein dotted the i's and crossed the t's. The Mandelstam–Planck polemic ceased. The idea that the intensity of light scattering is related to the fluctuations induced by thermal molecular motion in a medium received general acceptance. Already in Ref. [15] (1913), L I Mandelstam considered the scattering of light from the fluctuations on a liquid surface, having completely abandoned the postulate of optical homogeneity of a medium, which he had adopted in Refs [9, 10]. Here are just two citations of Ref. [15]:

“As is well known, the statistical consideration of the second principle of thermodynamics leads to the notion that a system which is in the state of thermodynamic equilibrium does not nevertheless stay at rest, but fluctuates constantly about its equilibrium position.”

“The surface of a liquid, which would be, for instance, planar under ideal equilibrium, is persistently deformed due to irregular thermal motion. When a light beam is reflected from such a surface, along with regular (specular) reflection, diffuse reflection is also bound to occur.”

Today these statements are evident. However, as the foregoing suggests, the way to them proved to be by no means an easy one for the physicists early in the 20th century.

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