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Abstract

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PHYSICS

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ON SOME FEATURES OF DIFFUSE SCATTERING

(Presented by Academician I. V. Obreimov, 28 III 1967)

The experimental study of absorption spectra (and the determination of $k(\lambda)$, the absorption coefficient in Bouguer's law) is a more or less simple problem if a homogeneous sample of the substance of sufficient size is available. However, the situation becomes more complicated if the substance for spectral analysis is obtained in the form of small particles and obtaining the spectrum of an individual particle is impossible for technical reasons. The study of the light scattered by an aggregate of a large number of particles gives characteristics F that depend not only on the properties of the substance, but also on the particle sizes a_i , their shape, mutual arrangement, etc. The scattering function F may correspond to various time-averaged characteristics of light fluxes that arise as a result of the interaction of incident monochromatic light with an aggregate of absorbing and scattering particles; here we have in mind characteristics that do not depend on the intensity of the incident light. In the case of scattering by particles arranged in a plane-parallel layer illuminated from one side, F may denote: R , the diffuse reflection coefficient (the ratio of the reflected light flux to the incident one), T , the transmission coefficient (the ratio of the transmitted light flux to the incident one), the angular distribution of reflected and transmitted light, etc.

To determine $k(\lambda)$ from experimentally measured characteristics F , a_i , etc., it is necessary to know the dependences connecting these quantities; in the general case this problem is mathematically so complex that so far no encouraging results have been obtained along this path. The greatest successes in this direction have been achieved using model representations of the scattering medium ^(1, 2). Another problem is also of interest: the determination of F from known $k(\lambda)$, a_i , etc.; although it is mathematically simpler than the preceding one, its solution in analytic form also does not yet appear possible, even for comparatively simple scattering systems.

The present work is devoted to establishing certain features of the dependence of the scattering function F on k , a_i , l , ε , ω (l is the size of the region occupied by the scattering particles, ε is the dielectric constant of the substance of the particles, ω is the cyclic frequency of the scattered light).

As is known, the coordinate-dependent part of the electric $\mathbf{E}(\mathbf{r}) \exp(-i\omega t)$ and magnetic $\mathbf{H}(\mathbf{r}) \exp(-i\omega t)$ fields of a monochromatic wave is determined by the equations ⁽³⁾

$$\Delta \mathbf{E} + \frac{\varepsilon(\omega, \mathbf{r})}{c^2} \omega^2 \mathbf{E} - \text{grad div } \mathbf{E} = 0,$$

$$\Delta \mathbf{H} + \frac{\varepsilon(\omega, \mathbf{r})}{c^2} \omega^2 \mathbf{H} + \frac{1}{\varepsilon(\omega, \mathbf{r})} [\nabla \varepsilon(\omega, \mathbf{r}) \text{ rot } \mathbf{H}] = 0. \quad (1)$$

Here $\varepsilon(\omega, \mathbf{r})$ does not depend on \mathbf{E} and \mathbf{H} (linear optics).

This system of homogeneous linear equations follows directly from the macroscopic Maxwell equations in the absence of currents and charges. The properties of the scattering system (system 1) determine the spatial-

dependence of the dielectric constant $\varepsilon(\omega, \mathbf{r})$. To find $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$, one should solve equations (1) taking into account the incident monochromatic radiation $\mathbf{E}_0(\mathbf{r}) \exp(-i\omega t)$, $\mathbf{H}_0(\mathbf{r}) \exp(-i\omega t)$. Having found $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$, one can determine at any point $\bar{\mathbf{s}} = \frac{c}{4\pi} [\mathbf{E}\mathbf{H}]$ —the mean (in time) energy-flux density. Knowing \mathbf{s} , it is easy to pass to

$$F = \frac{\int_{\Sigma'} \bar{\mathbf{s}} d\vec{\sigma}}{\int_{\Sigma} \bar{\mathbf{s}}_0 d\vec{\sigma}},$$

where in the denominator is the flux of the incident radiation through the surface Σ , and in the numerator the flux of the scattered radiation through the surface Σ' ; $d\vec{\sigma}$ is a surface element. Since equations (1) are linear and homogeneous, F does not depend on the intensity of the incident light.

Consider system 2, similar to system 1, but in which all geometrical dimensions are increased by a factor q , i.e. $\varepsilon(\omega, q^{-1}\mathbf{r})$. Let monochromatic radiation be scattered by system 2,

$$\mathbf{E}_0 \left(\frac{\mathbf{r}}{q} \right) \exp \left(-i \frac{\omega}{q} t \right), \quad \mathbf{H}_0 \left(\frac{\mathbf{r}}{q} \right) \exp \left(-i \frac{\omega}{q} t \right)$$

with frequency ω/a . The equations determining the coordinate part of the electric and magnetic fields of system 2 coincide with (1), but only the argument is not \mathbf{r} , but $\mathbf{r}_1 = q^{-1}\mathbf{r}$. Thus $\mathbf{E}(\mathbf{r})$, $\mathbf{H}(\mathbf{r})$, $\bar{\mathbf{s}}(\mathbf{r})$ at the point \mathbf{r} of system 1 coincide with $\mathbf{E}(\mathbf{r}_1)$, $\mathbf{H}(\mathbf{r}_1)$, $\bar{\mathbf{s}}(\mathbf{r}_1)$ at the corresponding point $q\mathbf{r}$ of system 2. Consequently, in system 1 and system 2 there is similarity of the scattered fluxes and equality of F for the corresponding Σ and Σ' . The absorption losses (referred to a unit volume per 1 sec.) at the point \mathbf{r} of system 1 are equal to

⁽³⁾ $-\operatorname{div} \mathbf{s}(\mathbf{r})$, while the losses at the corresponding point $q(\mathbf{r})$ of system 2 are $-q^{-1} \operatorname{div} \mathbf{s}(\mathbf{r})$, i.e. the absorption coefficient $k(\lambda)$ at the corresponding points of system 2 is q times smaller. The reflection coefficient at the boundaries of the particles and the binding medium in systems 1 and 2 is the same, since ε is the same.

Thus, under the transformations $k^{-1} \rightarrow qk^{-1}$, $l \rightarrow ql$, $a_i \rightarrow qa_i$, $\lambda \rightarrow q\lambda$ (λ is the wavelength of the incident light), $\varepsilon \rightarrow \varepsilon$, the scattering function F remains unchanged, i.e. $F(k^{-1}, l, a_i, \lambda, \varepsilon) = F(qk^{-1}, ql, qa_i, q\lambda, \varepsilon)$; consequently, F is a homogeneous function of degree zero ⁽⁴⁾

$$F = F[(ka)^{-1}, l/a, a_i/a, \lambda/a, \varepsilon]. \quad (2)$$

In the case where scattering occurs on identical particles $a_i = a$,

$$F = F[(ka)^{-1}, l/a, \lambda/a, \varepsilon]. \quad (3)$$

If for the binding medium $\varepsilon \neq 1$, then it also enters as an argument in F . According to Euler's theorem on homogeneous functions, F must satisfy the equation:

$$l \partial F / \partial l - k \partial F / \partial k + a \partial F / \partial a + \lambda \partial F / \partial \lambda = 0. \quad (4)$$

Formulas (2), (3), (4) have been obtained under the assumption of geometrical similarity of systems 1 and 2. However, if a large number of particles is concentrated in the scattering volume V , then one may expect that F will be the same for a broad class of systems differing only in the mutual arrangement of particles in the volume V , provided that this distribution is random in character. Indeed, experimentally measured values of F for systems with a large number of particles arranged in different ways (but in identical volumes) give coincident results. For example, the intensity of light scattered by fog remains unchanged, although the arrangement of particles in the fog changes continuously (the geometrical arrangement—

of particles corresponds to the maximum entropy of the system). Thus, F is a macroscopic characteristic of the system, arising as a result of statistical averaging of the scattering properties over a large number of particles. In a certain sense the scattering function F is analogous to thermodynamic functions.

What has been set forth above does not make it possible to determine the concrete form of F . Nevertheless, expressions (3), (4) are of essential importance, since any formula proposed for F must satisfy (3) and (4); if this is not so, then the proposed concrete formula is incorrect. Thus, (3), (4) restrict the class of functions admissible for F . For example, $F = F[(bk + da^{-1/2})r]$; $b, d = \text{const}$ cannot occur, whereas $F = F[(bk + da^{-1})r]$ encounters no objections from the standpoint of (3).

Let us consider several examples; for simplicity we shall assume that $\lambda a^{-1} \ll 1$ and that diffraction phenomena may be neglected; then $F = F[(ka)^{-1}, l/a, \varepsilon]$. Let the scattering particles be concentrated in a plane-parallel layer bounded by the planes $z = 0$ and $z = l$. In this case the expressions for R and T usually contain an exponential dependence on l . Under the assumption of complete homogeneity of the medium and complete diffuseness of the light [6],

$$R = R_\infty(1 - e^{-2Ll})/(1 - R_\infty^2 e^{-2Ll}), \quad T = (1 - R_\infty^2)e^{-Ll}/(1 - R_\infty^2 e^{-2Ll}). \quad (5)$$

Here R_∞ is the coefficient of diffuse reflection from an infinitely thick layer. The parameter L , according to (3), must have the form $L = a^{-1}f(ka, \varepsilon)$. If L is represented as a sum, then the only term $a^{-1}f_0(\varepsilon)$ will not depend on k , and $kf_1(\varepsilon)$ will not depend on a ; all the remaining terms depend both on k and on a . According to (3), $R_\infty = R_\infty(ka, \varepsilon)$, but it does not depend on k and a separately. If (5), for small k , $(1 - R_\infty)^2(2R_\infty)^{-1} \sim k$, then $R_\infty = 1 - \sqrt{ka} f_2(ka, \varepsilon)$, where $f_2(0, \varepsilon) > 0$.

For nonabsorbing ($ka = 0$) systems the expressions (5) are inapplicable, since $R_\infty = 1$. In this case $R(l/a, \varepsilon) + T(l/a, \varepsilon) = 1$, and the assumption of complete diffuseness of the incident, reflected, and transmitted light leads to

$$\left[T\left(\frac{l_1 + l_2}{a}, \varepsilon\right) \right]^{-1} = \left[T\left(\frac{l_1}{a}, \varepsilon\right) \right]^{-1} + \left[T\left(\frac{l_2}{a}, \varepsilon\right) \right]^{-1} - 1. \quad (6)$$

This functional equation is easily solved:

$$T\left(\frac{l}{a}, \varepsilon\right) = \left[1 + \varphi(\varepsilon)\frac{l}{a} \right]^{-1}, \quad R\left(\frac{l}{a}, \varepsilon\right) = \varphi(\varepsilon)\frac{l}{a} \left[1 + \frac{l}{a}\varphi(\varepsilon)\frac{l}{a} \right]^{-1}. \quad (7)$$

To estimate $\varphi(\varepsilon)$ in the case of dense packing of particles, we use model representations [6]. The model of a light-scattering medium is a set of la^{-1} planes with reflection coefficient $2r_0(1 - r_0)^{-1}$ from each; r_0 is the Fresnel reflection coefficient at the boundaries between the particles and the binding medium, averaged over the angle of incidence (and depending on the shape of the particles). Then $T(l/a, \varepsilon) = [1 + \delta(\varepsilon)2r_0(1 - r_0)^{-1}la^{-1}]^{-1}$, and it may be expected that, for a broad class of particle shapes, $\delta(\varepsilon)$ depends only weakly on ε .

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