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Abstract

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The Light Regime in the Depths of a Scattering Medium and the Spectroscopy of Dispersed Substances

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1. Studying the light regime in the depths of a scattering medium, V. A. Timofeeva ([1]) discovered a number of empirical regularities which may, in particular, be of significance for the absorption spectroscopy of dispersed substances. The present work is devoted to the theoretical substantiation and refinement of some of these regularities. In view of the need to take polarization effects into account, we shall use the terminology and apparatus connected with the description of the polarization state of a light beam by means of the Stokes vector-parameter ([2]).

2. Suppose that a homogeneous isotropic scattering medium fills the lower half-space $Z > 0$ and is illuminated from the side $Z < 0$. The optical properties of the medium are characterized by the absorption and scattering coefficients (cross sections) α and σ, referred to unit volume, and also by the scattering matrix $\frac{1}{4\pi} f_{ik}(\mu, \mu', \varphi, \varphi')\, d\Omega$, where $i, k = 1, 2, 3, 4$; $\mu = \cos\theta$; $\mu' = \cos\theta'$; θ and φ are the polar and azimuthal angles for the scattered beam (the z-axis serves as the polar axis); θ' and φ' are the same for the scattering beam, and $d\Omega$ is the element of solid angle formed by the scattered beam.

It is easy to show that, in the case of an isotropic medium, the scattering matrix must, in particular, possess the following integral properties:

$$\widetilde{F}_{12} \equiv \widetilde{F}_{13} \equiv \widetilde{F}_{14} \equiv \widetilde{F}'_{21} \equiv \widetilde{F}'_{31} \equiv \widetilde{F}'_{41} \equiv 0, \qquad \widetilde{F}_{11} \equiv \widetilde{F}'_{11} \equiv 1, \tag{1}$$

where

$$\widetilde{F}_{ik}(\mu') = \frac{1}{2} \int_{-1}^{+1} F_{ik}(\mu, \mu')\, d\mu, \qquad \widetilde{F}'_{ik}(\mu) = \frac{1}{2} \int_{-1}^{+1} F_{ik}(\mu, \mu')\, d\mu'; \tag{2}$$

$$F_{ik}(\mu, \mu') = \frac{1}{2} \int_{-1}^{+1} f_{ik}(\mu, \mu', \varphi - \varphi')\, d(\varphi - \varphi). \tag{3}$$

3. The propagation of light (or of fluxes of particles with spin 1/2) in a scattering medium is described by the transport equation (2)

$$\mu \frac{dS_i(z,\mu,\varphi)}{dz} =$$

$$= -kS_i(z,\mu,f) + \frac{\sigma}{4\pi}\sum_k \int_{-1}^{+1}\int_0^{2\pi} f_{ik}(\mu,\mu',\varphi-\varphi')\, S_k(z,\mu',\varphi')\, d\mu'\, d\varphi', \quad (4)$$

where $k = \alpha + \sigma$; $S_i(z,\mu,\varphi)$ are the components of the Stokes vector-parameter, characterizing the light flux in the direction μ, φ at depth z, with the vertical plane containing the direction μ, φ chosen as the reference plane (2).

From physical considerations it is obvious that, in the interior of a scattering medium, at a sufficient distance from its boundary, a stationary regime must be established in which the angular dependences of the quantities S_i do not change with depth and are free from the influence of the angle of inclination and the character of polarization of the light fluxes irradiating the outer boundary of the medium. At the same time, the azimuthal dependence of S_i must disappear. This assertion is valid if, in the scattering events, there is no degradation of the energy of the scattered particles or photons, and if the specific absorptivity of the medium

$$\beta \equiv \alpha/\sigma \quad (5)$$

is small. According to what has been said, in the interior of a weakly absorbing scattering medium there must be a separation of variables

$$S_i(z,\mu,\varphi) = R(z)s_i(\mu). \quad (6)$$

Substituting (3), (6), and (5) into (4), we obtain

$$-\frac{dR(z)}{dz} = -k'R(z); \quad (7)$$

$$(1+\beta)(1-\gamma\mu)s_i(\mu) = \frac{1}{2}\sum_k \int_{-1}^{+1} F_{ik}(\mu,\mu')S_k(\mu')\, d\mu', \quad (8)$$

where

$$\gamma \equiv k'/k \quad (9)$$

does not depend on μ.

It follows from (7) that

$$R(z) = e^{-k'(z-z_0)}. \tag{10}$$

Equation (8), generally speaking, may admit a number of values of k', but at sufficiently great depth all solutions will attenuate except the solution with the smallest k', which will correspond to the establishment of the stationary regime. Thus, in the interior the dependence $S_i(z)$ must be exponential, and the depth attenuation coefficient k' does not depend on the direction of the ray.

Integrating (8) with respect to μ and taking (1) into account, it is easy to show that for $\gamma = 0$, $\beta = 0$, and $\rho_i = \text{const} \cdot \delta_{i1}$, while the flux of radiation through a horizontal section is

$$\Phi \equiv \int_{-1}^{+1} \mu s_1(\mu)\, d\mu = 0.$$

If, however, the medium possesses absorption, then

$$\gamma = \frac{\beta}{1+\beta} \cdot \frac{1}{\bar{\mu}}, \tag{11}$$

where $\bar{\mu} = \Phi / \int_{-1}^{+1} s_1(\mu)\, d\mu$. For $\beta \ll 1$, i.e. $k \simeq \sigma$, it follows from (5) and (11) that

$$\alpha = k'\bar{\mu}. \tag{12}$$

4. Let us now consider the light regime for $\gamma \ll 1$. Put

$$s_i(\mu) = j_0 \left[\delta_{i1} + \gamma a_i(\mu) + \gamma^2 b_i(\mu) + \gamma^3 c_i(\mu) + ...\right]; \tag{13}$$

$$\beta = p\gamma + q\gamma^2 + r\gamma^3 + ... \tag{14}$$

Substituting (13) and (14) into (8) and collecting terms with like powers of γ, we obtain a system of equations for $a_i(\mu)$, $b_i(\mu)$, $c_i(\mu)$, etc. Integrating these equations with respect to μ, taking (1) into account, we find

$$p = 0, \qquad q = \frac{1}{2}\int_{-1}^{+1} \mu a_1(\mu)\, d\mu,$$

$$r = -q \cdot \frac{1}{2} \int_{-1}^{+1} a_1(\mu)\, d\mu + \frac{1}{2} \int_{-1}^{+1} \mu b_1(\mu)\, d\mu; \tag{15}$$

as a result, the equations themselves take the form:

$$a_i(\mu) = \mu \delta_{i1} + \frac{1}{2} \sum_k \int_{-1}^{+1} F_{ik}(\mu, \mu')\, a_k(\mu')\, d\mu',$$

$$b_i(\mu) = -q\delta_{i1} + \mu a_i(\mu) + \frac{1}{2} \sum_k \int_{-1}^{+1} F_{ik}(\mu, \mu')\, b_k(\mu')\, d\mu', \tag{16}$$

$$c_i(\mu) = (q\mu - r)\delta_{i1} - q a_i(\mu) + \mu b_i(\mu) + \frac{1}{2} \sum_k \int_{-1}^{+1} F_{ik}(\mu, \mu')\, c_k(\mu')\, d\mu'.$$

5. From (15) and (16) it is clear that the form of the functions $a_i(\mu)$, $b_i(\mu)$, $c_i(\mu)$ ..., and consequently the values of the coefficients q, r, ..., depend only on the form of the scattering matrix and uniquely determine the dependence of β on γ according to (14):

$$\frac{\beta}{q} = \gamma^2 \left(1 + \frac{2}{q}\gamma + ...\right), \tag{17}$$

whence

$$\gamma = \left(\frac{\beta}{q}\right)^t, \tag{18}$$

where

$$t \simeq \frac{1}{2}\left(1 - \frac{r\sqrt{\beta}}{q^{3/2} \ln \beta}\right), \tag{19}$$

or, taking (5) and (9) into account:

$$k' = \left(\frac{\alpha k}{q}\right)^+ \simeq \sqrt{\frac{\alpha k}{q}}. \tag{20}$$

From (10) and (13), for $\gamma \ll 1$ we find: for the intensity

$$I \equiv S_i = j_0 e^{k' z_0} [1 + \gamma a_1(\mu) + ...] e^{-k' z}; \tag{21}$$

for the degree of polarization [2]

$$P \equiv \frac{\sqrt{S_2^2 + S_3^2}}{S_1} \simeq \gamma\sqrt{a_2^2(\mu) + a_3^2(\mu)}; \tag{22}$$

for the degree of ellipticity [2]

$$g \equiv \frac{S_4}{S_1} \simeq \gamma a_4(\mu) \tag{23}$$

and for the angle of inclination of the plane of preferential polarization ψ_0 [2]

$$\operatorname{tg} 2\psi_0 = \frac{S_3}{S_2} \simeq \frac{a_3(\mu)}{a_2(\mu)}. \tag{24}$$

Thus, in the first approximation, the degree of polarization and the degree of ellipticity must be proportional to γ, while ψ_0 is independent of q. As for the shape of the body of intensities $I(\mu)$, as γ increases it is deformed, moving ever farther away from sphericity and becoming asymmetric, which corresponds to the increasing deviation from zero of $\overline{\mu}$, i.e., of the radiation flux Φ through a horizontal cross section.

Let us note that relations (10), (19), and (20) fully correspond to the results of the experiments of V. A. Timofeeva ([1]). As for expression (21), the information published by her is insufficient for its verification, and a more careful comparison with experimental data is necessary.

6. One of the main difficulties standing in the way of the development of spectroscopy of dispersed substances consists in the need for separate determination of α and σ of the scattering medium. In the case $\beta \gg 1$ this can be done by measuring its reflectance ([3]). Relations (12), (20), (21), and (22) indicate the possibility of a sufficiently reliable determination of α in the opposite case $\beta \ll 1$. V. A. Timofeeva ([1]) was the first to point out this possibility on the basis of an analysis of experimental material. It is very significant that, for $\beta \ll 1$, $\sqrt{\alpha k} \gg \alpha$, i.e., the use of relation (20) makes it possible to determine experimentally values of α much smaller than those accessible to measurement by other methods. It may be supposed that further theoretical and experimental investigations in this direction will create a solid basis for the development of absorption spectral analysis of dispersed substances. As is known, up to the present time such analysis has been practically unfeasible, with the exception of a few special problems whose experimental solution was found empirically.

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Note: Figure translations are in progress. See original paper for figures.