SPECIFIC FEATURES OF THE TEMPORAL STRUCTURE OF RADIATION IN DENSE SCATTERING MEDIA. PART III. THE USE OF DIFFUSION APPROXIMATIONS TO DESCRIBE PULSE SHAPE

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The experimental dependences of the duration of pulses and the angular structure of radiation in an optically thick scattering medium (τ = 10 to 70) are found to be essentially different from the results obtained using the diffusion approximations. Based on empirical data, a technique is proposed for describing the shape of the pulse polarization components using the formulas of the small-angle diffusion approximation, using the effective diffusion coefficient which increases as the photon dwell time in the medium is increased. The applicability limits of the asymptotic solutions are estimated.

In Part II of the present work[1] we have cited a few results obtained from experimental studies and computational modeling of pulse structure at different optical thicknesses τ of the scattering medium and different observation angles. Systematizing the results, which exhibit a complicated dependence on a number of geometric and optical parameters, it is desirable to be guided by inferences from approximate solutions of the nonstationary radiation transfer equation (RTE). Among the great number of techniques available for describing the propagation of pulsed radiation in the form of a narrow beam, of particular interest are approximations based on the concept of the diffuse character of the scattered radiation, including the diffusion approximation asymptotic limits[2,3] and the small-angle diffusion approximation (SADA).[4,5] Comparing our results with those obtained from the diffusion approximations we will look at the possibility of using them to estimate the pulse shape and stretching in optically thick ($\tau \gg 1$) scattering layers.

Asymptotic solutions describe the behavior of radiation under conditions of a weakly anisotropic and stationary angular intensity distribution, which is valid for $\tau \gg 1$ and at long times $u = \varepsilon ct \gg 1$, and, undoubtedly, can serve as a measure of adequacy of the results obtained at limitingly long times and large optical thicknesses. However, under our experimental conditions (τ = 10–70) the assumption of stationarity of the angular structure of the radiation is clearly not fulfilled. Figure 1 presents the angular structure of the brightness field (the component $I_{\parallel}$) at different times (in dimensionless units) measured by an azimuthal detector scanning (see Ref. 6, Fig. 1) at τ = 46 (solid curves) and τ = 15 (dashed curves). The figures near the curves correspond to values of the time u. The angular distribution changes with time and only on the trailing edge of the pulse (the time u = 32 corresponds to the 0.1 amplitude level at τ = 46) begins to approach $I(\mu) = 3(1 + 2\mu)/7$ (Ref. 7), its characteristic dependence for the asymptotic solution of the Milne stationary problem (dotted curves). The dependence of the pulse duration on the direction of observation vanishes only for $\tau > 60$.

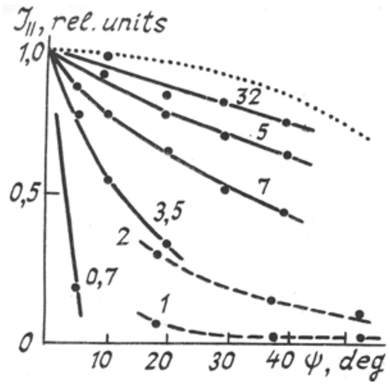

FIG. 1. Angular structure of the brightness field at different times.

A more adequate picture of the angular distribution of the radiation at different times is given by SADA, which holds in the cases where the variance of the angular intensity distribution exceeds the variance of the beam deflection angle in a simple scattering event, i.e.,

$$\overline{\gamma^2} \ll \overline{\theta^2} \ll 1, \quad (1)$$

where $\overline{\gamma^2} = \int \theta^2 g(\theta)\theta d\theta$, $\overline{\theta^2} = \int \theta^2 I(\theta)\theta d\theta$. To deal with actual scattering phase functions under SADA it is necessary to introduce certain effective parameters,[4] assuming that the radiation is scattered within the angle θ_0, chosen such that

$$\theta_0 \ll 1, \quad \beta = \int_{\theta_0}^{\pi} g(\theta)\, d(\cos\theta) \ll 1, \tag{2}$$

and the remaining part of the radiation is taken to be effectively absorbed. Then the true scattering and absorption coefficients σ_0 and κ_0 for the medium are replaced by their respective effective values:

$$\sigma = (1 - \beta)\sigma_0, \quad \kappa = \kappa_0 + \beta\sigma_0. \tag{3}$$

The medium must have an appreciable absorption coefficient κ_0, so that all the radiation scattered at $\theta > \theta_0$ can be treated as actually absorbed. These conditions are satisfied by certain hydrosols having extremely prolate scattering phase functions and appreciable absorption.[8] Cloud media hardly satisfy condition (2). Thus even for the cloud scattering phase function with $\bar{\mu} = 0.91$ (Ref. 9) and $\theta_0 = 0.35$ (20°) the fraction of the radiation scattered at large angles β is equal to 0.24. The parameter $\overline{\gamma_0^2} = \int_0^{\theta_0} \theta^2 g(\theta)\theta d\theta$ used in SADA becomes even more ambiguous, since its magnitude is a more sensitive function of θ_0 than the integrated phase function $(1 - \beta)$. Because of the low actual absorptivity typical of clouds,[9] ($\kappa_0/\sigma_0 \approx 10^{-3} \ll 1$] the radiation scattered at large angles, which is neglected in SADA may affect the value of the recorded signal at long times. This fact together with a complete ambiguity as to the choice of $\overline{\gamma_0^2}$ should make the use of SADA rather difficult. The dependences of the pulse width on the optical thickness of the scattering layer obtained in SADA are typical of the diffusion approximations:

$$\Delta t \approx \frac{1}{10}\frac{DZ^2}{c} = \frac{1}{10} T_0 \frac{\overline{\gamma_0^2}}{4}\tau. \tag{4}$$

Here $D = \sigma\overline{\gamma_0^2}/4$ is the diffusion coefficient; $T_0 = Z/c$ is the transit time of the layer. The numerical coefficient of 1/10 corresponds to a pulse width obtained by numerical computations of the Green's function[5] at the 0.5 amplitude level for a detector placed on the beam axis.

Let us now compare our experimental and calculated dependences of the pulse duration t on the optical thickness of the layer with the estimates given by the diffusion approximations. In Fig. 2 the value of $\tilde{\eta} = 10\Delta t(T_0\tau)$ is plotted along the ordinate. This is convenient because if relation (4) is fulfilled, $\tilde{\eta} = \overline{\gamma_0^2}/4 = \text{const}$. The experimental data for $I_\parallel$ and $I_\perp$ correspond to observation angles ψ of 0°(1), 30°(2), and 65°(3). Curves 4–7 illustrate calculations performed for different fields of view w and deviations from the beam axis α_0. Curve 8 presents the pulse width (underestimated by a factor of 3) given by the diffusion approximation[2,3] along the beam axis for $3(1-\bar{\mu}) = 0.42$ (the cloud model C1 at $\lambda = 0.45$ μm). Also shown for comparison are the results of numerical computations along the beam axis ($2\omega = 5°$) taken from Ref. 10 for scattering phase functions with $\bar{\mu} = 0.86$ (curve 9) and $\bar{\mu} = 0.95$ (curve 10). The dependence $\eta(\tau)$ has a dip at $\tau < 10$ where the pulse width for the chosen detector field of view ($\omega = 2°$) is determined by single scattering at the observation angle: $\Delta t = \frac{1}{2} Z\alpha\omega / c$ and is independent of τ. At higher τ the dependences for different ω and α converge to a straight line $\tilde{\eta}$ = const corresponding to the results of SADA for the diffusion coefficient computed from the total scattering phase function ($\theta_0 = 180°$).

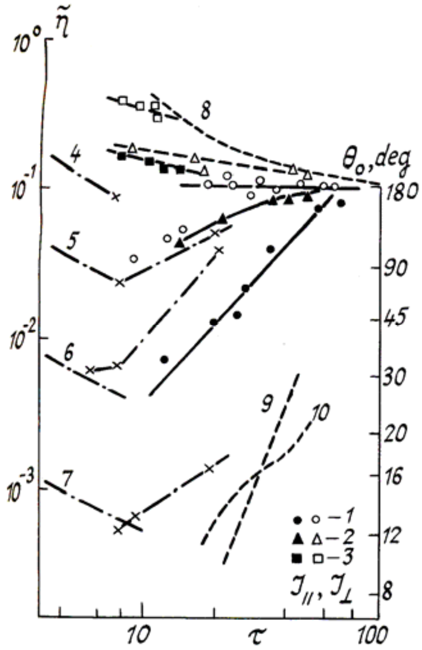

FIG. 2. The pulse width expressed in terms of the parameter $\tilde{\eta}$ as a function of the optical thickness of the layer. Experiment — curves 1–3. Calculation: α = 10°, *4* — ω = 80°, *5* — ω = 10°. *6* — ω = 1°, *7* — α = 2°, ω = 2°.

Within the investigated range of optical thicknesses the behavior of the asymptotic dependence is most closely followed by the broadening of the depolarized component ($I_\perp$) at large (ψ = 65°) observation angles off the beam axis. This is clear if we take account of the fact that the depolarized background is made up of photons which have undergone multiple scattering at large angles, is characterized by a weak angular structure, and can be described by the diffusion equation. When observed in the direction of the source the duration of $I_\perp$ grows linearly as τ is increased starting from τ = 20 (which corresponds to $\theta_0 = 180°$).

Note that the duration of the pulse at large optical thicknesses has been underestimated by a factor of 3 as compared to the pulse durations obtained by the asymptotic formulas.[2,3] Since the pulse durations at $\tau > 50$ become comparable to the time it takes the light to travel across the aerosol chamber, one of the possible reasons for this may be the limited size of the scattering volume bounded by chamber walls having low reflectivity.

The dependence of the broadening of the $I_{\parallel}$ component (which carries most of the energy of the total signal) on the optical thickness of the scattering layer differs significantly from the diffusion pattern. For $\tau \leq 10$, it can be one to two orders of magnitude smaller than that given by Eq. (4).

To remove the discrepancy between the SADA results and the experimental data it has been suggested by certain authors that the actual scattering phase function be replaced by a model one, which falls off more rapidly as $\theta \to 90°$. For example, Mooradian et al.[11] have proposed using a phase function of the form $g(\theta) = \frac{1}{\pi\overline{\gamma_0^2}} e^{-\theta^2/\overline{\gamma_0^2}}$. In this case $\overline{\gamma^2} = 1/\pi g(0°)$, which allows an adequate description of the experimental results[11] by SADA. However, this approach is highly arbitrary and depends on the phase function model chosen. Moreover, no constant value of the parameter $\overline{\gamma_0^2}$ can account for the observed dependence of $\tilde{\eta} = \overline{\gamma_0^2}/4$ on τ in the range of τ = 10–70. Therefore it appears better-founded to choose the parameter $\overline{\gamma_0^2}$ by varying the angle θ_0 bounding the effective part of the scattering phase function $g(\theta)$, thus making it possible to take into account the actual form of the phase function in the pulse width computations.

The necessity of varying the angle θ_0, as radiation penetrates into the medium, can be explained within the framework of the concept of the small-angle diffusion approximation. SADA assumes that the directional beam is attenuated solely as a result of the pure absorption κ, because scattering does not extract any energy from the beam ($\overline{\gamma^2}$ is small) but rather results in its angular and spatial spreading. Since the beam divergence increases with increase of the layer thickness, an ever-increasing part of the scattered radiation remains within the directional beam. This effect should be accounted for by decreasing the effective absorption κ, which can be done by increasing the angle θ_0 that limits that part of the phase function that makes the predominant contribution to the directional beam. As radiation penetrates into the medium and the angle θ_0 increases, the effective diffusion coefficient $D = \sigma\overline{\gamma_0^2}/4$ also increases. It is natural to assume that for the photons which form the directional beam $\sqrt{\theta_0}$ 'is proportional to the beam divergence θ^2. If the observations are made at an angle α in a direction close to the source, the majority of the photons, before entering the detector, would have to undergo a single scattering event at an angle close to α and multiple forward scattering at very small angles. Note that the effective angle θ_0 should not be smaller than the maximum angle $\theta_0 = \sqrt{\overline{\theta^2}} + \alpha + \omega$ at which the singly scattered photons enter the detector. These considerations explain the dependence of θ_0 on the detector parameters; however, in the stationary case the expression for $\overline{\theta^2}$ ($\overline{\theta^2} = 4DZ$) itself contains the parameter $\overline{\gamma_0^2}$, which can be determined provided θ_0 is known. This ambiguity makes it necessary to use empirical curves of the type shown in Fig. 2 to evaluate the temporal pulse stretching by formula (4). Estimating the angle θ_0 (right-hand scale, Fig. 2) and using the effective diffusion coefficient $D = \overline{\gamma_0^2}\sigma_0(1-\beta)/4$, we succeed both in describing the shape of the polarized pulse components and their amplitudes as functions of the optical thickness. Figure 3 compares the experimentally recorded pulses (solid lines) with those computed from the SADA formulas (dashed lines).[5] For the depolarized component $I_{\perp}$, the angle θ_0 was chosen to be 180°, and for the polarized component $I_{\parallel}$, it followed the dependence on r given in Fig. 2. The intensity values expressed as $W/m^2/s/sr$ were calculated in SADA. Since absolute intensities were not obtained experimentally, the scale was chosen assuming the equality of the $I_{\perp}$ amplitudes at τ = 46. It is evident that the fractional change in the polarized component amplitude is adequately described by the diffusion approximation. However, SADA results in a longer delay of the pulse maximum for the same pulse duration.

The change in θ manifests itself in the intersection of the $\tilde{\eta}(\tau)$ curves for different phase functions, which is clearly visible from the results of the numerical calculations[10] (curves 9 and 10, Fig. 2). A more prolate phase function with $\overline{\mu}$ = 0.95 is characterized by a more rapid growth of the parameter $\overline{\gamma_0^2}$. As a result, the pulse stretching for this phase function at $\tau < 25$ is larger than for a less prolate phase function. At the same time the value of $\overline{\gamma_0^2}$ (θ_0 = 180°) is larger for the phase function with $\overline{\mu}$ = 0.86, and the point of intersection of the dependences $\tilde{\eta}(\tau)$ corresponds to θ_0 = 16°, for which the values of $\overline{\gamma_0^2}(1-\beta)$ are the same for the two phase functions.

A series of calculations of the impulse response function (IRF) of the scattering layer for optical thicknesses $2 < \tau < 10$ in the paraxial region[13] for phase functions that differ strongly in their degree of prolateness ($\overline{\mu}$ = 0.7–0.98) have shown that the use of constant, time-independent parameter $\overline{\gamma_0^2}$ in SADA results in a pulse shape that falls off faster at long times than the asymptotic limit. Taking into account the fact that the angular variance of the photons is related to the time of arrival ($\overline{\theta^2} = 4(ct - Z)/Z$, Ref. 5), it can be assumed that the IRF at the time t is formed mainly by photon scattering within a certain effective part of the phase function (within the angles $0 < \theta < \theta_0 = \sqrt{\overline{\theta^2}}$), the effective range of angles increasing with the delay of the time of arrival. In this case, the dependence of θ_0 on the optical thickness and the observation angle is automatically accounted for by varying the photon arrival time. It has been shown elsewhere[13] that at thicknesses and times for which the angular photon spectrum does not differ appreciably from the angular width of the single scattering phase function, the SADA equations satisfactorily approximate the results obtained from calculations which make use of the photon arrival time chosen so that its form differs from that given in Ref. 5 by a numerical coefficient:

$$t = Z\left[1 + \theta_0^2/25\right]/c. \tag{5}$$

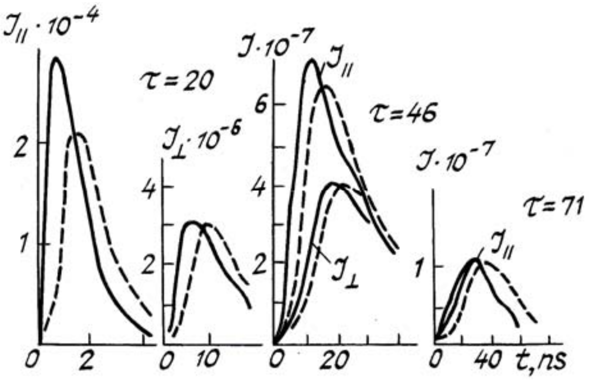

FIG. 3. Comparison of experimentally recorded pulses with those calculated by the SADA formulas

Expression (5) sets a lower bound on the effective angle θ_0 since, for a transverse displacement of the detector from the beam axis by the amount $r_\perp$, the observation time should exceed the arrival time of the first photon $\left(T_0 = \sqrt{Z^2 + r_\perp^2}/c\right)$. As a result, the range of the effective angles θ_0 should increase with increase of the observation angle. After choosing the angle θ_0, we determine the parameter $\overline{\gamma_0^2}$ appearing in the expression for the diffusion coefficient, proceed from there to the effective optical constants (3), and finally compute the IRF for the time $t(\theta_0)$, numerically integrating the expression for the Green's function.[5] For sufficiently long times, when $\theta_0 > 8°$, we use a two-parameter dependence of the form

$$t = \frac{Z}{c}\left[1 + \frac{\beta(8°)}{\beta(\theta_0)}\frac{\theta_0^2}{25}\right], \tag{6}$$

where $\beta(\theta)$ is given by Eq. (2) for a chosen scattering phase function. This dependence is evidence of the fact that the time delay depends on the degree of asymmetry of the phase function. The proposed technique implies that the actual phase function must be assigned with a high angular resolution $\Delta\theta = 0.5°$. Such an angular step is sufficient to determine the temporal structure of the IRF for optical thicknesses $2 < \tau < 10$, an angular displacement from the beam axis $\alpha \leq 10°$, and observation angles $\theta \leq 1.5\alpha$ with respect to the direction of the source.

When computing the pulse shape for the detector field of view w and the chosen experimental conditions, assuming a region where the detector field of view and the light beam intersect, it should be kept in mind that the shape of the leading edge is determined by the singly and multiply forward scattered radiation and cannot be described by the diffusion approximation for $\tau \leq 10$. Let us check on the validity of the SADA dependences for the trailing edge of the pulse. Figure 4a illustrates the calculated shape of a pulse transmitted through a layer with an optical thickness $\tau = 8$ for detector fields of view $\omega = 1°$ (solid line) and $\omega = 10°$ (dashed line). The detectors are placed at different angular distances from the beam axis and 6000 m away from the source. The time scale is counted off from the moment the first photon, arrives at the detector $T_0 = Z/c$. The greater the distance from the axis, the lower is the pulse amplitude and the longer is the time delay for the arrival of the peak pulse. At long times the pulse shapes overlap to form a single curve which decays as $t^{-5/2}$. Such a decay of the intensity in the asymptotic solution is typical of long times limited by the constraint $\tau \ll u \ll 3(1 - \bar{\mu})\tau^2$ (see Ref. 2). That the illumination at different points of a sphere with the radius Z tends to the same value as $t \to \infty$ to is quite clear from the optical reciprocity theorem.[7] This makes it different from the picture found in the transverse plane where, according to the experimental data[8] and small-angle approximation estimates,[5] the illumination maximum is somewhat shifted from the beam axis at large t.

The results obtained from the SADA computations[5] are given in Fig. 4b. The solid lines correspond to calculations by proposed relation (6), and the dot-dash lines show the dependence $\theta_0^2 = 5(ct - Z)/Z$. The figures near the solid curves indicate the angles θ_0 corresponding to the chosen dependence $t(\theta_0)$. As can be seen from the figure, the choice of the numerical coefficient determines whether the rate of decay of the intensity on the trailing edge of the pulse follows a $t^{-5/2}$ law and the radiation curves converge to a single profile for different angular distances from the detector to the beam axis. The introduction of the indicated dependence makes it possible to adequately describe the decay of the pulse within 1–1.5 orders of magnitude of the maximum value. Beyond these levels the SADA formulas yield a faster decay of radiation than that predicted by numerical calculations. It should be noted that within the range of optical thicknesses ($\tau \le 10$) studied the angular distribution of the pulse amplitudes depends strongly on the detector field of view.

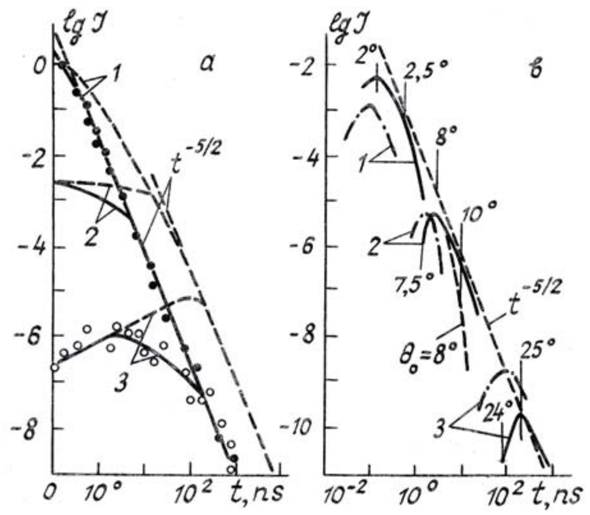

FIG. 4. Pulse shapes obtained by numerical calculations (a) and by SADA (b) for different detector angular displacements from the beam axis: 1 —α = 0.5°; 2 — α = 2°; 3 — α = 10%.

Rogozkin[14] has shown that the change of the numerical coefficient in Eq. (5) is related to specific applications of the diffusion approximations to scattering media in which the scattering phase functions decay with insufficient rapidity (slower than θ^{-4}) with increasing scattering angle. Moreover, the exact value of the coefficient depends on the form of the scattering phase function and, when the angle θ_0 is made to be time-dependent, the SADA solution agrees with the small-angle approximation. The expressions given in Ref. 14 are in qualitative agreement with our results.

Thus it is possible to distinguish optical thicknesses at which different approaches must be used to calculate the pulse duration.

1. For small τ, where the angular spreading of a beam due to multiple scattering does not exceed the angular aperture of the detector ($\overline{\theta^2} \ll \omega^2$), the pulse duration is governed by single scattering within the detector field of view, and is independent of the optical thickness. For the conditions of our model experiment, $\tau \le 10$.

2. In the transition range (τ = 10–70) the pulse shape as a function of time is described by SADA, assuming that the effective diffusion coefficient $D = \overline{\gamma_0^2}\sigma / 4$ increases with τ and the photon dwell times in the medium increase.

3. For $\tau > 70$, the dependence of the pulse duration on the observation angle vanishes, and asymptotic solutions become valid.

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