Light-reflection spectrum of a turbid medium. Role of interference effects

V. V. Afonin and Yu. M. Gal'perin

Ioffe Physicotechnical Institute, USSR Academy of Sciences

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The interference contribution to the spectrum $R(\omega)$ of unpolarized light reflected backwards from a turbid medium is considered. It is shown that this contribution is highly sensitive to the presence of internal degrees of freedom of the scattering particles. Thus, the back-reflection has a line spectrum even when the distance between the scattering-particle levels is much less than the reciprocal free-path time of the light wave. Analytic expressions are obtained for $R(\omega)$ at various relations between the frequency of the transition between the particle levels and the broadening due to the motion of the latter in real space. The angular dependence of the interference contribution to the back-reflection coefficient is analyzed. In contrast to the integrated intensity, the wings of the reflection spectrum are determined exclusively by multiple scattering. An investigation of $R(\omega)$ makes it therefore possible to separate in experiment the multiple-scattering contribution and to obtain information on the internal structure of the scattering particles.

1. INTRODUCTION

Very many studies have been made of wave propagation in media with random inhomogeneities. It is known[1–3] that on satisfaction of the condition

$$kl \gg 1 \tag{1}$$

(where $k = \Omega/c$ is the wave vector and l is the wave absorption length) the wave-energy transport is described in the zeroth approximation by intensity-transport equations in which interference effects are neglected.

Interference effects in multiple scattering of light have recently become the subject of intensive study. Interest in this problem is due to the possibility of observing for light waves weak-localization effects heretofore investigated for electrons in metals.[4,5] It has been found that experimental observation of these phenomena for light waves is preferable, for in the case of electrons, as a rule, the important inelastic processes are those leading to phase relaxation.

The principal interference contribution in scattering by stationary inhomogeneities was considered in Refs. 6–11. It is shown in them that the interference contribution to the total cross section is small relative to the parameter (1). For backscattering, however, it is of the order of the differential scattering cross section. The reason is that waves passing along the same trajectories in opposite directions acquire in the case of elastic scattering equal phases and add up coherently when scattered backwards.

A number of experiments on coherent backscattering of light in various media have already been performed by now.[12–16] The subjects of these references were the angular dependence of the scattered-light line as well as polarization effects. These effects were theoretically considered in Refs. 12–25.

The statement that the waves add up coherently in backscattering is correct only if one neglects processes that violate the invariance of the scattering to time reversal. In particular, interference is upset by the motion of the scattered particles. Since the forward and backward waves pass through the same inhomogeneities at different instants of time, motion of the particles causes the phase shifts of these waves to be different, and the interference is suppressed as a result. This phenomenon was considered in Ref. 26 for the case of sound scattering by bodies located on a wavy sea surface under conditions when single scattering by the surface and by the scatterer is significant. For multiple scattering in a volume this phenomenon was investigated in Ref. 17.

Our present purpose is to consider the spectrum of backscattered light. The form of this spectrum is determined by the nonstationary behavior of the scattering system [the spectrum for scattering by static inhomogeneities has the form $\delta(\omega)$, where ω is the frequency difference between the incident and scattered waves]. It must be noted here that, in contrast to the integrated intensity, the reflection-spectrum wings are governed exclusively by multiple scattering. A study of the spectrum of the reflection coefficient $R(\omega)$ permits therefore experimental separation of the contribution of the interference effects.

We shall assume that the scattering particles have an internal degree of freedom. We shall simulate this degree of freedom by a two-level system with distance $\hbar\omega_0$ between levels and a mobile mass center. Physically such a system can be, for example, the rotation of a scattering particle in a suspension. We assumed that $\hbar\omega_0 \ll T$, where T is the temperature of the scattering system. [If several levels turn out in a real situation to be in the region of energies lower than T, our results will be somewhat modified, namely, expressions (5)–(7) below for the phase difference must contain all the possible transitions.]

The invariance of the system of time reversal is thus violated in the system considered both on account of particle motion and on account of elastic scattering of the waves by the particles. If the condition $\omega_0\tau_{12} \gg 1$ is met, where τ_{12} is the characteristic wave relaxation time relative to a transition between the levels of the scattered particles, each scattering act with a transition between levels leads to collapse of the phase of the wave. If furthermore $\tau_{12} \approx \tau = l/c$, where l is the wave free path time and c is its velocity, the interference contribution to the multiple scattering is completely suppressed. The width of the scattered-wave spectrum is then of the order of ω_0. Of much greater interest is the case

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$$\omega_0\tau_{12}\ll 1, \qquad (2)$$

which we shall in fact consider hereafter.

In this situation it can be assumed that the light propagates in a medium with a dielectric constant $1+\delta\varepsilon(\mathbf{r},t)$. The correction $\delta\varepsilon$ is small, changes rapidly with change of the spatial variable, and has a weak time dependence.[1)] We examine now how this temporal change influences the spectrum of the reflection backwards. We disregard for simplicity its angular dependence (see the sections that follow), as well as all effects connected with light polarization.

To obtain the spectrum of the reflected light we must calculate the unequal-time correlator $\langle A(t)A^*(0)\rangle_{\delta\varepsilon}$, where A is the amplitude of the light wave and the angle brackets denote averaging over all possible configurations of the correction to the dielectric constant taken at some instant of time. Each of these amplitudes can be represented in the form $A=\Sigma_i A_i$, where A_i is the amplitude of the light wave propagating along the ith trajectory. These trajectories consist of straight-line segments along which the light wave propagates, and have kinks at the scatterer locations. We know that the interference contribution is determined by closed trajectories, and the main contribution to the correlator is made by amplitude products of form $A_+(t)A^*_-(0)$, which describe motion along one and the same trajectory but in opposite directions. In this case the difference between the starting times in the loop in the two directions is equal to t (we assume $t>0$). Let the light negotiate the given closed trajectory in a time t_0. Let the parameter of the coordinates $\mathbf{r}_1$ of the loop be the time t_1 needed for the light to travel clockwise from the intersection point to the point $\mathbf{r}_1$. Moving in the opposite direction, the light reaches the point $\mathbf{r}_1(t_1)$ at the time t_0-t_1. We assume that the light started to move along the loop clockwise at the instant $-t_0/2$, and in the opposite direction at the instant $t-t_0/2$. It reaches then the point $\mathbf{r}_1(t_1)t$ the respective instants of time $t-t_0/2$ and $t+t_0/2-t_1$. By virtue of the inequality (1) and the smallness of $\delta\varepsilon$, the amplitudes A have a semiclassical form and the correction to the phase of a plane wave moving clockwise can be written as

$$\varphi^+=-\frac{\Omega_k}{2}\int_{-t_0/2}^{t_0/2} dt_1\,\delta\varepsilon\left(\mathbf{r}_1(t_1),t_1-\frac{t_0}{2}\right), \qquad (3)$$

while for the wave moving in the opposite direction we have

$$\varphi^-=-\frac{\Omega_k}{2}\int_{-t_0/2}^{t_0/2} dt_1\,\delta\varepsilon\left(\mathbf{r}_1(t_1),t+\frac{t_0}{2}-t_1\right), \qquad (4)$$

where Ω_k is the frequency of the incident wave.

We now must average the quantity $\exp[i(\varphi^+-\varphi^-)]$ over all the realizations of $\delta\varepsilon$. We assume that the phase difference has a Gaussian distribution:

$$\langle\exp(i\Delta\varphi)\rangle_{\delta\varepsilon}=\exp[-{}^1/_2\langle(\Delta\varphi)^2\rangle_{\delta\varepsilon}]. \qquad (5)$$

The problem reduces thus to calculation of the correlators $\langle\delta\varepsilon(\mathbf{r}_1(t_1),t)\cdot\delta\varepsilon(\mathbf{r}_2(t_2),0)\rangle$ with different spatial and temporal arguments. Since the corrections to the dielectric constants depend strongly on their spatial arguments and are independent at different points of space, we assume that the coordinate correlator is a δ-function and depends little on the time as a parameter. (We want to describe quasielastic scattering.) In this case it can be represented in the form (cf. Ref. 21)

$$\langle\delta\varepsilon(\mathbf{x}_1(t_1),t)\delta\varepsilon(\mathbf{x}_2(t_2),0)\rangle_{\delta\varepsilon}\sim\frac{\Delta(t)}{k^2l}\delta(\mathbf{x}_1-\mathbf{x}_2)$$

$$\sim\frac{\Delta(t)}{k^2cl}\delta(t_1-t_2), \qquad (6)$$

$$\Delta(t)=\cos(\omega_0 t)\exp(-D_sk^2t). \qquad (7)$$

Here D_S is the diffusion coefficient of the scatterers, the factor $\cos\omega_0 t$ corresponds to a transition between their energy states, and $\exp(-D_sk^2t)$ describes motion in real space. We have assumed, here, of course, that during the time of motion along the loop the molecules manage to collide many times with one another or with the solvent molecules, i.e., the conditions

$$kL\ll 1,\qquad D_sk^2\tau\ll 1,\qquad D_sk^2t\ll 1$$

are met, where L is the scatterer mean free path.

Note that at $kL\gg 1$ the values of $\Delta(t)$ would vary like $\exp(-k^2\langle v_s^2\rangle t^2)$, where $\langle v_s^2\rangle$ is the mean squared scatterer velocity, corresponding to a Doppler broadening of the levels on account of the particle motion. The calculation is in this case similar to the one given below, and we omit it for brevity.

Ultimately we get

$$\frac{\langle(\Delta\varphi)^2\rangle}{2}\sim\int_{-t_0/2}^{t_0/2}\frac{dt}{\tau}[1-\cos[\omega_0(t+t_0-2t_1)]]$$

$$\times\exp[-D_sk^2(t+t_0-2t_1)]. \qquad (8)$$

Only trajectories on which $\langle(\Delta\varphi)^2\rangle_{\delta\varepsilon}\ll 1$ contribute to interference effects. (It follows here from the derivation that $t,t_0\gg\tau$. For an approximate discussion we assume for simplicity that all the free-path times are the same.) This is possible only if the second term of (8) differs little from unity.

We must now establish the dependence of the pre-exponential factor on the time t_0 of motion over the loop. We take into account the small inelasticity only to the extent that it leads to relaxation of the light-wave phase. The pre-exponential factor can therefore be estimated in the same way as in the elastic approximation. In this case both pre-exponentials are independent of the time t and are equal. Their dependence on the time of motion over the loop is determined by the probability of the light returning to the initial time after the time t_0, i.e., it is proportional to $t_0^{-1/2}$. We get as a result

$$\langle A(t)A^*(0)\rangle_{\delta\varepsilon}$$

$$\sim\int_\tau^\infty\frac{dt_0}{t_0^{1/2}}\exp\left\{-\int_{-t_0/2}^{t_0/2}\frac{dt_1}{\tau}[1-\cos(\omega_0(t+t_0-2t_1))\right.$$

$$\left.\times\exp(-D_sk^2(t+t_0-2t_1))]\right\}. \qquad (9)$$

To obtain a final expression for $R(\omega)$ we must take a Fourier transform with allowance for the parity of the correlator (9) under the substitution $t \to -t$.

Let us analyze the $R(\omega)$ spectral dependence for the parameters of greatest interest from our point of view. A more complete and accurate analysis will be given in the sections that follow. We begin with the simplest situation, in which the dominant phase-relaxation mechanism is motion of the scatterers in space. It is so fast that the characteristic time t_0 is insufficient for the scatterer to go over to another energy state. The phase relaxes in this case in a time $\tau_2 \sim (\tau/D_s k^2)^{1/2}$ (Ref. 17; see also Ref. 27), and the condition postulated above can be written in the form $(\omega_0/D_s k^2)^{3/2}(\omega_0\tau)^{1/2} \ll 1$. We have then

$$\langle(\Delta\varphi)^2\rangle \sim \tau_2^{-2}(t_0^2+2t_0|t|),$$

and

$$R(\omega) \sim \int_\tau^\infty \frac{dt_0}{t_0^{1/2}} \frac{\exp(-t_0^2/\tau_2^2)}{(\omega\tau_2)^2+(2t_0/\tau_2)^2} \sim \begin{cases} \omega^{-1/2}, & \omega\tau_2 \ll 1 \\ \omega^{-2}, & \omega\tau_2 \gg 1 \end{cases} . \tag{10}$$

This expression is valid in the frequency region $\tau^{-1} \gg \omega \gg D_s k^2$; it is important here that the characteristic times are $t_0 \gg \tau$.

Let us consider the inverse limiting case $1 \gg (\omega_0\tau)^{1/3} \gg D_s k^2/\omega_0$. In this situation the expression for $R(\omega)$ can be written in the form

$$R(\omega) \sim \int_{-\infty}^{\infty} dt\, e^{i\omega t} \int_\tau^\infty \frac{dt_0}{t_0^{3/2}} \times \exp\left\{-\int_{-t_0/2}^{t_0/2} \frac{dt_1}{\tau}[1-\cos(\omega_0(t+t_0-2t_1))] - \frac{1}{\tau_2^2}(t_0^2-2t|t|)\right\}. \tag{11}$$

It is seen from (11) that as $\tau_2 \to \infty$ the correlator $\langle A(t)A^*(t)\rangle$ is periodic in t, and therefore the interference effects are significant near the points $t_n = 2\pi n/\omega_0$, where $\langle(\Delta\varphi)^2\rangle \ll 1$. The physical reason for this is simple. If scatterer energy-level spacing is $\hbar\omega_0 \ll T$, the probability of finding a scatterer in a given state oscillates with a period $2\pi/\omega_0$. The light incident on the scatterer at instants of time that differ by t_n is scattered then by a molecule in the same state, and the interference contribution to the unequal-time correlator near these points is not small.

We make in (11) the change of variable $t = t_n + \delta t$. If the characteristic width of the peak is less than ω_0^{-1} the integral with respect to t can be represented as a sum of integrals around each peak, and by virtue of the rapid convergence of each term we can let the integration limits in them tend to $\pm\infty$:

$$\int dt \to \sum_{n=-\infty}^{\infty} \int_{-\infty}^{\infty} d\,\delta t.$$

We consider next for simplicity a situation in which the peak width is determined by scatterer transitions from one energy state to another (the second term of the expansion $\cos[\omega_0(t+t_0-2t_1)]$). This damping is nonlinear in the times t and t_0, and the characteristic phase-relaxation time due to these processes is $\tau_3 \sim \tau(\omega_0\tau)^{-2/3}$ (Refs. 17 and 28). We are now interested in the case $\tau_3 \ll \tau_2$. We retain in the phase relaxation due to the scatterer motion only to the extent to which it violates the correlator periodicity, i.e., we retain only the term $2|t_n|t_0/\tau_2^2$. The sum over n [which we denote by $F(\omega)$] is then easily calculated, and the integral over δt is simply Gaussian. Ultimately,

$$R(\omega) \sim \int_\tau^\infty \frac{dt_0}{t_0^2} F(\omega, t_0) \exp\left[-\frac{t_0^3}{3\tau_3^3} + \frac{1}{4t_0}\left(i\omega\tau_3^{3/2} - \frac{t_0^2}{\tau_3^{1/2}}\right)^2\right], \tag{12}$$

where

$$F(\omega, t_0) = \left[1-\exp\left(\frac{2\pi i\omega}{\omega_0} - \frac{2\pi t_0}{\omega_0\tau_2^2}\right)\right]^{-1} + \exp\left[-\frac{2\pi i\omega}{\omega_0} - \frac{2\pi t_0}{\omega_0\tau_2^2}\right]\left[1-\exp\left(-\frac{2\pi i\omega}{\omega_0} - \frac{2\pi t_0}{\omega_0\tau_2^2}\right)\right]^{-1}. \tag{13}$$

We consider the spectrum region in which peaks exist. They are due to poles of the function $F(\omega)$, and contributions to them are made by small enough trajectories $t_0 \ll \omega_0\tau_2^2$. Let us examine the form of an individual peak. To this end we put $\omega = n\omega_0 + \delta\omega$, $\delta\omega \ll \omega_0$ and examine for the time being only the region $\omega\tau_3 \ll 1$. In this case

$$F(\omega, t) \sim t_0/[t_0^2+(\delta\omega\tau_2^2)^2].$$

The integration region is cut off by $\min(\tau_3, \omega_0\tau_2^2)$ on the side of large t_0 and by $t_{max} = \max(\tau, \omega^2\tau_3^3)$ on for small t_0 (see the argument of the exponential). To be specific, we assume that $\tau_3 \gg \omega_0\tau_2^2$. Expression (13) can then be represented in the form

$$R(\omega) \sim \int_{t_{max}}^{\omega_0\tau_2^2} \frac{dt_0}{t_0} \frac{1}{t_0^2+\delta\omega\tau_2^2} \sim \frac{1}{(\delta\omega\tau_2^2)^2} \ln\left(1+\frac{\delta\omega\tau_2^2}{t_{max}}\right). \tag{14}$$

It is valid under the condition $t_{max} \ll \omega_0\tau_2^2$. It is seen from (14) that in the frequency region $\omega^2\tau_3^3 \ll \tau$ the peaks have equal heights and widths. The width is of the order of $D_s k^2$. The ratio $R(\delta\omega=0)/R(\delta\omega \approx \omega_0)$ is of the order of $(\omega_0\tau_2^2/\tau)^2/\ln(\omega_0\tau_2^2/\tau) \gg 1$.

In the frequency region $\omega^2\tau_3^3 \gg \tau$ the peaks begin to broaden and their amplitudes begin to decrease. The reason is that nonlinear damping begins to increase here. The characteristic peak width $\overline{\delta\omega}$ becomes of the order of $\omega^2\tau_3^3/\tau_2^2$ and they are resolved at $\overline{\delta\omega} \ll \omega_0$, i.e., to frequencies lower than $(\omega_0\tau_2^2/\tau_3^3)^{1/2}$. The peak amplitudes decrease in this frequency region like ω^{-4}. At higher frequencies the peaks coalesce to form a continuum. Expression (14) is no longer valid here. The continuum is described by trajectories with $t_0 \gg \omega_0\tau_2^2$, and F is of the order of unity in this region. In the upshot we have at frequencies $(\omega_0\tau_2^2/\tau_3^3)^{1/2} \ll \omega \ll 1/\tau_3$

$$R(\omega)\sim\int\limits_{\omega^2\tau_3^3}^{\tau_3}\frac{dt_0}{t_0^2}\sim\omega^{-2}. \qquad (15)$$

We proceed now to the highest-frequency region $\omega\tau_3\gg 1$. Following the change of variable $t_0\to t_0(\omega\tau_3^3)^{-1/2}$ it is expedient to reduce in this region expression (12) to the form

$$R(\omega)\sim\frac{1}{(\omega\tau_3^3)^{1/2}}\int\limits_a^\infty\frac{dt_0}{t_0^2}\exp\left[-\frac{(\omega\tau_3)^{3/2}}{4}\left(\frac{t_0^3}{3}+2it_0+\frac{1}{t_0}\right)\right],$$

$$a\sim(\omega\tau_3)^{-1/2}\ll 1. \qquad (16)$$

This integral has a saddle point in the complex plane and its modulus is of order unity, so that with exponential accuracy $|R(\omega)|\propto\exp[-(\omega\tau_3)^{3/2}]$.

One can thus expect in experiment a highly variegated behavior of the scattered-light line, depending on the ratio of the characteristic times of the problem. The following is noteworthy. It is known that the interference-contribution intensity averaged over the spectrum is determined by processes of low multiplicity and only a small temperature-dependent increment is determined by multiple scattering. At the same time, the scattering line wings are determined solely by multiple scattering. A study of the spectrum of the interference contribution to the reflection coefficient seems to us quite interesting for the analysis of phase-relaxation processes and also for the study of the scatterer spectrum, since it permits resolution of levels that differ by less than $\hbar/\tau_{12}$.

2. GENERAL EXPRESSIONS FOR THE INTERFERENCE CONTRIBUTION TO THE UNEQUAL-TIME CORRELATOR

We now formulate briefly the method of calculating $R(\omega)$. To make the results not too unwieldy, we confine ourselves to the scalar model customarily used for effective depolarization of the radiation. [Note that polarization effects are very important for the study of backward reflection.[21] This simple model, however, yields correct frequency dependences for $R(\omega)$ in the case of experiments with normally incident depolarized light.] In the context of this model the field is characterized by one component A of the vector potential. We must take into account effects connected with light scattering by two-level systems. This can be done by introducing into the equation for A an additional term $g_{ik}\psi_i^*\psi_k$:

$$-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}A+\Delta A-g_{ik}\psi_i^*\psi_k A=-\frac{4\pi}{c}j_0, \qquad (17)$$

where ψ_i and ψ_k are the scatterer-field operators over which averaging is to be carried out; i and k are the numbers of the states, and g_{ik} is the coupling constant. In this case we can write down the formal solution of Eq. (17) by expressing the potential A in terms of the extraneous current j_0:

$$A(\mathbf{r})=-c\int d^3\mathbf{r}_1\,D(\mathbf{r},\mathbf{r}_1)j_0(\mathbf{r}_1). \qquad (18)$$

Here $D(\mathbf{r},\mathbf{r}_i)$ is the Green's function of Eq. (17), for which we can write the usual integral equation that permits iteration in powers of $g_{ik}\psi_i^*\psi_k$. We must write down now the diagram expression for the correlator $A(\mathbf{r}_1)A^*(\mathbf{r}_2)$ and average it thermodynamically over the fields ψ. We assume a polarization interaction of the light with the scatterers. Writing down the bare propagator of the light in the form standard for bosons:

$$D_R^0(\Omega,\mathbf{k})=(\Omega-\Omega_k+i\delta)^{-1}-(\Omega+\Omega_k+i\delta)^{-1}, \qquad (19)$$

we can write, after averaging over the fields ψ, the following expression for the vertex:

$$g_{ik}=\Omega\alpha_{ik},$$

where $2\alpha_{ik}$ is a matrix made up of the molecule polarizability operators.

In the diagrams we represent the bare photon propagator by a thin solid line, and the bare vertex g corresponding to interaction of a photon with a scattering particle, by a light square. Since we are considering particles with two different internal states, the vertex g is a matrix over the states 1 and 2 of the scattering particle. The self-energy part of the photon Green's function is shown in Fig. 1, where the dashed lines represent the propagators of the scattering particles, and the dark square represents the total vertex part. Note that a diagram of type 2b does not enter in the averaged equations. It can be verified that in the limit of interest to us the total vertex part contains only diagrams corresponding to interaction between particles. We can therefore introduce an effective scattering-particle propagator represented by the diagrams of Fig. 1c (summation over the indices i and k is implied).

We shall study the quantity

$$I(\mathbf{r},t_+|\boldsymbol{\rho},t_-)=\langle A(\mathbf{r}+\boldsymbol{\rho}/2,t_++t_-/2)A^*(\mathbf{r}-\boldsymbol{\rho}/2,t_+-t_-/2)\rangle_\psi, \qquad (20)$$

where the angle brackets denote averaging over an ensemble of scatterers and over the density matrix of the radiation. In the general case, I is a matrix in the vector indices of A. In the case of monochromatic spactially coherent radiation we have $I=|A_0|^2\exp[i(\mathbf{k}\boldsymbol{\rho}-\Omega t_-)]$, where A_0 is the wave amplitude, k the wave vector, and Ω the frequency. For $\boldsymbol{\rho}=0$ and $t_-=0$ the value of I is proportional to the light intensity at the point $\mathbf{r}$ and at the instant t.

We assume that a plane monochromatic wave with frequency and wave vector $\mathbf{k}$ is incident on the boundary of the medium. To determine the scattered wave intensity $I(\mathbf{r},t_+|\mathbf{k},\Omega)$ ($\mathbf{k},\Omega$ are the Fourier components of I in the variables $\boldsymbol{\rho}$ and t_-) we must analyze the integral transport equation shown by Fig. 2a. In this figure the thick lines cor-

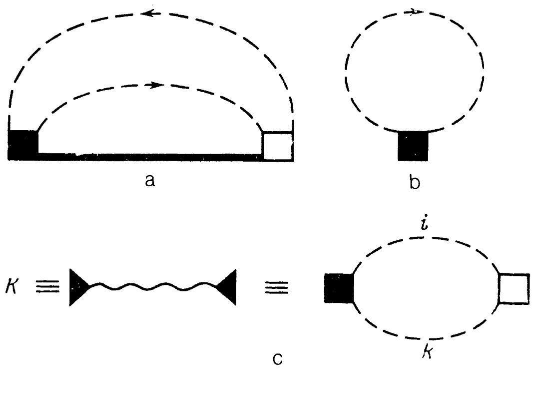

FIG. 1.

respond to averaged Green's functions of the waves, and the square corresponds to an irreducible four-point diagram.

In the case of a coherent wave the intensity I is independent of the variable t_+; to continue the calculation we shall find it expedient to transform to a momentum representation in terms of the variable $\mathbf{r}$ (we denote the corresponding momentum by $\mathbf{q}$). As a result we need to analyze the behavior of a four-point diagram with a momentum arrangement clear from Fig. 2b. It is known that the four-point diagram P has singularities when the difference momentum $\mathbf{q}$ and the total incoming momentum $\mathbf{Q} = \mathbf{k} + \mathbf{k}'$ are small. The first and second singularities lead to the need for summing ladder and fan diagrams, respectively. The net result is $P = \mathcal{L} + V$. It is precisely this relation which determines the interference effects that are of importance in backward scattering ($k' = -k$). To calculate this contribution we can iterate the equation of Fig. 2a.[17]

To make the final equation simple we confine ourselves to normal incidence of the wave on the medium. The reflection coefficient $R(\omega, \mathbf{s} = \mathbf{k}'/k)$ can then be expressed in terms of the values of $\mathcal{L}(\mathbf{q},\omega)$ and $V(\mathbf{q},\omega)$ obtained for the case of a spatially homogeneous region, with the aid of the specular-reflection method. Acting in the spirit of Refs. 17 and 21 we obtain $\tilde{R}(\omega) = R(\omega)/R_i$ at $|s_1| \ll 1$, where

$$\tilde{R}(\omega)=\frac{a}{l}\int d^3q\,\frac{q_z^2}{(q_z^2+l^{-2})^2}[\mathcal{L}(\mathbf{q},\omega)\,\delta(\mathbf{q}_\perp)+V(\mathbf{q},\omega)\,\delta(\mathbf{q}_\perp-k\mathbf{s}_\perp)],$$

$$R_i=\int d\omega\,R(\omega). \qquad (21)$$

Here $a \approx 1$, the medium occupies the half-space $z > 0$, and $\mathbf{q}$ stands for the difference momentum in the expression for $\mathcal{L}$ and for the total incoming momentum in the expression for V. The expression for $R_i^{(s)}$ has been quite well investigated.[17,21] It is known that the interference contribution to R_i is of the order of unity in the angle interval $\theta \approx (kl)^{-1}$ relative to the normal to the surface. Note that the multiple scattering is not separated in the expression for R_i and makes a contribution of the same order as the low-multiplicity processes. At the same time, multiple scattering makes the main contribution in the region of the spectral-line edge, as will be shown below.

Thus, to calculate the interference contribution to the scattering we must analyze $V(\mathbf{q},\omega)$ with $\mathbf{q} = \mathbf{k} + \mathbf{k}'$. The sum of the diagrams for this quantity is shown in Fig. 2c.

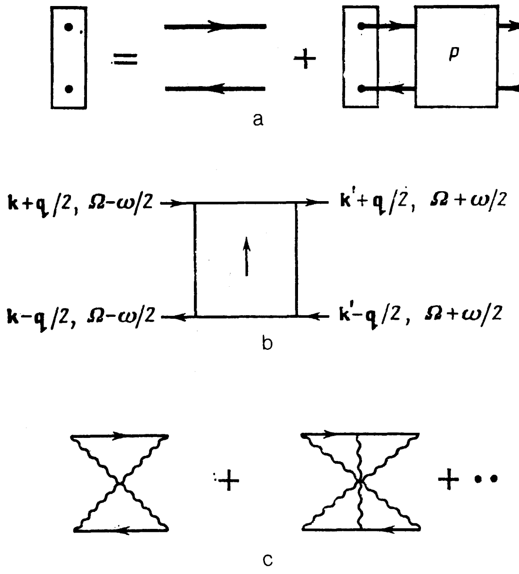

FIG. 2.

3. CALCULATION OF THE SUM OF FAN DIAGRAMS

To analyze $V(\mathbf{q},\omega)$ (to cooperon) we derive more specific expressions for the Green's functions. We determine first the propagator K of the scattering particles. To this end we must consider the dashed-line loop shown in Fig. 1c. There are three types of such a loop, corresponding to the indices 1 and 2 of the discrete levels and 12 of the off-diagonal part. It is expedient to calculate them by the Abrikosov–Maleev technique,[30,31] but first in the framework of the Matsubara technique, and followed by analytic continuation. As a result we have at $\hbar\omega \ll T$

$$K_\pm^R(\omega,\mathbf{p})=\frac{\hbar\omega}{T}\,\frac{\exp(\mp\hbar\omega/2T)}{2\,\mathrm{ch}(\hbar\omega_0/2T)}\,\frac{1}{\omega+iD_sp^2} \qquad (22)$$

$$K_{12}^R(\omega,\mathbf{p})=\mathrm{th}\left(\frac{\hbar\omega_0}{2T}\right)\left(\frac{1}{\omega-\omega_0+iD_sp^2}-\frac{1}{\omega+\omega_0+iD_sp^2}\right). \qquad (23)$$

It was assumed in the derivation of these expressions that the following conditions are met:

$$\omega,\ \omega_0 \ll \tau^{-1},\ \tau_s^{-1}, \qquad l^{-1} \ll p \ll L^{-1}, \qquad (24)$$

where τ_s is the scatterer free-path time and the $+$ and $-$ signs designate below the upper and lower levels of the pair, respectively. The physical meaning of these conditions is that the scattering-particle motion is diffuse. To calculate the self-energy part Σ of the photon Green's function we must substitute (22) and (23) in the standard expressions for Σ and sum over the channels. When the conditions $\Omega, T/\hbar \gg \omega_0$ are met the result is a standard expression for damping due to Rayleigh scattering:

$$1/\tau=1/\tau_+ + 1/\tau_- + 1/\tau_{12}. \qquad (25)$$

The values of τ_1, τ_2, and τ_{12} differ only by factors proportional respectively to the vertices g_+^2, g_-^2 and g_{12}^2; in the quasi-elastic approximation with condition (1) met the propagators K lead to contributions of the same order.

If the scatterers move ballistically and Doppler line broadening obtains, it is necessary to replace the last factors of (22) and (23) by $\langle(\omega + \mathbf{p}\mathbf{v}_s + i/\tau_s)^{-1}\rangle$ and $\langle(\omega + \mathbf{p}\mathbf{v}_s \pm \omega_0 + i/\tau_s)^{-1}\rangle$, respectively. Here $\tau_s = L/\mathbf{v}_s$ ($\omega, \omega_0 \gg \tau_s^{-1}$) and the angle brackets denote averaging over all the directions of the scattering velocities $\mathbf{v}_s$. All the subsequent calculations are performed similarly (cf. Ref. 27).

To determine $V(\mathbf{q},\omega)$ it is necessary to sum ladder diagrams in the particle–particle channel. Calculating this sum by the Matsubara technique and continuing the result analytically in analogy with the procedure used in Ref. 29, we arrive at the following integral equation for V:

$$V(\omega,\mathbf{q})=\tilde{V}_0(\omega,\mathbf{q})+\int\frac{d\omega'\,d^3k'}{2\pi}V(\omega',\mathbf{q})\times\frac{N(\omega)[N(\omega')-N(\omega'-\omega)]}{N(\omega')}\times D^R(\Omega-\omega',\mathbf{k}')D^A(\Omega-\omega+\omega',\mathbf{q}-\mathbf{k}')\times\sum_\alpha|g_\alpha|^2[K_\alpha{}^R(\omega-\omega',\mathbf{k}-\mathbf{k}')-K_\alpha{}^A(\omega-\omega',\mathbf{k}-\mathbf{k}')]. \tag{26}$$

Here $N(\omega)$ is the Planck function, and summation over α means summation over the channels. The quantity $\tilde{V}_0(\mathbf{q},\omega)$ is the contribution of a diagram with one crossing. In the approximation $\omega\tau\ll 1$ of interest to us we can put $\tilde{V}_0=V_0[1+(c\tau)^2(\mathbf{n}_k\mathbf{q})^2]^{-1}$, where $\mathbf{n}_k=\mathbf{k}/k$ and

$$V_0=iN(\omega)\sum_\alpha|g_\alpha|^2[K_\alpha{}^R(\omega,\mathbf{q})-K_\alpha{}^A(\omega,\mathbf{q})]. \tag{27}$$

The succeeding analysis of Eq. (26) is similar to that of Ref. 28: the functions $N(\omega)$ are expressed in semiclassical form, followed by integration with respect to $\Omega_{k'}$ with the quasielasticity of the scattering taken into account. This circumstance signifies the calculations considerably, since $|\mathbf{k}|=|\mathbf{k}'|$. It is convenient next to introduce an auxiliary function $\tilde{V}$ defined as

$$\tilde{V}(\omega,\mathbf{k},\mathbf{q})=V(\omega,\mathbf{k},\mathbf{q})[1+(c\tau)^2(\mathbf{n}_k\mathbf{q})^2]^{-1} \tag{28}$$

and change to a temporal representation with respect to the variable ω. The result is the integral equation

$$[1+(c\tau)^2(\mathbf{n}_k\mathbf{q})^2]\tilde{V}(t,\mathbf{k},\mathbf{q})=\tilde{V}_0+\int_{-\infty}^{t}\frac{dt}{\tau}\exp\left(\frac{t_1-t}{\tau}\right)\tilde{V}(t_1,\mathbf{k},\mathbf{q})J(2t-t_1), \tag{29}$$

where

$$J(t)=\frac{-4\Omega_k{}^2\tau}{\pi\hbar c^3}\int\frac{d\omega}{2\pi\omega}e^{i\omega t}\langle\operatorname{Im}K^R(\omega,\mathbf{k}-\mathbf{k}')\rangle_{\mathbf{n}_{k'}}, \tag{30}$$

and $t\geqslant 0$.

We shall consider hereafter only the hydrodynamic situation for the scattering particles. In this case

$$J(t)=(1-2D_sk^2|t|)[1-(\tau/\tau_{12})(1-\cos\omega_0t)]. \tag{31}$$

Expanding $\tilde{V}(\mathbf{k},\mathbf{q})$ in spherical harmonics, we readily verify that the principal role is played by a contribution that is independent of the angle between $\mathbf{k}$ and $\mathbf{q}$. The equation for this part is obtained from (29) by making the substitution $(c\tau)^2(\mathbf{n}_k\mathbf{q})^2\to(ql)^2/3$.

Equation (29) can be analytically solved by the Evans method (see Ref. 28). For $t\gg\tau$ it can be written in the form

$$\tilde{V}(\mathbf{q},t)=\int_{-\infty}^{t}\frac{dt_1}{\tau}\langle\tilde{V}_0(t_1)\rangle_{\mathbf{n}_k}\exp\left[-\int_{t_1}^{t}\frac{dt_2}{\tau}a(t_2)\right], \tag{32}$$

where

$$a(t)={}^1/_3(ql)^2+2D_sk^2t+(\tau/\tau_{12})(1-\cos\omega_0t)\times[1-{}^1/_3(ql)^2-2D_sk^2t]. \tag{33}$$

Note that expression (33) can be substantially simplified in two limiting cases, $D_sk^2\gg\omega_0(\omega_0\tau)^{1/3}(\tau/\tau_{12})^{2/3}$ and its inverse. In the first case the principal role in the wave phase relaxation is played by the motion of the scattering particles in real space. In this situation one needs retain only the first two terms of (33), while in the second case one can neglect all terms but unity in the square brackets. Note also that at the accuracy of interest to us we can neglect the differences between $\tilde{V}$ and V and between $\langle\tilde{V}_0\rangle$ and V_0.

4. ANALYSIS OF LIGHT REFLECTION FROM A HALF-SPACE

A. We begin the analysis with the simplest case, when light scattering with transitions between the levels of the scatterers is of no importance:

$$D_sk^2\gg\omega_0(\omega_0\tau)^{1/3}(\tau/\tau_{12})^{2/3}. \tag{34}$$

In this situation

$$V(\mathbf{q},t)=\int_0^\infty\frac{dt'}{\tau}\exp\left[-\frac{(ql)^2}{3}\frac{t'}{\tau}-D_sk^2\left(\frac{t'^2}{\tau}+\frac{2t'|t|}{\tau}\right)\right], \tag{35}$$

where we have used the explicit expression for V_0 at $\mathbf{k}=-\mathbf{k}'$.

The Fourier transform of this expression with respect to t, normalized to the integral over all ω, has at the accuracy of interest to us the form

$$V(\mathbf{q},\omega)=\frac{1}{D_sk^2}\int_0^\infty\frac{dt_1}{\tau}\exp\left[-\frac{(ql)^2}{3}\frac{t_1}{\tau}-D_sk^2\frac{t_1^2}{\tau}\right]\mathcal{F}(\omega,t_1), \tag{36}$$

where

$$\mathcal{F}(\omega,t)=\frac{1}{\pi}\frac{t/\tau}{(\omega/2D_sk^2)^2+(t/\tau)^2}. \tag{37}$$

To calculate the normalized contribution to the reflection coefficient in accord with Ref. 21, this expression must be integrated with respect to q_z with weight $q_z^2/(q_z^2+l^{-2})^2$, putting $\mathbf{q}_\perp=k\mathbf{s}_\perp$. We have then $|\mathbf{q}|=k\theta$, where θ is the angle between the scattered-light direction and the normal to the surface. Note that the integrand in (35) was obtained under the assumption that $q_zl\ll 1$. It makes sense therefore to integrate only over the region $q_z\lesssim l^{-1}$. This region makes the main contribution only if $t_1/\gg 1$, as is the case in the region of the spectral-line edge. Taking this into account and using the condition $kL\ll 1$ we arrive at the following expression, normalized to unity, for the interference contribution to the reflection coefficient:

$$\tilde{R}^{(V)}=\frac{\tau_D}{\pi j_0(\theta)}\int_0^\infty\frac{M(x)x\,dx}{x^2+(\omega\tau_D)^2}\exp\left[-(kl\theta)^2x-\frac{3\tau}{2\tau_D}x^2\right]. \tag{38}$$

where $\tau_D = (6D_s k^2)^{-1}$ and the function $M(x)$ was obtained by integrating with respect to q_z with the weight indicated above in absolute-value limits $\sim l^{-1}$; for $x \ll 1$ we have $M \sim 1$ and for $x \gg 1$

$$M(x) \approx (\pi/x^3)^{1/2}. \quad (39)$$

The function

$$f_0(\theta) = \int_0^\infty M(x)\,dx \exp[-(kl\theta)^2 x], \quad (40)$$

determines the angular dependence of the elastic scattering and is calculated in a number of papers (see, e.g., Ref. 21).

It can be seen from (38) that the characteristic width of the spectral line is determined by the parameter τ_D. The approximations above make it possible to consider the region $\omega\tau_D \gg 1$. We can use here the asymptotic expression (39) for $M(x)$, after which the integral in (38) can be readily analyzed. It can be seen that there exists a critical angle

$$\theta_c = (kl)^{-1}(3\tau/2\tau_D)^{1/4} \ll (kl)^{-1}. \quad (41)$$

At $\theta \ll \theta_c$ the absorption spectrum is independent of the angle. If

$$\omega\tau_D \ll (2\tau_D/3\tau)^{1/2}, \quad (42)$$

we have

$$\tilde{R}^{(V)}(\omega) = \tau_D/(2\pi)^{1/2} f_0(\theta)\,\omega^{1/2}. \quad (43)$$

If, however, the conditions inverse to (42) is met, we get

$$\tilde{R}^{(V)}(\omega) = \frac{\Gamma(1/4)}{2\pi^{1/2} f_0(\theta)} \frac{\tau_D}{(\omega\tau_D)^2 (3\tau/2\tau_D)^{1/4}}. \quad (44)$$

Thus, the farthest wing of the spectrum has a Lorentz shape and an effective width of order $\tau_D^{-3/4}\tau^{-1/4} \gg \tau_D^{-1}$.

At $\theta \gg \theta_c$ the spectrum begins to depend strongly on the scattering angle. If the condition

$$\omega\tau_D \ll (\theta_c/\theta)^2 (2\tau_D/3\tau)^{1/2} \quad (45)$$

is met the result coincides with (43). In the opposite limiting case

$$\tilde{R}^{(V)}(\omega) = \frac{\Gamma(1/4)}{f_0(\theta)} \frac{\tau_D\theta_c}{\theta(\omega\tau_D)^2 (3\tau/2\tau_D)^{1/4}}. \quad (46)$$

We have thus again a Lorentz wing with effective width $(\theta_c/\theta)\tau_D^{-3/4}\tau^{-1/4}$. Ultimately its spectrum becomes narrower when the angle is increased, and the angular width of the peak decreases for a given distance from the spectral-line center.

We compare now the width of the spectral line of interference scattering with the linewidth of ordinary diffuse scattering. At $kL \ll 1$ the frequency transfer in the first case is of order $D_s k^2 \approx \tau_D^{-1}$, which is smaller by a factor $(\tau/\tau_D)^{1/2}$ than the interference-line characteristic width. When the scattering particles have an internal degree of freedom, the characteristic scattering linewidth is of order ω_0. Since we shall assume that $\omega_0\tau \ll 1$, we see that in this case the interference-scattering width is larger.

B. We proceed now to the inelastic case, when the quasidiscrete character of the scattering-particle spectrum is of importance:

$$\omega_0/D_s k^2 \gg (\omega\tau)^{-1/3}. \quad (47)$$

(The remaining criteria are listed in detail in the discussion of the qualitative picture.) In this situation, $a(t)$ in (32) can be expressed as

$$a(t) \approx (ql)^2/3 + 2|t|/3\tau_D + (\tau/\tau_{12})(1-\cos\omega_0 t). \quad (48)$$

We shall see that allowance for the second term is important here, the inequality (47) notwithstanding.

It can be seen that expression (32) for the fan diagrams has in the vicinity of the points $t_n = 2\tau n/\omega_0$ singularities that lead to a quasidiscrete scattered-light spectrum. The interference contribution to the scattering can be not small only in these vicinities, where the rate of phase collapse is small. It is therefore natural to break up the region of integration with respect to t_1 into intervals $(2n-1)\pi/\omega_0 < t_1 < (2n+1)\pi/\omega_0$. It can be easily verified that in these vicinities the integrals with respect to t_1 converge rapidly and it is possible in the calculation of the contribution of an individual integral to iterate in the range $(-\infty, \infty)$ and also expand $\cos\omega_0 t$ in a series:

$$1-\cos\omega_0 t_1 \approx {}^1/_2\omega_0^2 (t-t_n)^2.$$

The expression for $\tilde{R}(\omega)$ takes ultimately the form

$$\tilde{R}(\omega) = f_E(\omega,\theta)/f_1(\theta), \quad (49)$$

where

$$f_E(\omega,\theta) = \frac{1}{2\pi^{1/2}\omega_0}\left(\frac{\tau_{12}}{\tau}\right)^{1/2} \sum_{n=-\infty}^{\infty} \exp\left(\frac{2\pi i\omega|n|}{\omega_0}\right) \times \int_0^\infty \frac{dx\,M(x)}{x^{1/2}} \exp\left[-\frac{2\pi|n|x}{\omega_0\tau_D} - \left(\frac{\omega}{\omega_0}\right)^2 \frac{\tau_{12}}{\tau}\frac{1}{x} + \frac{i\omega\tau x}{2} - \frac{x^3\omega_0^2\tau^3}{\tau_{12}}\right] \exp(-k^2l^2\theta^2 x), \quad (50)$$

while $f_1(\theta)$ is the integral of $f_E(\omega,\tau)$ over all the frequencies ω. When the contribution $x \ll \omega_0\tau_D$ is met the sum over n can be represented as

$$\omega_0\tau_D x/\pi(s^2+x^2), \quad (51)$$

where $s = (\omega - \omega_n)\tau_D$ is the dimensionless distance to the nearest maximum in the spectrum $(s \ll \omega_0\tau_D)$.

We obtain ultimately

$$f_E(\omega,\theta) = \frac{\omega_0\tau_D\tau}{\pi^{3/2}} \int_0^\infty \frac{dx\,x^{1/2} M(x)}{s^2+x^2} \times \exp\left[-(kl\theta)^2 x - \frac{3\omega^2\tau_{12}}{2\omega_0^2\tau x} + \frac{3}{2} i\omega\tau x - \frac{\omega_0^2\tau^3 x^3}{8\tau_{12}}\right]. \quad (52)$$

It is convenient next to introduce a time τ_φ that plays the role of the time of phase relaxation on account of inelastic scattering:

$$\tau_\varphi^{-3} = \omega_0^2/6\tau_{12}. \quad (53)$$

Introducing a new variable

$$x = \gamma z, \qquad \gamma(\omega) = (\tau_\varphi/\tau)(\omega\tau_\varphi/3)^{1/2}, \quad (54)$$

we easily reduce the argument of the exponential to the form $-\lambda\varphi(z)$, where

$$\lambda=(3^{1/2}/4)(\omega\tau_\varphi)^{3/2}, \tag{55}$$

$$\varphi(z)=z^3/3-2iz+1/z+z\theta^2/\theta_\omega^2, \tag{56}$$

$$\theta_\omega=2(\omega\tau)^{1/2}(3^{1/4}kl)^{-1}. \tag{57}$$

We begin with the case of small angles, $\theta\ll\theta_\omega$ If the condition

$$\omega\tau_\varphi\ll1 \tag{58}$$

is satisfied, the integral has no saddle-point peak. To analyze this case it is convenient to introduce

$$x_\omega={}^1/_4(\tau_\varphi/\tau)(\omega\tau_\varphi)^2. \tag{59}$$

We examine first the frequency region $x_\omega\gg1$. Note that expression (52) has meaning if

$$\omega\tau_\varphi\ll(\tau/\tau_\varphi)^{1/2}(\omega_0\tau_D)^{1/2}. \tag{60}$$

In this case

$$f_E(\omega)=\frac{\omega_0\tau_D\tau}{\pi s^2}\ln\left(1+\frac{s^2}{x_\omega^2}\right)^{1/2}. \tag{61}$$

The characteristic width of the peak in frequency units is thus of the order of x_θ/τ_D. For the width of the nth peak we have

$$\Gamma_n=\frac{3}{2}\left(\frac{\tau_{12}}{\tau}\right)\frac{n^2}{\tau_D}, \tag{62}$$

and the peaks are resolved at $\Gamma_n\ll\omega_0$. Hence

$$n\leqslant n_{max}=(\omega_0\tau_D)^{1/2}(2\tau/3\tau_{12})^{1/2}. \tag{63}$$

The peak envelope is given by

$$f(n\omega_0)=\frac{\omega_0\tau_D\tau}{2\pi x_{n\omega_0}^2}=\frac{2\tau}{9\pi}\left(\frac{\tau}{\tau_{12}}\right)^2\frac{\omega_0\tau_D}{n^4}. \tag{64}$$

In the frequency region $x_n\ll1$ the argument of the logarithm in (61) is $(1+s^2)^{1/2}$.

Another situation is realized for $x_\omega\gg\omega_0\tau_D$ if the condition $\omega_0\tau_D\tau/\tau_\varphi\ll1$ is met. In this case the function $x/(x^2+s^2)$ in (51) must be replaced by $2\pi^2/\omega_0\tau_D$. We obtain then

$$f_E(\omega)=\frac{2\pi\tau}{x_\omega}=2\pi\tau\left(\frac{3\tau}{\tau_{12}}\right)\left(\frac{\omega_0}{\omega}\right)^2. \tag{65}$$

At $\omega\simeq n_{\max}\omega_0$ expressions (64) and (65) become nearly equal with value on the order of $\tau/\omega_0\tau_D$. The characteristic width of the Lorentz wing is in this case $\sim\tau^2/\tau_\varphi^3$. Under the condition $\omega_0\tau_D/\tau_\varphi/\tau$, however, there is no Lorentz wing in the reflected-light spectrum. The peaks, remaining resolved, begin to decrease in amplitude rapidly (exponentially) [see (66)].

We turn now to the case of the highest frequencies, when conditions inverse to (58) are satisfied. In this situation the principal role in the integral with respect to x is played by the saddle point $z_0=\exp(i\pi/4)$, $\varphi''(z_0)=0$, and there exist therefore three steepest-descent directions. Passing the integration contour through the saddle point and proceeding in standard manner, we obtain

$$f_E(\omega)=\frac{\omega_0\tau_D\tau}{(\omega\tau_\varphi)^{1/2}}\frac{3^{1/6}\Gamma(^1/_3)}{s^2+i\gamma^2}\exp\left[-\left(\frac{2}{3}\right)^{1/2}(\omega\tau_\varphi)^{3/2}(1-i)-\frac{i\pi}{12}\right]. \tag{66}$$

The modulus of the reflection coefficient is thus

$$|\tilde{R}^{(V)}(\omega)|=\frac{\omega_0\tau_D\tau}{(\omega\tau_\varphi)^{1/2}}\frac{3^{1/6}\Gamma(^1/_3)}{(s^4+\gamma^4)^{1/2}}\exp\left[-\left(\frac{2}{3}\right)^{1/2}(\omega\tau_\varphi)^{3/2}\right]. \tag{67}$$

The line profile is ultimately "cut off" at values $\omega\sim\tau_\varphi^{-1}$.

We consider now the angular dependence of the reflection coefficient. It is necessary for this purpose to analyze expression (56) with account taken of the coefficient θ^2/θ_ω^2. The principal role is again played here by the saddle point $z_0=\exp(i\pi/4)$. In the upshot, the argument of the exponential acquires an additional factor $3\cdot2^{-5/2}(\theta/\theta_\omega)^2$, and the rest of the expression remains unchanged. The argument of the exponential is thus

$$-\left(\frac{3}{2}\right)^{3/2}\frac{\tau_{12}}{\tau}\left(\frac{\omega}{\omega_0}\right)^2(kl\theta)^2.$$

We note in conclusion that even in the lower-frequency region the peak widths and amplitudes also depend on the angle θ. This dependence begins to manifest itself at angles $\theta\gtrsim\theta_c$, where

$$\theta_c^2\sim(kl)^{-2}[\min(\tau_\varphi/\tau,\ \omega_0\tau_D)]^{-1}.$$

The corresponding expressions for $f_E(\omega,\theta)$ can also be easily obtained from (52).

[1)] It must be pointed out here immediately that our qualitative interpretation is purely illustrative. The point is that the spectral dependence of R is very sensitive to the method used to average the phase difference (5). Naturally, an accurate calculation, which will be carried out in the following sections, does not have such a leeway and is therefore necessary from our point of view.

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