diverges logarithmically. In conclusion we note that:

(1) The contribution to transport of the recollision part of the four-particle collision operator for real gases had previously been shown to diverge logarithmically for the quantum[6] as well as the classical case.[1–5]

(2) The present communication shows that the contribution to transport of the WC limit of the same recollision operator converges in the quantum as well as in the classical case.

(3) Taken together, conclusions (1) and (2) imply that the WC limit fails for the transport divergence problem.

(4) A rather curious behavior is uncovered for the quantum WC limit in which $\partial I(t)_{\mathrm{WC}}/\partial t$ is found to grow as $t^{1/2}$ and oscillate rapidly. This growth, although faster than logarithmic in t, is due to the spatial overlapping of broadening wave packets, as can be deduced with (23) and (24), and does not present any divergence problem to transport. The overlapping wave-packet phenomenon is clearly distinct from the logarithmic divergence in which "information" can be carried between separated wave packets.

PHYSICAL REVIEW VOLUME 169, NUMBER 1 5 MAY 1968

Spectrum of Light Doubly Scattered by an Opalescent Fluid*

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(Received 18 December 1967)

An expression is derived for the frequency spectrum of light which is scattered twice by thermal diffusion-type density fluctuations in a fluid. In the two limiting cases of forward and backward scattering, the shape of the spectrum is insensitive to the presence of a double-scattering component.

As has recently been demonstrated in numerous experimental investigations,[1,2] the spectral analysis of monochromatic light scattered by a fluid near its critical point makes possible the determination of the coefficient of thermal diffusion $D(T)$. If k is the wave number of the entropy fluctuation producing the density fluctuation which causes the light scattering, the width of the Lorentzian curve which describes the Landau-Placzek central peak is

$$\Gamma_k^{(1)}(T)=D(T)k^2. \quad (1)$$

Thus the interpretation of the frequency width of the experimentally observed spectrum of the scattered light is unambiguous, provided that it is certain that only one scattering process has been involved. The purpose of this paper is to point out that in the two limits of forward scattering and backward scattering such a rigorous criterion can be relaxed. In these cases the shape and width of the Landau-Placzek[3] central peak is insensitive to the double-scattering corrections, even when the fluid is opalescent to such an extent that the optical mean free path is comparable to the optical path length in the sample.

The demonstration of this result will be limited to the calculation of the correction for double scattering. Extension to higher multiples of scattering is straightforward. While no simple general result applies for backward scattering, the case of forward scattering is generally described by the qualitative result which we will now proceed to establish explicitly for the limited case of double scattering.

Successive scattering from the fluctuations of wave number $\mathbf{k}_1$ and $\mathbf{k}_2$ gives an observed scattering at wave number transfer $\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2$. By the convolution theorem the doubly scattered light will have width

$$\Gamma_{k_1,k_2}^{(2)}(T)=\Gamma_{k_1}^{(1)}(T)+\Gamma_{k_2}^{(1)}(T)=D(T)(k_1^2+k_2^2). \quad (2)$$

It is now trivially evident that backward scattering satisfies the condition $k_1^2+k_2^2=k^2$, regardless of the actual direction of $\mathbf{k}_1$. Hence all of the doubly scattered light observed in the backward direction has the same frequency width as the singly scattered component, and no correction is required for the frequency spectrum in this case. (As remarked above, this conclusion is limited to double scattering and is no longer true when higher-order corrections are important.)

For the general case of double scattering of light of wave number k_0 at net scattering angle θ, the transfer wave number is $\mathbf{k}$ where $k=|\mathbf{k}|=2k_0\sin\frac{1}{2}\theta$. Thus we require $\mathbf{k}_1+\mathbf{k}_2=\mathbf{k}$ and change variable to $\mathbf{k}'=\mathbf{k}_1-\frac{1}{2}\mathbf{k}$.

* Work supported in part by the Air Force Office of Scientific Research and carried out under the auspices of the Center for Theoretical Physics of the Department of Physics and Astronomy, University of Maryland.

[1] N. C. Ford, Jr., and G. Benedek, Phys. Rev. Letters **15**, 649 (1965) and in *Proceedings of a Conference on Critical Phenomena* (National Bureau of Standards, Washington, D. C., 1965).

[2] For an application of the method of Ford and Benedek (Ref. 1) to the determination of the temperature dependence of the thermal diffusion coefficient of CO_2 near its critical point see J. Osmundson, Ph.D. dissertation, University of Maryland, 1968 (to be published) and H. Swinney, Ph.D. dissertation, The Johns Hopkins University, 1968 (unpublished). See also H. Swinney and Herman Z. Cummings (to be published).

[3] L. D. Landau and G. Placzek, Physik. Z. Sowjetunion **5**, 172 (1934).

The resulting double-scattering breadth is then proportional to

$$k_1^2+k_2^2=|\mathbf{k}'+\tfrac{1}{2}\mathbf{k}|^2+|\mathbf{k}'-\tfrac{1}{2}\mathbf{k}|^2$$
$$=2k'^2+\tfrac{1}{2}k^2$$
$$=\tfrac{1}{2}k^2+2k_0^2+2K^2-4k_0K\cos\psi\,, \qquad (3)$$

where ψ is the angle between the vectors $\mathbf{K}=\mathbf{k}_0+\frac{1}{2}\mathbf{k}$ and $\mathbf{k}_0+\mathbf{k}_1$. We now assume that the wavelength of the light is much greater than the correlation length of the fluctuations in the fluid and that the double-scattering events are weighted equally in proportion to the differential solid angle $2\pi\sin\psi d\psi$. Thus the frequency spectrum of the double-scattering events is given by the integral over Lorentzians,

$$I_2(\omega,k)=-\mathrm{Im}\frac{1}{2\pi}\int_0^\pi\frac{\sin\psi d\psi}{\omega+iD(k_1^2+k_2^2)}$$
$$=\frac{1}{16\pi Dk_0K}\ln\frac{\omega^2+D^2[\frac{1}{2}k^2+2(k_0+K)^2]^2}{\omega^2+D^2[\frac{1}{2}k^2+2(k_0-K)^2]^2}, \qquad (4)$$

where $\omega/2\pi$ is the frequency shift of the doubly scattered light and we have substituted from Eq. (3). As a check on this result we note that the case of backward scattering is described by the limit $K\rightarrow 0$. In this case we can neglect terms of order K^2 within the logarithm, which then reduces to $\ln[(1+x)/(1-x)]\approx 2x$, where

$$x=8D^2k^2k_0K[\omega^2+D^2(\tfrac{1}{2}k^2+2k_0^2)]^{-1}$$
$$=8D^2k^2k_0K(\omega^2+D^2k^4)^{-1},$$

with $k=2k_0$. Equation (4) consequently becomes

$$I_2(\omega,k)=\frac{1}{\pi}\frac{Dk^2}{\omega^2+D^2k^4}$$
$$=-\mathrm{Im}\frac{1}{\pi}\frac{1}{\omega+iDk^2}$$
$$=I_1(\omega,k)\,, \qquad (5)$$

the frequency scattering for single scattering. Thus there is no double-scattering correction to the spectrum observed in the backward direction, as already remarked above.

It is evident from Eq. (4) that the double-scattering correction at a typical angle (e.g., $\theta=\frac{1}{2}\pi$) considerably modifies the spectrum. This correction can be avoided not only in the backward direction, as we have seen, but also in the forward direction. The simplying feature of the latter case is that the doubly scattered light is spread out over a frequency range of the order Dk_0^2 and is consequently very weak in the much smaller frequency interval of half-width $Dk^2\ll Dk_0^2$, where the singly scattered light is concentrated. In this case we can make the approximation $(k_0-K)^2=4k_0^2\sin^4(\frac{1}{4}\theta)\approx k^4/(64k_0^2)\ll k^2$, so Eq. (4) becomes

$$I_2(\omega,k)\approx\frac{1}{16\pi Dk_0^2}\ln\frac{64k_0^4}{\omega^2+\frac{1}{4}D^2k^4}. \qquad (6)$$

The strength of this spectrum at $\omega=0$ compared to the central strength of the spectrum of the singly scattered light is

$$\frac{I_2(0,k)}{I_1(0,k)}\approx\frac{k^2}{4k_0^2}\ln\frac{k_0}{k}. \qquad (7)$$

As a numerical example, for the particular case of scattering at $\theta\approx 11°\approx\frac{1}{5}$ rad, we have $k/k_0=\frac{1}{5}$, and the ratio takes on the value 10^{-2} ln 5=0.016. Hence, in the frequency interval in which the singly scattered light is concentrated, the strength of the doubly scattered light amounts to less than 2%.

In summary, Eq. (4) exhibits the spectrum which is expected for doubly scattered light when the basic fluctuations producing the scattering are controlled by thermal diffusion. Equation (4) simplifies in two special cases. In the backward direction, the double scattering has no net effect because it has the same spectrum as single scattering. In the forward direction (i.e., scattering angle of 11° or smaller) the double scattering has a different spectrum but its strength in the relevant frequency interval is sufficiently small as to be negligible. As a final remark, it should be cautioned that Eq. (4) cannot be applied directly to directions other than the two special cases which have led to the qualitative conclusions presented here. This is because the geometrical shape of the scattering sample also contributes a weighting factor for the individual scattering events which would have to be included in any quantitative application of Eq. (4). A further caution is that the results here are limited to the case of isotropic opalescence. This is the case when the correlation length is smaller than the optical wavelength, which is true except when very close to the critical point (i.e., extremely small values of $T-T_c$).