Coherent four-photon Rayleigh scattering of light

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A general analysis is presented of four-photon coherent interaction of light, as applied to scalar Rayleigh scattering in liquid and gaseous media. It is shown that the use of this type of interaction in active spectroscopy provides an appreciable gain (by several orders of magnitude) in sensitivity compared with the method of thermal scattering, both for the undisplaced (Rayleigh) line and for the Mandel'shtam-Brillouin doublet. From the experimental point of view, this method has also a number of other advantanges, it is shown how the expression for the "Landau-Placzek ratio" is generalized in the case of coherent four-photon scattering.

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The great progress made recently in molecular spectroscopy in the region of active Raman-scattering spectroscopy,[1] also called coherent anti-Stokes Raman spectroscopy,[2,3] suggests that the use of coherent four-photon processes can be promising also in the study of Rayleigh scattering of light. A particular case of such processes was considered in essence earlier,[4] in an analysis of four-photon scattering by ion-sound oscillations in a plasma for diagnostic purposes.

The first experimental demonstration of the advantage of four-photon Rayleigh scattering is the work of Pohl *et al.*[5] who reported observation of this scattering on an undisplayed Rayleigh line in an NaF crystal. Two crossed CO_2-laser beams were used to excite the temperature wave in the crystal; the sounding beam was produced by a helium-neon laser. The use of the four-photon scattering procedure has made it possible to lower the crystal temperature to 20°K with a single amplitude exceeding the noise level. The amplitudes of the excited temperature waves exceeded the mean squared value of the temperature fluctuations by 10^5–10^6 times. We note also a study by Pohl and Irniger,[6] which has a bearing on our problem, where they observed four-photon scattering by second-sound waves in the same NaF crystal.

In the present paper we present a general analysis of the problem of coherent four-photon scattering as applied to scalar type of Rayleigh scattering in liquid and gaseous media. As to scattering in the Rayleigh wing, due to anisotropic variations $\Delta\epsilon_{ik}$ of the permittivity the coherent four-photon processes correspond here to the optical Kerr effect (in the field of two pump waves) and to scattering of the sounding wave by the variations $\Delta\epsilon_{ik}$ induced by this effect. This phenomenon has in effect already been considered earlier,[7,8] in an analysis of the possibility of discriminating the non-resonant coherent background in observation of the resonant contribution to four-photon Raman scattering.

Two pumping light waves with frequencies ω_1 and ω_2 and with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, interacting with the medium, excite in the latter, as a result of absorption and electrostriction, a temperature wave and an acoustic wave of frequency $\Omega=\omega_1-\omega_2$ and wave vector $\mathbf{q}=\mathbf{k}_1-\mathbf{k}_2$. These waves scatter in the medium a third (sounding) wave $(\omega_3, \mathbf{k}_3)$, which can constitute a fraction of the radiation of one of the pump waves, generally speaking with a change of the propagation direction. The frequencies of the scattered waves are $\omega_s^{(\mp)}=\omega_3\mp\Omega$, and accordingly the scattering cross sections should be maximal when the scattering wave vector is $\mathbf{k}_s^{(\mp)}=\mathbf{k}_3\mp\mathbf{q}$ (the equivalent of the Laue condition). The scattering by the temperature wave corresponds to an isobaric contribution to the scattering, i.e., to the undisplaced (Rayleigh) line, and scattering by the sound wave corresponds to an adiabatic contribution to the scattering, i.e., to a Mandel'shtam-Brillouin shift. We present below quantitative estimates of this process.

The field $\mathbf{E}_s$ of the scattered wave satisfies the equation

$$\nabla^2\mathbf{E}_s-\frac{\varepsilon}{c^2}\frac{\partial^2\mathbf{E}_s}{dt^2}=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\left[\left(\frac{\partial\varepsilon}{\partial T}\right)_p T'+\left(\frac{\partial\varepsilon}{\partial p}\right)_s p'\right]\mathbf{E}_3, \tag{1}$$

where ϵ is the real part of the dielectric constant of the medium (it is assumed that the imaginary part $\epsilon''\ll\epsilon$ and that the damping of the scattered wave in the medium can be neglected), T is the temperature of the medium, S is the specific entropy, p is the pressure, and T' and p' are the deviations of the temperature and pressure from the equilibrium values and are due to the interaction of the medium with pump waves $\mathbf{E}_1$ and $\mathbf{E}_2$. Strictly speaking, the nonlinear polarization of the medium and the right-hand side of Eq. (1) should be written in the form

$$\mathbf{P}^{(\mathrm{nl})}=\left[\left(\frac{\partial\varepsilon}{\partial S}\right)_p S'+\left(\frac{\partial\varepsilon}{\partial p}\right)_s p'\right]\mathbf{E}_3, \tag{1a}$$

where the deviation of the entropy is $S'=c_p/T)T'-(\alpha/\rho)p'$, is the specific heat at constant pressure, ρ is the density of the medium, and $\alpha=V^{-1}(\partial V/\partial T)_p$ is the coefficient of the thermal volume expansion. Recognizing that

$$\left(\frac{\partial\varepsilon}{\partial S}\right)_p=\frac{T}{c_p}\left(\frac{\partial\varepsilon}{\partial T}\right)_p,\quad \left(\frac{\partial\varepsilon}{\partial p}\right)_s=\rho\beta_s\left(\frac{\partial\varepsilon}{\partial\rho}\right)_s$$

($\beta_S=-V^{-1}(\partial V/\partial p)_S$ is the adiabatic compressibility) and $\alpha^2T/\rho c_p\beta_S=\gamma-1$ $(\gamma=c_p/c_v)$, we get

$$\mathbf{P}^{(\mathrm{nl})}=\left\{\left(\frac{\partial\varepsilon}{\partial T}\right)_p T'+\left(\frac{\partial\varepsilon}{\partial p}\right)_s p'\left[1+\frac{\gamma-1}{(\rho\,\partial\varepsilon/\partial\rho)_s}\left(\frac{\partial\varepsilon}{\alpha\,\partial T}\right)_p\right]\right\}\mathbf{E}_3.$$

For most liquids (see[9], p. 44) the second term in the

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square brackets is small in comparison with unity, and this justifies the representation of $\mathbf{p}^{(nl)}$ in the form (1).

The deviations T' and p' satisfy the equations

$$\frac{\partial T'}{\partial t}-\chi\nabla^2T'=\frac{1}{\rho c_p}(\mathbf{E}_1+\mathbf{E}_2)(\dot{\mathbf{P}}_1+\dot{\mathbf{P}}_2), \tag{2}$$

$$\frac{\partial^2p'}{\partial t^2}-\nabla^2\left(\Gamma\frac{\partial p'}{\partial t}+v_s^2p'\right)=\frac{v_s^2}{8\pi}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_T\nabla^2(\mathbf{E}_1+\mathbf{E}_2)^2. \tag{3}$$

Here $\mathbf{P}_{1,2}$ is the polarization of the medium under the influence of the field $\mathbf{E}_{1,2}$, ρ is the density of the medium, v_s is the speed of sound, χ is the coefficient of the thermal diffusivity of the medium, $\Gamma=(1/\rho)(\frac{4}{3}\eta+\zeta)+\chi(\gamma-1)$ (η and ζ are the coefficients of the shear and bulk viscosity, $\gamma=c_p/c_v$). Putting furthermore

$$\mathbf{E}_l={}^1/_2(\mathbf{A}_l\exp[i(\mathbf{k}_l\mathbf{r}-\omega_lt)]+\text{c.c.}),\quad l=1,2,3, \tag{4}$$

where $\Omega\equiv(\omega_1-\omega_2)\ll\omega_{1,2}$, $\mathbf{q}\equiv\mathbf{k}_1-\mathbf{k}_2$, $q=2k\sin(\theta/2)$, $k=\omega n/c$, $n=\epsilon^{1/2}$, $\omega\approx\omega_1\approx\omega_2\approx\omega_3\approx\omega_s$, and θ is the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$, we obtain for the amplitudes T_0 and p_0 on the basis of (2) and (3) the expressions

$$T_0=\frac{\mu cn}{4\pi\rho c_p}\frac{\mathbf{A}_1\mathbf{A}_2^*}{\chi q^2-i\Omega}, \tag{5}$$

$$p_0=\frac{v_s^2}{8\pi}\frac{(\rho\,\partial\varepsilon/\partial\rho)_Tq^2\mathbf{A}_1\mathbf{A}_2^*}{\Omega^2-v_s^2q^2+i\Gamma q^2\Omega}, \tag{6}$$

where $\mu=\omega\epsilon''/nc$ is the coefficient of optical absorption in the medium.

Substitution of (4)–(6) in Eq. (1) leads to the following result. The scattering field $\mathbf{E}_s$ consists of four components—the Stokes and anti-Stokes components of the Rayleigh line and of the Mandel'shtam-Brillouin doublet, respectively:

$$\mathbf{E}_s={}^1/_2(\mathbf{A}_R^{(-)}\exp(-i\omega_s^{(-)}t)+\mathbf{A}_R^{(+)}\exp(-i\omega_s^{(+)}t)+\mathbf{A}_{M-B}^{(-)}\exp(-i\omega_s^{(-)}t)+\mathbf{A}_{M-B}\exp(-i\omega_s^{(+)}t)+\text{c.c.}),$$

where the amplitudes $\mathbf{A}_R^{(\mp)}$ (the Rayleigh components) and $\mathbf{A}_{M-B}^{(\mp)}$ (the Mandel'shtam-Brillouin components) satisfy the equations

$$\nabla^2\mathbf{A}_R^{(\mp)}+\varepsilon\left(\frac{\omega_s^{(\mp)}}{c}\right)^2\mathbf{A}_R^{(\mp)}=-\frac{\eta cn}{8\pi\rho c_p}\left(\frac{\omega_s^{(\mp)}}{c}\right)^2\left(\frac{\partial\varepsilon}{\partial T}\right)_p\frac{\mathbf{A}_3e^{i(\mathbf{k}_3\mp\mathbf{q})\mathbf{r}}}{\chi q^2\pm i\Omega}\begin{Bmatrix}\mathbf{A}_1\mathbf{A}_2^*\\ \mathbf{A}_1^*\mathbf{A}_2\end{Bmatrix}, \tag{7}$$

$$\nabla^2\mathbf{A}_{M-B}^{(\mp)}+\varepsilon\left(\frac{\omega_s^{(\mp)}}{c}\right)^2\mathbf{A}_{M-B}^{(\mp)}=-\frac{v_s^2}{16\pi}\left(\frac{\omega_s^{(\mp)}}{c}\right)^2\times\left(\frac{\partial\varepsilon}{\partial p}\right)_s\frac{(\rho\,\partial\varepsilon/\partial\rho)_Tq^2\mathbf{A}_3e^{i(\mathbf{k}_3\mp\mathbf{q})\mathbf{r}}}{\Omega^2-v_s^2q^2\mp i\Gamma q^2\Omega}\begin{Bmatrix}\mathbf{A}_1\mathbf{A}_2^*\\ \mathbf{A}_1^*\mathbf{A}_2\end{Bmatrix}. \tag{8}$$

In the Fraunhofer zone relative to the scattering volume, i.e., to the volume of the mutual intersection of light beams 1, 2, and 3, these equations have the following solutions:

$$\mathbf{A}_R^{(\mp)}=\frac{\mu cn}{32\pi^2\rho c_p}\left(\frac{\omega_s^{(\mp)}}{c}\right)^2\left(\frac{\partial\varepsilon}{\partial T}\right)_p\frac{V\delta_V(\mathbf{k}_3\mp\mathbf{q}-\mathbf{k}_s^{(\mp)})}{\chi q^2\pm i\Omega}\times\mathbf{A}_3\frac{\exp(ik_s^{(\mp)}r)}{r}\begin{Bmatrix}\mathbf{A}_1\mathbf{A}_2^*\\ \mathbf{A}_1^*\mathbf{A}_2\end{Bmatrix}, \tag{9}$$

$$\mathbf{A}_{M-B}^{(\mp)}=\frac{v_s^2}{64\pi^2}\left(\frac{\omega_s^{(\mp)}}{c}\right)^2\left(\frac{\partial\varepsilon}{\partial p}\right)_s\frac{(\rho\,\partial\varepsilon/\partial\rho)_Tq^2V\delta_V(\mathbf{k}_3\mp\mathbf{q}-\mathbf{k}_s^{(\mp)})}{\Omega^2-v_s^2q^2\mp i\Gamma q^2\Omega}\times\mathbf{A}_3\frac{\exp(ik_s^{(\mp)}r)}{r}\begin{Bmatrix}\mathbf{A}_1\mathbf{A}_2^*\\ \mathbf{A}_1^*\mathbf{A}_2\end{Bmatrix}, \tag{10}$$

where V is the scattering volume, $\mathbf{k}_s^{(\mp)}=\omega_s^{(\mp)}n\mathbf{m}/c$ ($\mathbf{m}$ is a unit vector in the scattering direction),

$$\delta_V(\boldsymbol{\varkappa})=\frac{1}{V}\int_V e^{i\boldsymbol{\varkappa}\mathbf{r}}dV,\qquad|\delta_V(\boldsymbol{\varkappa})|\leqslant\delta_V(0)=1.$$

From (9) and (10) it is seen that for specified directions of the wave vectors of the pump $\mathbf{k}_1$ and $\mathbf{k}_2$, the directions of the maximum scattering at the Stokes $\omega_s^{(-)}=\omega_3-\Omega$ and anti-Stokes $\omega_s^{(+)}=\omega_3+\Omega$ frequencies do not coincide and are determined by the conditions $\mathbf{k}_s^{(\mp)}=\mathbf{k}_3\mp\mathbf{q}=\mathbf{k}_3\mp(\mathbf{k}_1-\mathbf{k}_2)$. It follows, in particular, that the direction of the maximum for the Stokes (anti-Stokes) component goes over into the direction of the maximum for the anti-Stokes (Stokes) component if the pump beams 1 and 2 change places. Recognizing, furthermore, that according to (9) and (10) the spectral frame of the Stokes and anti-Stokes components are the same (mirror-symmetry in the case of the Rayleigh line), and the scattering cross sections (in the direction of the maxima) are equal (accurate to terms $\sim\Omega/\omega$), we consider hereafter coherent scattering at only the anti-Stokes frequency $\omega_s^{(+)}=\omega_3+\Omega=\omega_1+\omega_3-\omega_2$, which has a maximum in the direction $\mathbf{k}_s^{(+)}=\mathbf{k}_3+\mathbf{q}=\mathbf{k}_1+\mathbf{k}_3-\mathbf{k}_2$. The last condition is easiest to satisfy by choosing the vectors $\mathbf{k}_1$, $\mathbf{k}_2$, $\mathbf{k}_3$, and $\mathbf{k}_s^{(+)}$ lying in the same plane with angles θ_{13} (between $\mathbf{k}_1$ and $\mathbf{k}_3$), θ_{1s} (between $\mathbf{k}_1$ and $\mathbf{k}_s^{(+)}$), and θ (between $\mathbf{k}_1$ and $\mathbf{k}_{12}$)[1)] such as to satisfy the condition (it is recognized that $|\mathbf{k}_1|\approx|\mathbf{k}_2|\approx|\mathbf{k}_3|\approx|\mathbf{k}_s^{(+)}|$) $\cos\theta_{13}=\cos\theta+\cos\theta_{1s}-1$. We see therefore that $\theta_{13}\geqslant\theta$. In the experiment, it is advantageous to choose the scattering direction $\mathbf{k}_s^{(+)}$ along the bisector of the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$, i.e., to put $\theta_{1s}=\theta/2$. Then, given the angle θ, the "synchronization angle" is determined from the equation

$$\cos\theta_{13}=\cos\theta+\cos(\theta/2)-1. \tag{11}$$

The Rayleigh and the Mandel'shtam-Brillouin lines are well resolved if $\chi q^2\ll v_sq$ and $\Gamma q^2\ll v_sq$, which is equivalent to the condition that the absorption of sound with wavelength $\Lambda=2\pi/q$ be small. For most liquids and compressed gases this condition is known to be sufficiently well satisfied up to hypersonic frequencies ($q\sim10^5$ cm^{-1}). Assuming henceforth this condition to be satisfied, we consider separately the coherent scattering corresponding to the Rayleigh and Mandel'shtam-Brillouin lines.

A. Rayleigh line. According to (9), to observe the spectral shape of this line the difference Ω between the pump-wave frequencies should be tuned in the interval $0 \leq \Omega \leq \chi q^2$, and their spectral widths $\Delta\omega_{1,2}$ must satisfy the condition $\Delta\omega_{1,2} \ll \chi q^2$. The last condition restricts the minimal values of q, i.e., of the angle θ between the vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. If, for example, we use the emission of a helium-neon laser ($\lambda = 0.63\ \mu$m) with an emission line width $(\Delta\omega_{1,2}/2\pi c) \approx 10^{-5}$ cm^{-1}, then for typical liquids far from the critical point ($\chi \sim (1-0.5)\times 10^{-3}$ cm^2/sec) we obtain the condition $\theta \geq 60°$; the frequency tuning range $\chi q^2/2\pi c$ should then amount to ~10^{-4} cm^{-1} [at $\theta = 60°$ we have $\theta_{13} \approx 69°$ from (11)].

The cross section for coherent scattering in the direction of the maximum is, on the basis of (9),

$$\left(\frac{d\sigma}{do}\right)_R^{\rm coh} = \frac{|\mathbf{A}_R^{\mp}|^2 r^2}{|\mathbf{A}_3|^2} = \left(\frac{\omega}{c}\right)^4 \left(\frac{\mu}{4\pi\rho c_p}\right)^2 \frac{V^2 (\partial\varepsilon/\partial T)_p^2 I_1 I_2}{\Omega^2 + (\chi q^2)^2}, \qquad (12)$$

where $I_{1,2} = (cn/8\pi)|\mathbf{A}_{1,2}|^2$ is the intensity of the pump beams.[2)] The signal-power gain due to coherent four-photon scattering compared with thermal Rayleigh scattering is defined as the ratio

$$\eta_R = \frac{(d\sigma/do)_R^{\rm coh}\ (\Delta o)^{\rm coh}}{(d\sigma/do\, d\Omega)_R^{\rm therm} \Delta o\, \Delta\Omega}. \qquad (13)$$

Here

$$\left(\frac{d\sigma}{do\, d\Omega}\right)_R^{\rm therm} = \frac{(\omega/c)^4}{16\pi^2} V \left(\frac{\partial\varepsilon}{\partial T}\right)_p^2 \frac{T^2 \sin^2\varphi}{\rho c_p} \frac{\chi q^2/\pi}{\Omega^2 + (\chi q^2)^2} \qquad (14)$$

is the cross section of thermal Rayleigh scattering (see[9]) ($q = 2k\sin(\vartheta/2)$, ϑ is the thermal-scattering angle, φ is the angle between the vectors $\mathbf{A}_3$ and $\mathbf{k}_s$, T is the temperature of the medium in energy units), $\Delta\Omega$ is the scattered-radiation photoreceiver bandwidth and satisfies the conditions

$$\chi q^2 \gg \Delta\Omega \gtrsim \Delta\omega_{1,2}; \qquad (15)$$

$(\Delta o)^{\rm coh}$ is the solid angle in which the coherent radiation propagate, with the axis along the direction of $\mathbf{k}_s^{(+)} = \mathbf{k}_1 + \mathbf{k}_3 - \mathbf{k}_2$. In order of magnitude we have $(\Delta o)^{\rm coh} \sim (\lambda/a)^2 = (2\pi/ka)^2$, where a is the radius of light beams 1, 2, and 3; Δo is the solid angle from which the scattered radiation is gathered by the photoreceiver. This angle must satisfy the conditions[3)]

$$\frac{q/k}{(1-(q/2k)^2)^{1/2}} \gg (\Delta o)^{1/2}, \quad \Delta o \gtrsim (\Delta o)^{\rm coh}. \qquad (16)$$

Substituting in (13) expressions (12) and (14) (at $\varphi = \pi/2$) and putting $\Delta o \approx (\Delta o)^{\rm coh}$, $V = \pi k a^4/\xi$ (i.e., assuming that the volume V amounts to a fraction of the focal volume of each of the intersecting light beams 1, 2, and 3), we obtain for the gain

$$\eta_R = \frac{k\mu^2}{\xi\rho c_p} \frac{P_1 P_2}{T^2 (\chi q^2)^2} \left(\frac{\chi q^2}{\Delta\Omega}\right)^2, \qquad (17)$$

where $P_{1,2} = \pi a^2 I_{1,2}$ are the total powers of the pump light beams. At $P_1 = P_2 = 10^{-1}$ W, $\mu = 0.1$ cm^{-1}, $k = 1.5 \times 10^5$ cm^{-1}, $T = 300$ K, $\chi q^2 = 10^7$ sec^{-1}, $\rho = 1$ g/cm^3, $c_p = 1$ cal/g-deg, $\chi q^2/\Delta\Omega = 10$, and $\xi = 3$ we have $\eta_R \approx 10^5$. Increasing the laser powers P_1 and P_2 and the absorption coefficient[4)] μ leads to a strong increase of η_R. It must be borne in mind, however, that increases of $P_{1,2}$ and μ can heat the scattering volume of the medium. To avoid strong heating it is necessary to reduce the degree of focusing of the beams (i.e., to decrease $I_{1,2}$).

B. Mandel'shtam-Brillouin line. According to (10), to observe the spectral shape of this line of difference Ω the pump-wave frequencies must be tunable in the interval $0 \leq \Omega \leq \Gamma q^2$, and their spectral widths $\Delta\omega_{1,2}$ must satisfy the condition $\Delta\omega_{1,2} \ll \Gamma q^2/2$. Just as in the case of the Rayleigh line, the last condition restricts the minimal values of the angle θ. If we use here, too, a helium-neon laser with the same spectral line width 10^{-5} cm^{-1} then the minimum attainable value of the angle θ decreases to approximately 25°, because the kinematic viscosity $\nu = \eta/\rho$ of most liquids exceeds by approximately one order of magnitude the thermal diffusivity χ (at $\theta = 25°$ we have $\theta_{13} \approx 30°$). On the basis of (10), the cross section for coherent scattering in the direction of the maximum is

$$\left(\frac{d\sigma}{do}\right)_{M-B}^{\rm coh} = \frac{|\mathbf{A}_{M-B}^{(\mp)}|^2 r^2}{|\mathbf{A}_3|^2} = \frac{(\omega/c)^4}{2^8\pi^2\varepsilon c^2} \left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_s^2 \left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_T^2 \frac{\beta_s^2 v_s^2 q^2 V^2 I_1 I_2}{(\Omega - v_s q)^2 + (\Gamma q^2/2)^2}. \qquad (18)$$

Account is taken here of the fact that $\Gamma q^2 \ll v_s q$ and $\Omega > 0$, and also that $(\partial\epsilon/\partial p)_s = \beta_s(\rho\partial\epsilon/\partial\rho)_s$, where $\beta_s = -(1/V)(\partial V/\partial p)_s$ is the adiabatic compressibility.

The signal power gain $\eta_{M\text{-}B}$ due to coherent scattering compared with thermal Mandel'shtam-Brillouin scattering is determined by a relation similar to (13), with $(d\sigma/do)_R^{\rm coh}$ replaced by $(d\sigma/do)_{M\text{-}B}^{\rm coh}$ and $(d\sigma/do\, d\Omega)_R^{\rm therm}$ by

$$\left(\frac{d\sigma}{do\, d\Omega}\right)_{M-B}^{\rm therm} = \frac{(\omega/c)^4}{32\pi^2} V \left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_s^2 \beta_s T \sin^2\varphi \frac{\Gamma q^2/2\pi}{(\Omega - v_s q)^2 + (\Gamma q^2/2)^2}, \qquad (19)$$

which is the thermal Mandel'shtam-Brillouin scattering cross section corresponding to the anti-Stokes component of the doublet.[9] The receiver bandwidth $\Delta\Omega$ must now satisfy the condition $(\Gamma q^2/2) \gg \Delta\Omega \gtrsim \Delta\omega_{1,2}$; the solid angles $(\Delta o)^{\rm coh}$ and Δo have the same meaning as in the case of the Rayleigh component, and satisfy the conditions (16) (see also footnote 3).

Taking all the foregoing into account, we obtain for $\eta_{M\text{-}B}$ the expression

$$\eta_{M-B} = \frac{k\beta_s}{8\xi\varepsilon c^2 T} \left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_T^2 \left(\frac{v_s q}{\Gamma q^2/2}\right)^2 P_1 P_2 \left(\frac{\Gamma q^2}{2\Delta\Omega}\right). \qquad (20)$$

We have put here again $V = \pi k a^4/\xi$. At the same laser-beam powers $P_1 = P_2 = 10^{-1}$ W and $2v_s q/\Gamma q^2 = 10^2$, $\Gamma q^2/2\Delta\Omega = 10$, $\beta_s = 10^{-4}$ bar^{-1} and $\xi = 3$, the gain is $\eta_{M\text{-}B} \approx 10^3$.

It is of interest to examine in this case the generalization of the "Landau-Placzek ratio," which determines the ratio $(I_R/2I_{M-B})^{therm}$ of the frequency-integrated intensity of the thermal scattering in the Rayleigh line to the integrated intensity of the thermal scattering in the two lines of the Mandel'shtam-Brillouin scattering and equal to $L(\gamma-1)$, where

$$L=\left(\frac{1}{\alpha}\frac{\partial\varepsilon}{\partial T}\right)_p^2\Big/\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_s^2, \quad \alpha=\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p$$

is the coefficient of thermal volume expansion.[5)] Writing down an analogous ratio for the case of coherent scattering, namely

$$\left(\frac{I_R}{2I_{M-B}}\right)^{coh}=\int\left(\frac{d\sigma}{do}\right)_R^{coh}d\Omega\Big/2\int\left(\frac{d\sigma}{do}\right)_{M-B}^{coh}d\Omega$$

(it is assumed that the cross sections correspond to the directions of the maximum scattering), we obtain on the basis of (12) and (18) (recognizing that $\alpha^2T/\rho c_p\beta_s=\gamma-1$)

$$\left(\frac{I_R}{2I_{M-B}}\right)^{coh}=\left(\frac{I_R}{2I_{M-B}}\right)^{therm}\left(\frac{\mu/k_0}{v_s/c}\right)^2\frac{\Gamma/\chi}{\rho c_pT\beta_s(\rho\, d\varepsilon/\partial\rho)_T^2}\sin^{-2}\frac{\theta}{2}, \quad (21)$$

where $k_0=\omega/c$. We note that for most liquids at room temperatures $\rho c_pT\beta_s(\rho\partial\epsilon/\partial\rho)_T^2\sim 1$. It is seen from (21) that the ratio $(I_R/2I_{M-B})^{coh}$ can be varied in a wide range by changing the absorption coefficient μ and the angle θ.

The foregoing estimates show that there are indeed all grounds for hoping that the use of coherent four-photon Rayleigh scattering will uncover new experimental possibilities in the study of molecular scattering of light, particularly in the region near the critical liquid-vapor phase-transition point, where the experiments are difficult. In this region, the thermal Rayleigh scattering, while increasing significantly (critical opalescence), has a narrow forward directivity (small values of q) because of the abrupt increase of the fluctuation correlation radius. It becomes therefore extremely difficult to obtain information on the dynamic properties of the medium at large q (small wavelengths $\Lambda=2\pi/q$). When the considered coherent scattering is used, no upper bounds are imposed on q, so that the extraction of this information encounters no new difficulties.

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1) All the angles are reckoned in the same direction away from k_1.
2) To simplify the formulas it is assumed that beams 1 and 2 are linearly polarized perpendicular to the incidence plane.
3) The meaning of the first condition (16) is that the thermal-scattering angle interval $\Delta\vartheta\approx(\Delta o)^{1/2}$, which is determined by the angle aperture of the photoreceiver, should be small enough for the corresponding increment $\Delta q=k\cos(\vartheta/2)\Delta\vartheta$ to be small in comparison with $q=2k\sin(\vartheta/2)$. We note that if the two conditions (16) (or the two conditions (15)) are mutually incompatible, then to observe coherent and thermal scattering it is necessary to use photoreceivers with different values of $\Delta 0$ and $\Delta\Omega$.
4) The value of μ can be increased by adding to the investigated liquid a small amount of strongly absorbing material.
5) The "classical" Landau-Placzek ratio (in the form obtained by them) corresponds to $L=1$. In fact, L is not identically equal to unity, but is close to it for most liquids and gases (for details see [9]).

1. S. A. Akhmanov and N. I. Koroteev, Zh. Eksp. Teor. Fiz. **67**, 1306 (1974) [Sov. Phys. JETP **40**, 650 (1975)].
2. M. D. Levenson and N. Bloembergen, Phys. Rev. B **10**, 4447 (1974).
3. R. F. Begley, A. B. Harvey, and B. L. Byer, Appl. Phys. Lett. **25**, 387 (1974).
4. F. V. Bunkin, F. V. Kalinin, and P. I. Pashinin, Kvantovaya Elektron. (Moscow) **5**, 468 (1978) [Sov. J. Quantum Electron. **8**, 278 (1978)].
5. D. W. Pohl, S. E. Schwarz, and V. Irniger, Phys. Rev. Lett. **31**, 32 (1973).
6. D. W. Pohl and V. Irniger, Phys. Rev. Lett. **36**, 480 (1976).
7. D. Heiman, R. W. Hellwarth, M. D. Levenson, and G. Martin, Phys. Rev. Lett. **36**, 189 (1976).
8. A. F. Bunkin, S. G. Ivanov, and N. I. Koroteev, Dokl. Akad. Nauk SSSR **233**, 338 (1977) [Sov. Phys. Dokl. **22**, 168 (1977)].
9. I. L. Fabelinskiĭ, Molekulyarnoe rasseyanie sveta (Molecular Scattering of Light), Nauka, 1965.

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