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SPECTRAL WIDTH OF LIGHT SPONTANEOUSLY SCATTERED IN A PERFECT GAS

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The mean square frequency shift $\overline{\delta\omega^2}$ for spontaneous light scattering in a perfect gas is calculated. It is shown that the value of $\overline{\delta\omega^2}$ remains constant for variation in the pressure at a fixed value of $|\mathbf{q}|$, where $\mathbf{q}$ is the change in the photon wave vector in the scattering process. The value of $\overline{\delta\omega^2}$ for spontaneous scattering on anisotropic molecules and for spontaneous scattering on electrons in plasma is also calculated.

As is well known, the spectrum of light scattering in a gas undergoes very significant changes upon change in the density. So far as the scalar part of the scattering is concerned, at low gas densities (such that $ql \gg 1$) the spectrum of the scattered light is determined by the Doppler shift on moving molecules that have a Maxwellian velocity distribution. Here l is the free path length of the molecules, $q = |\mathbf{q}| = |\mathbf{k}_1 - \mathbf{k}_2|$ is the change in the photon wave vector in the scattering process. At high pressures (such that $ql \sim 1$) the spectrum of the scalar part of the scattering contains in place of the Doppler curve components of the Mandel'shtam-Brillouin doublet and a central entropy peak. In the intermediate case $ql \gg 1$ the spectrum has a rather complicated shape and should be calculated by solution of the Boltzmann equation—see, for example[1,2].

In the present paper, we call attention to the fact that the second moment of the spectral distribution $\overline{\delta\omega^2}$, expressed in units of q, remains constant over the entire range of pressures from $ql \gg 1$ to $ql \ll 1$. The quantity $\overline{\delta\omega^2}$ also possesses certain invariance properties, even in light scattering in a plasma.

We denote the normalized spectrum of light scattered through a given angle by $g(\omega - \omega_0)$, where $\int g(\omega - \omega_0)d\omega = 1$, and introduce the mean square frequency shift $\overline{\delta\omega^2}$:

$$\overline{\delta\omega^2} = \int\limits_{-\infty}^{+\infty} g(\omega-\omega_0)(\omega-\omega_0)^2\,d\omega. \tag{1}$$

We get the following expression for this quantity in the present work:

$$\overline{\delta\omega^2} = q^2kT\left(\sum_l N_{(l)}\alpha^2_{(l)}/m_{(l)}\right)\Big/\left(\sum_l N_{(l)}\alpha^2_{(l)}\right). \tag{2}$$

Here $N_{(l)}$, $\alpha_{(l)}$, $m_{(l)}$ are respectively the concentration, polarizability and mass of the molecules of the l-th component of a gas mixture; kT is the temperature in energy units.

For proof of formula (2), we introduce the normalized correlation function

$$\gamma(\tau) = \int\limits_{-\infty}^{+\infty} g(\delta\omega)e^{-i\delta\omega\tau}\,d\delta\omega; \tag{3}$$

here

$$\gamma(0) = 1,\quad \left.\frac{d\gamma}{d\tau}\right|_{\tau=0} = -i\overline{\delta\omega},\quad \left.\frac{d^2\gamma}{d\tau^2}\right|_{\tau=0} = -\overline{\delta\omega^2}. \tag{4}$$

In scattering in the classical region (for $h(\omega' - \omega) \ll kt$), the spectrum $g(\delta\omega)$ is symmetric and $\overline{\delta\omega} = 0$. The normalized correlation function $\gamma(\tau)$ for scattering on molecules with a scalar polarizability α is equal to

$$\gamma(\tau) = \frac{\langle\Sigma(t+\tau)\Sigma^*(t)\rangle}{\langle\Sigma(t)\Sigma^*(t)\rangle},\quad \Sigma(t) = \sum \alpha_s e^{i\mathbf{q}\mathbf{r}_s(t)}, \tag{5}$$

where the random function $\mathbf{r}_s(t)$ gives the location of the s-th scattering molecule at the instant of time t; the asterisk denotes the complex conjugate. Double differentiation of (5) gives

$$\overline{\delta\omega^2} = -\frac{1}{\langle\Sigma(t)\Sigma^*(t)\rangle}\left\langle\sum_{ss'}\alpha_s\alpha_{s'}\exp\{i\mathbf{q}[\mathbf{r}_s(t)-\mathbf{r}_{s'}(t)]\}[i\mathbf{q}\ddot{\mathbf{r}}_s - (\mathbf{q}\dot{\mathbf{r}}_s)(\mathbf{q}\dot{\mathbf{r}}_s)]\right\rangle \tag{6}$$

It is important that the quantities under the averaging sign in (6) refer to the same instant of time. With account of this, we have the following equations for an ideal gas (i.e., with neglect of terms $\sim Na^3$, where a is the dimension of the molecule):

$$\left\langle\sum_{ss'}\alpha_s\alpha_{s'}\exp\{i\mathbf{q}(\mathbf{r}_s-\mathbf{r}_{s'})\}\right\rangle = \sum_{ss'}\alpha_s\alpha_{s'}\delta_{ss'} = \sum_s\alpha_s^2, \tag{7.1}$$

$$\left\langle\sum_{ss'}\alpha_s\alpha_{s'}\exp\{i\mathbf{q}(\mathbf{r}_s-\mathbf{r}_{s'})\ddot{\mathbf{r}}_s\}\right\rangle = 0, \tag{7.2}$$

$$\left\langle\sum_{ss'}\alpha_s\alpha_{s'}e^{i\mathbf{q}(\mathbf{r}_s-\mathbf{r}_{s'})}(\dot{\mathbf{r}}_s)_i(\dot{\mathbf{r}}_s)_k\right\rangle = kT\delta_{ik}\sum_{ss'}\frac{\alpha_s\alpha_{s'}\delta_{ss'}}{m_s} = kT\delta_{ik}\sum_s\frac{\alpha_s^2}{m_s}, \tag{7.3}$$

substitution of which in (6) also gives Eq. (2), pertaining to scattering of the scalar type. We note the peculiar role of molecular collisions in the problem under study: they have little effect on the total light scattering cross section and on the value of $\overline{\delta\omega^2}$ (corrections $\sim Na^3$, which we neglect), but strongly affect the shape of the spectrum (corrections $\sim(ql)^{-1} \approx Na^2q^{-1}$ if we go in the direction of low density; these corrections are taken into account exactly in Eq. (2).

In the case $ql \gg 1$, the scattering spectrum is given by the superposition of the Doppler curves from the different components of the mixture and Eq. (2) becomes trivial.

In the case $ql \ll 1$, the spectrum contains the Mandel'shtam-Brillouin doublet and the central entropy-concentration part. If we assume the gas to be a single-component one, the lines of the doublet and the entropy part are infinitely narrow (i.e., we neglect their broadening) having the ratio of intensities according to the Landau-Placzek formula, while for the sound velocity, we have the usual expression $v_S = (c_p kT/c_V m)^{1/2}$; for such a model spectrum, the mean square width $\overline{\delta\omega^2}$ is also given by Eq. (2). Thus, Eq. (2) is in its way a generalization of the Landau-Placzek formula for the ideal gas to the case of arbitrarily large damping of hypersound and of a multicomponent mixture.

In the case of rotational Raman scattering of light the polarization structure of which in an ideal gas pertains to the symmetric trace-free type) one can expect a frequency shift $\delta\omega \sim \Omega_{\text{rot}} \sim \sqrt{kT/J}$, where $J \sim ma^2$ is the moment of inertia of the molecule; here we again consider the quasiclassical case when $\hbar\Omega_{\text{rot}} \ll hT$. In the general case of arbitrary anisotropy of the polarizability tensor α_{ik} and the inertial tensor J_{ik}, an explicit expression for the shape of the spectrum (even in the quasiclassical case) is lacking in the literature; there are only comparatively complicated formulas for the case of a molecule of the gyroscopic type (with coinciding axes for α_{ik} and J_{ik}), see[3]. It is seen that for the quantity $\overline{\delta\omega^2}$, it is possible even in the general case, to obtain an explicit formula (in the quasiclassical limit $\hbar\Omega_{\text{rot}} \ll kT$):

$$\overline{\delta\omega^2} = kT\frac{4\,\mathrm{Sp}(\tilde{\alpha}\tilde{\alpha})\,\mathrm{Sp}(J^{-1}) - 6\,\mathrm{Sp}(J^{-1}\tilde{\alpha}\tilde{\alpha})}{\mathrm{Sp}(\tilde{\alpha}\tilde{\alpha})}. \tag{8}$$

Here we have introduced the tensor $\tilde{\alpha}_{ik}$:

$$\tilde{\alpha}_{ik} = \alpha_{ik} - {}^1/_3\delta_{ik}\mathrm{Sp}\,\alpha, \tag{9}$$

by the same token, we have eliminated from consideration the scalar part of the scattering. At the same time, the total intensity of the trace-free part of the scattering that has been considered (the denominator in Eq. (8)) itself includes the contribution from the anisotropy fluctuation. The latter, in contrast to the rotational combination scattering, gives a very small shift of the frequency (see[4], Sec. 96). The derivation of Eq. (8), as also Eq. (2), is based on the double differentiation of the corresponding normalized correlation function and use of the following equality:

$$\langle \Omega_i \Omega_k \rangle = kT(J^{-1})_{ik}, \tag{10}$$

where $\mathbf{\Omega}$ is the angular velocity vector of molecular rotation. The applicability of Eq. (8) is also limited by the condition $Na^3 \ll 1$. Here the discrete lines of the rotational combination scattering can be shown to overlap because of collisions, and Eq. (8) will be valid as before.

In light scattering in a plasma, the scattering cross section dR/do of a unit volume integrated over the frequency also depends on q:

$$\frac{dR}{do} = N_e\frac{d\sigma_T}{do}\frac{(qr_D)^2 + z}{(qr_D)^2 + z + 1}, \tag{11}$$

where $d\sigma_T/do$ is the Thomson cross section, r_D the Débye radius, and z the ion charge; here the shape of the spectrum depends in complicated fashion on qr_D (see[5]). However, even in this case, it is possible to obtain an integrated expression[1] similar to Eq. (2):

$$\int \frac{d^2R}{do\,d\omega}(\omega - \omega_0)^2\,d\omega = q^2\frac{kT}{m_e}\frac{d\sigma_T}{do}; \tag{12}$$

here we have neglected those components on the right side of (12) which are proportional to m_i^{-1}, where m_i is the ion mass. We note that the dependence on the parameter qr_D drops out of Eq. (12). In a plasma, in contrast with a neutral gas, the scattering cross section, integrated over the frequency and referred to a single particle, changes materially with change in the pressure. In spite of this fact, the value of (12) remains invariant with an accuracy up to terms $\sim m_c/m_i$.

The quantity $\overline{\delta\omega^2}$ enters into the expression for the amplification coefficient in stimulated light scattering with broadband exciting radiation in the approximation of completely random phase. In fact, in the specified approximation, the intensity amplification coefficient $\beta[\text{cm}^{-1}]$ at a frequency is given by the convolution in frequency of the intensity distribution of the exciting radiation, and the amplification coefficient for monochromatic excitation. The latter quantity is connected with the frequency derivative of the cross section of spontaneous scattering (see, for example,[6]). As a result, we obtain

$$\beta(\omega) = \left(\frac{c}{\omega}\right)^2(2\pi)^3\frac{dR}{do}\int\frac{P(\omega')}{\hbar\omega'}\left[\exp\left\{\frac{\hbar(\omega' - \omega)}{kT}\right\} - 1\right]g(\omega - \omega')\,d\omega', \tag{13}$$

where $P(\omega)[\text{erg-cm}^{-2}\text{-sec}^{-1}\cdot(\text{rad/sec})^{-1}]$ is the spectral density of the power of the exciting radiation and $dR/do\ [\text{cm}^{-1}\,\text{sr}^{-1}]$ is the angular derivative of the scattering cross section per unit volume. For scalar scattering,

$$\frac{dR}{do} = \frac{1}{(4\pi)^2}\left(\frac{\omega_0}{c}\right)^4|\mathbf{e}_1\mathbf{e}_2^*|^2\sum_l N_{(l)}\alpha_{(l)}^2,$$

where $\mathbf{e}_1$ and $\mathbf{e}_2$ are the polarization unit vectors of exciting and scattered radiation. Assuming that $P(\omega)$ changes slowly over the entire range of the spectrum of spontaneous scattering, so that one can set

$$P(\omega') \approx P(\omega) + (\omega' - \omega)\,dP/d\omega$$

and taking it into account that $\hbar(\omega' = \omega) \ll kT$, we obtain

$$\beta(\omega) = -\frac{(2\pi)^3}{\omega}\left(\frac{c}{\omega}\right)^2\frac{dR}{do}\frac{1}{kT}\frac{dP}{d\omega}\overline{\delta\omega^2}. \tag{14}$$

It is interesting to note that the amplification coefficient from (14), after substitution of the expression (2) for $\overline{\delta\omega^2}$, turns out to be independent both of the temperature and of the free path length, and is proportional to q^2.

In certain cases, calculation of the energy put into the material in stimulated light scattering is of interest (see, for example, the problem of plasma heating due to the induced Compton effect[7]). In the random phase approximation, and for a broadband exciting radiation, the energy supplied to the material can also be expressed in terms of the mean square frequency shift for the case of spontaneous light scattering.

In conclusion, we note that measurement of the second derivative of the time correlation function of scattered radiation can be shown to be more attainable, due to the application of laser sources and methods of photostatistics in the recording process (see, for example,[8]).

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[1] Equations (11) and (12) are valid only under the assumption that the frequency of the incident light is much greater than the value of the plasma frequency.

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