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# Light scattering by particles suspended in a turbulent fluid†

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**Abstract.** The spectrum of light scattered by particles suspended in a turbulent fluid is investigated theoretically.

Laser light scattering by particles suspended in a fluid can be used to study the statistical properties of the fluid. A spectral measurement of a laser beam after interaction with particles suspended in a turbulent liquid has been recently performed (Pike *et al.* 1968) in order to gain information on the hydrodynamical velocity fluctuations of the medium. In that paper a simple model was used to connect the Doppler-shift broadening of the scattered light to the velocity fluctuations of the suspended particles. The experimental situation was such as to justify the assumption of particles following exactly the motion of the surrounding liquid.

Under the same hypothesis, we wish to give here a more complete theoretical treatment of the subject. The spectrum $I(\boldsymbol{k}, \omega)$ of scattered light will be obtained by introducing the 'microscopic density' of suspended particles, thus obtaining a dependence upon all-order velocity correlations. Furthermore, the moments of the spectral distribution will be easily shown to be related to equal space and time velocity correlations of the corresponding order.

The electric field scattered by $N$ particles in the direction $\boldsymbol{R}$ may be written as

$$\boldsymbol{E}(\boldsymbol{R}, t) = \mathscr{E}(\boldsymbol{k}) \exp\left\{\mathrm{i}\omega_0 \left(t - \frac{R}{c}\right)\right\} \sum_{j=1}^{N} \int_{\mathrm{V}} \mathrm{e}^{-\mathrm{i}\boldsymbol{k}.\boldsymbol{r}}\, \delta\{\boldsymbol{r} - \boldsymbol{r}_j(t)\}\, \mathrm{d}\boldsymbol{r} \tag{1}$$

where $\omega_0$ is the frequency of the incident light of wave number $\boldsymbol{k}_0$, $\mathscr{E}(\boldsymbol{k})$ the scattered amplitude associated with the single particle whose position is labelled with $\boldsymbol{r}_j(t)$, V is the scattering volume and $\boldsymbol{k} = \boldsymbol{k}_0 - k_0\boldsymbol{R}/R$. Under the hypothesis of homogeneity and stationariness, the associated intensity can be written as

$$\begin{aligned} I(\boldsymbol{k}, \omega) &= \int_{-\infty}^{+\infty} \mathrm{e}^{-\mathrm{i}\omega t} \langle \boldsymbol{E}(\boldsymbol{R}, t) \,.\, \boldsymbol{E}^*(\boldsymbol{R}, 0)\rangle\, \mathrm{d}t \\ &= A(\boldsymbol{k}) \int_{-\infty}^{+\infty} \mathrm{d}t\, \mathrm{e}^{-\mathrm{i}(\omega-\omega_0)t} \sum_{i=1}^{N} \sum_{j=1}^{N} \iint_{\mathrm{V}} \mathrm{d}\boldsymbol{r}\, \mathrm{d}\boldsymbol{r}'\, \mathrm{e}^{-\mathrm{i}\boldsymbol{k}.\boldsymbol{r}} \langle \delta\{\boldsymbol{r}' + \boldsymbol{r} - \boldsymbol{r}_j(t)\}\delta\{\boldsymbol{r}' - \boldsymbol{r}_i(0)\}\rangle \end{aligned} \tag{2}$$

where $A(\boldsymbol{k}) = |\mathscr{E}(\boldsymbol{k})|^2$ and the brackets stand for ensemble average.

The scatterer motion is described by

$$\boldsymbol{r}_j(t) = \boldsymbol{r}_{j0} + \int_0^t \boldsymbol{U}\{\boldsymbol{r}_j(t'), t'\}\, \mathrm{d}t' \tag{3}$$

and $\boldsymbol{U}(\boldsymbol{r}, t)$ represents the fluid velocity field and $\boldsymbol{r}_{j0} \equiv \boldsymbol{r}_j(0)$. Inserting equation (3) into equation (2) we obtain

$$\begin{aligned} I(\boldsymbol{k}, \omega) = A(\boldsymbol{k}) &\int_{-\infty}^{+\infty} \mathrm{d}t \exp\{-\mathrm{i}(\omega - \omega_0)t\} \\ &\times \sum_{i=1}^{N} \sum_{j=1}^{N} \left\langle \exp\left[-\mathrm{i}\boldsymbol{k} \,.\, (\boldsymbol{r}_{j0} - \boldsymbol{r}_{i0}) - \mathrm{i}\boldsymbol{k} \,.\int_0^t \boldsymbol{U}\{\boldsymbol{r}_j(t'), t'\}\, \mathrm{d}t'\right]\right\rangle. \end{aligned} \tag{4}$$

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By separating $\boldsymbol{U}(\boldsymbol{r}, t)$ into its mean and fluctuating parts $\boldsymbol{U}_0$ and $\boldsymbol{U}_1(\boldsymbol{r}, t)$, equation (4) yields

$$I(\boldsymbol{k}, \omega) = A(\boldsymbol{k}) \int_{-\infty}^{+\infty} \mathrm{d}t \exp\{-\mathrm{i}(\omega - \Omega)t\} \times \sum_{i=1}^{N} \sum_{j=1}^{N} \left\langle \exp\{-\mathrm{i}\boldsymbol{k} \,.\, (\boldsymbol{r}_{j0} - \boldsymbol{r}_{i0})\} \exp\left[-\mathrm{i}\boldsymbol{k} \,.\, \int_0^t \boldsymbol{U}_1\{\boldsymbol{r}_j(t'), t'\} \,\mathrm{d}t'\right] \right\rangle \tag{5}$$

where $\Omega = \omega_0 - \boldsymbol{k} \,.\, \boldsymbol{U}_0$, the Doppler shift associated with the drift motion being $-\boldsymbol{k} \,.\, \boldsymbol{U}_0$. The dependence of $I(\boldsymbol{k}, \omega)$ on every order correlation functions of the fluid velocity field is explicitly shown by expanding the exponential in equation (5)

$$I(\boldsymbol{k}, \omega) = A(\boldsymbol{k}) \int_{-\infty}^{+\infty} \mathrm{d}t \exp\{-\mathrm{i}(\omega - \Omega)t\} \sum_{i=1}^{N} \sum_{j=1}^{N} \left\langle \exp\{-\mathrm{i}\boldsymbol{k} \,.\, (\boldsymbol{r}_{j0} - \boldsymbol{r}_{i0})\} \times \left(1 - \mathrm{i}\boldsymbol{k} \,.\, \int_0^t \boldsymbol{U}_1\{\boldsymbol{r}_j(t'), t'\} \,\mathrm{d}t' - \tfrac{1}{2}\left[\boldsymbol{k} \,.\, \int_0^t \boldsymbol{U}_1\{\boldsymbol{r}_j(t'), t'\} \,\mathrm{d}t'\right]^2 + \dots\right) \right\rangle. \tag{6}$$

We now observe that it is possible to average separately over the suspended particles and over the velocities of the turbulent field. As a matter of fact, the ensemble average over the particles means the average over the initial positions and velocities which can be arbitrarily chosen. The averaging over the turbulent field yields the connection between the scattered intensity and the 'Lagrangian time correlations' of the velocities. For example, the quadratic term of equation (6) may be expressed, under isotropic conditions, as

$$\overline{-\tfrac{1}{2}\left(\boldsymbol{k} \,.\, \int_0^t \boldsymbol{U}_1\{\boldsymbol{r}_j(t'), t'\} \,\mathrm{d}t'\right)^2} = -\frac{k^2}{3} \overline{U_1{}^2} \int_0^t \mathrm{d}\tau (t - \tau) R_{\mathrm{L}}(\tau) \tag{7}$$

where $R_{\mathrm{L}}(\tau) = \overline{U_1(t) \,.\, U_1(t+\tau)} / \overline{U_1{}^2}$ is the second-order Lagrangian time correlation (Hinze 1959, pp. 42–9) and the bar indicates average over turbulence.

Let us now evaluate the moments of the intensity distribution defined as

$$M(n) \equiv \int_{-\infty}^{+\infty} \mathrm{d}\omega \, I(\boldsymbol{k}, \omega)(\omega - \Omega)^n. \tag{8}$$

The $M(n)$ are directly related with equal space and time correlations. Taking into account the property of the $n$th derivative of the δ-function we easily obtain expressions for even-order moments, while the odd ones vanish under isotropic conditions. We obtain, for example,

$$\begin{aligned} M(0) &= 2\pi A(\boldsymbol{k}) N \\ M(2) &= 2\pi A(\boldsymbol{k}) N k^2 \overline{U_{1l}{}^2} \\ M(4) &= 2\pi A(\boldsymbol{k}) N (k^4 \overline{U_{1l}{}^4} + k^2 \overline{U_{1l}'^2}) \end{aligned} \tag{9}$$

where $U_{1l}$ and $U_{1l}'$ are the components of the turbulent velocity and acceleration along an axis arbitrarily chosen. In fact, equations (9) have been obtained considering an axis parallel to $\boldsymbol{k}$ but the result is quite general owing to the isotropy of turbulence. We wish also to note that we have omitted the correlation terms which can be shown to vanish in an isotropic case (Hinze 1959, p. 40).

More generally one obtains

$$M(2n) = 2\pi A(\boldsymbol{k}) N k^{2n} \overline{U_{1l}{}^{2n}} \tag{10}$$

if one retains only the terms of highest order in $k$ which corresponds to assuming the wavelength of the light much smaller than the characteristic length of turbulence.

If, for example, one assumes the validity of the joint-Gaussian distribution hypothesis for the velocity field (Batchelor 1951, Chandrasekhar 1951), that is $\overline{U_{1l}^{2n}} = (2n-1)!!(\overline{U_{1l}^{2}})^n$, the $M(n)$ factorize in the following form:

$$\frac{M(2n)}{M(0)} = (2n-1)!!\left\{\frac{M(2)}{M(0)}\right\}^n. \tag{11}$$

Equation (11) is consistent with a Gaussian behaviour of $I(\boldsymbol{k}, \omega)$. This is, for example, the case of the experimental results reported in the paper by Pike *et al.* (1968).

## References

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