Study of the Scattering Regime Diagrams

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Based on a stochastic, wave-mechanical theory of Keller, the scattering regime diagrams for several relative complex refractive indexes of scatterers are presented. The criterion adopted here for demarcating the uncorrelated scattering regime from the correlated scattering regime is given by $|\gamma_2 - 1| \leq 0.1138$, where γ_2 represents the ratio of the actual extinction coefficient of scatterers to the extinction coefficient derived from Mie's theory. The diagrams were found to consist of three major regimes: one uncorrelated scattering regime and two correlated scattering regimes. The detailed profiles of each regime are considerably influenced by the values of the relative complex refractive index m of scatterers. Moreover, it was found that the demarcation curves are characterized by the presence of small and large size parameter limits: the small size parameter limit varies with the value of m, whereas the large size parameter limit does not depend on m and is given by $f_v = 0.07899$, where f_v represents the volume fraction of scatterers.

Nomenclature

A'	= integral defined by Eq. (8)
B'	= integral defined by Eq. (9)
${}^eB_\ell, {}^mB_\ell$	= Mie's coefficients
d_p	= representative diameter of scatterers
f_v	= volume fraction of scatterers ($=\pi d_p^3 n_p/6$)
f_{vL}^*	= volume fraction corresponding to the large size parameter limit on the demarcation curve
f_{vs}^*	= volume fraction corresponding to the small size parameter limit on the demarcation curve
$g(r)$	= radial distribution function
$\mathrm{Im}(z)$	= imaginary part of z
K_i	= quantity defined by Eq. (14)
K_r	= quantity defined by Eq. (14)
k	= effective propagation constant for a scattering medium ($=k_1 + k_2 i$)
k_0	= propagation constant for a scatterer-free medium
k_1	= real part of k
k_2	= imaginary part of k
m	= relative complex refractive index of scatterers ($=m_1 + m_2 i$)
m_1	= real part of m
m_2	= imaginary part of m
n_p	= number density of scatterers
$\mathrm{Re}(z)$	= real part of z
$S(0)$	= forward scattering amplitude
S_I	= imaginary part of $S(0)$
S_R	= real part of $S(0)$
z	= complex number
α	= size parameter ($=k_0 d_p$)
β	= extinction coefficient of scatterers
γ_1	= quantity defined by Eq. (4)
γ_2	= extinction coefficient ratio defined by Eq. (5)
η	= dimensionless radial distance
$\Phi(\eta)$	= hard-sphere potential

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Introduction

It is important for theoretical analysis of radiative transfer within a scattering medium to know whether the radiative properties of a cloud of scatterers can be given by simply summing up the radiative properties of spheres constituting the scattering medium. If it is possible, uncorrelated scattering is said to occur within the medium, otherwise, correlated scattering is said to occur. Qualitatively speaking, the above condition for uncorrelated scattering is satisfied whenever the distance between scatterers in a layer is much greater than the scatterer diameters and the wavelength of the incident radiation. On the other hand, correlated scattering arises when the number density of scatterers becomes high and the scattering due to an individual scatterer is influenced by neighboring scatterers. However, the quantitative measure for uncorrelated or correlated scattering is indeed requisite for actual computations. In order to meet this requirement, the scattering regime diagram, which demarcates the uncorrelated scattering regime from the correlated scattering one on the plane defined by the size parameter, and the volume fraction of scatterers has been proposed by several researchers.[1–4] The characteristic features of this diagram in the small size parameter region were understood to some extent throughout the study of Cartigny et al.,[3] but the effects of the relative complex refractive index of scatterers on this diagram have not yet been discussed in the small size parameter region, and also the diagram in the large size parameter region has been subject to controversial issues even now.[1,2,4]

The present study endeavors to give an answer to these subjects and to confirm the scattering regime diagram over the entire region of the size parameter on the basis of a stochastic, wave-mechanical theory of Keller.[5]

Theory

According to the stochastic, wave-mechanical theory proposed by Keller, the effective propagation constant k of a coherent wave propagating through a random distribution of scatterers is given by

$$k^2 = k_0^2 + \frac{[4\pi n_p S(0) i]}{k_0} - \left\{\frac{[4\pi n_p S(0)]^2}{k_0^3}\right\} \int_0^\infty e^{ik_0 r}[g(r) - 1] \sin k_0 r \, dr \tag{1}$$

where the Percus-Yevick distribution function governed by Eq. (2) was assumed for $g(r)$.[6]

$$g(\eta)e^{\Phi(\eta)} = 1 + 12f_v \int_0^\infty \int_{|\eta - s|}^{\eta + s} g(s) \times [1 - e^{\Phi(s)}][g(t) - 1]st \, dt \frac{ds}{\eta} \tag{2}$$

where $\eta = r/d_p$ and $\Phi(\eta)$ represents the hard-sphere potential [$\Phi(\eta) = \infty$ for $\eta < 1$ and 0 for $\eta \geqq 1$].

Equation (1) was derived from the second-order perturbation equation to a stochastic wave equation in media of discrete scatterers.

The extinction coefficient β can be evaluated from k as follows:

$$\beta = 2\mathrm{Im}(k) \tag{3}$$

where $k = k_1 + k_2 i$. In order to calculate k, we introduce the following quantities:

$$\gamma_1 = 2k_1/[4\pi n_p \mathrm{Re}[S(0)]/k_0^2] \tag{4}$$

$$\gamma_2 = 2k_2/[4\pi n_p \mathrm{Re}[S(0)]/k_0^2] \tag{5}$$

Note that γ_2 defined by Eq. (5) represents the ratio between the correlated extinction coefficient given by Eq. (3) and the uncorrelated extinction coefficient calculated from Mie's theory.[7] Substituting k_1 and k_2 derived from Eqs. (4) and (5) into the left side of Eq. (1) and picking up the real and imaginary parts yield the following results:

$$\gamma_1\gamma_2 = (32S_R/3Q_{ex}^2 f_v \alpha) - (64/Q_{ex}^2\alpha^3)[2S_R S_I A' + B'(S_R^2 - S_I^2)] \tag{6}$$

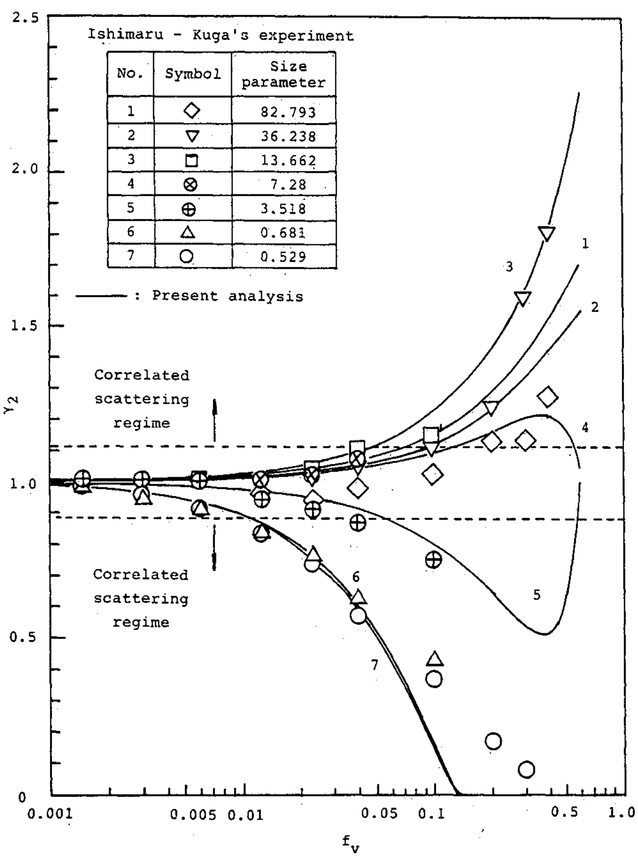

Fig. 1 Comparison between theory and Ishiumaru-Kuga's experiment for the extinction coefficient ratio γ_2.

$$\gamma_1^2 - \gamma_2^2 = (64\alpha^2/9Q_{ex}^2 f_v^2) - (64S_I/3Q_{ex}^2 f_v \alpha) - (128/Q_{ex}^2\alpha^3)[(S_R^2 - S_I^2)A' - 2S_R S_I B'] \tag{7}$$

where $Q_{ex} = 4\mathrm{Re}[S(0)]/\alpha^2$, and A' and B' are the integrals defined by

$$A' = \int_0^\infty [g(\eta) - 1] \sin 2\alpha\eta \cos 2\alpha\eta \, d\eta \tag{8}$$

$$B' = \int_0^\infty [g(\eta) - 1] \sin 2\alpha\eta \sin 2\alpha\eta \, d\eta \tag{9}$$

Furthermore, $S(0)$ is given by

$$S(0) = \frac{1}{2}\sum_{\ell=1}^{\infty} (-i)^{\ell+1}\ell(\ell+1)({}^eB_\ell + {}^mB_\ell) \tag{10}$$

where ${}^eB_\ell$ and ${}^mB_\ell$ are Mie's coefficients[7] and involve the size parameter and the relative complex refractive index of a scatterer as a parameter.

Equations (6) and (7) can be readily solved analytically because these equations constitute a quadratic equation with respect to γ_2^2 (or γ_1^2). Moreover, utilizing the two limiting expressions for $S(0)$ given by van de Hulst,[8] the following two limiting expressions for γ_2 can be obtained from Eqs. (6) and (7):

When $\alpha \to \infty$,

$$\gamma_2 = 1 + (3/2)f_v - (3/4)f_v^2 \tag{11}$$

When $\alpha \to 0$,

$$\gamma_2 = \left[3(1-f_v)^4 - (1+2f_v)^2\right]/2(1+2f_v)^2/\sqrt{1-3(m^2-1)f_v/(m^2+2)} \quad \text{for} \quad \mathrm{Im}(m) = 0 \tag{12}$$

$$\gamma_2 = \sqrt{2\left[\sqrt{(1-3f_vK_i)^2 + 9f_v^2K_r^2} - (1-3f_vK_i)\right]}/3f_vK_r \quad \text{for } \mathrm{Im}(m) \neq 0 \tag{13}$$

where

$$K_i = -[(m_1^2 - m_2^2 - 1)(m_1^2 - m_2^2 + 2) + 4m_1^2m_2^2]/[(m_1^2 - m_2^2 + 2)^2 + 4m_1^2m_2^2]$$

$$K_r = 6m_1m_2/[(m_1^2 - m_2^2 + 2)^2 + 4m_1^2m_2^2] \tag{14}$$

In the case of $\alpha \to 0$ and $\mathrm{Im}(m) = 0$, Twersky[9] obtained the following result based on the Percus-Yevick distribution function and the Rayleigh theory:

$$\gamma_2 = (1 - f_v)^4/(1 + 2f_v)^2 \tag{15}$$

It is of interest to note that Eq. (12) may be reduced to Eq. (15) under the condition of $f_v \to 0$ and $m \to 1$, though the discrepancy between the two expressions becomes significant as f_v increases.

Results and Discussion

Analysis of Ishimaru-Kuga's Experiments

In order to examine the validity of the present approach, we tried to evaluate γ_2 under the conditions corresponding to Ishimaru-Kuga's experiment.[10] The results are shown in Fig. 1, which indicates that the agreement between theory and experiment is good as long as f_v is smaller than about 0.1, but, for larger f_v, relatively large discrepancies occur between them. However, the accuracy of Keller's theory is satisfactory for the present purpose because the important demarcation

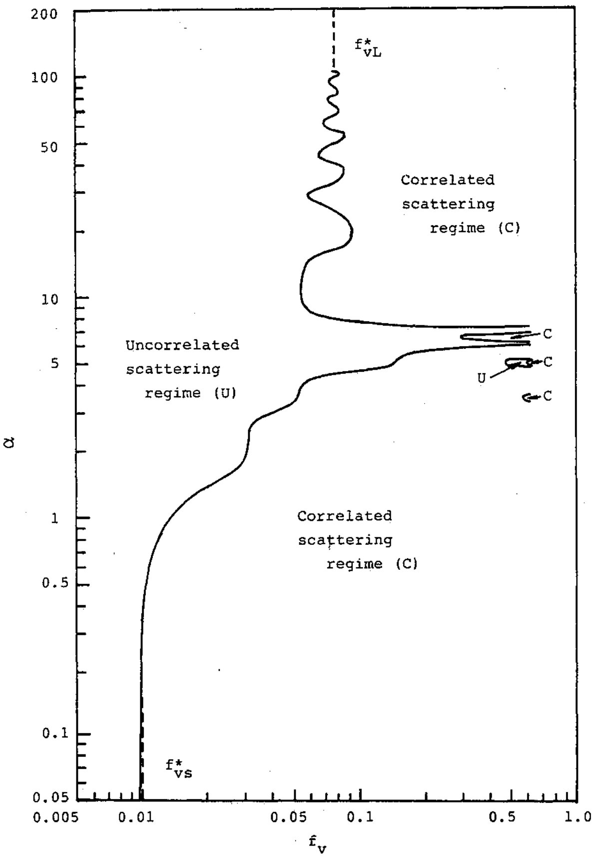

Fig. 2 The scattering regime diagram for $m = 1.192$.

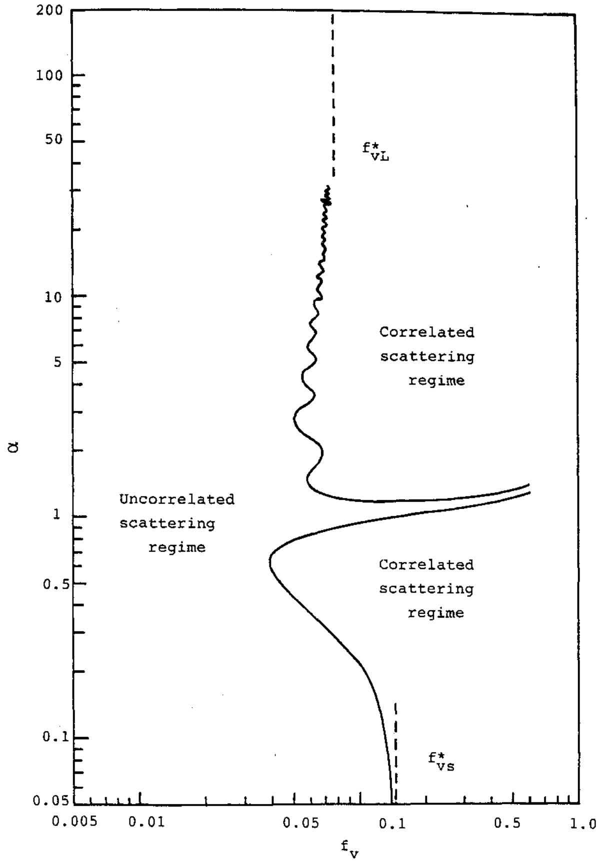

Fig. 4 The scattering regime diagram for $m = 2 + i$.

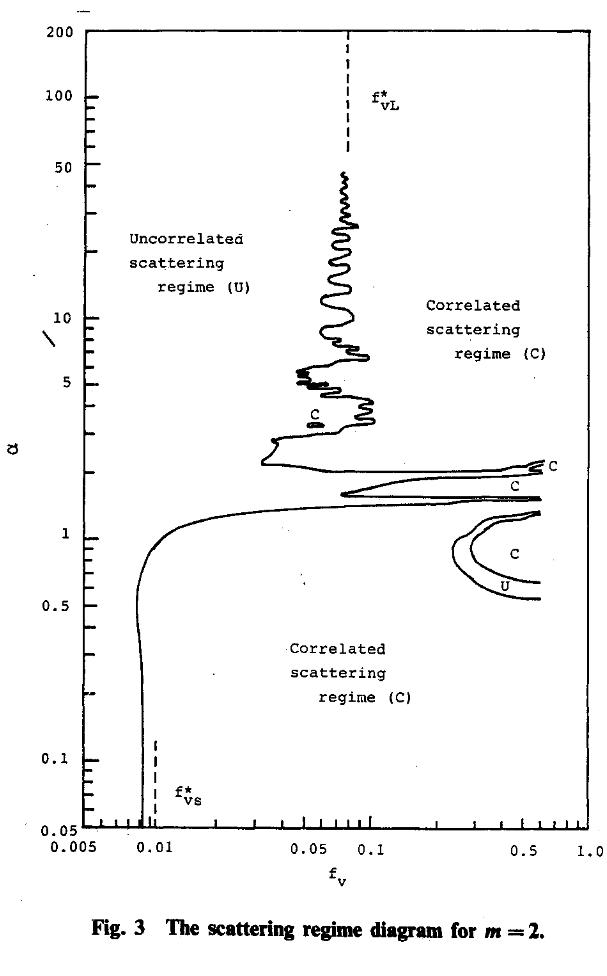

Fig. 3 The scattering regime diagram for $m = 2$.

curves on the scattering regime diagram are located within the small f_v region as will be shown later.

Evaluation of the Scattering Regime Diagrams

First, we must define the uncorrelated scattering regime quantitatively. Here, we adopted the following definition:

$$|\gamma_2 - 1| \leqq 0.1138 \tag{16}$$

This definition of the uncorrelated scattering regime is based on the Hottel et al.[1] criterion for the small size parameter region, i.e., $1 - \gamma_2 \leqq 0.1138$, and is extended here to include the larger size parameter region, where the observed extinction coefficient is greater than the extinction coefficient calculated from Mie's theory, and thus γ_2 becomes greater than unity, as shown in Fig. 1. Since γ_2 can be expressed in the analytical form, the value of γ_2 can be readily evaluated provided that the values of α, f_v, and m are given explicitly. Of these, the values of m were fixed at 1.192, 2 and $2 + i$, whereas the other parameters were widely changed. The integrals of A' and B' were evaluated within the region of η from 0 to 12.6 by utilizing an adaptive automatic integration method with a nine-point, Newton-Cotes formula. The Percus-Yevick distribution function[6,11] required was evaluated analytically for η from 0 to 5 and was calculated numerically by solving the Percus-Yevick integral equation within the region from 5 to 12.6.

The results thus obtained are shown in Figs. 2–4. As readily found, there exist three major regimes: one uncorrelated scattering regime and two correlated scattering regimes. The gross outline of the correlated scattering regime located within the relatively small size parameter region almost agrees with that previously obtained by Cartigny et al.[3]; though the details of the demarcation curve, which obviously depend on the values of m, are different from the previous study. More-

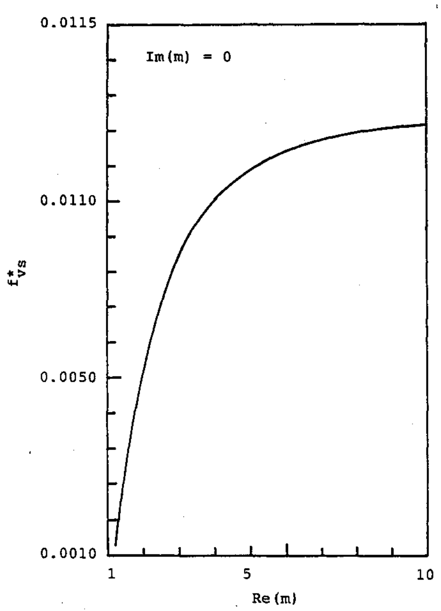

Fig. 5 The relationship between f^*_{vs} and Re(m) in the case of Im(m) = 0.

over, according to Cartigny et al.[3] and Twersky,[9] the value of the small size parameter limit f^*_{vs} on the demarcation curve should take a constant value, irrespective of m, but the present result shows that f^*_{vs} depends on the values of m. Here, it should be noted that the present approach holds true for any value of m; whereas the previous studies stand for the limiting case of m, i.e., $m \rightarrow 1$ alone. The relationship between f^*_{vs} and Re(m) for the case of Im(m) = 0 is shown in Fig. 5. In the case of Im(m) $\neq 0$, f^*_{vs} can be analytically obtained and is given by

$$f^*_{vs} = -0.09110564/K_i \tag{17}$$

As seen from Figs. 2-4, the uncorrelated scattering regime located within the intermediate size parameter region extends to a large f_v region. The existence of this extended region is also supported by the experiments of Ishimaru and Kuga.[10]

The correlated scattering regime located within the relatively large size parameter region is enclosed by two lines: one almost constant volume fraction line and one almost constant size parameter line. The vertical demarcation line in this region has already been predicted by Hottel et al.,[1] who gave $f_v = 0.27$ based on limited experimental results, but their result does not quantitatively agree with the present result. Again, the details of the demarcation curves are considerably influenced by the values of m, but the large size parameter limit f^*_{vL} on the demarcation curve never depends on m and takes a constant value 0.07899.

Conclusions

The major conclusions that can be drawn from the present study are as follows:

1) The scattering regime diagram consists of three major morphologically different regimes: one uncorrelated scattering regime and two correlated scattering regimes.

2) The details of each regime are considerably influenced by the values of the relative complex refractive index of scatterers m.

3) The value of the small size parameter limit on the demarcation curve also depends on the value of m, whereas the large size parameter limit takes a constant value 0.07899 not depending on the values of m.

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