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Optics of fractal clusters such as smoke

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Abstract. Scalar or vector light of wavelength $2\pi/k$ strikes N small refracting and absorbing spherules, each of radius a, which have coagulated into a sparse random cluster with fractal dimension D (for smoke, $D\approx 1{\cdot}78$). It is assumed that $ka \ll 1$ but the cluster size $R=aN^{1/D}$ may be larger than the wavelength. Using a mean field theory it is shown that multiple scattering is negligible for all N if $D<2$, and becomes important when $N\sim(ka)^{-D/(D-2)}$ if $D>2$. Cross-sections are calculated as functions of N, D and the complex refractive index of the spherules. If $D<2$ the scattering cross-section per spherule rises with N and saturates when $kR\gg 1$, at a value exceeding that of an isolated spherule by a factor of order $(ka)^{-D}$; if $D>2$ the same quantity increases as $N^{1-2/D}/(ka)^2$. For $D<2$ the absorption cross-section is N times that of a solitary spherule. These results are very different from those for spherules coagulating into compact solid spheres ($D=3$), and are important for the optics of sooty smoke, with $D\approx 1{\cdot}78$, which is used as an example throughout.

1. Introduction

Smoke is formed as small soot spherules [1] which stick together when they touch and thereby coagulate. Further aggregation leads to sparse random clusters with a fractal dimension of about 1·78 [2, 3]. This fractality has not been included in studies of the optics of smoke. The omission is important for the scattering and extinction of radiation by smoke lofted into the atmosphere by large fires, such as those produced by multiple nuclear explosions [4–7]. Our purpose here is to present a theory of the optics of smoke in which the importance of the fractal structure is fully brought out. Implications for the nuclear winter predictions will be discussed in a separate publication.

The central simplifying principle will be that the wavelength λ of the illuminating radiation, which is about $0{\cdot}5\,\mu$ for the visible and $10\,\mu$ in the infrared, greatly exceeds the spherule radius a, which is about $0{\cdot}005\,\mu$–$0{\cdot}05\,\mu$ for sooty smoke. In other words, the observation scale is much bigger than the inner fractal scale. However, the size R of a cluster (that is, the outer fractal scale) increases with aggregation (§ 2) as the number of spherules in a cluster increases, from a (when $N=1$) to values which may considerably exceed λ (when N is several thousand). We shall calculate the optical cross-sections as functions of N and show that the optics of clusters with $R\ll\lambda$ and $R\gg\lambda$ can be very different. Moreover the differences depend strongly on whether the fractal dimension D is less than 2 or greater than 2.

The complications of vector wave theory obscure the essential ideas and so this case will be treated last (§§ 6–8), after the scalar theory (§§ 3–5) (which applies to scattering of scalar sound waves and to neutron optics, and also as a good approximation for light).

2. Cluster statistics

Consider the ensemble consisting of all those clusters containing N spherules. The *size* R of these N-clusters is defined as the r.m.s. distance between spherules, that is, $\sqrt{2}$ times the radius of gyration. For clusters with fractal dimension D (which satisfies $1 \leqslant D \leqslant 3$ because each cluster is connected) the size is related to the spherule radius a by

$$R = aN^{1/D}. \tag{1}$$

(Strictly speaking we should write $R = \alpha a N^{1/D}$, but $\alpha = 1$ is a close approximation: for smoke ($D = 1{\cdot}78$), the simulations of Mountain and Mulholland [3] give $\alpha = 0{\cdot}96$; for a linear array ($D = 1$), $\alpha = (\frac{2}{3})^{1/2} = 0{\cdot}82$; for a random walk of touching spherules ($D = 2$), $\alpha = 2/\sqrt{3} = 1{\cdot}15$; for a close-packed 'sphere' of spherules ($D = 3$), $\alpha = 1{\cdot}11$.) Equation (1) embodies the sparseness of fractal clusters, because the number density of spherules decreases with aggregation as

$$\rho \sim \frac{N}{R^3} = \frac{1}{a^3 N^{3/D-1}}. \tag{2}$$

Fractality implies not only sparseness but also self-similarity and strong clustering. When applied to the optically important probability density $P(\mathbf{r})$ for finding a randomly chosen pair of spherules separated by a vector $\mathbf{r}_j - \mathbf{r}_l = \mathbf{r}$, self-similarity, together with isotropy, means that

$$P(\mathbf{r}) = \frac{1}{4\pi R^3} p(r/R) \tag{3}$$

where

$$\int_0^\infty \mathrm{d}x\, x^2 p(x) = 1 \quad \left(\text{normalization, and} \quad \int_0^\infty \mathrm{d}x\, x^4 p(x) = 1 \quad \text{size } R\right). \tag{4}$$

Strong clustering means that the number of particles in a sphere of radius $\delta (\ll R)$ centred on a spherule is proportional to δ^D, so that

$$p(x) \to \frac{1}{x^{3-D}} \quad \text{as} \quad x \to 0. \tag{5}$$

For detailed calculations we will take the convenient form

$$p(x) = \frac{A}{x^{3-D}} \exp(-bx) \tag{6}$$

whose constants, determined by (4), are

$$A = [(D+1)D]^{D/2}/\Gamma(D), \quad b = [(D+1)D]^{1/2}. \tag{7}$$

When $D=1{\cdot}78$, $A=4{\cdot}48$, $b=2{\cdot}22$. For comparison, the exact interspherule distance probability for a random walk ($D=2$) has

$$p(x)=\frac{8}{(2\pi)^{1/2}}\left((1+x^2)\int_1^\infty \mathrm{d}v \exp\left(-\tfrac{1}{2}x^2v^2\right)-\exp\left(-\tfrac{1}{2}x^2\right)\right). \tag{8}$$

This is derived by averaging the Gaussian probability distribution for $\mathbf{r}_j-\mathbf{r}_l$ (which has variance proportional to $|j-l|$ if j and l label positions along the walk) over the N values of j and l. It has the limiting forms

$$p(x) \begin{cases} \nearrow\ 4/x, & \text{as } x\to 0 \\ \searrow\ \dfrac{16\exp(-x^2/2)}{(2\pi)^{1/2}x^4}, & \text{as } x\to\infty. \end{cases} \tag{9}$$

We emphasize that provided $p(x)$ satisfies the conditions (4) and (5) (and also decays faster than x^{-1} as $x\to\infty$) its detailed form will not affect any of our conclusions.

Scaling breaks down at the inner fractal scale and we should take $p(x)=0$ for $x<2a/R$. However, for all our optical averages over pairs of spherules, integrals over $p(x)$ will converge with the limiting form (5). Therefore the small-x cut off would introduce a negligible correction and we henceforth ignore it.

3. Scalar waves: scattering equations

Each spherule will be considered to have the same refractive index

$$\mu=\mu_1+\mathrm{i}\mu_2. \tag{10}$$

For smoke, the transmission and absorption indices μ_1 and μ_2 depend on wavelength and on its physical and chemical nature. Representative values for soot (estimated from data of Chylek *et al.* [1]) are

$$\left.\begin{aligned} \mu&=1{\cdot}75+0{\cdot}3\mathrm{i}, && \text{visible} \\ \mu&=3+\mathrm{i}, && \text{infrared.} \end{aligned}\right\} \tag{11}$$

Scalar waves $\psi(r)$ satisfy

$$\nabla^2\psi(\mathbf{r})+k^2\mu^2(\mathbf{r})\psi(\mathbf{r})=0 \tag{12}$$

where $k\equiv 2\pi/\lambda$, $\mu(\mathbf{r})=\mu$ inside a spherule and zero outside, and ψ and $\nabla\psi$ are continuous. The incident wave

$$\psi_i=\exp(\mathrm{i}\mathbf{k}_\mathrm{i}\cdot\mathbf{r}) \tag{13}$$

interacts with the N-cluster and produces the scattered wave, whose asymptotic form is

$$\psi_\mathrm{s}=\frac{\exp(\mathrm{i}kr)}{r}f(\mathbf{k}_\mathrm{s},\mathbf{k}_\mathrm{i}) \tag{14}$$

when $\mathbf{k}_\mathrm{s}\equiv k\mathbf{r}/r$ and f is the scattering amplitude. We seek to calculate the ensemble averages of the differential cross-section

$$\mathrm{d}\sigma(\mathbf{k}_\mathrm{s},\mathbf{k}_\mathrm{i})/\mathrm{d}\Omega\equiv|f(\mathbf{k}_\mathrm{s},\mathbf{k}_\mathrm{i})|^2, \tag{15}$$

the scattering cross-section

$$\sigma_s(\mathbf{k}_i) \equiv \int\int d\Omega_s \frac{d\sigma}{d\Omega}(\mathbf{k}_s, \mathbf{k}_i) \tag{16}$$

(where Ω_s is the direction of $\mathbf{k}_s$), the extinction cross-section

$$\sigma_e(\mathbf{k}_i) = (4\pi/k)\,\mathrm{Im}\, f(\mathbf{k}_i, \mathbf{k}_i) \tag{17}$$

and the absorption cross-section

$$\sigma_a(\mathbf{k}_i) = \sigma_e(\mathbf{k}_i) - \sigma_s(\mathbf{k}_i). \tag{18}$$

The fundamental approximation is $ka \ll 1$ (small spherules); thus each spherule scatters isotropically and weakly. At this stage the Born approximation is the simplest approximation for individual scatterings but it is not adequate for our purposes because its failure to satisfy the optical theorem for real μ implies a false absorption in addition to the true absorption when μ is complex. Instead we write the scattering amplitude for a single spherule as

$$f_1 \equiv \tau/k \quad \text{with} \quad \tau \equiv \sin\eta \exp i\eta \tag{19}$$

where the s-wave phase shift η, obtained by solving (12) [8], is approximately

$$\eta \approx \tfrac{1}{3}(ka)^3(\mu^2 - 1). \tag{20}$$

For real μ, η is real and the identity $|\tau|^2 = \mathrm{Im}\,\tau$ guarantees that the optical theorem holds. For complex μ, for example smoke, the single-spherule cross-sections are

$$\sigma_s^{(1)} \approx \pi a^2 \tfrac{4}{9}(ka)^4|\mu^2 - 1|^2, \quad \sigma_a^{(1)} \approx \sigma_e^{(1)} \approx \pi a^2 \tfrac{4}{3} ka\, \mathrm{Im}\, \mu^2. \tag{21}$$

For sooty smoke all three cross-sections are much smaller than the spherule's geometrical size, and the absorption greatly exceeds the scattering.

For the cluster, with spherules centred at $\mathbf{r}_1 \ldots \mathbf{r}_N$, the total scattered wave can be written as the sum of wave from each spherule

$$\psi_s = \frac{\tau}{k} \sum_{j=1}^{N} \psi_j \frac{\exp(ik|\mathbf{r} - \mathbf{r}_j|)}{|\mathbf{r} - \mathbf{r}_j|} \rightarrow \frac{\exp(ikr)}{r} \tau \sum_{j=1}^{N} \psi_j \exp(-i\mathbf{k}_s \cdot \mathbf{r}_j) \tag{22}$$

where ψ_j is the total wave incident on the jth spherule. This in turn is the sum of the incident wave and those scattered to j from the other spherules:

$$\psi_j = \exp(i\mathbf{k}_i \cdot \mathbf{r}_j) + \tau \sum_{l \neq j} \psi_l \frac{\exp(ikr_{lj})}{kr_{lj}} \tag{23}$$

where $\mathbf{r}_{lj} \equiv \mathbf{r}_l - \mathbf{r}_j$. Now we write

$$\psi_j = c_j \exp(i\mathbf{k}_i \cdot \mathbf{r}_j) \tag{24}$$

where the coefficients c_j incorporate the effects of multiple scattering amongst the spherules. Thus, finally, we have the exact relations

$$f(\mathbf{k}_s, \mathbf{k}_i) = \frac{\tau}{k} \sum_{j=1}^{N} c_j \exp[i(\mathbf{k}_i - \mathbf{k}_s) \cdot \mathbf{r}_j] \tag{25}$$

where the c_j are determined by

$$c_j = 1 + \tau \sum_{l \neq j} c_l \frac{\exp(\mathrm{i}kr_{lj} + \mathbf{k}_\mathrm{i} \cdot \mathbf{r}_{lj})}{kr_{lj}}. \tag{26}$$

This equation is the basis of calculations by Jones [9] of the optics of small individual clusters.

4. Scalar waves: multiple scattering estimates

We seek conditions when multiple scattering can be neglected, that is when the $N-1$ terms of the summation in (26) can be neglected, giving $c_j \approx 1$. A crude preliminary bound for multiple scattering can be obtained by iteration, making the successive terms as large as possible by setting all separations r_{lj} equal to the smallest possible value, namely $2a$. This gives

$$c_j \sim (1 - N\tau/2ka)^{-1} \tag{27}$$

so that multiple scattering is certainly negligible if

$$N \ll 2/|\tau| \sim 1/(ka)^2. \tag{28}$$

In terms of the cluster size (1) this condition is

$$kR \ll 1/(ka)^{2/D-1}. \tag{29}$$

When $D<2$, therefore, clusters can be much bigger than the wavelength and still not scatter multiply. When $D>2$ this crude lower bound on N gives a cluster size much smaller than the wavelength and is not useful.

One procedure for getting better estimates might be to study the successive orders of multiple scattering obtained by iterating (26) and averaging each term separately. This is a notoriously difficult procedure because although successive scattering must be from different spherules, later rescatterings from the same spherule do occur, leading to infinite complexity in the scattering topologies which must be summed and averaged (for a survey see [10]). Moreover, the finite order multiple scattering approximations all violate the optical theorem. Another possibility is numerical inversion of the $N \times N$ matrix of coefficients in (26), but while this is feasible for small N [9] we are interested in $N \to \infty$ with ka and D variable.

Here we adopt a mean field approximation, in which all the multiple scattering coefficients c_j have the same value, c, given by averaging the $N(N-1)/2$ matrix elements in (26), so that

$$c = [1 - (N-1)\tau Q]^{-1} \tag{30}$$

where

$$Q \equiv \left\langle \frac{\exp(\mathrm{i}kr_{lj}) \cos(\mathbf{k}_\mathrm{i} \cdot \mathbf{r}_{lj})}{kr_{lj}} \right\rangle \tag{31}$$

with $\langle \quad \rangle$ denoting an average over all interparticle distances r_{lj} in the ensemble of N-clusters. Thus the wave ψ_j incident on each spherule is given by the same multiple of the incident wave. In terms of multiple scattering this is an infinite-order approximation, whose diagrammatic interpretation, for the non-fractal case of the effective refractive index of a statistically homogeneous distribution of scatterers, was discussed by Warner and Gubernatis [11].

A merit of the approximation is that it preserves the optical theorem exactly. From (25), the extinction cross-section is

$$\langle\sigma_e\rangle = \frac{4\pi N}{k^2}\operatorname{Im} c\tau = \frac{4\pi N[\operatorname{Im}\tau + |\tau|^2(N-1)\operatorname{Im} Q]}{k^2|1-(N-1)\tau Q|^2}$$
$$= \frac{4\pi N(\operatorname{Im}\tau + |\tau|^2(N-1)\langle(\sin kr_{lj}/kr_{lj})\cos(\mathbf{k}_i\cdot\mathbf{r}_{lj})\rangle)}{k^2|1-(N-1)\tau Q|^2}. \tag{32}$$

The scattering cross-section is

$$\langle\sigma_s\rangle = \frac{|\tau|^2|c|^2}{k^2}\sum_{j=1}^{N}\sum_{l=1}^{N}\left\langle\int\int d\Omega_s \exp[\mathrm{i}(\mathbf{k}_s-\mathbf{k}_i)\cdot\mathbf{r}_{lj}]\right\rangle$$
$$= \frac{4\pi N|\tau|^2(1+(N-1)\langle(\sin kr_{lj}/kr_{lj})\cos(\mathbf{k}_i\cdot\mathbf{r}_{lj})\rangle)}{k^2|1-(N-1)\tau Q|^2}. \tag{33}$$

The absorption cross-section is thus

$$\langle\sigma_a\rangle = \frac{4\pi N(\operatorname{Im}\tau - |\tau|^2)}{k^2|1-(N-1)\tau Q|^2} \tag{34}$$

which indeed vanishes, as the optical theorem demands, when the spherules are non-absorbing so that μ and hence η are real.

Within this approximation, multiple scattering can be neglected provided

$$(N-1)|\tau Q| \ll 1. \tag{35}$$

Use of (3) gives

$$Q = \frac{1}{k^2R^2}\int_0^\infty dx\, x^2 p(x)\frac{\exp(\mathrm{i}kRx)}{x}\frac{\sin kRx}{x}. \tag{36}$$

With the distribution (6) and (7) this can be integrated exactly and with (1) and $N \gg 1$ leads to

$$N\tau Q = \frac{2\mathrm{i}\tau[(D+1)D]^{D/2}}{(2ka)^D(2-D)(D-1)}\left\{\left(\frac{b^2}{4k^2R^2}+1\right)^{1-D/2}\right.$$
$$\left.\times\exp\left[-\mathrm{i}(2-D)\tan^{-1}\left(\frac{2kR}{b}\right)\right] - \left(\frac{b}{2kR}\right)^{2-D}\right\}. \tag{37}$$

When $kR \ll 1$, (19) and (20) give

$$|N\tau Q| \approx \frac{2^{1-D}|\mu^2-1|[(D+1)D]^{D/2}}{3(D-1)}(ka)^{3-D}\left(\frac{2kR}{b}\right)^{D-1} \ll 1 \tag{38}$$

confirming what is obvious, that multiple scattering is unimportant for small clusters of small spherules. The results for large clusters are less obvious; when $kR \gg 1$, (37) gives

$$\left.\begin{aligned} |N\tau Q| &\approx \frac{2^{1-D}[(D+1)D]^{D/2}|\mu^2-1|(ka)^{3-D}}{3(2-D)(D-1)}, && \text{if} \quad D<2 \\ |N\tau Q| &\approx \frac{D(D+1)|\mu^2-1|kaN^{1-2/D}}{6(D-2)(D-1)}, && \text{if} \quad D>2\ . \end{aligned}\right\} \tag{39}$$

The limit for $D<2$ is independent of N and predicts that, provided ka is small enough, multiple scattering is negligible at all stages of aggregation. The limit for $D>2$ shows that multiple scattering becomes important when $N \sim (ka)^{-D/(D-2)}$, i.e. $kR \sim (ka)^{-(3-D)/(D-2)}$. When $D=3$ this is $(\mu^2-1)kR \sim 1$ which is the correct condition for the scattering from a homogeneous refracting ball to depend on more than the single scattering from its separate parts. The dependences of $|N\tau Q|$ on ka and N in (39) survive for any $p(x)$ satisfying (4) and (5) (and decaying faster than x^{-1} as $x \to \infty$); that is, they do not depend on the particular form (6).

Intuitive arguments support the distinction between $D<2$ and $D>2$. When $D<2$ the cluster is geometrically transparent (projected area is $N\pi a^2$ rather than πR^2); when $D>2$ it is geometrically opaque. And if we pretend to assign a bulk 'refractive index' μ_c to the cluster, according to the elementary rule

$$\mu_c \sim 1+4\pi\left(\frac{N}{R^3}\right)\frac{\tau}{k^3} \sim 1+\frac{4\pi(\mu^2-1)}{N^{3/D-1}} \tag{40}$$

then the phase accumulated by a 'ray' passing through the cluster is

$$kR(\mu_c-1) \sim \tfrac{4}{3}\pi|\mu^2-1|kaN^{1-2/D}, \tag{41}$$

which vanishes as $N \to \infty$ if $D<2$ and grows if $D>2$ (but note that the 'focal length' $R/(\mu_c-1)$ diverges even if $D>2$: we do not expect fractal balls to form images for any $D<3$. Finally we observe that $D=2$ is the dimension of a Brownian trail [12] and in the quite different problem of scattering by fractal surfaces [13] those whose profiles were graphs of Brownian motion also separated different sorts of diffraction.

Nevertheless we cannot rule out the possibility that when $D<2$ the fluctuations in the matrix elements of (26), which our mean-field approximation has neglected, might conspire to cause non-negligible multiple scattering when N exceeds some large negative D-dependent power of ka.

5. Scalar waves: single scattering formulae

For single scattering from the N-cluster the amplitude is (25) with $c_j=1$. Straightforward averaging over the directions of separations between spherules gives, for the differential cross-section for scattering by θ:

$$\left\langle\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\theta)\right\rangle = N\frac{\mathrm{d}\sigma^{(1)}}{\mathrm{d}\Omega}\left(1+(N-1)\int_0^\infty \mathrm{d}x\, x^2 p(x)\frac{\sin(2kRx\sin(\theta/2))}{2kRx\sin(\theta/2)}\right) \tag{42}$$

where $\mathrm{d}\sigma^{(1)}/\mathrm{d}\Omega = |\tau|^2/k^2$ is the (isotropic) differential cross-section from a single spherule. With the fractal $p(x)$ given by (6) and (7) the cluster average can be evaluated exactly

$$\left\langle\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\theta)\right\rangle \bigg/ N\frac{\mathrm{d}\sigma^{(1)}}{\mathrm{d}\Omega} \equiv F_d(\theta) = 1 + \frac{(N-1)\sin\{(D-1)\tan^{-1}[2kR/b\sin(\theta/2)]\}}{(D-1)(1+4k^2R^2\sin^2(\theta/2)/b^2)^{(D-1)/2}2kR\sin(\theta/2)/b}. \tag{43}$$

This is the factor by which the scattering per spherule is enhanced by coherence due to the fractality. In the forward direction there is complete coherence, as for any structure, and in fact

$$F_d(\theta) \approx N \quad \text{when} \quad \theta \ll (kR)^{-1}. \tag{44}$$

The large-angle scattering, however, saturates with coagulation (increasing N) to the value

$$F_{\mathrm{d}}(\theta) \approx \frac{\sin[(D-1)\pi/2]}{(D-1)(2ka\sin(\theta/2)/b)^{D}} \quad \text{when} \quad \theta \gg (kR)^{-1}. \tag{45}$$

This saturation arises from the fractal structure and would not happen for homogeneously disordered spherules: when $D=3$ the limit (45) gives zero, whereas in reality (43) shows that the large-angle scattering decreases as $N^{-1/3}$. The main properties of $F_{\mathrm{d}}(\theta)$ are illustrated in figure 1. The asymptotic angle dependence (45) has been verified experimentally, using light and X-ray scattering, for systems with a variety of fractal dimensions [14, 15].

The scattering cross-section is the integral of (43) over directions, and the enhancement factor F_{s} for the scattering per spherule is easily calculated (from (43) or from (33) as $1+(N-1)\operatorname{Im} Q$ with Q given by 37)

$$\langle\sigma_{\mathrm{s}}\rangle/N\sigma_{\mathrm{s}}^{(1)} \equiv F_{\mathrm{s}} = 1 + \frac{2(N-1)}{(D-1)(2-D)X^{2}}\{(1+X^{2})^{1-D/2}\cos\{(2-D)\tan^{-1}X\}-1\} \tag{46}$$

where $X \equiv 2kR/b$. For small clusters this has the trivial limit

$$F_{\mathrm{s}} \approx N \quad \text{when} \quad kR \ll 1, \quad \text{i.e.} \quad N \ll (ka)^{-D}. \tag{47}$$

For large clusters the limiting behaviour depends on D:

$$\left.\begin{aligned} F_{\mathrm{s}} &\approx \frac{2\cos[(2-D)\pi/2]}{(D-1)(2-D)(2ka/b)^{D}}, && \text{if} \quad D<2; \\ F_{\mathrm{s}} &= \frac{\ln(4k^{2}a^{2}N/b)}{(2ka/b)^{2}}, && \text{if} \quad D=2; \\ F_{\mathrm{s}} &\approx \frac{2N^{1-2/D}}{(D-1)(2-D)(2ka/b)^{2}}, && \text{if} \quad D>2; \end{aligned}\right\} \tag{48}$$

when $kR \gg 1$, i.e. $N \gg (ka)^{-D}$.

The full curves in figure 2 illustrate the N- and D-dependence of F_{s}. Just as for the multiple scattering estimates of §4, what matters is whether $D<2$ or $D>2$. If $D<2$ the scattering per particle is enhanced by a factor which saturates to a large $(\sim(ka)^{-D})$ value that is independent of N. If $D>2$, coagulation continues to increase the enhancement factor, until the single-scattering approximation breaks down when $N \sim (ka)^{-D/(D-2)}$ (cf. (39)). $D=2$ shows the logarithmic behaviour typical of crossover dimensionalities.

The absorption cross-section is, from (34) (with $Q=0$) and (21), simply the sum of the absorptions of all the spherules, namely

$$\langle\sigma_{\mathrm{a}}\rangle = N\sigma_{\mathrm{a}}^{(1)} = (4\pi N/k^{2})\,(\operatorname{Im}\tau-|\tau|^{2})N\pi a^{2}\tfrac{4}{3}ka\operatorname{Im}\mu^{2}. \tag{49}$$

If the real and imaginary refractive indices are of comparable magnitudes, absorption greatly exceeds scattering, even for large clusters, by a factor which from (48), (46) and (21) is of order $1/(ka)^{3-D}$ if $D<2$ and $1/(N^{1-2/D}ka)$ if $D>2$ (which is less than unity only when N exceeds the single-scattering limit after (39)). For smoke the extinction by fractal clusters of non-transparent spherules is therefore dominated by absorption.

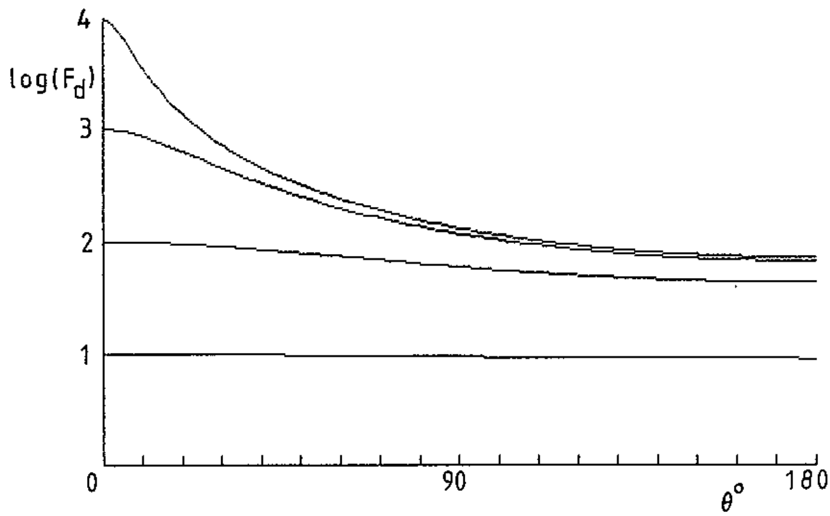

Figure 1. Coherent enhancement factor $F_d(\theta)$ for the differential cross-section per spherule (equation (43)), for $D=1\cdot78$, $ka=0\cdot111$ and $N=10, 100, 1000, 10000$, showing isotropic scattering for small N (cluster smaller than wavelength), and concentrated forward peak and saturation of large-angle scattering for large N. Logarithms are to base 10.

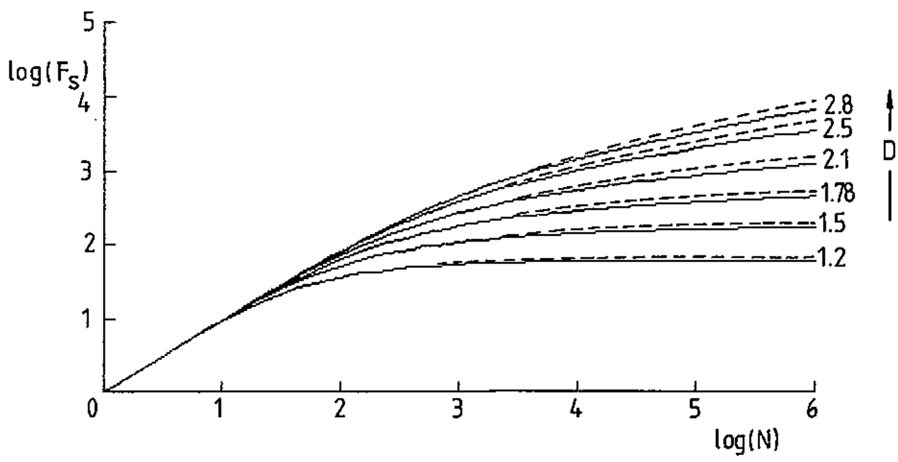

Figure 2. Coherent enhancement factors F_s for the scattering cross-section per spherule for scalar waves (full curves, from (46) and (1) and vector waves (broken curves, from (74) and (1)), for $ka=0\cdot111$, showing saturation with increasing N when $D<2$, and continuing increase when $D>2$. Logarithms are to base 10.

It is worth comparing the scattering and absorption for large ($kR \gg 1$) fractal clusters with $D<2$ (for example, smoke) with the corresponding predictions that would follow from the false assumption that coagulation produces large absorbing solid spheres ($D=3$). On this assumption the extinction cross-section of an N-cluster is given by geometrical optics as

$$\sigma_e^{\text{large solid}} = \pi R_{\text{solid}}^2 = \pi a^2 N^{2/3}. \tag{50}$$

The fractal extinction cross-section, given by (49), exceeds this by a factor

$$\langle\sigma_a\rangle/\sigma_e^{\text{large solid}}=\tfrac{4}{3}\,\mathrm{Im}\,\mu^2N^{1/3}ka=\tfrac{4}{3}\,\mathrm{Im}\,\mu^2kR_{\text{solid}}. \tag{51}$$

This exceeds unity for large solid spheres, so that the solid-coagulation assumption underestimates the extinction. A similar estimate for scattering shows that this is overestimated, except for *very* large clusters ($N>(ka)^{-3(4-D)}$) when fractals scatter more.

6. Vector waves: scattering equations

In this and the next two sections we follow the scalar-wave theory as closely as possible. The electronmagnetic fields satisfy not (12) but Maxwell's equations, and we will work entirely with the electric field $\mathbf{E(r)}$. Instead of (13) the incident field, with polarization unit vector $\mathbf{e}_i(\perp\mathbf{k}_i)$, is

$$\mathbf{E}_i=\mathbf{e}_i\exp(\mathrm{i}\mathbf{k}_i\cdot\mathbf{r}) \tag{52}$$

The cluster produces from this a scattered wave whose asymptotic form, involving a vector scattering amplitude, is

$$\mathbf{E}_s=\frac{\exp(\mathrm{i}kr)}{r}\,\mathbf{f}(\mathbf{k}_s,\mathbf{k}_i). \tag{52 a}$$

The scattering and absorption cross-sections are given by (16) and (18), the differential and extinction cross-sections being [16]

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}(\mathbf{k}_s,\mathbf{k}_i)=\mathbf{f}(\mathbf{k}_s,\mathbf{k}_i)\cdot\mathbf{f}^*(\mathbf{k}_s,\mathbf{k}_i) \tag{53}$$

and

$$\sigma_e=(4\pi/k)\,\mathrm{Im}\,\mathbf{f}(\mathbf{k}_i,\mathbf{k}_i)\cdot\mathbf{e}_i. \tag{54}$$

The fundamental approximation $ka\ll 1$ implies that each spherule scatters weak dipole radiation. At this stage we do not use the Rayleigh (i.e. Born) approximation because this fails to satisfy the optical theorem for real index μ. Instead we use a unitarity-preserving weak-scattering approximation to the exact Mie theory [17], in which a single spherule at the origin produces the scattered wave

$$\mathbf{E}_s=\frac{3\rho\exp(\mathrm{i}kr)}{2kr}\left[(\mathbf{n}\wedge\mathbf{e}_i)\wedge\mathbf{n}+[3\mathbf{n}(\mathbf{n}\cdot\mathbf{e}_i)-\mathbf{e}_i]\left(\frac{1}{(kr)^2}-\frac{\mathrm{i}}{kr}\right)\right] \tag{55}$$

where $\mathbf{n}\equiv\mathbf{r}/r$ and

$$\rho\equiv\sin\delta\exp(\mathrm{i}\delta) \tag{56}$$

in which δ is a p-wave phase shift given approximately, by solving Maxwell's equations, as

$$\delta\approx\tfrac{2}{3}(ka)^3\left(\frac{\mu^2-1}{\mu^2+2}\right). \tag{57}$$

For the single-spherule scattering amplitude the first (radiative) term in (55) gives

$$f_1(\mathbf{k}_s,\mathbf{k}_i)=\frac{3\rho}{2k^3}(\mathbf{k}_s\wedge\mathbf{e}_i)\wedge\mathbf{k}_s=\frac{3\rho}{2k}\left(\mathbf{e}_i-\frac{(\mathbf{k}_s\cdot\mathbf{e}_i)\mathbf{k}_s}{k^2}\right). \tag{58}$$

Instead of (21), the three cross-sections for complex μ are found using (16) and (54) to be

$$\frac{\mathrm{d}\sigma^{(1)}}{\mathrm{d}\Omega}(\mathbf{k}_\mathrm{i}, \mathbf{k}_\mathrm{s}) \approx k^4 a^6 \left|\frac{\mu^2-1}{\mu^2+2}\right|^2 [1-(\mathbf{k}_\mathrm{s}\cdot\mathbf{e}_\mathrm{i})^2/k^2],$$

$$\sigma_\mathrm{s}^{(1)} \approx \pi a^2 \tfrac{8}{3}(ka)^4 \left|\frac{\mu^2-1}{\mu^2+2}\right|^2, \quad \sigma_\mathrm{a}^{(1)} \approx \sigma_\mathrm{e}^{(1)} \approx \pi a^2 4ka \,\mathrm{Im}\left(\frac{\mu^2-1}{\mu^2+2}\right). \tag{59}$$

For unpolarized incident light, the directional factor in $\mathrm{d}\sigma^{(1)}/\mathrm{d}\Omega$ becomes $\frac{1}{2}(1+\cos\theta)$.

For the cluster the asymptotic scattered wave, analogous to (22), is, from (55),

$$\mathbf{E}_\mathrm{s} = \frac{3\rho}{2k^3 r}\sum_{j=1}^{N} (\mathbf{k}_\mathrm{s}\wedge\mathbf{E}_j)\wedge\mathbf{k}_\mathrm{s}\exp(-\mathrm{i}\mathbf{k}_\mathrm{s}\cdot\mathbf{r}_j) \tag{60}$$

where $\mathbf{E}_j$ is the total field incident on the jth spherule. This in turn is the sum of the incident field and those scattered to j from the other spherules:

$$\mathbf{E}_j = \mathbf{e}_\mathrm{i}\exp(\mathrm{i}\mathbf{k}_\mathrm{i}\cdot\mathbf{r}_j) + \frac{3\rho}{2}\sum_{l\neq j}\Bigg[(\mathbf{n}_{jl}\wedge\mathbf{E}_l)\wedge\mathbf{n}_{jl} + [3\mathbf{n}_{jl}(\mathbf{n}_{jl}\cdot\mathbf{E}_l)-\mathbf{E}_l] \times\left(\frac{1}{(kr_{jl})^2}-\frac{\mathrm{i}}{kr_{jl}}\right)\Bigg]\frac{\exp(\mathrm{i}kr_{jl})}{kr_{jl}}. \tag{61}$$

Now we introduce multiple scattering coefficients $\mathbf{d}_j$ (cf. (24)) by

$$\mathbf{E}_j \equiv \mathbf{d}_j \exp(\mathrm{i}\mathbf{k}_\mathrm{i}\cdot\mathbf{r}_j). \tag{62}$$

This gives finally the vector-wave equations, exact for small ka,

$$\mathbf{f}(\mathbf{k}_\mathrm{s}, \mathbf{k}_\mathrm{i}) = \frac{3\rho}{2k^3}\sum_{j=1}^{N}(\mathbf{k}_\mathrm{s}\wedge\mathbf{d}_j)\wedge\mathbf{k}_\mathrm{s}\exp[\mathrm{i}(\mathbf{k}_\mathrm{i}-\mathbf{k}_\mathrm{s})\cdot\mathbf{r}_j] \tag{63}$$

with $\mathbf{d}_j$ determined by

$$\mathbf{d}_j = \mathbf{e}_\mathrm{i} + \frac{3\rho}{2}\sum_{l\neq j}\left[(\mathbf{n}_{jl}\wedge\mathbf{d}_l)\wedge\mathbf{n}_{jl} + [3\mathbf{n}_{jl}(\mathbf{n}_{jl}\cdot\mathbf{d}_l)-\mathbf{d}_l]\left(\frac{1}{(kr_{jl})^2}-\frac{\mathrm{i}}{kr_{jl}}\right)\right]\exp(\mathrm{i}kr_{jl}-\mathrm{i}\mathbf{k}_\mathrm{i}\cdot\mathbf{r}_{jl}). \tag{64}$$

7. Vector waves: multiple scattering estimates

By analogy with the scalar case, we estimate the effects of multiple scattering by making the mean-field approximation that all multiple scattering coefficients are the same, and parallel to the incident polarization, i.e.

$$\mathbf{d}_j \approx d\mathbf{e}_j. \tag{65}$$

This amounts to replacing by their average values the matrix elements in (64), whose component along $\mathbf{e}_\mathrm{i}$ gives (cf. (30) and (31))

$$d = [1-\tfrac{3}{2}(N-1)\rho P]^{-1} \tag{66}$$

where

$$P \equiv \left\langle \frac{\exp(\mathrm{i}kr_{jl})}{kr_{jl}}\cos\mathbf{k}_\mathrm{i}\cdot\mathbf{r}_{jl}\left[1-(\mathbf{n}\cdot\mathbf{e}_\mathrm{i})^2 + [3(\mathbf{n}\cdot\mathbf{e}_\mathrm{i})^2-1]\left(\frac{1}{(kr_{jl})^2}-\frac{\mathrm{i}}{(kr_{jl})^2}\right)\right]\right\rangle \tag{67}$$

with $\mathbf{n} \equiv \mathbf{r}_{jl}/r_{jl}$.

Again this approximation preserves the optical theorem exactly. This is more surprising than in the scalar case, because P in (67) involves the near-zone (non-radiative) terms in the dipole field, and the scattering involves only the far field. To show it, we first write the extinction cross-section using (63) as

$$\langle\sigma_e\rangle=\frac{6\pi N}{k^2}\operatorname{Im}\rho d=\frac{6\pi N[\operatorname{Im}\rho+\frac{3}{2}|\rho|^2(N-1)\operatorname{Im}P]}{k^2|1-\frac{3}{2}(N-1)\rho P|^2}. \tag{68}$$

The scattering cross-section is

$$\langle\sigma_s\rangle=\frac{9|\rho|^2|d|^2}{4k^2}\sum_{j=1}^{N}\sum_{l=1}^{N}\left\langle\iint d\Omega_s \exp[i(\mathbf{k}_i-\mathbf{k}_s)\cdot\mathbf{r}_{lj}]\left[1-\left(\frac{\mathbf{k}_s\cdot\mathbf{e}_i}{k}\right)^2\right]\right\rangle$$

$$=\frac{6\pi N|\rho|^2\left(1+[3(N-1)/8\pi]\left\langle\iint d\Omega_s\exp(-i\mathbf{k}_s\cdot\mathbf{r}_{jl})\{1-[\mathbf{k}_s\cdot\mathbf{e}_i/k]^2\}\cos(\mathbf{k}_i\cdot\mathbf{r}_{jl})\right\rangle\right)}{k^2|1-\frac{3}{2}(N-1)\rho P|^2}. \tag{69}$$

An elementary calculation shows that the angular integral is precisely $4\pi\operatorname{Im}P$, so the absorption cross-section is

$$\langle\sigma_a\rangle=\frac{6\pi N(\operatorname{Im}\rho-|\rho|^2)}{k^2|1-\frac{3}{2}(N-1)\rho P|^2} \tag{70}$$

which indeed vanishes for non-absorbing spherules.

Within this approximation, multiple scattering can be neglected provided

$$\tfrac{3}{2}(N-1)|\rho P|\ll 1. \tag{71}$$

Use of (9) gives

$$P=\frac{1}{k^2R^2}\int_0^\infty dx\, x^2p(x)\frac{\exp(iX)}{x^2}\left\{\left[1-\frac{2}{X^2}+\frac{3}{X^4}+\frac{i}{X}\left(1-\frac{3}{X^2}\right)\right]\right.$$
$$\left.\times\sin X+\frac{1}{X}\left(1-\frac{3}{X^2}+\frac{3i}{X}\right)\cos X\right\} \tag{72}$$

where $X\equiv kRx$. Note that the isotropy of the cluster statistics has eliminated the dependence on the initial polarization $\mathbf{e}_i$. P is similar to the scalar-wave quantity Q (equation (36)) with the factor $\{\ \}$ replacing $\sin X$. Now, as $X\to 0$ the factor $\{\ \}$ vanishes like X, just as $\sin X$ does. Therefore the integral in (72) converges for fractal cluster distributions $p(x)$, and this implies that the vector-wave condition (71) depends on N, D and ka in the same way as the scalar-wave condition (35), apart from numerical factors (in the next section we shall see that these numerical factors are closely similar). The discussion based on (37)–(39) can therefore be taken over to the vector case, the main conclusion being that as for scalar waves multiple scattering can always be neglected when $D<2$ (provided of course that $ka\ll 1$).

9. Vector waves: single scattering formulae

For single scattering from the N-cluster the amplitude is (63) with $\mathbf{d}_j=\mathbf{e}_i$. Straightforward cluster averaging shows that the differential cross-section is given exactly by the scalar-wave formula (42) (the single-spherule differential cross-section being (59)), and the same discussion applies, based on (43)–(45) and figure 1.

The scattering cross-section is (69) with the modulus factor unity. The angular integral is $2\pi\,\mathrm{Im}\,P$ and (72) and the distribution (6) and (7) lead to

$$\langle\sigma_s\rangle/N\sigma_s^{(1)}\equiv F_s \tag{73}$$

where the enhancement factor F_s for the scattering per spherule is now

$$F_s=1+\frac{3(N-1)}{(D-1)X^2}[\![(1+X^2)^{1-D/2}\{\cos[(2-D)\tan^{-1}X]\}(T_0+T_2/X^2+T_4/X^4)$$
$$+\sin[(2-D)\tan^{-1}X](T_1/X+T_3/X^3)-(U_0+U_2/X^2+U_4/X^4)]\!] \tag{74}$$

in which $X\equiv 2kR/b$ and

$$\left.\begin{aligned}
&T_0\equiv\frac{1}{2-D}-\frac{2}{4-D}+\frac{2}{6-D}, && T_1\equiv\frac{4}{3-D}-\frac{4}{4-D}-\frac{8}{5-D}+\frac{8}{6-D},\\
&T_2\equiv\frac{2}{2-D}-\frac{4}{3-D}-\frac{10}{4-D}+\frac{24}{5-D}-\frac{12}{6-D}, && T_3\equiv\frac{8}{3-D}-\frac{24}{4-D}+\frac{24}{5-D}-\frac{8}{6-D},\\
&T_4\equiv\frac{2}{2-D}-\frac{8}{3-D}+\frac{12}{4-D}-\frac{8}{5-D}+\frac{2}{6-D}, && u_0\equiv\frac{1}{2-D},\\
&U_2\equiv\frac{2}{2-D}-\frac{4}{3-D}+\frac{2}{4-D}, && U_4\equiv\frac{2}{2-D}-\frac{8}{3-D}+\frac{12}{4-D}-\frac{8}{5-D}+\frac{2}{6-D}.
\end{aligned}\right\} \tag{75}$$

This is the vector equivalent of (46), and it has similar limiting behaviour. For small clusters, much cancellation leads to

$$F_s\approx N \quad \text{when} \quad kR\ll 1, \quad \text{i.e.} \quad N\ll(ka)^{-D}. \tag{76}$$

For large clusters, the limits depend on D:

$$\left.\begin{aligned}
&F_s\approx\frac{3\cos[(2-D)\pi/2]}{(D-1)(2-D)(2ka/b)^D}\left(1-\frac{4(2-D)}{(4-D)(6-D)}\right), && \text{if} \quad D<2,\\
&F_s\approx\frac{3\ln(4k^2a^2N/b)}{2(2ka/b)^2}, && \text{if} \quad D=2,\\
&F_s\approx\frac{3N^{1-2/D}}{(D-1)(D-2)(2ka/b)^2}, && \text{if} \quad D>2,
\end{aligned}\right\} \tag{77}$$

when $kR\gg 1$, i.e. $N\gg(ka)^{-D}$.

The dashed curves in figure 2 illustrate the N- and D-dependence of F_s. Evidently the vector and scalar scattering cross-sections are quantitatively as well as qualitatively similar, and the same remarks (following equation (48)) apply. The absorption cross-section is, likewise, simply the sum of the absorptions of all the spherules, namely

$$\langle\sigma_a\rangle=N\sigma_a^{(1)}=\frac{6\pi N}{k^2}(\mathrm{Im}\,\rho-|\rho|^2)=N\pi a^2 4ka\,\mathrm{Im}\left(\frac{\mu^2-1}{\mu^2+2}\right). \tag{78}$$

This is similar to (49) and the discussion from that equation to the end of § 5 applies here too.

10. Conclusions

Our theory shows that for both scalar and vector waves the scattering cross-section of a D-dimensional fractal cluster of N small spherules is different for $D>2$ and $D<2$. For $D>2$ the scattering per spherule for large clusters grows with aggregation as $N^{1-2/D}$. For $D<2$ (smoke) the surprising result is that the scattering per spherule increases to a constant saturation value which exceeds that of a solitary spherule by a factor of order $(ka)^{-(3-D)}$. The absorption cross-section is independent of D and simply the sum of absorptions of the separate spherules.

Any real smoke is an assembly of clusters of different sizes, with N as a random variable. If the smoke has n spherules per unit volume and has coagulated into a sparse distribution of n/N clusters with, on the average, N spherules each, then the absorption or scattering length (for exponential attenuation) of an incident wave, is

$$L=[(n/N)\sigma]^{-1} \tag{79}$$

where σ is the appropriate cross-section. Our results show that for absorption L is always independent of N, that is, of coagulation, and for scattering from smoke ($D<2$) L decreases with N to a constant value, unaffected by further coagulation. These predictions could be tested by experiments in which the absorption and scattering were measured for smoke coagulating under controlled conditions.

In obtaining these results, multiple scattering has been neglected. This was justified (for all N when $D<2$ and for $N \lesssim (ka)^{-D/(D-2)}$ when $D>2$) by estimating the multiple scattering using a mean field theory. Obviously this is an approximation and it would be very interesting to have an exact theory of the large N limit for fractal clusters, based (probably) on a renormalization analysis of the field fluctuations within a cluster.

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