Physica A 191 (1992) 352–357
North-Holland

PHYSICA A

Scattering and localization of light on fractals

Vladimir M. Shalaev[a,b,1], Martin Moskovits[a,b],
Andrey A. Golubentsev[b,c] and Sajeev John[b,c]

[a]*Department of Chemistry, 80 St. George Street, University of Toronto, Toronto, Ontario M5S 1A1, Canada*
[b]*The Ontario Laser and Lightwave Research Centre, Toronto, Ontario M5S 1A1, Canada*
[c]*Physics Department, 60 St. George Street, University of Toronto, Toronto, Ontario M5S 1A1, Canada*

Resonant Rayleigh scattering by fractal clusters is shown to be greatly enhanced. Localization of light occurs when the spatial scale of coherent multiple scattering is reduced to the wavelength. Anomalies in light absorption by fractals arise because of critical slowing down of the photons near a mobility edge.

The character of the propagation of waves of any kind and, in particular, of light in media with random refractive-index inhomogeneities is determined by the ratio of the wavelength λ to the radiation elastic mean-free path l. Light localization is an effect that arises entirely from coherent multiple scattering and interference when l reduces to λ [1,2]. This effect is similar to Anderson localization of electrons in disordered solids [3]. The interference of optical waves in multiple scattering manifests itself also in coherent backward scattering, which is a factor two more intense than the incoherent background [4,5]. Achieving the condition $l \sim \lambda$ needed for observation of localized light modes is experimentally rather difficult since usually $l \gg \lambda$.

Fractals clusters are thought to be promising media for the light localization. It was shown [6] that aggregation of initially isolated particles into fractal clusters leads to great enhancement of the scattering cross-section σ_s. The scattering cross-section per particle is enhanced by coherence due to fractality [6,7] (in comparison with that of an isolated particle). If the particles forming the cluster possess optical resonances with a high quality factor, an additional enhancement appears due to the resonance character of light scattering by collective eigenmodes of the fractal clusters [6].

A cluster is considered as a fractal set of N monomers: $N = (R_c/R_0)^D$ (D is the fractal dimension), with dipolar interaction between them determined by

[1] Phone: (416)978-5596; Fax: (416)978-8775; E-mail: vshalaev@alchemy.chem.utoronto.ca

$$L_X \sim R_0 (R_0^3 |X|)^{(d_0-1)/(3-D)} , \tag{4}$$

where d_0 is an index ($0 < d_0 < 1$) called the optical spectral dimension. The scaling condition $R_0 \ll L_x \ll R_c$ is fulfilled if

$$R_0^3 \delta, N^{-(3/D-1)/(1-d_0)} \ll R_0^3 |X| \ll 1 . \tag{5}$$

An additional scaling condition $L_X < \lambda$ is automatically satisfied by condition (1). Note that requirement (1) is compatible with (5) if $(R_0/\lambda)^{3-D} \ll 1$ for $D < 2$ (that is always valid when $R_0 \ll \lambda$) and if $N < (\lambda/R_0)^{D/(D-2)}$ for $D > 2$. For example, when $\lambda/R_0 \approx 30$, $D = 2.5$, the latter requirement restricts the number of particles in a cluster to $N < 10^7$. When these conditions are fulfilled multiple scattering is small for one cluster (but not for a whole sample consisting of many such fractal clusters).

The absorption σ_a and the scattering σ_s cross sections for a fractal cluster have the following scaling forms [6]:

$$\sigma_a \sim N \lambda^{-1} R_0^3 (R_0^3 |X|)^{d_0 - 1} , \tag{6}$$

$$\sigma_s = F_s N \sigma_s^{(0)} , \qquad F_s = f_s Q (R_0^3 |X|)^{d_0+1} , \tag{7}$$

where $Q \equiv |R_0^3 \operatorname{Im} \chi_0^{-1}|^{-1} = (R_0^3 \delta)^{-1}$ is the quality factor of the resonance ($Q \gg 1$), $\sigma_s^{(0)} = (8\pi/3) k^4 |\chi_0|^2$ is the scattering cross section of a solitary particle (k is the wave vector) and the coherent fractality factors f_s depend on D as follows:

$$f_s \sim \begin{cases} (\lambda/R_0)^D , & D < 2 , \quad (8) \\ (\lambda/R_0)^2 \ln[4\pi^2 N (R_0/\lambda)^2] , & D = 2 , \quad (9) \\ (\lambda/R_0)^2 N^{1-2/D} , & D > 2 . \quad (10) \end{cases}$$

According to (7) the scattering per particle is enhanced by coherence due to fractality factor f_s and as a result of the resonant character of the light scattering by the dipolar eigenmodes of the cluster ($\sigma_s \propto Q$).

Under condition (5) $|\chi_0^{-1}|^2 \approx |X|^2$ and one can rewrite (7) in the form

$$\sigma_s \sim N f_s (R_0^6/\lambda^4) Q (R_0^3 |X|)^{d_0 - 1} . \tag{11}$$

If the laser frequency is far from all eigenfrequencies of fractal modes or if the quality factor of the resonances is low (the limit of nonresonant scattering:

$\chi_i \approx \chi_0$) the quantity $Q(R_0^3|X|)^{d_0-1}$ in (11) will be approximately 1. This reduces to the result obtained earlier by Berry and Percival [7]: $\sigma_s \sim (R_0^6/\lambda^4) f_s N$.

It follows from (6), (11) that absorption is small in comparison with light scattering if $f_s(R_0/\lambda)^3 Q > 1$. For $D < 2$ this implies that $Q > (\lambda/R_0)^{3-D}$. Assuming $\lambda/R_0 \sim 10$, and $D \approx 1.78$ as for cluster–cluster aggregates, then $Q > 16$. Such a condition is fulfilled, for example, for silver and gold colloids where Q is of the order of 10 to 100. For $D > 2$ the corresponding condition $N^{1-2/D} Q > \lambda/R_0$ can be easily fulfilled even for very small clusters.

Let us consider now a trivial mixture of fractal clusters with intercluster distance of the order of the cluster size R_c (as in a gel). The elastic mean-free path in this case is $l \sim (\sigma_s n_c)^{-1} \sim R_c^3/\sigma_s$, where n_c is the concentration of clusters. Using (11) gives

$$l/\lambda \sim N^{3/D-1} (\lambda/R_0)^3 f_s^{-1} Q^{-1} (R_0^3|X|)^{1-d_0} . \tag{12}$$

Note that in general Q can depend on λ (as, for example, in the case of metal colloids when Q increases with increasing λ [6]). Let us, for the sake of simplicity, consider the case when such a dependence can be neglected (for example, for clusters consisting of particles with isolated resonance). It follows from (12) that l decreases with decreasing $|X|$ or, in accordance with (4), with increasing correlation length, L_X. The minimum value of l is reached for the highest possible (at least in the frame of our scaling approach) value of $L_X \sim \lambda$. That the optimum condition for localization is achieved when the correlation length is of the order of the wavelength, agrees with the conclusion reached in ref. [1]. Taking into account that $L_X \sim \lambda$ at $(R_0^3|X|)^{1-d_0} \sim (\lambda/R_0)^{D-3}$ (see (4)), one obtains from (12) and (8)–(10)

$$l/\lambda \sim \begin{cases} N^{3/D-1}/Q, & D < 2, \quad (13) \\ [\ln(Nk^2R_0^2)]^{-1} N^{1/2}/Q, & D = 2, \quad (14) \\ (\lambda/R_0)^{D-2} N^{5/D-2}/Q, & D > 2. \quad (15) \end{cases}$$

Let us now estimate the cluster sizes capable of fulfilling these conditions. For the cluster–cluster aggregation ($D \approx 1.78$) and $Q \sim 10^2$ the requirement $l \sim \lambda$ can be only reached for a sample consisting of rather small clusters $N \sim 10^3$ (that is, for a rather high concentration of clusters in the sample). However, for the media formed by clusters grown under diffusion limited aggregation (DLA; $D \approx 2.5$) the condition of the light localization $l \sim \lambda$ can be easily fulfilled (for example, at $\lambda/R_0 \sim 10$, $Q \sim 10$) for any size of clusters. Accordingly a most promising medium for observing light localization is a gel of metal fractal clusters grown under conditions of DLA. It is worth noting the nontrivial dependence of l on D: l increases with N for $D<2.5$, as $N^{3/D-1}$ if $D<2$, and as $N^{5/D-2}$ if $2<D<2.5$, while for $D=2$ one has a logarithmic behaviour typical of crossover dimensionalities. For $D>2.5$, l decreases with increasing N as $N^{5/D-2}$. Note also that for the parameter values used above absorption is weak as compared with scattering. Such a situation is made possible by the resonant enhancement of the scattering ($\sigma_s \propto Q$) which is absent for absorption (compare (6) and (7)). The physical reason for such a difference is related to the fact that scattering (in contrast to the absorption) is proportional to the second power of a local field, which results (after averaging over ensemble of fractals) in the proportionality of σ_s to Q [6]. For "nonresonant fractals" having comparable magnitudes of the real and imaginary refractive indices absorption usually exceeds scattering.

Let us assume that the laser frequency is close to a mobility edge. In this case one can determine the effective diffusion coefficient D_s, which takes into account the interference corrections to the classical diffusion constant [1]: $D_s \sim cl^2/a$, where $a \equiv \min\{\xi, L, l_{abs}\}$ and c is the light velocity. The quantity ξ is a coherence length that represents a scale on which the interference is significant and modifies the classical diffusion process, L is a sample length and l_{abs} is a characteristic length of the light absorption. Let us consider first the case of weak absorption $l_{abs}>L$. For the case of classical diffusion, the transmission coefficient T, defined as the ratio of the transmitted to the incident intensity, is equal to l/L, where l is the classical mean-free path. Using the equivalent form $T \sim D_s/cL$ with the effective rather than classical diffusion coefficient ($D_{clas} \sim cl$), one finds [1] $T \sim l^2/\xi L$ for $l \ll \xi \ll L$ and $T \sim l^2/L^2$ for $l \ll L \ll \xi$ with l given by (12)–(15). Such scale dependent light diffusion can be used as an experimental paradigm for the observation of light localization.

Now consider how the light diffusion in a fractal gel modifies the absorption near the mobility edge. The imaginary part of the medium effective susceptibility is given by $\kappa'' \sim n_c N R_0^3 (R_0^3|X|)^{d_0-1} \sim q(R_0^3|X|)^{d_0-1}$, where q is the volume filling fraction of the scatters (for $n_c \sim R_c^{-3} q \sim N^{1-3/D}$). Substituting the characteristic time of inelastic scattering $\tau_{abs} \sim (\omega\kappa'')^{-1}$ into the expression $\alpha \sim l_{abs}^{-1} \sim (D_s \tau_{abs})^{-1/2} \sim (\omega\kappa''/D_s)^{1/2}$, one obtains

$$\alpha \sim (q\omega/D_s)^{1/2}(R_0^3|X|)^{(d_0-1)/2} . \tag{16}$$

At $L_X \sim \lambda$ and $n_c \sim R_c^{-3}$ this gives

$$\alpha \sim N^{(D-3)/2D}(\omega/D_s)^{1/2}(\lambda/R_0)^{(3-D)/2} . \tag{17}$$

If l_{abs} acts as a long-distance cutoff for coherent wave interference ($l < l_{abs} < \xi$, L; $D_s \propto l_{abs}^{-1} \sim \alpha$), then one finds that $\alpha \propto (\kappa'')^{1/3}$ [1] or, using the expression for κ'',

$$\alpha \propto q^{1/3}(R_0^3|X|)^{(d_0-1)/3} . \tag{18}$$

These anomalies in light absorption by fractals arise because of the critical slowing down of the photons near the mobility edge. They, too, can be used as a means for observing light localization experimentally.

References

[1] S. John, Physics Today (1991) 32.
[2] S. John, Phys. Rev. Lett. 53 (1984) 2169.
[3] P.W. Anderson, Phys. Rev. 109 (1958) 1492.
[4] A.A. Golubentsev, Sov. Phys. JETP 59 (1984) 26.
[5] Y. Kuga and A. Ishimaru, J. Opt. Soc. Am. A 1 (1984) 831.
[6] V.M. Shalaev, R. Botet and R. Jullien, Phys. Rev. B 44 (1991) 12216.
[7] M.V. Berry and I.C. Percival, Optica Acta 33 (1986) 577.
[8] V.A. Markel, L.S. Muratov, M.I. Stockman and T. F. George, Phys. Rev. B 43 (1991) 8183.
[9] M.I. Stockman, T.F. George and V.M. Shalaev, Phys. Rev. B 44 (1991) 115.