Visibility–invisibility phase transition in a fractal cluster

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The fractal dimension $D=3/2$ is critical. At this value of D, the optical properties of a fractal cluster undergo abrupt changes. At $D>3/2$, the monomers of a cluster scatter light incoherently. At $D<3/2$, there is a huge increase in the scattering due to long-range correlations in the arrangement of particles of the fractal cluster.

A fractal cluster is an agglomerate of tiny solid particles of "monomers."[1] The primary distinguishing feature of a fractal cluster is scale invariance: The system remains self-similar over a wide range of the spatial scale. This scale invariance causes a slow (power-law) decrease in binary correlations in the arrangement of the monomer particles, so there can be a coherent scattering of light by the fractal cluster. The transition from incoherent to coherent scattering is very abrupt and is determined by the fractal dimension D of the cluster. In this letter we wish to examine this transition.

Let us consider the scattering of light by a fractal cluster consisting of nanometer-range metal particles. We assume that the length scale of the cluster, L, is comparable in order of magnitude to the wavelength of the incident light, λ. The frequencies which we are considering here are close to the frequencies of dipole surface plasmons in the isolated particles of the cluster. In other words, they correspond to the visible-to-UV part of the spectrum. An individual monomer is a spherical particle with a radius $R\ll\lambda\sim L$.

A fractal cluster with $L\gg R$ is a rather ephemeral structure, characterized by the presence of a large number of cavities with a power-law size distribution.[1] Near the frequencies of surface plasmons in the cluster, an ordinary renormalization of the wavelength of an incident photon occurs. The wavelength of the photon in the cluster, $\lambda_{\rm int}$, becomes far shorter than the wavelength of the incident photon, λ. For a typical photon trajectory in the cluster, there is typically a multiple rereflection from the cavity walls or a "cycling" involving a pair of monomers, as illustrated in Fig. 1. It is a straightforward matter to construct a simple equation (again, see Fig. 1) for the average t-matrix of the scattering of a photon by a monomer particle which reflects specifically this feature of the motion of a photon in a cluster. A dashed line here represents the potential of the interaction of a photon with a particle, $\mathscr{P}$. A dot is associated with a factor n_0 (n_0 is the average concentration of particles in the cluster). A light horizontal line represents the propagator of a free photon in the gauge with a zero scalar potential D^0. A heavy horizontal line represents the average photon propagator in the cluster D which satisfies the Dyson equation, represented in the same manner. The quantity Σ is the photon mass operator, which is determined by the last lines in Fig. 1.

The differential cross section for the elastic scattering of a photon by a cluster can

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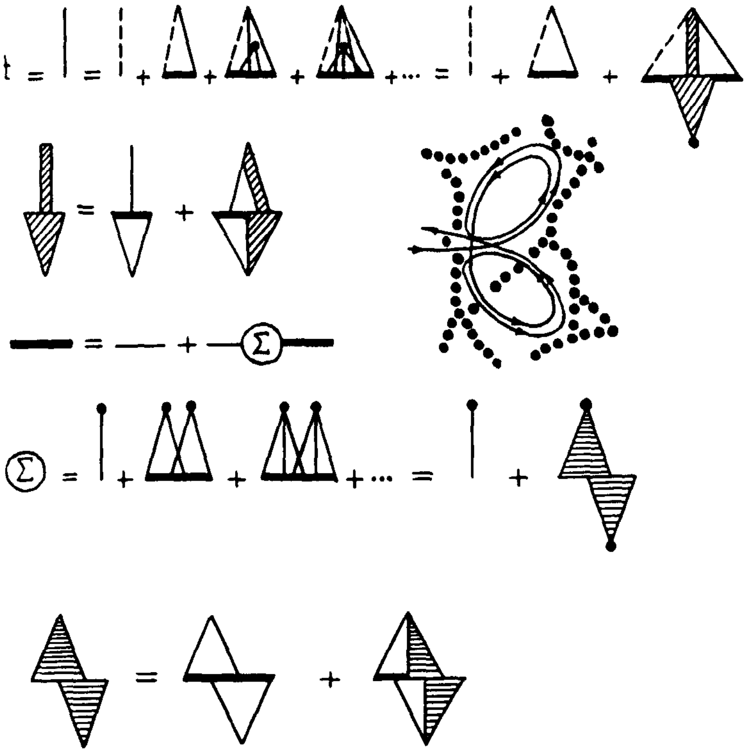

FIG. 1. Typical photon trajectory in a fractal cluster. Shown here are the perturbation-theory series for the average t-matrix of the scattering of a photon by a monomer, the Dyson equation for a one-particle photon propagator in the cluster, and the perturbation-theory series for the photon mass operator, constructed in accordance with the picture of the propagation of a photon in a fractal cluster.

be found by taking the average of the square modulus of the scattering amplitude, τ, for which several perturbation theories are available:[2,3]

$$\tau=e_{i\alpha}\int e^{-i\mathbf{k}_f\mathbf{r}}\sum_{\mathbf{a}}\mathscr{P}^{\mathbf{a}}_{\alpha\beta}(\mathbf{r},\mathbf{r}')e_{f\beta}e^{i\mathbf{k}_f\mathbf{r}'}d\mathbf{r}d\mathbf{r}'+e_{i\alpha}\int e^{-i\mathbf{k}_f\mathbf{r}}$$
$$\times\sum_{\mathbf{a}}\mathscr{P}^{\mathbf{a}}_{\alpha\beta}(\mathbf{r},\mathbf{r}_1)D^0_{\beta\gamma}(\mathbf{r}_1,\mathbf{r}_2)\sum_{\mathbf{b}}\mathscr{P}^{\mathbf{b}}_{\gamma\nu}(\mathbf{r}_2,\mathbf{r}')e_{f\nu}e^{i\mathbf{k}_f\mathbf{r}'}d\mathbf{r}d\mathbf{r}'d\mathbf{r}_1d\mathbf{r}_2+\cdots.$$

Here

$$\mathscr{P}^{\mathbf{a}}_{\alpha\beta}(\mathbf{r},\mathbf{r}')=\frac{\epsilon(\omega)-1}{4\pi}\delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}')\theta(R-|\mathbf{a}-\mathbf{r}|)\frac{\omega^2}{c^2}$$

is the potential of the interaction of a photon with a particle centered at point a,

$$D^0_{\alpha\beta}(\mathbf{r},\mathbf{r}')=\left(\delta_{\alpha\beta}-\frac{c^2}{\omega^2}\frac{\partial^2}{\partial r_\alpha \partial r'_\beta}\right)\frac{e^{-i\omega|\mathbf{r}-\mathbf{r}'|/c}}{|\mathbf{r}-\mathbf{r}'|} \qquad (1)$$

is the propagator of a free photon, ω is the frequency, c is the velocity of light in vacuum, $\mathbf{k}_i$ and $\mathbf{k}_f$ are the wave vectors of the incident and scattered photons, $\mathbf{e}_i$ and $\mathbf{e}_f$ are the corresponding polarization unit vectors, $\theta(x)$ is the Heaviside unit step function, and

$$\epsilon(\omega)=1-\omega_0^2/\omega^2$$

is the dielectric constant of the metal, where ω_0 is the classical plasma frequency of an electron gas in a metal.

Equations (1) are solved in the dipole approximation. In solving these equations, we make use of the condition $R\ll\lambda$ and the circumstance that the cavity size in which we are interested is on the order of the renormalized photon wavelength λ_{int}. Near the frequencies of surface plasmons, the latter wavelength is also far smaller than λ [under these conditions, we can ignore the first term in (1) for the vacuum propagator D^0, and we can seek a solution of the Dyson equation for D in the form $D_{\alpha\beta}(\mathbf{r},\mathbf{r}')\propto\nabla_\alpha\nabla'_\beta G(\mathbf{r},\mathbf{r}')$, see Ref. 2 for further details]. The t-matrix becomes

$$t^{\mathbf{a}}_{\alpha\beta}(\mathbf{r},\mathbf{r}')=\frac{\tau}{4\pi}\frac{\omega^2}{c^2}\,\delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}')\theta(R-|\mathbf{a}-\mathbf{r}|),$$

where the resonant factor τ characterizes a dipole surface plasmon in a particle in the neighborhood of other particles of the cluster. The dependence of $|\tau|^2$ on the dimensionless frequency $y=\omega/\omega_1$ ($\omega_1=\omega_0/\sqrt{3}$ is the frequency of a dipole surface plasmon in an isolated particle) is shown in Fig. 2. We easily see that with increasing D the surface plasmon becomes poorly defined. The characteristics of this plasmon depend only weakly on the correlations in the positions of the particles of the fractal cluster. These correlations were accordingly ignored in the system of equations in Fig. 1.

For the scattering cross section, the correlations play a decisive role. When we take the average of the square modulus of the amplitude τ, we find a perturbation-theory series for the differential cross section for elastic scattering, $d\sigma/d\mathbf{n}_f$ ($\mathbf{n}_f$ is a unit vector in the direction of the scattered photon). The structure of this series is clear from Fig. 3. The zigzag line here corresponds to a pairwise correlation

$$g(r)=-\frac{r^{D-3}}{4\pi n_0 L^D\Gamma(D)}\exp\left(-\frac{r}{L}\right),$$

where $\Gamma(x)$ is the gamma function, and the wavy lines represent the wave functions of the incident photon, $e_i\exp(-i\mathbf{k}_i\mathbf{r})$ and of the outgoing photon, $e_f\exp(i\mathbf{k}_f\mathbf{r})$. The double-headed hatched blocks are the same as in Eq. (1) for the mass operator Σ.

The first term in the expression for $d\sigma/d\mathbf{n}_f$ in Fig. 3 describes ordinary incoherent Rayleigh scattering of light by the monomers. The other terms stem from correlation effects and some unusual interference effects which occur in this case. For example, let us examine the second diagram on the right. Since the wavelength of the incident photon satisfies $\lambda\sim L\gg R$, we are incapable in principle of distinguishing the particular order in which the multiple traversal of the dimer by the photon occurs in the fractal

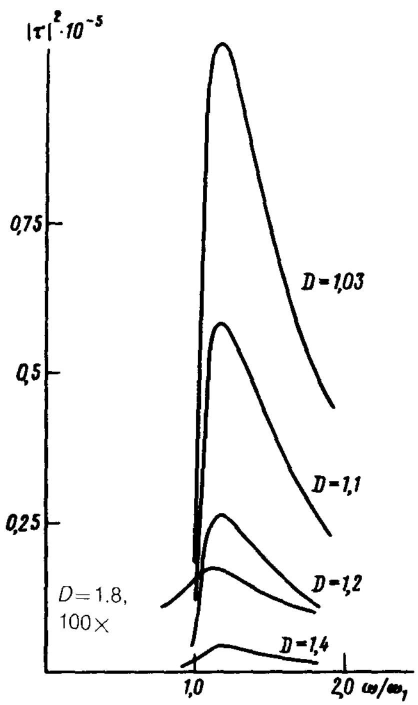

FIG. 2. Frequency dependence of the quantity $|\tau|^2$, which is a characteristic of the mechanism for the intensification of the scattering associated with a collective plasmon in a fractal cluster.

region (i.e., we cannot determine whether particle 1 comes first and then particle 2 or vice versa; this case is illustrated in Fig. 3). Furthermore, we are unable to distinguish which photon (the ith or fth) is executing this traversal. An interference of the probability amplitudes corresponding to these alternative possibilities stems from the second diagram in Fig. 3. From this standpoint, curiously, the first diagram, which describes Rayleigh scattering, is due exclusively to our ignorance of which photon (the ith or the fth) is interacting with the particle at the given instant.

The scattering is dominated by the diagrams of the subseries enclosed in parentheses. The greater the correlation of the particles in the diagram, the stronger the contribution of this diagram to the cross section. For a diagram of this series which contains N correlated particles ($N \gg 1$) the amplification factor, i.e., the ratio of its contribution to the cross section to the Rayleigh cross section, is

$$|\tau|^N N^{\frac{3-2D}{D}} N^2 \left\{ \frac{\sin[(D-1)\arctan(2k_i L \sin\frac{\theta}{2})]}{(D-1)2k_i L \sin\frac{\theta}{2}} \right\}^{N/2}, \tag{2}$$

where θ is the scattering angle. At $D < 3/2$ this is a huge quantity, while at $D > 3/2$ it is essentially zero. In other words, as D passes through the value 3/2 the entity ceases to be invisible (the Rayleigh scattering by nanometer-range particles is negligible) and becomes visible.

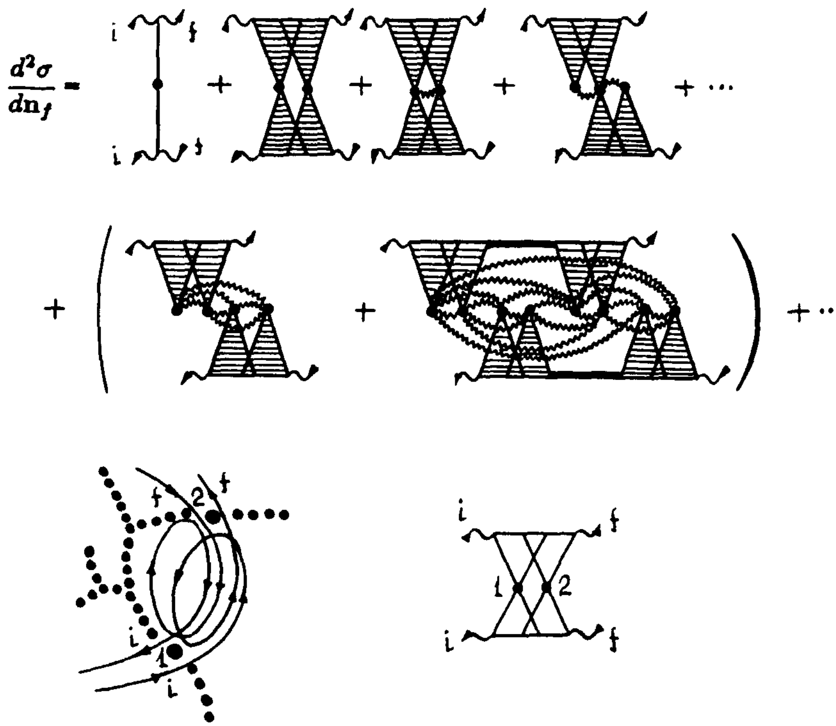

FIG. 3. Perturbation-theory series for the average differential cross section for the elastic scattering of a photon by a fractal cluster (not all of the 28 correlation functions are shown in the last diagram of the subseries enclosed in parentheses). Typical example of the processes whose amplitudes interfere, and a diagram describing the contribution of this interference to the scattering cross section.

The physical reason for the manifestation of coherence effects in the scattering at low values of D is either (on the one hand) the existence of long linear chains of monomers separated by a distance on the order of $\lambda_{\rm int}$ in the cluster, so that the waves scattered by the monomers interfere constructively—or (on the other) the presence of a large number of correlated cavities in the cluster which are at resonance with $\lambda_{\rm int}$, and which also scatter "in phase."

As can be seen from expression (2), the angular distribution of the scattered light shows an increased forward scattering, reminiscent of the scattering of light by a diffraction grating.

We are discussing this problem (i.e., the scattering of a photon by a system of small metal particles near the frequency of a surface dipole plasmon in the isolated particle) in pursuit of one independent goal: to show that interference effects occur in the scattering even if only a single photon ($L \sim \lambda$) is incident on the cluster, rather than a wavefront, as in the case with a diffraction grating. The physical reason for the interference in this case is an influx of photons into the cluster (near its resonant mode) which are not directly incident on the cluster, "from the side." If we attribute the intensification of scattering [the factor of $|\tau|^N$ in expression (2)] to a correlation mechanism, this intensification mechanism remains valid for a fractal cluster consisting of dielectric monomers, if the size of the cluster is greater than the wavelength of the photon and if a large number of photons are incident on the cluster.[4] Admittedly, in this case we will see distinctive features only in the differential scattering cross section; there will be no increase in the total scattering cross section.

The ability of a fractal cluster with a small value of D to exhibit an intense coherent scattering of light is intimately related to the possibility that strong local electromagnetic fields exist within the cluster. These could be either coherent electromagnetic fields of fluctuation origin or coherent fields induced in the cluster by external sources. Typical examples of entities in which we would expect strong local fields are a fractal system of cracks at the surface of certain metal catalysts[5] or the fractal skeleton of ball lightning, which is made up of chain aggregates of solid nanometer-range particles.[1]

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