

Theory and Numerical Simulation of Optical Properties and Selective Photomodification of Fractal Clusters

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Fractals, introduced by Benoit Mandelbrot, are scale self-similar mathematical objects possessing nontrivial geometrical properties [1,2]. There exist various physical realizations of fractals [3,4] and here we shall consider one of the most important such realizations, namely, fractal clusters. A fractal cluster is a system of interacting material particles called monomers. This system is self-similar (in statistical sense) with respect to scale transformation in an intermediate region of sizes r , $R_0 \ll r \ll R_C$, where R_0 is a characteristic separation between monomers and R_C is the cluster total size. A consequence of this is the scaling behavior in the intermediate region of the pair correlation function (density-density) $g(r)$, the number N of monomers in a cluster and its mean density $\bar{\rho}$:

$$g(r) = \frac{D}{4\pi R_0^3} \left(\frac{r}{R_0} \right)^{D-3}; \quad N \sim (R_C/R_0)^D; \quad \bar{\rho} \sim (R_C/R_0)^{D-3}, \quad (1)$$

where index D is the fractal dimension.

The linear optical (dipole) polarizabilities of fractals have been studied theoretically [5-8]. However, in [5-8], based on various modifications of the mean-field method, strong fluctuations in fractals

are not completely taken into account. In later papers [7,8] fluctuations play a decisive role. There the binary approximation was employed: the interaction of a monomer only with its nearest neighbor was accurately taken into account, while the effect of other monomers was simulated by the Lorentz field. Here we shall develop a theory of optical properties of fractals, taking full account of their fluctuation nature without the restriction of binary approximation.

Let us consider a model of cluster as a system consisting of N polarizable particles (monomers) located at the points \vec{r}_i . The total size of the cluster is supposed to be much less than the wavelength. The dipole moments \vec{d}_i induced on monomers obey the well-known system of equations:

$$d_{i\alpha} = \chi_0 E_{\alpha}^{(0)} - \chi_0 \sum_{j\beta} (i\alpha|W|j\beta) d_{j\beta} \quad (2)$$

where the Greek subscripts stand for tensor components and the Latin indices stand for ordinal numbers of monomers, χ_0 is the linear dipolar polarizability of an individual (isolated) monomer, $\vec{E}^{(0)}$ is the electric field of the external radiation and $(i\alpha|W|j\beta)$ is the dipolar interaction matrix:

$$(i\alpha|W|j\beta) = (1 - \delta_{ij}) [\delta_{\alpha\beta} - 3n_{ij}^{(ij)} n_{ij}^{(ij)}] r_{ij}^{-3} \quad (3)$$

Here $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ and $\vec{n}_{ij}^{(ij)} = \vec{r}_{ij}/r_{ij}$. The matrix W operates in $3N$ -dimensional space of the vectors \vec{d} and \vec{E} with components $(i\alpha|\hat{d}) = d_{i\alpha}$, $(i\alpha|\hat{E}) = E_{\alpha}$ and the basic system of equations may be written in the form:

$$(z + W)\hat{d} = \hat{E} \quad (4)$$

where the complex variable $z = -(X + i\delta) = \chi_0^{-1}$ is introduced.

Let us look at the dependence of X and δ on frequency ω for the particular case of an isolated resonance with frequency ω_m , relaxation rate (homogenous width Γ) and transition dipole matrix element d_{12} :

$$\chi_0 = \frac{|d_{12}|^2}{\hbar(\Omega + i\Gamma)}; \quad X = \frac{\hbar\Omega}{|d_{12}|^2}; \quad \Omega = \frac{\hbar\Gamma}{|d_{12}|^2} \quad (5)$$

It is clear that in this case X is a relative detuning and δ is a relative width of the resonance in an isolated monomer.

To find the properties of the exact solution, we perform a decomposition in terms of the eigenfunctions and eigenvalues of the operator W , which also turns out to be the most powerful computing method for carrying out our numeral simulations. Expressing the required quantities $d_{i\alpha}$ in terms of the eigenvalues w_n and components of cor-

responding eigenvectors $(n|U|i\alpha)$, one obtain from (4):

$$d_{i\alpha} = \chi_{\alpha\beta}^{(i)} E_{\beta}^{(0)}; \quad \chi_{\alpha\beta}^{(i)} = \sum_j (n|U|i\alpha) (n|U|j\beta) (z + w_n)^{-1} \quad (6)$$

From the form of the solution (6) the exact sum rules follow:

$$\int_{-\infty}^{\infty} \text{Im} \chi_{\alpha\beta}^{(i)}(X) dX = \pi \delta_{\alpha\beta}; \quad \int_{-\infty}^{\infty} \text{Re} \chi_{\alpha\beta}^{(i)}(X) dX = 0 \quad (7)$$

It can be seen that the absorption integral is conserved; it is the same as for an isolated monomer.

Let us also employ rotational symmetry. Then after averaging over the orientation of a cluster as a whole and over monomers in a cluster (denoted as $\langle \dots \rangle_0$) the polarizability tensor is reduced to a scalar: $\langle \chi_{\alpha\beta}^{(i)} \rangle_0 = \chi_{\alpha\beta}^{(i)}$.

Now we obtain from (6) an exact relation which is a counterpart of the optical theorem:

$$\frac{1}{N} \sum_i \chi_{\alpha\gamma}^{(i)*} \chi_{\beta\gamma}^{(i)} = \frac{1}{\partial N} \sum_i \text{Im} \chi_{\alpha\beta}^{(i)} \quad (8)$$

With the aid of (8) let us find the average (over a cluster) squared electric field \vec{E}_i which acts upon a monomer. This field determines the enhanced Raman scattering from the cluster, and also its photomodification. The acting (local) field is coupled to the solution of the system (4) in an obvious manner: $\vec{E}_i = \chi_0^{-1} \vec{d}_i$. For definiteness let us suppose that the external field $\vec{E}^{(0)}$ is directed along the z-axis. Then from (8) we obtain:

$$\frac{1}{N} \sum_i |\vec{E}_i|^2 / |\vec{E}^{(0)}|^2 = \delta (1 + X^2 / \delta^2) \text{Im} \chi_{zz} \quad (9)$$

The above exact results didn't use any specificity of fractals. Those results, however, will now be employed to describe the collective polarizability of fractals. Self-similarity, which is a fundamental property of fractality, means that a fractal reproduce itself when the spatial scale is changed. Since in general a fractal is a random object this reproduction has statistical meaning. A change of length scale brings about simultaneous change of the scale of eigenvalues w_n and the variable X . As a consequence of this, intermediate asymptotic values of observed quantities as function of w and X should be scalable.

Let us describe $\text{Im} \chi(X)$ (absorption). From (6) we see that scaling behavior of $\text{Im} \chi(X)$ may exist only if $|X| \gg \delta$. Then absorption is determined by those eigenstates for which $X = w_n$. These eigenstates are characterized by a coherence length L_X and are collective, i.e. delocalized over many monomers, in the intermediate region: $R_0 \ll L_X \ll R_C$. Taking into account dimensionality arguments and provided $|X| \gg \delta$ we

obtain in this region:

$$\text{Im}\chi(X) \sim R_0^3 (R_0^3 |X|)^{d_0-1} \quad (10)$$

where d_0 is an index which we call the optical spectral dimension. From the convergency requirement it follows that $d_0 > 0$.

Now, using a renorm-transformation, we can evaluate some transformation rules for R_0 , X and L_X . Such renorm-transformation means coarsing of the spatial precision with which a fractal is viewed and is performed in the following manner: we isolate in a fractal fluctuations with sizes on the scale of l_0 and then we consider monomers, which forms this fluctuations, as a new composite (renormalized) monomers with renormalized X . The renorm-transformation should conserve the total absorption of a fractal and the coherence length L_X . Thus we obtain:

$$|X| \propto R_0^{-(3d_0-D)/(d_0-1)}; \quad L_X \sim R_0 (R_0^3 |X|)^{(d_0-1)/(D-3)} \quad (11)$$

For $D=3$ (a trivial fractal) the expression for L_X is singular. This singularity has simple physical meaning, and to see this we consider the reciprocal of the function (11), which is the dispersion relation for the excitation of a fractal. For $D=3$ it follows from (11) that $|X|$ does not depend on L_X (the excitation wavelength). This fact exactly corresponds to the dispersion law (existence of a spectral gap) of long-wave surface plasmons to which the dipole excitation of a fractal tends as $D \rightarrow 3$.

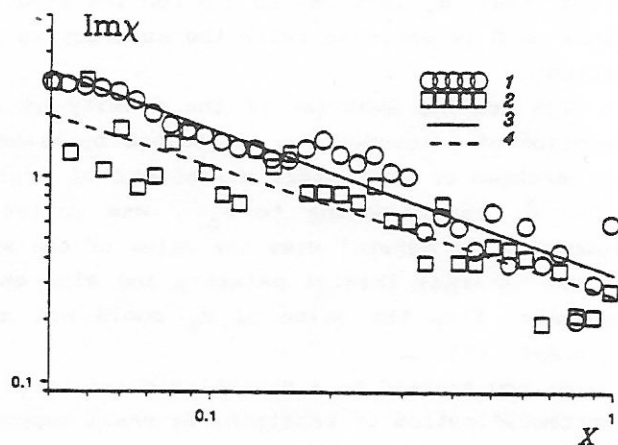
We now find the relation between the absorption of a fractal $\text{Im}\chi(X)$ and density of eigenstates $\nu(X) = \langle \sum_n \delta(X - w_n) \rangle$. In a strongly disordered system such as a fractal all collective eigenstates should with equal statistical weights contribute to the optical absorption. Hence, as it follows from (6), sum rules for absorption contour and provided that scaling of exists we can write:

$$\text{Im}\chi(X) \simeq \frac{\pi}{3} \nu(X) \sim |X|^{d_0-1} \quad (12)$$

Note, that relation (12) is not exact. It is violated at the wings of absorption contour and has a statistical sense in the collective region.

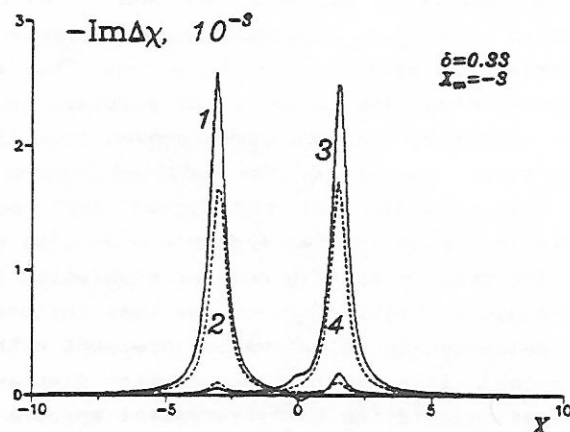
The fundamental predictions of the theory are the universal scaling behavior of absorption (10) and relation (12). These predictions (as well as other theoretical results) were tested by numeral simulation. At Fig.1 results of such simulation for random walks fractal clusters ($D=2$) are shown. From this figure it is clear that the absorption and density of states in fact obey Eq. (10), (12) with good accuracy. The values of optical spectral dimension were measured for

Fig.1. Fractal absorption as a function of X (for $X > 0$) on a double logarithmic scale built for an ensemble of random-walks clusters ($D=2$, number of clusters 75, average number of monomers in a cluster 30). 1 - found from the exact formula (6); 2 - expressed via the state density according to (12). The corresponding lines are obtained by linear regression. The value X is measured in units of R_0^{-3} , and χ - in units of R_0^3 , where R_0 is determined by (1).



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Fig.2. Change of the fractal absorption spectra induced by photo-modification (the plus sign corresponds to a decrease of absorption): 1,2 - parallel and 3,4 - normal polarization; 2,4 - numeral calculations via exact formula 1,3 - the analytical result of the binary approximation. The parameters are: $X_m = -3$, $\delta = 0.33$, $K = 0.25$. All values are measured in the same units as in Fig.1. For calculations is used the same ensemble of clusters as in Fig.1.



$D=1.8$ (self-avoiding random walks clusters), $D=2.0$ (random walks clusters), and $D=2.5$ (Witten-Sander clusters). The results are correspondingly $d_0 = 0.43(3)$, $0.38(3)$, $0.49(2)$ (from the absorption) and $d_0 = 0.54(8)$, $0.33(5)$, $0.51(6)$ (from states density). We note, that in

all cases $0 < d_0 < 1$, which corresponds to a normal dispersion law (11). For a trivial fractal no scaling behavior is observed. Also we conclude that d_0 is close to 0.5 for the studied fractals and no dependence on D is observed (with the accuracy we had in our numeral simulation).

The scaling behavior of the density of fractal eigenstates as a function of frequency was introduced by Alexander and Orbach [9] for the problem of mechanical vibrations of fractals ("fractons"), where index \bar{d} , corresponding to d_0 , was called the spectral (fracton) dimension. In general case the value of the spectral dimension depends on the internal fractal geometry and also on the interaction between monomers. Thus the value of d_0 should not necessarily coincide with \bar{d} in Ref. [9].

We now proceed to a numeral simulation of the threshold selective photomodification of fractals. We shall suppose the modification to be local: a given monomer is modified (evaporated, or, actually, deleted from a fractal) if the intensity of the local field exceeds a threshold $I_m = \text{const}$: $I_i = |\vec{E}_i|^2 > I_m$. For the modifying radiation we take the following parameters: $X = X_m$, $K = I/I_{th} - 1 > 0$ (I_{th} is threshold intensity of the modifying radiation for which the modification starts and is coupled with I_m by the amplification factor G : $I_{th} = I_m/G$), and linear polarization parallel to the z -axis. The modification condition was computed according to the exact solution (6). Those monomers for which this condition was met were removed from the fractal. After that the absorption spectra of the modified fractal were numerally determined for the parallel (z) and normal (xy) polarizations of the probe radiation. Finally the ensemble averaging was performed.

The results of this numeral simulation (differential spectra) are performed at Fig.2. Here we see that the photomodification is spectral and polarization-selective in agreement with experiment [10]. At Fig.2 a dashed line is also shown which displays the results of the theoretical calculation of differential spectra performed in binary approximation. Good agreement shows us that the photomodification at wings of the absorption contour has a binary nature (pairs which fit resonance condition are burned). For binary spectra look [7,8]. The binary approximation also makes it clear why we have two different picks for parallel and normal polarization of the probe radiation. This is a property of an absorption contour of a pair of monomers. Namely, for the z -polarization of the probe field the spectral hole is positioned at the modification frequency ($X = X_m$), and in the case of xy -polariza-

tion the hole is centered about the "mirror" frequency ($X = -X_m/2$ for $X_m < 0$ and $X = -2X_m$ for $X_m > 0$).

The more detailed results are going to be published in PHYS.REV.B.

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