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Theory and Numerical Simulation of Optical Properties of Fractal Clusters

by

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THEORY AND NUMERICAL SIMULATION OF OPTICAL PROPERTIES OF FRACTAL CLUSTERS

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Theory of linear optical properties of fractal clusters is developed. The theory is based upon the exact properties of dipole polarizability and assumption of the existence of scaling for the dipole excitations (eigenstates) of the fractal. This assumption is self-consistently validated by the results of the theory and is also confirmed by numerical stimulation in the framework of the Monte-Carlo method. Using exact relations and the scaling requirements, it is shown that the fractal absorption and density of eigenstates scale with the same exponent $d_o - 1$. The index d_o, which is called the optical spectral dimension, is the counterpart of the Alexander-Orbach fracton dimension $\bar{\bar{d}}$. The dispersion law for the fractal eigenstates is found: it is governed by d_o and the Hausdorff dimension D. Using this law, the condition of the scaling is determined. According to this, the scaling region occupies the center of the absorption band of the fractal. The spectral dimension d_o is found for three types of the fractals studied from the results of the numerical simulation. The wings of the fractal absorption contour are described by introducing the model of diluted fractals with the use of the binary approximation. The general conclusion is that large fluctuations of local fields dominate in determining the fractal optical spectra.

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1. Introduction

Fractals, as introduced by Benoit Mandelbrot over ten years ago, 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 what we believe to be one of the most important such realizations, namely, fractal clusters. Attention will be paid mainly to their optical properties.

A fractal cluster is a system of interacting material particles called monomers. This system is self-similar (in a statistical sense) with respect to scale transformations in an intermediate region of sizes r, $R_0 \ll r \ll R_c$, where R is a characteristic separation between nearest monomers, and R_c is the cluster total size. For the sake of brevity, such clusters will simply be called fractals, since other realizations of fractals will not be used in this paper.

Fractals are widespread in nature [4,5]. Products of a broad class of diffusion-controlled aggregation reactions in solutions and gases can be labelled as fractals. Accordingly, fractals are particles in colloidal solutions, sols and gels, and soot and smoke, and most macromolecules are fractals. Rough surfaces, disordered layers on surfaces, porous objects (microfilters including activated coal, porous glasses, aerogels, many heterogeneous catalysts, etc.) also possess fractal structure in an intermediate range of sizes.

A consequence of the scale self-similarity of fractals is a power dependence of correlation functions on coordinates. In particular, the pair (density-density) correlation function in the intermediate region has the scaling form

$$g(r) = \frac{D}{4\pi R_0^3}\left(\frac{r}{R_0}\right)^{D-3} \tag{1}$$

where the index D is called the fractal (external Haussdorff) dimension. Equation (1) is also a definition of the constant R_0. A consequence of (1) is the scaling behavior of the number N of monomers in a fractal and its mean density $\bar{\rho}$:

$$N \sim (R_c/R_0)^D \quad , \quad \bar{\rho} \sim (R_c/R_0)^{D-3} \quad . \tag{2}$$

A fractal is called nontrivial if D is less than the dimension of the space it is embedded in, or in our case of a cluster, if $D < 3$. As one can see from (2), the mean density of a

nontrivial fractal is asymptotically (for $R_c \gg R_0$) zero. This feature, together with strong pair correlation (1), is the reason for the great role that fluctuations play in a nontrivial fractal.

Among the physical properties of fractals, the optical properties have been the least studied. For example, in a set of proceedings [4] several years ago devoted to the physical properties of fractals, there are no papers on the optical properties. The linear optical (dipole) polarizabilities of fractals have been studied theoretically [6-9]. However, in Refs. [6,7] based on various modifications of the mean-field method, strong fluctuations (of density, local fields, etc.) in fractals are not completely taken into account. In later papers [8,9], fluctuations in the immediate surroundings of a monomer are seen to play a decisive role. There, the binary approximation was employed: the interaction of a monomer with only its nearest neighbor was accurately taken into account, while the effect of other monomers was simulated by the Lorentz field.

If monomers are high-quality optical resonators, for example, particles of coinage (e.g., Ag and Au) metals or other metals possessing strong luster, strongly-fluctuating local fields in a fractal can significantly exceed the exciting external field. This very feature brings about a giant enhancement of Raman scattering from fractals [8,9]. Nonlinearities, as usual, increase the effect of fluctuations and related to this are the huge magnitudes of nonlinear polarizabilities of fractals predicted in [10,11]. This prediction was then confirmed experimentally [12] for optical phase conjugation in a degenerate four-wave mixing process in fractal silver clusters. Photomodification of such clusters, which is selective in the wavelength and polarization of the radiation, was later observed [13]. These established effects are rich in physics and promise interesting applications. Therefore, they are worthwhile for more detailed theoretical study.

The aim of this paper is to develop a theory of optical properties of fractals, taking full account of their fluctuation nature without the restriction of the binary approximation. The theory is based on a scale-invariant approach. In Sec. 2 we present the basic equations, and in Sec. 3 we examine the polarizability in the collective region. The fractal optical response at the wings of the absorption contour is addressed in Sec. 4, results of numerical simulation are given in Sec. 5, and Sec. 6 provides a concluding discussion.

2. Basic equations and exact properties of solutions

Let us consider a model of a cluster as a system consisting of N polarizable particle (called monomers) located at the points $\mathbf{r}_i$, $i = 1 \ldots N$. The total size of the cluster is supposed to be much less than the wavelength. Therefore, the interaction between monomers at optical frequencies is principally the interaction of their transitional multipole moments. The main part of this work deals with interactions at distances which are much greater than the monomer size, which is valid for both the collective region (see Sec. 3) and diluted fractals in the binary region (see Sec. 4). In this case the most important part of the multipole interaction is the dipole one. However, if the monomers are conducting particles, the charge transfer between them (ohmic current) should also be considered. In Maxwell's equations, the dipole interaction in a continuous medium is described by the displacement current $\mathbf{j}_d = (\epsilon/c)(\partial \mathbf{E}/\partial t) = i\epsilon(\omega/c)\mathbf{E}$, where ϵ is the dielectric constant, $\mathbf{E}$ is electric field, ω is the optical frequency. The ohmic current $\mathbf{j}_\Omega = \sigma \mathbf{E}$, where is σ is the conductivity. The ratio $|j_\Omega/j_d|$ shows us how important the ohmic coupling is with respect to the dipolar one. Using the above expressions, in which ϵ is taken to be the dielectric permittivity of the monomer material, to describe the pair interaction between nearest monomers, we can obtain the upper estimate for $|j_\Omega/j_d|$. The reason why this estimate is the upper one is that the ohmic contact between monomers may have high resistance due to its small area, surface oxidation, spatial separation, etc., while the dipole interaction is determined by geometrical factors and, therefore, is the same order of magnitude as in the continuous medium. Using the familiar relation $\sigma = (\omega/4\pi)\mathrm{Im}\epsilon$, we obtain the estimate $|j_\Omega/j_d| \lesssim |\mathrm{Im}\epsilon/\mathrm{Re}\epsilon|$. In fact, in the visible portion of spectrum this ratio is small for most metals since $-\mathrm{Re}\epsilon$ is large, and for semiconductors and dielectrics since $\mathrm{Im}\epsilon$ is small. For example, for $\lambda = 650$ nm, $|j_\Omega/j_d| = 0.07$ and 0.02 for Ag and Si, respectively (the optical data are taken from Ref. [14]). We shall consider materials of the types indicated above and, therefore, shall neglect the ohmic coupling.

The dipole (transition) moments induced on monomers obey the well-known system of equations

$$d_{i\alpha} = \chi_0 E^{(0)}_{i\alpha} - \chi_0 \sum_{j=1}^{N}{}' \left[\delta_{\alpha\beta} - 3n^{(ij)}_\alpha n^{(ij)}_\beta\right] r^{-3}_{ij} d_{j\beta} \quad . \tag{3}$$

where the Greek subscripts stand for tensor components (summation over repeated indices is implied), and the Latin indices stand for ordinal numbers of monomers: α_0 is the dipolar polarizability of an individual (isolated) monomer, taken to be isotropic to avoid inessential complications; $\mathbf{E}^{(0)}$ is the electric field of the external (exciting) radiation at the site of the ith monomer; $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, and $\mathbf{n}^{(ij)} = \mathbf{r}_{ij}/r_{ij}$.

In the electrodynamics of continuous media, Eq. (3) is treated in the mean-field approximation, in which it is reduced to the integral equation for the mean dipole momentum $\mathbf{d}$, $\mathbf{d} = \alpha_0\mathbf{E}^{(0)} - \alpha_0 \int [\mathbf{d} - 3\mathbf{r}(\mathbf{rd})/r^2] r^{-3} g(r) d\mathbf{r}$. Taking (1) into account, one can easily see that for the nontrivial fractal the right-hand side of this equation diverges at small distances as r^{D-3}. It means that the most important is the contribution of the nearest monomers. This feature was exploited in recent work [8-11] in which the binary approximation has been developed to exactly take into account the field of the closest monomer, with the rest of monomers treated by the mean field. Due to the divergence at $r \to 0$, this approach is sensitive to the small-scale structure of the fractal, and the polarizability found does not obey scaling in the general case. In the present paper (Sec. 4), we introduce the model of diluted fractals, which have the simplest possible small-scale structure and obey scaling both at intermediate and small scales. In this model, the binary approximation is shown to correctly describe the wings of the absorption contour of the fractals.

We show below (Sec. 3) that the central part of the spectra can self-consistently be described as scale-invariant. This means that the polarizability is determined by collective (long-scale correlated) fluctuations of the local dipole momenta $\mathbf{d}_i$. Since these fluctuations dominate, the the mean-field approach is not applicable. In this respect, the fractals resemble systems near the phase transition point. In the collective region, the fractal optical properties are not sensitive to the small-scale structure, scaling takes place, and the diluted fractals has the same polarizabilities as the original ones.

To develop the general theory of the optical properties of fractals, we shall use the approach based on the expansion of polarizability in terms of the exact eigenvectors and eigenstates of the interaction operator (this is similar to the Lehman expansion in field theory). Also, such an expansion turns out to be the most efficient computing method for

carrying out our numerical simulations in Sec. 5.

To perform the above indicated expansion, let us represent (3) in the form of an operator equation by introducing the matrix W with elements

$$(i\alpha|W|j\beta) = \begin{cases} [\delta_{\alpha\beta} - 3n_\alpha^{(ij)} n_\beta^{(ij)}] r_{ij}^{-3}, & i \neq j; \\ 0, & i = j \end{cases} \quad . \tag{4}$$

which operates in a $3N$-dimensional vector space of the vectors $\hat{d}$ and $\hat{E}$ with components

$$(i\alpha|\hat{d}) = d_{i\alpha} \quad , \qquad (i\alpha|\hat{E}) = E_{i\alpha} \quad . \tag{5}$$

Then the basic system of equations (3) acquires the form

$$(Z + W)\,\hat{d} = \hat{E}^{(0)} \quad , \tag{6}$$

where the complex variable Z with real $(-X)$ and imaginary $(-\delta)$ parts is introduced,

$$Z \equiv -(X + i\delta) = \chi_0^{-1} \quad . \tag{7}$$

The symmetric real matrix W is diagonalized by the orthogonal transformation

$$UWU^{\mathrm{T}} = \mathrm{diag}(w_n) \quad , \quad UU^{\mathrm{T}} = 1 \quad . \tag{8}$$

where $\mathrm{diag}(w_n)$, is the diagonalized matrix consisting of eigenvalues w_n, and the superscript T denotes the transpose. The formal solution of the basic equation (6) has the form

$$\hat{d} = U^{\mathrm{T}} \mathrm{diag}\left(\frac{1}{Z + w_n}\right) U \hat{E}^{(0)} \quad . \tag{9}$$

Expressing the required quantities $d_{i\alpha}$ in terms of the eigenvalues w_n and components of the corresponding eigenvectors $(n|i\alpha)$, one obtains from (9)

$$d_{i\alpha} = \sum_{jn} (n|U|i\alpha)(n|U|j\beta)(Z + w_n)^{-1} E_{j\beta}^{(0)} \quad . \tag{10}$$

We consider below the clusters whose size R_c is much smaller than the radiation wavelength λ. Then the external field $E_{j\beta}^{(0)}$ may be considered homogeneous, i.e., not

depending on j. In this case the dipole moment induced on the ith monomer is expressed in terms of the corresponding linear polarizability $\chi^{(i)}_{\alpha\beta}$ as

$$d_{i\alpha} = \chi^{(i)}_{\alpha\beta} E^{(0)}_{\beta} \quad . \tag{11}$$

It is clear from (10) that $\chi^{(i)}_{\alpha\beta}$ is given by

$$\chi^{(i)}_{\alpha\beta} = \sum_{jn} (n|U^{\dagger}|i\alpha)(n|U|j\beta)(Z + w_n)^{-1} \, . \tag{12}$$

The polarizability as an analytical function of Z differs from that expressed in terms of frequency ω. In particular, this function is not "real" in the sense that $\chi^{(i)*}_{\alpha\beta}(X) \neq \chi^{(i)}_{\alpha\beta}(-X)$. Nevertheless, it can be shown directly from the solution (12) that the conventional Kramers-Kronig formula in X is valid,

$$\mathrm{Re}\chi^{(i)}_{\alpha\beta}(X) = \frac{1}{\pi}\mathbf{P}\int_{-\infty}^{\infty} \mathrm{Im}\chi^{(i)}_{\alpha\beta}(X')\frac{\mathrm{d}X'}{X' - X} \, , \tag{13}$$

where $\mathbf{P}$ denotes the principal value of the integral. We shall obtain also some other exact relations for $\chi^{(i)}_{\alpha\beta}$ which are a consequence of the fact that the eigenvalues w_n are real, the matrix U is orthogonal, and δ is positive corresponding to normal absorption (we assume no population inversion).

From the form of the solution (12), the exact sum rules follow:

$$\frac{1}{\pi}\int_{-\infty}^{\infty} \mathrm{Im}\chi^{(i)}_{\alpha\beta}(X)\mathrm{d}X = \delta_{\alpha\beta} \quad , \quad \mathbf{P}\int_{-\infty}^{\infty} \mathrm{Re}\chi^{(i)}_{\alpha\beta}(X)\mathrm{d}X = 0 \quad . \tag{14}$$

From (14) it can be seen that the absorption integral is conserved: it is the same as for an isolated monomer. The absorption cross section differs from $\mathrm{Im}\chi^{(i)}_{\alpha\alpha}$ only by a trivial factor $4\pi k$, where k is the wave vector. As a characteristic of absorption we shall use $\mathrm{Im}\chi^{(i)}_{\alpha\alpha}$.

We now obtain an exact relation which is a counterpart of the optical theorem. For this purpose, from (12) and taking (8) into account, we find a quadratic form of the polarizabilities:

$$\sum_{i} \chi^{(i)*}_{\alpha\alpha}\chi^{(i)}_{\alpha\beta} = \sum_{jj'n} (n|U^{\dagger}|j\alpha)(n|U|j'\beta)(Z^* + w_n)^{-1}(Z + w_n)^{-1} \quad . \tag{15}$$

Performing in (15) an elementary decomposition into simple fractions,

$$(Z^* - w_n)^{-1}(Z - w_n)^{-1} = -\mathrm{Im}(Z - w_n)^{-1} \quad \mathrm{Im}Z \tag{16}$$

and introducing the mean polarizability of a monomer in the cluster,

$$\chi_{\alpha\beta} = \frac{1}{N}\sum_i \chi^{(i)}_{\alpha\beta} \quad . \tag{17}$$

we obtain the required relation

$$\frac{1}{N}\sum_i \chi^{(i)*}_{\alpha\alpha}\chi^{(i)}_{\alpha\beta} = \frac{1}{\delta}\mathrm{Im}\chi_{\alpha\beta} \quad . \tag{18}$$

With the aid of (18), let us find the average (over a cluster) of the squared electric field $\mathbf{E}$ which acts upon a monomer. This field determines the enhanced Raman scattering from the cluster, and also its photomodification (see Sec. 4). The acting (local) field is coupled to the solution of the system (6) in an obvious manner:

$$\mathbf{E}_i = \chi_0^{-1}\mathbf{d}_i \quad . \quad E_{i\alpha} = \chi_0^{-1}\chi^{(i)}_{\alpha\beta}E^{(0)}_\beta \quad . \tag{19}$$

For definiteness, let us suppose that the external field $\mathbf{E}$ is directed along the z-axis. From (18) and (19) we obtain

$$\frac{1}{N}\sum_i |\mathbf{E}_i|^2/|\mathbf{E}^{(0)}|^2 = \delta\,(1 + N^2/\delta^2)\,\mathrm{Im}\chi_{zz} \quad . \tag{20}$$

This relation resembles the Callen-Welton formula. Earlier a result had been obtained with the use of the binary approximation (see Eq. (45) in Ref. [9]) which differs from (20) only by some notations. This result was the basis to develop a theory of enhanced Raman scattering from fractals.

We emphasize that the above relations (13), (14), (18) and (20) are exact. They are valid for an arbitrary cluster (fractal or not) without any averaging over cluster ensemble or orientation. It is only essential that the cluster consists of monomers with pair interactions between them which give a linear response to an external field and possess normal (not population-inverted) absorption.

Let us now invoke rotational symmetry. After averaging over the orientation of a cluster as a whole (denoted below as $\langle \ldots \rangle_0$), the polarizability tensor is reduced to a scalar,

$$\chi_{\alpha\beta} = \chi\delta_{\alpha\beta} \quad , \quad \chi = \frac{1}{3}\chi_{\alpha\alpha} \quad . \tag{21}$$

For the first moment of χ, we can find directly from (12)

$$\mathbf{P}\int_{-\infty}^{\infty} X\mathrm{Im}\chi(X)\mathrm{d}X = \mathbf{P}\int_{-\infty}^{\infty} \mathrm{Re}\chi(X)\mathrm{d}X + \frac{1}{3N}\sum_{ijn}\langle n|U|i\alpha\rangle w_n \langle n|U|j\alpha\rangle . \tag{22}$$

Accordingly to (14), the first term on the right side of (22) equals zero. The second term with the help of (8) is reduced to the form $\sum_{ij}\langle i\alpha|W|j\alpha\rangle$. Using the fact that the interaction tensor (4) becomes zero when convoluted over the indices α and β, one comes to the conclusion that the second term is also equal to zero. In such a way, we obtain the exact sum rule

$$\mathbf{P}\int_{-\infty}^{\infty} X\mathrm{Im}\chi(X)\mathrm{d}X = 0 \quad , \tag{23}$$

meaning that the absorption center of a cluster is at $X = 0$.

The universality of the above obtained relations is, in particular, due to the use of natural variables X and δ for the present problem. These variables, indeed, depend upon the frequency ω, but not in a universal way. In what follows, it is important to establish the region in which the spectral variable X is changing. Let us consider the dependence $X(\omega)$ in two different models.

First, let a monomer possess a single isolated resonance with frequency ω_0, relaxation rate (homogeneous width) Γ and transition dipole matrix element d_{12}. In this case we have

$$\chi_0 = -\frac{|d_{12}|^2}{\hbar(\Omega + i\Gamma)} \quad , \quad X = \frac{\hbar\Omega}{|d_{12}|^2} \quad , \quad \delta = \frac{\hbar\Gamma}{|d_{12}|^2} \quad , \tag{24}$$

where $\Omega = \omega - \omega_0$. One can see that X is a relative detuning and δ is a relative width of the resonance in an isolated monomer. We point out that the first of the relations (14) together with (23) reproduces the well-known dipole sum rule for the polarizability. Note that the minimum value of Γ is equal to the natural (radiative) linewidth $\Gamma_n = 4k^3|d_{12}|^2/3\hbar$.

Substituting $\Gamma = \Gamma_n$ into Eq. (24), we obtain for the resonance ($\Omega = 0$) the upper estimate $\mathrm{Im}\chi_0 \sim \lambda^3$, which is much greater than the volume of the monomer, in accord with the quantum-mechanical Breit-Wigner formula. In real clusters, the width Γ is determined by radiativeless dissipation and dephasing, and $\Gamma \gg \Gamma_n$. Thus, the magnitude of $\mathrm{Im}\chi_0$ is much smaller than λ^3, though it may still be much greater than the monomer size due to resonant enhancement (cf. the example below).

The second model, which is realistic, in particular, for clusters obtained by aggregation in colloidal solutions, considers the monomer as a macroscopic sphere with radius R_m, consisting of the material with a complex dielectric permittivity $\varepsilon \equiv \varepsilon' + i\varepsilon''$. The polarizability of such a sphere is given by the well-known expression

$$\chi_0 = R_m^3(\varepsilon - 1)(\varepsilon + 2)^{-1} \ . \tag{25}$$

For the metallic sphere, the permittivity is well described by the Drude formula

$$\varepsilon = \varepsilon_0 - \omega_p^2/\left[\omega(\omega + i\gamma)\right]^{-1} \ , \tag{26}$$

where ε_0=const is the contribution of the interband transitions (e.g., for silver $\varepsilon_0 \approx 5$), ω_p is the electron plasma frequency, and γ is the electron collision frequency. For most metals, in particular, for the noble metals, $\gamma \ll \omega_p$ and, consequently, $\varepsilon'' \ll \varepsilon'$. In this case, from (7) and (25), one can see that $X(\omega)$ turns to zero at the point of the surface plasmon resonance, $\omega = \omega_s$, where $\varepsilon' = -2$. From (26) it follows that $\omega_s \approx \omega_p(\varepsilon_0 + 2)^{-1/2}$. In the vicinity of the surface plasmon resonance, the dependence $X(\omega)$ reduces to (24) with the following parameter values: $\Omega = \omega - \omega_s$, $|d_{12}|^2 = 3R_m^3\hbar\omega_p/\left[2(\varepsilon_0 + 2)^{3/2}\right]$, $\Gamma = \gamma/2$. Also, it follows from (25) and (26) that for $\omega \ll \omega_s$, the variable X tends to -1. In the vicin of the frequency $\omega_d = \omega_p(\varepsilon_0 - 1)^{-1/2}$, the variable Z (7) experiences strong dispersion

$$Z \approx \frac{3}{2} R_m^{-3}(\varepsilon_0 - 1)^{-3/2}\omega_p/\left[\omega - \omega_d + i\gamma/2\right] \ . \tag{27}$$

It follows from (27) that in a small vicinity of ω_d the variable X changes sign, acquiring a large absolute value limited by $X_{max} = 3R_m^{-3}\omega_p/\gamma$.

3. Polarizability of a fractal in the collective region

To obtain the above exact results we did not use any specificity of fractals. Those results, however, will now be employed to obtain a description of the collective polarizability of fractals. Self-similarity, which is a fundamental property of fractality, means that a fractal reproduces 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 a 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 functions of w and X should be power-like (scaling).

We shall describe first $\mathrm{Im}\chi(X)$ (absorption) and later consider $\mathrm{Re}\chi(X)$ with the aid of (13). From (12) and taking (7) into account, we see that the universal scaling behavior of $\mathrm{Im}\chi(X)$ may exist only if $X \gg \delta$. Then

$$\mathrm{Im}\chi(X) = \frac{\pi}{3X} \sum_{ijn} \left\langle (n|U|i\alpha)(n|U|j\alpha)\delta(X - w_n) \right\rangle \quad . \tag{28}$$

where $\langle \ldots \rangle$ denotes an average over the ensemble of fractals. From (28) it follows that absorption is determined by those eigenstates for which $w_n = X$.

The eigenstates of any large system, including the fractal, are characterized by a coherence length (which turns out to be the wavelength in the trivial case $D = 3$), within which the excitations of monomers are strongly correlated. This coherence length is a function of the generalized frequency X, which we shall denote as L_X. The eigenstates are collective, i.e., delocalized over many monomers, if $L_X \gg R_0$. The scaling should exist in the intermediate region

$$R_0 \ll L_X \ll R_c \quad . \tag{29}$$

Our basic assumption is that the dependence of L_X on X allows the existence of a finite interval of X in which (29) is satisfied. Assuming so, we shall develop the scale-invariant theory of the collective excitations of fractals. This theory will eventually allow us to find the dependence of L_X from the scaling properties of relevant quantities and, in such a way, to self-consistently confirm the basic assumption. It is also supported by the results of the numerical simulation (Sec. 5).

In the scaling region (29), the form of the absorption contour $\mathrm{Im}\chi(X)$ should be power-like. The exact symmetry of the eigenstate problem of the W operator with respect to a sign change of w_n is absent. Consequently, parity in X is not held, suggesting that the indices for $X > 0$ and $X < 0$ could be different. However, the sum rule (23) together with the self-similarity of a fractal guarantees that these indices coincide, and the corresponding coefficients also coincide. Thus, in the region (29) (provided $|X| \gg \delta$), a symmetric scaling dependence of $\mathrm{Im}\chi$ is valid, with coefficients which are determined by R_0 and can be estimated from dimensionality arguments,

$$\mathrm{Im}\chi(X) \sim R_0^3 \left(R_0^3|X|\right)^{d_o - 1} \quad . \tag{30}$$

where d_o is an index which we call the optical spectral dimension. From the convergency requirement for the first of the sum rules (14), it follows that $d_o > 0$. We should emphasize that the symmetry of absorption in X is not an exact property, but is valid only in the collective region, and is violated in the binary region (see Sec. 4)

Let us consider the transformation properties with respect to a change in the minimum size R_0 (renorm-transformation). We take the initial value as $R_0 = 1$, i.e., we measure lengths in the unit of the initial size R_0 . Increasing R_0 means coarsing of the spatial precision with which a fractal is viewed. Such coarsing can be achieved in the following manner [15]. We first isolate in a fractal fluctuations with sizes on the scale of $l \gg 1$. We then consider monomers, which form such a fluctuation, as a new composite (renormalized) monomer. From the property of self-similarity, it follows that the obtained cluster, consisting of renormalized monomers, is a fractal with the same critical exponents, and $R_0 \sim l$. The renorm-transformation conserves the form of the absorption contour (30), provided the collectivity criterion (29) is met.

We now determine the renormalization law for X and find the coherence length L_X . The renorm-transformation should conserve the total absorption of a fractal. This requirement, taken together with (2) and (30), reduces to

$$\left(\frac{R_c}{R_0}\right)^D R_0^3 \left(R_0^3|X|\right)^{d_o - 1} \propto 1 \quad . \tag{31}$$

where the proportionality is understood in the sense of the dependence on R_0 (and not on R_c). From (31) it is clear that X transforms in the following way:

$$|X| \propto R_0^{-(3d_o-D)/(d_o-1)} \quad . \tag{32}$$

We note that in the case of a trivial fractal ($D = 3$), $|X| \propto R_0^{-3}$ independently of the index d_o, i.e., we have the well-known result $\mathrm{Re}\chi_0 \propto R_0^3$, which demonstrates the proportionality of the polarizability of a composite monomer to its volume. We should point out that the transformation law (32) is derived from general principles, and the single condition of its validity is the existence of the scaling behavior of absorption (see below).

For strongly-disordered systems such as fractals, Alexander [15] has assumed that collective states with a given frequency (whose role in this case is played by the variable X), are characterized by a single coherence length which is an analog of, simultaneously, the wavelength and localization radius. This property, called strong localization, is a consequence of the self-similarity of a fractal and its collective states. For a trivial (three-dimensional) system, this property in the general case can obviously be violated: the excitation localization radius can essentially exceed the wavelength, which is the situation of Anderson localization.

The coherence length L_X, as other observable quantities in the scaling regime, should not depend on both the maximum R_c and minimum length R_0. The latter requirement means that L_X should be an invariant of the renorm-transformation. Taking into account Eq. (32), one can make sure that it possible to construct only one quantity which possesses the above indicated properties and has the dimensionality of length.

$$L_X \propto R_0 (R_0^3 |X|)^{(d_o-1)/(3-D)} \quad . \tag{33}$$

Thus, for the present problem, Alexander's strong localization follows from scaling. The relation reciprocal to (33) is the dispersion law for the excitations of the fractal:

$$|X(L_X)| \propto R_0^{-3} (L_X/R_0)^{(3-D)/(d_o-1)} \quad . \tag{34}$$

This dispersion law for the collective dipole excitations of the fractal can be considered as a theorem based upon the general relations and the scaling assumption.

For the trivial system ($D = 3$), the expression (33) is singular, which is not surprising (see above). Nevertheless, this singularity has simple physical meaning, and to see this, we consider the dispersion law (34). For $D = 3$, it follows from (34) that $X(L_X)$ does not depend on L_X, i.e., on the excitation wavelength, $|X| \sim R_0^{-3}$. This estimate exactly corresponds to the dispersion law (existence of a spectral gap) of long-wave surface plasmons. Note that in the trivial limit ($D \to 3$) the dipole excitations of a fractal, which give contribution to the optical absorption, tend exactly to the surface plasmons since the radius of the fractal R_c is assumed to be much less than the radiation wavelength λ. To demonstrate this, consider an averaged trivial cluster as a microscopic sphere, for which the surface plasmon resonance condition is $\epsilon' = -2$ (cf. discussion of Eqs. (25)-(27)), where ϵ is the dielectric permittivity of the sphere material. Recalling that $\epsilon = 1 + 4\pi R_0^{-3}\chi_0$ and taking (7) into account, we obtain for the resonance $X = -\frac{4\pi}{3}R_0^{-3}$, in accord with the above estimate.

For a normal dispersion law, the "frequency" X should decrease with the wave vector, i.e., with an increase of L_X. Taking into account (34), this leads to the limitation $d_o < 1$, which we accept (as will be confirmed later by numerical simulations). We shall not use the renorm-transformations below and shall return to the original value $R_0 = 1$. Then the condition (29), which determines the scaling region, together with (2) and (33) acquires the form

$$N^{(3/D-1)/(1-d_o)} \ll X \ll 1 \quad . \tag{35}$$

Hence, this region is spread to small $|X|$ and, consequently, occupies the center of the absorption band (cf. (24)). Obviously, the necessary condition for scaling, $|X| \gg \delta$, is compatible with (35) only if the Q-factor of the resonance in a monomer is high:

$$Q = 1/\delta \gg 1 \quad . \tag{36}$$

As follows from (35), when a fractal tends to a trivial one ($D \to 3$), the region of collective behavior in X degenerates to a point corresponding to the frequency of surface plasmons (see above). Thus, a finite size of the band of collective states of a fractal is entirely due to its nontriviality.

The scaling region (35) can be physically realizable only if the interval of values of the function $X(\omega)$ (7) includes it. This interval depends on which monomers the cluster is built with. Let us discuss in this connection the above considered model of the spherical monomer (see (25)-(27) and the corresponding text). The monomer radius R_m is supposed to be on the order of $R_0 = 1$ (in the present units). There exist small values of X, including zero, near the frequency $\omega \approx \omega_s$, if the surface plasmon resonance is well pronounced, the condition of which $\gamma \ll \omega_s$ coincides with (36). In practice, it is well met for many metals, e.g., for silver $\gamma/\omega_s \lesssim 1/30$, with the resonant frequency ω_s corresponding to the near-UV wavelength of approximately 300 nm. From the discussion of Eqs. (25)-(27), it follows that in the most interesting band of $\lambda \gtrsim 300$ (i.e., soft-UV, visible and IR light) the variable X belongs to the interval from -1 to 0, which includes the whole scaling region (35). The variable X is in the interval from 0 to a large value of $\sim \omega_p/\gamma$ when ω is from ω_s to $\approx \omega_d$ (far-UV band), and large negative values of X occur for $\omega > \omega_d$.

To summarize the above obtain results, let us point out that we have initially assumed there to exist the region (29) of the collective fractal excitations. Starting from this point and using exact relations, we have shown that the absorption is given by the scaling law (30), found the scaling properties (34) of the variable X, and determined the dispersion law for the fractal excitations (33) (or (34)). From this law, we have determined the scaling conditions (35), which predict the final interval (in X or in ω) of the scaling. Thus, the initial assumption is self-consistently validated. Taking into account divergence at small r in the mean-field equations (see the discussion after Eq. (3)), the existence of the scaling means that in the region (35) the large-scale fluctuations dominate in determining the polarizability and cause failure of the mean-field approach. This situation is similar to the phase transition with X as the transition parameter, $X = 0$ being the transition point and the polarizability playing its usual role.

We now proceed to find the relation between the absorption of a fractal, $\mathrm{Im}\chi(X)$, and density of its eigenstates, $\nu(X)$, which is introduced in the conventional way as

$$\nu(X) = \frac{1}{N}\sum_n \left\langle \delta(X - w_n) \right\rangle \quad . \tag{37}$$

The density ν is normalized to one monomer, and the mean value of X weighted by $\nu(X)$

is equal to zero since the interaction tensor (4) is traceless:

$$\int_{-\infty}^{\infty} \nu(X)\mathrm{d}X = 3 \quad , \quad \int_{-\infty}^{\infty} X\nu(X)\mathrm{d}X = \mathrm{Tr}W = 0 \quad . \tag{38}$$

The value of the first integral in (38) is due to the vector character of the interaction between monomers.

Isolating in Eq. (28) the terms with $i = j$ using the orthogonality property of the matrix U, from Eq. (8) and comparing the obtained results with Eq. (37), we obtain

$$\mathrm{Im}\chi(X) = \frac{\pi}{3}\nu(X) + \frac{\pi}{3N}\sum_{i\neq j,n}\left\langle (n|U|i\alpha)(n|U|j\alpha)\right\rangle \quad . \tag{39}$$

It follows from (38) that the first term in this expression (39) satisfies both sum rules for $\mathrm{Im}\chi$, (14) and (23). Correspondingly, the zeroth and first moments in X of the second term in (39) are exactly equal to zero. Since this term should also have a scaling dependence on X (provided (35) is satisfied), it then equals zero. Hence, in the collective region

$$\mathrm{Im}\chi(X) = \frac{\pi}{3}\nu(X) \propto |X|^{d_o - 1} \quad . \tag{40}$$

i.e. both the absorption and eigenstate density scale with the same index d_o (the optical spectral dimension).

We emphasize that the relation (40) between the fractal optical absorption and the eigenstate density of its dipolar excitations has also the character of the theorem based upon exact sum rules and the scaling assumption. The physical meaning of Eq. (40) is clear. In a strongly-disordered system such as a fractal, all collective eigenstates with equal statistical weights contribute to the optical absorption. The relation (40), however, should only hold for nontrivial fractals in the spectral region (35). Note that at the wings of the absorption contour, which are described in the binary approximation (Sec. 4), it is violated In ordered systems, selection rules are valid, and only few states of corresponding symmetry contribute to the absorption.

The scaling behavior of the density of fractal eigenstates as a function of frequency was introduced by Alexander and Orbach [16] for the problem of mechanical vibrations

of fractals ("fractons"), where the index $\bar{\bar{d}}$, corresponding to d_s in (40), was called the spectral (fracton) dimension. In the general case, the value of the spectral dimension is determined by the internal fractal geometry, and, for a vector problem, also by the manner in which the fractal is embedded into the three-dimensional space. In principle, the spectral dimension may also depend on the interaction between monomers. Thus, the value of the optical spectral dimension d should not necessarily coincide with the vibrational spectral dimension in Ref. [16].

In Refs. [15-17] different ways are provided for finding an expression for the dispersion relation of fractal vibrations, which in our notation has the form

$$|X| \propto L_X^{-D/\bar{\bar{d}}} \tag{41}$$

which obviously differs from (34). In particular, for a trivial fractal ($D = 3$), the relation (41) predicts a gapless dispersion law, which is linear for $\bar{\bar{d}} = 3$. An inspection of the treatments of Refs. [15-17] shows Eq. (41) to be valid only for excitations of the Goldstone-type (acoustic phonons, magnons and so on), whose corresponding creation-annihilation operators in the long-wavelength limit turn to generators of symmetry groups of the given system (translations, rotations, etc.). The dipolar excitations are not of the Goldstone-type, but rather tend to surface plasmons, which do have a gap in the spectrum. In such a way, for the plasmon-like excitations considered in present paper the dispersion relation (34) is valid, and not (41), which holds for phonon-like excitations.

Finally, we briefly consider $\mathrm{Re}\chi(X)$. Since the kernel of the integral (Gilbert) transform (13) by itself does not provide convergence, both the collective region ($X \ll 1$) and wings of the absorption contour ($X \gtrsim 1$) give rise to $\mathrm{Re}\chi(X)$. Therefore, $\mathrm{Re}\chi(X)$ is not described by a scaling dependence even for X in the region (35). This conclusion can also be reached by starting directly from (12).

4. Wings of the absorption contour and model of diluted fractals

In the previous section the collective region was considered in which the eigenstates of a fractal are determined by interactions on large scales and spread over many monomers.

The observable quantities governed by such excitations are insensitive to small-scale details of fractal structure and the form of the interaction of monomers at small distances. The behavior of such quantities should be universal and scaling. Below we shall describe the wings of the fractal spectrum (the region $|X| \gtrsim 1$). In this case the external field interacts with those fractal excitations which, in accord with (33), are localized on a few monomers. Therefore, small-scale details are essential.

It follows from the above arguments that it is in principle impossible to formulate a universal description of the wings of a fractal spectrum. It thus seems appropriate to employ a model of a fractal with extremely simple structure on a small scale. For such a model we introduce dilute fractals which could be obtained by coarsing a structure of original fractals. Instead of the renorm-transformation described in Sec. 3, we shall use a simple and numerically efficient algorithm for the structure coarsing, called dilution, which consists of the following. Each monomer of an original fractal is randomly left at its position with some small probability $\beta \ll 1$, or is removed with the probability $1-\beta$. Then the fractal as a whole is reduced in size $(1/\beta)^{1/D}$ times. After that, we obtain a fractal which is characterized by unchanged critical exponents governing the collective region, with the pair correlation function (1) ($R_0 = 1$) being valid up to small distances on order of $r_0 = (\beta)^{1/D} \ll 1$. The dilution, indeed, does not change the polarizability of a monomer, i.e., it conserves the variable Z, and the total size of the fractal is diminished by a factor of r_0. The dilution (random decimation, in other words) eliminates the local structure of a fractal, such as connectivity, separation between nearest monomers, coordination numbers, etc., without affecting the collective region (Sec. 3).

The diluted fractals do not only provide a convenient theoretical model, needed, in particular, to check the numerical simulation in the spectral wings (see below). They also can be considered as an idealization for a rather wide class of fractal clusters in nature, which are formed by random binding of impurities to an original (nondiluted) fractal, with a small fraction of occupied binding sites. As examples, we may mention dye molecules or metallic microparticles in complexes with macromolecules, or in pores of glasses, and so on. Hence, the description of spectral wings in the model of diluted fractals is interesting by itself.

To describe the interaction at small distances, which is important for $|X| \gtrsim 1$, we shall use the binary approximation developed earlier [8-11]. For each monomer we shall exactly take into account its interaction with its nearest monomer neighbor, neglecting the effect of other monomers. The polarizability of a monomer in such a pair is given by the tensor $\chi_{\alpha\beta}^{(p)}$ [8],

$$\chi_{\alpha\beta}^{(p)} = (Z + r_n^{-3})^{-1}\delta_{\alpha\beta} + \left[(Z - 2r_n^{-3})^{-1} - (Z + r_n^{-3})^{-1}\right] n_\alpha n_\beta \quad . \tag{42}$$

where $\mathbf{r}_n$ is the radius vector of the nearest neighbor, $\mathbf{n} = \mathbf{r}_n / r_n$, and the superscript (p) stands for "pair". After averaging over $\mathbf{r}$, we obtain the mean polarizability of a monomer in the fractal as

$$\chi = \frac{1}{3}\int_0^\infty \left[2(Z + r_n^{-3})^{-1} + (Z - 2r_n^{-3})^{-1}\right]\rho(r_n)\mathrm{d}r_n \quad . \tag{43}$$

where $\rho(r_n)$ is the density of the distribution in distances between nearest neighbors. For $|X| \gg \delta$ it follows from (43) that (cf. (28))

$$\mathrm{Im}\chi = \frac{\pi}{9} r_n^4 \left[2\rho(r_n)\Theta(X) + 2^{1/3}\rho(2^{1/3}r_n)\Theta(-X)\right]\Big|_{r_n = |X|^{-1/3}} \quad . \tag{44}$$

where $\Theta(X)$ is the Heaviside unit step function.

At $|X| \gg 1$ the main contribution to (44) occurs at small distances r_n. Here the following asymptotic form, derived from (1), is valid,

$$\rho(r_n) \approx D r_n^{D-1} \quad , \tag{45}$$

which is universal for diluted fractals. Substitution of (45) into (44) yields the following expression for the far-wing absorption contour:

$$\mathrm{Im}\chi = \frac{2\pi D}{9}|X|^{-(1+D/3)}\left[\Theta(X) + 2^{D/3-1}\Theta(-X)\right] \quad . \tag{46}$$

As one can see, this is a scaling form but, in contrast to the collective region, the exponent is greater than unity, and the spectrum is asymmetric with respect to the sign of X (cf. (40) and the discussion of it). One can show by adding a third monomer to a pair of

nearest neighbors that the form of the absorption wing is given correctly by the binary approximation (46) in the leading term in $|X|$ (the relative magnitude of corrections is on the order of $|X|^{-2}$).

For the case $r_n \sim 1$, the distribution $\rho(r_n)$ cannot be found in a universal form since it is determined not only by (1) but also by higher correlation functions. To derive a rough estimate, let us neglect higher correlations. Then the number of monomers in a fixed vicinity of a given monomer obeys the Poisson distribution and, consequently,

$$\rho(r_n) = D r_n^{D-1} \exp(-r_n^D) \quad . \tag{47}$$

Substitution of (47) into (43) results in the expression

$$\chi = \frac{1}{3}\left[2S_D(-Z) - \frac{1}{2}S_D(\frac{1}{2}Z)\right] \quad . \tag{48}$$

where the function S_D of the complex variable Z is defined by means of the integral representation

$$S_D(Z) = -\frac{D}{3}\int_0^\infty y^{-(1+D/3)} \exp(-y^{-D/3}) \frac{dy}{Z-y} \tag{49}$$

and its analytic continuation. The function $S_D(Z)$ is analytic in the complex plane with the branch cut from 0 to B along the real axis; $Z = 0$ is a second-order branch point. The polarizability (48) satisfies all the exact relations obtained above.

The form of the expression (48) coincides with that previously derived [8,9], although the function $S_D(Z)$ is different. The difference is due to an incorrect averaging in [8,9] over configurations of a monomer pair in a fractal (see the text between Eqs. (14) and (15) in Ref. [9]). We emphasize that all the results of Refs. [8-11] remain valid, except that now the form (49) of the S_D-function should be used.

It can easily be shown that in the binary approximation the eigenstate density is symmetric in X and has the form

$$\nu(X) = \frac{1}{2}\int_0^\infty \left[2\delta(|X| - r_n^{-3}) + \delta(|X| - 2r_n^{-3})\right]\rho(r_n)dr_n \quad . \tag{50}$$

The relation (40) between $\mathrm{Im}\chi(X)$ and $\nu(X)$ in the binary region does not hold unless the polarizability (43) is symmetrized in X. Hence, the contribution from the binary region does not interfere with the normalization of the relation (40) in the collective region.

5. Results of numerical simulation

Three types of fractals were studied as obtained by the Monte-Carlo method with the use of a program generator of random numbers:

(i) Random walks ($D = 2$) were built on a cubic lattice with monomers placed at visited sites and subjected to 1000-fold ($\beta = 0.001$) dilution (random decimation – see Sec. 4). The fractals obtained after the dilution contained a mean number of monomers of $\langle N \rangle = 30$ (for the control, $\langle N \rangle = 15$).

(ii) Self-avoiding random walks [18] were generated with fixed length and random step direction using a trial-and-error method. Monomers were placed at nodes and taken to be hard spheres of diameter equal to the step. If any step brought about an intersection of spheres, then such a step was rejected and repeated. A 20-fold dilution ($\beta = 0.05$) was performed. The clusters thus obtained contain a mean number of monomer of $\langle N \rangle = 25$. Their fractal dimension $D = 1.8$ found with the use of Eq. (1) agreed reasonably with the published value of $D = 1.7$ [18].

(iii) The Witten-Sander clusters [19] were built by the simulation of a diffusion-controlled aggregation of monomers on a cubic lattice and were 20-fold diluted ($\beta = 0.05$) to obtain $\langle N \rangle = 25$. The fractal dimension was found as $D = 2.5$, in close agreement with the known value of $D = 2.51$ [19].

As the control, a gas of monomers ($D = 3$) was also used. The dilution for cases (ii) and (iii) above was not very high, nor was the number of monomers in the clusters studied very large due to limitations of existing computing facilities. However, as will be shown below, even for such fractals the polarizability per a monomer γ does not depend significantly on the number of monomers.

The polarizability was computed from (12) by diagonalization of the matrix (4) using the Jacobi method [20]. The principal advantage of this approach is that the relations (13), (14) and (18) are met exactly. Its merits as a computation method are due to the fact that upon performing the diagonalization once, one finds the solution for any X and δ by simple summation. For the sake of control we also used a direct solution of the original system by the Gauss and square-root methods. For all cases the three indicated

methods agreed perfectly. The calculated individual polarizabilities were averaged over a large ensemble of fractals (75 clusters for $\langle N \rangle = 30$ and $\langle N \rangle = 25$, or 150 for $\langle N \rangle = 15$). The statistical error achieved was less than one percent of the maximum magnitude.

The major portion of our results will be demonstrated for the random-walk fractals, since in this case the theoretical values of the parameters are exactly known: $D = 2$ and $R_0 = R_1(6\beta)^{-1/2}$, where R_1 is the lattice period. Besides, such fractals can easily be generated and diluted, and thus not limited by computer capabilities. Therefore, a high degree of dilution ($\beta = 0.001$) can be achieved. Other types of fractals will also be considered, but only to examine dependences on D for the cases indicated explicitly.

The linear polarizability χ as a function of X is shown in Fig. 1 for different values of the parameter δ. One can see that the absorption contour $\mathrm{Im}\chi(X)$ has the form of a wide peak, with its width decreasing and height growing with increase of $Q = 1/\delta$. In accord with the results obtained, the dependence on δ for $|X| \gg \delta$ is weak. The obvious increase of statistical noise with diminishing δ is in agreement with Eq. (18) (the optical theorem). The width of the fractal absorption contour greatly exceeds that of an individual monomer ($\sim \delta$). The qualitative behavior of the polarizability for fractals of the other types studied is close to that shown in Fig. 1.

The absorption contours calculated for the fractals with significantly different mean numbers of monomers are shown in Fig. 2. It can be seen that for $\langle N \rangle > 15$ the dependence of χ on $\langle N \rangle$ has practically leveled off. This behavior supports the idea of using a characteristic such as the polarizability of a fractal per one monomer.

The fundamental prediction of the theory is the universal scaling behavior of the absorption (30) and the eigenstate density (40) in the collective region (35). This prediction is tested in Fig. 3. From this figure it follows that the absorption and density of states, in fact, obey Eq. (40) with good accuracy expected from an estimate obtained with the aid of Eq. (18).

The values of the optical spectral dimension found from Fig. 3 and similar data for the other types of fractals together with corresponding statistical uncertainties and values of D are shown in Table 1. It follows from this table that for fractals of a given

type, the indices of $\mathrm{Im}\chi(X)$ and $\mathrm{re}\chi(X)$ differ by less than two standard deviations, which is statistically insignificant. For all the cases, the magnitudes of d_ρ deviate significantly (on the level of five to twenty standard deviations) from the limiting values of $d_\rho = 1$ and and $d_\rho = 0$. The fact that $d_\rho = 1$ is evident directly from Figs. 1 and 2: if $d_\rho = 1$ then the central portion of the absorption contour would be not a peak but constant, which is clearly not the case. We note the absence of a significant correlation between d_ρ and D. For the trivial ($D = 3$) fractal studied, no pronounced scaling behavior is observed, in correspondence with the theory (see, in particular, the collectivity criterion (35)).

It is shown above that for diluted fractals, the wings of the absorption contour ($|X| \gtrsim 1$) should correctly (in the leading order in $|X|$) be reproduced by the binary approximation. Regular computation of $\chi(X)$ in the framework of this approximation was carried out according to the following procedure. For each monomer the closest neighbor was found, the quantity $\chi_{zz}^{(p)}(X)$ (42) was calculated, then averaging over all the monomers of the given cluster and the ensemble of clusters was carried out. The absorption found this way is compared in Fig. 4 with the result computed by the Monte Carlo method using the accurate formula (12) ($\delta = 0.33$) One can see that the agreement is, indeed, very good for $|X| > 2$. Even at the center of the contour the differences are not large, which is due to the fact that δ is not very large. In fact, the maximal radius of coherence for the above indicated dimensions, which can be estimated from (33) by substituting $|X| = \delta$, is $L_X \approx 2$, and this does not strongly exceed $R_0 = 1$. Thus, in this case the universal scaling behavior is not developed, even at the center of the contour, as a consequence of the violation of the condition (36).

In Fig. 4 the result is also presented (dashed curve) for a calculation of the estimate provided by the analytic formula (48). Its agreement with the result obtained from (12) in the wings of contour is reasonably good, although at the center it is worse than the regular binary approximation (dotted curve).

Similar data for $\delta = 0.03$ are shown in Fig. 5. Comparing this with Fig. 4, we conclude that the agreement between the three shown curves at the wings ($|X| > 2$) remains good. However, the central peak can no longer be described by any version of the binary approximation. This peak is symmetric, whereas the binary approximation yields

an asymmetric contour, and given that it is scalable (cf. Fig. 3), it is certainly collective.

Lastly, let us describe one of the most important properties of fractals, namely the existence of high local fields which can significantly exceed the external field. Such fluctuations bring about giant enhancement of the Raman and nonlinear parametric scattering of light from fractals, which has earlier been predicted based on the use of the binary approximation [8-11].

From the optical theorem (20) and Eqs. (40) and (46), for the enhancement factor of the local field acting upon a monomer, one can derive the estimates, which are asymptotically exact in the sense of the dependence on $|X|$ and δ in the collective region as

$$G \equiv \frac{\langle |\mathbf{E}_i|^2 \rangle}{|\mathbf{E}^{(0)}|^2} \sim \frac{1}{\delta}|X|^{d_o+1} \quad . \tag{51}$$

and for the diluted fractals in the binary region as

$$G \sim \frac{1}{\delta}|X|^{1-D/3} \quad . \tag{52}$$

We recall that the existence of a nonvanishing collective region is the property of nontrivial fractals. From the estimates (51) and (52), it follows that for nontrivial fractals in both the collective and binary regions, the fluctuations of the local field are strong: the dispersion of this field is proportional to a large parameter, the Q-factor of an isolated monomer (36).

The enhancement factor G as a function of X for the fractals of the three types studied is shown in Fig. 6. In accord with the above presented theory, the curves in their central part, corresponding to the collective region, are symmetric and are described by the scaling formula (51). The factor G increases with $|X|$, in agreement with (51) and (52), reaching values on the order of 10 (for $\delta = 0.1$) at the wings. We point out that there exists no pronounced correlation between G and the fractal type, in accord with the fact that nearly the same values of d_o are found above for different fractals (see Table 1).

In such a way, the present theory and numerical simulation results show that large fluctuations of the local fields, and hence giant enhancement of the Raman and parametric scattering, are not a result of using the binary approximation in [8-11] but the general property of nontrivial fractals, whose monomers possess high-Q optical resonances.

6. Concluding discussion

In this paper we described the optical (dipole) polarizability of fractal clusters. The principal question investigated is how the self-similarity of a nontrivial fractal in coordinate space is transferred to the self-similarity of its dipole excitations and optical polarizability. Our approach to the problem of the fractal optical properties consists in using the exact relations (sum rules for the polarizability obtained in Sec. 2) and consequences of the scaling. The scaling of the optical polarizability is first introduced as an assumption, then the scale-invariant theory is developed (Sec. 3). Finally, the results of the theory determine conditions of the scaling and confirm its validity. Namely, the scaling behavior is shown to exist in the center of the absorption contour of the fractal. The wings of this contour are described in the binary approximation (Sec. 4). The numerical simulation results (Sec. 5) confirm the analytical theory. The new index, optical spectral dimension, governing the scaling of optical absorption and, also, the dispersion law and the density of eigenstates of the dipole excitations, is numerically determined for three types of the fractals studied.

Let us discuss the principal points of the present theory and main results obtained in some more detail, starting with the basic system of equation (3). The interaction between constituents of the fractal (called monomers) is described by the dipole-dipole interaction (see (3)). In the scaling region, where the polarizabilities are determined by the interaction on distances much greater than the monomer size, the dipole-dipole interaction is universal and asymptotically exact.

Instead of frequency ω, we have introduced the new variable X (7), in terms of which the problem of the optical responses is expressed in the universal way. The analytical properties and sum rules for the polarizability as a function of X differ from those with ω as the variable. To develop the theory, we introduce the decomposition (see Eq. (12)) of the polarizability in the terms of the exact eigenvectors and eigenvalues of the interaction operator. Using the analytical behavior of the polarizability given by this decomposition, tensor properties of the interaction and positiveness of absorption (absence of the population inversion), the exact sum rules (14),(18) and (23) are established.

At this point, the assumption about existence of the scaling behavior (see Eqs.

(29) and (30)) is invoked. Using the above indicated sum rules and the requirement of the scale invariance, the forms of the fractal absorption (30) and density of the eigenstates (40) are established. It is rigorously shown that both these quantities scale with the same index, which is denoted as $d_o - 1$, where d_o is called the optical spectral dimension. The index d_o is similar, though it differs in magnitude from the fracton spectral dimension $\bar{\bar{d}}$ introduced by Alexander and Orbach [16] (cf.: both these indices determine the density of eigenstates in the same manner, but $d_o \in [0, 1]$, while normally $\bar{\bar{d}} \in [1, 3]$).

To make the theory closed, using the found scaling properties (32) of X, we derive the dispersion law for the fractal dipole excitations (34). For this derivation, we have formally invoked Alexander's strong localization hypothesis that there exists a single coherence radius of the collective excitation. However, the validity of this hypothesis for the present problem follows directly from the assumption of scaling: only one quantity with the dimension of length, which obeys the required scaling properties, can be constructed from all parameters of the problem. With the aid of the dispersion law obtained, the condition of the scaling for the fractal absorption is found (Eqs. (35) and (36)). It is shown that this condition can be physically met for the realistic model of the monomer as a macroscopic sphere. Thus, the existence of the scaling is established in the self-consistent manner. More exactly, the scaling assumption is shown to be non-self-contradictive.

At the wings of the absorption contour ($|X| \gtrsim 1$), the fractal excitations are primarily two-particle, and the binary approximation [8,9] is applicable. These excitations are sensitive to small-scale details of fractal structure and, therefore, cannot be described in a universal form. In connection with this, a model for a diluted fractal is introduced which is characterized by an extremely simple structure at small distances, completely defined by the pair correlation (1), and thus by the Haussdorff dimension D. Though in the binary region there is no scaling in the general case (cf. Eq.(44)), the far wings of absorption (see Eq. (46)) scales with the index $1 + D/3$, which is completely different from the index $do - 1$ in the collective region (35). We should emphasize that the behavior in the binary region is not universal and is characteristic of the diluted fractals (for other fractals, there may be no scaling at all). The scaling behavior in the collective region (35) is completely universal and is the same for the diluted fractals as for the initial (before

(dilution) ones.

To complement the theory, we have carried out a numerical solution of the basic equations, including averaging over an ensemble of fractals. The polarizabilities of fractals over a wide spectral range have been computed (see Fig. 1). The central portions of the absorption contour and the eigenstate density obey scaling with the accuracy, which is expected from the calculation of dispersion (with the use of Eqs. (18),(19) and Fig. 6), thus confirming the theory. The values of the index d_o are found for the three types of the fractals studied (see Table 1). The indices for a given fractal found from the absorption and eigenstate density do not deviate significantly, in accord with Eq. (40). The values found for d_o differ strongly (on the level of many standard deviations) from the limiting values $d_o = 0, 1$. Hence, the optical spectral dimension d_o proves to be a new nontrivial index.

Lastly, fluctuations of the local fields (dipole momenta) have been computed (Fig. 6). These fluctuations proves to be large, much greater than the mean values of the corresponding quantities. This result is in accord with our understanding of the optical properties of fractals as determined by strong fluctuations. The scaling law and magnitude of the fluctuations in the collective region are universal in the sense that they are defined by d_o and do not depend on the small-scale structure of the fractal. The large fluctuations, in accord with [8-11], should bring about giant enhancement of the Raman and nonlinear parametric scattering of light from fractal clusters, which will be considered elsewhere.

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Table 1

The spectral dimensions d_o and estimates of their statistical uncertainty (on the level of one standard deviation) found from the absorption contour $\mathrm{Im}\,\chi(X)$ and eigenstate density $\nu(X)$ for different fractals as indicated. The values of D are given for convenience.

Fractal type:	Self-avoiding walks	Random walks	Witten-Sander's clusters
d_o from absorption contour:	0.43 ± 0.03	0.38 ± 0.03	0.49 ± 0.02
d_o from eigenstate density:	0.54 ± 0.08	0.33 ± 0.05	0.51 ± 0.06
D found:	1.8	2.0	2.5

Fig. 1. Fractal polarizability ($D = 2$, $N = 30$) for different parameters.

Fig. 2. Fractal absorption ($D = 2$) for different values of the mean number of monomers.

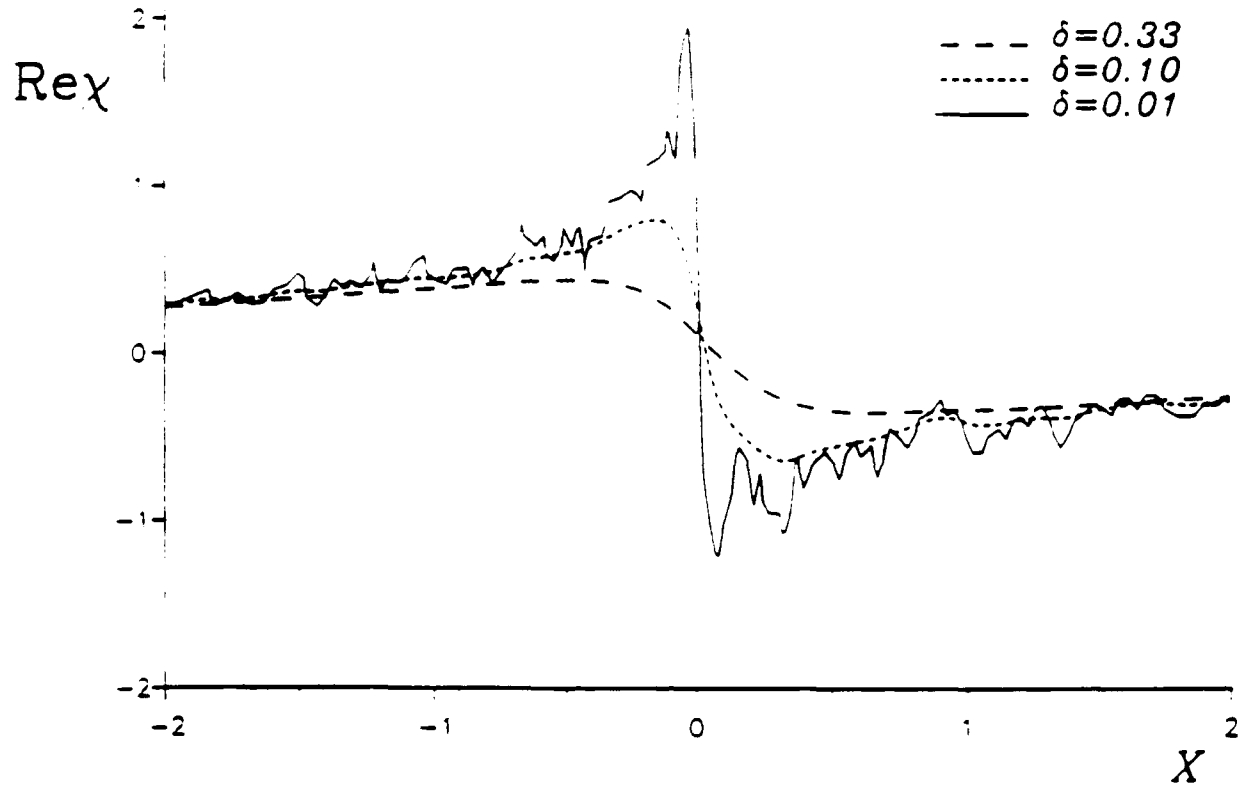
Fig. 3. Fractal absorption as a function of X (for $X > 0$) on a double logarithmic scale: 1 - found from the exact formula (12); 2 - expressed via the state density according to (40). The corresponding lines are obtained by linear regression. The values of the optical dimension obtained see in Table 1.

Fig. 4. Fractal absorption at $\delta = 0.33$: 1 - calculated from the exact formula (12); 2 - in the binary approximation using (42) and numerical averaging; 3 - in the binary approximation from the estimate formula (48).

Fig. 5. Same as Fig. 4 but for $\delta = 0.03$.

Fig. 6. Enhancement factor G (normalized by δ) of the local field intensity plotted against X for the three types of fractals studied ($D = 1.8$, 2.0, 2.5) with $\delta = 0.1$.

Fig. 1


Re χ
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δ=0.10
δ=0.01
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1
0
-1
-2
-2
-1
0
1
2
X

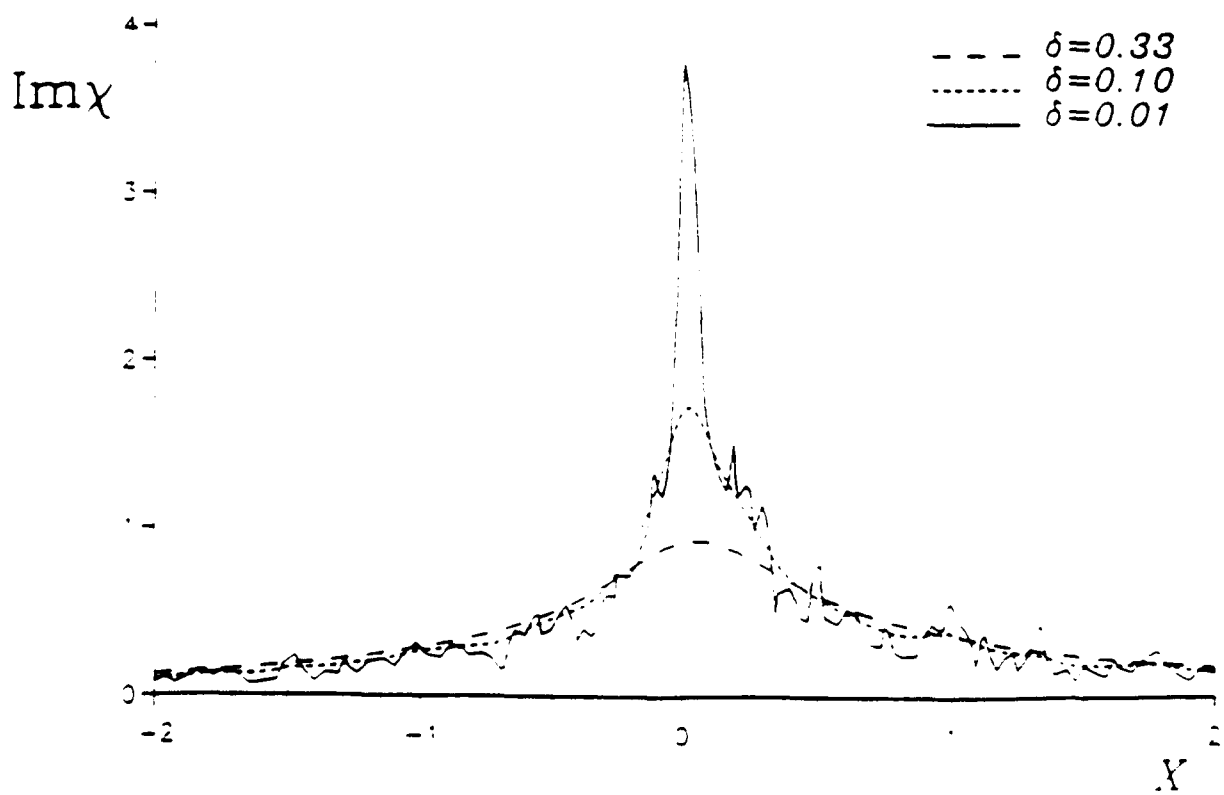

Im χ
δ=0.33
δ=0.10
δ=0.01
4
3
2
1
0
-2
-1
0
1
2
X

Fig. 2

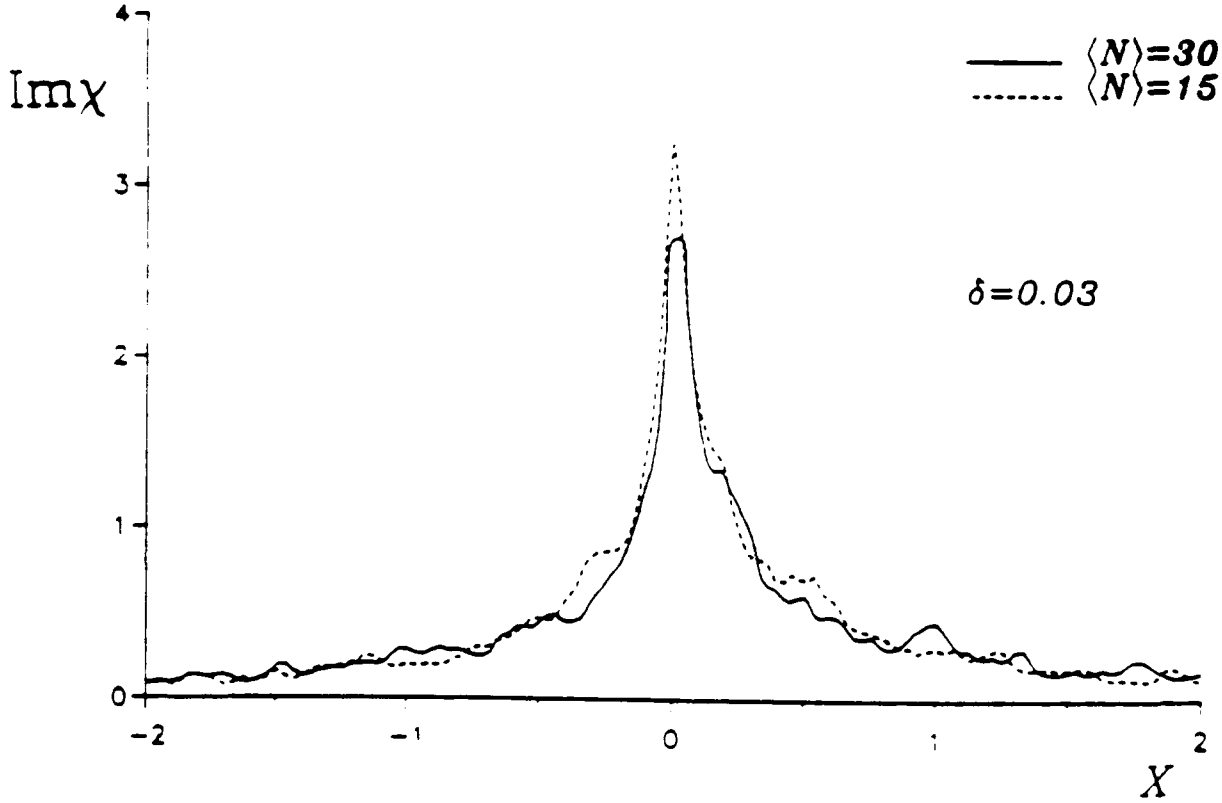


Fig. 3

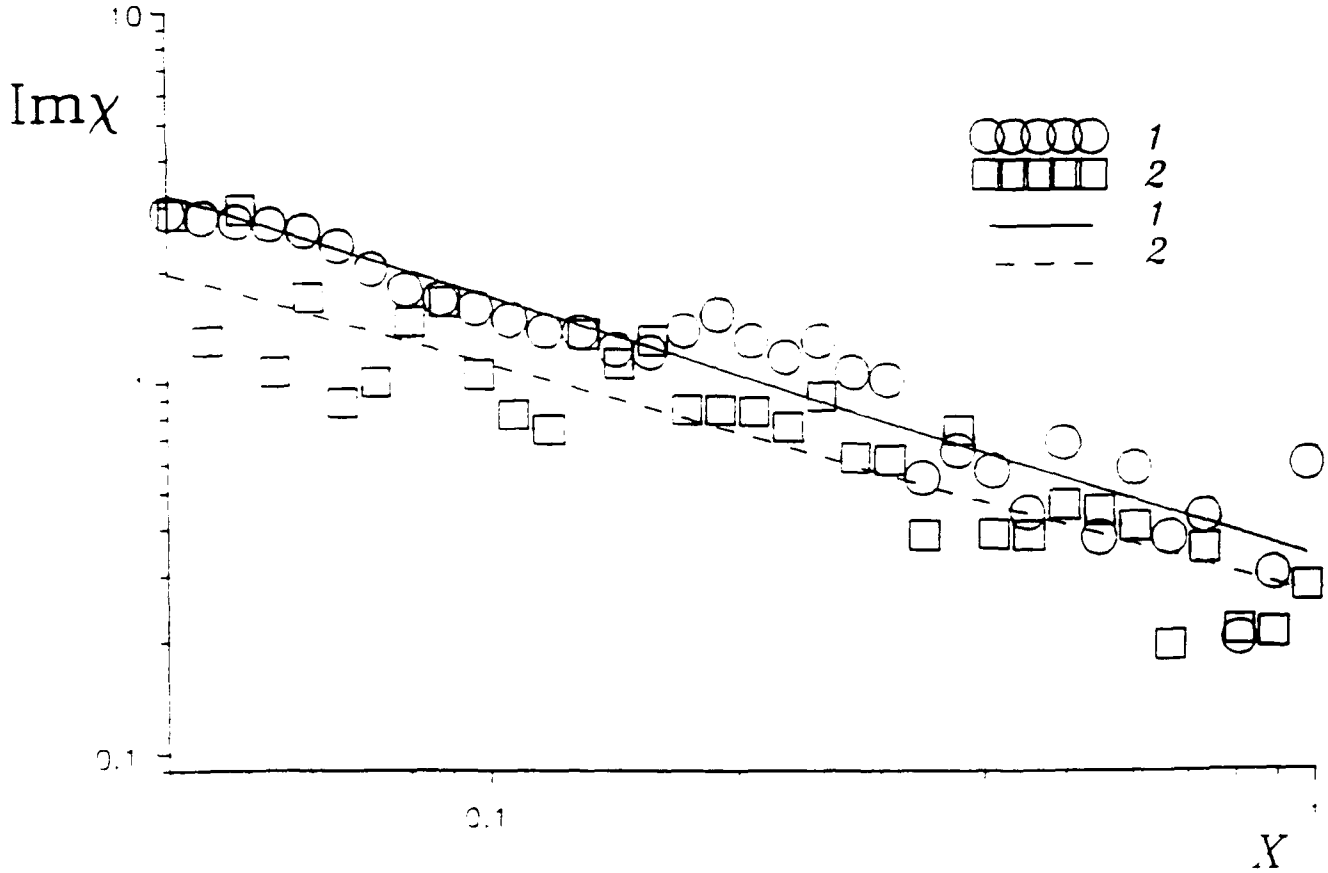


Fig. 4

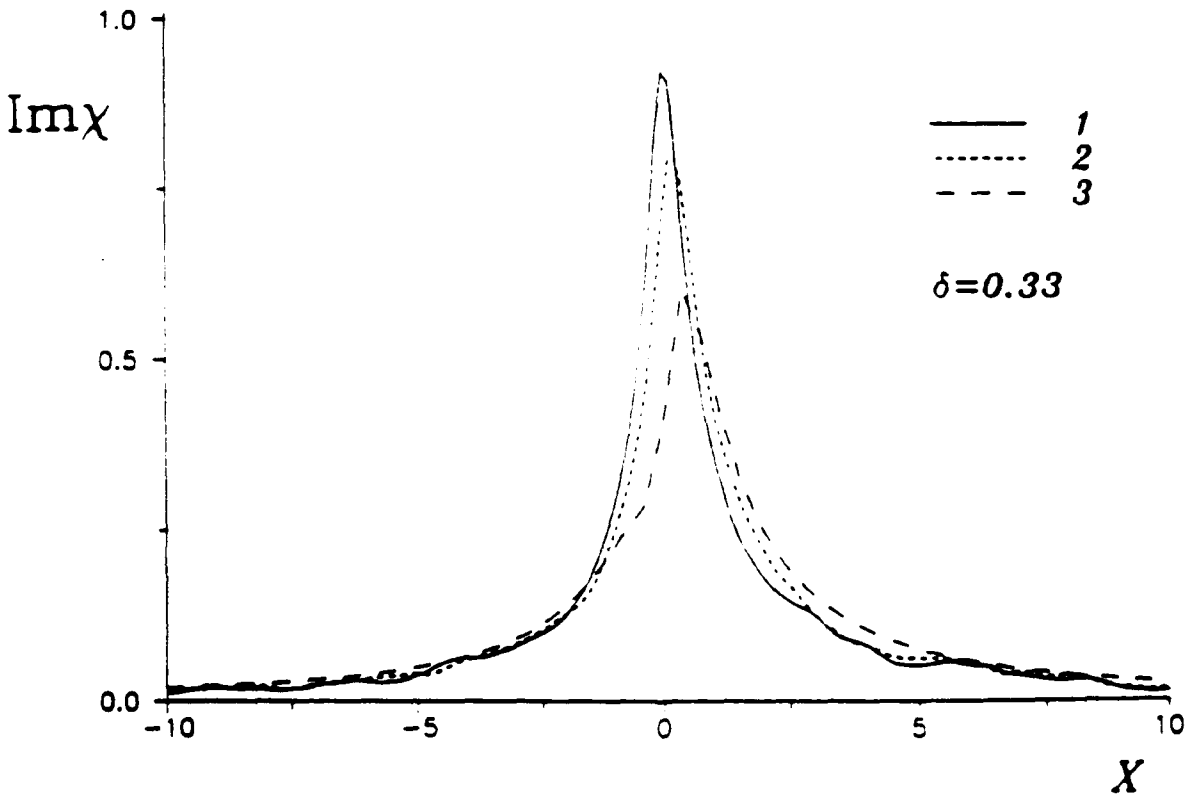


Fig. 5

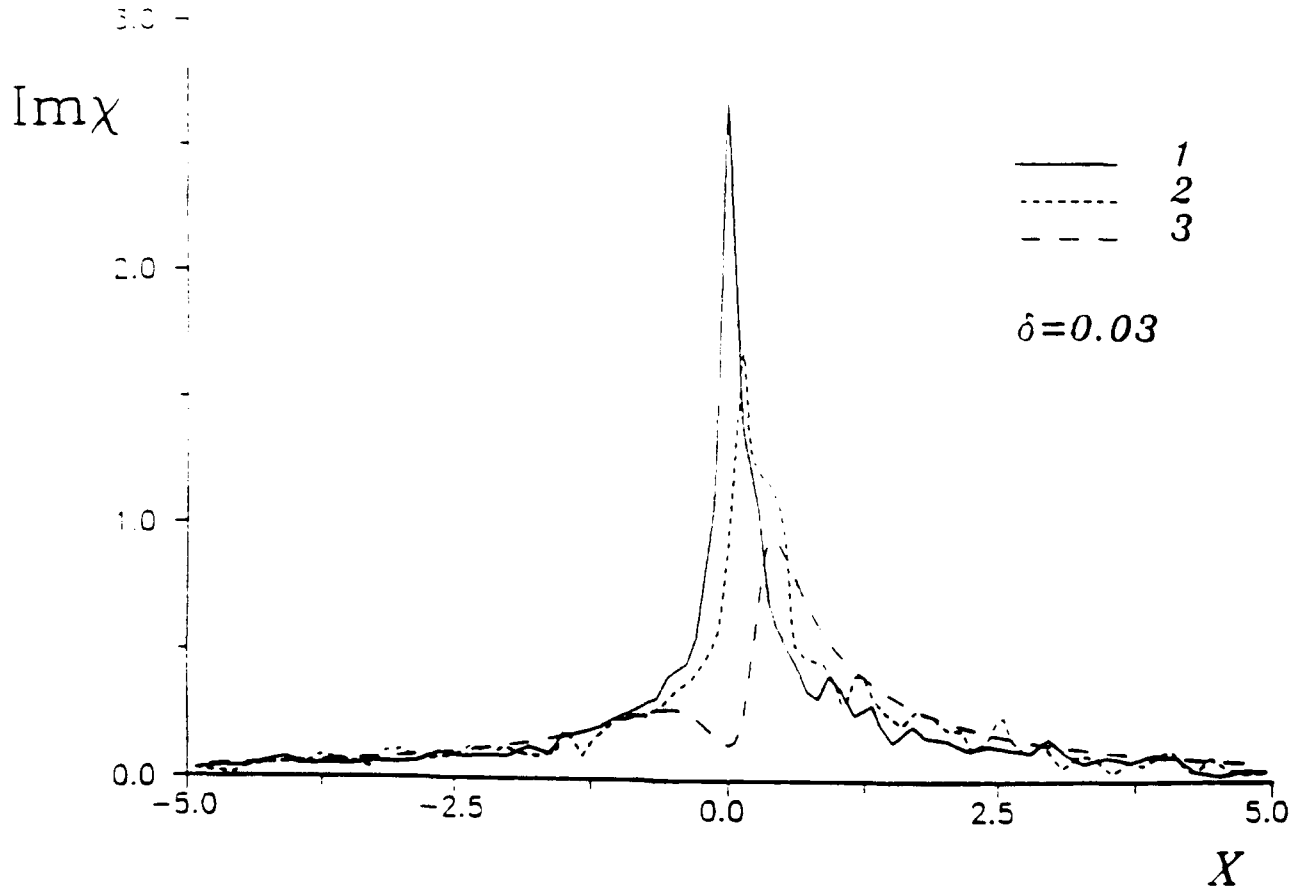
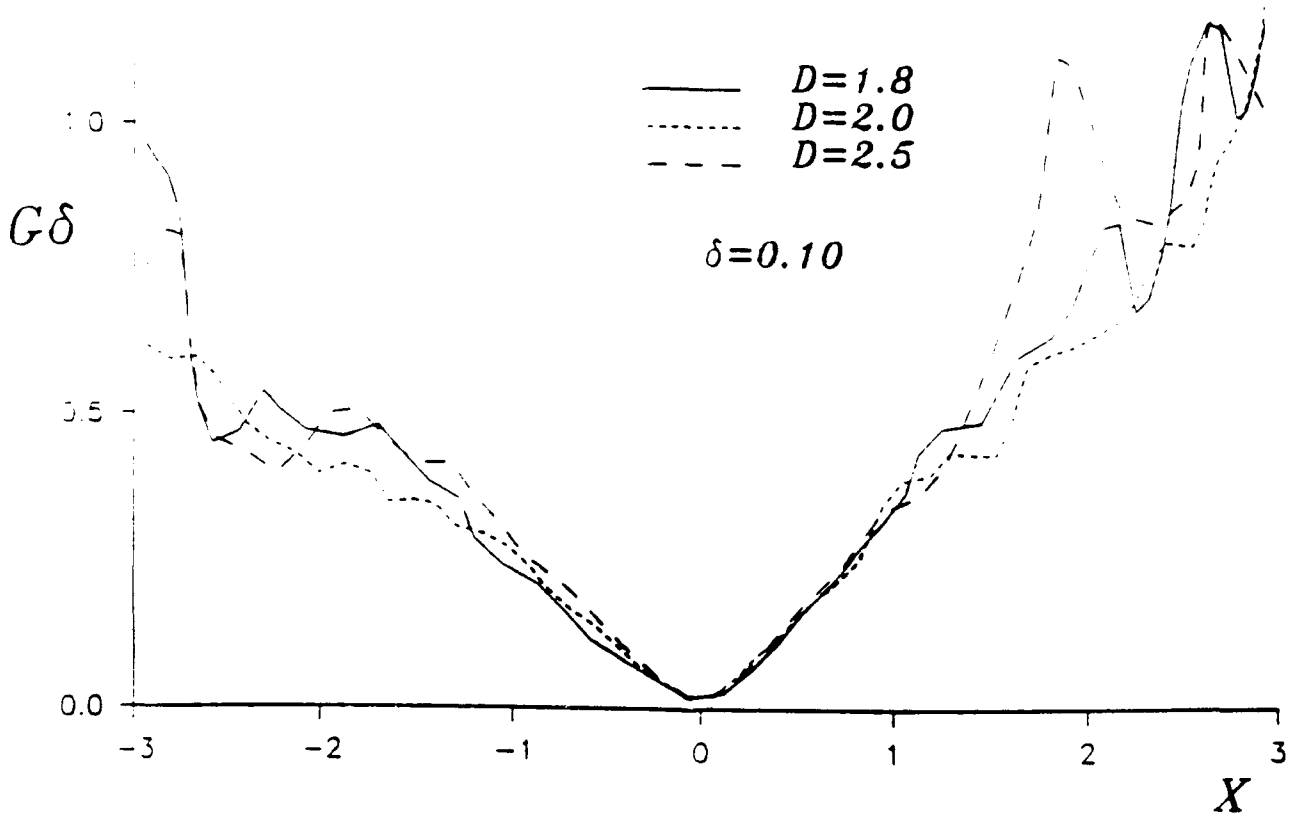


Fig. 6



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