Optical properties of fractals: theory and numerical simulation

V. A. Markel', L. S. Muratov, and M. I. Shtokman

Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR

A scale-invariant theory is derived for the linear optical properties of nontrivial fractals (fractal clusters) in the collective region, i.e., near the center of the absorption band. A model of dilute fractals is introduced and is used to find the shape of the wings of the absorption line in the binary approximation. Strong fluctuations of the local fields in nontrivial fractals are described. These fluctuations cause a giant intensification of the Raman and nonlinear parametric scattering of light by fractals. A theory is proposed for the spectrally selective and polarization-selective threshold photomodification of fractals. The linear spectra and photomodification of fractals have been simulated numerically by the Monte Carlo method. The results of this simulation are compared with the analytic theory. The values of the optical spectral dimension are found for fractals of the types studied.

1. INTRODUCTION

Mandelbrot has introduced the concept of "fractals," which are self-similar (scale-invariant) mathematical objects which have some nontrivial geometric properties.[1,2] Several physical realizations of fractals exist (see, in particular, the review by Zel'dovich and Sokolov[3] and the collection of papers in Ref. 4). In the present paper we will be discussing what we regard as one of the most important realizations: fractal clusters (see, for example, the review by Smirnov[5] regarding the properties of these entities).

By "fractal cluster" we mean a system of interacting material particles, called "monomers," which has the property of scale invariance (in the statistical sense) in an intermediate range of sizes r, namely, $R_0 \ll r \ll R_C$, where R_0 is a characteristic distance between nearest monomers, and R_C is the size of the cluster. For brevity we will call such clusters "fractals." This usage should not lead to any misunderstanding, since we will not be discussing other realizations of fractals here.

Fractals are quite common in nature.[4,5] Examples are a wide class of products of diffusion-controlled aggregation in liquids and gases; particles in colloidal solutions, sols, and gels; soot and smoke particles; and most macromolecules. Rough surfaces, disordered films on surfaces, and porous objects (microfilters and adsorbents, including activated charcoal, porous glasses, aerogels, several heterogeneous catalysts, etc.) also exhibit a fractal structure in an intermediate range of sizes in many cases.

A consequence of the scale invariance of fractals is a power-law dependence of the correlation functions on the coordinates. In particular, the binary density–density correlation function in the intermediate asymptotic region can be written in the 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 dimension" (more precisely, the "external Hausdorff dimension") of the fractal [expression (1) also fixes the definition of the constant R_0]. A consequence of (1) is that the number of monomers in the fractal, N, and the average density of the fractal, $\bar{p}$, have the behavior

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

A fractal is called "nontrivial" if D is smaller than the dimensionality of the space in which the fractal is embedded (i.e., if $D < 3$, in the case at hand). It can be seen from (2) that the density of a nontrivial fractal asymptotically (at $R_C \gg R_0$) approaches zero. This property, combined with the strong binary correlation in (1)—which results in a substantial binary interaction between monomers—is responsible for the major role of fluctuations in a nontrivial fractal.

Among the physical properties of fractals, the optical properties have been studied least (in the collection of papers in Ref. 4 on the physics of fractals, for example, there are no corresponding reviews). The linear optical (dipole) polarizabilities of fractals have been the subject of several theoretical papers.[6–9] Two of these studies,[6,7] which were based on various modifications of the mean field method, did not deal fully with strong fluctuations (in the density, local fields, etc.) in a fractal. In two papers coauthored by one of the present authors,[8,9] the fluctuations in the nearest neighborhood of a monomer played a governing role. Those papers, however, used the binary approximation: The interaction of a monomer with only its nearest neighbor was considered exactly; the effect of the other monomers was dealt with only by means of a Lorentz field.

If the monomers are high-Q optical resonators (e.g., microscopic particles of noble or other metals having a prominent luster), the strongly fluctuating local fields in a fractal may be significantly stronger than the exciting external field. It is this circumstance which is responsible for the huge Raman scattering by fractals.[8,9] As always, nonlinearities increase the role of fluctuations. This effect is pertinent to the huge size of the nonlinear optical polarizabilities of fractals which was predicted in Refs. 10 and 11.

This prediction has been verified experimentally for phase conjugation in degenerate four-wave parametric processes involving fractal clusters of silver.[12] Recent experiments have revealed[13] a photomodification of such clusters which is selective in terms of the wavelength and polarization of the light. The effects which have been observed are rich in physical content, hold the promise of significant applications, and thus deserve a deeper theoretical study.

Our purpose in the present paper is to derive a theory for the optical properties of fractals, dealing fully with their fluctuation properties, but going beyond the binary approximation. We focus on the scale-invariant theory.

2. BASIC EQUATIONS AND ACCURACY OF THEIR SOLUTIONS

We consider a model fractal of a system of N polarizable monomers with a dipole–dipole interaction at optical frequencies. The monomers are at the points r_j. The dipole moments d_i induced at the various monomers obey the well-known system of equations

$$d_{i\alpha}=\chi_0 E_{i\alpha}^{(0)}-\chi_0 \sum_{j=1;j\neq i}^{N} \frac{\delta_{\alpha\beta}-3n_{\alpha}^{(ij)}n_{\beta}^{(ij)}}{r_{ij}^3}d_{j\beta}, \tag{3}$$

where the Greek letters specify tensor components (a repeated index implies a summation), the Latin letters number the monomers, χ_0 is the dipole polarizability of an individual monomer (which is assumed to be isotropic, so that some unnecessary complications can be avoided), $E_i^{(0)}$ is the electric field of the external (exciting) radiation at the position of monomer i, $r_{ij} = r_i - r_j$, and $n^{(ij)} = r_{ij}/r_{ij}$.

For use below, we will rewrite Eq. (3) as an operator equation. To do this, we introduce the matrix W, with the elements

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

which acts in the $3N$-dimensional space of the vectors $\hat{d}$ and $\hat{E}^{(0)}$ with the components

$$(i\alpha|\hat{d})=d_{i\alpha}, \qquad (i\alpha|\hat{E}^{(0)})=E_{i\alpha}^{(0)}. \tag{5}$$

The basic system of equations then becomes

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

where we have introduced the complex variable z with a real part—X and an imaginary part—δ:

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

To find the accuracy properties of the solution, we make use of an expansion in the eigenfunctions of the operator W (the same expansion turns out to be the most powerful computational approach in a numerical simulation, as we will see below). The symmetric real matrix W is diagonalized by the orthogonal transformation

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

where $\text{diag}(w_n)$ is a diagonal real matrix of the eigenvalues w_n, and the superscript T means transposition. The formal solution of our basic equation (6) has the obvious form

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

Expressing the unknown values $d_{i\alpha}$ in terms of the eigenvalues w_n and the components of the corresponding eigenvectors $(n|U|i\alpha)$ on this basis, we find

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

We will be discussing the case in which the cluster dimensions R_C are much smaller than the wavelength of the radiation, λ. The external field $E_j^{(0)}$ can then be assumed to be uniform (independent of j). In this case the dipole moment induced at monomer i can be expressed in terms of the corresponding linear polarizability $\chi_{\alpha\beta}^{(i)}$ in the following way:

$$d_{i\alpha}=\chi_{\alpha\beta}^{(i)}E_{\beta}^{(0)}. \tag{11}$$

It is clear from (10) that the following expression holds for $\chi_{\alpha\beta}^{(i)}$:

$$\chi_{\alpha\beta}^{(i)}=\sum_{j,n} (n|U|i\alpha)(n|U|j\beta)(z+w_n)^{-1}. \tag{12}$$

As an analytic function of z, the polarizability $\chi_{\alpha\beta}^{(i)}$ is different from that in the expression in terms of the frequency ω. In particular, this function is not "real," in the sense that we have $\chi_{\alpha\beta}^{(i)*}(X)\neq\chi_{\alpha\beta}^{(i)}(-X)$. Nevertheless, the standard Kramers-Kronig formula holds here in terms of the variable X, as can be verified directly on the basis of solution (12):

$$\text{Re}\,\chi_{\alpha\beta}^{(i)}(X)=\frac{1}{\pi}\int_{-\infty}^{\infty} \text{Im}\,\chi_{\alpha\beta}^{(i)}(X')\frac{dX'}{X'-X}. \tag{13}$$

Here we are using the standard notation for an integral which is to be understood in the principal-value sense. We will also derive several other exact relations for $\chi_{\alpha\beta}^{(i)}$ below; these relations are consequences of the real nature of the eigenvalues w_n, the orthogonality of the matrix U in (8), and the positivity of δ in (7), which reflects the normal absorption in the monomer (we are assuming that there is no population inversion).

Some exact sum rules also follow from the form of solution (12):

$$\frac{1}{\pi}\int_{-\infty}^{\infty} \text{Im}\,\chi_{\alpha\beta}^{(i)}(X)dX=\delta_{\alpha\beta}, \qquad \int_{-\infty}^{\infty}\text{Re}\,\chi_{\alpha\beta}^{(i)}(X)dX=0. \tag{14}$$

We see, in particular, that the integral absorption[1] over X is conserved: It is the same as that of an isolated monomer.

We can now derive an exact relationship which is an analog of the optical theorem. For this purpose we find from (12), with the help of (8), a quadratic form of the polarizability summed over the monomers:

$$\sum_i \chi_{\gamma\alpha}^{(i)*}\chi_{\gamma\beta}^{(i)}=\sum_{j,j',n} (n|U|j\alpha)(n|U|j'\beta)(z^*+w_n)^{-1}(z+w_n)^{-1}. \tag{15}$$

Carrying out an elementary expansion in simple fractions in (15),

$$(z^*+w_n)^{-1}(z+w_n)^{-1}=-\text{Im}(z+w_n)^{-1}/\text{Im}\,z \tag{16}$$

and introducing the average polarizability of the monomer in a given cluster,

$$\chi_{\alpha\beta}=\frac{1}{N}\sum_i\chi_{\alpha\beta}^{(i)}, \tag{17}$$

we find the exact relationship which we need:

$$\frac{1}{N}\sum_i \chi_{\gamma\alpha}^{(i)*}\chi_{\gamma\beta}^{(i)}=\frac{1}{\delta}\text{Im}\,\chi_{\alpha\beta}. \tag{18}$$

Using (18), we can find the square of the local electric field E_i acting on a monomer, averaged over a cluster. This field determines the enhanced Raman scattering by a cluster and also the photomodification of the cluster (more on this

below). The local field is related to the solution of system (6) in the obvious way:

$$\mathbf{E}_i = \chi_0^{-1} \mathbf{d}_i, \qquad E_{i\alpha} = \chi_0^{-1} \chi_{\alpha\beta}^{(i)} E_\beta^{(0)}. \qquad (19)$$

We assume for definiteness that the exciting field is directed along the z axis. From (18) and (19) we find

$$\frac{1}{N} \sum_i |\mathbf{E}_i|^2 / |\mathbf{E}^{(0)}|^2 = \delta(1 + X^2/\delta^2) \operatorname{Im} \chi_{zz}. \qquad (20)$$

This relationship is reminiscent of the Callen-Welton theorem (or fluctuation dissipation theorem). A result which is the same as (20) aside from the notation has been derived previously in the binary approximation [see Eq. (45) in Ref. 9] and has served as the basis of a theory for the huge Raman scattering of fractals.

We wish to stress that Eqs. (13), (14), (18), and (20), derived above, are exact. They are valid for an arbitrary cluster (fractal or otherwise), without any averaging over realizations or orientations. The only important points are that the cluster consist of monomers between which there is a binary interaction, that these monomers exhibit a linear response to an external field, and that they have a normal absorption (without a population inversion).

We now make use of the symmetry under rotation. After we take an average over the orientations of the cluster as a whole (we denote this procedure by $\langle \ldots \rangle_0$), the polarizability tensor reduces to a scalar:

$$\langle \chi_{\alpha\beta} \rangle_0 = \chi \delta_{\alpha\beta}, \qquad \chi = \frac{1}{3} \chi_{\alpha\alpha}. \qquad (21)$$

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

$$\int_{-\infty}^{\infty} X \operatorname{Im} \chi(X) \, dX = \int_{-\infty}^{\infty} \operatorname{Re} \chi(X) \, dX$$
$$+ \frac{1}{3N} \sum_{i,j,n} (n|U|i\alpha) w_n (n|U|j\alpha). \qquad (22)$$

According to (14), the first term on the right side of (22) is zero. The second term can be put in the form $\sum_{i,j} (i\alpha|W|j\alpha)$ with the help of (8). Making use of the fact that interaction tensor (4) vanishes when we convolve in terms of the indices α and β, we reach the conclusion that the second term is also zero. We thus find the exact sum rule

$$\int_{-\infty}^{\infty} X \operatorname{Im} \chi(X) \, dX = 0, \qquad (23)$$

which means that the center of the absorption of the cluster is at the point $X = 0$.

The equations derived above are universal because (in particular) of the use of the variables X, δ, which are natural for this particular problem. These variables of course depend on the frequency ω, but not in a universal way. Although to do so is not necessary for the theory, it is useful to think of the ω dependence of monomers which have one prominent resonance, with a frequency ω_0, a transition dipole element d_{12}, and a relaxation rate (homogeneous width) Γ. We then have

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

where $\Omega = \omega - \omega_0$. We see that in this case X is the relative frequency deviation, while δ is the relative width of the resonance in an isolated monomer. The first of relations (14), combined with (23), results in the satisfaction of the well-known dipole sum rule for the polarizability.

3. POLARIZABILITY OF A FRACTAL IN THE COLLECTIVE REGION

In deriving the exact results above, we did not make use of the specific properties of fractals. Nevertheless, we will make substantial use of these results below in our description of the collective polarizability of a fractal.

The self-similarity which is a fundamental property of a fractal entity means that the fractal reproduces itself upon a variation in a spatial scale. Since a fractal is in general a random entity, this reproduction occurs in a statistical sense. A variation in the length scale implies a simultaneous change in the scale of the eigenvalues w_n and of the variable X. It follows that the intermediate asymptotic behavior of observable quantities as functions of w and X must be a power-law behavior.

We will first describe $\operatorname{Im} \chi(X)$ (the absorption), and then we will use (13) to examine $\operatorname{Re} \chi(X)$. Using (7), we see from (12) that a universal power-law behavior of $\operatorname{Im} \chi(X)$ can prevail only if $|X| \gg \delta$, in which case we can write

$$\operatorname{Im} \chi(X) = \left\langle \frac{\pi}{3N} \sum_{i,j,n} (n|U|i\alpha)(n|U|j\alpha) \delta(X - w_n) \right\rangle, \qquad (25)$$

where $\langle \ldots \rangle$ means an average over the ensemble of fractals. It follows in particular that the absorption determines the eigenstates for which $w_n = X$. These states are characterized by a coherence length L_X, and they are "collective" (i.e., delocalized among many monomers) in the intermediate asymptotic region:

$$R_0 \ll L_X \ll R_c. \qquad (26)$$

We will estimate the length L_X below on the basis of the transformation properties of a quantity upon a change in spatial scale.

In the collective region, (26), the absorption profile $\operatorname{Im} \chi(X)$ must be a power law. The eigenvalue problem of the operator W does not have an exact symmetry under a change in the sign of w_n. Consequently, there is no parity with respect to X. We thus cannot immediately rule out the possibility that the indices will be different for $X > 0$ and $X < 0$. However, sum rule (23), combined with the scale invariance of the system, guarantees that these indices are identical for the absorption and that furthermore the corresponding coefficients are equal to each other. A symmetric power-law dependence thus holds in region (26) (and at $|X| \gg \delta$). The coefficient of this power-law dependence is determined by R_0 and can be estimated from dimensionality considerations:

$$\operatorname{Im} \chi(X) \sim R_0^3 (R_0^3 |X|)^{d_o - 1}, \qquad (27)$$

where d_o is some index which we will call the "optical spectral dimension." From the requirement of convergence in the first of sum rules (14) we conclude $d_o > 0$. We wish to stress that the symmetry of the absorption with respect to X is an inexact property, prevailing only in the collective re-

gion (compare with the discussion below of the binary region).

Let us examine the transformation properties of quantities upon a change in the minimum scale R_0 (a renormalization transformation). We assume an initial value $R_0 = 1$ (i.e., we are expressing lengths in units of R_0). An increase in R_0 implies a coarsening of the spatial resolution with which we are looking at the fractal. This increase can be achieved in the following way:[14] In the fractal we distinguish fluctuations with a length scale $l \gg 1$. We combine the monomers involved in such a fluctuation into a new (renormalized) monomer. It follows from the property of self-similarity that the resulting cluster is a fractal, with the same critical exponents and with a value $R_0 \sim l$. The renormalization transformation conserves the absorption profile in (27), with a new parameter X [under the condition that the condition for a collective nature, (26), remains satisfied].

Let us determine the X renormalization law and the coherence length L_X. The total absorption of the fractal should be conserved under a renormalization transformation. This requirement reduces to the following requirement when we make use of (2) and (27):

$$\left(\frac{R_c}{R_0}\right)^D R_0{}^3 (R_0{}^3 |X|)^{d_o-1} \propto 1. \tag{28}$$

It is clear that X transforms in accordance with

$$|X| \propto R_0^{-(3d_o-D)/(d_o-1)}. \tag{29}$$

In the case of a trivial fractal ($D = 3$) we would have $|X| \propto R_0^{-3}$, regardless of the exponent d_o; i.e., we would have $\mathrm{Re}\,\chi_0 \propto R_0^3$. The effect is to reproduce the proportionality of the polarizability of the composite monomer to its volume.

For greatly disordered systems such as fractals, Alexander[14] has suggested that collective states with a certain value of the frequency (the role of the frequency is being played here by the variable X) are characterized by a single coherence length, which is simultaneously the wavelength and the localization radius of the excitation. This property is a consequence of the self-similarity of the fractal and its collective states. When we go over to the trivial (three-dimensional) system, the generality of this property is obviously destroyed: The excitation localization radius may be far larger than the wavelength (the case of Anderson localization).

Using (29), we can construct from X and R_0 the sole invariant of the renormalization transformation, which has the dimensionality of a length. According to the discussion above, this invariant must be equal to L_X:

$$L_X \sim R_0 (R_0{}^3 |X|)^{(d_o-1)/(3-D)} \tag{30}$$

In the $D = 3$ case, expression (30) has a singularity (not surprisingly, in view of the discussion above). This singularity nevertheless has a simple physical meaning. To demonstrate it, we consider the inverse of relationship (30), which is the dispersion relation for excitations of the fractal:

$$|X| \propto R_0^{-3} (L_X/R_0)^{(3-D)/(d_o-1)}. \tag{31}$$

For $D = 3$ we see that $|X|$ does not depend on L_X (the excitation wavelength). This situation corresponds precisely to

the dispersion relation (the presence of a spectral gap) for long-wavelength surface plasmons, into which dipole excitations of a fractal convert in the limit $D \to 3$.

With a normal dispersion relation, the "frequency" X should fall off with decreasing wave vector, i.e., with increasing L_X. Using (31), we see that this requirement leads to a limitation $d_o < 1$, which we will adopt below (this limitation is also supported by the numerical calculations, as we will see below). We will not make any further use of the renormalization transformation; we go back to our initial value $R_0 = 1$. Condition (26), which defines the collective region, then takes the following form, when we make use of (30):

$$N^{-(3/D-1)/(1-d_o)} \ll |X| \ll 1. \tag{32}$$

In other words, this region stretches out in the direction of small $|X|$ and thus occupies the center of the absorption band [cf. (24)]. The necessary condition for a power-law behavior, $|X| \gg \delta$, is evidently compatible with (32) only if the quality factor Q of the resonance at the monomer is high:

$$Q = 1/\delta \gg 1. \tag{33}$$

It follows from (32) that as the fractal approaches the state of a trivial fractal ($D \to 3$) the region of collective behavior in terms of X degenerates to a point, which is the point corresponding to the frequency of the surface plasmons. The finite width of the zone of collective states of the fractal is thus due entirely to its nontrivial nature.

We now seek the relationship between the absorption of the fractal, $\mathrm{Im}\,\chi(X)$, and the density of its eigenstates, $\nu(X)$, which we introduce in the usual way:

$$\nu(X) = \left\langle \frac{1}{N} \sum_n \delta(X - w_n) \right\rangle. \tag{34}$$

This density is normalized to a single monomer. Its mean value is zero because the trace of interaction tensor (4) is zero:

$$\int_{-\infty}^{\infty} \nu(X)\,dX = 3, \qquad \int_{-\infty}^{\infty} X\nu(X)\,dX = \mathrm{Sp}(W) = 0. \tag{35}$$

The value of the first integral in (35) is determined by the vector nature of the interaction between monomers.

Singling out the terms with $i = j$ in expression (25), making use of the orthogonality of matrix U in (8), and comparing the result with (34), we find

$$\mathrm{Im}\,\chi(X) = \frac{\pi}{3} \nu(X)$$
$$+ \left\langle \frac{\pi}{3N} \sum_{i \neq j, n} (n|U|i\alpha)(n|U|j\alpha) \delta(X - w_n) \right\rangle. \tag{36}$$

It follows from (35) that the first sum in this expression satisfies both sum rules for $\mathrm{Im}\,\chi$, (14) and (23). Correspondingly, the zeroth and first moments in terms of X of the second term in (36) are exactly zero. Since this term must also have a power-law dependence on X under condition (32), it is zero. In the collective region we thus have

$$\mathrm{Im}\,\chi(X) \approx \frac{\pi}{3} \nu(X) \propto |X|^{d_o-1}. \tag{37}$$

Relationship (37) has a transparent physical meaning. In a greatly disordered system such as a fractal, all the col-

lective eigenstates contribute to the optical absorption with equal weights. We wish to stress that relationship (37) between the absorption and the density of states is not exact. For example, it is easy to see that at the wings of the absorption profile described in the binary approximation (as discussed below) this relationship is violated. In ordered systems, the sum rules hold, and only a few states of the corresponding symmetry contribute to the absorption.

A power-law behavior of the density of eigenstates of a fractal as a function of the frequency was originally introduced by Alexander and Orbach[15] for a problem involving mechanical vibrations (fractons). The index $\bar{d}$, which corresponds to d_o in (37), was called a spectral (or "fracton") dimension in that earlier study. In general, the value of a spectral dimension is determined by the internal geometry of the fractal. For a vector problem it is also determined by the method by which the fractal is embedded in three-dimensional space.[14] In principle, a spectral dimension may also depend on the potential of the interaction between monomers. In terms of the magnitude of the optical spectral dimension, d_o thus does not necessarily have to be the same as the vibrational spectral dimension $\bar{d}$ in Ref. 15.

A dispersion-relation for oscillations of a fractal was derived in Refs. 14–16 in a variety of ways. In our notation that relation is

$$|X| \propto L_x^{-D/\bar{d}} \qquad (38)$$

and is clearly different from (31). In particular, for a trivial fractal ($D = 3$) relation (38) predicts a gap-free dispersion law which is linear at $\bar{d} = 3$. Analysis of Refs. 14–16 shows that the approach taken there is valid only for excitations of a Goldstone type (acoustic phonons, magnons, and so forth), whose creation and annihilation operators transform in the long-wavelength limit into the generators of symmetry transformations (translations, rotations, and so forth) of the system. The dipole excitations which are pertinent to the problem at hand are not Goldstone excitations, since they transform into surface plasmons, for which there is a gap in the spectrum [see the discussion of Eqs. (31) and (32) above]. For the problem at hand, it is thus dispersion relation (31), rather than (38), which holds.

We conclude this section of the paper with a brief discussion of $\text{Re}\,\chi(X)$. Since the kernel of Hilbert integral transformation (13) does not by itself ensure convergence, both the collective region ($|X| \ll 1$) and the wings of the absorption profile ($|X| \gg 1$) contribute to $\text{Re}\,\chi(X)$. Consequently, $\text{Re}\,\chi(X)$ is not described by a power law, even in region (32). The same conclusion could be reached by working directly from (12).

4. DILUTE FRACTALS IN THE BINARY APPROXIMATION

In the preceding section we examined the collective region, in which the natural excitations of the fractal are determined by the long-range interaction and span many monomers. The observable quantities determined by such excitations are insensitive to the structural details of the fractal, and they are also insensitive to the nature of the short-range interaction of monomers. They are characterized by universal power laws. In the present section of the paper we describe the wings of the spectrum of a fractal (the region ($|X| \gtrsim 1$). In this case the external field interacts with natural excitations of the fractal, which are localized at only a few monomers, according to (32). Consequently, observable quantities are generally sensitive to these details.

Since it is impossible in principle to formulate a universal description of the wings of the fractal spectrum according to the arguments above, we would like to use a model of a fractal with an extremely simple small-scale structure. As such a model we introduce "dilute" fractals, which are constructed by coarsening the structure of the original fractals. Instead of the renormalization transformation described above, we use a simple and efficient coarsening algorithm which we call "dilution" (hence the name of the model). This algorithm can be summarized as follows: Each monomer of the original fractal is either left in place, on a random basis with some probability $\beta \ll 1$, or taken out of the fractal, with a probability of $1 - \beta$. The fractal as a whole is then compressed in size by a factor of $(1/\beta)^{1/D}$. The result is a fractal which has the same critical exponents determining the intermediate asymptotic behavior, and it has the binary correlation function in (1) ($R_0 = 1$). This correlation function is valid to small distances on the order of $r_0 = (\beta)^{1/D} \ll 1$. Obviously, there is no change in the polarizability of the monomer (the variable z) upon dilution, while the overall radius of the fractal decreases by a factor of r_0. In the limit $\beta \to 0$, a repeated dilution would again generate a dilute fractal, differing only in the value of R_C.

Dilution (random decimation) eliminates local structure of the fractal (the connectedness, the distance to the nearest neighbor, the number of nearest neighbors, etc.) without altering the properties at intermediate scales $r_0 \ll r \ll R_C$ and thus without altering the behavior of quantities in the collective region (as discussed above).

To describe the short-range interaction, which is important at $|X| \gtrsim 1$, we adopt the binary approximation, which was used in Refs. 8–11. In other words, we take the interaction of each monomer with its nearest monomer into account exactly, while we ignore the effect of other monomers. The polarizability tensor of a monomer in a pair, $\chi_{\alpha\beta}^{(p)}$, is given by[8]

$$\chi_{\alpha\beta}^{(p)} = (z+r_n^{-3})^{-1}\delta_{\alpha\beta} + [(z-2r_n^{-3})^{-1} - (z+r_n^{-3})^{-1}]n_\alpha n_\beta, \qquad (39)$$

where $\mathbf{r}_n$ is the radius vector of this nearest neighbor, and $\mathbf{n} = \mathbf{r}_n/r_n$. After taking an average over $\mathbf{r}_n$, we find the polarizability of a monomer in the fractal:

$$\chi = \frac{1}{3}\int_0^\infty [2(z+r_n^{-3})^{-1} + (z-2r_n^{-3})^{-1}]p(r_n)dr_n, \qquad (40)$$

where $p(r_n)$ is the distribution with respect to distance to the nearest neighbor. Under the condition $|X| \gg \delta$ we find from (40) [cf. (25)]

$$\text{Im}\,\chi = \frac{\pi}{9}r_n^4[2p(r_n)\theta(X) + 2^{1/3}p(2^{1/3}r_n)\theta(-X)]|_{r_n=|X|^{-1/3}}, \qquad (41)$$

where $\theta(X)$ is unit step function.

Under the condition $|X| \gg 1$, small distances r_n dominate (41). At such distances we have the following asymptotic behavior, which follows from (1) and which is universal for dilute fractals:

$$p(r_n) \approx Dr_n^{D-1}. \qquad (42)$$

Substitution of (42) into (41) leads to an expression for the shape of the remote wing of the absorption profile of a dilute fractal:

$$\operatorname{Im}\chi = \frac{2\pi D}{9}\,|X|^{-(1+D/3)}[\theta(X)+2^{D/3-1}\theta(-X)].\qquad(43)$$

Although this is a power law, the power is greater than 1.0 in absolute value, and the spectrum is asymmetric under a change in the sign of X, in contrast with the behavior in the collective region [cf. (37) and the discussion which follows]. By adding a third monomer to the pair of nearest monomers, we can show that the binary interaction describes the wing shape in (43) correctly on the order of the parameter $1/X^2$.

In the case $r_n \sim 1$, the distribution $p(r_n)$ cannot be found in a universal form, since it is determined not only by (1) but also by higher-order correlation functions. To find a rough estimate, we ignore these higher-order correlations. The number of monomers in a fixed region around a given monomer then obeys a Poisson distribution, and we can write

$$p(r_n) = Dr_n^{D-1}\exp(-r_n^D).\qquad(44)$$

Substitution of (44) into (40) leads to the expression

$$\chi = \frac{1}{3}\left[2S_D(-z)-\frac{1}{2}S_D\!\left(\frac{z}{2}\right)\right],\qquad(45)$$

where the function of a complex variable $S_D(z)$ is defined by the integral representation

$$S_D(z) = -\frac{D}{3}\int_0^\infty y^{-(1+D/3)}\exp(-y^{-D/3})\,\frac{dy}{z-y}\qquad(46)$$

and its analytic continuations. The function $S_D(z)$ is analytic in the complex plane with a cut along the positive ray of the real axis; $z=0$ is a branch point. The polarizability in (45) satisfies all the exact relations derived above.

Expression (45) has the same shape as one derived previously,[8,9] but the function $S_D(z)$ is quite different. The reason lies in an incorrect averaging over the configurations of the pair of monomers in the fractal in Refs. 8 and 9 [an incorrect transformation from (14) to (15) in Ref. 9]. We wish to stress that all the results of Refs. 8–11 nevertheless remain valid if the function $S_D(z)$ as in (46) is substituted into them.

It is a straightforward matter to show that the density of eigenstates of a fractal is symmetric with respect to X in the binary approximation and is given by

$$\nu(X) = \frac{1}{2}\int_0^\infty [2\delta(|X|-r_n^{-3})+\delta(|X|-2r_n^{-3})]p(r_n)\,dr_n.\qquad(47)$$

The relationship between $\operatorname{Im}\chi(X)$ and $\nu(X)$ given by (37) is not valid in the binary region. It begins to hold, however, if we put the polarizability in (40) in a form symmetric with respect to X. The contribution of the binary region thus does not disrupt the normalization of relationship (37) for the collective region.

In the binary approximation, one can derive a theory for the threshold photomodification of fractals. We recall that this effect was observed experimentally by Karpov et al.[13] After intense radiation, with an intensity above a cer-

tain threshold, is applied, the spectra of the fractal clusters undergo irreversible changes: Persistent gaps, which are selective in terms of frequency and polarization, appear in them.

Let us assume that the modification is local and that a given monomer becomes modified if the field intensity at it (which we will take to be simply the square of the amplitude) exceeds a certain threshold $I_m = \mathrm{const}$:

$$|\mathbf{E}_i|^2 > I_m.\qquad(48)$$

In the binary approximation, a pair of nearest monomers constitutes a closed system, and the fields at these monomers are furthermore equal. We can thus use (20) and find the gain of the local field intensity:

$$G(X) = \frac{|\mathbf{E}_i|^2}{|\mathbf{E}^{(0)}|^2} = \frac{\operatorname{Im}\chi_{zz}^{(p)}(X)}{\operatorname{Im}\chi_0(X)}.\qquad(49)$$

The experimental data of Ref. 13 suggest that this modification of monomers is of the nature of a melting or evaporation. It is thus reasonable to assume that the modified monomer ceases to contribute to the polarizability of the fractal. In this case we find the following expression for the quantity of interest—the change in the polarizability (per monomer of the original fractal) as a result of the photomodification—from condition (48) with the help of (49):

$$\Delta\chi_{\alpha\beta}(X) = -\int_0^\infty \chi_{\alpha\beta}^{(p)}(X)\,\theta\!\left(\frac{G(X_m)}{G_{max}(X_m)}-\frac{I_{th}}{I}\right)p(r_n)\,dr_n.\qquad(50)$$

Here X_m is the value of the variable X for the frequency at which the photomodification was carried out, $G_{max} = 1 + X_m^2/\delta^2$ is the maximum possible value of the gain factor G at the given X_m, I is the intensity of the external field, and $I_{th} = I_m/G_{max}$ is its threshold value.

5. RESULTS OF A NUMERICAL SIMULATION

Fractals of three types, generated by the Monte Carlo method, were used (a software random-number generator was used to generate the fractals).

1. Random walks ($D = 2$) were constructed on a cubic lattice (the monomers were placed at the sites which were visited) and then diluted by a factor of 1000 ($\beta = 0.001$; see the discussion above). The clusters resulting from this dilution contained a mean number of monomers $\overline{N} = 30$ (in a control, there were $\overline{N} = 15$).

2. Self-avoiding random walks[17] were constructed by trial and error with a step of fixed length and a random direction. The monomers were placed at sites and were treated as mutually impenetrable spheres with a diameter equal to the step of the walk. If a certain step resulted in an intersection of spheres, it was discarded. A dilution by a factor of 20 was carried out ($\beta = 0.05$). The resulting clusters had a mean number of monomers $\overline{N} = 25$. Their fractal dimension, determined from function (1), was $D = 1.8$, in reasonable agreement with the known value[17] $D = 1.7$.

3. Witten-Sander clusters[18] were constructed through a simulation of a diffusion-controlled aggregation on a cubic lattice and then diluted by a factor of 20 to $\overline{N} = 25$ monomers. The value $D = 2.5$ which was found was close to the value 2.51 in the literature.[18]

As a control, we also used a gas of monomers ($D = 3$).

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The reason for the moderate dilutions in cases 2 and 3 and the reason why we did not use a very large number of monomers in the resulting clusters was a limitation in computation power. As we will see below, however, even for these fractals the polarizability per monomer, χ, is essentially independent of the number of monomers.

The polarizability was calculated in accordance with (12) through a diagonalization of matrix (4) by the Jacobi method.[19] That method has the advantage in principle that the solutions it generates automatically satisfy exact relationships (13), (14), and (18). It also has the advantage (as a computational method) that after going through the diagonalization one time for each cluster one can find solutions for arbitrary X and δ through a simple summation. As a control we also carried out a direct solution of the original system of equations, (3), by the Gauss elimination method and by the square-root method. In all cases, these three independent methods led to results which were in excellent agreement with each other. The individual polarizabilities which were calculated were averaged over a large number of realizations (75 or 150 in the $N = 15$ case). In this case the statistical error in the polarizability did not exceed 1% (of the value at the maximum).

We will demonstrate the basic results here for fractals constructed in the random-walk model, since the theoretical parameter values are well known in this case: $D = 2$, $R_0 = R_1 (6\beta)^{-1/2}$, where R_1 is the step of the lattice. Furthermore, in the construction of the fractals the limitation in terms of computation facilities was unimportant, so it was possible to achieve a large dilution ($\beta = 0.001$). Unless otherwise stipulated below, we will mean fractals of specifically this type. We will discuss other fractals in order to demonstrate behavior as a function of the fractal dimension ($D = 1.8$–2.5).

Figure 1 shows the results of a calculation of the linear polarizability χ as a function of the variable X for various values of the parameter δ. We see that the Im $\chi(X)$ absorption profile is a peak, whose width decreases, and whose height increases, with increasing $Q = 1/\delta$. In accordance with the results derived above, the δ dependence is weak at $|X| \gtrsim \delta$. The obvious increase in the statistical noise with decreasing δ agrees with (18) (the optical theorem). The width of the absorption profile of a fractal is significantly greater than the width of the resonance in an individual monomer ($\sim\delta$). Qualitatively, the behavior of the polarizability for fractals of other dimensions is similar to that shown in Fig. 1.

Figure 2 shows the results of an absorption calculation for fractals containing quite different mean numbers of monomers $\overline{N}$. We see that the dependence on $\overline{N}$ has already essentially reached saturation at $\overline{N} > 15$. This fact is a reason for using such a characteristic as the polarizability per monomer of a fractal.

A fundamental theoretical prediction is a universal power-law behavior as a function of $|X|$ (a scaling) of the absorption [see (27)] and of the density of states [see (37)] in the collective region, (32). This prediction is illustrated by Fig. 3. We see that a power-law behavior of these two quantities is indeed exhibited at small values of $|X|$, within the statistical noise. For the other fractals we find a similar agreement. It also follows from Fig. 3 that the absorption and the density of states satisfy relationship (37) quite accurately.

Here are the values of the optical spectral dimension (for $D = 1.8$, 2.0, and 2.5, respectively) found from Fig. 3 and from corresponding data for fractals of other types:[2] $d_o = 0.43(3)$, 0.38(3), and 0.49(2) in terms of the absorption; and $d_o = 0.54(8)$, 0.33(5), and 0.51(6), respectively, in terms of the density of states. It follows that for fractals of a common type the indices of the functional dependences Im $\chi(X)$ and $v(X)$ differ by less than two standard deviations; i.e., the difference is statistically insignificant. In all cases, we find $d_o < 1$, in agreement with the arguments above. Note also that there is no significant correlation
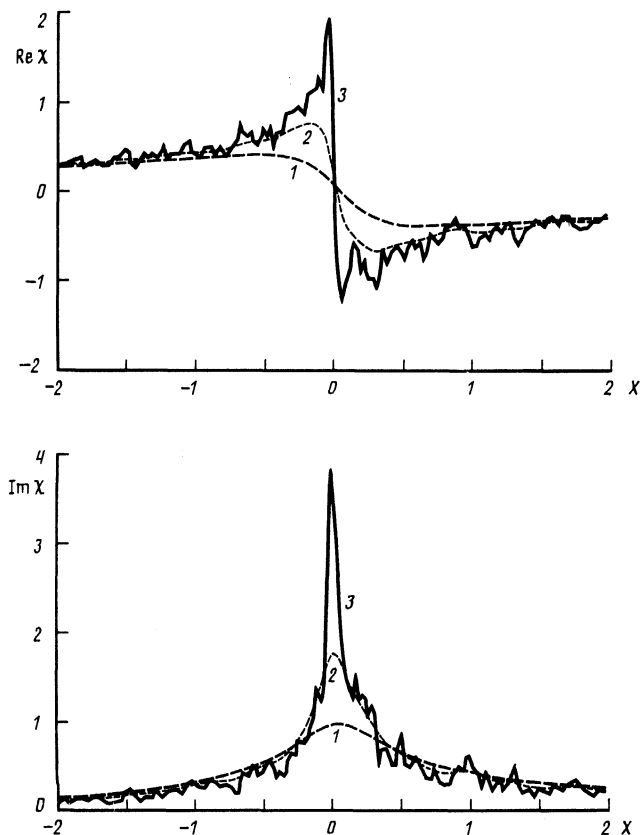

FIG. 1. Polarizability of a fractal ($D = 2$, $\overline{N} = 30$) for various values of the parameter δ: 1—0.33; 2—0.10; 3—0.01.
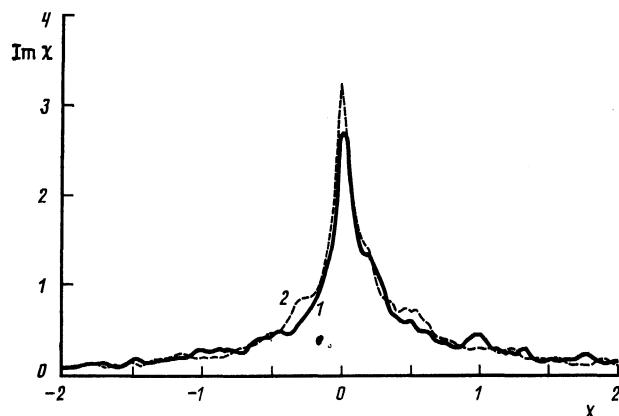

FIG. 2. The absorption of a fractal ($D = 2$) for $\delta = 0.03$ and two values of the mean number of monomers: 1—$\overline{N} = 30$; 2—$\overline{N} = 15$.
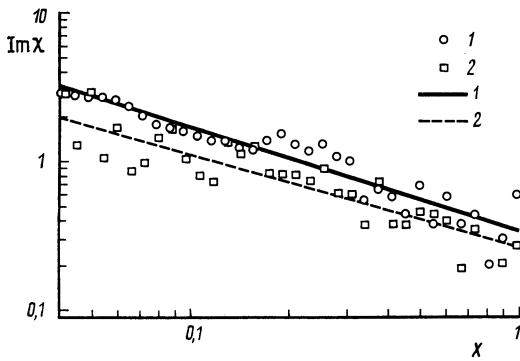
FIG. 3. Absorption of a fractal as a function of $X > 0$, in full logarithmic scale. 1—Found from exact expression (12); 2—expressed in terms of the density of states in accordance with (37). The corresponding straight lines were found by linear regression ($\delta = 0.01$).
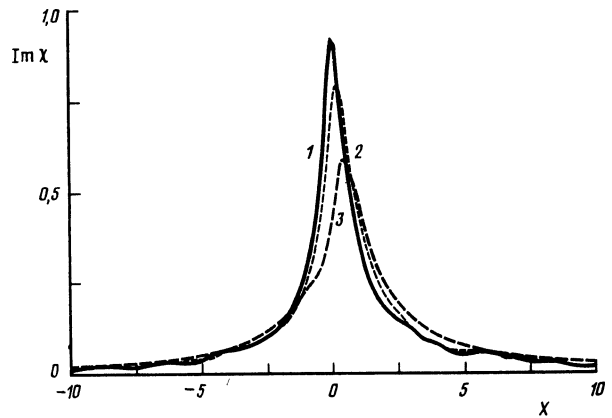

FIG. 4. Absorption of a fractal with $\delta = 0.33$. 1—Calculated from exact expression (12); 2—calculated in the binary approximation through the use of (39) and a numerical averaging; 3—calculated in the binary approximation from approximate expression (45).

between d_o and D. For a trivial fractal ($D = 3$), there is no obvious power-law behavior as a function of $|X|$, in agreement with the theory [see, in particular, the condition defining the collective region, (32)].

As was shown above, the wings of the absorption profile ($|X| \gtrsim 1$) for dilute fractals should be described correctly (within the parameter $1/|X|$) by the binary approximation. A systematic calculation of $\chi(X)$ in this approximation was carried out by the following numerical procedure. The nearest neighbor of each monomer was found. The quantity $\chi_{zz}^{(p)}(X)$ in (39) was calculated. An average was then taken over all monomers of the given cluster and over all clusters. Figure 4 compares the absorption calculated by this method with the result calculated by exact expression (12) ($\delta = 0.33$). We see that the agreement between the binary approximation and the exact calculation is indeed very good at $|X| > 2$. Even near the center of the profile the differences between the binary approximation and the exact calculation are small; the reason is that the value of δ is not very large. Specifically, the maximum coherence radius of the excitations, which can be found from (30) by substituting in $|X| = \delta$, is $L_X \sim 2$ for the dimensions given above. In other words, it is not much greater than $R_0 = 1$. In this case a universal scaling-invariant behavior does not develop, even near the center of the profile, because of the violation of condition (33).

Also shown in Fig. 4 is a result calculated from approximate analytic expression (45) (curve 3). Its agreement with the result of the exact calculation is also good in the region in which the binary approximation is valid (at the wings of the profile). The agreement at the center is poorer than for the systematic binary approximation (curve 2).

Figure 5 shows corresponding data for $\delta = 0.03$. It follows from a comparison of this figure with the preceding figure that all three curves agree well with each other at the wings of the profile ($|X| > 2$). The absorption peak near the center of the spectral profile, in contrast, is not described by the binary approximation in any of its modifications. This symmetric peak (the binary approximation predicts an asymmetric absorption profile) is of a collective nature. Its shape is a power law (cf. Fig. 3).

The numerical simulation of the threshold photomodification of clusters (see the discussion above) was carried out in the following way. For the external radiation (causing

the modification) the following were specified: the value of $X = X_m$, the extent to which the intensity exceeded the photomodification threshold ($K = I/I_{th} - 1$), and a linear polarization parallel to the z axis. Photomodification condition (48) was written in the form

$$|\mathbf{E}_i|^2/I > G_{max}/(1+K).$$

With $X = X_m$, the values of $|\mathbf{E}_i|^2/I$ were found from (12) and (19). Those monomers for which the photomodification condition was satisfied were eliminated from the cluster. After the elimination of these monomers, the absorption spectra of the cluster were determined numerically, for a linear z polarization (parallel) and for a linear xy polarization (perpendicular) of the probing radiation. An average was then taken over the ensemble of fractals.

Figure 6 shows the changes caused in the spectra by the photomodification as calculated by the numerical procedure described above and also as calculated in the binary approximation, in accordance with (50). We can draw some conclusions from these results. The photomodification is spectrally selective and also selective in terms of polarization; these conclusions agree with the experimental facts.[13] In the case of the z polarization of the probing radiation, the dip is observed near the modification frequency ($X = X_m$), while for
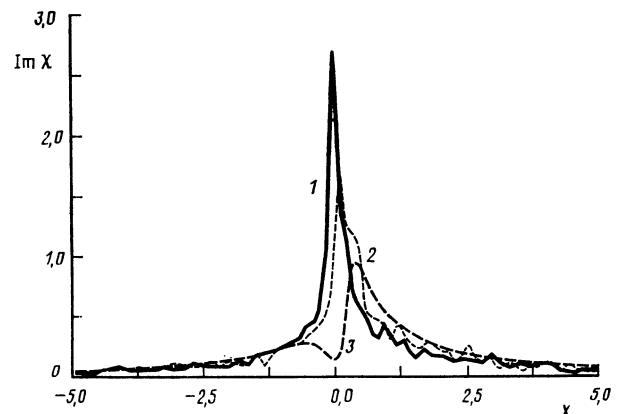

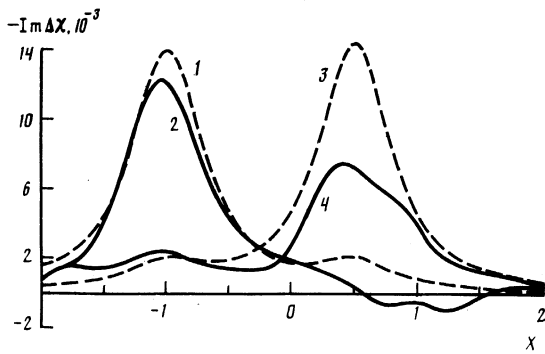FIG. 5. The same as in Fig. 4, but for $\delta = 0.03$.

FIG. 6. Changes in the cluster absorption spectra during photomodification for (1,2) parallel and (3,4) perpendicular polarizations of the modifying and probing radiation (the positive direction along the ordinate corresponds to a decrease in the absorption during photomodification). 2,4—Exact calculation (see the text proper); 1,3—calculation in the binary approximation ($X_m = -1$, $\delta = 0.33$, $K = 0.25$).

the xy polarization the dip is centered around the "mirror" frequency ($X = -X_m/2$).

It follows from Fig. 6 that the binary approximation reproduces the qualitative features found through the numerical calculations. It thus becomes possible to offer an interpretation of the observed spectral and polarization selectivity. According to (39) at negative deviations of the modifying radiation ($X_m < 0$), most of the pairs which reach resonance with the radiation are directed along the field (for these pairs, the vector n is parallel to the z axis). For the z polarization of the probing radiation, these pairs provide an absorption near $X = X_m$, while for the xy polarization of the probing radiation they provide an absorption near $X = -X_m/2$. The following established facts can also be explained on the basis of the binary approximation. As $|X|$ increases, the spectral and polarization selectivities intensify (the binary approximation becomes more valid in the process). The maximum selectivity of the modification is observed near the threshold, i.e., at small values of K.

6. CONCLUSION

Here we will briefly review the basic physical principles underlying this study and some of the results which we find of most interest. In this paper we have described the linear polarizabilities of fractals (or of fractal clusters). As the sole spectral variable $z = -X - i\delta$ we used the inverse polarizability of an isolated monomer, in (7), and the point $X = 0$ corresponded to the resonant frequency of the monomer. Exact relationships derived for the polarizability, (14), (18), and (23), and the scale invariance of fractals underlie the theory. In addition to the analytic study, we carried out a numerical solution of the basic equations, taking an average over the ensemble of fractals. In particular,we calculated the polarizabilities of fractals over a wide spectral range (Fig. 1).

The transformation properties [see (29)] of the variable X (the reciprocal polarizability of a renormalized monomer) under a change in the minimum scale R_0 (a renormalization transformation) were established. Using these properties, we found estimate (30) of the coherence length of normal excitations of the fractal. With that estimate, we were in a position to classify these excitations. Specifically, for monomers having a sufficiently high resonance Q [see

(33)] the overall spectral range breaks up into a collective region (32), and a binary region ($|X| \gtrsim 1$).

In region (32), which corresponds to the central peak of the absorption spectrum, the eigenstates of the fractal are of a collective nature, i.e., delocalized over a large number of monomers. The absorption and the density of states of the fractal are power-law functions of $|X|$, with identical powers (equal to $d_o - 1$, where d_o is the optical spectral dimension and lies in the interval $0 < d_o < 1$) and with numerically related coefficients [see (37) and Fig. 3]. This behavior is universal, independent of the structural details of the fractal at small scale (the coordination numbers, the connectedness, etc.). The dipole–dipole interaction of the monomers is asymptotically exact and universal at the distances of interest, on the order of $L_X \gg R_0$. From a comparison with the results of the numerical calculations, we determined the values of the optical spectral dimension, which turned out to be approximately the same for the fractals studied, $d_o \approx 0.5$.

In the wings of the absorption profile (at $|X| \gtrsim 1$) the states of the fractal are two-particle states. These states are sensitive to the small-scale structural details of the fractal and to the interaction between monomers, so they cannot be described in a universal way. In this connection, we introduced a model of a dilute fractal, for which the small-scale structure is extremely simple and is determined by binary correlation function (1), which depends on the Hausdorff dimension D. We wish to stress that the dilution of the fractal does not alter its properties in the collective region.

Dilute fractals are not merely a convenient theoretical model but in fact an idealization of a rather wide class of real clusters, formed by sparsely distributed impurities connected with the monomers of the original fractal (e.g., dye molecules in a complex with a macromolecule, microscopic metal particles in pores in a glass, etc.). The description of dilute fractals is thus of interest in its own right. Working in the binary approximation, we have derived the polarizability of dilute fractals, (40), and we have compared it with the results of numerical calculations (Figs. 4 and 5).

It has been shown (Fig. 6) that the threshold photomodification of fractals gives rise to comparatively narrow dips (with a width on the order of a few times δ) in their spectra. These dips have a well expressed dichroism; i.e., the photomodification is spectrally selective and polarization-selective. This result reproduces experimental data.[13] It indicates
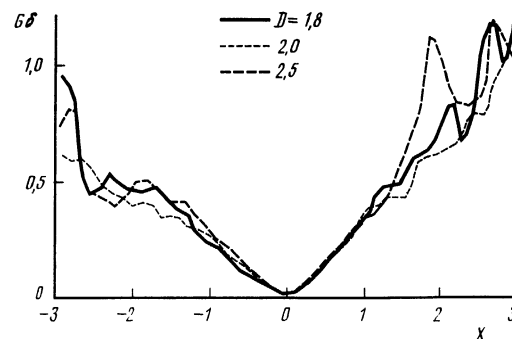

FIG. 7. The gain for the local field, $G = \langle |E_i|^2 \rangle / |E^{(0)}|^2$, as a function of X for fractals of three types ($D = 1.8$, 2.0, and 2.5) for $\delta = 0.1$ (actually plotted here is the produce $G\delta$, since this combination depends only slightly on δ).

a fundamental feature of the spectra of fractals: The broadening mechanism is anisotropic and basically nonuniform.

The selectivity of the photomodification makes fractal media extremely promising for use as carriers for dense optical data storage. At values $\delta = 0.1$–0.3, which are realistic values for the media which have been studied experimentally, one could burn several easily distinguishable dips in the spectral profile in each of two polarizations (z and xy). Consequently, it would be possible to store about 10 data bits at each point in the medium and thereby raise the storage density by an order of magnitude.

Finally, we would like to discuss an extremely important property of fractals: the presence of large fluctuations in the local fields—significantly larger than the mean field. Such fluctuations lead to a huge intensification of Raman scattering and of nonlinear parametric scattering (intrinsic or extrinsic) of light by fractals. This intensification was predicted previously on the basis of the binary approximation.[8-11]

From the optical theorem, (20), and from expressions (37) and (43) for the gain G (the factor by which the local field acting on a monomer is intensified), we find some estimates (which are asymptotically exact in terms of X dependence). In the collective region, (32), we find

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

while for dilute fractals, in the binary region ($X \gg 1$), we find

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

The existence of a collective region which is nondegenerate (i.e., of finite width) is a property of nontrivial factors. It follows from estimates (51) and (52) that for nontrivial fractals, in both the collective and binary regions, the fluctuations of the local fields are large: The variance of the field is proportional to a large parameter, namely, the Q of an isolated monomer [see (33)].

Figure 7 shows the gain G as a function of X for fractals of three types which we have studied here. In agreement with the theory presented above, the central part of these curves, which corresponds to collective excitations, is symmetric with respect to X and is described by expression (51). Toward the wings, the factor G increases, as follows from (51)

and (52), reaching values on the order of 10 (for $\delta = 0.1$). There is no obvious dependence of G on the type of fractal (this result agrees with the approximate equality of the values of d_o found above).

In summary, this systematic theory shows that the large fluctuations of the local fields (and thus the huge intensification of Raman and parametric scattering of light) which were predicted in Refs. 8–11 are not an artifact of the binary approximation but instead a general property of nontrivial fractals whose monomers are high-Q optical resonators.

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