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Grant N00014-90-J-1193

TECHNICAL REPORT No. 25

Scale-Invariant Theory of Optical Properties of Fractal Clusters

by

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Prepared for Publication

in

Scaling in Disordered Materials
Edited by J. P. Stokes
Materials Research Society, Pittsburgh

Departments of Chemistry and Physics
State University of New York at Buffalo
Buffalo, New York 14260

September 1990

UNCLASSIFIED
SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1a. REPORT SECURITY CLASSIFICATION
Unclassified

1b. RESTRICTIVE MARKINGS

2a. SECURITY CLASSIFICATION AUTHORITY

2b. DECLASSIFICATION / DOWNGRADING SCHEDULE

3. DISTRIBUTION / AVAILABILITY OF REPORT
Approved for public release; distribution unlimited

4. PERFORMING ORGANIZATION REPORT NUMBER(S)
UBUFFALO/DC/90/TR-25

5. MONITORING ORGANIZATION REPORT NUMBER(S)

6a. NAME OF PERFORMING ORGANIZATION
Depts. Chemistry & Physics
State University of New York

6b. OFFICE SYMBOL (If applicable)

7a. NAME OF MONITORING ORGANIZATION

6c. ADDRESS (City, State, and ZIP Code)
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Buffalo, New York 14260

7b. ADDRESS (City, State, and ZIP Code)
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Arlington, Virginia 22217

8a. NAME OF FUNDING / SPONSORING ORGANIZATION
Office of Naval Research

8b. OFFICE SYMBOL (If applicable)

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER
Grant N00014-90-J-1193

8c. ADDRESS (City, State, and ZIP Code)
Chemistry Program
800 N. Quincy Street
Arlington, Virginia 22217

10. SOURCE OF FUNDING NUMBERS

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO	WORK UNIT ACCESSION NO

11. TITLE (Include Security Classification)
Scale-Invariant Theory of Optical Properties of Fractal Clusters

12. PERSONAL AUTHOR(S)
Vadim A. Markel, Leonid S. Muratov, Mark I. Stockman and Thomas F. George

13a. TYPE OF REPORT

13b. TIME COVERED
FROM ________ TO ______

14. DATE OF REPORT (Year, Month, Day)
September 1990

15. PAGE COUNT
5

16. SUPPLEMENTARY NOTATION
Prepared for publication in Scaling in Disordered Materials, Edited by J. P. Stokes, Materials Research Society, Pittsburgh

17. COSATI CODES

FIELD	GROUP	SUB-GROUP

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FRACTAL CLUSTERS
OPTICAL PROPERTIES
SCALE-INVARIANT THEORY
EXTERNAL HAUSSDORF DIMENSION
DENSITY FLUCTUATIONS
LOCAL FIELDS

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Optical properties of fractal clusters are considered by means of a scale-invariant theory. Numerical simulations are carried out for three fractal types: self-avoiding walks, random walks and Witten-Sander's.

20. DISTRIBUTION / AVAILABILITY OF ABSTRACT
☒ UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT. ☐ DTIC USERS

21. ABSTRACT SECURITY CLASSIFICATION
Unclassified

22a. NAME OF RESPONSIBLE INDIVIDUAL
Dr. David L. Nelson

22b. TELEPHONE (Include Area Code)
(202) 696-4410

22c. OFFICE SYMBOL

DD Form 1473, JUN 86 Previous editions are obsolete.

SECURITY CLASSIFICATION OF THIS PAGE
UNCLASSIFIED

Scaling in Disordered Materials, Proceedings of Symposium W (Extended Abstracts)
Materials Research Society 1990 Fall Meeting, edited by J. P. Stokes

SCALE-INVARIANT THEORY OF OPTICAL PROPERTIES OF FRACTAL CLUSTERS

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INTRODUCTION

Fractals are scale self-similar mathematical objects possessing nontrivial geometrical properties [1,2]. There exist various physical realizations of fractals [3-5], and here we shall consider optical properties of what we believe to be one of the most important such realizations, namely, fractal clusters (to be named below simply as fractals).

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_0 is a characteristic separation between nearest monomers, and R_c is the cluster total size.

A consequence of the self-similarity is the scaling (power-like) form of the pair (density-density) correlation function $g(r)$, mean number N of the monomers in the the fractal and its mean density $\bar{\rho}$,

$$g(r) = (D/4\pi R_0^3)\,(r/R_0)^{D-3}\,, \quad N \sim (r/R_0)^D\,, \quad \bar{\rho} \sim (R_c/R_0)^{D-3}\,. \tag{1}$$

where the index D is the fractal (external Haussdorff) dimension. A fractal is called nontrivial if $D < 3$. As one can see from Eq. (1), the mean density of a nontrivial fractal is asymptotically (for $R_c \gg R_0$) zero. This feature, together with strong pair correlation, is the reason for the great role that the density fluctuations play in the fractals. Below we shall show that large correlated fluctuations of local fields play a decisive role in determining optical properties of fractals. In particular, there exists scaling of the optical absorption and the density of the dipole excitations (eigenstates) of the fractal.

The linear optical (dipole) polarizabilities of fractals have been studied theoretically [6-9]. However, the scale-invariant theory, which is the subject of the present report, has not been published in the literature.

FRACTAL OPTICAL PROPERTIES AND SCALING

Let us consider a model of a cluster as a system consisting of N polarizable particle (called monomers) located at the points $\mathbf{r}_i$, $i = 1 \ldots N$, with the dipole interaction between them, and $R_c \ll \lambda$, where λ is the light wavelength. The light-induced dipole (transition) moments $\mathbf{d}_i$ on monomers obey the well-known system of equations

$$d_{i\alpha} = \chi_0 E_\alpha^{(0)} - \chi_0 \sum_{j=1}^{N}{}' \left[\delta_{\alpha\beta} - 3n_\alpha^{(ij)} n_\beta^{(ij)}\right] r_{ij}^{-3} d_{j\beta}\ . \tag{2}$$

where the Greek subscripts stand for tensor components (summation over repeated indices is implied), and the Latin indices stand for ordinal numbers of monomers; χ_0 is the dipolar polarizability of an individual (isolated) monomer: $\mathbf{E}^{(0)}$ is the electric field of the external (exciting) radiation: $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$: and $\mathbf{n}^{(ij)} = \mathbf{r}_{ij}/r_{ij}$.

The fractal specificity of the considered problem is due to the divergence at small r in the mean-field analog of Eq.(2), $\mathbf{E}^{(0)} - \chi_0 \int [\mathbf{d} - 3\mathbf{r}(\mathbf{rd})/r^2] r^{-3} g(r) \mathrm{d}\mathbf{r}$ (cf. Refs. [8,9]). This divergence is an indication of the dominating role of large fluctuations and existence of scaling in fractals. In this brief report we can only outline the theory and then briefly discuss the results obtained.

The main object of the theory is the optical (dipole) polarizability $\chi(X)$ of the fractal (per one monomer) as a function of the variable $X = \mathrm{Re}\chi_0^{-1}$. The polarizability is formally found as a decomposition in terms of the exact eigenvectors and eigenvalues of Eq. (2). Using this decomposition, the exact sum rules for the polarizability are obtained as

$$\frac{1}{\pi}\int_{-\infty}^{\infty} \mathrm{Im}\chi_{\alpha\beta}^{(i)}(X)\mathrm{d}X = \delta_{\alpha\beta} \ , \quad \mathbf{P}\int_{-\infty}^{\infty} \mathrm{Re}\chi_{\alpha\beta}^{(i)}(X)\mathrm{d}X = 0 \ , \quad \mathbf{P}\int_{-\infty}^{\infty} X\mathrm{Im}\chi(X)\mathrm{d}X = 0 \ . \tag{3}$$

where $\mathbf{P}$ denotes the principal value of integral.

The next step is to assume the scaling of dipole excitations of the fractal. Then the function $\mathrm{Im}\chi(X)$ can be shown to have the scaling form (note that $\mathrm{Re}\chi$ does not scale), if the resonance in the monomer has a high Q-factor, $Q = [R_0^3 \mathrm{Im}\chi_0^{-1}]^{-1}$. From (3) it follows that $\mathrm{Im}\chi$ scales as the function of $|X|$, thus having the form

$$\mathrm{Im}\chi(X) \sim R_0^3 \left(R_0^3|X|\right)^{d_o - 1} \ , \tag{4}$$

where the index d_0 is called the optical spectral dimension, $0 \le d_o \le 1$. Then, using the fact that $\mathrm{Im}\chi$ describes absorption and the total absorption of a fractal in the scaling region should not depend on choice of the minimum length R_0, from Eq. (4) we obtain the scaling transformation law $|X| \propto R_0^{-(3d_o - D)/(d_o - 1)}$. Using this, one can make sure that it possible to construct only one quantity L_X which has dimensionality of length and is invariant under the scaling transformation,

$$L_X \sim R_0 (R_0^3 |X|)^{(d_o - 1)/(3-D)} \ . \tag{5}$$

This quantity, in accord with Alexander's strong localization hypothesis [10], should coincide with the coherence length of the fractal excitations. Equation (5) defines, thus, the dispersion law for the dipole excitations of the fractal, with $1/L_X$ playing the role of wavevector. The conditions of scaling $R_0 \ll L_X \ll R_c$ and $X \gg R_0^{-3}Q^{-1}$ with the aid of Eqs. (5) and (1) are rewritten as

$$R_0^{-3}Q^{-1}, \ R_0^{-3}N^{(3/D-1)/(1-d_o)} \ll X \ll R_0^{-3} \ . \tag{6}$$

Hence, if the necessary condition $Q \gg 1$ is met, scaling occurs at small $|X|$. In the vicinity of the frequency ω_0 of the resonance in the monomer, the variable X is proportional to detuning $\Omega = \omega - \omega_0$, where ω is the light frequency. Hence, the scaling region does exist, occupying the center of the fractal absorption band. Equation (6) can be considered as a self-consistent validation of the scaling assumption.

Using the above mentioned exact decomposition of χ and Eqs. (3), one can show for the scaling region (6) that the following relation exists between $\mathrm{Im}\chi$ and the density $\nu(X)$ of the dipole excitations (eigenstates) of the fractal:

$$\mathrm{Im}\chi(X) = \frac{\pi}{3}\nu(X) \propto |X|^{d_o - 1} \ . \tag{7}$$

Thus, the density of states and optical absorption scale with the same exponent $d_o - 1$, and the index d_0 is the counterpart for the dipole excitations of the vibration (fracton) spectral dimensionality $\bar{\bar{d}}$ introduced by Alexander and Orbach [11]. The dipole excitations tend to surface plasmons in the trivial ($D = 3$) limit, while vibrations tend to phonons. The surface plasmons possess a spectral gap, while phonons do not, which is the reason why the dispersion law (5) differs from its counterpart [10,11]. For the same reason, the index d_o does not numerically coincide with $\bar{\bar{d}}$ (cf. $0 \leq d_o \leq 1$, while $0 \leq \bar{\bar{d}} \leq 3$).

The failure of the mean-field approach and existence of scaling imply that fluctuations of the local fields in the fractals play a decisive role in determining their optical properties. From the above mentioned expansion of χ in the terms of the fractal eigenstates, it is possible to obtain the exact relation (an analog of the optical theorem) describing the relative dispersion G of the local field $\mathbf{E}_i$ acting upon the i^{th} monomer in the fractal.

$$G \equiv \left\langle |\mathbf{E}_i|^2 \right\rangle / |\mathbf{E}^{(0)}|^2 = Q^{-1} \left(1 - Q^2 X^2\right) \mathrm{Im}\chi_{zz} \quad . \tag{8}$$

In the scaling region (6), from Eq. (8) it follows that

$$G \sim Q \, R_0^3 |X|^{d_0 - 1} \, . \tag{9}$$

Thus, the fluctuations are large: the dispersion of $\mathbf{E}_i$ is proportional to $Q \gg 1$, reaching its maximum at the wings of the scaling region, i.e., for $|X| \lesssim 1$.

NUMERICAL SIMULATION.

Three types of fractals have been generated by the Monte-Carlo method: random walks ($D = 2$), self-avoiding random walks ($D \approx 1.7$) and the Witten-Sander clusters [12] ($D \approx 2.5$). The mean number of monomers in the cluster was $N = 25 - 50$, and the number of clusters was from several hundred to thousand. For each cluster the fundamental system (2) was solved, and averaging over different fractals was performed. The numerical results show that the absorption and density of states obey the scaling prediction (4) and (7). The example of such results is presented in Fig. 1. The the values found for the d_o-exponent from the figure and similar data for other fractals are given in Table 1.

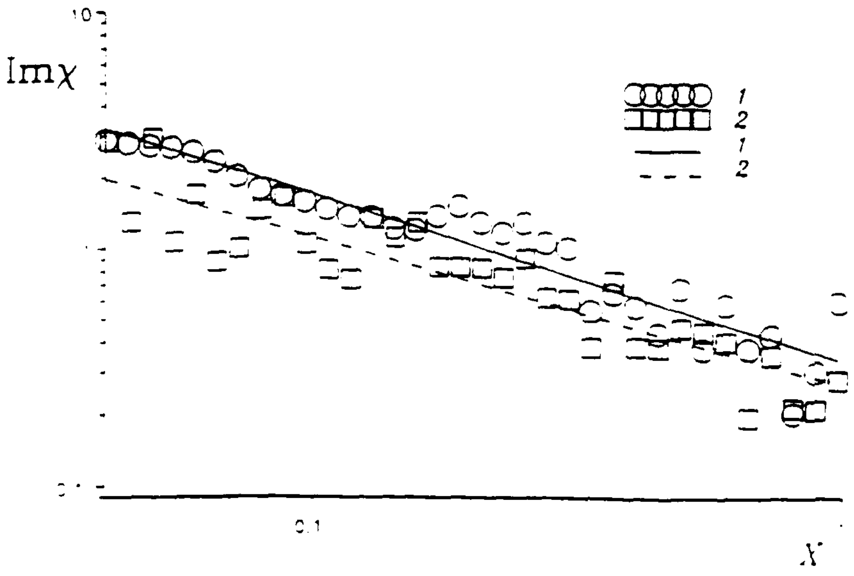

Fig. 1
Double-logarithmic plot of $\mathrm{Im}\chi(X)$ calculated directly (circles) and expressed through the eigenstate density with the aid of Eq. (7) (squares). The straight lines are found by the linear regression.

Table 1

Fractal type:	Self-avoiding walks	Random walks	Witten-Sander's
d_o found from Imχ:	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

CONCLUDING DISCUSSION

The scaling of optical absorption in nontrivial fractals is suggested by the failure of the mean-field approach. Assuming scaling (4), we have finally found the condition (6) of it, which means that the scaling assumption is self-consistent. Scaling is confirmed by numerical simulation, and the values of d_o are found for three types of fractals. Also, scaling is intimately coupled to large fluctuation of local fields in nontrivial fractals. Such fluctuations, as shown in the framework of the binary approximation [13], bring about giant enhancement of the nonlinear parametric scattering of light from fractals. The selective photomodification of fractals is also due to these fluctuations [14]. The features mentioned indicate the fractals to be promising media for nonlinear optics and optical information processing.

ACKNOWLEDGMENTS

This research was supported by the office of Naval Research and the National Science Foundation under Grant CHE-8922288.

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14. T. T. Rantala, M. I. Stockman and T. F. George, in the present Proceedings.

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