Localization of collective dipole excitations on fractals

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Dipole optical excitations on fractals are numerically simulated. It is shown that dipole eigenmodes of fractals are localized. The corresponding localization length dependence on the generalized frequency parameter (dispersion law) has been calculated. The value of optical spectral dimension found from the dispersion relations is in agreement with the result obtained earlier from calculations of the density of eigenstates.

Objects with irregular geometry are ubiquitous in nature, and their dynamical properties are of general interest. The emergence of fractal geometry was a significant breakthrough in the description of irregularity. It is now well established that excitations of fractals also possess fractal properties. In particular, for vibrations on fractals the dispersion relation has the scaling form[1] $L \propto \omega^{-\tilde{d}/D}$, where ω is the excitation frequency, $\tilde{d}$ is the fracton dimension, D is the fractal (Hausdorff) dimension, and L is the coherence length. Dynamical excitations of random fractals should be characterized by a single unique length scale L which combines the roles of a wavelength and a localization length. In the trivial limit ($D = \tilde{d} = d_{\mathrm{ES}}$, where d_{ES} is the dimension of the embedding space) the relation reproduces the dispersion law of running waves (phonons) with L as the wavelength.

However, it is clear that the dispersion law for vibrations on fractals can only be valid for excitations of the Goldstone type which do not have a gap in the spectrum and become running waves in the trivial limit. For non-Goldstone excitations on fractals[2,3], such as dipole ones (e.g., of the plasmon type), one can expect a dispersion law which possesses a spectral gap for $D \to d_{\mathrm{ES}}$ and therefore it should be different from that for vibrations.

Let us consider a cluster as a fractal set of N polarizable monomers [$N = (R_c/R_0)^D$, where R_c is the gyration radius and R_0 is the characteristic distance between neighbor monomers] with a light-induced dipole-dipole interaction between them determined by the complex polarizability χ_0 of the isolated monomer. We define $Z \equiv \chi_0^{-1}$, $X \equiv -\mathrm{Re}Z$, and $\delta \equiv -\mathrm{Im}Z$. In the theory, X plays the role of a natural spectral variable (generalized frequency parameter) and δ is a positive parameter which describes dielectric losses. For monomers having an isolated resonance $\chi_0 = -\omega_m R_m^3(\Omega + i\Gamma)^{-1}$ ($\Omega \equiv \omega - \omega_0$, where ω and ω_0 are the laser and the resonant frequencies, ω_m and R_m are the characteristic excitation frequency and the monomer's size, respectively, and Γ is the homogeneous width of the resonance) one finds that $X = R_m^{-3}\Omega/\omega_m, \delta = R_m^{-3}\Gamma/\omega_m$. If monomers are spherical particles with $\chi_0 = R_m^3(\epsilon - 1)/(\epsilon + 2)$, then $X = -R_m^{-3}(|2\epsilon + 1|^2 - 9)/(4|\epsilon - 1|^2)$, and $\delta = 3R_m^{-3}\epsilon''/|\epsilon - 1|^2$. We assume here and below that $\lambda \gg R_m, R_0$.

An external electrical field, whose value at the site of the ith monomer ($i = 1, 2, ..., N$) is equal to $\tilde{E}_\alpha^i = E_\alpha^{(0)} \exp[-i\omega t + i\mathbf{k}\cdot\mathbf{r}_i]$, induces the transitional dipole moment $\tilde{d}_\alpha^i = d_\alpha^i \exp[-i\omega t + i\mathbf{k}\cdot\mathbf{r}_i](\alpha = x, y, z)$. We introduce the $3N$-dimensional vectors $|d)$ and $|E^{(0)})$ with components $(i\alpha \mid d) = d_{i\alpha}$ and $(i\alpha \mid E^{(0)}) = E_\alpha^{(0)}$. Similar notations will be also used for other vectors. The equation for $\mid d)$ acquires the form

$$Z \mid d) = \mid E^{(0)}) - V \mid d), \tag{1}$$

where

$$(i\alpha \mid V \mid j\beta) = \frac{a^{ij}\delta_{\alpha\beta} - 3b^{ij}n_\alpha^{(ij)}n_\beta^{(ij)}}{r_{ij}^3} \times \exp(ikr_{ij} - i\mathbf{k}\cdot\mathbf{r}_{ij}), \tag{2}$$

$$a^{ij} = 1 - ikr_{ij} - (kr_{ij})^2,$$
$$b^{ij} = 1 - ikr_{ij} - \tfrac{1}{3}(kr_{ij})^2.$$

Here V is the dipole-dipole interaction operator ($V = 0$ for $i = j$), $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, and $\mathbf{n}^{ij} = \mathbf{r}_{ij}/r_{ij}$. The interaction (2) includes the near-zone (nonradiative), transitional, and far-zone (radiative) terms of the dipole field. It was shown in Ref. 4 that if $R_0^3|X| \gg (R_0/\lambda)^{3-D}$ (this condition is always fulfilled at $\lambda \gg R_0$) for $D < 2$ and $R_0^3|X| \gg (R_0/\lambda)N^{1-2/D}$ for $D > 2$ then the transitional and far-field zones (i.e., monomers positioned at

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distances $r_{ij} \sim \lambda$ and $r_{ij} \gg \lambda$ from the given ith one) contribute negligibly to the local field. In this case one can reduce V in (1) and (2) to the Hermitian near-zone dipole-dipole interaction operator

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

Note that for small clusters ($R_c \ll \lambda$) the substitution of W for V is quantitatively applicable without invoking additional assumptions discussed above. Introducing eigenvectors $\mid n)$ and the corresponding eigenvalues w_n of the W operator, one can present a solution[2,5] of (1) (with the change $V \to W$) for the cluster polarizability $\chi^{(c)}$ ($d_\alpha^{(c)} = \sum_i d_\alpha^i = \chi_{\alpha\beta}^{(c)} E_\beta^{(0)}$) in the form[6]

$$\chi_{\alpha\beta}^{(c)} = \sum_n a_{n,\alpha} \chi_{n,\beta}^{(c)}, \quad \chi_{n,\alpha}^{(c)} = a_{n,\alpha} (\chi_0^{-1} + w_n)^{-1},$$

$$a_{n\alpha} = \sum_i (i\alpha \mid n). \quad (3)$$

Thus, a strong dipole interaction leads to the renormalization of the problem. Rather than N dipole moments in a cluster one should consider $3N$ dipolar eigenmodes having polarizability $\chi_{n,\alpha}^{(c)}$ [$(n \mid d) \equiv d_n = \chi_{n,\alpha}^{(c)} E_\alpha^{(0)}$] and contributing to the total cluster polarizability with the weight $a_{n,\alpha}$. These modes are shown[2] to be localized on fractals within the coherence length L_X given by

$$L_X \sim R_0 (R_0^3 |X|)^{(d_o - 1)/(3-D)}, \quad (4)$$

where d_o is an index called the optical spectral dimension ($0 < d_o < 1$).[2] Formula (4) can be obtained[2,3] using the self-similarity of a fractal and the idea that collective excitations of large coherence length L_X should be invariant with respect to scale transformation $R_0 \to R_0'$. It is clear that these scaling arguments are valid only under the condition $R_0 \ll L_X \ll R_c, \lambda$, which we assume to be fulfilled.

In order to clarify the meaning of formula (4) let us rewrite it in the following equivalent form:

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

Factor $R_0^3 (R_0^3 |X|)^{d_o - 1}$ in (5) represents the imaginary part of the polarizability (absorption) averaged over the ensemble of clusters per one particle, $\mathrm{Im}\chi^2$. Taking this into account, one can see the physical meaning of Eq. (5)

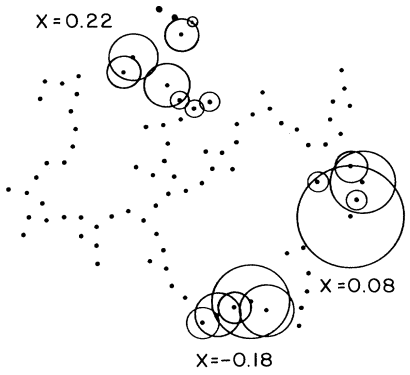

FIG. 1. Localized dipole modes on the fractal.

[and, accordingly, Eq. (4)]: Because of the scaling, there exists a coherence length L_X such that for the monomers within L_X the total polarizability (to be exact, its imaginary part, giving absorption and having dimensionality of length cubed) neither depends on the smallest scale R_0 nor on the largest scale R_c, and is determined by the coherence length itself. Thus the localization of dipole excitations on fractals expressed by dispersion law (4) is a direct consequence of the scale invariance (self-similarity).

Note that the localization of dipole excitations on fractals is a nontrivial fact since the long-range dipole-dipole interaction for "usual" media ($D \to 3$) leads to the logarithmic divergence and the excitations spread over the entire sample unless there is mutual compensation of the local fields due to high symmetry of the system. In Fig. 1 three different eigen dipole modes of the fractal are presented. Each mode is determined by certain value of the dimensionless spectral variable $R_0^3 |X|$ ($R_0 \equiv 1$), and it is excited at the corresponding frequency of the external (laser) field. The cluster was simulated by the cluster-cluster aggregation.[7] Points in the figure correspond to the centers of particles touching each other and forming the cluster. Radii of the circles drawn around the particles give the values of dipole moments induced on them. These dipole moments were calculated by determining the eigenvectors of the interaction operator W and substituting them to Eq. (3). It is clear from the figure that strong localization of the collective dipole modes occurs.

The coherence length L_X of the eigenmodes with a given eigenvalue w_n is defined as[3]

$$L^2(X) = \frac{\langle \sum_n \delta(X - w_n) \{ \sum_i (i\alpha \mid n)^2 r_i^2 - [\sum_i (i\alpha \mid n)^2 \mathbf{r}_i]^2 \} \rangle}{\langle \sum_n \delta(X - w_n) \rangle} \quad (6)$$

where $\langle \cdots \rangle$ denotes averaging over the ensemble of clusters. This definition has a clear quantum-mechanical analogy with $(i\alpha \mid n)$ as the wave function [$\sum_i (i\alpha \mid n)^2 = 1$]. In order to find the localization length at certain value of $X = X_0$ we used in our simulations the following formula which is equivalent to (6) at small values of decay constant δ:

$$L^2(X_0) = \frac{1}{m} \sum_{n_k=1}^{m} \left\{ \sum_{i=1}^{N} (i\alpha \mid n_k)^2 r_i^2 - \left[\sum_{i=1}^{N} (i\alpha \mid n_k)^2 \mathbf{r}_i \right]^2 \right\} \quad (7)$$

where $w_{n_k} \in [X_0 - \delta/2, X_0 + \delta/2]$ and $n_k = 1, ..., m$. The summation in (7) is over n_k for all realizations in the ensemble.

We have examined the cluster-cluster aggregates (fractal dimension $D \simeq 1.78$). The clusters in our simulations were generated using the Monte Carlo method and well-known procedures. Then, following Ref. 2, the clusters have been subjected to dilution (random decimation), which consists of the following. The ith ($i = 1, ..., N$) monomer is randomly retained in the cluster with some small probability β or removed with probability $1 - \beta$. This procedure simplifies the fractal structure at small scales and reduces the total number N of monomers in the cluster on average by a factor β, simplifying greatly the numerical simulations. At the same time, the resulting (diluted) fractal is characterized by the same fractal dimension as the original one. In most cases, 32-fold decimation ($\beta \simeq 0.03$) has been performed. Some simulations have been done with β as small as 10^{-3} for comparison. The results of the computations clearly show that the optical properties of fractals in the scaling range do not depend on the dilution, as expected. Finally averaging over a large ensemble of fractals (200 clusters) has been performed.

The localization length L_X as a function of the dimensionless spectral variable $R_0^3|X|(R_0 \equiv 1)$ for both positive and negative X is shown in Fig. 2 in a double-logarithmic scale. The number of particles in the diluted cluster is $N = 128$ (for smaller number of particles strong "finite-size" effect was obtained in our simulations). The calculations were made on the basis of formula (7) by using eigenvectors of the operator of dipole interaction W. The calculated points lie along straight lines, having slopes -0.53 ± 0.07 and -0.56 ± 0.06 for $X > 0$ and $X < 0$, respectively. The corresponding value of the optical spectral dimension, in accordance with (4), is $d_o = 0.33 \pm 0.08$. This value agrees well with $d_o = 0.3 \pm 0.1$ found in Ref. 4 from the density of states.

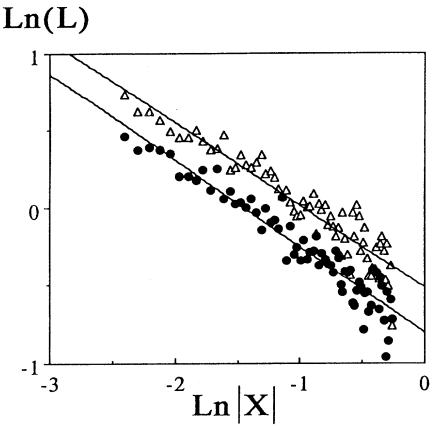

FIG. 2. Localization length L_X of dipole excitations on fractals as a function of the frequency parameter X for $X > 0$ (triangles) and $X < 0$ (circles).

Summarizing, strong localization of dipole collective excitations on fractals occurs under resonant light excitation conditions. The localization of the optical excitations on fractals results in very high local fields leading to the huge enhancement of resonant Rayleigh,[4] Raman,[8] and, especially, nonlinear scattering such as degenerate four-wave mixing.[9] The localization length dependence on the generalized frequency parameter (dispersion law) has scaling dependence (4) with the exponent expressed in terms of the optical spectral dimension and Hausdorff dimension. Our direct simulations of the localization length for cluster-cluster aggregates confirm the dispersion law predicted in Ref. 2 and generalized in Ref. 3 for arbitrary polar excitations and in Ref. 4 for arbitrary cluster size.

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