Inhomogeneous localization of polar eigenmodes in fractals

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A pattern of localization, called inhomogeneous localization, is found for dipolar eigenmodes (surface plasmons or eigenstates of the corresponding Schrödinger equation) of fractal clusters. At any given frequency, individual eigenmodes are dramatically different from each other, their sizes vary in a wide range, and their internal geometry may be topologically disconnected and singular at the small scale. These properties differ principally from the results reported for vibrational eigenmodes of fractals, which is attributed to the long-range interaction and non-Goldstonian nature of the polar modes.

The problem of the localization of elementary excitations in disordered condensed media has attracted a great deal of attention, particularly due to its universality and formidability. The localization is important for such various physical phenomena as metal-insulator (Anderson) transitions,[1] anomalous vibrational relaxation,[2] and enhanced optical responses of fractal clusters[3] (from an extensive literature, only a few examples most pertinent to the present paper are cited). A nontrivial geometrical structure of localized eigenmodes described by a multifractal statistics[4] has recently been established for vibrations of fractals ("fractons").[2,5–7] Localized excitations of fractal drums at low frequencies have been found.[8]

In this paper we consider another class of excitations, namely, polar excitations ("surface" plasmons) of fractals. As we show, they not only possess a nontrivial geometry different from that for fractons (a disconnected topology and singularity at the small scale), but also are extremely inhomogeneous at any given frequency in the whole spectral range. These principal differences are due to the long range of the dipole interaction and non-Goldstonian nature of the polar modes.

Surface plasmons in fractal clusters determine strong fluctuations and enhancement of the local optical fields[9] responsible for their enhanced nonlinear-optical responses. Earlier, Alexander and Orbach suggested that vibrational excitations in fractals are strongly localized.[10] We have formulated a strong-localization hypothesis for surface plasmons and on its basis obtained various scaling relations between the dispersion relation of the plasmons, optical absorption, and eigenmode density.[11] Strong localization of the surface plasmons has been confirmed by a subsequent numerical simulation[12] and observed in experiments with fractal clusters in plane.[13] However, high-resolution calculations of the linear responses and dispersion relations[14] have revealed substantial disagreement with the strong-localization predictions. We have also commented[15] that the experimental data of Ref. 13 do not necessarily imply a strong localization. These findings can be understood on the basis of the present results.

We study analytically and numerically both the localization and internal geometry of the surface plasmons. This plasmon problem maps to Schrödinger equation in the tight-binding representation with the corresponding hopping amplitudes. We have shown that (i) the eigenmodes (surface plasmons or eigenstates) at any given frequency [determined by the spectral variable X (Ref. 11)] are extremely inhomogeneous, and their localization radii L_n have a wide distribution between the radius of the cluster R_c and the minimum radius l_X which is expected to scale with X. (ii) The eigenmodes (especially in spectral wings) may consist of spatially disconnected but coherent "hot spots." The pair-correlation function $G_X(r)$ of the eigenmodes scales with the distance r, indicating self-similarity (fractality) of the average internal geometry of these modes. The Hausdorff dimension D_e of the modes is different from that of the underlying clusters and can be negative, which implies a singularity at $r \to 0$. The spatial behavior of individual eigenmodes is chaotic and does not possess the exponential tails associated with the localization, found earlier for quantum percolation.[6]

Consider a fractal cluster consisting of N monomers positioned at points $\mathbf{r}_i$, $i=1,\ldots,N$. The monomers are subjected to an external-wave electric field $\mathbf{E}^{(0)}$ oscillating at the optical frequency. We assume that the total size of the cluster, R_c, is much less than the light wavelength λ. Thus the field $\mathbf{E}^{(0)}$ is the same at each monomer. This field polarizes monomers, inducing oscillating dipole moments $\mathbf{d}_i$ which are random quantities due to the random structure of the fractal. The polarization causes local electric fields $\mathbf{E}_i = Z\mathbf{d}_i$, where $Z=\alpha_0^{-1}$, and α_0 is the dipole polarizability of an isolated monomer, supposed to be scalar for the sake of simplification. These local fields obey the well-known system of equations, $E_{i\alpha}=E_{0\alpha}-Z^{-1}\Sigma_j W_{i\alpha,j\beta}E_{j\beta}$, where the Greek subscripts denote vector indices, with the summation over the repeated indices implied, and W is the dipole-interaction tensor,

$$W_{i\alpha,j\beta}=[r_{ij}^2\delta_{\alpha\beta}-3(r_{ij})_\alpha(r_{ij})_\beta]r_{ij}^2, \tag{1}$$

for $i\neq j$, and $W_{i\alpha,j\beta}=0$ for $i=j$. Introducing a $3N$-dimensional vector $|E)$ with the components $(i\alpha|E)=E_{i\alpha}$ (and similar for other vectors), we obtain a single equation in $3N$-dimensional space,[11]

$$|E)=|E_0)-Z^{-1}W|E), \tag{2}$$

where the dipole-interaction operator is defined by its matrix elements as $(i\alpha|W|j\beta)=W_{i\alpha,j\beta}$. Similar to Ref. 11, we introduce the spectral variable $X=-\mathrm{Re}Z$ that is convenient to

use instead of the frequency and a parameter $\delta = -\,\mathrm{Im}Z$ determining the dissipation in a monomer.

The solution of Eq. (2) is determined by the eigenvalues w_n and eigenmodes $|n)$ of the W operator,[11]

$$(W - w_n)|n) = 0. \tag{3}$$

Such dipolar eigenmodes are traditionally called surface plasmons. Equation (3) has the form of a Schrödinger equation for vector (spin 1) particles on a (fractal) lattice $\{\mathbf{r}_i\}$ in the tight-binding approximation, where $|n)$ are the eigenkets. Thus, the present results are valid also for the quantum tight-binding vector problem.

We will study the spatial localization and delocalization of the surface plasmons or eigenkets (both are called below as eigenmodes). Each of these eigenmodes has intensity (quantum probability) $(i\alpha|n)^2$ at an ith monomer. It is natural to introduce the pair-correlation function of these intensities in space for an nth eigenmode,

$$g_n(r) = \sum_{i,\alpha,j,\beta} \delta(r - |\mathbf{r}_i - \mathbf{r}_j|)(i\alpha|n)^2 (j\beta|n)^2. \tag{4}$$

We can also average this correlation function over all eigenmodes with eigenvalues close to some spectral point X within spectral width δ. Assuming the homogeneous nature of spectral broadening for isolated monomers, the corresponding weight factors $\nu_n(X)$ can naturally be chosen in a Lorentz form, $\nu_n(X) = [(X - w_n)^2 + \delta^2]^{-1}$. We note that the eigenmode density is $\nu(X) = \Sigma_n \nu_n(X)$.[11] In such a way, we obtain for the spatial pair-correlation function $G_X(r)$ at a spectral point ("frequency") X the expression

$$G_X(r) = \left\langle \sum_n g_n(r)\, \nu_n(X)/\nu(X) \right\rangle. \tag{5}$$

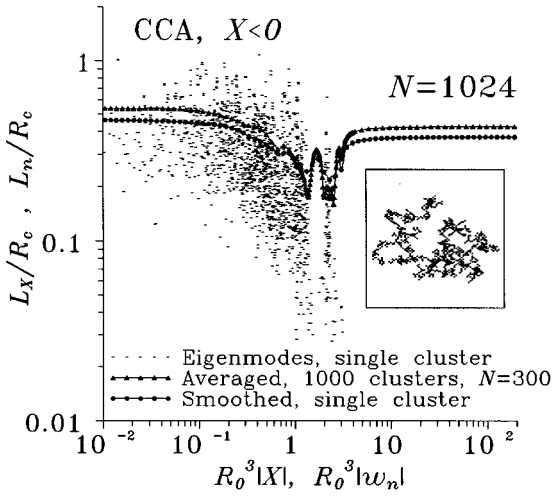

FIG. 1. The localization radii L_n of the eigenmodes vs their normalized eigenvalues w_n for a CCA cluster [whose two-dimensional (2D) projection is shown in the inset]. The "dispersion relations" $L_X \equiv L_X^{(2)}$ calculated for the given cluster and for the ensemble of 1000 clusters are also shown.

From Eq. (5), we can find the average localization radius $L_X^{(q)}$ of the eigenmodes calculated from the qth moment of the distribution, $L_X^{(q)} = [\frac{1}{2}\int_0^\infty G_X(r) r^q \mathrm{d}r]^{1/q}$.

Self-similarity of a fractal associated with strong fluctuation of its density at all scales suggests that eigenmodes of any size are scattered from the inhomogeneities of the corresponding scales and, consequently, may be strongly localized. The strong localization implies that the eigenmodes with $w_n \approx X$ are characterized by a single size L_X which is

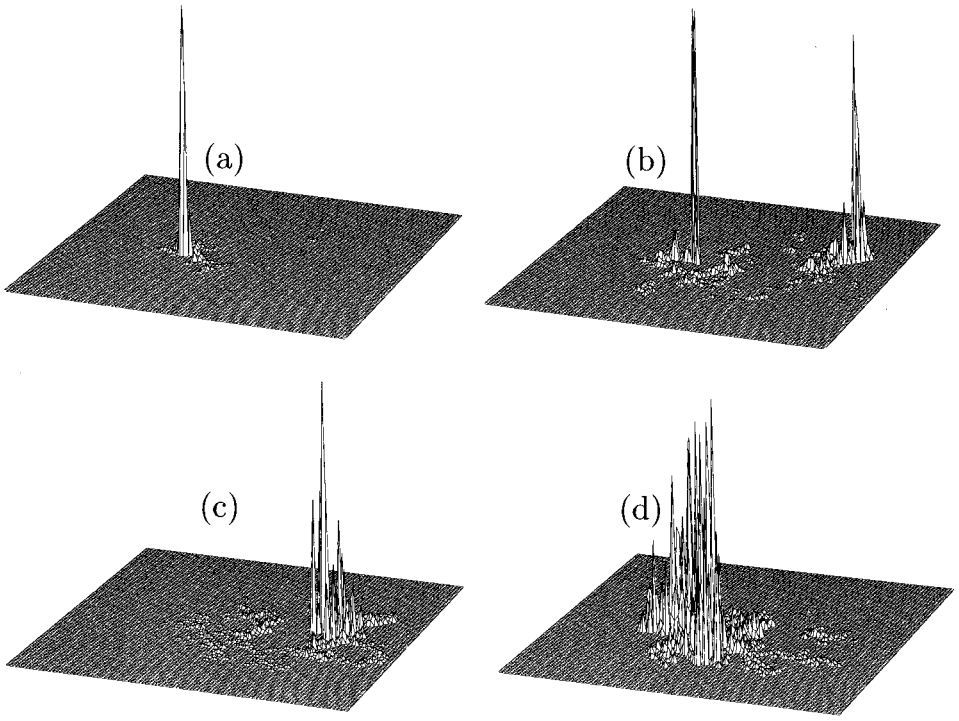

FIG. 2. The spatial intensity distribution of four eigenmodes for the cluster shown in Fig. 1, displayed over a 2D projection of the cluster. The values of $R_0^3 w_n$ and L_n/R_c for each eigenmode shown in the figure are (a) $R_0^3 w_n = -1.28$ and $L_n/R_c = 0.034$, (b) $R_0^3 w_n = -1.20$ and $L_n/R_c = 0.89$, (c) $R_0^3 w_n = -0.097$ and $L_n/R_c = 0.21$, and (d) $R_0^3 w_n = -0.099$ and $L_n/R_c = 0.60$.

simultaneously their wavelength and localization radius.[10] A possible form of $G_X(r)$ in this case is given by the superlocalization ansatz

$$G_X(r) \propto \exp\{-(r/L_X)^{d_\phi}\}, \tag{6}$$

where d_ϕ is the superlocalization exponent. In this case, $L_X^{(q)} \sim L_X$; i.e., it does not depend on R_c and can only weakly depend on q.

An alternative to the strong localization is the scaling correlation of the eigenmode intensities,

$$G_X(r) \propto r^{D_e - 1}, \tag{7}$$

where D_e is an index. If $D_e > 0$, it can be interpreted as the Hausdorff dimension of the excitation intensity as a measure. A negative value of D_e would imply an essential singularity at the small scale. Distribution (7) is applicable in the intermediate region of distances $R_c \gg r \gg l_X$, where R_c is the cluster radius, and l_X is a minimum correlation radius for the given frequency X. As one can see from Eqs. (1)–(3), the change of X is equivalent to the change of the spatial scale (by a factor $\propto |X|^{1/3}$), under which the underlying fractal cluster is invariant. This scaling argument suggests that D_e should not depend on X; i.e., D_e should be an index characteristic of a given type of fractals and not of a position in the spectrum. Distribution (7) implies that the ensemble of the eigenmodes at any given frequency possesses self-similarity as the underlying fractal support does (in contrast, individual eigenmodes break the scale invariance due to the localization). In the case of Eq. (7), the effective localization radius $L_X^{(q)}$ significantly depends on the power q,

$$L_X^{(q)} \approx l_X \left[\frac{D_e}{D_e + q} \frac{1 - (l_X/R_c)^{D_e+q}}{1 - (l_X/R_c)^{D_e}} \right]^{1/q}, \tag{8}$$

and it also depends on the maximum-scale size R_c.

Numerical calculations have been made using cluster-cluster aggregates (CCA's), both original and diluted (DCCA),[11] and random-walk and random-gas[14] clusters, employing the Lanczos diagonalization algorithm for $N = 300$ and $N = 1024$. The Monte Carlo simulation has invoked ensembles of 1000 clusters with $N = 300$.

In Fig. 1 for a single $N = 1024$ CCA cluster, we present the localization radii of the eigenmodes $L_n \equiv \frac{1}{2}\int_0^\infty g_n(r) r^2 dr$ versus their normalized eigenvalues (''eigenfrequencies'') $R_0^3 w_n$, where R_0 is the minimum characteristic distance between the monomers. We see that the eigenmodes at any frequency form an extremely heterogeneous distribution between the maximum size R_c and some minimum size $\sim l_X$ (a similar distribution has been found for two-dimensional projected clusters.[15]) Clearly, the eigenmodes in Fig. 1 cannot be characterized by a single dispersion relation. The minimum cutoff length l_x increases with $|X| \to 0$ in a manner that does not contradict the scaling ansatz of Ref. 11, $l_x \sim R_0 (R_0^3 |X|)^{(1-d_o)/(3-D)}$, where $d_o \leqslant 1$ is the optical spectral dimension, and D is cluster's Hausdorff dimension.

Further insight into the internal geometry of the eigenmodes can be obtained from the spatial distribution of the intensities of four sample eigenmodes shown in Fig. 2. For $R_0^3 |X| \approx 1$, two extreme cases of a strong [Fig. 2(a)] and weak [Fig. 2(b)] localization are shown. In the first case, all the intensity is concentrated on virtually a single monomer. The weak-localization mode has a disconnected topology, consisting of two highly localized hot spots separated by a distance on the order of and limited by the total size R_c. This is likely to be due to the long range of the dipole interaction. As $R_0^3 |X|$ decreases (i.e., the excitation frequency approaches the plasmon resonance of the monomers), the minimum size of the eigenmodes increases [Fig. 2(c)], while the maximum size remains as $\sim R_c$ [Fig. 2(d)]. The internal structure of the eigenmodes is irregular with strong spatial fluctuations.

The excitation-intensity correlation function $G_X(r)$ of Eqs. (4) and (5) averaged over a 1000-member ensemble of $N = 300$ CCA clusters is shown in Fig. 3(a) and the corresponding localization radii $L_X^{(q)}$ in Fig. 3(b). The general

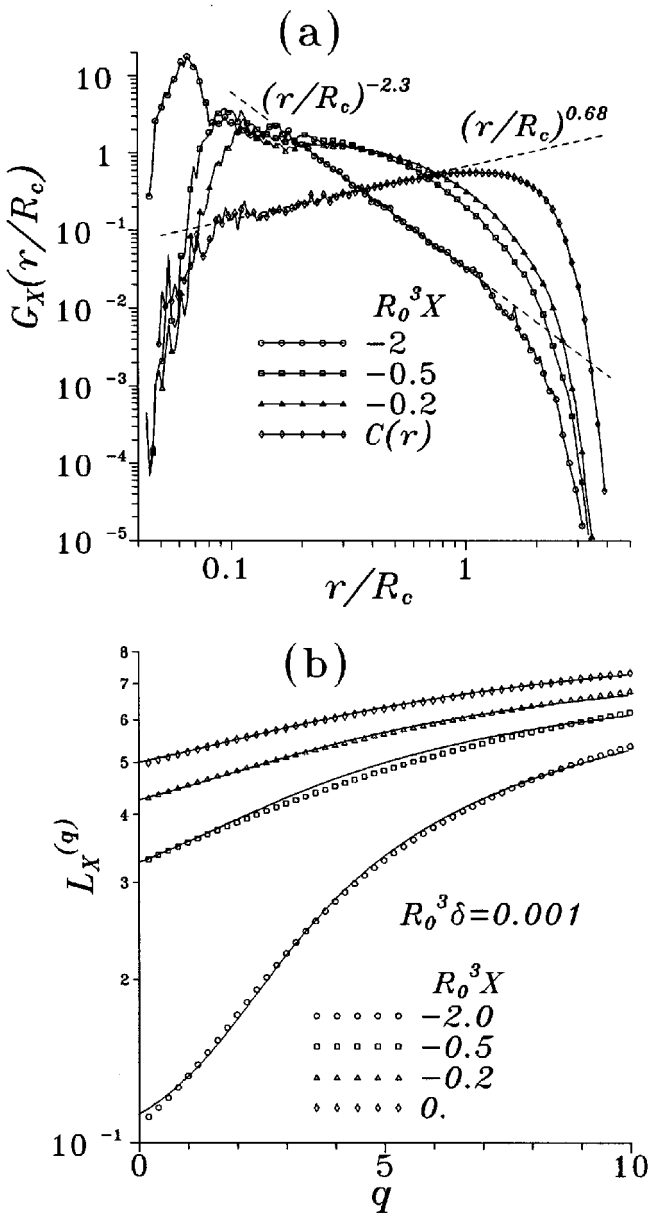

FIG. 3. Pair-correlation function $G_X(r/R_c)$ (a) and localization radius $L_X^{(q)}$ as a function of q (b) for CCA clusters, found by Monte Carlo simulation for the values of X shown. The solid lines in (b) are obtained from Eq. (8) for the values of $l_X/R_c = 0.07$, 0.22, 0.29, and 0.33 for $R_0^3 X = -2$, -0.5, -0.1, and 0.

form of $G_X(r)$ is incompatible with the strong localization in general and superlocalization form (6) in particular. The dropoff of $G_X(r)$ for $R_c \gg r \gg R_0$ is consistent with the scaling behavior of (7) for $R_0^3|X|=2$ and is slower for smaller $|X|$. We attribute the lack of scaling for these smaller $|X|$ to comparatively large values of l_X [see the caption to Fig. 3(b)], which leave a little space for the scaling. In all cases, $G_X(r)$ differs substantially from the support-density correlation function $C(r)=\langle N^{-2}\Sigma_{ij}\delta(r-|\mathbf{r}_i-\mathbf{r}_j|)\rangle$ [see Fig. 3(a)]. This implies that there exists a substantial correlation within the individual eigenmodes, in addition to the density correlation $C(r)\propto r^{D-1}$ inherent in the fractal support.

From $G_X(r)$ for $R_0^3X=-2$ [Fig. 3(a)], we find that $D_e=-1.3$. A negative value of D_e indicates that there is a singularity of the eigenmodes at the minimum scale, which is consistent with the previous spectroscopic results of Ref. 14. Thus, D_e for CCA's cannot be treated as the Hausdorff dimension. In the extreme scaling case where $l_X \ll R_c$, one gets from Eq. (8) that $L_X^{(q)} \sim l_x$ for $q \lesssim -D_e$ (which signifies the small-scale singularity), and $L_X^{(q)} \to R_c$ as $q \to \infty$. This type of behavior is actually observed for $R_0^3X=-2$ in Fig. 3(b). In this case, there exists a multitude of radii characterizing the eigenmodes at a given frequency, in accordance with Fig. 1. This situation is analogous to multifractality,[4,7] but multifractality does not necessarily imply the heterogeneity of individual eigenmodes at a given frequency.

The results for DCCA clusters (data not shown) are similar to those shown for CCA's in Fig. 3 except for one significant difference. Namely, we have found $D_e=0.18$. Consequently, D_e can be treated as the excitation Hausdorff dimension. In contrast to fractals, a principally different situation exists for random clusters consisting of noncorrelated monomers (data not shown). In this case, we have found that $G_X(r) \approx C(r)$ for $r \gg l_X$. The variation of the localization radii $L_X^{(q)}$ with q is negligible, and $L_X^{(q)} \approx R_c$. These results mean that the excitations are delocalized over the whole cluster, and there is no nontrivial correlation in their internal geometry. Thus, fractality of the support and not just its disorder is a determining factor of the inhomogeneous localization.

To summarize briefly, we have found a pattern of localization of dipolar eigenmodes characteristic of fractals that we call the inhomogeneous localization. Distinct from vibrational excitations,[5–8] individual polar eigenmodes even at the same frequency are dramatically different from each other. They possess localization radii varying in a wide range, a singularity at a small scale, and may have a disconnected topology. The pronounced distinction from the situation with vibrational eigenmodes is supposedly due to two facts: (i) The dipole interaction is a long-range one, and (ii) the dipolar eigenmodes are non-Goldstonian, in contrast to vibrations.

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