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Optical free-induction decay in fractal clusters

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It is shown that optical free-induction decay (OFID) in fractals is governed by a power-law time dependence. The scale invariance of the OFID in the time domain results from the self-similar distribution of the dipolar eigenmodes in fractals.

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A spatial scaling (self-similarity) of fractal clusters results under certain conditions in scaling in the time domain. Martin *et al.*, who studied the dynamics of the sol-gel transition probed by the use of light scattering, reported on the observation of power-law time decay of the intensity autocorrelation function.[1] The power-law decay of the correlation of the detected photons at different instants of time indicates in this case a fractal time set in the scattered field.[1]

Fractal time processes often imply a self-similar distribution of the eigenfrequencies.[2] Optical eigenmodes of fractal clusters (referred to below for the sake of brevity as fractals) were studied in a number of papers.[3–7] In particular, it was shown in Refs. 5–7 that a distribution of the dipolar eigenstates of a random fractal possesses a scaling.

The time decay of the optical free induction (macroscopic dipolar moment) is typically described by an exponential or "stretched" exponential dependence with some characteristic time τ.[8] In the present paper we show that optical free-induction decay (OFID) in fractals is described by a power-law (rather than exponential) time dependence, and, thus, there is no characteristic time scale. The scaling of OFID in fractals is a consequence of the self-similar distribution of the dipolar eigenmodes.

Let us consider a fractal cluster consisting of polarizable particles (monomers). The number of monomers N in the cluster is given by $N = (R_c/R_0)^D$, where R_c is the gyration radius, R_0 is a typical separation between neighbor monomers, and D is the fractal (Hausdorff) dimension. The polarizability of an isolated particle is χ_0. We assume that the particles possess an optical resonance at ω_0 with a high quality factor. If such a cluster is irradiated by an external field with frequency close to ω_0, then large transitional dipole moments are induced on the particles and, accordingly, strong dipole-dipole interaction occurs, resulting in the formation of collective eigenmodes. At $|\omega - \omega_0| \ll \omega_0$ the density of the modes for random fractal clusters with $N \to \infty$ is given by the power-law dependence[5,6]

$$\rho(\omega - \omega_0) \sim N\Delta^{-d_o}|\omega - \omega_0|^{d_o - 1}, \qquad (1)$$

where d_o is the optical spectral dimension ($0 < d_o < 1$) introduced in Ref. 5. The quantity $\Delta = R_m^3\omega_0\langle w_n^2\rangle^{1/2}$ characterizes the width of the spectral range within which the modes are distributed. Here w_n are the eigenvalues of the interaction operator $\hat{W}$, i.e., $\hat{W}|n) = w_n|n)$, and $\hat{W}$ has the following matrix elements in the coordinate representation: $(i\alpha|\hat{W}|j\beta) \equiv W_{\alpha\beta}^{(ij)} = [\delta_{\alpha\beta} - 3n_\alpha^{(ij)}n_\beta^{(ij)}]r_{ij}^{-3}$ if $i \neq j$ and $W_{\alpha\beta}^{(ij)} = 0$ if $i = j$. (The Greek indices α, β label the Cartesian components, summation over repeated Greek indices is implied, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, and $\mathbf{n}^{(ij)} = \mathbf{r}_{ij}/r_{ij}$.) The quantity R_m is a typical charge displacement (which is of order of the size of a monomer) associated with the dipolar moment induced on the monomer.

The interaction operator in the presented form is valid in general if the wavelength λ is much greater than the size of the cluster $kR_c \ll 1$. For the sake of simplicity we assume first that this condition is fulfilled. However, as will be discussed below, the main results obtained in this work remain valid for the arbitrary size of the cluster.

Let us assume that the dipolar eigenmodes are excited under steady-state conditions by a laser beam and that suddenly, at $t = 0$, the excitation is terminated. Then, at $t > 0$, the modes will freely oscillate with their eigenfrequencies. We will consider the system behavior for times t, which are much less than a characteristic time, γ^{-1}, of irreversable relaxation: $\gamma t \ll 1$. At $t = 0$, all the modes are in phase, and the macroscopic dipolar moment is proportional to the number of particles (accordingly, the radiated intensity $\propto N^2$). However, for $t > 0$, since the frequencies of the freely oscillating dipoles associated with different modes are different, the modes gradually become out of phase. First, the modes at the two opposite sides of the spectrum, having the largest difference in frequencies ($\sim 2\Delta$), are dephased, then those which are closer to the central frequency, ω_0, and, therefore, have a smaller frequency difference, and so on. This process of the mode dephasing developing with time results in decay of the macroscopic dipolar moment. The phenomenon, by analogy with magnetic induction decay, has been called optical free-induction decay.[8]

The decay is described in general by the integral

$$f(t) = \int_{-\infty}^{\infty} d\Omega\ \rho(\Omega)\phi(\Omega)\exp\{-i\Omega t\}\ , \qquad (2)$$

where the density of the eigenmodes, $\rho(\Omega)$, is given by

(1), and $\phi(\Omega)$ is the weight that each mode contributes to the macroscopic dipolar moment (Ω is the detuning from the central frequency ω_0). The weight function $\phi(\Omega)$ depends in general on the way the system was excited before $t = 0$.

The main contribution to the integral in (4) is given by the frequency range for which $|\Omega|t < 1$ and $|\Omega|t \sim 1$. Accordingly, if we consider the time for which $\Delta t \gg 1$, then all the frequencies Ω contributing significantly to the integral are such that $|\Omega| \ll \Delta$. The modes for which $|\Omega| \geq \Delta$ become out of phase by the time when $\Delta t \gg 1$ and, therefore, their contribution to the macroscopic dipole is of no importance. We assume that the [redacted] of the eigenmodes have been excited at $t = 0$ and $\phi(\Omega)$ is a reasonably smooth function with a characteristic scale of changing of Δ. Then, for $|\Omega| \ll \Delta$ the weight-function $\phi(\Omega)$ may be approximated by its value at $\Omega = 0$ when performing the integration in (2). Using (1), one obtains the following estimation for $f(t)$ in (2):

$$f(t) \sim N\phi(0)(\Delta t)^{-d_o} \quad (\Delta t \gg 1). \tag{3}$$

Thus, for the time t satisfying to the condition $\Delta t \gg 1$, the decay of the optical free-induction in fractals is characterized by a power-law (rather than exponential) time dependence.

Below we demonstrate this general result for a fractal set of harmonic oscillators. The transitional dipole moment $\tilde{\mathbf{d}}^{(i)}$ induced by an external field $\tilde{\mathbf{E}}^{(0)} = \mathbf{E}^{(0)}\exp(-i\omega t)$ on the ith particle obeys in this case the following system of equations:

$$\ddot{\tilde{d}}_\alpha^{(i)} + 2\gamma\dot{\tilde{d}}_\alpha^{(i)} + \omega_0^2\tilde{d}_\alpha^{(i)} = R_m^3\omega_0^2\left(\tilde{E}_\alpha^{(0)} - \sum_j W_{\alpha\beta}^{(ij)}\tilde{d}_\beta^{(j)}\right). \tag{4}$$

Under steady-state conditions, (4) reduces to the system of algebraic equations

$$d_\alpha^{(i)} = \chi_0 E^{(0)} - \chi_0\sum_j W_{\alpha\beta}^{(ij)}d_\beta^{(j)}, \tag{5}$$

where $d_\alpha^{(i)} = \chi_{\alpha\beta}^{(i)}E_\beta^{(0)}$ is the dipole amplitude, and χ_0 is defined as

$$\chi_0 = (R_m^3\omega_0^2)/[\omega_0^2 - \omega^2 - 2i\gamma\omega]$$
$$\approx (\frac{1}{2}R_m^3\omega_0)/[\omega_0 - \omega - i\gamma]\ .$$

We will use the latter expression for χ_0 below. In the case of a harmonic oscillator, it is valid for $|\omega_0 - \omega| \ll \omega$. Note that Eqs. (4) and (5) describe also a set of two-level particles for which the latter expression for χ_0 is exact if the system is far from saturation.[8]

The solution of (5) for a cluster polarizability, $\chi_{\alpha\beta} = \sum_i \chi_{\alpha\beta}^{(i)}$, in terms of the eigenfunctions of the interaction operator has the form[5,9]

$$\chi_{\alpha\beta} = N\sum_n \frac{(\psi_\alpha|n)(n|\psi_\beta)}{\chi_0^{-1} + w_n}, \tag{6}$$

where

$$|\psi_\alpha) \equiv \frac{1}{\sqrt{N}}\sum_i |i\alpha). \tag{7}$$

Let us assume that at $t < 0$ a system was excited by the field with frequency which was far beyond all the eigenvalues w_n (nonresonant excitation), i.e., $\chi_0^{-1} \gg \Delta$. Then, under steady-state conditions, all of the dipole moments induced on the particles have the same amplitude. For $t = 0$, the dipole values are given by

$$\tilde{d}_\alpha^{(i)}(0) = \chi_0\sum_{j,n}(i\alpha|n)(n|j\beta)E_\beta^{(0)} = \chi_0 E_\alpha^{(0)}.$$

For $t > 0$ the modes freely oscillate (slowly decaying with a characteristic time γ^{-1}) with eigenfrequencies ω_n, so that for $\tilde{d}_\alpha^{(i)}(t)$ one obtains

$$\tilde{d}_\alpha^{(i)}(t) = e^{-\gamma t}\chi_0\sum_{j,n}\exp\{-i\omega_n t\}(i\alpha|n)(n|j\beta)E_\beta^{(0)}, \tag{8}$$

where $\omega_n = \omega_0(1 + R_m^3 w_n)^{1/2}$. Assuming that $R_m^3\Delta \ll 1$, one finds for the eigenfrequencies

$$\omega_n \approx \omega_0 + \Omega_n, \quad \Omega_n \equiv \frac{1}{2}\omega_0 R_m^3 w_n. \tag{9}$$

The macroscopic dipolar moment, $P(t)$, is defined as $P_\alpha(t) \equiv N\mathrm{Re}\langle\tilde{d}_\alpha^{(i)}(t)\rangle$, where the angular brackets denote averaging over an ensemble of clusters. Performing first the averaging over orientations of a cluster as a whole and using (8), we find $P_\alpha(t) = \chi_0 E_\alpha^{(0)}\mathrm{Re}[f\exp\{-\gamma t - i\omega_0 t\}]$, or in the explicit form

$$P_\alpha(t) = \chi_0 E_\alpha^{(0)}e^{-\gamma t}\{f_1\cos\omega_0 t + f_2\sin\omega_0 t\}, \tag{10}$$

where $f \equiv f_1 + if_2$ is given by

$$f(t) = \left\langle\frac{1}{3}N\sum_n\exp\{-i\Omega_n t\}(\psi_\alpha|n)(n|\psi_\alpha)\right\rangle. \tag{11}$$

To obtain (10) and (11), we have taken into account that for a nonresonant excitation at $t < 0$, one can neglect the imaginary part of χ_0. The macroscopic dipole moment in (10) and (11) satisfies the initial conditions $P_\alpha(0) = N\langle d_\alpha^i(0)\rangle = N\chi_0 E_\alpha^{(0)}$ and $\dot{P}_\alpha(0) = 0$.

The function $f(t)$ in (11) determines optical free-induction decay [cf. (2)]. Provided all the modes contribute to the signal with an equal weight (see below), then

$$f = \left\langle\sum_n\exp\{-i\Omega_n t\}\right\rangle = \int\exp\{-i\Omega t\}\rho(\Omega)\ d\Omega\ .$$

As follows from (6) and (7), and (9) and (11), function $f(t)$ can be expressed in terms of the Fourier transform of

$$\chi(\omega) \equiv \chi'(\omega) + i\chi''(\omega) \equiv 1/3\langle\chi_{\alpha\alpha}\rangle\ ,$$

$$f(t)e^{-\gamma t}e^{-i\omega_0 t} = -\frac{2i}{\pi R_m^3\omega_0}\int_{-\infty}^{\infty}\chi(\omega)e^{-i\omega t}d\omega\ , \tag{12}$$

in agreement with the general theory of a linear optical response.[10] Note that $P(t)$ in the form (10) and (11) can also be obtained for a wide band excitation when $\tilde{E}^{(0)}(t) \propto E^{(0)}\delta(t)$ and one has, accordingly,

$$P(t) \propto \int_0^\infty f(\tau)\tilde{E}^{(0)}(t-\tau)d\tau$$
$$\propto f(t) \propto \int \chi(\omega)\exp(-i\omega t)d\omega\ .$$

As follows from comparison of (12) and (2) in both the considered cases, nonresonant steady excitation followed by a sudden termination of the excitation and a wide-band excitation by a δ pulse, a spectral dependence of the product $\rho(\Omega)\phi(\Omega)$ in (2) is given by polarizability $\chi(\omega)$. Note, however, that in general a system can be "prepared" in such a way that only a part of the modes are excited at $t = 0$.

Further simplification in (12) can be done by using the equality $\int_{-\infty}^{\infty}\chi'(\omega)e^{-i\omega t}d\omega = i\int_{-\infty}^{\infty}\chi''(\omega)e^{-i\omega t}d\omega$, which is valid for $t > 0$ and can readily be proved by performing a Fourier transform of χ' and χ'' in (6) and (7). This results in the following formula for $P_\alpha(t)$:

$$P_\alpha(t) = \frac{4\chi_0 E_\alpha^{(0)}}{\pi R_m^3\omega_0}\mathrm{Re}\int_{-\infty}^{\infty}\chi''(\omega)e^{-i\omega t}d\omega. \qquad (13)$$

Since for $\Delta t \gg 1$ the modes for which $|\Omega| \equiv |\omega_0 - \omega| \geq \Delta$ become out of phase, only the central modes with $|\Omega| \ll \Delta$ contibute significantly to the integral in (13) [cf. with the discussion preceding formula (3)]. For random fractals the eigenmodes in the central part of the spectral contour contribute to the imaginary part of the polarizability with an equal weight so that $\chi''(\omega)$ is simply proportional to the density of eigenstates in this region: $\chi''(\omega) = (\pi R_m^3\omega_0/6)\rho(\omega-\omega_0)$.[5] Thus, provided $\Delta t \gg 1$, one can substitute the spectral density $\rho(\omega-\omega_0)$ for $\chi''(\omega)$ in (13). Using (1), (10), and (13), one readily obtains

$$P_\alpha(t) = \frac{2}{3}\chi_0 E_\alpha^{(0)}\mathrm{Re}\left(e^{-i\omega_0 t}\int_{-\infty}^{\infty}\rho(\Omega)e^{-i\Omega t}d\Omega\right)$$
$$\propto t^{-d_0}\ (\Delta t \gg 1)\ , \qquad (14)$$

in agreement with that predicted by (3).

The intensity radiated by a freely decaying system is determined by the expression $I \equiv \langle[\mathrm{Re}\sum_i \tilde{d}_\alpha^i]^2\rangle$. Using (6) and (7), and (10) and (11), one finds for I

$$I = \frac{1}{2}[\chi_0 E^{(0)}e^{-\gamma t}]^2F^2, \qquad (15)$$

where $F^2 \equiv F_1^2 + F_2^2$ and

$$F_1^2 = \left\langle\left(\frac{1}{3}N\sum_n \cos(\Omega_n t)(\psi_\alpha|n)(n|\psi_\alpha)\right)^2\right\rangle,$$

$$F_2^2 = \left\langle\left(\frac{1}{3}N\sum_n \sin(\Omega_n t)(\psi_\alpha|n)(n|\psi_\alpha)\right)^2\right\rangle. \qquad (16)$$

Note that in general $F^2 \neq |f|^2$ [cf. (11) with (16)].

In our simulations we have examined ballistic cluster-cluster aggregates ($D \approx 1.9$). The clusters were generated using the Monte Carlo method and well-known procedures. Then, the clusters have been subjected to dilution (random decimation), which consists of the following:[5] The ith ($i = 1, 2, ..., N$) monomer is randomly retained in the cluster with some small probability β or removed with probability $1-\beta$. The fractal as a whole is in turn reduced in size $\beta^{-1/D}$ times, so that the value of R_0 remains the same. This procedure reduces the total number of particles on average by a factor of β. The fractal structure at small scales is significantly simplified by the procedure. The important aspect is that the resultant (diluted) cluster is characterized by the same fractal dimension as the original one. The clusters ($N = 32768$) in our simulations have been subjected to the dilution with $\beta = 64$ so that after the dilution each cluster consists of $N = 512$ particles. Averaging over an ensemble of 100 random clusters has been conducted. In the simulations, the eigenvalues and eigenvectors of the interaction operator $\hat{W}$ for the generated aggregates have been found and then the macroscopic dipolar moment, $P(t)$, and the intensity radiated by freely decaying fractals, $I(t)$, have been calculated using (10) and (11) and (15) and (16), respectively.

In Figs. 1 and 2 we present the results of the simulations of optical free-induction decay in fractals. The dependence $f_1(t)/N$ on t and a double-logarithmic plot of $F(t)/N$ are shown in Figs. 1 and 2, respectively. As follows from (11) and (16), $f_1(0)/N = F_1(0)/N = 1$, and $f_2(0) = F_2(0) = 0$. Thus, at $t = 0$, one has $f = N$ and $F^2 = N^2$, i.e., all the dipolar moments are in phase. At $t > 0$, the quantities $f_1(t)$ and $F(t)$ decay and exhibit the power-law dependence ($\propto t^{-d_0}$) within a certain time

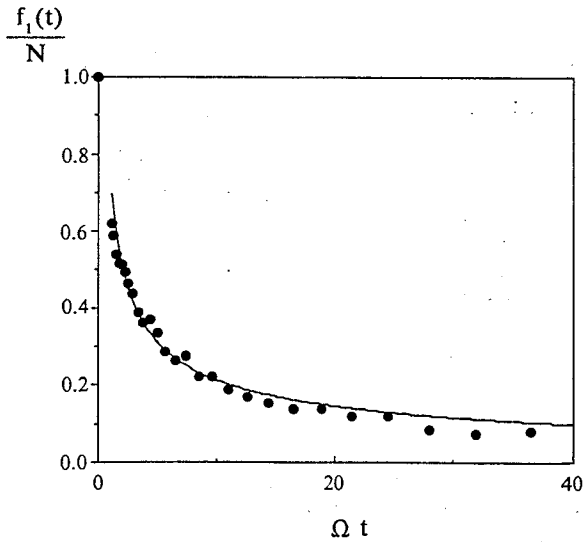

FIG. 1. Function $f_1(t)/N$ describing the decay of the optical free induction. The solid line is the $0.75(\Omega t)^{-0.55}$ dependence. The exponent gives the value of the optical spectral dimension $d_0 = 0.55$. The simulations are made for ballistic cluster-cluster aggregates having the fractal dimension $D \approx 1.9$.

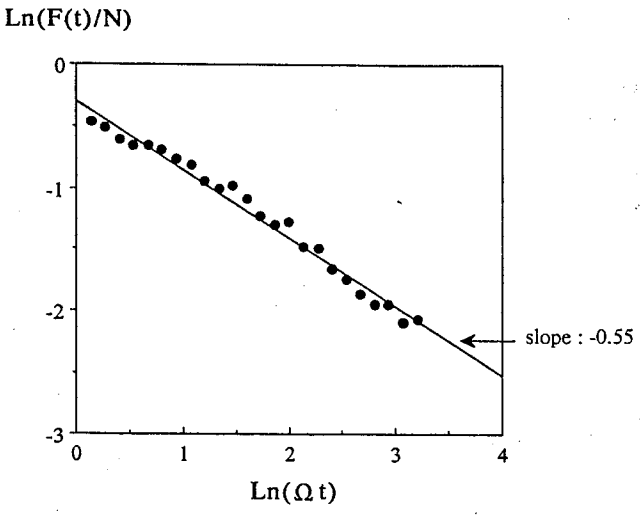

FIG. 2. Double-logarithmic plot of $F(t)/N$ characterizing a square root of the intensity (per particle) radiated by ballistic cluster-cluster aggregates. The slope obtained is equal to $-d_o$, where $d_o = 0.55$ is the optical spectral dimension.

interval, the scaling region. For large times (not shown in the figure) $f(t)$ falls to zero, reflecting the complete dephasing of the dipole moments and the radiated intensity, $I \propto F^2$, tends to the nonzero steady-state value $I_s \propto F_s^2 \sim F^2(0)/N = N$. The quantity $F_s^2 \sim N$ characterizes the intensity radiated by N independent dipoles.

The exponent in the power-law dependence, $f_1(t)$ and $F(t)$, obtained for the scaling region in the simulations is -0.55. In our work[13] we reported for the ballistic cluster-cluster aggregates the value $d_0 = 0.53 \pm 0.1$. Thus, the exponent found in the simulations coincides within the error interval with $-d_0$, in accordance with the theoretical predictions [see Eqs. (3) and (14)].

We have assumed above that the size of a cluster is smaller than the wavelength. However, since the dipolar modes are localized in fractals in regions that are significantly smaller than the wavelength,[5,7,11] the reduced Hermitian operator, $V_{\alpha\beta}^{(ij)} = W_{\alpha\beta}^{(ij)}$, if $kr_{ij} < 1$, and $V_{\alpha\beta}^{(ij)} = 0$, otherwise, can be used instead of the exact non-Hermitian operator of the dipole-dipole interaction of particles in a cluster of an arbitrary size.[12] Accordingly, the scaling properties of collective dipolar excitations remain valid for a cluster larger than the wavelength. In particular, the power-law decay of the macroscopic dipole moment occurs in large fractal clusters as well. Note, however, that the intensity radiated by a large cluster at small values of t is proportional to $N(kR_0)^{-D}$ for $D < 2$ and $N^{2-2/D}(kR_0)^{-2}$ for $D > 2$, rather than N^2, as in the case of a cluster smaller than the wavelength.[12,14] This is because the complete coherence for large clusters occurs only within a range of an order of the wavelength.

The dipolar eigenmodes, which are considered above, are non-Goldstone-type excitations. As shown in our work[6], Goldstone-type vibrational excitations having an energy gap in the spectrum, e.g., optical phonons, also possess a self-similar distribution of eigenmodes in the form (1). The exponent d_o in this case is half the spectral dimension $\bar{\bar{d}}$ (i.e., $d_o = \frac{1}{2}\bar{\bar{d}}$), which determines the scaling of gapless Goldstone-type vibrational excitations,[15] e.g., acoustic phonons.[15] The theory of OFID developed in this work can also be applied to the decay of the Goldstone-type optical phonons. Accordingly, the power-law OFID can be observed for the optical phonons on a fractal lattice.

To summarize, power-law time decay of the optical free-induction is shown to occur in fractals, resulting in scale invariance of the macroscopic dipolar moment and the radiated intensity in the time domain. The temporal scaling is a consequence of the self-similar distribution of the dipolar eigenmodes in the frequency domain.

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