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SCATTERING FROM FRACTALS

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INTRODUCTION AND SCOPE

The realization that structures in Nature often can be described by Mandelbrot's "fractals" has led to a revolution in many areas of physics. The interaction of waves with fractal systems has, understandably, become intensely studied since scattering is the method of choice to probe delicate fractal structures such as chainlike particle aggregates. Not all of these waves are electromagnetic: neutron scattering, for example, is an important complementary tool to structural studies by x-ray and light scattering. Since the phenomenology of small-angle neutron scattering (SANS), as it is applied to fractal systems, is identical to that of small-angle x-ray scattering (SAXS), it falls within the scope of this Working Paper.

A growing body of work indicates that fractal structure may be important in electrochemical reactions at rough surfaces. Although this area has far reaching consequences, it is not addressed here.

The importance of scattering from fractals can be illustrated by the example of nuclear winter. Studies on the effects of loading the atmosphere with light-absorbing particles by the nuclear ignition of cities supposed that the soot material could be assumed spherical in morphology--the so-called spherical chicken approximation. All sensible aerosol scientists know that soot particles are highly ramified; in fact, physicists have been claiming that soot is fractal. It is a characteristic of fractals to influence large volumes of space for their small mass, hence the specific extinction cross sections for fractal aggregates are large as well. (Although specific absorbance is relatively insensitive to mass for fractal dimensions less than 2, recent calculations by Jenny Nelson show that the scattering efficiency is not.) Here fractal structure is not in our favor because fractal soot as opposed to spherical soot implies a deeper global cooling. Fortunately, the water condensation on soot may mitigate these effects by collapsing the soot aggregates by capillary pressure, as shown by Mulholland and coworkers at NIST.

Other, less morbid examples of applied fractal scattering exist, such as in monitoring the growth of a surface layer by analyzing scattering from surface roughness. Fractal concepts greatly simplify the analysis, making in situ scattering a feasible quality control technique.

Mass fractals are characterized by a geometric scaling between mass M and radius R, $M \sim R^D$, implying an algebraically decreasing density,

$$\rho \sim M/V \sim R^D/R^3 \sim R^{D-3} \quad .$$

Here D is the mass fractal dimension, which for connected objects ranges from 1 to 3. Surface fractals obey a similar relation between area S and the radius of the attending body, but it is more appropriate to consider the apparent or measured area as a function of the "tile" size r used to cover the body (the resolution of the measurement),

$$S(r) \sim S_o\, r^{2-D_s} \quad ,$$

where D_s is the surface fractal dimension ranging from 1 to 2, and S_o is a prefactor relating surface area measurements for different resolutions r. The lack of an intrinsic length scale in these power-law relations implies "self similarity" or "self affinity", i.e. invariance under dilation.

The scattering properties of fractals have been reviewed by Martin and Hurd.[1] The scattered intensity is simply the Fourier transform of the density correlation function $g(r) = \langle\rho(0)\rho(r)\rangle$,

$$I(q) = \int_V g(r)\, e^{iq.r}\, d^3r \quad ,$$

where V is the volume of the illuminated scatterer(s) and $q = 4\pi/\lambda \sin(\theta/2)$ is the scattering wave vector (λ is the wavelength of the radiation and θ is the scattering angle). Quite generally, the power-law density of a mass fractal implies a scaling form for g(r), which is (ignoring prefactors)

$$g(r) = r^{D-3}\, h(r/R) \quad ,$$

where h(r/R) is a function characterizing the finite size of the fractal object with characteristic size R. For small r/R, h(r/R) approaches 1, and for large r/R it approaches 0. These assumptions imply an intensity,

$$I(q) \sim q^{-D} \qquad (qR \gg 1).$$

Surface fractals, with roughness on a length scale below R_s, scatter as a power law, too, due to the fact that the correlation function takes the form

$$g(r) \sim 1 - [S(r)/V]\, r + \ldots \qquad (r \ll R_s) ,$$

hence

$$I(q) \sim [S_o/V]\, q^{-(6-D_s)} \quad (qR_s \gg 1).$$

Note that the specific surface area prefactor survives in the final intensity. Similarly, in the case of a (mass) fractal aggregate of N identical particles, the prefactor to q^{-D} involves N.

STATE OF THE ART

Mass Fractals. Soon after T.A. Witten pointed out the possibility of using scattering to probe the power-law correlations of mass in a fractal, the program was accomplished by D.W. Schaefer and coworkers for fractal aggregates. However, it became evident that a detailed theoretical approach would be necessary to explain the scattering from metallic fractals, such as gold colloids studied by D. A. Weitz et al. at Exxon. Numerical and analytical calculations had been attempted for aggregates of few primary particles by F. Borghese and others, but only recently have the simplifications offered by fractal geometry been fully exploited.[2] Confirmation of our understanding still relies heavily on computation.

Even for nonabsorbing materials, the mass-fractal scattering problem cannot be considered completely solved, particulary with respect to aggregates since there is no theory for their finite size function h(x) in spite of empirical and "reasonable" approximations. Nevertheless, the list of measurable quantities from scattering by aggregates is impressive[3] and still growing with further theoretical understanding. Teixeira recently showed how the mass N of a fractal aggregate can be measured by the scattering prefactor. Several clever regular-fractal models have appeared by P.W. Schmidt and D.L. Jaggard that can be solved exactly as a check on theory.

Surface Fractals. Fractal concepts rescued a great backlog of ill-understood scattering data from the trash can after the publication of the seminal paper on surface-fractal scattering.[4] Although it has taken some time to sort out the details, such as the Wong-Bray prefactor relating to surface area recently verified experimentally by Hurd et al., the fractal school fits in with more traditional developments[5] of rough surface scattering. This connection has been advanced recently by S. Sinha et al. and by P.-z. Wong and A. Bray.

Dynamic Scattering. The unique structure of fractal materials has deep implications on their vibrational modes. For vibrational wavelengths encompassed by the scaling regime, the normally freely propagating phonons become localized "fractons," as originally proposed in 1982 by S. Alexander and R. Orbach. Such localized modes are a bottleneck to the propagation of heat, and it is thought that fractons explain the thermal conductivity anomalies of some disordered materials. Light and neutron scattering studies from aerogels and silica aggregates, notably by E. Courtens and coworkers, have convinced many that this is indeed the case. Detailed lineshapes have been put forward by Orbach and Entin-Wohlman to fit the data of Courtens.

A separate area having to do with dynamic scattering is in the study of turbulence using dynamic light scattering. Turbulence is thought to be a self-similar cascade of ever smaller eddies; dynamic light scattering has been used by W. Goldburg et al. to study the velocities present in the cascade.

Multiple Scattering. Many fractal materials are porous, disordered ceramics and glasses, which tend to scatter radiation very strongly. Techniques have been developed to gain meaningful information from experiments in the strongly multiple-scattering limit. SANS from ceramic green bodies has been studied by K.A. Hardman-Rhyne and N.F. Berk from this point of view to obtain the characteristics of the interstitial pores. While ceramic green bodies may not be fractal in structure, similar techniques have been proposed for the study of phase-separation-leached glasses (e.g. Vycor) that are reputedly fractal. Moreover, dynamic techniques have been developed to measure diffusive transport through porous glasses (or in any situation where multiple scattering dominates) by groups at Exxon and the University of

Pittsburgh, wherein the dynamic structure factor is averaged over many scattering wave vectors.

Instruments. Although small-angle x-ray scattering and light scattering instruments are widely available and well refined, recent advances in their capabilities have contributed to our understanding of fractal systems.

A small-angle light scattering instrument using an array of annular solid state detectors has been demonstrated[6] that reaches extremely low angles, $\theta \sim 0.2^\circ$ or $q = 400\ cm^{-1}$, corresponding to a spatial resolution of 25 microns (an order of magnitude greater than typical). The instrument has been used in the study of fractal aggregate structure.

For x-ray work, synchrotron radiation appears to be the wave of the future. A Bonse-Hart camera that has been implemented at Brookhaven by the Exxon group[7] has shown its utility in obtaining information on fractal systems over a very wide range of lengths.

Neutron scattering is re-emerging in the USA, but has not caught up with Europe where it enjoys great health. State-of-the-art small-angle and inelastic scattering are available at ILL in Grenoble, and several other centers in Europe are operating reliably. In the United States, facilities for SANS are available at Argonne (IPNS), Los Alamos (LANSCE), the University of Missouri (MURR), and the National Institute of Standards and Technology (NIST). SANS at LANSCE is new and suffering the usual ills of a new user facility; the highly reliable MURR is not funded as a user facility but is available on a case-by-case basis. HFIR at Oak Ridge has been restarted and should be at full power soon. Chalk River in Canada and NIST in Gaithersburg have the capabilities to perform inelastic scattering experiments.

EMERGING FUTURE DIRECTIONS

A number of new theoretical techniques have emerged to characterize fractals in terms of moment expansions, including multifractals, wavelet transforms and multipole expansions. The last has proven effective in the analysis of scattering data,[8] but it is too early to say what new viewpoint on scattering is offered by other expansions. Multifractal analysis is particularly appealing for insight into the outstanding question of "DLA" structure and the "fractal antenna" problem. In a similar development, the use of higher order correlation functions has been suggested by Z.-Y. Chen et al. as a means to squeeze more information about fractals from dynamic light scattering.

The proliferation of multifractal analysis underscores the fact that typical fractal systems cannot be characterized by a single parameter such as the fractal dimension. The simplest scaling behavior involves prefactors, at least. For surface scattering, this prefactor S_0 contains useful information concerning specific surface area (at a given resolution), whereas for mass fractal scattering the prefactor relates to the mass. There have been no experiments to verify the latter relationship in detail.

Regarding instrumentation, there is great room for improvement. Synchrotron sources are currently used by few although they offer the greatest experimental advantage in exploring scaling structures over a wide range of lengths. Neutron scattering is crucial for the fundamental study of certain materials, especially ceramics and glasses involving an ever larger number of researchers. In the rapidly developing area of the dynamics of fractals, demand will probably increase for inelastic neutron scattering. European neutron technology currently surpasses the US in most respects.

Many observers are reluctant to recognize fractals as a legitimate area of physics, materials science or chemistry. Indeed, unlike its role in mathematics, fractals is not really a "field" purveying a well defined set of physical rules, such as thermodynamics. Instead it is a useful "category" for grouping together otherwise unrelated problems. The subsequent cross fertilization is the real value in fractals.

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