Complex dielectric response of metal-particle clusters

P. M. Hui
Department of Physics, Ohio State University, Columbus, Ohio 43210

D. Stroud
*Department of Physics, Ohio State University, Columbus, Ohio 43210**
and Laboratoire de Physique Theorique des Liquides, Universite de Paris VI, 4, Place Jussieu, 75005 Paris, France
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We discuss the complex dielectric response of metal-particle clusters. A self-consistent theory is introduced, which leads to a cubic equation for the cluster dielectric constant. The model is one in which the particles form fractal clusters. Both the electric and magnetic dipole absorptions are found to be enhanced by this fractal clustering. In the low-frequency and long-wavelength limits, analytic expressions for the enhancement factors are obtained. The model is applied to small particle composites, for which a red shift in the Mie resonance is obtained. For superconducting particles, the absorption in the superconducting state α_s is found to be greater than that of the normal state at frequencies slightly higher than the gap, provided the fractal dimension is so low that the electric dipole absorption predominates. At very low frequencies, fractal clustering also leads to an enhancement in the diamagnetic susceptibility of superconducting small-particle composites.

I. INTRODUCTION

The electromagnetic properties of small metal-particle composites have attracted much attention in recent years.[1] A particularly intriguing experimental observation is that far-infrared absorption in such composites is vastly enhanced over predictions based on the assumption that the composites are random and can be treated with classical electromagnetic theory in the long-wavelength limit.[2–4] The unsuccessful theories treat the composite as a homogeneous dielectric with an effective dielectric function usually calculated using either the Maxwell-Garnett theory (MGT)[5] or the effective-medium approximation (EMA).[6] A number of mechanisms have been proposed for this anomalous enhancement, namely, eddy-current losses in small metal particles,[7] distribution of particle sizes,[8] absorption by oxide layers surrounding the particles,[9] absorption due to size quantization of electronic energy levels in small metal particles,[10] and clustering of small metal particles into clumps[11] to an extent vastly in excess of what would be expected based on an uncorrelated distribution of small metal particles in the composite.

Recent experimental work[12] has made the clustering explanation seem much more plausible than any other. A concurrent theory[13] has shown that the anomalous absorption can be accounted for if certain quite general geometrical assumptions are made about clusters within the composite. The purpose of this paper is to show that another kind of cluster geometry, namely, fractal clusters, can also produce enormous enhancement of far-infrared absorption by small metal particles. Fractal clusters have been reported in colloidal gold suspensions by Weitz and collaborators.[14] In addition, such clusters are the natural results of many growth processes resulting from irreversible kinetic aggregation.[15] While the particular small metal-particle composites for which infrared studies have been conducted do not show evidence of fractal clusters, such clusters must exist in many experimental situations, possibly including such exotic materials as interstellar dust.

Our model calculations are based on a differential effective medium approximation previously applied to the conductivity of porous rocks.[16] The complex dielectric function for the cluster in this approach is determined by a cubic equation. The composite is constructed from the cluster by embedding the fractal clusters in an insulating host, and the absorption coefficient is determined by applying the Mie theory to a suspension of such clusters. In the low-frequency and long-wavelength limit, both electric and magnetic dipole absorptions per unit volume of metal are found to be enhanced relative to those of isolated particles. The enhancement factors are $(R/a)^{(5/2)(3-d_f)}$ and $(R/a)^{(1/2)(1+d_f)}$, respectively, where a is the particle radius, R the cluster radius, and d_f the fractal dimension of the cluster.[17] Isolated fractal clusters produce a strong red shift in the Mie resonance of the cluster relative to that of isolated particles. This red shift is not experimentally observed, but may be less important than meets the eye, because only a few fractal clusters are needed to produce a huge far-infrared absorption relative to isolated particles, while the spectral weight of the Mie resonance may still be dominated by isolated particles.

Finally, we also obtain the intriguing result that clusters with a fractal dimension less than $\frac{5}{3}$ are not dense enough to exclude electromagnetic radiation—that is, their electromagnetic skin depth increases faster than their radius.

The same approach can also be applied to superconducting clusters. If the metal particles in the superconducting cluster are assumed to be described by the Mattis-Bardeen dielectric function, then agreement with experiment in the superconducting case implies that the far-infrared absorption must be dominated by *electric di-*

pole absorption. Magnetic dipole absorption (i.e., eddy current absorption) usually produces much enhancement over electric dipole absorption in isolated particles, but electric dipole absorption can exceed magnetic in large clusters of low fractal dimension. Such clusters can therefore explain both the anomalous enhancement in the normal state and the relative magnitudes of superconducting and normal absorption observed experimentally. Our conclusions are thus similar to those drawn by Curtin and Ashcroft[13] for a distribution of nonfractal clusters whose concentrations span the percolation threshold.

We turn now to the body of the paper. Section II presents our model and describes some analytic results. The model is applied to dilute metal-insulator composites in Sec. III. A brief discussion follows in Sec. IV.

II. THE MODEL

All the essential results of our model flow from a picture of a metallic cluster whose conductivity is a monotonically decreasing function of radius (or number of cluster particles). The basis idea is to design a cluster which is self-similar on the average, whose metallic portion is connected at every stage of its evolution, and which therefore remains conducting no matter how small its metallic volume fraction. One such cluster is shown schematically in Fig. 1 at various stages in its development. It happens to be two dimensional but a three-dimensional version can easily be designed also. As illustrated, the cluster's linear dimension and metallic mass at the nth stage are, respectively,

$$R=2^n a \ , \qquad M=\rho(3/4)^n R^2 \ , \tag{1}$$

where a is the radius of an elementary square and ρ is the mass density of the metallic component. The fractal dimension is

$$d_f=2+\ln\tfrac{3}{4}/\ln 2=1.585 \ . \tag{2}$$

The conductivity of this cluster can be estimated using the two-dimensional effective medium approximation repeatedly; the result is

$$\sigma=\sigma_0(2f-1)^n=\sigma_0(\tfrac{1}{2})^n \ , \tag{3}$$

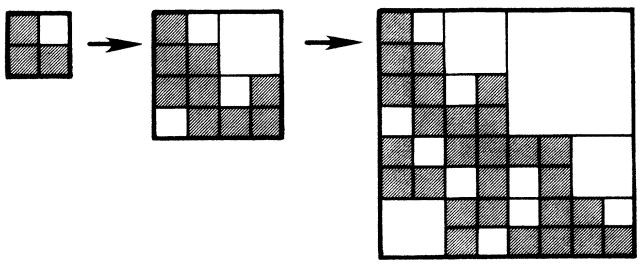

FIG. 1. Schematic diagram showing one possible way of building up a fractal cluster. In each step, the linear dimension is doubled and $\frac{3}{4}$ of the new volume is filled by the clusters of the previous step.

where $f=\frac{3}{4}$ is the areal fraction of metal in the basic cluster and σ_0 is the conductivity of the metal component. Thus the conductivity can be expressed as a power of the radius,

$$\sigma=\sigma_0(R/a)^{-1} \ . \tag{4}$$

The power -1 is special to the fractal dimension of the cluster and the amount by which the area is rescaled at each iteration, but the variation of conductivity as an inverse power of the linear dimension is general for fractal clusters. Within the effective medium approximation the cluster conducts at any stage n, no matter how small the areal fraction of metal.

Next we outline an apparently more exact procedure for generating self-similar (or other) clusters, using a differential effective medium approach in three dimensions. Consider a cluster of radius R, with volume fraction of metal $f(R)$ and of insulator $1-f(R)$. We start by adding metal and insulator to the cluster in such a way that the radius is increased by an amount δR. With R', V', and M' denoting the radius, volume, and metallic mass of the enlarged cluster, we have

$$R'=R+\delta R \ , \tag{5}$$

$$V'=A(R+\delta R)^3 \ , \tag{6}$$

$$M'=\rho f(R+\delta R)V' \ , \tag{7}$$

where ρ is the mass density of metal, and A is a constant ($4\pi/3$ for a sphere). For simplicity, we assume here that the insulator is massless; the same result [Eq. (12)] is found, however, if we consider a finite density for the insulator. To first order in δR, the change in volume δV and in mass δM are thus

$$\delta V=3AR^2\delta R \ , \tag{8}$$

$$\delta M=\rho[3AR^2 f(R)\delta R+AR^3 f'(R)\delta R] \ , \tag{9}$$

where $f'(R)=df(R)/dR$.

To make the theory self-consistent, we divide δV into two parts, δV_1 and δV_2. δV_1 is chosen to contain all the added metal, and to have the same metal concentration $f(R)$ as the cluster to which it is being added:

$$\frac{\delta M_1}{\delta V_1}=\rho f(R)=\frac{\rho}{\delta V_1}[3AR^2 f(R)\delta R+AR^3 f'(R)\delta R] \ . \tag{10}$$

Equation (10) implies that the remaining volume δV_2, which contains only insulator, is given by

$$\delta V_2=-\frac{f'(R)}{f(R)}AR^3\delta R \ . \tag{11}$$

The *volume fraction* of added insulator, η, is given by

$$\eta\equiv\frac{\delta V_2}{V'}=-\frac{f'(R)}{f(R)}\delta R \ . \tag{12}$$

An application of the EMA leads to a new conductivity of the enlarged cluster,

$$\sigma(R+\delta R)=\sigma(R)(1-\tfrac{3}{2}\eta) \ . \tag{13}$$

Equation (13) is an exact expression to order η for the change in conductivity due to an infinitesimal addition of insulator, so long as the conductivity of the cluster is isotropic before and after the addition (it does not assume that the insulator in δV_2 is added in the form of spheres, or any other particular shape).

Substitution of (12) into (13) results in a differential equation for $\sigma(R)$,

$$\frac{d\sigma(R)}{dR}=\frac{3}{2}\sigma(R)\frac{f'(R)}{f(R)}\ , \tag{14}$$

which can be integrated to give

$$\frac{\sigma(R)}{\sigma(R_0)}=\left[\frac{f(R)}{f(R_0)}\right]^{3/2}\ . \tag{15}$$

If, in particular, a is taken to be the radius of a single small metal particle, then $\sigma(a)$ is the conductivity of such a particle and $f(a)$ is unity, whence

$$\sigma(R)=\sigma(a)[f(R)]^{3/2} \tag{16}$$

which gives the conductivity of a cluster of radius R.

At finite frequencies, the far-infrared absorption is described by a complex cluster dielectric function $\epsilon(R)$, for which a differential equation similar to (14) can also be derived. To first order in η, one obtains the following *exact* equation for the change in the dielectric function of the cluster:

$$\epsilon(R+\delta R)-\epsilon(R)=3\eta\epsilon(R)\frac{\epsilon_i-\epsilon(R)}{\epsilon_i+2\epsilon(R)}\ , \tag{17}$$

where $\epsilon(R)$ is the cluster dielectric function at radius R and ϵ_i is the dielectric constant of the insulator. Substituting for η, we find a differential equation,

$$\frac{d\epsilon(R)}{dR}=-\frac{3f'(R)}{f(R)}\epsilon(R)\frac{\epsilon_i-\epsilon(R)}{\epsilon_i+2\epsilon(R)}\ , \tag{18}$$

which can be integrated to give

$$\frac{\epsilon(R)}{\epsilon(a)}\left[\frac{\epsilon_i-\epsilon(a)}{\epsilon_i-\epsilon(R)}\right]^3=\frac{1}{[f(R)]^3}\ . \tag{19}$$

Equation (19) is a cubic equation for $\epsilon(R)$. Similar equations have been obtained in a differential effective medium treatment of sedimentary rocks.[16] Note that Eqs. (15) and (19) do not depend on the validity of the MGT or EMA at large η, since η is always small in this treatment.

We now specialize this treatment to composites in which the metal particles are distributed in fractal clusters. For a fractal cluster of radius R and fractal dimension d_f, the volume V and mass M are given (in three dimensions) by

$$V_{\rm fr}=AR^3,\quad M_{\rm fr}=BR^{d_f}\ , \tag{20}$$

where A and B are constants. The volume fraction $f_{\rm fr}(R)$ of metal in the cluster is thus

$$f_{\rm fr}(R)=\left[\frac{R}{a}\right]^{d_f-3}\ , \tag{21}$$

from which, using Eqs. (15) and (19),

$$\sigma(R)=\sigma(a)\left[\frac{R}{a}\right]^{-(3/2)(3-d_f)}\ , \tag{22}$$

$$\frac{\epsilon(R)}{\epsilon(a)}\left[\frac{\epsilon_i-\epsilon(a)}{\epsilon_i-\epsilon(R)}\right]^3=\left[\frac{R}{a}\right]^{3(3-d_f)}\ . \tag{23}$$

Equation (22) shows how the conductivity of a cluster decreases with increasing radius. It is this decrease in conductivity which leads to an enhancement of electric dipole absorption. The magnetic dipole absorption is enhanced because of the increase of radius of the cluster relative to that of single particles. The remaining Eq. (23) determines $\epsilon(R)$ and can easily be solved numerically.

We now consider several results following from Eq. (22). First, in the low-frequency and long-wavelength limit, the electric dipole absorption of the cluster per unit *volume* is proportional to ω^2, where ω is the frequency, and inversely proportional to the conductivity. The electric dipole absorption α_e of the composite per unit *mass of metal* is given to within a proportionality constant by

$$\alpha_e\sim\frac{\omega^2}{\sigma(a)}R^{(3/2)(3-d_f)}R^3R^{-d_f}\sim\frac{\omega^2}{\sigma(a)}\left[\frac{R}{a}\right]^{(5/2)(3-d_f)} \tag{24}$$

which shows that α_e is increased by a factor of $(R/a)^{(5/2)(3-d_f)}$ relative to the unclustered absorption when the particles form fractal clusters in a composite material. This factor can be enormous for a large cluster or small d_f.

Similar considerations in the same limit for the magnetic dipole absorption α_m per unit mass of metal give

$$\alpha_m\sim\omega^2\sigma(R)R^2R^{3-d_f}\sim\omega^2a^2\sigma(a)\left[\frac{R}{a}\right]^{(1/2)(1+d_f)}\ . \tag{25}$$

The enhancement by a factor of $(R/a)^{(1/2)(1+d_f)}$ results from clustering of particles. In all cases, the increase in cluster radius more than compensates for the decrease in cluster conductivity (which tends to reduce magnetic dipole absorption).

Finally, the electromagnetic skin depth $\delta(R)$ of the cluster is inversely proportional to the square root of the conductivity $\sigma(R)$. If $\delta(a)$ is the skin depth of a fractal cluster of radius R, then it follows from Eq. (22) that

$$\delta(R)/R=\delta(a)/a\,(R/a)^{(1/4)(5-3d_f)} \tag{26}$$

which implies that if $d_f<\frac{5}{3}$, the cluster is not sufficiently dense to screen out an applied electromagnetic field: $\delta(R)/R\to\infty$ as $R\to\infty$. In Eq. (26) $\delta(a)$ is the skin depth of a single metallic particle of frequency ω.

III. APPLICATION TO COMPOSITES

To illustrate the effects just described, we model a composite by embedding fractal metallic clusters in an insulating host. For simplicity, we take the dielectric constant of the insulator to be unity. Equation (23) can then be solved for the cluster dielectric constant in terms of R and d_f.

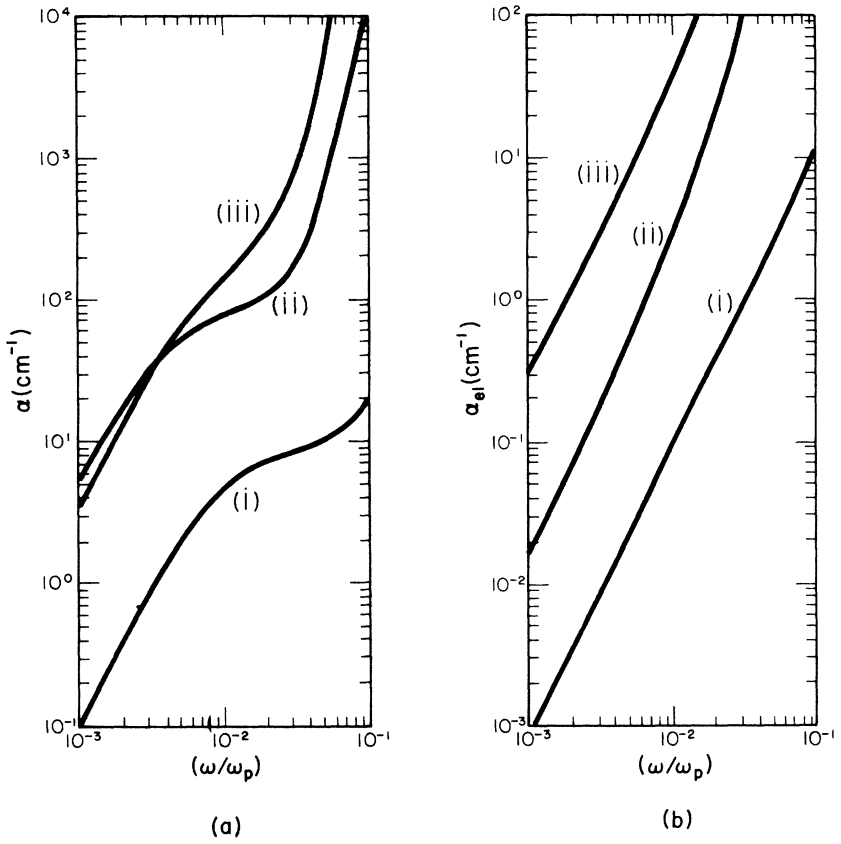

FIG. 2. Absorption coefficient in the presence of fractal clustering. In (a) both electric and magnetic dipole absorption are included and in (b) only the electric dipole absorption is included. Curves (i): no clustering. Curves (ii): $R=10a$ and $d_f=2.5$. Curves (iii): $R=10a$ and $d_f=2.0$. R is the radius of the cluster and d_f is the fractal dimension; a is the radius of a single metal particle.

The extinction coefficient can be calculated from the Mie theory. If f_{cl} is the concentration by volume of clusters in the composite, and the clusters are assumed all identical and spherical, then the extinction coefficient α_{tot} is given by the optical theorem:[18]

$$\alpha_{tot}=\frac{4\pi}{k^2}\mathrm{Re}S(0)\ , \tag{27}$$

$$S(0)=\tfrac{1}{2}\sum_{l=1}^{\infty}(2l+1)(a_l+b_l)\ , \tag{28}$$

where $S(0)$ is the forward scattering amplitude and a_l and b_l are the electric and magnetic multipole coefficients describing scattering from the clusters. These are expressed in terms of spherical Bessel functions (exact expressions may be found, for example, in Ref. 7). While the extinction is due to both absorption and scattering, for the particles of greatest interest (typically of radius 100 Å or so) the extinction is entirely due to absorption in the long-wavelength limit.

Model calculations based on this approach are shown in Fig. 2. The metallic component is assumed to be described by a Drude dielectric function

$$\epsilon(\omega)=1-\frac{\omega_p^2}{\omega(\omega+i/\tau)}\ , \tag{29}$$

where ω_p is the plasma frequency and τ is a characteristic relaxation time. We use parameters $\omega_p\tau=100$ and $\omega_p a/c=1$, where c is the speed of light. The latter condition would correspond to approximately 100-Å Al particles; however, such particles would probably have relaxation times somewhat shorter than that assumed by $\omega_p\tau=100$ because of surface scattering from the boundary of the small metal particles. The results presented are based on the Mie expansion [Eqs. (27) and (28)] including both a_1 and b_1 in the full Bessel-function form. In all calculations, the concentration by volume of metal in the composite is 0.01.

Figure 2(a) shows the absorption due to a_1 and b_1, while for comparison, Fig. 2(b) shows the absorption due to a_1, i.e., electric dipole, only. The curve labeled (i) denotes no clustering, while (ii) and (iii) correspond to different fractal dimensions. For the fractal dimensions shown, absorption is dominated by the magnetic dipole coefficient b_1, as would be predicted from the asymptotic formulas (24) and (25). The enhancement factors obtained numerically at very low frequencies indeed correspond to those calculated analytically.

The low-frequency limit requires that the electromagnetic skin depth be large in comparison with particle size. This requirement is not always satisfied as the frequency

increases, and the magnitude of the enhancement therefore varies from that predicted by Eqs. (24) and (25). This deviation from asymptotic behavior can be clearly seen in the figures.

Figure 3 shows the behavior of the imaginary part of the composite dielectric constant at higher frequencies. Here, we choose the parameters such that the "quasistatic limit" is valid (we assume $\omega_p\tau=100$ and $\omega_p a/c \ll 1$), and consider the effects of fractal clustering in this limit. All the curves in the figure describe a composite of 1% by volume of metal particles. Curve (a) is the simple Maxwell-Garnett approximation, equivalent to assuming no clustering. Curve (b) shows the effect of fractal clustering in which $d_f=2.5$ and the radius of the cluster is $R=10a$. We calculate the dielectric constant of the clusters by solving Eq. (23) and then calculate the composite dielectric constant in the Maxwell-Garnett approximation. For comparison, in curve (c), we show a similar calculation in which the EMA is used to calculate the cluster dielectric constant, while the MGT is used to compute the composite dielectric constant from that of the cluster.

The most striking feature of Fig. 3 is the strong red shift in the Mie resonance of the fractal cluster, relative to that calculated from the Maxwell-Garnett approximation (appropriate to isolated metal particles in an insulating host). As noted in the Introduction, such a strong red shift has not been detected in any reported experiments, though small red shifts have been described. But if the metal in the composite is largely present as isolated particles, then most of the oscillator strength of the Mie resonance will *not* be red shifted, while the few metal particles that *are* grouped into fractal clusters will predominate in the absorption at far-infrared frequencies. Note also that a range of fractal dimensions and cluster radii will further broaden that part of the Mie peak due to clusters. If the clusters are not in the form of fractals, then curve (c) of Fig. 3 shows that the Mie resonance due to such clusters is much broadened even without considering distributions of cluster sizes.

It is of interest to contrast the absorption of fractal clusters in the superconducting (S) and normal (N) states.[19] Experiments show a striking difference in far-infrared absorption between N and S particles of Sn. This difference evidently depends on the method of preparation. To treat this absorption, we model the S particles by a Mattis-Bardeen form of the conductivity,[20]

$$\sigma_s(\omega)=\frac{iG_0}{\omega}+\sigma_s'(\omega)\ , \tag{30}$$

$$G_0=\sigma_n\frac{\pi\Delta}{\hbar}\ , \tag{31}$$

in which σ_n is the conductivity of the S particles in their normal state, Δ is the superconducting energy gap, and $\sigma_s'(\omega)$ is a complex function whose real part vanishes below $\hbar\omega=2\Delta$. (An analytic expression is quoted in Ref. 20.)

The ratio of the absorption of a cluster in the superconducting state to that in the normal state, α_s/α_n, is shown in Fig. 4. In all calculations, we use $\alpha_n=10^6\ \Omega^{-1}\,\mathrm{cm}^{-1}$.

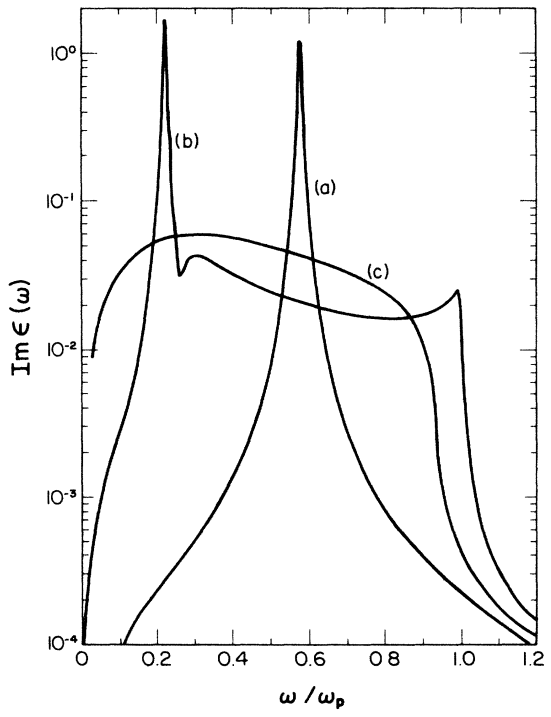

FIG. 3. Imaginary part of the composite dielectric constant: (a) Maxwell-Garnett approximation with no clustering; (b) fractal clustering with $R=10a$ and $d_f=2.5$ with differential EMA for the cluster dielectric constant; and (c) nonfractal clustering with EMA for the dielectric constant. In both (b) and (c) the concentration of metal in each cluster is $10^{-(3-d_f)}=0.316$; the difference between the curves arises from the different ways the metal is distributed within the clusters.

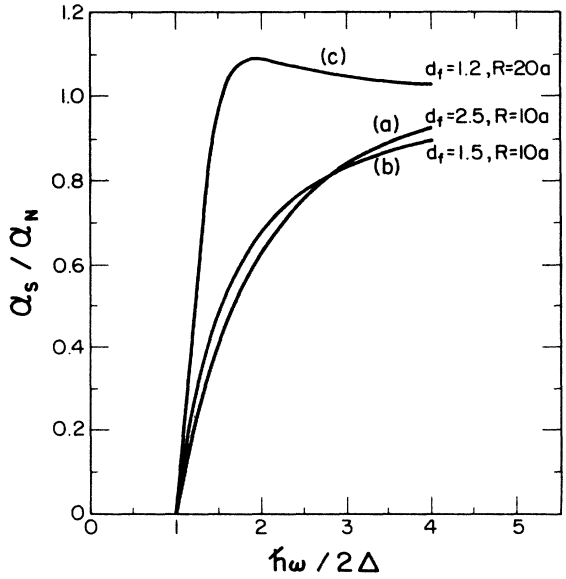

FIG. 4. The ratio of the absorption of a cluster in the superconducting state to that in the normal state, α_s/α_n, for different values of fractal dimensions and cluster sizes. Curves (a) and (b) are cases in which the magnetic dipole absorption is dominant. Curve (c) corresponds to the case in which the fractal dimension is so low that the electric dipole absorption predominates. In case (c), α_S becomes greater than α_n at frequencies slightly above $2\Delta/\hbar$.

Curves (a) and (b) correspond to the case in which the magnetic dipole term is dominant. In curve (c), the fractal dimension is so low that the electric dipole absorption predominates. The behavior in curve (c) is in qualitative agreement with experiment.[21] It is evident that the experimentally observed difference between S and N can be accounted for only if electric dipole absorption is dominant. If this is the case, then the fractal cluster mechanism can account for the result if the clusters have fractal dimension so low that the electric dipole mechanism is the dominant channel. However, since the required fractal dimension is very low, it seems unlikely that this mechanism is the explanation for the experimental results. The present work should be thought of in the spirit of a model calculation which describes the electromagnetic response of fractal clusters should they exist in any given composite.

Another intriguing signature of fractal clustering would be an enhancement of diamagnetic susceptibility χ_m in the superconducting state. For frequencies much lower than the superconducting gap 2Δ, the conductivity in the superconducting state can be written as[20]

$$\sigma=\frac{iA}{\omega}\sigma_n, \quad \hbar\omega \ll 2\Delta \ . \tag{32}$$

The coefficient b_1 can be calculated in this low-frequency limit as[7]

$$b_1 \sim -\frac{1}{45i}\left[\frac{\omega r}{c}\right]^5\left[\frac{4\pi\sigma_n A}{\omega^2}\right] \tag{33}$$

for a particle of radius r. Hence, the contribution per unit volume of the particle to the diamagnetic susceptibility χ_m is given by

$$\chi_m=-\frac{1}{10}\left[\frac{r^2}{c^2}\right]\sigma_n A \ . \tag{34}$$

The dependence on r^2 of χ_m is the same as that in the magnetic dipole absorption. Hence, in the case of fractal clustering, we expect an enhancement factor of $(R/a)^{(1/2)(1+d_f)}$ in the diamagnetic susceptibility. Such an enhancement would be another experimental signal that the superconducting particles are present in the form of clusters, rather than as isolated entities.

IV. DISCUSSION

We have presented in this paper an analysis of the far-infrared response of small metal particles on the assumption that these particles are grouped into clusters of fractal dimensionality. Application of a differential effective medium approximation leads to the conclusion that such clusters will have an effective conductivity which decreases as a power law in the radius, and which also decreases more rapidly for clusters of lower fractal dimension. Explicit calculations show in a simple way that such clusters will have an enormously enhanced electric dipole absorption coefficient per unit volume of metal, relative to that of isolated particles. The magnetic dipole absorption coefficient is also greatly enhanced by clustering. Observed far-infrared absorption by both normal and superconducting small particles can both be explained only if it is assumed that there exist a few clusters of low fractal dimensionality, such that their absorption coefficients are predominantly due to the electric dipole mechanism.

We have also shown that clustering will affect other dynamical properties of small metal particles. In small superconducting particles, for example, the diamagnetic susceptibility is considerably enhanced by the grouping of such particles into fractal clusters. If such an enhancement were observed, that would be a strong signal that clusters exist, and would tend to serve as corroboration of the clustering theory of far-infrared absorption.

Finally, we consider the question of the existence of clusters such as are assumed in this paper, and the possible application of our method to the properties of clusters in other contexts. Ample experimental evidence now exists that fractal clusters can form in colloidal suspensions of small metal particles. A large body of theoretical studies also shows that fractal clusters are the natural result of many irreversible growth processes which might easily influence the development of small metal particles. The fractal dimension of the resulting clusters depends on the growth mechanism. As for other potential applications, one can imagine various circumstances in atmospheric physics or astrophysics in which absorption by clusters of metal particles is of importance; the present work shows that it will be necessary to have a clear picture of the geometry of such clusters in order to calculate such absorption reliably.

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*Permanent address.

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