Far-infrared absorption by fine-metal-particle composites

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A number of mechanisms for absorption of far-infrared radiation by small metal particles are explored. These mechanisms include the magnetic and electric-dipole absorption from needle-shaped particles and the electric dipole absorption by coated particles. Although the absorption coefficient of small-particle composite is enhanced by these mechanisms, the calculations cannot explain the experimental absorption.

I. INTRODUCTION

In this paper we explore mechanisms which could influence the far-infrared absorption by small metal particles, an absorption which is anomalously large. We investigate several models that include the effects of coatings on particle surfaces or of clustering of individual grains into needle-shaped structures, but find that they cannot explain the data. The far-infrared absorption thus remains mysteriously large.

Several workers have measured the far-infrared absorption coefficient of small metal particles embedded at a low volume fraction in an insulating host[1-5] with a number of common features emerging from the experiments. First, the frequency dependence of the absorption is quadratic at low frequencies but tends to level off at higher frequencies. Second, except in the case of Al-KCl composite systems,[4] the absorption coefficient is approximately linear in metal volume fraction. Third, the magnitude of the absorption increases rather slowly with increasing particle size. Fourth, the magnitude of the absorption is quite similar for different types of metals but is substantially larger for metal particles than for insulating ones. Finally, the absorption is substantially larger than the dipolar absorption given by classical electromagnetic theory. These results may be summarized by writing the experimental absorption coefficient as

$$\alpha = K f \omega^2 ,\tag{1}$$

where f is the metal volume fraction, ω the frequency, and K is a "constant" which depends only weakly on frequency, particle size, and material. When α and ω are both expressed in cm^{-1}, the experimental values for K are typically in the range 0.1 to 1

cm. In contrast, theoretical values for K are in the range $10^{-7}-10^{-2}$ cm. The simplest classical theory of this absorption treats the particles as isolated metal spheres. The metal is characterized by a Drude complex dielectric function,

$$\epsilon_m = 1 - 4\pi\sigma_m\tau_m + 4\pi i\frac{\sigma_m}{\omega} ,\tag{2}$$

where σ_m is the dc conductivity of the metal and τ_m is the relaxation time of the conduction electrons. (In this equation and in the rest of this paper we assume that we are in the low-frequency limit, where $\omega\tau_m \ll 1$.) The metal particles are embedded in an insulating host, which has a real and constant dielectric function ϵ_i. In this theory, the low-frequency absorption coefficient is given by [see Eq. (17), Ref. 2]

$$\alpha = \sqrt{\epsilon_i}f\frac{\omega^2}{c^2}\left[\frac{9c\epsilon_i}{4\pi\sigma_m} + \frac{2\pi a^2\sigma_m}{5c}\right] ,\tag{3}$$

where a is the average particle radius. Equation (3) is of the same form as Eq. (1),

$$\alpha = f\omega^2(K_e + K_m) .\tag{4}$$

The first term is the electric dipolar absorption, which increases with decreasing particle conductivity. If the conductivity of the particle approaches that of a metal $\sim 10^6\ \Omega^{-1}cm^{-1}$, the constant K_e is much smaller than experiment: $K_e \sim 10^{-7}$ cm. The second term is the magnetic dipolar absorption (or eddy-current loss), which increases with increasing conductivity. For metal particles with radii bigger than 50 Å, the eddy-current loss dominates the electric dipole loss. In addition, this loss is quadratic at low frequencies but levels off at higher frequencies, as does the observed far-infrared absorption. How-

ever, the constant K_m for isolated spherical particles is smaller, by at least a factor of 10, than experiment. More importantly, the long-wavelength eddy-current loss grows as the square of the radius, a size dependence which is never experimentally observed.

To explain the anomalous far-infrared absorption, Simanek[6] has proposed a model in which the metal particles, each coated with an insulating oxide layer, are stuck together to form needlelike structures. (Electron micrographs of small particles often show such structures, which probably result from electrostatic attraction between particles.[7]) The absorption, according to this model, occurs in the oxide which coats the particles. There are a number of difficulties with this model: (a) Russell et al.[3] and Carr et al.[4] have observed far-infrared absorption in small particles which is larger than the predictions of this model. (b) All the absorption occurs in the oxide but oxides themselves do not show high far-infrared absorption.[4] (c) Unoxidized small particles have equally large absorption.[3] (d) Because the metal particles are assumed to be coated with an insulator, the composite could never become conducting, even if the filling fraction and geometry allowed percolation. In an elaboration of Simanek's model, which overcame objection (d) above, Ruppin[8] calculated the absorption from conducting clusters of particles and obtained a larger absorption than Simanek, but at the cost of a stronger frequency dependence to the absorption than experimentally observed.

Measurements of the far-infrared absorption in composites containing superconducting small particles[9,10] support a conclusion that the absorption occurs in the metallic part of the particles. When these samples are cooled below the transition temperature of the superconductor, the absorption increases (by about 50%) for frequencies near the energy gap 2Δ of the superconductor. This increase is opposite to expectation, because a bulk superconductor has $\sigma_s(\omega)=0$ for $\omega < 2\Delta$ and $\sigma_s(\omega) < \sigma_n(\omega)$ at higher frequencies. Only when $\omega \gg 2\Delta$ does $\sigma_s = \sigma_n$ (here σ_s is the conductivity of the material when superconducting, and σ_n is the same quantity in the normal state). According to Eq. (3), the eddy-current absorption should be smaller in the superconducting than in the normal state, because $K_m \sim \sigma$. In contrast, the electric dipole absorption should be larger in the superconducting state, as long as the limits which give Eq. (3) hold. However, if the electric dipole absorption is the dominant mechanism, the magnitude of the theoretical absorption is smaller by a factor of 10^7 than experi-

ment. (A simple calculation[10] based on quasiparticle tunneling between grains disagrees with experiment for $\omega > 2\Delta$; the predicted absorption of the superconductor is very close to but always smaller than that of the normal metal.)

In the remainder of this paper, we will explore a model for the far-infrared absorption of normal-metal—insulator composites that allows the particles to have a coating with a nonzero dc conductivity and that in addition considers the effects of having the individual grains organized into clusters. The coating gives rise to a large absorption; when the particles stick together the absorption is further increased. However, the calculations still fail to reproduce experiment.

II. EDDY-CURRENT ABSORPTION

Because the eddy-current absorption usually dominates the calculated absorption of isolated metal particles, we first investigated this absorption in needle-shaped clusters. Our calculation is relatively simple. The electric field $\vec{E}$ is obtained from $c\nabla \times \vec{E} = -\partial \vec{B}/\partial t$; the induced current is computed from the electric field. The current gives rise to a magnetic moment, $\vec{m}$, given by

$$\vec{m} = \frac{1}{2c} \int (\vec{J} \times \vec{r})d^3r \ . \tag{5}$$

The magnetic moment of both spheres and needles is given by Landau and Lifshitz.[11]

For a sphere, the long-wavelength result for the induced magnetic moment per unit volume is $m/V = B_{in}(k_m a)^2/40\pi$; for an infinite cylinder the same quantity is $m/V = B_{in}(k_m a)^2/16\pi$ when the ac magnetic field is perpendicular to the axis and $m/V = B_{in}(k_m a)^2/32\pi$ when the field is parallel to the axis. Here, a is the cylinder radius, B_{in} is the magnetic field strength, and $k_m = \sqrt{\epsilon_m}\omega/c$ is the wave vector of light inside a particle characterized by dielectric function ϵ_m. If the cylinder axis directions are distributed randomly, an angular average in the long-wavelength limit gives for the effective permeability of the composite

$$\mu_g = 1 - \frac{5\pi f \sigma_m \tau_m a^2 \omega^2}{6c^2} + i\frac{5\pi f \sigma_m a^2 \omega}{6c^2} \ , \tag{6}$$

where f is the volume fraction of metal. The eddy-current absorption is proportional to the imaginary part of this permeability,

$$\alpha_m = \sqrt{\epsilon_i} f \frac{\omega^2}{c^2} \frac{5\pi a^2 \sigma_m}{6c} \ . \tag{7}$$

Although cylinders have almost twice as much eddy-current loss as spheres [Eq. (3)], the magnitude of the loss remains smaller than experimental measurements. We conclude that eddy-current losses cannot account for the far-infrared absorption in small particles. This conclusion is reinforced by the weak size dependence of the observed absorption: Never is the a^2 behavior predicted by theory found.

When the particles string together to form a loop, the eddy-current absorption can be very large, because the current loops would have large areas and correspondingly large dipole moments. The absorption due to randomly oriented loops of radius R, when $|k|R \ll 1$, is

$$\alpha_l = f\omega^2 K_m \frac{5}{6} p_l \left[\frac{R}{a} \right]^2 . \tag{8}$$

Here, p_l is the probability that a metal grain is in a loop. If $R \sim 15a$ and $p_l = 10^{-2}$, then the absorption is enhanced by a factor of 2: $\alpha = 2f\omega^2 K_m$. If all the particles were tied up in loops, i.e., $p_l = 1$, then the absorption could easily be 2 orders of magnitude larger than the absorption from isolated spheres. However, such ringlike structures are extremely unlikely in low-volume-fraction composites. If the composite were near its percolation transition so that relatively large clusters of particles were occurring, and if the clusters formed a winding structure with a substantial self-inductance, then there could be large local magnetic fields and large absorption. This idea is discussed further in Sec. V, below.

III. ELECTRIC DIPOLE ABSORPTION IN COATED PARTICLES

In the preceding section, we showed that magnetic dipole absorption could not generally account for experimental measurements on small-particle systems. In this section we examine ways of enhancing the electric dipole absorption, the first term in Eq. (3). A substantial enhancement is required, because the electric dipole term for isolated particles is, in most cases, a factor of 10^7 smaller than experiment. The electric dipolar absorption increases with de-

creasing particle conductivity [as long as the limits which gave Eq. (1) continue to hold]. If the metal particle (characterized by complex dielectric function ϵ_m) were surrounded by a coating (with ϵ_c), then the particle dielectric constant is given by[12]

$$\epsilon_p = \epsilon_c \frac{3\epsilon_m - 2\zeta(\epsilon_m - \epsilon_c)}{3\epsilon_c + \zeta(\epsilon_m - \epsilon_c)} , \tag{9}$$

where ζ is the volume fraction of the coating, $\zeta = 1 - (a/a + t)^3$, a is the radius of the metal core and t is the thickness of the coating ($\zeta = 3t/a$ if $t \ll a$). If, as is likely, $|\epsilon_m| \gg |\epsilon_c|$ we find $\epsilon_p = (a/t)\epsilon_c$ and

$$\alpha = \sqrt{\epsilon_i} f \frac{\omega^2}{c^2} \frac{9c\epsilon_i t}{4\pi\sigma_c a} , \tag{10}$$

where σ_c is the dc conductivity of the coating. If $\sigma_c = 1 \ \Omega^{-1} \text{cm}^{-1} = 10^{-6}\sigma_m$, and $t \sim 0.1a$, the absorption is enhanced by 5 orders of magnitude.

IV. ELECTRIC DIPOLE ABSORPTION IN NEEDLELIKE CLUSTERS

When the individual metal grains stick together to form needlelike structures, the electric dipole absorption will increase. We assume for the model described here that the dielectric function of these structures will be dominated by the interfacial boundary between the individual grains forming a needle. The needle conductivity will be substantially smaller than the conductivity of the metal due to a high interparticle resistance. The composite in this case consists of a collection of randomly oriented spheroids with depolarization factor L along the symmetry axis, volume fraction f_L, and complex dielectric function ϵ_n distributed in an insulating matrix with dielectric function ϵ_i. Many approximations could be used to find the effective dielectric function of the material; for the low concentrations considered here, the Maxwell-Garnett theory, generalized for a collection of randomly oriented nonspherical inclusions is sufficient. This effective dielectric function is obtained from the expressions for local fields in Refs. 13 – 15:

$$\epsilon_g = \epsilon_i + \epsilon_i \frac{\left\langle f_L \left[\dfrac{1}{\epsilon_i + L(\epsilon_n - \epsilon_i)} + \dfrac{4}{2\epsilon_i + (1-L)(\epsilon_n - \epsilon_i)} \right] (\epsilon_n - \epsilon_i) \right\rangle}{\left\langle 3(1 - f_L) + f_L \epsilon_i \left[\dfrac{1}{\epsilon_i + L(\epsilon_n - \epsilon_i)} + \dfrac{4}{2\epsilon_i + (1-L)(\epsilon_n - \epsilon_i)} \right] \right\rangle} . \tag{11}$$

The angular brackets in Eq. (11) denote an average over the values of depolarization factor L, which occur with concentration f_L. The depolarization factor of the needle is small for an electric field parallel to its long axis; when the semimajor axis (b) substantially exceeds the semiminor axis (a), then[11]

$$L = \left[\frac{a}{b}\right]^2 [\ln(2b/a) - 1] . \tag{12}$$

As $a/b \rightarrow 0$, $L \rightarrow 0$. A small value for the depolarization factor means that surface charges will not screen out the applied field, leading to a large absorption.

The dielectric constant of a needle-shaped particle may be calculated in either of two ways; both give essentially the same results. First, if the needle is constructed of coated spheres, the effective dielectric function of the needle can be found from Eq. (9). Second, if we envision the particle to be a series connection of metal grains each surrounded by a poorly conducting coating, then an equivalent-circuit analysis shows that the needle dielectric function, ϵ_n, is obtained from

$$\frac{1}{\epsilon_n} = \frac{a}{(a+t)\epsilon_m} + \frac{t}{(a+t)\epsilon_c} , \tag{13}$$

where ϵ_m and ϵ_c are, respectively, the complex dielectric functions of the metal core and of the coating, a is the metal radius and $t \ll a$ is the coating thickness. If $|\epsilon_m| \gg |\epsilon_c|$, then $\epsilon_n = (a/t)\epsilon_c$.

Capacitive coupling between the metal grains dominates the resistive coupling when $\omega > 4\pi\sigma_c/\epsilon_c$. This coupling increases the conductivity of the needle-shaped cluster and (because of the absorption from induced electric dipoles) decreases the absorption.

To show explicitly that the needle-shaped particles have enhanced electric dipole absorption, we consider the case when the following limits hold: (a) $L |\epsilon_n| \gg \epsilon_i$, (b) $L \ll 1$, (c) $f \ll 1$, and (d) $\omega\tau_m \ll 1$. The quantity τ_m in limit (d) is the electronic scattering time in the metal grains; this limit ensures that the frequency-dependent conductivity is close to the dc value. When limits (a) and (b) do not contradict each other (the usual case), Eq. (11) reduces to

$$\epsilon_g = \epsilon_i \left[1 + \frac{f}{3L} + if\frac{\omega\epsilon_i}{12\pi\sigma_n L^2}\right] . \tag{14}$$

This expression should be compared with the effective dielectric function for a collection of coated spheres,

$$\epsilon_g = \epsilon_i \left[1 + 3f + 3if\frac{\omega\epsilon_i}{4\pi\sigma_p}\right] , \tag{15}$$

where σ_p is the effective conductivity of the coated sphere from Eq. (9). The small depolarization factor of the needle-shaped particle increases both the real and imaginary parts of the effective dielectric function. The absorption coefficient is proportional to the imaginary part of these dielectric functions, so that

$$\alpha_n = \alpha_{\text{sphere}}(\sigma_n/\sigma_p L^2) . \tag{16}$$

As frequency increases, $4\pi\sigma_n/\omega$ decreases until the limit $L |\epsilon_n| \gg \epsilon_i$ does not hold. The frequency dependence of the absorption then becomes complicated. Even in the case of spheres, if $|\epsilon_p| \ll \epsilon_i$, then $\epsilon_g = \epsilon_i + \frac{3}{2}f\epsilon_p$ giving $\alpha \sim \sqrt{\omega}$; similarly in the case of needle-shaped particles, when $L |\epsilon_n| \ll 1$, $\epsilon_g = \epsilon_i + \frac{5}{3}\epsilon_n$, giving a square-root behavior.

Figure 1 gives calculated values of the absorption coefficient for the various cases discussed above. Table I gives values for the coefficient K appearing in Eq. (4). We find a frequency-independent K for $\omega < 40$ cm^{-1} only for spherical particles and then only when $\sigma_c > 1$ Ω^{-1}cm^{-1}. In general, K is a constant (i.e., the absorption is quadratic in frequency) only for very low frequencies, in contrast to experiment. Table I gives ω_c, the frequency at which de-
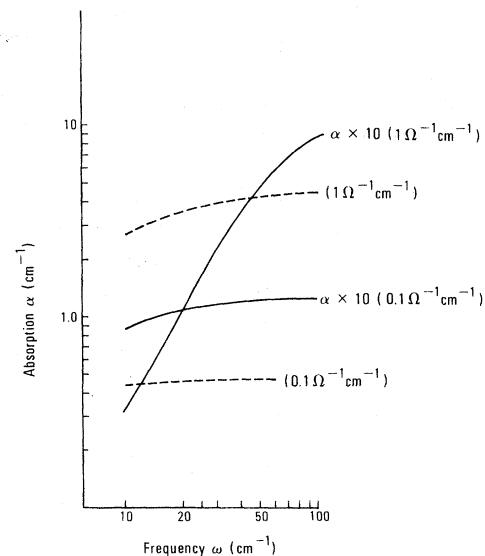

FIG. 1. Calculated far-infrared absorption coefficient of a small-particle composite containing 0.01-volume-fraction coated particles in an insulator. Curves are shown for spherical particles (solid) and for needle-shaped particles (dashed) that have depolarization factors of 0.01. Other parameters are shown in Table I.

TABLE I. Numerical results for electric dipole far-infrared absorption. Values of parameters used: $\epsilon_i = 4.8$, $\zeta = 0.3$, $\epsilon_n = \epsilon_p$ [Eq. (11)], $\sigma_m = 3.6 \times 10^5$ $\Omega^{-1} cm^{-1}$, $\tau_m^{-1} = 400$ cm^{-1}, $f_L = 0.01$, and $L = 0.01$.

σ_c ($\Omega^{-1} cm^{-1}$)	Sphere			Needles ($L = 0.01$)		
	K_e ($\omega \ll \omega_c$) (cm)	ω_c (cm^{-1})	α (30 cm^{-1}) $f = 0.01$ (cm^{-1})	K_e ($\omega \ll \omega_c$) (cm)	ω_c (cm^{-1})	α (30 cm^{-1}) $f = 0.01$ (cm^{-1})
10^{-2}	3.1	0.1	0.12	1015	0.01	0.044
10^{-1}	0.31	1	0.12	101.5	0.1	0.44
1	0.03	10	0.23	10	1	3.9
10	0.003	40	0.028	0.1	10	7.41

viations from the quadratic frequency dependence are noticeable in our calculations. The table also provides values for the absorption coefficient at 30 cm^{-1} for a volume fraction of 1% metal. For spheres, the absorption never equals the experimental value. For needles the 30-cm^{-1} absorption for $1 < \sigma_c < 10$ $\Omega^{-1} cm^{-1}$ is comparable to experiment; however, the shape of the curve shown in Fig. 1 differs radically from experiment.

V. THE PERCOLATION THRESHOLD

The real part of the dielectric function of a composite containing needle-shaped inclusions which have depolarization factor L is enhanced by a factor $(9L)^{-1}$ over a composite containing spherical grains. These inclusions may be viewed as modeling the clusters which form in a composite as a percolation threshold is approached. At the percolation point, the inclusions span the sample; $b \to \infty$ in Eq. (12); $L \to 0$; and the dielectric constant [Eq. (14)] becomes large. It is known that the real part of the dielectric constant of a composite diverges as the percolation threshold is approached.[16] (It has been shown elsewhere[13] that a conductivity threshold is a sufficient but not a necessary condition for a divergence in the dielectric constant. Furthermore, the conductivity threshold need not be a percolation threshold.) As $L \to 0$, the condition $L |\epsilon_m| \gg \epsilon_i$ does not hold, but when $L = 0$, Eq. (11) may be simplified directly to find

$$\epsilon_g = \epsilon_i \left[1 + f_n \frac{(\epsilon_n - \epsilon_i)(\epsilon_n + 5\epsilon_i)}{\epsilon_i [\epsilon_n (3 - 2f) + \epsilon_i (3 + 2f)]} \right] .$$

$$(17)$$

In the usual case, $|\epsilon_n| \gg \epsilon_i$, yielding

$$\epsilon_g = \epsilon_i + \frac{f \epsilon_n}{3 - 2f} .$$

$$(18)$$

The conductivity in this limit is metallic,

$\sigma_g = f \sigma_n / (3 - 2f)$. The absorption coefficient would no longer be quadratic; instead it would vary as $\sqrt{\omega}$, as is typical for substances with finite dc conductivity.[17]

We have used in this section a quasielectrostatic approach, which seems reasonable because $|k| a \ll 1$. However, if the clusters which occur near percolation form a winding structure with substantial self-inductance there could be large ac magnetic fields with a correspondingly large absorption, similar to that from the loops described in Sec. II. Although such a structure probably does occur, these effects are likely to be important only very near percolation. Continuous loops which form near percolation are also thought to affect the diamagnetic susceptibility χ of granular superconducting materials.[18,19] When the London penetration depth exceeds the grain size, supercurrents in the loops are the dominant factor in the diamagnetism. Scaling arguments imply that $\chi \sim (f - f_c)^u$, where the exponent u is found[19] from a calculation based on a two-dimensional fractal to be $u \approx 0.77$. With a positive exponent, the susceptibility does not diverge at the percolation point. A second way to infer the amount of "windingness" in a cluster is to compare two lengths[20,21]: The cluster coherence length ξ, which diverges as $\xi \sim (f - f_c)^{-\nu}$, and the path length along the conducting backbone L_B, which diverges as $L_B \sim (f - f_c)^{-\zeta}$. In three dimensions $\nu \sim 0.9$ while $\zeta \sim 1$. The similarity of these exponents implies that the effects of winding structures should occur only very near percolation. To have $L_B = 2\xi$, we must have $f - f_c \approx 10^{-3}$.

VI. CONCENTRATION DEPENDENCE

Most samples have an absorption which increases linearly with metal concentration, as would be expected for the dilute metal concentrations. However, the absorption in aluminum particles is nearly quadratic[4] in f. The self-similar model[14] provides a

quadratic term in the concentration, but one which is not larger than the linear term. For example, it is easy to show using this model that the effective dielectric constant ϵ_{ss} of a collection of spherical particles is given by the solution to

$$\left[\frac{\epsilon_m - \epsilon_{ss}}{\epsilon_m - \epsilon_i}\right]\frac{\epsilon_i}{\epsilon_{ss}} \simeq (1-f)^3 , \tag{19}$$

and the absorption coefficient α_{ss} is

$$\alpha_{ss} = \frac{K_e f \omega^2}{(1-f)^6} = K_e f \omega^2 (1+6f) . \tag{20}$$

Although the second term occurs, it is not large enough to explain experiment.

VII. CONCLUSIONS

Our model for the far-infrared properties of small metal particles has shown ways in which the strength of the electric dipole absorption could be increased above the value for isolated metal spheres. The most successful model viewed the metal grains as being clustered into needle-shaped particles with the magnitude and frequency dependence of the absorption being controlled by the quality of the contact between grains. Our model is consistent with the weak dependence on both particle size and conductivity observed experimentally but cannot predict the magnitude of the absorption while retaining the proper frequency dependence. Thus the far-infrared absorption in small metal particles still remains a mystery.

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