FAR INFRARED ABSORPTION IN GRANULAR SUPERCONDUCTORS*

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ABSTRACT

Measurements of far infrared transmission and reflection have been done on composites consisting of small tin particles (approximately 250Å radius) embedded randomly in an insulating matrix of KCl, with volume fraction of tin ranging from 0.001 to 0.03. These experiments measure the optical properties from 3 cm^{-1} to 50 cm^{-1} and at temperatures from 1.7K to 27K, thus covering the energy gap frequency and the transition temperature of the tin. The results of these experiments show an enhanced absorption around the gap frequency at superconducting temperatures.

INTRODUCTION

In this paper we present the results of far infrared absorption measurements on superconducting-insulator composites. Previous work by Tanner et al[1] indicated no change in the absorption spectrum, $\alpha(\omega)$, for very small (radius 70Å) Sn particles imbedded in KCl at temperatures below T_c(bulk). Patton[2] has considered the effect of particle size on superconductivity and finds that for small particles of diameter less than the coherence length (ξ), fluctuations exist which smooth out the energy gap structure in a fairly wide temperature region about T_c. This smoothing would account for the lack of structure in $\alpha(\omega)$ for such small particles. However, for particles whose diameter approaches the coherence length (ξ~1000Å) we would expect a reasonably well defined gap to emerge with corresponding structure in $\alpha(\omega)$. We have therefore chosen to study larger particles with mean radii on the the order of 250Å.

EXPERIMENT

Because we wish to observe the composite at temperatures well below T_c, we have chosen Sn with T_c(bulk)=3.7K since temperatures of ~1.7K can easily be reached with pumped helium. Also, the gap frequency (2Δ) is 9.7 cm^{-1}, which falls within the range of our instrumentation.

The small Sn particles were made by smoke evaporation, as described by Granqvist and Buhrman[3]. Sn was evaporated from an alumina coated molybdenum boat in an atmosphere of 75% argon and 25% oxygen. The gas pressure ranged from 0.1 to 1 Torr. The oxygen formed an insulating layer of tin oxide on the outer surface to help avoid cold welding between particles. The particles collected on a glass surface surrounding the boat. By varying the gas pressure, mean particle radii ranging from 250Å to 400Å were

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produced. The particle size distribution was measured with a transmission electron microscope (see Fig. 1).

The Sn particles were thoroughly mixed with finely ground ($1\mu m < d < 10\mu m$) KCl powder and then pressed under vacuum (10 kbar) into a solid disk. The sample was reground and repressed 3 to 5 times to help achieve uniformity. X-ray mappings (using a scanning electron microscope) of the surface of a typical metal-insulator composite showed a reasonably uniform distribution of metal with little evidence of clustering.

Transmission and reflection measurements of these samples were made on a lamellar grating interferometer[4] with a mercury arc lamp as a source and a germanium bolometer operating at 1.2K as a detector. The composite under study was anchored to a copper block which housed a heater and several carbon resistors for thermometry. Measurements were made at 1.7K, 3.3K, 4.2K and 27K.

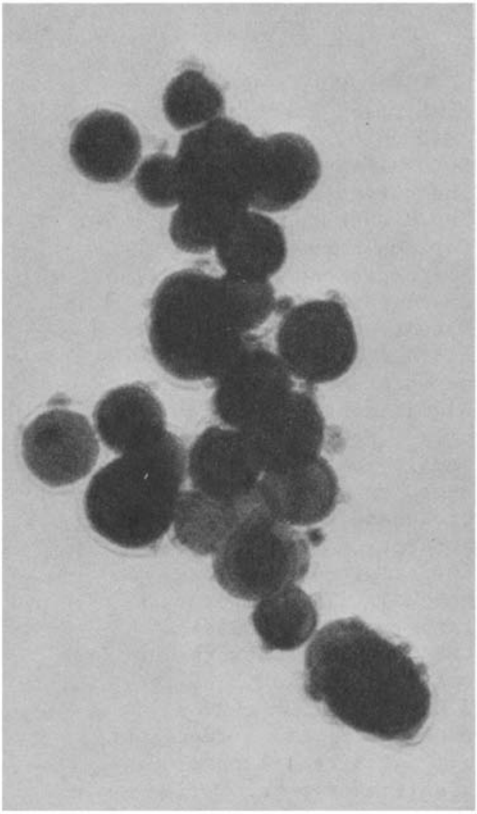

Fig. 1. Transmission electron micrograph of Sn particles. The mean particle radius is 250Å.

An expression for the absorption coefficient $\alpha(\omega)$, of a material can be easily derived in terms of the transmittance of the disc-shaped sample, $T(\omega)$. The expression, which allows for multiple internal reflections of the light, is:

$$\alpha(\omega) = -\frac{\ln T(\omega)}{x} - \frac{2\ln(1-R)}{x} \qquad (1)$$

where x is the sample thickness and R is the reflectance. We have estimated the reflectance from $R = (n-1)^2/(n+1)^2$ with $n = 2.2$ being the refractive index of KCl. Note that in our low resolution measurements the interference pattern produced by the multiple internal reflections is averaged out and that the internally reflected power is small enough to allow discarding all but its lowest order contributions to $\alpha(\omega)$.

RESULTS

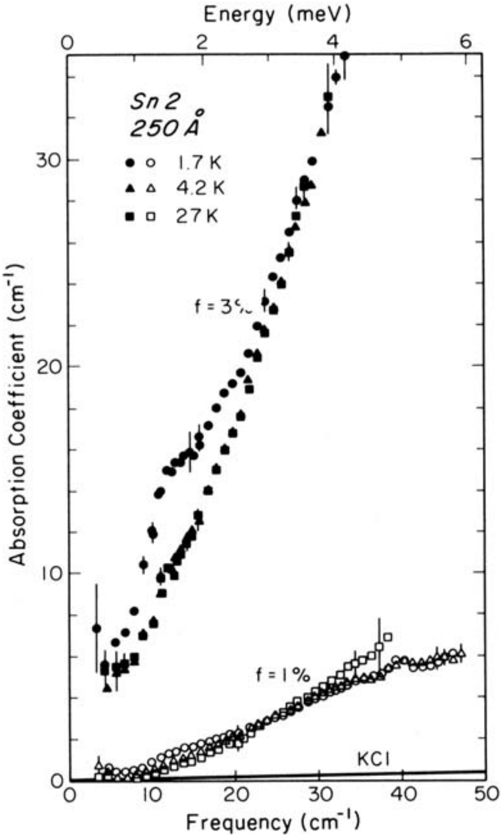

Figure 2. Far infrared absorption in Sn-KCl composite systems. Data are shown for two concentrations and three temperatures.

Figure 2 shows the absorption coefficient over 3-40 cm^{-1} for 250Å radius Sn small particles in KCl. The curves for T=4.2K and T=27K each represent $\alpha(\omega)$ for the normal state. The $\sim\omega^2$ dependence is typical of small-particle composites.[5] Despite the wide temperature difference between the two measurements, there is no significant difference in $\alpha(\omega)$. However, when the sample temperature was reduced below 4.2K, the broadband far infrared absorption increased (i.e. the sample became more opaque). The temperature range where the onset of this increased absorption was observed was 3.5K to 3.0K. The 1.7K curve shown in Figure 2 represents $\alpha(\omega)$ at a temperature significantly below T_c. These three curves show clearly that $\alpha(\omega)$ is unaffected by the superconducting transition for $\hbar\omega >> 2\Delta$. However, for $\hbar\omega \sim 2\Delta$, there is a significant absorption associated with the transition to the superconducting state. This point is illustrated more clearly in Figure 3, where we have plotted the difference between the superconducting and normal state absorption coefficients.

DISCUSSION

The effect described in the preceeding section - an enhanced absorption in the superconducting state - is not predicted by the commonly used theories for inhomogeneous media. The two models[6-9] commonly applied to the problem of infrared absorption in composites are the Maxwell-Garnett theory (MGT) and the effective medium (or self-consistent) theory (EMT). For metal volume fraction, f, on the order of 1% and for particle radii small compared to the skin depth, the two theories do not yield significantly different results. The far infrared abosrption in these limits is[1]

$$\alpha(\omega) = \frac{f\omega^2}{c}\left(\frac{9}{4\pi\sigma_1} + \frac{2\pi a^2\sigma_1}{5c^2}\right), \quad (2)$$

where $\sigma_1(\omega)$ is the real part of the complex conductivity of the metal and a is the particle radius. The first term in brackets arises from electric dipole absorption while the second arises from eddy currents induced in the particle. For metallic values of the conductivity (i.e. $\sigma_1 \sim 10^5 \Omega^{-1}\mathrm{cm}^{-1}$) the second term dominates the far infrared absorption for particle radii greater than 25Å. This theory gives the correct shape of $\alpha(\omega)$ for the normal state absorption, even though it falls short in magnitude by more than a factor of ten.[1,5,10]

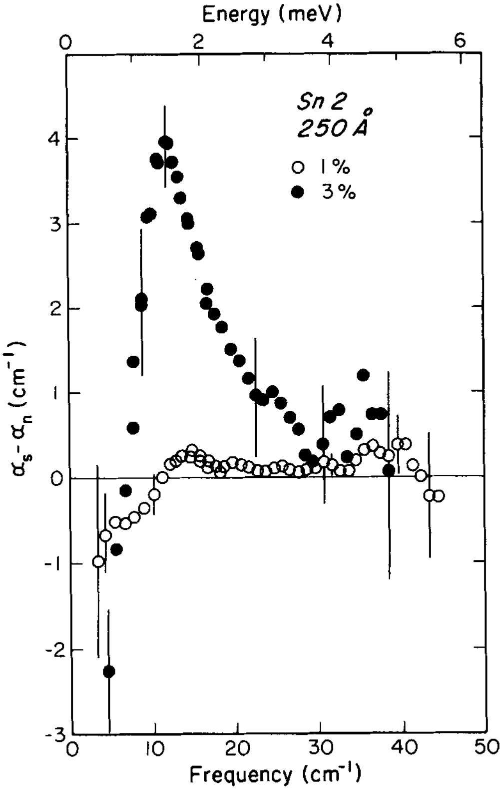

Figure 3. Difference between the absorption in the superconducting state and in the normal state in Sn-KCl composites. Data are shown for two concentrations.

We have calculated the far infared absorption in superconducting composites using the BCS expressions for $\sigma_1(\omega)$ in the superconducting state[11] within both the MGT and EMT. The results of these simple theories give an absorption coefficient which is zero for $\hbar\omega<2\Delta$, where 2Δ, is the full energy gap of the superconductor, and then rises, with an initially non-zero slope, approaching the normal state result for $h\omega>>2\Delta$. This result is the expected one, since the spheres, behaving much as in a bulk specimen, are not absorbing below 2Δ because no electronic excitations are possible. Such a simple theory is not capable of explaining the enhanced absorption which is experimentally observed.

CONCLUSIONS

These experiments have found an enhanced far infared absorption in composite superconductors at frequencies near the superconducting energy gap. A simple model for composites using BCS values for the frequency dependent conductivity does not explain the observed infrared absorption spectrum. Possible explanations may be based on size effects, electron hopping mechanisms from particle to particle (tunneling) or clustering of particles into a (non-spherical) macroparticle.

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