LETTER TO THE EDITOR

Effective-medium theory for granular high- $T_{\rm c}$ superconductors—electromagnetic properties in the microwave region

T C Choy† and A M Stoneham

Theoretical Physics Division and Material Physics and Metallurgy Division, Harwell Laboratory, AEA Technology, Oxfordshire OX11 0RA, UK

Received 14 December 1989

Abstract. We extend to finite frequencies our recently developed effective-medium theory (EMT) for granular high- $T_{\rm c}$ superconductors (HTS). Here we present results for the electromagnetic properties in the microwave region, which is of potential significance for early commercial applications of HTS materials. Most of our discussions centre on generalisations of classical Mie scattering theory to *spherical* superconductor grains. However, there are important corrections to previous works on EMT at finite frequencies by Stroud and Pan and also Mahan, which we discuss. Some illustrative numerical results are presented, deferring lengthy numerical computations and comparisons with experiments, to be published elsewhere.

In a recent paper [1], we have developed an effective-medium theory (EMT) which shows considerable promise as a model for the description of the electromagnetic (EM) properties of granular oxide superconductors. In that paper we discussed the essential modifications of conventional EMT by the explicit inclusion of the magnetic polarisability calculated from London's [2] theory for a spherical grain. Our results in that paper pertain to the DC region, and we illustrated the model with numerical results, using the further hypothesis of weak-link Josephson junctions for the grains, which show good agreement with observed experimental behaviour [3] for the temperature profiles of the resistivity and magnetic susceptibility near $T_{\rm c}$.

In the present note we report on the extension of our work to finite frequencies, briefly alluded to in [1]. The development of this model to microwave frequencies is the key subject here. This is strongly motivated by the current research and market indications of the likelihood for the application of oxide superconductors in passive microwave engineering, where the widely publicised critical current problem is of no significance.

Let us first specify three regimes for the EM properties of oxide granular superconductors, estimated from measurements of the superconducting and EM paramaters. These regimes are characterised by the skin depth δ and the London penetration λ_L with respect to the wavelength λ of the radiation. For operation at 85 K these are approximately:

[†] Now at: Department of Physics, Monash University, Clayton, Victoria 3168, Australia.

- (I) <3.5 GHz, Meissner current regime;
- (II) 3.5 GHz to 10⁵ GHz, eddy current regime;
- (III) $> 10^5$ GHz, displacement current regime;

where we have classified these regimes by the dominant currents at that frequency range. The estimates are for grains with a bulk T_c of 91.6 K, grain size of $\sim 1 \, \mu m$, normal resistivity of 200 $\mu\Omega$ cm and London penetration $\lambda_L(0)$ of 1400 Å as in [1]. As indicated by London [2], the ratios of these currents are given by

$$1: \sqrt{2}\lambda_{L}/\delta: \lambda_{L}/\lambda \tag{1}$$

although a further limitation comes from our considerations of the static zone $a \ll r \ll \lambda$ situation [4]. For situations in which the radiation zone $a \ll \lambda \ll r$ is appropriate [4], the theoretical framework is more complicated. As we shall see, it involves the full apparatus of Mie scattering theory [5] with suitable modifications for London electrodynamics [1], as well as special corrections to the magnetic and electric integrals for the polarisabilities. At the higher infra-red frequencies, the normal dielectric properties take over and superconductivity loses its significance, apart from the characteristic frequencies associated with the intergrain Josephson junctions and superconducting gap, which are not the concern of our discussions here.

We begin our discussions by noting that the London equations at finite frequencies ω [2] generalise as a vector Helmholz equation for W = (E, B, or J):

$$\nabla^2 \mathbf{W} + \mathbf{k}^2 \mathbf{W} = 0 \tag{2}$$

where the generalised wave vector k defined by

$$\mathbf{k}^2 = \mathbf{k}_0^2 - i\sigma\mu\omega - 1/\lambda_L^2 \qquad \mathbf{k}_0^2 = \omega^2\varepsilon\mu$$
 (3)

includes the electromagnetic skin depth $\delta = \sqrt{2}(\sigma\mu\omega)^{-1/2}$ and the London penetration length λ_L . One can immediately obtain the necessary modifications to classical Mie theory [5], by analytic continuation of the formulas of Mie [5] and Debye [6]. Initially, however, we shall avoid these complications by considering a quasi-static approximation for regime I. In the near-field and DC limit, this naturally recovers the previous results [1], e.g. in calculating the magnetic polarisability. Therefore, in regime I, we can obtain the polarisabilities at finite frequencies, for spherical grains, using a quasi-static approximation; see, e.g., Landau and Lifshitz [7]. Following similar arguments to those used in [1], we extend our EMT for the electric permittivity ε and the magnetic permeability μ to finite frequencies:

$$c_{g}\left(\frac{\varepsilon_{g}-\varepsilon}{\varepsilon_{g}+2\varepsilon}\right)+c_{v}\left(\frac{\varepsilon_{0}-\varepsilon}{\varepsilon_{0}+2\varepsilon}\right)+c_{s}=0$$
(4)

and

$$c_{\rm g}\left(\frac{\mu-\mu_{\rm g}}{\mu_{\rm g}+2\mu}\right)+c_{\rm v}\left(\frac{\mu-\mu_{\rm 0}}{\mu_{\rm 0}+2\mu}\right)+\frac{c_{\rm s}}{2}Z_{\rm L}\left(\frac{a}{\lambda_{\rm L}}\right)=0\tag{5}$$

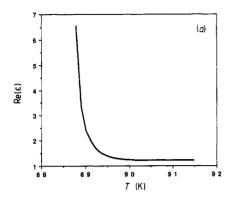
using similar notation to [1], where

$$Z_1(x) = 1 - (3/x)(\coth(x) - 1/x).$$
 (6)

Here we shall assume for simplicity (which can later be improved) that the complex (normal) grain permittivity and permeability are

$$\varepsilon_{g} = \varepsilon_{0} - i\varepsilon'_{g} \qquad \mu_{g} = \mu_{0} - i\mu'_{g}. \tag{7}$$

The imaginary parts, as expected, arise from the skin depth δ of the normal grains, and are given by



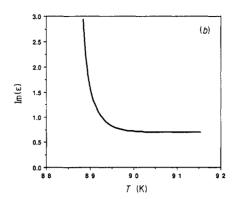
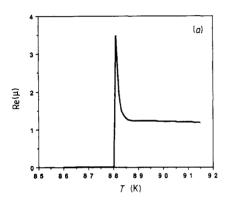


Figure 1. (a) Real part of permittivity ε in the quasi-static limit (regime I), in units of free space ε_0 . The parameters for the calculations of this letter are for the model granular superconductor described in figure 2 of [1]. (b) Imaginary part of permittivity ε in the quasi-static limit (regime I), in units of free space ε_0 times $3 \times 10^7 \lambda$ (in metres).



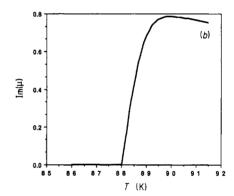


Figure 2. (a) Real part of permeability μ in the quasi-static limit (regime I), in units of free space μ_0 . The behaviour contrasts with that of the DC permeability [1] which shows no divergence. (b) Imaginary part of permeability μ in units of free space μ_0 times $3 \times 10^7 \lambda$ (in metres). The bulk T_c is 91.6 K.

$$\varepsilon_{\rm g}'/\varepsilon_0 = \mu_{\rm g}'/\mu_0 = \sigma/\omega\varepsilon_0. \tag{8}$$

The quantity c_s is the superconducting fraction, which we have obtained from a weak-link Josephson junction model (see [1] for details) which will not be discussed any further in this letter. Equations (4) and (5) are *uncoupled* quartic equations, which can easily be solved for negative imaginary parts of ε and μ . In figures 1 and 2 we show the results for the complex permittivity ε and permeability μ respectively. We also show in figure 3 the complex impedance $Z = \sqrt{(\mu/\varepsilon)}$, which is an important quantity for transmission line and microwave cavity applications.

In the remainder of this letter, we shall discuss the changes necessary in regime II. First, as pointed out earlier, the quasi-static or Rayleigh scattering approximation, used in deriving equations (4) and (5), is no longer valid. The electromagnetic scattering coefficients have to be predicted by Mie theory [5], suitably modified for London

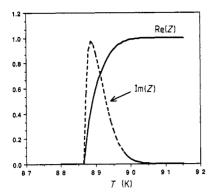


Figure 3. Complex impedance Z, in the quasi-static limit (regime I), e.g. for an ideal HTs transmission line. Real Z is in units of the free space impedance $Z_0 = 376.6 \,\Omega$. Imaginary Z is in the units Z_0 times $1.6 \times 10^{-7}/\lambda$ (in metres). Note that in this regime our calculations show that the impedance is zero (i.e. lossless transmission) below about 88.5 K.

electrodynamics as stated in equations (2) and (3). These can be obtained directly from the Mie formulas:

$$a_{l} = \frac{\eta_{s} \psi_{l}'(x) \psi_{l}(y) - \eta \psi_{l}(x) \psi_{l}'(y)}{\eta_{s} \psi_{l}(y) \xi_{l}'(x) - \eta \xi_{l}(x) \psi_{l}'(y)}$$
(9)

and

$$b_{l} = \frac{\eta \psi_{l}'(x)\psi_{l}(y) - \eta_{s}\psi_{l}(x)\psi_{l}'(y)}{\eta \psi_{l}(y)\xi_{l}'(x) - \eta_{s}\xi_{l}(x)\psi_{l}'(y)}$$
(10)

where $x = \eta k_0 a$, $y = \eta_s k_0 a$, using the standard notation [8]. Here we have expressed the wavevector k in terms of the complex refractive index η of the medium that we seek to calculate. For a superconducting grain, η_s follows from equation (3):

$$\eta_s^2 = 1 - 2i/(k_0 \delta)^2 - 1/(k_0 \lambda_L)^2. \tag{11}$$

These modifications to Mie theory are straightforward, although various asymptotic expansions, as given for example by Van der Hulst [8], are no longer valid. They must be rederived with care. In most cases, we have to resort to numerical calculations. In particular, we can see that in regime II, the consideration of the first two dipole terms a_1 and b_1 alone is inadequate. However, we shall not discuss these technical details here any further.

Of greater importance, as a matter of principle, are the modifications to EMT associated with the polarisability calculations [1]. These are formulated in terms of the integral

$$I_i^{\mathsf{E}} = \int_{v_i} \mathrm{d}\mathbf{r} \, \mathbf{E}_i \, \mathrm{e}^{-\mathrm{i}\mathbf{k} \cdot \mathbf{z}} \tag{12}$$

for the *i*th grain of volume v_i , using the same notation as [9]. Stroud and Pan [9] evaluated this integral in the far field (radiation zone):

$$I_i^{\rm E} = (4\pi/k^3)(\varepsilon/\delta\varepsilon_i)S_i(0) \tag{13}$$

where $\delta \varepsilon_i = \varepsilon - \varepsilon_i$ and where $S_i(0)$ is the forward scattering amplitude of the scattered wave due to the *i*th grain:

$$S_i(0) = \sum_{l} (a_l + b_l). \tag{14}$$

Note that we have reverted to Gaussian units here to conform with the results of [9]

and [10]. We found that a fundamental correction to this integral is necessary for superconducting grains. Instead of (13), our results are given by

$$I_i^{\rm E} = \left(\frac{1+\mu}{1-\varepsilon\mu}\right) \frac{4\pi}{k^3} \frac{\varepsilon}{\delta\varepsilon_i} S_i(0) \tag{15}$$

for the electric integral and

$$I_i^{\rm B} = \left(\frac{1+\varepsilon}{1-\varepsilon\mu}\right) \frac{4\pi}{k^3} \frac{\mu}{\delta\mu_i} S_i(0) \tag{16}$$

for the magnetic integral corresponding to (12). Using these expressions for the polarisabilities in the effective-medium theory [1], we immediately see that there is a coupling between the electric permittivity ε and magnetic permeability μ , a situation like that for ferrite composites as studied by Mahan [10]. This is probably unique to superconductivity due to London electrodynamics, but it may also be important for magnetoelectric composites [7]. As it happens, these corrections are significant in the microwave regime II and they seriously complicate our numerical calculations. We note finally that, in regime III, the dielectric properties dominate and we can use Stroud and Pan's result (13), which is valid there.

In conclusion we have presented an extension of our EMT to finite frequencies. Illustrative numerical results in regime I are shown for the complex permittivity ε and magnetic permeability μ respectively. In the microwave regime II, we argued that fundamental corrections to the EMT discussed by Stroud and Pan [9] and also Mahan [10], with a coupling between ε and μ , are important. These follow from equations (15) and (16) which are our corrections to their polarisabilities for *spherical* superconducting grains. Detailed numerical calculations and comparison with experiments will be presented elsewhere.

This work was supported by the Underlying Research Programme of the UKAEA.

References

- [1] Choy T C and Stoneham A M 1990 J. Phys.: Condens. Matter 2 939-51
- [2] London F 1950 Superfluids vol 1 (New York: Wiley)
- [3] Härkönen K, Tittonen I, Westerholm J and Ullakko K 1987 Phys. Rev. B 39 7251-4
- [4] Jackson J D 1975 Classical Electrodynamics 2nd edn (New York: Wiley)
- [5] Mie G 1908 Ann. Phys., Lpz. 25 377
- [6] Debye P 1909 Ann. Phys., Lpz. 30 59
- [7] Landau L D, Lifshitz E M and Pitaevskii L P 1984 Electrodynamics of Continuous Media (London: Pergamon)
- [8] Van der Hulst H C 1981 Light Scattering by Small Particles (New York: Dover)
- [9] Stroud D and Pan F P 1978 Phys. Rev. B 17 1602-20
- [10] Mahan G D 1988 Phys. Rev. B 38 9500-2