

Random mixtures with orientational order, and the anisotropic resistivity tensor of high- T_c superconductors

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(Received 23 August 1990; accepted for publication 4 September 1990)

By generalizing effective-medium theory to the case of orientationally ordered but positionally disordered two-component mixtures, it is shown that the anisotropic dielectric tensor of oxide superconductors can be extracted from microwave measurements on oriented crystallites of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ embedded in epoxy. Surprisingly, this technique appears to be the only one which can access the resistivity perpendicular to the copper-oxide planes in crystallites that are too small for depositing electrodes. This possibility arises in part because the real part of the dielectric constant of oxide superconductors has a large magnitude. The validity of the effective-medium approach for orientationally ordered mixtures is corroborated by simulations on two-dimensional anisotropic random resistor networks. Analysis of the experimental data suggests that the zero-temperature limit of the finite-frequency resistivity does not vanish along the c axis, a result which would imply the existence of states at the Fermi surface, even in the superconducting state.

I. MOTIVATION

When new oxide superconductors are synthesized, the first crystals which can be produced are usually microcrystals which are too small to allow measurement of the anisotropic resistivity tensor by techniques which require contacts. Microwave techniques do not require external contacts but, on the other hand, large volumes and aligned microcrystals are necessary to get enough absorption sensitivity. In the measurements of Ref. 1 a permanently aligned sample was produced by mixing the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ powder with epoxy resin, which is an insulator, and using a magnetic field to orient the large-susceptibility c -axis direction along the magnetic field. Measurements of the ab plane dielectric constant is straightforward to interpret from measurements which use the alternating microwave magnetic field to drive eddy currents in the ab plane. To obtain c -axis results, on the other hand, it is preferable to place the sample directly in the alternating microwave electric field. In this case, however, the measurements are strongly influenced by the epoxy and it becomes necessary to analyze the data through the effective-medium approach presented below.

Our aim then in this paper is to obtain the dielectric tensor of the superconducting grains from the experimentally measured¹ dielectric constants of the epoxy and of the epoxy-oxide superconductor mixture. Since the experiments are done for dilute mixtures, far from a percolation threshold, the effective-medium approximation (EMA) should be adequate. This approach has been successfully applied to a great variety of problems.² In the case of superconducting ceramic samples, the anisotropy of the superconducting grains has been considered, but always in the context of randomly aligned crystallites.³ In the above-mentioned experiments the alignment of the grains is *not* random and the anisotropy occurs in two ways: the grains are prolate spher-

oids and, furthermore, they have different dielectric constants along the axes. The geometry and the dielectric anisotropy have, however, the same principal axes. Since there is negligible orientational disorder, the anisotropy of the superconducting grains reflects itself in the properties of the mixture.

In Sec. II, analytical results are derived and then verified in the special case of two dimensions by comparisons with numerical simulations. Section III interprets the experimental results of Ref. 1 and comments on the microwave resistivity along the c axis which seems particularly interesting for the physics of the superconducting state of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

II. ANISOTROPIC EFFECTIVE-MEDIUM APPROXIMATION

The mixtures of interest here are dilute, and hence we are in a situation of weak disorder where effective-medium theory should be a reliable approach. To apply this method to the present case, we furthermore need the wavelength and penetration depth of the applied external field to be larger than the size of the inhomogeneities, a condition which is further discussed in Sec. III. In the present section, we generalize the usual approach to the case of oriented mixtures. In Sec. II A, we consider a discrete model, in Sec. II B a continuous one, and in Sec. II C, the results are specialized to two dimensions and compared with computer simulations. While the results are written down for the conductivity, the ac case of interest in Sec. III is easily obtained by replacing conductivity by complex dielectric constant.^{4,5}

A. Discrete model

While our goal is to obtain results for continuous models, in this section we derive the results for random resistor networks because numerical simulations are easy to perform for this problem,⁶ hence allowing a useful check of the anisotropic generalization of EMA proposed here.

Consider a very simple model of anisotropic random

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resistor network. This network consists of bonds of type 1 and 2 occurring with probabilities p and $(1 - p)$, respectively. The conductivities of those bonds are σ_i^{\parallel} and σ_i^{\perp} ($i = 1, 2$), depending on the direction of the bond with respect to the externally applied electric field. From this statement it is obvious that the macroscopic conductivity will be direction dependent as well. By a straightforward generalization of the effective-medium approach to this kind of networks,⁷ one obtains the following relation between the conductivities of a two-dimensional (2D) square network:

$$p \frac{\sigma_1^{\parallel} - \sigma^{\parallel}}{\sigma^{\parallel} + (\sigma_1^{\parallel} - \sigma^{\parallel})f(\alpha)} + (1 - p) \frac{\sigma_2^{\parallel} - \sigma^{\parallel}}{\sigma^{\parallel} + (\sigma_2^{\parallel} - \sigma^{\parallel})f(\alpha)} = 0,$$

$$p \frac{\sigma_1^{\perp} - \sigma^{\perp}}{\sigma^{\perp} + (\sigma_1^{\perp} - \sigma^{\perp})f(1/\alpha)} + (1 - p) \frac{\sigma_2^{\perp} - \sigma^{\perp}}{\sigma^{\perp} + (\sigma_2^{\perp} - \sigma^{\perp})f(1/\alpha)} = 0, \quad (1)$$

where $\alpha = \sigma^{\perp}/\sigma^{\parallel}$ is the anisotropy factor of the effective medium and

$$f(\alpha) = \frac{1}{\pi} \arccos \frac{\alpha - 1}{\alpha + 1} \quad (2)$$

is obtained from the lattice Green's function. If the bonds corresponding to each compound were randomly oriented we would have only one equation for the effective-medium conductivity which would obviously be isotropic.

B. Continuous model

The EMA approach can also be applied to more realistic systems than random resistor networks. In particular, it has been extensively used for isotropic continuous systems, consisting in a collection of spheres of two different compounds, both of them characterized by isotropic properties (conductivity, resistivity, or dielectric constant), and which fill completely the volume under consideration. In Ref. 6 the conductivity of continuum percolation models has been obtained by means of random walks in two and three dimensions and for conducting-insulating and normal-superconducting composites. Their results are compared with EMA theories and show a reasonable agreement. As explained in Sec. I we are interested in systems where anisotropy arises from both geometry and intrinsic electrical properties, corresponding to the experimental conditions of Ref. 1.

We follow the generalized effective-medium approach of Ref. 8, which applies as long as inclusions have an ellipsoidal shape so that the field inside the grains is along the applied one. The relation between the conductivity tensors, which is analogous to Eq. (1), reads

$$\sum_i v_i (1 - \delta \sigma_i \cdot \Gamma_i)^{-1} \delta \sigma_i = 0, \quad (3)$$

where the sum runs over the different grains forming the macroscopic system, v_i is the volume of the i th grain, $\delta \sigma_i = \sigma_i - \sigma$ the difference between its conductivity tensor

and that of the effective medium, and Γ_i is defined by⁸

$$\Gamma_i^{\alpha\beta} = - \oint_s \frac{\partial}{\partial r_\alpha} G(\mathbf{r} - \mathbf{r}') n'_\beta d^2 r', \quad (4)$$

$G(\mathbf{r} - \mathbf{r}')$ being the Green's function, solution of the equation

$$\nabla \cdot \sigma \cdot \nabla G(\mathbf{r} - \mathbf{r}') = - \delta(\mathbf{r} - \mathbf{r}'). \quad (5)$$

The integral in (4) extends over the surface of the i th grain, and n'_β is the β th component of a unit vector perpendicular to the surface.

Keeping in mind the measurements of Ref. 1, we consider material 1, which consists of ellipsoids which are symmetric under rotation along the c (parallel) direction, and material 2, which consists of spherical grains. The conductivity tensors have the same symmetries as the geometry of the corresponding material. The spheroids are assumed to be aligned along their symmetry axis so that the macroscopic effective medium conductivity is anisotropic as well. Defining p as the volume fraction of material 1, Eq. (3) can be rewritten as

$$p \frac{\sigma_1^{\parallel} - \sigma^{\parallel}}{1 - (\sigma_1^{\parallel} - \sigma^{\parallel})\Gamma_1^{\parallel}} + (1 - p) \frac{\sigma_2 - \sigma^{\parallel}}{1 - (\sigma_2 - \sigma^{\parallel})\Gamma_2^{\parallel}} = 0,$$

$$p \frac{\sigma_1^{\perp} - \sigma^{\perp}}{1 - (\sigma_1^{\perp} - \sigma^{\perp})\Gamma_1^{\perp}} + (1 - p) \frac{\sigma_2 - \sigma^{\perp}}{1 - (\sigma_2 - \sigma^{\perp})\Gamma_2^{\perp}} = 0, \quad (6)$$

depending, as in Eq. (1), on the direction of the applied electric field. The different Γ 's appearing in (6) are obtained from their definition (4) and from the Green's function (5). Introducing $m = a^{\perp}/a^{\parallel}$ as the aspect ratio of the spheroids in three dimensions (3D), one obtains, following Ref. 8,

$$\Gamma_1^{\parallel} = - \frac{1}{\sigma^{\parallel}} \alpha^{1/2} \frac{m^2}{(m^2 - \alpha)^{3/2}} \left(\frac{(m^2 - \alpha)^{1/2}}{\alpha^{1/2}} - \arcsin \sqrt{\frac{m^2 - \alpha}{m^2}} \right), \quad (7)$$

$$\Gamma_1^{\perp} = - \frac{1}{2\sigma^{\parallel}} \alpha^{-1/2} \frac{m^2}{(m^2 - \alpha)^{3/2}} \left(- \frac{(m^2 - \alpha)^{1/2} \alpha^{1/2}}{m^2} + \arcsin \sqrt{\frac{m^2 - \alpha}{m^2}} \right), \quad (8)$$

which for spheres, $m = 1$, reduces to

$$\Gamma_2^{\parallel} = - \frac{1}{\sigma^{\parallel}} \left(\frac{\alpha^{1/2}}{(\alpha - 1)^{3/2}} \ln(\sqrt{\alpha - 1} + \sqrt{\alpha}) - \frac{1}{\alpha - 1} \right), \quad (9)$$

$$\Gamma_2^{\perp} = - \frac{1}{2\sigma^{\parallel}} \left(- \frac{\alpha^{-1/2}}{(\alpha - 1)^{3/2}} \ln(\sqrt{\alpha - 1} + \sqrt{\alpha}) + \frac{1}{\alpha - 1} \right). \quad (10)$$

For 2D disks, on the other hand, one obtains

$$\Gamma^{\parallel} = - \frac{1}{\sigma^{\parallel}(1 + \sqrt{\alpha})}, \quad (11)$$

$$\Gamma^{\perp} = - \frac{1}{\sigma^{\parallel}\sqrt{\alpha}(1 + \sqrt{\alpha})}. \quad (12)$$

It is easy to see that these Γ 's depend only on the microscopic geometry and on the effective-medium conductivity outside the grains. For the most simple case of isotropic spheres one has that the various Γ 's depend only on dimensionality and on the effective conductivity through $\Gamma = -1/(\sigma d)$. In that case Eq. (6) becomes identical to the isotropic limit of the random resistor result (1).

We have assumed that all the spheroids are aligned along their c axis, but this is not what is reported in the experimental data where there is a 3° misalignment around the c axis. This misorientation can be taken into account by writing the Γ in terms of an arbitrary angle θ with respect to the direction of the field and averaging over the solid angle.

C. Numerical simulations

Simulations have been done for 100×100 square lattices using the algorithm of Frank and Lobb⁹ based on the Y - Δ transformation in electrical circuits. Keeping the top and bottom rows of bonds with very large conductivity compared with all the bulk bonds preserves the effect of anisotropy.

Figure 1 is a plot of the conductivity of the network as a function of the concentration of type-1 bonds for $\sigma_1^{\parallel} = 10^5$, $\sigma_1^{\perp} = 10^3$, $\sigma_2^{\parallel} = \sigma_2^{\perp} = 1$. For small concentrations, which is the region we are interested in, the EMA represented by the solid line is in reasonable agreement with the simulations. At the percolation threshold, $p = 0.5$, the simulations seem to suggest that there is a difference between parallel and perpendicular directions, but using more detailed finite-size scaling analysis, we have checked that the macroscopic conductivity at that point is indeed isotropic, as expected. Above

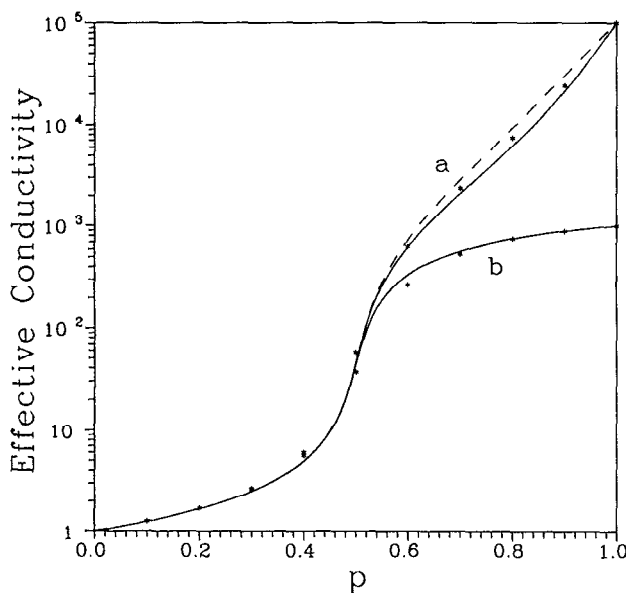


FIG. 1. Effective conductivity as a function of the concentration of type-1 bonds ($\sigma_1^{\parallel} = 10^5$, $\sigma_1^{\perp} = 10^3$, $\sigma_2^{\parallel} = \sigma_2^{\perp} = 1$). The * represent the simulations and the lines the effective-medium approximation. Solid lines correspond to discrete EMA, Eqs. (1), and dashed lines to continuous EMA, Eqs. (6)–(12). Curves near the label a represent the parallel effective conductivity and those near b the perpendicular conductivity. Note that for the latter case both effective-medium approximations coincide.

the two-dimensional percolation threshold the agreement for the parallel conductivity is less satisfactory, but nevertheless, the EMA and simulations agree within 20% over the whole range of concentrations. As a physical interpretation of the disagreement for values of p above 0.5, we suggest that in that regime the smallest conductivity limits the conduction process so that the effect of the largest conductivity on the overall conductivity becomes a small perturbation which is difficult to properly treat within the EMA. An additional complication comes from the appearance of an effective percolation threshold around $p = 1$, as discussed below.

As is well known, isotropic effective-medium theory is accurate when the ratio of the conductivities is not too large. In our case, a new ingredient, anisotropy, has been added and the limitations of the theory must be discussed. Figure 2 is a plot of the results of simulations for an isotropic “host” ($\sigma_2^{\parallel} = \sigma_2^{\perp} = 1$) and varying degrees of anisotropy, the largest of the conductivities being fixed at 10^5 . At small concentrations of the anisotropic material, the macroscopic conductivity is weakly dependent on microscopic anisotropy. Also, anisotropy is easier to see when one of the anisotropic conductivities approaches that of the host. The EMA results do not appear on the figure, but we can mention that when the anisotropy of the system is weak (parallel and perpendicular conductivities are about the same order of magnitude), the agreement is very good. However, when increasing the anisotropy ratio or the ratio between the conductivities of the anisotropic material and that of the host, the agreement is lost mainly around and above the percolation threshold (0.5). The worst disagreement for the simulations of Fig. 2 is by a factor of about 1.5 and it occurs for very large anisotropies near $p = 1$. Indeed, when only one of the conductivities is much larger than the other two, the medium looks like

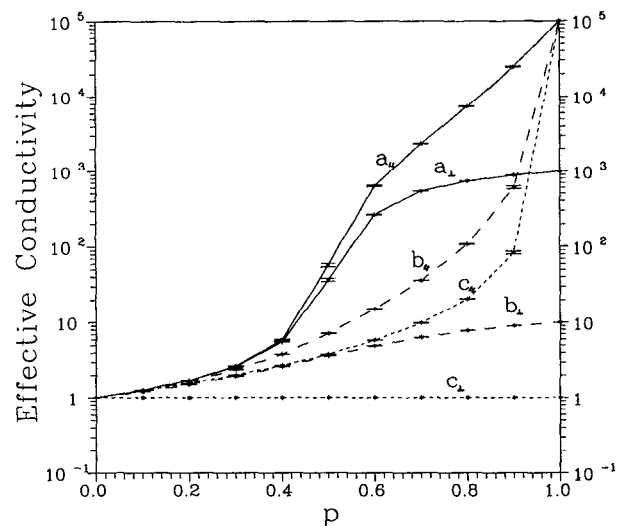


FIG. 2. Effective conductivity obtained from numerical simulations on 100×100 random resistor networks as a function of the concentration of higher-conductivity bonds. Error bars correspond to statistical errors in the sets of realizations. In the different curves, the parallel conductivity of type-1 bonds ($\sigma_1^{\parallel} = 10^5$) as well as that of type-2 bonds ($\sigma_2^{\parallel} = \sigma_2^{\perp} = 1$) are kept constant while the perpendicular conductivity is varied: (a) solid line, $\sigma_1^{\perp} = 10^3$, (b) long-dashed line, $\sigma_1^{\perp} = 10^1$, (c) short-dashed, $\sigma_1^{\perp} = 1$. The lines are guides to the eye.

parallel one-dimensional strings of highly conducting material, and as such it has an apparent percolation threshold near $p = 1$, as can be seen from cases (b) and (c) on the figure. In particular, the fluctuations are larger near $p = 1$, as expected in such a case, and the effective-medium approximation in this limit gives worse results. The numerical simulations of this section show that the EMA works very well in two dimensions. It should be even more reliable in the three-dimensional case where fluctuations are smaller.

III. INTERPRETATION OF THE EXPERIMENTAL RESULTS

The reported experimental results of Poirier *et al.*¹ are 17-GHz microwave absorption measurements of the temperature-dependent dielectric tensor of a sample consisting of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ crystals embedded in epoxy resin. The single-crystal grains are spheroids 3–6 μm in axis length and the concentration of the powder is about 15% in volume. The crystals were aligned along the c axis with a precision of 3° . The dielectric properties of the epoxy were also measured.

Our previous discussion has been about conductivity. At frequencies which are larger than zero, but small enough that the grain size is smaller than the skin depth, it suffices to replace conductivity by dielectric constant in the formulas.^{4,5} In the experiment of Poirier *et al.*, in the microwave regime, the estimated skin depth is slightly larger than the grain size. Finally, numerical simulations of Ref. 6 for superconducting isotropic spheres embedded in a normal-conducting medium¹⁰ confirm that the value of the density used in the experiment is far enough from the percolation threshold to avoid possible problems with the EMA.

To fit the experimentally measured dielectric constant of the composite, shown in Fig. 3, knowing that of the epoxy, shown in Fig. 1 of Ref. 1, we first impose in Eqs. (6)–(10) that the real part of the dielectric constant of the supercon-

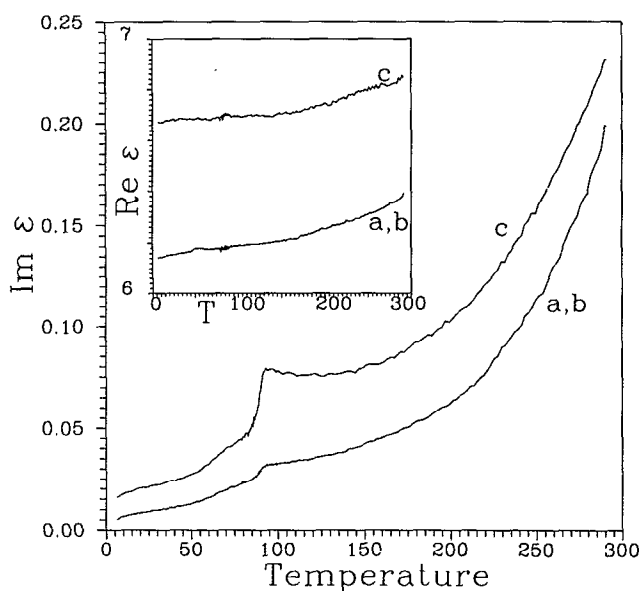


FIG. 3. Imaginary and real (inset) parts of the dielectric tensor from Ref. 1. The measurements are for both the parallel components (c axis) and the perpendicular ones (a - b plane).

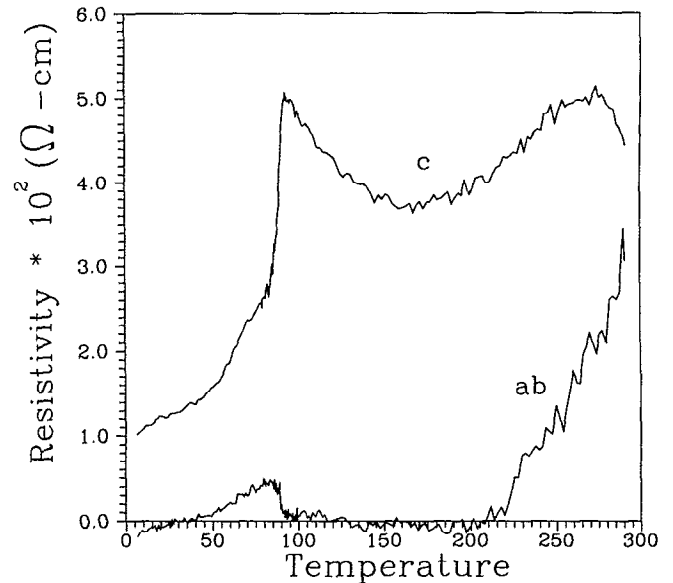


FIG. 4. Resistivity along the different axis extracted from the 17-GHz experimental data plotted in Fig. 3 via the EMA of Eqs. (6)–(12).

ductor be very large and negative in the superconducting state ($T < T_c$) and very large and positive in the normal state¹¹ ($T > T_c$). This allows us to adjust the density and the aspect ratio of the spheroids to p about¹² 18% and m about 1.1. Once these parameters are known, the parallel and perpendicular complex dielectric constants of the superconductor can be obtained. From these dielectric constants, we extract the resistivities in both principal axis directions, as shown in Fig. 4. Note that the high-resistivity result is the one with less noise. The resistivities in the present problem are in fact almost directly proportional to the conductivities, or equivalently to the imaginary part of the dielectric constant, because the magnitude of the real part of the dielectric constant is large.

One can check that the resistivity obtained for the c direction is of the same order of magnitude as the data obtained from the zero-frequency measurements of Ref. 13 at least for $T > T_c$. The measured value of the dc resistivity¹³ near T_c in the a - b plane is about two orders of magnitude smaller than along the c axis but it does not agree so well with that extracted from the microwave experiment using the effective-medium approximation (Fig. 4). One of the reasons for this is undoubtedly that the errors in the measurements lead to errors in the calculated resistivity in the a - b plane which are of the same order of magnitude as the extracted resistivity itself. Also, as discussed in the previous section, for the small values of the density considered here the distinction between the parallel and perpendicular conductivities is more difficult and the macroscopic conductivity scales mainly as the smallest value, in this case, the c axis. Note also that at high temperatures, where both parallel and perpendicular to c -axis resistivities become comparable, they tend to merge and it becomes difficult to separate them.

Perhaps the most interesting result from the point of view of the physics of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is the c -axis resistivity for $T < T_c$ in Fig. 4. While absorption at finite frequency

occurs in superconductors¹⁴ even for $T < T_c$, for an s -wave superconductor it usually vanishes exponentially at zero temperature. The residual zero-temperature resistivity observed in Fig. 4 could come from nodes in the superconducting gap,¹⁵ or from parasitic effects such as weak links or other phases, as discussed in Ref. 1. However, the highly anisotropic nature of the zero-temperature absorption makes the latter two parasitic effects less likely. Skin-depth effects not accounted for in the EMA would also tend to make the absorption smaller instead of large as observed. We are thus lead to favor the hypothesis of the existence of states at the Fermi energy in the superconducting gap or from other sources. The hypothesis would also be consistent with the measurements¹ on the same samples performed in the alternating magnetic field configuration. Indeed, when eddy currents flow in the a - b plane, there is negligible low-temperature absorption, but when they flow in a plane which includes the c axis there is sizeable absorption in the zero-temperature limit.

Other complications in the interpretation of the data may come from the fact that in principle one should also take into account the misalignment of the crystals, but explicit calculations of these corrections show that here they do not appreciably affect the final result. When analyzing experiments, more accurate results can in principle be obtained by considering models which include the nonuniform distribution of shapes, the skin-depth effect, correlations between the different grains,^{16,17} or by developing, in analogy with Ref. 18, interpolation formulas which are not available yet for the anisotropic problem discussed here.

IV. CONCLUSIONS

In this paper we have studied, in the framework of the effective-medium approximation, how the microscopic anisotropy of oriented crystallites manifests itself in the macroscopic dielectric properties of the medium in which they are embedded. The results in the case of two dimensions have been checked by computer simulations. In the parameter range of interest here, we have seen that the effective-medium approximation can be applied confidently to obtain good estimates of the desired quantities.

Keeping in mind the limitations of the EMA, this approach has been applied to extract the temperature-dependent resistivities of the high-temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ from microwave measurements performed by Poirier *et al.*¹ on crystallites embedded in epoxy. The a - b plane resistivity is directly available to these experiments when a pure alternating magnetic field is applied perpendicular to the plane. Surprisingly, mainly because of the large value of the real part of the dielectric constant of these materials, we have shown here that this method is also a good probe of the resistivity in the direction of largest resistivity when the alternating electric field is applied to the sample. We remind the reader that the resistivity of powders is dominated by the low-resistivity component of the tensor. While the resistivity tensor of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ has now been accurately measured on large crystals, when new materials are synthesized and only microcrystallites are available, microwave absorption measurements on crystallites embedded

and oriented in epoxy may be the method of choice to obtain the first estimates of the complete anisotropic resistivity tensor. Furthermore, the experiments of Ref. 1 appear to date to be the only ones which have measured the finite frequency resistivity below T_c in the c direction. Perhaps the most striking result is that even in the limit of zero temperature the finite frequency resistivity along the c direction does not seem to vanish, as discussed in Sec. III. This would be consistent with tunneling experiments,¹⁹ for example, which suggest the existence of states at the Fermi surface even in the superconducting state.

ACKNOWLEDGMENTS

We are indebted to G. Quirion and M. Poirier for the data of Fig. 3 and for numerous illuminating discussions. A.D.-G. is supported by a Postdoctoral Fellowship from the Spanish Ministry of Education and also by a grant of CICyT (No. PB 87-0782) for a previous visit to Sherbrooke. This work was supported by the Natural Sciences and Engineering Research Council of Canada and by the Ministère de l'Éducation du Québec.

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