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## Cell Percolation Model for Electrical Conduction of Granular Superconducting Composites

### I. The Resistivity of an Individual Cell of the Composite

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A simple model is given to determine the resistivity of the elongated granular superconducting composites in the normal and superconducting states. The model is constructed from uniform cells to fill the entire volume of the composite wire or tape. For the uniform cells a random distribution is assumed. The individual cells consist of cylindrical or parallelepiped elongated superconducting filaments surrounded by a normal matrix. In the calculation of the resistivity-temperature curve of an individual cell the proximity effect is taken into account.

Es wird ein einfaches Modell zur Bestimmung des Widerstands der langgranularen supraleitenden Zusammensetzungen im normal- und supraleitenden Zustand angegeben. Das Modell ist aus einheitlichen Zellen konstruiert, die das gesamte Volumen des Drahtes oder Bandes füllen. Für die einheitlichen Zellen wird eine statistische Verteilung angenommen. Die individuellen Zellen bestehen aus langen zylindrischen oder parallelepipedisch geformten supraleitenden Drähten, die von einer normalen Matrix umgeben sind. Bei der Berechnung der Widerstands-Temperaturkurve einer individuellen Zelle wird der Nahwirkungs-Effekt berücksichtigt.

### 1. Introduction

The methods of production of commercial multifilamentary superconducting composites are very complicated and expensive, and they have some disadvantages mainly in mechanical aspects. The alternative way to make superconducting composites is the in situ filamentary technique [1], when the mixed solid constituents are melted together, cooled at a controlled rate and the filaments are formed by subsequent drawing and/or flattening. In this case the discontinuous superconducting filaments have a random distribution.

In the cooled mixture the shape and size of the superconducting inclusions depend on the fabrication and the cooling processes but in many cases they have a quasi-spherical symmetry and a very small size ( $\approx 10 \mu\text{m}$ ) [2]. After the drawing and/or the flattening of the cast of the composite the filaments can be considered as cylinders or thin parallelepipeds.

For the superconductivity of these materials with discontinuous superconducting filaments there are some explanations. As has been shown in highly reduced wires there is a very low resistance when the filaments do not touch [3]. Through the random contacts a continuous network can be formed [4], that is the filaments percolate in superconducting state. In the development of the percolative chains the proximity effect of the filaments can be taken into account, too [5].

Our aim is to give a simple model to calculate the resistance of an in situ superconducting tape in normal and superconducting states. The elongated and flattened

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filaments are assumed to be parallel to the direction of elongation. According to the Carr model [6, 7] in a strongly in situ elongated superconductor the filaments are very close to each other, they have numerous neighbouring filaments and the whole material can be considered as a continuum which has a quasi-translational symmetry.

To construct a uniform cell around a filament the distances between the neighbouring filaments must be divided into halves. The uniform model consists of such uniform cells with the only filament placed coaxially. When the non-uniformities of the filaments do not play a great role the superconducting tape can be treated as a continuum with anisotropic properties and the resistivity can be calculated on the basis of the determination of the resistivity of a uniform cell alone.

## 2. Normal State Resistivity of in situ Composite

We consider the uniform cell of the superconducting tape as a parallelepiped shown in Fig. 1, where  $x_1, y_1, z_1, x_f, y_f, z_f$ , are the length, the width, and the thickness of the cell and the filament, respectively. The directions of elongation and flattening are  $x$  and  $y$ , respectively. In our model a reasonable assumption for the filaments is  $x_f \gg y_f \gg z_f$ . In the case of a superconducting wire  $x_f \gg y_f, x_f \gg z_f$ , and  $y_f \approx z_f$  are valid but that does not alter our discussion in the least. The resistivities of the matrix and the filaments are  $\varrho_m$  and  $\varrho_f$ , respectively.

The resistance  $R_x$  in direction  $x$  of one cell can be calculated from the integral

$$\frac{1}{R_x} = \int_0^{z_1} \int_0^{y_1} \frac{dy dz}{\int_0^{x_1} \varrho(x, y, z) dx}, \quad (1)$$

and in the same way the resistances  $R_y$  and  $R_z$  can be deduced, where  $\varrho(x, y, z)$  is the resistivity function in the whole cell. Solving the integrals we get the following expressions:

$$\begin{aligned} \frac{1}{R_x} &= \frac{y_1 z_1 - y_f z_f}{\varrho_m x_1} + \frac{y_f z_f}{\varrho_m (x_1 - x_f) + \varrho_f x_f}, \\ \frac{1}{R_y} &= \frac{x_1 z_1 - x_f z_f}{\varrho_m y_1} + \frac{x_f z_f}{\varrho_m (y_1 - y_f) + \varrho_f y_f}, \\ \frac{1}{R_z} &= \frac{x_1 y_1 - x_f y_f}{\varrho_m z_1} + \frac{x_f y_f}{\varrho_m (z_1 - z_f) + \varrho_f z_f}. \end{aligned} \quad (2)$$

Using the fact that the volume of the superconducting composite does not change significantly under drawing and/or flattening, we can write

$$x_1 y_1 z_1 = x_0 y_0 z_0 = x_f y_f z_f / c = x_{f0} y_{f0} z_{f0} / c, \quad (3)$$

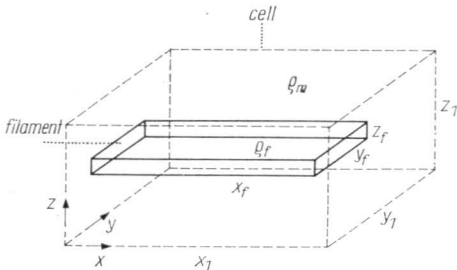


Fig. 1. The uniform cell of an elongated and flattened granular superconductor

where  $x_0, y_0, z_0$  and  $x_{f0}, y_{f0}, z_{f0}$  are the starting dimensions of the formed uniform cell and filament, respectively, and  $c$  is the superconducting volume ratio in the cell. Let us introduce the relations

$$E = x_1/x_0 = x_f/x_{f0} \quad \text{and} \quad k = y_1/z_1 = y_f/z_f \quad (4)$$

to characterize the elongation and the flattening, assuming that the matrix and the filaments are deformed simultaneously. If the macroscopic starting dimensions of our composite are  $X_0, Y_0, Z_0$ , and there are  $N$  superconducting inclusions placed at  $n_x, n_y, n_z$  in directions  $x, y, z$ , we can express the starting dimensions of the cell by the equations

$$x_0 = X_0/n_x, \quad y_0 = Y_0/n_y, \quad z_0 = Z_0/n_z, \quad (5)$$

while for  $n_x, n_y, n_z$  the equation  $N = n_x n_y n_z$  exists. For the superconducting concentration the expression

$$c = N \frac{x_{f0} y_{f0} z_{f0}}{X_0 Y_0 Z_0} \quad (6)$$

can be derived. Using starting quasi-spherical symmetry which has been confirmed experimentally in many cases [2], we can apply the notation  $L_0 = X_0 = Y_0 = Z_0$ ,  $l_0 = x_0 = y_0 = z_0$ , and  $l_{f0} = x_{f0} = y_{f0} = z_{f0}$  for each dimension as an average value over all the grains. With the aid of (3) to (6) the length, the width, and the thickness of the cell and the filament are

$$\begin{aligned} x_1 &= c^{-1/3} l_{f0} E, & x_f &= l_{f0} E, \\ y_1 &= c^{-1/3} l_{f0} (k/E)^{1/2}, & y_f &= l_{f0} (k/E)^{1/2}, \\ z_1 &= c^{-1/3} l_{f0} (kE)^{-1/2}, & z_f &= l_{f0} (kE)^{-1/2}, \end{aligned} \quad (7)$$

where all parameters of the dimensions are macroscopic, namely the elongation  $E$ , the flattening  $k$ , the superconducting volume ratio  $c$ , and the superconducting grain size  $l_{f0}$ .

The resistances of the cell in different directions can be expressed with these macroscopic parameters

$$\begin{aligned} R_x^{-1} &= \frac{l_{f0}}{E^2} F(\varrho_m, \varrho_f, c), \\ R_y^{-1} &= \frac{E l_{f0}}{k} F(\varrho_m, \varrho_f, c), \\ R_z^{-1} &= E k l_{f0} F(\varrho_m, \varrho_f, c), \end{aligned} \quad (8)$$

where

$$F(\varrho_m, \varrho_f, c) = \frac{1}{\varrho_f + \varrho_m(c^{-1/3} - 1)} + \frac{c^{-1/3} - c^{1/3}}{\varrho_m}. \quad (9)$$

Taking into account of the distribution of the grain size we have to solve the integrals

$$R_i = \frac{\int_0^m R_i(l_{f0}) L(l_{f0}) dl_{f0}}{\int_0^m L(l_{f0}) dl_{f0}}; \quad i = x, y, z, \quad (10)$$

where  $m$  and  $L(l_{f0})$  are the maximum and the distribution function of the grain size, respectively. In many cases the distribution function can be represented well by a logarithmic normal distribution, but it always depends strongly on the production procedures. Nevertheless we use the average value of the dimensions in our calculations [8].

So the anisotropic resistivities of a uniform cell are

$$\begin{aligned}\varrho_x &= R_x y_1 z_1 / x_1 = R_x l_{f0} / (E^2 c^{1/3}), \\ \varrho_y &= R_y x_1 z_1 / y_1 = R_y l_{f0} E / (k c^{1/3}), \\ \varrho_z &= R_z x_1 y_1 / z_1 = R_z l_{f0} E k c^{-1/3}.\end{aligned}\quad (11)$$

### 3. Resistivity of a Superconducting Uniform Cell

The average cell resistivities given above are good approximations for the normal bulk composite when the resistivities of the constituents do not differ strongly. In superconducting state the resistivities of the filaments will be zero and the current will find an advantageous path in the cell. In such a case the resistivities must be calculated from the average over the parts of the cell carrying the current. A more exact model is given by Carr [6, 7] taking into account the dependence of the properties of the superconducting composite on the current densities.

Our aim is to examine the dependence of the uniform cell resistivities on the temperature below the critical temperature  $T_c$  of the filaments. The transition curves of in situ composite superconductors have a very strange behaviour contrary to the continuous multifilamentary superconductors. Before the vanishing of the resistance they have a plateau. Our aim is to interpret qualitatively this plateau using the uniform cell model.

In our discussion we take into consideration the appearance of the Josephson jackets around the superconducting filaments below  $T_c$ . The characteristic thickness of the Josephson jacket is the coherence length  $\zeta$  of the superconducting filament,

$$\zeta = \left( \frac{h v_F \lambda_e}{12 \pi^2 k_B T} \right)^{1/2}, \quad (12)$$

where  $h$ ,  $v_F$ ,  $\lambda_e$ ,  $k_B$ , and  $T$  are the Planck constant, the Fermi velocity, the electron mean free path, the Boltzmann constant, and the temperature, respectively. As we can see from (12), the coherence length  $\zeta$  increases with decreasing temperature, and through  $\lambda_e$  it decreases with increasing impurity concentration. In this way the effective superconducting concentration  $c_{\text{eff}}$ , namely the filaments and their Josephson jackets together, increases with decreasing temperature in an elongated and/or flattened in situ superconductor,

$$c_{\text{eff}} = c \left( 1 + 2 \frac{\zeta}{x_f} \right) \left( 1 + 2 \frac{\zeta}{y_f} \right) \left( 1 + 2 \frac{\zeta}{z_f} \right). \quad (13)$$

In developing the bulk superconductivity these overlapping uniform cells percolate, so the in situ composite will be a macroscopic superconductor when the effective concentration reaches the percolative threshold value  $c_p$ . The plateau is created by the enhancements of the filament proximity effect and the cell percolation.

In the uniform model the following replacements are needed:

$$\varrho_f = 0, \quad x_f \rightarrow x_f + 2\zeta, \quad y_f \rightarrow y_f + 2\zeta, \quad z_f \rightarrow z_f + 2\zeta, \quad (14)$$

to introduce zero resistivity inside the Josephson jacket. Using (2), (7), (11), and (14) we get the resistivities  $\varrho_{xs}$ ,  $\varrho_{ys}$ ,  $\varrho_{zs}$  of a uniform cell below  $T_c$ ,

$$\begin{aligned}\varrho_{xs}^{-1} &= \varrho_m^{-1} \frac{E c^{1/3}}{l_{f0}} \left[ \frac{\frac{l_{f0}^2}{c^{1/3}E} - \left( l_{f0} \frac{k^{1/2}}{E^{1/2}} + 2\zeta \right) \left( \frac{l_{f0}}{(kE)^{1/2}} + 2\zeta \right) c^{1/3}}{l_{f0}E} + \right. \\ &\quad \left. + \frac{\left( l_{f0} \frac{k^{1/2}}{E^{1/2}} + 2\zeta \right) \left( \frac{l_{f0}}{(kE)^{1/2}} + 2\zeta \right)}{\frac{l_{f0}E}{c^{1/3}} - l_{f0}E - 2\zeta} \right], \\ \varrho_{ys}^{-1} &= \varrho_m^{-1} \frac{k c^{1/3}}{l_{f0}E} \left[ \frac{\frac{l_{f0}^2}{c^{1/3}(kE)^{1/2}} - (l_{f0}E + 2\zeta) \left( \frac{l_{f0}}{(kE)^{1/2}} + 2\zeta \right) c^{1/3}}{l_{f0} \frac{k^{1/2}}{E^{1/2}}} + \right. \\ &\quad \left. + \frac{(l_{f0}E + 2\zeta) \left( \frac{l_{f0}}{(kE)^{1/2}} + 2\zeta \right)}{\frac{l_{f0}k^{1/2}}{c^{1/3}E^{1/2}} - l_{f0} \frac{k^{1/2}}{E^{1/2}} - 2\zeta} \right], \\ \varrho_{zs}^{-1} &= \varrho_m^{-1} \frac{c^{1/3}}{l_{f0}kE} \left[ \frac{\frac{l_{f0}^2 k^{1/2}}{c^{1/3}E^{1/2}} - \left( l_{f0} \frac{k^{1/2}}{E^{1/2}} + 2\zeta \right) (l_{f0}E + 2\zeta) c^{1/3}}{\frac{l_{f0}}{(kE)^{1/2}}} + \right. \\ &\quad \left. + \frac{\left( l_{f0} \frac{k^{1/2}}{E^{1/2}} + 2\zeta \right) (l_{f0}E + 2\zeta)}{\frac{l_{f0}}{c^{1/3}(kE)^{1/2}} - \frac{l_{f0}}{(kE)^{1/2}} - 2\zeta} \right].\end{aligned}\quad (15)$$

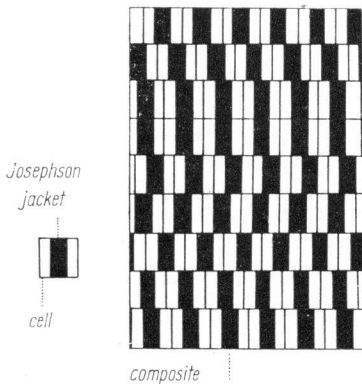


Fig. 2

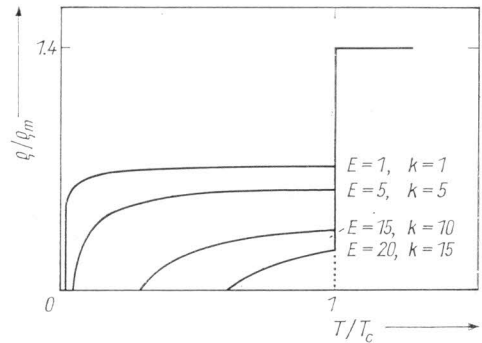


Fig. 3

Fig. 2. A two-dimensional example for a non-superconducting in situ granular wire with superconducting cells

Fig. 3. The superconducting transition curve of a uniform cell at different values of elongation  $E$  and flattening  $k$

The uniform cell goes into a superconducting state in the different directions when the Josephson jacket reaches the corresponding walls of the cell. To obtain the zero resistivities in the different directions the following coherence lengths are appropriate:

$$\begin{aligned}\zeta_x &= \frac{1}{2} l_{f0} E (c^{-1/3} - 1), \\ \zeta_y &= \frac{1}{2} l_{f0} (k/E)^{1/2} (c^{-1/3} - 1), \\ \zeta_z &= \frac{1}{2} l_{f0} (kE)^{-1/2} (c^{-1/3} - 1).\end{aligned}\quad (16)$$

The anisotropy on account of the elongation and/or flattening manifests itself in the direction dependent coherence lengths. Of course with these values of the coherence length the composite does not surely become a macroscopic superconductor in the corresponding direction because of the random cell distribution as can be shown in Fig. 2 in a plane, for example.

The critical temperatures of a uniform cell can be derived from the coherence lengths,

$$\begin{aligned}T_{cx}^* &= \frac{h v_F \lambda_e}{3\pi^2 k_B l_{f0}^2 E^2 (c^{-1/3} - 1)^2}, \\ T_{cy}^* &= \frac{h v_F \lambda_e E}{3\pi^2 k_B l_{f0}^2 k (c^{-1/3} - 1)^2}, \\ T_{cz}^* &= \frac{h v_F \lambda_e k E}{3\pi^2 k_B l_{f0}^2 (c^{-1/3} - 1)^2}.\end{aligned}\quad (17)$$

Comparing the temperatures, the relations  $T_{cz}^* \gg T_{cx}^*$  and  $T_{cz}^* \gg T_{cy}^*$  are obvious in a strongly elongated and flattened in situ superconductor. Although the uniform cells are superconducting below these temperatures the superconductivity of the whole composite is a percolative task.

To demonstrate the developing of the cell superconductivity the resistivities of the cell in any direction are plotted in Fig. 3 against the temperature. The resistivities of the cell are calculated from (11) and (15) in the temperature range  $T > T_c$  and  $T_c \geq T \geq T_{ci}^*$ , respectively. Below the temperature  $T_{ci}^*$  the resistivities are considered as zero. In the calculation we used the following data chosen reasonably:  $c = 0.15$ ,  $l_{f0} = 10^{-5}$  m,  $\lambda = 2.5 \times 10^{-7}$  m,  $v_F = 1.5 \times 10^6$  ms $^{-1}$ ,  $\rho_f/\rho_m = 10^3$ , and the parameters  $E$  and  $k$ .

#### 4. Conclusions

As we can see in Fig. 3 the dependence of the resistivities of our uniform cell on the temperature is very similar to the curves which can be obtained in experiments in the case of conventional granular superconductors [1 to 7] and some high  $T_c$  superconductors [9, 10]. The transition curve has a plateau depending on the elongation and/or flattening. Increasing the deformations, the temperature  $T_{ci}^*$  comes closer and closer to the critical temperature  $T_c$  of the filament. First of all through the electron mean free path the increasing impurity changes the transition curves slightly emphasizing the plateau.

In a real granular superconductor the character of transition curve is the same but the zero value of the resistance appears at a different temperature  $T_c^{**}$  taking into account the percolation of the uniform cells. The calculation of the resistance-temperature curve of the percolating uniform cells can be found in the next part of this paper.

In the treatment of this topic the resistivity increase due to the size effect can be neglected in the normal state of the composite as has been shown previously [11 to 13].

### References

- [1] C. C. TSUEI, *Science* **180**, 57 (1973).
- [2] C. J. LOBB, *Tech. Rep.* **16**, (1980).
- [3] A. DAVIDSON, M. R. BEASLEY, and M. TINKHAM, *IEEE Trans. Magnetics* **11**, 276 (1975).
- [4] A. DAVIDSON and M. TINKHAM, *Phys. Rev. B* **13**, 3261 (1976).
- [5] C. C. TSUEI and L. R. NEWKIRK, *J. Mater. Sci.* **8**, 1307 (1973).
- [6] W. J. CARR, *J. appl. Phys.* **54**, 5911 (1983).
- [7] W. J. CARR, *J. appl. Phys.* **54**, 5917 (1983).
- [8] C. C. KOCH and G. R. LOVE, *J. appl. Phys.* **40**, 3582 (1969).
- [9] S. YOMO, C. MURAYAMA, H. TAKAHASHI, N. MORI, K. KISHIO, K. KITAZAWA, and K. FUEKI, *Japan. J. appl. Phys.* **26**, L603 (1987).
- [10] J. BÁNKUTI, M. GÁL, G. HORVÁTH, I. KIRSCHNER, K. G. SÓLYMOS, and K. TORKOS, *Japan. J. appl. Phys.* **26**, 1073 (1987).
- [11] G. HORVÁTH and J. BÁNKUTI, *phys. stat. sol. (a)* **110**, 549 (1988).
- [12] J. BÁNKUTI and G. HORVÁTH, *phys. stat. sol. (a)* **111**, K185 (1989).
- [13] G. HORVÁTH and J. BÁNKUTI, *Cryogenics* **29**, 931 (1989).

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