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Central Research Institute for Physics, Hungarian Academy of Sciences, Budapest¹) (a) and Department of Low Temperature Physics, Eötvös University, Budapest (b)

Cell Percolation Model for Electrical Conduction of Granular Superconducting Composites

II. Percolation Conductivity of the Uniform Cells²)

By

G. Horváth (a) and J. Bánkuti (b)

The percolation of the electrical conductivity of the uniform cells is studied in an in-situ elongated granular superconducting composite on the basis of the uniform cell model improved previously. The critical temperatures are determined in the macroscopic superconducting state of the two- and the three-dimensional composites.

In einer in-situ-verlängerten granularen supraleitenden Zusammensetzung wird auf der Grundlage des kürzlich verbesserten Modells einheitlicher Zellen die Perkolation der elektrischen Leitfähigkeit untersucht. Die kritischen Temperaturen des makroskopischen Supraleitungszustands der zwei- und drei-dimensionalen Zusammensetzungen werden bestimmt.

1. Introduction

The limits of the conventional technologies and the advantages of the new in-situ technology of the production of the superconducting composites are discussed in our previous paper [1] and elsewhere [2 to 4].

In the recent cell percolation model based on the uniform cells [1] the macroscopic superconductivity of the elongated multifilamentary in-situ granular superconductors is examined. The use of a uniform cell for the description of the properties of the whole material is supported by the Carr model [5, 6] and the superconductor is treated as an anisotropic continuum. The cylindrical or parallelepiped individual uniform cells consist of an elongated filament and the normal matrix around it and they fill the entire volume of the composite wire or tape. The filaments are assumed to be parallel with the elongation direction.

In the calculation of the resistivity of an individual cell the Josephson jacket around the filaments and the increase of the effective superconducting concentration are taken into account. The resistivity—temperature curves of a uniform cell in different directions are very similar to the curves obtained for the conventional granular superconductors [2 to 4] and for some high- T_c superconductors [7, 8]. They have a significant plateau and a critical temperature T_{ci}^* below the critical temperature T_c of the filament. The length of the plateau and the value of the critical temperatures depend on the elongation and slightly on the impurities.

¹⁾ P.O.B. 49, H-1525 Budapest, Hungary.

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In a real granular superconductor in spite of the superconductivity of the uniform cells the macroscopic superconductivity appears only at a lower temperature $T_{\rm ci}^{**}$ ($< T_{\rm ci}^{*}$) because of the random distribution of the superconducting filaments. The transition curve of an in-situ superconducting composite can be divided into the following parts according to lowering temperatures:

- 1. Normal state in the temperature range $T > T_{\rm e}$, when the filaments, the uniform cells, and the conductor are in normal state.
- 2. Submicroscopic superconducting state in the temperature range $T \stackrel{e}{\geq} T > T_{ei}^*$, when the filaments are in superconducting state, and the uniform cells and the conductor are in normal state.
- 3. Microscopic superconducting state in the temperature range $T_{\text{ci}}^* \geq T > T_{\text{ci}}^{**}$, when the filaments and the uniform cells are in superconducting state, and the conductor is in normal state.
- 4. Macroscopic superconducting state in the temperature range $T_{ci}^{**} \ge T > 0$ when the filaments, the uniform cells, and the conductor are in superconducting state.

At present we examine the macroscopic conduction properties of the elongated filamentary granular superconducting wires or tapes. In the development of the macroscopic superconductivity the percolation of the uniform cells must be taken into account and we get the macroscopic critical temperatures from the percolation of the uniform cells in two and three dimensions.

2. Cell Percolation in Two Dimensions

Let the elongated filamentary in-situ superconductor be in the microscopic superconducting state ($T_c^* \ge T > T_c^{**}$) in a given direction. This means in our uniform cell model that the Josephson jackets around the filaments have reached the walls of the cells in the given direction. To fill randomly the entire two-dimensional conductor with uniform cells including a filament coaxially there are two possibilities, namely the random translations of the uniform cells along one of its sides. The configuration of the translation along the filaments does not give a percolation task and the macroscopic superconductivity of the conductor is obvious. The translation perpendicular to the filaments, as can be seen in Fig. 2 of [1], is closer to a real random distribution of the uniform cells and we examine this arrangement further on.

Accordingly in our model the uniform cells are randomly translated at right angles to the direction of the superconducting bands of the uniform cells. The percolation problem of this arrangement can be formulated in the following way: Oblongs with breadth s and length b are randomly thrown in the cells of dimension [(a/2) + s] b of a channel with breadth (a/2) + b and in every cell only one oblong is orientated in the axis of the channel.

We examine the one-dimensional percolation of these oblongs. It is evident that in the case of $s \ge [(a/2) + s]/2$ there is a percolation chain of the oblongs in a channel of arbitrary length, but in the case of $0 \le s \le [(a/2) + s]/2$ the percolation chain in an infinite channel will be disconnected undoubtedly. For the band of breadth s in an infinite channel the critical percolation threshold is

$$s^{\infty} = \frac{a}{2}.\tag{1}$$

In a finite channel the percolation of the *i*-th and the (i + 1)-th cells depends on the ratio of the oblong breadth s and the cell breadth a and on their relative positions. If the distance of the *i*-th oblong from the wall of the channel is x_i , the following cases can be distinguished when the percolation of the neighbouring cells occurs:

I.
$$s \leq \frac{a}{4}$$
 if 1. $0 \leq x_i \leq s$, then $0 \leq x_{i+1} \leq x_i + s$,
2. $s < x_i < \frac{a}{2} - s$, $x_i - s \leq x_{i+1} \leq x_i - s$,
3. $\frac{a}{2} - s \leq x_i \leq \frac{a}{2}$, $x_i - s \leq x_{i+1} \leq \frac{a}{2}$;
II. $\frac{a}{4} < s < \frac{a}{2}$ 1. $0 \leq x_i \leq \frac{a}{2} - s$, $0 \leq x_{i+1} \leq x_i + s$; (2)
2. $\frac{a}{2} - s < x_i < s$, $0 \leq x_{i+1} \leq \frac{a}{2}$,
3. $s \leq x_i \leq \frac{a}{2}$, $x_i - s \leq x_{i+1} \leq \frac{a}{2}$;
III. $\frac{a}{2} \leq s \leq a$ 1. $0 \leq x_i \leq \frac{a}{2}$, $0 \leq x_{i+1} \leq \frac{a}{2}$.

The probabilities corresponding to the oblongs in the appropriate positions in the i-th cell are

$$P_{iI}^{(1)} = \frac{2s}{a}, P_{iI}^{(2)} = \frac{a-4s}{a}, P_{iI}^{(3)} = \frac{2s}{a},$$

$$P_{iII}^{(1)} = \frac{a-2s}{a}, P_{iII}^{(2)} = \frac{4s-a}{a}, P_{iII}^{(3)} = \frac{a-2s}{a}.$$
(3)

The probabilities corresponding to the oblongs in the appropriate positions in the (i + 1)-th cell are

$$P_{(i+1)I}^{(1)} = \frac{2(x_i + s)}{a}, \qquad P_{(i+1)I}^{(2)} = \frac{4s}{a}, \qquad P_{(i+1)I}^{(3)} = \frac{a - 2x_i + 2s}{a},$$

$$P_{(i+1)II}^{(1)} = \frac{2(x_i + s)}{a}, \qquad P_{(i+1)II}^{(2)} = 1, \qquad P_{(i+1)II}^{(3)} = \frac{a - 2x_i + 2s}{a}.$$
(4)

The probability for $x_i \in (x_i, x_i + dx_i)$ is

$$dP(x_i) = \frac{2 dx_i}{a}.$$
 (5)

Using (3) to (5) we can calculate the probability of the percolation of the *i*-th and (i + 1)-th

cells for the case K (K: I, II, III) and position k (k: 1, 2, 3) when $x_i \in (x_i, x_i + dx_i)$: $P_{iK}^{(k)} P_{(i+1)K}^{(k)} dP(x_i)$. Hence the probability of the percolation of the K-th ratio of s/a is

$$P_K = \sum_{k=1}^{3} \int_{x_i k_{\min}}^{x_i k_{\max}} P_{iK}^{(k)} P_{(i+1)K}^{(k)} \, \mathrm{d}P(x_i) \,. \tag{6}$$

Performing the calculations the concrete values of the probabilities in the cases mentioned above.

$$P_{\rm II} = \frac{8s}{a^3} \left[3s^2 + 2\left(\frac{a}{2} - 2s\right)^2 \right],$$

$$P_{\rm II} = \frac{8}{a^3} \left[\left(\frac{a}{2} + s\right) \left(\frac{a}{2} - s\right)^2 + \frac{a}{2} \left(2s - \frac{a}{2}\right) \right],$$

$$P_{\rm III} = 1.$$
(7)

The relations $P_{\rm I}$ < 1 and $P_{\rm II}$ < 1 can be realized easily from (7).

To get the probability of the formation of a superconducting chain from n cells we use the probabilities of the neighbouring cells,

$$P_{nK} = P_K^{n-1} \,, \tag{8}$$

where n-1 is an exponent. In the case of an infinite chain $(n \to \infty)$ there is no percolation at the superconducting bands of s < a/2, i.e. the probabilities go to zero, $P_{nK} \to 0$ (K: I, II).

If we have n_{\perp} cells in the direction of the translation of the uniform cells and n_{\parallel} cells in the direction of the percolation, it is clear that the number of the parallel percolation chains is n_{\perp} . The breadth of these percolation chains is equal to the breadth of the filament together with its Josephson jacket perpendicular to the direction of the superconducting chains.

As we have shown [1] the breadth of the filament together with its Josephson jacket depends on the temperature. The condition for the development of the superconducting percolation path is that the breadth of the superconducting stripe reaches the value a/2,

$$s(T_c^{**}) = \frac{a}{2}. \tag{9}$$

The condition of the microscopic superconducting state can be given similarly,

$$s'(T_c^*) = b. (10)$$

The bulk composite becomes a superconductor below the temperature T_c^{**} , and in the temperature range $T_c^{**} < T < T_c^{*}$ the probability of the bulk superconductivity is P_{nI} or P_{nII} depending on the parameters s and a.

From our consideration the temperature $T_{c_1}^*$ for the bulk superconductivity in the other direction is lower, because the condition is

$$s(T_{c}^{*}) = a, \tag{11}$$

and the temperature $T_{c_1}^{**} = T_{c}^*$ is obviously higher than $T_{c_1}^*$.

3. Cell Percolation in Three Dimensions

In a three-dimensional in-situ composite we have uniform cells of measures $x_1y_1z_1$ and superconducting prisms of measures $s_1s_2s_3$ as they are shown in Fig. 1. Let the uniform cell be in the microscopical superconducting state at first in the direction z as can be concluded reasonably in an in-situ composite elongated in the direction x and flattened in the plane xy. The percolation problem can be considered in analogy to the two-dimensional case, accordingly we place randomly oblongs of measures s_1s_2 in the cells of measures $[(x_1/2) + s_1][(y_1/2) + s_2]$ of the cross-section xy (Fig. 2a). The centres of the oblongs c_i can move randomly in a subcell of measures $(x_1/2)(y_1/2)$ and the condition of the percolation of the i-th and (i + 1)-th cells is that the centre c_{i+1} of the (i + 1)-th cell falls into the parallelepiped of measures $2s_12s_2$ around the centre c_i of the i-th cell as can be seen in Fig. 2b.

Similarly to the two-dimensional case the different possibilities for the percolation according to the positions of the oblongs (in this case nine possibilities) can be calculated numerically.

The probability for $x_i \in (x_i, x_i + dx_i)$ and $y_i \in (y_i, y_i + dy_i)$ is

$$dP(x_i, y_i) = \frac{4 dx_i dy_i}{x_i y_i}.$$
 (12)

In this case the probability of the percolation of the *i*-th and (i + 1)-th cells corresponding to the K-th relations of s_2/y_1 and s_2/y_2 , (K: I, ..., IX), and J_i -th, k-th positions of x_i and y_i , respectively, is $P_{iJ_iK}^{(k)}P_{(i+1)J_iK}^{(k)} dP(x_i, y_i)$, (k, l: 1, 2, 3). At last the probability of the *i*-th and (i + 1)-th cells corresponding to K-th relations is

$$P_{k} = \sum_{k,l=1}^{3} \int_{x_{l}^{(k)}}^{x_{l}^{(k)}} \int_{y_{l}^{(k)}}^{y_{l}^{(k)}} P_{iJ_{l}K}^{(k)} P_{(i+1)J_{l}K}^{(k)} \, \mathrm{d}P(x_{i}, y_{i}) \,. \tag{13}$$

Using (12) and (13), the percolation probabilities are

$$\begin{split} P_{\rm I} &= \frac{128s_1s_2}{x_1^3y_1^3} \left[\frac{9}{2} \, s_1^2 s_2^2 + \, 3s_2^2 \left(\frac{x_1}{2} - 2s_1 \right)^2 + \, 3s_1^2 \left(\frac{y_1}{2} - 2s_2 \right)^2 \right. \\ &\quad + \, 2 \left(\frac{y_1}{2} - 2s_2 \right)^2 \cdot \left(\frac{x_1}{2} - 2s_1 \right)^2 \right], \\ P_{\rm II} &= \frac{32s_1}{x_1^2y_1} \left[\, 3s_1^2 + \, 2 \left(\frac{x_1}{2} - 2s_1 \right)^2 \right] \\ &\quad \times \left[\left(2s_2 - \frac{y_1}{2} \right)^2 + \frac{2}{y_1} \left(\frac{y_1}{2} - s_2 \right) \left(\frac{y_1^2}{4} - s_2^2 \right)^2 \right], \\ P_{\rm III} &= \frac{16}{x_1^2y_1^2} \left[\left(2s_2 - \frac{y_1}{2} \right)^2 + \frac{2}{y_1} \left(\frac{y_1}{2} - s_2 \right) \left(\frac{y_1^2}{4} - s_2^2 \right)^2 \right] \\ &\quad \times \left[\left(2s_1 - \frac{x_1}{2} \right)^2 + \frac{2}{x_1} \left(\frac{x_1}{2} - s_1 \right) \left(\frac{x_1^2}{4} - s_1^2 \right)^2 \right], \\ P_{\rm IV} &= \frac{8}{y_1^3} \left(\frac{y_1}{2} - s_2 \right) \left(\frac{y_1^2}{4} - s_2^2 \right) + \frac{4}{y_1^2} \left(2s_2 - \frac{y_1}{2} \right)^2, \end{split}$$

$$P_{V} = \frac{24s_1^3}{x_1^3} + \frac{16s_1}{x_1^3} \left(\frac{x_1}{2} - 2s_1\right)^2,$$

$$P_{VI} = \frac{8}{x_1^3} \left(\frac{x_1}{2} - s_1\right) \left(\frac{x_1^2}{4} - s_1^2\right) + \frac{4}{x_1^2} \left(2s_1 - \frac{x_1}{2}\right)^2.$$

$$P_{VII} = 1,$$

$$P_{VII} = \frac{32s_2}{x_1^3} \left[3s_2^2 + 2\left(\frac{y_1}{2} - 2s_2\right)^2\right]$$
(14)

$$\begin{split} P_{\text{VIII}} &= \frac{32s_2}{x_1 y_1^2} \left[3s_2^2 + 2\left(\frac{y_1}{2} - 2s_2\right)^2 \right] \\ &\quad \times \left[\left(2s_1 - \frac{x_1}{2} \right)^2 + \frac{2}{x_1} \left(\frac{x_1}{2} - s_1\right) \left(\frac{x_1^2}{4} - s_1^2\right)^2 \right], \\ P_{\text{IX}} &= \frac{24s_2^3}{y_1^3} + \frac{16s_2}{y_1^3} \left(\frac{y_1}{2} - 2s_2\right)^2. \end{split}$$

The inequality $P_K < 1$ ($K \neq VII$) can be realized easily.

The probability of the formation of a superconducting chain from n cells in the case K is

$$P_{nK} = P_K^{n-1}; \quad K: I, ..., IX.$$
 (15)

The infinite superconducting percolation chain can be formed only in the case K = VII: $\lim_{n \to \infty} P_{nVII} = 1$. In other cases an infinite superconducting percolation chain cannot be

formed: $\lim_{n \to \infty} P_{nK} = 0 \ (K \neq VII).$

The critical temperature T_c^{**} can be determined by the following equations:

$$s_2(T_c^{\prime **}) = y_1/2, \qquad s_1(T_c^{\prime **}) = x_1/2.$$
 (16)

The real critical temperature T_c^{**} will be the lower one of the temperatures $T_c^{'**}$ and $T_c^{''**}$,

$$T_{c}^{**} = \min \left\{ T_{c}^{'**}, T_{c}^{"**} \right\}, \tag{17}$$

while the critical temperature T_c^* can be obtained from the equation

$$s_3(T_c^*) = z_1$$
 (18)

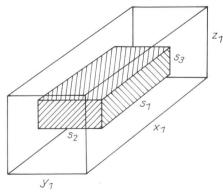


Fig. 1. Uniform cell of the three-dimensional model composite investigated. The dark parallelepiped (with dimensions s_1 , s_2 , s_3) in the middle of the cell (with dimensions x_1 , y_1 , z_1) is the Josephson jacket surrounding the superconducting filament

The conditions for the microscopic superconductivity in directions y and x are determined by the equations

$$s_2(T_{c_1}^{\prime*}) = y_1$$
 and $s_1(T_{c_1}^{\prime\prime*}) = x_1$. (19)

4. Application of Three-Dimensional Cell Percolation on the Uniform Model

The concrete measurements of the superconducting prism mentioned above can be written in following forms:

$$s_1 = x_f + 2\xi, \quad s_2 = y_f + 2\xi, \quad s_3 = z_f + 2\xi,$$
 (20)

where x_f , y_f , z_f , and ξ are the measurements of the filament and the coherence length, respectively.

The corresponding critical temperatures can be calculated from the equations

$$z_{\rm f} + 2\xi(T_{\rm cz}^*) = z_1, \qquad y_{\rm f} + 2\xi(T_{\rm cy}^*) = y_1, \qquad x_{\rm f} + 2\xi(T_{\rm cx}^*) = x_1, y_{\rm f} + 2\xi(T_{\rm cy}^{**}) = y_1/2, \qquad x_{\rm f} + 2\xi(T_{\rm cx}^{**}) = x_1/2.$$
(21)

Using the measurements of the uniform cell given in [1], the critical temperatures will be

$$T_{cy}^{**} = \frac{4hv_{F}\lambda_{e}E}{3\pi^{2}k_{B}l_{f0}^{2}k(c^{-1/3} - 2)^{2}},$$

$$T_{cx}^{**} = \frac{4hv_{F}\lambda_{e}}{3\pi^{2}k_{B}l_{f0}^{2}E^{2}(c^{-1/3} - 2)^{2}}.$$
(22)

The temperatures T_{ci}^* can be obtained from [1]. As we can see,

$$\lim_{c \to \frac{1}{8} - 0} \left\{ T_{cx}^{**}, T_{cy}^{**} \right\} = \infty , \qquad (23)$$

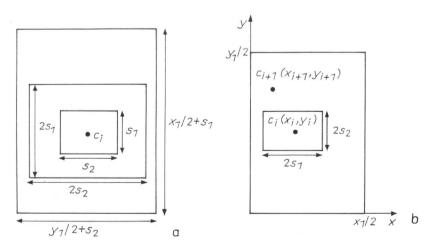


Fig. 2. Determination of the probability of percolation between two neighbouring uniform cells. Relative positions of the centres of the percolating i-th and (i + 1)-th cells

and from [1],

$$\lim_{c \to 0} \left\{ T_{cx}^*, T_{cy}^*, T_{cz}^* \right\} = 0 ,$$

$$\lim_{c \to 1} \left\{ T_{cx}^*, T_{cy}^*, T_{cz}^* \right\} = \infty .$$
(24)

So the temperatures $T_{ci}^{**} = \infty$ can be interpreted in the case of concentrations $c \ge 1/8$. In a superconducting tape elongated in direction z and flattened in the plane xy the inequalities

$$T_{cx}^* \le T_{cz}^*, \qquad T_{cy}^* \le T_{cz}^*, \qquad T_{cy}^* < T_{cy}^{**}, \qquad T_{cx}^* < T_{cx}^{**}$$
 (25)

can be realized. Consequently the uniform cell turns to the microscopic superconducting state in direction z first of all.

The macroscopic superconducting state can be set in at a temperature $T \leq T_c^{**} = \min\{T_{cx}^{**}, T_{cy}^{**}\}$.

The minimum value of the temperatures $T_{\rm ex}^{**}$, $T_{\rm ey}^{**}$ depends on the deformation of the superconducting tape.

In the case of $k < E^3$ the temperature $T_{\rm cx}^{**}$ is the lower one, thus $T_{\rm c}^{**} = T_{\rm cx}^{**}$ and, depending on the concentration, we get

$$c > \frac{1}{8} : T_{c}^{**} > T_{cz}^{*},$$

$$c < \frac{1}{8} : \begin{cases} T_{c}^{**} > T_{cz}^{*} & \text{if } E(kE)^{1/2} < \frac{2 - 2c^{1/3}}{1 - 2c^{1/3}}, \\ T_{c}^{**} < T_{cz}^{*} & \text{if } E(kE)^{1/2} > \frac{2 - 2c^{1/3}}{1 - 2c^{1/3}}. \end{cases}$$

In the case of $k > E^3$ the temperature T_{cy}^{**} is the lower one, thus $T_{c}^{**} = T_{cy}^{**}$ and, depending on the concentration, we get

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$$c < \frac{1}{8} : \begin{cases} T_{c}^{**} > T_{cz}^{*} & \text{if } k < \frac{2 - 2c^{1/3}}{1 - 2c^{1/3}}, \\ T_{c}^{**} < T_{cz}^{*} & \text{if } k > \frac{2 - 2c^{1/3}}{1 - 2c^{1/3}}. \end{cases}$$

Supposing in three dimensions the initial quasi-spherical symmetry for the bulk material consisting of N^3 uniform cells the number of the percolation chains can be N^2 and each chain formed from N uniform cells. As we mentioned previously the probability of the formation of a percolation chain in the K-th case is P_K^{N-1} , thus the number of the percolation chains is

$$N_{\rm p} = N^2 P_{\rm K}^{N-1} \,. \tag{26}$$

Obviously the number of percolation chains does not decrease below N, because that means the lack of the percolation chains, therefore the critical temperature T_{cK}^{**} in the K-th case can be given by

$$N_{p}(T_{cK}^{**}) = N^{2} P_{K}^{N-1}(T_{ck}^{**}) = N = n^{1/2},$$
(27)

where n is the average number of filaments running through the cross-section of the conductor. From this the critical temperatures can be determined using the probabilities (14),

$$P_{K}(T_{cK}^{**}) = N^{1/(N-1)} = n^{1/(2(1-n^{1/2}))}.$$
(28)

At given values of E, k, c, and l_{f0} with decreasing temperatures the in-situ granular superconductor passes through the different possible percolation cases mentioned above.

In the case of $T_{cz}^* > T_{cK}^{**}$ first the uniform cells go into the superconducting state and afterwards the whole composite becomes a superconductor by the further decrease of temperature.

In the case of $T_{cz}^* < T_{cK}^{**}$ the bulk composite and the cells will be superconducting at the same time when the temperature of the cell superconductivity has been reached.

5. Conclusions

On the basis of our uniform cell model the probabilities of the formation of percolation chains can be given in two and three dimensions in an elongated and/or flattened in-situ granular superconducting composite. The critical temperatures of the macroscopic superconducting state can be derived from these probabilities.

With decreasing temperature the different cases of the macroscopic superconductivity are realized and evidently the different macroscopic superconducting states have different superconducting parameters, namely critical magnetic fields and critical currents, too.

The diversities of the transition curves of granular superconductors can be interpreted easily taking into account the diversities of the deformation parameters E, k, the concentration c, and measurements of the superconducting grains.

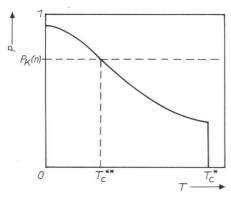


Fig. 3. Determination of the macroscopic critical temperature T_c^{**} from the temperature dependence of the percolation probability $P_K(n)$. T_c^{**} is the microscopic critical temperature

At the parameters given above, in three dimensions the temperature dependence of the probabilities can be calculated and T_c^{**} will be pointed out by $P_K(n)$ as is shown in Fig. 3. The critical temperature of the macroscopic superconductivity is

$$\begin{split} T_{\rm c}^{**} &= 0 \quad \text{if} \quad P_K(n) > P(T=0) \,, \\ T_{\rm c}^{**} &= T_{\rm cz}^{*} \quad \text{if} \quad P_K(n) = P(T_{\rm cz}^{*}) \,, \\ 0 &\leq T_{\rm c}^{**} \leq T_{\rm cz}^{*} \quad \text{if} \quad P(T=0) \leq P_K(n) \leq P(T_{\rm cz}^{*}) \,, \end{split}$$

where T_{cz}^* is the critical temperature of the microscopic superconductivity.

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