Reentrant superconductivity mechanisms in amorphous carbon-silicon films containing tungsten

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(Received 25 February 2000; revised manuscript received 5 June 2000)

A model—based on Josephson junctions array and Coulomb blockage—is presented that is capable to explain reentrant resistive peaks in diamondlike carbon-silicon films containing tungsten. Such systems show a main transition together with one or two reentrant or quasireentrant ones: computer simulations for one reentrant resistive peak have been performed that confirm with clear evidence the experimental data.

I. INTRODUCTION

Reentrant or quasireentrant superconductivity phenomena in disordered metals have been the object of experimental and theoretical research for many years.[1] The interest shown for these phenomena is due to their fundamental importance in highlighting new aspects of the quantistic nature of electrons in solids. The reentrant superconductivity (RS) has been observed in magnetic superconductors of the type $HoMo_6S_8$ and $ErRh_4B_4$,[2] and it appears as a superconductor-dielectric transition at a certain temperature $T_r < T_c$, where T_c is the critical temperature of the superconductor. For $T < T_r$ the system remains in the normal state down to $T = 0$ K. The superconducting transition at $T = T_c$ may be complete or incomplete. In the latter case (the so-called ''quasireentrant phenomenon''), it consists of a resistance minimum of varying depth at $T < T_c$. This phenomenon—indicated as the ''traditional'' one in the following—has been often observed in nonhomogeneous metallic films in which the growth is of Volmer-Weber type or in granular metals.[1] Granular metals consist of superconducting metallic grains—approximately the same dimension—embedded in a dielectric matrix. At the present time, the RS and quasi-RS phenomena in magnetic superconductors or traditional ones are studied in great detail and their microscopic mechanisms have been clearly understood.[2] In traditional granular superconductors, the transition to the dielectric state at $T < T_c$ is due to the Coulomb interaction between Cooper pairs, described by a charging energy E_c. This explanation has been proposed by Abeles[3] and it is known as the ''Coulombian blockage.''

During the last ten years a different kind of reentrant superconductivity phenomenon has been revealed, observed in amorphous carbon-silicon films containing tungsten, grown on a dielectric substrate by the classical plasma decomposition method.[4,5] In more detail, after having become superconductors, some samples show a small but clear increase in resistance when going down in temperature, followed by a return of the resistance to the former nearly zero value. The result is one or more resistive peaks in a range of temperatures that should correspond to the superconducting state. It looks like an attempted superconducting-dielectric transition in which the system, far from becoming an insulator and remaining in this state for lower temperatures, soon becomes superconducting again. The present paper is devoted to the qualitative analysis of the possible microscopic mechanisms for the reentrant superconductivity phenomenon in its peak manifestation. The proposed mechanisms have been used as a base of the computer simulation in the case of one or two peaks.

II. EXPERIMENT

Films were grown on dielectric substrates by plasma decomposition of siloxane vapors in a dc diode reactor.[4,5] At the same time, during film growth, W atoms were deposited by using magnetron sputtering in order to vary the electrical conductivity from the insulating to metallic behavior. W concentrations were ranging from 1–2 at. % (corresponding to a resistivity of 10^{14}–10^{16} Ω cm) to 50–60 at. % (corresponding to 2×10^{-4} to 4×10^{-4} Ω cm).[6] For the samples of interest that show reentrant peaks, resistance measurements were based on the four-probes dc technique, with a current of 0.1 mA. Electrical contacts were prepared from silver paint. Magnetic susceptibility measurements were performed with an ac susceptometer, based on the mutual inductance technique. The source field used for measurements can be set at two different frequencies, 80 Hz and 2.5 kHz. Data can be collected both on warming and on cooling from 1.4 K to room temperature.

III. RESULTS

In these films the Anderson dielectric-metal transition (DMT) is obtained for relative tungsten concentrations $x_c \simeq 0.25$–0.30.[5] The metallic phase for $x > x_c$ presents a granular system in which, according to measurements by electron microscope, the grains dimensions are distributed between 100 and 1000 Å . The Poole-Frenkel effect and hopping conductivity measurements at the dielectric phase for $x < x_c$ show that the matrix is characterized by a dielectric constant $\kappa = 19$.[6]

On the metallic side of DMT ($x > x_c$), the superconducting transition is observed at a temperature T_c varying from 2 to 4 K, depending on the metal concentration.[6] Although large, the main superconducting transitions are sharp and show no evidence of different superconducting phases. Reentrant transitions of the new type in carbon-silicon films containing tungsten manifest themselves as one or two normal-state ''peaks'' at $T < T_c$ (Figs. 1 and 2) (Figs. 9 and 10 in Ref. 6) and a steady superconducting state at low

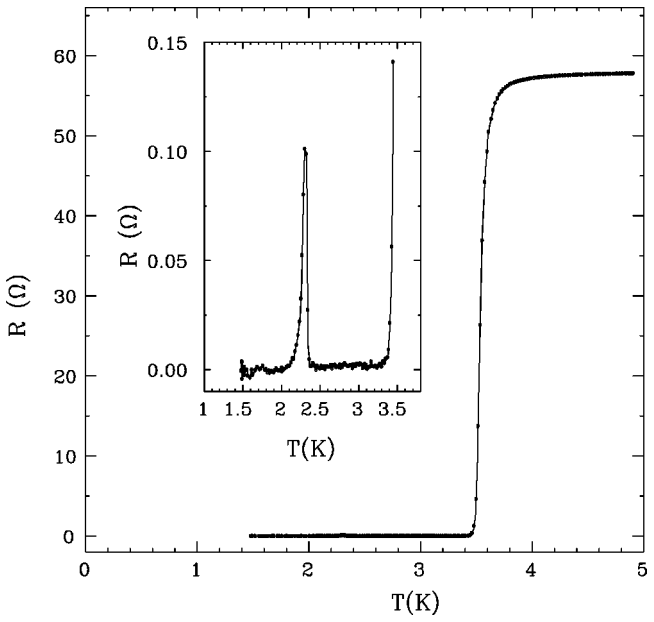

FIG. 1. Superconducting resistive transition with one reentrant peak in diamondlike carbon-silicon films containing tungsten ($x > x_c$) (Ref. 7).

temperatures—at least down to $T=16$ mK. Peaklike reentrant transitions have been studied in detail by our group[6,7] and we established:

1. Reentrant peaks group by a single or two narrow ones—$\simeq 0.1$ K wide—and in all samples peaks are observed at $T>1$ K.

2. Peaks are associated to the quasireentrant superconducting phenomenon, i.e., the main superconducting transition is often incomplete, a nonzero resistance appearing in the temperature regions bounded by the main transition and the peaks themselves.

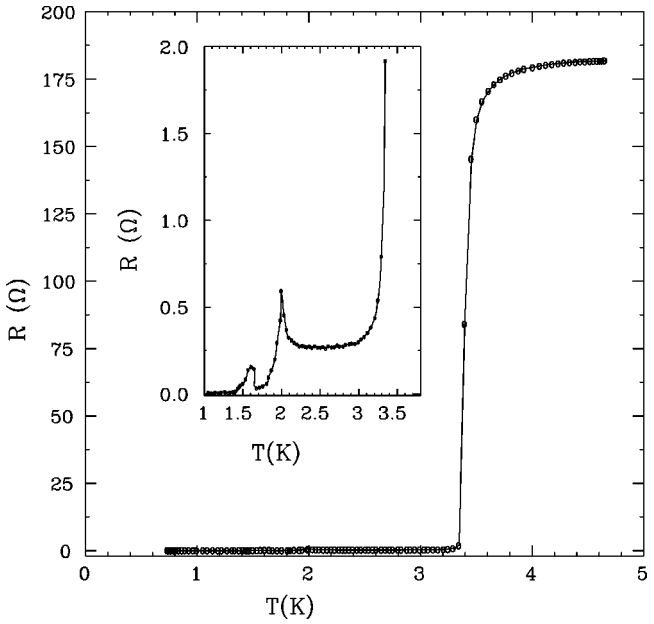

FIG. 2. Quasireentrant resistive transition with two reentrant peaks in diamondlike carbon-silicon films containing tungsten ($x > x_c$) (Ref. 7).

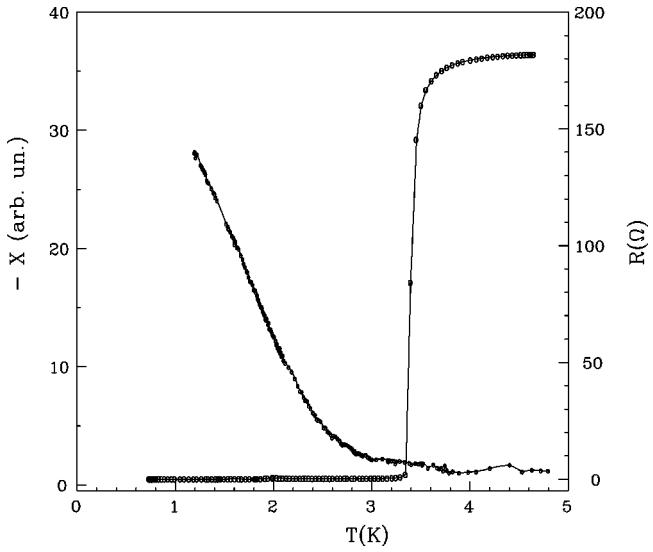

FIG. 3. Typical main superconducting transition in resistance R and in magnetic susceptibility χ in carbon-silicon films containing tungsten for $x > x_c$.

3. The height of each peak is very small when compared to the main transition one: it is of the order of $10^{-3}R_n$, R_n being the normal resistance immediately over T_c.

4. Reentrant peaks are not observed in magnetic susceptibility measurements carried out using a resolution $\Delta\chi/\chi \simeq 10^{-6}$.

5. The superconducting main transition in susceptibility χ is shifted by ~ 1.5 K towards zero with respect to the main transition in resistance R (Fig. 3).

6. A strong non-ohmic behavior of the system is observed in proximity of the peaks.

7. The critical magnetic field for the reentrant peaks is always less than the main superconducting transition one.

IV. DISCUSSION

In principle, the superconducting phase in the aforementioned films may be constituted by any type of superconductor:

(i) experiments on amorphous thin W films[8] have shown the bulk T_c [12 mK (Ref. 9)] can be increased of two orders of magnitude up to 2 or 3 K. It has been suggested[9] that this enhancement may be due to the presence of tungsten oxide having T_c close to 3 K. Another explanation of the enhanced superconductivity in the amorphous W films might be derived from Ginzburg's surface-state superconductivity;[10]

(ii) tungsten-carbon compounds W_2C and W_xC_{1-x} for $0.45<x<0.5$ are superconductors showing a T_c in the range 2–10 K;[11]

(iii) tungsten-silicon compound $W_{0.6}Si_{0.4}$ is also a superconductor with a $T_c=2.8$ K.[8]

The superconducting inclusions are most likely of the (ii) type since carbides have been detected that lead to critical temperatures of the main transitions as high as 3.9 K for certain W concentration. In order to explain the reentrant peaks phenomenon, the concepts to which we shall refer are the same used for granular metals, in which the superconducting to dielectric transition is decided by two relevant energies. The Coulombian blockage is due to the character-

istic energy $E_c^{i,j}$, expressed by the capacitance matrix C_{ij} describing the electrostatic coupling between Cooper pairs on grains i and j:

$$E_c^{i,j}=\frac{1}{2}\frac{Q_iQ_j}{C_{ij}}, \tag{1}$$

where Q_i, Q_j are, respectively, the charge excess due to Cooper pairs (charge $2e$) in the grains i and j. Diagonal elements of the matrix C_{ij}^{-1} provide the charging energy $(2e)^2/2C_{ii}$, where C_{ii} is the self-capacitance of the grain i. Off-diagonal elements with $i=j\pm1$ describe the charging energy for the nearest-neighbors interaction.

In granular metals, the grains are weakly coupled by tunnel junctions with characteristic energy:

$$E_J(T)=\frac{\pi\hbar}{4e^2}\frac{1}{R_n}\Delta(T)\tanh\left(\frac{\Delta(T)}{2k_BT}\right), \tag{2}$$

where $\Delta(T)\simeq\Delta(0)(1-T/T_c)^{1/2}$ is the superconducting gap and R_n is the resistance for single-electron tunneling between neighboring grains. This means that a granular metal can be seen as a disordered Josephson junction array (JJA). The behavior of this system at low temperatures is governed by the competition between the two above-mentioned energies E_c and E_J. In the limit $E_c>E_J$, the Coulombian blockage pins Cooper pairs to the grains and at low temperatures ($T<E_c/K_B$) the array is insulating. In the opposite limit $E_J>E_c$, the Josephson coupling between the grains allows the transport of Cooper pairs and the array is superconducting. The theoretical analysis of the superconducting-dielectric transition in JJA has been made for a periodic system, in which the metallic grains are placed at the vertices of a crystal lattice.[12–14] The phase diagram showing the transition from superconducting to dielectric state has been mapped in the plane $k_BT_c/E_J-E_c/E_J$, either if the main contribution to Coulomb blockage is given by the self-capacitance C_{ii},[12,13] or by intergranular capacitance $C_{ij}(i\neq j)$.[14] The analysis developed in Refs. 12 and 13 is able to describe the main features of the superconducting-dielectric transition that has been observed in granular metals like Al, Bi, In, etc.[1] However, these models do not predict a peak existence: once the reentrance to the dielectric state appears, the system remains in this state until low temperatures.

In recent years, a certain interest has been oriented toward two-dimensional periodical networks artificially fabricated by planar technology (see Refs. 15 and 16 and references therein). These systems are ordered ones and clearly they can be better described by the theory[12–14] for a periodic JJA. A control parameter that can be used to induce phase transition in JJA is the magnetic field. In this case, the transition dynamics is determined by vortices motion, whereas at the dielectric side of DMT dynamics is determined by $2e$ charge excitation. It is important to pay attention to the fact that, in periodical networks, the superconductor to dielectric transition has a macroscopical character. On the contrary, for us it is worthwhile to pay attention to the small height of each peak ($10^{-3}R_n$), showing that changes in the metallic system occur at the mesoscopic level only, with relatively few grains involved.

In order to build a qualitative model explaining the peak existence, it is important to remark that superconducting susceptibility transition is shifted about 1.5 K towards lower temperatures with respect to the one in resistivity (Fig. 3). This fact constitutes the physical base for using the percolative model to explain the superconducting transition in a granular system carbon-silicon-tungsten. The transition shift in R in respect to the one in χ indicates that a superconducting path linking sample electrodes appears at higher temperatures with respect to the diamagnetic transition. In order to obtain a superconducting shunt in a percolative system, it is enough to have a path consisting of superconducting connected grains; it can have a low dimensionality, being practically quasiunidimensional. On the contrary, to obtain the diamagnetic transition, most of the grains must be organized in big superconducting clusters. To break the superconducting path across the sample it suffices to restore the normal state in a small number of grains, thus cutting the superconducting links on a mesoscopic level; the resulting variation in the volume of the superconducting phase produces no sensible change in our measurements of susceptibility. So as to build a qualitative physical picture, consider what happens in a granular superconductor for temperatures lower than T_c. Suppose the grains consist of a material becoming a superconductor at the bulk transition temperature T_{co}. Let us analyze the superconducting state appearance when going down in temperature, starting from $T\lesssim T_{co}$. The complex order parameter ψ can be written in the form $\psi=\Delta e^{-i\phi}$, where Δ represents the amplitude and ϕ is the phase. It is well known that disorder can destroy the superconducting state by decreasing the amplitude Δ and by destroying the phase coherence between superconducting electrons. The system under examination is on the metallic side of DMT, so it is nonhomogeneous since the critical cluster linear size $L(x)$ near the percolation threshold is much bigger than the coherence length ξ. For similar systems, the effective temperature T_c of the superconducting transition is less than T_{co} for the coherence phase suppression between weakly linked superconducting clusters of metallic grains.[1] In Refs. 17–19, the strongly disordered ($\gamma\sim1$) superconductors behavior has been studied near the percolation threshold, in two and in three dimensions, on the base of BCS models. The parameter that characterizes the disorder is $\gamma=(k_Fl)^{-1}$, k_F being the Fermi wave vector and l indicating the mean free path for the elastic scattering. It has been shown that the thermodynamic fluctuations near T_c become stronger with increasing disorder.[20,21] This explains the decrease in T_c with respect to T_{co}.

For the three-dimensional case

$$\frac{T_c}{T_{co}}=1-\frac{\sigma\min}{\sigma}\frac{k_BT_{co}}{\varepsilon_F}, \tag{3}$$

where σ_{min} is the Mott's minimum conductivity in a metal, σ is the normal conductivity just over the superconducting transition temperature. For a dielectric thickness S_{ij} between two neighboring grains of about 10–20 Å, electrons are able to tunnel from one grain to a neighboring one. This effect defines all the system properties. In a granular system, a superconducting transition occurs in two steps. First, when the system reaches the temperature $T=T_{co}$, the supercon-

ducting gap appears in each grain, but the order parameter phases of different grains are not correlated. Then, at a lower temperature $T=T_c<T_{co}$, the tunneling of Cooper pairs establishes long-range phase coherence and all the system undergoes the electronic phase transition to the superconducting state. This transition consists in the formation of a big superconducting cluster or even a macroscopic one (infinite cluster), partially or completely shunting the sample. It is worth paying attention to the fact that this explanation for the transition into the dielectric state does not take into account the screening effect between charged grains due to single electrons, whose number is proportional to the factor $\exp[-\Delta(T)/k_BT]$ and is high in the temperature range near T_c. The screening effect strongly decreases the effective energy of the electrostatic interaction between the grains. When going down in temperature, the single electrons number decreases according to an exponential law so that at $T\sim\frac{1}{2}T_c$ the screening effect becomes practically negligible. In this case, at the temperature

$$T_b\simeq\frac{E_c}{k_B} \tag{4}$$

if $E_c>zE_J$, where z is the nearest neighbors average number, at least a few grains may undergo the transition into the dielectric state, destroying the macroscopic superconducting cluster that shunts the sample. For temperatures lower than Eq. (4), the system behavior is governed by the concurrence of the Josephson coupling energy $E_J(T)$ that gets higher and the Coulombian energy E_c, whose effective value becomes practically constant. As a result, the dielectric state established in any cluster at a temperature T_b is removed at a temperature T_{ub} defined by the following equality:

$$zE_J(T_{ub})=E_c, \tag{5}$$

where z should be the number of the nearest neighbors. More correctly, the unblockage condition in a regular grains lattice is given by[1,14]

$$zE_J(T_{ub})=E_c+k_BT. \tag{6}$$

Nevertheless, for temperatures close to T_{ub} the following condition, $k_BT\ll zE_J,E_c$, holds so that the thermal energy k_BT can be neglected. This is especially true for disordered systems for which the z value is unknown and may be treated as a free parameter. The equality (5) holds only if $zE_J(0)>E_c$. Otherwise, the system remains blocked in the dielectric state down to $T=0$ K. According to our model, the reentrant peak is the manifestation of the normal state for only a few clusters in the small temperature range:

$$T_{ub}<T<T_b \tag{7}$$

The small peak height $\sim10^{-3}R_n$ reveals the dielectric state does not appear in the whole system, but only in a few clusters involving a relative number of grains not bigger than $\sim10^{-3}$. Experimental data obtained for different samples of carbon-silicon films with W show three different situations in the temperature range $T<T_c$: reentrant phenomenon with one peak, the same with two peaks or the absence of any peak.

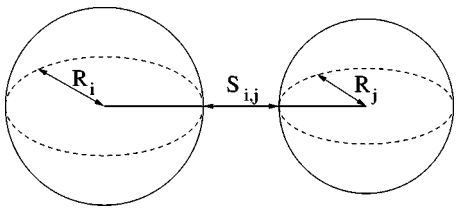

FIG. 4. Two neighboring grains in the dielectric medium separated by the distance $S_{i,j}$.

To prove that the proposed mechanisms are responsible for the reentrant superconductivity with ''peaks,'' we have performed a numerical simulation for the case of one peak. To evaluate the parameters necessary for numerical simulation, we used the experimental temperature values $T_r=2.36$ K and $T_{ub}=2.16$ K obtained for a typical sample chosen at random among the ones showing one peak, where T_r and T_{ub} correspond, respectively, to the beginning and the end of the peak when going down in temperature. To simulate the system behavior, a numerical sample has been generated consisting of nonoverlapping spherical grains, with radii and intergranular distances Gaussianly distributed around the respective mean values. Then we devised an algorithm (to be published elsewhere) to check the presence or the absence—in each temperature interval—of at least one superconducting path between the sample electrodes. Consider two grains i, j (Fig. 4), R_i and R_j represent the respective radii and $S_{i,j}=d_{i,j}-(R_i+R_j)$ the intergranular dielectric thickness. The relations used to compute the Coulombian energy $E_c^{i,j}$ and the Josephson one $E_J^{i,j}$ are Eqs. (1) and (2); the capacitance of two neighboring grains has been computed in planar approximation:[3]

$$C_{i,j}=\frac{\alpha\pi\kappa(R_{min}^{i,j})^2}{S_{i,j}}, \tag{8}$$

where $R_{min}^{i,j}$ indicates the minimum radius among the ones of two neighboring grains, $\kappa=19$ is the dielectric constant, $\alpha=0.3$–0.5 is the correcting factor that takes into account the fact the capacitors surface is not flat.[3] For we are interested in electrons tunneling between nearest-neighboring grains, we considered only the case $j=i\pm1$, with the capacitance represented by a flat capacitor whose plates are the grains. Having chosen the mean values for grains radii $\overline{R^i}=200$ Å on the base of experimental data and for nearest-neighboring grains distance $\overline{s_{i,j}}\lesssim10$ Å, the virtual sample has been generated and the Coulombian energy $E_c^{i,j}=Q^2/2C_{i,j}$ is practically known for each grains pair. In our numerical simulation, the effective Coulombian energy E_c at $T\lesssim\frac{1}{2}T_c$ turns out to be practically temperature independent, the screening effect of the grains being negligible. On the contrary, the Josephson energy $E_J^{i,j}$ depends from temperature and its value is given by Eq. (2), where $\Delta(T)=\Delta(0)(1-T/T_c)^{1/2}$. To compute $\Delta(0)$, we used the BCS relation

$$2\Delta(0)=3.53k_BT_c. \tag{9}$$

Then, in the Josephson energy expression there remains only one unknown quantity, namely the resistance $R_n^{i,j}$ for single electrons tunneling between neighboring grains. $R_n^{i,j}$ must be

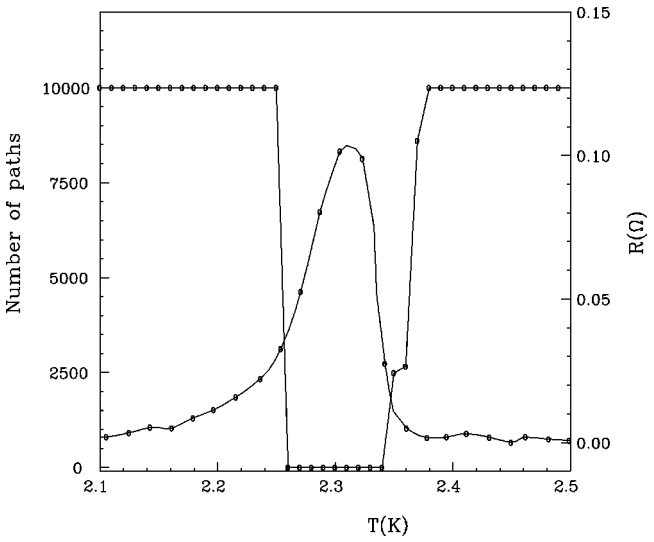

FIG. 5. Percolation paths number vs temperature obtained from computer simulation. An upper cutoff of 10 000 paths has been used. Comparison with the experimental reentrant resistive peak shows good agreement.

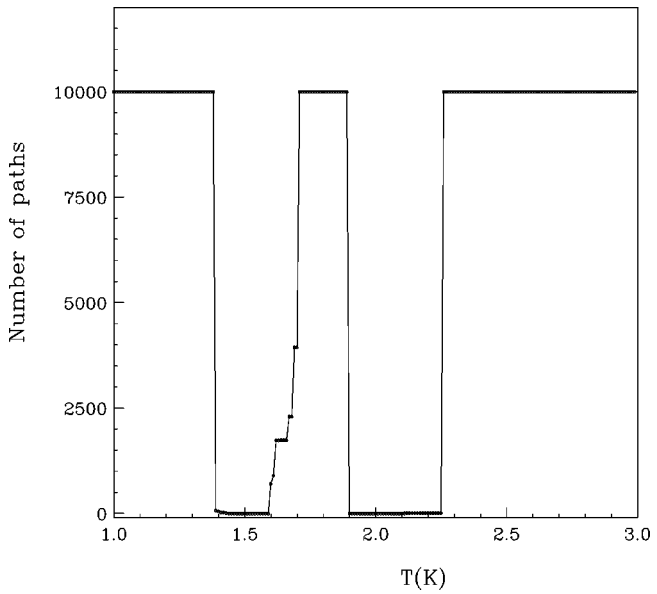

FIG. 6. Percolation paths number vs temperature obtained from computer simulation for a system containing two different grains distributions.

surely dependent from grains radii and distances. For this quantity we have chosen the following form:

$$R_n^{i,j} = A \frac{1}{\alpha \pi (R_{min}^{i,j})^2} \exp\left(\frac{S_{i,j}}{\delta}\right), \tag{10}$$

where A is a constant and δ is a characteristic exponent for tunneling barrier that may be defined using Eq. (7) considering $\overline{S_{i,j}} = \delta$. The constant A in Eq. (9) has been evaluated by using the mean values for grains radii, the mean distance $\overline{S_{i,j}}$, and the experimental result for the unblockage temperature T_{ub}. In our numerical example the mean value for $\overline{R_n}$ is equal to 15.9 kΩ. The obtained value for $\overline{R_n}$ demonstrates the grains system is on the metallic side for DMT: in disordered systems of grains DMT corresponds to the value $\overline{R_n} \sim \hbar/e^2 \sim 25.8$ kΩ.[1] Once all the relevant energy parameters are known, for each value of temperature the algorithm can search for the number of superconducting paths linking one electrode to the other. The algorithm compares the Josephson energy with the Coulombian one. In conclusion, Cooper pairs tunneling between nearest-neighboring grains can be allowed only if Josephson energy E_J is greater than the Coulombian ''gap.'' If such condition holds, we say two nearest-neighboring grains are connected. If not, we say grains are not linked. For each temperature, the algorithm counts the number of macroscopic paths crossing the whole sample and linking one electrode to the other. For we are interested only in determining the eventual temperature interval in which there is not any superconducting path crossing the whole sample, an upper limit on paths counting has been set and fixed on 10 000 paths. Varying the temperature, our simulation showed the inexistence of any superconducting path between the electrodes for T varying from 2.25 to 2.35 K, nicely corresponding to the experimental range ($T_{ub} = 2.16$ K and $T_r = 2.36$ K) (Fig. 5). A similar numerical simulation can be developed for the case of two reentrant peaks (Fig. 2). Following the same approach, the presence of two peaks may be due to the existence of two different kind of grains, composed for instance by W_2C and W_xC_{1-x} for $0.45 < x < 0.5$ or by $W_{0.6}Si_{0.4}$ and W_2C having slightly different dimensions. In fact, different sizes yield different blockage temperatures. The numerical analysis varying simulation parameters shows that in order to obtain two close reentrant peaks it is enough to allow the overlap of the radii distribution for the two grains kinds. To run the numerical simulation, we have employed the experimental data turned out for a typical sample among ones showing two peaks. The characteristic temperatures for such sample were

$$T_b^I = 2.1 \text{ K}; \quad T_{ub}^I = 1.9 \text{ K}; \quad T_b^{II} = 1.65 \text{ K}; \quad T_{ub}^{II} = 1.45 \text{ K}, \tag{11}$$

where the apices I and II select the peak among the existing two. These data allow us to determine the average value of the radii distribution, which are given by

$$\overline{R^1} = 220.1 \text{ Å } \overline{R^2} = 248.3 \text{ Å}, \tag{12}$$

where apices 1 and 2 refer to the grains kind. The searching for superconducting paths across the whole sample is the same applied to the case of one peak: the dielectric state unblockage is governed by the equality (5) corresponding to the particular grain kind. The simulation results show the absence of superconducting paths in two temperature ranges, 1.4–1.5 K and 1.9–2.1 K in good agreement with experimental data (Fig. 6).

V. CONCLUSIONS

The agreement between simulation results and the experimental values allows us to suppose that the mechanisms explaining the reentrant phenomenon with one or two peaks are connected with the Coulombian blockage, due to the intergranular capacitance of two nearest-neighbors grains, and the unblocking process indicated by the equality (5). The micro-

scopic mechanisms responsible for the reentrant peak can be tested in an independent way by the numerical simulation just developed and by the evaluation of the critical current for each superconducting path, consisting of a chain of grains mutually connected, to obtain the global critical current. The algorithm we developed may in fact be extended to find out not solely peaks position in the temperature scale, but also the temperature dependence of resistance for each peak, their current-voltage characteristics, and finally the temperature dependence of their critical magnetic field. These aspects will be the object of future investigations on this interesting reentrant phenomenon.

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