Percolation in Superconductive Networks

J. Simonin and A. López
Centro Atómico Bariloche, Comisión Nacional de Energía Atómica, CP 8400 Bariloche R.N., Argentina
(Received 4 February 1986)

The percolation properties and upper critical field of a disordered superconductive square network are calculated by use of the de Gennes–Alexander theory. It is suggested that photolithographic techniques be used to build such artificially disordered systems. The critical exponent of the upper critical field is calculated and found to be $k=0.93\pm0.06$ on the metallic side of the percolative transition.

PACS numbers: 74.40.+k, 74.60.Ec

Since the pioneering papers of de Gennes[1,2] and Alexander[3,4] on inhomogeneous superconductors, a large amount of both theoretical and experimental work has been done in the field of superconductive micronetworks. Application of the model to the case of regular networks of thin wires gave quite important and interesting results.[5–9] In particular, in the case of large (infinite) regular networks, theory and experiment complemented each other nicely, allowing for the first experimental access to the amusing properties of the spectrum of these systems. As is known[4–7] the (H,T) phase diagram is the envelope of the energy spectrum of an electron in the same geometry. The disordered systems which gave rise to the original ideas of de Gennes and Alexander were inhomogeneous superconductors prepared by coevaporation of In and Ge on a glass substrate.[4] Present-day technology allows for the manufacture of disordered samples of a different kind. The photolithographic samples of Ref. 8 could be artificially disordered either by laser irradiation of an originally regular lattice or by computer design of the mask used in the process.[10] Although this is not the original problem of inhomogeneous superconductors, it is another example in which theory and experiment could go hand in hand, since the de Gennes–Alexander approach can be applied to a model system of exactly the same characteristics as that prepared in the laboratory.

In this Letter we study the (H,T) phase diagram of a square network when nodes are removed at random (site percolation). The model[2] assumes that the cross section of the wires (or critical temperature T_{cw}) is small enough so that there is no magnetic response due to the wires themselves; magnetic properties depend on the topology of the network and on whether the superconductive state extends over all the system.

We write the Ginzburg-Landau free energy[2–5] (up to second order in the order parameter) as a sum over links of the network:

$$F=sE_c\sum_{ab}\int_a^b dl\{-|\psi|^2+\xi^2|[i\,d/dl+(2\pi/\phi_0)A(l)]\psi|^2\}, \tag{1}$$

where s is the cross section of the wires, E_c [$\sim(1-t)^2$, with $t=T/T_{cw}$ the reduced temperature] the condensation energy per unit volume, ξ [$=\xi_0/(1-t)^{1/2}$] the temperature-dependent coherence length, ψ the complex order parameter normalized by its zero-field value, and l the coordinate along the link ab. $A(l)$ is the projection of the vector potential along the wire and ϕ_0 ($=ch/2e$) the flux quantum. The linearized Ginzburg-Landau equation[5] gives the order parameter in the strand as a function of its values at the nodes:

$$\psi(l)=[\exp(i\gamma_{al})/\sin(\mathscr{L}_{ab})][\psi_a\sin(\mathscr{L}_{ab}-l)+\psi_b\exp(-i\gamma_{ab})\sin(l)], \tag{2}$$

where $\mathscr{L}_{ab}$ is the length of the strand normalized by ξ and γ_{al} is $2\pi/\phi_0$ times the circulation of the vector potential along the link.

Equation (2) allows us to write the free energy as a function of the values of the order parameter at the nodes:

$$F=sE_c\sum_{ab}\left\{\frac{\xi}{\sin(\mathscr{L}_{ab})}\{(\psi_a\psi_a^*+\psi_b\psi_b^*)\cos(\mathscr{L}_{ab})-[\psi_a\psi_b^*\exp(i\gamma_{ab})+\psi_a^*\psi_b\exp(-i\gamma_{ab})]\}\right\}. \tag{3}$$

The free energy is thus isomorphic to an electronic tight-binding Hamiltonian and can be diagonalized by solving the corresponding system of linear equations:

$$sE_c\sum_z\{[\xi/\sin(\mathscr{L}_{az})][\psi_{an}\cos(\mathscr{L}_{az})-\psi_{zn}\exp(-i\gamma_{az})]\}=f_n\psi_{an}, \tag{4}$$

where n stands for the eigenstate and the sum is over the nearest neighbors (z) of a given node (a). The thermal probability ρ_n takes the usual form:

$$\rho_n=\exp(-F_n/k_BT)/Z;\quad Z=\sum_n\exp(-F_n/k_BT). \tag{5}$$

The (H,T) phase diagram within the mean-field solution follows from looking at the point at which the lowest eigenvalue becomes zero.[2-7] Some characteristics of the weak-link arrays deduced from Eqs. (3) and (4) are as follows:

(1) The mean-field solution at zero field always gives $T_c - T_{cw}$ and corresponds to a constant order parameter throughout the network ($\psi_a = \psi_b = \ldots = 1$) independent of the kind of disorder introduced. This should imply that the classical percolation characteristics of the network determine the magnetic response, as assumed in earlier works,[1,4,11] but a more complete calculation, with the corresponding thermal probabilities instead of the mean-field solution, must consider the nature of the excited states.

(2) The dead-end links, i.e., those not belonging to a closed loop, are not affected by the field since the exponential factor in Eqs. (3) and (4) can be absorbed by a change of gauge, and thus tend to push the field-dependent critical temperature [$T_c(H)$] towards T_{cw}, its value at zero field.[2,5]

(3) In agreement with (1) these systems do not have two characteristic temperatures at zero field, one for the superconductive transition of the nodes and a lower one for the onset of a coherent state, as occurs for arrays of superconductive grains coupled by Josephson junction.[12] In our weak-link arrays these transitions happen at once. This property becomes apparent if we rewrite the free energy, Eq. (3), as

$$F = s\sum_{ab}[E_c\xi\cot(\mathscr{L}_{ab})(|\psi_a|^2+|\psi_b|^2) - J^c_{ab}|\psi_a||\psi_b|\cos(\beta-\alpha-\gamma_{ab})], \tag{6}$$

where α (β) is the phase of the order parameter at node a (b) and $j^c_{ab} = 2E_c\xi/\sin(\mathscr{L}_{ab})$ plays the role of critical current for the links, whose supercurrent is given by

$$J_{ab} = J^c_{ab}|\psi_a||\psi_b|\sin(\beta-\alpha-\gamma_{ab}). \tag{7}$$

Equation (6) explicitly shows the main difference between these systems and Josephson-coupled superconductive grains: The nodes are not massive and thus the Josephson part of the free energy of the links [second term on the right-hand side of Eq. (6)] provides all the condensation energy; the nodes cannot become superconducting by themselves.

Equations (3) and (4) are, in general, difficult to solve. They become more tractable for simple geometries, like the lasso[2] and the boleadora,[5] and for regular networks, like the single ladder[6] and the square net.[7] These regular networks all have links of equal length ($L_{ab} = L$) and a well defined elementary loop of enclosed area L^2. Thus, because of the special magnetic field dependence of the free energy [Eqs. (3) and (4)], the (H,T) phase diagram is periodic with period H_0 such that $L^2H_0 = \phi_0$.

For the square lattice Eq. (4) reads

$$[sE_c\xi/\sin(\mathscr{L})][z_a\psi_{an}\cos(\mathscr{L}) - \textstyle\sum_z \psi_{zn}\exp(-i\gamma_{az})] = f_n\psi_{an}, \tag{8}$$

where the sum is over the nearest neighbors of node a, whose number is z_a. We introduce disorder by removing a fraction q of the nodes at random, i.e., a fraction $p = 1-q$ of the nodes are present. A link ab is present only if both nodes, a and b, are present.

At $p = 1$ we have $z_a = 4$ and Eqs. (8) are isomorphic to the tight-binding equations of an electronic system in the same geometry and twice the applied field. The energy spectrum for this problem has a complex magnetic field dependence and was first computed by Hofstadter.[13] The phase diagram of the superconductive square lattice (at $p = 1$) is straightforward to obtain[7] from it. For rational values of H/H_0, the eigenstates are extended. For irrational H/H_0, Eq. (8) corresponds to a critical point in the parameter space of the equivalent 1D incommensurate Hamiltonian studied by Aubry.[14] This point separates extended from localized states, and Aubry conjectured that for it the states are power-law localized.[7,14]

When $1 > p > p_c$ (p_c = classical percolation threshold = 0.59) we are on the "metallic" side of the percolative transition: There is an infinite cluster in the sample. The lowest free-energy eigenstate is still extended (at $H = 0$) and the peridocity of the phase diagram in H is conserved as a result of the fact that higher-order loops are integer multiples of the elementary loop. We analyze two aspects of the (H,T,p) phase diagram: (a) the (H,T) phase boundary as a function of $p > p_c$; and (b) the nature of the excited states at $H = 0$.

To do this we run a tridiagonalization procedure[15,16] over the free energy obtaining effective one-dimensional free energy. We analyze it by means of numerical diagonalization, continued fractions, and decimation techniques.[7] The "numerical" samples are lattices of 100×100 sites and the tridiagonalization is followed through 512 steps. We study the response of the "infinite" cluster. Finite clusters are not taken into account since they play no role in the coherent superconductive transition. We find the following:

(a) The (H,T,p) phase boundary looking at the mean-field solution. The lowest eigenvector is analyzed by the continued-fraction method and its localization length obtained. This localization length can be interpreted as equivalent to the Ginzburg-Landau

coherence length in a continuous medium, which appears as the size of the nucleation regions. In the present case the dead ends act as nucleation centers.

Figure 1 shows the phase boundary for one sample at three different concentrations ($p=1$, 0.9, and 0.7). On the vertical axis we plot $\mathscr{L}^2=(L/\xi_0)^2(1-t)$ and the abscissa is L^2H/ϕ_0. When the disorder increases the field-dependent critical temperature tends to T_{cw} and the localization length decreases. The latter also decreases when the applied field is increased towards $H_0/2$. These characteristics correspond to the effects of the dead-end links and higher-order loops that appear with increasing disorder.

For $H/H_0<0.05$ the nucleation field (H_{c2}) depends linearly on $1-t$, as is usual for massive superconductors; beyond $H=0.1H_0$ the structure of the array produces a departure from the linear law.

The derivative of H_{c2} with respect to T increases with decreasing p, as can be seen in Fig. 1, and diverges for $p=p_c$. This has been theoretically analyzed by means of scaling arguments[4,11] and it was proposed that $dH_{c2}/dT\sim(p-p_c)^{-k}$, where the critical exponent k is a function of known critical indices. According to Ref. 11 its value is $k=0.57$, whereas Ref. 4 predicts $k=0.87$. It should be noticed that H_{c2} has different behavior according to whether p is larger or smaller than p_c. As discussed above dH_{c2}/dT at T_{cw} is well defined for $p>p_c$, whereas for $p<p_c$ one has $H_{c2}\sim(1-t)^{1/2}$ so that k as defined loses sense. In that case one defines the critical index through $H_{c2}\sim(1-t)^{1/2}(p_c-p)^{-k}$, where k is now related to the behavior of the diamagnetic susceptibility χ of the clusters. This follows from the relation $H_{c2}\sim(1-t)/\chi^{1/2}$, valid only for finite systems.[11] In this region ($p<p_c$, region I of Ref. 11) there are numerical experiments by Bowman and Stroud[17] using an XY model which are in rough agreement with the scaling predictions of Ref. 11. Although these authors use a different model, one expects the critical indices to be universal. The constant-amplitude approximation, which leads in the present systems to an XY model, is appropriate to study finite systems for which the nucleation has been found to be uniform for small fields,[7,11] but not for large samples where vortex penetration is present at any field. In the same spirit we should also mention the results of John and Lubensky[18] for granular superconductors.

We have fitted the linear expression

$$H_{c2}L^2/\phi_0=A(L/\xi_0)^2(1-t) \tag{9}$$

to the initial part of the phase diagram, obtaining A as a function of p for $p>p_c$. From this we calculate the critical exponent

$$dH_{c2}/dT-A\phi_0/\xi_0^2T_{cw}=B(p-p_c)^{-k}. \tag{10}$$

A log-log plot of A vs $p-p_c$ is shown in Fig. 2 for one of the samples. Repeating this for ten samples we obtain an average critical index $k=0.93\pm0.06$, which is in rough agreement with both the predictions of Deutscher, Grave, and Alexander[4,11] and of John and Lubensky.[18] Experimental data, also from Deutscher, Grave, and Alexander,[4] give $k=0.6\pm0.05$ for inhomogeneous InGe films. This discordance could be due to the three-dimensional nature of the InGe samples. New experiments, using microlithography techniques, should provide an experimental test of the theory. We want to stress once more the importance that these experiments could have, since they would provide a controlled approach to disorder in quantum systems.

(b) Equation (8) could be written in the following

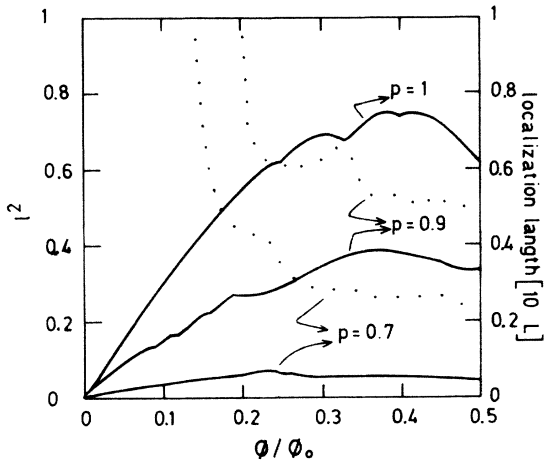

FIG. 1. Critical field (full line) and localization length (or range of nucleation) (dotted line) as a function of $\mathscr{L}^2$ for the disordered square lattice at concentrations $p=1.0$, 0.9, and 0.7.

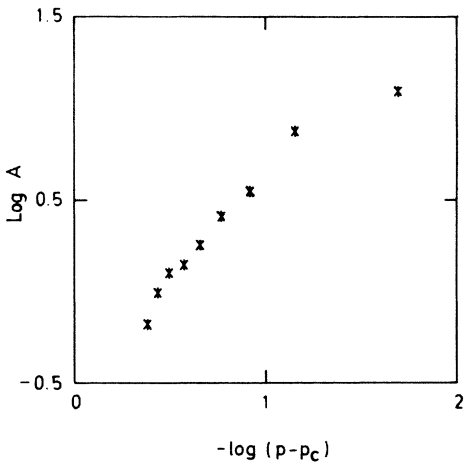

FIG. 2. Log-log plot of the initial slope of the critical field, parameter A in Eq. (9), vs $p-p_c$. The nucleation-field critical exponent k, Eq. (10), is obtained this way.

generalized form at $H=0$:

$$z_a E\psi_a - \sum_{z=1}^{z_a} \psi_z = w\psi_a. \quad (11)$$

Several physical situations give systems of equations similar[3] to Eq. (11), among these tight-binding Hamiltonians. The pecularity of the "superconductive type" of Hamiltonian is that the site energy ($=z_a E$) is proportional to the number of nearest neighbors. For the particular case we are interested in, $E=1$ ($T=T_{cw}$) and thus the lowest eigenstate corresponds to ψ constant throughout the lattice and w equal to zero. In contrast with other tight-binding models,[15,16] this property does not change when disorder is introduced through removal of sites or bonds. Thus in this model classical and quantum percolation are coincident. A similar response to disorder has been found in elastic media by John, Sompolinsky, and Stephen.[19]

Unfortunately the very methods used in Refs. 15 and 16 to study quantum percolation fail to give an answer about the nature of the excited states. The tridiagonalization procedure does not show convergence of the parameters of the effective Hamiltonian towards constant values (or periodic repetition), a property used in Refs. 15 and 16 to estimate the quantum percolation threshold. Continued-fraction and decimation technique analysis[7] of the tridiagonalized Hamiltonian show that states with energy (w) near zero extend over all of the sample, but this is not a conclusive result: The localization could be larger than the size of the samples.

Elucidation of the nature of these excited states will provide a better understanding of quantum percolation. They are also test cases to check numerical methods. In superconducting networks these excited states modify the pure geometrical arguments used to study the critical exponents and the response of these networks under sizable external currents.

One of us (J.S.) thanks the Consejo Nacional de Investigaciones Científicas y Técnicas for financial support.

[1]P. G. de Gennes, C.R. Acad. Sci. B **292**, 9 (1981).
[2]P. G. de Gennes, C.R. Acad. Sci. B **292**, 279 (1981).
[3]S. Alexander, Phys. Rev. B **27**, 1541 (1983).
[4]G. Deutscher, I. Grave, and S. Alexander, Phys. Rev. Lett. **48**, 1497 (1982).
[5]H. J. Fink, A. Lopez, and R. Maynard, Phys. Rev. B **26**, 5237 (1982).
[6]J. Simonin, D. Rodrigues, and A. Lopez, Phys. Rev. Lett. **49**, 944 (1982).
[7]J. Simonin, C. Wiecko, and A. Lopez, Phys. Rev. B **28**, 2497 (1983).
[8]B. Pannetier, J. Chaussy, and R. Rammal, J. Phys. Lett. **44**, L853 (1983), and Phys. Rev. Lett. **53**, 1845 (1984).
[9]M. Tinkham, D. W. Abraham, and C. J. Lobb, Phys. Rev. B **28**, 6578 (1983).
[10]B. Pannetier, private communication.
[11]R. Rammal, T. C. Lubensky, and G. Toulouse, J. Phys. Lett. **44**, 65 (1983).
[12]D. Kimhi, F. Leyvraz, and D. Ariosa, Phys. Rev. B **29**, 1487 (1984).
[13]D. R. Hofstadter, Phys. Rev. B **14**, 2239 (1976).
[14]S. Aubry, in *Bifurcation Phenomena in Mathematical Physics and Related Topics,* edited by C. Bardos and C. Bessis (Reidel, New York, 1980), pp. 163–184.
[15]R. Raghavan and D. C. Mattis, Phys. Rev. B **23**, 4791 (1981).
[16]J. Simonin, unpublished.
[17]D. R. Bowman and D. Stroud, Phys. Rev. Lett. **52**, 299 (1984).
[18]S. John and T. C. Lubensky, Phys. Rev. Lett. **55**, 1014 (1985).
[19]S. John, H. Sompolinsky, and M. J. Stephen, Phys. Rev. B **27**, 5592 (1983).