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Manifestation of percolation in high temperature superconductivity

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ABSTRACT

Emergent advanced electronic and magnetic functionalities in novel materials appear in systems with a complex lattice structure. The key point is understanding the intrinsic effect of lattice fluctuations on the relevant electronic features in the range of 10–100 meV near the Fermi level in new materials which is needed to develop advanced quantum nano-devices. This requires the control of structural inhomogeneity at multiple scales. Here we report some of the known advances in the field of percolative superconductivity. The necessity of the review is based on the growing consensus that the lack of an understanding of high temperature superconductivity is due to the few information on lattice fluctuations. In particular they could control the pseudo-gap phase, the electronic duality of holes in Fermi arcs and electrons in small Fermi pockets, multiple condensates in different points of the *k*-space. Moreover the emerging lattice granularity in cuprates shifts the search for the superconducting mechanism from a homogeneous superconductivity to a percolative superconductivity, therefore it is the scope of this review to provide further data to this kind of research.

1. Introduction

Modern materials have a strong tendency to inhomogeneity and phase separation [1–3]. This intrinsic aspect of advanced devices is often complicated by a high degree of structural and electronic disorder [4,5].

Phase separation and disorder are considered as the main obstacles for a full understanding of desperate phenomena occurring in modern materials such as high temperature superconductivity. This apparently unavoidable features of modern materials is believed to be an intrinsic manifestation of a multi-band electronics that is often described with multi-Hubbard-models, are known to have a richer phase diagram than simpler single band models [6–11].

Since experiments are showing that the multi-band nature of many advanced quantum nanomaterials and interfaces is accompanied by an ineluctable high level of inhomogeneity, driven by misfit strain and dopants distribution, it is becoming more urgent to identify the essential heterogeneities [12].

Not all the authors convey on the definition of what could be considered an essential heterogeneities, nevertheless it was already considered a central problem in the early days of high temperature superconductivity by the same Nobel prize winner for the high T_c discovery [13], and before [14].

Here we will focus especially on the inhomogeneity which is described by mixing real space and *k*-space information, therefore putting more emphasis on the two-component physics in the real space and in the momentum representation [15].

Several techniques have allowed to spatially resolve the *k*-space information with spatial nanometer to micron-scale resolution such as scanning tunneling microscopy (STM) and scanning nano-diffraction (SμXRD), to mention some of them. Although the extensive application of STM has given compelling evidences of multi-gap heterogeneity in all the classes of high temperature superconductors [16,17], SμXRD has provided insight in the structural aspects of the heterogeneities found respectively in cuprates [18–22] and pnictides [23] on a scale not accessible by STM. The question therefore of which could be the optimum inhomogeneity for a multi-band system in order to display the highest critical temperature or even new functions, has led some authors to propose new recipe based on the experimental approach used. Some definitions of optimum inhomogeneity have been focusing on *k*-space ordering and correlations [24–26], others more on *r*-space ordering and correlations [19,27,28].

Generally, the progressive identification of the underling in homogeneities in multi-band system and their correlations in both *k*-space and *r*-space, has been the fertile ground for theories. In this view, the emergent properties of a system are not only a mere

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outcome of the chemistry of the system, but also depend on how the different components of a system connect to each other, and on the pattern displayed [29,30]. These patterns also show an unexpected advantage for the control [31,32].

An optimum inhomogeneity, showing a special pattern is the one with scale-free distributions, often arising in systems with multiple distinguishable phases driven far from equilibrium [19]. If the multiple phases are close to the atomic limit, the word "superstripes" has been in the course of the years used to describe this particular state of matter [5].

The search of the key ingredients for the optimum inhomogeneity for the highest critical temperature is a daunting task, but in this review we attempt to give some contribution to this problem, by focusing on some relevant issues observed and discussed in superconducting materials in the last three decades such as percolative superconductivity or more general about disorder and superconductivity. Although, this review is far from being exhaustive about this extensive topic both theoretically and experimentally, we hope that is enough to further stimulate interest around this problem.

2. Relevant materials

To get an overview of how often the problem is discussed in literature and for which systems, a table is made of papers that speak explicitly of percolative superconductivity in different superconducting materials (Table 1).

Table 1
Papers that speak explicitly of percolation superconductivity and materials.

Material	References
YBCO	[18,20,36,39,43,58,60,74,76,77,84,85,89,94,95]
Au–YBCO	[67]
La_2CuO_4	[6,18,19,54,86]
MgB_2	[45,54,61,71]
FeTeSe	[1]
FeTeS	[59]
KFeSe	[1,23,38,50]
LaSrCuO	[36,76,85]
LaBaCuO	[57,76]
La_2NiO_4	[3]
$La_2(Zn, Mg)xCu_{1-x}O_4$	[110]
Cuprates	[13,40,41,109]
BSCCO	[22,36,78,85]
Manganites	[2]
$LaMnO_3$	[3]
Pb	[82,99]
k-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]$ Halogen	[108,114]
InGe	[116]
$CeCoIn_5$	[3]
$NbSe_2$	[51]
Nb_3Ge	[90]
ErBaCuO	[53,56]
MBCOH, (M = Y, Er, Eu)	[69]
BSCO	[36]
BLCO	[55]
PrCeCuO	[56]
BaFeCoAs	[49,102]
$Ba_{0.6}K_{0.4}Fe_2As_2$	[38]
$(Fe_{0.92}Co_{0.08})_2As_2$	[38]
$SmFeAsO1-xFx$	[102]
$Tl_2Ba_2CuO_6$	[36]
Doped amorphous carbon	[37]
Granular Sn–Ge films	[52]
Sn–Ge thin film	[112]
Al–Ge films	[65]
Sn, and Pb–Ag films	[99]
Na surface doped WO_3	[81]
Bi–2223	[87]
Bi–2212	[87]
$SrTiO_3$	[84]
$Nd_{2-x}Ce_xCuO_{4-y}$	[91]
$YBa_2SnO_{5.5}$	[95]

3. Percolative superconductivity

Percolation theory can be used in physics as a statistical analysis tool to analyze spatial inhomogeneous materials under the right circumstances [62,63,66,68,72,73,75,79,80,83,88,92,93,96–98,100,101,103–107,111,113,115,117]. First, one needs to have a physical property which depends on connectivity between clusters. Another ingredient is that there is a uniform chance that two clusters are connected, and this probability is a function of certain physical order parameter. As a function of the order parameter one can then use percolation theory to calculate a transition between connected and unconnected states, i.e., a phase transition. The remainder of this paper concerns examples of how percolation superconductivity is discussed in literature.

4. Superconductor–Insulator transitions in granular superconductors

Ref. [74] discusses percolation to describe critical current in polycrystalline superconductors. The grain boundaries that form between the grains are considered weak links, whose critical current density is a function of relative angles. In cuprates this effect is strengthened due to the *d*-wave character of the superconductivity [118]. One can then setup a network of superconductors with different critical currents to calculate the critical current of the total system. A relation for the individual grain boundary critical current is used which is well established for YBCO. Under certain circumstances this problem can be reduced to a percolation problem, and the general solution shows similar behavior.

Ref. [33] formulates a theoretical model for the I–S transition on granular superconductors. A 2D network of random resistors is setup, connecting the grains. It is assumed that two neighboring grains are either Josephson coupled, such that a supercurrent can flow through the junction, or not, so a tunneling current can flow with a finite resistance R_n. A criteria to determine if two junctions are coupled is given, which takes into account both thermal and quantum fluctuations. It depends on the distance r_{ij} between resistors, where $r_{ij} = 2\,\bar{l}\,\xi_{ij}$. ξ_{ij} is uniformly distributed between 0 and 1, and $\bar{l}$ is the average distance between grains. $\bar{l}$ is a function of sheet thickness, because thinner sheets mean larger average distance between grains. For the resulting random resistor network, one can calculate an effective resistance $R(T)$ of the whole network. This $R(T)$ as a function of sheet thickness is compared with experiment on granular Pb films [34], in the temperature range around T_c, and the fitting agrees well with the data. It is done for several values of $\bar{l}$, which can be interpreted as a measure for disorder.

Ref. [64] also describes a model to explain granular superconductors which is similar to the model described in Ref. [33]. One difference is that coupling occurs when the Josephson energy is larger than the thermal energy, and charging energy is neglected. The second is the parameter which is stochastic. In Ref. [33], it is assumed that the distance between neighboring grains is uniformly distributed, while in Ref. [64] it is assumed that the normal state resistance of the various junctions obeys a normal distribution. A critical temperature is obtained by assuming it occurs at the temperature where the percentage of bonds is equal to p_c of the network.

Ref. [35] also addresses theory of granular superconductivity, but the approach it differently. The model [35] assumes a Random Josephson Network (RJN) where neighboring grains are connected with a random probability p, which is analyzed with field theoretical methods. The phase diagram is explored as a function of p, B, T, in particular the S–I transition with order parameter p at a low temperature at 0 magnetic field and at a finite magnetic field (called transition I and II respectively). The diamagnetic susceptibility $\chi(1)$ and the mean square fluctuations of the total magnetic moment $\chi(2)$ of large clusters are calculated for both transitions.

Experimental papers [36–38] report on magnetization measurements on respectively cuprates, sulfur doped amorphous carbon films and iron pnictides. All papers found anomalous behavior in the magnetization hysteresis loops, which they explain with percolative superconductivity.

Ref. [45] uses percolation theory in a different way. In granular superconductors, B_{c2} is a function of the direction of the applied magnetic field. The material MgB_2 is discussed, which is anisotropic. If one lowers temperature at a fixed external magnetic field, some clusters will start to get superconducting, and at some point there are enough superconducting clusters to form a percolative path. The transition then happens at a critical angle, θ_c, where θ is the angle between the external magnetic field direction and the *c*-axis. It is assumed that the boron planes are randomly distributed.

Then the isotropic $J_c(B, B_{c2})$ relation is made anisotropic trough B_{c2}; $J_c(B, B_{c2}(\theta))$. So given a known B one can then calculate all J_c's on every grain and invoke a random resistor network to calculate the critical current of the whole sample.

The idea is that J_c of the whole material goes to zero even below B_{c2}, because due to the anisotropy some grains are not superconducting and a percolative path cannot be formed.

The calculated $J_c(B)$ is compared with experimental data and fits the curves well. Also the broadening of resistive transitions with increasing magnetic field is explained.

5. Fluctuations

In cuprates it is found that above T_c in the underdoped region, there is still a small gap in the density of states around the Fermi level [119]. The next examples explain this with the presence of non-percolating superconducting islands. It implies that a fraction of the material is still superconducting, causing the pseudogap, but no bulk supercurrent can be established because the islands are not connected. This picture is sometimes called fluctuations. There exists then a temperature T^* below which superconducting islands start to form, and T_c is the temperature where the superconducting islands percolate or become phase coherent.

Ref. [39] presents the results of measurements of specific heat on YBCO and finds a sharp dependence of the specific heat jump as a function of doping in the optimal doped region, where T_c is approximately constant. But the specific heat jump should be proportional to the density of states at the Fermi level, which should hardly change because the critical temperature is almost constant in the measured doping region, so BCS theory does not work in this case. The authors argue that the behavior can be explained by assuming the existence of superconducting islands in an insulating matrix that percolate between underdoped and optimally doped region. So, when the doping is decreased starting from optimal doping, there still exists macroscopic phase coherence, so T_c is unchanged, but the size of the percolating cluster does decrease in this region, and the specific heat jump decreases correspondingly. However, if the doping falls below a critical value, the cluster breaks up into smaller clusters and now also T_c starts to decrease.

Ref. [70] did transport measurements on amorphous IO as a function of temperature, just below the SIT transition. It was found that the resistance is simply activated, i.e., $R(T) \sim \exp[T_0/T]$ below approximately 10 K. Above that the resistance reverts to variable range hopping. This is done for different samples with different annealing times, which thus have a different degree of disorder. For the most disordered sample that was measured, a VRH resistance dependence was found even in the temperature region below 5 K. The fact that the resistance reverts from simply activated to VRH as temperature is increased cannot be reconciled with conduction mechanisms such as nearest neighbor hopping (it should be the other way around). The paper suggests that it can be explained with the presence of non-percolating superconducting islands below the Anderson transition, which cause the electrons to tunnel through the superconducting islands. This can be modeled with a random network of SIS junctions, which predicts a simply activated $R(T)$ curve on the I side of the SIT transitions up to a temperature T_0 larger than T_c, and VRH above that.

Ref. [40] explains the pseudogap phenomena differently, not with preformed superconducting islands, but arguing that the pseudogap and superconductivity are competing phenomena. It is formulated as follows: "first, highest level ab initio density functional calculations are done on both undoped and doped cuprates. With this it is found that the holes arising from doping are out of the CuO_2 plane, and has an apical O p_z orbital that delocalizes over a plaque of four surrounding copper sites. For $x \approx 0.05$–0.06, a percolating pathway comprised of adjacent plaquettes is formed. This leads to a Cu $d_{x^2-y^2}$/O p_σ metallic band of states delocalized inside this path of percolating plaquettes which are well described with standard band methods. Undoped Cu sites remain localized d^9 spins with AF coupling. Superconducting *d*-wave cooper pairing of metallic Cu $d_{x^2-y^2}$/O p_σ electrons occurs by coupling to the anti-ferromagnetic undoped d^9 spins at the surface of the percolating metallic path. Thus the onset of metallic percolation is also the onset of superconductivity. At $x \approx 0.271$, undoped d^9 spins no longer have an undoped d^9 neighbor leading to the vanishing of the superconducting pairing and no superconductivity for $x > 0.271$. The pseudogap arises from the splitting of degenerate p_x and p_y states at the Fermi level that reside inside isolated plaquettes (not part of the percolating plaquette path). The magnitude of this psuedogap depends on the distance between other isolated plaques with a certain attenuation length".

With this model a number of properties can be predicted simply by counting plaques (susceptibility, resistivity, ARPES data, and specific heat). The main result is that this theory fits the experimental data for the pseudogap well.

In Ref. [41] the same model is used to also explain the normal-state resistance, which is linear as a function of temperature over the large temperature range of $10 < T < 1000$ K.

ef. [85] implies the existence of a local transition temperature as a function of position r, i.e., $T_c(r)$. As T is lowered below the maximum T_c (which is called T^*), the superconducting islands grow and at some critical temperature called the global T_c, the superconducting regions percolate. The paper also gives a quantitative relation between $T_c(r)$ and the charge distribution $P(\rho)$, which in turn depends on the doping, so implicitly the connectivity probability p is a function of both T and $\langle\rho\rangle$. This provides a phase diagram as a function of doping and temperature, and predicts a transition between superconducting, pseudogap and non-superconducting phases.

6. Vortex percolation

Percolation theory can also be applied to the Bragg glass–vortex liquid transition [42]. In the Bragg glass state vortices have only small lateral displacement. If one assumes an order parameter which represents the amount of lateral displacement a vortex can have per unit vortex length, the lateral displacement as a function of that order parameter can be such that the vortices start to wind around each other, which is the case in a vortex liquid. Ref. [42] shows that this effect can be modeled with a percolation problem.

Ref. [43] uses percolation theory to explain a different aspect of vortices in HTS, namely depinning. Vortices form around local non-superconducting pinning sites (usually caused by spatial defects), which is thermodynamically preferred (i.e., you do not 'waste' superconducting space for the vortex). In a real material with irregular defects, a complex thermal equilibrium is formed where the loss of condensation energy is optimized. Now, if one applies a current, these vortices experience a Lorentz force and try to move, but they can only do this if the current is strong enough to overcome the pinning force, and if there is a non-superconducting region nearby. This paper discusses the case where one has a superconducting sea with normal-state islands, connected by weak links. Vortices can only move to other islands which are coupled to the island it resides on. This again is a percolative problem, where the number of phase-coherent neighbors is a critical parameter which is argued to depend on the fractality D of the normal phase clusters (which is half of the power law dependence between the perimeter and the area of an island, perimeter $\sim$ area$^{d/2}$). In an YBCO film this parameter is measured to be non-unity, but equal to 1.44 (this is done by measuring the perimeter and area of many normal phase clusters).

7. Percolation threshold resonance in superconductivity

In Ref. [18] an experiment is described where nanofocused XRD measurements were done on $La_2CuO_{4.1}$. It was found that there are domains in the superconductor which have a locally altered lattice structure. Particularly, oxygen interstitials in the spacer layer form stripes. A second phase was observed in Ref. [19] and associated to the self-organization of local lattice distortions. By using different quenching processes, two distinct phases can be made, named the $T_c = 40$ K phase and the $T_c = 16 + 32$ K phase (which has two transition temperatures, this is measured with a single-coil inductance method, which shows two peaks in the derivative of $1/\omega(T)^2$). The diffraction peak intensity of the nanofocused XRD measurements were plotted as a function of real space for both phases. This map turns out to be very inhomogeneous, so there are local ordered oxygen interstitial and lattice distortions domains. Then a correlation function was calculated, and it showed a power law behavior (with an exponential cut-off) for both phases. Also the intensity probability distribution showed power law behavior for both phases from which it is concluded that the network of these ordered oxygen interstitial domains is scale free with exponential cut-off. It is suggested that the scale free structure promotes superconductivity, possibly with percolation effect.

Since scale-free self-organization in a system are often associated to the proximity to critical transition, it should be considered not a surprise that the system is very sensitive to an external driving source such X-ray illumination [32]. The opportunity to tune by X-rays continuous exposition of the system, the oxygen interstitials self-organization in the spacer layers near room temperature, and also at lower temperature the local lattice distortions [19,48], enables the possibility to pattern superconductors with X-rays. The percolative evolution of the oxygen interstitials order has been also theoretically investigated [47].

Ref. [120] works out a model of high T_c superconductivity with the Ising model. Here an array of clusters is defined where each cluster has a spin. There is an adjacency matrix which defines which clusters are connected, and only connected clusters can interact and exchange spin. Given a certain coupling, at a critical temperature the spins get a net magnetization as a function of magnetic field, and this is assumed to be equivalent to the formation of cooper pairs. Now, the clusters are determined as follows: a square grid of sites is defined, and the chance that a site is occupied is p. Then clusters are defines as a collection of sites which have a distance $d \leqslant \sqrt{2}$. Two clusters are considered connected if the minimum distance between the two clusters $d \leqslant \sqrt{5}$. The critical temperature as a function of p is calculated, and it had a sharp maximum at p_c, where p_c is the p where the clusters percolate. Thus, in this model there is a resonance in the critical temperature at the percolation threshold. It is speculated that the variable p can be related to the amount of doping in cuprates.

Then in Ref. [28] the same model is used on a scale free network (such that the degree distribution is power law with exponential cutoff, $p(\theta) = \theta^{-\gamma} e^{-\theta/\xi}$, and the network can be generated as a function of the power law and the exponent). The resulting critical temperature turned out to strongly depend on the parameters of the scale free distribution. Particularly, only if $\gamma < 3$, then T_c diverges as the exponent ξ diverges.

Ref. [44] studies the Bose-Hubbard model (analyzed with mean field theory) on a percolative network to study the Mott transition. The Bose-Hubbard model has a connectivity term to indicate which sites can have paired interaction, and this network is defined in such a way that there is a power law degree distribution with exponential cutoff, like in Ref. [30]. It is again found that the Mott transition depends on the exponent and power of the degree distribution. In particular it is found that if the exponential cut-off diverges and if the power is smaller than 3, the Mott insulating phase disappears. This study is repeated on apollonian networks as an example of this statement, because it has a power law degree distribution with no exponential cut-off and power law smaller than 3, so the Mott insulator phase is expected to disappear in the limit of an infinite network, and indeed it does.

8. Other applications of percolation theory

Ref. [46] proposes an experimental technique where a change in certain properties (resistance or superfluid properties) is measured when a probe is held (charged AFM or STM tip) near the surface. This procedure will allow spatial resolution of the local propagation of coherence in disordered superconductors, and, more generally, in disordered systems manifesting the BKT (Berezinsky–Kosterlitz–Thouless) transition. This BKT transition is driven by the unbinding and proliferation of vortex–antivortex pairs at the critical temperature T_{BKT} (smaller than T_c), which destroy global phase coherence.

A numerical simulation is done to make specific predictions about what the outcome of such measurements would be.

To this end, the *XY* model is used. The quantity helicity modulus is studied on this model, as it undergoes a BKT phase transition. The coupling between nearest neighbors J_{ij} is the exponential of a uniform distribution between -5 and 5 times J_0. dY_{ij} is the quantity that represents the difference between undisturbed Y and Y_{ij} for which a bond between i and j is broken. dY is the sum over all dY_{ij}'s. By plotting dY_{ij} one can see a formation of paths, so the coherence propagates along ramified networks in the *XY* model, similar to percolation networks. Simulations indicate that percolation on this network is lost as T approaches T_{BKT}. In other words, the effect of breaking interactive bonds in the *XY* model has the most impact on the helicity when done along percolative paths. In this sense the *XY* model predicts a 'hidden' percolative network.

Ref. [102] discusses the use of fast neutrons (0.1 MeV) to bombard FeAs like samples. This neutron irradiation causes impurities in the sample, on the nanometer scale (from point defects to several nanometers). It is shown that as a function of irradiation normal state resistance increases, critical temperature decreases and $B_{c2}(T)$ decreases (at least in the region near T_c). Further, if temperature is normalized to T_c, the critical field $B_{c2}(T/T_c)$ hardly depends on neutron irradiation. Critical current and bulk pinning is strengthened in most of the T and B range.

It is concluded that fast neutrons could be useful to create pinning defects and increase critical currents. Furthermore, the paper uses percolation theory to explain these effects. It is argued that two types of weak links can form between the grains, ones that can carry current at high fields and ones that do not. This is used to explain the magnetization and critical current data.

9. Conclusion

The problem of percolation in superconductivity has been reviewed. Although considered as a mere problem of material science, recently has been started to be considered not only an intrinsic aspect of modern materials but also an advantage for their manipulation. In fact, a current challenge in science and technology is not only to observe matter on ever smaller scale, but also to direct its functionality at the relevant length, time and energy scales. A recent report by the basic energy science advisory committee in USA, directing matter and energy: five challenges for science and the imagination, emphasizes this advent of control science. In relation to this it is important to recall that the exploitation of conventional semiconductors took off when materials scientists have learned how to grow crystals that are nearly perfect. Modern semiconductor technology is dependent on the ability to accurately place dopants in such crystalline materials and manipulate electrical transport with external fields in nano-scaled device structures. Percolation therefore provides not only a crucial basic aspect of modern materials, as shown by many works provided here, but also an opportunity for which new control tools are needed.

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