The Role of the Short Coherence Length in Unconventional Superconductors

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Abstract: A short coherence length is a distinctive feature of many cases of unconventional superconductivity. While in conventional superconductors, it is many orders of magnitude larger than the basic inter-particle distance, a short coherence length is common to superconductors as diverse as the cuprates, the picnites and granular superconductors. We dwell particularly on the last, because their simple chemical structure makes them a favorable material for exploring fundamental phenomena such as the Bardeen-Cooper-Schrieffer (BCS)-to-Bose–Einstein condensation cross-over and the effect of the vicinity of a Mott metal-to-insulator transition.

Keywords: coherence length; granular superconductivity; Mott transition; BCS–BEC cross-over

1. The Coherence Length in Conventional versus Unconventional Superconductivity

The importance of the role played by the short coherence length in the physical properties of the high Tc cuprates has been at the heart of my collaboration with Alex Muller [1].

In the framework of the Bardeen–Cooper–Schrieffer theory of superconductivity, Cooper pairing involves a very large number of pairs interacting within the radius of one pair. This radius is the coherence length of the superconducting state, over which the superconducting order can only vary slowly. The number of pairs within a coherence volume is of the order of the square of the Fermi energy divided by the energy gap. In a typical metal, this number is of the order of $10^6$ to $10^8$. This very large number is the reason why the superconducting state is so robust in the face of local perturbations of the crystal lattice, such as impurities or grain boundaries. It characterizes conventional superconductivity.

Can superconductivity persist if the number of pairs within a coherence volume is of order unity, and if yes, what are the properties of such an unconventional superconducting state? This question is important from both a theoretical and practical point of view. It is in fact at the heart of the possible applications of the high temperature superconductors discovered by Bednorz and Muller.

The cuprates are, of course, the most famous example of short-coherence-length superconductors. It is about 1.2 nanometers or three lattice spacings in the CuO planes. This immediately explains why superconducting properties such as the critical current density are so sensitive to defects such as grain boundaries that have little impact in conventional, long-coherence-length conventional superconductors [1]. The coherence length is even shorter along the direction perpendicular to these planes. At optimum doping, the number of pairs in a coherence volume is only of order 10, and the condensation energy per coherence volume is only a few times larger than the critical temperature. This puts severe limits on the possibility of designing effective pinning centers.

The short coherence length is also an indication regarding the type of interaction that is at the origin of the superconductivity in the cuprates. Regrettably, we do not yet have a full theoretical understanding of this mechanism. Is it basically of the same kind as that in conventional superconductors, namely, an interaction between charge and lattice displacement, or is it of a different nature, primarily involving the spin’s degrees of freedom? Alternatively, is it a mixture of both? The short coherence length favors this possibility, as do local contractions of the Cu–O–Cu bond lengths [2,3].
In any case, the classification of conventional versus unconventional superconductivity according to the number of pairs in a coherence volume has the advantage of being very general. It does not presume anything regarding the pairing mechanism or the symmetry of the superconducting order. A well-known transition from conventional to unconventional superconductivity is the cross-over regime between BCS and Bose–Einstein condensations. It indeed occurs when the number of pairs per coherence volume is of order unity. It is continuous, as shown by Nozieres and Schmitt-Rink [4], following the earlier work of Leggett [5]. In these theoretical papers, the control parameter is the strength of an attractive electron–electron interaction. Experimentally, we do not know how to control this parameter. However, we can use geometrical constraints to directly reduce the number of pairs per coherence volume and, in this way, experimentally study the cross-over regime. This has been achieved by studying granular superconductivity.

2. Granular Superconductivity

What happens to superconductivity when the volume of a grain becomes smaller than the coherence volume? At first, not much, as long as the number of pairs in the grain remains much larger than unity. However, if that number becomes of order unity, or smaller, the superconductivity is quenched. This happens when the distance between discrete electronic levels becomes smaller than the energy gap. For aluminum, for instance, this occurs below a size of about 5 nanometers.

A more interesting question is, what happens if we take such small, non-superconducting grains and start to couple them together through weak tunnel barriers? It turns out that this is, in fact, a very fundamental problem, which I have called “transition to zero dimensionality” [6].

As long inter-grain coupling is very weak, the granular system is non-superconducting and insulating. When the coupling is strong, namely, when the level broadening is much larger than the distance between the discrete levels in the individual grain, we expect to recover BCS superconductivity. However, what will happen in between? At what coupling strength will this happen? What will be the properties of the emerging superconductor?

This is not a gedanken experiment, but one that has actually been realized. When aluminum is evaporated in a vacuum system in the presence of a finite vapor pressure of oxygen, aluminum oxide forms and is segregated at Al grain boundaries, because the metal and the oxide are immiscible. Additionally, there exists only one aluminum oxide, Al$_2$O$_3$, which simplifies the situation. As the oxygen vapor pressure is increased, more oxide is formed, the tunnel barriers become less transparent and the electronic coupling between neighboring Al grains becomes weaker [6]. In this way, one can go continuously from weak to strong coupling.

The continuous transition predicted by Legget, Nozières and Schmitt-Rink, and studied recently in more detail by Strinati and coworkers [7], does occur. It takes place when the coherence length is of the order of a few grain sizes. The ratio of the gap to the superconducting critical is higher in weakly coupled grains than in strongly coupled ones.

However, the emerging superconductor has some unexpected properties:

(i) The critical temperature of the granular superconductor goes through a maximum, which is higher than that of the bulk material [6];
(ii) The emerging superconductor allies a very small superfluid density and a relatively high critical temperature [8];
(iii) Additionally, free spins appear [9].

3. Granular versus Atomically Disordered Superconductors

These features are related to the nature of the metal-to-insulator transition, which is of the Mott type [9], while in atomically disordered superconductors, it is of the Anderson type. Surprisingly, the effect of disorder is stronger in the case of “homogeneous” disorder than in the granular case. In the case of homogeneous disorder such as in NbN$_x$, where it is created by
introducing N vacancies, Anderson localization drives the metal-to-insulator transition [10]. It occurs when the product $k_Fl$, where $k_F$ is the Fermi wave vector and $l$ is the mean free path, is of order unity. Enhanced electron–electron interactions do occur, but disorder is the leading factor.

In the granular case, Coulomb interactions are the leading factor driving the metal-to-insulator transition. They result from the grain’s electrostatic energy. As shown by Antoine Georges and co-workers, conduction through a weakly coupled grain is governed by the combined effect of this charging energy and of the splitting between the electronic levels of the isolated grain [11]. A Kondo resonance occurs when these two energy scales are of the same order. Surprising effects can then occur, for instance, when the charging energy is somewhat larger than the energy level splitting. At temperatures lower than the charging energy, a multi-level Kondo effect allowed by level broadening can restore metallic conduction. When the temperature becomes lower than the level broadening, the conduction becomes insulation-like. However, at even lower temperatures, lower than the energy level splitting, the Kondo resonance re-instates a metal-like conductivity. This predicted non-monotonous conductivity behavior has indeed been observed in nano-scale granular aluminum [12]. It can be considered as a precursor of a Mott transition. This is supported by the presence of free spins inferred from magneto-resistance measurements and, more directly, by muon spin rotation experiments.

4. Superconductivity near a Mott Transition

At a Mott transition, which occurs when the Coulomb energy is of the same order as the band width, the density of states at the Fermi level remains finite [13]. At the same time, the effective band-width reduces and tends towards zero. If the system is superconducting, the finite density of states allows the critical temperature to remain finite up to the transition and the gap to remain well defined, while the superfluid density tends towards zero.

These two features—the absence of sub-gap states and, at the same time, a very small superfluid density—are indeed observed in granular aluminum near the metal-to-insulator transition. They are not observed in atomically disordered superconductors near their transition, where the critical temperature collapses and many sub-gap states appear, because the transition is of the Anderson type. The vicinity of a Mott transition is what allows the successful use of thin film granular superconductors to produce circuit elements that have, at the same time, a large kinetic inductance and low losses [14].

It is interesting to note that a transition to zero dimensionality, first identified many years ago in granular aluminum in our original paper [6], has recently been invoked to explain the behavior of very thin NbN films [15]. These two-dimensional films, originally considered to be homogeneous, may in fact be granular. The same conclusion was reached in an extensive review of other nominally homogeneous two-dimensional films near the superconductor-to-insulator transition [16]. While the 2D homogeneous case was thought to be more accessible to theoretical analysis than the 3D granular “dirty” case, it may turn out in the end that the issue of granularity cannot be avoided near the superconductor-to-insulator transition.

5. Conclusions

The continuing in-depth study of granular superconductivity offers the possibility of studying a system that is chemically much simpler than the cuprates and, at the same time, shares with them some fundamental properties. Both are in a BCS-to-BEC cross-over regime, as attested to by their short coherence length (a few lattice spacings in the cuprates and a few grain sizes in granular Al) and the large value of the strong coupling factor. In both cases, one is close to a Mott insulating state—spins are present as expected [14]. The experimental results suggest that there is a close relationship between a Mott transition and the BCS-to-BEC cross-over. However, while the theories of the BCS–BEC cross-over [7] and of the Mott transition [13] are separately well developed, a comprehensive approach involving both of them is still lacking and needed for a comparison with experiments. While the
The cross-over regime is not favorable for some applications such as for high temperatures, high currents and high fields, it is favorable for some others such as high-kinetic-inductance elements for qubit circuits.

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