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Cooper, John R.; Babić, Dinko; Loram, J. W.; Lo, Wai; Cardwell, D. A.

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GROUND STATE SUPERCONDUCTING PHASE FLUCTUATIONS AS A PRECURSOR FOR STRONG CRITICAL FLUCTUATIONS IN HIGH- T_c SUPERCONDUCTORS

J. R. COOPER^a, D. BABI $\acute{C}^{b,c}$, J. W. LORAM^a, WAI LO^{a,d} and D. A. CARDWELL^a

 a^a Interdisciplinary Research Centre in Superconductivity, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom

 b Institute of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

 c Department of Physics, Faculty of Science, University of Zagreb, Bijenička 32, HR-10001 Zagreb, Croatia

 d Texas Center for Superconductivity, University of Houston, Houston, U.S.A.

Dedicated to Professor Boran Leontić on the occasion of his 70th birthday

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We analyse the reversible magnetisation and heat capacity of YBa₂Cu₃O_{7−δ} in the "vortex liquid" state and find that both properties are reasonably well described by the 3D XY critical-fluctuation model. The free-energy density in the "vortex liquid" state has a particularly simple form over a wide range of fields (H) and temperatures (T) . This leads us to a picture in which the presence of critical fluctuations in high- T_c superconductors is directly linked to the remarkably small number of overlapping Cooper pairs at $T=0$ and $H=0$ rather than low dimensionality or high temperatures.

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1. Introduction

The discovery of high- T_c superconductors (HTS) in 1986 [1] generated great enthusiasm concerning possible large-scale applications of superconductivity at liquidnitrogen temperatures. Considerable progress has been made in a relatively short time and superconducting tapes are now being produced commercially on a large scale for use in superconducting magnets and other devices. One of the limiting

factors for high-field applications is the irreversibility line $H_{irr}(T)$, which separates regions of irreversible and reversible magnetisation. In many cases $H_{irr}(T)$ is fairly close to the vortex-solid melting line $H_m(T)$ [2, 3], at which there is a weak first-order phase transition [4, 5] that is believed to correspond to the disappearance of the vortex lattice and a transition to the "vortex liquid" phase. In general, $H_{irr}(T) \leq H_m(T)$ [3, 6]. The name "vortex liquid" should not be taken too literally, because although neutron diffraction studies [7] show that the ordered vortex lattice disappears at $H_m(T)$, as yet there is no direct evidence for the presence of line vortices, i.e. a liquid, above $H_m(T)$. In the "vortex liquid" state the diamagnetic response still indicates the presence of superconducting pairing, the resistivity is finite and ohmic but considerably lower than the extrapolated normalstate value even in very high magnetic fields. The qualitative difference between the magnetoresistance curves of HTS, which broaden in a magnetic field, and those of conventional superconductors, which simply shift to lower temperatures, was an early indication of a crucial difference between the two classes of superconductors.

The details of the phase transition at $H_m(T)$, and the nature of the "vortex liquid" state, are only partly understood. However there is evidence that for YBa₂Cu₃O_{7−δ} (YBCO(7-δ)), which for $\delta = 0$ has the highest $H_m(T/T_c)$ of all HTS, both the phase transition and the properties of the "vortex liquid" are reasonably well described within the framework of the 3D XY critical-fluctuation model [2, 3, 8, 9]. The region in the $H-T$ phase diagram where mean-field theory fails and critical-fluctuation theory gives an adequate description of the experimental data is unexpectedly large. For YBCO(7- δ) it typically extends over a region $0.8 - 0.85 \le t \le 0.99$, where $t=T/T_c$ (note that in the 3D XY picture T_c is not field-dependent), and above $H \approx 10 \text{ kOe} (= 796 \text{ kA/m})$, with only slight dependence on the doping δ [3, 8]. 3D XY scaling, but with a different scaling function, also has been found [9, 10] in $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi2212) which is highly anisotropic and has particularly low values of $H_m(T/T_c)$. This suggests that the fluctuation behaviour outlined above is an inherent property of HTS.

In conventional superconductors the behaviour mentioned above is hardly detectable and $H_{irr}(T)$ is very close to the upper critical field $H_{c2}(T)$, which makes any dissipative "vortex liquid" region negligibly small. Even in conventional superconductors with very low flux-line pinning, such as amorphous $Nb₃Ge$ thin films, $H_{irr}(T)$ stays close to $H_{c2}(T)$ for three-dimensional samples (where the film thickness is larger than the coherence length). In two-dimensional samples the values of $H_{irr}(T)$ are lower but they are still high in comparison with those of HTS, when measured in units of $H_{c2}(T)$ [11]. Moreover, it is not clear that critical fluctuations of the 3D XY class could account for the disappearance of the vortex lattice at $H_{irr}(T)$ in conventional superconductors. Probably the transition is of a different type, e.g. melting that is governed by a classical process described by the Lindemann criterion [12].

Several papers (e.g. [13, 14]) have discussed why HTS are so different from conventional superconductors, especially with respect to their high susceptibility to critical fluctuations. The obvious reasons are that the superconducting transition temperatures of HTS are an order of magnitude higher than those of conventional

superconductors and that, being anisotropic, HTS have reduced dimensionality, both of which enhance critical fluctuations. In this paper we focus on the third cause of strong fluctuations in HTS, the small values of the in-plane coherence length $\xi_{ab}(T)$ [13, 14]. We give arguments that there is a fundamental difference between HTS and conventional superconductors with regard to the number of overlapping Cooper pairs, i.e. the number of pairs in the coherence volume. This number is small in HTS but very large in conventional superconductors, which then causes the build-up of the superconducting phase to be quite different in the two classes of superconductors. In this picture the extreme susceptibility of HTS to critical fluctuations is due to the small number of Cooper pairs in a coherence volume even at $T=0$ and $H=0$. This makes the superconducting phase less stable at elevated temperatures and magnetic fields.

2. Discussion

2.1. Experimental implications

The 3D XY approach to the properties of HTS in a magnetic field has been discussed in detail by Fisher, Fisher and Huse [15] and by Schneider and Keller [16]. In the 3D XY picture the free-energy density f_s which properly describes the thermodynamics of HTS in the fluctuation-dominated regime has the form

$$
f_s = f_n - \frac{Bk_B T}{V_c} G\left(\frac{H\xi^2}{\Phi_0}\right) ,\qquad (1)
$$

where f_n is the normal-state free-energy density, B a constant of order unity, $V_c =$ $\xi_{ab}^2 \xi_c$ the temperature-dependent coherence volume, ξ a coherence scale, Φ_0 the magnetic-flux quantum, and G an unknown scaling function. For H \parallel c, $\xi = \xi_{ab}$, and for H || ab, $\xi = (\xi_{ab}\xi_c)^{1/2}$. The temperature dependence of the coherence lengths in the 3D XY universality class is given by $\xi_{ab,c} = \xi_{ab,c}^0 (1-t)^{-2/3}$ (the theoretical value [17] of the exponent is $\nu = 0.669 \pm 0.002$ while the measured value [18] for liquid ⁴He is $\nu = 0.672 \pm 0.001$.

In our recent work on the reversible magnetisation $M = -\partial f_s/\partial H$ of YBCO(7- δ) [2, 3, 8], after making small corrections for paramagnetic Curie terms determined by fitting susceptibility data well above T_c to a Curie law, we have indeed found excellent validation of the 3D XY scaling expected from Eq. (1) in a large portion of the H-T plane right down to $H_{irr} \approx H_m$. Furthermore, a similar scaling approach to the heat capacity of YBCO [9] also supports the above form of f_s . Further analysis of our experimental $M - H$ data [3, 8] led us to an explicit expression for f_s , namely

$$
f_s = f_n - F(T) + \frac{Bk_B T}{\xi_{ab}(T)\xi_c(T)\sqrt{\Phi_0/H}}
$$
\n(2)

 $(F(T))$ is essentially field independent), which holds throughout the "vortex liquid" state investigated (although for YBCO(7) there are some deviations at

high fields, see Fig. 1). Equation (2) provides a simple physical result, namely the field-dependent part of f_s is the thermal energy k_BT divided by a fielddependent coherence volume $\xi_{ab}(T)\xi_c(T)(\Phi_0/H)^{1/2}$. As shown in Fig. 1, our reversible-magnetisation data with $H \parallel c$ plotted in the standard 3D XY way (i.e. $M/TH^{1/2}$ vs. $H/(1-t)^{4/3}$) collapse on to a single curve for all δ values investigated $(0.03 \pm 0.02 \le \delta \le 0.35 \pm 0.02)$ when we take into account the measured values of the anisotropy parameter $\gamma(\delta) = \xi_{ab}(\delta)/\xi_c(\delta)$ and choose appropriate values for the parameter $D(\delta) = \xi_{ab}^{0}(\delta)/\xi_{ab}^{0}(\delta = 0)$. The value $\xi_{ab}^0(\delta = 0) = 1.26$ nm was known from the previous work [2, 14], this leads

Fig. 1. 3D XY plot of the reversible magnetisation M_{rev} of YBCO(7- δ) for $0.03 \leq \delta \leq 0.35$, from Ref. [8], where $A = 2\Phi_0^{1/2} (\xi_{ab}^0 (\delta = 0))^2/Bk_B = 0.07$ $KG^{-1}Q^{-1/2}$ (1 $Qe = 79.58$ A/m). M_{rev} shows an $H^{-1/2}$ dependence over the whole doping range, providing evidence for the field-dependent term in Eq. (2). In the inset we show the δ dependences of the anisotropy parameter γ determined from other measurements [3] and the parameter $D = \hat{\xi}_{ab}^{0}(\delta)/\xi_{ab}^{0}(\delta=0)$. The latter is chosen so that data for samples with different values of δ collapse on to the same curve.

to $B \approx 1.5$. Note also that the values of D used are consistent with estimates of ξ_{ab}^0 obtained from measurements of the fluctuation diamagnetism well above T_c . The results in Fig. 1 suggest that 3D XY scaling of the reversible magnetisation could be

general in HTS; otherwise the YBCO(7- δ) samples with different oxygen contents, i.e. different critical temperatures and anisotropies, would not behave in the same way. Moreover, the validity of Eq. (2) over a substantial region of the fluctuationdominated part of the $H-T$ plane suggests that the correct theoretical model of the "vortex liquid" phase should give the same simple relationship between the thermal energy and the coherence volume that is observed experimentally. However we should emphasise that the present results have been obtained over a limited range of γ (a factor of 2) and that highly anisotropic Bi2212 has a different scaling function [9, 10].

Fig. 2. $f(H,T) - f(0,T)$ vs. $H^{1/2}$ plots derived from the specific heat data of of Ref. [20] (a melt-processed YBCO(7) sample with large aligned crystallites and H | c). The melting line $H_m(T)$ derived from these data, which agrees with our magnetisation results, is shown by the full line. The temperatures are indicated. These plots are linear in the "vortex liquid" state with zero intercept at $H = 0$, as expected from Eq. (2).

A natural extension of the magnetisation work is to investigate Eq. (2) with respect to other thermodynamic properties, such as the specific heat [19] $C =$ $-T\partial^2 f_s/\partial T^2$. We have done this by comparing the predictions of Eq. (2) with several independent studies of the heat capacity of YBCO, including single crystals [4, 5] and melt-processed samples with large aligned crystallites [20]. We have found

that the free-energy-density data derived from these measurements do fit Eq. (2). As an example in Fig. 2 we show the free energy density results for melt-processed YBCO(7- δ) [20] for $\delta \approx 0$, plotted against $H^{1/2}$. The magnitude of f_s agrees with Eq. (2) if $B = 1.5$ and $\xi_{ab}^0 = 1.39$ nm. Consequently plots of $C(H, T)/T - C(0, T)/T$ vs. $H^{1/2}$ should also be linear. This is also observed experimentally, but the slopes of these plots do not agree with the full second derivative of Eq. (2). as shown in Fig. 3. However, agreement can be restored if we replace the thermal energy k_BT in the numerator of the field-dependent part of Eq. (2) with a constant of order $k_{\rm B}T_{\rm c}$. In this case the second derivative of f_s then varies as $(1-t)^{-2/3}$. As also shown

Fig. 3. The slopes of $C(H,T)/T - C(0,T)/T$ vs. $H^{1/2}$ plots derived from data in Ref. [20]. Full squares represent the $(1-t)^{-2/3}$ dependence (left-hand scale), where $t = T/T_c$, and $T_c = (89.4 \pm 0.3)$ K. This is expected from the second derivative of Eq. (2) if the k_BT term in the numerator is replaced by k_BT_c . It can be seen that the measured slopes of C/T vs. $H^{1/2}$ are consistent with a $(1-t)^{-2/3}$ dependence within the scatter of the data since they fit a straight line through the origin. On the other hand the slopes are not proportional to the full second derivative of Eq. (2) (including the k_BT term) since the open points (right-hand scale) do not lie on a straight line through the origin.

in Fig. 3, within the experimental error, the slopes of $C(H, T)/T - C(0, T)/T$ vs. $H^{1/2}$ plots are consistent with this $(1-t)^{-2/3}$ dependence. This replacement does not affect the reversible-magnetisation and free-energy-density plots due to their weaker sensitivity on the k_BT term in the numerator of the field-dependent part of Eq. (2). One possible way of justifying the above replacement is to say that the origin of the pronounced fluctuations in HTS is to be sought in the ground state of the system, i.e. at $T=0$ and $H=0$. We discuss this possibility in the next section. We cannot entirely rule out a more prosaic scenario, namely that there is a small extra field-dependent term in f_s , not included in Eq. (2), which has large temperature derivatives and so has a strong effect on $C(H, T)$ with little effect on $M(H, T)$ and $f_s(H,T)$, but this seems to be a very artificial "ad hoc" interpretation.

2.2. Number of overlapping superconducting pairs at $T=0$ and $H=0$

The superconducting condensation-energy density $U_0 = H_c^2(0)/8\pi$ is the appropriate measure of the strength of superconductivity, as it equals the free-energydensity difference between the superconducting and the normal states of a superconductor at $T = 0$. The quantity $U_0 \xi_0^3$, where ξ_0 is the BCS coherence length, gives the number of superconducting pairs N_p (each carrying energy Δ_0) within the coherence volume, i.e. $N_p = U_0 \xi_0^3 / \Delta_0$. Since at $T = 0$ each pair can be considered to occupy a volume ξ_0^3 , N_p is also the number of pairs that spatially overlap a given pair. N_p can be derived from experimental data for U_0 , ξ_0 and T_c via the parameter $K = U_0 \xi_0^3 / k_B T_c$. Within standard, weak-coupling s-wave BCS theory $K = \alpha N_p$, where $\alpha = \Delta_0/k_B T_c = 1.76$. For electrons in a 3D parabolic energy band, weak-coupling BCS theory gives $K = (\alpha/\pi^5)(E_F/\Delta_0)^2$, where E_F is the Fermi energy, and therefore N_p is very large in conventional superconductors.

Before we proceed with further analysis of the findings of Sect. 2.1, let us discuss the importance of N_p in a qualitative way. Superconductivity is a manybody quantum-mechanical state, determined both by the amplitude of the order parameter, i.e. Δ_0 , and also the phase φ which describes the coherence of the many-body state formed by individual Cooper pairs. Both Δ_0 and φ are manybody properties, but Δ_0 is more closely connected with the individual properties of one superconducting pair, whereas φ is connected with the quantum-mechanical coherence of the many-body system. The phase φ is very sensitive to N_p because if there are not enough overlapping pairs to build up coherence, φ will show significant deviations from its mean value, giving temperature-independent fluctuations $\delta\varphi$. Tinkham [21] pointed out a useful analogy for this process, as follows. A coherent electromagnetic field in lasers can be built up only if there are enough photons. Each photon has an energy $\hbar\omega$ (a single-photon property) but a fully coherent state described by a single, macroscopic phase can be established only if a large number of them contribute to a beam of light. This property of needing many particles in superposing individual states to form a macroscopically coherent state is a general property of many-body coherent systems.

The above discussion is essential for understanding our analysis from Sect. 2.1.

Namely, by combining our magnetisation data with values of U_0 from the specificheat data of Loram et al. $[22]$ we were able to calculate the parameter K for YBCO(7- δ) [3]. The coherence volume depends on the model used, e.g. for the anisotropic effective mass model it is $(\xi_{ab}^0)^2 \xi_c^0$ and for a layered 2D model $(\xi_{ab}^0)^2 d$, where d is the c-axis lattice parameter. Disregarding the details of the model K is always found to be very small (0.2 in the former [3] and 0.8 in the latter case) and basically independent of δ . There are two main implications of this result. The first is that the universal behaviour of the fluctuation-dominated reversible magnetisation shown in Fig. 1 is not just a coincidence but must be linked to the fundamental parameter K , implying that we are dealing with the same physical mechanism for YBCO(7- δ) over a large doping range. The second is that the number N_p is remarkably small in HTS. This is in a great contrast with the value of N_p in conventional superconductors, where this number is very large since E_F >> Δ_0 . This might shed more light on the observed differences between HTS and conventional superconductors, which in the present picture arise from different properties of their ground states at $T=0$ and $H=0$. In conventional superconductors the superconducting phase peaks sharply around its mean value and there are only appreciable phase fluctuation when external perturbations, such as temperature or a magnetic field, are very strong. The consequence of this is that critical fluctuations can occur only over a very restricted region of the H-T plane. In HTS, on the other hand, the temperature-independent phase uncertainty $\delta\varphi$ is considerable even at $T = 0$ and $H = 0$, which results in an intrinsic instability of the phase. Furthermore, in this picture the thermal energy plays a second-order role, compared with the number of pairs within a coherence volume, in defining a macroscopic phase. Thus, the replacement of k_BT in Eq. (2) with a constant of order k_BT_c which was required to explain the specific-heat data (Sect. 2.1) now becomes more meaningful. It means that the field-dependent part of Eq. (2) represents the increase in $\delta\varphi$ as the correlation volume $\xi_{ab}\xi_c(\Phi_0/H)^{1/2}$ shrinks with increasing magnetic field, without a significant contribution from the thermal energy which thus does not appear in the numerator of the field-dependent part of Eq. (2).

The above discussion does not take into account the probable d-wave symmetry of the order parameter in HTS, which may well have some important effects on our qualitative model. For d-wave superconductivity there are nodes of zero gap at certain points of the Fermi surface. As pointed out by Kosztin and Leggett [23], Cooper pairs near these nodes have large values of ξ_0 . Because $\xi_0 = \hbar v_F / \pi \Delta_0$ (v_F is the Fermi velocity), there will be a complicated mixture of two types of pairs: "short" ones made up of $\vec{k} \uparrow$ and $-\vec{k} \downarrow$ states well away from the nodes, and "long" ones near the nodes. The overlap of these pairs is then more complicated than in the simple analysis presented here, and could result in somewhat larger values of N_p and, consequently, smaller phase fluctuations $\delta \varphi$. This might be another possible reason for the unusual increase of H_{irr} at low temperatures [24]. We believe, however, that N_p calculated within a d-wave approach would never reach the value obtained in conventional superconductors, and that our main conclusions still hold reasonably well.

2.3. Some other evidence for phase fluctuations in high- T_c superconductors

Besides the work on the thermodynamics of HTS described in previous sections of this paper there is other evidence for strong dephasing phenomena in HTS. More specifically, there were several reports [25–30] of unusual ohmic magnetoresistance of Bi2212 and $YBa₂Cu₃O₇ / PrBa₂Cu₃O₇$ multilayers deep in the "vortex liquid" state. Here we concentrate on the results obtained in the group of Boran Leontić at the University of Zagreb, Croatia [28–30]. This work has been done using the group's own Bi2212 single crystals as a further investigation of the important finding of Raffy et al. [25], who as early as in 1991 showed that the magnetoresistance of Bi2212 has a remarkable functional form suggesting that phase decoherence is induced in the "vortex liquid" state by a magnetic field . Namely, at a constant temperature the magnetoresistance shows no flux-flow-like behaviour, characteristic of conventional superconductors, but agrees extremely well with the dependence $R(H, T) \propto \Psi(1/2 + H(T)/H) + \ln(H/H(T))$, where $\Psi(x)$ is the digamma function and $\tilde{H}(T)$ the decoherence field. The above dependence is actually well known, it describes the Maki-Thompson [31] magnetic-field-induced de-phasing of nondissipative, coherent quasi-particle pairs above T_c in 2D systems (and Bi2212 is the most 2D of all HTS). Early reports on this behaviour [25–29] did not focus on the fact that originally the formalism was developed for fluctuations at temperatures $T>T_c$. More recently Leontić et al. have given [30] a semi-quantitative model which to some extent reconciles the original theory developed for $T>T_c$ with its applicability to the "vortex liquid" .

For the present paper it is important to note that because of the remarkably small number of overlapping pairs the phase of the superconducting order parameter is intrinsically weak in the "vortex liquid" state of HTS, and that dissipation occurs by its further weakening in higher magnetic fields. Although it is difficult to compare the thermodynamic and transport phenomena directly, the approach presented here is conceptually compatible with that of Leontić et al. Namely, it is the presence of large phase fluctuations that makes HTS so remarkably different from conventional superconductors.

3. Summary and conclusions

Many properties of high- T_c superconductors are different from those of conventional superconductors. The existence of a large fluctuation-dominated state called the "vortex liquid" is specific to high- T_c superconductors and is only partially understood. We have presented evidence that the thermodynamics of high- T_c superconductors in the "vortex liquid" state is successfully described in terms of 3D XY critical fluctuations. More detailed analysis of experimental data for the reversible magnetisation of YBCO(7- δ) and heat capacity of YBCO(7), which we took as a representative of high- T_c materials, lead us to an intriguing picture regarding the origin of the large critical fluctuations observed in these materials. In

this picture the high transition temperatures and decreased dimensionality of high- T_c compounds are not the primary cause of the observed fluctuation behaviour, although they could still be important in specific cases. On comparing the number of overlapping pairs N_p which build up the coherent superconducting state at $T=0$ and $H=0$ we have found a remarkable difference between conventional and high- T_c superconductors. In conventional superconductors the large value of N_p results in a superconducting phase which is strongly peaked around its mean value, and hence mean-field theory is applicable over most of the H-T phase diagram. In contrast, the small value of N_p in high- T_c materials causes an appreciable spread of the superconducting phase from its average even in the ground state. At elevated temperatures and magnetic fields this leads to strong fluctuations. The d-wave symmetry of the superconducting pairing in high- T_c materials could increase N_p significantly, but we believe that even in this case N_p still does not reach the value characteristic of conventional superconductors. Finally, we found similarities between our approach to the thermodynamics of high- T_c compounds and the description of the unusual magnetoresistance of high- T_c materials by Leontić et al. Both models suggest that there are fluctuations of the superconducting phase in its ground state, which are then manifested fully in the "vortex liquid" state.

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FLUKTUACIJE FAZE SUPRAVODLJIVOG UREĐENJA U OSNOVNOM STANJU KAO PREDUVJET JAKIH KRITIČNIH FLUKTUACIJA U VISOKOTEMPERATURNIM SUPRAVODIČIMA

Analiziramo reverzibilnu magnetizaciju i toplinski kapacitet YBa₂Cu₃O_{7−δ} u stanju "tekućine vrtloga", i nalazimo da su oba svojstva razumno dobro opisana 3D XY modelom kritičnih fluktuacija. U velikom području polja (H) i temperatura (T) , gustoća slobodne energije u stanju "tekućine vrtloga" ima osobito jednostavan oblik. To nas vodi do slike u kojoj je prisustvo kritičnih fluktuacija u visokotemperaturnim supravodičima neposredno povezano s vrlo malim brojem prekrivajućih Cooperovih parova pri $T = 0$ i $H = 0$, prije nego s niskom dimenzionalnošću ili visokim temperaturama.