On the consequences of the uncertainty principle on the superconducting fluctuations well inside the normal state

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Abstract

We first argue that the collective behaviour of the Cooper pairs created by thermal fluctuations well above the superconducting transition temperature, $T_C$, is dominated by the uncertainty principle which, in particular, leads to a well-defined temperature, $T_C^*$, above which the superconducting coherence vanishes. On the grounds of the BCS approach, the corresponding reduced-temperature, $\varepsilon_C \equiv \ln(T_C^*/T_C)$, is estimated to be around 0.55, i.e., above $T_C^* \simeq 1.7T_C$ coherent Cooper pairs cannot exist. The implications of these proposals on the superfluid density are then examined using the Gaussian-Ginzburg-Landau approximation. Then we present new measurements of the thermal fluctuation effects on the electrical conductivity and on the magnetization in different low- and high-$T_C$ superconductors with different dopings which are in excellent agreement with these proposals and that demonstrate the universality of $\varepsilon_C$.

74.20.-z Theories and models of superconducting state
74.20.De Phenomenological theories (two-fluid, Ginzburg-Landau, etc.)
74.40.+k Fluctuations (noise, chaos, nonequilibrium superconductivity, localization, etc.)
The behaviour of the superconducting fluctuations well inside the normal state, when the superconducting coherence length, $\xi(T)$, becomes of the order of its amplitude extrapolated at $T = 0$ K, $\xi(0)$, is a long-standing open problem which interest has been considerably enhanced by the discovery of the high temperature cuprate superconductors. Recent measurements of the superconducting fluctuations above $T_C$ in various high-$T_C$ and low-$T_C$ superconductors suggest the existence of a well-defined reduced-temperature, $\varepsilon_C \equiv \ln(T_C/T)$, above which the fluctuation effects vanish. These results were explained phenomenologically by introducing in the Gaussian-Ginzburg-Landau (GGL) approach a so-called “total-energy” cutoff, instead of the conventional momentum cutoff always used until now. The first aim of this Letter is to analyze the physical origin of such a total-energy cutoff. We will see here that it can be easily understood in terms of the uncertainty principle, which imposes a limit to the shrinkage above $T_C$ of the superconducting wave function when the temperature increases. Then, we will probe experimentally the universality of $\varepsilon_C$ by extending our previous measurements of the fluctuation effects in the high reduced-temperature region to very different low- and high-$T_C$ superconductors with different dopings, including for the first time a type-I superconductor.

The Heisenberg uncertainty principle was applied in 1953 to superconductors by Pippard to relate the size of the “wave packet formed by the electronic states”, $\xi_0$, to the normal-superconducting transition temperature, $T_C$. In terms of the BCS approach, the Pippard proposal suggests that the minimum size of a Cooper pair is of the order of $\xi_0$. By taking into account that the superconducting coherence length, $\xi(T)$, is the characteristic distance over which the density of Cooper pairs may vary, it may be concluded that even above $T_C$, where the Cooper pairs are created by thermal fluctuations, $\xi(T)$ must verify

$$\xi(T) \gtrsim \xi_0,$$

where $\xi_0$ is the actual superconducting coherence length at $T = 0$ K. Equation (1) provides then a constraint for the existence and behaviour of coherent Cooper pairs which must apply to any theoretical description of the superconducting state formation, including those non-BCS-like which are being proposed for cuprate superconductors. In fact, this condition may be directly obtained by applying the uncertainty principle to the Cooper pairs localized in a coherent volume. In other words, Eq. (1) accounts for the limits imposed by the uncertainty principle to the shrinkage, when the temperature increases, of the superconducting wave function.

The most outstanding consequence of Eq. (1) is that it naturally leads to a reduced-temperature, $\varepsilon_C$, determined by $\xi(\varepsilon_C) \simeq \xi_0$, above which coherent Cooper pairs cannot exist. The value of $\varepsilon_C$ will depend on each particular approach through the $\varepsilon$-dependence of $\xi(\varepsilon)$, where $\varepsilon \equiv \ln(T/T_C)$ is the reduced-temperature. For instance, by using the mean-field reduced-temperature dependence of the coherence length $\xi(\varepsilon) = \xi(0)\varepsilon^{-1/2}$, then $\varepsilon_C \simeq (\xi(0)/\xi_0)^2$. On the grounds of the BCS approach in the clean limit, $\xi(0) = 0.74\xi_0$, and so $\varepsilon_C \simeq 0.55$ in these superconductors. In fact, this
striking result probably also holds at a qualitative level in moderately dirty BCS superconductors (when $\ell \lesssim \xi_0$, $\ell$ being the mean free path of the normal carriers) because one may expect that both the Ginzburg-Landau (GL) coherence length amplitude and the actual superconducting coherence length at $T = 0$ K will be affected by impurities to a similar extent (see, e.g., Ref. [8], §7-2).

What consequences has Eq. (1) on the collective behaviour of the Cooper pairs at reduced-temperatures below $\varepsilon^C$? Probably the simplest way to address this question at a qualitative level is to introduce Eq. (1) in the conventional GGL framework, in spite that the latter formally applies only when $\varepsilon \ll 1$. In fact, another central point in this Letter will be to probe experimentally if the introduction of Eq. (1) in the GGL approximation extends its applicability up to $\varepsilon^C$. On the grounds of the GGL approach, Eq. (1) leads to

$$\xi^{-2}(\varepsilon) + k^2 \lesssim \xi_0^{-2},$$

where the left-hand term is the GGL total energy per superconducting carrier (in units of $\hbar^2/2m^*$, where $m^*$ is the superconducting carriers effective mass) of the fluctuating mode with wave vector $k$. Actually, as stressed before for Eq. (1), one may directly obtain Eq. (2) by naively applying the uncertainty principle to the spatial extension of the superconducting fluctuations and taking into account that at finite temperatures the energy balance when creating the fluctuations must include both the uncertainty principle and the thermal agitation. This crude reasoning is to some extent similar to the well-known textbook procedure used to estimate the minimum size of atoms by balancing the Coulomb and the Heisenberg localization energies. Note that for $\varepsilon \ll \varepsilon^C$ [and using $\xi(0)$ instead of $\xi_0$] Eq. (2) reduces to the widely used kinetic-energy or momentum cutoff condition $k^2 \leq c \xi^{-2}(0)$, where $c$ is a cutoff constant of the order of unity [1, 2, 5]. Note also that any cutoff condition is not built-in in the standard GGL equations, and it must be added to them “by hand”. Adequate extensions of the standard GGL equations to further Gor’kov perturbative orders could perhaps reproduce the total-energy cutoff, but we are not aware of any successful attempt of this.

The deep influence of Eq. (2) on the superconducting fluctuations in the normal state below $\varepsilon^C$ is well illustrated by the reduced-temperature dependence of the superfluid density, $\langle n_s(\varepsilon) \rangle$, on which any observable will depend. $\langle n_s(\varepsilon) \rangle$ is defined here on the grounds of the conventional GL approach as the spatially- and thermally-averaged squared modulus of the GL wave function [1, 2]:

$$\langle n_s(\varepsilon) \rangle \equiv \langle |\Psi|^2 \rangle = \sum_k \langle |\Psi_k|^2 \rangle,$$  

where $\Psi_k$ is the wave function of the fluctuating mode with wave vector $k$. On the grounds of the Gaussian approximation for the fluctuations of both the amplitude and phase of the order parameter, and for zero applied magnetic field, $\langle |\Psi_k|^2 \rangle$ is given
by the well-known result (see, e.g., Eq. (8.27) of Ref. [2]):

$$\langle |\Psi_k|^2 \rangle = \frac{2m^*\xi^2(0)}{\hbar^2} \frac{k_BT}{\varepsilon + k^2\xi^2(0)}.$$  \hspace{1cm} (4)

This familiar expression of the amplitude average of each fluctuating mode already illustrates the need of a total-energy cutoff: $\langle |\Psi_k|^2 \rangle$ increases (instead of decreasing) when the temperature increases well above $T_C$. This nonphysical behaviour appears for any $k$-value at reduced-temperatures above $\varepsilon + k^2\xi^2(0) = 1$. It cannot be eliminated, therefore, by the momentum cutoff condition. In contrast, a total-energy cutoff will remove, if $\varepsilon^C \leq 1$, this nonphysical behaviour (see also later).

From Eqs. (3) and (4) the superfluid density for any cutoff criterion may be directly calculated by just imposing on the $k$-summation the corresponding limits for the modulus of $k$. Such a limit is infinity for no cutoff, $\sqrt{(\varepsilon^C - \varepsilon)/\xi(0)}$ for the total-energy cutoff, and $\sqrt{c}/\xi(0)$ for the momentum cutoff. As it is well known, without cutoff the above integrations diverge at every temperature [1,2]. With the total-energy cutoff, we get for 2D-films of thickness $d$ (and also for extremely anisotropic layered superconductors with effective interlayer separation $d$):

$$\langle n_s(\varepsilon) \rangle_{E}^{2D} = m^* k_B T \frac{1}{2\pi^2 \hbar^2 d} \ln \left( \frac{\varepsilon^C}{\varepsilon} \right),$$ \hspace{1cm} (5)

and for 3D-bulk isotropic superconductors:

$$\langle n_s(\varepsilon) \rangle_{E}^{3D} = m^* k_B T \frac{\sqrt{\varepsilon}}{\pi^2 \hbar^2 \xi(0)} \left( \sqrt{\frac{\varepsilon^C - \varepsilon}{\varepsilon}} - \arctan \sqrt{\frac{\varepsilon^C - \varepsilon}{\varepsilon}} \right).$$ \hspace{1cm} (6)

The corresponding expressions under a momentum cutoff may be obtained from Eqs. (5) and (6) by replacing $\varepsilon^C$ by $c + \varepsilon$.

The reduced-temperature dependence of the superfluid density under these different cutoff conditions is shown in Fig. 1(a). We have chosen for representation $\varepsilon^C = 0.55$ as it results from the simplest estimate using the BCS theory in the clean limit (or, in the case of the momentum cutoff, $c = 0.55$). All the other parameters entering in Eqs. (5) and (6), including $\xi(0)$, are absorbed by the normalization chosen in the plot. As clearly illustrated by the figure, the conventional momentum cutoff predicts a nonphysical increase of the superfluid density at high $\varepsilon$’s, which is a consequence of the high-$T$ divergence of Eq. (4). In contrast, under the total-energy cutoff $\langle n_s(\varepsilon) \rangle$ vanishes at $\varepsilon = \varepsilon^C$. The corresponding fall-off is remarkably sharp, following in the close vicinity of $\varepsilon^C$ a power law-like behaviour with respect to $|\tilde{\varepsilon}|$, with $\tilde{\varepsilon} \equiv \ln(T/T^C) = \varepsilon - \varepsilon^C$: In the 2D superconductors $\langle n_s(\varepsilon) \rangle_{E}^{2D} \propto |\tilde{\varepsilon}|$, and in the 3D superconductors $\langle n_s(\varepsilon) \rangle_{E}^{3D} \propto |\tilde{\varepsilon}|^{1.5}$, in both cases with 5% or better accuracy for $|\tilde{\varepsilon}| \leq 0.1$ if $\varepsilon^C \geq 0.3$. Also noticeable is that such a rapid fall-off occurs when the coherence length competes with the size
of the individual Cooper pairs, as illustrated by the $\xi(\varepsilon)/\xi_0$ scale shown in Fig. 1(a) (whose numerical values correspond to the BCS clean limit): When $\xi(\varepsilon) > 2\xi_0$, which roughly corresponds to $\varepsilon \lesssim 0.15$, the consequences of the uncertainty principle on the superconducting fluctuations are inappreciable, and the momentum cutoff condition provides a good approximation to $\langle n_s(\varepsilon) \rangle$. However, for $2\xi_0 > \xi(\varepsilon) > \xi_0$ (corresponding to $0.15 < \varepsilon < \varepsilon^C$), i.e., when $\xi(\varepsilon)$ competes with the size of the individual Cooper pairs, the uncertainty principle will dominate the collective behaviour of these Cooper pairs.

The above results strongly suggest, therefore, that any superconducting fluctuation effect in the normal state will vanish above $\varepsilon^C$. However, probably the best way to probe these results is to extend our first experiments on the fluctuation effects in the high-$\varepsilon$ region [3,4] to different superconducting materials with different $\xi(0)$ values and also with various pairing states and maybe different pairing mechanisms. Some examples of the fluctuation effects on the magnetization [the so-called fluctuation magnetization, $\Delta M$] and on the electrical conductivity [the so-called paraconductivity, $\Delta \sigma(\varepsilon)$] obtained in different low- and high-$T_C$ superconductors are presented in Fig. 1(b). The experimental setups and procedures used in our present experiments are similar to those described in Refs. [3] and [4]. Other aspects, including the preparation of the samples, are going to be published elsewhere. Let us stress here, however, that the samples used in the paraconductivity experiments were single crystals and epitaxial thin films. The electrical resistivity versus temperature was measured with a four-terminal arrangement by using a conventional low-frequency (37 Hz) ac lock-in amplifier phase sensitive technique. To be able to determine $\Delta M$ in the high-$\varepsilon$ region, the magnetization measurements were performed with a SQUID magnetometer (Quantum Design, model MPMS) and by using quite big polycrystalline samples (with masses up to a few grams). Let us also note here that in analyzing all these measurements the background fitting region was always localized well above $\varepsilon^C$. So, the results presented in Fig. 1(b) are only moderately affected by the background uncertainties. These measurements cover almost two orders of magnitude in reduced magnetic fields $h \equiv H/H_{c2}(0)$, where $H_{c2}(0)$ is the upper critical magnetic field amplitude (extrapolated to $T = 0$ K). In fact, they cover both the zero field limit ($h \ll \varepsilon$) and the finite field regime. In this last case the $\Delta M(\varepsilon)_h$ data could be also affected by dynamic and non-local electrodynamical effects [1,2]. These examples also cover quite different superconductors, including moderately dirty (PbIn8%, i.e., Pb-8 at. % In) and clean (all the others), type I (Pb) and II (all the others), 3D-bulk low-$T_C$ (isotropic PbIn8% and Pb; moderately anisotropic MgB$_2$) and 2D-layered high-$T_C$ (the optimally-doped Bi-2212, i.e., Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ with $\delta \simeq 0.16$, and in the inset the underdoped LaSCO/0.1, i.e., La$_{1.9}$Sr$_{0.1}$CuO$_4$).

The hallmark of the suppression of the superfluid density by the total-energy cutoff, i.e., the rapid fall-off at a well-defined reduced-temperature, $\varepsilon^C$, of the superconducting fluctuation effects, is present in all the experimental curves in Fig. 1(b). The dashed and the solid lines in the main Fig. 1(b) and in its inset correspond to the GGL predictions in the zero-field limit under a conventional momentum cutoff and, respectively, a total-
energy cutoff. The corresponding formulas may be found in Refs. [3, 4]. Note that for our present purposes we must substitute \( c \) by \( \varepsilon_C \) in the expressions under the total-energy cutoff, and that in the case of the 2D-multilayered superconductors studied here we must multiply the corresponding single-layered expressions by \( N \), the number of superconducting layers per periodicity length. The \( \xi(\varepsilon)/\xi_0 \) scales in Fig. 1(b) and in its inset illustrate that the behaviour of the superconducting fluctuations is dominated by the localization energy when the superconducting coherence competes with \( \xi_0 \), the size of the Cooper pairs. The comparison between the two \( \Delta M(\varepsilon)_{\text{h}} \)-curves for PbIn8\% in Fig. 1(b) shows that the data for \( h = 0.1 \) are, below \( \varepsilon \simeq 0.1 \), appreciably affected by finite-field (or Prange) effects, but their \( \varepsilon_C \) still remains unchanged well within the experimental uncertainties. A more detailed comparison with the theoretical results in this finite-field regime will be published elsewhere.

The \( \varepsilon_C \)-values for all the compounds studied here, included those of Fig. 1(b) and also the ones measured in Refs. [3, 4], are presented in Fig. 2 as a function of \( \xi(0) \). In this figure, Tl-2223 and Y-123 stand for the optimally-doped Tl\(_2\)Ba\(_2\)Ca\(_2\)Cu\(_3\)O\(_{10}\) and Y\(_1\)Ba\(_2\)Cu\(_3\)O\(_{7-\delta}\) with \( \delta \simeq 0.05 \), LaSCO/0.14 and 0.25 stand for the optimally-doped La\(_{1.86}\)Sr\(_{0.14}\)CuO\(_4\) and, respectively, overdoped La\(_{1.75}\)Sr\(_{0.25}\)CuO\(_4\), and PbIn18\% stands for the Pb-18at.\%In alloy. As it may be clearly seen, \( \varepsilon_C \) varies less than a factor 3, while these measurements cover almost two orders of magnitude in coherence lengths, in amplitudes of the fluctuation effects at \( \varepsilon = 0.01 \) (on both \( \Delta M \) and \( \Delta \sigma \)), or (in the case of \( \Delta M \)) in applied reduced magnetic fields \( [h \equiv H/H_{c2}(0)] \) covers in these experiments the range \( 2 \times 10^{-3} \lesssim h \lesssim 2 \times 10^{-1} \). Therefore, these results provide strong experimental evidence that the suppression of the superconducting coherence above a well-defined temperature \( T_C \) in the normal state is due to a universal mechanism. These results do not exclude, indeed, a possible variation of \( \varepsilon_C \) in extremely dirty superconductors or in the high reduced magnetic field region (when \( h \to 1 \)), where different non-local and pair-breaking effects could appear [1, 2].

In conclusion, the experimental results and the analyses presented here suggest that the behaviour of the superconducting fluctuations at high reduced-temperatures is dominated by the uncertainty principle, which imposes a limit to the shrinkage of the superconducting wave function. These ideas provide a physical meaning to the “total-energy” cutoff heuristically introduced before [3, 4] to extend the applicability of the mean-field–like approximations to describe the superconducting fluctuations above \( T_C \) from \( \varepsilon \ll 1 \) up to \( \varepsilon_C \). It would be also interesting to extend these analyses to other superconductors, very in particular to the heavy fermions whose superconducting state coupling is based on antiferromagnetic spin fluctuations. Our present results may also have implications beyond the superconducting fluctuations issue. For instance, the striking fact that even in cuprates with different dopings (including the underdoped) \( \varepsilon_C \) takes the value that we have directly obtained by using the BCS relationship between \( \xi(0) \) and \( \xi_0 \) provides a constraint to any theoretical description of the superconducting state formation in these compounds, whose implications will deserve further analysis.
The possible implications of these results on the so-called zero-dimensional superconductors [1, 2], when $\xi(T)$ becomes bigger than the dimensions of the material in all directions, will also deserve further analysis. In fact, our present results suggest that the uncertainty principle constraint on the superconducting wave function will provide the last limit to the smallness of an isolated superconductor. They also suggest that the uncertainty principle must be taken into account when describing the short-wavelength thermal fluctuations around any phase transition with a quantum order parameter, so that the classical cutoff condition, $k \lesssim \xi^{-1}(0)$, must be substituted by a total-energy cutoff which takes into account the shrinkage at high reduced-temperatures of the quantum wave function.

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[5] Previous works on the high-$\varepsilon$ behaviour of the paraconductivity in LTSC and HTSC have been quoted in Refs. [3] and [4].

[6] In cuprate superconductors, the thermally activated Cooper pairs well inside the normal state may directly concern the formation of the superconducting state itself. See, e.g., Orenstein J. and Millis A.J., Science, 288 (2000) 468. See also, Batlogg B. and Varma C.M., Physics World, February issue 2000, p. 33.
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[8] See, e.g., de Gennes P.G., *Superconductivity of Metals and Alloys* (W.A. Benjamin, New York) 1966, sec. 1-4.

[9] The conventional GGL approach, including the $\varepsilon^{-1/2}$ dependence of $\xi(\varepsilon)$, is formally valid only in the $\varepsilon$-region $\varepsilon_{LG} \lesssim \varepsilon \ll 1$, where $\varepsilon_{LG}$ is the so-called Levanyuk-Ginzburg reduced-temperature [see, e.g., Vidal F. and Ramallo M.V., in *The Gap Symmetry and Fluctuations in High-$T_C$ Superconductors*, edited by J. Bok, et al. (Plenum, New York) 1998, p.443]. Nevertheless, the conventional GGL approximation regularized through a momentum cutoff was unsuccessfully used by different authors beyond the $\varepsilon \ll 1$ condition [9]. Our present results strongly suggest that the limitation of the shrinkage of the superconducting wave function is the dominant effect when $\varepsilon$ approaches $\varepsilon^C$, and that both the GGL approach under a total-energy cutoff and the mean-field critical exponent of $\xi(\varepsilon)$, $x = -1/2$, remains qualitatively valid even up to $\varepsilon^C$. Other attempts to explain beyond the conventional momentum cutoff approach the superconducting fluctuations in the short-wavelength region, also unsuccessful at high reduced-temperatures, are commented in the footnote 19 of Mosqueira J. et al., *Phys. Rev. B*, **65** (2002) 174522.

[10] See, e.g., Feynman R.P., Leighton R.B. and Sands M., *The Feynman Lectures on Physics, Quantum Mechanics, Volume III* (Addison-Wesley, Reading) 1965, sec. 2-4.
\[ \frac{\xi(\epsilon)}{\xi_0} = \ln \left( \frac{T}{T_c} \right) \]

\[ n_s(\epsilon) = n_s(\epsilon = 0^+) \]

\[ \xi(\epsilon) \]

\[ \xi(\epsilon) / \xi_0 \]

\[ \frac{\langle \eta_s(\epsilon) \rangle}{\langle \eta_s(\epsilon = 0^+) \rangle} = 10^{-2} \]

\[ \epsilon = \ln \left( \frac{T/T_c}{T_c} \right) \]

Figure 1: (a) Reduced-temperature dependence of the superfluid density above \( T_c \), calculated on the grounds of the GGL approach for zero applied magnetic field and in the 3D and 2D limits under both the momentum and the total-energy cutoffs. (b) Some examples of the reduced-temperature dependence of the fluctuation-induced magnetization in the zero field limit (open symbols) and in the finite field regime (solid symbols), and of the fluctuation-induced electrical conductivity, measured in different superconductors. In the anisotropic superconductors, the magnetic field is applied perpendicular to the in-plane directions. The dashed and solid lines in the panel (b) and its inset correspond to the GGL calculations in the zero field limit under a momentum cutoff and, respectively, a total-energy cutoff. The \( \xi(\epsilon) / \xi_0 \) scales in both (a) and (b), whose values correspond to the BCS clean limit, illustrate that the behaviour of superconducting fluctuations is dominated by the uncertainty principle when \( \xi(\epsilon) \) competes with \( \xi_0 \), the size of the Cooper pairs.
Figure 2: Values of $\varepsilon^C$, the reduced-temperature at which the superconducting fluctuations above $T_C$ vanish, obtained from measurements of the thermal fluctuation effects on the magnetization (circles) or on the electrical conductivity (squares) in different superconducting materials, as a function of their Ginzburg-Landau coherence length amplitude $\xi(0)$. The data points for Tl-2223, Bi-2212, Y-123, MgB$_2$, PbIn18% and PbIn8% were taken from Refs. [3] and [4]. In the case of the anisotropic superconductors, $\xi(0)$ corresponds to the in-plane coherence length amplitude. The magnetization data always correspond to $h \leq 0.2$. The error bars represent well all the experimental uncertainties, included those associated with the background estimation. The line $\varepsilon^C = 0.55$ corresponds to the simplest theoretical estimate using the BCS model in the clean limit.