Comment on “Diamagnetism and Cooper pairing above $T_c$ in cuprates”

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It is shown that the magnetization rounding measured by L. Li and coworkers above the superconducting transition in optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$ crystals under magnetic fields up to 14 T [Phys. Rev. B 81, 054510 (2010)] may be explained at a phenomenological level in terms of the mean field Gaussian-Ginzburg-Landau (GGL) approach for layered superconductors. This result challenges the claims of L. Li and coworkers, who write “[…] we are observing the phase-disordering mechanism, rather than Gaussian mean-field fluctuations”, but it is in full agreement with earlier magnetization measurements by different authors in optimally-doped YBa$_2$Cu$_3$O$_{7-\delta}$ under lower magnetic fields. The adequacy of the mean-field Ginzburg-Landau descriptions is further confirmed when analyzing the magnetization data reported below $T_c$ by L. Li and coworkers.

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In Ref. 1 it is claimed that the magnetization rounding measured in that work above $T_c$ (the so-called precursor diamagnetism) in several families of high temperature cuprate superconductors (HTSC) provides thermodynamic evidence of the so-called phase fluctuation scenario, a conclusion also stressed in a Viewpoint on that paper by Kivelson and Fradkin. In the case of the prototypical optimally doped (OPT) YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO), in Ref. 2 it is noted that it “[…] should be the least susceptible to the phase-disordering mechanism for the destruction of long-range phase coherence at $T_c$ (and hence the best candidate for Gaussian fluctuations among cuprates). However, the torque measurements reveal that $T_c$ in OPT YBCO is also dictated by large phase fluctuations.” To support these conclusions, Li and coworkers provide in Fig. 8 of their paper magnetization data up to magnetic fields of 14 T. Then, without presenting any quantitative comparison with earlier Gaussian-Ginzburg-Landau (GGL) approaches for the precursor diamagnetism in superconductors, they write that the “[…] significant diamagnetism surviving to intense fields, at temperatures up to 40 K above $T_c$ is strong evidence that we are observing the phase-disordering mechanism, rather than Gaussian mean-field fluctuations”.

In principle, the credibility and relevance of the conclusions summarized above could be strongly enhanced by the well-known experimental advantages of the OPT YBCO crystals, which allow a reliable extraction of the precursory diamagnetism. Also, as stressed in Refs. 1 and 2, the torque magnetometry technique used in these measurements allows the use of large applied magnetic fields. However, in this Comment we show that, contrary to the claims of Refs. 1 and 2, these interesting experimental results may be easily explained at a quantitative level in terms of the GGL approach for multilayered superconductors confirming similar conclusions obtained by different authors from measurements under lower applied magnetic fields. Let us also note already here that, in our opinion, the opportunity of our present Comment is enhanced by the fact that the questionable conclusions of Ref. 1 are being cited by not few authors as a strong experimental support of the phase fluctuation scenario for the HTSC at present a still open and debated issue of the physics of these superconductors.

Some of the arguments presented in Ref. 1 against the GGL scenario for the precursor diamagnetism in OPT YBCO may be in fact easily refuted without the need of detailed calculations. For instance, when analyzing the magnetization versus temperature curve presented in their Fig. 8(b), these authors write: “[…] the curve at 14 T reveals the existence of the large fluctuating diamagnetism associated with the vortex liquid. This point [...] highlights the major difference between the diamagnetism in hole-doped cuprates and low-$T_c$ superconductors. In the latter, increasing $H$ in the fluctuation regime above $T_c$ rapidly squelches the (Gaussian) fluctuation signal altogether.” However, as the in-plane coherence length amplitude, $\xi_{ab}(0)$, of OPT YBCO is around 1 nm, the reduced magnetic field, $h \equiv H/[(\phi_0/2\pi\mu_0)^{2/3}]$, corresponding to 14 T is near 0.05 (in this expression $\phi_0$ is the flux quantum and $\mu_0$ the vacuum permeability). By taking a look at Fig. 8.5 of Tinkham’s textbook one may already conclude that also in the conventional metallic low-$T_c$ superconductors such a relatively weak reduced field has an almost unobservable influence on their precursor diamagnetism, in full agreement with the results of the GGL approach in presence of finite magnetic fields (the so-called Prange regime). In fact, the results of Soto and coworkers on the precursor diamagnetism in Pb-In alloys, published eight years ago, provide a direct experimental demonstration that even the precursor diamagnetism onset, that in the zero-field limit is located at temperatures about 1.7$T_c$, is little affected by reduced magnetic fields below 0.1.

The adequacy of the GGL approach for multilayered superconductors to account for the results of Ref. 1 on OPT YBCO may be easily confirmed at a quantitative level by just comparing these data with the theoretical expression for the fluctuation-induced magnetization for fields applied perpendicular to the $ab$ layers (denoted $M_{ab}$, as in Ref. 3) proposed in Ref. 14 for layered superconduc-
tors under a total-energy cutoff:

\[ M_d(\varepsilon, h) = -f k_B T \frac{h}{2\phi_0 s} \frac{B_{LD}}{\varepsilon} \left( 1 + \frac{B_{LD}}{\varepsilon} \right)^{-1/2} \]  

(2)

The data points in Fig. 1(a) correspond to those presented in Fig. 8(a) of Ref. [1] for the \(M_d\) dependence on the applied magnetic field above \(T_c\). The solid lines correspond to Eq. (1), evaluated by using the \(T_c\) value proposed in Ref. [1] (92 K), the coherence length amplitudes in Ref. [7], \(\xi_{ab}(0) = 1.1 \text{ nm}\) and \(\xi_c(0) = 0.1 \text{ nm}\), and \(f = 0.5\) which is well within the one observed in twinned OPT YBCO crystals. As it may be seen, the agreement is reasonably good, taking into account the unavoidable uncertainties associated with the determination of both the corresponding background contributions and the \(T_c\). The agreement extends also to the as-measured magnetization (denoted \(M_{\text{eff}}\), as in Ref. [1]) versus temperature curve presented in Fig. 8(b) in Ref. [1], which corresponds to an applied magnetic field of 14 T. The analysis of this curve on the grounds of Eq. (1) is presented in our Fig. 1(b). The solid line was obtained by adding the background proposed in Fig. 8(b) of Ref. [1] (dotted curve) and the fluctuation-induced magnetization calculated from Eq. (1) by using the same parameter as before for Fig. 1(a). As one may appreciate, the agreement is excellent even in the high reduced-temperature region, including the temperature for the onset of fluctuation effects. Our present results are also consistent with earlier analyses of Lee, Klemm and Johnston and of Ramallo, Torrón and Vidal, although in their case without penetrating the high reduced-temperature region. Surprisingly, in Refs. [1] and [2] it was not presented any quantitative analysis of their experimental results on the grounds of the GGL approaches. In addition, earlier results obtained under lower field amplitudes by other authors, as those published in Refs. [4] to [11], were also overlooked.

Although both the experimental results and the, always qualitative, analyses on the diamagnetism in OPT YBCO presented by Li and coworkers are centered on the behaviour above \(T_c\), for completeness we present here a detailed quantitative analysis, in terms of the Ginzburg-Landau scaling proposed by Ullah and Dorsey of the data below \(T_c\) provided by these authors. This approach corresponds to the lowest-Landau-level (GL-LLL) approximation for 3D superconductors. For applied magnetic fields up to 5 T, the adequacy of this GL-LLL approach to explain the diamagnetism around \(T_c\) observed in OPT YBCO was first demonstrated by Welp and coworkers and then by other authors. All these results were also overlooked by Li and coworkers.
Let us note also that the seemingly anomalous non-monotonic profile of the $M_d$ vs. $H$ isotherms that may be observed below $T_c$ in Fig. 8(a) of Ref. 1 is compatible with the conventional (GL-like) fluctuation scenario: in YBCO the slope of the upper critical field close to $T_c$ is $\mu_0 dH_{c2}/dT|_{T_c} \approx 3 T/K$. Then, $\mu_0 H_{c2}$ just below $T_c$ is in the Tesla range. In particular, at 91, 90, and 89 K it is about 3, 6, and 9 T respectively. A simple inspection of the corresponding $M_d(H)$ reveals a change from concave to convex just at these field values. This change may be then attributed to a transition from the mixed state (in which $|M_d|$ decreases with $H$) to the normal state at higher fields (in which, due to conventional fluctuation effects, $M_d$ is not null and behaves like in the isotherms above $T_c$).

We must also remark here that the differences between the diamagnetism behavior in the OPT YBCO and in a low-$T_c$ superconductor may be easily attributed to the differences between their superconducting parameters. For instance, in the case of the NbSe$_2$ compared in Ref. 28 with the HTSC, $T_c$ is one order of magnitude smaller than in OPT YBCO, while the coherence lengths are one order of magnitude larger. Then, for similar reduced temperatures and magnetic fields, the fluctuation magnetization above $T_c$ is a factor $\sim$300 smaller as may be easily estimated from the GGL approach. A thorough comparison between the diamagnetism above $T_c$ in HTSC and low-$T_c$ superconductors, and on the corresponding influence of the presence of $T_c$ inhomogeneities, is presented in Ref. 30 and in the references therein.

In summary, we have analyzed at a quantitative level, in terms of the mean-field GGL approach for multi-layered superconductors, the experimental results of Li and coworkers on the diamagnetism above $T_c$ in OPT YBCO. These analyses lead to conclusions just opposite to those stressed in Ref. 1 (and Ref. 2), but provide a nice quantitative confirmation, up to magnetic field amplitudes of 14 T, of earlier conclusions of different authors later extended to high-reduced temperatures through the introduction of a total-energy cutoff. The adequacy of the extended GGL scenario to account at a phenomenological level for the precursor diamagnetism (including its onset) of this prototypical cuprate superconductor. Although not addressed in Ref. 1, for completeness we have also analyzed quantitatively, in terms of the Ginzburg-Landau scaling proposed by Ullah and Dorsey, the data below $T_c$ provided in that work. This comparison demonstrates the adequacy of the GL scaling for the data located in the critical region around $T_c(H)$, extending up to 14 T the earlier conclusions for lower field amplitudes. As these analyses above and below $T_c$ have been performed by using the same parameter-values, our results represent a stringent check of consistency of the validity of the mean-field-like GL scenario for the fluctuation diamagnetism observed above and below the superconducting transition in OPT YBCO. It would be useful to extend these analyses to the results of Ref. 1 on other HTSC families with different doping.
levels. However, as we have already stressed when commenting other results of these authors, non-optimally-doped compounds will be more affected by chemical disorder (intrinsic to their non-stoichiometric nature) than OPT YBCO, and the superconducting fluctuation effects will be entangled with those associated with the corresponding $T_c$-inhomogeneities.

Finally, let us stress that the results summarized here try to contribute, by presenting quantitative analyses on the grounds of the mean-field-like GL approaches, to clarify the debate on the phenomenological descriptions of the superconducting transition in HTSC, but do not pretend to bridge that debate even in the case of the prototypical OPT YBCO. In fact, the majority of the theoretical works published in the last few years on that issue, including some of the most influential, propose different unconventional (non-Ginzburg-Landau) scenarios, the most popular being the one based on phase-disordering. Even in the case of the conventional GL-like scenarios, the possible presence of different, direct or indirect, superconducting fluctuation contributions is a long standing issue still open at present.

However, our present Comment further stresses that, in any case, before adopting any superconducting fluctuation scenario for the HTSC it will be crucial to perform thorough quantitative analysis of the experimental results, in some cases taking into account the presence of chemical disorder.

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11. The OPT YBCO crystals are particularly well suited to probe the superconducting fluctuations around $T_c$, mainly due to the availability of large single crystals with a high chemical and structural quality. This minimizes the possible effect of $T_c$-inhomogeneities which could mimic the fluctuation effects (see main text). In addition, its normal-state magnetization is linear in an extended temperature region, allowing a reliable background extraction. See W.C. Lee and D.C. Johnston, Phys. Rev. B 41, 1904 (1990), and also Refs. 6 and 8 and references therein.
12. However, it is worth noting that well below $T_c$ the analysis of the magnetic torque in terms of the magnetization perpendicular to the ab layers may be affected by the angular slippage from the crystal c-axis of the reversible magnetization vector, see J. Mosqueira, R.I. Rey, A. Wahl, and F. Vidal, Phys. Rev. B 84, 134504 (2011).
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14. As explained in detail in Refs. 8 and 9 in HTSC with two superconducting planes in their periodicity length (which in YBCO equals the c-axis lattice parameter), the effective periodicity length in the GGL scenario becomes $s = c$ only if the Josephson couplings $\gamma_1$ and $\gamma_2$ between planes with interdistances $d_1$ and $d_2$ differ among them by orders of magnitude (formally when $\gamma_1/\gamma_2 \to \infty$). Instead, for $BL_D \sim 0.15$ as in OPT YBCO, $s = c/2$ is an excellent approximation for any $\gamma_1/\gamma_2 \lesssim 30$ (to be compared with $d_2/d_1 \sim 2.5$ in that compound). For further confirmations of the need to consider in HTSC each CuO$_2$ plane per periodicity length as an independent superconducting plane, see e.g., D.C. Ling, G. Yong, J.T. Chen, and L.E. Wenger, Phys. Rev. Lett. 75, 2011 (1995); M.V. Ramallo, Europhys. Lett. 65, 249 (2004).
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