Unusual Interplay between Superconductivity and Field-Induced Charge Order in $YBa_2Cu_3O_y$

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We present a detailed study of the temperature (T) and magnetic field (H) dependence of the electronic density of states (DOS) at the Fermi level, as deduced from specific heat and Knight shift measurements in underdoped $YBa_2Cu_3O_y$. We find that the DOS becomes field independent above a characteristic field $H_{\rm DOS}$, and that the $H_{\rm DOS}(T)$ line displays an unusual inflection near the onset of the long-range 3D charge-density wave order. The unusual S shape of $H_{\rm DOS}(T)$ is suggestive of two mutually exclusive orders that eventually establish a form of cooperation in order to coexist at low T. On theoretical grounds, such a collaboration could result from the stabilization of a pair-density wave state, which calls for further investigation in this region of the phase diagram.

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There is now compelling evidence that high-T_c superconductivity in cuprates competes with a charge-density wave (CDW) over a substantial range of carrier concentrations [1–7]. This CDW is most prominent in underdoped $YBa_2Cu_3O_y$, where it causes a major reconstruction of the Fermi surface [8–13]. In zero or low magnetic fields, the CDW order is two dimensional and short ranged, but it becomes both long ranged and correlated in all three dimensions (3D) in high magnetic fields [1,2,6,14–16]. Several consequences of the competition between the CDW and superconducting orders have already been highlighted, including the strong field (H) and temperature (T) dependence of the CDW in the superconducting state [1,2,4,5, 14–16], a diminution of T_c [17], and a severe reduction of the upper critical field $H_{c2}(T=0)$ [18,19].

Here, our specific heat and spin susceptibility measurements reveal an unforeseen effect of this competition: upon cooling, the temperature dependence of the field $H_{\rm DOS}(T)$ above which the electronic density of states (DOS) at the Fermi level saturates displays a clear inflection when the field-induced long-range CDW order develops [6,14–16]. $H_{\rm DOS}(T)$ then sharply increases below ∼10 K, tending towards $H_{c2}(0)$ for $T \to 0$ [19,20]. This results in an unusual S shape of $H_{\rm DOS}(T)$, which is suggestive of two mutually exclusive orders that eventually establish a form of cooperation in order to coexist at low temperature. These results raise the question of whether the nature of the superconducting state is altered in order to allow for this collaboration, in which case the low-temperature phase might correspond to the predicted pair-density wave (PDW) order [21–32].

The magnetic field dependence of the electronic specific heat of the superconducting phase, $C_{\rm es}(T,H) = C_{\rm p}(T,H) - \gamma_{\rm R}T - C_{\rm ph}(T)$ has been measured in underdoped $YBa_2Cu_3O_y$ single crystals [33] with a number of doped holes per planar Cu $p = 0.098$ ($T_c = 57.3$ K), 0.109 ($T_c = 61.5$ K) and 0.119 ($T_c = 65.6$ K), with γ_R being the residual Sommerfeld coefficient, obtained by extrapolating C_p/T towards $T = 0$ (for $H = 0$), and $C_{\rm ph}(T)$ being the phonon contribution (see the Supplemental Material [34]). As shown in Figs. 1(a) and 1(b) (see also Fig. S2 in Ref. [34]), $C_{\rm es}/T$ becomes field independent above a characteristic field $H_{\rm DOS}(T)$. For $H \leq H_{\rm DOS}(T)$, $C_{\rm es}/T$ decreases approximately linearly, except at 2.5 K, for which a more complex behavior is found. As previously reported by Kemper *et al.* [36], $C_{\rm es}/T$ displays a possible $\sqrt{H}$ dependence at low field and very low temperature (characteristic of d-wave superconductors), but the presence of a Schottky anomaly (which has been subtracted from the data [34]) is hindering any detailed discussion of this field dependence.

Quantum oscillations (QOs) are observed in $C_{\rm es}/T$ for a $p = 0.109$ crystal for $\mu_0 H \geq 25$ T and $T \leq 6$ K [Fig. 2(a)]. The oscillations, here observed down to a field value

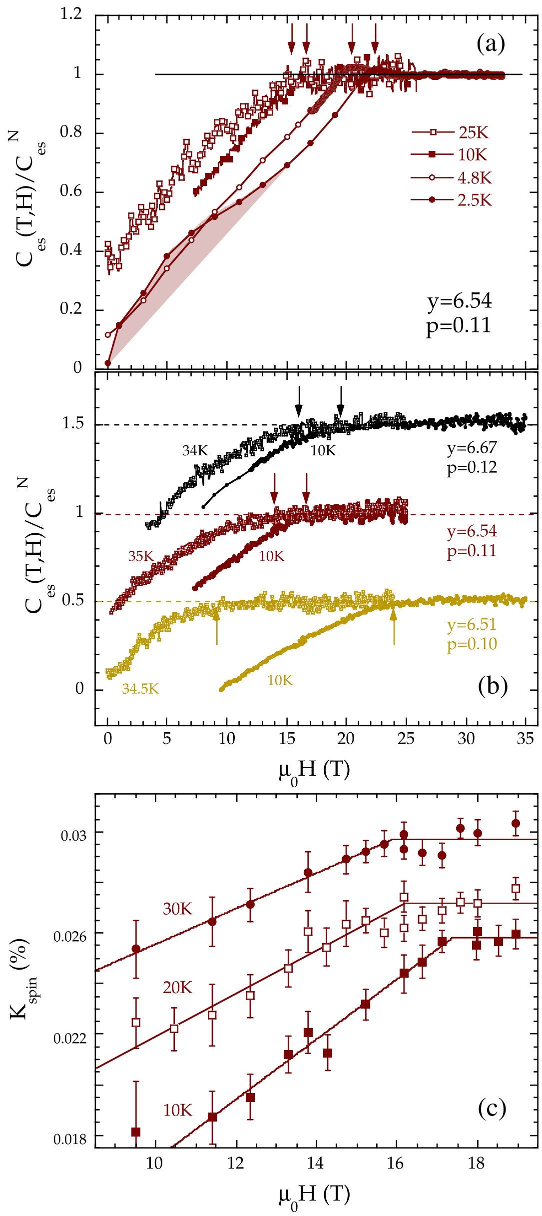

FIG. 1. (a),(b) Magnetic field dependence of the electronic specific heat, $C_{\mathrm{es}}(T,H)/T$, normalized to its value at the field H_{DOS} above which data saturate (see arrows), C_{es}^N/T. [The curves in panel (b) have been shifted for clarity, by $+0.5$ for $p = 0.119$ and by -0.5 for $p = 0.098$.] At 2.5 K, the data markedly deviate from a linear dependence [shaded area in panel (a); see the text for details]. (c) Field dependence of the ^{17}O Knight shift for $p = 0.109$. Solid lines are fits to the data, as explained in the main text (see Ref. [34] for the determination of the error bars).

significantly smaller than that in Ref. [37], can be well described by the Lifshitz-Kosevich (LK) formula (see Ref. [34] for details). As this formula changes sign for $T/\mu_0H \sim 0.11 \times m_e/m^*$, the π phase shift of the oscillations observed for $T = 2.3 \pm 0.3$ K and $B \sim 31$ T (see the vertical solid line) directly implies that the effective mass $m^* = (1.5 \pm 0.2)m_e$ for $p = 0.109$. The (solid) green line is then obtained by introducing the frequency ($F = 530$ T) and warping term ($t_w = 15$ T) used in Ref. [37]. As shown, the QOs abruptly disappear below ∼25 T (for all temperatures), and a small change of the parameters [$F = 520$ T, $t_w = 22$ T; see the red lines in Fig. 2(a)] predicts almost undetectable oscillations below ∼25 T, suggesting that this dampening could be due to the presence of a node around 25 T. However, torque measurements indicate that there is no node in this field range, and that QOs can persist well below 25 T [38]. Note that this "onset" field is close to the field H_{scat}, below which the thermal conductivity κ_{xx} abruptly decreases [18], marking the onset of strong *scattering* by vortices, which are also expected to lead to a significant dampening of the QOs.

In two dimensions, the Sommerfeld coefficient in the normal state γ_N is directly related to m^* through $\gamma_N \sim 2.9 \times (m^*/m_e)$ mJ/mol K^2/pocket (in two-layer systems), and, assuming that the Fermi surface is constituted of one single (electron) pocket per $\mathrm{CuO_2}$ layer, one expects $\gamma_N = 4.4 \pm 0.5$ mJ/mol K^2 in very reasonable agreement with the C_{es}/T value obtained for $H \geq H_{\mathrm{DOS}}$: $C_{\mathrm{es}}^{\mathrm{N}}/T = C_{\mathrm{es}}(T \to 0, H \geq H_{\mathrm{DOS}})/T = 4.8 \pm 0.6$ mJ/mol K^2 [see Fig. 2(b) and Fig. S1 in Ref. [34]]. Note, however, that we did not take into account the residual specific heat γ_R, which hence seems not to be directly related to the reconstructed FS. A similar $C_{\mathrm{es}}^{\mathrm{N}}/T$ (i.e., m^*) value is measured for $p = 0.119$, but $C_{\mathrm{es}}^{\mathrm{N}}/T$ increases to 7.5 ± 0.5 mJ/mol K^2 for $p = 0.098$, indicating that the effective mass sharply increases to 2.60 ± 0.15, in good agreement with the change in m^* deduced from quantum oscillations in transport and skin depth measurements [12,13] [gray circles in Fig. 2(b)].

We now turn to the saturation of the specific heat above H_{DOS}, the field for which C_{es}/T reaches its maximum value in either temperature or field sweeps [see also Fig. S1(b) in Ref. [34]]. At low temperature ($T \simeq 2$–3 K), earlier C_p [20] and NMR Knight shift [19] measurements show that the saturation of the DOS coincides with $\mu_0H_{\mathrm{scat}} \equiv \mu_0H_{c2}(0) \sim 24$ T [18] [see Fig. S4(a) in Ref. [34]]. What do we expect at higher temperatures? As is well known in high-T_c cuprates, thermal fluctuations cause the vortex lattice to melt into a vortex liquid at temperatures well below the mean-field transition field H_{c2}^{MF}. As the line tension of vortices vanishes at the melting transition [39,40], the normal and vortex-liquid states are the same phase and are connected by a smooth crossover [39,40]. Nevertheless, the specific heat is still expected to present a smeared anomaly in the vicinity of the former H_{c2}^{MF} line, where most of the ordering energy comes out and the DOS reaches its normal-state value. We indeed find that a clear saturation of $C_p(H)$ persists upon heating [Figs. 1(a) and 1(b)], but we observe that H_{DOS} rapidly decreases with temperature (Figs. 2 and 3).

Note that an "overshoot" is observed in the field dependence of C_{es}/T at low temperatures [see Fig. 1(a)

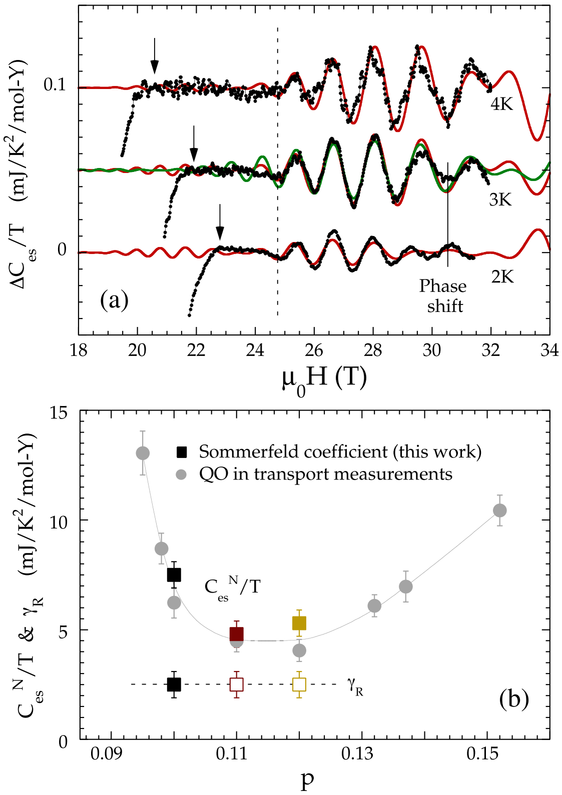

FIG. 2. (a) Magnetic field dependence of the specific heat at low temperature ($p = 0.109$; a smooth polynomial background has been removed from the data, and each temperature has been shifted by 0.05 for clarity; arrows indicate the $H_{\rm DOS}$ values). The clear quantum oscillations can be very well described by the standard Lifshitz-Kosevich formula with $F \sim 530$ T and a warping term of $t_w \sim 15$ T (red lines), or with $F \sim 520$ T and $t_w \sim 22$ T (green line). (b) $C_{\rm es}^{\rm N}/T$ (closed squares) and residual C_p/T values (γ_R, open squares) as a function of the doping rate, together with the effective mass deduced from quantum oscillations in transport and skin depth measurements (gray circles) [12,13].

and Fig. S4(a) in Ref. [34]]. This "overshoot" is reminiscent of the mean-field specific heat jump at $H_{c2}^{\rm MF}$. Indeed, in the absence of thermal fluctuations, a specific heat jump $\Delta C_p/T = -\mu_0(\partial M/\partial T)_H(dH_{c2}^{\rm MF}/dT)$ is observed at $H = H_{c2}^{\rm MF}$ (M being the reversible magnetization in the superconducting state), and a smeared "overshoot" is still expected to be observed in the presence of thermal fluctuations if $dH_{\rm DOS}/dT \neq 0$ (and the slope of the magnetization is changing rapidly close to $H_{\rm DOS}$). The observation of such an overshoot at $T/T_c \sim 1/30$ is very unusual, as $dH_{c2}^{\rm MF}/dT \to 0$ at low temperature, but here it is thermodynamically consistent with the strong temperature dependence of $H_{\rm DOS}$ below 10 K.

Upon further heating, we find that this decrease of $H_{\rm DOS}(T)$ is followed by a plateau with $\mu_0 H_{\rm DOS} \simeq 15$ T in the 10–30 K range—that is, in the vicinity of the onset field of long-range CDW order for both $p = 0.109$ and $p = 0.119$ [Figs. 3(b) and 3(c)], as detected by sound velocity [6,41], x-ray scattering experiments [14–16], and thermal Hall conductivity κ_{xy} measurements [42]. Finally, above ~30 K, $H_{\rm DOS}$ further decreases (tending towards zero for $T \to T_c$), giving rise to an unexpected S shape of the $H_{\rm DOS}(T)$ line. Note that for $p = 0.119$ the $H_{\rm DOS}(T)$ line well agrees with the line below which the intensity of the (short-range) CDW diffraction peaks becomes field dependent [open diamonds in Fig. 3(c)], marking the onset of the superconducting phase [4]. For $p = 0.098$, in which the CDW onset field is much larger [41] and the CDW much weaker [43], we do not observe such a plateau. Nevertheless, a change of slope of $H_{\rm DOS}(T)$ remains visible [see Fig. 3(a)] on entering into the CDW phase.

In order to confirm these results, we have measured the spin part of the ^{17}O Knight shift $K_{\rm spin}$ in a similarly doped $p = 0.109$ single crystal [2,19,44]. $K_{\rm spin}$ is proportional to the uniform spin susceptibility $\chi_{\rm spin} = \chi_{\rm spin}(q = 0, \omega = 0)$ at planar sites: $K_{\rm spin} = K_{\rm total} - K_{\rm orb} = A/(g\mu_B)\chi_{\rm spin}$, where $K_{\rm total}$ is the measured total Knight shift, $K_{\rm orb}$ the orbital shift, A the hyperfine coupling constant, g the Landé factor, and μ_B the Bohr magneton. As discussed in Ref. [19], the field dependence of $\chi_{\rm spin}$ in the superconducting state is expected to reflect changes in the DOS, even though $\chi_{\rm spin}$ may not be related to the DOS in a simple way. As shown in Figs. 1(c) and 3(b), Knight shift measurements unambiguously confirm a saturation of the DOS above $H_{\rm DOS}$. The $K_{\rm spin}(H)$ data have been fitted by a linear increase up to $H_{\rm DOS}$ and to a constant value beyond. In order to avoid any arbitrary choice for $H_{\rm DOS}$, we have fitted the whole field range, with $H_{\rm DOS}$ itself being a fitting parameter. As shown in Fig. 3 [see also Figs. S4(a) and S4(b) in Ref. [34]], the fits lead to $H_{\rm DOS}$ values in agreement with those obtained in C_p measurements. In particular, the NMR data confirm the nearly constant $\mu_0 H_{\rm DOS} \simeq 16$–17 T for 10 K $\lesssim T \lesssim$ 30 K. Note that $K_{\rm spin}$ is not expected to present any overshoot, clearly indicating that the S shape of the $H_{\rm DOS}$ line is not related to the presence of such an overshoot in C_p. Even though the maximum accessible field was limited to 20 T in this NMR experiment, it is important to stress that the saturation is beyond error bars. Furthermore, our data in much higher fields ($\mu_0 H \simeq 28$ T) show identical $K_{\rm spin}$ values (see Fig. S5 in Ref. [34]), which demonstrates that the saturation of $K_{\rm spin}$ has indeed been reached at ~17 T.

In principle, the constant DOS could be due to entering into a new type of gapless superconducting state, but we fail to see any theoretical support for such a scenario. Furthermore, our low-T data exclude the possibility that the saturation results from an accidental compensation between a decrease of the DOS due to the opening of a CDW gap and an increase of the DOS in the still-existing superconducting phase. If this were the case, $C_{\rm es}/T$ would saturate near $\mu_0 H_{\rm CDW} \simeq 17$ T at 2.5 K for $p = 0.109$, whereas we observe that it saturates close to $\mu_0 H_{c2}(0) \simeq 24$ T. Finally, as the vortex melting line lies significantly below $H_{\rm DOS}$ (for $T \neq 0$, see Ref. [20] for

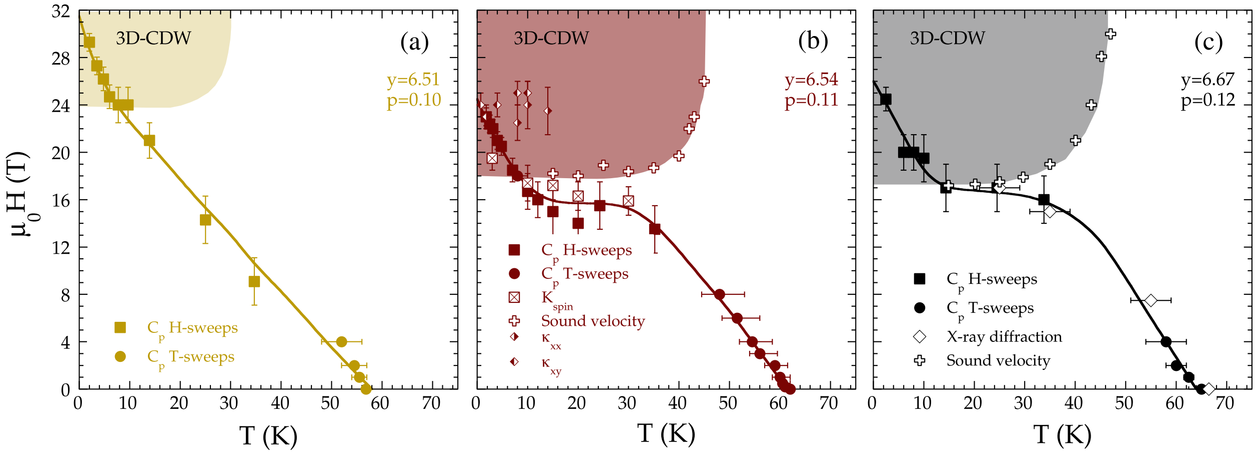

FIG. 3. H_{DOS} versus temperature for the indicated doping contents. The solid circles and squares have been derived from T (see Refs. [20,34]) and H sweeps [see Figs. 1(a) and 1(b)] of the specific heat, respectively, and the crossed squares have been deduced from $K_{\mathrm{spin}}(H)$ [see Fig. 1(c)]. As shown, a clear "plateau" is observed in $H_{\mathrm{DOS}}(T)$ for $p = 0.109$ [panel (b)] and $p = 0.119$ [panel (c)] in the vicinity of the onset of the long-range 3D-CDW [41,42] (open crosses and shaded areas), highlighting the interplay between those two competing orders. For $p = 0.098$ [panel (a)], a change of slope is observed when H_{DOS} crosses H_{CDW}. The field H_{scat} marking the onset of scattering by vortices, as deduced from thermal conductivity measurements (diamonds [18,42]), has also been reported for $p = 0.11$. Open diamonds in panel (c) correspond to the fields below which the intensities of the CDW diffraction peaks decrease [4]. Lines are guides to the eyes.

$p = 0.109$ as an example), the saturation of the DOS cannot be related to the melting of the vortex solid. Thus, our measurements indicate that there is an intrinsic saturation of the DOS for $H \geq H_{\mathrm{DOS}}(T)$. Note that superconducting fluctuations persist well above H_{DOS} (for $T \neq 0$, see the sketch in Fig. 4). The plateau apparently reflects a separation between the field scale deduced from probes sensitive to the DOS and the one deduced from probes sensitive to vortex scattering. The two field scales merge as $T \to 0$, but a detailed discussion of the onset of those fluctuations is beyond the focus of the present Letter. The phase diagrams of Figs. 3(b) and 3(c) leave little doubt that the unusual S shape of $H_{\mathrm{DOS}}(T)$ directly results from the influence of three-dimensional (3D) long-range CDW order on superconductivity (and not from, e.g., a disorder-driven Griffiths superconducting phase [45,46]).

The presence of this plateau suggests that the high-field CDW and superconductivity initially repel each other, as if they were mutually exclusive orders that cannot coexist [47]. However, the upswing of the $H_{\mathrm{DOS}}(T)$ line below $\sim$10 K suggests that superconductivity eventually finds a way to accommodate the presence of the 3D long-range CDW order. Our measurements do not offer a direct clue on what microscopically characterizes the "collaborative" state between CDW and superconducting orders for $T \leq$ 10 K and $\mu_0 H \gtrsim 15$ T. However, because unusual effects are seen in the DOS, it is likely that some characteristics of the superconducting state have changed. Some sort of collaboration between CDW and superconducting orders was also proposed in Ref. [48]. However, details of the phase diagram differ from ours, as we do not see any signature of a second vortex solid phase in our C_p and NMR data (see also Ref. [19]).

In this context, it is interesting to note that recent theoretical works [21,23,25,27], motivated by

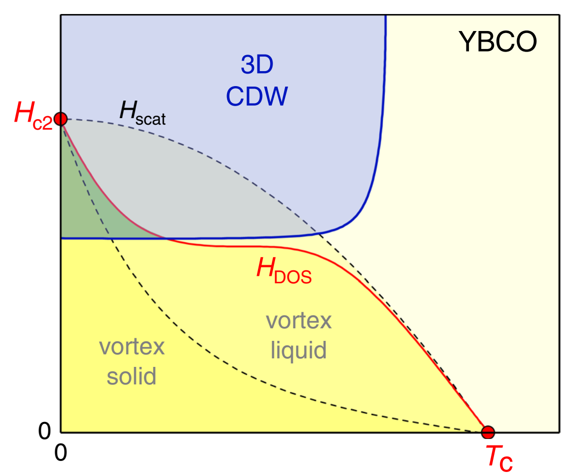

FIG. 4. Sketch of the $H-T$ diagram of underdoped $YBa_2Cu_3O_y$, emphasizing the interplay between the superconducting and CDW orders. Specific heat and Knight shift measurements show that the density of states at the Fermi level reaches its normal-state value above H_{DOS} (see Fig. 1). Different shades of yellow tentatively depict the intensity of local superconducting fluctuations, with emphasis on the field scale H_{scat} deduced from thermal conductivity measurements [18,42]. The greenish region corresponds to the $H_{\mathrm{CDW}} \leq H \leq H_{\mathrm{DOS}}$ field range, in which superconductivity might be impacted by the presence of 3D CDW order.

scanning-tunneling microscopy of the vortex cores [26], suggested that high magnetic fields may reveal a pair-density-wave (PDW) state in which spatial variations of the superconducting- and CDW-order parameters are intertwined (see Ref. [24] for a review). While it has been proposed that the 3D CDW is actually a consequence of a primary PDW order [23], the above-proposed interpretation of our data seems to fit more naturally with the view that the PDW order is stabilized only in the coexistence region by the presence of an independent 3D CDW order [25,27]. However, in order to discriminate between theories, our results should be compared with predictions for the field dependence of the DOS from different theoretical models.

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