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# How ‘pairons’ are revealed in the electronic specific heat of cuprates

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Understanding the thermodynamic properties of high- $T_c$  cuprate superconductors is a key step to establish a satisfactory theory of these materials. The electronic specific heat is highly unconventional, distinctly non-BCS, with remarkable doping-dependent features extending well beyond  $T_c$ . The pairon concept, bound holes in their local antiferromagnetic environment, has successfully described the tunneling and photoemission spectra. In this article, we show that the model explains the distinctive features of the entropy and specific heat throughout the temperature-doping phase diagram. Their interpretation connects unambiguously the pseudogap, existing up to  $T^*$ , to the superconducting state below  $T_c$ . In the underdoped case, the specific heat is dominated by pairon excitations, following Bose statistics, while with increasing doping, both bosonic excitations and fermionic quasiparticles coexist.

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*Introduction* More than thirty years after the discovery of cuprate superconductivity by Bednorz and Müller [1], the challenge persists to describe their transport, spectroscopic and thermodynamic properties in a coherent and satisfactory way. In particular, the thermodynamic properties are of high interest giving access to the fundamental excitations of the system at equilibrium.

The pioneering studies of the specific heat [2] showed the inherent difficulty to separate the electronic from the phonon contribution in a variety of cuprates near the superconducting transition (see the review of Fisher et al.[3] and references therein). However, the innovative work of Loram et al. [4, 5] showed convincingly that the electronic part of the specific heat  $C_e(T)$  is highly unconventional and deviates markedly from the BCS behavior [6].

Below  $T_c$ , low temperature measurements have demonstrated the d-wave symmetry of the order parameter [7–12]. The global shape of  $C_e(T)$  is strongly doping dependent and electronic signatures extend well beyond the critical temperature, especially in the underdoped case (see Fig. 1). The pseudogap (PG) in the normal state, revealed by RMN [13, 14] tunneling [15] and angle-resolved photoemission spectroscopy (ARPES) [16, 17] experiments, is also evidenced by the specific heat [18, 19]. Its relation to the superconducting state (preformed pairs, coexisting or competing orders) is still debated. Also, it remains to be clarified whether the pseudogap is present all along the superconducting (SC) dome or ends at a quantum critical point for  $p \approx 0.2$  above which (i.e. in the overdoped regime) a Fermi liquid behavior and BCS superconductivity would be recovered.

A key question is whether these features in the spe-

cific heat can be well understood in the framework of a ‘preformed pair’ model, wherein  $T^*$  is the onset of pair formation, and not a competing gap as in [19]. In this article, we use the pairon model to calculate the electronic specific heat of cuprates, as a function of temperature ( $T$ ) and hole doping ( $p$ ). The model allows to describe the main features observed in the specific heat experiments as a function of  $T$  and  $p$ . We show that the superconducting transition is governed by pairon excitations following Bose statistics throughout the phase diagram. Whereas such excitations dominate at low doping, there is a coexistence of both pairon and quasiparticle excitations in the overdoped regime.

*Pairon model* We have recently proposed that superconductivity in cuprates can be explained by the formation of a new quantum object. The pairons are bound pairs of holes which form in their local antiferromagnetic environment [20]. They naturally reconcile antiferromagnetism and Cooper pairing, two normally antagonistic phenomena. The binding energy  $\Delta_p$  is dictated by the spin exchange interaction  $J$  and its doping dependence is linear with  $p$ , in agreement with many experiments including ARPES and tunneling [21]. Condensation of pairons arises due to their mutual interaction, giving rise to a collective quantum state with superconducting properties.

At odds with the BCS case, the critical temperature is not directly proportional to the gap energy but rather to the pair-pair interaction  $\beta_c \simeq 2.2 k_B T_c$ . This interaction parameter depends on both the density of pairons, which follows the doping, and on their binding energy  $\Delta_p$ , the

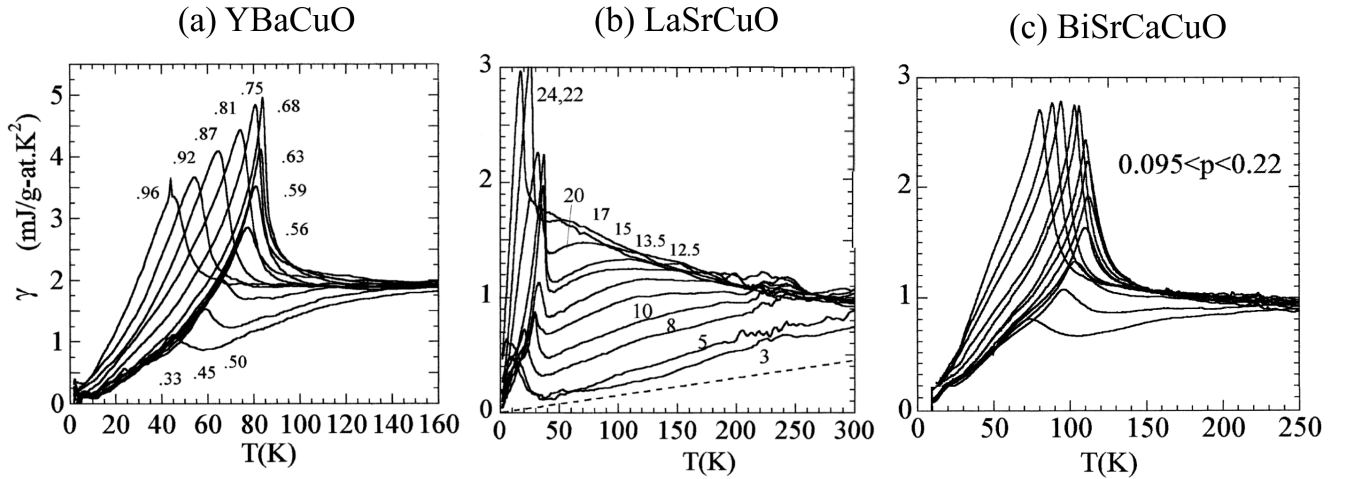


FIG. 1: (Color online)  $\gamma$  coefficient measured as a function of temperature for three different cuprates,  $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{6+\delta}$  (a),  $La_{2-x}Sr_xCuO_4$  (b) and (20% Pb doped, 15% Y doped)  $Bi_2Sr_2CaCu_2O_{8+\delta}$  (c), by Loram et al. [19].

latter having the characteristic temperature scale  $T^*$ :

$$\beta_c = C \frac{(p - p_{min})}{p_c} \Delta_p \quad (1)$$

where  $p_{min} \approx 0.05$  is the value at the SC onset and where  $C = 0.9$  for  $Bi_2Sr_2CaCu_2O_{8+\delta}$ , as deduced from fits of tunneling data [20]. The critical doping  $p_c \approx 0.27$  is directly related to the pairon size [20]. While the gap energy is analogous to the Cooper pairing between two fermions, the quantity  $\beta_c$  arises from an additional four fermion term in the hamiltonian [22] which couples different pair states. This explains why a pairing gap can persist above  $T_c$ , without SC coherence, being linked to the higher temperature  $T^*$ .

In our picture, the increase of the pairon density with doping opposes the decrease of their binding energy, giving rise to the dome shape for the critical temperature. Both parameters depend on a single energy scale, the antiferromagnetic exchange energy  $J$  [20] and one length scale  $\xi_{AF}$  [23]. The pairon model hamiltonian allows to calculate the spectral function as well as the density of states in excellent agreement with the experimental tunneling spectra [24] as well as ARPES, as a function of temperature and doping [25].

*Elementary excitations* In conventional superconductors, superconductivity arises due to electron-electron interaction via phonon exchange resulting in an energy gap  $\Delta_p$  in the excitation spectrum. As shown in the energy diagram Fig. 2a, the elementary excitations from the ground state are quasiparticles of energy  $E_k$ , i.e. exotic fermions following Fermi-Dirac statistics. This results in the familiar quasiparticle density of states with a temperature dependent gap, the order parameter, which vanishes at the critical temperature.

In cuprates, as mentioned previously, condensation of

pairons is due to pairon-pairon interaction which leads to a collective quantum state having long range SC properties. The total ground state energy (per pair)  $E_c$  is given by  $E_c = -\Delta_p - \beta_c$ , where the first term is analogous to BCS and the second term arises due to the mutual interaction between pairons (see the energy diagram Fig. 2b). At zero temperature, all pairons belong to the superconducting ground state with energy  $E_c$ . As the temperature increases, pairons are excited out of the condensate ground state with an occupation number given by Bose-Einstein statistics.

It is these thermal excitations of pairons that break long range SC coherence and not quasiparticle excitations. As a result the condensation energy weakens with temperature and, at the critical temperature, the effective interaction energy is zero. This is precisely the pseudogap state where incoherent pairons, with energy gap  $\Delta_p(T_c)$ , survive (as indicated in Fig. 2b) whereas superconductivity no longer exists. Further rising temperature implies the familiar pair breaking into quasiparticles thus leading to the normal state near  $T^*$ .

Thus, contrary to BCS where only fermionic excitations are responsible for the destruction of the SC order, here the bosonic character of pairons is the key effect. This conclusion was already discussed in the context of tunneling and ARPES spectra [24, 25] and now will be borne out in the present study of the entropy and the specific heat.

In a Bose picture, the density  $n_c$  of condensed pairons is given by

$$n_c(T) = n_0 - A \int_{\delta}^{\Delta_0} P_0(\varepsilon_i) f_{BE}(\varepsilon_i) d\varepsilon_i \quad (2)$$

where  $A$  is a normalization coefficient,  $n_0$  is the density of pairons at  $T = 0$  ( $\propto p/2$ ),  $P_0(\varepsilon_i)$  is the density of pairon

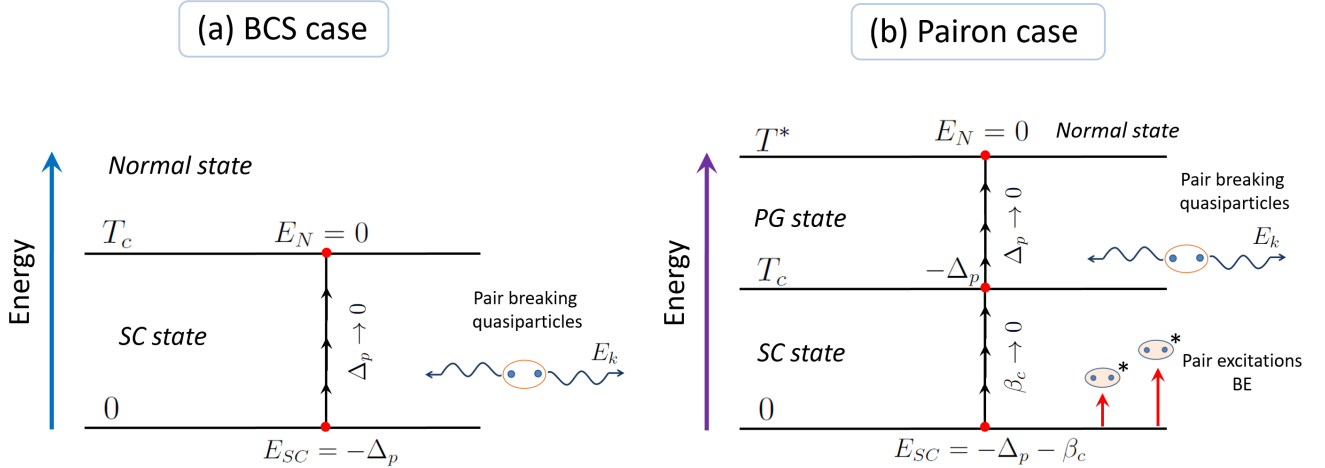


FIG. 2: (Color online) a) Energy diagram in the BCS case. The elementary excitations are quasiparticles (fermionic type). b) Energy diagram in the pairon model where two kinds of excitations are present, bosonic (pairon excitation) and fermionic (quasiparticles) types. While in the BCS case the condensate is described by a single parameter  $\Delta_p$ , two energy scales,  $\Delta_p$  and  $\beta_c$ , are needed to describe the cuprates.

excited states, and  $f_{BE}(\varepsilon) = 1 / (\exp(\frac{\varepsilon - \mu_b}{k_B T}) - 1)$  is the Bose-Einstein distribution. As in our previous work, we have chosen a Lorentzian form for the density of pairon excited states:  $P_0(\varepsilon_i) = \sigma_0^2 / [(\varepsilon_i - \beta_c)^2 + \sigma_0^2]$ , where  $\sigma_0$  is the width of the distribution. Although this distribution is written differently from our previous work, it is in fact a change in variable, as detailed below.

The upper limit of integration  $\Delta_0$  is the maximum energy of a pairon while the lower cut-off is a gap in the excitation spectrum of the pairons ( $\delta \sim 2\text{meV}$ ) [22]. Since we assume a Bose-Einstein like condensation at the critical temperature,  $\mu_b = 0$  below  $T_c$  while  $\mu_b(T)$  must conserve the total number of pairons above  $T_c$ . The constraint that  $n_c(T = T_c) = 0$  imposes the value of the normalization coefficient  $A$ .

Once the condensate is completely depleted, the total energy of the system is  $\sim \Delta_p(T_c)$ , the pseudogap state. In the underdoped regime, up to the optimal doping, the antinodal gap varies very little below  $T_c$ , and consequently the total energy at the transition is nearly equal to the antinodal gap  $\Delta_p(T = 0)$ . This clearly illustrates the difference between cuprates and conventional BCS superconductors: the gap is not the order parameter.

*Entropy calculation* In a conventional superconductor, the elementary excitations of the condensed state are quasiparticles arising from the dissociation of Cooper pairs. Here the increase of the entropy originates from two fundamental processes, namely pairon excitations following Bose statistics and the dissociation of pairons into quasiparticles following Fermi-Dirac statistics. These two fundamental processes are included in

the concise expression of the total entropy  $S$ :

$$S = \sum_i n_i(\varepsilon_i, T) S_i(\varepsilon_i, T) \quad (3)$$

where  $n_i$  is the density of excited pairons with energy  $\varepsilon_i$  and  $S_i$  is the associated entropy term. For every pairon excitation energy  $\varepsilon_i$  there is a set of binding energies  $\Delta_i$ , associated with Cooper pairs decaying as quasiparticles of energy  $E_k^i = \sqrt{\varepsilon_k^2 + \Delta_i^2}$ . We therefore write:

$$S_i(\varepsilon_i, T) = \sum_{\vec{k}} S(E_k^i, T) \quad (4)$$

The constitutive equation between the pairon energy and the associated Cooper pairs,  $\varepsilon_i(\Delta_i)$  is needed. Although phenomenological, we have used with success the equation  $\varepsilon_i = \Delta_i - \Delta_p(T, \theta)$ . This equation gives the excitation energy of pairons with respect to the associated Cooper pairs of energy  $\Delta_i$ .  $\Delta_p(T, \theta)$  is assumed to be the average value of the excitation spectrum, at the angle  $\theta$  on the Fermi surface.

In our previous work, we chose  $\Delta_p(T, \theta) = \Delta_p(T) \cos(2\theta)$  (except for some angular corrections, due to the spatial extension of the pairons [23]), mainly present in the underdoped regime), with  $\Delta_p(T)$  given by the BCS formula (brown curve, upper right of Fig. 3). However, such an expression leads to a discontinuity of the specific heat at  $T^*$  which is absent in the experiments. A precise fit to the data is obtained using a smooth function which softens the variation of  $\Delta_p(T)$ , as illustrated in Fig. 3 (upper right, blue curve).

In Eq. 4,  $S(E_k^i, T)$  is the usual entropy expression for

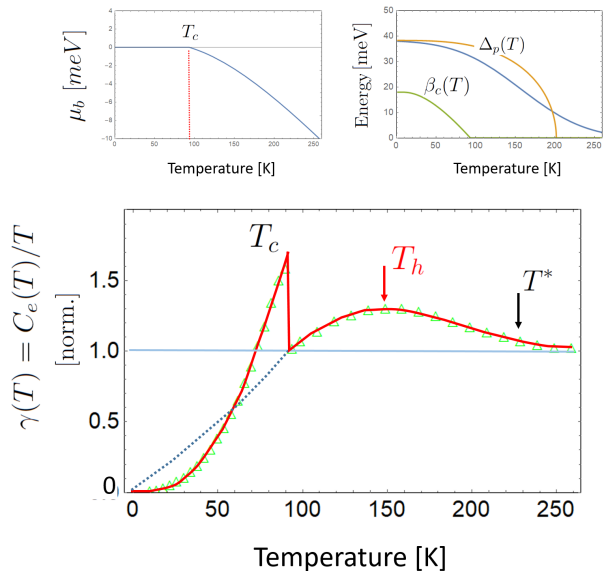


FIG. 3: (Color online) Lower curve: Electronic specific heat as a function of temperature at optimum doping, where are indicated the three characteristic temperatures  $T_c$ ,  $T_h$  and  $T^*$ . Upper curve, left: chemical potential  $\mu_b(T)$  of the pairons. Upper curve, right: Condensate energy  $\beta_c n_c(T)$  which vanishes at the critical temperature (red curve); Dependence of the gap function in the antinodal direction used in our calculations (blue curve) as a function of temperature compared to a gap function following a BCS-type dependence (orange curve).

fermions with energy excitation spectrum  $E_k^i$ .

$$S(E_k^i, T) = -2k_B [f(E_k^i) \ln(f(E_k^i)) + (1 - f(E_k^i)) \ln(1 - f(E_k^i))] \quad (5)$$

with  $f(E)$  the Fermi-Dirac function. Summing over all  $\vec{k}$  values, gives

$$S_i(\varepsilon_i, T) = -k_B N_n \int_0^\infty d\epsilon_k \int_0^{2\pi} d\theta S(E_k^i, T), \quad (6)$$

where  $N_n$  is the normal density of states at the Fermi level. Finally, we replace the discrete sum over the states by an integral  $\sum_i \approx \int d\varepsilon_i P_0(\varepsilon_i)$ , and get

$$S = \int_\delta^{\Delta_0} d\varepsilon_i P_0(\varepsilon_i) n_i(\varepsilon_i, T) S_i(\varepsilon_i, T), \quad (7)$$

with  $P_0(\varepsilon)$  being the density of pairon excited states.

*Results for the entropy and the specific heat* The entropy as a function of temperature for different doping values is plotted in Fig. 4. At high temperature ( $T \gg T^*$ ), the entropy  $S(T)$  is linear as a function of temperature, which is characteristic of metallic behavior. This result can be seen as a limit of Eq. 4 where the gap energy vanishes and  $S_i$  equals the free electron entropy,

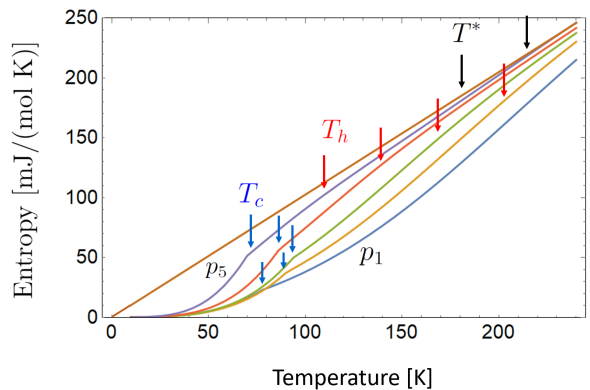


FIG. 4: (Color online) Entropy as a function of temperature for different doping values ( $p_1 = 0.11$ ,  $p_2 = 0.135$ ,  $p_3 = 0.16$ ,  $p_4 = 0.185$ ,  $p_5 = 0.21$ ). Red, blue, and black arrows respectively indicate the position of the critical temperature  $T_c$ , the hump temperature  $T_h$ , and the pseudogap temperature  $T^*$ .

and using particle conservation  $\sum_i n_i(\varepsilon_i, T) = n_0$ . At lower temperature ( $T < T^*$ ),  $S(T)$  departs from the linear asymptote and stays below the metallic value. This is a direct consequence of the formation of pairons which, as a result of increasing order, lowers the entropy relative to the metallic case.

Furthermore, a smooth but marked change of the  $S(T)$  curvature is observed at the temperature  $T_h$ , located between  $T^*$  and  $T_c$ . It corresponds to a maximum of quasiparticle excitations which occurs at the *inflection* point of the gap function  $\Delta_p(T)$  (see Fig. 3, right upper curve). At even lower temperature, the progressive condensation of pairons is manifested by the lowering of the entropy with an abrupt change in the slope of  $S(T)$  at  $T_c$ . Below  $T_c$ , the entropy decreases rapidly to zero and vanishes at absolute zero.

The specific heat is obtained directly from the entropy according to the relation  $C_e(T) = T \frac{dS}{dT}$ . The standard quantity  $\gamma(T) = C_e(T)/T$  is plotted in Fig. 3, lower curve, for optimum doping. At low temperature,  $\gamma(T)$  is very small and then increases rapidly due to pairon excitations and reaches a maximum at the transition  $T_c$ , where all pairons are excited out of the condensate,  $n_c(T_c) = 0$ , as illustrated in Fig. 3, right upper curve. The sharp peak in  $\gamma(T)$ , followed by the discontinuity, is therefore associated with the disappearance of the condensate.

At higher temperature, a smooth hump is observed at  $T_h$  corresponding to a maximum of quasiparticle excitations, seen as the point of inflection in the entropy. This effect has been discovered and studied by Matuzaki et al. [26] and is discussed below. Finally, a constant is recovered for  $T \gg T^*$  corresponding to the metallic behavior. Note that the return to the normal state, corresponding to the vanishing of the pseudogap state, is smooth and

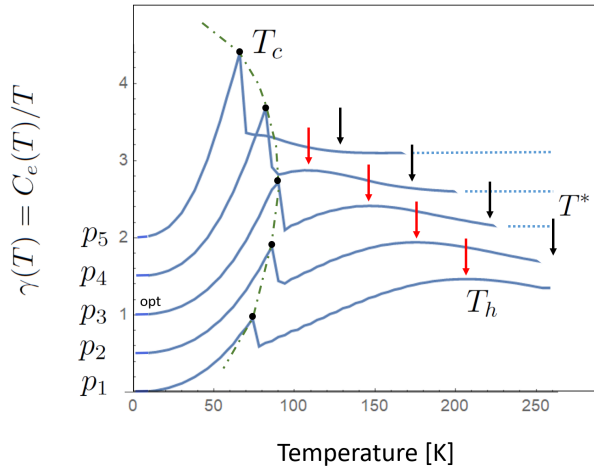


FIG. 5: (Color online) Lower curve: Electronic specific heat as a function of temperature for different doping values all over the phase diagram ( $p_1 = 0.11$ ,  $p_2 = 0.135$ ,  $p_3 = 0.16$ ,  $p_4 = 0.185$ ,  $p_5 = 0.21$ ).  $T_c$ ,  $T_h$  and  $T^*$ , are respectively indicated by the dashed curve and red and black arrows. The dotted horizontal line is the asymptotic limit  $\gamma(T) = \gamma_N$  reached in the normal state. The curves have been shifted vertically for clarity.

continuous.

Returning to the question of the SC state, the dashed curve in Fig. 3 is obtained for incoherent pairons with no superconducting transition. Examining  $\gamma(T)$  one sees clearly the conservation of the relative area compared to the background. While BCS superconductivity emerges from the normal state (wherein  $\gamma(T) = \gamma_N$ ,  $\gamma_N$  being the gamma coefficient in the normal state), here it emerges from an incoherent PG state of preformed pairons. We stress that this incoherent state is responsible for the unconventional background of  $S(T)$  and  $C_e(T)$  observed in the experiments.

We have also calculated  $C_e(T)$  for various doping values along the phase diagram, as summarized in Fig. 5. In agreement with experiments we find that the shape of the curves is quite different from underdoped to overdoped regimes. While the parameters underlying each curve vary monotonically, there is no simple homothetic relation between the curves. It is evident that  $T_c$  follows a dome shape in agreement with the phase diagram, see Fig. 6. With increased doping,  $T_c$  and  $T^*$  approach each other as deduced from ARPES or tunneling spectroscopy experiments [21]. Finally, they merge at the maximum doping  $p_c = 0.27$ , the critical point in our phase diagram.

The third characteristic temperature  $T_h$  [26] is a prominent feature of the present calculation. As seen in Fig. 3, for optimum doping, the hump is roughly midway between  $T^*$  and  $T_c$ . At this doping, the quasiparticle formation is quite distinct from the SC transition at  $T_c$ . However for larger  $p$ , the hump moves down towards  $T_c$

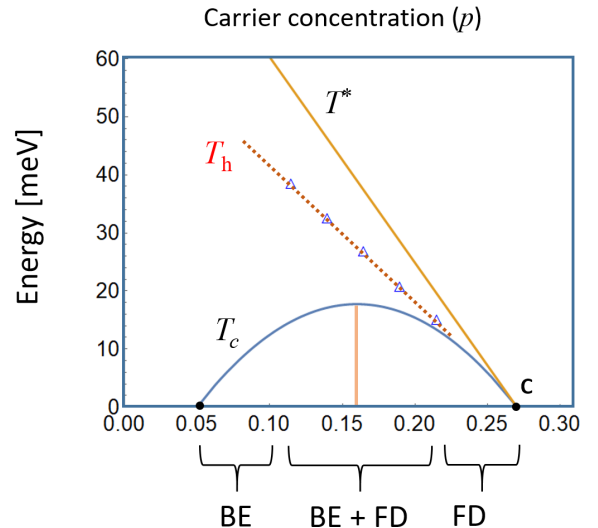


FIG. 6: (Color online) Phase diagram showing the superconducting dome  $T_c$  ( $\propto \beta_c$ ), the pseudogap temperature  $T^*$  ( $\propto \Delta_p$ ) as a function of hole doping  $p$ . The specific heat ‘hump’ temperature  $T_h$ , scaling perfectly with  $T^*$ , is also plotted. The type of excitations which dominate in the different doping regime is indicated below. Bose-Einstein (BE) excitations in the underdoped regime, Bose-Einstein and quasi-particle fermionic excitations (BE+FD) around the optimum doping, and fermionic type excitations (FD) in the highly overdoped region.

and merges progressively with the ‘bosonic’ discontinuity at  $T_c$ . In the overdoped regime,  $T_h$  is no longer visible. This behavior is particularly evident in experimental results on  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (Fig. 1) and also in (20% Pb and 15% Y doped)  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , although the effect is less pronounced.

*Discussion* It is remarkable that the entropy and the electronic specific heat can be described by the same set of equations (Eq. 3, 4 and 5), regardless of the carrier concentration all along the phase diagram. Although the detailed shape of  $C_e(T)$  varies significantly as a function of doping, quite surprisingly, it can be explained by the same underlying mechanisms. Moreover, the conclusions are in agreement with our previous work, where we deduced a universal phase diagram involving the most relevant parameters, the pairon binding energy and their mutual interactions [20]. The fundamental length scale  $\xi_{AF}$  and energy scale  $J$ , the antiferromagnetic exchange energy, are at the heart of the wide range of phenomena as seen in tunneling and ARPES. Such a simplified picture is now in qualitative agreement with the specific heat.

In our view, there is no abrupt transition as a function of doping from underdoped to overdoped sides, but a continuous evolution. The apparent change of behavior seen in the specific heat, just as in tunneling and ARPES

experiments, reflects the dual nature of the excitations of the system, fermionic and bosonic type. Above  $T_c$ , the characteristic temperature  $T_h$  corresponds to the point where the change of population of quasiparticle excitations reaches a maximum, which occurs at the inflection point of  $\Delta_p(T)$ . Above this temperature, as illustrated in Fig. 3,  $\Delta_p(T)$  is rapidly decreasing.

It is close to the node that quasiparticle excitations from decaying pairons dominate. This is the Fermi-arc contribution included in our calculation, being directly proportional to  $T_c/T^*$ , as we have shown in [25]. In the underdoped regime, for  $T < T_c \ll T_h$  pairon excitations dominate in entropy and the specific heat (Fig. 4 and Fig. 5). However, the composite nature of pairons is key. As the doping increases,  $T_h$  decreases, then more quasiparticle excitations are present at lower temperature. Finally, on the higher doping range, ( $T_h \sim T_c$ ), there is a coexistence of both types of excitations.

The hump in the specific heat curve is also associated with the number of the pairon energy states. On the lower-temperature side of the hump, the specific heat increases because of a large increase of entropy, according to Eq.4 as more pairon excited states become populated. Interestingly, this effect is similar to a Schottky anomaly. However, in a standard Schottky anomaly, the decrease of the specific heat on the right side of the hump is the result of the high population of the excited energy levels. In our case, it is due to quasiparticle decay of pairons. The hump in the specific heat can thus be considered as an unconventional Schottky anomaly.

The present study shows that the temperature dependence of the thermodynamic quantities can be well described by assuming an average gap  $\Delta_p(T)$ , describing the pairon excited states, which decreases very smoothly and vanishes at the typical temperature  $T^*$  (Fig. 3). This mean field gap is not due to an hypothetical competing order and shows no sign of discontinuity neither is it due to SC fluctuations, the energy scale being too large. Rather, the gap originates from excited pairs above  $T_c$ , in agreement with the conclusion raised by Wen et al. [27] from specific heat measurements in  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$ . Therefore in our model, there is no phase transition associated with the characteristic temperature  $T^*$ .

However, the shape of  $\Delta_p(T)$  does imply that a residual pairon density persists above  $T^*$ . This residual density is likely to be too small to have any significant effect on the tunneling and ARPES spectra. Nevertheless, it remains an interesting and open question as to their possible detection above  $T^*$ .

In their stimulating work, Curty et al.[28] also address the calculation of the specific heat and the interpretation of the phase diagram. They consider an attractive Hubbard-like hamiltonian with a d-wave local pairing on adjacent sites and then determine thermodynamic properties in a Ginzburg-Landau/Monte Carlo

approach. Their work reveals that the superconducting state emerges from an incoherent phase of pairs. The present work is in qualitative agreement with a number of their conclusions. Based on the idea of incoherent preformed pairs, we see that two energy scales, without a discontinuity of behavior for a wide range of doping, fits the specific heat and entropy. Curty et al. also stress the absence of a quantum critical point under the dome. This is also the case in our work, as illustrated in Fig. 6, since the pseudogap persists for all  $p$  values, even in the overdoped regime up to the maximum value  $p_c$ .

*Conclusion* We have shown that the thermodynamic properties of cuprates can be very well described by the formation of pairons, bound pairs of holes in their anti-ferromagnetic environment. Superconductivity emerges from an incoherent state of pairons, the pseudogap state, as a result of their mutual interactions. Two fundamental temperature scales can be clearly identified in specific heat experiments. They correspond to the mean binding energy of pairons  $\Delta_p$ , related to the pseudogap temperature  $T^*$ , and the interaction energy  $\beta_c$ , which directly proportional to the superconducting critical temperature. The peak at the critical temperature in the specific heat is explained in terms of pairon excitations following Bose-Einstein statistics which deplete the condensate. In the underdoped regime, pairon excitations qualitatively dominate in the specific heat up to the critical temperature. As the doping increases, the contribution of quasiparticle fermionic excitations becomes more and more pronounced even below  $T_c$ : the Fermi-arc phenomenon. Therefore, both types of excitations coexist, particularly in the overdoped regime, a unique feature of cuprate superconductivity.

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