Specific-heat jump at the superconducting transition and the quantum critical nature of the normal state of pnictide superconductors

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Recently it was discovered that the jump in the specific heat at the superconducting transition in pnictide superconductors is proportional to the superconducting transition temperature to the third power, with the superconducting transition temperature varying from 2 to 25 K including underdoped and overdoped cases. Relying on standard scaling notions for the thermodynamics of strongly interacting quantum critical states, it is pointed out that this behavior is consistent with a normal state that is a quantum critical metal undergoing a pairing instability.

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At present it is widely believed that the "high" T_c superconductivity observed in pnictide superconductors[1] is explained by the classic Bardeen-Cooper-Schrieffer (BCS) mechanism with the caveat that the pairing glue is likely not phonons but instead related to magnetic fluctuations. There is abundant evidence for the opening of a gap in the spectrum of electronic excitations at the superconducting (SC) transition temperature (T_c) as related to the binding of electrons in Cooper pairs. However, for the BCS formalism to be valid (including the nonphonon glue possibility) one also has to assume that the normal state is a Fermi liquid. T_c is after all determined by the balance of the free energy of the superconducting and normal states and one has to understand the nature of both states in order to find out why the transition happens at a particular temperature. More specifically, the pairing instability in classic BCS is governed by the electronic pair susceptibility which is a four-point linear-response function. It is a specialty of the Fermi liquid that the information on the pair susceptibility is entirely contained in the single fermion response but this will *a priori* not be the case in any form of non-Fermi-liquid matter.[2] This issue is recognized both in the context of the "quantum critical" heavy fermion superconductors[3] and the optimally doped cuprate superconductors[4] where it is well established that the normal states are poorly understood non-Fermi liquids. The situation in the pnictides is less clear. Their normal states are bad metals showing quite large, strongly temperature-dependent resistivities and other anomalous transport properties, and it appears that this is not just caused by the low carrier density.[5] The normal state is still rather poorly characterized empirically and it has been hypothesized that it might be quantum critical, perhaps driven by the vanishing of the antiferromagnetism and/or structural phase transition of the parent compounds under influence of doping.[6]

The thermodynamics of strongly interacting quantum critical states is governed by simple scaling behaviors that are applicable also when a microscopic understanding of the critical state is completely absent.[7,8] Here I want to draw attention to recent measurements, revealing a surprising scaling of the jump of the specific heat at the superconducting transition versus T_c in the 122 pnictide family, involving the full doping range where superconductivity occurs. In Fig. 1 I reproduce the results by Bud'ko, Ni, and Canfield (BNC),[9] and include newer data by Mu *et al.*,[10] revealing that the specific-heat jump shows a scaling behavior $\Delta C_p = A T_c^3$, where A is a constant over a dynamical range of more than a decade with T_c varying between 3 and 35 K. To explain such a scaling behavior within the realms of conventional BCS theory one needs extreme fine tuning. Here I want to point out that this scaling finds a natural explanation in terms of the normal state being in some fermionic quantum critical phase that undergoes a pairing instability.

Let us first consider the problems of principle one encounters rationalizing Fig. 1 in terms of conventional Fermi-liquid-based pairing theory. The jump in the specific heat at the transition finds its origin in the fact that the superconducting gap opens up exponentially fast. The specific heat just above the transition reflects the number of degrees of freedom that contributes to the entropy at the temperature $\simeq T_c$ and since these are determined by the renormalized Fermi energy of the metal the Sommerfeld expression C_p

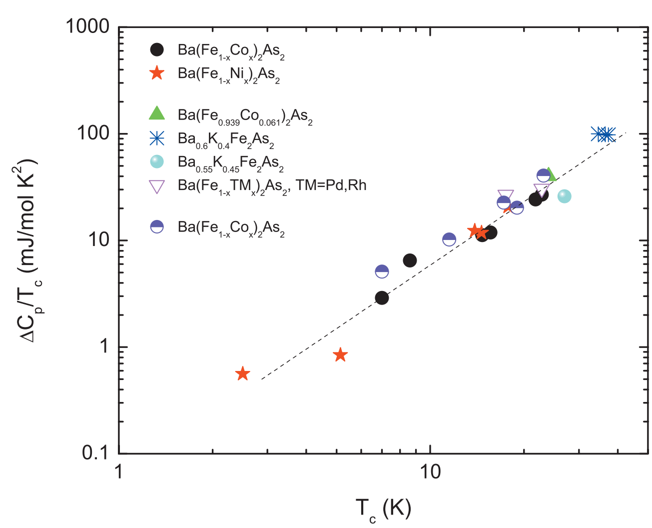

FIG. 1. (Color) The scaling behavior of the ratio specific jump at the superconducting transition and the superconducting transition temperature T_c $\Delta C_p/T_c$ versus T_c in the 122 pnictides, as reproduced from Bud'ko *et al.* (Ref. 9), with the independent results by Mu *et al.* (Ref. 10) (purple half-filled circles) analyzed in a similar fashion and added.

$=\gamma T$ determines the specific heat, where $\gamma = N_0 \sim 1/E_F$. When the SC gap opens, these degrees of freedom suddenly disappear from the energy window $\simeq T_c$ and therefore the specific-heat jumps by an amount $\Delta C_p = BC_p^m(T_c) = B\gamma T_c$, where B is a constant of order unity depending on the details of the thermal evolution of the gap (in weak-coupling *s*-wave BCS, $B=1.14$). Therefore, the ratio $\Delta C_p/(k_B T_c) \simeq N_0$, the density of states in the metal at the temperature T_c. Although numerical factors do depend on complicating factors such as multigap superconductivity, strong-coupling effects and so forth, the scaling of the jump with temperature will not change since it is governed by the Fermi energy, the largest scale of the Fermi liquid. Within this conventional interpretational framework, the "BNC scaling" revealed by Fig. 1 would imply that the density of states in the metal would actually vary like T_c^2. As Bud'ko *et al.* argue, this is quite hard to understand because a Fermi-liquid normal state would imply that local-density approximation band structure should at least yield a qualitative impression of the density of states of the metal in the doping range of the superconducting dome. These calculations however indicate that the density of states should evolve quite smoothly.[11] In fact, one has to deal with a severe "naturalness" problem relying on the conventional BCS interpretation. Internal consistency requires that the coupling constant that determines T_c is itself determined by the density of states of the metal: $\lambda = N_0 V$, where V is the strength of the glue-mediated attractive interaction. The problem can be directly inferred from the weak-coupling BCS expression for the transition temperature $k_B T_c \simeq \hbar\omega_0 \exp(-1/\lambda)$. Writing $N_0 = C_0 T_c^2$ and inserting $\lambda = VC_0 T_c^2$ one finds the condition $T_c^2 \ln(\frac{\hbar\omega_0}{k_B T_c}) \simeq \frac{1}{C_0 V}$. Assuming that the glue retardation scale $\hbar\omega_B$ is doping independent $1/V$ should vary precisely like $T_c^2 \ln(\hbar\omega_B/k_B T_c)$ over a range where T_c varies by more than an order of magnitude. Alternatively, assuming it is due to the retardation scale one has to require that $\hbar\omega_0 \simeq k_B T_c \exp(1/VC_0 T_c^2)$: these are most unnatural fine tuning conditions indeed. This fine tuning problem becomes only worse using more fanciful expressions like the McMillan formula. As a limiting case, consider the ultrastrong-coupling case of Dynes and Allen[12] where $T_c = 0.183\sqrt{\lambda\langle\omega_0^2\rangle}$; this would turn into the extreme fine tuning condition $1 = 0.183\sqrt{C_0 V\langle\omega_0^2\rangle}$.

This paradox finds its origin in the assumption that the normal state is a Fermi liquid. The Fermi liquid is exceptional in the regard that everything is eventually governed by the scale of the Fermi energy. This is obvious for the specific heat but it is also underlying the standard BCS theory. Staying within the realms of a pairing instability, the superconducting transition is governed by the criterium $1 - V\chi'_{pp}(\omega = 0, \vec{q} = 0) = 0$, where V is the interaction strength and χ'_{pp} is the zero frequency, zero momentum real part of the electronic pair susceptibility. For the special case of noninteraction fermions this susceptibility becomes marginal in a scaling sense.[2] The imaginary part is independent of frequency and its magnitude is therefore set by $1/E_F$. By accounting for retardation via the Kramers-Kronig transform[2] $\chi'(\omega=0) = \int_0^{2\omega_B}\chi''(\omega)/\omega d\omega = N_0\int_0^{2\omega_B} d\omega/\omega$ one recovers the "BCS logarithm" that is responsible for the exponential dependence of the gap and T_c on the coupling constant. The relation between the density of states measured by the specific-heat jump and the coupling constant as of relevance to T_c is therefore unique for the Fermi liquid: for *any* other fluid fermion state there will not be a direct relation between these two quantities. The apparent paradox of the previous paragraph can therefore be seen as strong evidence that the normal state of the pnictides is not a Fermi liquid.

Let me now discuss why the scaling of Fig. 1 is suggestive of a quantum critical state. In fact, besides standard scaling arguments one just needs that the system behaves BCS like in the sense that a pairing gap rapidly opens in the spectrum of, now quantum critical, electronic excitations at T_c. This is phenomenologically implied by the very fact that the specific heat jumps. Therefore, the jump measures the normal-state specific heat at T_c, associated with the electrons that pair up in the superconductor: $C_p(T_c) \simeq A_C T_c^3$. It appears that the only way one can explain this scaling behavior without running into other fine-tuning issues is by asserting that the specific heat in the metallic state over the whole superconducting range has a "universal" form $C_p = A'_C T^3$, being just probed at different temperatures (the T_c's). As I will discuss in more detail, this has far reaching and unexpected consequences, and a direct experimental check of this assumption would be desirable. However, it might well appear to be experimentally impossible to disentangle an electronic specific heat from a phonon background with the same T^3 temperature dependence.

This T^3 specific heat is in turn is a rather famous property of the thermodynamics of a strongly interacting quantum critical system.[13] Dealing with a scale invariant (conformal) quantum system one learns from thermal field theory that at finite temperature the scale invariance is broken by the finite radius of the imaginary time circle $R_\tau = \hbar/(k_B T)$. When the fixed point is strongly interacting (obeying hyperscaling) the singular part of the free energy acquires the scaling form,[7,8]

$$F_s = -\rho_0\left(\frac{T}{T_0}\right)^{(d+z)/z} f\left[\frac{r}{(T/T_0)^{\,y_r/z)}}\right], \qquad (1)$$

where d and z are the number of space dimensions and the dynamical critical exponent, respectively, while T_0 is the high-energy cutoff. The crossover function f is governed by the zero-temperature coupling constant r with scaling dimension y_r and since there is no singularity at $r=0$, $T>0$ it expands as $f(x\to 0) = f(0) + xf'(0) + \cdots$. Since the specific heat $C_p = -T(\partial^2 F/\partial T^2)$ it follows,[8]

$$C_p = A_{cr}\left(\frac{T}{T_0}\right)^{d/z}, \qquad (2)$$

where $A_{cr} = \rho_0 f(0)(d+z)d/z^2$. The specialty of the specific heat of a strongly interacting quantum critical system is the fact that its temperature is governed by the engineering dimensions d and z, as rooted in the finite-size scaling. In pnictides it is reasonable to take $d=3$ and consistency with the BNC scaling suggests that $z=1$ reflecting an "emergent Lorentz invariance." Notice also that it requires that nonsingular contributions to the electronic free energy are absent. This is not unreasonable given that we are dealing with fermionic quantum critical matter: it is hard to reconcile fermion statistic with the notion that some electrons stay in a Fermi liquid and others go critical—electrons are after all indistinguishable.

If the above makes sense, we are likely dealing with some unknown form of fermionic quantum criticality and it is *a priori* impossible to make definitive statements regarding the constant A'_C. In 1+1 D it is set by the central charge of the two-dimensional conformal field theory (CFT) but this is much less understood in higher dimensions. The only example where its magnitude for a strongly interacting quantum critical state in higher dimensions is known is the maximally supersymmetric Yang-Mills theory in the large N limit at zero chemical potential. Its thermodynamics is according to the string theoretical Anti-de-Sitter (AdS)/CFT correspondence governed by black hole thermodynamics in an Anti-de-Sitter space time with one extra dimension.[13,14] As in the previous paragraph, the T^d temperature dependence is fixed by scaling but it turns out that the specific heat in the large N limit is just 3/4 of the "Debye" specific heat associated with N free fields,

$$C_p \simeq \frac{4\pi^4}{15} R N^2 \left(\frac{T}{T_0}\right)^3. \tag{3}$$

To give an impression of the numbers in the game, I estimate the prefactor A'_C from the data in Ref. 9 to be $\simeq 26$ mJ/mol K^4. Assuming $N=3$, this just becomes the Debye specific heat for phonons in $d=3$ and the UV cutoff (Debye) temperature becomes $T_0=900$ K. Alternatively, taking $N=1$ it follows that $T_0=432$ K. It is unlikely that the pnictide critical metal has any direct dealings with this zero density large N gauge theory, but this example illustrates that the gross magnitude of A'_C is in first instance determined by the UV cutoff scale which appears to fall in a reasonable regime for the electron system under consideration.

What is the relationship between the specific heat and the superconducting transition temperature when the normal state is a quantum critical metal? On general grounds one expects that the fine tuning problems encountered in the Fermi-liquid case disappear since the "Fermi energy as common denominator" for specific heat and T_c is no longer a factor. This can be illustrated using the simple scaling theory for "BCS" pairing in a quantum critical normal state as recently discussed by She and myself.[2] This departs from the assumption that a truly conformal fermionic state is perturbed by an external retarded bosonic mode causing attractive interactions with the consequence that the BCS gap equation is still valid. The information on the fermion system enters through the fermionic pair susceptibility with a form that is fixed by the conformal invariance and parameterized by an anomalous dimension η_p and dynamical critical exponent z. The superconducting transition temperature is now determined by,[2]

$$k_B T_c \simeq \hbar\omega_0 \left[1 + \frac{1}{\tilde{\lambda}}\left(\frac{2\omega_0}{T_0}\right)^{2-\eta_p/z}\right]^{-z/(2-\eta_p)}, \tag{4}$$

where ω_0 and $\tilde{\lambda}$ represent the glue frequency and pairing strength, respectively, The UV cutoff scale T_0 also enters through the normalization of the dimensionless coupling $\tilde{\lambda} \sim V/T_0$, where V is the dimensionful coupling. By varying V and/or ω_0 one can vary T_c at will while the specific-heat jump automatically tracks the BNC scaling. The fine tuning problem of conventional BCS has completely disappeared.

Without claiming it to be an unique explanation, the scenario in the above is at least consistent with the strong constraints posed by the BNC scaling. It does have however a quite surprising and far reaching consequence for the physics of the pnictides. It suggests that the normal state is a quantum critical *phase* extending over the whole superconducting doping range: this scenario revolves around the notion that there is a metal phase with a specific heat that is doping independent. The prevailing view is that when quantum criticality is relevant for pnictides, it should be tied to the isolated quantum critical point (QCP) associated with the disappearance of the magnetism and/or lattice distortion. After all, this QCP seems at least in the 112 system coincident with the doping level where T_c is maximal.[15–18] As well documented in the heavy fermion systems, a quantum critical metal "fan" as function of increasing temperature or energy is centered at such a zero-temperature QCP. In the cuprates the situation is less clear[19] but a similar analysis as presented here indicates that there is certainly not a "universal" quantum critical thermodynamics.[8] However, the BNC scaling appears to be inconsistent with a metallic state that is controlled by an isolated QCP on the doping axis. The expectation would be that in a doping regime close to optimal doping the transition would go directly from the quantum critical metal to the superconductor, but farther out in the "wings" of the superconducting dome the metal would first crossover to a stable, scale-full state with the transition to the superconductor happening at lower temperature. The emergence of such a scale (Fermi energy, pseudogap, whatever) should show up as a failure of the BNC scaling when T_c's become low. A loophole is that the crossover temperatures might increase very slowly in moving away from the QCP. However, this argument excludes the magnetic quantum critical point as the cause of the quantum criticality. The thermal transition to the finite-temperature magnetic order can only happen at a temperature below the quantum critical crossover temperature, and Fig. 1 contains a number of points at doping levels where the superconducting transition is (much) lower than the antiferromagnetic transition. This does not imply that the magnetic/structural QCP is irrelevant for the superconductivity. It might well be that, as in the Fermi liquid, the critical fluctuations of the bosonic order parameter are a source of strong retarded attractive interactions also in the quantum critical metal.[20]

In conclusion, thermodynamics is a powerful source of information dealing with quantum critical states of matter since it is subjected to strong scaling principles that makes it possible to arrive at phenomenological insights even when a more microscopic understanding is completely absent. If the present claim based on thermodynamics is correct that pnictide metals are quantum critical, this should have far reaching ramifications for other experiments on the normal state. It is hoped that this work will form a source of inspiration for a concerted effort to study this normal state in much further detail. A special challenge in this regard is of course a direct measurement of the electronic specific heat of the normal state to confirm that the electrons that are responsible for the superconductivity contribute a T^3 term to the specific heat. From the estimates in the above it follows that this contribution should be small compared to the T^3 contribution coming from the acoustic phonons.

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