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Zaanen, J.; Hosseinkhani, B.

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Thermodynamics and quantum criticality in cuprate superconductors

J. Zaanen and B. Hosseinkhani

Instituut Lorentz for Theoretical Physics, Leiden University, P.O.Box 9506, 2300 RA Leiden, The Netherlands

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We will present elementary scaling arguments focused on the thermodynamics in the proximity of the quantum critical point in the cuprate superconductors. Extending the analysis centered on the Grüneisen parameter by Rosch, Si, and co-workers to the cuprates, we demonstrate that a combination of specific-heat and chemical potential measurements can reveal the nature of the zero temperature singularity. From the known specific-heat data it follows that the effective number of time dimensions has to equal the number of space dimensions, while we find a total of six scaling laws governing the temperature and density dependence of the chemical potential, revealing directly the coupling constant scaling dimension.

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The possible existence of quantum phase transitions (QPT's) in a variety of condensed matter systems is attracting much interest.¹ The cuprate high- T_c superconductors have played a prominent role in this development since it has been suspected for a long time^{2–5} that the state realized at the doping, where the superconducting transition temperature is maximal (x_{opt}), is controlled by a continuous QPT. This suspicion is mainly motivated by the observation of the “wedge” in the doping (x) temperature (T) plane set by the “pseudogap” [$T_{SG}(x)$] and “coherence”^{6,7} [$T_{coh}(x)$] crossover temperatures, bordering a “quantum-critical” (QC) region characterized by power-law behaviors. It is believed that this signals a QPT from a poorly understood “pseudogap” phase at low dopings to a Fermi liquid at high dopings. Although direct evidence appeared for the presence of scale invariance of the quantum dynamics in the QC regime,⁸ it is unclear if this “critical state” is truly critical in the sense that it is characterized by universality and hyperscaling.¹ Given that apparently fermionic degrees of freedom are involved, this is, from a theoretical point of view, far from obvious because the fermion signs obscure the analogy with thermal phase transitions.⁹ One would like to establish empirically the presence of scaling laws, revealing universality. Such evidence is lacking in the cuprates.

Thermodynamics has played a pivotal role in establishing the nature of the classical critical state. In a recent paper, Zhu *et al.*¹¹ showed that the thermodynamic singularity structure of QPT's has quite interesting observable consequences. They argued that in the case of a QPT, where pressure takes the role of a zero-temperature control parameter (“coupling constant” r), the Grüneisen parameter (ratio of thermal expansion and specific heat C) is particularly revealing with regard to the presence of universality. This was subsequently applied successfully to the QPT's in several heavy fermion intermetallics.¹² Here we will adapt and extend their scaling analysis to the particular situation encountered in the cuprate superconductors. The electronic specific heat of the cuprates is known,¹³ and using simple scaling arguments we will argue that its “normal” appearance (i.e., $C = \gamma T$ with γ constant) in the QC and over-doped regime has actually a profound consequence: it implies that the effective number of time dimensions associated

with the universality class (z , the dynamical critical exponent) has to be equal to the number of space dimensions (d). The quantum ($T=0$) singularity resides elsewhere: the chemical potential μ . We find a large set of scaling relations between its temperature dependence and its density dependence (i.e., the inverse electronic compressibility) while it also relates directly to the doping dependence of the pseudogap scale T_{SG} . The chemical potential can be measured, in principle, with the required accuracy and such experiments can decide if a genuine quantum phase transition is taking place in the cuprates.

Thermodynamics is, of course, in the first instance associated with temperature. A classical phase transition is driven by temperature, but this is profoundly different for a quantum phase transition. The QPT is driven by a zero temperature control parameter r , and the path integral formalism shows that temperature takes the role of a finite size,¹ as the compactification radius of the imaginary time dimension $L_\tau = \hbar / (k_B T)$. The essence of the Zhu *et al.* scaling analysis¹¹ is that one has to determine the dependence of the free energy relative to variations of the coupling constant to learn about the quantum singularity. However, standard thermodynamics associated with variations of temperature gives additional information of the finite-size scaling variety. Their combination yields a powerful phenomenological scaling tool box.

Following Zhu *et al.*,¹¹ our analysis rests on a single theoretical assumption. It is assumed that the QPT is associated with an unstable fixed point at zero temperature, reached by tuning a single zero temperature variable y such that $r = (y - y_c) / y_c$ measures the distance from the critical point residing at y_c . Since temperature T corresponds with L_τ it enters the singular part of the free energy density F_s as a finite size under a scale transformation $x \rightarrow bx$,

$$F_s(r, T) = b^{-(d+z)} F_s(b^y r, b^z T), \quad (1)$$

where d is the space dimensionality and z the dynamical exponent, while hyperscaling is assumed in order to relate the finite size to the scaling dimension y_r of the coupling constant ($y_r = 1/\nu$, where ν is the correlation length expo-

ment). Equation (1) is equivalent to the following scaling forms for the free energy density,¹¹

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

$$= -\rho_0 \left(\frac{T}{T_0}\right)^{(d+z)/z} f\left(\frac{r}{(T/T_0)^{y_r/z}}\right), \quad (2)$$

where ρ_0 and T_0 are nonuniversal constants, while $f(x)$ and $\tilde{f}(x)$ are universal scaling functions. Since there is no singularity at $r=0$, $T>0$, $f(x \rightarrow 0) \simeq f(0) + x f'(0) + (1/2)x^2 f''(0) + \dots$ while $\tilde{f}(x) = \tilde{f}(0) + g(x)$, where $g(x)$ describes the low temperature thermodynamics of the phases to the left or right side of the QPT. When the phase is fully gapped $g(x) \sim e^{-1/x}$ while for a massless phase $g(x) = c x^{y_0+1}$ such that y_0 corresponds with its specific heat exponent ($y_0=1$ for a Fermi liquid, and 2 for a “nodal liquid” characterized by d -wave-like “Dirac cones”).

We find it convenient to parametrize the exponents in terms of d, z , and the zero-temperature analog of the specific heat exponent α , characterizing a thermal phase transition,

$$\alpha_r = 2 - \frac{d+z}{y_r}. \quad (3)$$

In analogy with classical criticality, we expect this exponent to be a fraction of unity. Following Zhu *et al.*, we will consider the specific heat $C = -T(\partial^2 F / \partial T^2)$ and the quantity $\eta_r = (\partial^2 F / \partial r \partial T)$, revealing the dependence of the entropy on the coupling constant. However, we will extend the analysis by also including the “coupling constant susceptibility” $\chi_r = \partial^2 F / \partial r^2$, which is the quantity that is actually most sensitive to the zero-temperature singularity.

From the scaling forms Eq. (2) and the above definitions it follows that the singular parts of various measurable quantities have the following temperature dependence in the quantum critical state ($r=0$):

$$C_{cr}(T, r=0) = \rho_0 f(0) \frac{(d+z)d}{z^2} \left(\frac{T}{T_0}\right)^{d/z},$$

$$\eta_{r,cr}(T, r=0) = -\frac{\rho_0 f'(0)}{T_0} \frac{1 - \alpha_r}{2 - \alpha_r} \frac{d+z}{z}$$

$$\times \left(\frac{T}{T_0}\right)^{[d(1-\alpha_r)-z]/[z(2-\alpha_r)]},$$

$$\chi_{r,cr}(T, r=0) = -\rho_0 f''(0) \left(\frac{T}{T_0}\right)^{-[(d+z)\alpha_r]/[z(2-\alpha_r)]}. \quad (4)$$

On the other hand, in the massless phase characterized by a specific heat exponent y_0 at low temperatures in the vicinity of the QPT,

$$C_{cr}(T \rightarrow 0, r) = \frac{\rho_0 c}{T_0} y_0 (y_0 + 1) r^{(2-\alpha_r)(d-y_0 z)/(d+z)} \left(\frac{T}{T_0}\right)^{y_0},$$

$$\eta_{r,cr}(T \rightarrow 0, r) = -\frac{\rho_0 c}{T_0} \frac{(d-y_0 z)}{d+z} (y_0 + 1)(2 - \alpha_r)$$

$$\times r^{(2-\alpha_r)(d-y_0 z)/(d+z)-1} \left(\frac{T}{T_0}\right)^{y_0},$$

$$\chi_{r,cr}(T \rightarrow 0, r) = -\rho_0 \tilde{f}(0) (d+z-1)(2 - \alpha_r) r^{-\alpha_r}$$

$$- c \rho_0 \frac{(d-y_0 z)}{d+z} \left[(2 - \alpha_r) \left(\frac{d-y_0 z}{d+z}\right) - 1 \right]$$

$$\times r^{(2-\alpha_r)(d-y_0 z)/(d+z)-2} \left(\frac{T}{T_0}\right)^{y_0+1}. \quad (5)$$

From the above equations one directly infers the main results from Zhu *et al.*¹¹ the “Grüneisen ratio” $\Gamma_r = \eta_r / C \sim T^{-y_r/z}$ in the quantum critical state while in the massless phase it becomes exactly $(d-y_0 z)/(y_0 y_r) r^{-1}$, i.e., it acquires a universal amplitude expressed entirely in terms of the exponents. The significance of the coupling constant susceptibility χ_r is immediately clear from Eqs. (4) and (5). Its temperature dependence reveals that it is more singular than η_r , which is in turn more singular than C . In addition, its temperature-independent part diverges in the approach to the critical point with the exponent α_r , in direct analogy with the divergence of the specific heat with α in the approach to a thermal phase transition.

Let us now apply the above scaling laws to the specific context encountered in the cuprates. By restricting ourselves to thermodynamics we have to assume very little in addition to Eq. (1): (i) In the cuprates the relevant zero-temperature direction is the electron density varied by the doping p . The reduced coupling constant corresponds, therefore, with $x = (p - p_c)/p_c$. (ii) Recently, evidence has been accumulating showing that the overdoped state is a Fermi liquid, characterized by $y_0=1$.^{6,7,10} (iii) We rely on the specific heat as measured by Loram and co-workers.¹³ Since the superconductivity appears to hide the critical behavior, the regime of interest is at high temperature.

Given the assumption that electron density is the zero temperature control parameter it follows from elementary thermodynamics that the quantities η_r and χ_r relate to μ ,

$$\eta_{cr,x} = \left. \frac{\partial S_{cr}}{\partial x} \right|_{\mu} = - \left. \frac{\partial \mu}{\partial T} \right|_x,$$

$$\chi_{cr,x} = \left. \frac{\partial^2 F_{cr}}{\partial x^2} \right|_{\mu} = \frac{\partial \mu}{\partial x} = \frac{1}{n^2 \kappa}, \quad (6)$$

where κ is just the electronic compressibility and n the total electron density. Notice that when pressure is the control parameter, $\chi \sim \partial^2 F / \partial p^2 \sim \partial V / \partial p$ refers to the total compressibility.

Let us now turn to the measured electronic specific heat of the cuprates.¹³ In fact, the remarkable property of the measured specific heat is its uninteresting appearance. In the overdoped regime it is indistinguishable from the specific heat of a conventional BCS superconductor. At high temperatures, $C = \gamma T$ with a temperature-independent γ as in a

Fermi liquid, and at the superconducting transition the specific heat shows a BCS-like anomaly. Upon decreasing doping, all that happens is that the pseudogap scale manifests itself quite clearly in the form of a decreasing γ , a fact exploited by Loram *et al.* to study the doping dependence of the pseudogap temperature T_{PG} . Above T_{PG} γ is temperature independent and connected smoothly with the specific heat in the overdoped regime, showing no noticeable doping dependence.

It seems to be a reflex to assume that the “metallic” appearance of the γ above T_{PG} is just revealing that a Fermi-liquid state is reestablished at high temperatures, but this is actually quite unreasonable. Recently, evidence has been accumulating that on the overdoped side a “coherence” crossover occurs: one can identify a temperature T_{coh} below which transport shows Fermi-liquid signatures^{7,10} while photoemission reveals that the quasiparticles become underdamped.⁶ T_{coh} emerges at optimal doping and increases with increasing doping in the overdoped regime. It is no wonder that the low-temperature specific heat in this Fermi-liquid regime is conventional, but why is it so that it remains conventional above T_{coh} ? Stronger, why is it unaltered at temperatures greater than T_{PG} even in the strongly underdoped regime?

Let us reconsider the scaling of the specific heat in the QC regime, Eq. (4). The remarkable fact is that its temperature dependence is predicted to be uninteresting. Its temperature exponent is just given by the ratio of the number of space (d) and effective time (z) dimensions. In the quantum critical regime of the cuprates $C \sim T$ and this means that $d=z$, the number of space dimensions equals the number of time dimensions. At these high temperatures, it seems reasonable to assume that $d=2$, with the implication that $z=2$, signaling diffusion.

There is a nontrivial consistency with the observation that the specific heat is not sensitive to the crossover from the quantum critical to the Fermi-liquid regime at T_{coh} . From Eq. (5) it follows that the specific heat in a massless state knows about the proximity of the QPT via the factor $r^{(2-\alpha_r)(d-y_0z)/(d+z)}$, governing the divergence of the quasiparticle mass. The exponent contains the combination of the dimensions $d-y_0z$ and when $d=z$ and $y_0=1$ as in the Fermi liquid the exponent vanishes and the specific heat becomes insensitive to the zero temperature singularity. The specific heat is expected to be just the same at all temperatures and dopings as long as $T > T_{PG}$ despite the fact that other properties demonstrate large scale changes in the physics.

To further stress this point, let us consider what happens in the pseudogap regime $T < T_{PG}$. The measured specific heat shows that in between the superconducting T_c and T_{PG} $C \sim T^2$ and thermodynamically it can be viewed as a “nodal liquid” characterized by $y_0=2$. Insisting that $d=z$ it follows from Eq. (5) that $C \sim r^{-(2-\alpha_r)/2} T^2$. From Eq. (2) it follows immediately that the pseudogap scale $T_{PG} \sim r^{z/y_r} = r^{(2-\alpha_r)/2}$; it just means that $C \sim T^2/T_{SG}$ which is consistent with experiment. Notice that this would fail when $d \neq z$.

Because α_r is expected to be small, T_{PG} is expected to be weakly sublinear in x when $d=z$. In a recent paper,¹⁴ the behavior of T_{PG} for small x has been determined in 123

samples where the superconductivity has been suppressed by Zn doping. T_{PG} turns out to be indeed weakly sublinear in x , suggesting that α_r is in the range 0.2–0.3, i.e., a reasonable value for a strongly interacting unstable fixed point.

Up to this point we have presented the case that if a QPT is present at optimal doping, the quantum singularity is largely hidden from the specific heat for specific reasons ($d=z$, the Fermi liquid). To establish the presence of this singularity one has to look elsewhere and the remedy is obvious: the thermodynamic potential. Assuming $d=z$ one finds an interesting collection of scaling behaviors for $\partial\mu/\partial T$ and the inverse compressibility χ_x .

Omitting nonuniversal factors and including the specific heat for completeness, these become in the quantum-critical regime,

$$C_{cr} \sim T, \quad \frac{\partial\mu}{\partial T_{cr}} \sim -T^{-\alpha_r/(2-\alpha_r)}, \quad \chi_{cr,x} \sim -T^{-2\alpha_r/(2-\alpha_r)}. \quad (7)$$

Hence, by measuring the temperature dependences of the chemical potential and the compressibility in the *high-temperature* quantum critical regime one obtains directly the “quantum alpha” characterizing the nature of the quantum singularity. Notice that the incompressibility should be precisely twice as singular as the temperature derivative of μ .

The Fermi-liquid regime ($y_0=1$) is not particularly revealing,

$$C_{FL,cr} \sim T, \quad \frac{\partial\mu}{\partial T_{FL,cr}} = 0, \quad \chi_{FL,cr,x} \sim x^{-\alpha_r}. \quad (8)$$

The critical part of $\partial\mu/\partial T$ vanishes because the prefactor contains $d-y_0z$ as does the temperature-dependent part of χ_n . Only the temperature-independent part of the inverse compressibility reveals directly the quantum singularity.

In the pseudogap regime ($y_0=2$) this changes drastically. Parametrizing matters in terms of the pseudogap scale $T_{PG}(n) \sim x^{(2-\alpha_r)/2}$,

$$C_{PG,cr} \sim \frac{T^2}{T_{SG}(x)}, \quad \frac{\partial\mu}{\partial T_{SG,cr}} = \frac{C_{SG,cr}}{x},$$

$$\chi_{PG,cr,x} \sim Ax^{-\alpha_r} - B \frac{C_{SG,cr}T}{x^2}. \quad (9)$$

The second and third lines reflect the workings of the “generalized Grueneisen parameters” as realized by Zhu *et al.*¹¹ $\partial\mu/\partial T$ is clearly “one order more singular” in n than the specific heat, but the temperature-dependent part of the incompressibility is actually “twice as singular.”

To summarize, using elementary power-counting arguments, we have discovered an empirical strategy that should make it possible to decide if the “quantum criticality” of the cuprates has to do with universality. We have found that the temperature and density dependences of the chemical potential in the various regimes should obey *six* scaling laws, which are all governed by a single fundamental scaling dimension (α_r). As an input, we have used the specific heat

data to argue that the effective number of space (d) and time (z) dimensions characterizing the critical state have to be the same.

We are not aware of chemical potential and electronic compressibility measurements of the cuprates having the required accuracy. However, this does not appear to represent a problem of principle. In the experimental literature one finds a variety of methods to measure these quantities;^{15,16} a prime example is the vibrating Kelvin probe method that allows for high accuracy measurements of the chemical potential which was Rietveld *et al.* used by van der Marel some time ago to determine the density dependence of the superconducting T_c .¹⁷ We suggest using these experimental methods to estab-

lish once and for all the presence or absence of a genuine quantum phase transition in the cuprates.

We notice that our scaling relations might also be put to the test in the context of the metal-insulator transition in the two-dimensional electron gas. Using various ingenious techniques,^{18,19} the electronic compressibility has been measured in the proximity of this quantum phase transition and it would be highly interesting to focus on the temperature dependence of the chemical potential. In a future publication we will address this problem in more detail.

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