

Critical Fluctuations and Conserved Dynamics in a Strange Ferromagnetic Metal

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The origin of the strange metallic behavior observed in a wide range of quantum materials is an open challenge to condensed matter physics. Historically, strange metals were uniquely associated with antiferromagnetic quantum critical points (QCPs), but a new generation of materials reveals their association with uniform order parameters, such as ferromagnetism, valley, or nematic order, suggesting a deeper common denominator. At a QCP, order parameter fluctuations are characterized by the dynamical critical exponent z , which quantifies the space-time scaling asymmetry. Here, we report the observation of a divergence in the Grüneisen ratio at the QCP of the strange-metal ferromagnet CeRh_6Ge_4 with a dynamical critical exponent $z = 3$, signaling that the underlying quantum singularity involves a conserved degree of freedom. Yet the magnetization of this easy-plane ferromagnet is not conserved. We argue that the $z = 3$ strange criticality requires a description beyond the Landau paradigm, proposing a link with the gauge modes of the small-to-large Fermi surface transition and the associated gauge charge of the delocalizing heavy electrons.

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The strange metal state that develops near quantum phase transitions is a long-standing mystery [1,2]. These unusual metallic states exhibit fundamental departures from the conventional Fermi liquid physics of metals, with a temperature dependence of the resistivity $\Delta\rho \sim T$ [$\Delta\rho = \rho(T) - \rho(0)$] consistent with a “Planckian” scattering rate [3–6] and a logarithmically divergent heat capacity coefficient $C/T \sim -\log T$ [5,7,8]. Moreover, their close association with superconductivity suggests that the strange metal provides the highly entangled quantum fabric of unconventional and high temperature superconductivity.

Today, a new generation of strange metals challenges our understanding of quantum materials. In early experiments, strange metals were uniquely associated with antiferromagnetic settings, as observed in heavy-fermion, high-temperature, and iron-based superconductors. However, recent experiments on heavy fermion materials [9], as well as iron selenide [10,11], twisted bilayer graphene, and tungsten diselenide [12,13], reveal that strange metals also develop in the vicinity of quantum phase transitions associated with fluctuating uniform order, such as spin and valley ferromagnetism or nematic order. These developments motivate new inquiries into the role of the critical fluctuations in strange metal physics.

The study of ferromagnetism as a quantum phase transition has a long legacy [14–16]. Hertz considered the Stoner ferromagnetic (FM) instability of metals as a quantum critical point (QCP), and later works revealed that a 3D Hertzian FM QCP would exhibit an anomalous $\Delta\rho \sim T^{5/3}$ resistivity characteristic of non-Fermi liquid behavior [17]. However, quantum criticality in a FM metal is predicted to be interrupted by a first order transition due to various non-analyticities originating from coupling

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between the soft modes of magnetization and the itinerant electron gas [18–20].

The discovery of quantum criticality and strange metal behavior in FM Kondo lattices [9,21] has launched a new debate. Since both the Planckian resistivity $\Delta\rho \sim T$ and continuous transition are ostensibly inconsistent with a Hertzian transition, what then is the nature of the critical point? Should we regard the quantum phase transition as the development of critical spin polarization waves, or does it involve a Kondo delocalization of the electrons? Belitz and Kirkpatrick have argued that the inclusion of spin-orbit interactions in noncentrosymmetric systems will re-establish a continuous quantum phase transition within the framework of Hertzian criticality [22]. This idea has spurred additional theoretical debate [23] on whether higher order non-linearities within the Hertzian picture might still give rise to a first order transition. An experimental method that can effectively delineate between a local or an itinerant FM quantum criticality [24] is thus of paramount importance for the unfolding understanding of strange metals.

Generally, a QCP is distinguished by the development of long-range correlations in both space and time. At a critical point the correlation length ξ diverges as a power law of the tuning parameter r according to $\xi \sim r^{-\nu}$. The temporal correlation time ξ_τ also diverges, but it does so at a different rate, so that $\xi_\tau \sim (\xi)^z \sim r^{-\nu z}$, where z is the dynamical critical exponent. A classic experimental probe of the divergence in the temporal correlation time is provided by the Grüneisen parameter $\Gamma = \beta/C$, which is the ratio of the volume thermal expansion coefficient $\beta(T) = d \log V/dT$, where V is the volume, to the specific heat C . General scaling arguments [25–27], independent of microscopic considerations, show that at a QCP the critical component of the Grüneisen parameter diverges with the same scaling dimension as the inverse tuning parameter, i.e., $\Gamma_c \sim r^{-1} \sim \xi_\tau^{1/\nu z}$. Under the usual hyperscaling assumption of quantum criticality [25,28], the correlation time is cut off by thermal fluctuations and scales with the Planck time, $\xi_\tau \sim (\hbar/T)$, from which it follows that $\Gamma_c \sim T^{-1/\nu z}$. In this way, temperature-dependent measurements of the Grüneisen parameter can be used to characterize a QCP. Since theory provides firm predictions for the relationship between dynamical critical exponent and the lifetime of the underlying critical modes, a measurement of Γ_c can therefore, shed light on the nature of the transition.

While the application of hydrostatic pressure to CeRh_6Ge_4 allows for the realization of a FM QCP in a clean stoichiometric compound without introducing disorder [9], only a limited range of experimental probes can be utilized to probe quantum criticality under pressure. Since measurements of the Grüneisen parameter at the FM QCP of CeRh_6Ge_4 can provide vital information about the nature of the quantum criticality, this motivates the search for dopants that tune the FM order [29,30], while preserving the

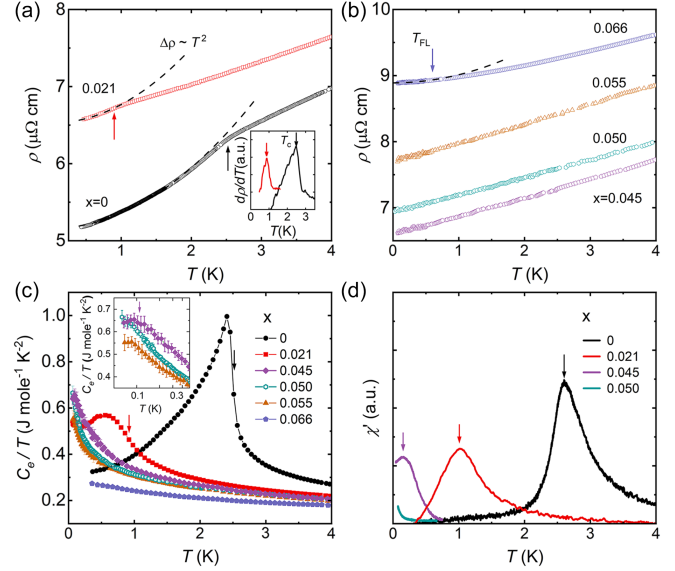


FIG. 1. Temperature dependence of the resistivity of $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ with the current along the c axis for (a) $x \leq 0.021$ and (b) $x \geq 0.045$. The ferromagnetic transitions at T_C are detected in the $x = 0$ and $x = 0.021$ samples as shown by the vertical arrows, which for the resistivity are clearly observed in the derivative in the inset of (a). $\rho(T)$ for $x = 0$ is shifted vertically by $3.6 \mu\Omega \text{ cm}$ for clarity. The dashed lines show the $\Delta\rho \sim T^2$ behavior present below T_{FL} . Temperature dependence of the (c) electronic specific heat coefficient C_e/T (after subtracting the data of LaRh_6Ge_4), of $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ single crystals down to 0.06 K , and (d) the real part of the ac susceptibility χ' , where the ferromagnetic transition corresponds to a peak marked by vertical arrows. The inset of (c) magnifies the low temperature C_e/T below 0.5 K .

intrinsic quantum critical behaviors. Although the substitution of Si for Ge is the most obvious choice for inducing chemical pressure, upon suppressing ferromagnetism with Si doping there is no longer a T -linear resistivity, and anomalous behaviors occur at the critical concentration [29]. Here we instead dope the transition metal site by synthesizing $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ single crystals, where we find that 5% Co doping can induce a FM QCP while still exhibiting the same strange metal behaviors in the resistivity and specific heat.

Single crystals of $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ were grown using a Bi flux method [9,31]. Figures 1(a) and 1(b) display the low temperature electrical resistivity $\rho(T)$ of $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$, where the current is applied along the c axis. For $x = 0.021$, T_C is detected at a lower temperature of 0.9 K , most clearly in the derivative displayed in the inset. At higher concentrations ($x \geq 0.045$), no transition is resolved in $\rho(T)$, and instead there is a linear temperature dependence, much like the strange metal behavior in pressurized CeRh_6Ge_4 [9]. On the other hand, transitions are detected in the electronic specific heat as C_e/T [Fig. 1(c)] with $x = 0.021$, as well as for $x = 0.045$ when measuring down to 0.06 K . Meanwhile at a higher

concentration of $x = 0.050$ no transition is observed and C_e/T diverges, while for $x = 0.055$, C_e/T flattens at the lowest measured temperatures. Upon further increasing the concentration with $x = 0.066$, there is the emergence of $\Delta\rho \sim T^2$ behavior at low temperatures [dashed line in Fig. 1(b)], which together with the flattening of C_e/T , signals the onset of a heavy Fermi liquid state. Similarly, upon measuring the ac susceptibility χ' down to 0.07 K [Fig. 1(d)], the characteristic peak corresponding to T_C is present at 0.15 K for the $x = 0.045$ sample, but is absent for $x \geq 0.050$. Consequently, these results suggest that Co doping continuously suppresses T_C to a QCP at a critical concentration $x_c \approx 0.05$, at which there are the same strange metal behaviors in $\rho(T)$ and C_e/T as at the hydrostatic pressure induced FM QCP [9]. Here the difference from the Si-doped case may be due to the smaller disorder effect, where for Co doping $\rho(0) \approx 7 \mu\Omega\text{cm}$ at x_c , as compared to $\rho(0) \approx 12 \mu\Omega\text{cm}$ for Si [29].

Given that Co doping preserves the FM quantum criticality and strange metal behavior, a wider range of probes are accessible at the QCP, such as the linear thermal expansion. The thermal expansion was measured parallel and perpendicular to the c axis using high-resolution dilatometers in a ^3He refrigerator down to 0.3 K [32]. The temperature dependence of the linear thermal expansion coefficient as α/T , is displayed in Fig. 2 both parallel ($\alpha_{\parallel c}$) and perpendicular ($\alpha_{\perp c}$) to the c axis down to 0.3 K. Above around 3 K, there are similar positive values for all concentrations, but upon cooling there is a pronounced drop in those samples exhibiting magnetic transitions, which crosses to negative α/T for both directions, before reaching a sharp minimum. For $x = 0.045$, the onset of the drop in α/T is observed, which is a precursor to the transition occurring at lower temperatures [Figs. 1(c) and 1(d)]. Meanwhile markedly different behavior is found for samples with $x \geq x_c$, whereby α/T continues to increase upon cooling. Above the critical concentration, α/T diverges with a power law dependence $\alpha/T \sim T^{-n}$, where for $x = x_c$, $n \approx 0.7$ for both directions. Note also the magnitude of $\alpha_{\perp c}$ is consistently two-three times larger than $\alpha_{\parallel c}$, which may also reflect the anisotropy of the crystal-line-electric field ground state and corresponding quasi-1D hybridization [33,34]. Figure 3 shows the temperature- x phase diagram for $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ obtained from combining these different measurements. Here the T_C from thermal expansion results are determined from where there is a sign change of the volume thermal expansion coefficient $\beta = 2\alpha_{\perp c} + \alpha_{\parallel c}$ [35,36], and there is good agreement with the T_C obtained from $\rho(T)$, C_e/T , and χ' . Together these strongly suggest that Co doping suppresses T_C to an FM QCP at $x_c \approx 0.05$.

The behavior at the critical concentration $x_c \approx 0.05$ is highlighted in Fig. 4. In panel (a), $\rho(T)$ and the electronic specific heat coefficient C_e/T are displayed down to 0.06 K. Here $\rho(T)$ remains linear down to the base

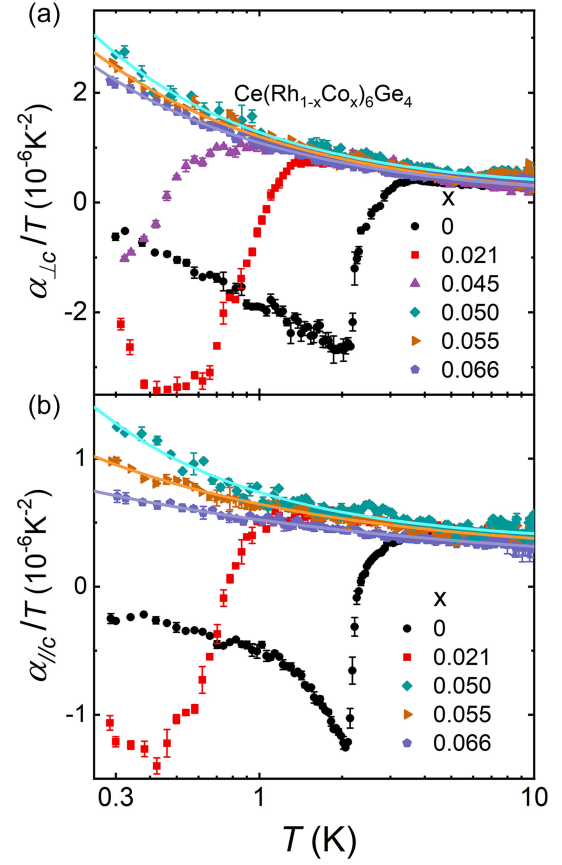


FIG. 2. Temperature dependence of the linear thermal expansion coefficient (a) perpendicular ($\alpha_{\perp c}$), and (b) parallel ($\alpha_{\parallel c}$), to the c axis. For samples exhibiting magnetic transitions, there is a drop of α/T upon cooling for both directions. For $x = 0.050$, 0.055, and 0.066, α/T continues to increase with decreasing temperature, which can be described by a power law behavior $\alpha/T \sim T^{-n}$ as shown by the solid lines, where the corresponding n for $\alpha_{\perp c}$ are 0.70(6), 0.60(4), and 0.61(3), respectively, while for $\alpha_{\parallel c}$ they are 0.70(6), 0.47(6), and 0.33(5).

temperature, mirroring the observation in pressurized CeRh_6Ge_4 over almost two decades of temperature [9]. Meanwhile C_e/T exhibits a logarithmic divergence $C_e/T \sim \log(T^*/T)$ below around 4 K, where the data down to 0.5 K are described by a characteristic temperature [17] $T^* = 48.9$ K (red solid line). At lower temperatures there is a change of slope leading to a more divergent C_e/T , which can be accounted for by either a logarithmic divergence with $T^* = 5.0$ K, or a power law divergence $\sim T^{-0.31}$. A similar change of slope is also observed in CeRh_6Ge_4 under hydrostatic pressure (below which $T^* = 2.3$ K is deduced) [9], in $\text{YbNi}_4(\text{P}_{0.92}\text{As}_{0.08})_2$ [21] and in stoichiometric as well as 5% Ge-substituted YbRh_2Si_2 [5,48]. At very low temperatures below 0.08 K, C_e/T may slightly flatten. This could signal the emergence of a heavy Fermi liquid state (corresponding to a greatly enhanced Sommerfeld coefficient $\gamma \approx 700$ mJ/mol K²), situating the sample very slightly above the QCP.

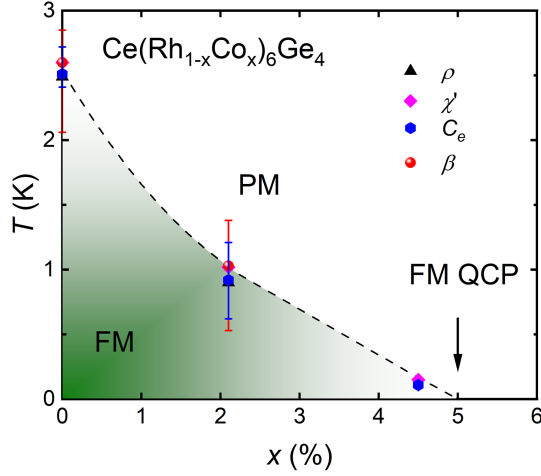


FIG. 3. Temperature-doping phase diagram of $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ determined from resistivity, ac susceptibility, specific heat, and thermal expansion measurements. The T_C for the specific heat and thermal expansion are determined from the midpoints and change of sign of $\beta(T)$, respectively, while the error bars correspond to the transition widths. The ferromagnetic transition is suppressed by doping, and is no longer detected for $x = 0.050$, which is therefore in close proximity to a ferromagnetic QCP.

Figure 4(b) shows the temperature dependence of β/T , with the corresponding critical component of the Grüneisen ratio $\Gamma_c(T) = \beta_c(T)/C_e(T)$ shown in the inset, where the critical component $\beta_c(T)/T$ has the temperature independent contribution subtracted [25,26]. Clear divergences are observed in both quantities, demonstrating a QCP in $\text{Ce}(\text{Rh}_{1-x}\text{Co}_x)_6\text{Ge}_4$ [25–27], with best fits $\beta(T)/T \sim T^{-0.67(4)}$, and $\Gamma_c(T) \sim T^{-0.66(9)}$.

At first sight, our findings of $\nu z = 3/2$ inferred from the exponent in $\Gamma_c(T)$ of $\approx -2/3$ might be interpreted as the observation of a classic Hertzian FM QCP, with $z = 3$ [25] (and $\nu = 1/2$, following [14]). However, this interpretation cannot be reconciled with the strange metal behavior [9], for on the one hand a Hertzian QCP gives rise to a $T^{5/3}$ resistivity [17] while on the other, it is inconsistent with the local moment character of the phase transition, evidenced by the large S_0 in the heat capacity $C/T = S_0/T^* \log(T^*/T)$ [9].

Moreover, CeRh_6Ge_4 is an easy-plane ferromagnet, in which the critical magnetic modes lie in the basal plane, and on rather general grounds we do not expect $z = 3$, because the in-plane magnetization is not conserved. We recall that in an itinerant ferromagnet, collective magnetic modes are overdamped with a dynamical susceptibility that has the form $\chi^{-1}(q, \omega) \sim m + v^2 q^2 + i\omega/D(q) + O(\omega^2)$, where m is the quantum critical tuning parameter and $D(q)$ is the momentum-dependent decay rate of the magnetization at wave vector q in the underlying electron sea. Away from criticality, $m > 0$ is finite giving rise to a dynamical susceptibility with a Lorentzian lineshape with a width

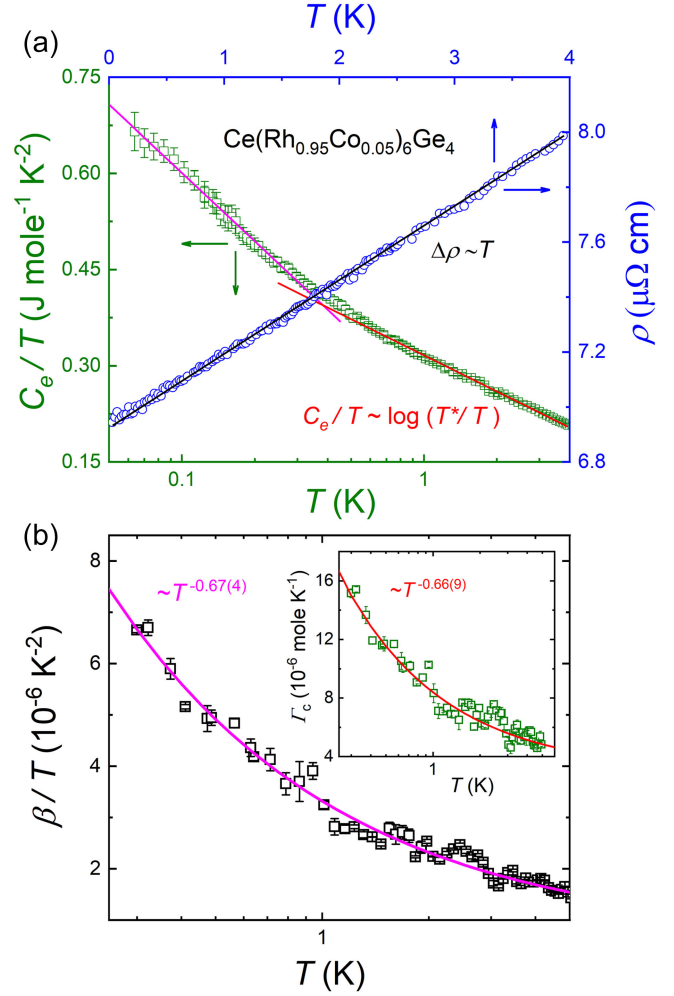


FIG. 4. (a) Temperature dependence of the specific heat coefficient (left axis) and resistivity (right axis) of a sample close to the critical concentration, $\text{Ce}(\text{Rh}_{0.95}\text{Co}_{0.05})_6\text{Ge}_4$, measured down to 0.06 K. Clear strange metal behavior is observed, with the resistivity showing a linear-temperature dependence, while C_e/T continues to increase with decreasing temperature, following a logarithmic temperature dependence (red line), which exhibits a slope change below around 0.5 K. (b) Temperature dependence of the volume thermal expansion (as β/T), together with fit to a power law $\beta/T \sim T^{-n}$ shown by the solid line with $n = 0.67(4)$. The inset shows the temperature dependence of the critical component of the Grüneisen ratio Γ_c that diverges at low temperatures, with an exponent $n = 0.66(9)$.

$\tau^{-1} \sim mD(q)$. If the magnetization is conserved, then the decay rate $D(q) \propto q$ vanishes linearly with momentum. However, in an easy-plane ferromagnet the magnetization is not conserved, so that $\tau^{-1}(q = 0) \sim mD(0)$ is finite, leading to a constant rate of decay of the magnetization at $q = 0$, away from criticality. At the QCP where m vanishes, the decay rate $\tau^{-1}(q) \sim q^z \propto q^2 D(q)$ now determines the dynamical critical exponent. Thus, if the magnetization is conserved $z = 3$ [14,49], but since an easy-plane magnetization is not conserved $\tau^{-1}(q) \sim q^2 D(0)$ implies $z = 2$.

Further symmetry considerations reinforce the departure in CeRh_6Ge_4 from the $z = 3$ expected in a Hertzian model of ferromagnetism. Here, the noncentrosymmetric crystal removes the Kramers degeneracy of the Fermi surface, which as shown in Supplemental Material [36] leads to an excitation gap for in-plane spin fluctuations of conduction electrons. This gives rise to an effective vacuum for the critical magnetic modes which become undamped, giving rise to a reduction of the dynamical critical exponent. For a complete gap, this leads to $z = 1$, predicting $\Gamma_c \sim T^{-2}$, but in the presence of touching points between spin-split Fermi surfaces (the example treated in [36]), a residual damping leads to $z = 2$, predicting $\Gamma_c \sim T^{-1}$. These are both in disagreement with the observed exponent. Together, these arguments rule out the magnetism as the origin of the underlying criticality in our experiment and in so doing implies the criticality lies beyond the Landau paradigm.

An appealing alternative scenario lies in a possible connection between the observed $z = 3$ criticality and the small-to-large Fermi surface transition of a strange metal [36]. The quasiparticle charge density, described by the Fermi surface volume, undergoes a transition in a Kondo-breakdown QCP [50–53]. Notably, while quantum oscillation experiments indicate localized $4f$ electrons at ambient pressure [54], an abrupt reconfiguration of the electronic structure in the vicinity of the QCP has been recently signalled in CeRh_6Ge_4 from thermopower measurements [55]. The essence of the small to large Fermi surface transition is an Anderson-Higgs process in which the phase fluctuations of the hybridization between conduction and f electrons are absorbed into the f -electron gauge field, giving them charge [56–58]. Unlike a relativistic field theory where critical modes propagate at the speed of light, here they move within a dissipative Fermi sea, separating into two components: amplitude modes, which like zero sound in a Landau Fermi liquid lie above the particle-hole continuum, forming a $z = 1$ mode, and gauge modes associated with local conservation of f charge, which lie inside the particle-hole continuum, coupling to the conserved charge of the quasiparticles and giving rise to a $z = 3$ criticality [59,60]. The key point here is that phase fluctuations of the hybridization are overdamped through their coupling to charge via particle-hole excitations. Since the total charge density is a constant of motion, the over-damped dissipation rate vanishes linearly as $q \rightarrow 0$. This leads to a dynamical susceptibility for hybridization fluctuations of the form $\chi_\phi^{-1}(q, \omega) \sim q^2 + i\omega/q$. One can see from this that $\chi_\phi \sim q^{-2}f(\omega/q^3)$, corresponding to $z = 3$ dynamical scaling. Thus by ruling out magnetism, we are led to identify these soft phase fluctuations, mixed with gauge modes, as the most likely origin of the singular thermodynamics and the observed $z = 3$ divergence of the Grüneisen parameter.

This framework allows us to unify various recent observations about strange metal criticality. For instance,

we are able to understand the observation of an almost identical divergence in the Grüneisen parameter $\Gamma_c \sim T^{-0.7}$ in antiferromagnetic YbRh_2Si_2 [26], despite a completely different magnetic structure, as a natural consequence of the conserved dynamics of the small-to-large Fermi surface transition. Based on our arguments, we expect a $z = 3$ gauge mode to develop as a universal precursor of partial Mott localization, independently of the mechanism or whether the transition involves staggered antiferromagnetism [1,61], ferromagnetism, valley [62], or nematic [10] order. More generally, these results underscore the pivotal role of charge-conserving modes [63–65] in a diverse range of strange metal systems. A future measurement of the Grüneisen parameter at the strange metal nematic critical point of sulphur-doped $\text{FeSe}_{1-x}\text{S}_x$ [10] will provide an important test of these ideas.

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Data availability—The data that support the findings of this article are openly available [66].

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