Quantum critical scaling and superconductivity in heavy electron materials

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Quantum critical scaling and superconductivity in heavy electron materials

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We use the two fluid model to determine the conditions under which the nuclear spin-lattice lattice relaxation rate, T_1, of candidate heavy quantum critical superconductors can exhibit scaling behavior and find that it can occur if and only if their "hidden" quantum critical spin fluctuations give rise to a temperature-independent intrinsic heavy electron spin-lattice relaxation rate. The resulting scaling of T_1 with the strength of the heavy electron component and the coherence temperature, T^*, provides a simple test for their presence at pressures at which the superconducting transition temperature, T_c, is maximum and is proportional to T^*. These findings support the previously noted partial scaling of the spin-lattice relaxation rate with T_c in a number of important heavy electron materials and provide additional evidence that in these materials their optimal superconductivity originates in the quantum critical spin fluctuations associated with a nearby phase transition from partially localized to fully itinerant quasiparticles.

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A tantalizing hint that the spin fluctuations seen in the nuclear spin relaxation rate for a number of unconventional superconductors might be the magnetic glue responsible for their superconductivity appears in a scaling relation between that rate and the optimal superconducting transition temperature, T_c, that was first noted by Curro *et al*[1]. In the present communication we focus on understanding this scaling relation for one important member of this family, the heavy electron materials, for which some experimental results are given in Fig. 1[1,2]. As may be seen in Fig. 2, finding such a relation appears at first sight highly problematic because the scaling covers a range of temperatures ($T_c < T < T^*$) in the normal state in which both hybridized localized spins and the itinerant heavy electron Kondo liquid contribute to the spin-lattice relaxation rate. However, we find that rigorous Curro T_c scaling can become possible if three conditions are met: (1) the maximum in T_c occurs at the pressure p_L, at which the line marking the boundary between partially localized and fully itinerant behavior for heavy electron quasiparticles, T_L, intersects with T_c, so that $T_c^{\max} = T_L(p_L)$; (2) at p_L the total spin-lattice relaxation rate scales with the coherence temperature, $T^*(p_L)$; and (3), that at p_L, the materials possess identical values of the product $(3T_c/2T^*)(T_1^{KL}/T_1^{SL})$ where T_1^{KL} and T_1^{SL} are the intrinsic temperature independent spin-lattice relaxation rates of the Kondo and spin liquids respectively.

If the first condition is met, T_c will scale with the coherence temperature, $T^*(p_L)$[3]. The second scaling relation requires that the intrinsic spin-lattice relaxation rate of the heavy electron Kondo liquid be independent of temperature over much of the relevant temperature region, which will be the case if, and only if, it is caused by magnetic quantum critical spin fluctuations[4]. We present a simple way to test whether the second condition is met, and find that is satisfied for a number of heavy electron superconductors. The third condition tells us that Curro scaling cannot be universal, since since it involves two material-specific parameters, $3T_c/2T^*$ and T_1^{KL}/T_1^{SL}, but, as may be seen in Fig. 1, these ratios are sufficiently similar for a number of materials that Curro scaling is approximately valid.

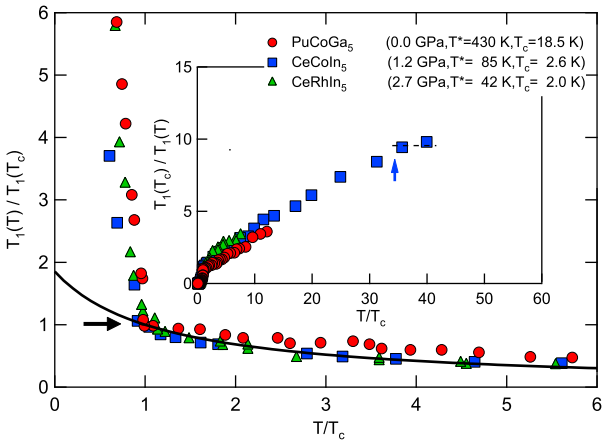

FIG. 1: (Color online) The scaling of the spin-lattice relaxation rate with temperature normalized by the superconducting transition temperature for a number of heavy electron superconductors at pressures near that at which T_c is maximum[1,2]. The solid line represents an approximate scaling relation for $T_c < T < T^*/2$, see Eq. (6). The analysis is extended to higher temperatures in the insert, where the arrows indicate T^*. The pressure, p_L, at which T_c is maximum is not known for $PuCoGa_5$; it is 1.4 GPa for $CeCoIn_5$ and 2.4 GPa for $CeRhIn_5$, values that differ somewhat from those for which data is currently available.

The phenomenological two-fluid model, which explains so many other aspects of heavy electron behavior[4–12], is also key to understanding the remarkable scaling of their dynamic behavior. In it, below T^*, the collective hybridization of the Kondo lattice of f-electron local moments with the background conduction electrons gives rise to an itinerant heavy electron Kondo liquid (KL) of strength (or volume fraction), f, that coexists with the hybridized local moment spin liquid, of strength $1-f$, with[5]

$$f(T,p) = f_0(p)\left(1-\frac{T}{T^*}\right)^{3/2}. \qquad (1)$$

The parameter $f_0(p)$ provides a quantitative measure of hybridization effectiveness; as may be seen in Fig. 2, it is unity at the delocalization quantum critical point (QCP) that marks a zero temperature transition from partially localized to fully itinerant behavior, and typically increases with increasing pressure for Ce-based compounds. For $f_0 > 1$, the model enables one to determine the pressure and temperature dependence of the delocalization line[5], T_L, shown in Fig. 2, that begins at the delocalization QCP and marks, at finite temperatures, the end of the collective hybridization process at $f(T_L,p) = 1$. It follows from Eq. (1) that the delocalization temperature takes the form:

$$T_L(p) = T^*(p)\left[1-f_0(p)^{-2/3}\right]. \qquad (2)$$

Since partial localization ($f < 1$) competes with superconductivity, the two-fluid model predicts that the maximum in the superconducting transition temperature, T_c^{max}, will be found at the pressure p_L, at which the delocalization line, T_L, intersects T_c; experiments on $CeCoIn_5$ and $CeRhIn_5$[13,14] show that this is the case for these materials for which p_L=1.4 GPa and 2.4 GPa, respectively. For pressures less than p_L, partial quasiparticle localization reduces the number of heavy electrons able to become superconducting and so suppresses T_c, while for pressures greater than p_L, it is physically appealing to assume that the attractive interaction between heavy electron quasiparticles becomes increasingly less effective[15].

Note that the quantum critical pressure, $p_{QC} \neq p_L$, as shown in Fig. 2. However, for candidate quantum critical superconductors, $f_0(p_L)$ is not far from its value at the quantum critical point, $f_0(p_{QC}) = 1$, so we can obtain a simple relation between T_c^{max} and $T^*(p_L)$,

$$\frac{T_c^{\max}}{T^*(p_L)} = 1 - f_0(p_L)^{-2/3} \approx \frac{2}{3}\left[f_0(p_L) - 1\right]. \qquad (3)$$

Since $f_0(p_L)$ is a material sensitive parameter, we see that the ratio of T_c to T^* will in general vary from one material to another and that T_c/T scaling will occur only for materials with comparable values of $f_0(p_L)$. This is roughly the case for the materials shown in Table I, in which the value of f_0 is estimated from other experiments

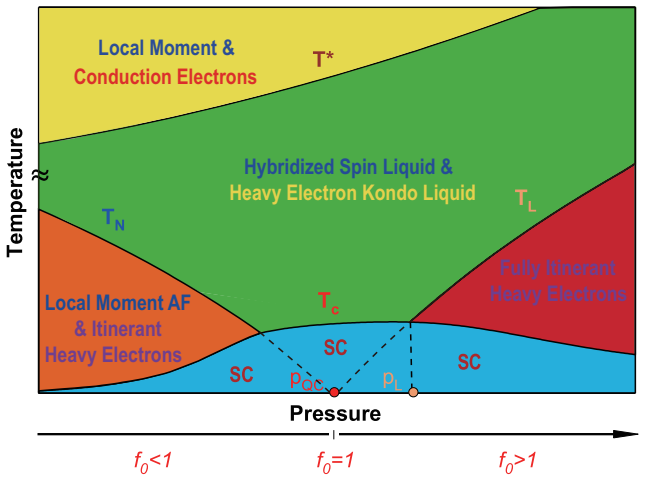

FIG. 2: (Color online) A schematic phase diagram for heavy electron superconductors that is based on the application of the two-fluid model to the pressure-induced changes in the behavior of $CeCoIn_5$ and $CeRhIn_5$[13,14]. It shows the coherence temperature, T^* ($> 30T_c$), at which itinerant heavy electrons emerge to form a Kondo liquid, and the delocalization line, T_L, that marks the boundary between fully itinerant and partially localized heavy electron quasiparticles. Note that T_c is maximum at the pressure, p_L, at which $T_c = T_L$, and, as shown in the text, this provides a link between T_c and T^*. Importantly, at p_L, between T^* and T_c, both the hybridized local spin liquid and the itinerant Kondo liquid contribute to the spin lattice relaxation rate.

for $CeCoIn_5$, $CeRhIn_5$ and URu_2Si_2 and assumed to be near unity at the optimal pressure p_L for $PuCoGa_5$.

We turn next to the two-fluid expression for the spin-lattice relaxation rate[4],

$$\frac{1}{T_1} = \frac{1-f(T)}{T_1^{SL}} + \frac{f(T)}{T_1^{KL}}, \qquad (4)$$

where T_1^{SL} and T_1^{KL} are the intrinsic spin-lattice relaxation times of the hybridized local moment spin liquid and the itinerant Kondo liquid. Eq. (4) tells us that T/T^* scaling will be found if and only if both intrinsic spin-lattice relaxation rates are independent of temperature, with all the measured temperature dependence of $1/T_1$ originating in the strength of the heavy electron component, $f(T)$. For the spin-liquid, experiment suggests that $1/T_1^{SL}$ becomes almost temperature independent as one approaches T^* for materials that are at or near the quantum critical or localization pressure [cf the insert in Fig. 1] and it is reasonable to assume that this behavior continues below T^*. Importantly, the intrinsic Kondo liquid spin-lattice rate will be nearly independent of temperature over a wide range of temperatures provided the spin fluctuations responsible for its spin-lattice relaxation rate, $1/T_1^{KL}$, exhibit the magnetic quantum critical behavior[4,19] that has been shown to give rise to $T_1^{KL}T \sim (T + T_0)$ with T_0 being small and going to zero at the QCP.

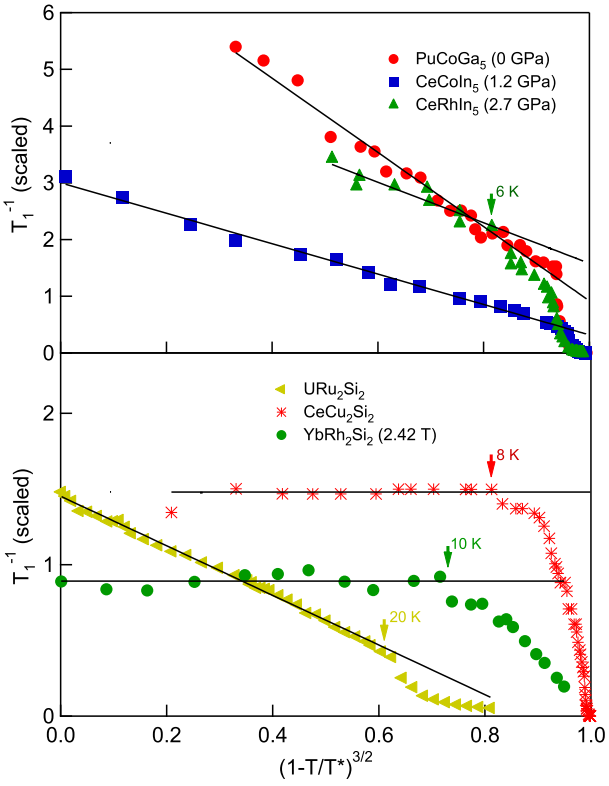

FIG. 3: (Color online) Probing quantum critical scaling behavior in a number of heavy electron materials[1,2,16–18] by plotting the spin-lattice relaxation rate against $(1-T/T^*)^{3/2}$. The arrows indicate the temperature where the scaling fails. Note that temperature decreases from T^* to zero as one goes from right to left on the horizontal axis. While in most of these materials the spin-lattice relaxation rate decreases as the temperature falls below T^* and scales with the strength of the heavy electron component down to some low temperature (indicated by arrows), note that $CeCu_2Si_2$ and $YbRh_2Si_2$ exhibit markedly different behavior.

For any material for which T^* is known, it is straightforward to test the extent to which these conditions are met for the intrinsic spin-lattice relaxation rates: one has only to plot $1/T_1$ vs the strength of the heavy electron component, $(1-T/T^*)^{3/2}$, for temperatures below T^*. On rewriting Eq. (4) as

$$\frac{1}{T_1} = \frac{1}{T_1^{SL}} + \left(\frac{1}{T_1^{KL}} - \frac{1}{T_1^{SL}}\right) f_0(p) \left(1-\frac{T}{T^*}\right)^{3/2}, \quad (5)$$

we see that to the extent that one finds linear behavior, one can determine directly both $1/T_1^{SL}$ and the product $f_0(p)\left(1/T_1^{KL} - 1/T_1^{SL}\right)$ from such a scaling plot. Our results for a number of heavy electron materials are given in Fig. 3, where the expected linear scaling is found between T^* and a cut-off temperature, T_x, for each material shown there, while the corresponding results for the intrinsic spin lattice relaxation rates are given in Table I.

We further note that to the extent that the ratio, T_1^{KL}/T_1^{SL}, is similar for different superconductors, one can obtain two simple non-universal scaling formulae for the spin-lattice relaxation rate. The first, applicable at temperatures from T_c up to $\sim T^*/2$, concerns the scaling behavior with T_c depicted in Fig. 1. Assuming two-fluid scaling behavior persists down to T_c, the strength of the heavy electron component is given by $f(T) \approx f_0(p_L)(1-3T/2T^*) \approx 1-3(T-T_c)/2T^*$ at p_L, and one easily finds,

$$\begin{aligned}\frac{T_1(T_c)}{T_1(T)} &= f(T) + [1-f(T)]\frac{T_1^{KL}}{T_1^{SL}} \\ &\approx 1 + g^*\left(\frac{T}{T_c} - 1\right), \end{aligned} \quad (6)$$

where g^* is a non-universal parameter and equal to $(3T_c/2T^*)(T_1^{KL}/T_1^{SL}-1)$, which is calculated and listed in Table I for each material. A suggested scaling line with $g^* = 0.27$ is plotted in Fig. 1 for comparison with experiment. We see that the overall agreement looks good despite the lack of universality.

A second scaling expression emerges if one consider the spin-lattice relaxation rate normalized at a fixed temperature, say $0.2T^*$:

$$\frac{T_1(0.2T^*)}{T_1(T)} \approx t_0 + 1.4(1-t_0)\left(1-\frac{T}{T^*}\right)^{3/2}, \quad (7)$$

where $t_0 = T_1(0.2T^*)/T_1(T^*)$. As shown in Table I, $t_0 \sim 2.7$ for $PuCoGa_5$ and $CeCeCoIn_5$, and is somewhat lower for the other member of the Ce115 family, $CeRhIn_5$. The solid lines in Fig. 4(a) shows that $t_0 = 2.7$ provides a surprisingly good fit to the data for the materials shown there, despite the fact that the hyperfine coupling constants and spin liquid contributions may differ from material to material and not all of these are at the pressure p_L at which T_c is maximum.

We now look into the details of the scaling behavior for individual materials. In Fig. 3(a), one sees that the T^* scaling continues down to the superconducting transition in $CeCoIn_5$ ($p = 1.2$ GPa) and $PuCoGa_5$. This proves that for $CeCoIn_5$ at p_L the spin fluctuations responsible for the intrinsic Kondo liquid T_1 exhibit quantum critical magnetic behavior, and suggests a possibly similar situation in $PuCoGa_5$. We note that recent elastic moduli measurements have observed an anomalous softening of the bulk modulus in $PuCoGa_5$ and suggested that valence fluctuations may play a critical role in its superconductivity[20]. Combining their results and the NMR measurement[1] suggests that both valence and spin fluctuations may be involved to give rise to the particularly high T_c in this interesting compound.

For $CeRhIn_5$ at $p = 2.7$ GPa, a pressure that is larger than its experimental value of $p_L = 2.4$ GPa, we see that scaling behavior is found down to a temperature $T_x \sim T_L$, its delocalization temperature, that is calculated to be 5.7 K using $T^* = 42$ K and $f_0(2.7\text{ GPa})$=1.24 estimated from other experiments[15]; the loss of scaling below T_x must be

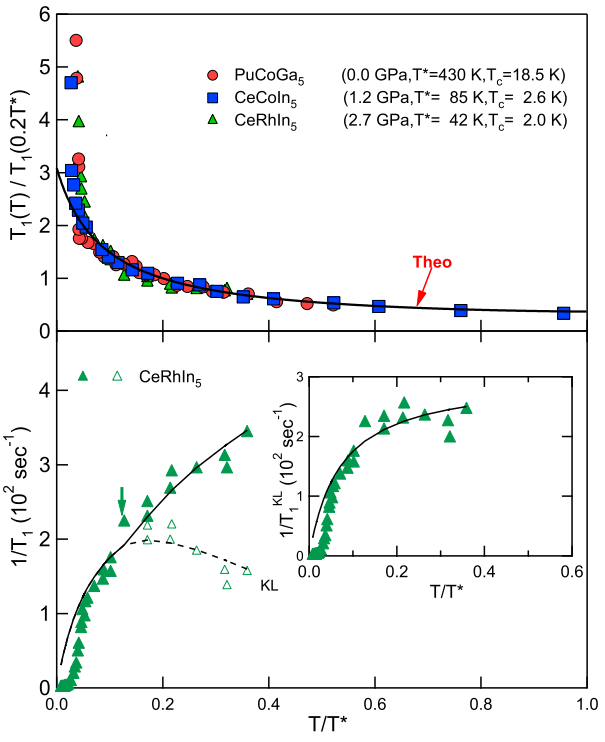

FIG. 4: (Color online) Comparison of theory and experiment for the spin-lattice relaxation rate in several heavy electron superconductors. (a) The scaling of T_1 as a function of T/T^*. The solid line is the proposed scaling formula with $t_0 = 2.7$. (b) The total $1/T_1$ and the Kondo liquid contribution for $CeRhIn_5$ at 2.7 GPa . The inset shows the intrinsic Kondo liquid term $1/T_1^{KL}$ derived after subtracting the spin liquid contribution. The (solid and dashed) lines are fit to experiment using $T_1^{KL}T \sim (T + T_0)$ with $T_0 = 3$ K for $CeRhIn_5$.

attributed to the intrinsic Kondo liquid. As may be seen in Fig. 4, a reasonable fit to the experimental data over the entire temperature region may be obtained if we take $T_1^{KL}T \sim (T + T_0)$ with $T_0 \sim 3$ K providing a measure of the distance away from the quantum critical pressure.

We next examine the scaling behavior, shown in Fig. 3(b), of three other extensively studied heavy electron materials, URu_2Si_2, $CeCu_2Si_2$ and $YbRh_2Si_2$. For URu_2Si_2, we see that as was the case for $CeRhIn_5$ at $p = 2.7$ GPa, T^* scaling works down to the delocalization temperature, $T_L \sim 20$ K (calculated with earlier estimates of $f_0 = 1.6$[5]), a temperature that is slightly above the hidden order transition temperature at 17.5 K. This tells us two things: that the hidden order state emerges from a fully formed itinerant heavy electron Kondo liquid; and that its physical origin is likely an attractive quasiparticle interaction induced by their coupling to quantum critical spin fluctuations, an interaction that, in this case, leads to the hidden order state, rather than superconductivity at much lower temperatures.

The behavior of the spin-lattice relaxation rate of both $CeCu_2Si_2$ and $YbRh_2Si_2$ at ambient pressure is anomalous. Although a number of experiments have suggested a $T^* \sim 70$ K for $CeCu_2Si_2$, we see in Fig. 3(b) that its spin-lattice relaxation rate $1/T_1$ continues to be temperature-independent down to ~ 8 K[17], below which spin density wave fluctuations are observed in neutron scattering experiments[21]. We encounter a similar anomaly in $YbRh_2Si_2$ for which $T^* \sim 50$ K[18,22]. Assuming the two-fluid model is applicable, this either requires a non-accidental cancellation ($T_1^{SL} \approx T_1^{KL}$) of the independent spin liquid and Kondo liquid contributions to $1/T_1$ over a substantial temperature range ($T \gg T_0$), or a quite different physical picture for spin-lattice relaxation in these two materials. At this stage, we cannot make explicit predictions on the nature of the QCP in each material.

In summary, we have shown that an analysis based on the two-fluid model leads to a number of non-universal quantum critical scaling relations in the spin-lattice relaxation rate of heavy electron materials at the pressure, p_L, at which T_c is maximum and explains the quantum critical spin fluctuation origin of the observed Curro T/T_c scaling. It shows that the total spin-lattice relaxation rate will scale with the strength of the heavy electron component, $f(T/T^*)$, and hence with T/T^* only if quantum critical spin fluctuations determine the intrinsic heavy electron spin-lattice relaxation rate, and yields a simple relation between T^* and T_c at p_L. It provides the hitherto missing link between the observation of Curro scaling and its physical origin and enables one to identify materials in which quantum critical spin fluctuations provide the pairing glue. Our results support a strategy for finding higher transition temperature heavy electron superconductors that is based on increasing $T^*(p_L)$[3] and raise the interesting question of whether a similar link can be found for other unconventional superconductors for which Curro scaling has been observed.

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TABLE I: Results of an analysis of the two-fluid parameters and the spin-lattice relaxation rate in several heavy electron superconductors at indicated pressures. The unit for all the spin-lattice relaxation rates is $\sec^{-1}$. T_x is the cut-off temperature below which the scaling breaks down and $t_0 = T_1(0.2T^*)/T_1(T^*)$. $f_0(p)$ is determined by Eq. (3) assuming $p = p_L$ for $PuCoGa_5$ and from previous estimates for all other materials.

	p (GPa)	T_c (K)	T^* (K)	T_c/T^*	$f_0(p)$	$1/T_1^{SL}$ ($\sec^{-1}$)	$1/T_1^{KL}$ ($\sec^{-1}$)	T_1^{KL}/T_1^{SL}	T_x	t_0	g^*	refs.
$PuCoGa_5$	0	18.5	430	0.043	1.068	746	134	5.6	$\sim T_c$	2.68	0.30	[1,3]
$CeCoIn_5$	1.2	2.6	85	0.031	1.023	300	38	7.9	$\sim T_c$	2.77	0.27	[2,12,13]
$CeRhIn_5$	2.7	2.0	42	0.048	1.244	516	228	2.3	$\sim T_L$	1.99	-	[2,14]
URu_2Si_2	0	1.5	75	0.02	1.6	2.91	0.86	3.4	$\sim T_{HO}$	-	-	[16]