Approaching quantum criticality in a partially geometrically frustrated heavy-fermion metal

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Quantum phase transitions have captured the interest of a large community in condensed-matter and atom physics research. These transitions occurring strictly speaking only at absolute zero temperature, are found in insulating and metallic systems, and also in ultracold dilute atomic gases in optical lattices [1, 2, 3]. The common feature of these very different material classes lies in the fact that the competition between low-energy scales can be tuned by a nonthermal parameter, such as pressure, magnetic or electric field, and chemical composition for the condensed-matter systems. In heavy-fermion materials, the strong exchange $J$ between $f$-electrons and conduction electrons can lead to quenching of the $f$-electron-derived (nearly) localized magnetic moments via the Kondo effect or, if $J$ becomes weaker, to long-range magnetic order via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediated by the conduction electrons [4]. In addition to the suppression of magnetic order by Kondo entanglement with conduction electrons, it has been suggested that magnetic order can be suppressed by quantum fluctuations [5, 6] which may be enhanced on general grounds by reducing the effective dimensionality [7] or, more specifically, by geometric frustration. Here we report on the observation of a quantum phase transition in a partially frustrated antiferromagnetic metallic system. In antiferromagnetic CePdAl the magnetic Ce ions form a network of equilateral triangles in the (001) plane, similar to the kagomé lattice, with one third of the Ce moments not participating in long-range order [8]. The Néel temperature
$T_N = 2.7 \text{ K}$ can be driven to zero upon replacing 14.4% of Pd by Ni. Here the specific heat $C$ exhibits a $C/T \propto -\log T$ dependence. Within the Hertz-Millis-Moriya model of quantum criticality, this behavior can be attributed to two-dimensional critical antiferromagnetic fluctuations arising from the decoupling of three-dimensional magnetic order by frustration. The intermediate planes of frustrated moments are a possible candidate for a two-dimensional spin-liquid. The simultaneous presence of magnetic order, geometric frustration, and Kondo effect in this system might thus entail a new route to quantum criticality.

Quantum critical behavior has been observed in a number of heavy-fermion (HF) systems[9]. In the canonical HF system CeCu$_{6-x}$Au$_x$ where a magnetic-nonmagnetic quantum phase transition (QPT) can be tuned by composition or pressure, quasi-two-dimensional (2D) magnetic fluctuations arise out of 3D long-range antiferromagnetic (AF) order, as shown by inelastic neutron scattering [10]. Furthermore, the QPT in this system is associated with unusual energy/temperature scaling of the dynamical susceptibility arising from critical fluctuations [11, 12], also observed in UCu$_{5-x}$Al$_x$ [13]. These findings prompted the development of alternative models of quantum criticality [14, 15] going beyond the Landau-Ginzburg-Wilson model of classical second-order phase transitions and its extension to QPTs at zero temperature by Hertz [16], Millis [17] and Moriya [18] (HMM model). These models were further nurtured by a comprehensive set of experiments on YbRh$_2$Si$_2$ that showed a drastic change of the Hall effect at a field driven QPT, incompatible with the HMM model [19]. In the model of local quantum criticality [15] 2D fluctuations are a prerequisite for the validity of the model. The issue of dimensional crossover in metallic quantum-critical magnets was discussed theoretically by Garst et al. [20]. Recently, several quantum-critical systems of reduced dimensionality were compared with cubic Ce$_3$Pd$_{20}$Si$_6$ [7].

Geometric frustration is a possible means to access quantum critical behavior in magnets, with the possible appearance of spin liquids, e.g., in kagomé lattices [21, 22]. Consequently, a more general (‘global’) phase diagram of quantum criticality was predicted [5, 6, 23]. However, quantum phase transitions induced by geometric frustration have been studied hitherto for insulating systems only. Hence it is of considerable interest to study quantum criticality in HF metals with partial frustration. First experiments were done recently on Ce$_2$Pt$_2$Pb, a realization of the Shastry-Sutherland model [24], and partially frustrated YbAgGe [25]. In the latter system and Ce$_2$Pd$_3$Sn [26] magnetic-field induced QPTs were studied.

In this paper we investigate the quantum critical behavior of partially frustrated CePdAl, a HF system with a distorted kagomé lattice, see Fig. 1(a). In its hexagonal ZrNiAl-type crystal structure, the magnetic Ce ions form a network of equilateral corner-sharing triangles in the $ab$ plane [27, 28]. The compound exhibits a strong magnetic anisotropy with the susceptibility ratio $\chi_c/\chi_{ab} \approx 14$ which is attributed to crystalline-electric-field and exchange anisotropies [28]. CePdAl exhibits Kondo-lattice properties as evidenced by an increase of the electrical resistivity towards low temperature yielding a Kondo temperature $T_K \approx 5 \text{ K}$ [29], and orders antiferromagnetically below $T_N = 2.7 \text{ K}$. 

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Figure 1: (a) View of the \( ab \) plane of CePdAl. The Ce and Pd(2) atoms form a plane at \( z = 0 \), the Al and Pd(1) atoms form a plane at \( z = 1/2 \). (b) View of the \( ab \) plane, showing one of the three symmetry-related magnetic structures. Only Ce atoms are shown for clarity. (c) Kagomé lattice for comparison.
A partially ordered magnetic state of CePdAl was revealed by neutron diffraction measurements on powder samples \cite{8}, with one third of the Ce moments not participating in the long-range order. The magnetic structure of CePdAl consists of three inequivalent Ce sites, with a magnetic ordering vector $Q = (\frac{1}{2}, 0, \tau)$ where $\tau \approx 0.35$ weakly depends on temperature \cite{30}. Within a single kagomé-like layer ferromagnetic chains separated by non-ordered Ce(2) atoms are coupled antiferromagnetically (see Fig. 1(b)). A recent thorough $^{27}\text{Al}$ NMR study indicated that the partially ordered state is indeed stable down to at least 30 mK (Ref. \cite{31}).

The nearest-neighbor (nn) and next-nearest neighbor (nnn) Ce-Ce distances within the basal plane are $d_{ab1} = 3.73$ Å and $d_{ab2} = 5.25$ Å, respectively \cite{8, 32}. The interplane nn Ce-Ce distance is $d_{\perp} = 4.24$ Å \cite{8}. Neglecting the interplane interactions, the magnetic structure has been described in terms of a purely 2D model with nn ferromagnetic ($J_1$) and nnn AF ($J_2$) interactions within the $ab$ plane \cite{33}, see Fig. 1(b). The competition between Kondo and RKKY interactions was modeled in this approach by a $T$-dependent on-site parameter $\Delta_i(T)$, and was studied in detail by variational Monte Carlo simulations \cite{34}. Unfortunately, a model incorporating the interplane coupling $J_{\perp}$ to be compared with the experimentally determined magnetic structure does not exist up to now. Previous work on CePdAl showed that $T_N$ can be suppressed by hydrostatic pressure \cite{29} or partial substitution of Pd by Ni \cite{35, 36}, suggesting the possibility for a QCP.

Polycrystalline samples of CePd$_{1-x}$Ni$_x$Al were prepared by arc-melting appropriate amounts of the pure elements Ce (Ref. \cite{37}), Pd(99.95), Ni(99.95), Al(99.999) under argon atmosphere with titanium gettering. To achieve homogeneity, the samples were remelted several times. The total weight loss after preparation did not exceed 0.5%. The samples were investigated in the as-cast state since annealing may cause a structural change \cite{38}. They were characterized by powder x-ray diffraction, revealing the single-phase ZrNiAl structure ($P\bar{6}2m$) of the parent compounds. Atom absorption spectroscopy was used to determine the actual Ni concentrations $x$ that are quoted throughout this paper. The lattice constants $a$ and $c$ and the unit-cell volume $V$ approximately follow Vegard’s law in the concentration range investigated ($x < 0.15$). Specific-heat measurements were performed in the temperature range $0.05 \leq T \leq 2.5$ K using the standard heat-pulse technique. A Physical Properties Measurement System (PPMS, Quantum Design) was used to obtain data at higher temperatures for some samples. The dc magnetic susceptibility $\chi$ was measured at 0.1 T in the zero-field-cooled field-heated mode in a vibrating sample magnetometer (VSM, Oxford Instruments). A sample-dependent residual background contribution $\chi_0 \approx 2 \cdot 10^{-4} \mu_B$/f.u. independent of $T$ was subtracted from the data.

The specific heat $C$ is shown as $C/T$ vs. $\log T$ in Fig. 2(a). The pure CePdAl compound exhibits a sharp anomaly at the Néel temperature $T_N = 2.7$ K in agreement with literature data \cite{39}. The anomaly broadens and moves to lower $T$ with increasing Ni content $x$ indicating a suppression of the antiferromagnetic (AF) transition. For $x = 0.144$, the $C/T$ vs. $\log T$ data follow a straight line in Fig. 2(a) over almost two decades of temperature in the range $0.05 \leq T \leq 3$ K, i.e., $C/T = a \log(T_0/T)$, with $a = 0.705$ J/mol K$^2$ and $T_0 = 11.7$ K. The non-Fermi-liquid behavior in the form of
Figure 2: (a) Specific heat $C$ of CePd$_{1-x}$Ni$_x$Al plotted as $C/T$ versus log $T$. (b) Specific heat $C$ of CePd$_{0.856}$Ni$_{0.144}$Al plotted as $C/T$ versus $T^{1/2}$. 
a logarithmic divergence of $C/T$ versus $T$ extends over nearly two orders of magnitude in $T$ (0.05 – 3 K). The HMM model [16, 17, 18] predicts for 2D AF fluctuations a logarithmic dependence of $C/T$ near the QCP as observed for CePd$_{1-x}$Ni$_x$Al, while the HMM prediction for 3D antiferromagnets, $C/T \propto \gamma - a \sqrt{T}$ for $T \to 0$, is clearly not compatible with our data, as can be seen from the plot of $C/T$ vs $\sqrt{T}$ in Fig 2(b).

We use the specific-heat data to obtain $T_N$ as the temperature where $C(T)$ is a maximum for low $x$. For larger Ni concentrations, however, the small specific-heat anomaly associated with the onset of magnetic order resides on a large $T$-dependent background $C_{bg}$, similar to the behavior of CeCu$_{6-x}$Au$_x$ close to the QCP [40]. Hence we use $C(T)$ of the allegedly quantum-critical sample with $x = 0.144$ as ‘background’ that is subtracted from the data of the samples close to $x_c$, and take $T_N$ as the temperature of the maximum of the resulting $\Delta C(T)$ curve. Of course, we cannot pin down the critical concentration with absolute accuracy. If the critical concentration would be slightly above 0.144, we would estimate for $x = 0.144$ a maximum possible broad anomaly at $T_N$ to $\approx 0.15$ K for this sample. The resulting $T_N(x)$ is shown in Fig. 3. Up to $x = 0.1$, $T_N(x)$ nicely follows a straight line as expected for the 2D AF QCP in the HMM model. The deviations towards the QCP, possibly signaling a restoration of three-dimensionality, will be discussed below.

One might want to compare $T_N(x)$ with the pressure dependence of $T_N$ of pure CePdAl [41, 29, 30] via the changes of the unit-cell volume by external hydrostatic pressure $p$ or by chemical pressure exerted by the smaller Ni atoms. While $V(x)$ is directly accessible from our x-ray diffraction data, for $V(p)$ one needs the compressibility which can be roughly estimated from the bulk moduli $B_V$ of the pure constituents as described in Ref. [42]. Apart from this uncertainty, $T_N(p)$ data of different authors [41, 29, 30], while agreeing on a reduction of $T_N$ with $p$, differ by a factor of $\approx 2$. We therefore cannot conclude whether the suppression with $x$ is solely due to a volume effect or (if the strong $T_N(p)$ dependence of Goto et al. [29] invalid) is mediated partly by a change of the electronic structure induced by Ni substitution.

In geometrically frustrated insulating magnetic systems with stable moments, frustration arises from (near) cancelation of competing exchange interactions at the site of a given moment. In this case the frustration parameter is defined as $f = |\Theta_{CW}| / T_N$, where $\Theta_{CW}$ is the Curie-Weiss temperature derived from the susceptibility in the paramagnetic regime. Using this definition, it could be shown for the poorly metallic $RXCu_4$ system with $R$ = rare-earth element and $X$ = In or Cd that there is an approximate inverse relation between $f$ and the electrical conductivity $\sigma$, i.e., higher $\sigma$ leads to lower $f$ [43]. This might explain why relatively few frustrated metallic systems have been investigated so far. In CePd$_{1-x}$Ni$_x$Al $|\Theta_{CW}|$ increases slightly with $x$ (by a factor of $\approx 2$), while $T_N$ decreases all the way to $T_N = 0$ between $x = 0$ and $x = 0.144$ [30]. The apparent increase of the ratio $|\Theta_{CW}| / T_N$ with $x$ thus is mostly due to a reduction of $T_N$ and rather reflects the competition between Kondo effect and RKKY interactions than an increased geometric frustration. In fact, a similar apparent increase of $|\Theta_{CW}| / T_N$ can be inferred from data for other HF systems approaching quantum criticality that do not exhibit geometric frustration.

Fig. 4 shows the low-temperature data of the inverse susceptibility $1/(\chi - \chi_0)$ vs. $T$.
Figure 3: Néel temperature $T_N$ vs. Ni concentration $x$ of CePd$_{1-x}$Ni$_x$Al.
for \( x = 0, 0.125 \) and 0.144 \[33\]. The data reveal that, at low \( T \), a marked difference in the \( T \) dependence develops. While \( \chi^{-1}(T) \) varies superlinearly for \( x = 0 \) approaching \( T_N \) from \( T > T_N \), the data for \( x = 0.144 \) can be described with an exponent \( \alpha \approx 0.9 \). This might point to an anomalous scaling exponent \( \alpha < 1 \) as observed for CeCu_{6-x}Au_x \[11\] [12]. Note that this sublinear exponent is only observed close to the QCP. Already for \( x = 0.125 \) the susceptibility exponent is \( \alpha = 1.2 \) signalling the finite \( T_N = 0.374 \). Indeed, sublinear exponent is expected in the model of local quantum criticality [14].

To address the role of frustration concerning the QCP in CePdAl, we first discuss this issue in terms of the HMM model. While the magnetic structure of CePdAl is three-dimensional in nature with the ordering wave vector \( Q = (\frac{1}{2} 0 \tau) \), the specific heat \( C/T \propto \log(T_0/T) \) for \( x \approx x_c \) and the linear \( T_N(x) \) dependence are indeed in accord with 2D AF quantum fluctuations in the HMM scenario. For a single kagomé-like \( ab \) plane, the magnetic structure can be described by ferromagnetic chains, as mentioned above, that couple only weakly antiferromagnetically by the nnn interaction \( J_2 \), because of the frustration of the nn interactions \( J_1 \) at the Ce(2) sites [33, 34]. With recurrence to the magnetic structure of CePdAl [8, 30] we can try to identify possible 2D AF planes: (1) the kagomé \( ab \) planes cannot be taken as origin of the two-dimensional antiferromagnetic fluctuations because the interlayer exchange \( J_\perp \) between Ce atoms is expected to be stronger than \( J_2 \). (2) Taking \( J_\perp \) into account, the magnetic order can be viewed as a stacking of ferromagnetic chains staggered along the \( c \)-direction and interrupted by the frustrated Ce(2) moments thus leading to 2D ferromagnetic planes oriented roughly perpendicularly to \( Q \). However, 2D ferromagnetic fluctuations will give rise to a specific heat \( C/T \propto T^{-\frac{3}{2}} \), which is not supported by our data. (3) A triple-Q structure is in principle compatible with the neutron-scattering data [8]. However, the magnetic component \( \tau \) along the \( c \) direction is, in fact, incommensurate and varies slightly with \( T \) \((\tau = 0.3535 - 0.3585 \) between 0.35 and 2.7 K) [30]. This \( T \)-dependent incommensurability is not easy to explain within a triple-Q structure expected to lock to \( \tau = \frac{1}{3} \). Furthermore, 2D magnetic structures are not easily formed in the triple-Q order of CePdAl. (4) Planes spanned by the ferromagnetic chains in the \( ab \) plane and the perpendicular \( c \) direction are indeed antiferromagnetic, they are separated by frustrated Ce(2) moments and could give rise to 2D AF fluctuations. Hence of the possibilities listed, only the last one presents a viable explanation for the \( C/T \propto \log(T_0/T) \) behavior.

While in the classical case of an anisotropic 3D system geometric frustration might give way to strictly 2D order, in a quantum system zero-point fluctuations might restore the original dimensionality [44]. This, however, is not always the case. For example, it was shown in the context of a Bose-Einstein condensation of magnetic triplets in BaCuSi_2O_6 [45] that geometrical frustration yields a reduction of the spatial dimensionality even close to the QCP [40] [44]. The partial magnetic frustration in CePdAl appears to reduce the dimensionality by forming frustrated planes from one third of the Ce moments, decoupling the antiferromagnetic planes of the other Ce moments as argued above. CePdAl might be a unique example where frustration provides a rationale of how a system might find its way from 3D magnetic ordering to 2D criticality, a microscopic link that is missing for CeCu_{6-x}Au_x, where the origin of 2D fluctuations remains unclear. Thus CePdAl might also be a very promising candidate to test the recent predictions for the
Figure 4: Inverse susceptibility $(\chi - \chi_0)^{-1}$ of $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$ versus temperature $T$ for $x = 0$, 0.125 and 0.144 in the low temperature region below 30 K, where a distinct difference in curvature emerges when approaching the QCP. A sample-dependent residual background contribution $\chi_0$ independent of $T$ (see text) was subtracted from the data. The lines show a fit with a power law $1/(\chi - \chi_0) \propto T^{\alpha} + \Theta^{\alpha}$ with $\alpha = 1.2$ for $x = 0.125$ and $\alpha = 0.9$ for $x = 0.144$. 
2D-3D crossover in terms of the critical quasiparticle theory \cite{47}. The deviations of $T_N(x)$ from a linear $x$ dependence close to $x_c$ (cf. Fig. 3) are compatible with such a crossover.

While we have discussed the possibility of 2D fluctuations confined to AF planes, the intermediate frustrated planes of Ce(2) atoms certainly deserve attention in their own right as well. The perspective of a 2D spin liquid of the these Ce(2) atoms on a rectangular lattice (lattice constants $d_{\perp} = 4.24\text{Å}$ and $d_{Ce(2)} = 7.2170\text{Å}$ perpendicular and parallel to the kagomé planes) is fascinating. It is important to point out that in the present treatments of the HMM model quantum fluctuations arising from geometric frustration are not included. Hence other scenarios are conceivable to explain the quantum critical behavior of CePdAl. Indeed, the susceptibility exponent $\alpha < 1$ for $x = 0.144$ could possibly be due to anomalous scaling. Detailed experiments including inelastic neutron scattering to determine scaling relations between the energy, temperature and field dependencies of quantum critical fluctuations as done, e.g., for CeCu$_{6-x}$Au$_x$ \cite{12}, are necessary to narrow down the origin of the unusual quantum critical behavior of CePdAl. As a further interesting point, CePdAl offers the possibility of investigating the Kondo effect of geometrically frustrated metals in detail, which may lead to a better understanding of the ‘global phase diagram’ \cite{5,6} of quantum criticality in metallic systems.

In conclusion, we have investigated systematically the effect of chemical pressure on the specific heat of CePd$_{1-x}$Ni$_x$Al alloys for Ni concentrations ranging up to $x = 0.144$, where the magnetic order is completely suppressed, giving rise to a QCP. Here the $T\log (T_0/T)$ dependence of the specific heat suggests, if interpreted within the HMM scenario of conventional quantum criticality, the evolution of 2D magnetic fluctuations. The partial geometric frustration of Ce moments in this system may be instrumental in establishing the unusual 2D-like character of quantum criticality, despite the fact that the magnetic order itself is 3D. On the other hand, the partial order might lead to novel types of excitations close to the QCP not contained in the HMM model. In this respect, our results present important clues on how a two-dimensional spin-liquid-like collective of frustrated moments immersed into long-range antiferromagnetic order reacts to the suppression of magnetic order to zero temperature near quantum criticality.

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