Quantum critical behavior in a CePt ferromagnetic Kondo lattice

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Most of the studies of heavy fermions near a quantum critical point (QCP) have been carried out on antiferromagnetic systems. In this paper we report on the disappearance of ferromagnetic (FM) order in CePt as seen by the ac magnetic susceptibility and the electrical resistance measurements under pressures P up to 15 GPa. The pressure dependence of the Curie temperature $T_C(P)$ follows a typical Doniach's phase diagram for Kondo lattice systems and it is consistent with a QCP at the critical pressure $P_C \approx 12.1$ GPa. The resistance in the vicinity of the QCP shows clearly a non-Fermi liquid (NFL) and beyond that a Fermi liquid (FL) state is recovered. A semiquantitative discussion on the nature of the vanishing of T_C in a FM Ce Kondo lattice is given for the first time.

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I. INTRODUCTION

In recent years, a growing interest has focused on the search and study of new heavy-fermion (HF) metals located close to a quantum critical point (QCP). The properties in the vicinity of this quantum phase transition are determined by quantum fluctuations, leading to anomalous temperature dependence of the thermodynamic and transport properties.[1]

Usually the physics of magnetic Kondo lattices (KL) can be described by Doniach's phase diagram[2] supplemented in the noncritical side by the concept of a coherence line, which marks the onset of Fermi-liquid behavior.[3] In principle, the physical scenario of a QCP is independent of the type of magnetic order, but the physical laws describing some parameters at low temperatures and in the vicinity of the QCP can be different if the magnetic order is ferromagnetic or antiferromagnetic (AF), respectively.[1,4–6] In particular, this is the case in the non-Fermi-liquid region above the QCP.

An interesting question in the context of QCPs is the role of the local magnetic moments.[7,8] On one hand, they can be quenched by the Kondo compensation at a finite Kondo temperature when the critical control parameter $(J/W)_C$ is reached; in this case they do not play a role at the quantum critical point. On the other hand, they can persist right down to $T_C=0$ and become quenched only at the QCP, where the Kondo temperature vanishes too. In this latter case the local moments take part in the magnetic critical fluctuations, and their dimensionality will determine the critical behavior.[9]

In comparison to many AF Ce-based heavy-fermion (HF) compounds, only a small number of FM Kondo lattices (FM-KL) have been investigated close to the magnetic instability. This is due to the scarcity of FM-KL systems, either because the few known FM systems often appear to have peculiar phase diagrams in the sense that there is also an AF phase neighboring or embedding the FM state.[10–12] As is well known, archetypes of FM Kondo lattices are considered CePdSb and CeAg compounds. Their investigation at high pressures[13] revealed a pressure dependence of T_C that could be well described by the Doniach diagram for KL,[2] i.e., T_C rises initially, passes through a maximum value, and decreases again. However, in both compounds the ferromagnetic signal in the susceptibility measurements disappeared at a certain pressure while T_C had not yet reached zero. This seems to be another indication for a change of the type of magnetic long-range order in FM-KL before the paramagnetic state is reached—or maybe, for a first-order phase transition. In addition, recent works have shown that the suppression of ferromagnetism in low-T_C clean itinerant ferromagnets is generally of first order.[14] Therefore a direct transition from the FM ordered phase to the paramagnetic state appears to be an exception.

CePt [orthorhombic CrB (B33) structure] is known as a FM compound with a tendency to Kondo screening.[15] Previous study under pressure up to 3.2 GPa revealed that its Curie temperature $T_C(P=0) \approx 6$ K increases with pressure.[16] It is common to compare changes under mechanical compression to the influence of chemical pressure exerted by alloying with isoelectronic atoms of a different size. So if Pt is replaced by Ni the lattice parameters decrease, while T_C increases at the same time until it reaches a maximum value of 8.8 K before it steeply drops and disappears close to $x \approx 0.9$.[17] Isostructural CeNi is known to be an intermediate valent compound. The lattice cell volume does not follow a Vegard's law between CePt and CeNi, decreasing more rapidly beyond about 80% Ni. This means that electronic effects play an important role in the alloys, and as a consequence there will be no one-to-one correspondence to the pressure experiments. However, the assumption appears to be justified that T_C can be driven to zero if only the pressure is high enough. In the following, we report on electrical resistance and susceptibility measurements in stoichiometric polycrystalline CePt in a pressure range which was considerably ex-

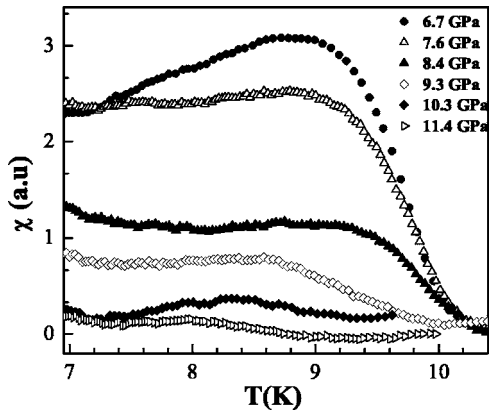

FIG. 1. The ac-susceptibility signal at higher pressure in the range of temperature where the maximum in the signal is observed.

tended compared to the former investigation by Itoh *et al.*[16] Our aim is to follow $T_C(P)$ and find out if it will break down in a similar rapid way as in the alloys. Moreover, by analysis of the electrical resistance data we intend to find details characterizing the type of QCP if this can be reached.

II. EXPERIMENTAL DETAILS

The CePt sample material was prepared by arc melting in purified argon and subsequently heat treated at 700 °C for one week. In a similar way a LaPt sample was prepared. Both materials have been analyzed by x-ray diffraction and showed the right lattice structure free from foreign phases within the limits of resolution of about 5%. Two different kinds of pressure techniques have been used. First, the ac four-probe electrical resistance of CePt was measured in a conventional Bridgman anvil technique using steatite as a pressure transmitting medium and a lead strip as an internal manometer. With this type of pressure cell fitted into a dipstick dilution refrigerator, a temperature range of 40 mK to room temperature could be covered. In a second type of experiment we have determined the magnetic ordering temperature by ac-susceptibility measurements $\chi(T)$ in a diamond anvil cell using a nonmagnetic CuBe gasket filled with a commonly used alcohol mixture as a liquid transmitting medium. The mechanical properties of this CuBe gasket limit the attainable pressures to below ≈11.6 GPa. Again Pb metal is used as an internal manometer. In addition, the electrical resistance of a polycrystalline LaPt sample was measured at ambient pressure and used in the following as an evaluation of the electron-phonon contribution to the CePt resistivity.

III. RESULTS AND DISCUSSION

A. Susceptibility and electrical measurements

In Fig. 1 we plot the ac susceptibility signal at higher pressure. The $\chi(T)$ curves show a distinct maximum at all pressures associated with T_C (see Fig. 3). The shape of $\chi(T)$ does not change with pressure, but the absolute values decrease continuously by an order of magnitude up to the highest pressures. This is evidence that the FM order remains, and the moments decrease under high applied pressure.

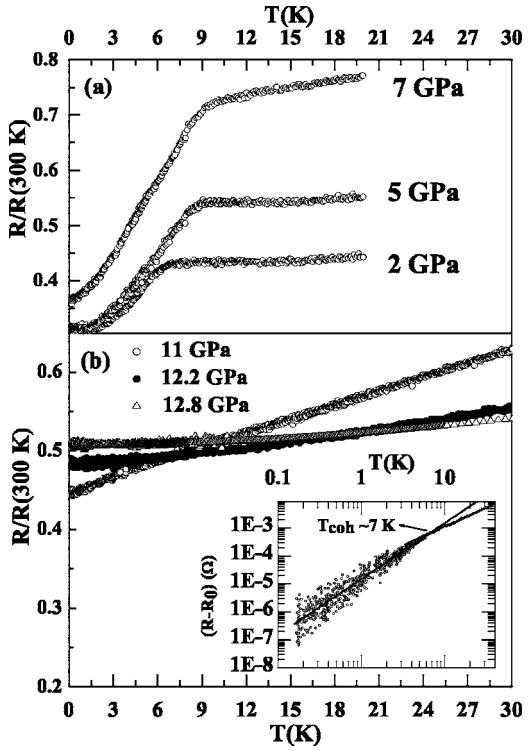

FIG. 2. The normalized resistance R(T) data $R/R(300\ \mathrm{K})$ of CePt at different pressures: (a) in the FM phase; (b) close to QCP, still showing a magnetic transition (11 GPa), or, respectively, incipient Fermi-liquid behavior (12.2 GPa and 12.8 GPa). Inset: At 12.2 GPa a clearcut $T^{1.34}$ dependence is found above ~7 K.

In a similar way, the normalized resistance measurements $R(T)/R(300K)$ are plotted in Fig. 2. As seen in Fig. 2(a), T_C is associated with a clear-cut bend in the resistance curves, especially at low pressures. The precise position of T_C is more reliably determined from the second derivative of the smoothed resistance data and is included in Fig. 3.

As can be seen from Fig. 3 the results for $T_C(P)$ taken from susceptibility and resistance measurements are in good

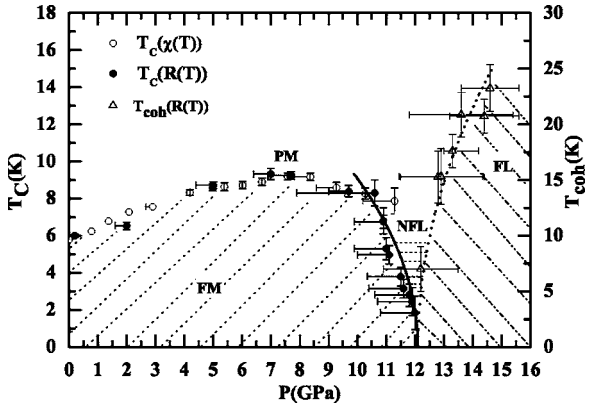

FIG. 3. Phase diagram of CePt as a function of pressure. The data points are from $R(T)$ and $\chi(T)$ measurements. The lines calculated from a power law belong to the mean-field contributions (see text) using P_C=12.1 GPa for the QCP.

agreement. The turnover to a decrease of T_C found in the susceptibility measurements around 9 GPa is confirmed. Moreover, it is followed by a steep drop of T_C between 11 and 12.1 GPa. The initial pressure coefficient $d \ln T_C/dP \approx 1.15 \times 10^{-1}$ GPa^{-1} is somewhat smaller than that found by Itoh *et al.*[16] A comparison with the chemical pressure data of Gignoux *et al.*[17] for $CeNi_xPt_{1-x}$ can be done assuming a constant compressibility of 1.2×10^{-2} GPa^{-1}.[16] It turns out that under pressure, T_C drops only at much higher volume compression than under Ni substitution supporting the assumption of effective electronic changes in the alloys leading to a crossover to intermediate valence. For pressures higher than 12 GPa there is no longer a sign of an ordered magnetic state.

Instead, $R(T) \propto T^n$ with $n=2$, as in a nonmagnetic Fermi liquid (FL), from the lowest measuring temperatures up to the coherence temperature T_{coh}. Beyond that n starts to deviate to smaller values. For instance at 12.2 GPa a power-law fit was possible with $n=1.34(1)$ from about 7 K up to at least 35 K [inset of Fig. 2(b)]. The pressure dependence of T_{coh} is included in Fig. 3. It can be seen that the temperature range for the Fermi-liquid behavior characterized by $R \propto T^2$ is increasing with pressure. By reducing the pressure, it was possible to again enter the magnetic phase and to reproduce some of the Curie temperatures; the shape of the $R(T)$ curves was recovered too. The transition from magnetic order to a paramagnetic state thus is fully reversible.

A remarkable result appears in the resistance curves for 11 GPa ($T_C \sim 5.3$ K) and 12.2 GPa ($T_{coh} \sim 7$ K), just below and above the pressure at which T_C vanishes [see Fig. 2(b)]. Above these temperatures, for both pressures $R(T)$ is nearly linear over two decades of the temperature as is often found in the non-Fermi-liquid (NFL) regime. Altogether our data give strong evidence for the existence of a quantum critical point in CePt separating a FM from a paramagnetic (PM) state at approximately $P_C = 12.1(1)$ GPa. This defines P_C with precision and allows us to obtain with good accuracy the shift exponent ψ describing the pressure dependence of $T_C(P)$ close to the critical pressure. In terms of the distance $\delta = |P - P_C|$ to the QCP, the best fit is given by $T_C = |\delta|^{1/2}$ (solid line in Fig. 3), such that the shift exponent takes the mean-field value $\psi = 1/2$.[18]

B. Ferromagnetic phase

The dome shape of the $T_C(P)$ curve in Fig. 3 can be qualitatively explained in the framework of the Doniach model[2] for KL systems. In this model the phase diagram results from the competition between the Kondo effect and the magnetic ordering of local moments through the RKKY interaction. Notice that in ferromagnetic transition-metal compounds where T_C can be driven to zero, the coupling of quantum critical fluctuations to soft particle-hole excitations can change the transition to first order.[14] We do not find any direct experimental evidence for discontinuous behavior, either in the exponent n of the T^n dependence of the resistance[19] shown below in Fig. 6(a), or in the susceptibility where an abrupt change is expected.[20] The only insinuation of such behavior is the sharp drop of T_C close to P_C.

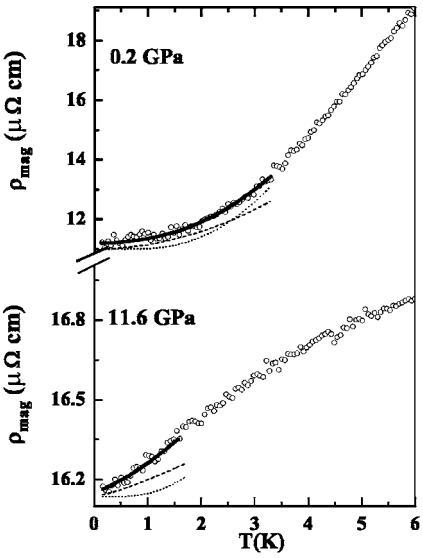

FIG. 4. Magnetic contribution (ρ_{mag}) to $\rho(T)$ of CePt and fits (solid lines) including scattering from spin waves with an excitation gap. The respective components, ρ_{SW} (dotted lines) and BT^n (dashed lines) according to Eqs. (1) and (2), are also shown (these components are shifted up by ρ_0 in order to show a clear overview of the fits).

A new aspect for our analysis of quantum criticality in CePt is added by a study of the magnetic contribution to the resistivity (ρ_{mag}) in the FM phase. This contribution is obtained by subtracting the phonon part given by $\rho(T)$ measured on the nonmagnetic, isostructural compound LaPt. As in previous work,[16] we have assumed that the resistivity of LaPt in the relevant temperature range does not change with pressure. This is justified by the observation that at higher temperatures, around 300 K, the resistance of LaPt only changes by 2% from $P=0$ to $P=2.4$ GPa.[16] The result of this separation for temperatures below 6 K is shown in Fig. 4. Since the $\chi(T)$ data show that the type of magnetic ordering of CePt remains FM, presumably up to the QCP (Ref. 21), collective ferromagnetic excitations (magnons) are present at temperatures below $T_C(P)$.

In anisotropic ferromagnets the dispersion relation of the hydrodynamic spin-wave modes is given by $\omega(k) = \Delta + Dk^2$, where D is the spin-wave stiffness and Δ the spin-wave gap. The origin of this gap arises from the high anisotropy of the crystalline structure of CePt (see Fig. 5) and of the electronic states (f states) responsible for the magnetism. The gapped magnon modes give rise to a temperature-dependent resistivity such as that for $k_B T \ll \Delta$, which is given by[22]

$$\rho_{SW} = bT\Delta e^{-(\Delta/k_B T)}\left[1 + 2\frac{k_B T}{\Delta}\right], \quad (1)$$

where b is a constant for a given material that depends on the spin-wave stiffness D. In fact, $b \propto 1/D^2$ such that the softer the magnons the larger this contribution to the resistivity. Then the resistivity data of CePt yield, in addition to T_C, the spin-wave gap $\Delta(P)$ and the general trend of $D(P)$. The values of Δ are obtained from the fit of the spin-wave contribu-

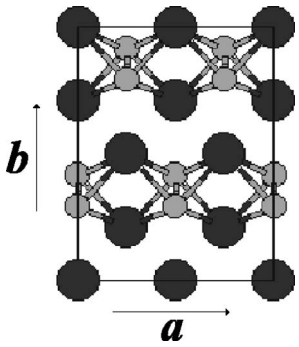

FIG. 5. Lattice structure of CePt showing the large crystalline anisotropy of this compound: large circles Ce, small circles Pt.

tion to the magnetic part of the electrical resistivity ρ_{mag} by the sum of three terms:

$$\rho_{\mathrm{mag}}(T) = \rho_0 + BT^n + \rho_{\mathrm{SW}}(T), \tag{2}$$

where ρ_0 is the residual resistivity, BT^n is the term due to electron-electron scattering, and ρ_{SW} is the spin-wave contribution of Eq. (1). The effective exponent n is plotted in Fig. 6(a). It takes the values $n=2$ for FL and $1<n<1.5$ in the NFL critical regime.[23–26]

The resistance data have been fitted in temperature ranges where the approximation of Eq. (1) is valid, i.e., for $T_{\mathrm{fit}} < 0.55T_C$, taking into account Eqs. (1) and (2), as shown in Fig. 4. The spin-wave contribution to ρ_{mag} is dominant up to 2 GPa ($n=2$). Above 5 GPa ($1<n<1.5$) it is still considerable so as to allow us to obtain reliable values for $\Delta(P)$ as well as for the coefficient $b(P)$.

Figure 6(b) shows the variation with pressure of the gap $\Delta(P)$ and $T_C(P)$, indicating a clear correlation between them. In particular, close to the critical pressure P_C, Δ goes down abruptly with pressure as does T_C. The drop of $\Delta(P)$ beyond

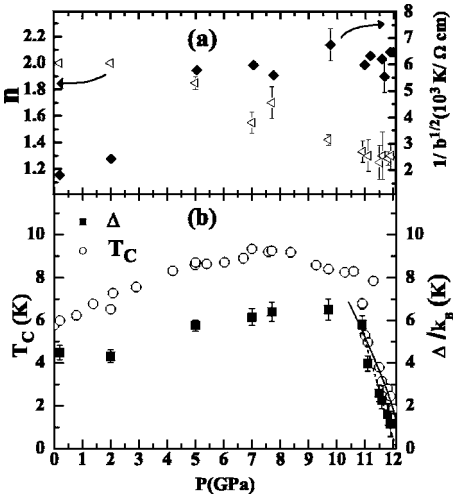

FIG. 6. (a) Pressure dependence of the exponent n and parameter b related to the spin-wave stiffness ($D \propto 1/\sqrt{b}$) of a FM CePt system. (b) $\Delta(P)$ compared to $T_C(P)$. The critical line close to P_C is fitted by Eq. (3) as appropriate for a 2D FM with a vanishing gap (solid line). The linear fit of the gap close to P_C is also shown (dashed line).

$P \sim 10.5$ GPa is well described by a linear fit, $\Delta \approx 4.58|P_C^{\mathrm{gap}} - P|$ leading to a critical pressure for the gap $P_C^{\mathrm{gap}} \approx 12.1$ GPa, which coincides with the critical pressure obtained previously. The coefficient $b(P)$, obtained from the same fit as the gap, is also shown in Fig. 6(a) but in the form relating it to the spin-wave stiffness ($D \propto 1/\sqrt{b}$). $D(P)$ has an initial strong increase and saturates beyond ~ 10.3 GPa, up to the critical pressure. This is unexpected behavior since the magnons should become softer as the QCP is approached. A similar correlation between the Néel temperature, T_N, the spin-wave gap Δ_{AF}, and the spin-wave stiffness was also seen by Continentino *et al.* in the antiferromagnetic heavy fermion compound $CeCoGe_{2.25}Si_{0.75}$ close to a QCP.[9,18]

Also shown in Fig. 6(b) is a fit of the critical line of CePt using a 2D FM anisotropic Heisenberg model. This system has a gap in the spectrum of spin-wave excitations. In this case the vanishing of T_C at P_C can be related to the vanishing of the gap at the same critical pressure. The appropriate expression is given by

$$k_B T_C = \frac{2}{\pi} \frac{\Gamma}{\ln\left(1 + \frac{\Gamma}{\Delta}\right)}, \tag{3}$$

such that $T_C \to 0$ when $\Delta \to 0$. The parameter Γ depends on the spin-wave stiffness D and is assumed to be constant in the region of the fit. The gap used in this equation is $\Delta(P) = 4.58|P - 12.1|$, which is obtained directly from the fit of the resistivity data. Then the only free adjustable parameter in Eq. (3) is the constant Γ which turns out from the fit to be $\Gamma/k_B = 14.2$K. Within the Heisenberg model of localized spins $\Gamma = J\pi^2/4$ and we obtain a ferromagnetic exchange constant $J = 5.75$ K. This value is consistent with the Curie temperature of CePt giving support for the analysis above, which implies a two-dimensional (2D) character for the spin fluctuations in CePt and also that the local magnetic moments remain unquenched up to the QCP. The latter point, whether the moments persist or are quenched at the zero-temperature phase transition, has been the subject of recent debate in the literature.[2,7–9]

Unfortunately there are no data on the *magnetic structure* of CePt in polycrystals or single crystals; a comparison with the known magnetic structure of the isostructural compound PrPt[27] is not sufficiently conclusive. The *lattice structure* itself, however, suggests a tendency to 2D character. This is because the compound is built up of layers of Ce triangular prisms containing Pt stacked along the c axis as shown in Fig. 5. A reduced dimensionality of magnetic fluctuations, from 3D to 2D, has also been considered to explain the vanishing of T_N at a QCP in the nearly AF system $CePd_2Si_2$, for which superconductivity was detected embedded in the NFL state. In this compound the 2D fluctuations of the magnetic moments play an important role not only in accounting for the suppression of magnetism but possibly also for the generation of the superconducting state.[7] In the case of CePt, however, the electrical resistance curves close to P_C do not show any evidence of a superconducting state, at least down to 50 mK.

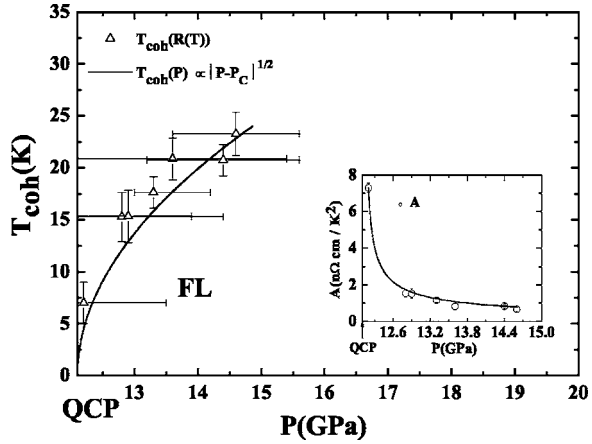

FIG. 7. Closeup of the coherence line. Inset: Pressure dependence of the coefficient A of the T^2 term of the resistivity above the QCP (P_C=12.1 GPa).

C. Above the QCP: Crossover to the Fermi-liquid state

To our knowledge CePt is the first Ce ferromagnetic KL compound that shows NFL behavior in the electrical resistance induced by higher pressures. As mentioned before, at P=12.2 GPa, which is the experimental point in the nonmagnetic side nearest to the estimated P_C, a careful analysis of $R(T)$ shows a dependence like $T^{1.34(1)}$ over two decades in temperature. In general, the spin-fluctuation model yields a resistance with a temperature dependence $R(T) \propto T^{\beta}$ with $\beta = (d+2)/z$, $1<\beta<2$ and $d+z>4$. Here d is the dimensionality of the spin fluctuations and z the dynamic exponent.[20,28] So for a system near a ferromagnetic instability at P_C, with z=3, one expects to find β=5/3 in three dimensions and β =4/3 in two dimensions.[20] Even considering the error bar of the exponent $\beta \sim 1.34(1)$ determined by the fit, this value is closer to that predicted for the case of 2D rather than of 3D ferromagnetic spin fluctuations.

At pressures above P_C=12.1 GPa, the electrical resistance curves develop a temperature dependence $R(T) \propto AT^2$, as appropriate to a FL state, from the lowest temperatures up to a limiting temperature T_{coh}. In the inset of Fig. 7 the coefficient A of the T^2 contribution is plotted as a function of pressure. The coefficient $A(P)$ exhibits a rapid decrease from pressures close to but above the QCP up to 13.3 GPa, after which it remains nearly constant. The behavior of both $T_{\mathrm{coh}}(P)$ and $A(P)$ indicates that the system is being tuned away from the quantum critical region and is in agreement with the theoretical predictions for the nonmagnetic side of the phase diagram of heavy fermions.[18] We find that in the vicinity of the QCP, the coefficient $A(P) \propto 1/|\delta|^{0.6}$ as shown in Fig. 7. This is in reasonable agreement with the value n=1/2 for a ferromagnetic QCP.[28] On the other hand, the experimental values of T_{coh} fall in a range of sufficiently high temperatures that they could be attributed to a mean-field term proportional to $|\delta|^{1/2}$.

IV. CONCLUSIONS

We have studied the quantum criticality of a FM CePt Kondo lattice using electrical resistance and susceptibility measurements. The FM order is suppressed by application of high pressures, very possible with a QCP, around $P_C \sim 12.1$ GPa, separating the FM phase from the paramagnetic phase. Beyond the QCP a Fermi-liquid state is recovered and a coherence line is determined. The analysis of the temperature dependence of the resistance near but above the QCP using the critical spin fluctuation theory is consistent with a dynamic exponent z=3 and d=2 as appropriate to FM fluctuations in a 2D system. Our results suggest a distinctive nature for the vanishing of long-range magnetic order in ferromagnetic f-electron systems as compared to ferromagnetic transition-metal-based materials. They also point out the relevance of the Doniach model for CePt.

In the ferromagnetic phase we have shown that an important contribution to the resistivity arises from the scattering of conduction electrons by ferromagnetic spin waves with a gap in the dispersion relation. The pressure dependence of the gap is correlated with that of the critical temperature; both seem to vanish at the same critical pressure, while the stiffness D remains finite. These results suggest an alternative scenario to the spin fluctuation theory, where the ferromagnetic instability is related to a soft spin-wave mode in a two dimensional material. A qualitative understanding of these results can be obtained within the f-d hybridization model of Endstra *et al.*[29] for similar heavy-fermion systems. As pressure increases, so does the f-d hybridization V_{df}. This in turn increases the effective exchange $J \propto |V_{df}|^4$ among the f spins causing the observed pressure variation of the stiffness ($D \propto J$). On the other hand, the anisotropy is strongly connected to the f character of the wave function of the local electrons. As V_{df} increases there is a strong admixture of these states into the d states which have quenched orbital moments. Consequently as V_{df} increases, the wave functions of the electrons responsible for the magnetism become much less anisotropic decreasing the spin-wave gap. This eventually vanishes at the critical pressure driving the system to a magnetic instability. This model together with the layered, quasi-two-dimensional structure of our compound (Fig. 5) and the spin-wave analysis provide a qualitative understanding of our results.

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