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Quantum criticality in $CePt_{1-x}Ni_xSi_2$

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Abstract. We report measurements of the specific heat, electrical resistivity, and magnetic susceptibility for $\operatorname{CePt}_{1-x}\operatorname{Ni}_x\operatorname{Si}_2$ from which we develop a T-x phase diagram that includes a quantum critical point near $x_{cr} \approx 0.125$ and accompanying non-Fermi-liquid behavior in a "v"-shaped region. This phase diagram is strikingly similar to that of CePtSi₂ under applied pressure P, suggesting that $\operatorname{CePt}_{1-x}\operatorname{Ni}_x\operatorname{Si}_2$ provides a model system in which a T-P-x phase diagram can be smoothly generated, thereby allowing a systematic study of the influence of disorder on quantum criticality.

1. Introduction

In strongly correlated electron systems, it is often possible to tune antiferromagnetic (AFM) order to zero temperature T by an external parameter: e.g., pressure P or chemical substitution x. For many such systems, the resulting quantum critical point (QCP) is protected by a dome of unconventional superconductivity (SC), above which normal state non-Fermi liquid (NFL) behavior is seen in a "v"-shaped region. As the tuning parameter is increased, a heavy fermion (HF) ground state often emerges [1]. While many QC materials fall into this category, a handful of others exhibit even richer phase diagrams. For instance, two domes of SC are observed for CeCu₂Si_{2-x}Ge_x, one of which is associated with an AFM QCP while the other is associated with a valence QCP [2]. It is also well known that P and x, while often similar, do not have identical effects: e.g., most systems which exhibit P induced SC near a QCP do not show SC when tuned by x. As a result, the connection between T - x and T - P phase diagrams is unclear: i.e., the influence of disorder on QC behavior remains an open question.

We were thus motivated to consider the isoelectronic substitution series $\text{CePt}_{1-x}\text{Ni}_x\text{Si}_2$. The compound CePtSi_2 is a HF AFM, for which P suppresses the Néel temperature T_N and a QCP is accessed near 1.5 GPa. The QCP is surrounded by a dome of SC that is followed by a "v"-shaped NFL region [3, 4]. In contrast, CeNiSi₂ exhibits intermediate valence (IV) behavior [5]. For $\text{CePt}_{1-x}\text{Ni}_x\text{Si}_2$, we find pronounced lattice compression with increasing xand commensurate suppression of T_N towards a putative QCP near $x_{cr} = 0.125$, where NFL behavior is observed in a "v" shaped region. In the vicinity of the QCP the characteristic temperatures T_1 and T_2 , which are associated with an interplay between Kondo physics and crystalline electric field effects, respectively, collapse to a single value. This result is similar to what is observed for CePtSi₂ when it is tuned toward its QCP by P. We also find that near the QCP, the magnetic susceptibility $\chi(T)$ is anomalously enhanced, suggesting ferromagnetic (FM) fluctuations. Finally, with increasing x above the QCP, a FL region is eventually recovered. This substitution series is of great interest, as it shares a striking resemblance to the T - P phase diagram of CePtSi₂ and, therefore, provides a model QC system in which a T - P - x phase diagram can be smoothly constructed.

2. Experimental Details

Polycrystalline specimens of $\text{CePt}_{1-x}\text{Ni}_x\text{Si}_2$ were synthesized by arc melting stoichiometric quantities of Ce (99.99%), Pt (99.95%), Ni (99.99%), and Si (99.9999%) under an Ar atmosphere. The resulting boules were flipped and remelted several times in order to encourage homogeneity and subsequently annealed in evacuated quartz tubes at 900 °C for two weeks. X-ray diffraction (XRD) measurements were performed on powdered specimens. The resulting patterns were fitted by means of Rietveld analysis [6] implemented in the program suite GSAS [7]. We investigated the specific heat C(T) for T = 0.4 K - 50 K in zero applied magnetic field H and the electrical resistivity $\rho(T)$ for T = 2 K - 300 K in H = 0 using a Quantum Design Physical Properties Measurement System. Magnetic susceptibility $\chi(T)$ measurements were carried out for T = 2 K - 300 K and H = 1000 Oe using a Quantum Design Magnetic Properties Measurement System.

3. Results

Rietveld analysis of the XRD data reveals that samples across the entire chemical substitution series crystallize in the CeNiGe₂-type orthorhombic structure (space group Cmcm). This structure is nearly tetragonal, consists of stacked Ce and Pt-Si layers along the b-axis, and is closely related to several other prototypical structures which have received intense scrutiny in the context of strongly correlated electron physics and unconventional SC (e.g., ThCr₂Si₂-type). The XRD patterns also include several peaks which are attributable to impurity phases, which makes a quantitative analysis of the low T transport and thermodynamic data challenging. Subsequent attempts to reduce or remove the impurity phases by further annealing were unsuccessful. In order to identify the doping region where the Néel temperature might be suppressed to T = 0, we estimated the effective "chemical pressure" using the formula $P(x) = B_0/B'_0[(V(x)/V_0)^{-B'_0} - 1],$ where $B_0 = 119$ GPa and $B'_0 = 3$ are the P = 0 values of the isothermal bulk modulus and its pressure derivative for CePtSi₂ [8], V_0 is the unit cell volume for CePtSi₂, and V(x) is the unit cell volume for $CePt_{1-x}Ni_xSi_2$ as obtained from XRD measurements (Fig. 1a). From this calculation, we estimate that between x = 0.1 - 0.2 the lattice contraction might produce a chemical pressure that is similar to the critical pressure $P_{cr} \approx 1.5$ GPa in CePtSi₂ (Fig. 1b).

The C(T)/T data are shown in Fig. 1c. For x = 0, C/T increases with decreasing T until a peak is observed near $T_N = 1.9$ K, marking the onset of AFM ordering. We also find a large value for C/T at low T, consistent with the previous conclusion that this system exhibits strong electronic correlations due to hybridization between the f- and conduction electron states. This point of view is further supported by the observation that the Kondo effect removes a significant fraction of the magnetic entropy S_{maq} , giving $S_{maq} \approx 0.4$ Rln2 at T_N . As Ni is substituted for Pt, T_N is suppressed to lower T and is no longer observable near the critical concentration x_{cr} = 0.125, where we suggest that a QCP is found (Fig 2f). This result is in agreement with our estimate for x_{cr} from the bulk modulus of CePtSi₂, showing that x and P are nearly equivalent tuning parameters. In the vicinity of the QCP, C/T exhibits approximately logarithmic behavior at low T, revealing the onset of NFL behavior. We point out that for several of these curves, broad humps are seen between 1-2 K which may be related to magnetic impurity phases: e.g., $CePt_2$ which exhibits antiferromagnetism near $T_N = 1.6$ K. If an impurity $CePt_2$ phase is present, then we estimate that it is no more that 10% of the total sample, by comparing to the jump in C/T at T_N for pure CePt₂ [9]. For $x_{cr} < x, C/T = \gamma$ recovers FL behavior at low T and γ is gradually reduced with increasing x, revealing that the strong electronic correlations that result in an enhanced effective mass m^* for low x are weakened with increasing x. Finally, in order to estimate the single ion Kondo scale T_K , we consider that for a spin 1/2 system, approximately

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Figure 1. (a) Unit cell volume V versus concentration x from powder x-ray diffraction measurements. (b) Chemical pressure P calculated from the bulk modulus B of pure CePtSi₂ (see text) versus x. (c) Specific heat divided by temperature C/T versus T. (d) Magnetic entropy S_{mag} versus T.

1/2Rln2 develops at roughly $0.8T_K$ [10]. From this estimate, we see that T_K smoothly increases with increasing x, as is expected with a gradual crossover from HF to IV behavior (Fig. 2e).

The $\rho(T)$ data are shown in Figs. 2(a,b). The x = 0 T-dependence is similar to previous results, where $\rho(T)$ initially increases with decreasing T due to spin fluctuation scattering and subsequently evolves through two maxima near $T_1 \approx 6$ K and $T_2 \approx 35$ K, which are associated with Kondo lattice behavior and crystalline electric field effects [3, 4]. With increasing x, T_1 increases slightly, while the hump associated with T_2 moves to higher T and broadens significantly. Between x = 0.1 - 0.2, T_1 and T_2 merge into a single peak which then moves to higher T with increasing x (Fig. 2e). We point out that this behavior is in good agreement with what is observed for CePtSi₂ under applied pressure. In order to search for SC in the vicinity of the QCP, we measured $\rho(T)$ for x = 0.1, 0.15, 0.2, and 0.25 down to 100 mK. These measurements revealed no evidence for SC.

The $\chi(T)$ data are shown in Fig. 2c where, up to x = 0.4, Curie-Weiss (CW) behavior given by the expression $\chi(T) = C/(T - \Theta)$ is observed, resulting in effective magnetic moments $\mu_{eff} \approx$ 2.5(4) μ_B for $x \leq 0.4$, as expected for localized Ce³⁺ ions. The CW temperatures Θ are plotted in Fig. 2d, where we find that they are strongly peaked in the vicinity of the QCP, reflecting an anomalous enhancement of $\chi(T)$ which may be related to the emergence of FM fluctuations. It is interesting to note that for several other systems (e.g., YbRh₂Si₂ [11] and β -YbAlB₄ [12]) FM may also play a role near the QCP.

Taken together, these measurements reveal a striking similarity between the T - P phase diagram for CePtSi₂ and the T - x phase diagram for CePt_{1-x}Ni_xSi₂, as summarized in Fig. 2(d,e,f). Thus, the CePt_{1-x}Ni_xSi₂ substitution series may provide a platform on which the influence of P and x can be smoothly connected in a T - P - x phase diagram. Such studies are of interest because they will allow for greater understanding of the influence of disorder on QC and associated phenomena (e.g., unconventional SC).



Figure 2. (a,b) Normalized electrical resistivity ρ/ρ_0 versus temperature T for CePt_{1-x}Ni_xSi₂ and the nonmagnetic analogue compounds LaPtSi₂ and LaNiSi₂. (c) Inverse magnetic susceptibility χ^{-1} versus T measured in an applied magnetic field H = 1000 Oe.(d) Curie-Weiss temperature Θ obtained from fits to $\chi(T)$ (see text) versus x. The dotted line is a guide to the eye. (e) Temperatures T_1 , T_2 , and T_{max} where the electrical resistivity exhibits either a maximum or a hump and Kondo temperature T_K (see text). (f) Phase diagram of $\text{CePt}_{1-x}\text{Ni}_x\text{Si}_2$ where T_N is the Néel temperature obtained from specific heat measurements and T_{FL} is the temperature where C/T becomes constant at low T, consistent with the onset of Fermi liquid behavior. The dashed lines are extrapolations for T_N and T_{FL} . The solid grey bar near $x_{cr} \approx 0.125$ denotes the putative quantum critical region.

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