Universal Scaling and a Novel Quantum Critical Behavior of $CeRhSb_{1-x}Sn_x$

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We observe and explain a universal scaling $\rho\chi =$ const for the electrical resistivity ρ with the inverse magnetic susceptibility χ^{-1} for the Kondo insulator $CeRhSb_{1-x}Sn_x$. In the regime where the Kondo gap disappears ($x > 0.12$), the system forms a non-Fermi liquid (NFL), which transforms into a Fermi liquid at higher temperature. The NFL behavior is associated with the presence of a novel quantum critical point (QCP) at the Kondo insulator–correlated metal boundary. The divergent behavior of the resistivity, the susceptibility, and the specific heat has been observed when approaching the QCP from the metallic side and is interpreted as due to the competition between the Kondo and the intersite magnetic correlations.

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The compounds CeRhSb and CeRhSn, both with strongly correlated $4f$ electrons, form unprecedented types of ground state: CeRhSb represents a *Kondo semiconductor* with a gap $\Delta \simeq 7$ K [1] and CeRhSn is a *non-Fermi* (non-Landau) quantum liquid [2].

The fundamental question is whether by studying $CeRhSb_{1-x}Sn_x$ the two states can be joined through a corresponding *quantum critical point* (QCP) or by a phase boundary. In that respect, the quantum critical points between antiferromagnetic metal and either superconducting [3] or paramagnetic [4] heavy-fermion states have been successfully identified and play an important role in the discussion of the origin of exotic superconductivity [3]. Also, the existence of the QCP between the Kondo insulator (KI) and an itinerant antiferromagnet has been proposed [5] as a function of either the Kondo coupling or pressure.

In this Letter we show directly that a transition of this type indeed exhibits a quantum critical behavior and a formation of a *singular* non-Fermi liquid (NFL) state as a function of the number of carriers. The number of the carriers per $4f$ electron is a crucial factor when examining the formation of the collective Kondo-lattice singlet state [6]. To address these basic topics, we have studied the series $CeRhSb_{1-x}Sn_x$, in which the number of valence electrons diminishes by one per formula unit when substituting Sn for Sb. The characterization of such QCP allows us also to compare the insulator-metal transitions for the *Mott-Hubbard* systems and for the *Anderson-Kondo* lattices and show they represent different universality classes of quantum materials.

We first demonstrate, on the basis of experimental-data correlation, a novel type of scaling between the electrical resistivity ρ and the magnetic susceptibility χ in the *quantum-coherence regime* for the Kondo semiconductors $CeRhSb_{1-x}Sn_x$, with $x \leq 0.12$. The reference resistivity data are displayed in Figs. 1(a) and 1(b). We are interested in the properties below the temperature T_{max}, where the resistivity has a maximum. For $T > T_{max}$ a pronounced $\ln T$ behavior of ρ should be noted for all the samples studied

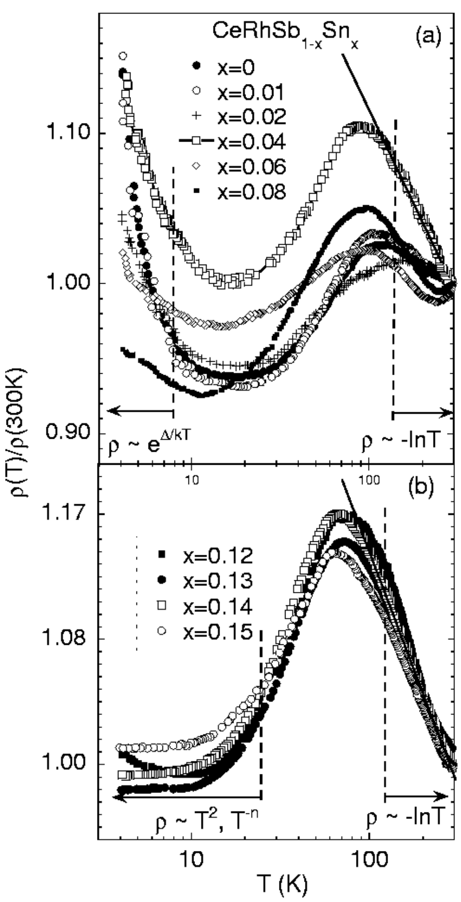

FIG. 1. Temperature dependence of the relative resistivity ρ for $CeRhSb_{1-x}Sn_x$ systems for (a) $x \leq 0.08$ and (b) $0.12 \leq x \leq 0.15$. The regimes, where $\rho(T) \sim \exp(\Delta/k_BT)$, $\sim -\ln T$, and $\sim T^2$, are marked by the corresponding arrows.

[7]. For $T \ll T_{\max}$ a pronounced exponential increase of $\rho \sim \exp(\Delta/T)$, representing a nondegenerate semiconductor, can be fitted well for $x \leq 0.12$. The sample preparation and the measurement techniques used are described in [8]. The stable monophase samples can be synthetized only for $0 \leq x \leq 0.2$ and $0.8 \leq x \leq 1$.

In Fig. 2 we show representative data (for $x = 0$) of the scaling of $1/\rho$ with χ; the inset provides the detailed behavior. One should note that in order to obtain such good scaling, we have to subtract from the measured χ the impurity Curie-law contribution (nC/T), with n in the range 0.004–0.008 (depending on the sample), which represents a standard procedure in these and related compounds [8]. Also, then $\chi \rightarrow 0$ with $T \rightarrow 0$, demonstrating that the Kondo semiconductors can be regarded at $T = 0$ as either *heavy quasiparticle band insulators* (with a band gap renormalized by the electron correlations [9]) or equivalently as *collective Kondo-lattice insulators* [10], with a binding of carriers into the spin-singlet ground state. The scaling law $\rho \sim \chi^{-1}$ speaks in favor of the latter explanation. Also, the vanishing χ distinguishes the Kondo insulators from the Mott-Hubbard insulators, for which the localized electrons have unpaired spins and thus order magnetically when $\Delta > 0$. In the regime $x \leq 0.06$ the value of $\rho\chi$ is universal and equal to 0.1 $\mu\Omega$ cm emu/mol. The details of the scaling will be presented elsewhere.

To characterize the metallic phase we have plotted in Fig. 3 the resistivity as a function of T^2. This dependence is well obeyed at higher temperatures $T > 10$ K. In the T range below about 10 K (see the inset), a clear deviation from the Landau-Pomeranchuk-Baber law $\rho = \rho_0 + AT^2$ for the Fermi liquid can be observed, particularly for the concentration $x \simeq 0.13$, where the gap has just disappeared. The fitting to the $\sim T^{-n}$ dependence, with $n \simeq 0.01$, should be regarded only as an indicator of incipient *NFL behavior,* requiring an additional test. Alternatively, these low-T data can be represented by the ln$T$ dependence.

To determine thermodynamic properties of the NFL in the vicinity of the critical point, we show in Fig. 4 the temperature dependence of $\chi(T)$ for (a) $x \leq 0.12$ and for (b) $0.13 \leq x \leq 0.16$. In the metallic phase (b), a clear dependence $\chi = aT^{-m}$ is observed (see the inset, where the double logarithmic scale was utilized). The values of the parameters are listed in Table I. The quantities χ^{-1} and ρ for $x \geq 0.13$ do not obey the type of relation shown in Fig. 2 for the KI state. Instead, χ diverges, and ρ deviates from the Fermi-liquid (FL) behavior. The divergence of χ signals quantum phase transition to a singular NFL phase with divergent χ, very small magnetic moment, and the presence of magnetic hysteresis [11]. The onset of magnetism signals in turn a spontaneous symmetry breakdown at $T = 0$ needed for the *quantum critical point* to be well defined. So, whereas in Mott-Hubbard systems the insulating phase is magnetic (very often antiferromagnetic) and the metallic phase can be either antiferromagnetic or paramagnetic, the sequence of phases here is that the Kondo insulating phase is nonmagnetic (diamagnetic) and the metallic phase is magnetic, with a very small moment, as is the case in many heavy-fermion systems.

We have also performed the specific heat (C_p) measurements on the sample with $x = 0.13$. The data in the temperature range 10–20 K can be fitted to the dependence $C_p/T = \gamma + \beta T^2$, with $\gamma = 63.3$ mJ/K^2 mol. Below

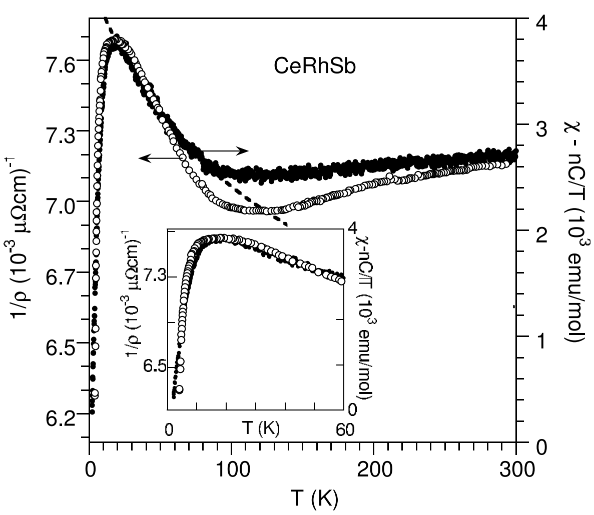

FIG. 2. Temperature dependences of the inverse resistivity ρ^{-1} (left scale) and of the paramagnetic susceptibility χ (with the impurity contribution nC_0/T, $n = 0.004$, subtracted) for CeRhSb. The dashed line displays the Curie-Weiss contribution to χ. The inset: ρ^{-1} and χ on an expanded scale.

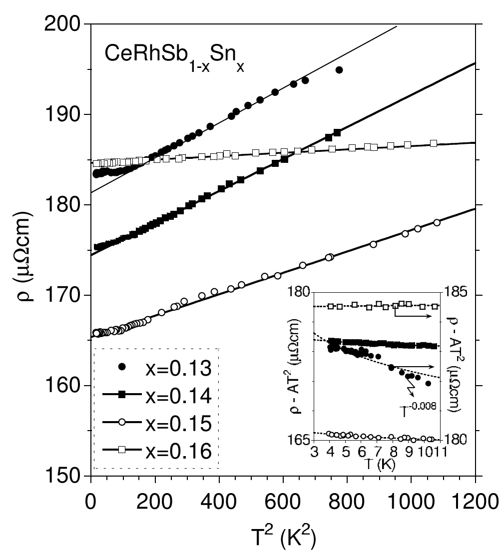

FIG. 3. Resistivity vs T^2 for the samples with $0.13 \leq x \leq 0.16$. The inset: non-Fermi-liquid-type scaling T^{-n}, $n \sim 0.01$, for the sample $x = 0.13$, i.e., close to the critical point where the Kondo gap disappears. The lines are the fitted curves.

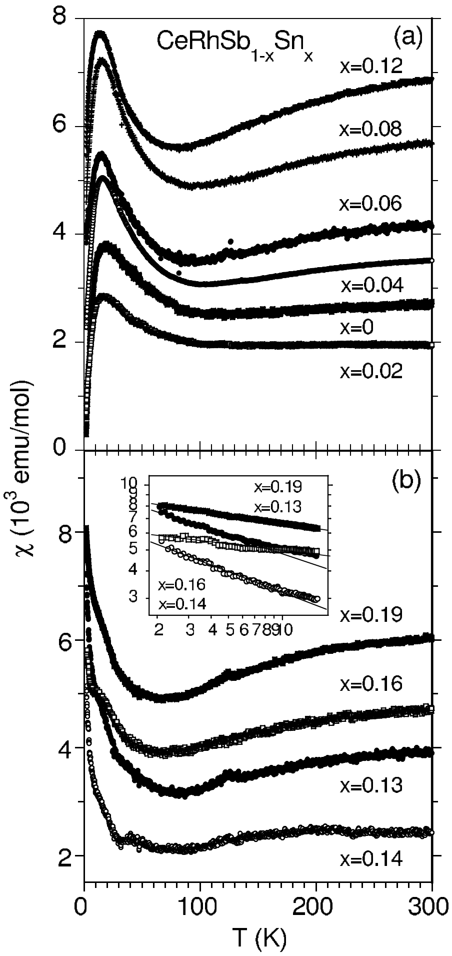

FIG. 4. Temperature dependence of ac susceptibility for (a) $x \leq 0.12$ and (b) $x \geq 0.19$. The inset illustrates the scaling $\chi \sim T^{-m}$ on the doubly logarithmic scale.

10 K the data start deviating from this dependence and the difference can be fitted to the relation $C_p/T = bT^{-s}$, with $b = 143.6$ mJ/K$^{1.7}$, and $s \simeq 0.3$. So again, as in the case of the resistivity, the NFL critical regime appears for $T \rightarrow 0$ from the gross Fermi-liquid-like state.

Two additional features of the analysis should be mentioned. The $\chi(T)$ data for $T > 15$ K and in the metallic phase ($x \geq 0.13$) can be parametrized in the form $\chi(T) = \chi_0 + C/(T - \Theta)$, with $\chi_0 \simeq (1.7\text{–}3.5) \times 10^{-3}$ emu/mol, a value that lies in the range for the heavy fermions (cf. Table I). Also, the value of the resistivity coefficient A decreases systematically with the increasing x and for $x = 0.15$ acquires the typical value 1.15×10^{-2} $\mu\Omega$ cm/K^2. The value of A remains finite when the system gradually transforms to the NFL state.

From our measurements an overall phase diagram can be drawn, as shown in Fig. 5. The solid line on the left is a guide to the eye representing the Kondo gap values $\Delta(x)$—filled circles, whereas the solid square on the right marks the point when the deviation from the FL behavior in ρ show up for $x = 0.13$. The empty squares mark the temperature [11], where a magnetic hysteresis disappears. It should be mentioned that the magnetic moment observed in the metallic samples with the help of the SQUID magnetometer is small and amounts to $(0.04\text{–}0.05)\mu_B/$Ce atom in the field of 5 T.

From the evolution of the x dependence of χ and ρ the following *physical picture* emerges clearly. First, the activated behavior of the conductivity is related to the fact that the nonzero carrier concentration n_c for $x \leq 0.12$ is caused by the thermal excitation across the Kondo gap Δ. The dependence $\rho \sim \exp(\Delta/T)$ is obeyed even for $T > \Delta$, in agreement with the well known fact that the Kondo semiconductors are systems with a low carrier concentration. Likewise, the magnetic susceptibility $\chi \sim n_c$, as the heavy carriers are activated from the Kondo-singlet bound state with $\chi \approx 0$. An elementary analysis shows that for quasiparticles $\rho = m^*/(n_c e^2 \tau) \sim n_c^{-1}$, so $\rho\chi =$ const, if only the carrier lifetime τ and the effective mass m^* do not change appreciably with either T or x (this is the case for $x \leq 0.06$). In this manner, the observed scaling is easily understood, at least in the low-temperature regime $T \leq 10$ K.

The system remains insulating at $T = 0$ when $x > 0$. This means that the *collective Kondo-lattice state* survives up to $x = 0.12$. The atomic disorder is not crucial because the gap value decreases with the increasing x. The susceptibility maximum in Fig. 4(a) takes place at $T_\chi = 18.6$ K for $x = 0$ and reduces to $T_\chi = 13.6$ K for $x = 0.12$. It is roughly equal to 2Δ ($x = 0$). The maximum presence reflects the situation when magnetic fluctuations are the strongest and the combined effect of thermal noise and spin-spin interaction becomes comparable to the effective Kondo coupling. At the critical point ($x \approx 0.12$) the Kondo-singlet state cannot be formed at any $T > 0$ and is outbalanced by the spin-spin interaction already at $T = 0$ and hence the susceptibility becomes divergent at $T = 0$.

TABLE I. Susceptibility and the resistivity parametrizations of the samples in the *metallic* regime ($x \geq 0.13$).

	$\chi = \chi_0 + C/(T - \Theta)$			$\chi = aT^{-m}$		$\rho = \rho_0 + AT^2$	$\rho - AT^2 \sim T^{-n}$
x	χ_0 (10^3 $\frac{\text{emu}}{\text{mol}}$)	Θ (K)	C ($\frac{\text{emu K}}{\text{mol}}$)	$10^3 a$	m	A (10^2 $\mu\Omega$ cm/K^2)	$10^3 n$
0.13	2.6	−7.0	0.043	6.0	0.17	1.78	8.0
0.14	1.7	−9.1	0.021	4.3	0.17	1.69	2.8
0.16	3.5	−7.6	0.038	6.0	0.09	0.186	1.3
	$15 \leq T \leq 90$ K			$T < 20$ K		$30 \geq T \geq 10$ K	$T < 10$ K

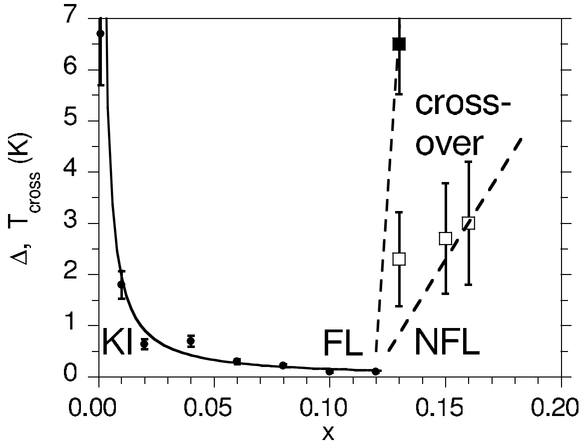

FIG. 5. Schematic phase diagram for the $CeRhSb_{1-x}Sn_x$ system. For details see main text.

This picture is also supported by the circumstance that χ systematically increases with $x \to x_c$. Thus, the evolution with increasing x is a practical realization of the *Doniach scenario* [12], albeit as a function of the electron concentration. However, this scenario should be modified to account for the itineracy of f electrons for $T < T_{max}$. As a result, for temperatures below that of the resistivity maximum (cf. Fig. 1), we enter the coherence regime for heavy-fermion metals. However, for the Kondo insulators the collective singlet bound state is gradually formed for $T < T_\chi$ and the *reentrant* localization effects appear with the further temperature decrease. Such a reentrant metallic behavior appears also for the Mott systems, except in a reverse order due to the difference in the nature of the magnetic ground state in those two systems [13].

In conclusion, we have shown that the Kondo semiconductors $CeRhSb_{1-x}Sn_x$, with $x \leq 0.12$ are characterized not only by a gap in conductivity and a vanishing paramagnetic susceptibility for $T \to 0$, but also by universal scaling law $\rho\chi = \text{const}(x)$. Also, the system undergoes the nonmagnetic Kondo insulator-magnetic metal transition at $x \approx 0.12$. On the metallic side, a novel quantum critical point and NFL behavior has been discovered for samples with $x \to 0.12+$ that is specified by the power laws ($T^{-\alpha}$) for the resistivity, the susceptibility, and the specific heat at lower temperatures. The simultaneous divergences of both ρ and χ at QCP illustrate nicely the circumstance that both the onset of the KI gap and the magnetic critical fluctuations' appearance coexist there. The transition is markedly different from the Mott-Hubbard transition. The non-Fermi-liquid behavior develops as $T \to 0$ from an overall Fermi-liquid state. Also, the quantum critical behavior described here as a function of the carrier number differs from that appearing at the paramagnetic heavy-fermion—antiferromagnetic metal boundary [4] and therefore defines a different class of quantum materials. Also, while the pressure-induced Kondo gap destruction in the stoichiometric system leads directly to a heavy Fermi liquid [5], we observe here a unique singular NFL state for $x > 0.12$ at low temperature. The role of atomic disorder in its formation should be studied further, particularly at very low temperatures.

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