

Electronic Griffiths phase and quantum interference in disordered heavy-fermion systems

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We investigated the specific heat and electrical resistivity of disordered heavy-fermion systems $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$. Results show that pronounced non-Fermi-liquid behavior in these Kondo disordered compounds originates from approaching metal-insulator transition rather than from proximity to magnetic instability. Power-law divergence of the local Kondo temperature distribution, $P(T_K)$, in the limit of $T_K \rightarrow 0$, and clear signature of the quantum interference corrections in the resistivity detected deep below the onset of Kondo coherent state, point to electronic Griffiths phase formation in the studied compounds.

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In the last three decades, a variety of new f -electron materials whose properties are fundamentally different from the predictions of Fermi-liquid (FL) theory have been discovered. Among several theoretical models that account for non-Fermi-liquid (NFL) behavior, there exist those directly associated with a quenched disorder in the crystal structure [1–8]. Usually, experimental data of such disordered f -electron systems are considered arbitrarily in the light of either Kondo-disorder model (KDM) [3,9,10] or magnetic Griffiths phases (MGP) [6,7,11,12]. Divergence of the thermodynamic properties in both scenarios is a result of a broad distribution of the characteristic energy scales, $P(E)$, which for enough strong disorder, acquires power-law dependence in the limit of $E \rightarrow 0$, regardless of whether the spin fluctuation is constrained to the single spin-flip mechanism (KDM) or some number of spins, which are locked together by their mutual interaction in clusters (MGP) [8].

On the other hand, it is known that induced by disorder, strong elastic scattering of conduction electrons is responsible for quantum interference effect, which reflects an approach to a disorder-driven metal-insulator transition (MIT). As a consequence, quantum interference of scattered electron waves produces fluctuations of the local density of states (LDOS) in conduction band [13] and causes the low-temperature increase in the resistivity due to modification of the electrons propagation through a random potential [14]. Remarkably, the fluctuations of the LDOS in conduction bands was also considered in the electronic Griffiths phase (EGP) approach to KDM as a dominating mechanism which is responsible for the NFL behavior of disordered Kondo systems [10].

Although there is still a growing number of compounds exhibiting NFL behavior induced by disorder [3,15–20], to the best of our knowledge, there are only a few examples including the Te-doped semiconductor FeSb_2 [21] and quasicrystalline $\text{Au}_{51}\text{Al}_{34}\text{Yb}_{15}$ [22], in which the emergence of the EGP was reported, while only in one Kondo-disordered compound, UCu_4Pd , quantum interference contribution to the resistivity was detected [23]. Here, we investigate two disordered

Kondo systems, $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, with pronounced NFL properties and minimum in the temperature dependence of electrical resistivity caused by quantum interference effects, simultaneously. This highlights quantum interference as a primary mechanism responsible for the enhancement of the LDOS fluctuations and EGP formation with power-law distribution of the T_K .

Earlier measurements performed on polycrystalline samples revealed that the system $\text{Ce}_2\text{Rh}_{1-x}\text{Co}_x\text{Si}_3$ evolves from the antiferromagnetic ground state in Ce_2RhSi_3 with T_N around 7 K [24] via a quantum critical point for $x = 0.6$ to a paramagnetic state in Ce_2CoSi_3 with pronounced mixed-valence properties [25,26]. Remarkably, $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ exhibits NFL properties, similar to those of $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, from which it follows that departure from the Fermi liquid is extended widely in paramagnetic range. Specific-heat coefficient C/T and susceptibility χ of these compounds was found to increase with lowering temperature in a manner characteristic of Griffiths phase power law $C/T \sim \chi \sim T^{(\alpha-1)}$ with $\alpha \approx 0.6$ and 0.5 for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively [27]; however, the question of the microscopic origin of the spin fluctuators involved remains.

In this study, we focus on the low-temperature results of the specific heat and electrical resistivity derived from our own measurements on single-crystalline $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$. The crystal structure and composition analysis of these two compounds was given in preliminary studies presented in Ref. [27] (see also the Supplemental Material [28]).

Figure 1(a) displays the magnetic contribution to the specific heat over temperature ratio, C_m/T [29], for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, as a function of T on a log-log plot. As seen, the C_m/T of the studied compounds increases with increasing temperature, while below around 4 K it acquires almost perfect power-law dependence. Remarkably, both KDM (solid lines) and MGP models (dashed lines), which are described in detail in the Supplemental Material [28], very well reproduce the derived specific-heat data below 20 K. Both of them might be expressed by an analogous relation: $C_{\text{MGP,KDM}}/T = \gamma_c + \int_0^\Lambda P(E)C/T d\Delta$, where γ_c is a regular term, Λ is a crossover scale, while C is a specific

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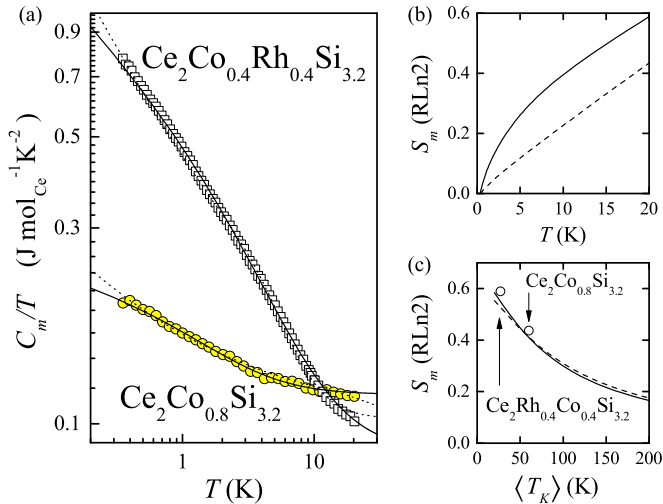


FIG. 1. (a) Specific-heat coefficient C_m/T of $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$ as a function of temperature. Dotted lines correspond to the MGP model with $\gamma_c = 0.102 \text{ J mol}^{-1} \text{ K}^{-2}$, $\Lambda = 100 \text{ K}$, $\alpha = 0.53$, $a = 0.0073$, and $\gamma_c = 0.103 \text{ J mol}^{-1} \text{ K}^{-2}$, $\Lambda = 17 \text{ K}$, $\alpha = 0.36$, $a = 0.046$ for Rh-free and Rh-doped compounds, respectively. The distribution is approximated by a power law $P(E) = aE^{(\alpha-1)}$. Solid lines represent KDM with $\gamma_c = 0.118 \text{ J mol}^{-1} \text{ K}^{-2}$, $\Lambda = 9 \text{ K}$, $\langle \lambda \rangle = 0.19$, $w = 0.0293$, and $\gamma_c = 0.086 \text{ J mol}^{-1} \text{ K}^{-2}$, $\Lambda = 22 \text{ K}$, $\langle \lambda \rangle = 0.165$, and $w = 0.035$ for Rh-free and Rh-doped compounds, respectively. Here, we use full form of distribution $P(E)$ [28], while ε_F was set to 1 eV. (b) Magnetic entropy S_m of $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ (dashed line) and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$ (solid line), calculated as an integral of C_m/T in the range 0.35–20 K. (c) S_m as a function of the average Kondo temperature $\langle T_K \rangle$ derived from KDM between 0.35 and 20 K for $\varepsilon_F = 1 \text{ eV}$ and $w = 0.03$ (solid line) and $w = 0.035$ (dashed line). Arrows indicate positions of the experimental values of the S_m for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$.

heat, which depending on the model considered is associated with either the tunneling clusters C_{Sch} (MGP) or single-ion spin fluctuation C_K (KDM). In this model the former specific-heat contribution was approximated by the Schottky specific heat C_{Sch} , by the analogy to tunneling two-level systems considered in the glasses [30]. In turn, the contribution to the specific heat C_K of nearly free magnetic moments with low T_K 's can be described by the resonant-level model for a Kondo impurity [31]. It is worth noting that C_K for spin impurity 1/2 has the shape of the broadened Schottky peak, therefore it is not surprising that heat-capacity data can be very well described by both considered models, making it difficult to conclude what is the microscopic origin of the NFL behavior in investigated f -electron compounds.

To distinguish between different Griffiths phases we estimate the typical size of the Griffiths droplets using entropy analyses. As shown in Ref. [8], if available entropy per Griffiths cluster is $S_{cl} = k_B \ln 2$, the entropy per mole of spin should be reduced from the value $R \ln 2$ by a factor of the order of typical number of spins per cluster N_ξ . The entropy, S_m , of $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$ calculated between 0.35 and 20 K reaches values of $0.4R \ln 2$ and $0.6R \ln 2$, respectively [see Fig. 1(b)], which might indicate that the underlying physics in considered compounds is related rather to the

single spin fluctuator. Here, to estimate the value of N_ξ more precisely, the idea of Miranda and Dobrosavlević was slightly modified (see Supplemental Material [28]). Consequently, the expected contribution to the entropy S due to tunneling clusters per mole of Ce^{+3} spins in the temperature interval 0.35–20 K can be calculated using the formula [28]

$$SN_\xi = R \int_{0.35 \text{ K}}^{20 \text{ K}} \frac{\int_0^\Lambda P(E) C_{\text{Sch}}/T dE}{\int_0^\Lambda P(E) dE} dT, \quad (1)$$

where the numerator represents the total contribution of the specific heat due to tunneling clusters with $E \leq \Lambda$, while the denominator defines the total number of tunneling clusters with $E \leq \Lambda$. Due to contribution of the regular term γ_c resulting from the droplets with $\Lambda > T$ and paramagnetic environment, the hypothetical experimental value of the entropy S associated with tunneling clusters would be given by the formula $S = S_m - S_p$ where S_p corresponds to $C_m/T = \gamma_c \approx 0.1 \text{ J mol}^{-1} \text{ K}^{-2}$. This yields entropy S equal to $0.09R \ln 2$ for Rh-free and $0.25R \ln 2$ for Rh-doped compounds. In turn, the right-hand side of this equation calculated for the parameters derived from the fitting (see caption, Fig. 1) are $0.56R \ln 2$ and $0.85R \ln 2$ for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively. As a result, from the ratio $0.85R \ln 2/S$ it was found that N_ξ is around 3, for $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, which leaves rather little room for large cluster formation. In turn, for the second compound the ratio $0.56R \ln 2/S$ yields $N_\xi \approx 6$, which from this point of view makes $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ a much better candidate for MGP formation. However, from the ‘‘Kondo necklace’’ picture of Doniach [32] one can deduce that the $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ should be considered as a compound distant from any magnetic phase transition. This, in turn, means that the local dimensionless Kondo coupling constant, λ , would be too large to allow the Ruderman-Kittel-Kasuya-Yoshida (RKKY) exchange interaction to stabilize local order and droplet formation.

Therefore, we consider a model with randomly distributed Kondo temperature $T_K(\lambda) = \varepsilon_F \exp(-1/\lambda)$ (ε_F is a Fermi energy), which results from the narrow Gaussian distribution of λ with an average $\langle \lambda \rangle$ and rms width w [3]. By analogy to the analyses performed within the MGP model, we split the C_m/T into a part which is responsible for the NFL behavior and a constant contribution γ_c resulting from the sites whose T_K is larger than a crossover scale Λ [33]. As shown in Fig. 1(a) (solid lines), the considered model provides a very good description of the data with $\langle \lambda \rangle = 0.19$, $w = 0.0293$ and $\langle \lambda \rangle = 0.165$, $w = 0.035$, for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively. On the other hand, fitting the experimental data below 4 K using power-law form $\gamma_c + aT^{(\alpha-1)}$ yields values of α equal to 0.54 for Rh-free and 0.48 for Rh-doped compounds, which turn out to be close to that determined in Ref. [27]. The resulting value of the average Kondo temperature $\langle T_K \rangle = 60 \text{ K}$ for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ is near the $T_K = 50 \text{ K}$ derived from the uniform Kondo model [34]. In turn, the $\langle T_K \rangle = 27 \text{ K}$ for $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$ is apparently smaller than for the former compound. Interestingly, as shown in Fig. 1(c), experimental values of S_m agree very well with those theoretically determined as a function of $\langle T_K \rangle$ for the above-derived parameters of the distribution $P(T_K)$.

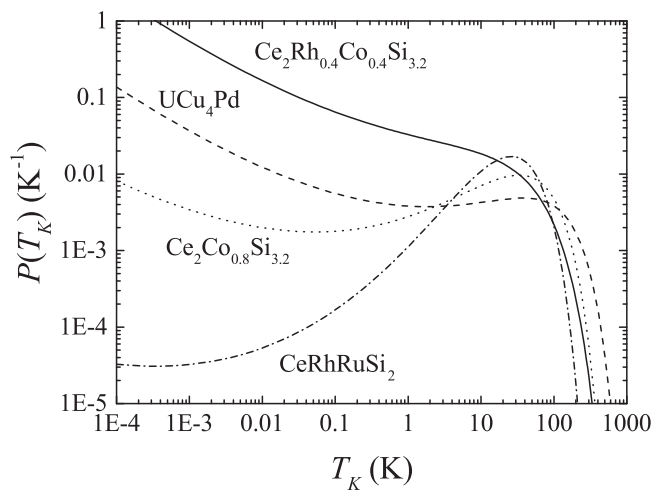


FIG. 2. Distribution of T_K 's derived from the specific-heat analysis. Solid and dotted lines represent $P(T_K)$ for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively. $P(T_K)$ of UCu_4Pd [3] (dashed line) and CeRuRhSi_2 [16] (dashed-dotted line) are shown for comparison.

The distribution of $P(T_K)$ of the considered compounds is shown in Fig. 2, together with the $P(T_K)$ of UCu_4Pd and CeRuRhSi_2 . The common feature of all these compounds is a finite probability of finding low- T_K spins, which generates the NFL behavior of thermodynamic properties. Moreover, at low T_K the probability functions of the investigated and UCu_4Pd compounds have an asymptotic power-law form characteristic of EGPs.

Figures 3(a) and 3(b) show the evolution of the low- T resistivity of $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively, with a magnetic field applied along the crystallographic ab

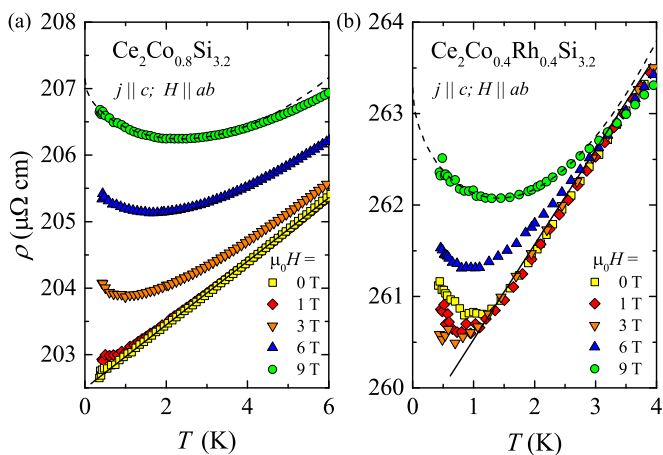


FIG. 3. Evolution of $\rho(T)$ at low T with external magnetic field for (a) $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and (b) $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively. Solid lines represent AT dependence with $A = 0.46 \mu\Omega \text{ cm K}^{-1}$ for the first compound and $A = 1 \mu\Omega \text{ cm K}^{-1}$ for the second compound. The dashed lines are fitted to the experimental data by relation $\rho_0 + A_{\text{FL}}T^2 - A_{\text{dis}}T^{1/2}$ (see the text for symbol description) with ρ_0 , A_{dis} , A_{FL} equal to $207 \mu\Omega \text{ cm}$, $0.77 \mu\Omega \text{ cm K}^{-1/2}$, $0.052 \mu\Omega \text{ cm K}^{-2}$ and $263 \mu\Omega \text{ cm}$, $1.5 \mu\Omega \text{ cm K}^{-1/2}$, $0.22 \mu\Omega \text{ cm K}^{-2}$ for $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$ and $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, respectively.

plane. The linear dependence of the zero-field resistivity observed in a broad temperature range is an evident manifestation of the NFL behavior. It might seem that this observation is in contrast to the KDM, which predicts negative linear-in- T contribution to the resistivity [9]. However, it is worth emphasizing that this model assumes only the incoherent nature of the transport caused by only a small fraction of the spins with $T_K < T$, which are not effectively frozen [4]. In turn, the temperature dependence of electrical resistivity $\rho(T)$ of the studied compounds exhibits Kondo-lattice behavior with a characteristic maximum of the resistivity at coherent Kondo temperature T_{coh} (see Fig. 2 in Ref. [27]). This indicates that nonmagnetic disorder inherent in the crystal structure does not kill the coherence entirely, although we should mention that the drop in resistivity below T_{coh} is significantly smaller than in clean heavy-fermion (HF) compounds. Therefore, it may suggest that disorder-induced distribution of the RKKY exchange interaction between some Ce^{+3} spins has to be also considered in parallel to the distribution of the Kondo interaction. Interestingly, the increase of the nonmagnetic disorder does not change the power-law decay of all the even moments of the RKKY interaction, but due to quantum interference correction results in a gradual enhancement of the amplitude of the RKKY interaction upon approaching Anderson localization [35]. Consequently, there is a finite probability to find the randomly located unscreened spins, for which the RKKY will dominate over the Kondo interaction. Since the effective interactions between such spins will be random in magnitude and sign, it might provide more complex ground-state properties of the disordered HF systems including random singlet, nanoscale ferromagnetism, spin-liquid, or spin glass formation [1, 36–39]. It would be especially relevant to the disordered HF systems close to the magnetic phase transition. This also may concern $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$, for which resistivity below around 1.5 K deviates from the linear-in- T dependence, more likely caused by the quenching of the free unscreened spins of Ce^{+3} by other local moments.

Remarkably, the quantum interference correction that modifies the RKKY interaction is of the same nature as that of the quantum correction to the conductivity in disordered electronic systems. Such corrections usually have very small values, making the anomaly difficult to observe experimentally. This is particularly relevant to the disordered HF, where despite enhanced elastic scattering, the dynamics of the resistivity variation with temperature below T_{coh} is still relatively strong, due to inelastic scattering of conduction electrons [40]. Interestingly, we found that zero-field resistivity of $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$ increases below around 1 K in a manner similar to the disordered electron systems. The position of the minimum in the $\rho(T)$ shifts toward lower temperatures at initial fields, while for $B > 3$ T the upturn in $\rho(T)$ becomes more prominent. As regards $\text{Ce}_2\text{Co}_{0.8}\text{Si}_{3.2}$, its resistivity gradually departs from the perfect linear-in- T behavior with increasing magnetic field, which undoubtedly manifests the field-induced FL in line with previously obtained C/T data [27]. However, it is worth underlining that above a field of 3 T, ρ does not follow the AT^2 relationship expected for the FLs. In turn, the sample develops a low- T upturn in ρ that becomes more evident with increasing magnetic field, and this same minimum that appears in the $\rho(T)$ shifts toward higher temperatures,

in a manner similar to $\text{Ce}_2\text{Co}_{0.4}\text{Rh}_{0.4}\text{Si}_{3.2}$. Interestingly, in a strong magnetic field of 9 T, in which the resistivity minima of the compounds studied becomes most evident, $\rho(T)$ can be described by the relation $\rho_0 + A_{\text{FL}}T^2 - A_{\text{dis}}T^{1/2}$, where the first term corresponds to the residual and spin disorder resistivity, the second term is a FL contribution due to collisions between conduction electrons, and the last term is a diffusion Altshuler-Aronov correction resulting from quantum interference effects in disordered metallic systems [14]. The correction comprises the exchange-like ($j = 0$) term and the Hartree-like ($j = 1$) term, which correspond to the different values of the total spin j of an electron and a hole, and contribute to the total correction with opposite signs [14]. Although the Hartree term is responsible for a decrease of the resistivity with lowering temperature its contribution is usually smaller than that resulting from the exchange term. Moreover, under the magnetic field applied the singularity of the Hartree term with $S_z = \pm 1$ is cut off by a Zeeman splitting effect of the conduction band. This leads to the positive magnetoresistivity, which decreases with the increasing temperature, which means that the correction is even more evident than in zero magnetic field [41]. Remarkably, this behavior closely resembles that reported for the crystallographically disordered AIB₂-type structure compounds U_2CoSi_3 [42] and U_2NiSi_3 [43]. It was shown that this singular $T^{1/2}$ term, observed also in many disordered conductors independent of their ground-state properties [42–50], represents the electron-electron interaction correction, and

reflects, similar to the weak localization correction, the approach to the disorder-driven MIT.

In summary, we demonstrated that the singular thermodynamic and transport properties of the investigated disordered HF systems arise from the disorder-induced MIT, rather than from the proximity of any magnetically ordered phase. This incipient electron localization manifests itself directly as a quantum interference correction to the resistivity in the coherent-Kondo state. Remarkably, quantum interference of elastically scattered electron waves is not only responsible for the low- T upturn in the resistivity but also causes the LDOS fluctuations in conduction bands [13], which results in the enhancement of the T_K fluctuation. In turn, this effect is crucial for EGP formation, leading to the power-law divergence of $P(T_K)$ in the limit $T_K \rightarrow 0$, and the NFL behavior in the thermodynamic properties. This highlights the importance of the elastic scattering of conduction electrons and quantum interference effects as a primary source of the NFL behavior of the investigated disordered HF systems. We believe that these findings will prompt further research into the interplay between strong electronic correlations and disorder.

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