Metallic Ground State and Glassy Transport in Single Crystalline URh_2Ge_2: Enhancement of Disorder Effects in a Strongly Correlated Electron System

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We present a detailed study of the electronic transport properties on a single crystalline specimen of the moderately disordered heavy-fermion system URh_2Ge_2. For this material, we find glassy electronic transport in a single crystalline compound. We derive the temperature dependence of the electrical conductivity and establish metallicity by means of optical conductivity and Hall effect measurements. The overall behavior of the electronic transport properties closely resembles that of metallic glasses, with at low temperatures an additional minor spin disorder contribution. We argue that this glassy electronic behavior in a crystalline compound reflects the enhancement of disorder effects as a consequence of strong electronic correlations.

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The interplay of disorder and electronic correlations represents a central issue of the physics of strongly correlated electron systems (SCES). The properties of SCES are extraordinarily sensitive to small levels of crystallographic disorder. This is attributed to the electronic correlations, which enhance the effect of the disorder [1]. Correspondingly, disorder has been proposed to greatly modify or control the physical properties of SCES in very many cases [2,3]. A thorough understanding of this issue, however, be it from the material physics point of view [4] or concerning the process of localization in correlated electron systems [5,6], is lacking.

Heavy-fermion metals represent an archetypical class of intensively studied SCES [7]. For heavy fermions, disorder effects have been found to affect the behavior close to magnetic instabilities, as in Ce-based compounds [8] or in U intermetallics as UCu_4Pd or $(U, Th)Pd_2Al_3$ [9]. Especially, the electronic transport properties of these U heavy fermions have been interpreted in terms of non-Fermi-liquid (NFL) behavior. Rather than exhibiting a generic heavy-fermion metallic resistivity, with a $\ln(T)$ dependence at high temperatures T and a crossover below T_{coh} into a coherent state with a metallic resistivity $\rho = \rho_0 + AT^2$, for these materials a resistivity deviating from Fermi liquid predictions is found, with $\rho = \rho_0[1 - a(T/T^*)^n]$, $n \sim 1$–1.5 [10]. Yet, the resistivities of these and various related materials are highly unusual in other aspects as well, as they remain—for metallic systems—large (many hundred $\mu\Omega$ cm) and exhibit a negative temperature coefficient of the resistivity (TCR), $d\rho/dT < 0$, down to lowest temperatures [11–13].

Metallurgically, these U compounds with a negative TCR are ''moderately'' disordered, i.e., crystallographic randomness on the atomic level of $\sim$10 vol %. Under such circumstances, the question about the origin of the anomalous transport arises: Is the negative TCR predominantly the result of quantum spin fluctuations and hence an indication for a NFL [9]? Or is it the result of the combined influence of strong electronic correlations and disorder-induced localization, which should be treated in the framework of Anderson vs Mott-Hubbard localization [14,15]. In order to address such issues we have performed a detailed study of the electronic transport properties of a U heavy-fermion compound with moderate and known disorder, namely, URh_2Ge_2 [16].

Previously, we have established this material as the first 3D random-bond, heavy-fermion Ising-like spin glass [16]. Further, we have determined type and level of the crystallographic disorder. The system crystallizes in the tetragonal $CaBe_2Ge_2$ lattice ($P4/nmm$). Disorder is present on the nonmagnetic ligand sites in the form of moderate bond length disorder, which likely is the result of $\sim$5%–10% Rh/Ge random site exchange [17]. In contrast, the U ions occupy translationally invariant positions on an ordered tetragonal sublattice. The bond length disorder, in as-grown single crystals, generates the spin glass ground state below $T_f \simeq 9$ K.

The electronic transport even in single crystalline URh_2Ge_2 follows that of an archetypical moderately disordered U heavy-fermion compound [12]. The absolute resistivity ρ along a and c axes ranges from 200 to 800 $\mu\Omega$ cm, with large sample-to-sample variations, while the TCR is negative up to well above 300 K. Below $\sim$10 K, in close resemblance to UCu_4Pd, the resistivity exhibits a NFL-like temperature dependence $\rho = \rho_0[1 - aT]$. While in UCu_4Pd the negative TCR has been interpreted within the framework of the disordered Kondo or Griffiths phase scenario, for various reasons in the spin glass URh_2Ge_2 this does not reflect a NFL ground state in the spirit of Refs. [2,9]. First, a single ion Kondo-like behavior of the resistivity $\propto \ln T$ is not observed. Second, the ρ values in URh_2Ge_2 by far exceed those obtained from the Ioffe-

Regel criterion, indicating substantial electronic localization. Third, while the models in Ref. [9] assume a distribution of Kondo temperatures T_K from 0 to some finite value, with the finite freezing temperature T_f in URh_2Ge_2, such a wide T_K distribution is unlikely. Finally, the anisotropic response of ρ to an annealing treatment, yielding a heavy-fermion metallic behavior along the a axis and a negative TCR along the c axis, cannot be reconciled with single ion Kondo scattering models or their extensions as put forth in Ref. [9]. Thus, the mechanism which actually controls the electronic transport has not been resolved.

In order to elucidate the physical mechanisms behind this highly unusual behavior, we have performed a thorough study of the electronic transport properties of single crystalline spin glass URh_2Ge_2. In this Letter, we present evidence for glassy electronic transport in single crystalline URh_2Ge_2. We extract a generic temperature dependence of the electrical conductivity σ and establish the metallicity of the material by means of optical conductivity and Hall effect measurements. Based on its temperature and magnetic field dependence, we distinguish between two conductivity components. The overall behavior of σ resembles that of metallic glasses; hence, we argue that it is primarily governed by disorder-induced localization [15]. In addition, at low T there is a secondary contribution from spin disorder scattering.

For our experiments we have investigated an as-grown single crystalline specimen URh_2Ge_2 previously used in an x-ray-absorption fine structure study [17]. Other pieces of this single crystal, with slightly different absolute ρ values, have been studied in Refs. [12,16]. The spin glass ground state below $T_f \simeq 9$ K has been established via susceptibility measurements. In Fig. 1(a) we plot the resistivity ρ, measured along the a and c axes. We observe the archetypical behavior of moderately disordered U heavy-fermion compounds [12]. The crystalline anisotropy is reflected in the anisotropy of ρ along the a and c axes.

In Fig. 1(b) we plot the reduced conductivity $\Delta\sigma = \sigma - \sigma_0$ as a function of T [12]. After multiplying the a axis data with a constant factor 1.77, $\Delta\sigma$ vs T for a and c axes superimpose over the full temperature range 1.5–300 K. This proves that the electronic transport along the two directions is governed by the same mechanisms with a generic T dependence.

The T dependence of $\Delta\sigma$ closely resembles the behavior of 3D amorphous metals. We quantify the resemblance by describing our data in terms of the corresponding localization theory [15], which successfully accounts for the conductivity of paramagnetic metallic glasses, amorphous ferromagnets, or icosahedral U spin glasses [18,19]. For weak electronic correlations (in URh_2Ge_2 at sufficiently high T) the T dependence of σ is attributed to the superposition of incipient localization, destroyed by inelastic scattering with phonons and electrons, and electronic interaction effects. It is given by [18]

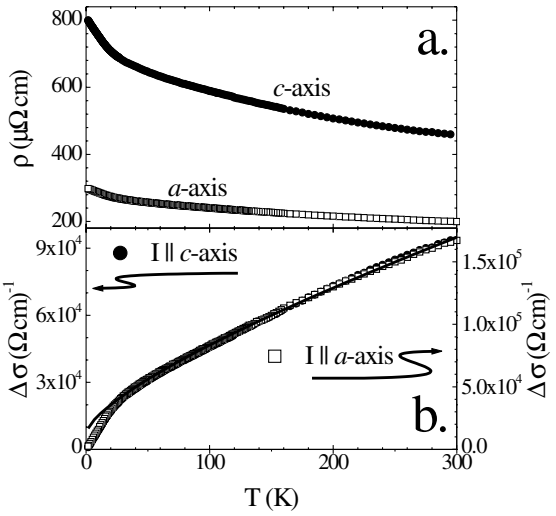

FIG. 1. (a) The resistivity ρ and (b) the reduced conductivity $\Delta\sigma$ as function of temperature of single crystalline URh_2Ge_2, measured along the tetragonal a and c axes. The solid line in (b) represents the result of a fit; for details, see the text.

$$\Delta\sigma(T) = \frac{e^2}{2\pi^2\hbar}(3\sqrt{b + c^2T^2} - cT - 3\sqrt{b} + d\sqrt{T}), \quad (1)$$

with fit parameters $b = 1/D\tau_{so}$, $c = \sqrt{1/4D\beta}$, $\beta = \tau_i T^2$ [20], and $d = 0.7367\sqrt{k/D\hbar}$ [diffusion coefficient D, spin-orbit (τ_{so}) and inelastic (τ_i) scattering times]. Above 30 K the T dependence of $\Delta\sigma$ is well described by Eq. (1), thus validating our statement on the close resemblance to the behavior of amorphous metals. In Fig. 1(b) we include the result of a fit to the data as a solid line, using parameters [21] along the a/c axis of $D = 0.54(20)/1.7(5) \times 10^{-6}$ m^2/s; $\tau_{so} = 82(68)/82(56) \times 10^{-12}$ s; $\beta = 50(29)/38(15) \times 10^{-10}$ sK^2. The values of the diffusion coefficient D are smaller by about an order of magnitude than those of common metallic glasses [18]. With the Einstein relation $\sigma_0 = e^2 D N(E_f)$, it reflects the much larger density of states $N(E_f)$ in URh_2Ge_2 [12]. Conversely, the inelastic scattering times τ_i are larger by an order of magnitude in URh_2Ge_2, implying that the inelastic diffusion lengths L_i are of the same order of magnitude as in metallic glasses [$L_i = \sqrt{D\tau_i} \sim O(10^{-7})$ m at low T] [18].

In applying Eq. (1) to URh_2Ge_2 we assume a metallic carrier density. To verify this assumption we have performed optical conductivity and Hall effect measurements. In Fig. 2 we plot the result of the optical conductivity study, with the real part $\sigma_1(\omega)$ along the crystallographic a axis obtained via the Kramers-Kronig analysis of the reflectivity (inset of Fig. 2). Within experimental resolution, there is no temperature dependence of σ_1, reflecting the weak overall temperature dependence of $\rho(a)$ [Fig. 1(a)]. The extrapolation of σ_1 to zero frequency yields a value of about 3000 $(\Omega\,\mathrm{cm})^{-1}$, in good agreement with the value extracted from $\rho_0^{-1}(a)$.

The overall behavior of $\sigma_1(\omega)$ qualitatively and quantitatively resembles that of other heavy-fermion metals [22], with a Drude-like conductivity at low ω and a maximum in σ_1 in the midinfrared regime from optical interband excitations. This observation verifies the metallic ground state of URh_2Ge_2 and disproves semiconductor or Kondo insulator scenarios to account for the electronic transport of URh_2Ge_2. Hence, the negative TCR must be the result of the crystallographic disorder.

To quantitatively characterize the metallic ground state of URh_2Ge_2, we have measured the Hall constant R_H along a and c axes (Fig. 3). The Hall constant has been derived from data taken below 1 T. In this field range, both R_H as the susceptibility χ are constant in field. For both directions R_H is dominated by anomalous Hall contributions. This is illustrated in Fig. 3 by including χ (measured in $B = 0.05$ T). Down to lowest temperatures R_H essentially follows χ, i.e., $R_H = R_0 + \chi R_S$ (R_0: ordinary Hall contribution; R_S: anomalous Hall coefficient). Assuming a spherical Fermi surface in a one band model, from these data we derive a carrier density n of ~ 3 carriers per unit cell for both axes.

Starting from the periodic Kondo lattice Anderson model, Kontani and Yamada [23] predicted a T dependence of the anomalous Hall contribution in heavy fermions $\propto \chi$ above T_{coh}. In URh_2Ge_2 coherence is suppressed with the crystallographic disorder, and thus $T_{\mathrm{coh}} = 0$. Accordingly, the prediction of Ref. [23] for $T > T_{\mathrm{coh}}$ would describe our experimental observation.

Alternatively, in disordered media the side jump effect contributes to the anomalous Hall contribution such that $R_H = R_0 + \chi\rho^2 R_S$ [24], with ρ as the total electrical resistivity. In Fig. 3 we include the normalized T dependence of $\chi\rho^2$ (dashed lines). It also reproduces the overall behavior of R_H and yields a carrier density of one to two carriers per unit cell.

Conversely, $R_H(T)$ of URh_2Ge_2 clearly is at variance with the Skew scattering prediction of Fert and Levy [25], $R_H = R_0 + \chi\rho_{\mathrm{mag}} R_S$ (ρ_{mag}: magnetic resistive contribution). Within this model, ρ_{mag} approaches zero for $T \to 0$, resulting in a drastic decrease of R_H at low temperatures and which is not observed in URh_2Ge_2.

Integrating the optical conductivity (Fig. 2) with respect to the effective metallic (Drude) component (i.e., from 0 up to about 1000 cm^{-1}) we obtain a plasma frequency of about 7000 cm^{-1}. Assuming a band mass $m_b = m_e$ we estimate a free charge carrier concentration of about 5×10^{20} cm^{-3}. The corresponding estimated R_H constant is about a factor of 10 larger than measured. Such a discrepancy can be easily accounted for by a too small band mass as well as by an underestimation of the effective free charge carriers spectral weight. Extending the integration of $\sigma_1(\omega)$ up to about 1 eV would lead to perfect agreement, within the simple assumption $m_b = m_e$, with the measured Hall constant (the dashed line in Fig. 2 represents the corresponding fit of the Drude component to the conductivity of URh_2Ge_2).

To further characterize the electronic transport in URh_2Ge_2 we have performed longitudinal and transversal magnetotransport experiments between 2 and 300 K in fields up to 5 T. As a representative, in Fig. 4 we depict the longitudinal magnetoresistivity (MR) for the magnetic field $B \parallel c$, $[\rho(B) - \rho(B = 0)]/\rho(B = 0) = \Delta\rho/\rho$, of

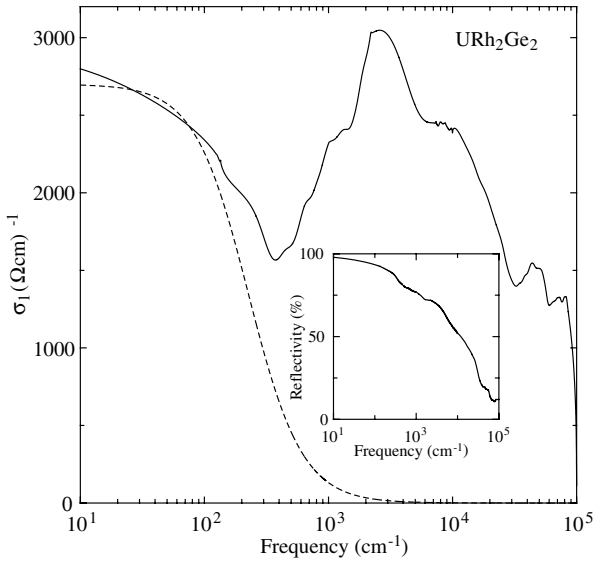

FIG. 2. The real part σ_1 of the a axis optical conductivity of URh_2Ge_2 at 10 K. The dashed line represents the result of a fit of the Drude component to the spectrum. The inset depicts the corresponding reflectivity spectrum.

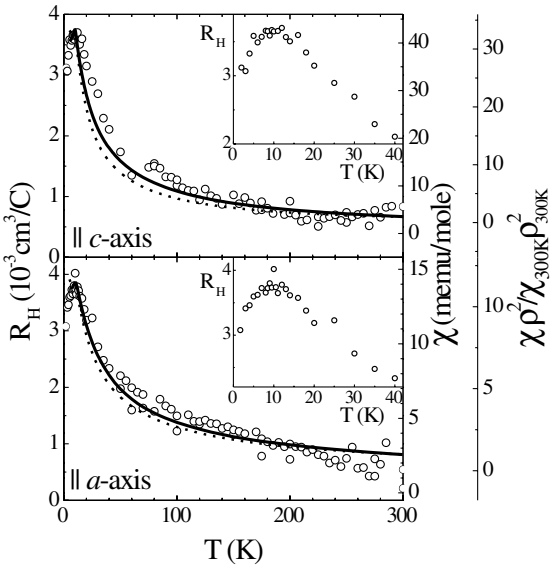

FIG. 3. The Hall constant R_H of URh_2Ge_2 along the a and c axes. The insets depict the low temperature regime, with the spin glass freezing transition causing the cusplike anomaly. The magnetic susceptibilities χ and the products $\chi\rho^2$ are displayed as solid and dashed lines, respectively; for details, see the text.

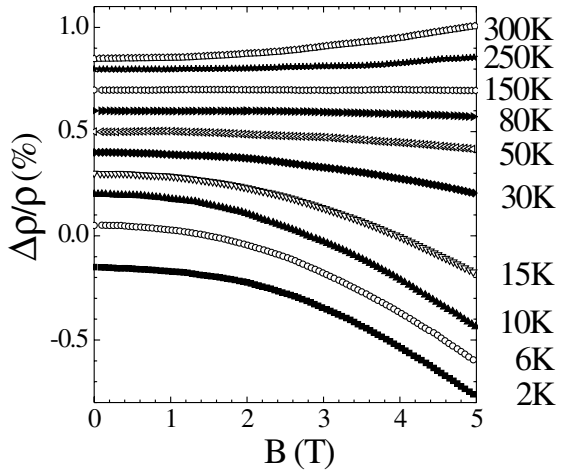

FIG. 4. The longitudinal magnetoresistivity of single crystalline URh_2Ge_2 between 2 and 300 K, measured for the $B \parallel c$ axis. Data are offset for clarity.

URh_2Ge_2. For the other experimental geometries the size of the MR is smaller by 1 order of magnitude than for the case depicted in Fig. 4, but otherwise very similar in behavior [26]. At all temperatures, for fields up to 5 T the MR is small ($< 0.7\%$) and follows essentially a B^2 dependence. While at low T, the magnetoresistivity is negative, surprisingly, it changes sign as temperature increases to above $\sim$100 K.

We have previously demonstrated that the negative MR at low T stems from the reduction of spin disorder scattering [12]. In contrast, a fundamentally different process is required to account for the positive MR at high T. For metallic glasses, in the limit $g\mu_B B/k_B T \ll 1$ both spin-orbit and interaction effects yield a positive $\Delta\rho/\rho \propto B^2$ [15,27], just as observed for URh_2Ge_2 above $\sim$100 K. While a full quantitative analysis fails because of parameter interdependency, with the above values for D and β the magnitude of the MR at $T \gtrsim 100$ K is reproduced using reasonable values for the interaction parameter γF_σ between 0 and 3 [27]. Therefore, we ascribe the positive MR at high T to incipient localization effects and electronic interactions in the spirit of Eq. (1) [15,18].

To summarize, with our study on URh_2Ge_2 we have for the first time fully characterized the electronic transport properties of a moderately disordered heavy-fermion compound. Our quantitative analysis demonstrates that the electronic transport in our single crystalline strongly correlated electron system with moderate, nonmagnetic site disorder closely resembles the behavior of metallic glasses. Thus, we find an amorphous behavior in an essentially crystalline material, reflecting the enhancement of disorder effects with electronic correlations.

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