Nernst Effect: Evidence of Local Kondo Scattering in Heavy Fermions

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A distinctly temperature-dependent Nernst coefficient, ν, which is strongly enhanced over that of $LaCu_2Si_2$, is observed between $T = 2$ and 300 K for $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$. The enhanced $\nu(T)$ is determined by the asymmetry of the on-site Kondo (conduction electron $-4f$ electron) scattering rate. Taking into account the measured Hall mobility, μ_H, the highly unusual thermopower, S, of these systems can be semiquantitatively described by $S(T) = -\nu(T)/\mu_H(T)$, which explicitly demonstrates that the thermopower originates from the local Kondo scattering process over a wide temperature range from far above to well below the coherence temperature ($\approx$20 K for $CeCu_2Si_2$). Our results suggest that the Nernst effect can act as a proper probe of local charge-carrier scattering. This promises to impact on exploring the unconventional enhancement of the thermopower in correlated materials suited for potential applications.

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Strongly correlated electron systems in various material classes like d- and f-electron-based intermetallics, organic charge-transfer salts, and transition-metal oxides have been in the focus of condensed-matter research for the past three decades. These systems give rise to a number of intriguing emerging phenomena, such as high-T_c superconductivity [1,2], colossal magnetoresistance [3], and heavy-fermion (HF) behavior [4], including HF superconductivity [5]. Apart from the fundamental interest, it is the potential for technical applications which has been fueling the exploration of strongly correlated materials.

Because of their giant thermoelectric power (TEP), Kondo systems are considered especially promising for thermoelectric applications at cryogenic temperatures [6]. In the case of dilute Kondo alloys, e.g., (La, Ce)Al_2 [7], the giant TEP derives from the on-site scattering of the conduction electrons by the local $4f$ electrons [8–10]. For dense Kondo or HF systems, on the other hand, the enhanced TEP is commonly ascribed to a weakly dispersive band of heavy quasiparticles, caused by the coherent hybridization between the $4f$ electrons and the conduction bands [11,12]. This view is strictly correct only at sufficiently low temperatures, $T \ll T_{\mathrm{coh}}$, the temperature below which phase coherence develops in the Kondo scattering. On the other hand, research aiming at thermoelectric applications focuses on the giant TEP at finite temperatures. In this case, a devoted theoretical treatment must account not only for the Kondo scattering, but also for the crystal-electric-field (CEF) splitting of the $4f$ states and their hybridization with the conduction electrons [9,10].

In this Letter, we present an experimental study of the Nernst effect combined with Hall-effect measurements, which allow for a semiquantitative description of the complex TEP behavior of a canonical HF metal based on local scattering processes in a wide temperature range, from room temperature to $T < T_{\mathrm{coh}}$. For this study, we have chosen the stoichiometric HF compound $CeCu_2Si_2$ and its 20 at.% La-doped variant. As previously shown, their TEP [13–15] exhibits a broad positive peak at $T \approx 160$ K, followed by a pronounced negative one at $T \approx 20$ and $\approx$12 K, respectively, see Fig. 1(b). These subsequent extrema are reflecting the single-ion Kondo temperatures (i) $T_{K,\mathrm{high}}$ of the $J = 5/2$ Hund's rule multiplet state of Ce^{3+}, involving higher CEF states, and (ii) T_K of the CEF doublet ground state [16]. T_K obtained from the TEP agrees well with the value estimated from the molar electronic entropy ($S_e \approx R\ln 2$). It also coincides with the position of the low-temperature peak in the electrical resistivity curve, $\rho(T)$, which is a good empirical measure of the coherence temperature, i.e., $T_{\mathrm{coh}} \approx 20$ K for $CeCu_2Si_2$; see inset of Fig. 2. Such a concurrence of T_K and T_{coh} has already been stated for $CeCu_2Si_2$ [17] as well as the isostructural HF metals $YbRh_2Si_2$ [18] and $CeNi_2Ge_2$ [19]. For comparison, we also present results on the nonmagnetic reference compound $LaCu_2Si_2$.

Polycrystalline $Ce_{1-x}La_xCu_2Si_2$ samples with $x = 0$, 0.2, and 1.0 were prepared as described elsewhere [15]. The measurements of the TEP $S(T)$, Nernst coefficient $\nu(T)$, and Hall coefficient $R_H(T)$ were carried out between 2 K and room temperature, with a typical sample dimension of $5 \times 2 \times 0.5$ mm^3. For the Nernst effect and TEP measurements, we employed one chip resistor (2000 Ω) as heater and one thin ($\phi = 25$ μm) chromel-$AuFe_{0.07\%}$ thermocouple for detecting the temperature gradient. The Nernst coefficient at each fixed temperature was measured in magnetic fields applied in opposite directions to cancel out the longitudinal TEP component. This procedure was further repeated at least five times to improve the resolution, from which we estimated the measurement errors at typical temperatures [cf. Fig. 1(a)]. No obvious

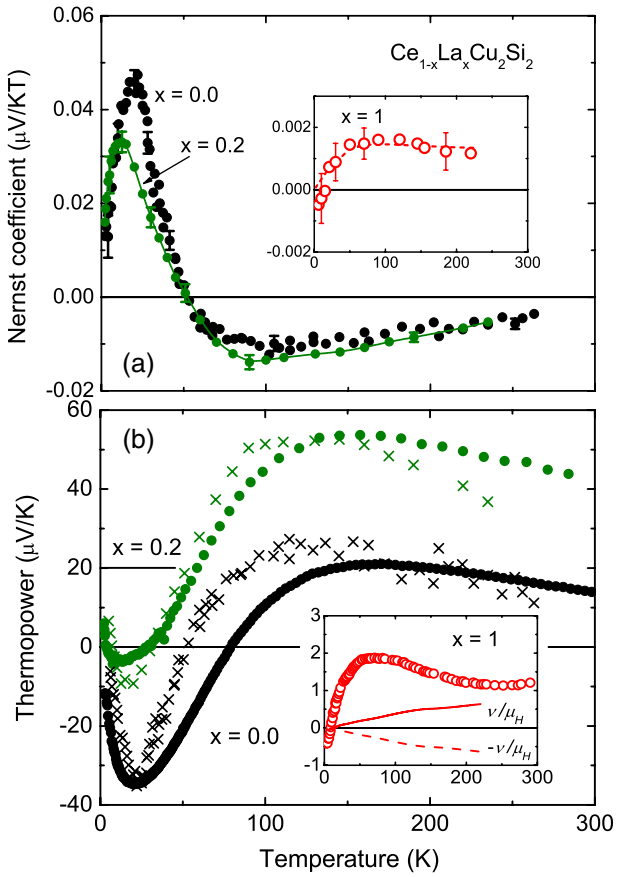

FIG. 1 (color online). (a) Nernst coefficient $\nu(T)$ of $CeCu_2Si_2$, $Ce_{0.8}La_{0.2}Cu_2Si_2$ (main panel) and $LaCu_2Si_2$ (inset). Dashed line in the inset was calculated for $LaCu_2Si_2$ under the assumption of dominating acoustic-phonon scattering, with $\tau(\epsilon) \propto \epsilon^r$ and $r = -(1/2)$; cf. Eq. (2) [23]. (b) Thermopower $S(T)$ compared to the ratio $-\nu/\mu_H$ vs T (crosses) for the same compounds as in (a). For $Ce_{0.8}La_{0.2}Cu_2Si_2$ the vertical axis is shifted upward by 20 μV/K for the sake of clarity. In contrast to the cases of $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$, no agreement is observed for $LaCu_2Si_2$ between $\pm(\nu/\mu_H)$ vs T and $S(T)$ (inset). Axes in the insets are in the same units used for the respective main panels.

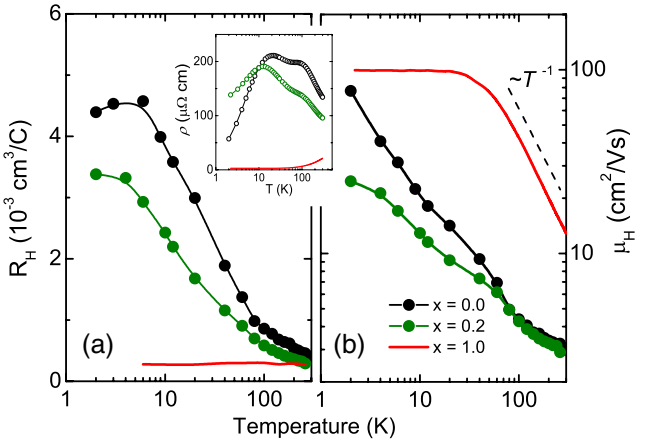

FIG. 2 (color online). Hall coefficient $R_H(T)$ in a semilog plot (a) and Hall mobility $\mu_H(T)$ in a double-log plot (b) for $CeCu_2Si_2$, $Ce_{0.8}La_{0.2}Cu_2Si_2$, and $LaCu_2Si_2$. Inset displays the temperature dependence of the electrical resistivity for the same compounds in a semilog representation.

magnetic-field dependence of $\nu(T)$ is observed up to 7 T and down to 2 K for the samples under investigation. This holds true also for $S(T)$ and $R_H(T)$.

As displayed in Fig. 1(a), the Nernst coefficient $\nu(T)$ is similar for $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$. It exhibits a broad negative peak at $T \approx 100$ K, and a positive peak at 20 and 12 K, respectively. By contrast, $\nu(T)$ of $LaCu_2Si_2$ [inset of Fig. 1(a)] shows a completely different temperature profile, with its values less than 2 nV/KT over the whole T range measured, spanning vertically a window that is a factor of 30 smaller than for $CeCu_2Si_2$. Note that $\nu(T)$ of the two Ce-based systems bears large resemblance to their TEP vs temperature results [cf. Fig. 1(b)], except for the opposite sign [20]. The complex TEP behavior as observed in $CeCu_2Si_2$ has been explained by Zlatić *et al.* [9,10] who consider the renormalized f-electron spectral weight distribution as a function of temperature in response to the interplay of multiple energy scales, namely the single-ion Kondo effect, the CEF splitting, and the strength of the hybridization of the conduction band with the local $4f$ states.

Following the description given by Mott [22], the thermoelectric tensor is determined by the logarithmic energy derivative of the electrical conductivity tensor. For a degenerate electron system, the diagonal term, i.e., the TEP is described by

$$S = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\left[\frac{\partial \ln\tau}{\partial \epsilon} + \frac{\partial \ln N}{\partial \epsilon}\right]_{\epsilon=\epsilon_F}, \quad (1)$$

with $\tau(\epsilon)$ denoting the relaxation time of the conduction electrons, and $N(\epsilon)$ denoting the electronic density of states (DOS) at the Fermi level ϵ_F. The scattering events usually encountered involve a power-law dependence of the scattering time $\tau(\epsilon) \sim \epsilon^r$, with $r \approx 1$, which implies a weak energy dependence of $\tau(\epsilon)$. Therefore, one of the prevailing strategies for designing efficient thermoelectric materials is to search for a largely asymmetric DOS at ϵ_F. This may be realized in materials with complex band structure and/or significant correlation effects [6]. The first term in Eq. (1), which is due to asymmetric scattering, has been practically ignored in most experimental and theoretical investigations, particularly on conventional, i.e., uncorrelated or only weakly correlated systems.

Like its diagonal counterpart (TEP), the off-diagonal terms of the thermoelectric tensor define the Nernst coefficient, which can be expressed by the derivative of the Hall angle $\tan\theta_H$ [21,23,24],

$$\nu = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{1}{B}\frac{\partial \tan\theta_H}{\partial \epsilon}\bigg|_{\epsilon=\epsilon_F}. \quad (2)$$

The Nernst effect is sensitive to the details of the scattering processes in which the conduction electrons are involved. For example, for a simple solid with a single conduction band, $\tan\theta_H$ is proportional to τ, $\tan\theta_H = eB\tau/m^* = \mu_H B$, where B denotes the applied magnetic field, m^* the effective mass, and μ_H (defined as R_H/ρ) the Hall mobility of the charge carriers [21,25]. The majority of Nernst investigations in the literature have focused on semiconductors (showing a relatively large response) in order to figure out the dominating scattering mechanism for the charge carriers [25]. Except for the cuprates [24], the Nernst effect has been generally believed to be negligibly small in metallic systems [23].

According to Eqs. (1) and (2), the Nernst coefficient ν would naturally be linked to the first term in Eq. (1), as long as $\tan\theta_H$ is related to τ. In a Kondo system, it is known that the skew scattering derived anomalous Hall coefficient $R_H = \xi\rho_{\mathrm{mag}}\tilde{\chi}$ [26,27], with ξ being a parameter determined by the phase shift of the Kondo scattering, ρ_{mag} the magnetic contribution to resistivity, and $\tilde{\chi}$ the reduced susceptibility. Within the single-impurity Anderson model, $\tilde{\chi}$ is identical to the renormalized f-electron DOS N_f [26] and $N_f \propto 1/\tau$, i.e., the conduction-electron scattering rate [9]. It is, therefore, expected that $\tan\theta_H \propto 1/\tau$ in a Kondo system, assuming ρ_{mag} dominates the measured ρ. Combining Eqs. (1) and (2) and adopting the different relationships between $\tan\theta_H$ and τ as mentioned above, one obtains

$$S = \pm\frac{\nu}{\mu_H} + \left(-\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \ln N}{\partial \epsilon}\bigg|_{\epsilon=\epsilon_F}\right), \qquad (3)$$

with the sign + and − before ν/μ_H corresponding to $\tan\theta_H \propto \tau$ (simple one-band metal) and $\tan\theta_H \propto 1/\tau$ (Kondo system), respectively. Note that, in deriving Eq. (3), the detailed function relating $\tan\theta_H$ and τ is not needed. Clearly, by measuring both $\nu(T)$ and $\mu_H(T)$, Eq. (3) enables one to separate the "scattering" contribution from the "band" contribution to the TEP [cf. Eq. (1)]. Labeling the latter term S_{DOS} yields

$$\nu = \pm(S - S_{\mathrm{DOS}})\tan\theta_H/B, \qquad (4)$$

which explicitly explains the so-called Sondheimer cancellation in the Nernst effect of simple metals [23,24]: the band term S_{DOS}, which is usually dominating the TEP, is cancelled out in a Nernst measurement. Consequently, in simple metals, ν is almost negligible.

We now turn to the electrical transport properties of $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$, in comparison to the ones of the nonmagnetic reference system $LaCu_2Si_2$. For the two Ce-based systems, the Hall coefficient $R_H(T)$ [Fig. 2(a)] is positive and increases with decreasing temperature, approaching a shallow maximum at $T < 5$ K. By contrast, $R_H(T)$ of $LaCu_2Si_2$ shows a small positive, nearly constant value over the whole temperature range, indicating a dominating hole pocket in the valence band. The Hall mobility $\mu_H(T)$ for both $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$ [Fig. 2(b)] increases monotonically upon decreasing temperature. Following the discussion by Coleman *et al.* [26] as well as Fert and Levy [27], this indicates a monotonic increase of N_f (or $1/\tau$) and is characteristic of the local Kondo scattering.

As displayed in Fig. 1(b), $-\nu/\mu_H$ vs T (crosses) shows reasonable agreement, concerning both magnitude and sign, with the measured $S(T)$ over the whole temperature range for both $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$, an observation which is in line with $\tan\theta_H \propto 1/\tau$ expected for a Kondo system. Here, solely the scattering term in Eq. (3), $-\nu(T)/\mu_H(T)$, is apt to describe reasonably well the measured TEP in a wide range of temperature, i.e., from room temperature to $T < T_{\mathrm{coh}}$. As already mentioned, T_{coh} is taken from the position of the low-T peak in $\rho(T)$ (inset of Fig. 2) which overlaps with a positive maximum in $\nu(T)$ [Fig. 1(a)] and a negative one in $S(T)$ [Fig. 1(b)]. The absence of a significant contribution of S_{DOS} to the TEP highlights the importance of the local Kondo scattering process over a large temperature range above 2 K in $CeCu_2Si_2$ and $Ce_{0.8}La_{0.2}Cu_2Si_2$. This parallels the observation by Aliev *et al.* [28] that the Hall effect of $CeCu_2Si_2$ is strongly enhanced upon cooling from far above to well below T_{coh}, which they ascribed to the formation of the local Kondo resonance. This conclusion is strongly supported by Coleman *et al.* [26], who calculated the anomalous Hall effect in a Kondo-lattice system in terms of the on-site skew scattering within the framework of a local Fermi-liquid model.

The preceding analysis does not apply to $LaCu_2Si_2$: as seen in the inset of Fig. 1(b), for this compound, $\pm\nu(T)/\mu_H(T)$ cannot explain the measured $S(T)$. Assuming that the dominating electron scattering in $LaCu_2Si_2$ is due to acoustic phonons, with the scattering exponent $r = -(1/2)$, one can calculate the expected $\nu(T)$ for this material, following Eq. (2) [23]. Indeed, this calculation [dashed line, inset of Fig. 1(a)] yields a good agreement with the measured $\nu(T)$ above 30 K. It is consistent with the carrier mobility $\mu_H(T)$ [Fig. 2(b)], which exhibits a T^{-1} dependence above 50 K, characteristic of conduction-electron scattering in a metal being dominated by acoustic phonons. Therefore, assuming the validity of Mott's formula, the band term S_{DOS} [i.e., the second term in Eq. (3)] has to contribute significantly to the TEP of $LaCu_2Si_2$, presumably along with a phonon-drag term which partly adds to the broad $S(T)$ hump at $T \approx 50$ K.

Successfully mapping the measured $S(T)$ by $\nu(T)$ [divided by $\mu_H(T)$] in the two Ce-based systems over a wide T range reveals that both quantities can be described solely by a single relaxation time $\tau(\epsilon)$ of the conduction electrons. Apparently, this is based on the fact that the anomalous Hall effect in a Kondo lattice is due to the formation of the Kondo resonance, which guarantees that the Hall angle can be described by N_f ($\propto 1/\tau$), as

discussed by Coleman *et al.* [26], down to well below T_{coh}. The latter case is totally different from that of, e.g., a magnetic semiconductor, where a huge Nernst coefficient has also been observed [29]. In such a system, due to the absence of the Kondo effect, the skew-scattering-derived anomalous Hall angle $\tan\theta_H$ and the relaxation time τ of the conduction electrons are independent of each other. Therefore, unlike the case of the Kondo lattice, no connection can be built between $S(T)$ and $\nu(T)$. Recent measurements of $\nu(T)$, $S(T)$, $R_H(T)$, and $\rho(T)$ over an extended temperature range were reported by Matusiak *et al.* for the HF metal Ce_2PdIn_8 [30]. There, the enhanced Hall coefficient $R_H(T)$, which is largely different from the typical behavior expected for Kondo scattering, is ascribed to antiferromagnetic spin fluctuations causing a highly anisotropic scattering time [30]. For Ce_2PdIn_8, the Nernst coefficient has a similar temperature profile as $R_H(T)$, strikingly different from the canonical behavior of $CeCu_2Si_2$ discussed above.

To substantiate our conclusion that the Nernst effect is evidencing the single-ion Kondo effect, we refer to Zlatić *et al.* [10], who calculated the thermoelectric response of a periodic Anderson model by considering the on-site Kondo scattering. This way, they showed that the "band" term, $(\partial/\partial\epsilon)(\ln N_c)|_{\epsilon=\epsilon_F}$, due to the renormalized conduction-band DOS, N_c, is negligibly small compared to the scattering term. The latter is related to the renormalized f-electron DOS, N_f, by

$$\frac{\partial}{\partial\epsilon}[\ln\tau(\epsilon)]|_{\epsilon=\epsilon_F} \simeq \pm\frac{2LN_f(\epsilon_F)Z_f^{-1}}{n_c}, \tag{5}$$

where L denotes the degeneracy of the f-electron spin state, Z_f the renormalization factor, N_f the renormalized f-electron DOS, and n_c the number of conduction electrons per site. Equation (5) explicitly links the asymmetric Kondo scattering of conduction electrons to the renormalized f spectral weight, and microscopically explains that, while the TEP of the Kondo lattice is determined by the scattering approach in a wide range of temperature, it is adequately described by the huge quasiparticle DOS at even lower temperatures, $T \ll T_{\mathrm{coh}}$. In addition, the "energy resolution" of the Nernst measurement, determined by the applied temperature difference δT during the measurement, has also to be considered. Because we applied a $\delta T < 0.02T$ (for example, $\delta T < 0.2$ K at $T = 10$ K) in our measurements, δT is much smaller than the Kondo temperature ($T_K \approx 20$ K for $CeCu_2Si_2$), which determines the energy width of the Kondo resonance. This makes it possible for the Nernst effect to resolve the asymmetry of the Kondo scattering.

In conclusion, we showed that the highly unusual, enhanced TEP [13–15] as well as the Nernst coefficient of the Kondo-lattice system $CeCu_2Si_2$ and its 20 at.% La-doped variant can be well explained by the highly asymmetric Kondo scattering process of conduction electrons, without resorting to the electronic structure of the heavy quasiparticles. This strongly indicates that the single-ion Kondo physics applies well to the Kondo-lattice system $CeCu_2Si_2$ down to $T < T_{\mathrm{coh}}$. A similar conclusion was recently drawn for $CeNi_2Ge_2$ from calorimetric results on the quasibinary alloys $(Ce_{1-x}La_x)Ni_2Ge_2$ [19]. Our results indicate the Nernst effect to be a promising probe for directly detecting asymmetric scattering processes in correlated-electron systems and, therefore, to play an important role in exploring unconventional thermoelectric materials for potential applications.

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