Resistivity and Thermopower of Heavy-Fermion Systems

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The characteristic temperature dependence of the electronic transport properties of heavy-fermion systems can be reproduced within the self-consistent second-order U-perturbation treatment of the periodic Anderson model. This approach, within which explicit calculations are possible in the limit of a large spatial dimension d for correlated lattice electrons, properly fulfills the Luttinger sum rules. Here we present the first explicit calculations of transport quantities and show that an excellent reproduction of the typical experimental results is possible by simply using the $d=\infty$ result for $d=3$.

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Heavy-fermion (HF) systems (e.g., $CeAl_3$, $CeCu_2Si_2$, $CeCu_6$, UBe_{13}) and also some of the less "heavy," more intermediate valent (IV) systems (e.g., $CePd_3$, $CeIn_3$) usually have a small residual resistance at $T=0$ K, a rapid increase of the resistance for low temperatures $T \ll T_K$, a maximum (of the magnitude of 50–100 $\mu\Omega$ cm) at some low temperature $T_{\max}$, which is of the same order of magnitude as the low (Kondo) temperature scale T_K characteristic of the HF system, and a resistivity (logarithmically) decreasing with increasing temperature for higher temperatures $T > T_{\max}$ [1–3]. The negative temperature coefficient (NTC) of the resistivity in the high-temperature regime is usually interpreted as a manifestation of the Kondo effect in a concentrated system [1,2]. Thus a crossover from a high-temperature regime, in which the conduction electrons are incoherently (impuritylike) scattered from the (periodic) arrangement of rare-earth or actinide ions, to a coherent low-temperature state, in which heavy quasiparticles with infinite lifetime for $T=0$ are formed, is commonly assumed to be most characteristic of HF systems and is most clearly reflected in the resistivity behavior described above.

But there exist a number of metals containing rare-earth or actinide ions with an unstable f shell, which also exhibit a rapid increase of the resistance for low temperatures, but then the resistivity curve only flattens and no NTC is obtained [3]. This different behavior is observed in most Yb-based systems (e.g., $YbAl_3$, YbCuAl, $YbCu_2Si_2$), but also in some of the Ce-based systems (e.g., $CeRh_2$, $CeSn_3$) and in UPt_3 [1,2]. Therefore, the resistivity maximum and the NTC are not necessarily the features which are most characteristic of HF materials, but may be only one of two possibilities for the temperature dependence of the resistance.

The thermoelectric power $Q(T)$ (or the Seebeck coefficient) of HF and IV systems also shows characteristic anomalies when compared to the thermopower of usual metals: $Q(T)$ is absolutely very large, of the order of 50–100 μV/K (compared to 1–5 μV/K usually), and it always has an extremum at some low temperature $T_{\max}$, which is roughly of the magnitude T_K and thus of the temperature of the resistivity maximum (if there is any). But the giant magnitude and the extremum in $Q(T)$ usually exists also for IV or HF systems which do not have the resistivity maximum (e.g., in $YBCu_2Si_2$ [4] and $YbAl_3$ [5]). In some (but not all) HF systems (e.g., $CeAl_3$ [6]) the thermopower changes sign and exhibits a further extremum for very low temperatures $T \ll T_{\max}$.

In this Letter we show that all mentioned aspects concerning the temperature dependence of the electronic transport properties of HF and IV systems can be explained within one single transport theory. We show that the qualitative tendency and also the absolute magnitude of resistance and thermopower can be understood within the framework of the periodic Anderson model (PAM). No additional scattering mechanism (phonons, etc.) is necessary to account for the observed behavior. The correlation between the f electrons, and thus the electron-electron interaction, is responsible for the temperature dependence of the transport quantities both in the low- and in the high-temperature regime.

We study the basic version of the PAM, which reads

$$H=\sum_{\mathbf{k}\sigma}\epsilon(\mathbf{k})c^{\dagger}_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}+\sum_{\mathbf{R}\sigma}[E_f f^{\dagger}_{\mathbf{R}\sigma}f_{\mathbf{R}\sigma}+\tfrac{1}{2}Uf^{\dagger}_{\mathbf{R}\sigma}f_{\mathbf{R}\sigma}f^{\dagger}_{\mathbf{R}-\sigma}f_{\mathbf{R}-\sigma}+V(c^{\dagger}_{\mathbf{R}\alpha}f_{\mathbf{R}\sigma}+f^{\dagger}_{\mathbf{R}\sigma}c_{\mathbf{R}\sigma})]\,. \quad (1)$$

Thus no realistic, but only a twofold, (spin) degeneracy is taken into account for the f and the band-electron systems, which are coupled by the hybridization V. To calculate the dynamical conductivity we start from the Kubo formula

$$\sigma_{xx}(\omega)=\frac{1}{\Omega}\frac{1}{\omega}\chi''_{j_xj_x}(\omega+i0)\,, \quad (2)$$

where $\Omega=Na^d$ is the volume of the d-dimensional system (N the number of lattice cells, a the lattice constant), ω is the frequency of the electrical ac field, and

$$\chi_{j_xj_x}(z)=-i\int_0^{\infty}e^{izt}\langle[j_x(t),j_x(0)]\rangle dt \quad (3)$$

denotes the current-current response function. The current operator x component which is consistent with the PAM (for constant, not $\mathbf{k}$-dependent, hybridization) is given by [7]

$$j_x=\frac{e}{\hbar}\sum_{\mathbf{k}\sigma}\frac{\partial\epsilon(\mathbf{k})}{\partial k_x}c^{\dagger}_{k\sigma}c_{\mathbf{k}\sigma}\,. \quad (4)$$

Inserting (4) into (3) and (2) we are left with the calculation of two-particle Green functions of the type $\langle\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}, c^{\dagger}_{\mathbf{k}'\sigma'} c_{\mathbf{k}'\sigma'}\rangle\rangle_z$.

Of course we need a suitable approximation to explicitly calculate these two-particle Green functions. But, as has been emphasized in particular by Martin [8], it is important for a proper transport theory of the PAM that the approximation used fulfills the Luttinger theorem and the related sum rules [9]. According to the Luttinger theorem the self-energy imaginary part vanishes at the Fermi energy μ according to $\mathrm{Im}\Sigma(E+i0)\sim(E-u)^2+(\pi T)^2$. Then for $T=0$, one has (heavy) quasiparticles with an infinite lifetime at the Fermi surface, and this property must be fulfilled within an approximation to account for a proper reproduction of the observed coherent ground state. But away from the Fermi surface there should be a finite lifetime even at $T=0$, and at the Fermi energy $\mathrm{Im}\Sigma(E+i0)$ should become finite with increasing temperature. It is not trivial that an approximation really reproduces these properties. Many of the successful theories of the PAM [10–13] map the PAM for $U\rightarrow\infty$ onto an effective one-particle two-band model, i.e., a correlation-free problem with renormalized parameters. These theories can properly account for the coherent ground state and the heavy effective mass of the quasiparticles, but so far they have been able to account for finite lifetime effects only in the very low temperature ($\sim T^2$) regime [10,11] and are not yet able to describe the crossover from the coherent zero-temperature ground state to an incoherent high-temperature state. Other transport theories for the PAM [7,14,15] may violate the Luttinger theorem; thus their applicability to the (coherent) low-temperature regime is questionable. Therefore, in a recent transport treatment two different approximations were used [16], namely, the slave-boson mean-field theory for the low-temperature regime, but an effective single-impurity model for high temperatures. Thus a transport theory for the PAM that is valid for low and high temperatures and describes the crossover between these two regimes does not yet exist in spite of attempts for more than ten years.

The simplest approximation, which automatically fulfills the Luttinger theorem and the resulting sum rules, is provided by the self-consistent second-order perturbation theory (SOPT) in terms of the Coulomb correlation U. Though the SOPT should be valid only in the weak-coupling regime of small U and U is considered to be relatively large for HF systems (of the order of 5–10 eV), meaningful results, in particular large enhancements of the effective mass, can be obtained for the PAM, as has been emphasized by Yamada and Yosida [17]. But for technical reasons explicit computations within the SOPT, in particular studies of the temperature dependence of actual physical quantities like the resistivity, were not possible until recently. Such SOPT calculations become possible in the limit of large dimensions, $d\rightarrow\infty$ [18], as introduced by Metzner and Vollhardt [19]. Following [19] we study a simple-cubic tight-binding dispersion

$$\epsilon(\mathbf{k})=2t\sum_{l=1}^{d}\cos(k_l a) \tag{5}$$

in the limits $d\rightarrow\infty$, $t\rightarrow 0$, keeping $dt^2=\text{const}$. Only with this scaling do lattice models for correlated electron systems remain nontrivial and keep all their essential properties, while actual computations are greatly simplified. We have shown, in particular, that for $d=\infty$ the SOPT of the PAM reproduces quasiparticles with infinite lifetime and a heavy effective mass for low temperatures, a characteristic low-temperature scale, and the crossover from a coherent ground state to an incoherent-scattering high-temperature state [18], and we have demonstrated that the limit $d=\infty$ is able to approximately describe three-dimensional systems [20], probably because the local approximation of a site-diagonal, $\mathbf{k}$-independent self-energy, which is correct for infinite d (not only within the SOPT, but in general) [19,21,22], is already fairly good for $d=3$.

Here we present the first application of this $d=\infty$ approach for the PAM to the calculation of the temperature dependence of transport quantities. As the vertex corrections are obtained from the self-energy diagrams by cutting Green-function lines, the vertex corrections must be local when the self-energy is local; but as in the site representation the current operator is a nonlocal operator (connecting nearest-neighbor sites), the current vertex corrections must vanish for $d=\infty$, as has been mentioned already by Khurana [23]. Therefore one can expect that neglecting the vertex corrections to the conductivity should be a good approximation for three-dimensional systems. Then we obtain in the limit $\omega\rightarrow 0$ for the static conductivity

$$\sigma_{xx}(\omega=0)=\frac{e^2a^{2-d}}{2\pi\hbar}t^2\int_{-\infty}^{\infty}dE\left(-\frac{df}{dE}\right)L(E)\,, \tag{6}$$

with

$$L(E)=\frac{2}{N}\sum_{\mathbf{R}\mathbf{R}'\sigma}\mathrm{Im}G^{c}_{\mathbf{R}\mathbf{R}'\sigma}(E+i0)\mathrm{Im}G^{c}_{\mathbf{R}'\mathbf{R}\sigma}(E+i0)\,, \tag{7}$$

$$G^{c}_{\mathbf{R}\mathbf{R}'\sigma}(z)=\frac{1}{N}\sum_{\mathbf{k}}\frac{e^{i\mathbf{k}\cdot(\mathbf{R}-\mathbf{R}')}}{z-V^2/[z-E_f-\Sigma(z)]-\epsilon(\mathbf{k})} \tag{8}$$

the band electron Green function, $\Sigma(z)$ the f-electron self-energy, and $f(E)$ the Fermi energy. The thermopower Q can also be calculated from the function $L(E)$ according to

$$Q=\frac{\int dE(-df/dE)(E-\mu)L(E)}{eT\int dE(-df/dE)L(E)}\,. \tag{9}$$

These equations are still exact for large d (within the linear-response theory), provided that one inserts the exact f-electron self-energy. $\Sigma(z)$ is the quantity one cannot determine exactly for the PAM even for $d=\infty$, and

we have calculated it within the SOPT [18]. Measuring energies (and temperatures) in units of the effective width of the unperturbed conduction band, which is centered around zero energy, i.e., setting $2t^2d=1$, we have calculated the temperature dependence of the resistivity $\rho(T)$ for the parameters $E_f=-0.5$, $U=1$, $V=0.4$, and different choices for the total number of electrons per lattice site $n_{\rm tot}$. The results are shown in Fig. 1 for $n_{\rm tot}=0.2$, 0.4, 0.6, and 0.8 (corresponding to less than half filling, as the maximum number of electrons per site is 4). Obviously, for $n_{\rm tot}=0.8$ (and also for larger values $0.8\leq n_{\rm tot}<2$) we obtain the typical behavior with a small residual resistance, a rapid increase $\sim T^2$ for very low T, an almost linear T dependence for $T<T_K$, a maximum near $T_{\rm max}=0.1$ ($\approx T_K$ for this parameter choice), and a NTC for higher temperatures. For $n_{\rm tot}=0.6$, however, we obtain the other possible characteristic behavior, namely, a rapid increase of $\rho(T)$ for low T but then a flattening, and no NTC. Further decreasing $n_{\rm tot}$ gradually leads to a resistivity behavior characteristic of normal metals. The (T-dependent) chemical potential μ has been determined self-consistently for a given $n_{\rm tot}$. For $n_{\rm tot}=0.2$, μ is far below the effective f-level position $\tilde{E}_f$, i.e., we are in the almost empty f-level regime qualitatively describing La or (because of particle-hole symmetry) Lu compounds. Increasing $n_{\rm tot}$ moves μ nearer to $\tilde{E}_f$ and increases the valence. For $n_{\rm tot}=0.6$ we are in a weakly IV regime and for $0.8\leq n_{\rm tot}<2$ we are in the strongly IV and real Kondo lattice regime, in which case we obtain the characteristic resistivity curve with a maximum and a NTC. The appearance or nonappearance of a NTC in the resistivity $\rho(T)$ curve seems to be correlated with whether the Fermi energy falls into the lower, ascending branch of the effective f-electron density-of-states peak around $\tilde{E}_f$ (being the case for $n_{\rm tot}\leq 0.6$), or whether it lies in the central region of this peak, near the hybridization (quasi) gap [18] (being the case for $0.8\leq n_{\rm tot}$). There is a corresponding correlation with the valence and the effective mass; for otherwise similar parameters (systems), a NTC is more likely to occur in the system with the larger valence or a larger effective mass $m_{\rm eff}$. This is in qualitative agreement with experimental results, for instance the very recent transport measurements for $YbInCu_4$ and $YbAgCu_4$ [24]; in the IV system $YbInCu_4$, no NTC is observed in the $\rho(T)$ curve, whereas in the isostructural $YbAgCu_4$ (with a larger valence and an about 3 times larger $m_{\rm eff}$) a NTC occurs. But the absolute value of $m_{\rm eff}$, which in our model is determined by the parameters V and U, is not essential for the high-temperature resistivity slope. Therefore, no NTC may occur in a HF system like UPt_3 with a relatively large $m_{\rm eff}$, and an IV system like $CePd_3$ with a much smaller $m_{\rm eff}$ may exhibit the NTC in the $\rho(T)$ curve. It is interesting that we obtain not only the characteristic behavior of very low temperatures but also that of intermediate temperatures (roughly corresponding to room temperature) within the PAM alone, i.e., without additional scattering mechanisms. According to Eq. (6) the static conductivity is calculated in units e^2a^{2-d}/hd, where h is Planck's constant. Therefore, in a strict limit $d\rightarrow\infty$ the conductivity would vanish because of the factor $1/d$ (resulting from the factor t^2 from the two current operators). But, of course, we are interested in the nonvanishing transport quantities in leading order in $1/d$. Setting simply $d=3$ and inserting a lattice constant of several Å, our resistivity unit is of the magnitude of a few mΩ cm, i.e., the maximum of the resistivity (~ 0.04) corresponds to 100 $\mu\Omega$ cm, which is just the magnitude which is typical for metallic HF and IV systems.

Corresponding results for the temperature dependence of the thermopower are shown in Fig. 2. We observe the extremum in $Q(T)$ approximately at the same temperature $T_{\rm max}$, where the resistivity has its maximum, and this extremum in $Q(T)$ is also present when $\rho(T)$ does not have the maximum and the NTC, in complete agreement with the experimental findings. We obtain a change of the sign of $Q(T)$ for relatively high temperatures, and for low temperatures we do not always find a change of the

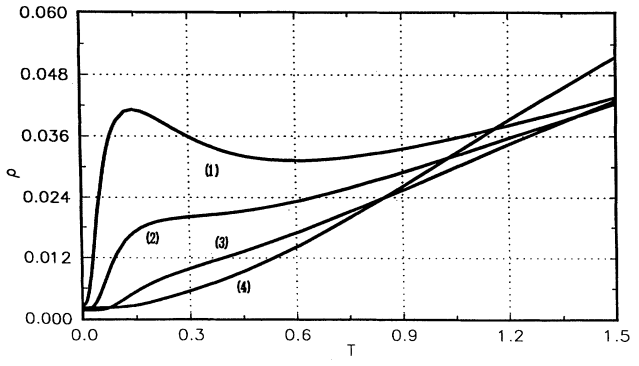

FIG. 1. Temperature dependence of the resistivity obtained for the PAM within the SOPT for $d=\infty$ and the parameters $E_f=-0.5$, $V=0.4$, $U=1$. (1) $n_{\rm tot}=0.8$, (2) $n_{\rm tot}=0.6$, (3) $n_{\rm tot}=0.4$, (4) $n_{\rm tot}=0.2$.

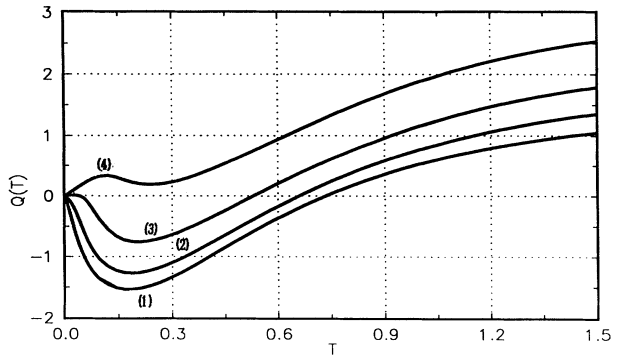

FIG. 2. Temperature dependence of the thermopower $Q(T)$; parameters as in Fig. 1.

sign, but only for certain parameters [cf. curve (3)]. Obviously our treatment can qualitatively explain and reproduce all the main and most characteristic features concerning the temperature dependence and the absolute magnitude of the resistivity and the thermopower of IV and HF systems.

Our conclusions are the following: (1) For nearly the whole temperature regime, i.e., for low and intermediate T, the transport quantities of HF and IV systems can be understood within a treatment of the PAM, which properly reproduces the Luttinger theorem, i.e., Fermi liquid behavior for very low T, but a finite lifetime for intermediate and higher T. (2) Both observed resistivity behaviors, with or without a NTC, can be obtained within the same treatment by changing slightly only one parameter. (3) The relevant scattering mechanism responsible for the T dependence of $\rho(T)$ and $Q(T)$ is the scattering of the conduction electrons from the correlated f-electron system, i.e., the scattering from local spin fluctuations, which are implicitly contained within the correlated f-electron system. (4) Vertex corrections are not important for a qualitative reproduction of the typical electronic transport behavior. (5) The large d limit for correlated lattice electron systems can be used to calculate measurable physical quantities like resistivity, and it can describe three-dimensional systems.

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