

Impurity resistivity in systems with a fluctuating valence

E. P. Fetisov¹⁾ and D. I. Khomskii

P. N. Lebedev Physics Institute, Academy of Sciences of the USSR

(Submitted 6 February 1986)

Pis'ma Zh. Eksp. Teor. Fiz. **43**, No. 7, 334–336 (10 April 1986)

The temperature dependence of the impurity resistivity in compounds with an intermediate valence and with heavy fermions is examined. At low temperatures the resistivity falls off as $\rho_{\text{imp}}(T) \simeq \rho_0 - aT^2$, but at high temperatures the resistivity of the system with impurities in an incoherent regime may fall below the resistivity of the pure system.

In standard metallic systems the resistivity $\rho(T)$ usually obeys Mathiessen's rule: It is represented as a sum of the temperature-dependent resistivity of the pure metal $\rho_p(T)$ and the temperature-independent resistivity of the impurity component ρ_{imp} . There are many deviations from this rule, but they are usually small, roughly on the order of several percent.

In systems with valence fluctuations (compounds with an intermediate valence and with heavy fermions), the situation is quite different. In the first place, the residual resistivity in them is usually quite large, much larger than the scattering by an impurity corresponding to the geometric cross section. Of less importance is the fact that ρ_{imp} depends strongly on the temperature, with variations on the order of unity.¹

This behavior has recently been demonstrated particularly clearly by Lawrence *et al.*² In measuring the resistivity of CePd₃ with an La impurity they found that at low temperatures the impurity resistivity is described by the relation $\rho_{\text{imp}} = \rho_0[1 - (T/T_f')^2]$, where $T_f' = 31$ K is the scale temperature of the order of magnitude of the degeneracy temperature of the heavy f component. It is important that

essentially the same scale value, $T_f = 45$ K, determines the resistivity of pure CePd₃, $\rho_p = \rho_M (T/T_f)^2$, where ρ_M is the Mott scale resistivity on the order of the maximum metallic resistivity (in these systems, it is typically $\rho_M \sim 100\text{--}300 \mu\text{m}\cdot\Omega\cdot\text{cm}$). Similar systematic features in the behavior of the resistivity of pure and doped systems are seen in many other substances of this class, including compounds with heavy fermions such as CeAl₃ and CeCu₆ (Refs. 1 and 3).

To account for the observed behavior of $\rho_{\text{imp}}(T)$, Lawrence *et al.*² used the Kondo lattice model and assumed that at low temperatures, when the system is in a coherent regime (the scattering by various Kondo centers is brought into phase), the absence of a spin in any lattice site (upon substitution of La for Ce, for example) is equivalent to the appearance of an additional Kondo scattering (the “Kondo-hole” model). It has so far not been determined, however, to what extent these hypotheses are legitimate. Apart from the resistivity, no other characteristics (magnetic susceptibility, specific heat) show any evidence of additional “Kondo” behavior; furthermore, a Kondo hole of this sort would, as noted by Lawrence *et al.*,² probably exhibit a ferromagnetic exchange interaction with the conduction electrons.

In the present letter we offer a simple explanation of the observable temperature dependence of the resistivity in systems with valence fluctuations, which can easily be demonstrated phenomenologically. We can show that the low-temperature thermodynamic properties of these systems are well described in a two-component model, with masses^{4,5} $m_d \sim m_0$ and $m_f \gg m_0$. In this physical model, which is very similar to the model for transition metals, the resistivity is due primarily to the scattering of light d electrons into the heavy f band. This mechanism was frequently used to explain the resistivity of transition metals and of the alloys based on these metals⁶ (e.g., Al₅ compounds⁷). The resistivity of a “pure” system is $\rho_p = AT^2$, where $A \sim T_f^{-2}$. In the scattering by impurities the conductivity is described by the standard expression

$$\sigma = \frac{n_d e^2}{m_d} \int \left(-\frac{\partial f}{\partial \epsilon} \right) \tau(\epsilon) d\epsilon, \quad \tau^{-1}(\epsilon) = \frac{2\pi}{\hbar} c |M|^2 N_f(\epsilon), \quad (1)$$

where c is the impurity concentration, $N_f(\epsilon)$ is the state density in the f band, $f(\epsilon)$ is the Fermi function, and $|M|^2$ is the square of the matrix element of the f - d scattering, which is averaged over the angles. For $N_f(\epsilon)$ the expression is usually written in the form

$$N_f(\epsilon) = \frac{2}{\pi} \frac{\Gamma}{(\epsilon - \epsilon_f)^2 + \Gamma^2} \quad (2)$$

(the “resonant-level” model); here ϵ_f is the position of the f level relative to the Fermi level, and Γ is the width of the f band. The scale temperature is $T_f \simeq (\Gamma^2 + \epsilon_f^2)^{1/2}$. In the cases of interest to use we have $\epsilon_f \lesssim \Gamma$ and $T_f \simeq \Gamma$.

From (1) and (2) we easily find that at low temperatures

$$\rho_{\text{imp}}(T) = \rho_0 \left[1 - \frac{\pi^2}{3} \frac{T^2(\Gamma^2 + 3\epsilon_f^2)}{(\Gamma^2 + \epsilon_f^2)^2} \right] \simeq \rho_0 \left[1 - \frac{\pi^2}{3} \left(\frac{T}{T_f} \right)^2 \right], \quad \rho_0 = \frac{4m_d c |M|^2 \Gamma}{n_d e^2 \hbar (\Gamma^2 + \epsilon_f^2)}$$

if the change in the chemical potential with the temperature is taken into account. First, we see that because of the large state density in the f band, the residual resistivity is very large, $\rho_0 \sim T_f^{-1}$ (these equations can be used only at $\rho_{\text{imp}} < \rho_M$). We also see that the impurity resistivity falls off quadratically with the temperature, with the same scale value of T_f which determines the resistivity of the pure system. A decrease in ρ_{imp} with increasing T is quite natural in the model: At $T > T_f$ the electrons with energies of $\sim T$, which are not at resonance, contribute to the conductivity, and the efficiency of the impurity d - f scattering decreases.

Our conclusions are in complete agreement with the experimental results of Ref. 2 [even the quantitative agreement is reasonably good, although it should not be overestimated: $T_f = 45$ K for CePd₃, found from the $\rho_p(T)$ curve, would give $T'_f = 26$ K, according to (3), while the experiment gives² $T'_f = 31$ K]. The result in (3) also accounts for the correlation found in CeCu₆ (Ref. 8) between the decrease in ρ_0 and the increase of the coefficient A of the temperature-dependent part of the resistivity, $\rho = \rho_0 + AT^2$: the second term in (3) renormalizes A , $A \rightarrow A - (\pi^2/3) (\rho_0/T_f^2)$. A simultaneous pressure-induced decrease of ρ_0 and A with increasing T_f in Ce_{0.9-x}La_xTh_{0.1} can also be explained qualitatively. Here A decreases faster than ρ_0 (Ref. 9): in our case $A \sim T_f^{-2}$ and $\rho_0 \sim T_f^{-1}$.

A decrease in ρ_{imp} with T at high temperature, $T \gtrsim T_f$, at which an independent incoherent scattering by f centers occurs, may cause the resistivity of the system with impurities to drop below that of the pure system. In this regime, the resonance scattering by f ions with a fluctuating valence has a large cross section (on the order of the unitary limit) and the replacement of such ions by impurity ions in the absence of valence fluctuations leads to a reduction of the total resistivity. Such a behavior has actually been observed in many systems such as CeCu₆:La (Ref. 3) or CeBe¹³:La (Ref. 10).

In summary, the simple arguments developed above have enabled us to explain the basic effect of impurities on the resistivity of the systems with a fluctuating valence. In the approach considered by us, other kinetic characteristics such as thermal conductivity, thermal emf, and other characteristics, in which a specific temperature dependence arises, can also be easily studied.

¹Engineering-Physics Institute, Moscow.

²A similar conclusion that the impurity resistivity is large in systems with heavy fermions was reached by B. Z. Spivak in the model of a single-component heavy Fermi liquid.

¹D. Wohlleben and B. Wittershagen, Adv. Phys. **34**, 403 (1985).

²J. M. Lawrence, J. D. Thompson, and Y. Y. Chen, Phys. Rev. Lett. **54**, 2537 (1985); **55**, 1702 (1985).

³Y. Onuki, Y. Shimizu, M. Nishihara, Y. Machii, and T. Komatsubara, J. Phys. Soc. Jpn. **54**, 1964 (1985).

⁴D. Jaccard, J. Flouquet, and J. Sierro, J. Appl. Phys. **57**(1), 3084 (1985).

⁵E. P. Fetisov and D. I. Khomskii, Solid State Comm. **56**, 403 (1985).

⁶N. F. Mott, Adv. Phys. **13**, 325 (1964).

⁷A. S. Aleksandrov and V. F. Elesin, Fiz. Tverd. Tela **22**, 1151 (1980) [Sov. Phys. Solid State **22**, 669 (1980)].

⁸A. Amato, D. Jaccard, E. Walker, and J. Flouquet, Solid State Comm. **55**, 1131 (1985).

⁹J. M. Lawrence, J. D. Thompson, Z. Fisk, and B. Ballogg, J. Appl. Phys. **57**(1), 3131 (1985).

¹⁰J. P. Kappler, G. Krill, M. F. Ravet, M. J. Besnus, and A. Meyer, in: Valence Fluctuations in Solids, ed. L. M. Kalikov, W. Hanke, and M. B. Maple, North-Holland, Amsterdam, 1981, p. 271.

Translated by S. J. Amoretti