Competition between Kondo effect and RKKY interactions in heavy-fermion systems: Transport properties

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Following Abrikosov's pseudofermion approach, transport properties such as thermal conductivity, electrical conductivity, and Lorenz ratio of highly correlated systems have been studied by taking into account the effect of impurity-impurity interactions [Ruderman-Kittel-Kasuya-Yosida (RKKY)] in the presence of a crystalline electric field in heavy-fermion systems. It is found that the RKKY contribution becomes significant at low temperatures. At these low temperatures large values of the Lorenz number are obtained which are in close agreement with the experimental results reported for Ce-based heavy-fermion systems. [S0163-1829(97)03930-1]

I. INTRODUCTION

Recently much interest has been focused on Kondo lattices or heavy-fermion systems in f-electron systems of rare-earth and actinide compounds.[1] In general these systems are characterized by unusual low-temperature thermodynamic and transport properties.[2] At low temperatures[3] an additional characteristic feature is the presence of coherence which is found to be responsible for exhibiting a peak in the specific heat[4] $[C(T)/T]$ near or below 1 K in systems such as $CeCu_2Si_2$, Upt_3, URu_2Si_2, and a sharp negative peak in the Hall coefficient around $\simeq 0.2$ K in the case of $CeCu_6$, and superconducting behavior in some of these systems.[5] It is well known that in cerium Kondo compounds in Kondo lattices there is a competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction; the former tends to suppress the magnetic moment with decreasing temperature and the latter, tends to give a magnetic ordering between the different rare-earth atoms. The neutron-scattering experiments reveal that these two competing mechanisms operating in these systems might explain the low-temperature behavior of the heavy-fermion systems (HFS's).

In an earlier paper[6] the authors investigated the problem of resistivity in HFS's in the presence of a crystalline electric field (CEF). It was found that in these systems, by considering the spin-flip scattering of electrons through Kondo- and RKKY-type terms, an extreme in resistivity at low temperatures below 50 K is manifested in the presence of a CEF. The nature of the extreme depends upon whether the RKKY coupling between the impurities is positive (ferromagnetic) or negative (antiferromagnetic). Ruvalds and Sheng[7] have shown that the spin-flip scattering of conduction electrons by a pair of magnetic impurities depends strongly on the temperature and a sharp drop in magnetoresistivity towards low temperatures is attributed to a consecutive spin-flip scattering of an electron of two adjacent impurities as mentioned above. Due to the presence of well-localized f electrons in these systems, the RKKY exchange interaction is expected to cause magnetically ordered states, not only ordinary ferromagnetic and antiferromagnetic states, but also orbital ordered states in $CeBe_6$ often called quadrupole ordered states. Even superconductivity has been argued to be realized by the RKKY exchange interactions[8] in these systems. The study of the competition between normal Fermi liquids and magnetic states or the competition between the Kondo effect and RKKY exchange interactions is required in order to understand a variety of ground states in these f-electron systems.[9,10] Further, it is pointed out that the strong temperature variation in the intermediate temperature range $(1<T<100\,\mathrm{K})$ is due to elastic spin flipping of an electron from the impurity pairs, coupled by RKKY interactions.[7]

Thermal conductivity, electrical conductivity, and Lorenz number are important transport properties of the HFS's and are sensitive to the various physical processes. Their study provides an effective check on the success of a particular model. Previously these properties were considered theoretically by Fulde *et al.*,[11] Bhattacharjee and Coqblin,[12] and others[13] particularly for Ce systems. The reduced Lorenz ratio $L(T)/L_0$ measured for Ce-Kondo compounds shows large values;[12] five times the Sommerfield value L_0 for CeB_6 and $CeCu_6$ at low temperatures while it is reported to lie between $2L_0$ and $4L_0$ for $CeAl_2$ and $CeLa_{1-x}Cu_6$.

In this paper an attempt has been made to study the thermal and electrical conductivities and the Lorenz ratio in these systems in which the Kondo scattering and RKKY coupling have been treated on equal footing in order to learn about their relative strength from a comparison of the results with experimental data. The plan of this paper is as follows. Section II briefly considers the formalism based upon Abrikosov's[14] pseudofermion method and presents the key results for the thermal conductivity, electrical conductivity, and Lorenz ratio. In Sec. III parameters are chosen and calculations are given. Finally, in Sec. IV the results obtained are presented and discussed.

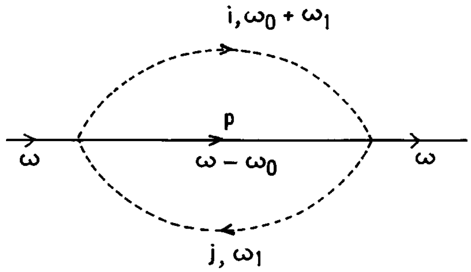

FIG. 1. Second-order self-energy diagram of conduction electrons in the presence of a CEF. The labels i j indicate different CEF split labels. The solid line is the propagating bare electron.

II. FORMALISM

The Hamiltonian of the system[6] in which electron gas interacts with an array of magnetic impurity spins distributed over a lattice may be written as

$$H = H_0 + H_1, \tag{1}$$

where

$$H_0 = \sum_{k,\sigma} E_k C^{\dagger}_{k\sigma} C_{K\sigma} - I \sum_{\substack{i_1, j_1 \\ (i \neq j)}} S^z_{i_1} S^Z_{j_1},$$

$$H_1 = -\frac{J}{N} \sum_{\substack{k_1, k_2, j \\ \sigma_1, \sigma_2 \\ \beta_1, \beta_2}} \langle \sigma_1 \beta_1 | \mathbf{s} \cdot \mathbf{S} | \sigma_2 \beta_2 \rangle e^{i(\mathbf{k}_1 - \mathbf{k}_2) \cdot \mathbf{R}_j}$$

$$\times C^{\dagger}_{K_1 \sigma_2} C_{K_2 \sigma_2} d^{\dagger}_{j\beta_1} d_{j\beta_2} - \frac{I}{2} \sum_{\substack{i_1 j_1 \\ (i_1 \neq j_1)}} (S^+_{j_1} S^-_{i_1} + S^-_{j_1} S^+_{i_1}).$$

H_0 represents the unperturbed part of the Hamiltonian which includes free electron energy $E_k = \hbar k^2/2m$ and the part of the impurity-impurity interactions that conserves the spins. $C^{\dagger}_{k\sigma}(C_{k\sigma})$ here represents the creation (destruction) operator with momentum state k and spin σ and $\mathbf{S}_{j_1}$ represents the impurity spin at the site $\mathbf{R}_{j_1}$. Since the heavy fermions are characterized for localized spin states corresponding to the impurity atoms, a coupling I is expected between impurities which are dominated by the RKKY indirect exchange mechanism via conduction electrons. The main interest is in the process of the spin-flip scattering of conduction electrons via impurities. H_1 is the interaction part of the Hamiltonian, where the first term in it represents the interaction between the magnetic impurity ion and conduction electrons represented by a coupling constant $J. \langle \cdots \rangle$ represents the matrix element of the transition between $\sigma_1 \beta_1$ and $\sigma_2 \beta_2$; $d^{\dagger}_{j\beta}(d_{j\beta})$ are the creation (destruction) operators corresponding to the impurity written in the Abrikosov's pseudofermion frame work.[14] The second term in H_1 represents the RKKY interaction responsible for spin-flip processes. The results obtained would be valid for an arbitrary number of impurity levels, however, we will concentrate on an impurity with the lowest two level structure.

As the first step the lowest (second) order contribution in the self-energy in the presence of a CEF is found[6] (Fig. 1), which after summing over the frequencies and in the lowest

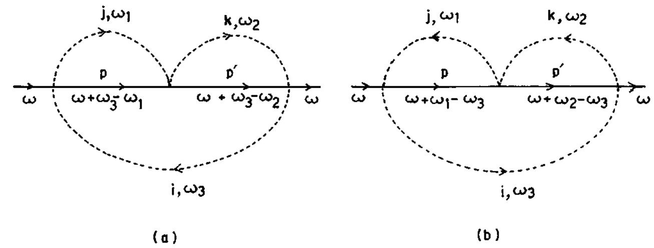

FIG. 2. (a) and (b) The third-order self-energy diagrams for exchange Kondo scattering in the presence of a CEF. Solid and dashed lines correspond to electron and impurity propagators.

two level approximation scheme yields the following inverse relaxation time in second order:

$$\frac{1}{\tau^{(2)}(\omega)} = 2\pi N(0) \left(\frac{J}{N}\right)^2 \left[M_e^2 + M_i^2 \left\{ 1 + \frac{1}{2} \tanh\left(\frac{\delta}{2T}\right) \times [D(\omega,\delta) - D(\omega,-\delta)] \right\} \right]. \tag{2}$$

Here

$$\delta = \delta_{ij} = \delta_i - \delta_j$$

and

$$D(\omega,\delta) = \tanh\left(\frac{\omega - \delta}{2T}\right) - \tanh\left(\frac{\omega}{2T}\right),$$

and M_e and M_i are elastic and inelastic contributions from the matrix element. Furthermore, the functions

$$g(x) = P \int_{-D}^{D} \frac{d\varepsilon}{\varepsilon - x}$$

and

$$h(x) = P \int_{-D}^{D} \frac{\tanh(\varepsilon/2T)}{\varepsilon - x} \, d\varepsilon \tag{3}$$

have been introduced while arriving at the above result for the reciprocal relaxation time given by Eq. (2).

Next, the Kondo contribution arising from the third-order self-energy diagrams, shown in Figs. 2(a) and 2(b), and after summing for the two lowest levels, yields the following expression:

$$\frac{1}{\tau^{(3)}(\omega)} = \pi N(0) \left(\frac{J}{N}\right)^2 \left[2\left(\frac{J}{N}\right) N(0) M_e^3 h(\omega) + \frac{1}{2}\left(\frac{J}{N}\right) \times N(0) M_e M_i^2 (1 + e^{\omega/T}) \operatorname{sech}^2(\delta/2T) \{ h(\omega - \delta) + h(\omega + \delta) \} + \left(\frac{J}{N}\right) N(0) M_e M_i^2 \{ [g(\omega - \delta) - g(\omega + \delta)] \tanh(\delta/2T) + [h(\omega - \delta) + h(\omega + \delta)] + (1 + e^{\omega/T}) \operatorname{sech}^2(\delta/2T) h(\omega) + 2h(\omega) \} \right]. \tag{4}$$

Further, higher order contributions to the self-energy arise from the RKKY interaction in IJ^2 corresponding to the dia-

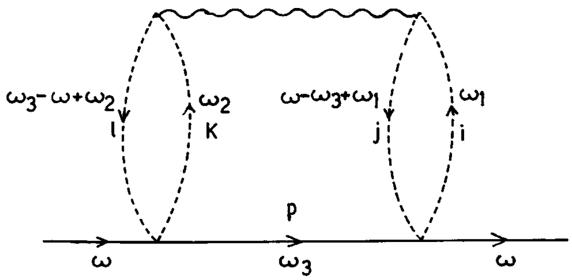

FIG. 3. The lowest order diagram of the self-energy of the conduction electron due to the presence of an impurity-impurity interaction in the presence of a CEF. The wiggly line represents the RKKY interaction.

gram shown in Fig. 3. After performing frequency sums, taking the limit $\lambda\to\infty$, and summing over the lowest two levels, one gets

$$\frac{1}{\tau_{\mathrm{RKKY}}(\omega)}=Il_2\pi N(0)f^t(2K_fR)\left(\frac{J}{N}\right)^2\frac{1}{4T}\left[\operatorname{sech}^2\left(\frac{\omega-\delta}{2T}\right)+\operatorname{sech}^2\left(\frac{\omega+\delta}{2T}\right)\right]\tanh^2\left(\frac{\delta}{2T}\right). \tag{5}$$

The total inverse relaxation time may be written as

$$\tau^{-1}(\omega)=\tau_0^{-1}\{(\tau^{(2)}_{(\omega)})^{-1}+(\tau^{(3)}_{(\omega)})^{-1}+[\tau_{\mathrm{RKKY}}(\omega)]^{-1}\}, \tag{6}$$

$$\tau_0^{-1}=2\pi N(0)\left(\frac{J}{N}\right)^2M_e^2,$$

$$\gamma^2=(M_i/M_e)^2,$$

where $\tau^{(2)}(\omega)$, $\tau^{(2)}(\omega)$, and $\tau_{\mathrm{RKKY}}(\omega)$ are the second-order, third-order, and RKKY contributions to the inverse relexation time.

A. Thermal conductivity

Thermal conductivity K is given by[11]

$$K=\frac{N}{mT}\int_{-\infty}^{+\infty}d\omega\,\omega^2(-\partial f/\partial\omega)\tau(\omega),$$

where N represents the total number of atoms per unit volume and m is the effective mass of electrons. Substituting for $\tau(\omega)$ from Eq. (6) after the approximation ($\omega,T\ll\delta$) in the third-order contribution, one is able to write

$$K=[K^{(2)}+K^{(3)}+K_{\mathrm{RKKY}}]. \tag{7}$$

The second-order contribution in $K^{(2)}$ is

$$K^{(2)}=K_0\left\{1-\frac{\gamma^2(\delta/T)}{\sinh(\delta/T)}\left[1+\left(\frac{\delta}{\pi T}\right)^2\right]\right\}, \tag{8}$$

where

$$K_0=\frac{N\pi^2T}{3m}\,\tau_0.$$

The third-order contribution $K^{(3)}$ corresponding to Kondo exchange scattering in the limit ω, $T\ll\delta$ is found to be

$$K^{(3)}_{(\omega,T\ll\delta)}=K_0\Bigg[\{1+\gamma^2[1+\operatorname{sech}^2(\delta/2T)]\}\alpha_1\ln|T/D| -\alpha_1\gamma^2(\delta/D)\tanh(\delta/2T)+\alpha_1\gamma^2\times\left\{\frac{T}{\delta}\ln|\delta/T|+16\left(1+\frac{\delta}{2\pi T}\operatorname{Im}\psi^{(1)}(i\delta/2\pi T)\right)\times\left(1+\frac{2}{3}(\delta/T)^3\right)+\frac{8}{6}\pi^2\right\}\{1+\operatorname{sech}^2(\delta/2T)\}\Bigg], \tag{9}$$

where $\alpha_1=(J/N)M_eN(0)$ is a constant.

Here $\psi^1(z)$ denotes the trigamma[15,16] function. Next the contribution to the thermal conductivity, in Eq. (7), is from the RKKY interactions which one can obtain using Eq. (6) as

$$K_{\mathrm{RKKY}}=32K_0\frac{I}{T}\frac{\tanh^2(\delta/2T)}{\sinh^3(\delta/4T)}\alpha_2\frac{1}{\pi^2}\times\left[\left\{\frac{3}{4}\left(\frac{\delta}{T}\right)^2+\pi\right\}\sinh(\delta/4T)-\frac{1}{2}\left\{\frac{1}{8}\left(\frac{\delta}{T}\right)^3+\frac{\pi}{2}\left(\frac{\delta}{T}\right)\right\}\cosh(\delta/4T)\right]. \tag{10}$$

Here $\alpha_2=(I_2/M_e^2)f^t(2K_fR)$ is treated as constant.

B. Electrical conductivity

The electrical conductivity σ is given as[11]

$$\sigma=\frac{Ne^2}{2m}\int_{-\infty}^{+\infty}d\omega(-\partial f/\partial\omega)\tau(\omega),$$

$$\sigma=[\sigma^{(2)}+\sigma^{(3)}+\sigma_{\mathrm{RKKY}}]. \tag{11}$$

Here $\sigma^{(2)}$, $\sigma^{(3)}$, and σ_{RKKY} are the second-order, third-order, and RKKY contributions to electrical conductivity in the presence of a CEF. δ is given below. For $\sigma^{(3)}$ we have used an approximation corresponding to the limit $\omega,T\ll\delta$:

$$\sigma^{(2)}=\sigma_0\left[1-\gamma^2\frac{(\delta/T)}{\sinh(\delta/T)}\right], \tag{12}$$

where

$$\sigma_0=\frac{Ne^2}{2m}\,\tau_0,$$

$$\sigma^{(3)}=\sigma_0\Bigg[\left\{1+\left(\operatorname{sech}^2(\delta/2T)+\frac{1}{2}\right)\gamma^2\right\}\alpha_1\ln\left|\frac{D}{T}\right|-\alpha_1\gamma^2\times[1+\operatorname{sech}^2(\delta/2T)]\left\{\frac{T}{4\delta}\ln\left|\left(\frac{D}{T}+\frac{\delta}{T}\right)\Big/\left(\frac{D}{T}-\frac{\delta}{T}\right)\right|+4\left(1+\frac{\delta}{2\pi T}\operatorname{Im}\psi^{(1)}(i\delta/2\pi T)\right)\right\}+\gamma^2\frac{2\delta}{D}\alpha_1\tanh(\delta/2T)\Bigg], \tag{13}$$

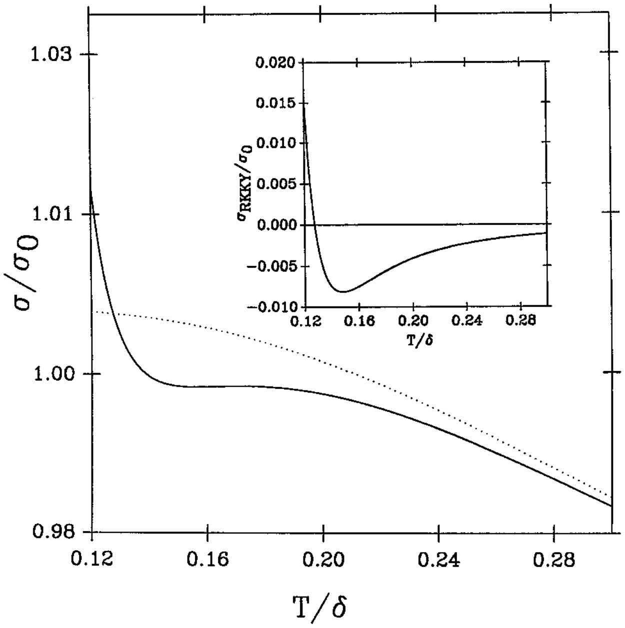

FIG. 4. Electronic conductivity $[\sigma(T)/\sigma_0]$ in the presence of a CEF; $\delta=90$ K and the RKKY interaction parameter $I=-0.01$ eV. The dashed curve shows the contribution from second- and third-order exchange scattering and the solid curve shows the total contribution, i.e., on addition of RKKY interaction and exchange scattering contributions. The inset shows the RKKY contribution to electrical conductivity $(\sigma_{\mathrm{RKKY}}/\sigma_0)$ for the same CEF and I values.

$$\sigma_{\mathrm{RKKY}}=\frac{I}{2T}\,\alpha_2\sigma_0\tanh^2(\delta/2T)\,\frac{1}{\sinh^3}\,\sinh^3(\delta/4T)\times\left[\cosh(\delta/4T)+\sinh(\delta/4T)-\frac{\delta}{4T}\cosh(\delta/4T)\right]. \tag{14}$$

C. Lorenz number

The Lorenz number is given by[12]

$$L=\frac{K}{\sigma T},$$

$$L_0=\frac{\pi^2}{3}\left(\frac{k_B}{e}\right)^2, \tag{15}$$

where e is the electron charge, k_B a Boltzmann constant, and L_0 is the Sommerfield value of the Lorenz number.

III. CALCULATIONS

Calculations for the electrical and thermal conductivities and the Lorenz number have been carried out, by considering the second- and third-order exchange and RKKY interaction contributions separately for two CEF(δ) values 90 and 140 K. Graphs for $\sigma(T)/\sigma_0$, $K(T)/K_0$, and $L(T)/L_0$ have been plotted as a function of T/δ for which the ratio of the square of the inelastic to elastic scattering matrix elements $[(M_i/M_e)^2=\gamma^2]$ is equal to 0.1, J is chosen to be 0.1 eV, $D=10\,000$ K, $N(0)=0.2$ eV^{-1}, $\alpha_1=0.001$ eV, $\alpha_2=0.01$, and RKKY coupling $I=\pm 0.01$ eV have been chosen. Plots obtained for different orders and CEF values, with I positive and negative for the above mentioned values are shown in Figs. 4, 5, and 6, respectively, for electrical and thermal conductivity and the Lorenz ratio, the values of which vary from 0.1 to 0.3 in the range T/δ, are found to exhibit the correct trend with respect to the results reported in the literature for Ce-based heavy-fermion systems.

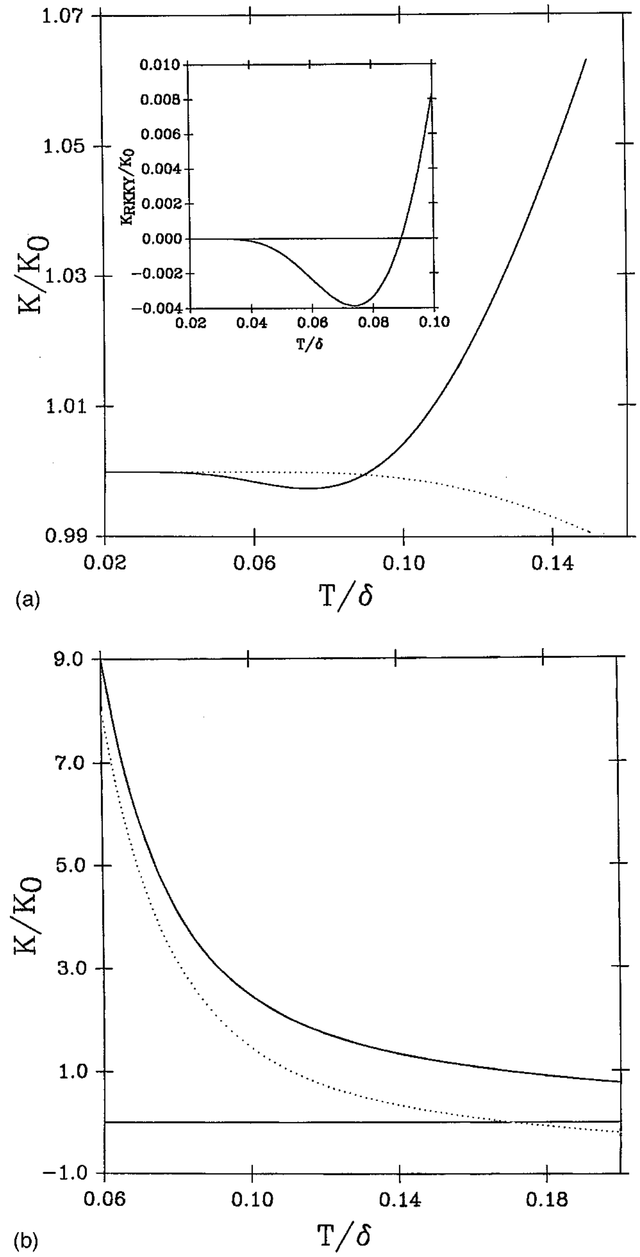

FIG. 5. (a) Electronic thermal conductivity $[K(T)/K_0]$ with a CEF; $\delta=90$ K and the RKKY coupling $I=0.01$ eV. The dashed curve represents the second-order contribution and the solid curve is obtained on adding the RKKY interaction contribution in the second-order exchange contribution. The inset shows the RKKY interaction contribution for the same values of CEF and I. (b) Theoretical plots for electronic thermal conductivity (K/K_0). The dotted curve represents the third exchange and RKKY interaction contribution and the solid curve, i.e., total contribution, is obtained on addition of the second-order exchange contribution to it, for CEF $(\delta)=90$ K and RKKY exchange coupling $I=-0.01$ eV.

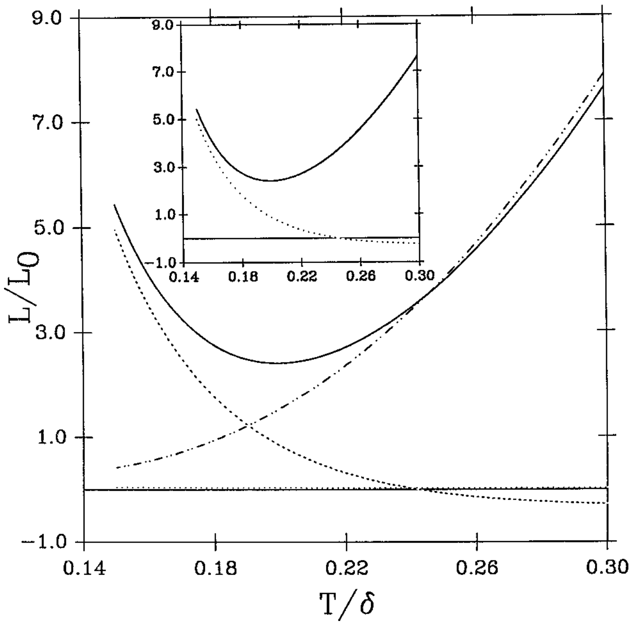

FIG. 6. Theoretical plots for Lorenz ratio $[L(T)/L_0]$ from second-order (dotted line) and third-order exchange (dashed line). RKKY interaction (dotted-dashed line) and total contribution (solid line) with CEF $(\delta)=90$ K and the RKKY coupling $I=0.01$ eV. The inset shows the theoretical plots for the Lorenz ratio from second-plus third-order exchange (dotted line) and total (solid line) contribution for the same CEF and I values.

IV. RESULTS AND DISCUSSION

Contributions to electrical $[\sigma(T)/\sigma_0]$ and thermal $[K(T)/K_0]$ conductivities and Lorenz number $L(T)/L_0$ from different orders, i.e., second, third, RKKY contributions, and total contributions to these, in the presence of a CEF have been calculated systematically. The extrema in the total electrical conductivity occurs around 20 K for $\delta=140$ K and around 13 K for $\delta=90$ K (Fig. 4), while in the thermal conductivity it appears around 6.8 K with $\delta=90$ K [Fig. 5(a)] and around 10.5 K with $\delta=140$ K. Thus the increase in the CEF (δ) values increases the temperature at which the extrema is found. This extrema is minimum for I positive (i.e., for ferromagnetic RKKY coupling) and maximum for I negative (i.e., antiferromagnetic RKKY coupling). Calculations for the Lorenz ratio $[L(T)/L_0]$ also show a minimum around 17 K for $\delta=90$ K and around 27 K for $\delta=140$ K (Fig. 6). Here L_0 is the Sommerfeld value of the Lorenz ratio[12] and is equal to 2.45×10^{-8} W Ω K^{-2}. Furthermore, the calculated value of the Lorenz number at low temperatures is large compared to its Sommerfeld value L_0 and is consistent with the results reported for CeB_6 and $CeCu_6$, which was not explained earlier.[12] From Fig. 6 the role of the third-order exchange interaction in explaining the large L/L_0 ratio at low temperatures is obvious.[17] The clear deviations of thermal and electrical conductivities in HFS's from the conventional Kondo behavior at low temperatures provides a good testing ground for the role of RKKY processes considered here. The Kondo exchange and RKKY interactions show their distinct influence in all properties. It is important to note from Figs. 4 and 5, that in these properties at low temperature the RKKY interaction contribution becomes significant in their total contribution. Furthermore, extrema obtained at finite temperature in these properties in the absence of an external magnetic field can be regarded as the clear cut manifestation of the presence of an internal CEF, which plays a significant part in these systems.[6] Qualitatively the calculations for these properties are quite consistent with the experimental results in the literature.[12,18]

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