From an antiferromagnet to a heavy-fermion system: $CeCu_5Au$ under pressure

H. Wilhelm, S. Raymond, D. Jaccard, O. Stockert†, and H. v. Löhneysen†
Département de Physique de la Matière Condensée, Université de Genève, Quai Ernest-Ansermet 24, 1211 Geneva 4, Switzerland
† *Physikalisches Institut, Universität Karlsruhe, 76128 Karlsruhe, Germany*

The electrical resistivity $\rho(T)$ of single crystalline $CeCu_5Au$ under pressure was measured in the temperature range 30 mK< T < 300 K. Pressure suppresses the antiferromagnetic order (T_N = 2.35 K at ambient pressure) and drives the system into a non–magnetic heavy–fermion state above P_c = 4.1(3) GPa. The electrical resistivity shows a deviation from a T^2 dependence of a Fermi–liquid in the pressure range 1.8 GPa $\leq P \leq$ 5.15 GPa. The $\rho(T)$–curves can be compared with those of $CeCu_{6-x}Au_x$ at different Au concentrations. Just before the long–range magnetic order vanishes, a possibly superconducting phase (at T_c = 0.1 K and P = 3.84 GPa) occurs, pointing to a coexistence of antiferromagnetic order and superconductivity. This new phase is only seen in a narrow pressure interval ΔP = 0.4 GPa.

[phase transition, high pressure, electrical resistivity, non–Fermi–liquid]

1. Introduction

In many heavy–fermion (HF) metals antiferromagnetic (AFM) quantum critical phenomena have been observed. Ternary Ce–based compounds like $CePd_2Si_2$ [1], $CeRu_2Ge_2$ [2], and $CeCu_{6-x}Au_x$ [3] can be tuned to a quantum critical point (QCP) either by pressure or doping. According to the spin–fluctuation theory [4,5,6] the temperature dependence of the electrical resistivity $\Delta\rho(T) \propto T^n$ shows a deviation from the conventional Fermi–liquid (FL) behaviour ($n = 2$) in a limited temperature range. Furthermore, the Néel temperature T_N approaches zero in a characteristic way: $T_N \propto (g_c - g)^m \rightarrow 0$, where g and g_c are the tuning parameter (like concentration x or pressure P) and its critical value. For AFM fluctuations the exponent is $m = 2/3$. This prediction has not been found unequivocally in experiment. Recent high pressure experiments on $CeRu_2Ge_2$ [2] gave $m = 0.70 \pm 0.08$ but reported variations on other compounds are mainly linear in x or P (for more details see Ref. [7] and references therein).

The anomalous exponents found for the temperature dependence of $\rho(T)$ of $CePd_2Si_2$ and $CeNi_2Ge_2$ ($n = 1.2$ [1] and 1.37 [8], respectively) as well as the logarithmic temperature dependence of the specific heat $C/T \propto \ln(T^\star/T)$ observed in $CeCu_{6-x}Au_x$ (see e. g. Ref. [9]) point to a fundamental breakdown of FL theory [10]. The results on high quality samples of $CePd_2Si_2$ [1] and $CeNi_2Ge_2$ [8] have shown that such a non–Fermi–liquid (NFL) behaviour occurs in a small temperature interval. Furthermore, close to the QCP superconductivity emerges in some cases. On the other hand, it is not clear if disorder will modify or even produce the various NFL properties. In a recent theoretical explanation [11] it was argued that the anomalies in $\rho(T)$ can be attributed to the interplay between quantum critical AFM fluctuations and impurity scattering in a conventional FL. Such a consideration, however, is not relevant for the $\Delta\rho(T) \propto T$ behaviour, observed in $CeCu_{5.9}Au_{0.1}$ [3], where the anisotropy of the spin fluctuations seems to play an important role [12,13].

The stoichiometric $CeCu_5Au$ compound has the highest T_N = 2.35 K of all $CeCu_{6-x}Au_x$ alloys with $0.1 \leq x \leq 1.0$ [14] where T_N varies linearly with x. At the critical concentration $x_c = 0.1$ the system loses its long–range magnetic order and the Kondo effect dominates the RKKY interaction (T_K = 6.2 K in $CeCu_6$ [15]). At very low temperature (T < 0.3 K) a FL behaviour is found in $CeCu_6$ [16]. The $T_N(x)$ variation seems to be related to the increase of the unit–cell volume upon doping, but changes in the band structure have to be considered if the different $T_N(x)$ and $T_N(P)$ dependence at equal volume have to be explained. Since in the alloys a certain disorder and a structural transition at low temperature (for $x < 0.15$) exist, measurements on the stoichiometric and single crystalline $CeCu_5Au$ [17] offer a unique possibility to study the pure pressure or volume effect on the magnetic ordering temperature.

Here we present electrical resistivity measurements on single crystalline $CeCu_5Au$ under high pressure (P < 8 GPa). The four–point resistance was measured on a sample in a clamped high pressure cell which was cooled down in a dilution refrigerator (T > 30 mK). Details of the high pressure set–up can be found elsewhere [2].

2. Results and Discussion

Representative electrical resistivity curves $\rho(T)$ of $CeCu_5Au$ are shown in a semi–logarithmic plot in Fig. 1. The low temperature part of the ambient pressure curve is identical to that reported in Ref. [9]. Below T_N = 2.35 K the antiferromagnetically ordered phase is entered, clearly visible by the pronounced cusp in $\rho(T)$. A negative logarithmic slope is present above T_N up to 10 K, reflecting the presence of the Kondo effect (T_K = 1.8 K [18]). A maximum in $\rho(T)$ develops at higher temperature ($T_{max}^{high} \approx$ 60 K). It becomes less pronounced and

shifts slightly down in temperature as pressure is applied. At a moderate pressure of $P = 2.98$ GPa a second maximum at $T_{\rm max}^{\rm low} = 3.5$ K appears, which is well separated from the entrance into the AFM state at $T_{\rm N} = 1.4$ K. It might be related to an already enhanced Kondo effect and could point to the development of a coherent state. The $\rho(T)$–curve at $P = 3.84$ GPa shows a peculiar low temperature behaviour. The entrance into a magnetically ordered phase at $T_{\rm N} \approx 1$ K is still visible but at $T = 0.1$ K the resistivity drops suddenly by more than 10%, indicating the occurrence of a new phase. Its possible nature will be discussed below. At this pressure the two maxima in $\rho(T)$ are still present whereas at higher pressure only $T_{\rm max}^{\rm high}$ remains and a FL behaviour is observed at low temperature. We mention that the residual resistivity exhibits a strong pressure dependence (*cf.* Fig. 1) and details will be given elsewhere [19].

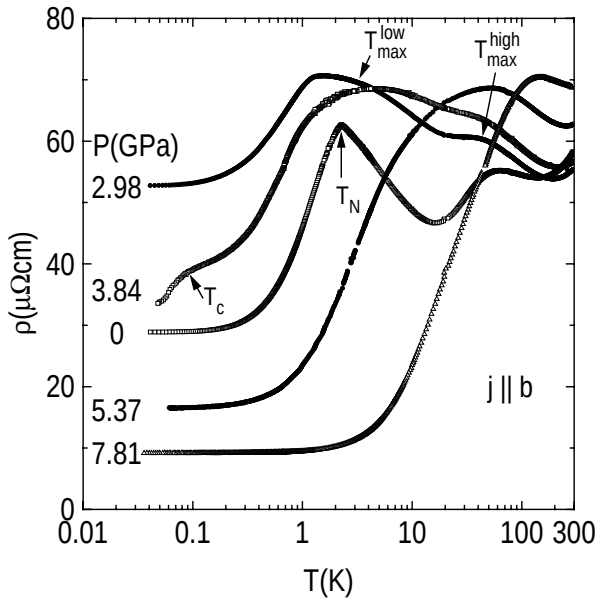

FIG. 1. Temperature dependence of the electrical resistivity of $CeCu_5Au$ at selected pressures. The curve at $P = 3.84$ GPa shows a sudden drop at $T = 0.1$ K, pointing to a new low temperature phase which exists only in a small pressure range ($\Delta P = 0.4$ GPa).

In Fig. 2 the pressure dependence of the characteristic temperatures in $\rho(T)$ of $CeCu_5Au$, obtained on two different pieces cut from the same single crystal, are shown. The open squares show that in one experiment the magnetic order was still visible at $P = 3.84$ GPa, where a pronounced drop in $\rho(T)$ occurred at $T_{\rm c} = 0.1$ K ($\diamond$). Traces of this drop were also found at 4.19 GPa. In a second experiment (filled symbols in Fig. 2) no signs of magnetism above 3.2 GPa and of a drop in resistivity around 3.8 GPa have been found. The $T_{\rm N}(P)$ dependence scales to zero like $T_{\rm N} \propto (P_{\rm c} - P)^m$ at a critical pressure $P_{\rm c} = 4.1 \pm 0.3$ GPa with an exponent $m = 0.68 \pm 0.11$ (inset Fig. 2). This value is in good agreement with $m = 2/3$, predicted within the spin fluctuation theory [6].

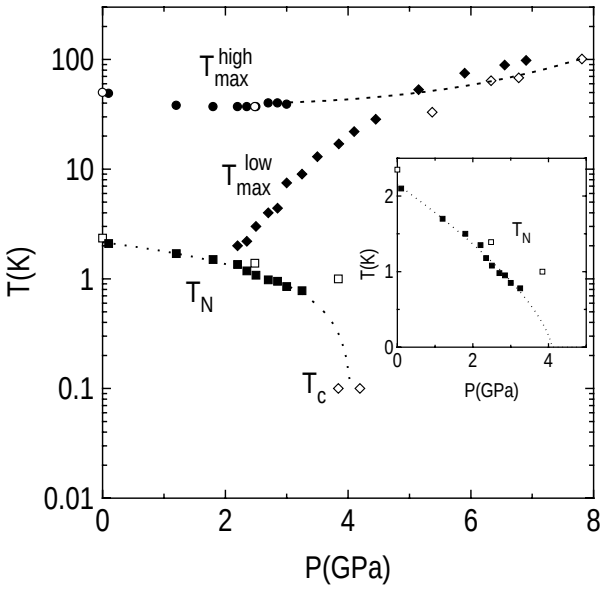

FIG. 2. Pressure dependence of the characteristic temperatures in $\rho(T)$ of $CeCu_5Au$ in a semilogarithmic plot. The Néel temperature ($T_{\rm N}$) scales to zero at $P_{\rm c} = 4.1(3)$ GPa. Two maxima in $\rho(T)$ at $T_{\rm max}^{\rm low}$ and $T_{\rm max}^{\rm high}$, related to the Kondo effect, seem to merge above 4 GPa. In one experiment (open symbols) indications of a new phase (possibly superconducting) below $T_{\rm c} = 0.1$ K ($\diamond$) was found. In the inset the $T_{\rm N}(P)$ dependence is shown in a linear plot.

The Kondo temperature in $CeCu_5Au$ at low pressure is small in comparison to the crystal field (CF) splitting $\Delta_{\rm CF}^{(1)} \approx 100$ K and $\Delta_{\rm CF}^{(2)} \approx 160$ K [20]. Therefore, as often observed in this situation for other compounds, the magnetic resistivity has two maxima at $T_{\rm max}^{\rm low}$ and $T_{\rm max}^{\rm high}$, whose high temperature sides are a sign of the Kondo scattering on the ground state and excited CF levels, respectively, and whose low temperature sides reflect the onset of a coherent heavy–fermion state and the freezing of scattering from CF levels. The pressure variation of $T_{\rm max}^{\rm low}$ might point to the possibility of an enhanced screening (induced by pressure) of the magnetic moments by the conduction electrons and thus to an strengthened role of the Kondo effect. Consequently, the anomaly at $T_{\rm max}^{\rm low}$ has to be related to $T_{\rm K}$. Both anomalies in $\rho(T)$ seem to merge above 4 GPa, indicating the entrance into an intermediate valence regime where the Kondo temperature becomes of the order of the CF splitting.

In the case of two excited CF levels Hanazawa *et al.* [21] have introduced a second Kondo temperature at high temperature $T_{\rm K}^{\rm h} = \sqrt[3]{T_{\rm K}\Delta_{\rm CF}^{(1)}\Delta_{\rm CF}^{(2)}}$. With the assumption that $\Delta_{\rm CF}^{(1)}$ and $\Delta_{\rm CF}^{(2)}$ are hardly changed at low pressure (i. e. $P < 4$ GPa), the $T_{\rm K}^{\rm h}$ values can be calculated if $T_{\rm K}$ is known. For some $CeCu_{6-x}Au_x$ compounds $T_{\rm K}$ has been

determined [22]. To transform the $T_K(V(x))$–dependence in a $T_K(P)$–dependence the relative unit–cell volumes $V(x)/V_0$, with V_0 the unit–cell volume of $CeCu_5Au$ at ambient pressure, have to be transformed into the corresponding pressure values. With the Murnaghan equation of state (EOS) [23] a $T_K(P)$ relation, applicable for $CeCu_5Au$, can be deduced (using $B_0 = 110$ GPa and $B_0' = 4$). This (linear) function then yields the $T_K^h(P)$ dependence. The $T_K^h(P)$ values are practically identical to $T_{max}^{high}(P)$ in the pressure range $2 < P < 3$ GPa. At lower pressure the agreement is not so good which might be related to the presence of magnetic order.

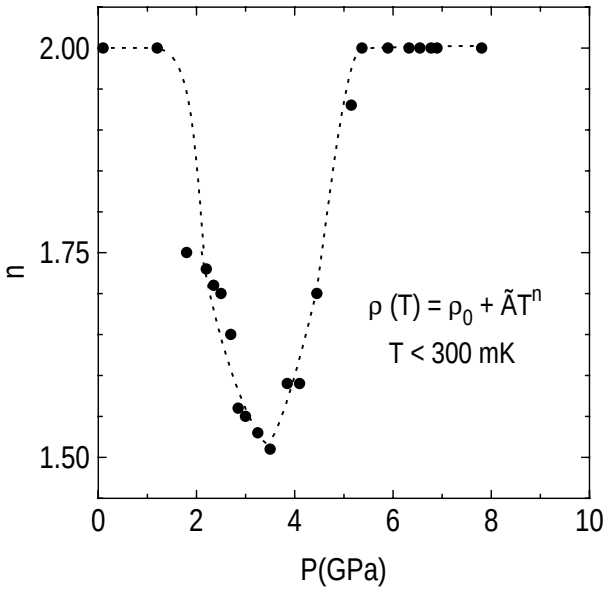

FIG. 3. The exponent n used in the power law of eq. (1) to describe the $\rho(T)$ data of $CeCu_5Au$ below $T = 0.3$ K. Around $P = 3.5$ GPa a clear deviation from a FL ($n = 2$) behaviour is observed. The dashed line is a guide to the eye.

Applying pressure to stoichiometric and single crystalline $CeCu_5Au$ offers the possibility to study the low temperature properties close to the magnetic instability and to compare them with the $CeCu_{6-x}Au_x$ solid solution (with $x \rightarrow x_c$). The electrical resistivity of $CeCu_5Au$ below 0.3 K can be described with

$$\rho = \rho_0 + \tilde{A}T^n \qquad (1)$$

at all pressures. The exponent n and the coefficient $\tilde{A}$ are fitting parameters. The only "constraint" to the fit was the fixation of the upper temperature limit $T_A = 0.3$ K. It is a compromise between an as narrow as possible temperature interval (30 mK$< T <$ 300 mK) and the reliability of the deduced parameters, i. e. n and $\tilde{A}$. Figure 3 illustrates how the deviation from a FL description ($n = 2$) evolves with pressure. Below 1.2 GPa $n = 2$ is consistent with residual electron–magnon scattering in a magnetic system [25]. Then, at 1.8 GPa, n suddenly attains a value of 1.75 and decreases further as pressure increases. At 3.5 GPa $n = 1.51$ is reached, close to the critical value $n = 3/2$, predicted by theory [6]. Increasing pressure further, leads to a higher n value which finally reaches $n = 2$, well inside the non–magnetic region ($P \geq 5.37$ GPa) and comparable to $CeCu_6$ [24]. The minimum in n vs P is not an artefact of the limited temperature interval in the fitting procedure as the fits for various T_A values (up to 0.6 K) showed always a minimum in $n(P)$ around 3.5 GPa, where n is the smaller ($n = 1.2$) the higher the T_A limit (0.6 K) was chosen. In the high pressure non–magnetic region however, the FL value $n = 2$ was found in temperature intervals which became enlarged with pressure.

This behaviour immediately raises the question whether these results can be compared to the observations in $CeCu_{6-x}Au_x$ with different Au concentrations. It is clear that the unit–cell volume variation is a crucial parameter. Thus, a correspondence of the x values in $CeCu_{6-x}Au_x$ to the pressure values in $CeCu_5Au$ is needed. Using the relation $V(x) = 420.225\ Å^3 + 13.988x$, deduced from x–ray data [15] and e. g. the Murnaghan EOS, a relation between x and P can be obtained. For $CeCu_5Au$, this results in a bulk modulus $B_0 = 110$ GPa (with $B_0' = 4$) leading to the correspondence $x = 0.5 \Leftrightarrow P = 1.8$ GPa, $x = 0.1 \Leftrightarrow P = 3.4$ GPa, and $x = 0 \Leftrightarrow P = 3.85$ GPa which should be taken as a guide rather than as a strict prediction.

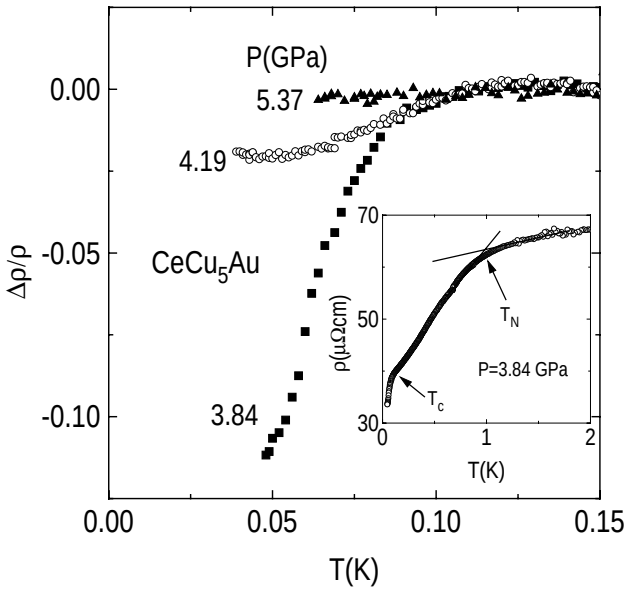

FIG. 4. Electrical resistivity $\rho(T)$ of $CeCu_5Au$ normalized at $T = 0.1$ K at pressures close to the magnetic instability. The strong drop in $\rho(T)$ at $T = 0.1$ K and $P = 3.84$ GPa is interpreted as the entrance into a (probably) superconducting phase. Inset: The anomaly in $\rho(T)$ at $\approx$ 1 K is interpreted as a sign of magnetic order.

Now the peculiar low temperature behaviour of the

$\rho(T)$ curves recorded close to the magnetic instability are of particular interest (Fig. 4). At $P = 3.84$ GPa the entrance into a magnetically ordered phase at $T_N \approx 1$ K is still visible (inset Fig. 4) but at $T = 0.1$ K, $\rho(T)$ drops suddenly more than 10%. This effect can be suppressed if a small magnetic field ($\vec{B} \parallel \vec{c}$, $B = 0.2$ T) is applied. Traces of this transition are also present at $P = 4.19$ GPa, where the resistivity starts to decrease, but not as strong as at the preceding pressure. No signs of a magnetically ordered phase were found. Hence, if magnetism and superconductivity coexist, it occurs in a very narrow pressure range. Well above this pressure, no anomalies in the low temperature part of $\rho(T)$ were found (see curve at $P = 5.37$ GPa in Fig. 4).

If the new phase should be found unequivocally to be a superconducting phase, the measurements show that low residual resistivity is not an important ingredient for superconductivity for $CeCu_5Au$. Just before $\rho(T)$ of $CeCu_5Au$ starts to decrease, the resistivity is close to 40 $\mu\Omega$cm (at 0.1 K and $P = 3.84$ GPa). Furthermore, as in the other pressure–induced HF superconductors, superconductivity then would emerge in the vicinity of the magnetic instability. Therefore, it is an intriguing possibility that AFM spin fluctuations may provide the attractive interaction between quasi–particles which is required to form Cooper–pairs [1]. However, additional experiments are necessary to clarify this point.

3. Conclusion

The electrical resistivity measurements on single crystalline $CeCu_5Au$ showed that the long–range magnetic order is suppressed at $P_c = 4.1(3)$ GPa. Close to the magnetic instability the electrical resistivity (below $T = 0.3$ K) deviates from a Fermi–liquid behaviour. At $P = 3.84$ GPa the pronounced drop in $\rho(T)$ at $T_c = 0.1$ K might point to the existence of a superconducting phase. At this pressure an AFM order is still present ($T_N \approx 1$ K), leading to the possibility to study the coexistence of AFM order and superconductivity as well as NFL behaviour close to a quantum critical point.

4. Acknowledgments

This work was partially supported by the Swiss National Science Foundation.

References

[1] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, R. K. W. Haselwimmer, and G. G. Lonzarich, Nature **394**, 39 (1998).
[2] H. Wilhelm, K. Alami-Yadri, B. Revaz, and D. Jaccard, Phys. Rev. B **59**, 3651 (1999).
[3] H. v. Löhneysen, T. Pietrus, G. Portisch, H. G. Schlager, A. Schröder, M. Sieck, and T. Trappmann, Phys. Rev. Lett. **72**, 3262 (1994).
[4] J. A. Hertz, Phys. Rev. B **14**, 1165 (1976).
[5] A. J. Millis, Phys. Rev. B **48**, 7183 (1993).
[6] T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. **64**, 960 (1995).
[7] A. Huxley, S. Kambe, C. Pfleiderer, H. Suderow, C. Thessieu, A. Buzdin, J. Flouquet, L. Glémot, I. Fomin, and J.-P. Brison, J. Phys. Soc. Jpn. **65**, Suppl. B, 1 (1996).
[8] P. Gegenwart, F. Kromer, M. Lang, G. Sparn, C. Geibel, and F. Steglich, Phys. Rev. Lett. **82**, 1293 (1999).
[9] H. v. Löhneysen, A. Neubert, A. Schröder, O. Stockert, U. Tutsch, M. Loewenhaupt, A. Rosch, and P. Wölfle, Eur. Phys. J. B **5**, 447 (1998).
[10] P. Coleman, Physica B **259-261**, 353 (1999).
[11] A. Rosch, Phys. Rev. Lett. **82**, 4280 (1999).
[12] A. Rosch, A. Schröder, O. Stockert, and H. v. Löhneysen, Phys. Rev. Lett. **79**, 159 (1997).
[13] O. Stockert, H. v. Löhneysen, A. Rosch, N. Pyka, and M. Loewenhaupt, Phys. Rev. Lett. **80**, 5627 (1998).
[14] T. Pietrus, B. Bogenberger, S. Mock, M. Sieck, and H. v. Löhneysen, Physica B **206&207**, 317 (1995).
[15] H. G. Schlager, A. Schröder, M. Welsch, and H. v. Löhneysen, J. Low Temp. Phys. **90**, 181 (1993).
[16] G. R. Stewart, Z. Fisk, amd M. S. Wire, Phys. Rev. B **30**, 482 (1984); A. Amato, D. Jaccard, J. Flouquet, F. Lapierre, J. L. Tholence, R. A. Fisher, S. E. Lacy, J. A. Olsen, and N. E. Phillips, J. Low Temp. Phys. **68**, 371 (1987).
[17] O. Stockert, A. Schröder, H. v. Löhneysen, N. Pyka, E. Garcia-Matres, R. v. d. Kamp, S. Wezel, and M. Loewenhaupt, Physica B **259-261**, 383 (1999).
[18] C. Paschke, C. Speck, G. Portisch, and H. v. Löhneysen, J. Low Temp. Phys. **97**, 229 (1994).
[19] H. Wilhelm, S. Raymond, D. Jaccard, O. Stockert, and H. v. Löhneysen, to be published.
[20] B. Stroka, A. Schröder, T. Trappmann, H. v. Löhneysen, M. Loewenhaupt, and A. Severing, Z. Phys. B **90**, 155 (1993).
[21] K. Hanazawa, K. Yamada, and K. Yoshida, J. Magn. Magn. Mater. **47&48**, 357 (1985).
[22] H. v. Löhneysen, M. Sieck, O. Stockert, and M. Waffenschmidt, Physica B **223&224**, 471 (1996).
[23] F. D. Murnaghan, Proc. Natl. Acad. Sci. USA **30**, 244 (1944).
[24] S. Yomo, L. Gao, R. L. Meng, P. H. Hor, C. W. Chu, and J. Susaki, J. Magn. Magn. Mater. **76&77**, 257 (1988).
[25] B. Coqblin, in *The Electronic Structure of Rare–Earth Metals and Alloys: The Magnetic Heavy Rare–Earths*, edited by B. Coqblin, Academic Press 1977.