Probing the phase diagram of CeRu₂Ge₂ by thermopower at high pressure

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The temperature dependence of the thermoelectric power, S(T), and the electrical resistivity of the magnetically ordered CeRu₂Ge₂ (T_N =8.55 K and T_C =7.40 K) were measured for pressures p < 16 GPa in the temperature range 1.2 K < T < 300 K. Long-range magnetic order is suppressed at $p_c \approx 7.8$ GPa. Pressure drives S(T) through a sequence of temperature dependences, ranging from a behavior characteristic for magnetically ordered heavy fermion compounds to a typical behavior of intermediate-valent systems. At intermediate pressures a large positive maximum develops above 10 K in S(T). Its origin is attributed to the Kondo effect and its position is assumed to reflect the Kondo temperature T_K . The pressure dependence of T_K is discussed in a revised and extended (T, p) phase diagram of CeRu₂Ge₂.

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I. INTRODUCTION

A large number of heavy fermion (HF) or unstable valence compounds and alloys revealed a complicated temperature (T) dependence of the thermoelectric power (S). This is due to competing interactions present in these systems and the sensitivity of S to details of the band structure. Depending on the strength of the hybridization between 4f and conduction electrons, the exchange interaction J leads to longrange magnetic order, magnetic Kondo systems, HF, or intermediate-valence (IV) behavior. As a measure for the strength of J of these four regimes one can use the Kondo temperature T_K and the Néel temperature T_N (or the Curie temperature T_C). Sakurai and co-workers¹ used the ratio of T_K to T_N to classify the S(T) data of different compounds into these regimes, each showing a characteristic S(T) dependence. Ce-based representatives of each regime investigated by S(T) are CeAu₂Si₂,² CeAl₂,³ CeAl₃,⁴ and CeRu₂Si₂ (Refs. 5 and 6) or CeNi₂Si₂.^{7,8}

Typical features in S(T) of these regimes can be seen in a single compound if *J* is increased, e.g., by alloying. Increasing *x* in the pseudobinary alloy Ce(Pb_{1-x}Sn_x)₃ (Ref. 9) and in the solid-solution CeRh_{2-x}Ni_xSi₂ (Ref. 7) tunes the systems from trivalent to intermediate valent, i.e., enhancing *J*. Also the interatomic distances seem to control the shape of S(T). This results from an analysis of the S(T) data of many Ce M_2X_2 compounds, with *M* a transition metal and X=Si or Ge.¹⁰ Apparently, in the series Ce M_2Si_2 , where the corrected interatomic distances decrease for M=Au, Cu, Rh, Ru, and Fe, the increase of *J* is accompanied with a characteristic variation of S(T) at low temperatures.

This systematic volume dependence suggests that S(T) is very sensitive to pressure. Based on the pressure-induced changes in S(T), Link *et al.*¹¹ sketched a sequence of S(T)dependences that shows the evolution of characteristic features in S(T) with pressure. The S(T) of a magnetically ordered system, such as CeCu₂Ge₂, shows one negative and one positive peak below ≈ 20 K and above about 100 K, respectively.¹⁰ In general, pressure will suppress the negative peak and lead to the appearance of new features at low temperatures. As pressure has tuned the system into the IV regime, only one maximum well above room temperature remains, like in CeNi2Si2 at ambient pressure.8 Similarly, the application of pressure on the nonmagnetic HF compounds CeCu₂Si₂ ($T_K \approx 20$ K) (Ref. 12) and CeAl₃ ($T_K \approx 5$ K) (Ref. 13) reduces the magnitude of the negative peak and leads to the development of two additional positive peaks around 20 K and even lower temperatures. The analysis of these pressure-induced S(T) dependences has led to the conjecture that the pressure-induced maximum in S(T) located at approximately 20 K is related to the Kondo effect.¹¹ The origin of the high-temperature maximum, present already at low pressure, is due to the crystal-field separation of the Ce 4felectron energy level from the ground state.¹⁴ Zlatić and co-workers¹⁵ achieved a qualitative understanding of the experimental results, using the Coqblin-Schrieffer model and assuming a splitting of the 4f states in the presence of a crystalline electric field.

The magnetically ordered CeRu₂Ge₂ offers the possibility to map the pressure dependence of the characteristic features in S(T) since its pressure-induced transition into the HF regime was intensively studied.^{16–21} The ambient pressure S(T)curve¹⁷ contains no maximum below room temperature, which is a consequence of the large energy separation of the crystal-field levels [Δ_1 =500 K and Δ_2 =750 K (Refs. 22 and 23)]. Assuming that the crystal-field levels are not strongly influenced by pressure, two distinct positive peaks at low and high temperatures in S(T) at intermediate pressures are expected. This could provide insight into the pressure dependence of T_K . Moreover, the S(T) data might reveal information about the pressure-induced change from a magnetically ordered compound (at p=0) to a HF Kondo-lattice compound (at high pressure), equivalent to $CeRu_2Si_2$ [$T_K=24$ K (Ref. 24)] at ambient pressure.



FIG. 1. Top view of the inner part of the pressure chamber before closing. Close to the heater two thermocouples (<u>Au</u>Fe and Chromel) are located on top of the sample. The Au wire connected at the opposite edge of the sample is chosen as a reference point for the two thermovoltages $V_{\underline{AuFe}}$ and $V_{\underline{Chromel}}$. Lead is used as pressure gauge. Au wires establish the connection through the pyrophyllite gasket. Steatite serves as pressure transmitting medium.

II. EXPERIMENTAL DETAILS

The CeRu₂Ge₂ and LaRu₂Ge₂ samples have been prepared by arc-melting stoichiometric amounts of Ce, La, Ru, and Ge under argon atmosphere. The purity of the elements was 99.99%, except for Ge, which had a purity of 99.999%. The x-ray diffraction pattern could be indexed according to the ThCr₂Si₂ structure (*I*4/*mmm*) with lattice parameters *a* =4.2685(4) Å and *c*=10.048(3) Å for CeRu₂Ge₂ and *a* =4.314(2) Å and *c*=10.129(6) Å for LaRu₂Ge₂. Samples of the CeRu₂Ge₂ ingot have been used in earlier electrical resistivity, $\rho(T)$,^{16,17} and specific heat¹⁸ pressure studies.

A clamped Bridgman anvil cell with synthetic diamonds was used to measure S(T) and $\rho(T)$ in the temperature range 1.2 K < T < 300 K. The pressure chamber was made of a nonmetallic gasket (pyrophyllite, $\phi_{int}=1$ mm) and two steatite disks served as the pressure transmitting medium. Electrical leads were attached to the sample (cross section of 14×108 μ m²) in such a way that a four-point $\rho(T)$ and, in a separate run, a S(T) measurement could be performed (Fig. 1). The pressure dependence of the superconducting transition temperature of Pb yielded the pressure.²⁵

A heater (Chromel wire), located close to the small edge of the sample, produced a temperature gradient ΔT along the sample. The opposite edge of the sample remained at T_0 , the temperature of the pressure cell,²⁶ and served as a reference for the two thermocouples AuFe (with 0.07 at. % Fe) and Chromel. The two measured thermovoltages were V_{AuFe} = $(S_{AuFe}-S)\Delta T$ and $V_{Chromel}=(S_{Chromel}-S)\Delta T$. The absolute thermoelectric power of the sample, *S*, at $T_0+\Delta T/2$ is given by

$$S = S_{AuFe} + \frac{S_{Chromel} - S_{AuFe}}{1 - V_{Chromel} / V_{AuFe}}.$$
 (1)



FIG. 2. Magnetic contribution $\rho_{mag}(T)$ to the electrical resistivity of CeRu₂Ge₂ at different pressures. Two different antiferromagnetic phases occur below T_N and T_L . A ferromagnetic ground state is present below T_C and low pressure. No traces of magnetic order are observed for p > 7 GPa above 1.2 K. Inset: raw data of $\rho(T)$ in a linear plot. Incoherent Kondo scattering is apparent for p = 5.7 GPa.

The absolute thermopower of AuFe and Chromel is assumed to be pressure independent. This seems to be a good assumption since the absolute value of S_{AuFe} at 12 GPa and 4.2 K is only 20% smaller than at ambient pressure.²⁷ However, small pressure-induced changes in S(T) of the sample should be interpreted carefully.

III. RESULTS

Figure 2 shows the magnetic part, $\rho_{mag}(T)$, of $\rho(T)$ of CeRu₂Ge₂ at several pressures. It was obtained by subtracting a phonon contribution, approximated as $\rho_{ph}(T)$ =0.12 $\mu\Omega$ cm/K \times T, from the raw data shown in the inset to Fig. 2. The slope $\partial \rho(T) / \partial T = 0.12 \ \mu\Omega \ cm/K$ was deduced from our $\rho(T)$ measurement of the nonmagnetic reference compound LaRu₂Ge₂ at ambient pressure for T > 70 K. This approximation had to be used, since the measured value of $\rho(T)$ of LaRu₂Ge₂ is slightly overestimated, presumably due to microcracks. At low temperature $\rho_{mag}(T)$ is dominated by the magnetic phase transitions, manifested by several discontinuities in $\rho_{mag}(T)$. The transition temperatures were defined by the intersection of two tangents drawn to the $\rho(T)$ curve below and above the kink. A high-temperature peak below room temperature evolves for pressures in the range 7.0 $\leq p \leq 10.4$ GPa. It is due to the interplay of the Kondo and crystal-field effects. At intermediate temperatures and pressures (e.g., at about 20 K and 5.7 GPa in the inset to Fig. 2), incoherent Kondo scattering is clearly increasing. In contrast to other compounds like CePd₂Ge₂,²⁷ CeCu₂Ge₂,²⁸ and



FIG. 3. Temperature dependence of the thermoelectric power S(T) of CeRu₂Ge₂ at selected pressures. The entrance into the magnetically ordered states is indicated by T_N , T_C , and T_L , as deduced from the $\rho(T)$ data. T_K and T_S label the center of broad, pressure-induced maxima. The inset shows the low-temperature part of S(T) at low pressure.

CeCu₅Au,²⁹ its contribution cannot be deconvoluted due to its modest magnitude.

The interpretation of the high-temperature maximum in $\rho_{mag}(T)$ as a result of Kondo exchange interaction between the conduction electrons and the crystal-field split ground state of the Ce³⁺ ions is supported by the evolution of the positive peak in S(T) below 300 K for p > 3.4 GPa (see Figs. 3 and 4). Its position T_S corresponds to a fraction of the crystal-field splitting as in many other Ce-based compounds and alloys. In CeRu₂Ge₂, T_S first decreases linearly with pressure (-23 K/GPa), attains a minimum around 200 K at about 9 GPa, and then starts to increase. Based on this $T_S(p)$ variation a maximum in S(T) is expected to occur at about 384 K at ambient pressure, well above the limit of our setup. The amplitude of the peak grows linearly with pressure [6.4 μ V/(K GPa)] and attains a maximum value of 55 μ V/K at about 10 GPa (Fig. 4).

The complicated S(T) below 10 K (inset to Fig. 3) is very likely caused by the onset of magnetic order and to the opening of a spin gap [Δ =14 K (Refs. 17 and 30)]. Pressureinduced changes of the Fermi surface due to the periodicity of the magnetic ordering (magnetic superzone effects) might be the explanation for the different low-temperature S(T) dependences for p < 3.4 GPa. At 5.7 GPa, S(T) is positive over the entire temperature range and a Kondo maximum at about T_K =12 K (incoherent scattering on the ground state) occurred (Fig. 3). The position of the maximum has shifted considerably towards higher temperatures in S(T) recorded at 7.0 GPa, supporting its assignment to the Kondo effect. A trace of this maximum can even be anticipated around 40 K at 8.0 GPa as a weak shoulder at the high-temperature maximum entered at $T_S \approx 200$ K (Fig. 4). At sufficiently high



FIG. 4. Thermoelectric power S(T) of CeRu₂Ge₂ above the critical pressure. S(T) is dominated by a high-temperature maximum. The inset shows the ambient pressure S(T) data of CeRu₂Ge₂ (Ref. 17) and its nonmagnetic counterpart LaRu₂Ge₂.

pressure, both peaks merge. Then the Kondo maximum dominates the high-temperature maximum as the crystal-field effect disappears when the system enters the IV regime. At 15.6 GPa, the S(T) maximum is well above room temperature. Thus, the overall influence of pressure on S(T) of CeRu₂Ge₂ fits rather well the behavior sketched in Ref. 11.

The effect of magnetic ordering in S(T) is obvious as CeRu₂Ge₂ is compared with the nonmagnetic LaRu₂Ge₂ (inset to Fig. 4). LaRu₂Ge₂ has a rather small S (-3.5μ V/K at 300 K) that decreases almost linearly in magnitude with decreasing temperature and reaches $S \approx 0$ at $T \approx 10$ K.^{31,32} Thus, the low-temperature anomalies in S(T) of CeRu₂Ge₂ at ambient pressure are caused by the occurrence of magnetic order. This implies that S(T) of CeRu₂Ge₂ above T_N can be regarded as the superposition of the pressure independent linear-in temperature electron-phonon term represented by LaRu₂Ge₂ and the incipient contribution characteristic of several HF compounds. That contribution has a negative peak centered at 80 K (ascribed to spin interactions¹¹) and a positive peak above room temperature (caused by the interplay of Kondo and crystal-field effects). At ambient pressure, the absolute value of S is small [as for, e.g., $CeAu_2Si_2$ (Ref. 33)] because the compound is far below the critical pressure.

IV. DISCUSSION

Figure 5 shows a revised and extended (T,p) phase diagram of CeRu₂Ge₂ based on the present S(T) and $\rho(T)$ data as well as $\rho(T)$ and calorimetric experiments performed on samples of the same batch.^{16–18} The different magnetic phases in CeRu₂Ge₂ are assumed to be the same as those found in the solid-solution CeRu₂(Si_{1-x}Ge_x)₂ for $0 \le x \le 1.^{34,35}$ For details about the magnetic structures the





FIG. 5. (T,p) phase diagram of CeRu₂Ge₂ obtained from electrical resistivity (half open symbols) (Refs. 16 and 17) calorimetric open symbols (Ref. 18), and the combined $\rho(T)$ and S(T) (bold symbols) measurements. At low pressure a paramagnetic (PM) to antiferromagnetic (AFM I) phase transition occurs at T_N and a subsequent transition into a ferromagnetic phase (FM) takes place at T_C . The FM ground state is suppressed at 2.3(2) GPa and a second antiferromagnetic order is suppressed at $p_c \approx 7.8$ GPa. T_ρ is the position of a maximum in $\rho(T)$ whereas T_S and T_K represent the center of peaks in S(T). The half-filled diamonds indicate $T_K(p) \propto 1/\sqrt{A^{17}}$. They are shifted by 1.4 GPa towards lower pressure (see text).

reader is referred to Ref. 30 and references therein. The pressure on CeRu₂Ge₂ or replacing Ge by Si in CeRu₂(Si_{1-r}Ge_r)₂ is equivalent, since the unit-cell volume seems to be the crucial parameter to change J. This was concluded from the common (T, V) phase diagram of both systems.^{17,20} The main observation of interest is that the long-range magnetic order is suppressed at a critical pressure p_c , and a HF behavior equivalent to CeRu₂Si₂ at ambient pressure is expected for $p > p_c$. The S(T) data suggest that long-range magnetic order is suppressed between 5.7 and 7 GPa (Fig. 3). Thus, the transition pressure is similar to that deduced from calorimetric measurements $[p_c \approx 6.9 \text{ GPa (Ref. 18)}]$ but is quite low compared to that obtained by electrical resistivity [$p_c = 8.7$ GPa (Refs. 16 and 17)]. This is partly due to the criterion used for the determination of p_c . As far as $\rho(T)$ data of Ref. 17 are concerned, the pressure where the anomaly in the $\tilde{A}(p)$ occurs (at p=7.8 GPa) seems to be a better choice of p_c than the $T_N(p) \rightarrow 0$ extrapolation (p=8.7 GPa). A is an adjustable parameter in a fit of $\rho(T) = \rho_0 + \tilde{A}T^n$ to the data below 1.5 K,¹⁷ with ρ_0 the measured residual resistivity and the exponent *n*.

Knowing the (T, p) phase diagram one is tempted to label the features in S(T) (Fig. 3). It seems that a discontinuity in the slope of S(T) can be used as definition of T_N for p

FIG. 6. Comparison of S(T) of CeRu₂Ge₂ at 7.0 GPa and CeRu₂Si₂ at ambient pressure (Ref. 37). Both curves reveal a pronounced high-temperature maximum and a moderate positive contribution to S(T) at about 25 K. Inset: The low temperature part of S(T) of CeRu₂Si₂ contains an additional positive peak below 1 K.

 ≤ 2.1 GPa, similar to the case of CeCu₂Ge₂.¹⁰ At p \geq 5.7 GPa, however, T_N yields no signature in S(T) as, e.g., in the case of CeAl₂ [T_N =3.8 K (Ref. 3) at p=0]. From the $\rho(T)$ data recorded at 5.7 GPa, it is inferred that T_N is located slightly below the maximum of the broad peak centered at 12 K. The signature of the FM transition is obscure. If it is related to the strong decrease of S(T) below about 7 K (for $p \leq 0.9$ GPa) then the rather constant S(T) between 2 and 5 K found at 2.1 GPa (inset to Fig. 3) would indicate that the ferromagnetic order is already suppressed. The maximum at about 3 K in the 5.7 GPa data seems to be caused by T_L . The assignment of the remaining, strongly pressure-dependent pronounced maximum as T_K is then quite obvious for p \geq 5.7 GPa. It might be speculated whether the broad feature below 2 K (for $p \leq 3.4$ GPa) is a signature of T_K or related to the magnetic order.

The interpretation of this pronounced low-temperature feature as signature of the Kondo effect is supported by the comparison of S(T) of CeRu₂Ge₂ at 7.0 GPa and CeRu₂Si₂ at ambient pressure⁶ (Fig. 6). It is important to note that in both cases S(T) was measured perpendicular to the c axis of the tetragonal crystal structure. The low-temperature maximum in CeRu₂Si₂ occurs at 25 K close to the value of T_{K} =24 K extracted from specific heat measurements using a singleimpurity model.²⁴ Reminiscent of this is the weak maximum in S(T) at 28 K for CeRu₂Ge₂ at 7.0 GPa. This agreement is not accidental since also the high-temperature maximum occurs at the same temperature T_s and has a similar magnitude as for $CeRu_2Si_2$. In S(T) of $CeRu_2Si_2$ it coincides with the temperature of the first excited crystal-field level (Δ_1 =220 K), deduced from a Schottky anomaly in the specific heat.²⁴ Such a perfect agreement of calorimetric and transport data is fortuitous since it depends on T_K , the crystal-field splitting, and the degeneracy of the crystal-field levels.³⁶ However, it applies also to CeCu₆ (Ref. 37) and CePd₂Si₂.¹¹ Thus, the peak in S(T) of CeRu₂Ge₂ at T_S can be ascribed to the interplay between incoherent Kondo scattering and crystal-field effects. This comparison shows that the first excited crystal-field level in CeRu₂Ge₂ decreased from about 500 K (Refs. 22 and 23) to about 200 K for pressures in the vicinity of p_c and explains why a high-temperature maximum in $\rho(T)$ (Fig. 2) was only seen in a certain pressure range above p_c . Furthermore, the resemblance of both S(T) curves confirms the assumption of $p_c < 7.0$ GPa for the sample studied in the present experiment.

The qualitative and quantitative agreement of both data sets can even be used to state that the occurrence of an inflection point in S(T) of CeRu₂Ge₂ at about 50 K might imply the change of regime as in CeRu₂Si₂.⁶ Slightly above this temperature, short-range AFM intersite correlations emerge in CeRu₂Si₂ at ambient pressure.^{38,39} In addition a μ SR investigation⁴⁰ revealed magnetic correlations below 1 K. These quasistatic correlations involve very small magnetic moments of the order of $10^{-3} \mu_B$. The origin of this weak static magnetism well below T_K might be due to the formation of HF magnetism involving the renormalized quasiparticles.⁴⁰ The presence of such correlations might be responsible for the positive contribution to S(T) below 1 K (inset to Fig. 6) and can be expected to occur also in CeRu₂Ge₂ below 1 K.

The very rapid increase of T_K with pressure is corroborated by the reported pressure dependence of the A coefficient,¹⁷ which was deduced from a fit of $\rho(T) = \rho_0$ $+AT^{2}$ to the data for T < 0.5 K. Assuming that the relation $A \propto 1/T_{K}^{2}$ holds, we calculated $T_{K}(p)$ and added it to the phase diagram (Fig. 5). In order to do so, these values were normalized in such a way that at $p = p_c + 0.6$ GPa (with p_c =7.8 GPa) the value of T_K of CeRu₂Si₂ at ambient pressure was obtained. The small pressure offset corresponds to the fact that long-range magnetic order in $CeRu_2(Si_{1-x}Ge_x)_2$ vanishes at $x_c \approx 0.06$.⁴¹ Furthermore, the calculated $T_K(p)$ data had to be shifted towards lower pressure to account for the different p_c values (assumption: $\Delta p_c = -1.4$ GPa). The normalization and the pressure shift are in part arbitrary because the S(T) contribution gives only an estimation of T_K and the p_c value of the sample studied by S(T) cannot be deduced precisely. But the pressure variations are reliable and noteworthy. Note here that one compares the energy scale T_K with the ground-state excitations, implying that only one energy scale exists and that notably T_K is finite at p_c . The $T_K(p)$ dependence revealed the interesting finding that two pressure ranges exist with quite different slopes in the T_K vs p plot. Comparable $\partial T_K / \partial p$ values are observed for pressures ranging from about 6.5 GPa-7.5 GPa and above 8.5 GPa. A much larger slope is found at intermediate pressures (7.5 GPa $\leq p \leq 8.5$ GPa), i.e., around p_c . The transition from a large to a small $\partial T_K / \partial p$ value is reminiscent of the change in the slope of T_K vs p observed in CeRu₂Si₂ at about 1 GPa.⁴² A weaker than exponential variation of $T_K(p)$ due to intersite interaction, proposed in Ref. 43, is not seen. If such a trend exists it seems to be restricted to pressures well below p_c .

The extrapolation of $T_K(p)$ seems to merge with $T_S(p)$ at pressures above 11 GPa, indicating the entrance into the IV regime (Fig. 5). Here, $k_B T_K$ exceeds the crystal-field splitting and the entire sixfold degeneracy of the Ce-4 f^1 multiplet is recovered. So far, similar pressure dependences of two maxima in $\rho(T)$ were found for CeCu₂Ge₂,²⁸ CePd₂Si₂,⁴⁴ CePd₂Ge₂,²⁷ and CeCu₅Au.²⁹ The pressure $p_v \approx 11$ GPa for CeRu₂Ge₂, where $T_K \approx T_S$, might define the region where the valence of the Ce ion starts to increase.

Band structure calculations⁴⁵ supported, e.g., by de Haasvan Alphen (dHvA) measurements⁴⁶ have shown that the $4f^1$ Ce electron does not participate in the Fermi surface of CeRu₂Ge₂ as is the case for CeRu₂Si₂.⁴⁷⁻⁴⁹ The comparison between the dHvA frequency branches of CeRu₂Ge₂ and the theoretical branches of LaRu₂Ge₂ implies that the 4f electron in CeRu₂Ge₂ is fully localized in the ground state.⁴⁵ One of the original motivations of the present transport investigation was to follow the promotion of this 4f electron. However, the data do not show any distinct pressure or a pressure range where the $4f^1$ electron becomes delocalized in CeRu₂Ge₂. Neither the residual resistivity²⁸ nor a plot of S/T at 1.5 K reveals a distinct feature related to the promotion of the $4f^1$ electron. S/T at 1.5 K has a marked maximum around 6 GPa but it seems to be correlated to T_L . It is also noteworthy that close to p_c , S/T vs T is without any anomaly and does not confirm the predictions of Ref. 50. Thus, the question can be asked whether the nature of the pressure-induced quantum discontinuity is of second order or if it really exists in CeRu₂Ge₂.

V. CONCLUSION

The influence of pressure on the temperature dependence of the thermoelectric power S(T) of CeRu₂Ge₂ was measured up to 16 GPa. The various magnetic phase transitions below 10 K yield a complex S(T) behavior. A large positive peak in S(T) centered at T_S just below room temperature starts to develop at pressures above 3 GPa. It is ascribed to the interplay between incoherent Kondo scattering and crystal-field effects. A pressure-induced low-temperature maximum in S(T) at T_K develops in the range 5.7 GPa $\leq p \leq 8$ GPa. It is interpreted as a signature of the Kondo effect, since its position shows a similar pressure dependence as $1/\sqrt{A(p)}$. This implies that only one energy scale seems to exists in $CeRu_2Ge_2$. A revised (T,p) phase diagram, based on transport and calorimetric investigations on samples of one batch, suggests that in CeRu₂Ge₂ long-range magnetic order is suppressed at $p_c \approx 7.8$ GPa. Well above p_c , the $T_K(p)$ and $T_S(p)$ dependences merge at a pressure $p_v \approx 11$ GPa, defining a pressure range where the Ce valence starts to increase.

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- ¹J. Sakurai, H. Takagi, S. Taniguchi, T. Kuwai, Y. Isikawa, and J.-L. Tholence, J. Phys. Soc. Jpn. Suppl. B 65, 49 (1996).
- ²A. Amato and J. Sierro, J. Magn. Magn. Mater. **47-48**, 526 (1985).
- ³D. Jaccard and J. Sierro, in *Valence Instabilities*, edited by P. Wachter and H. Boppart (North-Holland, Amsterdam, 1982), p. 409.
- ⁴D. Jaccard and J. Flouquet, J. Magn. Magn. Mater. **47-48**, 45 (1985).
- ⁵A. Amato, D. Jaccard, J. Sierro, F. Lapierre, P. Haen, P. Lejay, and J. Flouquet, J. Magn. Magn. Mater. **76-77**, 263 (1988).
- ⁶A. Amato, D. Jaccard, J. Sierro, P. Haen, P. Lejay, and J. Flouquet, J. Low Temp. Phys. **77**, 195 (1989).
- ⁷E. V. Sampathkumaran, R. Vijayaraghavan, A. Adam, Y. Yamamoto, Y. Yamaguchi, and J. Sakurai, Solid State Commun. **71**, 71 (1989).
- ⁸E. M. Levin, R. V. Lutsiv, L. D. Finkel'shtein, N. D. Samsonova, and R. I. Yasnitskii, Sov. Phys. Solid State 23, 1403 (1981).
- ⁹J. Sakurai, H. Kamimura, and Y. Komura, J. Magn. Magn. Mater. 76-77, 287 (1988).
- ¹⁰D. Jaccard, K. Behnia, and J. Sierro, Phys. Lett. A **163**, 475 (1992).
- ¹¹P. Link, D. Jaccard, and P. Lejay, Physica B 225, 207 (1996).
- ¹²D. Jaccard, J. M. Mignot, B. Bellarbi, A. Benoit, H. F. Braun, and J. Sierro, J. Magn. Magn. Mater. **47-48**, 23 (1985).
- ¹³C. Fierz, D. Jaccard, J. Sierro, and J. Flouquet, J. Appl. Phys. 63, 3899 (1988).
- ¹⁴A. K. Bhattacharjee and B. Coqblin, Phys. Rev. B **13**, 3441 (1976).
- ¹⁵ V. Zlatić, B. Horvatić, I. Milat, B. Coqblin, G. Czycholl, and C. Grenzebach, Phys. Rev. B 68, 104432 (2003).
- ¹⁶H. Wilhelm and D. Jaccard, Solid State Commun. **106**, 239 (1998).
- ¹⁷H. Wilhelm, K. Alami-Yadri, B. Revaz, and D. Jaccard, Phys. Rev. B **59**, 3651 (1999).
- ¹⁸F. Bouquet, Y. Wang, H. Wilhelm, D. Jaccard, and A. Junod, Solid State Commun. **113**, 367 (2000).
- ¹⁹T. C. Kobayashi, T. Miyazu, K. Shimizu, K. Amaya, Y. Kitaoka, Y. Nuki M. Shirase, and T. Takabatake, Phys. Rev. B **57**, 5025 (1998).
- ²⁰S. Süllow, M. C. Aronson, B. D. Rainford, and P. Haen, Phys. Rev. Lett. **82**, 2963 (1999).
- ²¹A. Demuer, C. Marcenat, J. Thomasson, R. Calemczuk, B. Salce, P. Lejay, D. Braithwaite, and J. Flouquet, J. Low Temp. Phys. **120**, 245 (2000).
- ²²R. Felten, G. Weber, and H. Rietschel, J. Magn. Magn. Mater. 63-64, 383 (1987).
- ²³A. Loidl, K. Knorr, G. Knopp, A. Krimmel, R. Caspary, A. Böhm, G. Sparn, C. Geibel, F. Steglich, and A. P. Murani, Phys. Rev. B 46, 9341 (1992).
- ²⁴M. J. Besnus, J. P. Kappler, P. Lehmann, and A. Meyer, Solid State Commun. 55, 779 (1985).

- ²⁵B. Bireckhoven and J. Wittig, J. Phys. E **21**, 841 (1988).
- ²⁶D. Jaccard, E. Vargoz, K. Alami-Yadri, and H. Wilhelm, Rev. High Pressure Sci. Technol. 7, 412 (1998).
- ²⁷H. Wilhelm and D. Jaccard, Phys. Rev. B **66**, 064428 (2002).
- ²⁸D. Jaccard, H. Wilhelm, K. Alami-Yadri, and E. Vargoz, Physica B **259–261**, 1 (1999).
- ²⁹ H. Wilhelm, S. Raymond, D. Jaccard, O. Stockert, H. v. Löhneysen, and A. Rosch, in *Science and Technology of High Pressure*, edited by M. H. Manghanani, W. J. Nellis, and M. F. Nicol (Universities Press, Hyderabad, India, 2000), p. 697; cond-mat/ 9908442 (unpublished).
- ³⁰S. Raymond, P. Haen, R. Calemczuk, S. Kambe, B. Fåk, P. Lejay, T. Fukuhara, and J. Flouquet, J. Phys.: Condens. Matter **11**, 5547 (1999).
- ³¹The tiny oscillation below 10 K is very likely an artefact of the measurement. The origin of the weak anomaly ($\approx 1 \ \mu V/K$) close to 70 K is not evident; a similar structure was also observed in LaPd₂Si₂ (Ref. 32).
- ³² Y. Bando, J. Sakurai, and E. V. Sampathkumaran, Physica B 186-188, 525 (1993).
- ³³P. Link and D. Jaccard, Physica B **230-232**, 31 (1997).
- ³⁴P. Haen, F. Mallmann, M. J. Besnus, J. P. Kappler, F. Bourdarot, P. Burlet, and T. Fukuhara, J. Phys. Soc. Jpn. Suppl. B **65**, 16 (1996).
- ³⁵P. Haen and T. Fukuhara, Physica B **312-313**, 437 (2002).
- ³⁶S. Maekawa, S. Kashiba, M. Tachiki, and S. Takahashi, J. Phys. Soc. Jpn. 55, 3194 (1986).
- ³⁷A. Amato, Ph.D. thesis, University of Geneva, 1988.
- ³⁸L. P. Regnault, W. A. C. Erkelens, J. Rossat-Mignod, P. Lejay, and J. Flouquet, Phys. Rev. B **38**, 4481 (1988).
- ³⁹J. Rossat-Mignod, L. P. Regnault, J. L. Jacoud, C. Vettier, P. Lejay, J. Flouquet, E. Walker, D. Jaccard, and A. Amato, J. Magn. Magn. Mater. **76-77**, 376 (1988).
- ⁴⁰A. Amato, R. Feyerherm, F. N. Gygax, A. Schenck, J. Flouquet, and P. Lejay, Phys. Rev. B 50, 619 (1994).
- ⁴¹P. Haen, H. Bioud, and T. Fukuhara, Physica B **281-282**, 59 (2000).
- ⁴² K. Payer, P. Haen, J.-M. Laurant, J.-M. Mignot, and J. Flouquet, Physica B **186-188**, 503 (1993).
- ⁴³J. R. Iglesias, C. Lacroix, and B. Coqblin, Phys. Rev. B 56, 11820 (1997).
- ⁴⁴A. Demuer, A. T. Holmes, and D. Jaccard, J. Phys.: Condens. Matter 14, L529 (2002).
- ⁴⁵H. Yamagami and A. Hasegawa, J. Phys. Soc. Jpn. **63**, 2290 (1994).
- ⁴⁶C. A. King and G. G. Lonzarich, Physica B **171**, 161 (1991).
- ⁴⁷ H. Aoki, M. Takashita, N. Kimura, T. Terashima, S. Uji, T. Matsumoto, and Y. Ōnuki J. Phys. Soc. Jpn. **70**, 774 (2001).
- ⁴⁸Y. Ōnuki, R. Settai, and H. Aoki, Physica B **223-224**, 141 (1996).
- ⁴⁹G. Zwicknagl, Adv. Phys. **41**, 203 (1992).
- ⁵⁰I. Paul and G. Kotliar, Phys. Rev. B **64**, 184414 (2001).