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TRANSPORT PROPERTIES OF $CeRu_2Ge_2$ AT HIGH PRESSURE

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The electrical resistivity and magnetoresistivity of $CeRu_2Ge_2$ were measured up to P = 10 GPa and down to T = 30 mK. Two magnetic transitions at T_C = 7.40 K and T_N = 8.55 K were found. The first one decreases with pressure ($\partial T_C/\partial P = -2.1$ K/GPa), whereas T_N first increases ($\partial T_N/\partial P$ = 0.7 K/GPa), and starts to decrease above $P \approx$ 5 GPa. Another magnetic phase appears at $T_L < T_N$ beyond $P \approx$ 3.4 GPa. The magnetic to non-magnetic transition takes place at a critical pressure $P_c \approx 8.7$ GPa. © 1998 Elsevier Science Ltd. All rights reserved

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1. INTRODUCTION

In the ternary cerium compounds CeM_2X_2 with X=Ge or Si and M a transition metal from Ru to Ag, a long range magnetic order appears at low temperature [1–5]. The only exception in this series is the heavy fermion compound $CeRu_2Si_2$ [6, 7] which can be considered to have a paramagnetic ground state although very small static moments ($\mu \approx 10^{-3}\mu_B$) below 2 K were found by μ-SR experiments [8]. Substituting a small amount of Si by Ge (x = 0.05) induces antiferromagnetic order in $CeRu_2(Si_{1-x}Ge_x)_2$ [9]. Higher Ge content x in this solid solution shifts T_N upwards and for $x \geq 0.8$ a ferromagnetic ground state occurs [10]. For $CeRu_2Ge_2$ itself (T_C = 7.5 K [11]) a magnetic moment of $\mu = 1.9\mu_B$ and a slightly enhanced low-temperature electronic specific heat γ = 20 mJ/mol K^2 were found [12]. At higher temperature presumably an antiferromagnetically modulated phase exists up to T_N = 8.6 K [13]. Substitution of Si leads to an increase of the unit-cell volume. This can be interpreted as introducing chemical pressure which decreases quantum fluctuations and favours long range magnetic order. Hence, reducing the volume by applying hydrostatic pressure on $CeRu_2Ge_2$ should enhance Kondo spin or valence fluctuations resulting in a suppression of the cerium moments at a critical pressure P_c. First results which support this idea were reported by Uwatoko et al. [13] who have measured the electrical resistance $R(T)$ up to P = 1.05 GPa. In this communication we report results on $CeRu_2Ge_2$ obtained by electrical resistivity $\rho(T)$ for 30 mK< T < 300 K and magnetoresistivity (MR) measurements at T = 100 mK for quasi-hydrostatic pressures up to 10 GPa.

2. EXPERIMENTAL

The sample was prepared by melting in stoichiometric amounts Ce(4N), Ru(4N), and Ge(5N) in an arc furnace under argon atmosphere. The sample was melted several times to achieve good homogeneity and annealed at 800°C for 10 days. X-ray diffraction analysis showed that the compound crystallised in the body-centred tetragonal $ThCr_2Si_2$ structure (a = 4.268Å, c = 10.048Å) and no parasitic phases were detected. A resistivity measurement at ambient pressure with the current in the basal plane of the tetragonal unit cell revealed a resistivity ratio ρ/ρ_0 = 116 ($\rho_0 = 0.3\mu\Omega$cm) emphasising the high quality of the sample. The $\rho(T)$-curves at high pressure were measured using a Bridgman type of high pressure cell with non-magnetic tungsten carbide anvils [14]. The pressure was determined via the pressure dependence

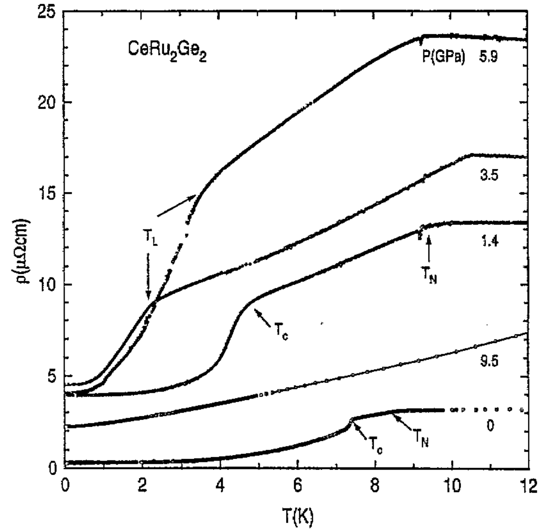

Fig. 1. Low temperature resistivity of $CeRu_2Ge_2$ at selected pressures ($j \perp c$). A ferro- (T_c), an antiferromagnetic (T_N) and a different modulated phase (T_L) are present.

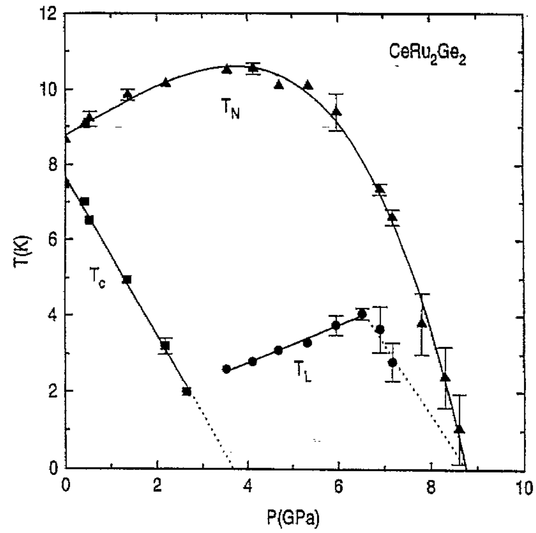

Fig. 2. Ordering temperatures T_C, T_N, and T_L of $CeRu_2Ge_2$ as a function of pressure. The error bars represent the limits for each transition temperature. At low pressure the symbol size is larger than the estimated error.

of the superconducting transition of Pb [15].

3. RESULTS

Figure 1 shows the temperature dependence of the electrical resistivity of $CeRu_2Ge_2$ below 12 K at various pressures. At ambient pressure two distinct transitions are present. The change of slope of $\rho(T)$ at $T_N = 8.55$ K is a clear sign for a transition at which the paramagnetic high-temperature phase transforms into a presumably antiferromagnetically ordered one. At $T_C = 7.40$ K a second transition into the ferromagnetic ground state takes place. The occurrence of a transition above T_C was already found by susceptibility and thermal expansion [11] as well as $\rho(T)$ measurements [13]. Specific heat experiments performed on the same sample showed a small anomaly ($C/T = 2\text{J/molK}^2$) at T_N and a huge jump ($C/T = 8\text{J/molK}^2$) at T_C. Applying a small pressure decreases T_C whereas T_N shifts upwards as can be seen from the $\rho(T)$-curve recorded at 1.4 GPa. At a first glance the shape of the $\rho(T)$-curve plotted for $P = 3.5$ GPa seems to be similar to those recorded at lower pressure. However, from the results described below, it is concluded that above $P = 3.4(3)$ GPa a different magnetic phase below a characteristic temperature T_L has developed. The reason of labelling the transition T_L will be given in the discussion. Further pressure increase forces T_L to shift towards higher temperature and T_N to decrease (see $\rho(T)$-curve at $P = 5.9$ GPa). The magnetic to non-magnetic phase transition has already taken place at $P = 9.5$ GPa since no anomalies are present in this $\rho(T)$-curve.

To deduce the ordering temperatures from the resistivity curves we used two different criteria. Firstly, we drew a straight line to the $\rho(T)$-curves above the region where $\rho(T)$ changes remarkably and took the first deviation of the data from this line as criterion for the occurrence of the phase transition. Secondly, we used the temperature derivative of $\rho(T)$ to estimate at which temperature a pronounced change of the slope takes place. The critical temperatures obtained from the ambient pressure $\rho(T)$-curve with these methods are in very good agreement with the temperatures, at which the anomalies in our specific heat measurements occurred. This gives additional confidence in the criteria used in determining the critical temperatures from the $\rho(T)$-curves at high pressure. At low pressure the two criteria gave almost identical values for a given ordering temperature. However, at higher pressures (i.e. above $P \approx 6$ GPa) the transitions are not longer pronounced and the two criteria gave rather different values for each ordering temperature.

In Fig. 2 the average of the two values for each ordering temperature is plotted as a function of pressure. A different pressure dependence is found for the critical temperatures T_N and T_C. The former one first increases, reaches a maximum of $T_N = 10.7$ K at $P = 4.1$ GPa and then decreases. The initial pressure dependence is 0.7K/GPa. The extrapolation $T_N \to 0$ to estimate where the magnetic to non-magnetic phase transition occurs yields a critical pressure $P_c \approx 8.7$ GPa. In

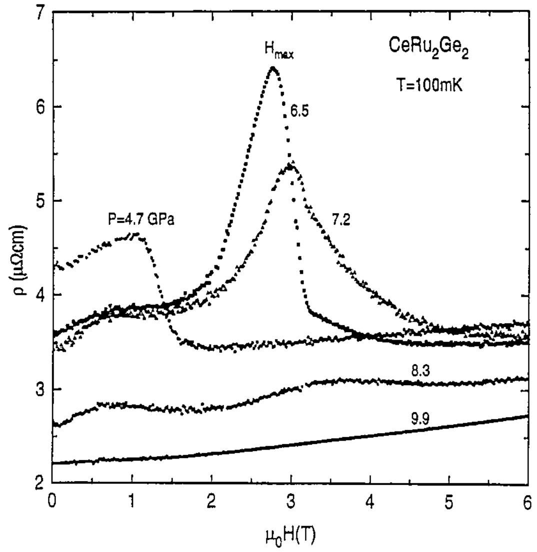

Fig. 3. High-field magnetoresistivity ($j \perp c$, $H \parallel c$) of $CeRu_2Ge_2$ at T = 100 mK and various pressures.

contrast to this, T_C decreases with a rate of 2.1K/GPa in the pressure range up to P = 2.7 GPa and extrapolates to zero for P = 3.7 GPa. For higher pressure $CeRu_2Ge_2$ seems to be in a different magnetically ordered ground state and the characteristic temperature T_L increases ($\partial T_L/\partial P$ = 0.5 K/GPa). Above P = 6.5 GPa, T_L decreases and no clear evidence for such a transition is found beyond P = 7.2 GPa. The initial pressure dependence of T_N and T_C given above are in qualitative agreement with those reported by Uwatoko [13].

In addition, transverse MR measurements ($H \parallel c$ and $j \perp c$) at T = 100 mK were performed. At low pressure (i.e. in the ferromagnetic phase) $\rho(H)$ increases slightly in a magnetic field up to B = 8 T. In Fig. 3, the MR-curves at high pressure are depicted. A feature starts to develop in the $\rho(H)$-curve at P = 4.7 GPa. It becomes a pronounced peak at H_{max} for P = 6.5 GPa and its relative increase between zero field and H_{max} is as high as 76%. Above this pressure the peak amplitude starts to decrease and the MR is without any feature at P = 9.9 GPa. The peak position H_{max} shifts up in field with pressure. A linear pressure dependence $H_{max}(P)$ was fitted in the range 4.7< P < 8.3 GPa. Its extrapolation down to zero yields P = 3.1 GPa which is in the vicinity of the value deduced from the $T_C(P)$-dependence (P = 3.7 GPa, Fig. 2). The fact that both $T_C(P)$ and $H_{max}(P)$ extrapolate to zero at almost the same pressure (P = 3.4(3) GPa) can be interpreted as a sign for a new magnetic ground state, characterised by T_L. Another point worth mentioning is the pressure induced decrease of ρ in zero field (-50% between 4.7 and 9.9 GPa). This reflects that even at 100 mK the resistivity is rather pressure dependent and far away from the pressure independent residual value.

The low temperature part of the $\rho(T)$-curves can be described according to a Fermi-liquid relation $\rho = \rho_0 + AT^2$ up to a temperature $T_A <\approx 0.8$ K. The coefficient A is small ($A \approx 0.1 \mu\Omega\text{cm/K}^2$) at low pressure and reaches a maximum $A \approx 1.3 \mu\Omega\text{cm/K}^2$ at $P \approx$ 7.8 GPa. On crossing the magnetic to non-magnetic phase boundary the coefficient A is expected to show a maximum at the critical pressure P_c. The small difference (less than 0.9 GPa) we find for both, P_c and the position of the peak in A is most likely due to the experimental uncertainty of the technique used. Furthermore, in this pressure region and in a slightly larger temperature interval the data could also well described using a power law $\rho = \rho_0 + AT^n$ with $n < 2$. An exponent smaller than two is pointing to possible non-Fermi liquid effects. To clarify these points additional studies are necessary.

4. DISCUSSION

The pressure dependence of the different ordering temperatures can be compared in a qualitative way with those extracted from specific heat measurements on the solid-solution of $CeRu_2(Si_{1-x}Ge_x)_2$ [10]. For this comparison one should keep in mind that pressure acts in the opposite way than the increase of the Ge-content x. For high x-values ($x \geq 0.8$) an antiferromagnetic and a ferromagnetic phase exist. Starting at x = 1.0 (T_C = 7.40 K) T_C decreases down to 5 K at x = 0.8. However, T_N increases up to $T_N \approx$ 10 K. This behaviour can be compared to the pressure dependence of the transition temperatures of $CeRu_2Ge_2$ shown in Fig. 2. Qualitatively the same variation of T_N and T_C is found if the pressure range up to $P \approx$ 4 GPa is considered. In the range $0.1 < x < 0.6$ a different modulated antiferromagnetic phase could exist which is concluded from neutron experiments on $CeRu_2(Si_{0.9}Ge_{0.1})_2$ [16]. Furthermore, these measurements also showed that at a temperature $T_L \approx$ 2 K stronger effects in the intensities occur than at T_N. The antiferromagnetic order ($T_N \approx$ 10 K at x = 0.6) decreases down to $T_N \approx$ 5 K (x = 0.1) whereas T_L (which is assumed to be the ordering temperature for x < 0.6) first slightly increases and finally decreases if the lower limit of the Ge-content (x = 0.1) is reached. A similar dependence of T_N and T_L is found for $CeRu_2Ge_2$ at pressures above $P \approx$ 5 GPa (Fig. 2). Finally, for Ge-content x < 0.05 the solid-solution $CeRu_2(Si_{1-x}Ge_x)_2$ showed no long range order like in $CeRu_2Si_2$ [9]. This situation is most likely the case for $CeRu_2Ge_2$ above a critical pressure $P_c \approx 8.7$ GPa.

The variation of the ordering temperature described so far is in qualitative agreement with the model proposed by Doniach [18]. In Kondo compounds a strong competition exists between the Kondo effect (T_K) and the RKKY-interaction (T_{RKKY}) which tends to support magnetic order. The derived phase diagram gives T_K and the magnetic ordering temperature T_N (in some cases also T_C) as a function of $| JN(E_F) |$, where J is the exchange constant and $N(E_F)$ the density of states of conduction electrons at the Fermi energy. The value $| JN(E_F) |$ can be tuned either by chemical pressure, like the variation of the Ge-content x in the solid-solution $CeRu_2(Si_{1-x}Ge_x)_2$, or external pressure. In many Ce compounds the ordering temperature initially increases with pressure, i.e. with increasing $| JN(E_F) |$, then passes through a maximum and finally tends to zero [19]. Regarding the phase diagram plotted in Fig. 2 it is clear that at low pressure a competition between ferro- and antiferromagnetic interactions exists but it appears that T_N is the appropriate ordering temperature to use in Doniach's model. At higher pressure and especially in the vicinity of P_c where T_N approaches T_L a strong competition between two (different) couplings raises the question of the nature of the magnetic interaction.

5. CONCLUSION

The electrical resistivity $\rho(T)$ of $CeRu_2Ge_2$ was measured in the temperature range 30 mK$< T <$ 300 K for quasi-hydrostatic pressures up to 10 GPa. The ferromagnetically ordered ground state (T_C = 7.40 K) is suppressed by applying $P = 3.4(3)$ GPa ($\partial T_C/\partial P = -2.1$ K/GPa) and above this pressure a differently ordered magnetic structure seems to exist, characterised by the ordering temperature T_L. Initially, the antiferromagnetic interaction is enhanced ($\partial T_N/\partial P = 0.7$ K/GPa) but above $P \approx 5$ GPa T_N starts to decrease. The magnetic to non-magnetic phase transition takes place at $P_c \approx 8.7$ GPa. The occurrence of a presumably differently ordered magnetic phase below T_L is supported by the appearance of a pronounced peak at H_{max} in the transverse magnetoresistivity measured at T = 100 mK.

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