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Specific Heat and Transport Properties of UPt_3

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Specific heat, resistivity, thermal conductivity and thermoelectric power measurements are reported for the normal and superconducting regimes of the heavy-fermion superconductor UPt_3. A comparison is made with uranium-based spin-fluctuation and cerium-based Kondo-lattice compounds.

Introduction

UPt_3 has been discovered to be the first compound in which strong spin-fluctuation phenomena and superconductivity coexist. The spin-fluctuation effects are most clearly proven by the $T^3 \ln T/T^*$ term in the specific heat [1–3] with T^* between 10 and 30 K. Superconductivity is observed below 0.5 K with large slope of the upper critical field vs. T curves. An analysis of those data results in values for the effective mass of the pairing electrons of about 180 times the free electron mass [3, 4]. In Meissner effect experiments [5] it was shown that the superconducting volume fraction at 350 mK is at least 30%. Flux-pinning effects presumably prevent the observation of the full Meissner effect. Nevertheless, superconductivity is considered to be a bulk property of UPt_3. The coexistence of spin-fluctuation phenomena and superconductivity has been taken as an indication for triplet superconductivity [2]. Subsequent papers on the anisotropy in the temperature dependence of the upper critical field near T_c, determined in resistivity measurements with the field parallel and perpendicular to the current direction [4, 6], as well as on the ultrasonic attenuation in the superconducting state [7], were considered to support this indication. One should note that for the other U-based heavy fermion superconductor UBe_{13} [8–10] evidence for triplet pairing has been deduced from specific heat, nuclear magnetic resonance and thermal conductivity data in the superconducting state.

The existing specific heat data on UPt_3 in the superconducting state reveal a broad anomaly just below the resistive transition temperature with values for the relative jump in the specific heat of 1/4 to 1/3 of the BCS result [2, 3]. In this contribution we shall report on recent specific heat measurements that have been performed with improved accuracy on the sample discussed in [1].

Resistivity measurements in the normal state [11], carried out on single-crystalline samples along different crystallographic directions between 1.5 K and 300 K, reveal pronounced spin-fluctuation effects that are partly suppressed under pressure. A T^2-dependence of the resistivity could not be observed above 1.5 K. In the whole temperature range the resistivity of UPt_3 is anisotropic with the resistivity along the c-axis being almost 50% lower than perpendicular to this axis. This anisotropy will not be addressed in the present study, since all experiments have been performed on polycrystalline samples. For the preparation method of these samples we refer to [10]. A survey of spin-fluctuations and superconductivity in UPt_3 has recently been presented in [12].

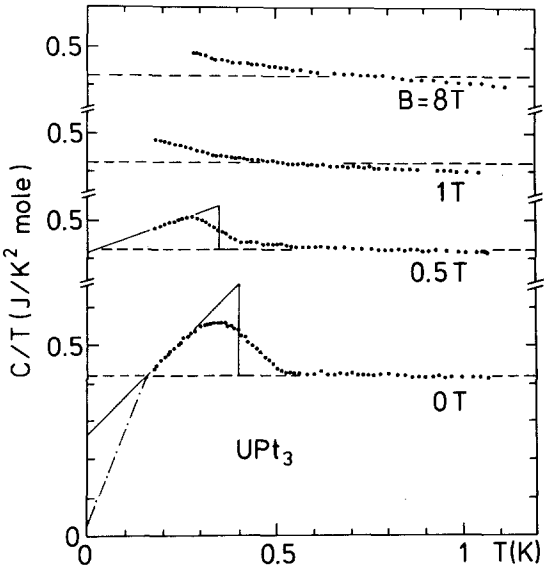

Fig. 1. The specific heat of polycrystalline UPt_3 at temperatures below 1.1 K in fields up to 8 T; the dashed curve represents the equation $C/T=\gamma^*+\beta^* T^2+\delta T^2 \ln(T/1\,\mathrm{K})$, with $\gamma^*=422\,\mathrm{mJ/K^2 mol}$, $\beta^*=-4.18\,\mathrm{mJ/K^4 mol}$ and $\delta=1.54\,\mathrm{mJ/K^4 mol}$ [15]. Schematic extrapolation of zero-field data by dash-dotted line satisfies entropy balance

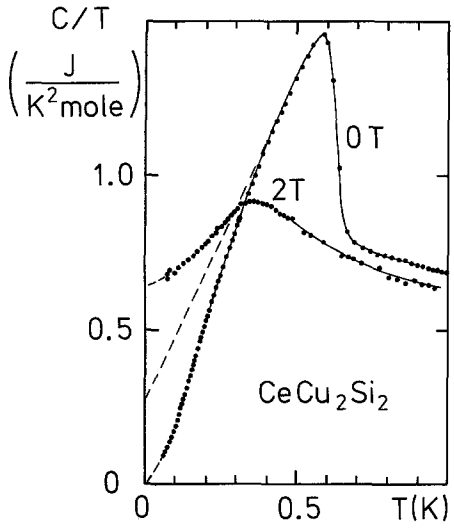

Fig. 2. C/T vs. T for a polycrystalline $CeCu_2Si_2$ sample with nominal Cu excess of 10 at %. Solid lines are guides to the eye, dashed lines are extrapolations to $T=0$ from low-T data and from data taken at $T \lesssim T_c$, respectively

Specific Heat

Specific heat experiments have been performed between 0.17 and 1.1 K. The minimum sample temperature in the used set-up has been limited by self-heating due to U-decay. The specific heat of the annealed polycrystalline UPt_3 sample is shown in Fig. 1 as C/T vs. T in different fields up to 8 T. In this experiment, the broad anomaly that was observed before [2, 3] is reproduced at zero field. The height of the peak as measured is about 30% of the specific heat extrapolated from above T_c. The relative jump height is increased to about 60% if the broad transition is replaced in the usual way by a sharp one. This procedure yields a calorimetric transition temperature $T_c \simeq 0.40$ K, in contrast to the resistive T_c of 0.52 K which marks the *onset* of superconductivity. Well below T_c, C/T is almost linear in T extrapolating to a finite $\gamma(T\to 0)$ value of about $260\,\mathrm{mJ/K^2 mole}$. By such an extrapolation, however, the entropy balance is not met, if the normal state specific heat is extrapolated to $T=0$ by using the dashed line in Fig. 1 (see below). This suggests that the low-T specific heat in the superconducting state should decrease faster than given by the straight line as is indicated in a schematic way by the dash-dotted line in Fig. 1. It is interesting to note that in $CeCu_2Si_2$, there is also [13] a nearly T^2-dependence of $C_s(T)$ just below T_c and a continuous change towards a nearly T^3-behaviour at very low temperature, see Fig. 2.

Further comparison with the latter compound suggests that UPt_3 is in between those $CeCu_2Si_2$ samples with pronounced specific heat jumps and other ones ("gapless superconductors") without significant jumps, but with giant linear terms $C_s=\gamma T$ as T goes to zero [14]. *If* there exists such a linear term in the low-temperature specific heat of superconducting UPt_3, too, it will probably not exceed a few percent of that in the normal state, according to the analysis shown in Fig. 1. In a field of 0.5 T, the transition is shifted downwards, and the jump is reduced by some 40% of its zero-field value. Again, very similar behavior was reported for $CeCu_2Si_2$ [14]. The weak temperature dependence of C/T in the normal state follows the description of the specific heat on the same sample (cylinder: length $\cong$ 8 mm, diameter $\cong$ 6 mm) that was deduced [15] for temperatures between 1.3 K and 16.5 K. Although there is clearly a field effect on the specific heat below 1 T, we cannot observe much difference between the 1 T and higher field results. Therefore, the results at 2, 4 and 8 T (not all shown in Fig. 1) are almost identical within the experimental uncertainty of $\pm 2\%$. In order to explain the field effects below 1 T one should take into account the impurities contained in the samples as observed in low-field DC magnetization studies [15]. Apart from these spurious effects, the overall temperature dependence of the specific heat below $T=30$ K shows that there are, except for the $T^3 \ln T/T^*$-term, no other pronounced features in

the electronic structure of UPt_3 near the Fermi energy.

Transport Properties

In order to look in some more detail into the electronic structure we have performed transport measurements down to 80 mK. Resistivity curves for UPt_3 are shown in Figs. 3 and 4 in the temperature regions below 1,000 K and 6 K, respectively. The temperature for the onset of superconductivity is 0.48 K, the width of the transition is about 0.1 K, in good agreement with other results for polycrystalline material [3]. The detailed behaviour of the unannealed polycrystalline sample below 1 K is shown in the inset of Fig. 4. The residual resistivity ρ_0 is 3.9 μΩcm; after annealing $\rho_0 = 2.9$ μΩcm, which is close to the values of 1.7 and 3.0 μΩcm reported for single crystals parallel to the c-axis and b-axis, respectively [11]. A quadratic temperature dependence $\rho(T) = \rho_0 + A \cdot T^2$ seems to hold up to 2 K, with a coefficient $A = 1.38$ μΩcm/K^2 before annealing and 1.7 μΩcm/K^2 thereafter, whereas for the single-crystalline samples studied before [11] a T^2-dependence was only suggested for temperatures below 1.5 K.

Thermoelectric power, $S(T)$, measurements have been performed between 0.2 K and 350 K, see Fig. 5a and b. There is a monotonous decrease of $S(T)$ below $T = 4$ K in the normal state, deviating, however, from the linear behaviour as expected in the absence of "fine details" in the electronic bandstructure. The main feature of $S(T)$ is a pronounced positive peak at $T \cong 8$ K. Its position nearly coincides with the maximum in the temperature derivative of the resistivity and can be taken as an estimate for the spin-fluctuation temperature [11]. $S(T)$ changes sign at $T = 24$ K and reaches a nearly constant value of -11 μV/K above $T \approx 100$ K where also the resistivity starts to saturate.

In Fig. 6a, the thermal conductivity, κ, is given as a function of temperature up to 200 K and compared with the electronic part as calculated from the measured electrical resistivity, assuming the validity of the Wiedemann-Franz law. In Fig. 6b, we show the Lorenz ratio, $L = \kappa\rho/T$, obtained from the measured $\kappa(T)$ and $\rho(T)$ data. Depending on the temperature regime, the Lorenz ratio in pure simple metals is either smaller than or comparable to $L_0 = 2.45 \times 10^{-8}$ V^2/K^2, the Sommerfeld value. In systems with small electronic mean free path, for example in the presence of strong electron-paramagnon scattering, phonons can dominate the heat transport, giving rise to a substantial increase of L above the Sommerfeld value. Since the phonon conductivity

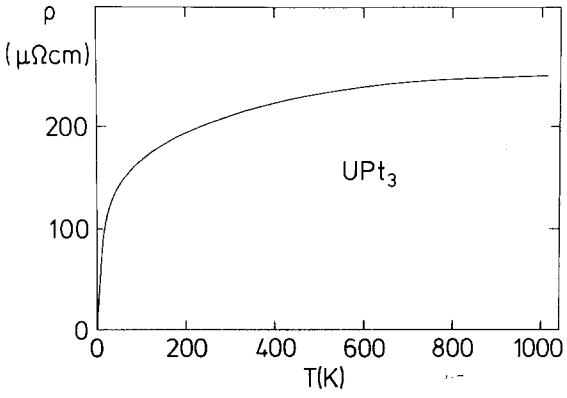

Fig. 3. Resistivity versus temperature curve up to 1,000 K for a polycrystalline UPt_3 sample

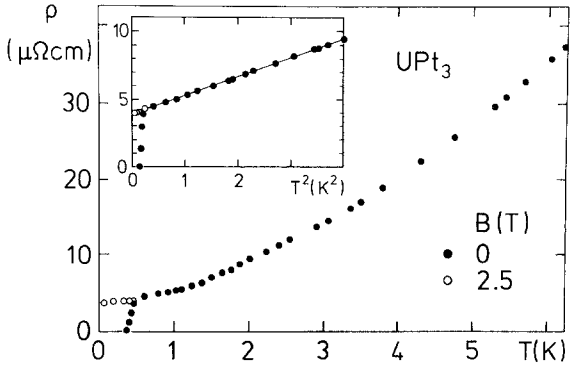

Fig. 4. Temperature dependence of the resistivity of a polycrystalline UPt_3 sample below 6 K. Inset shows data below 2 K versus a T^2 axis

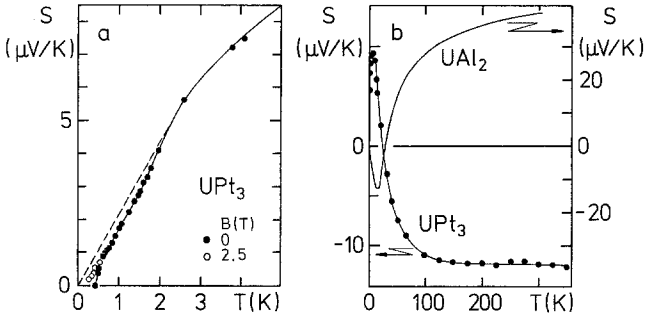

Fig. 5a and b. Thermoelectric power, S, of polycrystalline UPt_3 for $T < 5$ K (**a**) and for temperatures up to 350 K (**b**). In (**b**) the results for UAl_2, taken from [23], are also shown

usually has a maximum at $(0.1\text{–}0.2)\,\theta_D$, with θ_D the Debye temperature, this leads to a corresponding peak in $L(T)$. In the case of UPt_3 we find a maximum value $L_{max} = 5.5\,L_0$ at a temperature $T_{max} = 24\,K$, which is slightly higher than $0.1\,\theta_D$ ($\theta_D = 217$ K has been deduced from sound velocity measurements [16]). Because the total heat conductivity is the sum of an electronic part, κ_e, and a

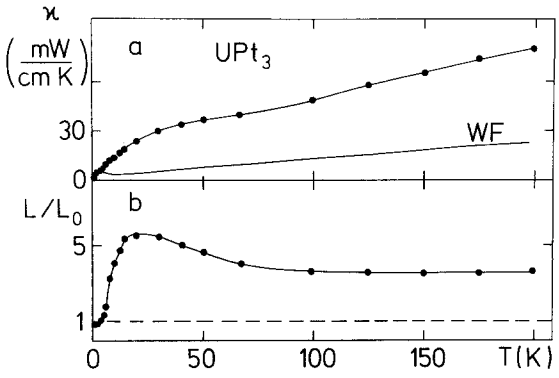

Fig. 6. a Thermal conductivity of polycrystalline UPt_3 for temperatures up to 200 K as measured and as calculated from the electrical resistivity by applying the Wiedemann-Franz law; **b** The experimentally determined value for the Lorenz ratio $L=\kappa(T)\rho(T)/T$, divided by the Sommerfeld value for this ratio, $L_0=2.45\times10^{-8}(V/K)^2$

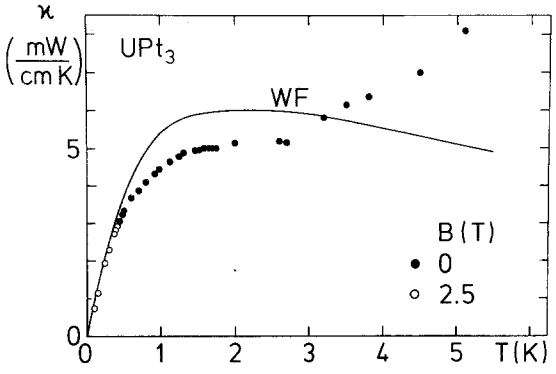

Fig. 7. Thermal conductivity of polycrystalline UPt_3 below 6 K; the full curve represents the Wiedemann-Franz law

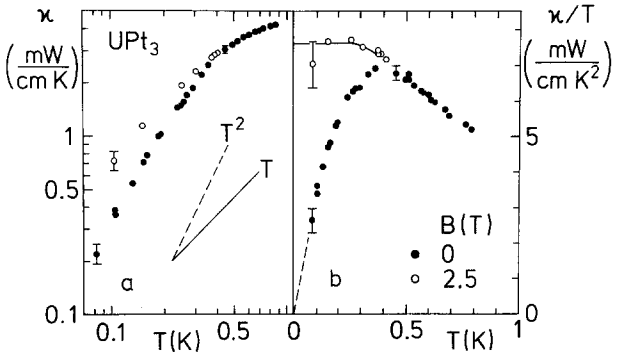

Fig. 8a and b. Temperature dependence of the thermal conductivity of polycrystalline UPt_3 in a double-logarithmic plot (**a**), and as κ/T vs. T (**b**)

phonon part, κ_p, the measured quantity $L(T)$ can be expressed by: $L=L_e(1+\kappa_p/\kappa_e)$, where $L_e(=\kappa_e\rho/T)$ is the electronic Lorenz ratio. Since at higher temperatures L_e should approach L_0, this analysis shows that $\kappa_p \gtrsim 4\kappa_e$ at T_{max}, which is not unreasonable in view of the already rather short electronic mean free path as inferred from the electrical resistivity of 120 μΩcm at this temperature (see Fig. 3). A much larger peak in $L(T)$ at a similar temperature has been found earlier for $CeCu_2Si_2$, where $\rho(T_{max})$ exceeds the value observed for UPt_3 by almost a factor of two [17].

The phonon mean free path l_p at T_{max}, as calculated from the difference between the measured $\kappa(T)$ and the calculated electronic part is approximately 60 Å, as derived by inserting the sound velocity $v_s=4\times10^5$ cm/s [7, 16] and a lattice specific heat C_p of approximately 0.27 Ws/Kcm^3 in the expression $\kappa_p=l_p v_s C_p/3$. This value for l_p can be compared to a value of the order of 10 Å for the electronic mean free path l_e at this temperature. The low-temperature normal-state heat conduction data, which are displayed in Fig. 7 in more detail, show two salient features: *(i)* at the lowest temperatures the Wiedemann-Franz law holds fairly well as is expected for elastic residual scattering, though the measured thermal conductivity is somewhat smaller than expected from the Wiedemann-Franz law, implying that the Lorenz number L is smaller than L_0 by about 8%; *(ii)* above 2.5 K, an intersection is found between the measured curve and the curve calculated from the resistivity which we attribute to the existence of an additional and large phonon contribution to the high-T conductivity.

Very similar thermal conductivity results were found for $CeAl_3$ and normal-state $CeCu_2Si_2$ [18, 19]. The low-temperature normal-state results for UPt_3, obtained in a field of 2.5 T are shown in Figs. 8a and b, together with data in the superconducting state at zero field. At temperatures below the superconducting transition, there is a difference between the normal-state (κ_n) and superconducting-state (κ_s) values. This difference increases towards lower temperatures where κ_s follows a power law with an exponent between 1 and 2. The temperature dependence of κ_s shows negative curvature for $T\geqq0.2$ K when plotted as κ_s/T vs. T (Fig. 8b). As is indicated by the dashed line, the data points at the very temperature end are consistent with $\kappa_s=\alpha T^2$, $\alpha=32$ mW/K^{3}cm.

A T^2 term in $\kappa_s(T)$ has also been found for the two other heavy-fermion superconductors UBe_{13} ($\alpha=0.37$ mW/K^3 cm, Ref. 10) and $CeCu_2Si_2$ ($\alpha=1.8$ mW/K^3 cm, Refs. 13 and 19). In the latter system an additional term linear in T exists. While it is not yet clear how these two contributions have to be interpreted for $CeCu_2Si_2$, the quadratic temperature dependence of κ_s in UBe_{13} was ascribed to the thermal excitations of a superconductor with strong gap anisotropy [10]. The same might hold for UPt_3: We

estimate that well below T_c, $\kappa_s(T)$ must be of dominant *electronic* origin, despite the importance of the phonon conductivity as inferred in the normal state for $T > 3\,K$ [20].

Concluding Remarks

Apart from the superconducting properties below 0.5 K and apart from distinct anisotropy effects in single-crystalline samples [11], the specific heat, resistivity, and thermoelectric power data of polycrystalline UPt_3 are remarkably similar to those of the "spin-fluctuation" compound UAl_2 [21–23], see e.g. Fig. 5b. The characteristic temperature of UAl_2 is about two times that of UPt_3. Compared to the Ce-based Kondo-lattice systems mentioned before, the U-based spin-fluctuation compounds show different behaviour in, at least, three respects, namely in (i) the $T^3 \ln T/T^*$-contribution to the low-temperature specific heat [15], (ii) in the absence of a low-temperature peak in $\rho(T)$, and (iii) in the absence of a high-temperature extremum in the thermoelectric power. Furthermore, the low-temperature extremum in the thermoelectric power of the uranium compounds corresponds to a maximum in $\partial\rho/\partial T$ rather than in $\rho(T)$ as is observed in the case of the cerium compounds. A very large quadratic term exists in the thermal conductivity $\kappa_s(T)$ well below T_c. It is dominantly of electronic nature and appears to be consistent with the contribution of the thermal excitations in a superconductor with strongly anisotropic pairing.

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References

1. Frings, P.H., Franse, J.J.M., Boer, F.R. de, Menovsky, A.: J. Magn. Magn. Mater. **31–34**, 24 (1983)
2. Stewart, G.R., Fisk, Z., Willis, J.O., Smith, J.L.: Phys. Rev. Lett. **52**, 679 (1984)
3. Visser, A. de, Franse, J.J.M., Menovsky, A., Palstra, T.T.M.: J. Phys. F **14**, L191 (1984); Physica B (in press)
4. Chen, J.W., Lambert, S.E., Maple, M.B., Fisk, Z., Smith, J.L., Stewart, G.R., Willis, J.O.: Phys. Rev. B **30**, 1583 (1984)
5. Palstra, T.T.M., Kes, P.H., Mydosh, J.A., Visser, A. de, Franse, J.J.M., Menovsky, A.: Phys. Rev. B **30**, 2986 (1984)
6. Willis, J.O., Fisk, Z., Smith, J.L., Chen, J.W., Lambert, S.E., Maple, M.B.: Proc. LT17. Eckern, U., Schmid, A., Weber, W., Wühl, H. (eds.), pp. 245–246. Amsterdam: North-Holland 1984
7. Bishop, D.J., Varma, C.M., Batlogg, B., Bucher, E., Fisk, Z., Smith, J.L.: Phys. Rev. Lett. **53**, 1009 (1984)
8. Ott, H.R., Rudigier, H., Rice, T.M., Ueda, K., Fisk, Z., Smith, J.L.: Phys. Rev. Lett. **52**, 1915 (1984)
9. MacLaughlin, D.E., Cheng Tien, Clark, W.G., Lan, M.D., Fisk, Z., Smith, J.L., Ott, H.R.: Phys. Rev. Lett. **53**, 1833 (1984)
10. Jaccard, D., Flouquet, J., Fisk, Z., Smith, J.L., Ott, H.R.: (to be published)
11. Visser, A. de, Franse, J.J.M., Menovsky, A.: J. Magn. Magn. Mater. **43**, 43 (1984)
12. Franse, J.J.M., Frings, P.H., Visser, A. de, Menovsky, A., Palstra, T.T.M., Kes, P.H., Mydosh, J.A.: Physica **126** B, 116 (1984)
13. Steglich, F., Bredl, C.D., Lieke, W., Rauchschwalbe, U., Sparn, G.: Physica **126** B, 82 (1984)
14. Bredl, C.D., Spille, H., Rauchschwalbe, U., Lieke, W., Steglich, F., Cordier, G., Assmus, W., Herrmann, M., Aarts, J.: J. Magn. Magn. Mater. **31–34**, 373 (1983)
15. Frings, P.H., Franse, J.J.M.: (to be published); see also Ref. 3.
16. Visser, A. de, Franse, J.J.M., Menovsky, A.: J. Phys. F. (in press)
17. Franz, W., Griessel, A., Steglich, F., Wohlleben, D.: Z. Phys. B – Condensed Matter and Quanta **31**, 7 (1978)
18. Ott, H.R., Marti, O., Hulliger, F.: Solid State Commun. **49**, 1129 (1984)
19. Sparn, G., Lieke, W., Gottwick, U., Steglich, F., Grewe, N.: Proc. Int. Conf. on Valence Fluctuations, Cologne 1984, Müller-Hartmann, E., Wohlleben, D. (eds.). J. Magn. Magn. Mater. (in press)
20. An upper limit of the phonon part to $\kappa_s(T)$ can be obtained by taking into account scattering of the phonons from grain boundaries only ("size effect"). Using the average grain size of 50 μm as determined for our polycrystalline sample, we estimate that at the lowest temperature the phonons contribute only a few % to the heat transport in the superconducting state
21. Trainor, R.J., Brodsky, M.B., Culbert, H.V.: Phys. Rev. Lett. **34**, 1019 (1975)
22. Buschow, K.H.J., Daal, H.J. van: AIP Conf. Proc. **5**, 1464 (1971)
23. Armbrüster, H., Franz, W., Schlabitz, W., Steglich, F.: J. Phys. (Paris) **40**, C4-150 (1979)

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