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Single crystals of several heavy-fermion systems grown by floating zone method

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A vertical floating zone method was successfully use to grow single crystals of heavy fermion systems UPt_3 and UNi_2Al_3. Metallurgical aspects of the crystal preparation are discussed. Superconducting and magnetic properties of the crystals were studied by means of electrical resistivity and heat capacity measurements down to 350 mK. In the best case, the resistivity of UPt_3 exhibits a transition to superconductivity between 530 mK and 560 mK and a sharp double transition in heat-capacity data. The heat capacity data of UNi_2Al_3 show sharp peaks corresponding to superconducting or to magnetic transition below 1.1 K and 5 K.

1. INTRODUCTION

The interplay between magnetism and superconductivity is still a fascinating subject of heavy–fermion physics. The heavy–fermion superconductors show many indication of unconventional superconductivity. Among them UPt_3 is the most promising candidate for unconventional superconductivity. UNi_2Al_3 is a new heavy fermion system which displays coexistence of superconductivity and antiferromagnetic order. Metallurgical problems have inhibited up till present the growth of superconducting bulk single crystal. In general, superconducting properties of UPt_3 [1] and UNi_2Al_3 [2] are very sensitivity to nominal composition and heat treatment of as–grown crystal. Our crystals were grown by the vertical floating zone method in a mirror furnace. It is known for this method that the melt composition tends to self–adjust at the beginning of a growth run, until steady state is reached, allowing the growth of incongruently melting materials.

2. RESULTS

2.1. UPt_3

UPt_3 forms congruently from the melt. It crystallizes in hexagonal close-packed structure ($MgCd_3$–type) and belongs to the space group $P6_3/mmc$. With decreasing temperature, the heavy–fermion, antiferromagnetic ($T_N = 5.5$ K [3]) and superconducting state ($T_c = 0.5$ K [4]) appear successively and coexist. Two superconducting transition were found on crystals of good quality [1]. Furthermore, a complex superconducting phase diagram with three superconducting phases exists [5].

All our UPt_3 crystals were grown under a continuously Ti–gettered argon atmosphere (underpressure 200 mbar) with the same crystal growth conditions: feed and seed translation close to 7 mm/hour, feed and seed rotation 35 rpm. The average length of crystals was 40 mm with diameter of 5 mm. Laue patterns taken on the surface of all crystals were of good quality, with sharp spots and without any indication of a mosaic–like–spread structure. The crystal #1 was grown from UO_2 contaminated polycrystalline bars. The surface of crystal was covered with thin layer of UO_2 (EMPA results). Our heat capacity measurements and resistivity measurements (Fig.1.) revealed that the crystal is not superconductive at least down to 350 mK. The heat treatment (950 °C, 4 days) resulted only in reduction of residual resistivity at about 1/3 of the residual resistivity on as–grown crystal.

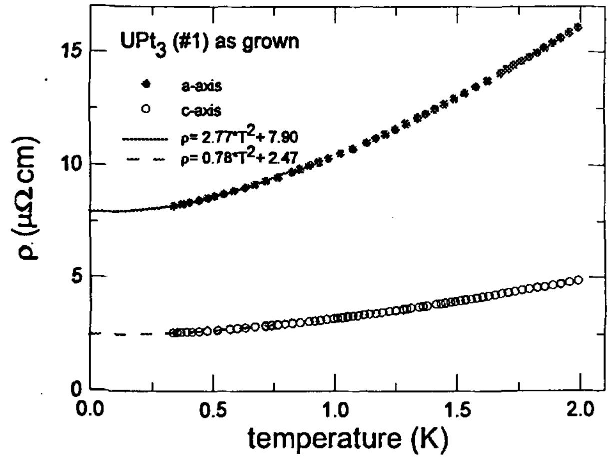

Figure 1: Temperature dependence of resistivity.

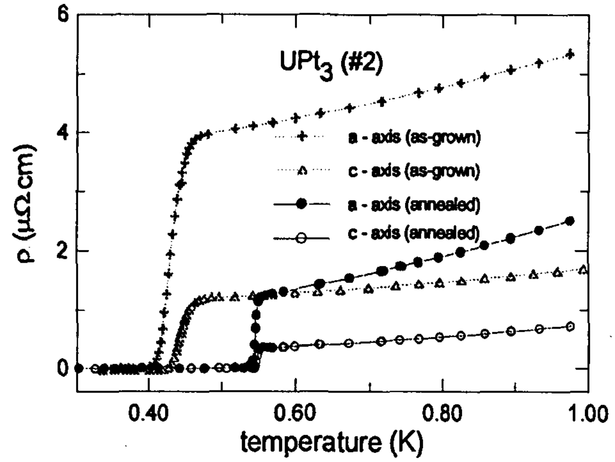

Figure 2: Temperature dependence of resistivity.

The crystal #2 was grown from pure starting materials. The surface of crystal was metallic bright. No second phase was detected by EMPA. Our heat capacity measurements and resistivity measurements (Fig.2.) revealed that the crystal is superconductive. The heat treatment (950 ° C, 4 days) improved the superconductive properties of the crystal #2 (Fig.2.). The resistivity exhibits a transition to superconductivity between 530 mK and 560 mK (Fig.2.). Our heat-capacity measurements showed a sharp double transition with two superconducting transitions at 0.490 mK and 0.535 mK.

2.2. UNi_2Al_3

UNi_2Al_3 is expected to form incongruently from the melt. It decomposes peritectically at about 1200 °C into UAl_2 and melt. This renders it rather difficult to obtain single crystal with good superconductive properties. Only very recently, Noriaki Sato [5] obtained a superconducting single crystal by Czochralski method. UNi_2Al_3 crystallizes in hexagonal $PrNi_2Al_3$ structure belonging to the space group P6/mmm. It undergoes antiferromagnetic transition at 5 K and superconducting transition at 1 K [2, 5]. Neutron scattering measurements show UNi_2Al_3 to be unique among heavy fermion superconductors displaying long range magnetic ordering which is incommensurate with its crystal lattice [6].

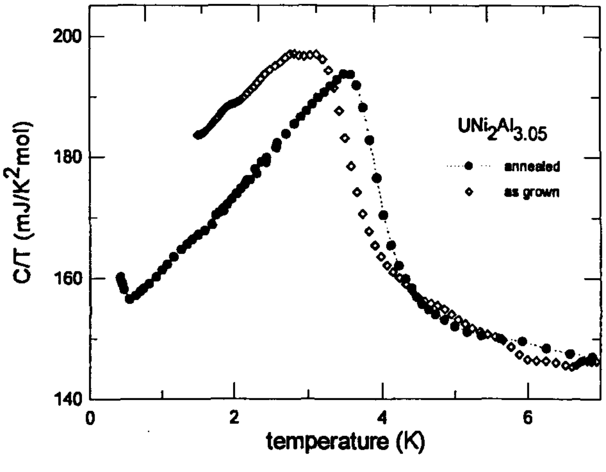

Figure 3: Temperature dependence of specific heat.

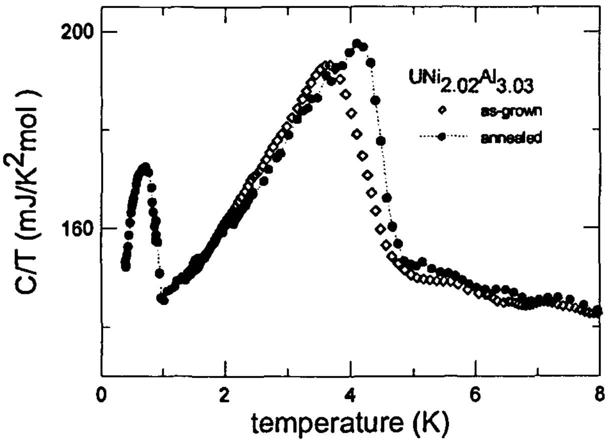

Figure 4: Temperature dependence of specific heat.

Our single crystals $UNi_2Al_{3.05}$ and $UNi_{2.02}Al_{3.03}$ were grown with the same condition as UPt_3 crystals; only in the case of the second one the feed and seed rotation were 25 rpm. The length of crystals was above 40 mm and diameter was 5 mm. Both crystals had a tendency of cracking and the surface of crystals was partially covered with NiAl layer. The Laue pictures showed a good quality of the crystals. The specific heat data (Fig.3 and Fig.4) show that $UNi_{2.02}Al_{3.03}$ undergoes antiferromagnetic transition at higher temperature as $UNi_2Al_{3.05}$ and the anomaly in heat capacity data is sharper. The annealing (950 °C, 4 days) improved magnetic properties of both crystals and $UNi_{2.02}Al_{3.03}$ became superconductive.

3. CONCLUSIONS

A vertical floating zone method was successfully use to grow single crystals of heavy-fermion systems UPt_3 and UNi_2Al_3. An appropriate nominal composition and heat treatment of as-grown crystals have essential importance for the quality of the crystals.

REFERENCES

[1] K. Bakker "Pressure effect in uranium-based heavy fermion compounds, PhD thesis, Amsterdam 1993.
[2] C. Geibel et al. Z. Phys. B 83 (1991) 305.
[3] G. Aeppli et al. Phys. Rev. Lett. 60 (1988) 615.
[4] A. de Visser et al. Physica B 186-188 (1993) 212.
[5] N. Sato et al. submitted to J.Phys.Soc. of Japan.
[6] A. Schröder et al. Phys. Rev. Lett. 72 (1994) 136