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STRUCTURE, TRANSPORT AND THERMAL PROPERTIES OF UCoGa

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Abstract

By means of neutron powder diffraction, we find that UCoGa crystallizes in the hexagonal ZrNiAl structure and orders ferromagnetically at low temperatures with magnetic moments stacked along the c axis. The magnetic-ordering temperature is reflected in anomalies in the temperature dependencies of the electrical resistivity and the specific heat at $T_C = 47$ K. Furthermore, the strong anisotropy in the electrical resistivity for $i \parallel c$ and $i \perp c$ indicates a significant contribution of the magnetic anisotropy to the electrical resistivity.

I. INTRODUCTION

UCoGa belongs to the large group of UTX compounds (T = transition metal, X = p-electron metal) and is reported to crystallize in the hexagonal ZrNiAl structure¹. Previous bulk magnetic investigations²⁻⁴ revealed magnetic order at about 47 K, but the results did not allow to conclude whether the ground state is ferro- or antiferromagnetic⁴. The purpose of the present contribution is to determine the structural parameters and to clarify the type of ground state as well as to investigate the bulk transport and thermal properties of single-crystalline UCoGa.

II. EXPERIMENTAL

For the present investigations we have used two kinds of samples: polycrystalline material and a small single crystal. The polycrystal, which has been used in the neutron-diffraction experiments, was prepared by arc-melting appropriate amounts of the constituting elements with a purity of at least 99.99%. The small single crystal is the same as was used in a previous investigation⁴. Neither the polycrystal nor the single crystal have been annealed.

For neutron-diffraction experiments, the polycrystal was ground and

enclosed with helium gas in a sealed vanadium tube, which was mounted on the cold finger of a closed-cycle refrigerator. This set up was installed in the High Intensity Powder Diffractometer HIPD at Los Alamos spallation pulsed neutron source LANSCE. Data have been taken on 6 detector banks ($2\theta = \pm 40, \pm 90, \pm 153$) at 60, 35 and 10 K. The diffraction patterns were analyzed using the Rietveld refinement program GSAS⁵. The magnetic intensities were also analyzed by extracting integrated intensities for individual peaks and fitting to models for the magnetic structure.

The temperature dependence of the electrical resistivity was measured between 4.2 and 300 K with the standard four-point ac method on two small bar-shaped single crystals of typical size $0.5 \times 0.5 \times 2 \text{ mm}^3$, where the largest distance coincides with the c axis for the first sample and is perpendicular to the c axis for the second one. The large error in the determination of the geometrical factor makes a reliable estimate of the absolute resistivity values impossible.

At 4.2 K, the field dependence of the electrical resistivity in magnetic fields up to 35 T has been measured in the Amsterdam High-Field Installation with $i \parallel B \parallel c$ -axis.

The temperature dependence of the specific heat has been measured between 4.2 and 100 K making use of both standard adiabatic and the relaxation-time method.

III. CRYSTAL AND MAGNETIC STRUCTURE

UCoGa crystallizes in the hexagonal ZrNiAl structure (space group: P62m). The structural parameters of UCoGa have been derived by neutron powder diffraction at 60 K and the results of the refinement are listed in Table 1. The absence of any unindexed reflection in the parent phase suggests that there is no impurity in the present sample. However, the reduced χ^2 drops by 5% when a slightly lower atomic fraction of Co and slightly higher fraction of Ga (of the order of 1% in both cases) are assumed. This result indicates a small deviation from the exact 1:1:1 stoichiometry. For UCoGa, the nearest interuranium distance d_{U-U} is found within the hexagonal basal plane (see Fig.1.) and is determined by $d_{U-U} = a\sqrt{3x^2-3x+1}$, where a is the lattice parameter and x is the U position parameter.

In order to clarify the magnetic structure, we have performed neutron-diffraction experiments at 35 and 10 K, well below the magnetic ordering temperature indicated by bulk magnetization results²⁻⁴. We do not observe any additional purely magnetic reflections at these temperatures, but an additional magnetic contribution to the nuclear reflections is found. This indicates that the magnetic unit cell is the same as the nuclear one but, by no means that antiferromagnetism is not possible. Possible magnetic structures have been

derived from magnetic space-group analysis. U atoms in the unit cell lie in the mirror planes at $x=0$, $y=0$ and $x=y$ (note, that x and y are making an angle of 120°). The moment corresponding to U in a certain mirror plane must be perpendicular or parallel to that mirror plane. The maximal non-isomorphic Shubnikov magnetic subgroups of P62m are shown in Fig. 1. Clearly, there are two antiferromagnetic non-collinear structures with moments in the basal plane and only one ferromagnetic (collinear) model with U moments along the c direction. The fact, that we do not observe any $00l$ magnetic contribution does not mean that the magnetic structure must be ferromagnetic since the two non-collinear models also have no $00l$ magnetic contributions. This is easily understood since the net moments in the plane perpendicular to $00l$ is zero for the non-collinear structures.

The integrated intensities have been corrected for the Lorentz factor⁶ and for absorption. We find no magnetic contribution to the 110 reflection, which excludes structure 2(b) as this gives noticeable intensity to this reflection. This is easily understood since the net moments in the $hh0$ plane are perpendicular to that plane which, in turn, gives zero magnetic contribution through the expression of $\sin\eta$ where η is the angle between the magnetic moment and the reciprocal lattice vector (in this case $\eta=0$). On the other hand, both structures, 2(a) and 2(c) are possible and the lower reduced χ^2 of the ± 90 detector banks suggests structure 2(c), the ferromagnetic one, to be more likely. However, the difference is only marginal and, in fact, if the results obtained on the ± 40 and ± 153 detector banks are included into the refinement, structure 2(a) is slightly more likely. However, due to inconsistencies in the magnetic appearance in the + and - detector banks, which are less pronounced in the ± 90 banks, we believe structure 2(c) to be the correct one, which is corroborated by the magnetization results. For the ferromagnetic structure 2(c) we deduce U magnetic moments of $0.74 \pm 0.03 \mu_B$ per atom, which is in good agreement with the value of $0.78 \mu_B/\text{f.u.}$ obtained from the high-field magnetization⁴. Note that model 2(a) yields much larger U magnetic moments of about $1.04 \pm 0.04 \mu_B$ per atom.

IV. TRANSPORT AND THERMAL PROPERTIES

The onset of magnetic ordering at $T_C = 47$ K is reflected by a maximum in the temperature derivative of the electrical resistivity at this temperature (see Fig. 2) for both $i \parallel c$ -axis and $i \perp c$ -axis. However, while for $i \parallel c$ -axis an appreciable reduction of the electrical resistivity with decreasing temperature is found, we observe an almost flat resistivity behaviour for $i \perp c$ -axis. The observed strong anisotropy in the resistivity correlates well with the magnetic anisotropy found in bulk magnetization measurements. At 300 K, very rough estimates of resistivities yield values around $150 \mu\Omega\text{cm}$ for $i \parallel c$ -axis, while

twice as large values for $i \perp c$ -axis are found. Below 40 K, the electrical resistivity follow a quadratic temperature dependence for both orientations, which is shown for $i \parallel c$ -axis in the inset of Fig. 2. For UCoGa, we may roughly estimate the pre-factors A to be about 0.216 and $0.138 \mu\Omega\text{cm}/\text{K}^2$ for $i \parallel c$ -axis and $i \perp c$ -axis, respectively.

For this compound, we find at 4.2 K (Fig.3.) for $i \parallel B \parallel c$ -axis an increase of the electrical resistivity with increasing magnetic field, which is in contrast to the large reduction of the electrical resistivity in isostructural antiferromagnetic UTX compounds upon application of sufficiently high magnetic fields⁷. The different behaviour of UCoGa may be taken as a further support for a ferromagnetic ground state of this compound. At the highest fields applied, we observe a slight saturation tendency. In general, UCoGa reflects a more 'normal' and expected magnetoresistance behaviour, however, the total increase of the resistivity (about 27% in 35 T) is surprisingly large.

As can be seen in Fig.4., the magnetic ordering of UCoGa is reflected in the specific heat by a maximum at $T_C = 47$ K. After subtraction of a phonon contribution determined by a Debye-function with $\theta_D = 195$ K, the magnetic entropy connected with ordering is considerably lower than $R\ln 9$ or $R\ln 10$, expected for the localized f^2 or f^3 configuration, respectively.

By linear extrapolation of C_p/T vs T^2 to $T = 0$ K, we derived the coefficient γ of the electronic contribution to the specific heat, to be about $48 \text{ mJ}/\text{molK}^2$.

V. CONCLUSIONS

UCoGa, which crystallizes in the hexagonal ZrNiAl structure, orders ferromagnetically below $T_C = 47$ K with ordered $5f$ moments of about $0.74 \mu_B$ stacked along the c axis. The rather low value of the ordered moments, which amounts to only half of the values found in isostructural UNiX compounds⁹, confirms the expected trends arising from $5f$ - d hybridization¹⁰ and points to a larger delocalization of the $5f$ electrons in UCoGa. For UCoGa, a significant anisotropy in the temperature dependence of the electrical resistivity has been found. Strongly anisotropic transport properties have also been detected in other UTX compounds¹¹, which suggests that they are caused by the general anisotropy due to the crystal structure and the Fermi-surface anisotropy.

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Table 1: Refined structural parameters for UCoGa at 60 K

Space group:		P62m				
U	3g	x	0	1/2	x = 0.580018 ± 0.000026	
Co ₁	2c	1/3	2/3	0		
Co ₂	1b	0	0	1/2		
Ga	3f	x	0	0	x = 0.239151 ± 0.000034	
Lattice parameters				R factors		
a = 666.456 ± 0.010 pm				R _{wp} = 3.00%		
c = 392.653 ± 0.006 pm				R _p = 2.13%		
				reduced χ ² = 3.53		

Figure captions

Fig. 1: The maximal non-isomorphic Shubnikov magnetic subgroup of P62m. In (a), the uranium moments are perpendicular to the mirror planes, while in (b) and (c), the moments are parallel to the mirror planes. Note that in the models (a) and (b) the moments are located within the hexagonal basal plane, while in (c) they are ferromagnetically coupled along the c axis. For clarity, only uranium atoms are shown. The dashed lines with arrows indicate the nearest uranium-uranium links.

Fig. 2: Temperature dependence of the electrical resistivity of UCoGa for $i \parallel c$ -axis and $i \perp c$ -axis normalized to the values at 300K. In the inset the low-temperature detail for $i \parallel c$ -axis is shown in the representation ρ/ρ_{300K} vs. T^2 .

Fig. 3: Field dependence of the electrical resistivity of UCoGa at 4.2 K in the configuration $i \parallel B \parallel c$ -axis.

Fig. 4: Temperature dependence of the specific heat of UCoGa.

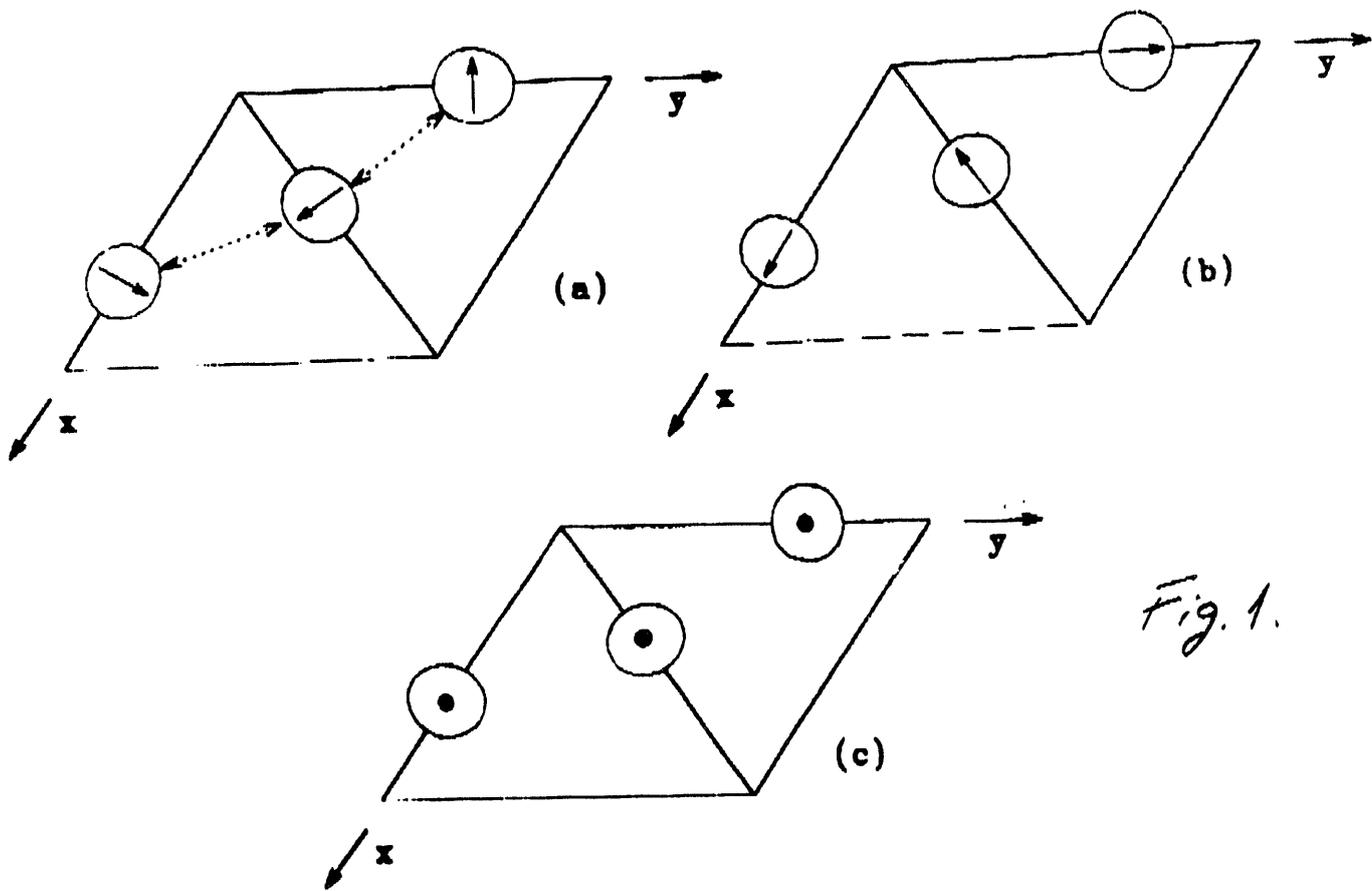
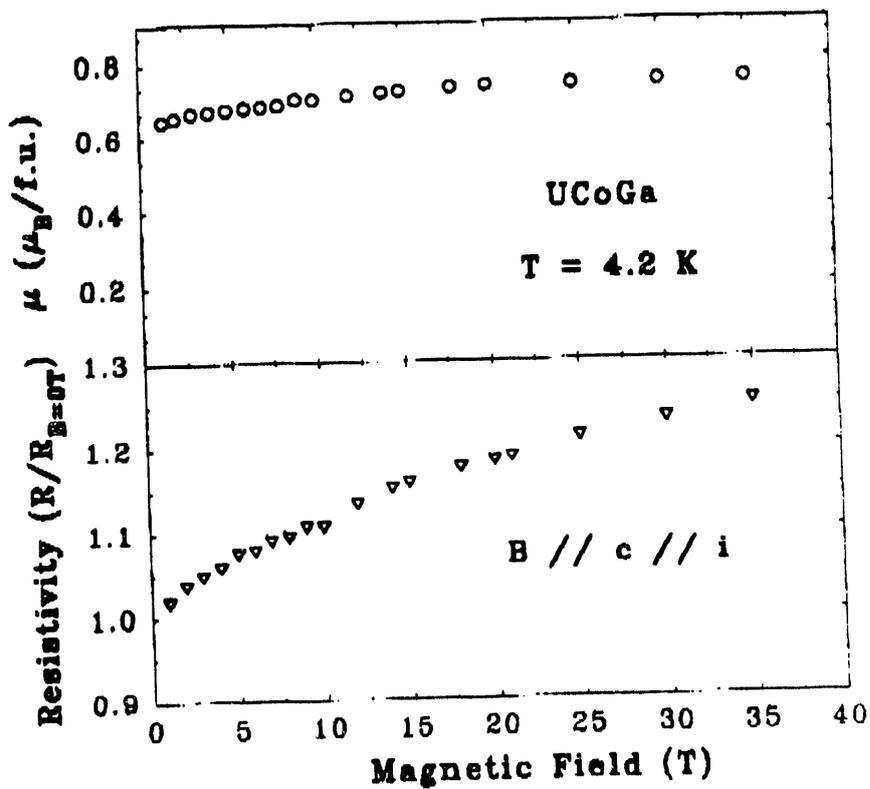


Fig. 1.

Fig. 3.



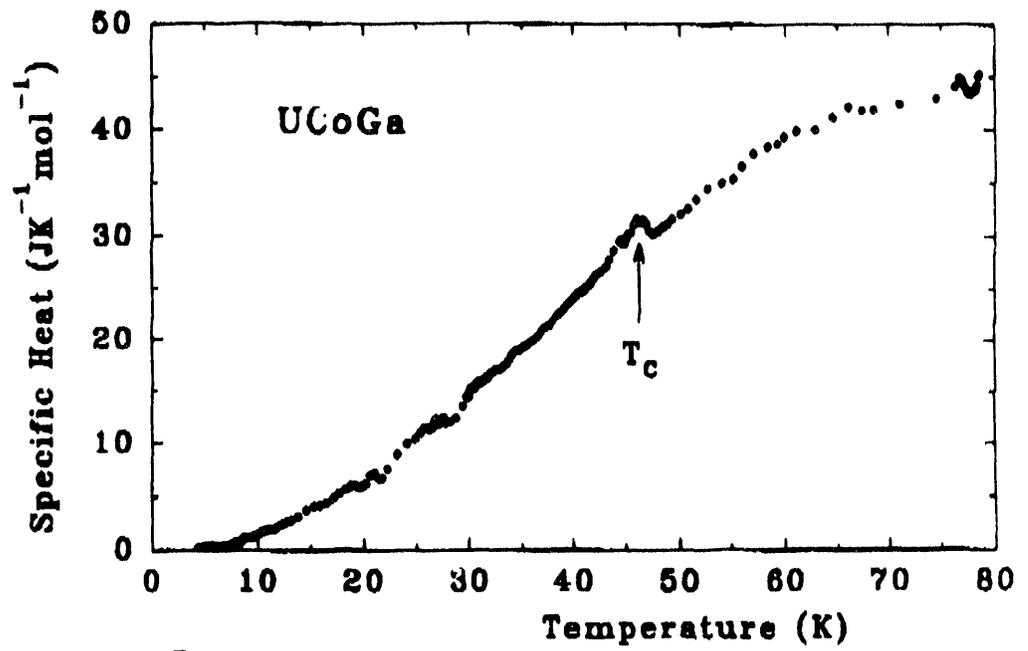


Fig. 4.

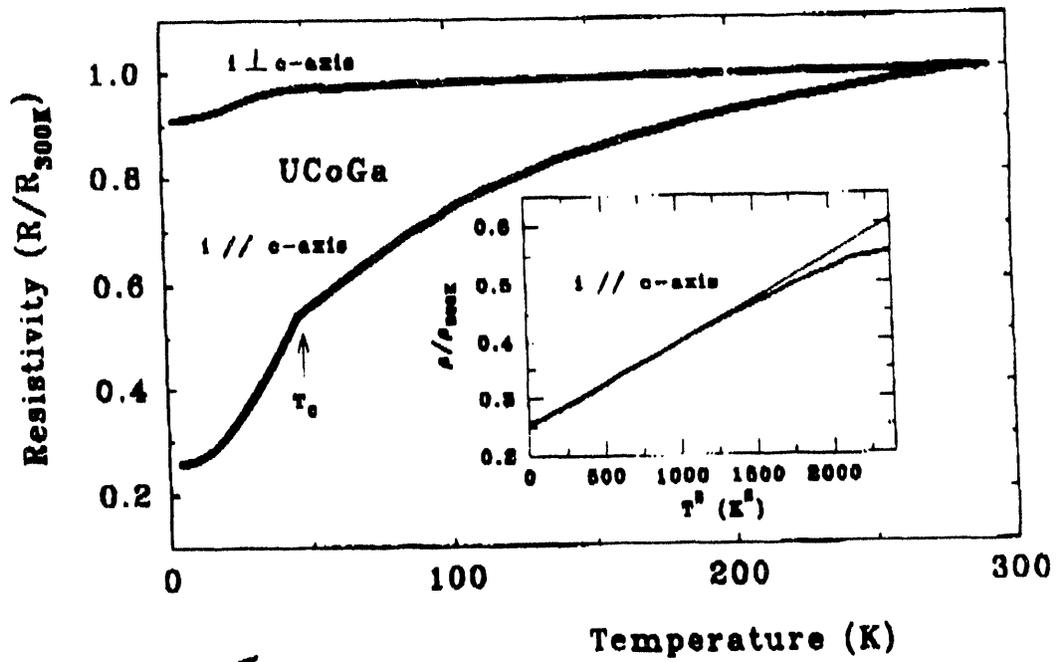


Fig. 2

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