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## Study on the U-Co-Ge ternary system at 973K

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The recent discovery of the coexistence of ferromagnetic order and pressure induced superconductivity in UCoGe [1] has boosted up explorative investigations on the crystal-chemistry and low-temperature physical properties of novel intermetallic phases in the U-Co-Ge ternary system. These efforts resulted in the characterization of six intermediate phases, which have hitherto been reported in the literature, namely: UCoGe [2], UCo<sub>2</sub>Ge<sub>2</sub> [3], UCo<sub>6</sub>Ge<sub>6</sub> [4], U<sub>3</sub>Co<sub>2</sub>Ge<sub>7</sub> [5], U<sub>3</sub>Co<sub>4</sub>Ge<sub>7</sub> [6] and U<sub>2</sub>Co<sub>17</sub>yGe<sub>y</sub> with  $1.3 \leq y \leq 3$  [7].

Our systematic study on the U-Co-Ge system was carried out in the whole concentration range of the ternary phase diagram. The syntheses were performed by melting appropriate amounts of the elements in an arc furnace under high-purity argon atmosphere. The samples were annealed at 973 K for 3 weeks in sealed silica tubes, followed by air quenching down to room temperature. The products were characterized by powder X ray diffraction. The microstructure and the chemical compositions were studied on polished surfaces using a scanning electron microscope coupled with an energy dispersive spectrometer (SEM-EDS). The crystal structures were determined from single crystal X-ray diffraction data. DC magnetic measurements were carried out using a SQUID magnetometer. The electrical resistivity was measured employing a standard DC four-point technique.

The assessment of U-Co-Ge isothermal section at 973 K confirmed the formation of the previously reported compounds. However, the crystal structure of UCo<sub>6</sub>Ge<sub>6</sub> was found different from that reported in the literature. In addition, our study revealed the existence of as many as twelve novel compounds: UCo<sub>3</sub>yGe<sub>y</sub> with  $0.2 \leq y \leq 0.4$ , UCo<sub>4.5</sub>Ge<sub>0.5</sub>, U<sub>2</sub>Co<sub>3</sub>Ge, U<sub>3</sub>Co<sub>6</sub>Ge<sub>2</sub>, U<sub>3</sub>Co<sub>12</sub>xGe<sub>4</sub> with  $0.0 \leq x \leq 2.0$  [8], U<sub>15</sub>Co<sub>55</sub>Ge<sub>30</sub>, U<sub>6</sub>Co<sub>30</sub>Ge<sub>19</sub>, U<sub>34</sub>Co<sub>4</sub>xGe<sub>33</sub> with  $0.0 \leq x \leq 3.0$ , UCo<sub>1-y</sub>Ge<sub>1+y</sub> with  $0.6 \leq y \leq 0.7$ , UCoGe<sub>2</sub>, U<sub>4</sub>Co<sub>1-y</sub>Ge<sub>6-x+y</sub> with  $x = 0.6$  and  $0.0 \leq y \leq 0.5$  and UCo<sub>1</sub>xGe<sub>2</sub> with  $0.22 \leq x \leq 0.45$  [9,10]. In this communication we describe the crystallographic features as well as the magnetic and electrical transport properties of U<sub>4</sub>Co<sub>1</sub>yGe<sub>6-x+y</sub> and UCo<sub>1-y</sub>Ge<sub>1+y</sub>.

UCo<sub>1-y</sub>Ge<sub>1+y</sub> ( $0.6 \leq y \leq 0.7$ ) crystallizes with a unit cell related to the AlB<sub>2</sub> type with the lattice parameters  $a = 4.111(5)$  Å and  $c = 7.641(5)$  Å (fig. 1a). The structure is composed of [U<sub>6</sub>] trigonal prisms, alternatively centred either by the (Co,Ge) atoms or the Ge atoms. The presence of modulations in the crystal structure makes its refinement difficult. The magnetic measurements revealed a ferromagnetic transition (fig. 2a), at the Curie temperature varying along the homogeneity range ( $38 \text{ K} \leq T_c \leq 48 \text{ K}$ ). In the paramagnetic region, the magnetic susceptibility follows a modified Curie-Weiss law with the effective magnetic moment of  $2.40(1)$  μB/(U atom). The electrical resistivity of UCo<sub>1-y</sub>Ge<sub>1+y</sub> is about  $360$  μΩcm at room temperature and shows rather minor temperature variation (fig. 2b). Below 75 K, it slightly increases with decreasing temperature reaching  $375$  μΩcm at liquid helium temperature. The ferromagnetic phase transition is hardly resolvable from the resistivity curve.

Fig.1: Projections on the (a,b) plane of the crystal structures of a) UCo<sub>1-y</sub>Ge<sub>1+y</sub> and b) U<sub>4</sub>Co<sub>1-y</sub>Ge<sub>6-x+y</sub>

U<sub>4</sub>Co<sub>1-y</sub>Ge<sub>6-x+y</sub> ( $x = 0.6$  and  $0.0 \leq y \leq 0.5$ ) adopts the hexagonal Er<sub>4</sub>(Ga,Ge)<sub>7</sub> type structure (space group P<sub>6</sub>m<sub>2</sub>) with the lattice parameters  $a = 8.016(1)$  Å and  $c = 4.021(1)$  Å. This crystal structure is a derivative of the AlB<sub>2</sub> type. The U atoms form [U<sub>6</sub>] trigonal prisms. Six over eight of them are occupied by the Ge atoms, one is centred by the Co atom and the eighth prism is empty, due to the proximity of two Ge atoms that are strongly shifted from the center of their prisms (fig. 1b). The compound shows a ferromagnetic transition at  $T_c = 39$  K (fig. 2a). At higher temperatures a modified Curie-Weiss law is observed for the magnetic susceptibility, with the effective magnetic moment of  $2.76(4)$  μB/(U atom). In the paramagnetic state the resistivity is almost temperature independent being of about  $275$  μΩcm (fig. 2b). The magnetic transition manifests itself as a distinct kink at the resistivity curve, below which the resistivity sharply decreases down to  $175$  μΩcm at 4 K.

Fig.2: Temperature dependencies of (a) the magnetization and (b) the resistivity of  $U_4Co_{1-y}Ge_{6-x+y}$  (blue symbols) and  $UCo_{1-y}Ge_{1+y}$  (red symbols). The ferromagnetic phase transitions are marked by arrows.

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**Primary author:** Mr SOUDÉ, Arnaud (Université Rennes 1)

**Co-authors:** Dr KACZOROWSKI, Dariusz (INTiBS); Dr NOËL, Henri (Université Rennes 1); Dr PASTUREL, Mathieu (Université Rennes 1); Dr TOUGAIT, Olivier (Université Rennes 1)

**Presenter:** Mr SOUDÉ, Arnaud (Université Rennes 1)

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