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STRUCTURAL AND MAGNETIC PROPERTIES OF THE U₃M₄Ge₁₃ (M = Ru, Os, Rh, Ir) TERNARY GERMANIDES

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Abstract. - U₃M₄Ge₁₃ and U₃Os₄Ge₁₃ germanides crystallize in a superstructure of the cubic Yb₃Rh₄Sn₁₃-type structure and exhibit no magnetic order above 4.2 K. On the contrary, U₃Ir₄Ge₁₃ orders ferromagnetically near Tc= 15-17 K whereas U₃Rh₄Ge₁₃ orders antiferromagnetically at 22 K.

In the last years, many ternary stannides, germanides and silicides with formulae RE₃M₄X₁₃ (RE = rare earth, M = Ru, Os, Rh, Ir and X = Si, Ge, Sn) were prepared [1, 2, 3]. These compounds crystallize in the cubic Yb₃Rh₄Sn₁₃-type (space group Pm3n) or in derived structure [4]. Some of these materials have remarkable superconducting properties.

Recently, it was established that URu₂Si₂ shows both a magnetic phase transition at 17.5 K and a superconducting transition at 0.8 K [5]. These interesting results have stimulated us to investigate other new uranium ternary compounds. We report here the structural, magnetic and electrical properties of the new ternary germanides U₃M₄Ge₁₃ with M = Ru, Os, Rh, Ir.

All samples were prepared by melting stoichiometric amounts of the binary germanide UGe₃, noble metals Ru, Os, Rh, Ir and germanium. The resulting ingots were annealed in evacuated quartz tubes at 850 °C for two weeks. The samples were characterized by X-ray diffraction and microprobe analysis.

X-ray powder diffraction shows that the U₃Ru₄Ge₁₃ and U₃Os₄Ge₁₃ germanides crystallize in the cubic Yb₃Rh₄Sn₁₃-type structure. The lattice parameter (a) is respectively equal to 8.939 Å for M = Ru and to 8.949 Å for M = Os. However a crystal study performed by Weissenberg and precession photographs on the U₃Os₄Ge₁₃ compound reveals a modification of the primitive cubic structure. Splitting of the diffraction spots, well seen at high θ values on Weissenberg films for the three axis, could be explained by a small tetragonal distorsion of the lattice and twinning of the crystal. In the family of ternary stannides, Gd₃Rh₄Sn₁₃ crystallizes in a tetragonal cell with a' ∼ a√2 and c' ∼ a [4]. As the difference between a'/√2 and c' is very small, twin formation has to be expected in this tetragonal phase. In the Yb₃Rh₄Sn₁₃-type or in the derived structure, the Ru and Os atoms occupy the trigonal prism formed by six Ge(2) atoms. The RuGe₆ or OsGe₆ prisms, share corners, to form a three-dimensional network, which generates icosahedral and cubo-octahedral sites which are occupied by the Ge(1) and U atoms. In these ternary germanides, the U-U distance (∼ 4.47 Å) is greater than the critical value of 3.4-3.6 Å known as the Hill limit [6]. Due to the decrease of the 5f-wave functions overlap, the U atoms bear a stable magnetic moment.

Above 300 K, the thermal variation of the reciprocal susceptibility χₘ⁻¹ of U₃Ru₄Ge₁₃ and U₃Os₄Ge₁₃ can be fitted to a linear Curie-Weiss law with an effective magnetic moment of 2.86 μ₆ / U atom for M = Ru and 3.24 μ₆ / U atom for M = Os. Above 4.2 K, no magnetic order has been detected for U₃Os₄Ge₁₃ compound. Below 6-8 K, χₘ tends to saturate for U₃Ru₄Ge₁₃ and the thermal variation of the electrical resistivity exhibits a minimum around T = 10-20 K and then increases at lower temperature. These results suggest a Kondo-like behaviour for U₃Ru₄Ge₁₃.

The X-ray powder patterns of U₃Rh₄Ge₁₃ and U₃Ir₄Ge₁₃ show that these germanides adopt a derived structure of Yb₃Rh₄Sn₁₃-type. Some diffraction lines corresponding to the Pm3n space group are split including the (222) one, and so a simple tetragonal distortion is excluded. So far, the crystal structure of these two germanides is unknown.

The magnetic susceptibility of U₃Ir₄Ge₁₃ follows a Curie-Weiss like dependence in the temperature range 300 K < T < 500 K with μeff = 2.92 μ₆ / U. This compound orders ferromagnetically near 15-17 K.

The thermal variation of the reciprocal magnetic susceptibility of U₃Ru₄Ge₁₃ (Fig. 1) shows a Curie-Weiss behaviour above 250 K with μeff = 2.96 μ₆ / U and θ = -70 K. The thermal dependence of the magnetic susceptibility reveals a maximum at 22 K suggesting an antiferromagnetic ordering of the U atoms (Fig. 1). The magnetization curves of U₃Rh₄Ge₁₃ observed at T = 2 K and 6 K in fields up to 6 T are displayed in figure 2. A rapid change of the magnetization between 3 and 4 T resembling a metamagnetic transition is observed at 6 K with a hysteresis phenomena. At 2 K, two jumps in the magnetization curve are clearly visible. In addition, the electrical resistivity of U₃Rh₄Ge₁₃ decreases at 1.7-1.8 K showing a zero-resistance value close to 1.5 K. The occurence of this anomaly which depends on the sample preparation could be due to the onset of a superconducting...
transition. A single crystal study is necessary in order to confirm if this transition is intrinsic or due to small amounts of precipitates not detected by X-rays.