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STRUCTURAL AND MAGNETIC PROPERTIES OF THE $U_3M_4Ge_{13}$ ($M = Ru, Os, Rh, Ir$) TERNARY GERMANIDES

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Abstract. – $U_3M_4Ge_{13}$ and $U_3Os_4Ge_{13}$ germanides crystallize in a superstructure of the cubic $Yb_3Rh_4Sn_{13}$-type structure and exhibit no magnetic order above 4.2 K. On the contrary, $U_3Ir_4Ge_{13}$ orders ferromagnetically near $T_c \simeq 15$-17 K whereas $U_3Rh_4Ge_{13}$ orders antiferromagnetically at 22 K.

In the last years, many ternary stannides, germanides and silicides with formulae $RE_3M_4X_{13}$ ($RE =$ rare earth, $M = Ru, Os, Rh, Ir$ and $X = Si, Ge, Sn$) were prepared [1, 2, 3]. These compounds crystallize in the cubic $Yb_3Rh_4Sn_{13}$-type (space group $Pm3n$) or in derived structure [4]. Some of these materials have remarkable superconducting properties.

Recently, it was established that $URu_2Si_2$ 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 $U_3M_4Ge_{13}$ with $M = Ru, Os, Rh, Ir$.

All samples were prepared by melting stoichiometric amounts of the binary germanide $UGe_3$, 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 $U_3Ru_4Ge_{13}$ and $U_3Os_4Ge_{13}$ germanides crystallize in the cubic $Yb_3Rh_4Sn_{13}$-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 $U_3Os_4Ge_{13}$ compound reveals a modification of the primitive cubic structure. Splitting of the diffraction spots, well seen at high $\theta$ values on Weissenberg films for the three axes, could be explained by a small tetragonal distortion of the lattice and twinning of the crystal. In the family of ternary stannides, $Gd_3Rh_2Sn_{13}$ crystallizes in a tetragonal cell with $a' \sim a\sqrt{2}$ and $c' \sim a$ [4]. As the difference between $a'/\sqrt{2}$ and $c'$ is very small, twin formation has to be expected in this tetragonal phase. In the $Yb_3Rh_4Sn_{13}$-type or in the derived structure, the Ru and Os atoms occupy the trigonal prism formed by six Ge(2) atoms. The RuGe$_6$ or OsGe$_6$ prisms, share corners, to form a three-dimensional network, which generates icosahedral and cuboctahedral sites which are occupied by the Ge(1) and U atoms. In these ternary germanides, the U-U distance ($\sim 4.47$ 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 $\chi_{\text{m}}^{-1}$ of $U_3Ru_4Ge_{13}$ and $U_3Os_4Ge_{13}$ can be fitted to a linear Curie-Weiss law with an effective magnetic moment of 2.86 $\mu_B / U$ atom for $M = Ru$ and 3.24 $\mu_B / U$ atom for $M = Os$. Above 4.2 K, no magnetic order has been detected for $U_3Os_4Ge_{13}$ compound. Below 6-8 K, $\chi_{\text{m}}$ tends to saturate for $U_3Ru_4Ge_{13}$ and the thermal variation of the electrical resistivity exhibits a minimum around $T \approx 10-20$ K and then increases at lower temperature. These results suggest a Kondo-like behaviour for $U_3Ru_4Ge_{13}$.

The X-ray powder patterns of $U_3Rh_4Ge_{13}$ and $U_3Ir_4Ge_{13}$ show that these germanides adopt a derived structure of $Yb_3Rh_4Sn_{13}$-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 $U_3Ir_4Ge_{13}$ follows a Curie-Weiss like dependence in the temperature range 300 K $< T < 500$ K with $\mu_{\text{eff}} = 2.92 \mu_B / U$. This compound orders ferromagnetically near 15-17 K.

The thermal variation of the reciprocal magnetic susceptibility of $U_3Rh_4Ge_{13}$ (Fig. 1) shows a Curie-Weiss behaviour above 250 K with $\mu_{\text{eff}} = 2.96 \mu_B / U$ and $\theta_m = -70$ 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 $U_3Rh_4Ge_{13}$ 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 $U_3Rh_4Ge_{13}$ decreases at 1.7-1.8 K showing a zero-resistance value close to 1.5 K. The occurrence 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.


