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Structure and magnetic properties of $U_2Co_{15}Ge_2$ and related compounds

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Abstract

Ternary compounds of $U_2T_{15}Ge_2$ (T≡Fe, Co and Ni) are crystallized in the hexagonal Th_2Ni_{17}-type structure. The Curie temperatures were determined for $U_2Fe_{15}Ge_2$, $U_2Co_{15}Ge_2$ and the alloy $U_2Co_{15-x}Fe_xGe_2$ ($x=3$, 6, 9, 12), and were found to be as high as 540–750 K. The magnetocrystalline anisotropy of $U_2Fe_{15}Ge_2$ and $U_2Co_{15}Ge_2$ was studied on single crystals.

1. Introduction

Investigation of actinide compounds has proved that actinide atoms often cause a strong magnetic anisotropy in numerous compounds [1]. Since the radioactivity of the natural and depleted U is extremely low, the U compounds with the elements of high magnetization and sufficiently high Curie temperature, such as Fe, Co and Ni, attract much interest for use as permanent magnet and magneto-optical recording materials. Among the binary U systems with these elements, however, only UCo_5 orders ferromagnetically above room temperature [2]. Up to now, much attention has been paid to the Fe-rich ternary intermetallic $UFe_{10}Si_2$ with a tetragonal $ThMn_{12}$-type structure [3].

In this paper, some magnetic properties of the ternary compound $U_2Co_{15}Ge_2$ and related compounds, which have Curie temperatures well above room temperature and which crystallize in hexagonal Th_2Ni_{17}-type structure, are presented.

2. Experimental details

The polycrystalline samples were prepared by melting the elements of at least 99.9% purity in stoichiometric amounts in an arc furnace under an Ar atmosphere purified by Ti. After arc melting, the samples were wrapped in Ta foils and annealed at about 950 °C in evacuated silica ampoules for one week. The single crystals of $U_2Co_{15}Ge_2$ and $U_2Fe_{15}Ge_2$ were grown by the Czochralski pulling method in a tri-arc furnace.

The Curie point was determined from the temperature dependence of the magnetization for the polycrystalline samples in a low magnetic field using a vibrating sample magnetometer. The magnetic field dependence of the magnetization was measured for the single crystals oriented from 0 to 10 T at 4.2 K. The density was obtained by gravimetric measurements.

3. Results and discussion

The phase analysis by X-ray diffractometry revealed a nearly single phase with the hexagonal Th_2Ni_{17}-type structure for $U_2Fe_{15}Ge_2$, $U_2Co_{15}Ge_2$ and $U_2Ni_{15}Ge_2$. The lattice parameters and densities are given in Table 1. The densities calculated by assuming the number of molecules to be two are in good agreement with the measured values. This shows that the hexagonal Th_2Ni_{17}-type structure is stabilized in ternary compounds of U, though isomorphous binary U_2M_{17} (M≡Fe, Co, Ni) cannot be formed. The occurrence of this phase is similar to one of $SmTiFe_{11}$ [4], $RFe_{10}M_2$ (R≡rare earth;

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TABLE 1. Crystallographic data of the ternary germanides

Compound	a (Å)	c (Å)	d_{expt}	d_x
$U_2Fe_{15}Ge_2$	8.419	8.355	9.44	9.45
$U_2Co_{15}Ge_2$	8.303	8.180	10.0	10.2
$U_2Ni_{15}Ge_2$	8.282	8.118	10.2	10.3

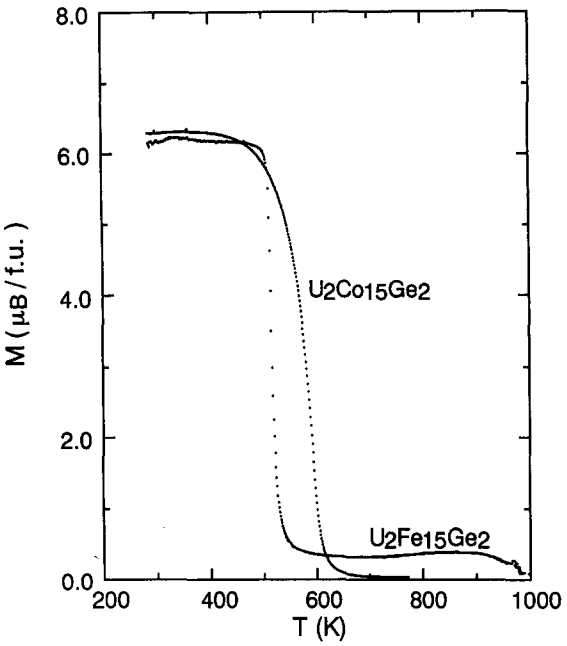

Fig. 1. Temperature dependence of the magnetization of $U_2Co_{15}Ge_2$ and $U_2Fe_{15}Ge_2$ at 0.1 T.

M≡Si, Ti, V, Cr, Mo, W) [5] or $UFe_{10}Si_2$ with a tetragonal $ThMn_{12}$-type structure. In the Th_2Ni_{17}-type structure, four atomic sites of 4f, 6g, 12j and 12k are available for occupation by Ge atoms. However, further investigations are necessary for the structural determination.

The temperature dependence of the magnetization shows that $U_2Fe_{15}Ge_2$ and $U_2Co_{15}Ge_2$ order ferromagnetically at temperatures as high as 540 K and 650 K, respectively, as is shown in Fig. 1, whereas $U_2Ni_{15}Ge_2$ is non-magnetic at room temperature. Berlureau *et al.* [6] reported the crystallographic data and Curie temperature for $U_2Fe_{15}Ge_2$, which are in good agreement with ours. The germanide with Co has a higher ordering temperature than that with Fe, which is similar to the Curie temperatures of R_2Co_{17} and R_2Fe_{17}: 1100–1200 K with Co and 300–500 K with Fe [7]. The Curie temperature of $U_2Fe_{15}Ge_2$ is higher than that of R_2Fe_{17}, despite the lower Fe content, which may suggest the contribution of the U sublattice to the magnetic properties. However, in comparison with $UCo_{10}Si_2$ and $UFe_{10}Si_2$, the ordering temperature of the compound with Co is lower by approximately 90 K [8].

It is expected that pseudoternary compounds of $U_2(Co,Fe)_{15}Ge_2$ exhibit some improvement in magnetic properties. The $U_2Co_{15-x}Fe_xGe_2$ ($x=3$, 6, 9, 12) alloys prepared contain a considerable amount (about 10%–20%) of extraneous phases, mainly α-Fe and α-Co. The lattice parameter of the Th_2Ni_{17}-type structure was obtained by X-ray reflection. As shown in Fig. 2, both parameters a and c do not follow the Vegard law. The Curie point was determined from the temperature dependence of the magnetization, though the measurement of the saturation moment was not allowed, owing to the existence of the extraneous phases. The Curie temperatures of the Co-rich quasi-ternaries ($x=3$, 6, 9) were as high as 700 K or more.

The field dependences of the molecular magnetic moment for single crystals $U_2Co_{15}Ge_2$ and $U_2Fe_{15}Ge_2$ are presented in Fig. 3. No differences in the magnetization resulting from an increasing or decreasing magnetic field were observed. One can see that the easy axis of magnetization is in the basal plane, the hard axis is the c axis for both germanides, and the magnetocrystalline anisotropy of $U_2Co_{15}Ge_2$ is almost equal to that of $U_2Fe_{15}Ge_2$. The hard direction curve of $U_2Fe_{15}Ge_2$ suggests some field-induced phase transition. The saturation moments at 4.2 K are $27.9\mu_B$/f.u. for $U_2Fe_{15}Ge_2$ and $14.2\mu_B$/f.u. for $U_2Co_{15}Ge_2$.

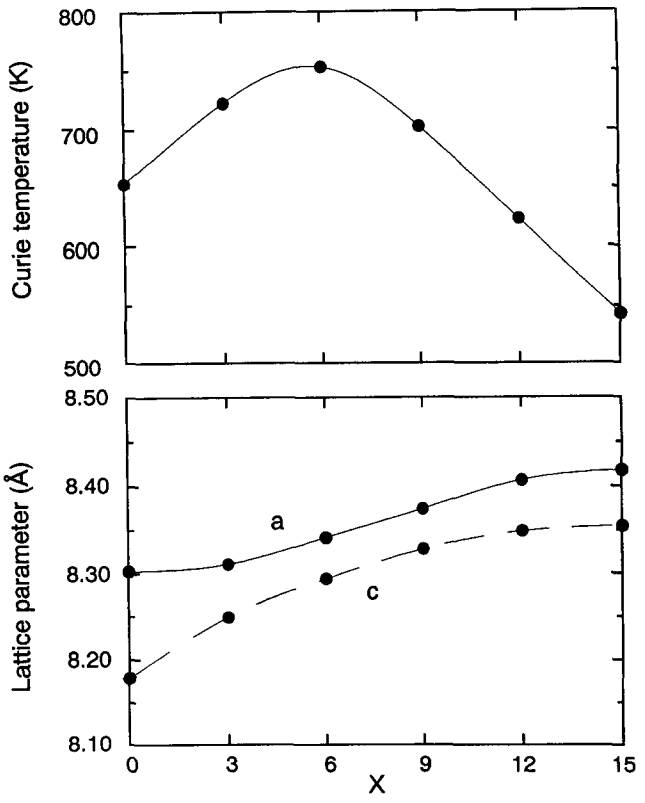

Fig. 2. Lattice parameter and Curie temperature of $U_2Co_{15-x}Fe_xGe_2$.

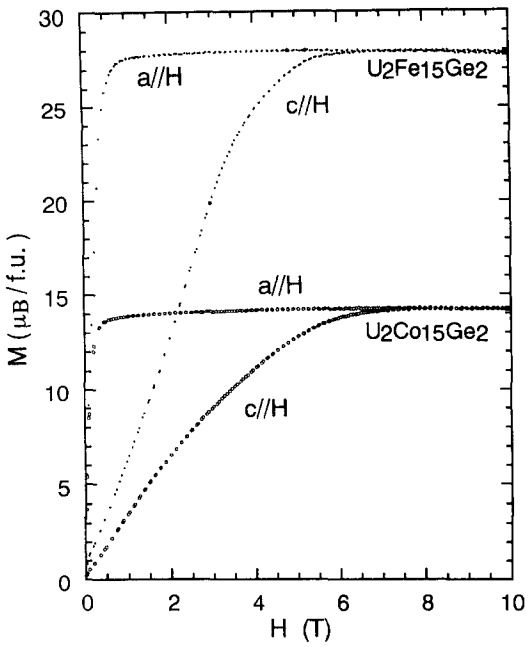

Fig. 3. Magnetization of $U_2Co_{15}Ge_2$ and $U_2Fe_{15}Ge_2$ along the *a* axis and *c* axis at 4.2 K.

The nearest-neighbour U–U separations are 4.86 Å in $U_2Fe_{15}Ge_2$ and 4.79 Å in $U_2Co_{15}Ge_2$, assuming the 2b and 2d positions in the Th_2Ni_{17}-type structure are occupied by U atoms. Since the separations exceed the critical distance of about 3.50 Å given by the Hill criterion [9], the U might be magnetic. In fact, in the $ThMn_{12}$-type $UFe_{10}Si_2$, whose nearest-neighbour U–U separation of 4.72 Å is shorter than that for the germanides, the presence of not only the Fe–Fe but also the U–Fe exchange interaction is suggested from the magneto-optical properties [10] and the magnetocrystalline anisotropy on aligned powder samples [11, 12].

The U germanides investigated with the hexagonal Th_2Ni_{17}-type structure are found to have Curie temperatures as high as 540–750 K, high magnetization values and fairly strong magnetocrystalline anisotropy. However, the following questions remain unsolved: (1) the preference of the Ge atom to occupy sites in the hexagonal Th_2Ni_{17}-type structure; (2) the contribution of U atoms to the magnetic properties. To answer these, the compounds substituted for U, Fe or Co atoms by non-magnetic atoms should be investigated, in addition to more sophisticated characterization for $U_2Fe_{15}Ge_2$ and $U_2Co_{15}Ge_2$.

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