Vol. 32 (2001) *ACTA PHYSICA POLONICA B* No 10 Special Issue

MAGNETIC ORDERING IN $Ce_2Co_{1-x}Au_xSi_3$ COMPOUNDS*

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(Received June 21, 2001)

Polycrystalline samples of $Ce_2Co_{1-x}Au_xSi_3$ for x equal to 0.4, 0.6, 0.8 and 1.0 have been studied by neutron diffraction. All compounds exhibit the hexagonal crystal structure derived from the AlB_2-type. For $x = 0.4$ the Ce magnetic moments equal to 0.6(1) μ_B form a simple antiferromagnetic structure and are parallel to the b-axis. In $Ce_2Co_{0.2}Au_{0.8}Si_3$ a short range magnetic order is observed. For $x = 0.6$ and 1.0 the Ce-moments do not order up to 1.5 K.

PACS numbers: 61.12.–q, 75.25.+z, 75.30.–m, 75.50.Ee

1. Introduction

The magnetic properties of cerium intermetallic compounds have been known to be controlled by sensitive balance between the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and the Kondo effect. The competition between these two mechanisms has been viewed in the literature in light of Doniach's one-dimensional Kondo necklace model [1–3].

The ternary intermetallic compounds Ce_2TSi_3 (T — nd electron metals) crystallize in a derivative of the hexagonal AlB_2-type structure with the Ce magnetic atoms fully ordered on the aluminium site 1a: 0,0,0 and the non-

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[*] Presented at the XII School of Modern Physics on Phase Transitions and Critical Phenomena, Lądek Zdrój, Poland, June 21–24, 2001.

magnetic atoms (T/Si) both on the boron site $2d$: $\frac{1}{3},\frac{2}{3},\frac{1}{2}$. These systems are of particular interest because they have complex magnetic properties: a long-range in Ce_2RhSi_3 [4] and a short-range in Ce_2PdSi_3 [5,6] antiferromagnetic order.

Investigated in this work the $Ce_2Co_{1-x}Au_xSi_3$ compounds exist in the whole region of concentration x. X-ray data indicate that these compounds crystallize in a $(2a, 2a, c)$ variant of the AlB_2 structure [7,8].

Macroscopic measurements suggest interesting properties of these compounds. The Ce_2CoSi_3 is a non magnetic Kondo-lattice compound [7,8]. With increasing the gold content the properties of $Ce_2Co_{1-x}Au_xSi_3$ change from characteristic for a non magnetic Kondo regime to the ones characteristic for a magnetically ordered RKKY- interaction dominated region. The temperature dependence of the specific heat give the sharp peak at 3.8 K for $x = 0.4$, one broad at 4 K for $x = 0.6$ and at 3.2 K for $x = 1.0$ and two broad peaks close to 4 and 7 K for $x = 0.8$. The plots of low-temperature magnetic susceptibilities and magnetization data indicate magnetic transitions around these temperatures [8].

This work reports the neutron diffraction data for $Ce_2Co_{1-x}Au_xSi_3$ ($x = 0.4$, 0.6, 0.8 and 1.0) compounds on the basis of which the crystal and magnetic structure parameters are determined.

2. Experiment and results

The experiment was performed on the same samples as were used in our previous study [8].

Neutron diffraction patterns were measured in the temperature range 1.5–10 K using the E6 diffractometer installed at the BERII reactor at the Berlin Neutron Scattering Centere, Hahn–Meitner Institute in Berlin. The incident neutron wave length was 2.441 Å. The data processing was performed by the Rietveld-type FULLPROF program [9].

Diffraction patterns collected in the paramagnetic state at 10 K for $Ce_2Co_{0.6}Au_{0.4}Si_3$ is shown in Fig. 1(a). The processing of neutron data confirmed that the space group is P6/mmm (No 191). The atoms occupy the following positions:

$$\begin{array}{rcrl}
\mathrm{Ce_1} & \text{in} & 1a: & 0,0,0; \\
\mathrm{Ce_2} & \text{in} & 3f: & \frac{1}{2},0,0; \\
\text{Co, Au randomly} & \text{in} & 2d: & \frac{1}{3},\frac{1}{3},\frac{1}{2}; \\
\mathrm{Si} & \text{in} & 6\mathrm{m}: & x,y,\frac{1}{2}\,.
\end{array}$$

The crystal structure of these compounds is shown in Fig. 2.

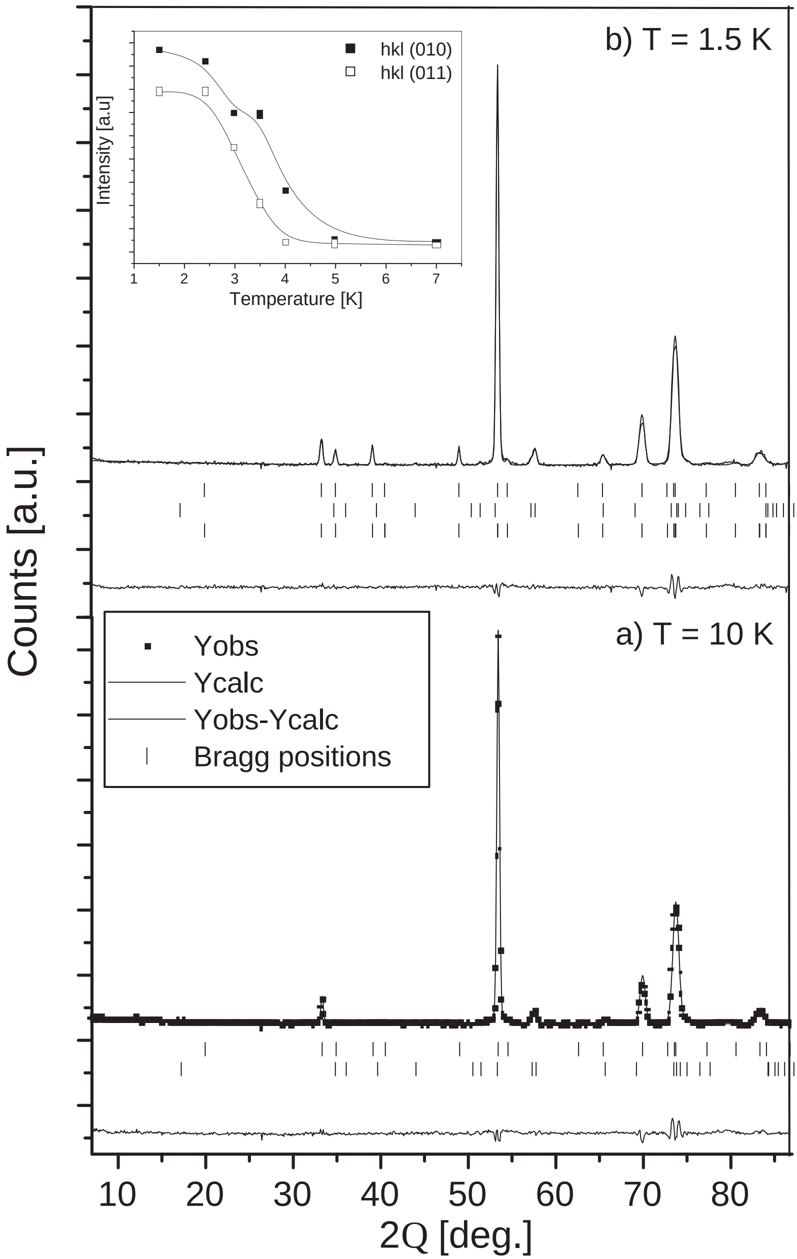

Fig. 1. Observed and calculated neutron diffraction patterns of $Ce_2Co_{0.6}Au_{0.4}Si_3$ at (a) 10 K and (b) 1.5 K. The inset shows the temperature dependence of the 100 and 011 magnetic peaks intensities.

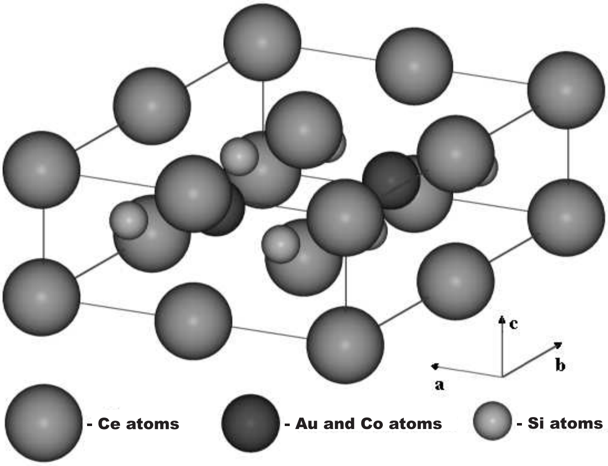

Fig. 2. Crystal structure of $Ce_2Co_{1-x}Au_xSi_3$ compounds.

For $Ce_2Co_{0.6}Au_{0.4}Si_3$ compound at low temperature on the 1.5 K pattern additional peaks of a magnetic origin are observed (see Fig. 1(b)). These peaks are indexed on an orthorhombic magnetic unit cell with lattice parameters derived from the hexagonal cell:

$$a_m = \frac{a}{2}, \quad b_m = \frac{\sqrt{3}a}{2}, \quad c_m = c$$

where a and c are the lattice parameters of the hexagonal crystal unit cell. The magnetic unit cell defined above contains only two cerium atoms located at the positions: (0,0,0) and $(\frac{1}{2}, \frac{1}{2}, 0)$. Only one type of antiferromagnetic order is possible in this lattice. The magnetic moments equal to 0.6(1) μ_B are coupled antiferromagnetically. An absence of the M(010) reflection indicates that cerium moments are aligned along the a-axis. The temperature dependence of the integral intensity of the 100 and 011 reflections give the Néel temperature 4.3 K which is in agreement with the specific heat data (3.8 K) [8].

For the $Ce_2Co_{0.2}Au_{0.8}Si_3$ compound at 1.5 K broad peaks at 2θ equal to 10^o, 36^o and 49^o are observed (Fig. 3). These peaks are similar to those observed in Ce_2PdSi_3 [5,6] and suggest a short range magnetic order. The intensity of these peaks decreases down to zero at 6 K.

For the $Ce_2Co_{0.4}Au_{0.6}Si_3$ and Ce_2AuSi_3 compounds at low temperature on the 1.5 K patterns there are no additional peaks of a magnetic origin which suggests an absence of a magnetic order at 1.5 K.

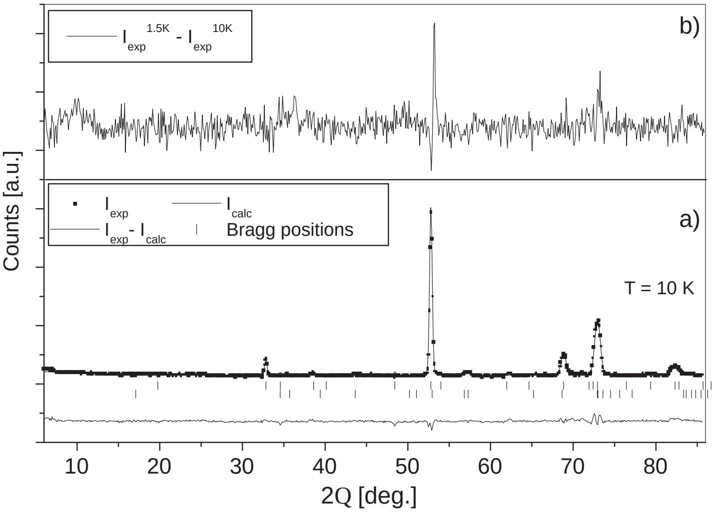

Fig. 3. (a) Observed and calculated neutron diffraction pattern of $Ce_2Co_{0.2}Au_{0.8}Si_3$ at 10 K. The squares represent the experimental points, solid lines the calculated profiles and difference bewteen observed and calculated patterns. The bars indicate the nuclear Bragg peaks. (b) Differential pattern at 1.5 and 10 K.

3. Discussion

The neutron diffraction experiment confirms that the $Ce_2Co_{1-x}Au_xSi_3$ (x = 0.4, 0.6, 0.8 and 1.0) compounds have the hexagonal structure of Ce_2CoSi_3 type. In the case of the compound with $x = 0.4$ the neutron diffraction data confirm the existence at low temperatures of a long-range magnetic order of cerium moments. The determined magnetic structure is similar to that observed in the isostructural Ce_2RhSi_3 [4]. A determined Ce magnetic moment equal to 0.6(1) μ_B is smaller that observed in Ce_2RhSi_3 (1.3(1) μ_B). In both these compounds the Ce magnetic moments in an ordered state are reduced, as compared to the value for the free Ce^{3+} ion value (g_J=2.14 μ_B).

The same tendency has been observed in a large number of cerium intermetallic compounds (see, for example Ref. [10]) and is commonly attributed either to valence fluctuations or to the Kondo effect [11]. For $Ce_2Co_{0.2}Au_{0.8}Si_3$ the short-range antiferromagnetic correlations similar to those observed in Ce_2PdSi_3 [5,6] are observed. In $Ce_2Co_{0.4}Au_{0.6}Si_3$ and Ce_2AuSi_3 the magnetic order is not detected up to 1.5 K. It is in contradiction with the macroscopic data (temperature dependence of the specific heat and magnetic susceptibility) which indicate an existence of the magnetic order in these compounds at low temperatures [8].

The obtained results for $x = 0.4$ are in good agreement with the results of the magnetization measurements (see Fig. 6 in Ref. [8]). The magnetization curve at $T = 2$ K has a metamagnetic transition with $H_{\mathrm{cr}} \simeq 10$ kOe typical for an antiferromagnet. For $x = 0.8$ and 0.9 the H_{cr} is near 1 kOe [8], in spite of fact that we have found only short range order for $x = 0.8$.

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