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Magnetic structure of $TbAgCu_4$

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The magnetic susceptibility of $TbAgCu_4$ has been measured between 4.2 and 400 K. The results are $T_N = 11.5 \pm 0.5$ K, $\theta_p = 9$ K, and $\mu_{eff} = 10.3\ \mu_B$. Neutron diffraction experiments were performed on polycrystalline specimen at temperatures 4.2–20, 77, and 300 K. The magnetic spin structure was determined to be like that of FeO: the magnetic spins align ferromagnetically in sheets parallel to the (111) plane and couple antiferromagnetically with adjacent sheets. The magnetic spin axis is directed along [111]. The magnetic moment for Tb is obtained to be $9 \pm 0.4\ \mu_B$.

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INTRODUCTION

The intermetallic compounds RM_5 formed between the rare earth metals (R) and the noble metals (M) have either the hexagonal $CaCu_5$-type structure or the cubic $AuBe_5$-type structure. In general, the structure is hexagonal for the light R metals and cubic for the heavy R metals and the tendency is well understood in terms of the radius ratio (1). The R atoms are situated on a face centered cubic lattice in the $AuBe_5$-type structure. The magnetic properties of the cubic RCu_5 compounds are antiferromagnetic for R = Tb and Dy and ferromagnetic for Ho, Er and Tm (2).

The effect of replacement of Cu with Ag in RCu_5 have been investigated by Takeshita et al.(3). The structure of $RAgCu_4$ is also cubic $AuBe_5$ for R= Nd to Tm. The magnetic properties are antiferromagnetic for R = Nd to Tb, and ferromagnetic for Dy and Ho. They reported the following magnetic properties for $TbAgCu_4$: T_N = 16.5 K and μ_{eff} = 9.38 u_B.

In this paper, we present experimental results of neutron diffraction studies on $TbAgCu_4$ together with some magnetic properties.

EXPERIMENTAL PROCEDURES

The specimens of $TbAgCu_4$ were prepared by melting the starting materials in an arc furnace under a purified argon gas atmosphere. The purities of the raw materials were: Tb 99.9 %, Ag 99.99 % and Cu 99.999 %. There was the weight loss of about 0.1 % during melting. The product was checked by X-ray powder diffraction technique at room temperature and the $AuBe_5$-type structure was confirmed. The obtained lattice parameter, a, is 7.161 A which is a little larger than 7.148 A in (3). There was no evidence from the X-ray results for the presence of any impurity phase.

The magnetic susceptibility was measured using a magnetic balance in the range of temperature between 4.2 and 400 K in a field of 2 kOe. The temperature was measured with a platinum thermocouple which was in contact with the specimen.

The neutron diffraction experiment was performed using the TOG diffractometer installed in the JRR-3, JAERI at temperatures: 4.2 K, 4.2 - 20 K. 77 K and 300 K. The neutron wavelength employed was 1.04 A, obtained from the (002) planes of a PG monochromating crystal. The polycrystalline specimen was crushed to a particle size smaller than 1 mm and put into a thin-walled aluminum capsule, 8 mm in diameter and 40 mm in length. The temperature of the specimen was measured using a Ge resistance thermometer.

RESULTS AND DISCUSSIONS

The magnetic susceptibility of $TbAgCu_4$ is shown in Fig. 1 as a function of temperature. With the rise of temperature from 4.2 K, the susceptibiltiy increases gradually at first, steeply around 8 - 9 K, peaks at 11.5 K and then decreases steadily. At higher temperatures the inverse of the magnetic susceptibility shows a linear variation with temperature. $TbAgCu_4$ is thus antiferromagnetic with $T_N = 11.5 \pm 0.5$ K, θ_p = 9 K and μ_{eff} = 10.3 u_B. These values may be compared with those previously reported (3), and the agreement is quite good except for a large discrepancy in T_N, which was reported as 16.5 K.

Fig. 2 shows the neutron diffraction patterns obtained at 77 K and 4.2 K. The pattern at 77 K consists of nuclear reflections from an $AuBe_5$-type lattice and the pattern at 300 K is essentially the same as that at 77 K. In the pattern at 4.2 K, there are many additional peaks compared to the 77 K pattern. All of these new peaks at 4.2 K may be indexed with half-odd indices which signifies the formation of a superstructure with a period twice that of the original lattice.

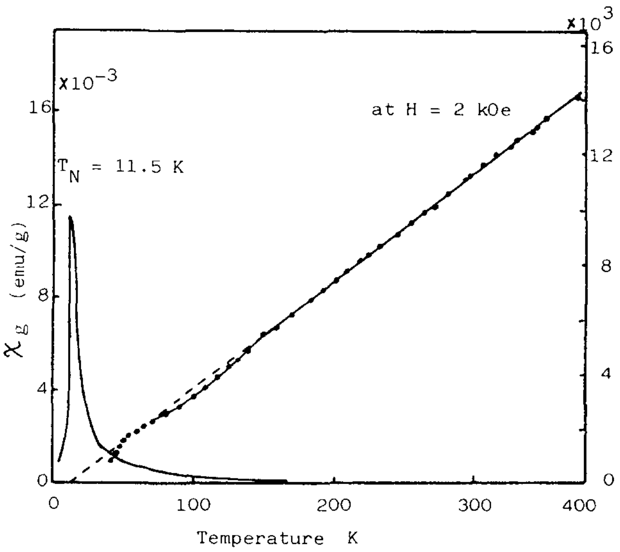

Fig. 1 Temperature dependence of magnetic susceptibility and its reciprocal of $TbAgCu_4$

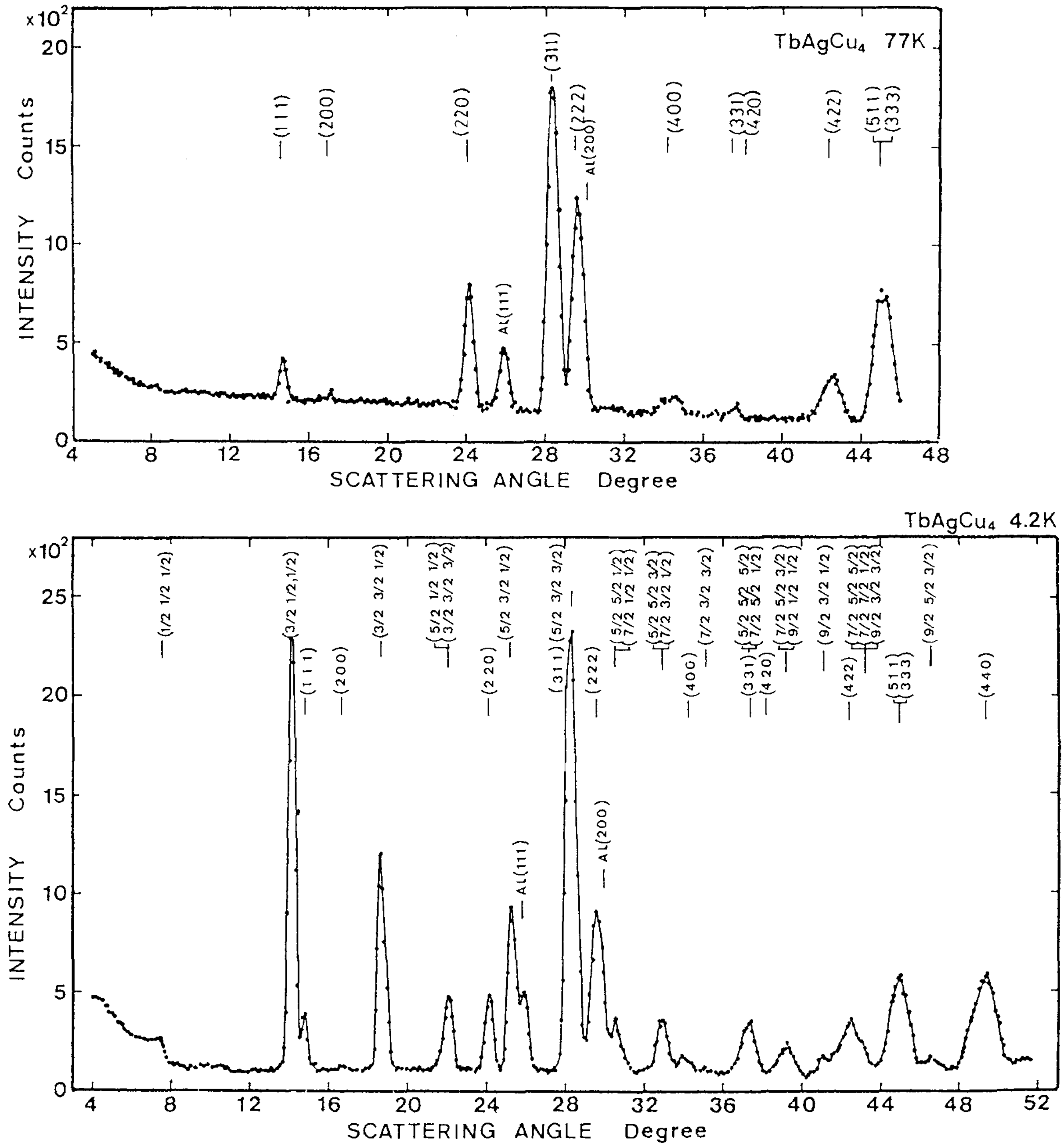

Fig. 2. Neutron diffraction patterns for $TbAgCu_4$ at 4.2 K and 77 K.

Another feature of the 4.2 K pattern is the fact that the distinct (1/2 1/2 1/2) superstructure peak is absent, except for a weak diffuse scattering ranging from 5 ° to 8 °. This kind of double magnetic cell was first observed in the antiferromagnetic state of FeO, which is an NaCl-type compound with the Fe atoms on a face centered cubic lattice (4). In FeO, the magnetic moments are in ferromagnetic sheets parallel to (111) planes and the direction of magnetic moment is antiparallel for neighbouring sheets. The magnetic moments are directed along [111] axis.

The intensities of neutron reflections in $TbAgCu_4$ were calculated on the basis of the FeO-type spin structure and the results are shown in Table 1 together with the observed values. Some systematic errors were observed for the nuclear lines most likely due to too large particle size. A correction was applied by averaging the values of two measurements taken with the specimen rotated around the vertical axis by 90 °. In this way, a better agreement, which the crystallographic R factor was about 6 %, was obtained for the nuclear lines. The intensities(cal.) for the nuclear lines were calculated by using the value, x = 5/8, for the parameter of Cu atom position. The intensities of the half-odd lines agree well with those calculated for magnetic reflections using the magnetic form factor of Tb^{3+} by A.J. Freeman and J.P. Desclaux (5). The R factor was about 8 %. Hence, the magnetic structure of $TbAgCu_4$ is concluded to be similar to that of FeO; the magnetic moment of the Tb ions are ferromagnetically coupled in the sheets parallel to the (111) planes with antiferromagnetic coupling between adjacent sheets. The magnetic moment are directed along the [111] axis. The value of magnetic moment derived from the magnetic peaks is $9 \pm 0.4\ \mu_B$ per Tb ion.

TABLE I Observed and calculated intensities of $TbAgCu_4$ in the antiferromagnetic state. (4.2 K)

h k l	2θ degree	Intensity obs.	Intensity calc.
1/2 1/2 1/2	7.4	2	
3/2 1/2 1/2	14.1	65	69
1 1 1	14.7	5	6
2 0 0	16.7		
3/2 3/2 1/2	18.6	27	20
3/2 3/2 3/2, 5/2 1/2 1/2	22.0	12	9
2 2 0	24.1	19	18
5/2 3/2 1/2	25.2	27	28
5/2 3/2 3/2			11
3 1 1	28.3	100	89
2 2 2	29.6	49	42
5/2 5/2 1/2, 7/2 1/2 1/2	30.5	10	9
5/2 5/2 3/2, 7/2 3/2 1/2	32.9	13	11
4 0 0	34.2	4	7
7/2 3/2 3/2	35.1		1
5/2 5/2 5/2, 7/2 5/2 1/2	37.3		7
3 3 1	37.4	11	3
4 2 0	38.2		2
7/2 5/2 3/2, 9/2 1/2 1/2	39.2	8	7
9/2 3/2 1/2	41.1	5	4
4 2 2	42.4	12	14
7/2 5/2 5/2, 7/2 7/2 1/2	43.2	4	5
9/2 3/2 3/2			
5 1 1, 3 3 3	44.9	49	49
9/2 5/2 3/2	46.5	2	2
4 4 0	49.3	53	54

The structure of $TbAgCu_4$; the space group O_h^7 - Fd3m
4 Tb; 000
4 Ag; 1/4 1/4 1/4
16 Cu; xxx; $\bar{x}\bar{x}x$; $\bar{x}x\bar{x}$, $x\bar{x}\bar{x}$
with x = 5/8

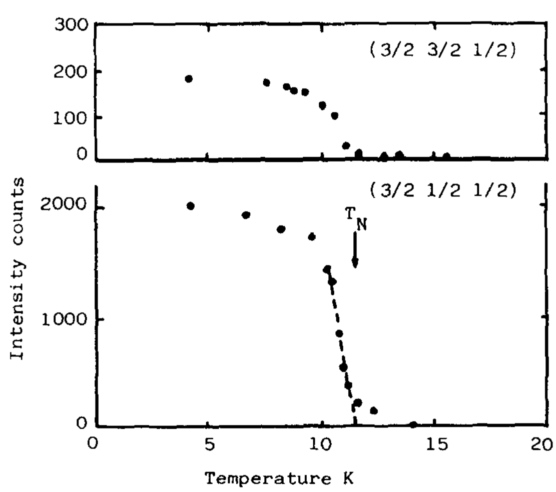

Fig. 3. Temperature dependence of magnetic reflection lines (3/2 3/2 1/2) and (3/2 1/2 1/2)

The variation with temperature of the reflections (3/2 1/2 1/2) and (3/2 3/2 1/2) was observed in the interval from 4.2 to 20 K. The results are shown in Fig. 3. The intensities of the two peaks decrease gradually with temperature at first, rapidly above 9 K and finally disappear around 14 K. We define the Néel temperature as being the point of the intersection of the T axis and the line extrapolated from the linear part of the intensity vs temperature curve as shown in Fig.3. The Neel temperature is determined to be 11.5 K, which coincides with the peak in the magnetic susceptibility. In the present case, no discontunuity was observed around 16.5 K.

In $TbAgCu_4$, the exchange interactions, J_1 between nearest neighbours and J_2 between next nearest neibours may be expressed by the following relations:

$$T_N\, q = -6J_2$$

$$\theta_p\, q = 12J_1 + 6J_2$$

and

$$q = (3/2)(g-1)^2 j(j+1).$$

We obtain:

$$J_1 = 0.244 \text{ K} \quad ; \quad J_2 = -0.278 \text{ K}$$

As T_N AND θ_p are very close, J_1 is close to $-J_2$.

In the monooxides of the transition metals: MnO, FeO, CoO and NiO, a spontaneous distortion of the lattice is observed in the magnetically ordered state. I the present neutron diffraction experiment, no evidence of distortion was observed in $TbAgCu_4$. More precise information will be obtained by X-ray diffraction at low temperature.

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