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MAGNETIC TRANSITIONS IN $\text{CrAs}_{1-x}\text{Sb}_x$ ($x \geq 0.6$)

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Abstract. – Four magnetic phases were found by neutron diffraction measurements in $\text{CrAs}_{0.4}\text{Sb}_{0.6}$. The low temperature phase is the CrSb type antiferromagnet with the magnetic moment of $2.06 \pm 0.07 \mu_B / \text{Cr}$ being almost parallel to the *c*-axis. This low temperature phase transforms with a spin reorientation to the *c*-plane at $T_1 = 112$ K into the high temperature AF phase with $T_N(T_2) = 140$ K. The precursor magnetic short range order is observed up to $T_3 = 320$ K.

CrSb is a collinear antiferromagnet with the NiAs type crystal structure, and CrAs is an itinerant helimagnet with the MnP type structure. The mixed crystals $\text{CrAs}_{1-x}\text{Sb}_x$ ($x \leq 0.5$), which show the transition from the MnP type to the NiAs type, exhibit a double spiral spin structure like CrAs [1]. In the region of $0.6 \leq x \leq 1$, all compounds retain the NiAs type down to 4.2 K, and have been considered to take the CrSb type magnetic structure [1]. X-ray and magnetic measurements revealed that the two kinds of transitions with temperature occur in the Sb-rich compounds, and suggested that a magnetic state different from that of CrSb type comes up in higher temperature region [2]. In this paper, we studied the magnetic structure of these compounds by neutron and X-ray diffractions for $\text{CrAs}_{0.4}\text{Sb}_{0.6}$, and found the precursor phase of magnetic short range order and the spin reorientation just below the Néel temperature.

The samples were prepared by the procedure described in the previous work [2]. The X-ray powder patterns of $\text{CrAs}_{0.4}\text{Sb}_{0.6}$ at various temperatures show the NiAs type structure and two kinds of transitions on increasing temperature. As shown in figure 1, the first transition at $T_1 = 120$ K is accompanied with a discontinuous change of lattice parameters and next one at $T_3 = 330$ K is of the second kind. In both transitions, the *a*-axis is stretched and *c*-axis is contracted in cooling process. The similar results have been obtained for the other Sb-rich compounds. A small discontinuous change in the lattices is found even for CrSb itself.

Powder neutron diffraction experiments were performed at the temperature from 10 to 450 K, by using the KID diffractometer installed at JRR-2 reactor in JAERI. The incident neutron beam with the wavelength of 1.005 Å was obtained by the reflection of the PG(002). The typical powder diffraction pattern corresponding to each phase is shown in figure 2. All nuclear reflections are analysed by the NiAs type crystal structure. The magnetic reflection peaks, which are obtained by making the difference pattern, are indexed with the same unit cell as the chemical one and have

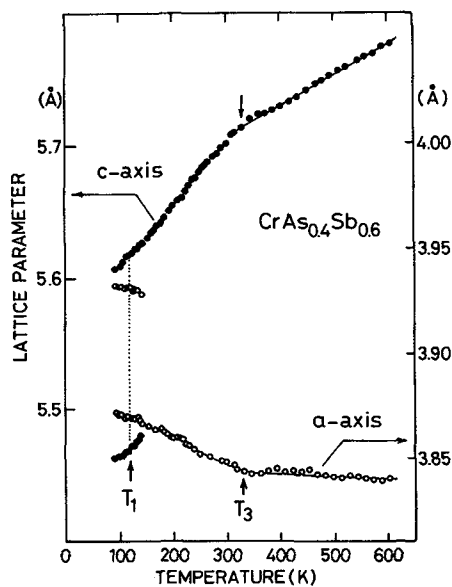


Fig. 1. – Temperature dependence of the lattice parameter of $\text{CrAs}_{0.4}\text{Sb}_{0.6}$.

odd ls. The magnetic scattering intensities at 10 K are almost explained by the CrSb type spin structure: the Cr magnetic moments along *c*-axis are coupled ferromagnetically in the *c*-plane and antiferromagnetically between successive layers. The magnetic moment of Cr ion is found to be $2.06 \pm 0.07 \mu_B$ by using a magnetic form factor of Cr^{+3} [3]. If we take notice of the very faint (001) magnetic reflection, the magnetic moment is estimated to be $\mu = 2.08 \mu_B / \text{Cr}$ with the inclination of less than 6° against the *c*-axis. The lower part of figure 3 shows that the very small (001) Bragg reflection becomes abruptly to the medium Bragg peak at around 90 K and then changes into a diffuse scattering after taking the maximum intensity at $T_1 = 112$ K. The measurements of this peak under slowly drifting temperature show the hysteresis of about 5 K for cooling and heating process. Then this spin reorientation is

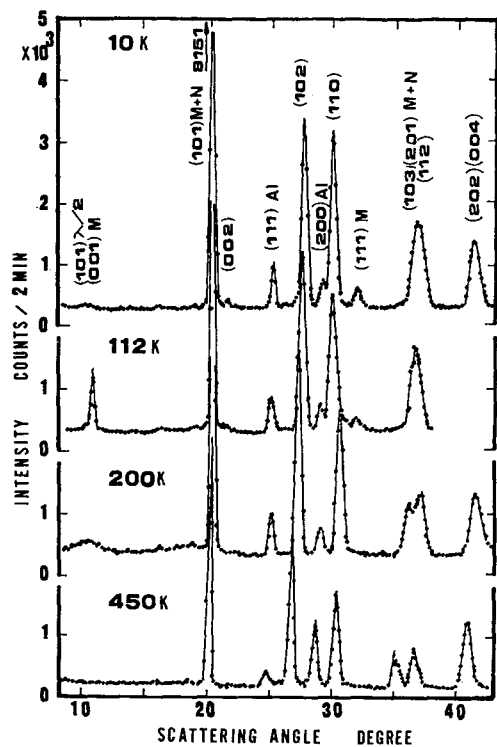


Fig. 2. - Typical powder neutron diffraction pattern corresponding to the low temperature state ($T < T_1 = 112$ K), the high temperature state ($T_1 \leq T \leq T_2$ (T_N) = 140 K), the precursor short range order state ($T_2 < T \leq T_3 = 320$ K) and paramagnetic state ($T > 320$ K).

considered to correspond to the first order transition of lattice parameters. The diffraction pattern of the high temperature AF phase at 112 K, shown in figure 2, can be analysed by a spin structure with antiparallel coupling of ferromagnetic c -plane along c -axis. The magnetic moment and its tilting angle from the c -axis at 112 K are about $1.5 \mu_B / \text{Cr ion}$ and 47° , respectively. The upper part of figure 3 shows the temperature dependence of the integrated intensity of (111) and the magnetic part of (101), both of which give the Néel temperature $T_N = T_2 = 140$ K. The diffuse scattering, obtained by a deconvolution of the magnetic scattering around the (001) Bragg angle assuming a Gaussian peak shape for the Bragg part, also gives the maximum integrated intensity at about $T_2 = 140$ K. This diffuse scattering might be attributed to such an anisotropic short range order of the high temperature AF that the ferromagnetic coupling in each c -plane is not broken so much but the antiferromagnetic stacking along the c -axis is cutted into the distributed lengths with temperature. The paramagnetic phase, as an usual mag-

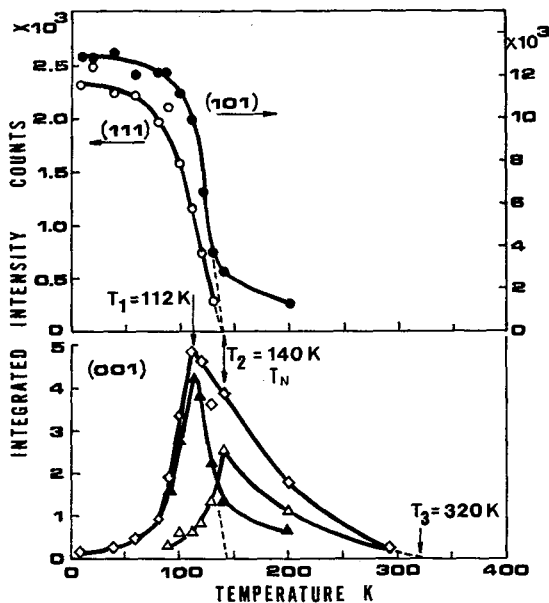


Fig. 3. - Temperature dependence of the intensity of the (111), (o) the magnetic part of the (101), (●) and the (001), (◇) which is separated into the Bragg scattering, (▲) and the diffuse scattering, (△).

netic disorder, is confirmed to be above $T_3 = 320$ K which correspond to be second kind transition point observed by X-ray.

The precursor state of magnetic short range order of $\text{CrAs}_{0.4}\text{Sb}_{0.6}$ is observed from 320 to 140 K and then it grows into the CrSb like long range order state with the spins largely inclined against the c -axis. And finally, this material becomes to the CrSb type antiferromagnet with the spins almost parallel to the c -axis, through the spin reorientation at 112 K. This spin reorientation may be due to a change in the magnetic anisotropy and the magnetic moment, because the great discontinuous change in the lattice parameters are observed. The behaviour of (001) Bragg peak is not found in the sample of $x = 0.7$ but in $x = 0.65$.

Acknowledgment

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