MAGNETIC PROPERTIES OF Cr2S3-xSex and CrS1.17-xSex SYSTEMS

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MAGNETIC PROPERTIES OF $Cr_2S_{3-x}Se_x$ and $CrS_{1.17-x}Se_x$ SYSTEMS

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Abstract. – It was found that the transition from ferrimagnetism to antiferromagnetism takes place at about $x = 1.8$ in $Cr_2S_{3-x}Se_x$ system and the slope of the concentration dependency of the magnetization changes at $x = 0.065$ in $CrS_{1.17-x}Se_x$ system and that the lattice parameters discontinuously changed at about the magnetic transition point.

1. Introduction

It has been known that Cr_2S_3 has a NiAs structure with an ordered arrangement of vacant chromium site and is ferrimagnetic with a Curie temperature of sbout 130 K and its spontaneous magnetization has broad maximum about 77 K and decreases during cooling and becomes constant [1]. On the other hand, Cr_2Se_3 also has the same crystal structure as Cr_2S_3 and is antiferromagnetic with a Neel temperature of about 43 K [2]. If the $Cr_2S_{3-x}Se_x$ system forms a solid solution, we can expect the transition from ferrimagnetism to antiferromagnetism at a certain value of x. Similar results are also expected in the $CrS_{1.17-x}Se_x$ system with a NiAs structure. It is known that $CrS_{1.17}$ is ferrimagnetic with a Curie temperature of about 307 K (T_c). Its spontaneous magnetization disappears suddenly during cooling at 153 K (T_t) and the compound becomes antiferromagnetic with a spiral configuration of spins [3]. Hence, in the present studies, magnetic and crystallographic analyses of these compounds were undertaken. In the following section the results of the measurements and a discussion of the magnetic transition are given.

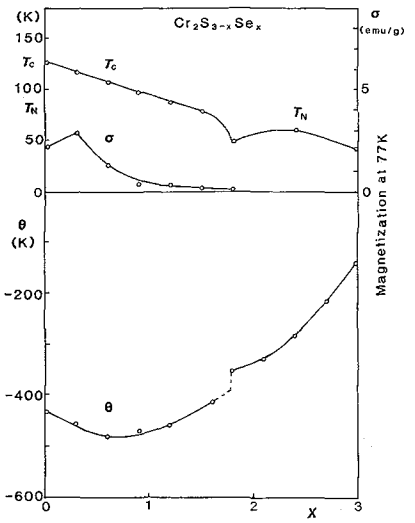

Fig. 1. – Curie and paramagnetic temperature, T_c and θ and magnetization at 77 K, σ, *versus* concentration in $Cr_2S_{3-x}Se_x$.

2. Experimental results and discussion

The specimens used were prepared by the usual ceramic method. The magnetic properties of the specimens were measured by means of a vibrating sample magnetometer. The results are shown in figures 1 and 2. As seen from the figures, it was found that the magnetic transition from ferrimagnetism to antiferromagnetism takes place at about $x = 1.8$ in a $Cr_2S_{3-x}Se_x$ system. The inverse susceptibility *versus* temperature plots of the $Cr_2S_{3-x}Se_x$ system are almost parallel for different x in high temperature for the prepared specimens. The Curie constant obtained from the slope of $1/\chi - T$ curve yields an effective magneton number per chromium ion of about 3.8. Assumung a Lande factor $g = 2$, the mean spin quantum number S is about 3/2 for each value of x. Therefore, the spins are considered to be canted and the canting angle changes with x in the ferrimagnetic range of x.

The X-ray data, obtained by means of a diffractometer at room temperature, are shown in figure 2. The

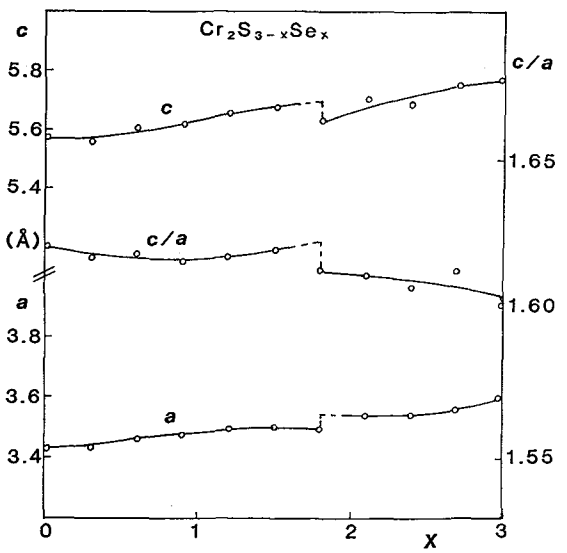

Fig. 2. – Lattice parameters *versus* concentration in $Cr_2S_{3-x}Se_x$.

hexagonal lattice parameters of the $Cr_2S_{3-x}Se_x$ system, a and c, increase monotonically with increasing x, but show a discontinuity at about $x = 1.8$ corresponding to the x value of the magnetic transition. Therefore the c/a ratio also shows a discontinuity at about $x = 1.8$. According to Kittel's [4] exchange inversion theory, it is considered that the crystallographic discontinuity causes the magnetic transition from Fr. state to A.F. one at $x = 1.8$.

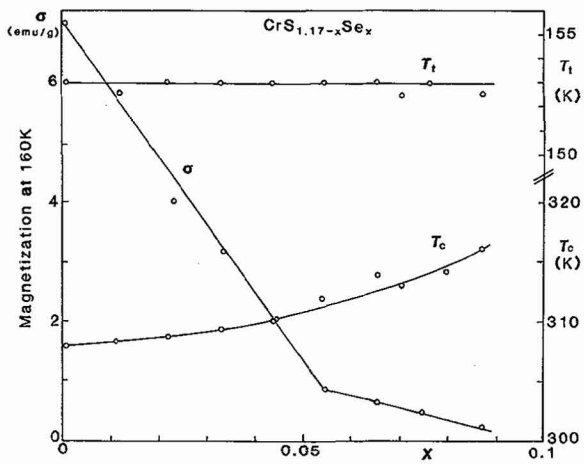

Fig. 3. – Curie and transition temperature, T_c and T_t and magnetization, σ, at 160 K in $CrS_{1.17-x}Se_x$.

On the other hand, the experimental results in the $CrS_{1.17-x}Se_x$ system are shown in figure 4. In this system, the specimens with $x = 0.1$ do not form solid solution. As shown from the figure, T_t is constant, T_c increases slightly, but the magnetization decreases with increase of x. The hexagonal lattice parameters of $CrS_{1.17-x}Se_x$ system, a and c, increase with increasing x and show a discontinuity at about $x = 0.065$. At $x = 0.065$, the slope of the σ *versus* χ curve changes. It

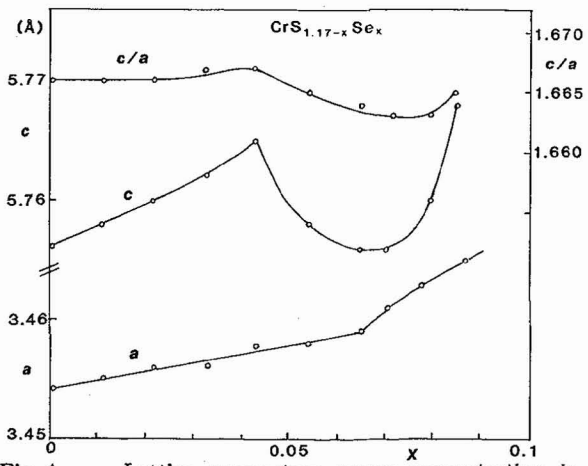

Fig. 4. – Lattice parameters *versus* concentration in $Cr_2S_{1.17-x}Se_x$.

is not clear for the lack of the data of the temperature dependence of inverse susceptibility or others, but the change of the slope might be associated with a change in the concentration dependency of the canting angle of the spins. The magnetic properties of $Cr_2S_{3-x}Se_x$ and $CrS_{1.17-x}Se_x$ systems are mainly explained by Hirone and Adachi's [5] theory related to the magnetic properties of substance with NiAs structure as shown in figure 5. According to this theory, three kinds of exchange interaction, J_1, J_2 and J_3 are applied in $Cr_2S_{3-x}Se_x$ system and the change of magnetic properties can be expressed by the change of J_2/J_1 and J_3/J_1 corresponding to the change of concentration of x. That is, previous results on the pressure effect on magnetic transition temperatures and exchange strictions for Cr chalcogenides [6] indicated that the exchange interactions in them are very sensitive to the lattice parameters. J_1, J_2 and J_3 change through the x dependence of a and c, since a, c and c/a change with x. More precise calculation will be done by application of exchange interactions, taking into account the chromium vacancies as in the studies of Hashimoto *et al.* [7] related to Cr_2Te_3 and Dwight *et al.* [3] related to Cr_5S_6.

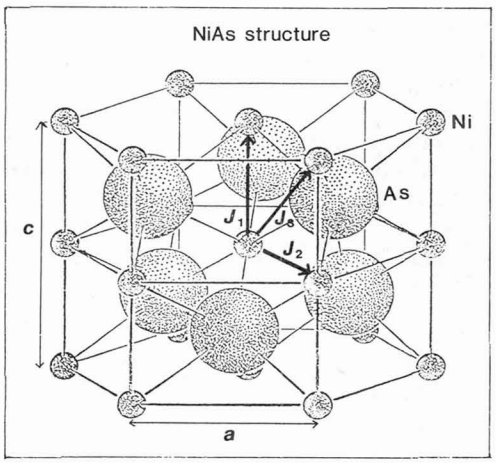

Fig. 5. – NiAs structure and exchange interaction J_1, J_2 and J_3.

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