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Neutron Diffraction Study of FeSn

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A neutron diffraction study on the powdered sample of FeSn has been made in order to determine the magnetic structure of this compound. The magnetic unit cell is twice as large as the chemical cell, being doubled along the c-axis. The moments of iron atoms are ferromagnetically coupled within a c-plane, while they are coupled antiferromagnetically to those on the adjacent c-planes. The moments lie in the c-plane. The atomic magnetic moment of Fe is obtained to be $1.5_5 \pm 0.1\ \mu_B$ at liquid nitrogen temperature.

§ 1. Introduction

The intermetallic compounds consisting of Fe and IVb group elements in the periodic table have been investigated by a number of workers by means of the Mössbauer effect and neutron diffraction as well as magnetic and electrical measurements.

One of the points of interest in these studies is the composition dependence of the Fe atomic moment μ_{Fe} in these compounds. In a given system the value of μ_{Fe} decreases with the increase of the concentration of the metalloid atoms, but the rate of decrease is different for different systems. For instance μ_{Fe} is zero in FeSi (B 20),[1,2] while it is 1.7 μ_B in FeGe (B 35).[3] So that it is interesting to know the value of μ_{Fe} in FeSn (B 35), which is isomorphous with FeGe.

Further the Mössbauer measurement of FeSn by Yamamoto[4] revealed that the quadrupole splitting was negative, while it was positive in FeGe.[5] Yamamoto has suggested that the spin axis of FeSn would lie in the c-plane provided that the direction of the maximum field gradient in FeSn is the same as in FeGe, because the spin axis of the latter compound is directed along the c-axis with the plus sign of the quadrupole splitting. The neutron diffraction study has been planned in order to bear out this suggestion.

§ 2. Experimental Procedures and Results

Iron powder (99.9 % purity) and tin pellet (99.999% purity) were mixed in the desired proportions, sealed in evacuated silica tubes and were sintered at 650°C for a week. The compound FeSn thus obtained was crushed into powder, sealed again in evacuated silica tubes and annealed at 600°C for 3 days and quenched in water.

X-ray and magnetic measurements were made prior to the neutron diffraction experiment. The Debye-Scherrer pattern showed that the substance was of the B 35 type structure, as had been reported by Ehret and Gurinsky.[6] The lattice constants, $a=5.28_8$Å and $c=4.44_2$Å, were in good agreement with their values.

In a preliminary magnetic measurement, an anomaly which was attributable to a very small quantity of an unknown ferromagnetic impurity with a Curie temperature at about 500°C was observed on the susceptibility *vs.* temperature ($\chi_g - T$) curve. This impurity was carefully removed with a small magnet, and then the magnetic measurement was tried again. The result obtained is shown in Fig. 1. Since the

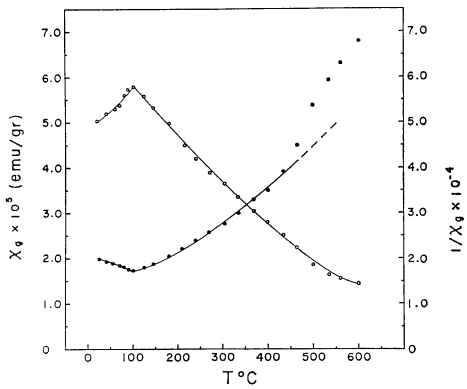

Fig. 1. Magnetic susceptibility (open circles) and its reciprocal (black circles) of FeSn as a function of temperature.

$\phi_g - T$ curve shows no anomaly near 500°C, it is considered that this impurity has been removed completely. But there is observed another anomaly on the $1/\chi_g - T$ curve above 500°C, and it is not known whether it is due to a phase transformation or to other impurities. It is to be

noted, however, that the impurity quantity, if any, was too small to make any appreciable influence on the X-ray and the neutron diffraction patterns.

The Néel temperature, the asymptotic Curie temperature and the effective Bohr magneton number are found from magnetic measurement to be 100°C, −158°K and 4.5 μ_B, respectively. These values are in qualitative agreement with Yamamoto's results.[4]

The neutron diffraction patterns of the powdered sample were taken with the TOG-ND diffractometer at JRR-3*. Monochromatic neutrons of wavelength 1.05Å obtained by a lead single crystal were used. The cassette was made of aluminum and had a specimen volume of a circular cylinder, 3 cm in diameter and 5 cm in length. The first to third collimators were set at 15′—∞—30′.

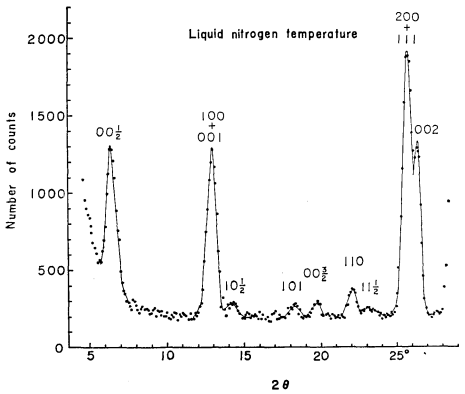

Fig. 2. Neutron diffraction pattern of FeSn at liquid nitrogen temperature.

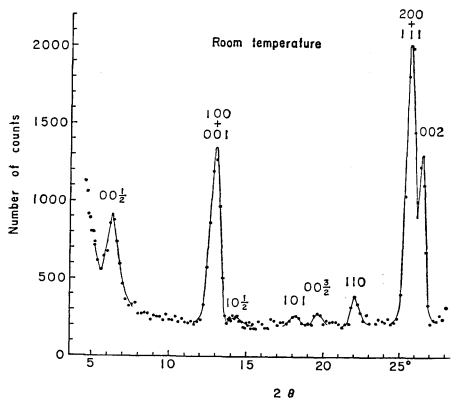

Fig. 3. Neutron diffraction pattern of FeSn at room temperature.

* Installed at the H-7 experimental hole of JRR-3, under the cooperation of Tohoku University, Osaka University and Japan Atomic Energy Research Institute.

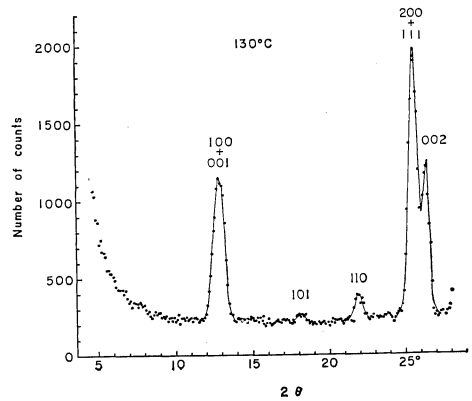

Fig. 4. Neutron diffraction pattern of FeSn at 130°C.

The measurements were made at liquid nitrogen temperature, room temperature and at 130°C, and the results are given in Figs. 2, 3 and 4, respectively. Four magnetic diffraction lines which could be indexed as $(00^1/_2)$, $(10^1/_2)$, $(00^3/_2)$ and $(11^1/_2)$ in terms of the original chemical unit cell were found. In particular, the $(00^1/_2)$ reflection was observed to be strong at liquid nitrogen temperature and became zero at 130°C, which is higher than the Néel temperature (100°C). The other magnetic diffraction lines also disappeared at 130°C their intensity decreasing with increasing temperature.

In the present neutron data, no diffraction lines which were to be attributed to contaminations, for instance $FeSn_2$, were observed.

§ 3. Analysis of the Experimental Data

The magnetic unit cell should be twice as large as the chemical cell, being doubled along the *c*-axis, As the magnetic $(00^1/_2)$ reflection was observed to be strong, it is quite evident that the magnetic moments of Fe atoms lie in the *c*-plane and are coupled ferromagnetically within a *c*-plane, while they are coupled antiferromagnetically to those on the adjacent *c*-planes.

The intensities of the $(00^1/_2)$ and $(00^3/_2)$ reflections are independent of the moment direction within a *c*-plane, while those of the $(10^1/_2)$ and $(11^1/_2)$ reflections depend on it. There are two possible moment directions (that is, the [100] and [210] directions) within a *c*-plane. The relative intensities have been calculated in each case of the two moment directions. The difference in the calculated intensities in these two cases, however, turned out to be small, and it was not possible to choose one or the other for the

moment direction because of the limited accuracy of the present measurement. On the assumption that the moment direction is along the [100], the antiferromagnetic structure of FeSn is illustrated in Fig. 5.

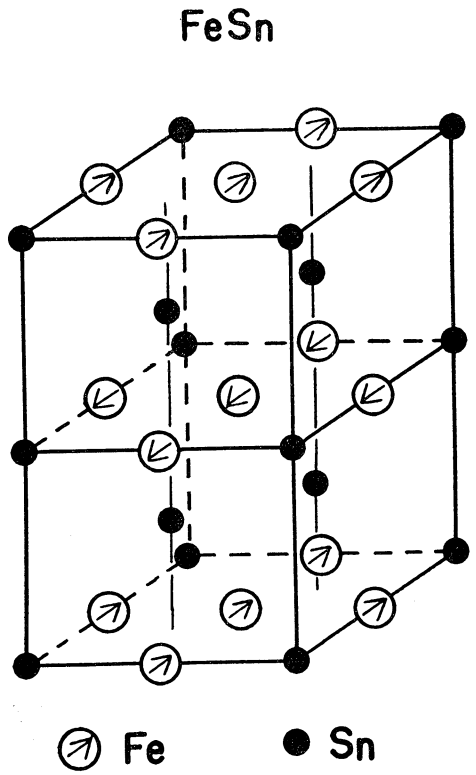

Fig. 5. The antiferromagnetic structure of FeSn. The c-axis is vertical.

The magnetic moment of an Fe atom was calculated from the intensity ratio of the $(00^1/_2)$ reflection to (100)+(001) reflection and using the spherical form factor of Fe due to Weiss and Freeman.[7] It turned out to be $\mu_{Fe}=1.5_5\pm0.1\ \mu_B$ at liquid nitrogen temperature.

The calculated intensities on the basis of the above assumption and the observed intensities are compared in Table I.

§ 4. Discussions

In the present study, it have been found that FeSn is antiferromagnetic with the layer magnetic structure having Fe moments in the c-plane. Watanabe and Kunitomi have shown that FeGe which is isomorphous with FeSn has an antiferromagnetic layer structure having moments along the c-axis. It is interesting to consider the difference between these two compounds.

Table I. The observed and calculated intensities of various reflections. All reflections are normalized in terms of the (100)+(001) reflection. The calculation has been made on the assumption that the moment direction is along the [100] and $\mu_{Fe}=1.5_5\ \mu_B$ at liquid nitrogen temperature and $\mu_{Fe}=1.1_6\ \mu_B$ at room temperature.

hkl	$2\theta°$	$\frac{I_{hkl}}{I_{100}+I_{001}}\times100$			
		cal.	obs.		
			liq. nit	room temp.	130°C
100+001	12.93°	100	100	100	100
101	18.29	2.6	4.8	5.0	4.8
110	22.07	10.0	13.6	12.2	13.3
200+111	25.59	132.0	137.6	127.4	141.7
002	26.29	70.0	73.7	78.0	76.3
		liq. nit. temp.		room temp.	
		cal.	obs.	cal.	obs.
$00^1/_2$	6.37	75.6	75.6	41.8	41.8
$10^1/_2$	14.33	5.2	5.4	2.9	2.6
$00^3/_2$	19.71	5.6	5.9	3.1	5.9
$11^1/_2$	23.17	1.7	3.0	0.9	0

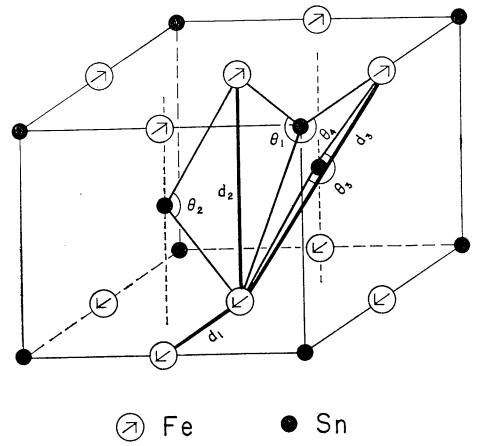

Fig. 6. The chemical unit cell of FeSn. Fe-Fe distances and the Fe-Sn-Fe angles are given as follows:
$d_1=2.64$ Å, $d_2=4.44$ Å, $d_3=5.17$ Å, $\theta_1=59°$, $\theta_2=111°$, $\theta_3=147°$ and $\theta_4=75°$.

The Fe-Fe distances and the Fe-Sn-Fe angles are illustrated in Fig. 6. The nearest neighbors arrange ferromagnetically with direct Fe-Fe coupling. The second and third neighbors are coupled antiferromagnetically due to the super-exchange interaction *via* intervening Sn atoms. As mentioned above the direct Fe-Fe interaction

is ferromagnetic, while the superexchange Fe-Sn-Fe interaction is antiferromagnetic. A similar conclusion has also been obtained for $FeSn_2$,[8] FeGe[3] and $FeGe_2$.[9~11]

The exchange interactions in FeSn are compared with that in FeGe. The molecular fields, $H^{\pm}$ acting on Fe atoms on + and − sublattices can be written in terms of the sublattice magnetization $M^{\pm}$, as

$$H^{\pm}=-(A_2+A_3)M^{\mp}-\Gamma M^{\pm}, \qquad (1)$$

where Γ, A_2 and A_3 are the molecular field coefficients associated with the first, second and the third neighbors (See Fig. 6). These molecular field coefficients are positive if the interaction is antiferromagnetic, while they are negative if the interaction is ferromagnetic. Putting $A=A_2+A_3$, we obtain

$$\left.\begin{aligned}\Gamma&=\frac{1}{C}(\theta_p-T_N)\ ,\\ \text{and}\qquad A&=\frac{1}{C}(\theta_p+T_N)\ ,\end{aligned}\right\} \qquad (2)$$

where C, θ_p and T_N are the Curie constant, asymptotic Curie and the Néel temperature, respectively. On substituting C (2.55 deg emu/mole), θ_p (−158°K) and T_N (373°K), Γ and A are found to be −208 mole/emu and 84 mole/emu, respectively. If we make a similar calculation of Γ and A values for FeGe with the aid of the experimental data in ref. 5), we find that the direct Fe-Fe interaction in FeGe is stronger than that in FeSn, and the superexchange Fe-Ge-Fe interaction is also stronger than the Fe-Sn-Fe interaction. This difference between the strength of the interactions in FeGe and FeSn may be attributable to the difference in the lattice constants of these compounds and also to the bonding nature of Ge and Sn atoms.

The magnetic moment of an Fe atom in FeSn is $1.5_5\pm 0.1\mu_B$ at 77°K from the present neutron diffraction study. The corresponding value in FeGe[3] was found to be $1.6_7\pm 0.1\mu_B$. On the other hand, the internal magnetic fields at Fe nuclei as obtained by means of the Mössbauer effect measurements in FeSn[4] and FeGe[5] were 163 kOe and 155 kOe at 79°K, respectively. Then the ratio of internal field to magnetic moment becomes 105 kOe/μ_B for FeSn and 93 kOe/μ_B for FeGe. Since the values of this ratio for other intermetallic compounds in these systems range from 100~180 kOe/μ_B, we find that the values obtained above for equiatomic compounds roughly correspond to the lower limit of this range.

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