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On the Neutron Diffraction Study of FeGe

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A powder neutron diffraction study has been made of FeGe. Published neutron diffraction data do not show any antiferromagnetic diffraction lines, while Mössbauer effect measurements do show that this substance is an antiferromagnetic with a Néel temperature at 400°K. The present study has revealed the presence of $(10\frac{1}{2})$ line, which is consistent with the Mössbauer data. The Fe spins on a c-plane are coupled ferromagnetically and directed along the c-axis, while they are coupled antiparallel to those on next Fe-containg c-planes.

§ 1. Introduction

Fe-Ge system has been studied extensively by many authors, and interesting results have been obtained with respect to the magnetic properties. There is, however, some inconsistency between various neutron diffraction data, or between neutron diffraction and the Mössbauer data. An example of this is found in the data on the equiatomic compound FeGe.

Ohoyama *et al.*[1] reported the results of their X-ray and magnetic measurement, from which they concluded that FeGe was of the hexagonal B35 type structure, with the lattice constants $a=5.00_3$Å and $c=4.05_3$Å; they suggested that the substance was an antiferromagnetic with a Néel temperature at 410°K: In a subsequent paper, Kanematsu *et al.*[2] revised the Néel temperature to be 400°K, although they found another susceptibility anomaly at 340°K.

Neutron diffraction studies of the system Fe-Ge, including FeGe, have been made at two places[3,4]; both of these studies revealed no antiferromagnetic Bragg reflections in FeGe above 140°K.

Mössbauer measurements have been made of FeGe by Tomiyoshi *et al.*[5] and Nikolaev *et al.*[6]; both of these studies showed a finite value of the internal field caused by the Zeeman splitting due to sublattice magnetizations at low temperatures. The observation of the temperature variation of the splitting located the Néel temperature at 400°K in the former study, while at 411°K in the latter study.

Although the above-mentioned neutron diffraction studies covered a wide range of scattering angles, they seemed to lack in sufficient accuracy to find possibly weak magnetic reflections. So we undertook to make a careful measurement, putting stress on scattering through relatively small angles.

§ 2. Experimental Procedures and Results

Iron powder (99.9% purity) and germanium powder (seven-nine purity) were mixed in the desired proportion, sealed in evacuated silica tubes and sintered at 700°C for a week. The product of this treatment was annealed at 650°C for a week and quenched in water.

The X-ray diffraction pattern showed that the substance was of the B35 type structure, as Ohoyama *et al.*[1] have reported. The lattice constants, $a=5.00$Å and $c=4.05$Å, were in agreement with their values.

The neutron diffractometer used was the JAERI diffractometer at JRR-2 (CP-5 type).[7] The cassette was made of aluminium and had a specimen volume of a circular cylinder, 2 cm in diameter and 5.5 cm in length. The measurements were made at liquid nitrogen temperature, room temperature and at 120°C. The first to third collimators were set at 15′–∞–30′. The wave length of the incident neutrons was very close to 1.00Å.

The results of the measurements are given in Figs. 1, 2 and 3. The main difference between the runs taken at various temperatures lies in the intensity of the small peak at 2θ (scattering angle)=15°. This line could be indexed to be $(10\frac{1}{2})$ in terms of the original chemical cell. The decrease in intensity of this line with rising temperature indicates that the line is of magnetic origin. The line at $2\theta=24°50'$ could be indexed as the (211) line of the contaminant $FeGe_2$ phase,

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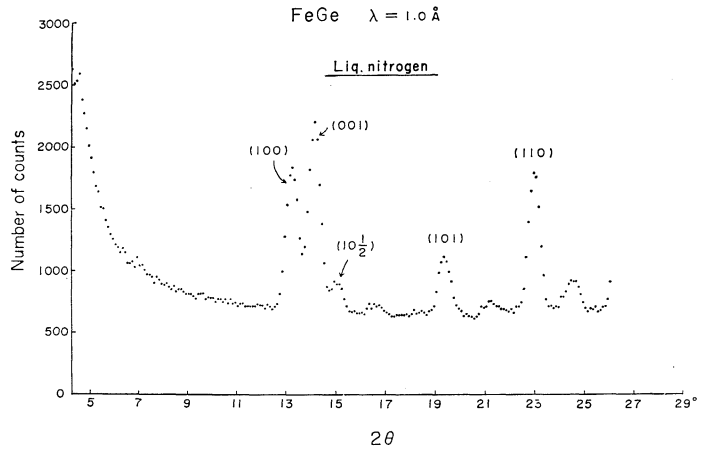

Fig. 1. Neutron diffraction pattern of FeGe at liquid nitrogen temperature.

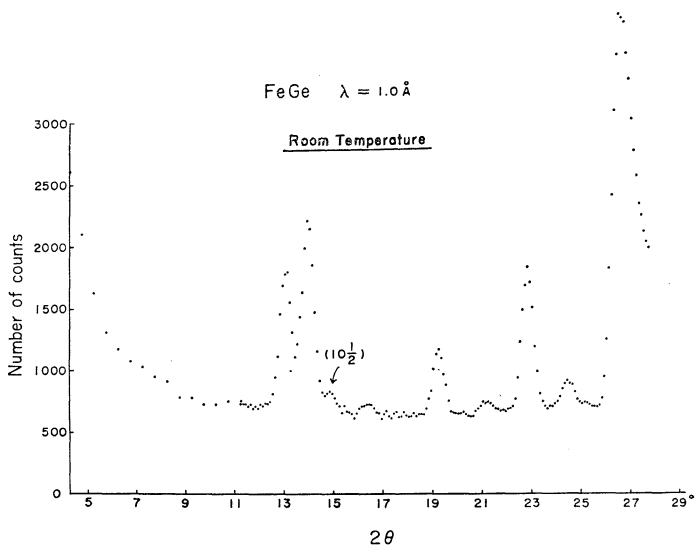

Fig. 2. Neutron niffraction pattern of FeGe at room temperature.

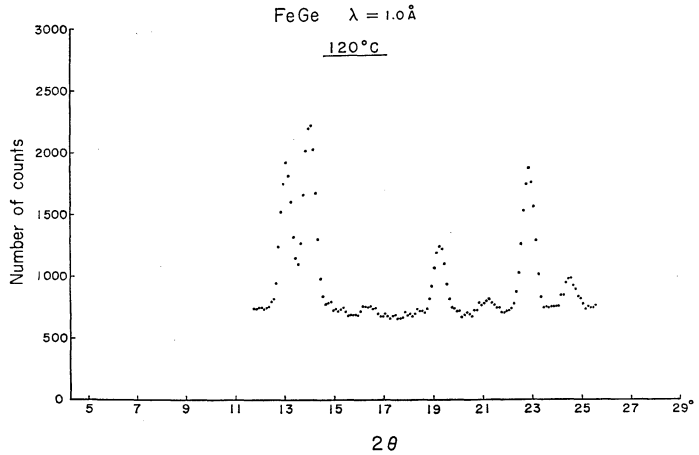

Fig. 3. Neutron diffraction pattern of FeGe at 120°C.

but the origin of the lines at 16°45′ and 21°25′ wer not easily known.

§ 3. Analysis of the Experimental Data

Although the magnetic line observed in the present study is only one, this is enough for determining the magnetic structure of FeGe. The magnetic cell should be twice as large as the chemical cell, being doubled along the c-axis. The simplest model consistent with the experimental data is one in which Fe moments on a given c-plane are coupled ferromagnetically, while they are coupled antiferromagnetically to the moments on the next Fe-containing c-planes (Fig. 4). Since $(00\frac{1}{2})$ line, which is to appear at

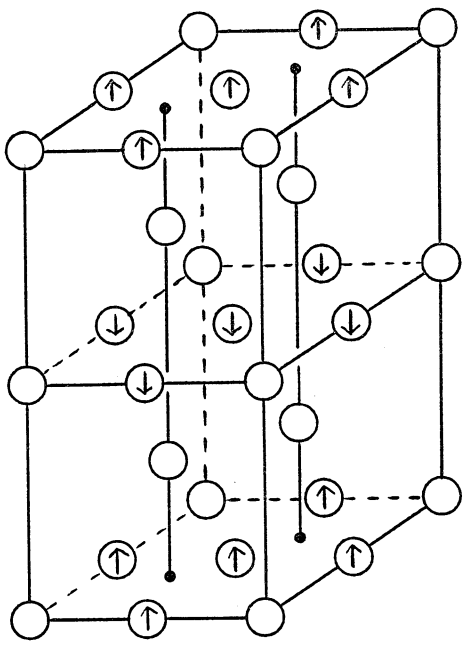

Fig. 4. Magnetic structure of FeGe. c-axis is vertical. Circles with arrows indicate Fe atoms with spin moments. Plain circles indicate Ge atoms.

$2\theta=7°$ provided the moments lie in the c-plane, is absent, the Fe moments should be directed along the c-axis. The value of the Fe moment was calculated on the basis of the above model and using the spherical form factor of iron due to Weiss and Freeman.[8] It turned out to be $\mu_{Fe}=1.67\pm0.05\mu_B$ at liquid nitrogen temperature, $1.28\pm0.04\mu_B$ at room temperature and 0.90 ± 0.03 at 120°C. The temperature dependence of μ_{Fe} is shown in Fig. 5. The value extrapolated to 0°K was then $1.70\pm0.05\mu_B$.

The relative intensities of various nuclear and magnetic lines are given in Table I. $(11\frac{1}{2})$ and $(10\frac{3}{2})$ lines are not observed, probably being hidden behind the contaminant $FeGe_2$ (211) peak.

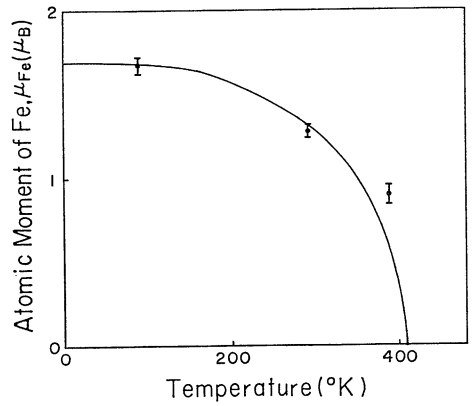

Fig. 5. Temperature dependence of the atomic moment of Fe, μ_{Fe}. The solid line gives the Brillouin function for $S=1.0$. The Néel temperature $T_N=410$°K is taken from the result of the Mössbauer effect measurement; cf. ref. 6.)

Table I. Relative intensities of various lines in FeGe.

hkl	cal.	obs.			2θ
		liq. nit.	room t.	120°C	
100	75.7	73.6	72.6	75.6	13°16′
001	100	100	100	100	14°10′
101	40.0	34.5	31.4		19°28′
110	67.6	78.4	75.2		23°4′
$10\frac{1}{2}$	15.0*	15.0	8.8	4.4	15°2′
$11\frac{1}{2}$	4.5*	not discernible			24°10′
$10\frac{3}{2}$	1.1*				25°11′

* Based on the proposed magnetic structure in Fig. 4, and assuming $\mu_{Fe}=1.67\mu_B$.

§ 4. Discussion

Contrary to the neutron diffraction data by other authors, the present study has verified that FeGe is antiferromagnetic, in conformity with the Mössbauer data. The reason for the difference from the previous studies may be in that the only observable magnetic line $(10\frac{1}{2})$ was of relatively small intensity, which could have escaped from observation with less precision.

The value of the Fe moment can be compared with that expected from the Mössbauer experiment. The internal field at liquid nitrogen tem-

perature was measured as 155 *kOe* by the Mössbauer study.[5] Since the empirical ratio of the internal field to the atomic moment is 120 $\sim$160 kOe/μ_B for a number of Fe-containing intermetallic compounds, the Fe moment per atom is expected to be 1.0$\sim$1.3μ_B. The value actually found from the present study, 1.7μ_B, is slightly higher than this expectation but not unreasonable.

The magnetic structure in this compound is understood by assuming ferromagnetic direct Fe-Fe interaction in the *c*-plane and antiferromagnetic superexchange Fe-Ge-Fe interaction between *c*-planes. The Fe-Fe distance in the *c*-plane is 2.50Å, which is close to the interatomic distance in metallic iron. Fe-Ge distance between adjacent *c*-planes is 2.4Å. The presence of superexchange interaction *via* Ge is a marked contrast to the absence of superexchange interaction via Si, in Fe-Si-Fe.[9–11]

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