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CHAPTER III

EXPERIMENTAL PROCEDURE

III.1 Sample Preparation

The ternary alloy $Ni_{1.55}Mn\ Ge_{0.45}$ was prepared from nickel 99.9 %, manganese 99.95 % and germanium 99.999 % purity. The appropriate proportions of nickel, manganese and germanium with atomic ratio of 1.55 : 1 : 0.45 were mixed together and melted in an alumina crucible under argon atmosphere at 1550 C. The furnace used in this experiment was made by Metal Research Corporation. The heating element of this furnace is a molybdenum wire which was worked under atmosphere of hydrogen gas to prevent the oxidation of heating element at high temperature. This furnace can go to temperature as high as 1800 C. The resulting ingot was crushed to powder and remelted twice in order to ensure good homogeneity of the alloy. In view of the insignificant loss, no analysis was made of the alloy. By using a hand magnet, it is found that this alloy is not a ferromagnetic material.

III. 2 Neutron Diffraction Study of $Ni_{1.55}Mn\ Ge_{0.45}$

Neutron diffraction study of $Ni_{1.55}Mn\ Ge_{0.45}$ was carried out at the Office of Atomic Energy for Peace, Bangkok. Neutron diffraction patterns of powder $Ni_{1.55}Mn\ Ge_{0.45}$ were obtained from the conventional double axis neutron spectrometer at T R R - 1 reactor. The neutron spectrometer was supplied by Bhabha Atomic Research Center, India. An Al (111) single crystal of the dimensions of 1 X 3 X 6 $in.^3$, was used as monochromator, to obtain a beam of neutrons with a wavelangth of 1.167 Å. The layout of the neutron spectrometer is showned in Figure 1. The block diagram of electronic equipment for measuring neutrons is shown in Figure 2.

Neutron diffraction patterns at various temperatures were obtained in order to determine the magnetic properties of the sample. The experiment was carried out for magnetic structure studies at room temperature. The cylindrical container made of thin aluminum foil was used so as to avoid the contamination of peaks of aluminum in the diffraction pattern. Neutron diffraction at elevated temperatures was done for determining the magnetic transition temperature of the sample. For experiments above the room temperature, the sample was sealed in an evacuated cylindrical quartz tube. The sample was heated in air by using aluminium foil as heat reflector. The temperatures between the top and the bottom part of the sample were different by about a few degrees Kelvin. Neutron diffraction at temperature above the transition temperature was also carried out in order to confirm the nuclear structure of this compound. Neutron diffraction pattern at low temperature was obtained by using a liquid nitrogen cryostat supplied by Bhabha Atomic Research Center. A copper cylindrical container was used in this experiment. For this part of the study, the temperature of the sample was 90 K. The diffraction pattern at low temperature was used for confirming the magnetic structure of the

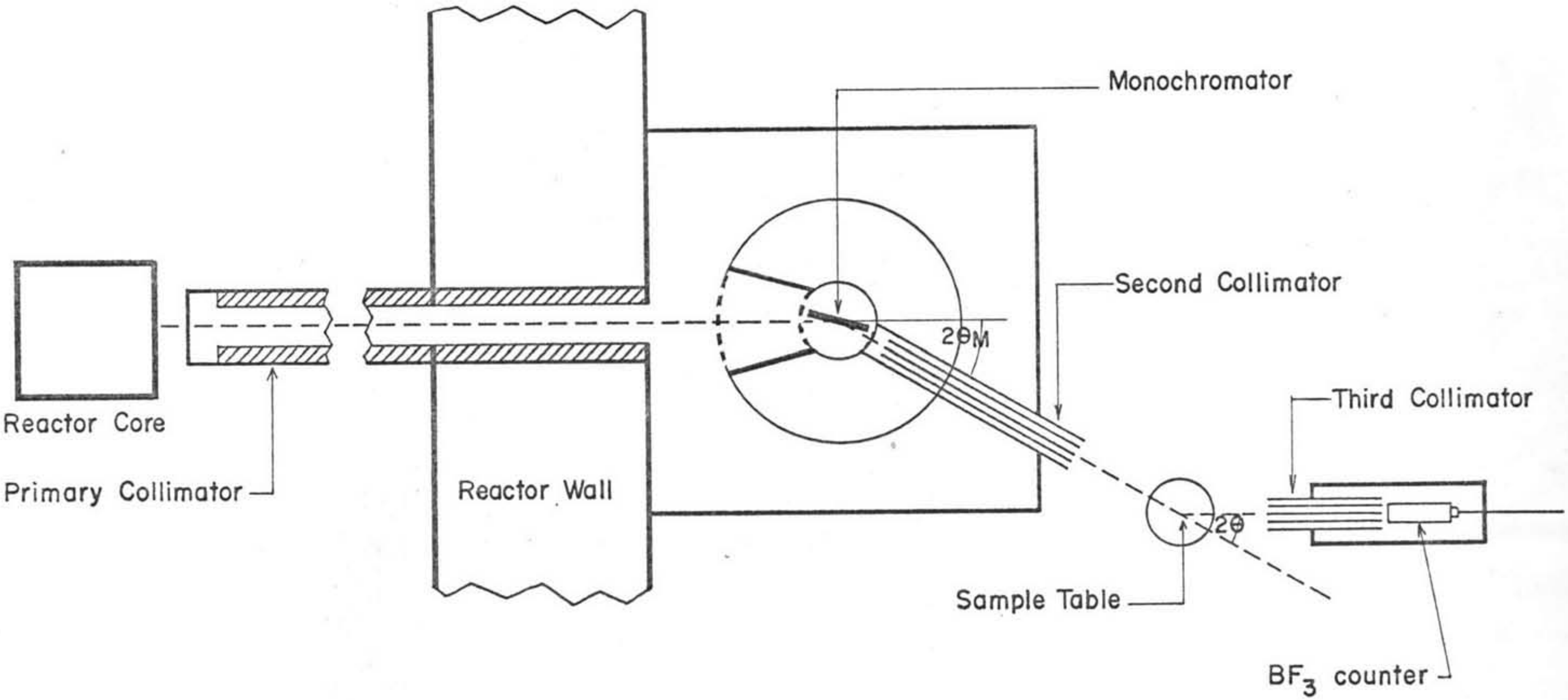

Fig. 1 Diagram of neutron spectrometer.

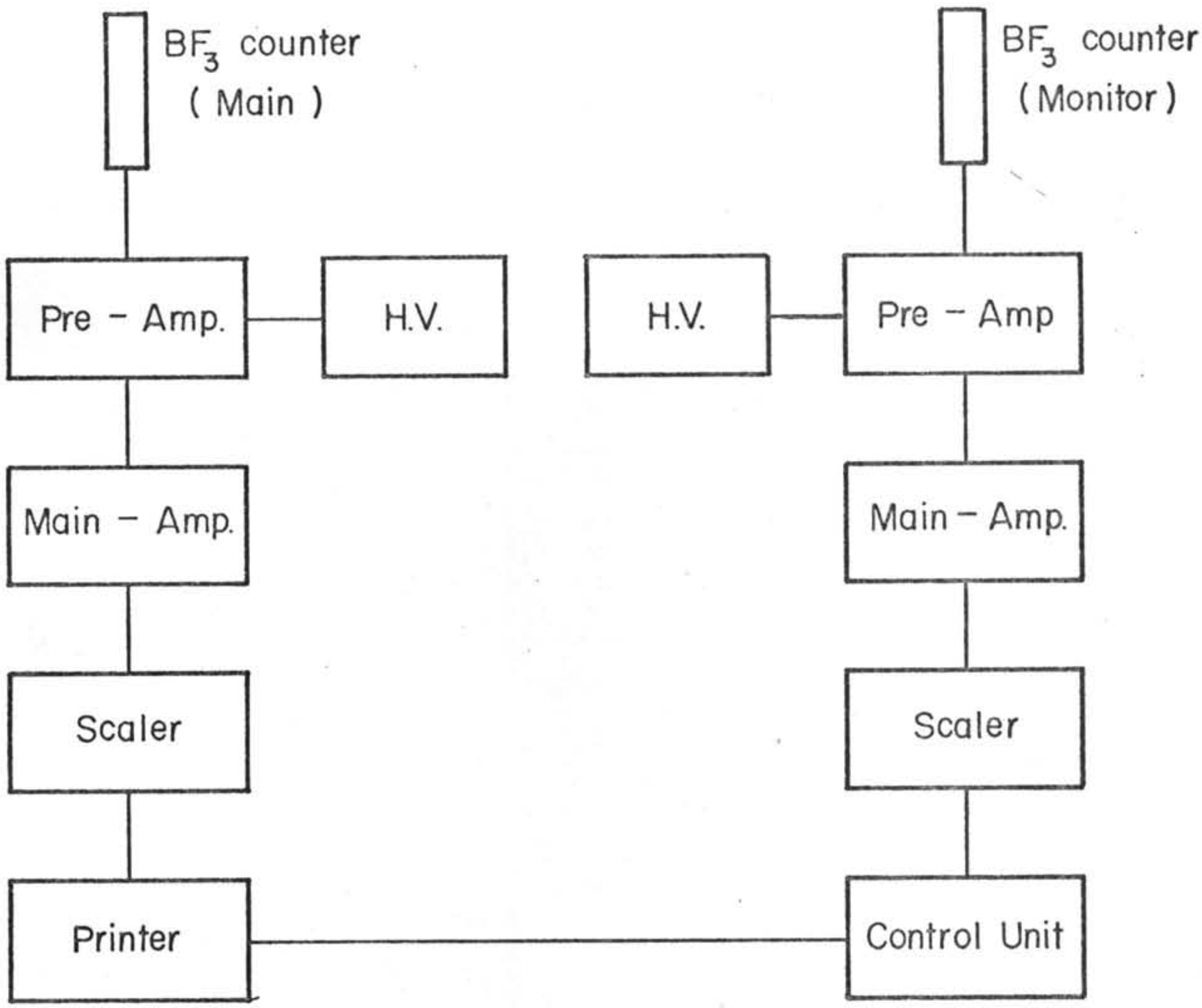

Fig. 2 Block diagram of electronics for neutron spectrometer.

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จุฬาลงกรณ์มหาวิทยาลัย

compound proposed at room temperature.

III.3 Neutron Diffraction Study at Room Temperature.

Neutron diffraction pattern of $Ni_{1.55}Mn\,Ge_{0.45}$ powder at room temperature is shown in Figure 3. The reflections obtained are indexed according to a cubic structure with lattic constant a = 6.762 Å and are in agreement with result found by Yu.V. Kuz'ma et.al[3]. Calculations of intensities of diffraction peaks were done, based on the Laves phase, $Mg\,Cu_2$ type structure in the manner described in Chapter II and III. In order to calculate the square of nuclear structure factor, we assumed that the 8 manganese atoms are located at A sites and the 16 atoms of nickel and germanium are located randomly at positions of the 16 B sites. The scattering amplitude of the B site is thus the average of the scattering amplitudes of nickel and germanium atoms. This scattering amplitude was found to be equal to 0.987×10^{-12} cm.

Since the nuclear structure factor of (200) plane of this compound is zero, the presence of (200) peak in neutron diffraction pattern at room temperature indicates that the compound is an antiferromagnetic material and this peak is a purely magnetic peak. In the calculation of magnetic intensities of various planes, the magnetic form factor of manganese reported by Takei et.al.[20] as shown in Figure 4, were used.

An analysis of the diffraction pattern obtained at room temperature shows that the 8 manganese atoms can be divided into C

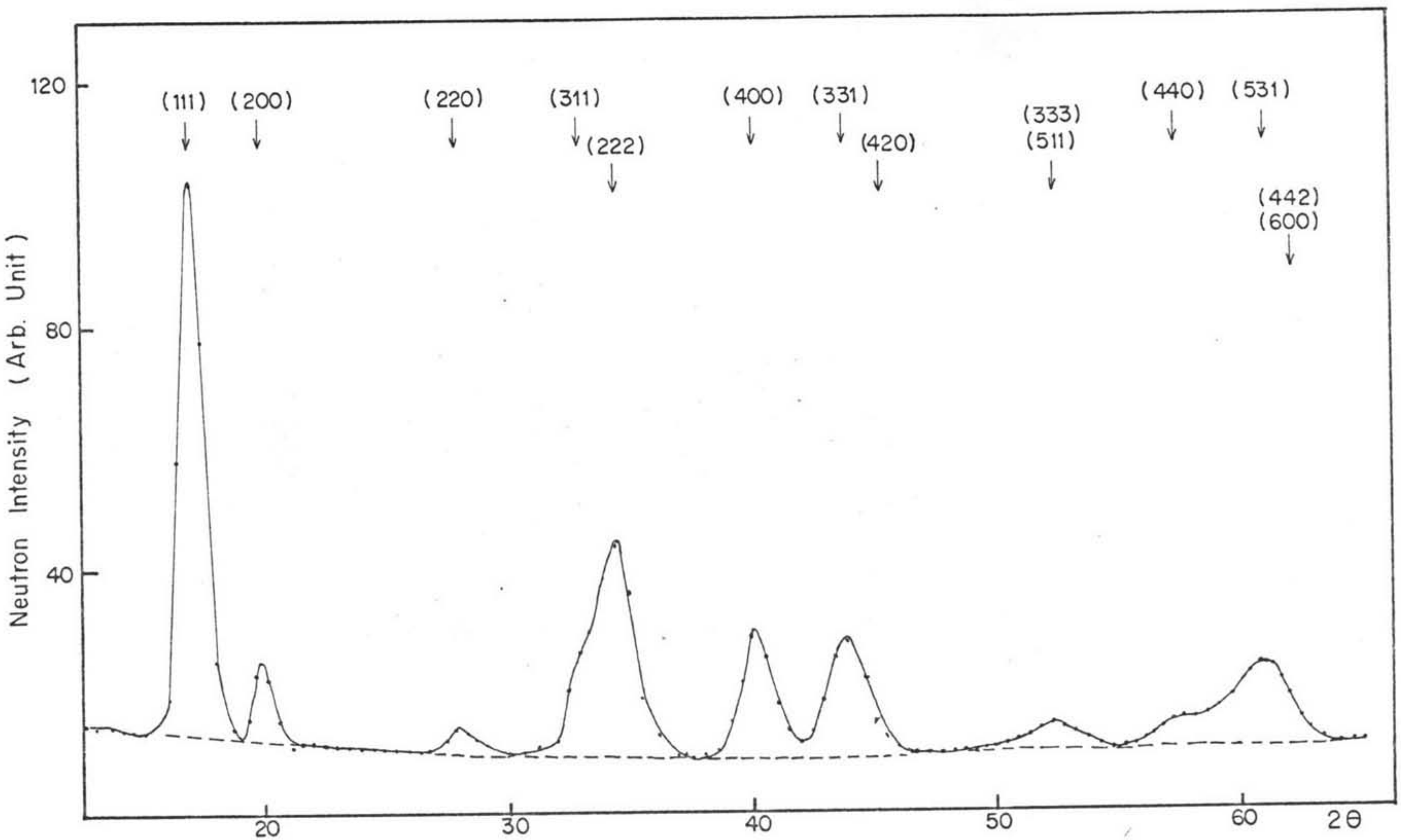

Fig. 3 Neutron diffraction pattern of $Ni_{1.55}MnGe_{0.45}$ at room temperature

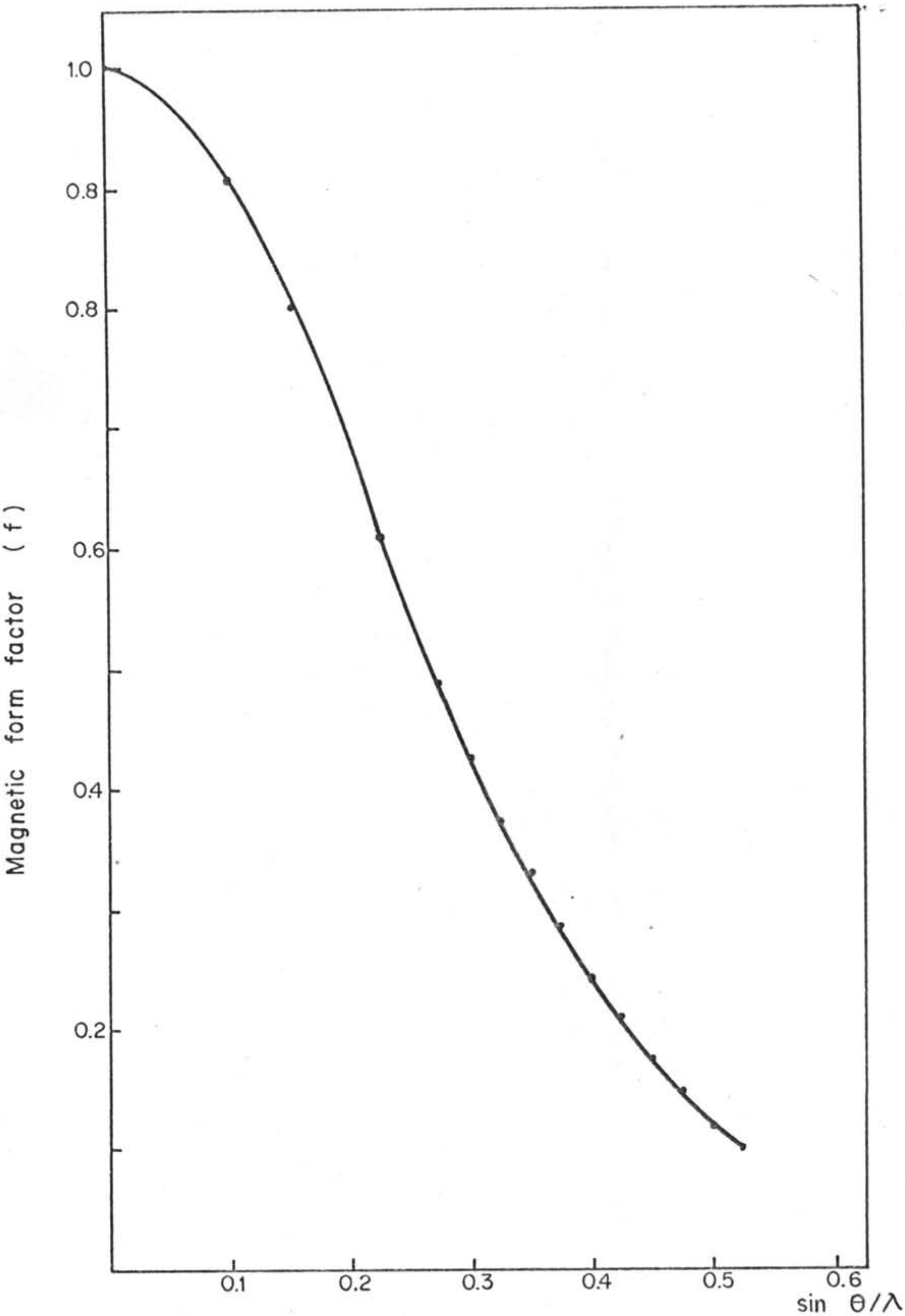

Fig. 4 Magnetic form factor of manganese.

and D sites:

C sites: $(0\ 0\ 0)\ (0\ \tfrac{1}{2}\ \tfrac{1}{2})\ (\tfrac{1}{2}\ 0\ \tfrac{1}{2})\ (\tfrac{1}{2}\ \tfrac{1}{2}\ 0)$

D sites: $(\tfrac{1}{4}\ \tfrac{1}{4}\ \tfrac{1}{4})\ (\tfrac{1}{4}\ \tfrac{3}{4}\ \tfrac{3}{4})\ (\tfrac{3}{4}\ \tfrac{1}{4}\ \tfrac{3}{4})\ (\tfrac{3}{4}\ \tfrac{3}{4}\ \tfrac{1}{4})$.

The manganese atoms in C sites are coupled ferromagnetically with each other but are coupled antiferromagnetically to manganese atoms in D sites. The magnetic unit cell has the same size as the chemical unit cell. It was found that the magnetic moment of each manganese atom of 3.6 Bohr magneton could be fitted to the observed relative intensities of the diffraction pattern. The calculated and observed relative intensities of $Ni_{1.55}Mn\,Ge_{0.45}$ *are shown in Table III. Temperature factors with the value B of* 1.7×10^{-16} *Å were included in the calculated relative intensities. The magnetic unit cell of* $Ni_{1.55}Mn\,Ge_{0.45}$ *is showned in Figure 5. It is clear that, the two nearest manganese atoms are coupled antiferromagnetically.*

III.4 Experiment at Low Temperature.

Neutron diffraction pattern at 90 K after correcting for the background of emptied copper container in liquid nitrogen cryostat is shown in Figure 6. It is obvious that the intensity of (200) plane at this temperature is higher than that of the room temperature. Neutron diffraction pattern at this temperature could be explained by the antiferromagnetic model with magnetic unit cell equals to chemical unit cell as found in part III 3. The magnetic moment of manganese atom was found to be 4 Bohr magneton. The agreement between the observed and calculated intensities is shown in Table IV. In

Table III Comparison between observed and calculated relative intensities of $Ni_{1.55}Mn\ Ge_{0.45}$ at room temperature.

hkl	$jL = j/\sin\theta \sin 2\theta$	F_n^2	jLF_n^2	F_m^2	$I = jL(F_n^2 + q^2F_m^2)e^{-W}$	$I_{calc.}$	$I_{obs.}$
111	181.24	98.69	17887	22.05	19416	279.0	279.6
200	102.33	0	0	39.58	2499	35.9	36.5
220	103.96	8.29	862	0	742	10.7	10.5
{311	153.91	34.36	5288	6.13	5081	214.0	222.5
{222	46.95	249.51	11714	16.95	9809		
400	26.85	348.79	9365	0	6959	100	100
{311	91.61	98.69	9041	2.9	6567	99.2	100.5
{420	89.90	0	0	8.08	334		
422	74.13	8.29	615	0	396	5.7	-
{333	22.27	34.36	765	2.2	482	27.7	25.7
{511	66.81	34.36	2296	2.2	1445		
{440	28.87	166.82	4816	0	2644	118.4	125.7
{531	107.16	98.69	10576	1.09	5554		
{600	13.09	0	0	2.02	9		
{442	52.35	0	0	2.02	6		

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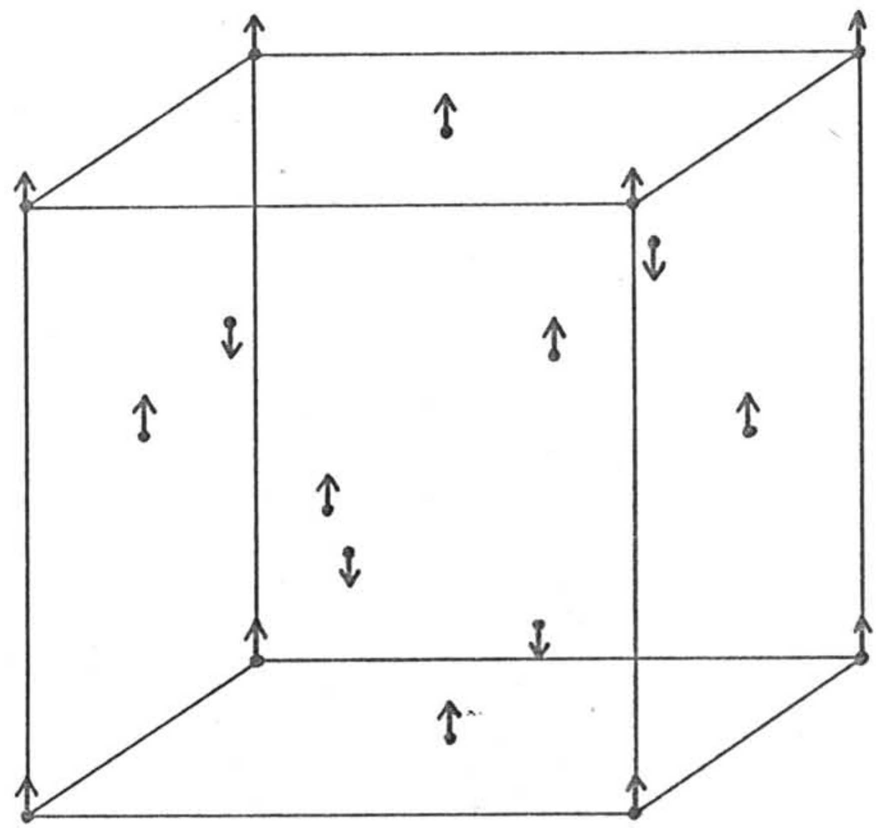

Fig. 5 The magnetic unit cell of $N_{1.55}MnGe_{0.45}$

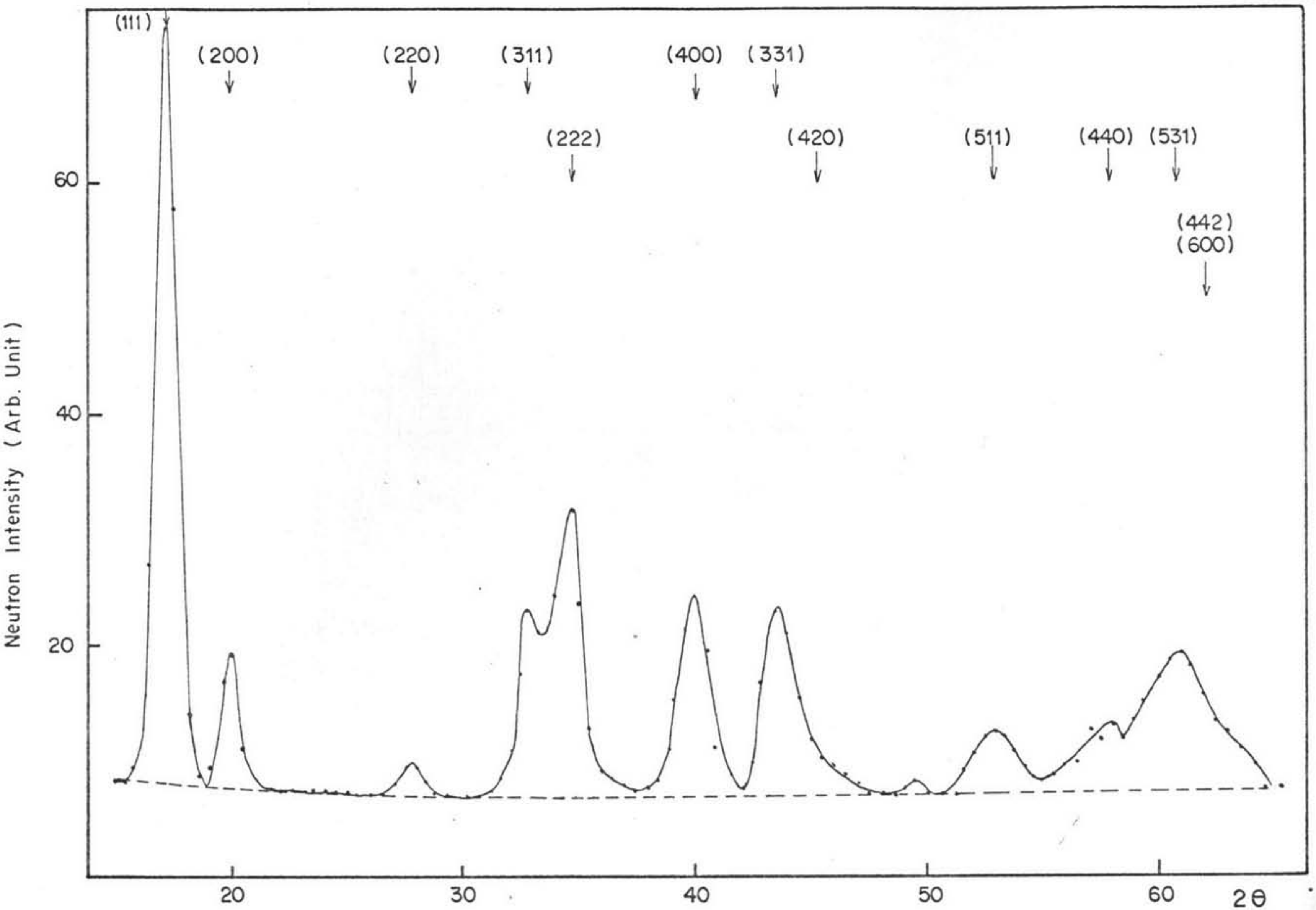

Fig. 6 Neutron diffraction pattern of $Ni_{1.55}MnGe_{0.45}$ at 90 K

Table IV Comparison between observed and calculated relative intensities of $Ni_{1.55}Mn\ Ge_{0.45}$ at 90 K

hkl	F_n^2	jLF_n^2	F_m^2	$jLq^2\ F_m^2$	$I = jL(F_n^2 + q^2F_m^2)e^{-2w}$	$I_{calc.}$	$I_{obs.}$
111	98.69	17887	27.22	3289	20755	246.1	240.6
200	0	0	46.87	3197	3114	36.9	38.7
220	8.29	862	0	0	818	9.7	10.7
{311	34.36	5288	11.35	1165	6003	206.7	197.8
{222	249.51	11714	20.92	655	11435		
400	348.79	9365	0	0	8434	100	100
{331	98.69	9041	5.38	329	8271	104.3	104.1
{420	0	0	9.98	598	525		
422	8.29	615	0	0	526	6.2	2.5
{333	34.36	765	2.71	40	674	32.0	44
{511	34.36	2296	2.71	121	2024		
{440	166.82	4816	0	0	3910	147.8	145.8
{531	98.69	10576	1.34	96	8472		
{600	0	0	2.49	22	17		
{442	0	0	2.49	87	69		

calculation of the intensity, temperature factors with the value B of 0.6 Å were included.

III.5 Neel Temperature of $Ni_{1.55}Mn\ Ge_{0.45}$ Determined by Neutron Diffraction Teachinque.

It is clear from parts III 3,and III 4that $Ni_{1.55}Mn\ Ge_{0.45}$ is an antiferromagnetic material with transition temperature or Neel temperature higher than room temperature. Neutron diffraction study of this compound at temperature above Neel temperature was carried out to confirm the nuclear structure. The neutron diffraction pattern at 573 K is shown in Figure 7. It is clear that the (200) peak has disappeared because the alloy becomes paramagnetic material at this temperature. The agreement between the observed and calculated intensities is shown in Table V.

In order to determine the Neel temperature of this alloy, neutron diffraction studies of (200) plane at various temperatures from 90 K up to 528 K were carried out. The intensity of this plane was found to decrease as the temperature increased. Intensities of (200) peak at various temperatures were plotted against temperatures as is shown in Figure 8. The extrapolation of this curve gave the Neel temperature of $Ni_{1.55}Mn\ Ge_{0.45}$ as 534 K.

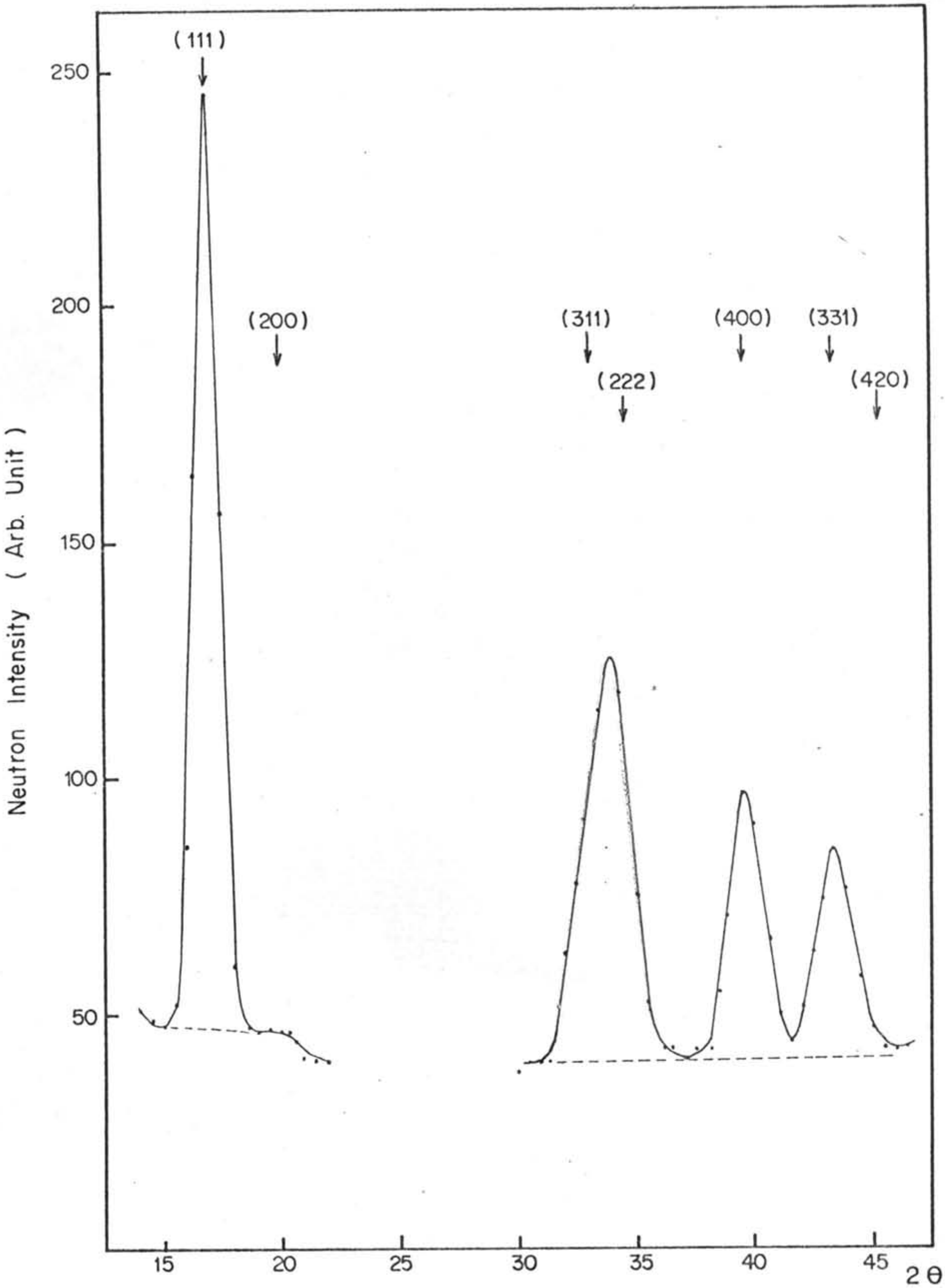

Fig. 7 Neutron diffraction pattern of $Ni_{1.55}MnGe_{0.45}$ at 573 K

Table V Comparison between observed and calculated relative intensities of $Ni_{1.55}Mn\ Ge_{0.45}$ at 573 K

hkl	$I_{obs.}$	$I_{calc.}$ (B = 2.4 A)
111	262.4	268.2
311 222	202.1	203.3
400	100	100
311	85.3	89.3

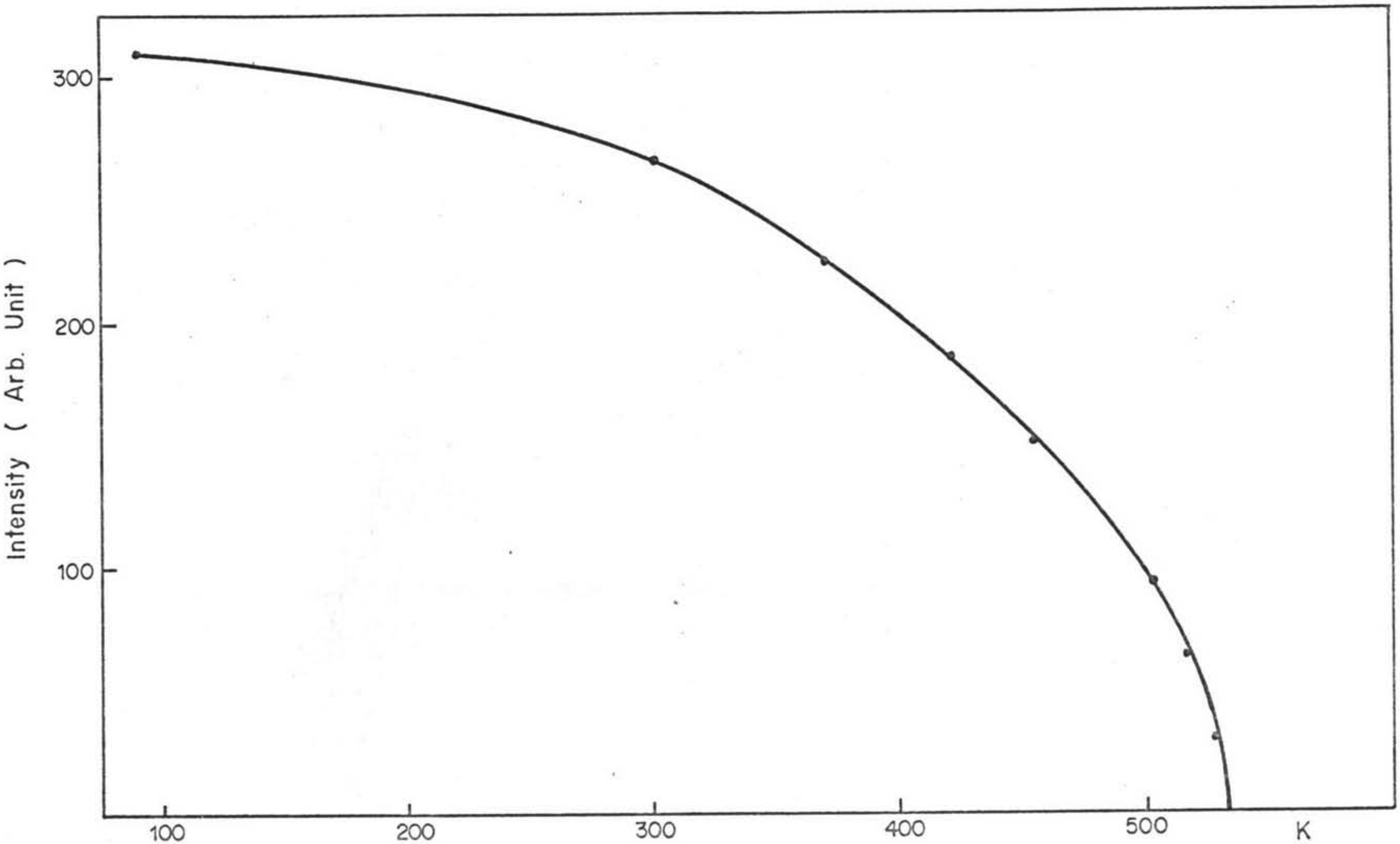

Fig. 8 Intensity of (200) plane at various temperatures.