International Atomic Energy Agency

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May 1980

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

Reproduced by the IAEA in Austria
May 1980
80-2414

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The nuclear scattering amplitude of uranium-235 isotope has been determined by the neutron diffraction method. Although the scattering cross section for slow neutrons of uranium-235 is very small (16.33 barns), the coherent scattering can be observed. The scattering amplitude of this isotope has been calculated from the measured diffraction intensities of the enriched metallic uranium as $b=(1.05\pm0.05)\times10^{-12}$cm. It may possibly give some more idea about the neutron-nucleus interactions of uranium-235 and some other elements which show the same scattering length for slow neutrons.

1. Introduction

In spite of the fact that coherent scattering amplitudes of neutrons have been measured for all of the elements, such measurements exist only for few individual isotopes. The importance of the study of the coherent scattering on uranium-235 and the determination of its amplitude arises from the fissionability of this nucleus compared to the stability and rigidity of the scattering nucleus of other elements under normal conditions. So it is important to know, besides the other factors, the magnitude of the scattered wave amplitude in the presence of the coherence and the nuclear scattering event of U-235 nucleus. Due to the fact that the resonance region is far from the low energy region for both the uranium-235 and uranium-238 isotopes, the scattering is mainly potential. The diffraction intensities represent the nuclear scattering only due to the paramagnetic behaviour of uranium element at room temperature, and so the magnetic contribution can not be observed on the diffraction intensities.

The nuclear coherent scattering depends on the spin of the compound nucleus, i.e. (i+1/2) and (i-1/2), the sign of the scattering amplitude and also the size of the nucleus and isotopic incoherence.

This paper is concerned with the measurement of coherent scattering amplitude of uranium-235 isotope by using 0.07 eV neutrons.

The experiments have been performed by the double crystal neutron diffractometer at TR-1, one megawatt reactor of

Çekmece Nuclear Research Center. The (111) plane of copper crystal has been used as neutron monochromator. The wavelength of the monoenergetic neutrons is 1.0824 A^{o}. During the experiment, several uranium samples with different enrichments have been used. All samples were metallic bars which show the powder character.

First, the neutron diffractometer had been calibrated with the natural powder nickel crystal.

As a control experiment, some measurements have been done with natural tungsten crystal in the powder form. The results obtained by this crystal are quite in agreement with those given in Ref.(1) and (2), (see Sec.3, Table II).

2. Theoretical Considerations

The coherent scattering amplitude of atoms can be expressed by means of the crystal structure factor[2] as

$$F^2_{hkl} = \left| \sum_j b_i e^{2\pi i (hx_j/a_o + ky_j/b_o + lz_j/c_o)} \right|^2 e^{-2W} \qquad (1)$$

where F_{hkl} is the amplitude of the diffracted neutron beam for the (hkl) reflection. a_o, b_o and c_o are the lattice parameters of the unit cell of the crystal and x_j, y_j, z_j the fractional coordinates of the atoms in the unit cell. e^{-2W} is the Debye-Waller factor. In Eq.(1) it has been assumed that the incident neutron beam has unit amplitude.

When the polycrystalline sample were used in an experiment, the most suitable geometrical shape for the sample is cylinder. The diameter of the cylinder must be smaller than the horizontal width of the monochromatic beam of neutrons.

In this case the number of neutrons P diffracted per minute into the detecting counter is given by[3]

$$\frac{P}{P_o} = \frac{\lambda^3 \, l \, V \rho'}{8\pi r \, S \, \rho} \times \frac{j \, N_c^2 \, F_{hkl}^2 \, e^{-2W} \, A_{hkl}}{\sin\theta \, \sin 2\theta} \qquad (2)$$

where P_o is the number of neutrons per minute in the incident monochromatic beam, λ the de Broglie wavelength of the neutrons, l the height of the counter slit, V the volume of the sample in the beam, r the distance from the specimen to the counter, ρ' the measured density of the specimen, ρ the theoretical density of the specimen, j the number of the cooperating planes for the particular reflection measured, N_c the number of unit cells per cm^3, A_{hkl} the absorption factor for the (hkl) planes, S the area of the sample in the beam and θ the glancing or Bragg angle of reflection. F_{hkl}^2 represents the geometrical part (first term in the right) of Eq.(1).

Comparison of the diffracted intensities of any substance containing nuclei of attested scattering amplitude give the simplest method of placing the intensities on an absolute scale. Under the same experimental conditions for the cylindrical specimens of standard dimensions the diffracted power is obtained from Eq.(2) as

$$P = \text{Cons.} \times \frac{\rho'}{\rho} \times \frac{j \, N_c^2 \, F_{hkl}^2 \, e^{-2W} \, A_{hkl}}{\sin\theta \, \sin 2\theta} \qquad (3)$$

Using Eq.(3) and Eq.(1) the structure factor F_{hkl}^2 and the average coherent scattering amplitude b_i, can be easily evaluated.

The amplitudes of the individual isotopes can be determined by using the system of equations[1]

$$\sum_j \alpha_{ij} b_j = b_i \qquad (4)$$

where b_i is the coherent scattering amplitude of the i-th mixture of isotopes and b_j is the coherent amplitude of the j-th isotope. α_{ij} is the content of the j-th isotope in the i-th mixture.

3. Experiment

From the diffraction intensities of enriched metallic uranium samples, the coherent scattering amplitude was determined.

Three differently enriched uranium and one natural uranium bars were used in the experiment*. The specifications of these samples are given in Table I.

Table I

Sample	Isotope				
	U-238	U-235	Wt.(gr)	Dia.(cm)	Heig.(cm)
Nat.Uran.	.9928	.0071	75.039	1.270	3.175
U-2	.90	.10	75.808	1.271	3.178
U-3	.60	.40	75.604	1.270	3.176
U-4	.20	.80	74.861	1.270	3.178

* The enriched samples were obtained from the Bureau Central de Measures Nucleaires through IAEA.

The natural nickel powder crystal has been used as standard. The coherent scattering amplitude of this substance is given[2] as $b=1.04x10^{-12}$cm. The constant in Eq.(3) had been determined by using this value.

In order to control the experimental conditions the coherent scattering amplitude of metallic powder tungsten in natural abundance has been measured. The value obtained for b_i is $(0.473 \quad 0.011)x10^{-12}$cm which is comparable with those of Ref. (1) and (2). Table II shows these values.

Table II

No	b_i(Nat.Tungsten)(cm)
Ref.(1)	$(0.4800 \pm 0.0025)x10^{-12}$
Ref.(2)	0.466 "
ÇNAEM	(0.473 ± 0.011) "

The ÇNAEM neutron diffraction equipment is given schematically in Fig.1. For this experiment, a special shaped shield has been designed and installed around the uranium samples. The shield was constructed from lead, borated-paraffine materials and cadmium plates.

Fig.1- The general layout of the experimental system.

In the experiment 0.07 eV energetic neutrons were used and the data were collected for the angles, $2\theta_B$, between 20° and 103° at the horizontal plane, where θ_B is the Bragg angle. The used proportional counter contains 96% enriched BF_3. Its sensitive length is 48.26 cm and the pressure 40.6 cm Hg.

A small proportional counter served as monitor. The monitor was placed in the monoenergetic neutron beam as seen in Fig.1.

All samples were put into the glass tubes in order to protect the reactor hall from the contamination of fission products.

The data obtained with the four uranium samples are reproduced in Fig.2. (a) represents the natural uranium diffraction data corresponding to 60 different (hkl) reflections. (b) shows the intensities obtained with the U-2 sample,one sees that the intensities are atenuated remarkably. (c) gives the U-3 sample's data,which shows that most of the diffraction peaks are disappered in the background. (d) shows the data of U-4 sample.Here only the most intense few reflections can be observed.

Fig.2- Diffraction data of the uranium samples.

Metallic uranium is in α phase at room temperature and has a base-centered orthorhombic crystal structure[4]. The lattice parameters of the unit cell are: a=2.854 A^o, b=5.869 A^o and c=4.955 A^o. In the diffraction data of uranium some of the extinctions are observed. These are for the (hkl) and (h0l) reflections when h+k and l are both odd. The space group for this crystal is Cmcm.

There are four uranium atoms in the unit cell in position c with a "y" value of approximately 0.105. The value of "y" is evaluated according to the lattice parameter. The final estimation for "y" values is given as 0.102 ± 0.002 by A.H.Cash et al.[5] and 0.105 ± 0.005 by Jacop and Warren[6].

Table III shows the measured and calculated all necessary characteristics of the uranium samples.

Table III

hkl	d (A°)	e^{-2W}	P_{obs}(Nat.)	P_{obs}(U-2)	$P_{cal.}$(U-2)	P_{obs}(U-3)	P_{obs}(U-4)
110	2.567	.975	st.		xb^2	-	-
021	2.525	.975	v st.	87.5	115.665	v w	v v w
002	2.476	.974	v st			v w	v v w
111	2.279	.967	st	25.5	33.834	v w	v w
022	1.279	.955	v w	v w	2.671	-	-
102	1.871	.954	v w	v w	1.871	-	-
112	1.783	.950	st	25.5	34.151	v w	v w
130	1.614	.939	v w	v w	4.951	-	-
131	1.534	.933	st	25.5	34.193	-	-
040	1.467	.926	m	v w	9.960	-	-
023	1.439	.924	m	w	23.441	-	v w
200	1.427	.922	m	m	12.271	v w	v w
041	1.407	.920	v w	v w	5.538	-	v w
113	1.389	.918	m	w	17.474	v w	-
132	1.352	.914	w	v w	6.957	-	-
220	1.283	.905	v w	v w	1.230	v w	-
042	1.262	.902	m			-	-
221	1.242	.899	m			w	w
004	1.239	.898	m	42.0	56.454	w	-
202	1.237	.898	m			-	v w
133	1.154	.884	w	m	27.166	v w	v w
024	1.141	.881	v w	v w	.975	-	-
222	1.140	.881	w	w	1.945	-	v w
114	1.116	.876	m	m	18.830	w	m
043	1.097	.872	v w	w	3.386	-	-
150	1.086	.870	w	w	13.955	-	-
151	1.060	.864	v w	-	-	-	-
240	1.023	.855	v w	v w	9.793	-	-
223	1.013	.852	m	m	23.486	-	w
152	.994	.847	m	m	23.555	-	w

134	.983	.844	w	-	-	v w	v w
060	.978	.842	w	w	2.740	-	-
061	.960	.837	w	-	-	-	-
242	.946	.832	m	m	16.851	w	-
310	.939	.830	m	m	6.763	-	-
204	.935	.829	m	m	10.757	w	w
311	.923	.825	w	w	7.880	-	-
062	.910	.820	w	v w	4.786	-	-
153	.907	.819	v w	v w	.497	-	-
312	.878	.808	v w	v w	11.949	-	-
243	.870	.805	v w	v w	4.367	w	v w
330	.856	.799	v w	-	1.440	-	-
063	.842	.793	m	st	4.714	w	-
006	.826	.786	v w	v v w	4.292	w	-
045	.821	.784	v w	w	1.973	-	-
260	.807	.777	w	w	3.865	-	-
116	.786	.766	m	st	9.857	-	-
225	.784	.766	m	st	14.775	w	w
172	.765	.755	v w	w	.134	-	-
333	.760	.752	v w	m	12.600	w	w
314	.748	.745	v w	-	-	-	w
350	.739	.740	v w	m	7.003	-	w
155	.732	.736	v w	w	.347	-	-
351	.731	.735	v w	w	.746	-	-
263	.725	.731	v w	m	7.422	w	-
046	.720	.728	v w	st	5.313	w	w
206	.715	.724	v w	st	6.858	-	w
400	.714	.724	v w	-	-	-	v w
245	.712	.723	v w	-	-	-	-

Note: The intensities of maximums have been compared with 200 cpm and 20 cpm for natural uranium and enriched samples respectively.

Table III,column IV shows the observed (hkl) intensities for natural uranium. Column III gives the Debye-Waller factors for each reflection.

Since there are too many diffraction maximums of metallic uranium in the angular interval considered,most of them were overlapping and it was not possible to determine the individual diffraction intensities precisely. On the other hand it could not be possible to determine the coherent scattering amplitude for U-235 isotope due to the abundance of this isotope in natural uranium,so it stays in the error limits. But, it made possible to observe the probable reflections which are expected from the metallic uranium structure and to get an approximate idea about their intensities.

Column V gives the observed intensities for the U-2 sample (10 % enriched). It was only possible to determine the intensities for several strong reflections for this sample numerically. From these intensities the coherent scattering amplitude was evaluated by means of Eq.(3). But unfortunately the experimental statistics is rather bad so the errors are large*.

Column VI shows the calculated P values for U-2 sample. Column VII shows the intensities corresponding to the U-3 sample (40 % enriched). As seen from the pattern in Fig.2, the fission event is dominant and many of the maxima have disappeared in the in the background. The observed intensities are so weak that no interpretation can be done satisfactorily.

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* The duration of the measurements were short due to the shut-down of the TR-1 Reactor which was to be extended.

Column VIII gives the intensities of the U-4 sample (80% enriched). Here the intensities are much weaker than those of the U-3 sample. Some small maxima emerge from the diffuse scattering pattern.

In the calculations of Debye-Waller factors, the Debye temperature has been taken as $\Theta=207^{o}K$[7]. The absorption term A_{hkl} is an other important factor, in particular for U-235. This factor has been determined through the method given by Bacon[2] for cylindrical samples. μR values were obtained by the transmission measurements. For these measurements the width of the cadmium slits was set to 0.15 cm at the two ends of the collimator put between the monochromator and the sample. Under these conditions the transmission through the specimen is equal to $e^{-2\mu R}$. From this quantity μR was determined. Glass tube corrections was done also.

From the measured value of μR, the absorption factor A_{hkl} have been determined by the aid of the Table 10, in Ref.(2). Since the variations of A_{hkl} with the Θ_B angle are small in the neutron case, a constant value for A_{hkl} was taken when Θ_B is between 0° and 30°.

Table IV shows the absorption coefficients and the interpolated A_{hkl} values for natural uranium and U-2 samples.

Table IV

Sample	μ (cm^{-1})	μR	A_{hkl}
Nat.Uran.	0.5285	0.3356	0.575
U-2	1.9659	1.2483	0.115

In spite of the fact that the used neutron counters were not too much sensitive against gamma rays and fast neutrons produced by fission in the samples, the background was high in the case of natural uranium sample. In the analysis of the data the background and the statistical fluctuations were considered too.

The increasing values of the background at the large angles as seen in Fig.2, are due to the effects of the laboratory background in that positions. In particular it increases remarkable after $\theta_B=25^o$. The incoherent (diffuse) scattering from U-235 is isotropic since it is a heavy nuclide.

Finally, the experimental values, P_{obs}, were compared with the values calculated from Eq.(2) or Eq.(3). This comparison gives the average values of b_i. By Eq.(4), and taking $0.85x10^{-12}$cm for the b_j value of U-238, the coherent scattering amplitude of U-235 was calculated for the most resolved intensities of the U-2 sample (Table III,column VI). The least-square method is applied to the b_j's in Table III. Fig.3, shows the graph of the b_j values. Thus the coherent scattering amplitude was evaluated for the U-235 isotope in the experimental error limits as $b_j=(1.05\pm0.05)x10^{-12}$cm.

Fig.3- Experimental results for b_j of uranium-235.

4. Experimental Results and Discussion

The measurements of coherent scattering amplitude for uranium was made in UO_2 by Willis[8] previously.

The coherent scattering amplitude and it's magnitude is important for uranium element and in particular for U-235 isotope, because it gives more information about the scattering condition of U-235 atoms in the unit cell of the uranium crystal. Although the distribution of isotope atoms among the sites in the unit cell is accepted as randomly, it may be worthwhile to study about it in order to get more information on this matter. On the other hand, the observation of coherent scattering for U-235 give some idea about the nuclear spin dependence also.

The experiment shows that there is a very weak coherent scattering of U-235 isotope of positive sign. The value of the coherent scattering amplitude b_j is $(1.05 \pm 0.05)\times 10^{-12}$ cm.

The spin of the U-235 nucleus is 7/2 and has rondom alignment at room temperature. The scattering seems strongly spin dependent and gives the spin incoherent scattering mostly. The compound nucleus formed with the slow neutron has two spin states as mentioned in Sec.1. The scattered neutrons can have either spin states which give two different scattering amplitudes. If the spin dependence is strong the unaligned nucleus spin has resulted a very weak coherent scattering. In the uranium case the number of slow neutrons incident is decreased due to the fission of the U-235 atoms in the sample.

The magnitude of scattering amplitude is almost unity for uranium-235.For other elements whose scattering cross sections for slow neutrons are 12-25 barns,the coherent scattering amplitude has about the same magnitude.

One onother result of the experiment is that some of the diffraction intensities show that the atoms doesn't keep the random distribution of the isotopes among the atomic sites in the unit cell even in the poorly enriched U-2 (10% U-235) sample. It is possible to see this at the (110) reflection of U-2 sample. It was expected that this reflection should be stronger than it is. Some other reflections may have same conditions but it is difficult to make an interpretation with this experimental statistics.

The other thing which attracts the attention is that some of the (hkl) reflection intensities obtained with the enriched uranium sample U-2 are stronger than expected.This can be interpreted by the existence of some preferred orientations in the sample or by the fact that the distribution of the isotopes in the unit cell random.

These important points are worthwhile to be investigated with suitable enriched uranium samples for the longer experimental periods.

The author would like to express her most gratefull thanks to Prof.Dr.F.Yeniçay, Prof.Dr.A.Y.Özemre and Dr.U.Adalıoğlu for their kind discussions and valuable critisisms on the minuscript. Thanks are also due to the technical personnel who helped me during the experiments and computer programmings.

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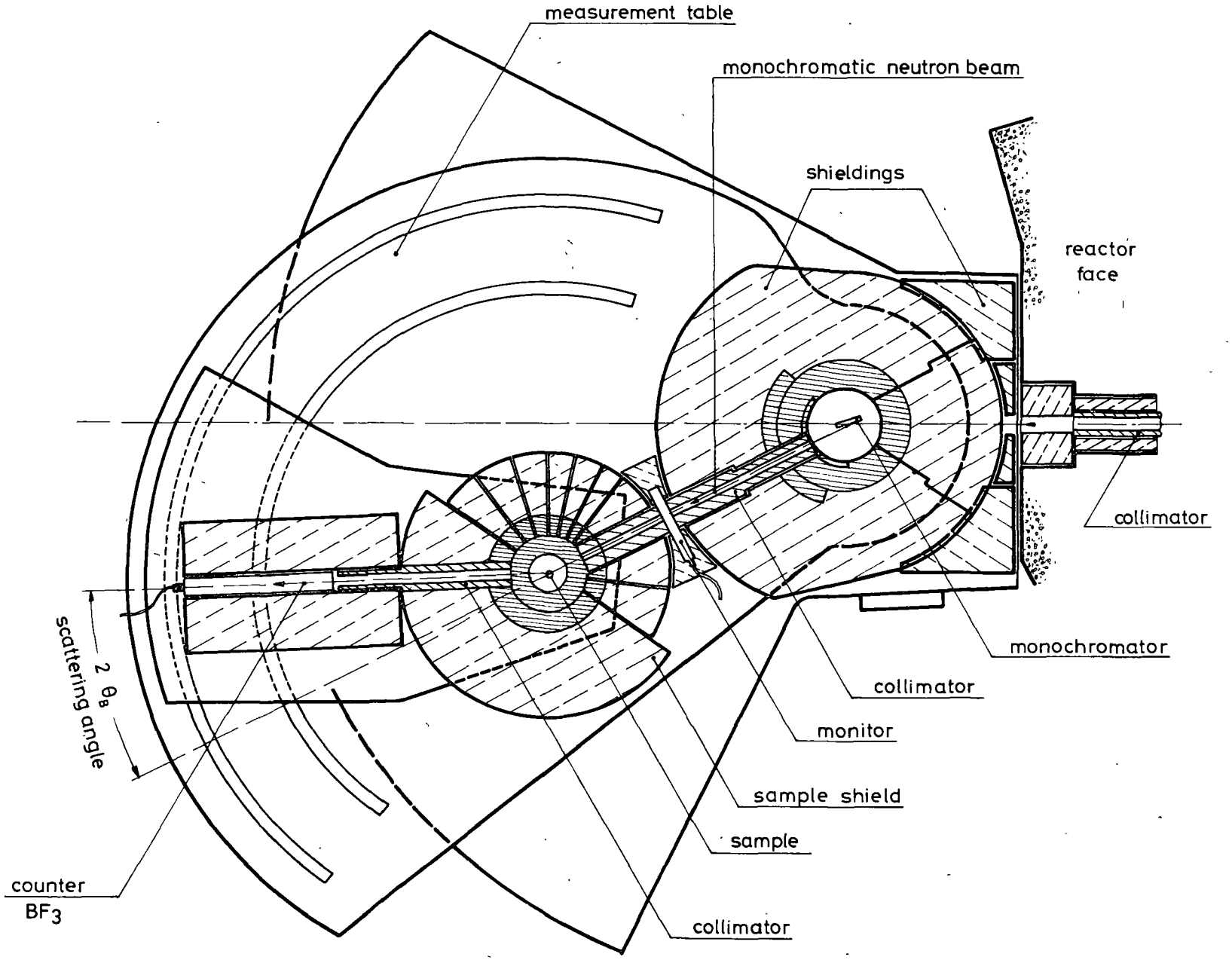
measurement table
monochromatic neutron beam
shieldings
reactor face
collimator
monochromator
collimator
monitor
sample shield
sample
collimator
counter
BF_3
2 θ_B
scattering angle

Fig. 1

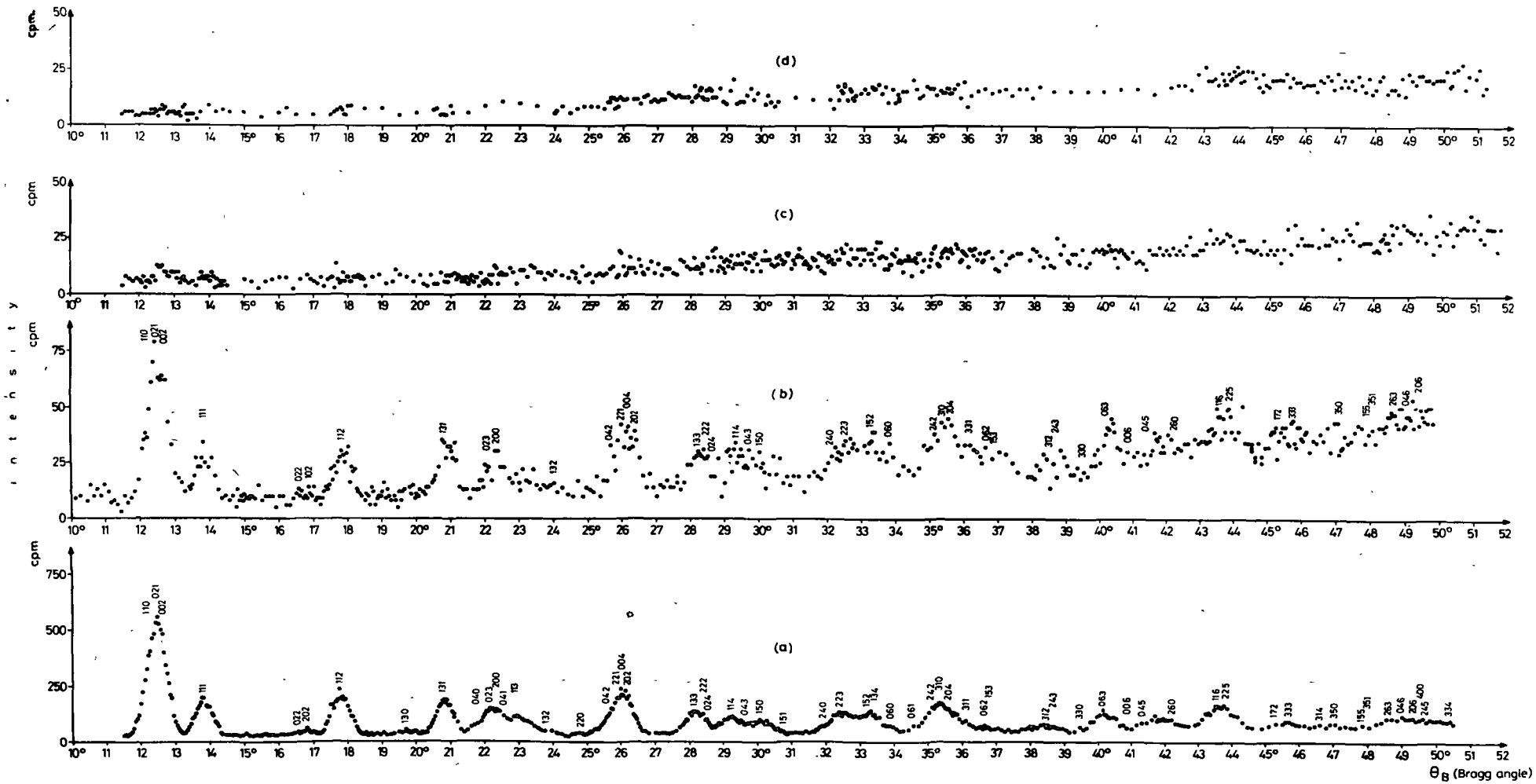

Fig. 2

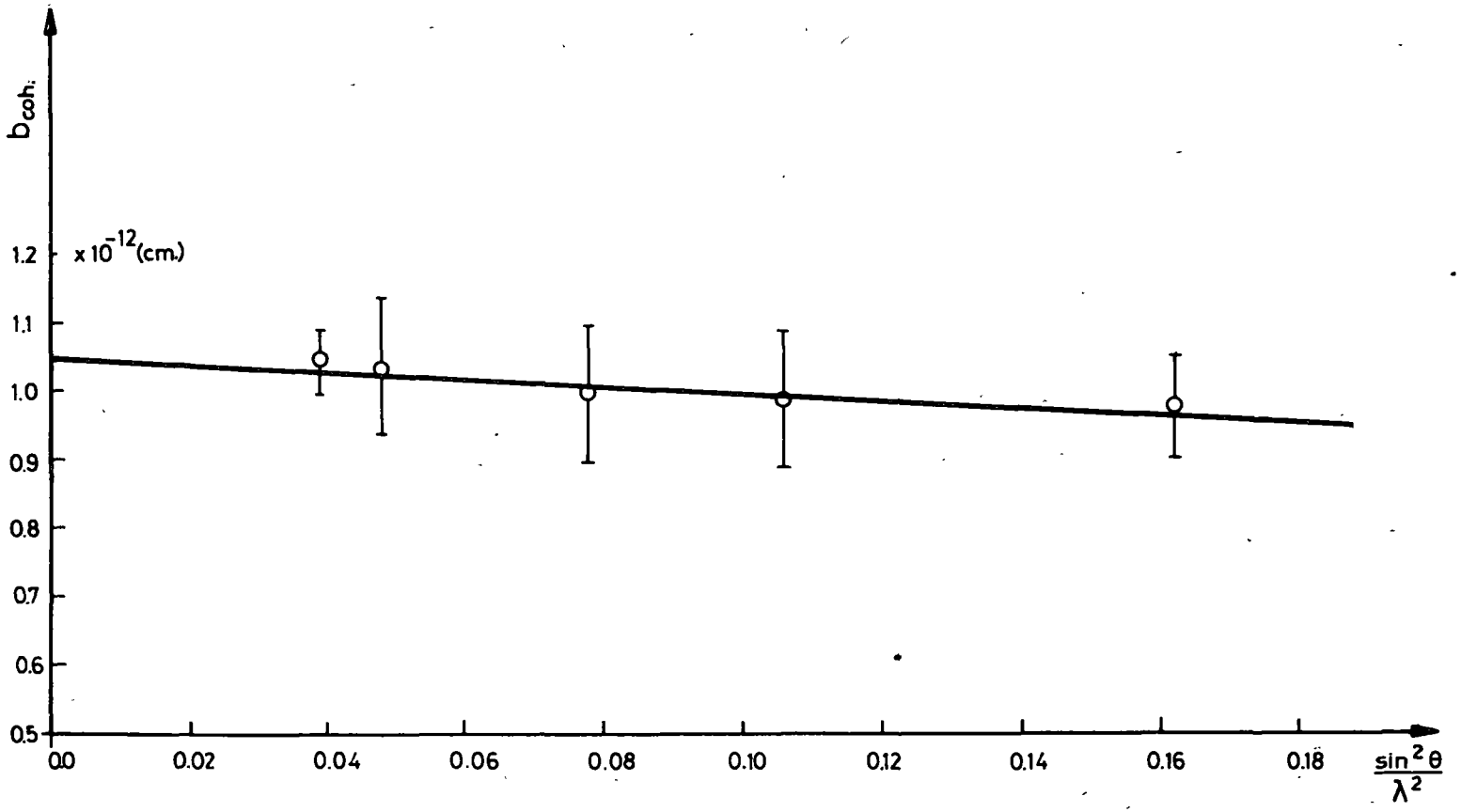
$b_{coh.}$
$\times 10^{-12}$(cm.)
1.2
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.0
0.02
0.04
0.06
0.08
0.10
0.12
0.14
0.16
0.18
$\frac{\sin^2\theta}{\lambda^2}$

Fig. 3