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THE DIFFRACTION OF NEUTRONS BY CRYSTALLINE POWDERS

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Introduction

Diffraction effects produced when a beam of slow neutrons falls on a crystalline substance have been demonstrated many times in the past decade. In general these experiments have made use of single crystals since then, the diffracted intensity is considerably enhanced over that when a crystalline powder is used. There are, however, several reasons for working with crystalline powders in spite of the much-reduced intensity obtainable, notably that (a) relative intensity data can be obtained without extinction and crystal perfection ambiguities and (b) substances can be examined which are generally not available in single crystal form. The performance of such experiments has, of course, become possible only since the advent of chain-reacting piles with their accompanying intense beams of collimated neutrons. The experiments to be described are more or less introductory to the general problem of the effect upon the intensity of diffraction of various nuclear and crystalline properties.

Experimental Technique

A schematic diagram of the equipment used in studying neutron diffraction effects is given in Figure 1. A collimated beam of slow neutrons is passed out through the radiation shielding of the Clinton pile and falls upon a monochromating single-crystal of sodium chloride. This crystal is oriented so as to Bragg-reflect neutrons of wavelength 1.08 augstroms (energy 0.069 electron volts). After collimation through slits of size 1 cm by 3.5 cm, the monochromatic beam strikes a flat plate of the crystalline powder under study mounted on a spectrometer table. Detection of the diffracted radiation is accomplished with an enriched $B^{10}F_3$ proportional counter of two-inch diameter and 20-inch length. The efficiency of this counter to neutrons of wavelength 1.08 augstroms is about 60 percent. The neutron current in the primary monochromatic beam is about 10^6 neutrons per minute and in the diffraction peaks about 30 - 100 neutrons per minute. In spite of rather heavy shielding of paraffin, B_4C and cadmium around the counter, the counter background (with the primary beam stopped off) amounts to 50 or 60 counts per minute. This background varies both in time and counter position and it has been found necessary to take alternate readings with and without a cadmium filter. Figure 2 shows a photograph of the diffraction spectrometer.

Experimental Results

I. Carbon Series

Three different structural forms of carbon namely: diamond, graphite and charcoal, have been examined and typical diffraction patterns for these are shown in Figures 3, 4 and 5. It is to be noted that an appreciable amount of diffuse scattering is present even in the case of diamond. This diffuse scattering has been studied over a considerably range of scattering angle and as is seen in Figure 6 it appears to be isotropic. From the intensity of the diffraction peaks and the diffuse scattering it is possible to calculate coherent and

diffuse scattering cross sections by use of equations

$$\frac{P_{hk\ell}}{P_o} = \frac{\lambda^3 \ell}{4\pi r}\left(\frac{h\rho'}{\rho}\right)\frac{e^{-\mu h \sec\theta}}{\sin^2 2\theta} \cdot j_{hk\ell} \cdot M^2 F^2_{hk\ell}$$

and

$$\frac{P_d}{P_o} = \frac{\ell \Delta\theta}{r}\left(\frac{h\rho'}{\rho}\right)\sec\theta\, e^{-\mu h \sec\theta} \cdot M \cdot S^2$$

where P_o is the incident beam intensity, $P_{hk\ell}$ the intensity in the $(hk\ell)$ diffraction peak, P_d the intensity of diffuse scattering, $F^2_{hk\ell}$ the coherent (Bragg) scattering cross section per molecule per unit solid angle, S^2 the diffuse scattering cross section per molecule per unit solid angle and other terms as defined in Compton and Allison. Using the experimental data for the carbon series, relative values of scattering cross section are obtained as shown in Table I.

Table I

Relative Cross Sections for Carbon Series

	Bragg Coherent	Diffuse	Total
Diamond	0.63	0.37	1.00
Graphite	0.69	0.56	1.25
Charcoal	-	1.46	1.46

All values relative to total scattering of diamond.

Thus even in the case of diamond the diffuse scattering seems to be over 30 percent of the total. The somewhat lower values for the diamond total cross section may be caused by extinction effects in the diamond sample. 800 mesh powder was used here and this may not have been fine enough to eliminate primary extinction. Experiments with 300 mesh powder showed a 15 percent change from the 800 mesh powder data. The considerably higher value for the charcoal total scattering may have been caused by the presence of occluded or adsorbed hydrogen and this point is being investigated more fully.

II. Calcium Series

Three calcium-containing compounds CaO, CaC_2 and CaS which contain zero spin and nearly monoisotopic nuclei have been examined. Typical patterns are shown in Figure 7. Relative values for the coherent and diffuse cross sections have been calculated and these are shown in Table II.

Table II

Experimental Cross Sections

	Ca	X	Diffuse	Total	Published Total Cross Section
CaO	0.46	0.16	0.92	1.54	2.8
CaC_2	0.31	0.27	2.9	3.8	4.0
CaS	0.22	0.04	2.1	2.4	2.2

(All cross sections are relative to that of diamond)

It is seen that the coherent scattering cross section for calcium varies by more than a factor of two from one compound to another. Moreover while the total cross sections for CaC_2 and CaS agree satisfactorily with the published values, that for CaO is only about one half the expected value. Again it is to be noted that the coherent Bragg scattering forms a small fraction of the total scattering being 0.4 for CaO and 0.11 for CaS. Such large values of the diffuse scattering seems to be in disagreement with current theories of inelastic diffuse scattering.

III. Determination of Phase Scattering

Theoretical considerations have indicated that the relative phase of scattering of neutrons may differ from one nucleus to another. In general this phase shift will be either 0^o or 180^o except when the neutrons have an energy very close to that of a nuclear resonance level. The diffraction pattern affords a convenient determination of this phase shift. Figure 8 shows typical patterns which indicate the relative phase shift upon scattering. Patterns are given for MgO, LiO, and MnO. These compounds all possess a NaCl -type structure and the relative intensities of the (200) and (111) diffraction peaks show immediately whether the compound nuclei are scattering in or out of phase with each other.

It is concluded that Mg and O scatter with the same phase while Mn and O (and Li and F) scatter out of phase with respect to each other. From considerations of this type along with experimental data on total reflection it is possible to group various nuclei as scattering with either positive or negative phase shift and this is done in Table III. The data shown in this table are composite taken both at Argonne Laboratory by Fermi and Marshall and at Clinton Laboratories.

Table III

Relative Phase of Scattering

Negative Phase Shift

Fe, Mg, O, Ba, Ca, S, F, Pb, N, C, Be, Na, K, Cl, Br, I, D

Positive Phase Shift

Mn, Li, H

As is seen in this table, only a few nuclei are known to scatter with a positive phase shift.

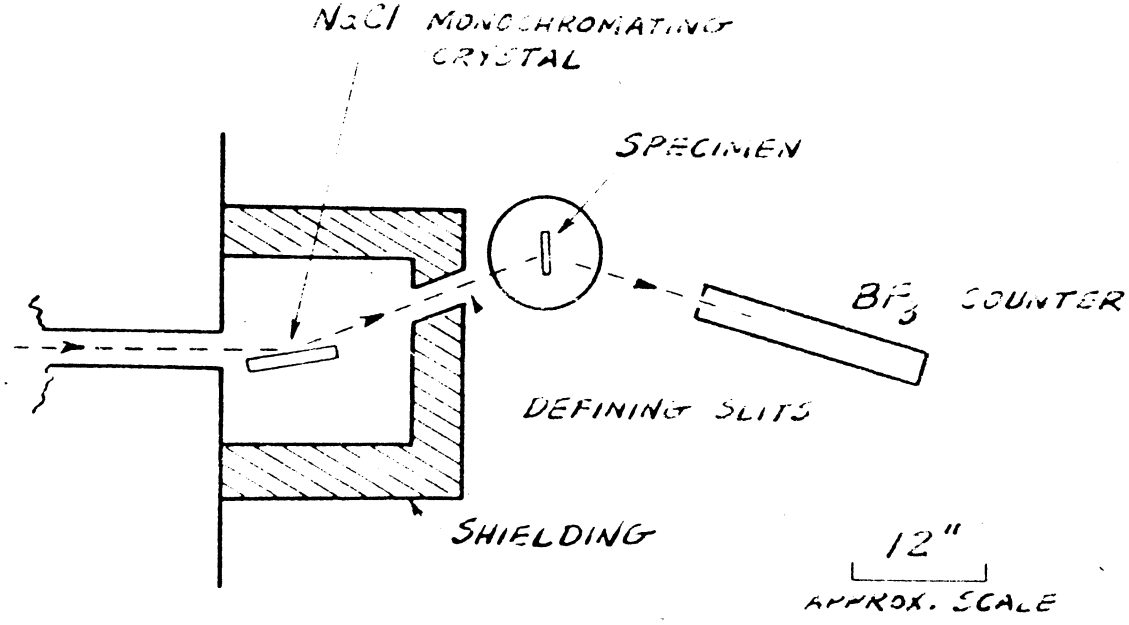


FIG. 1 - SCHEMATIC DIAGRAM OF NEUTRON DIFFRACTION SPECTROMETER

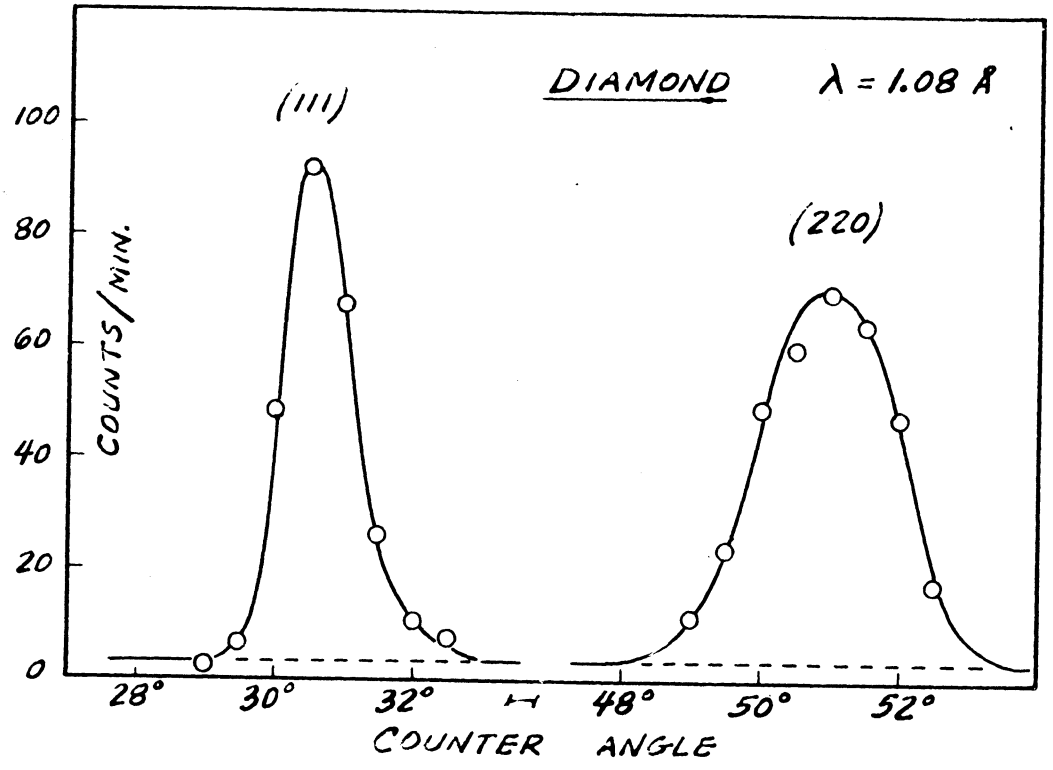

DIAMOND
λ = 1.08 Å
(111)
(220)
100
80
60
40
20
0
COUNTS/MIN.
28°
30°
32°
48°
50°
52°
COUNTER ANGLE

FIG. 3

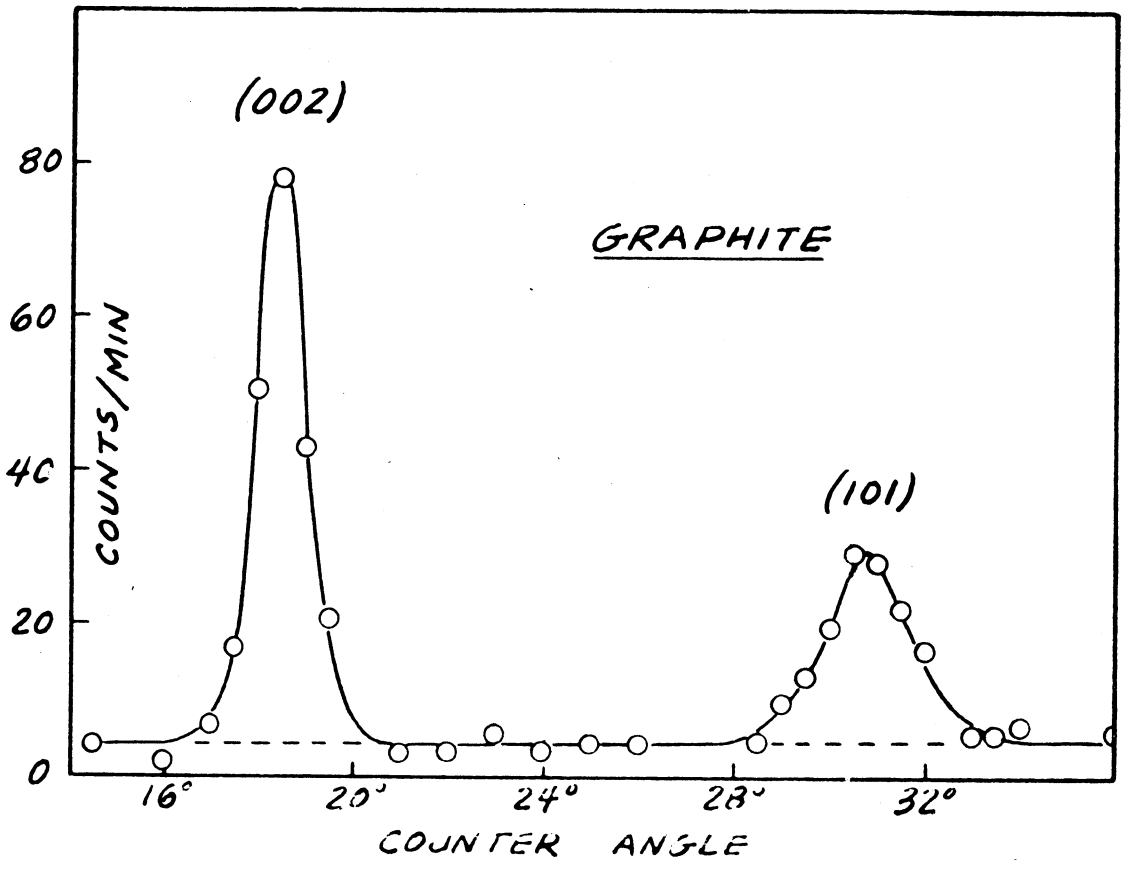

(002)
GRAPHITE
(101)
80
60
40
20
0
COUNTS/MIN
16°
20°
24°
28°
32°
COUNTER ANGLE

FIG. 4

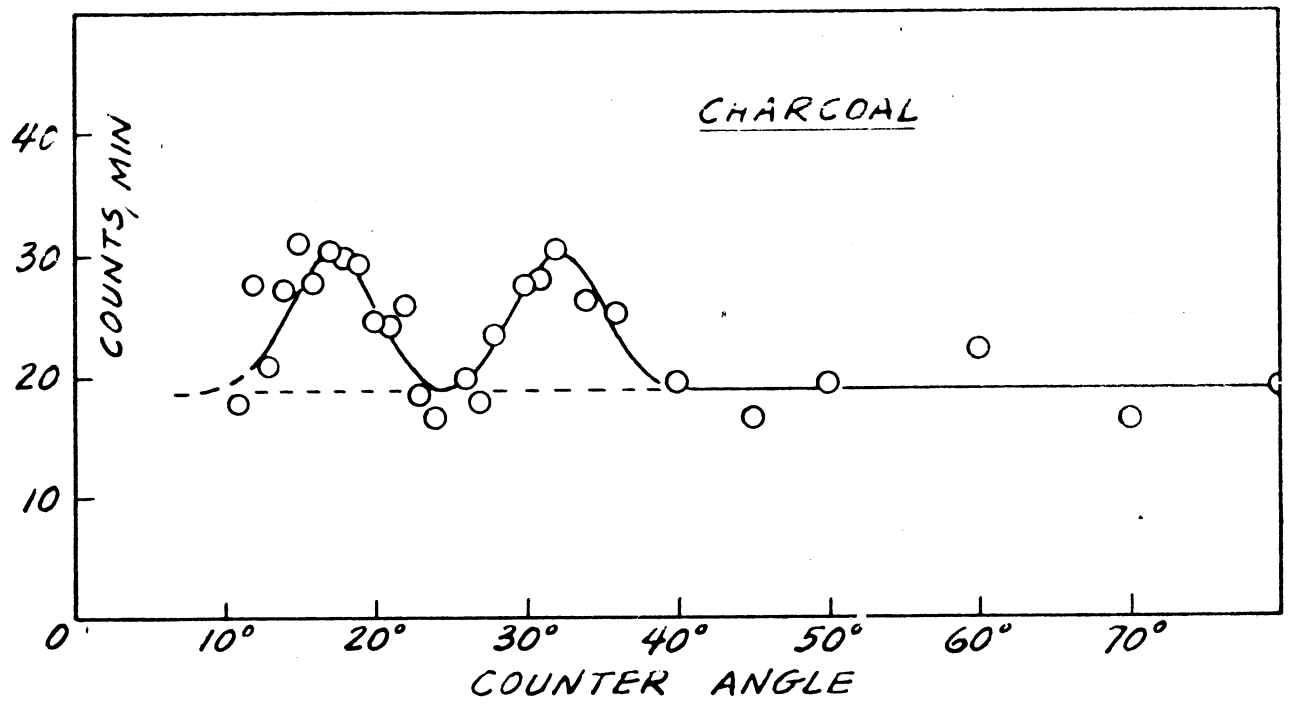

CHARCOAL
COUNTS/MIN
40
30
20
10
0
10°
20°
30°
40°
50°
60°
70°
COUNTER ANGLE

FIG. 5

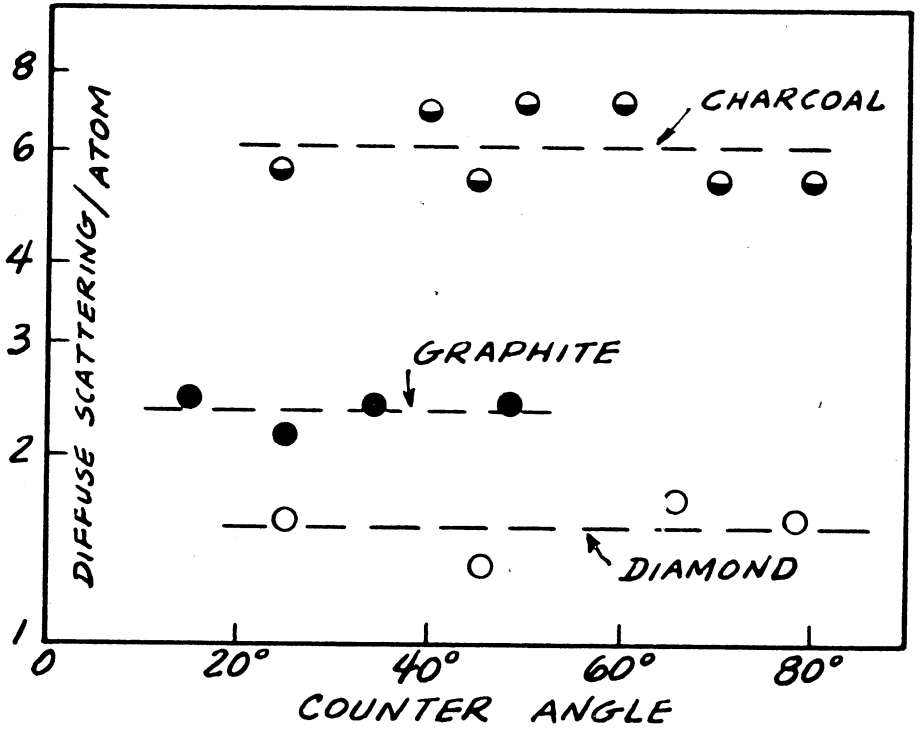

DIFFUSE SCATTERING/ATOM
8
6
4
3
2
1
0
20°
40°
60°
80°
COUNTER ANGLE
CHARCOAL
GRAPHITE
DIAMOND

FIG. 6

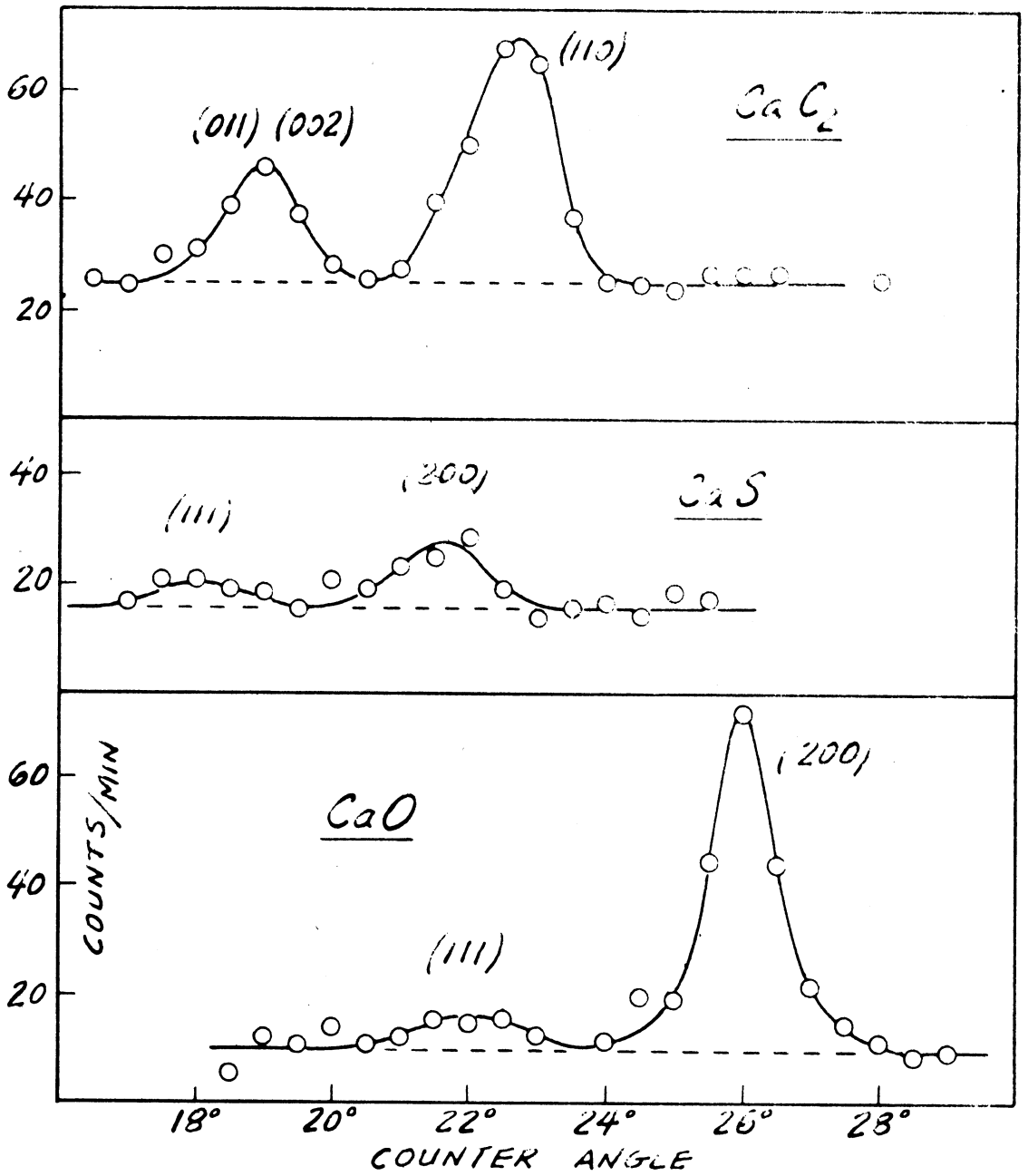
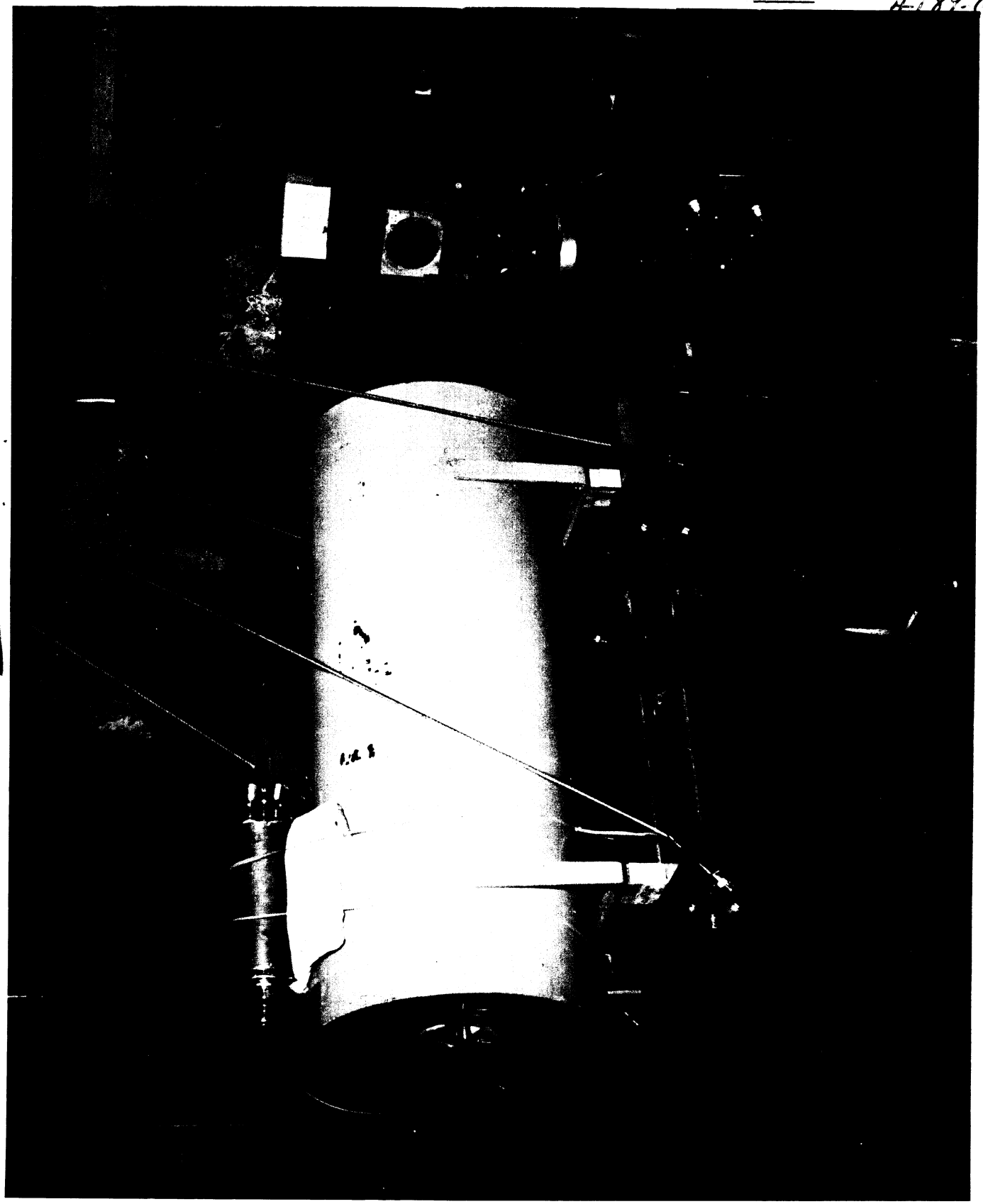

60
40
20
(011) (002)
(110)
CaC2
40
20
(111)
(200)
CaS
60
40
20
COUNTS/MIN
CaO
(200)
(111)
18°
20°
22°
24°
26°
28°
COUNTER ANGLE

FIG. 7



10

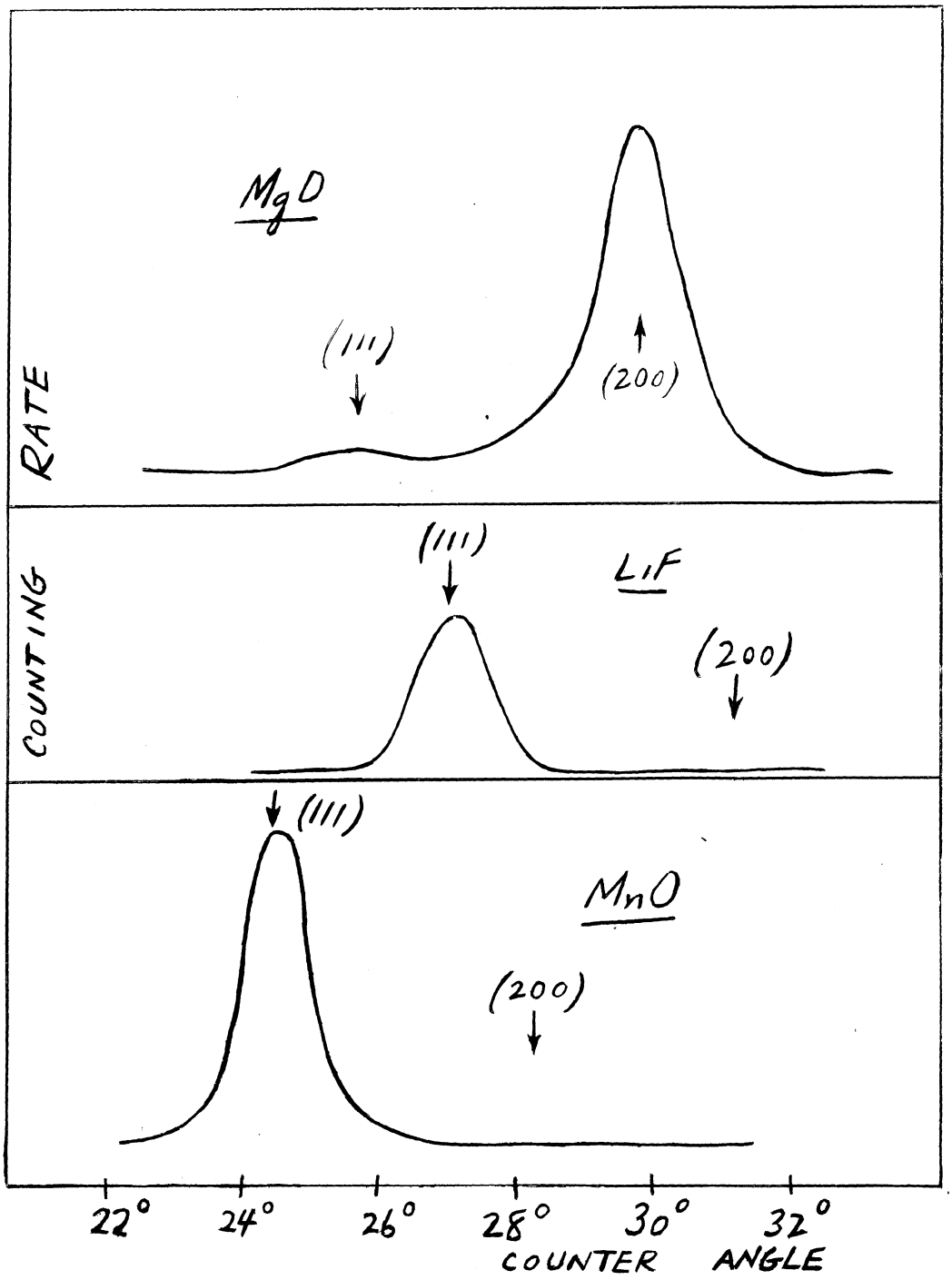

MgO
(111)
(200)
RATE
(111)
LiF
(200)
COUNTING
(111)
MnO
(200)
22°
24°
26°
28°
30°
32°
COUNTER ANGLE

FIG. 8