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A NEW APPARATUS FOR THE STUDY OF NUCLEAR BRAGG SCATTERING

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Abstract

A new monochromator system has been constructed which provides an energy resolution of 0.005 eV and an angular divergence of 0.4 arc seconds at an energy of 14.413keV. In conjunction with a highly perfect crystal of isotopically enriched $^{57}Fe_2O_3$, a beam of nuclear resonant photons was extracted from the synchrotron continuum with signal to noise ratio of 100:1, and an intensity of >2 quanta/sec.

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1. Introduction

The work of Mössbauer [1] showed that radiation having a very narrow energy width was produced by the recoil-free decay of nuclear excited states in solids. The nuclear excited states are produced as one product of a radioactive decay, and so only those nuclear species which are the product of a suitable decay scheme can be studied. The ready availability of highly intense x-ray beams from synchrotron sources in recent years has raised the possibility of directly exciting the resonance by nuclear photoabsorption [2]. The probability of absorption (and subsequent re-emission) is negligible except for photon energies very close to the resonance energy, and in the case of ^{57}Fe, this is at 14.413keV, in a bandwidth of about 10^{-9}eV. The problem of detecting the small number of photons within this bandwidth when using synchrotron radiation to excite such resonances is thus essentially one of signal to noise ratio.

Typical monochromators used for synchrotron radiation provide a bandpass of several eV, a factor of 10^9 larger than the width of the resonance. It is obvious that elastic scattering from the electrons in the material (which has no energy dependance on this scale) will dominate the observed intensities. It is possible to arrange the resonant atoms in a crystal lattice in such a way that for certain Bragg reflections only the resonant nuclei scatter in phase. Thus a perfect sample of such a crystal can suppress a large fraction of the unwanted electronic scattering. Indeed, the first observation of nuclear Bragg scattering using synchrotron radiation was made using two successive such 'pure nuclear'

reflections [3]. Even so, the signal to noise ratio achieved was only around unity. The number of systems for which two perfect isotopically enriched crystals are available is severely limited, and details of the resulting photon beam, whilst intrinsically interesting, are greatly complicated by the double nuclear scattering. We were thus prompted to try to develop an approach which might allow observation of the nuclear scattering in cases other than the magnetic systems used to date, in particular for situations where the electronic scattering is less perfectly suppressed, and to extend the capabilities to nuclei other than iron.

2. High Resolution Monochromators.

2.1 Resolution

The choice of a basic design for a high-resolution x-ray monochromator depends greatly on the expected application. This application had very clear requirements. It should provide the minimum possible bandwidth in order to minimize the effect of non-resonant scattering. The angular range of reflection for our sample crystal (a highly enriched perfect single crystal of $^{57}Fe_2O_3$) was a few arc seconds. Since radiation outside that range could only contribute to noise, an additional requirement is that the monochromator output beam should be collimated to a level similar to the crystal reflection width.

To minimize the bandpass of the monochromator requires the use of very high order Bragg reflections. The use of such high order reflections to generate

extremely narrow wavelength bands from a synchrotron source has been the subject of several investigations with the aim of observing inelastic x-ray scattering from phonons and other sub-eV excitations [4,5,6]. Those studies were able to choose freely the diffraction order utilized, since their choice of photon energy was arbitrary. The experiment by Graeff and Materlik [4] demonstrated that resolutions of millivolt order could in fact be observed. Their apparatus gave a resolution of 0.008 eV at an energy of 15.8 keV using the silicon (8,8,8) reflection. The study of inelastic x-ray scattering, however, requires the collection of large solid angles of radiation due to the diffuse and extremely weak nature of the scattering cross-sections. Consequently, a practical apparatus requires the use of focussing optics, as in the work of Dorner, Burkel and Peisl [5], and that of Siddons et al. [6]. The type of focussing optics used in those experiments would not be beneficial to the present experiment, since they would provide a divergent excitation beam. As pointed out above, only rays incident within the range of Bragg reflectivity of the resonantly scattering crystal can contribute to the signal. All other rays contribute only to the noise level. Another feature of these inelastic scattering experiments was their use of Bragg angles close to 90°. The reason for this choice of wavelength and diffraction order is simply that the influence of incident beam divergence on energy resolution is minimized at high Bragg angles. Since we are not free to choose an arbitrary wavelength, we must choose as high a diffraction order as possible, and control beam divergences so as to achieve the desired characteristics.

The design of choice for high resolution monochromators is the dispersive

double-crystal arrangement. In its classic configuration [7] it is somewhat unwieldy, since the monochromatic beam is deviated from the primary beam by four times the Bragg angle. A system in which the output beam is fixed in space (or at least in direction) is significantly easier to instrument. As pointed out by Beaumont and Hart [8], the availability of highly perfect crystals of silicon in large pieces makes possible the design of many-reflection systems with little cost in efficiency. A realization of the dispersive double-crystal apparatus using a pair of 2-reflection monoliths provides a fixed exit tunable beam of well-controlled divergence. Its resolution is essentially independant of the source characteristics. The highest order reflection available for diffraction at 14.413keV by silicon is the (11,5,3). For practical reasons we chose not to use this, but the (10,6,4). Not only is it easier to fabricate the two-reflection monoliths for this reflection, but reflections of even order have a better peak reflectivity. The bandwidth provided by this combination was .005eV, with an angular divergence of 0.4 arc seconds.

2.2. Thermal considerations

The NSLS storage ring produces a power load on the first optical element of 8 Watts per horizontal milliradian from a dipole source with a stored current of 100mA. This is always a problem for x-ray monochromators, but especially so for the high-resolution device described above. Changes in d-spacing causing changes in Bragg angle comparable to the Darwin width between one crystal element and another will seriously reduce the efficiency of the device. For

the (10,6,4) reflection at 14.413 keV, such changes in d-spacing occur for temperature differentials of roughly 0.2°K. With a power load of 8 Watts incident on one wafer of a 2-reflection monolith, it is quite difficult to maintain the temperature differential across the device to this level. Even though such changes can be compensated by suitable crystal design, time variations in the power load would render control unwieldy. It is also vital that the absolute energy of the transmitted beam remain constant (i.e. on resonance), and that is far more difficult to achieve if the temperature of the crystal elements is poorly defined. In view of the above, a third monolithic double reflector was inserted upstream of the high-resolution elements whose sole purpose was to intercept the bulk of the thermal load and prevent it from disturbing the thermal equilibrium of the succeeding stages. It does not need to provide resolution, but it should be efficient. A silicon (1,1,1) monolith was used. Its tolerance of thermal loads is much greater than that of the (10,6,4) device, and on the scale of millivolts it is a broadband device. Figure 1 shows the final optical arrangement comprising the three monoliths and the nuclear resonant sample, and figure 2 is a photograph of the crystals used.

2.3. Mechanical design

The entire apparatus was installed into an experimental hutch having dimensions 1.6 meters by 0.6 meters. This included rotation stages for all three monoliths capable of sub-arc second resolution plus a standard two-circle diffractometer and solid-state detector. The requirement for close angular con-

trol over the three optical elements led to a design based on sine bars. Each axis of the high resolution pair consisted of a precision spindle rigidly mounted to the chamber which enclosed them, coupled to a lever of length 0.56 meters. The free end of this lever was driven by a non-rotating micrometer head, which was in turn driven by a stepper motor. The combination was such that one motor step produced a rotation of the spindle of 0.2 arc seconds. The upstream monolith was mounted on a similar arrangement, but with a shorter lever arm (0.3 meters). It was surrounded by a lead cave in order to control scatter from the white beam incident on it. A moveable lead beamstop was arranged between the crystals of the high resolution pair, and moveable slits were situated just upstream and downstream of them (see figure 1). Thus any spuriously scattered photons were forced to undergo several scattering events before exiting the monochromator. The background level was thus rendered very low.

2.4 Alignment

Alignment of the three monoliths with the synchrotron beam is complicated by the fact that the only observable quantity is the intensity at the output of the monochromator whereas there are several adjustments, each of which causes intensity changes. The situation is clarified by reference to figure 3, which is a modified Dumond diagram illustrating schematically the angular ranges of reflection for the crystals and divergence of the synchrotron beam as a function of wavelength. The angle/wavelength passband of the entire system is

given by the extents of the small parallelogram enclosed by the two (10,6,4) traces. This small area can be made to move anywhere in the figure by suitable rotations of the two crystals (horizontal translation of the traces in the figure). If one of the (10,6,4) crystals is rotated, the rectangle traverses the (1,1,1) reflectivity range. If the synchrotron beam intensity is uniform (in angle) over this range, then such a scan should map out the silicon (1,1,1) Darwin-Prins curve. Figure 4 shows the result of just such a measurement. The theoretical width of the reflectivity curve is 3.5 arc seconds, whereas the width of the curve in figure 4 is around 7 arc seconds. However, simple geometric arguments based on figure 3 can show that rotation of one crystal by an angle ω causes the center of the parallelogram to move in angle by $\omega/2$, as observed.

The most direct algorithm for adjusting the monochromator is as follows. First the two (10,6,4)'s are set to give the correct wavelength. This setting (to first order, at least) depends only on their angle relative to each other, and not on their angle with any other component. Next, the (10,6,4)'s should be centered in the window of the (1,1,1) device, by rotating the (1,1,1) crystal relative to the (10,6,4)'s. The final step is to rotate the whole monochromator relative to the synchrotron beam so that the maximum of the vertical intensity profile is centered on the parallelogram. It is hardly necessary to point out the advantages of using monolithic double-reflectors in this system. A similar system based on seperate motions for each reflection would be extremely difficult to align.

3. Wavelength calibration

Since the resolution of this monochromator is very high, it becomes important to devise a method of calibration of the wavelength scale to a level similar to its resolution. Unfortunately, very few physical effects in the x-ray regime are sufficiently sharp or well-determined to justify their use as a calibration point. The Mössbauer line itself is such a feature, and would make an admirable wavelength standard: significantly better than the current x-ray wavelength standard (as was pointed out by Bearden [9]). However, examination of the literature reveals a range of values quoted for the energy of this line. Bearden [9] reported measurements made using calcite and quartz crystals in a double-crystal spectrometer. The two values obtained differed by 0.2 eV, a factor of forty times the resolution of our monochromator! Gerdau et al [3] quote a value which is 0.5 eV different from the mean of Bearden's values, although it derives from the same measurements. The difference must be due to the values of the fundamental constants used in converting from wavelength to energy. This emphasizes the fact that all diffraction-based spectroscopy essentially measures a wavelength relative to the lattice plane spacing of some reference crystal, which was (until recently) in turn compared to a standard wavelength (the Kα line of Tungsten [9]).

The best-determined length on the angstrom scale is in fact the silicon lattice constant [10,11]. This has been compared directly with the wavelength of a well-characterized laser line using simultaneous x-ray and optical interferometry.

thus breaking the circular relationship described above. It would appear sensible to use this fact in calibration. The output of our monochromator was passed to a two-circle diffractometer upon which was normally mounted our nuclear resonant sample. This diffractometer (a Huber 424) was said to have an absolute accuracy of less than 30 arc seconds. It was fitted with gear reducers and stepping motors which gave it a step size of 0.9 arc seconds. Our experience indicated that its reproducibility is much better than its accuracy, around ± 1 arc second. The principle purpose of a wavelength determination using this instrument would therefore be as an internal calibration. For the purposes of this calibration, the sample was replaced by a 0.5 m.m. thick silicon plate. This plate was cut in such a way that the $(\bar{1}0.6.4)$ planes were parallel to its major face, and the (1.1.1) planes were perpendicular to that face (see figure 5). The Bragg angle for the (10.6.4) reflection at 14.413 keV is 77.5 degrees. For the (1.1.1) reflection it is 7.9 degrees. Thus the crystal orientation for the $(\bar{1}0.6.4)$ reflection in Bragg case is only 4.6 degrees from the orientation for the (1.1.1) reflection in Laue case, and the $(\bar{1}.\bar{1}.\bar{1})$ reflection is also accessible by a rotation of 15.8 degrees from the (1.1.1). Figure 6 elucidates this geometry. The two (1.1.1) reflections have little value as a wavelength determination, but they provide an all-important zero of angle scale so that the Bragg angle for the $(\bar{1}0.6.4)$ reflection can be absolutely determined. Figure 7 shows a typical data set taken at the resonance energy. The angles given are those obtained relative to an arbitrary zero; the true zero being midway between the values at (a) and (b). The (10.6.4) value is, of course, offset by 90° from its nominal value. The value of the Bragg angle we obtained for the (10.6.4) reflection at the

Mössbauer resonance was 77.53350°, which corresponds to a wavelength of 0.860262Å. Various systematic errors were unaccounted for in these initial measurements, and so this value does not constitute an absolute determination of the wavelength of the Mössbauer line. Future measurements will take proper account of these errors and an improved value for the wavelength will result. Since the present level of knowledge of the line energy is around 0.2 eV, a search was still necessary initially to locate the exact resonance energy. Once it had been found, however, the above calibration scheme enabled us to return to the correct energy within a very short time.

4. Measurements

As described above, the absolute value of the resonance wavelength is rather poorly defined compared to the monochromator passband. It is therefore necessary to search in energy for the pure nuclear Bragg reflection which we are looking for. Although in principle not necessary, the specification of the diffractometer used required that we also search in Bragg angle. To minimize the area of this 2-dimensional scan, we must know very well the lattice constants of the sample. The value of the (1.1.1) (Rhombohedral indices) d-spacing of the Fe_2O_3 is even less well determined in the literature than the Mössbauer wavelength. We were able to determine it by measuring a series of Bragg angles for normal Bragg reflections (i.e. non-nuclear reflections) of type (h.h.h), for a wavelength calibrated using our silicon reference, as above. This same series of measurements also allowed us to set the zero of the sample

Bragg-angle scale. This is different from the zero in the wavelength calibration experiment since the crystal did not have a well-determined orientation relationship with the goniometer mount. The search for this peak .005eV and 3 arc seconds wide in a parameter space 1eV and 20 arc seconds wide took a considerable time. Once found, the resonant sample was removed and the wavelength determined as described above. Subsequent searches from this calibration point usually took around thirty minutes.

Figure 8 shows two rocking curves of the Fe_2O_3 crystal at the resonance energy. The upper curve is a normal Bragg peak (the allowed (6,6,6) reflection), whereas the lower is from the pure nuclear (7,7,7) reflection. The notable feature is the reflection widths, in that they are essentially the same. This implies that the width is characteristic of the strains in the crystal sample. By contrast, if we scan the same two reflections in wavelength, as in figure 9 (i.e. at fixed crystal angle while changing wavelength) we see the dramatic energy-dependance of the pure nuclear reflection. The non-nuclear (6,6,6) reflection shows the energy-dependance one would expect based on Bragg's law, whereas the pure nuclear (7,7,7) shows a very sharp response which is just the monochromator resolution function. Similar behaviour was also observed for the pure nuclear (5,5,5) reflection. We were not able in the time available to make velocity or time spectra, which are the usual test for Mössbauer sources, and so we must ask if our observations could arise from some other source. Possible candidates are magnetic scattering [12] and multiple diffraction [13]. Magnetic scattering does not have a strong energy dependance, and so the

data of figure 8 removes that possibility. Multiple diffraction is more difficult to discount, but several facts suggest strongly that our observation is not due to this phenomenon. First, any diffraction phenomenon in our crystal will be smeared to some extent by its grown-in strains, and this would be reflected in similar ways in both angular and energy scans. Comparison of figures 8 and 9 show the marked difference between angle and energy dependance of the pure nuclear reflection. Secondly, the occurrance of an accidental multiple diffraction point at exactly the same energy (to within a few parts in 10^5) as the nuclear resonance is highly unlikely, and the added coincidence of a similar peak observed at the pure nuclear (5,5,5) reflection is not reasonable.

There are other features of the data which are interesting from the point of view of resonant nuclear diffraction, but the reader is referred to reference [14] for a discussion of them.

5. Summary

A very high resolution multiple-crystal monochromator system has been built which provides an energy bandwidth of .005 eV at an energy of 14413 eV with a vertical beam divergence of 0.4 arc seconds. The photoexcitation of Mössbauer resonances is a new application for such devices, and their utilization opens up the possibility of extending such work to include previously inaccessible atomic species. This monochromator has been used to observe nuclear Bragg scattering

from a single pure nuclear reflection with a signal to noise ratio of 100:1. It is also intriguing to consider the application of this monochromator system, coupled with a similar dispersive double-crystal analyzer to inelastic x-ray scattering. Such a system would be ideal for probing inelastic scattering in the region of very small momentum transfer as well as the study of inelastic features which are sharp in momentum (i.e. having widths similar to Bragg reflection widths).

6. Acknowledgments

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Figure captions.

1. Disposition of monochromator components and nuclear resonant sample crystal, showing beam trajectory.

2. (a) Photograph of silicon monoliths used in the monochromator. (b) Photograph of perfect single crystal of $^{57}Fe_2O_3$.

3. Modified DuMond diagram showing angular ranges of reflection of the crystals and divergence of the synchrotron radiation beam as a function of wavelength.

4. Rocking curve produced by rotating one of the (10.6.4) crystals with all the other elements stationary. This is essentially the (1.1.1) Darwin-Prins curve with its angle scale multiplied by 2. The curve is simply a guide for the eye.

5. Orientation of the silicon wafer used for wavelength determination. The slot near the base of the crystal serves to allow mounting strains to relax before they can affect the active region of the wafer.

6. Geometry used for wavelength determination, showing orientation of crystal wafer for each of the three reflections measured.

7. Intensity profiles for the three reflections used for wavelength determination. The data points are joined by straight lines to guide the eye.

8. Rocking curves of the normal (6,6,6) Bragg peak (i.e. due to electronic scattering) and of the (7,7,7) pure Nuclear Bragg reflection, on resonance. The two curves have essentially the same shape which is dominated by the sample crystal perfection.

9. Energy scans at constant Bragg angle for the (6,6,6) and (7,7,7) reflections. The energy width of the (6,6,6) reflects the crystal perfection, and is consistant with Bragg's law, whereas the energy width of the (7,7,7) is just the passband of the monochromator, and verifies the pure nuclear nature of the reflection.

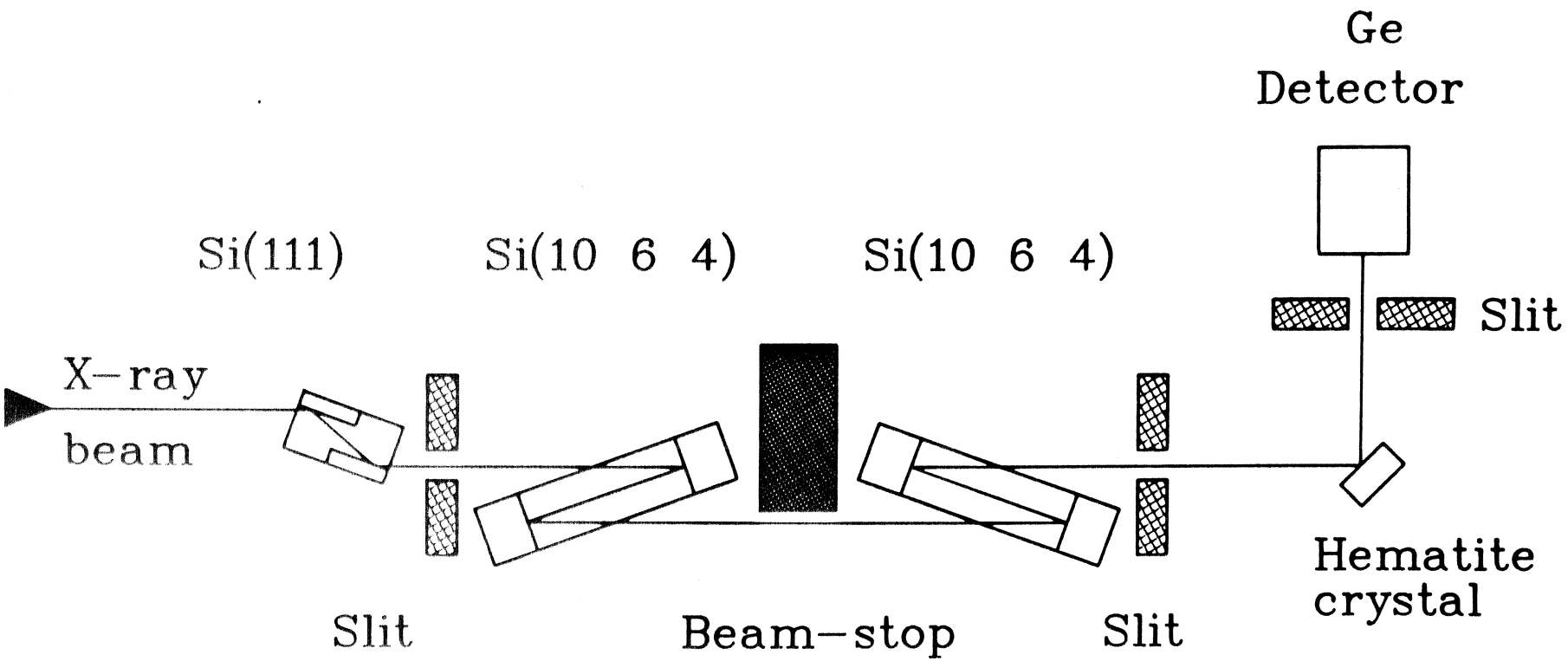
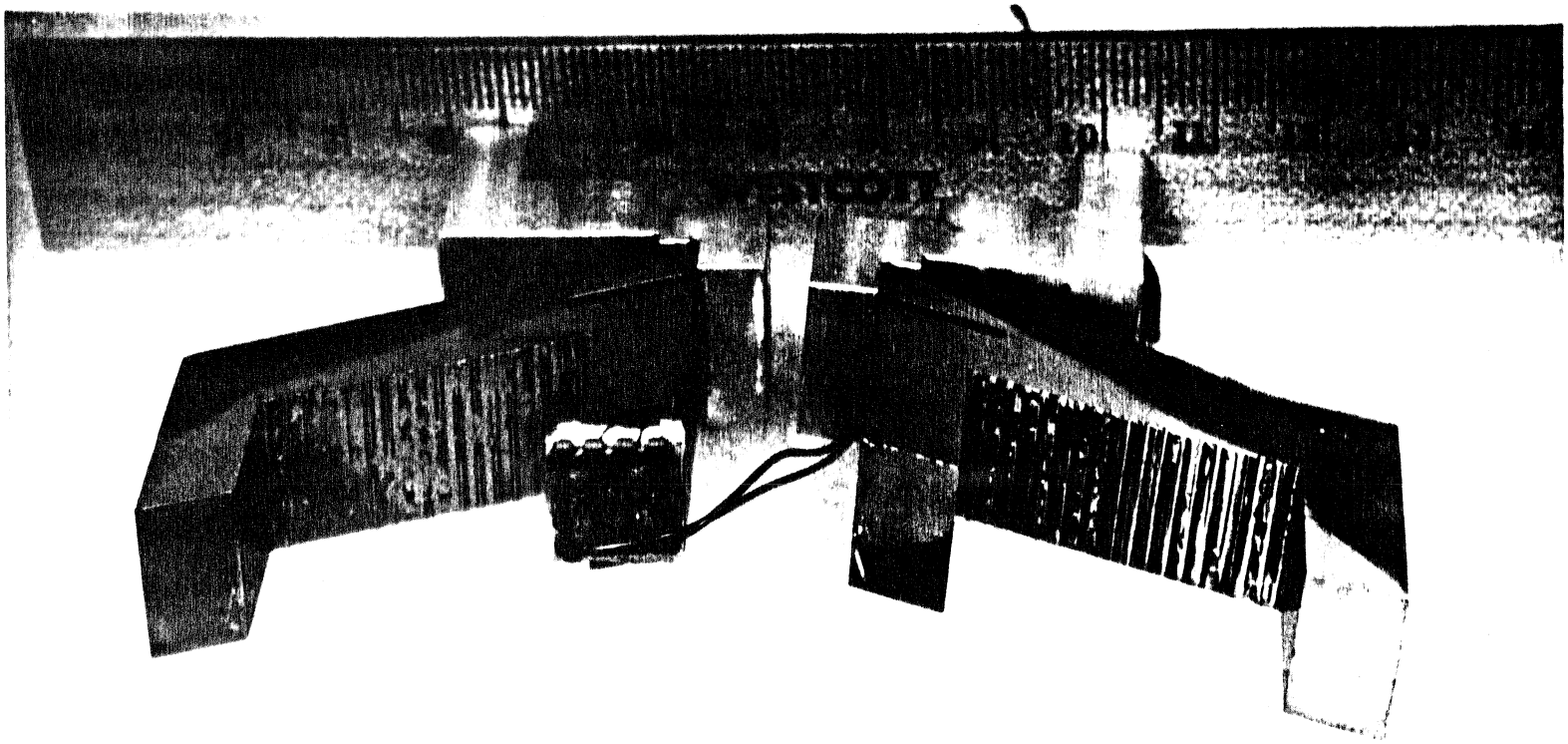

Ge
Detector
Si(111)
Si(10 6 4)
Si(10 6 4)
Slit
X-ray
beam
Hematite
crystal
Slit
Beam-stop
Slit

fig 1.



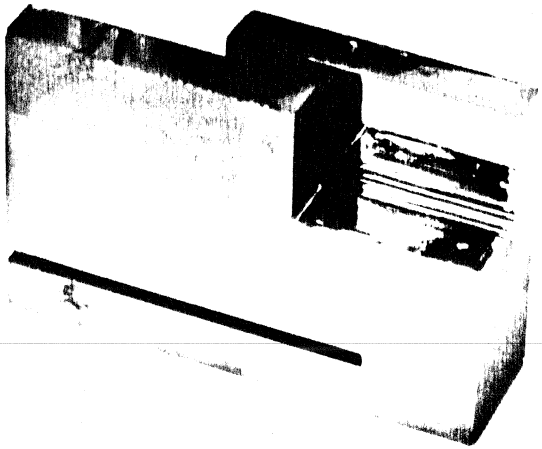


fig 2(a)

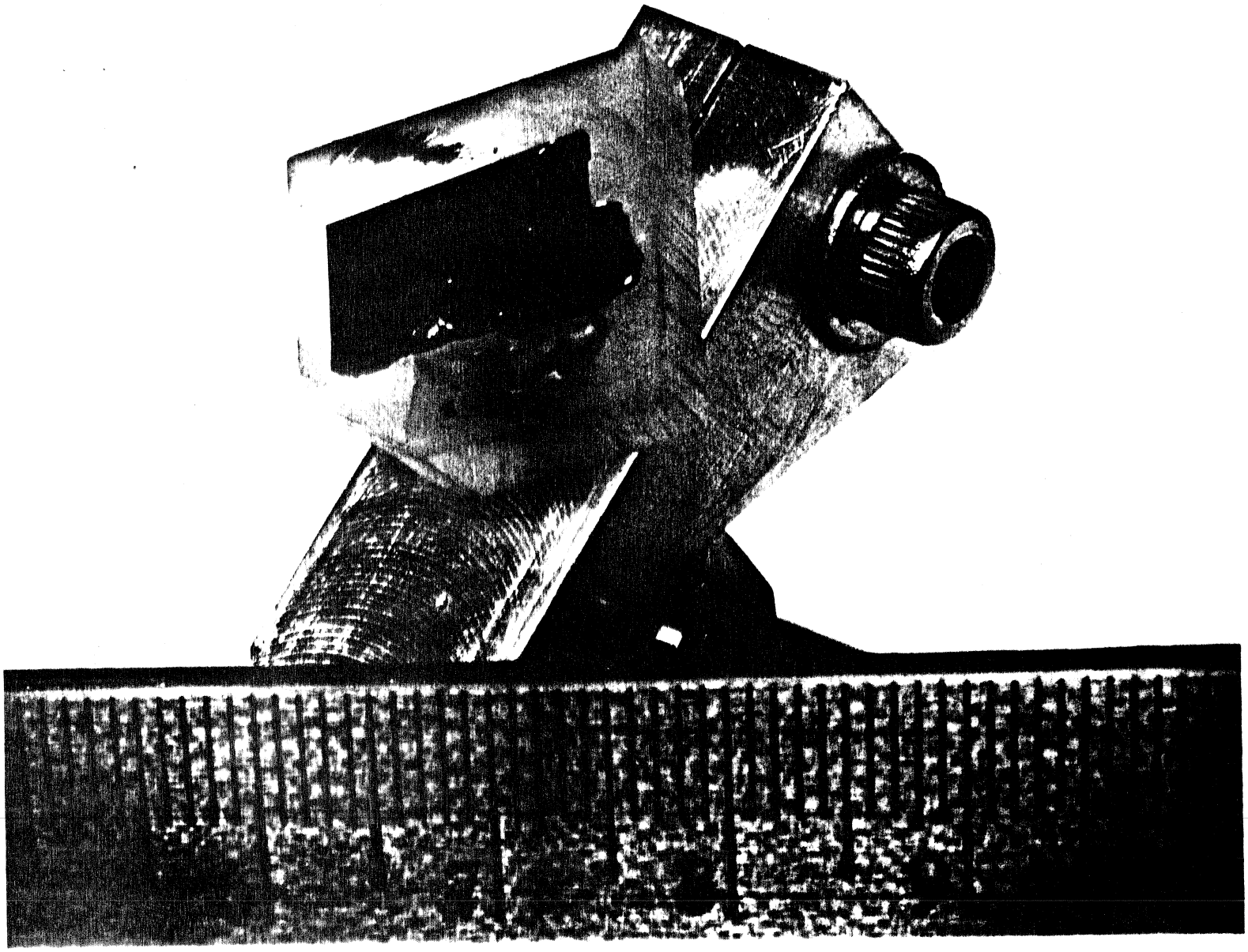


fig 2(b)

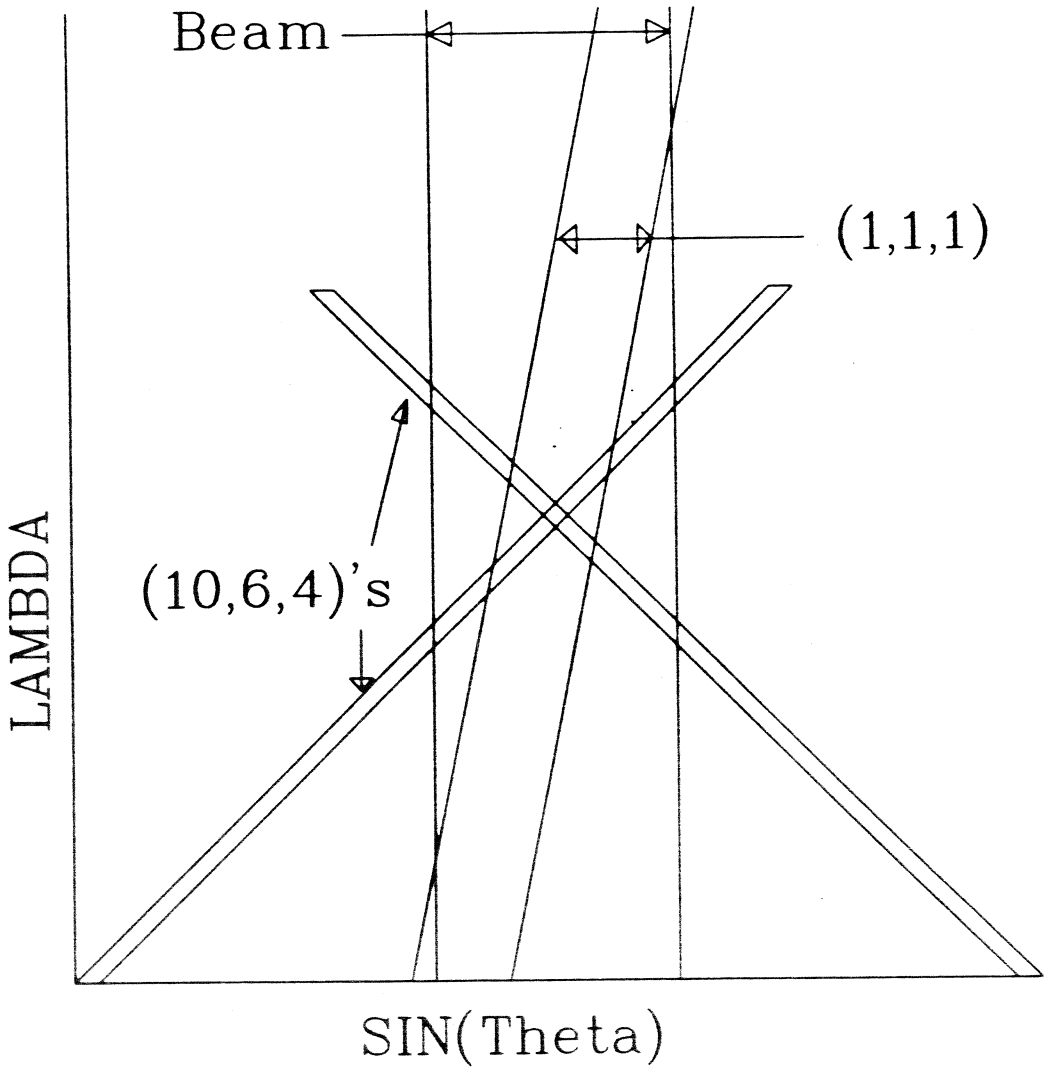


fig 3.

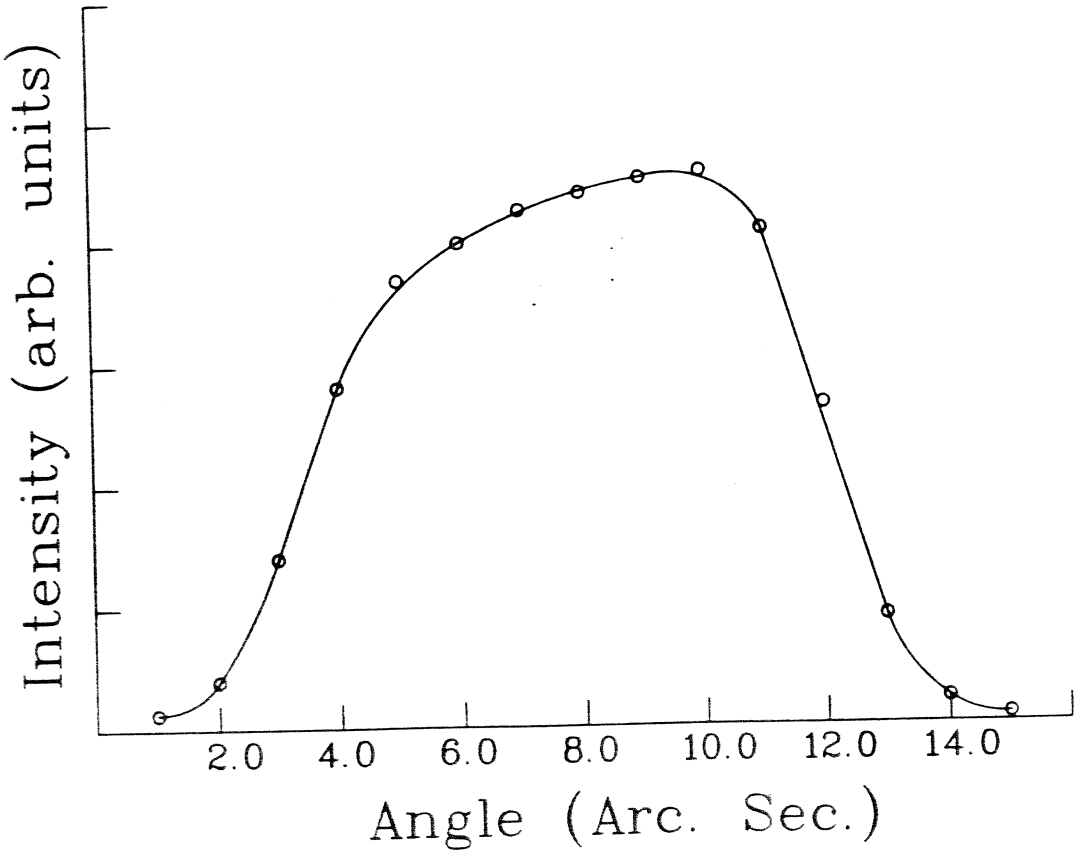


fig 4

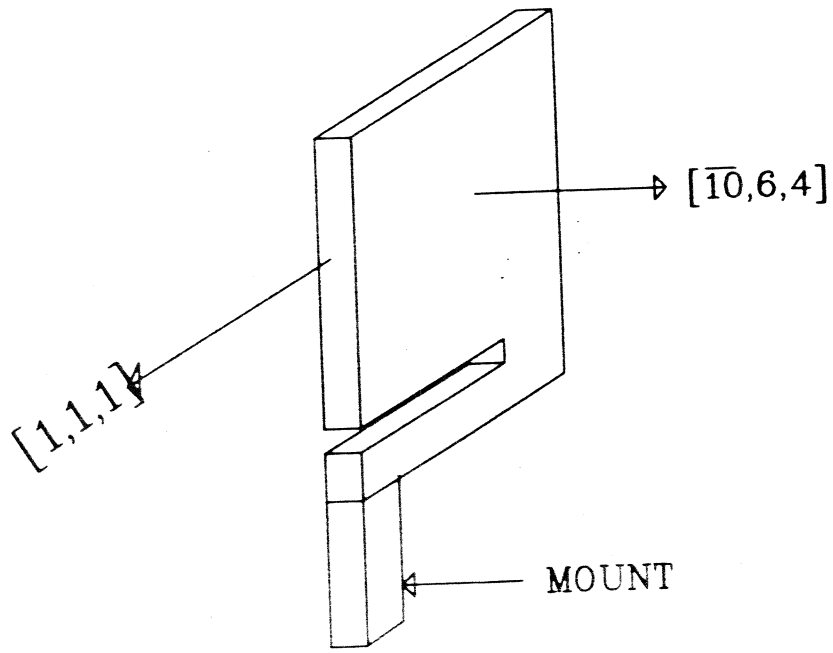
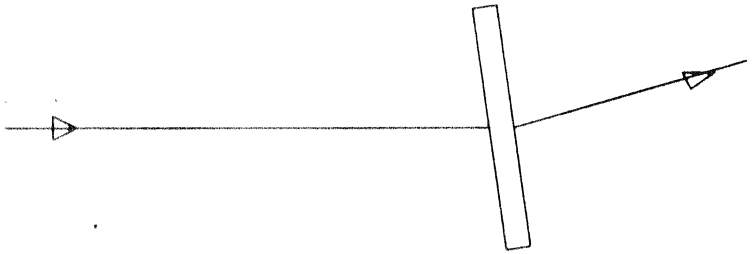
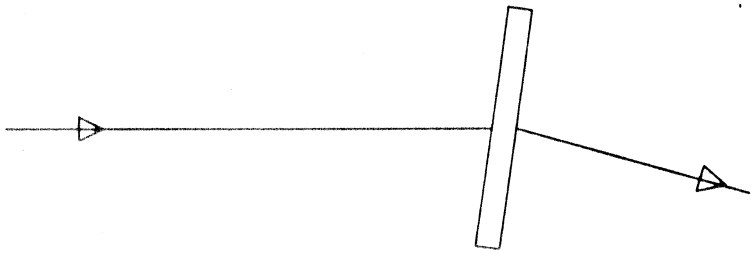


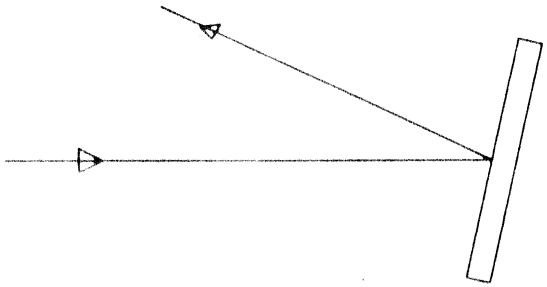
fig 5



(a) $(\bar{1},\bar{1},\bar{1})$ Laue case



(b) (1,1,1) Laue case



(a) $(\bar{1}0,6,4)$ Bragg case

fig 6.

4.38450
−11.38375
−15.96613
Intensity (arb. units)
0.001°
(a)
(b)
(c)
Omega (Degrees)

fig 7.

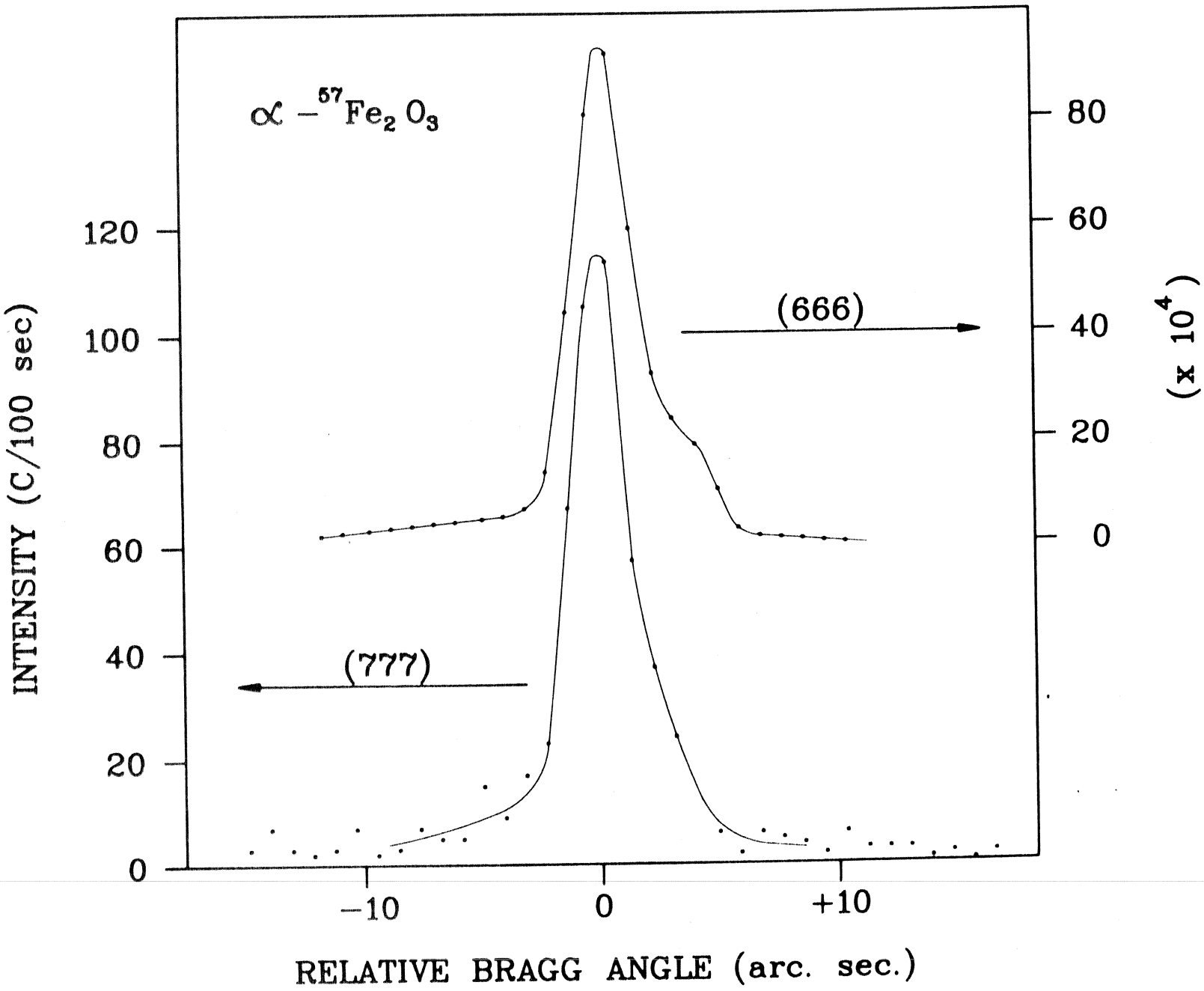

$\alpha - {}^{57}Fe_2O_3$
(666)
(777)
INTENSITY (C/100 sec)
(x 10^4)
120
100
80
60
40
20
0
80
60
40
20
0
−10
0
+10
RELATIVE BRAGG ANGLE (arc. sec.)

fig 8

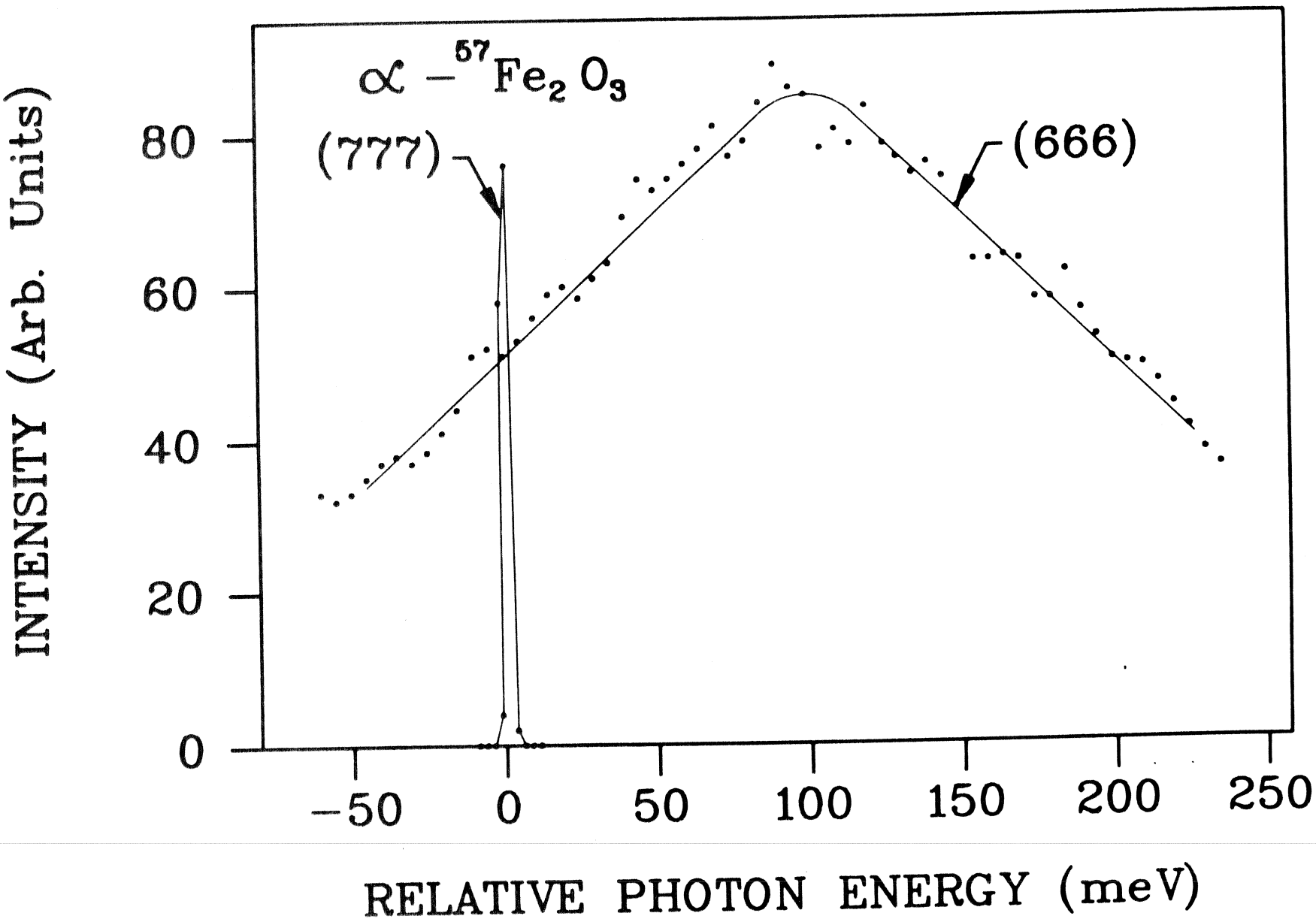

α − $^{57}Fe_2O_3$
(777)
(666)
INTENSITY (Arb. Units)
0
20
40
60
80
−50
0
50
100
150
200
250
RELATIVE PHOTON ENERGY (meV)

fig 9

DISCLAIMER