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High Energy-Resolution Inelastic X-ray Scattering

J. B. Hastings, D. E. Moncton, and Y. Fujii*

Brookhaven National Laboratory, Upton, New York 11973

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ABSTRACT

A brief review is presented of various aspects of high energy-resolution inelastic x-ray scattering based on synchrotron sources. We show what kinemetical advantages are provided by the photon probe and propose mirror and monochromator designs to achieve an optically efficient beam line for inelastic x-ray scattering.

*permanent address: Osaka University, Toyonaka, Osaka 560 Japan

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The development of x-ray scattering methods based on the unique properties of synchrotron radiation, and the concomitant demonstration of the enhanced beam brilliance provided by wigglers and undulators (see paper by G. Brown in these proceedings), has led to the consideration of new x-ray techniques which are not viable on conventional x-ray sources. Of particular interest in the context of the study of elementary excitations in condensed matter systems is the possibility of performing inelastic x-ray scattering experiments with energy resolution ranging from 1 eV down to 1 meV and perhaps ultimately to the regime below 1 μeV.

X-ray techniques will be able to compete in many problem areas traditionally accessed by neutrons, such as the study of phonons between 1 and 100 meV in bulk crystals. However, for certain applications, inelastic x-ray techniques will be unique. These areas include (1) the study of collective electronic excitations to which the neutron does not couple (see paper by S. Doniach in these proceedings), (2) the range of energy transfers above 200 meV, which is easily accessed with a 15 keV x-ray probe but difficult for the much lower-energy neutron, (3) excitations in disordered materials in which high neutron-energy transfer cannot be produced at small momentum transfer, and (4) the study of systems for which only small samples are available. In the last category, the study of the dynamics of two-dimensional systems should be revolutionized by inelastic x-ray scattering. It should be emphasized that there are, correspondingly, areas in which the neutron probe will remain unique. These include the regime of energy transfer below 1 meV and the study of magnetic excitations.

The kinematical advantages of the x-ray probe are described in Fig. 1 in terms of the energy and momentum transfer characterizing the excitations one wishes to study.[1] For a 14 meV neutron beam, the accessible range of energy and momentum is the parabolic area enclosed by the dashed curves; similarly the 100 meV neutron region is bounded by the dashed-solid curve. By comparison a 14.4 keV x-ray beam can access virtually the entire energy-momentum plane with the exception of a small sliver bounded by $Q(\text{Å}^{-1}) < 0.51\times10^{-6}$ E (meV). From this figure, it is apparent that inelastic x-ray scattering will be a viable technique if the necessary x-ray energy transfer resolution can be achieved in an instrument with sufficient flux to provide measurable signal rates. The remainder of this paper is devoted to these considerations.

We first address the question of energy resolution. The x-ray diffraction properties of perfect crystals are well known from the dynamical theory[2] and extensive experimental verification.[3] In the absence of absorption, the relative energy resolution of a particular Bragg reflection is given by

$$\Delta E/E = N d_{hk\ell}^2 \left|F_{hk\ell}\right| \qquad (1)$$

where N is the number of atoms per cm^3, and $d_{hk\ell}$ and $F_{hk\ell}$ are the spacing and structure factor respectively of the $hk\ell$ planes. In Fig. 2 the absolute energy resolution of the two classes of Bragg reflections in silicon is shown for the case of near back-scattering ($2\theta = 179°$). The back-scattering configuration provides the minimum absolute energy resolution possible with a given Bragg reflection, since the relative energy resolution $\Delta E/E$ is independent of the photon energy E used. The dynam-

ical theory is applied to the back-scattering geometry and measurements are made to verify its predictions in an article by Graeff and Materlik.[4] The back-scattering geometry is also advantageous for a number of other reasons which will be discussed shortly. From Fig. 2 it is apparent that one achieves the best energy resolution with the higher order Bragg reflections, and therefore with higher energy photons. Although counter-intuitive, this result is essential for the success of the back-reflection technique, since it leads to the requisite millivolt energy resolutions at photon energies which are most convenient (i.e. 10-15 keV).

Starting from Bragg's law, $\lambda = 2d\sin\theta$, one can immediately show that the geometrical bandpass for a monochromating crystal is

$$\Delta E/E = -\cot\theta\Delta\theta. \qquad (2)$$

In this expression $\Delta\theta$ is an angle deviation which can be taken to represent the range of angles of incidence due to beam spread, crystal mosaic distribution, or instability in setting the crystal angle. If the resulting energy width is larger than that given in Eq. 1 above, then the intrinsic resolution will not be achieved. For $\Delta E/E \approx 10^{-7}$, corresponding to $\Delta E = 1$ meV for a 10 keV photon, then $\Delta\theta \approx 10^{-2}$ arc sec if the scattering is carried out at conventional angles where $\cot\theta \approx 1$. Such a monochromator will then suffer from (1) the requirement of 10^{-2} arc sec angular stability; (2) the inability to view the entire photon source which subtends an angle of 10 arc sec, or to collect scattered radiation which will emanate from a sample crystal over considerably larger angles. The solution to these difficulties is to use the back-scattering geometry such that $\cot\theta \approx 10^{-2}$. For example, the use of the Si(880) reflection would give $\Delta E/E = 7.4 \times 10^{-7}$. For $2\theta = 179°$ the reflected pho-

ton energy would be E_i = 12.95 keV while $\Delta\theta$ = 17.5 arc sec. To collect the large solid angles necessary for the analyzer crystal, as well as to collect the horizontal divergence of a wiggler or bending magnet beam, a bent crystal is necessary. For the back-reflection geometry another advantage emerges: the ideal figure is spherical as shown in Fig. 3a. Such a spherical shape is easily and accurately obtained as shown long ago in the context small angle scattering by Hagström and Siegbahn.[5] In the present context a bender has been built [as sketched in Fig. 3(b)] and tested at the Stanford Synchrotron Radiation Laboratory.[6] The necessary figure accuracy was easily obtained.

The two remaining problems to solve are temperature stability and bending-induced strain. The thermal stability requirements are on the order of 10^{-1} to 10^{-2} °K and will require a thermally stabilized enclosure. This sensitivity may offer the advantage of providing an energy scanning mode as an alternate to scanning crystal angle. The final problem of bend-induced strain requires that the surface be mechanically strain-relieved [as shown in Fig. 3(c)], or that its d-spacing be graded as a function of distance into the sample. Some progress has been made on the first suggestion above at Bell Labs by George Kaminsky using anisotropic etching techniques.[7] Conventional fabrication techniques are not adequate, since for the high resolution required the tooth size in Fig. 3(c) is less than 0.2 mm wide. However for poorer resolution larger tooth dimensions are acceptable. Dorner et al.[8] have demonstrated the fabrication of such a crystal. (See paper by B. Dorner in these proceedings.)

The use of backscattering for both the monochromator and the analyzer of the high-energy-transfer-resolution spectrometer has been discussed above. The resolution of the instrument is determined by both the intrinsic resolution of the Bragg reflection used and the relevant angular divergences. Thus,

$$\frac{\Delta E}{E} = Nd^2F + \cot\theta\Delta\theta \qquad (3)$$

where the two terms on the right hand side are from Eqs. (1) and (2) respectively, as shown by Birr et al.[9] The best compromise is achieved by matching the intrinsic resolution with the term incorporating the divergence. To do so with Si(880) reflection, which gives $\Delta E \approx 10$ meV, requires a source size of 0.2 x 0.2 mm^2 for a source to monochromator distance of 2000 mm. Since this source size and distance are both considerably smaller than the relevant synchrotron parameters, an optical system is required to provide an image of the source at the entrance slit of the high-resolution monochromator.

The ideal optical system would (1) accept the full spatial extent of source, (2) fill but not overfill a 0.2 x 0.2 mm^2 aperture at the entrance to the high resolution spectrometer and (3) have an energy bandpass that accepts all photons with energies in the range $\Delta E/2 \leq E_0 \leq \Delta E/2$. Obviously all systems should accept as much horizontal angle as possible, typically a few milliradians with the new wiggler magnet sources. Three possible choices are discussed below (1) an 8:1 demagnifying ellipsoidal mirror; (2) two paraboloidal mirrors and a premonochromator and (3) a multilayer Kirkpatrick-Baez mirror pair.

The ellipsoidal mirror, shown schematically in Fig. 4(a) is the simplest system. At 8:1 demagnification it focuses an 1.6 x 0.5 mm^2 source onto a 0.2 x 0.2 mm^2 aperture. Ray tracing studies of such a mirror with 1 mrad horizontal acceptance show that almost no radiation falls outside the aperture. Thus this mirror would be an ideal choice. Since a Pt coating would provide a broad bandpass ($E_c \approx 16$ keV at 5 mrad grazing angle) criterium (3) would also be satisfied. The only drawback to such a system is that the total heat load of the source below 16 keV would impinge on the high-resolution monochromator.

The second system, sketched in Fig. 4(b) consists of two paraboloidal mirrors with a pre-monochromator in between them. The first mirror collimates the source and, by making the radiation more parallel allows the pre-monochromator with asymmetric Si(111) crystals to pass easily all photons in the high resolution bandwidth. The second mirror images the source onto the 0.2 x 0.2 mm^2 aperture. In a 1:1 geometry, this system overfills the aperture. However what is lost in spot size is made up by the increased horizontal acceptance (3-5 mrad) possible with the paraboloidal mirrors. Furthermore this optical system absorbs the thermal load at the pre-monochromator which is a considerably simpler device to cool.

The third system employs a concept first proposed by Kirkpatrick and Baez[10] as an optical system for an x-ray microscope. Two crossed mirrors that are cylindrical (spherical for our geometry) provide independent focusing in the vertical and horizontal planes. In our scheme shown in Fig. 4(c) the first mirror has a multilayer tungsten-carbon coating[11] and the second a Pt coating. This coating serves two purposes: (1) it in-

creases the grazing angle over that for a Pt reflector and (2) it monochromates with a modest resolution proportional to the number of layers. The increased angle provides a larger horizontal acceptance. This design also overfills the 0.2 x 0.2 mm^2 aperture, but as with the paraboloidal mirror design the increased horizontal acceptance compensates for the larger image. This system also removes the heat load from the high resolution monochromator and, as can be seen from the figure, is simpler than the pair of paraboloids.

Summarizing these systems, if the heat load on the high resolution monochromator can be tolerated, then the single ellipsoid is an ideal choice to match the source the spectrometer for both ease of operation and throughput. However if the heat load is intolerable then systems (b) or (c) are more appealing, with the final choice dependent on the performance of multilayers.

It is clear that the achievement of an optically efficient beam line for millivolt resolution inelastic x-ray scattering is a substantial technical challenge, and we are optimistic that it can be met. However, we note that any substantial improvement over the current level of about 1 eV energy resolution will result in important new opportunities for experimentation. Although we have concentrated the above discussion on a 10 meV instrument, considerable simplification results if one designs for 100 meV resolution. Finally mention should be made of the possibility of penetrating into the submicrovolt regime using nuclear resonance scattering.[12] These techniques seem ideally matched to x-ray undulator sources and will probably await the next generation of dedicated high-energy storage rings for their full exploitation.

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Figure Captions

Fig. 1. An illustration of the energy and momentum transfer regimes accessible using x-ray and neutron probes to study excitations by inelastic scattering. For 14 meV and 100 meV neutron energies, the accessible region is located within the respective inverted parabolas. For a 14.4 keV x-ray, virtually the entire energy-momentum plane is accessible except for a small region $Q(\text{Å}^{-1}) \leq 0.51 \times 10^{-6}$ E (meV).

Fig. 2. The absolute energy resolution versus the back-scattering ($2\theta \approx 179°$) photon energy for the two classes of Bragg reflections in silicon.

Fig. 3. The back-reflection monochromator. (a) Schematic illustration of a spherically bent strain-relieved back-scattering monochromator; (b) the bending concept (see Ref. 5); (c) the structure required for strain-relief of the crystal surface.

Fig. 4. Schematic diagrams of possible optical systems for a high energy resolution spectrometer. The spectrometer consists of a back-scattering monochromator and analyzer shown in panel (a). Panel (a) shows a 8:1 demagnifying ellipsoid matching the source to the monochromator. Panel (b) shows a double paraboidal mirror system imaging the source 1:1 with a pre-monochromator between the two mirrors. Panel (c) shows elevation and plan views of a Kirkpatrick-Baez mirror pair. The first, horizontally deflecting mirror has a multilayer coating.

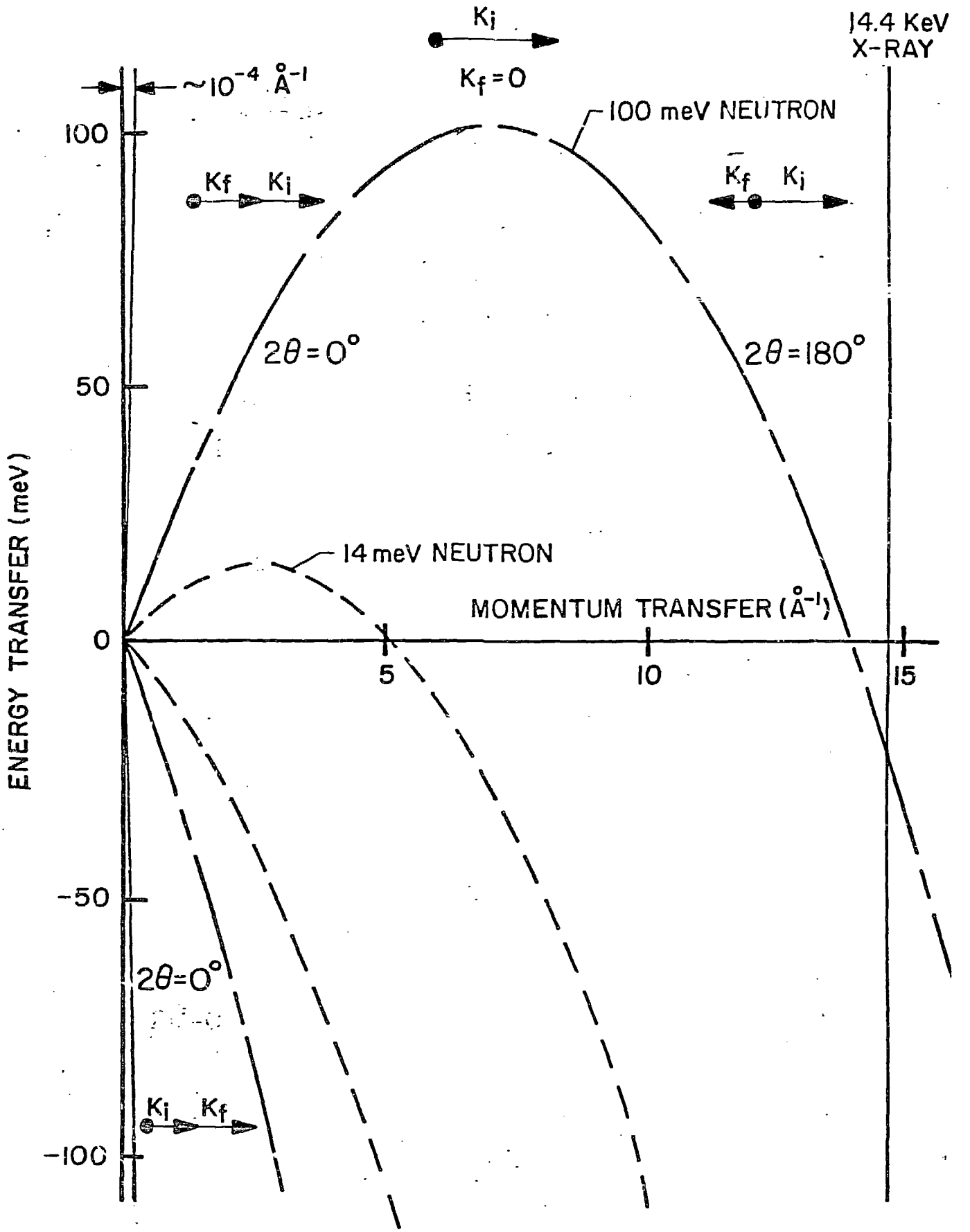
Ki
Kf = 0
14.4 KeV
X-RAY
~10^-4 Å^-1
100 meV NEUTRON
100
Kf Ki
Kf Ki
2θ = 0°
2θ = 180°
50
14 meV NEUTRON
MOMENTUM TRANSFER (Å^-1)
ENERGY TRANSFER (meV)
0
5
10
15
-50
2θ=0°
Ki Kf
-100

Figure 1

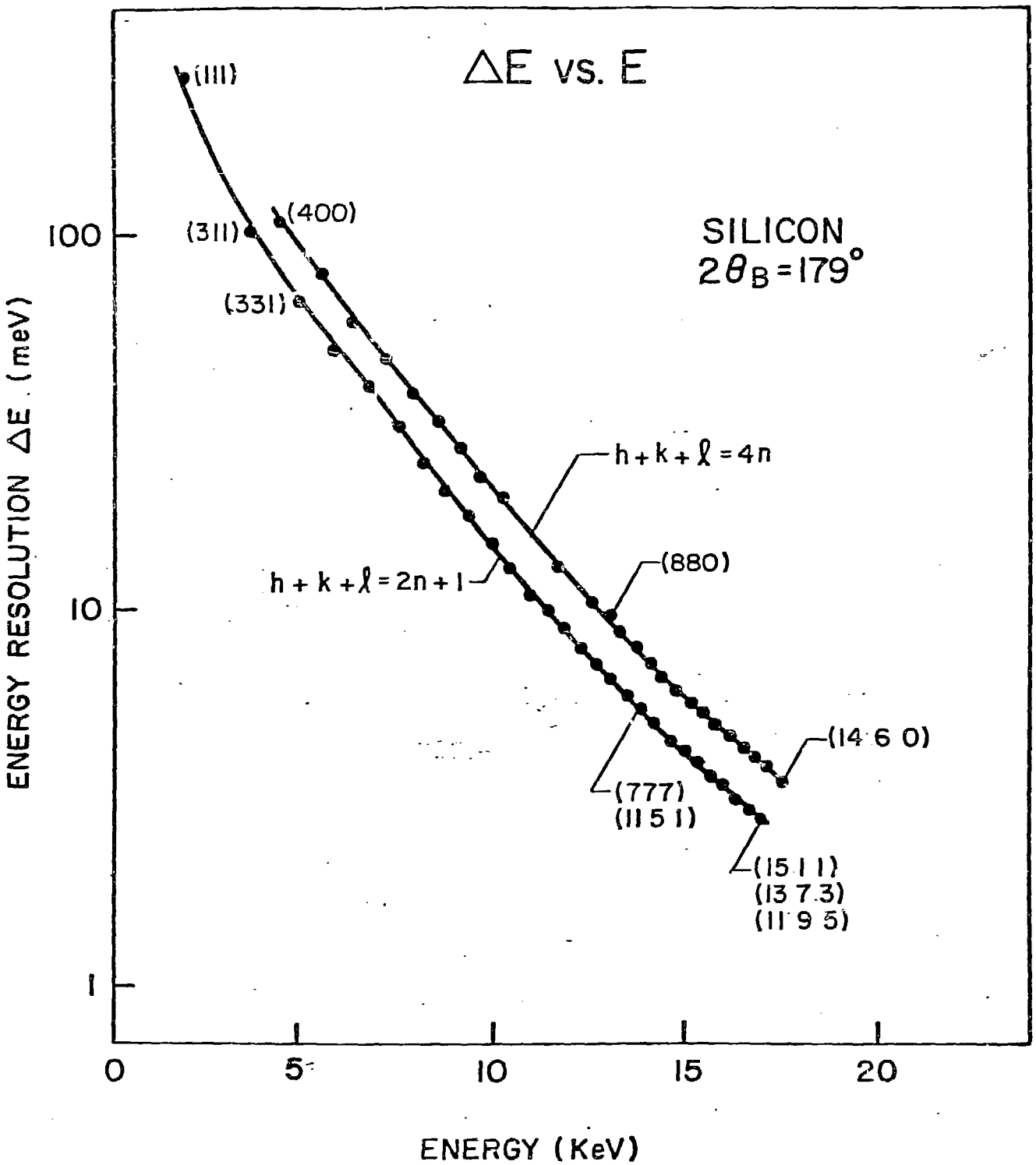
ΔE vs. E
SILICON
2θB = 179°
(111)
(400)
(311)
(331)
h + k + ℓ = 4n
(880)
h + k + ℓ = 2n + 1
(14 6 0)
(777)
(11 5 1)
(15 1 1)
(13 7 3)
(11 9 5)
ENERGY RESOLUTION ΔE (meV)
100
10
1
0
5
10
15
20
ENERGY (KeV)

Figure 2

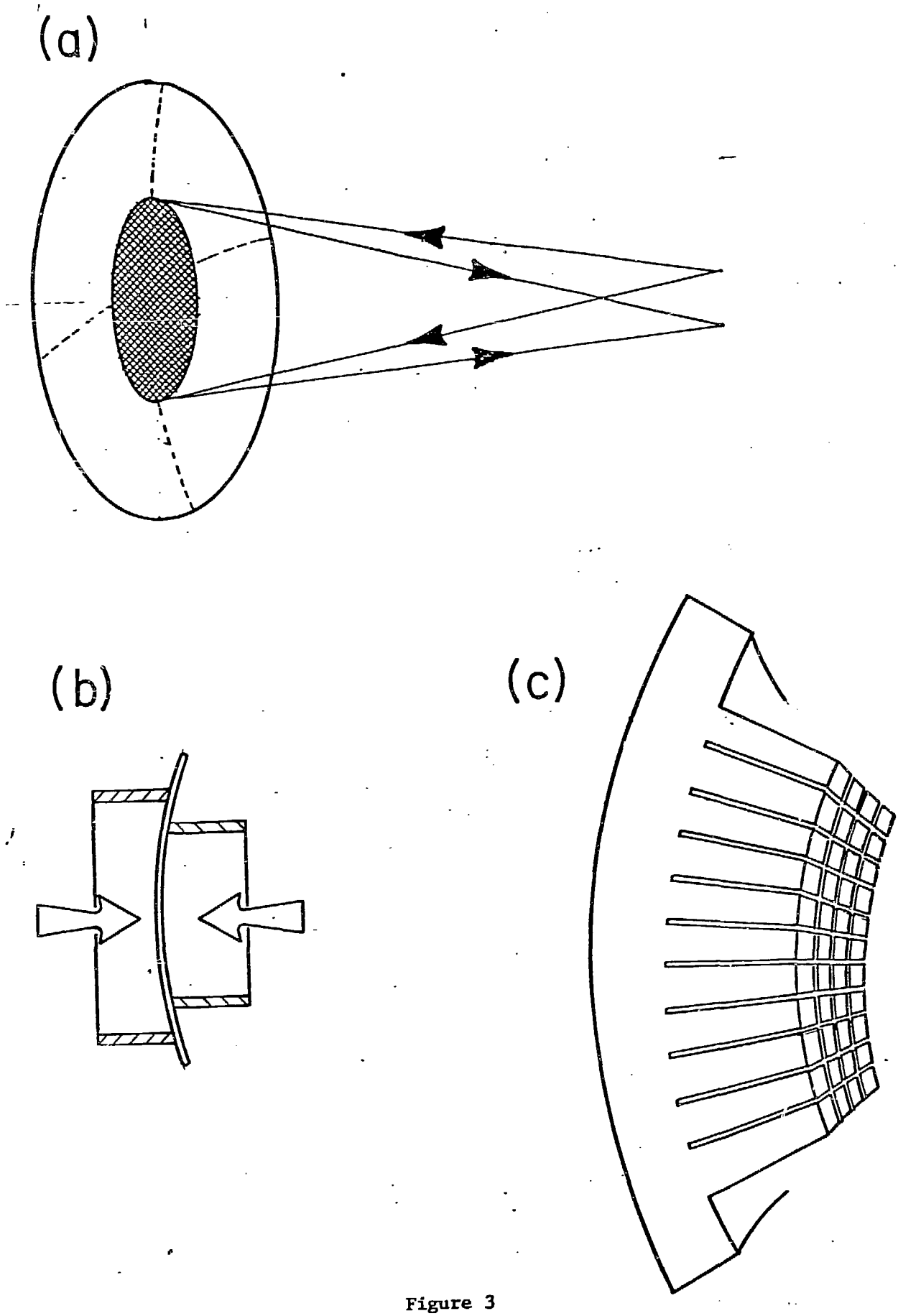

Figure 3

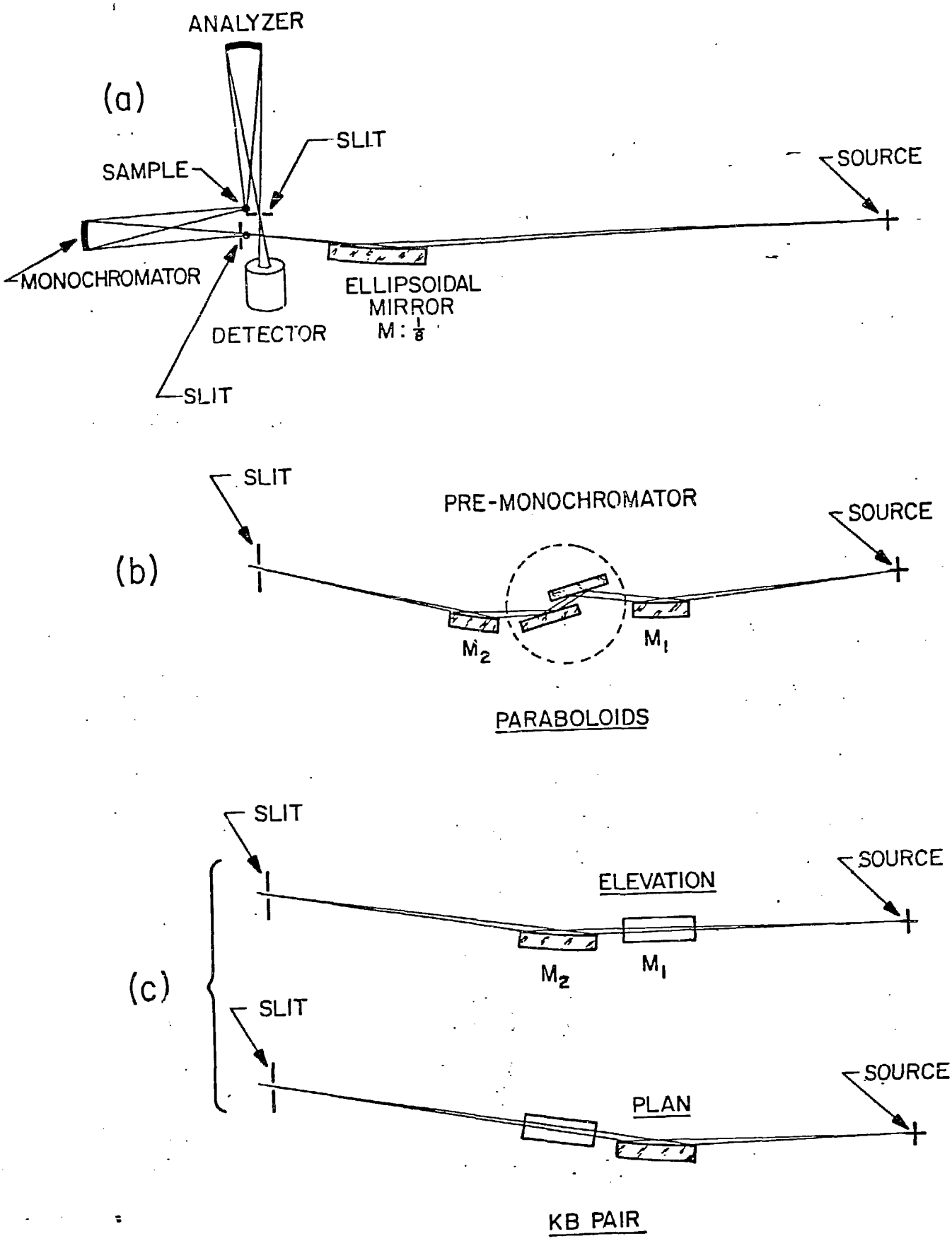
ANALYZER
(a)
SLIT
SAMPLE
SOURCE
MONOCHROMATOR
ELLIPSOIDAL
MIRROR
M : 1/8
DETECTOR
SLIT
SLIT
PRE-MONOCHROMATOR
SOURCE
(b)
M2
M1
PARABOLOIDS
SLIT
SOURCE
ELEVATION
(c)
M2
M1
SLIT
SOURCE
PLAN
KB PAIR

Figure 4