OPPORTUNITIES FOR INELASTIC X-RAY SCATTERING FROM ELECTRONIC EXCITATIONS AT THE ERL:

ANALYZER LIMITATIONS

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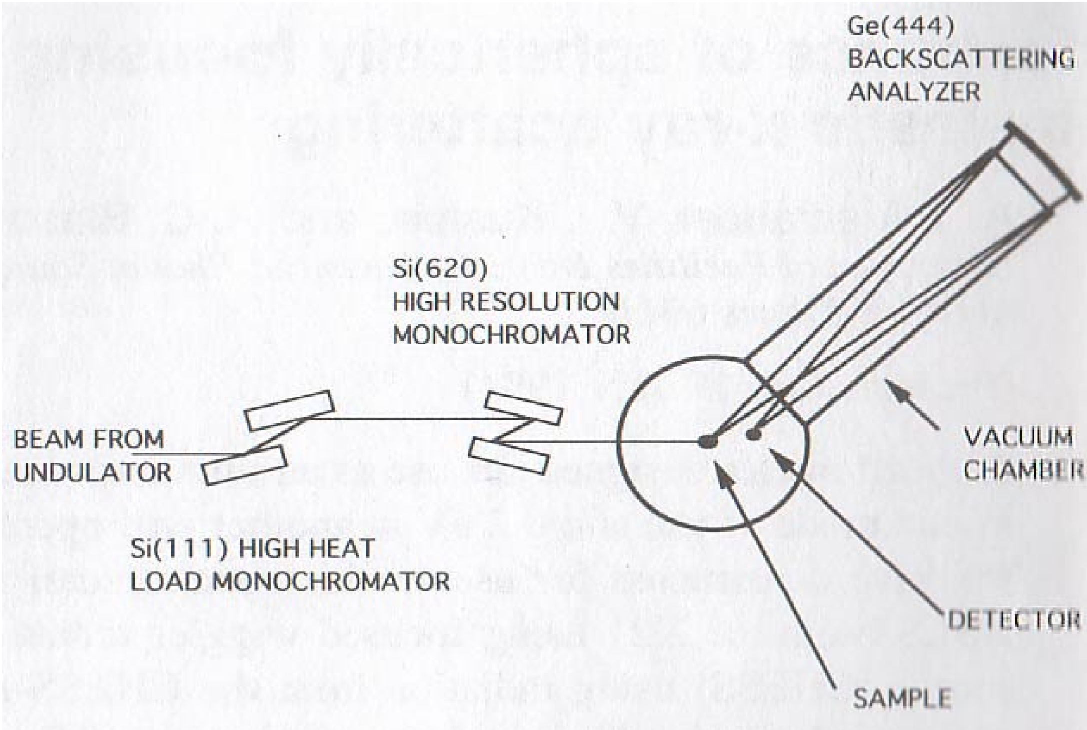
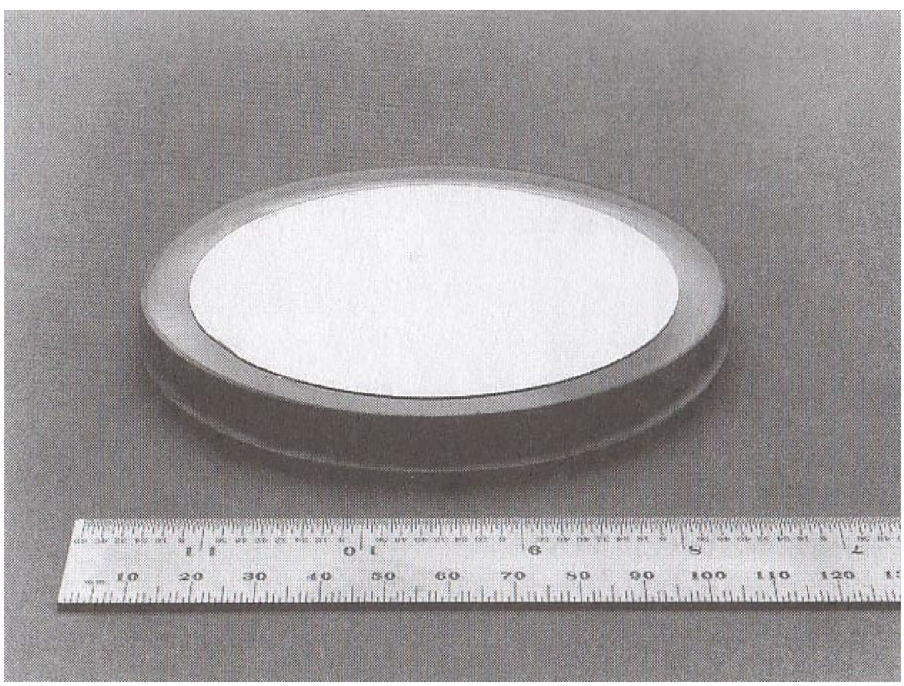
The study of electronic excitations by inelastic x-ray scattering (IXS-E) is hampered by extremely low scattering cross sections and by large x-ray absorption in high Z materials. Synchrotron studies for both non-resonant and resonant IXS-E are examined for their limitations, and it is concluded that the main limitations are set by the incident beam brilliance and by the efficiency of a momentum resolving analyzer. The high brilliance at high energies projected for the ERL will provide opportunities for new analyzers. Possible new directions for analyzer development will be charted.

Energy Recovery Linac Science Workshop, Cornell, Dec. 2-3, 2000





A.T. Macrander , P. A. Montano, D.L. Price, V. I. Kushnir,
R.C. Blasdell, C.C. Kao, B.R. Cooper, PRB 54, 305 (1996)
Ti and TiC

P.A. Montano and A.T. Macrander, J. Phys. Chem. Sol. 61, 415 (2000)
Cr

Performance of spherically focusing Ge(444) backscattering analyzers for inelastic x-ray scattering

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(Presented on 18 July 1994)

A spectrometer designed for use as an undulator source and having targeted resolutions of 0.01 eV in one mode of use and 0.2 eV in another will operate at the APS. We report here on analyzers that we have constructed for use on this spectrometer for 0.2 eV resolution. We have tested them at NSLS beamline X21 using focused wiggler radiation and at the Cornell high energy synchrotron source (CHESS) using radiation from the CHESS-ANL undulator. Analyzers were constructed by gluing and pressing 90-mm-diam, (111) oriented Ge wafers into concave glass forms having a radius near 1 m. An overall inelastic scattering resolution of 0.3 eV using the (444) reflection was demonstrated at CHESS. Recent results at X21 revealed a useful diameter of 74 mm at an 87° Bragg angle. © *1995 American Institute of Physics.*

I. INTRODUCTION

Since the early 1980's, inelastic x-ray scattering (IXS) has been considered both a desirable and achievable investigative tool with which to attack some of the fundamental problems in condensed matter physics.[1–4] Unlike inelastic neutron scattering, IXS is almost completely unhampered by the kinematic constraints arising from conservation of energy and momentum during the scattering process, and, unlike inelastic electron scattering, x rays penetrate the bulk sufficiently to rule out anomalies due to surface properties. However, cross sections for IXS are extremely small (e.g., 10^{-27} cm^2/eV for the double differential cross section of the plasmon in aluminum[5]) which implies that synchrotron radiation sources should be employed to obtain useful data. With the advent of the APS,[6] which is a third-generation synchrotron source, we expect that sufficiently great incident photon fluxes can be delivered and that spectra of inelastically scattered photons with reasonably good statistics for a wide range of samples can be obtained in scans lasting several hours.

The beamline optical layout will consist of a high-heat-load monochromator followed by a high-resolution monochromator. The diffuse scattering from a sample will be collected by a spherically focusing crystal analyzer.

II. BACKSCATTERING

Backscattering for the analyzer is a propitious geometry because (1) angular Darwin widths become very large, and (2) the derivative of the energy with respect to angle in Bragg's law goes to zero.[7] The consequence of (1) is that the angular acceptance of analyzers increases, and the consequence of (2) is that the resolution becomes less dependent on angular divergence. Because of absorption in the sample, it is favorable to work at as high an energy as possible. Backscattering from silicon at higher energies occurs from high-order reflections [e.g., (777)], which leads to resolutions of several meV. A spectrometer with a 3-m-long two-theta arm designed to employ high-order reflections from a silicon analyzer is under construction on sector 3 of the Synchrotron Radiation Instrumentation (SRI) Collaborative Access Team (CAT) at the APS. A resolution of several meV is useful for the study of phonons. However, it is not needed for the study of electronic excitations and results in an unnecessary band-pass collection penalty in that case.

III. Ge(444) ANALYZER

We have achieved a coarse energy resolution of several hundred meV by using a lower-order reflection [i.e., (444)] from an analyzer made of Ge with a focusing distance of 1 m. At this radius, neither the intrinsic energy width nor the efficiency of a bent Ge wafer is significantly reduced compared to the unbent case. This is demonstrated in Fig. 1 which is the result of a dynamical diffraction simulation.[8] A calculated analyzer resolution of 94 meV FWHM was obtained. Just as in the unbent case, the Darwin width in energy is almost independent of Bragg angle near backscattering.

The most successful procedure we have found to construct an analyzer is (1) first prepare a two-component epoxy mixture by pumping the air out of the mixture in a bell jar,

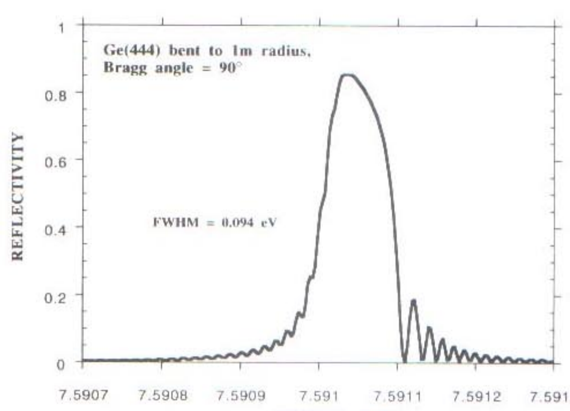

FIG. 1. Reflectivity as calculated using a dynamical matrix simulation method.

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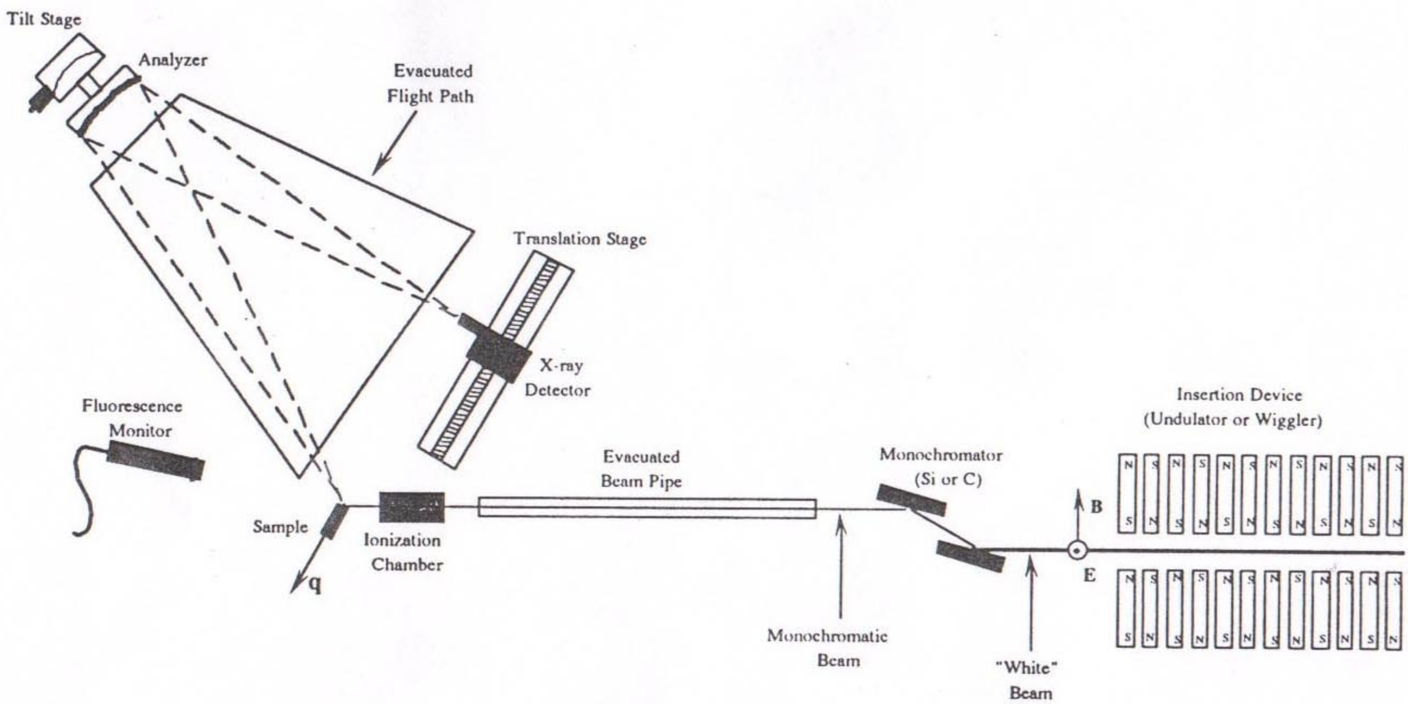

Figure 2.1: Schematic of an x-ray Raman spectromter.

P.M. Abbamonte, Ph.D. Thesis, U. of Illinois at U-C, 1999.

P.Abbamonte,C.A. Burns, E.D. Isaacs, P.M. Platzman, L.L. Miller, S.W. Cheong, M.V. Klein, PRL 83, 860 (1999)

La_2CuO_4 and $Sr_2CuO_2Cl_2$

Prototypical transition metal oxide:

V_2O_3

- Highly correlated electron system
- Mott-Hubbard system
- Metal insulator transition

E.D. Isaacs, P.M. Platzman, P. Metcalf, J. Honig, PRL 76,4211(1996)

ABSORPTION LENGTH AT 22.86 keV:

0.5 cm

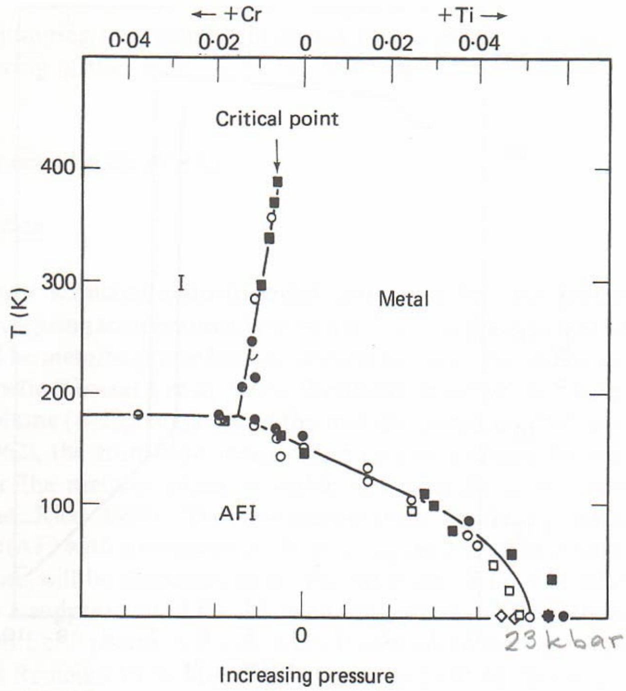

Fig. 6.3 Generalized phase diagram of the metal–insulator transition in V_2O_3 as a function of doping with Cr or Ti and as a function of pressure, showing the critical point (McWhan *et al.* 1971).

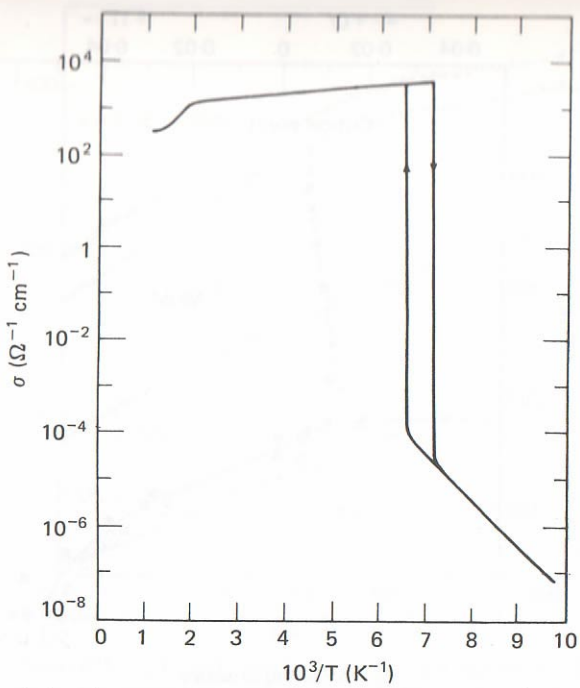

Fig. 6.2 Conductivity of a single crystal of V_2O_3 as a function of $1/T$, showing the metal–insulator transition (Föex 1946).

N.F. Mott, "Metal-Insulator Transitions", 2nd Ed., 1990

PHYSICAL REVIEW B VOLUME 19, NUMBER 2 15 JANUARY 1979

Curved-crystal x-ray monochromator efficiency

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This paper reviews factors relevant to the choice of a curved crystal for monochromatizing and focusing x rays efficiently. Results of numerical computation of reflectance by symmetrically curved ideally perfect crystals in several wavelength ranges are given. Some dimensionless numbers are defined that help to estimate reflectance without rigorous numerical computations in many cases.

I. INTRODUCTION

Details of the geometrical and physical optics of symmetric ideally perfect curved-crystal x-ray monochromators were recently described in two papers.[1,2] This paper will review a few highlights, add more quantitative results, and introduce some approximate formulas that may give a better qualitative understanding of the effects of varying the x-ray wavelength, the crystallographic parameters, and the radius of curvature of such monochromators. For very absorbing crystals and crystals with very short radii of curvature the reflected intensity approaches that for ideally mosaic crystals, while the usual dynamical theory for ideally perfect flat crystals applies for perfect curved crystals with very long radii. Examples of these extreme cases and of intermediate cases are given in this paper.

To obtain monochromatic line-to-line focusing, a set of confocal eliptical cylinders, as shown in cross section in Fig. 1, are the optimal loci and spacing of "Bragg planes." For point-to-point focusing the "planes" are also figures of revolution about the line between foci. The loci of planes of equal spacing are along circles through the two foci called "Rowland circles."[3] A reasonable volume of multilayer evaporated films conforming to these elliptical or ellipsoidal surfaces might, in principle, be fabricated but it is possible to come very close using a thin wafer of ideally perfect crystal such as silicon, germanium, or quartz, figured and then elastically bent tangent to a Rowland circle.[4] One way to do this for symmetric point-to-point focusing with unit magnification is shown in Fig. 2, where exact conformity to a Rowland circle is sacrificed in order to obtain a large solid angle through the use of a developable surface geometry.[1,5,6] The same bending geometry could be used in an asymmetric geometry but then different parts of the crystal would reflect a broader range of wavelengths, since they would intersect more widely separated Rowland circles.

II. NUMERICAL RESULTS

In the symmetric geometry, where the crystal planes are approximately parallel to the crystal surface, reflection of a flat or curved crystal can be computed using a modification of Abelés's 2×2 matrix formulation of the reflection and transmission problem for layered structures.[2,7] This method was used to compute the reflection spectra for various wavelengths with various Bragg planes of high-structure factor in quartz, silicon, and germanium. Experimental measurements of integrated reflectance by two different symmetric point focusing monochromators for Cu$K\alpha$ radiation made with very perfect quartz agree quite well with the computed values.[1,6]

The computed spectra are characterized by a main peak that is more or less like the Darwin band for a flat crystal. If the crystal radius is large that is all there is. If the radius is short planes below the first surface reflect at an angle that is enough larger than the angle at the surface that the wavelength of the center of the Darwin band there is appreciably longer than at the surface. This results in an exponentially diminishing tail on the long-wavelength side of the main reflec-

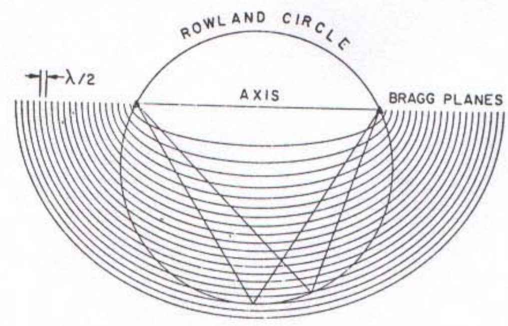
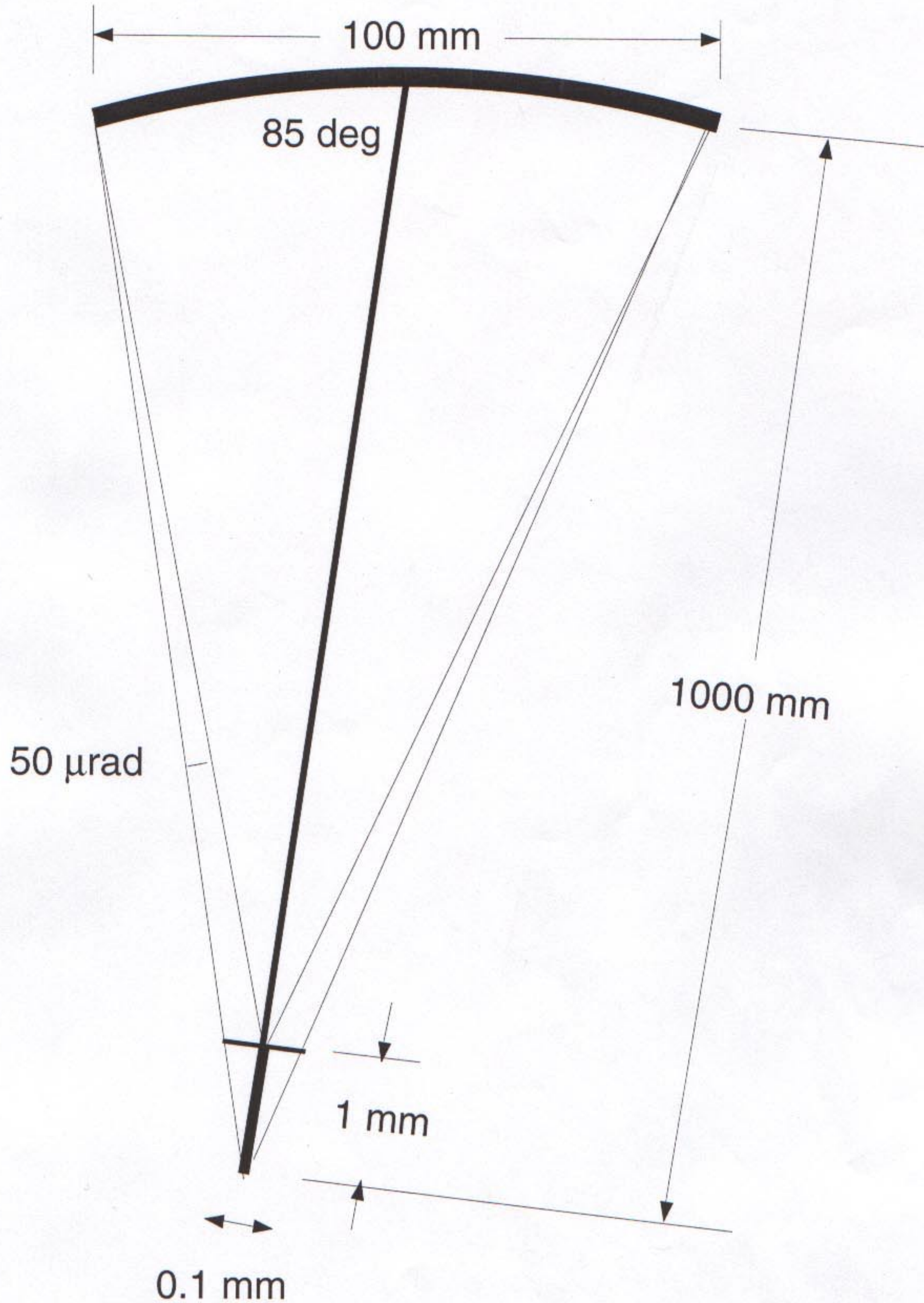
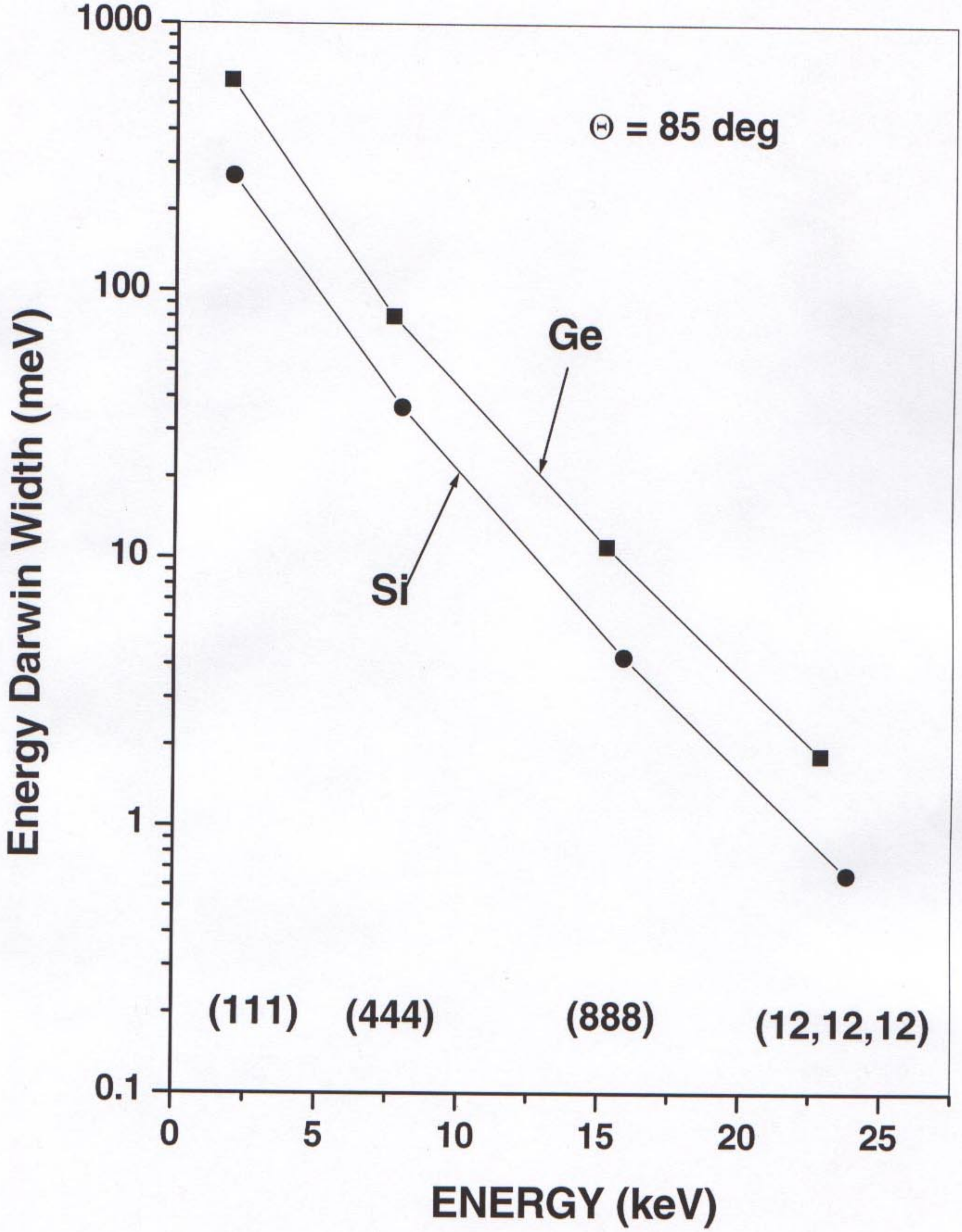
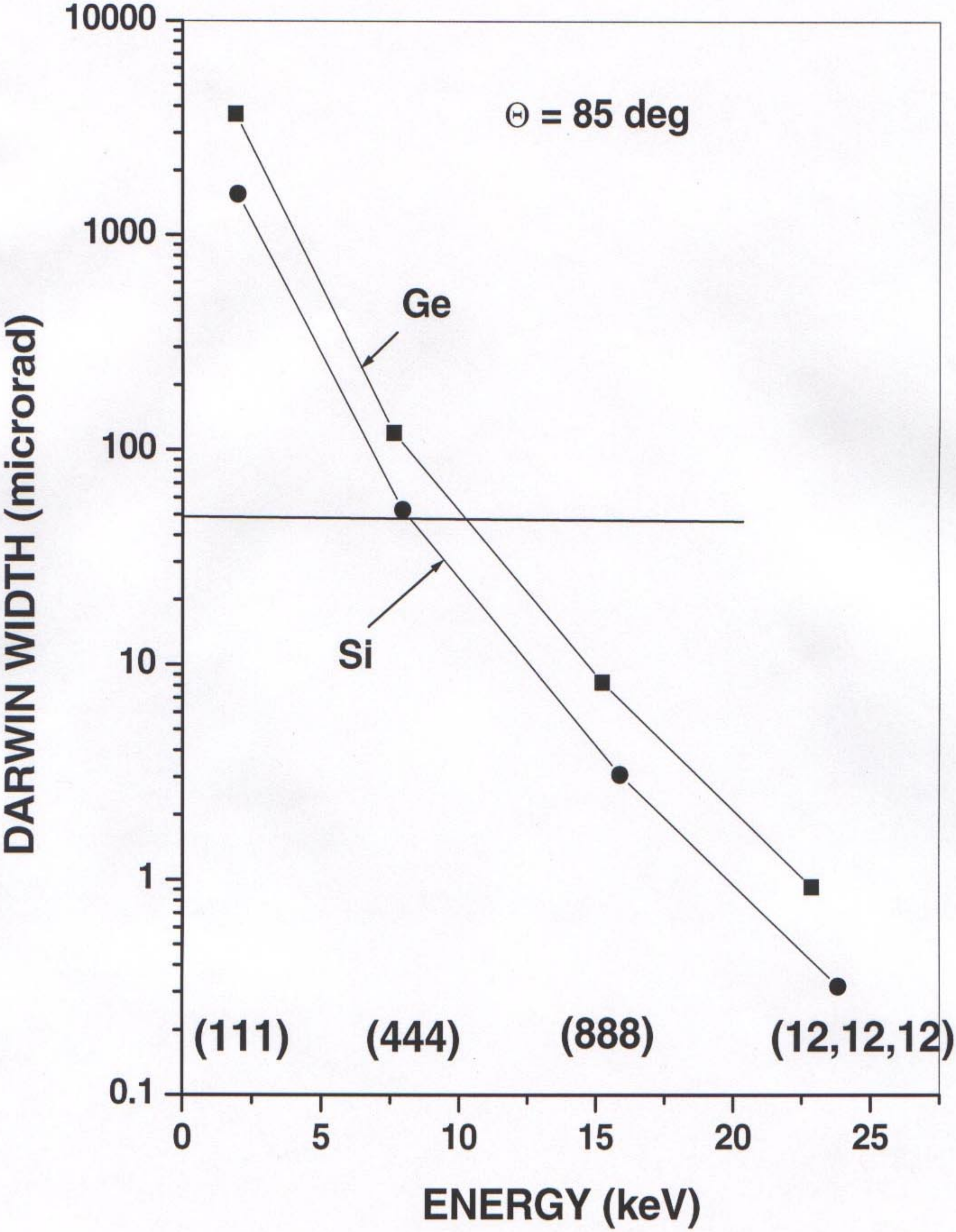


FIG. 1. Cross section of ideal ellipsoidal geometry for curved variable-spacing "Bragg planes" to give monochromatic point-to-point focusing. Spacing is constant along any circular arc terminating at conjugate foci.

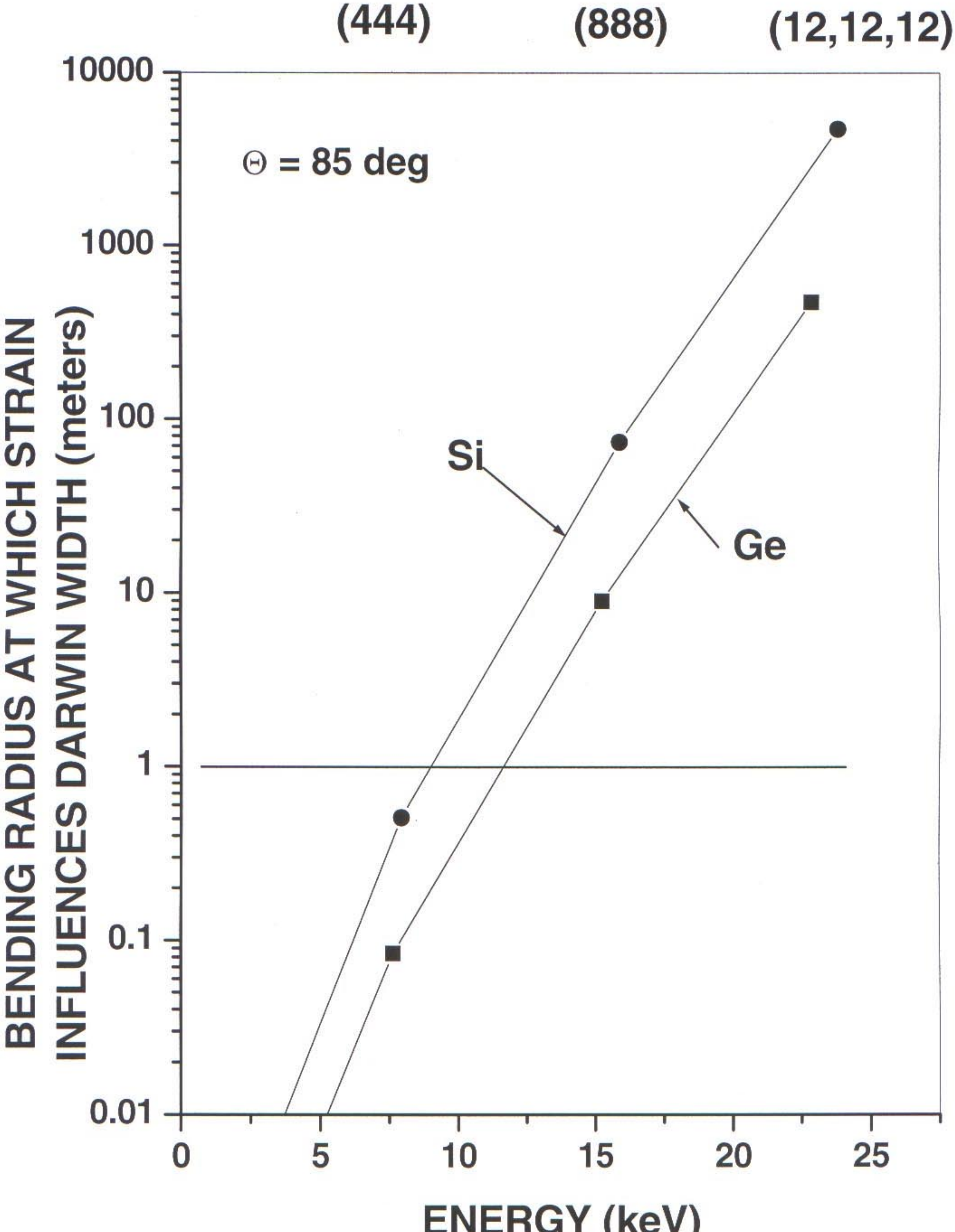

100 mm
85 deg
1000 mm
50 µrad
1 mm
0.1 mm



V.I. Kushnir : REFLEX



V.I. Kushnir : REFLEX



V.I. Kushnir : REFLEX

V.I. Kushnir and A.T. Macrander , NIM A 347, 331 (1994)

HOPG (0,0,16)
$c = 0.6708$ nm

$\gamma = 0.25$ deg

$\theta = 85$ deg
$E = 14.8$ keV

$\Delta E = E\gamma/\tan\theta =$ **5.6 eV**
$1/\mu = 5$ mm

A. K. Freund, NIM A 266, 461 (1988)

CRYSTAL ANALYZER FOR INELASTIC SCATTERING FROM ELECTRONIC EXCITATIONS: BETWEEN A ROCK AND A HARD PLACE

GOOD : ABSORPTION IN SAMPLE GOES DOWN WITH HIGHER ENERGY
BAD: ANGULAR DARWIN WIDTH GOES DOWN WITH HIGHER ENERGY

GOOD: ANGULAR DARWIN WIDTH GOES WAY UP AT BACKSCATTERING
BAD: SAMPLE AND DETECTOR CAN'T BE IN SAME PLACE; CAN'T DO RESONANCE IXS

GOOD: ANALYZER SHOULD BE CLOSE TO SAMPLE FOR SOLID ANGLE
BAD: BACKGROUND NOISE GOES UP FOR SAMPLE NEAR DETECTOR

OPPORTUNITIES AT ERL

HIGH ENERGY:
LOW ABSORPTION FOR HIGH Z MATERIALS

HIGH FLUX:
IXS CROSS SECTIONS ARE VERY SMALL

LOW DIVERGENCE:
FOCUSING NEEDED TO GET OFF BACKSCATTERING

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VOLUME 83, NUMBER 4 PHYSICAL REVIEW LETTERS 26 JULY 199

Resonant Inelastic X-Ray Scattering from Valence Excitations in Insulating Copper Oxides

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Inelastic x-ray scattering from TiC and Ti single crystals

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Inelastic X-Ray Scattering Study of the Metal-Antiferromagnetic Insulator Transition in V_2O_3

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Inelastic X-ray scattering study of Cr(110):
from low momentum transfer to the Compton scattering limit

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NSLS, X21, Kao et al.