THE BONSE-HART

USANS TECHNIQUE

FOR NEUTRONS

Michael Agamalian[a] and
Irina Pozdnyakova[b]

a) Oak Ridge National Laboratory, USA

b) Lebedev Physics Institute, Russia

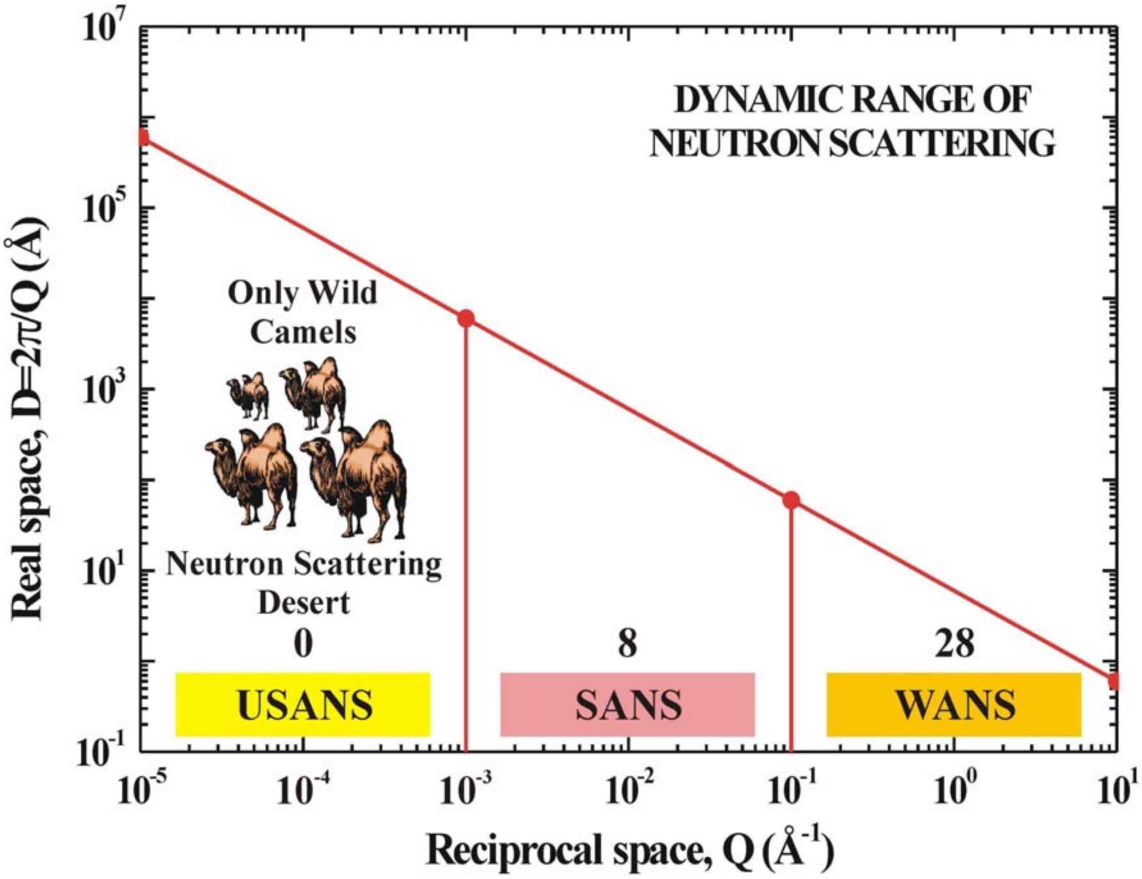
We present an overview of the most significant experimental and theoretical studies made in the past two decades, which resulted in adoption of the Bonse-Hart USAXS technique for neutrons.

Having over four orders of magnitude smaller absorption neutrons penetrate much deeper inside condensed matter revealing “internal” dynamical diffraction effects “invisible” for X-rays.

These effects have not been studied before more or less systematically, thus a serious experimental and theoretical research has been done to make performance of the USANS instrument comparable to its X-ray analog.

Application of the Bonse-Hart USANS instruments clearly demonstrated a real breakthrough to the µm-scale range of the neutron diffraction structural analysis. The new instrument extended a total dynamical Q-range of the neutron scattering by two orders of magnitude.

BEFORE 1996 THE USANS TECHNIQUE HAD LIMITED APPLICATION ONLY DUE TO THE LOW SIGNAL-TO-NOISE RATIO

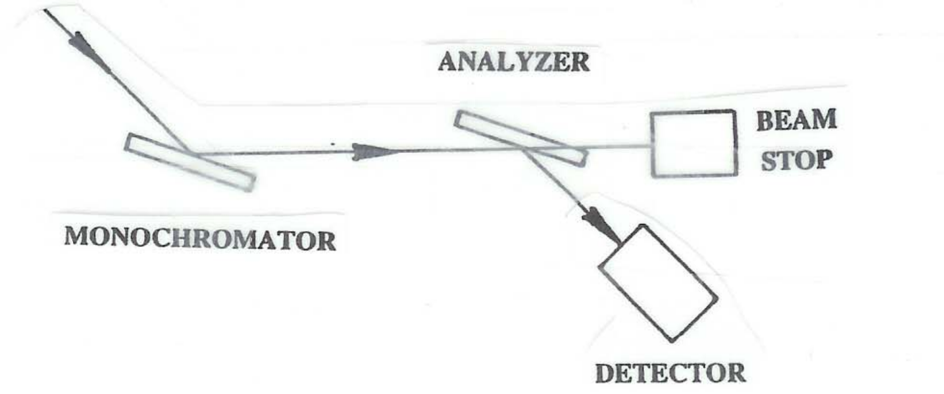


THE USANS RESOLUTION WAS JUST "AS GOOD AS IT GETS" FROM THE DARWIN THEORY:

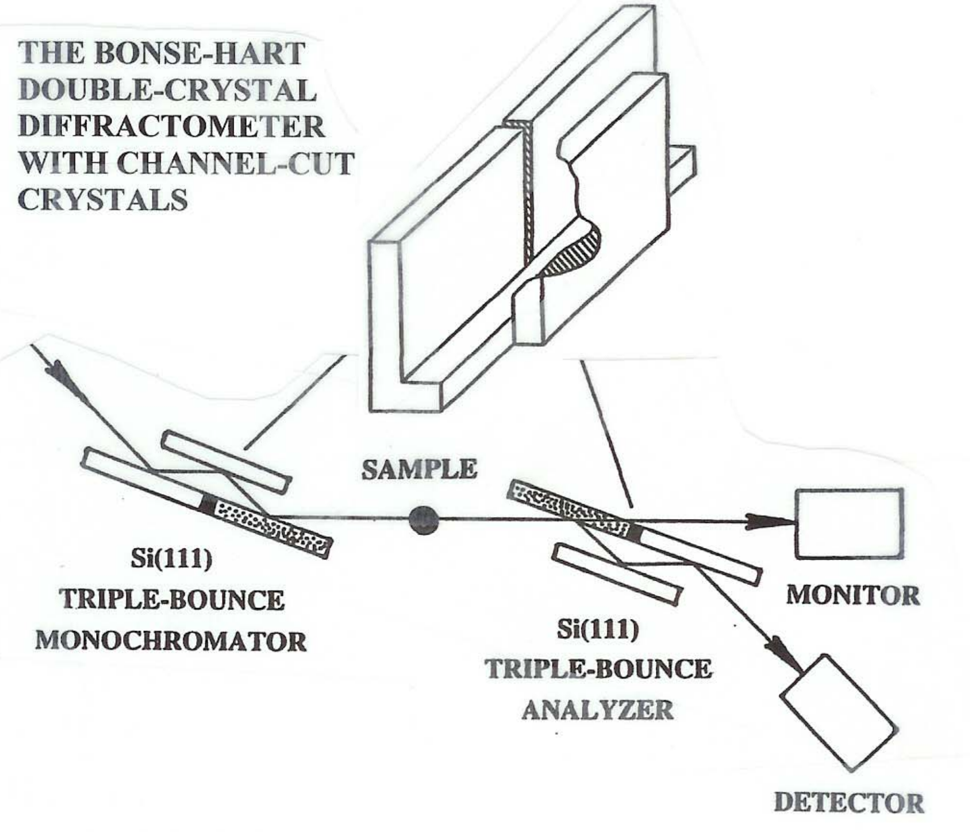
$$Q_{min} \approx 4\pi\delta\theta_D/\lambda \sim 2*10^{-5}\ \text{Å}^{-1}$$

THE BONS-HART DCD HAS PRACTICALLY THE SAME RESOLUTION, HOWEVER IMPROVES DRAMATICALLY THE SIGNAL-TO-NOISE RATIO

THE DOUBLE-CRYSTAL PARALLEL BRAGG ARRANGMENT

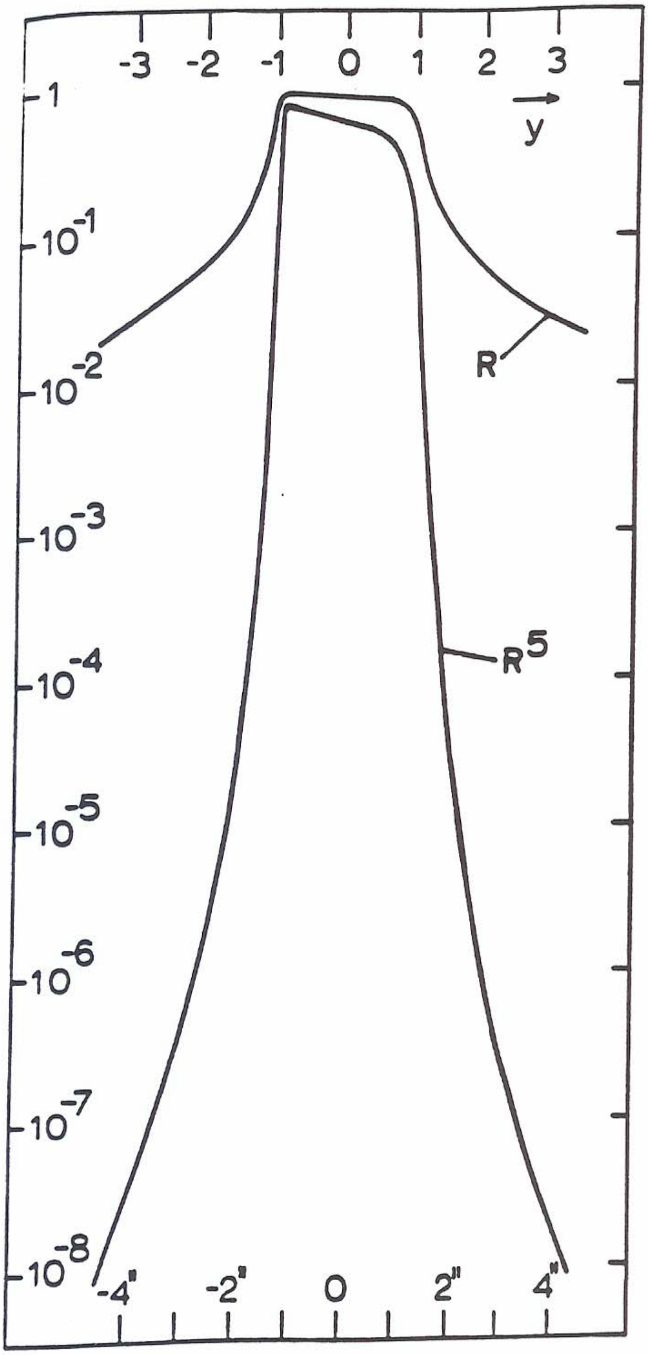


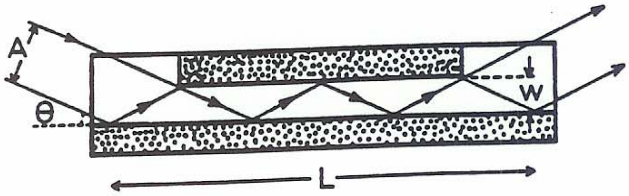
THE BONSE-HART DOUBLE-CRYSTAL DIFFRACTOMETER WITH CHANNEL-CUT CRYSTALS

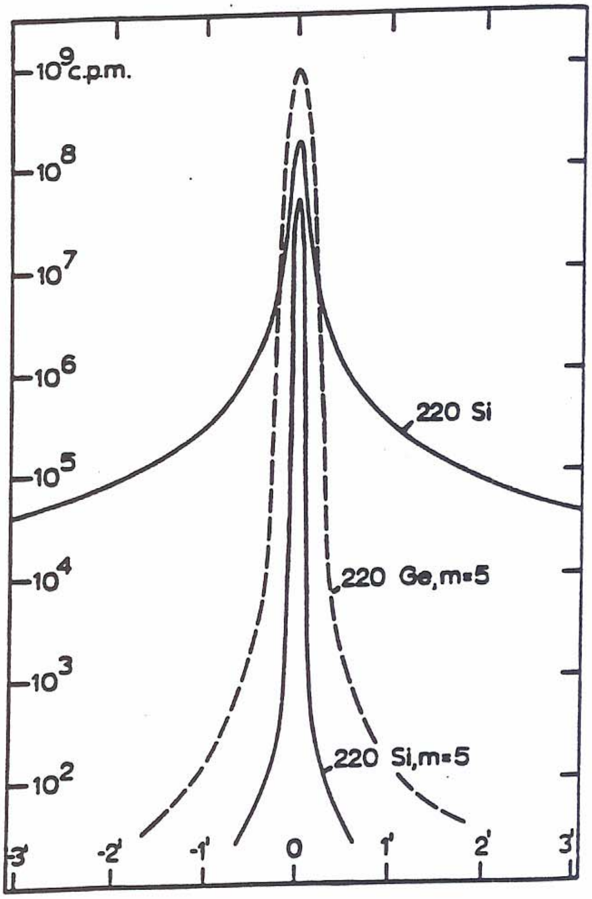


U. BONSE & M.HART

Cornell University, 1965







The best result obtained for Si(220): $I(Q=1.7\bullet10^{-4}\ \text{Å}^{-1})/I(0) \sim 10^{-5}$

THE RESULTS OBTAINED BY U. BONSE & M. HART IN 1965 ARE BASED ON THE FOLLOWING THEORETICAL COSIDERATION OF THE ROCKING CURVE FROM A DCD WITH CHANNEL-CUT CRYSTALS

The rocking curve is a convolution:

$$R(\Delta) = \int R_D{}^n(y) \cdot R_D{}^m(y + \Delta)\, dy,$$

where n and m are the numbers of reflections in the monochromator and analyzer channel-cut crystals correspondingly and $R_D(y)$ is the Darwin reflectivity function:

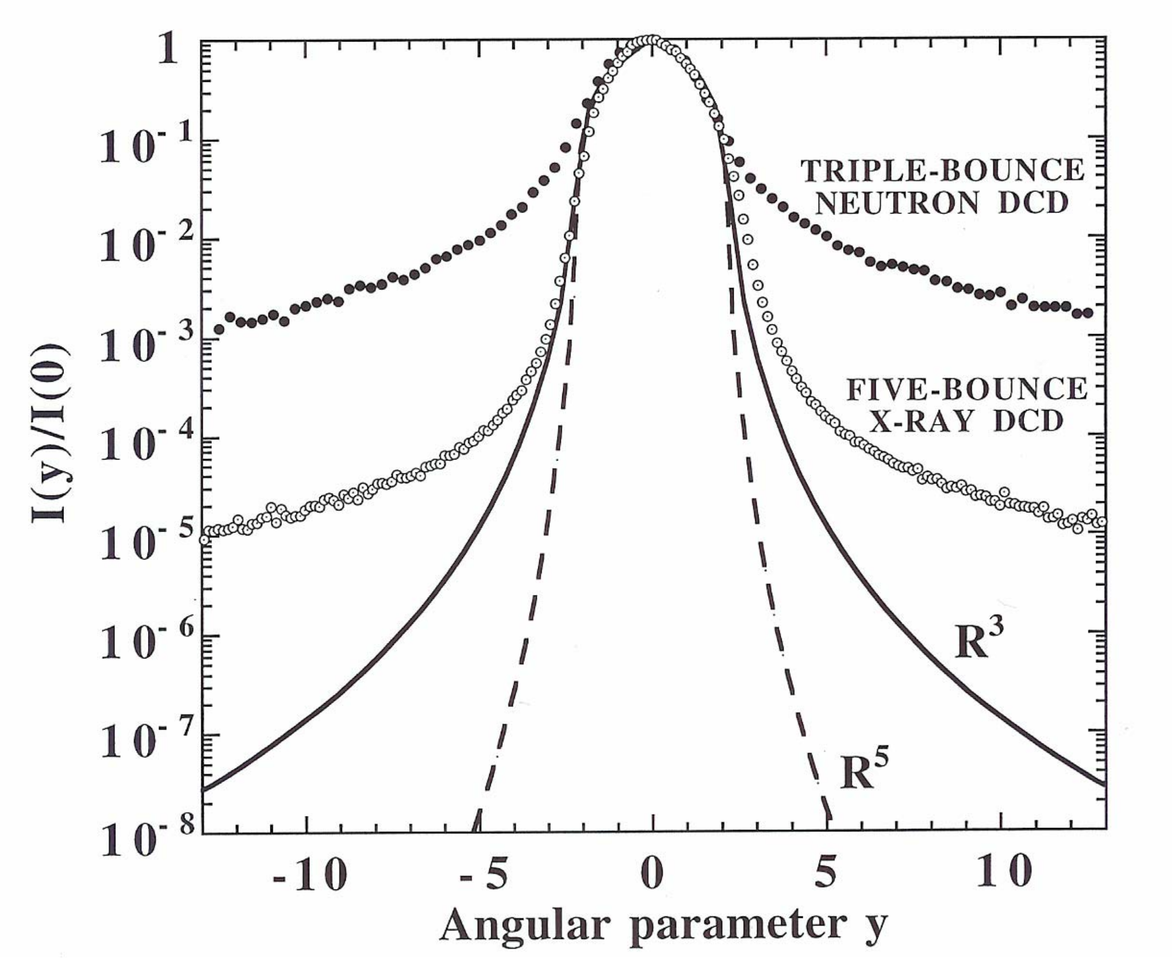
$$R_D(y) = 1,\ |y| < 1,$$

$$R_D(y) = \{|y| - (y^2 - 1)^{0.5}\}^2,\ |y| > 1,$$

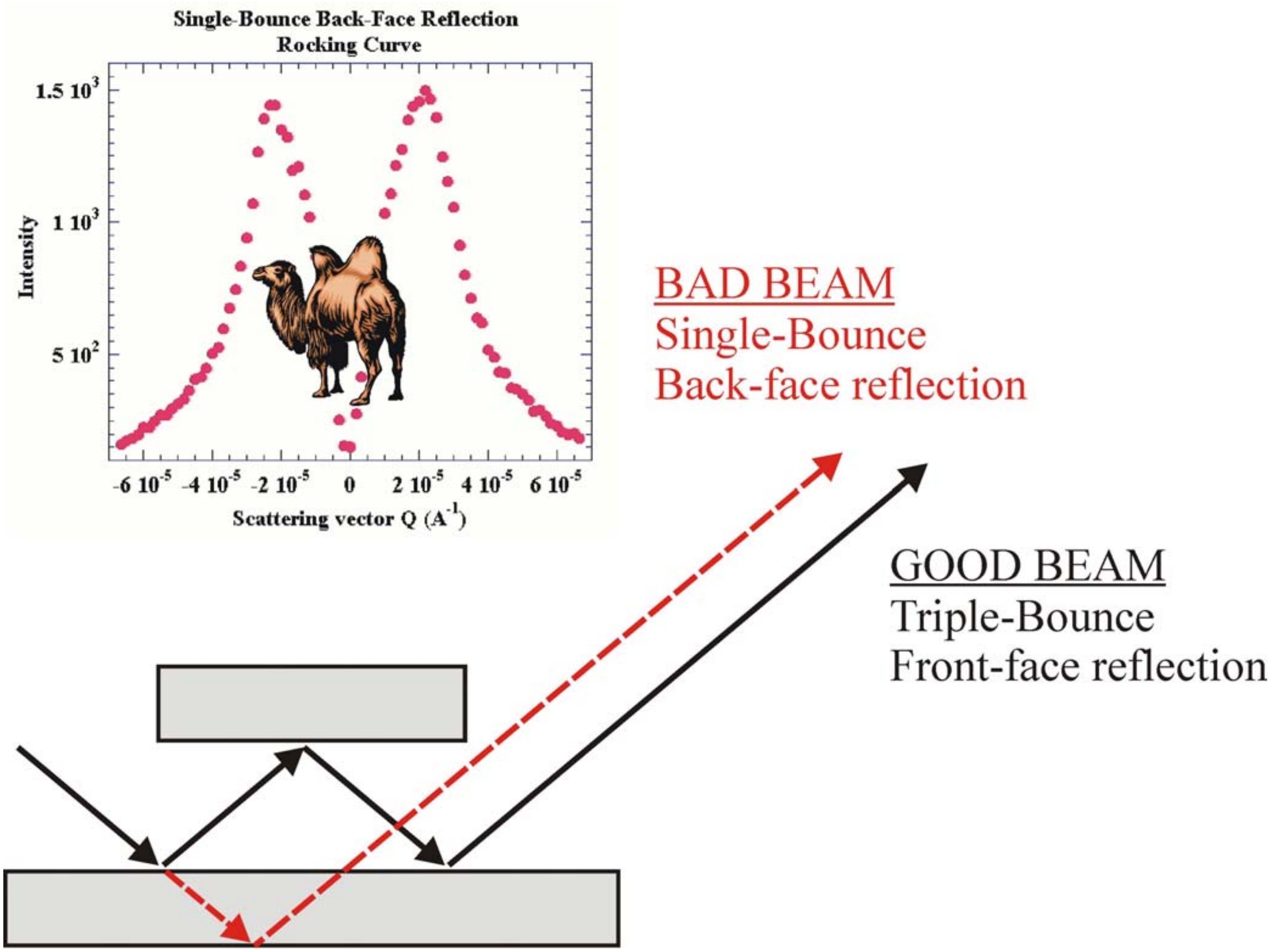
where $y = (\theta - \theta_B)/\delta\theta_D$, is deviation from the Bragg angle, θ_B, and $\delta\theta_D$ is the Darwin width. In case of X-rays the absorption effect has been taken into consideration.

X-RAY AND NEUTRON ROCKING CURVES

The best results obtained before 1995



CONTAMINATION OF THE FRONT-FACE TRIPLE-BOUNCE REFLECTION WITH THE BACK-FACE SINGLE-BOUNCE REFLECTION WAS THE MAIN PROBLEM FOR THE USANS INSTRUMENTS. THIS EFFECT DOES NOT OCCUR IN USAXS DCDs DUE TO ABSORPTION OF X-RAYS IN Si

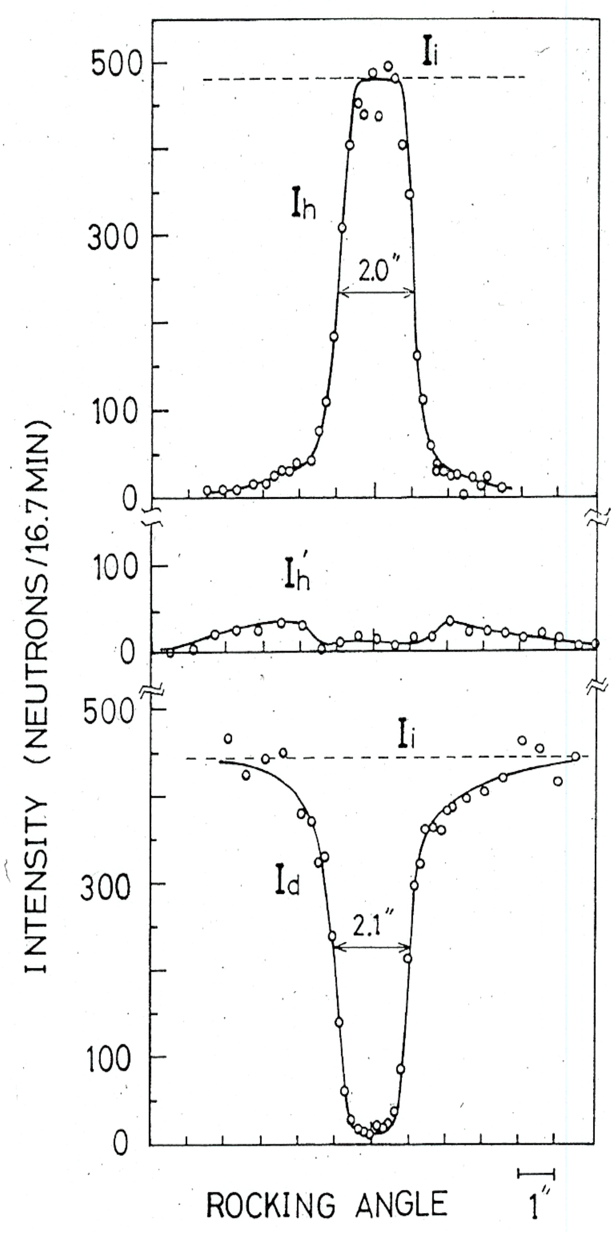


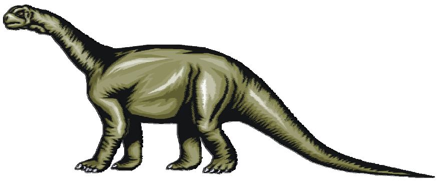
BAD BEAM
Single-Bounce
Back-face reflection

GOOD BEAM
Triple-Bounce
Front-face reflection

THE FIRST OBSERVATION OF BFRC (THE DINOSAUR -CAMEL)

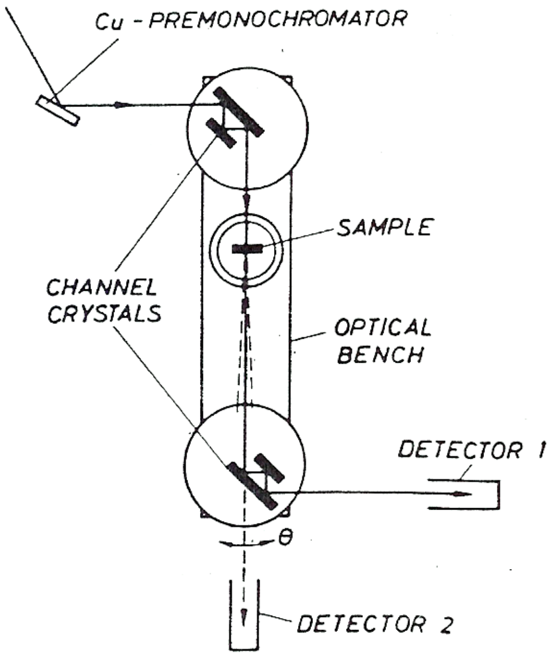
T. Takahashi *et al*, J. Phys. Soc. Japan, <u>45</u>, 1978

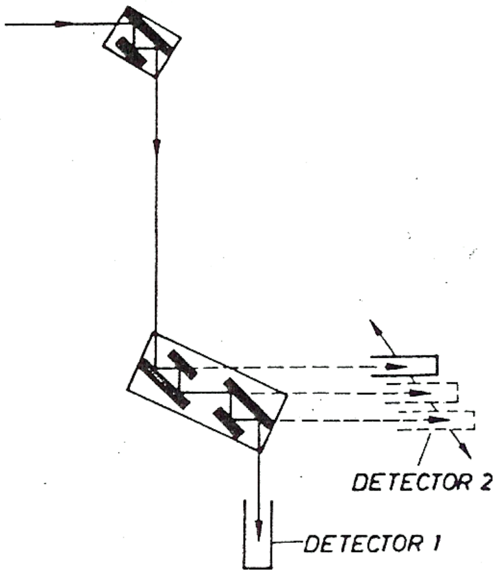




“TRANSMITTED” CAMEL?

D. Schwahn *et al*, NIM, A239, 1985





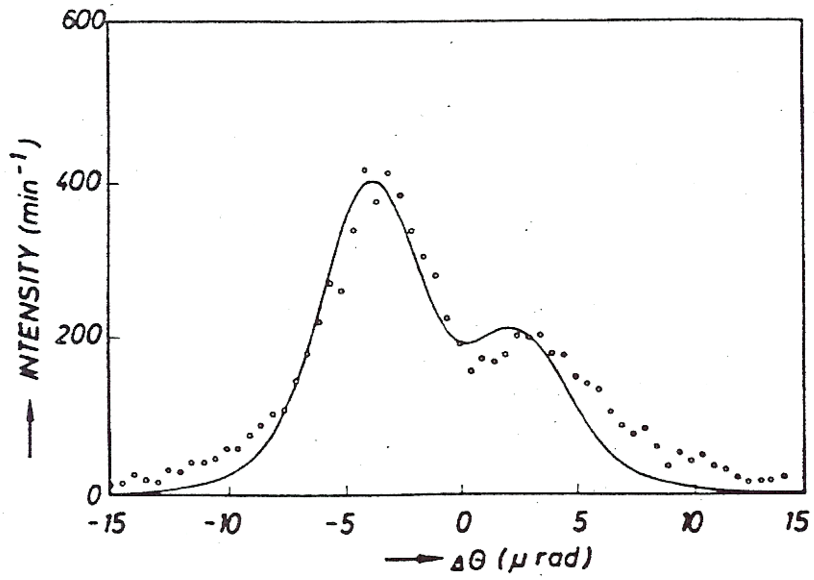
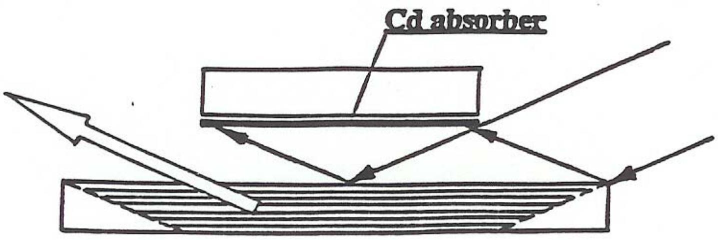
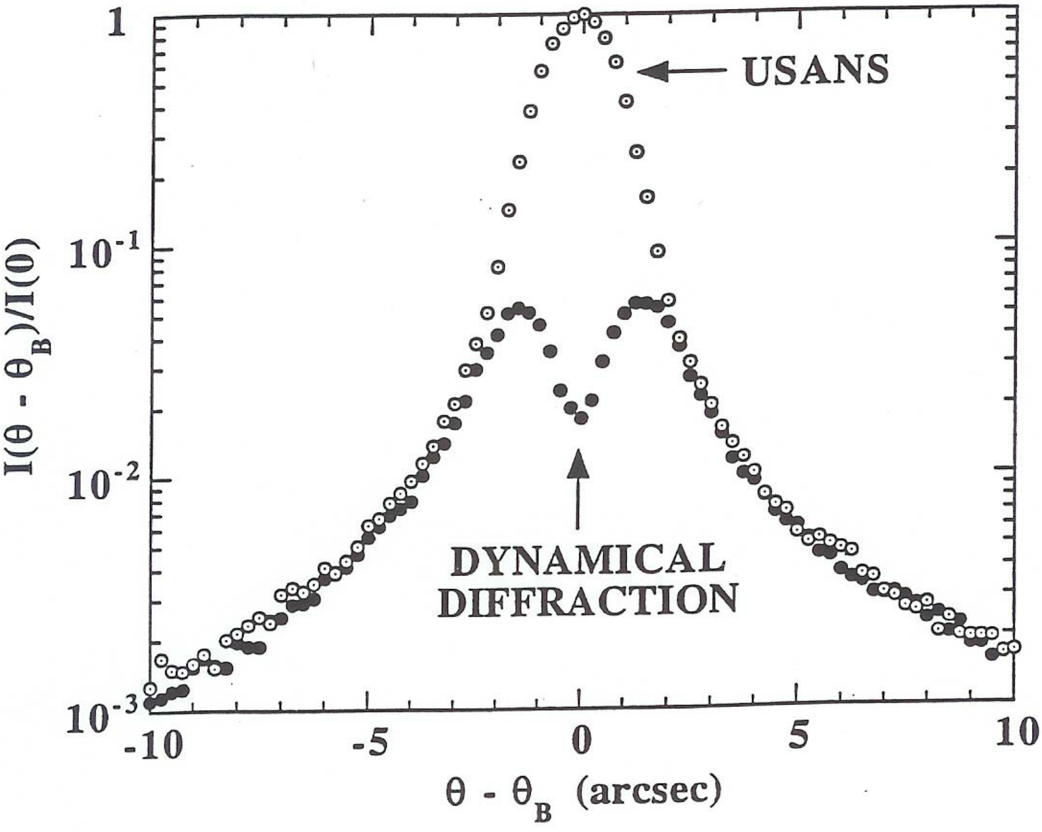


Fig. 5. Observed asymmetry of the transmitted intensity behind the fifth reflection (see fig. 2).

OBSERVATION OF A “CAMEL-SHAPED” ROCKING CURVE AT THE HFIR “MEMORIAL” USANS INSTRUMENT

M. Agamalian *et al*, J. Appl. Cryst., (1997), 30, 345.





THE DARWIN SOLUTION CONSIDERS A VERY THICK CRYSTAL WITH NEGLIGIBLY SMALL ABSORPTION

$(\mu_0 \approx 0; \quad \mu t_0 >> 0)$

THE EWALD SOLUTION CONSIDERS A THICK CRYSTAL WITH ZERO ABSORPTION $(\mu_0 = 0; \quad \mu t_0 = 0)$

The Darwin formula (Darwin, 1914):

$$R_D(y) = 1, \qquad |y| \leq 1$$

$$R_D(y) = [|y| - (y^2 - 1)^{0.5}]^2, \qquad |y| > 1$$

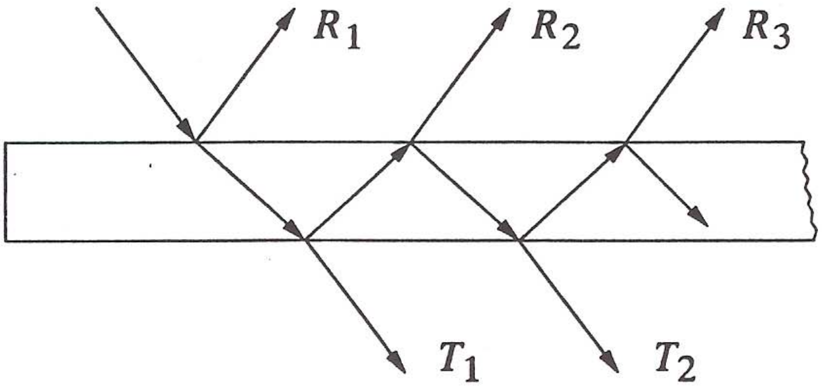
The Ewald formula (Ewald, 1917):

$$R_E(y) = 1, \qquad |y| \leq 1$$

$$R_E(y) = 1 - (1 - y^{-2})^{0.5}, \qquad |y| > 1$$

RELATIONSHIP BETWEEN THE DARWIN & EWALD FORMULAS

T. Takahashi *et al,* Phys. Lett., (1995) A200, 73.



This has been recently studied both theoretically and experimentally (Takahashi & Hashimoto, 1995, Phys.Lett., A200, 73.) and shown that for a transparent thick crystal (when the crystal thickness is much larger than the coherence length of neutrons) the Darwin formula gives the reflectivity only from the front-face, while the Ewald equation describes the total Bragg reflection from the front- and back-faces. Therefore, the simple relation between the **RE**(y) and **RD**(y) reflectivity functions can be written by the series:

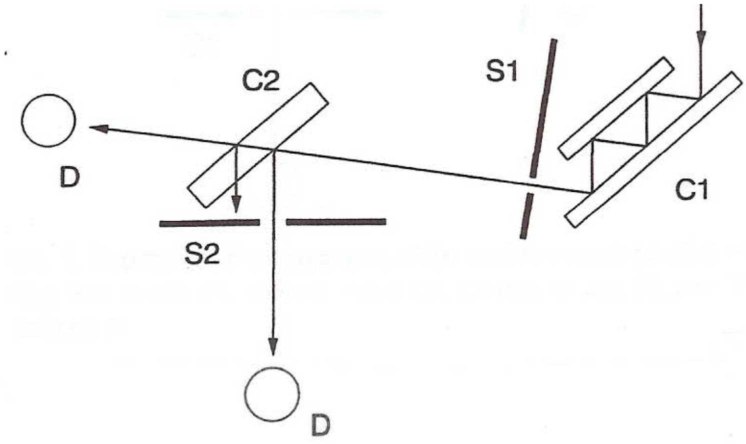
$$\mathrm{RE}(y) = \mathrm{RD}(y) + [1 - \mathrm{RD}(y)]^2 \mathrm{RD}(y)[1 + \mathrm{RD}(y)^2 + \mathrm{RD}(y)^4 + \ldots] =$$

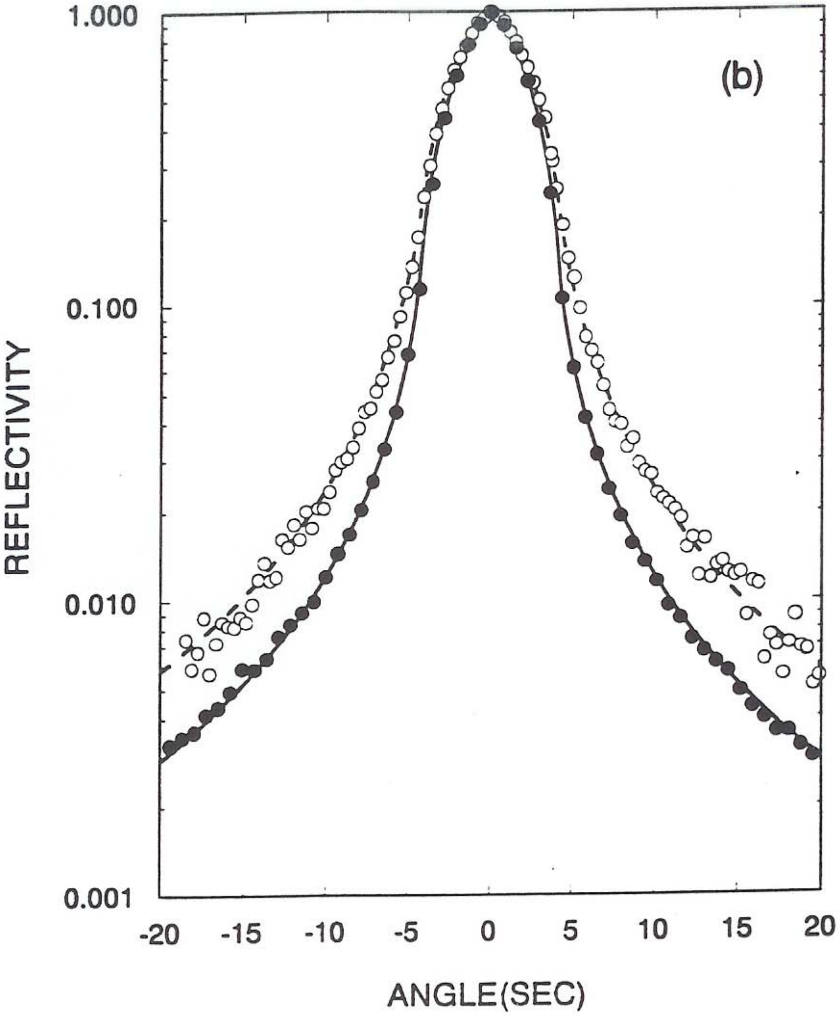
$$= 2\mathrm{RD}(y) / [1 + \mathrm{RD}(y)]$$

where the term $[1 - \mathrm{RD}(y)]^2 \mathrm{RD}(y)$ corresponds to the first back-face reflection.

EXPERIMENTAL PROF OF THE TAKAHASHI THEORY

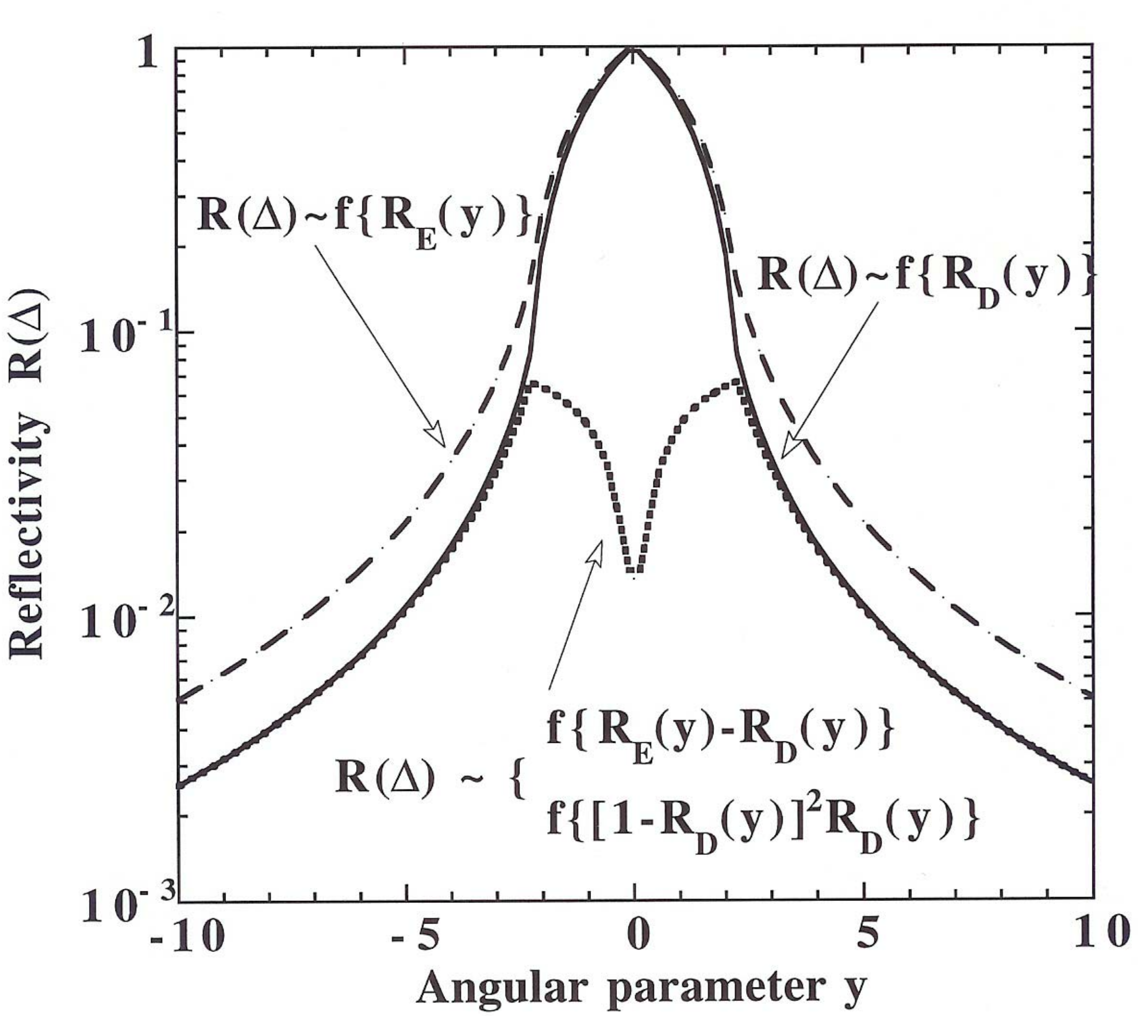
T. Takahashi *et al*, Phys. Lett., (1995) A200, 73.





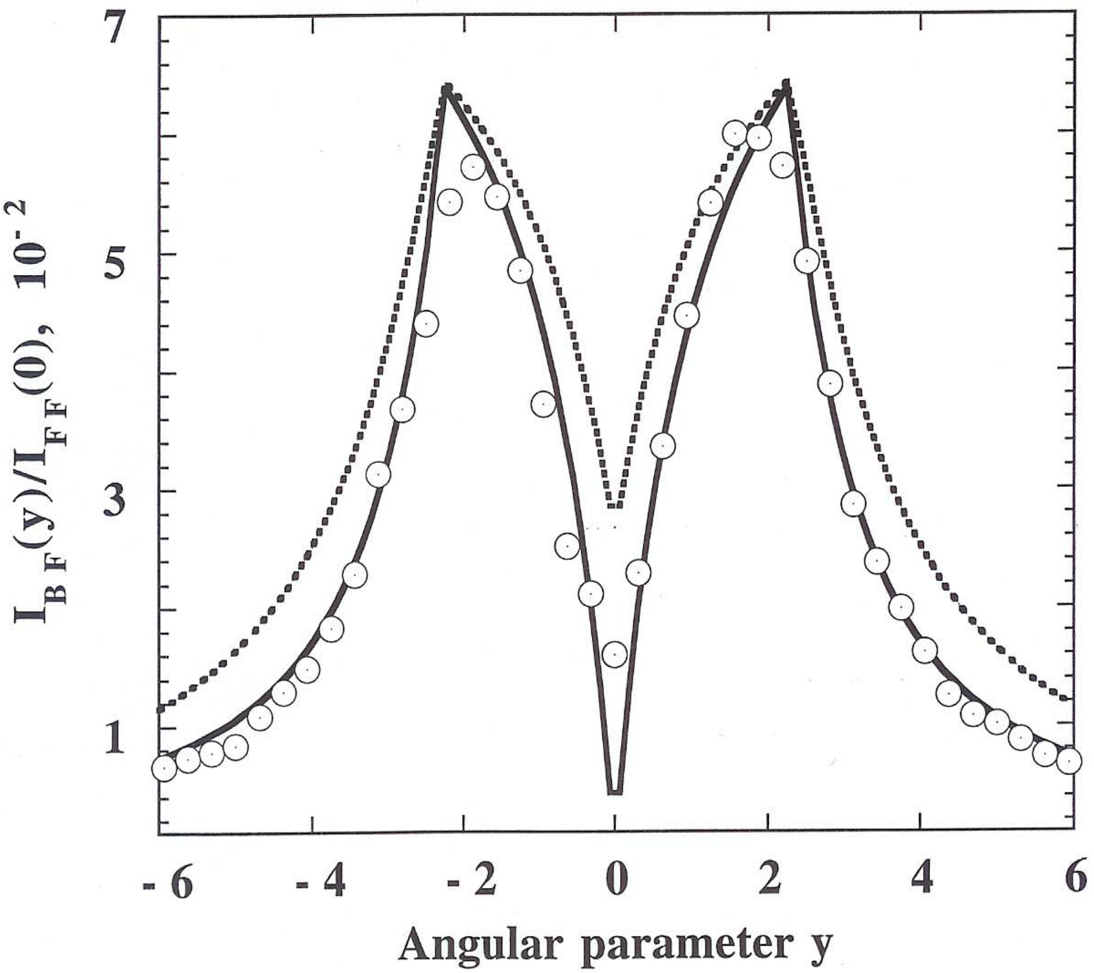
A "DARWIN-MINYS-EWALD" MODEL BASED ON THE TAKAHASHI THEORY, WHICH HAS BEEN ORIGINALLY USED TO FIT THE EXPERIMENTAL BFRC

M. Agamalian *et al*, J. Appl. Cryst., (1998), 31, 235



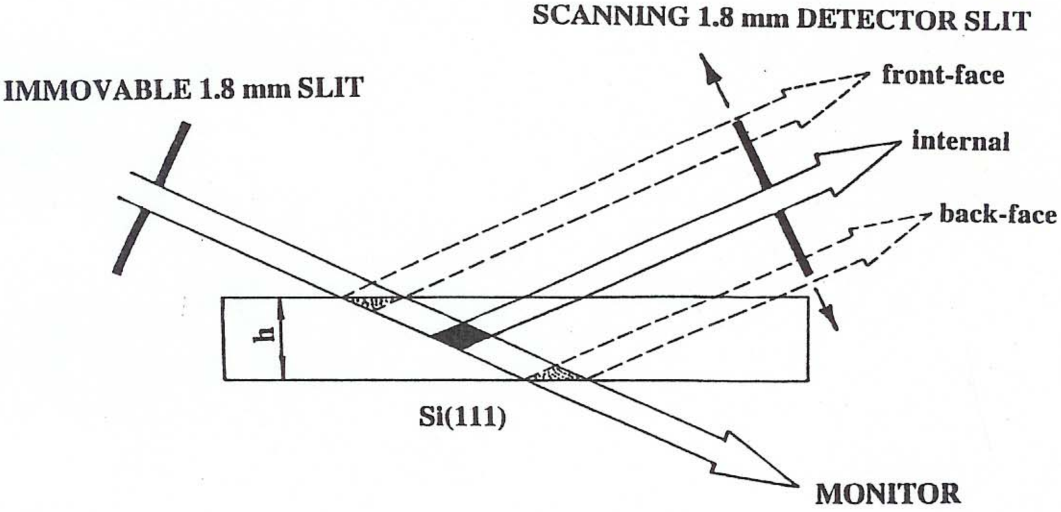
THE EXPERIMENTAL BFRC FITTED TO THE “DARWIN-MINYS-EWALD” MODEL

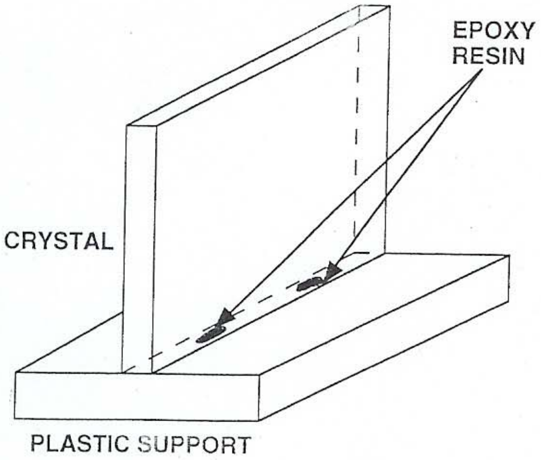
(solid and dotted lines are calculated for the triple- and single-bounce monochromators correspondingly)

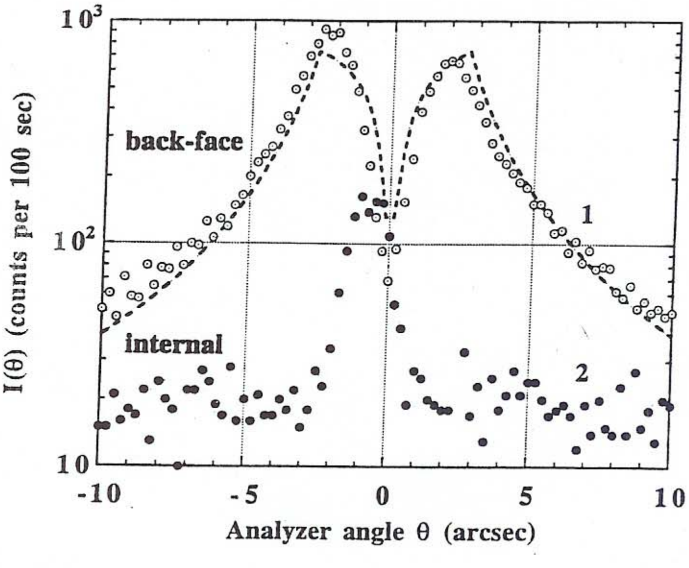


THE CLASSICAL EWALD AND DARWIN THEORIES DO NOT EXPLAIN THE EXPERIMENTALLY OBSERVED ASSYMETRY OF THE BFRC AND THE INTERNAL DIFFRACTION PEAK

M. Agamalian *et al*, J. Appl. Cryst., (1998), 31, 235

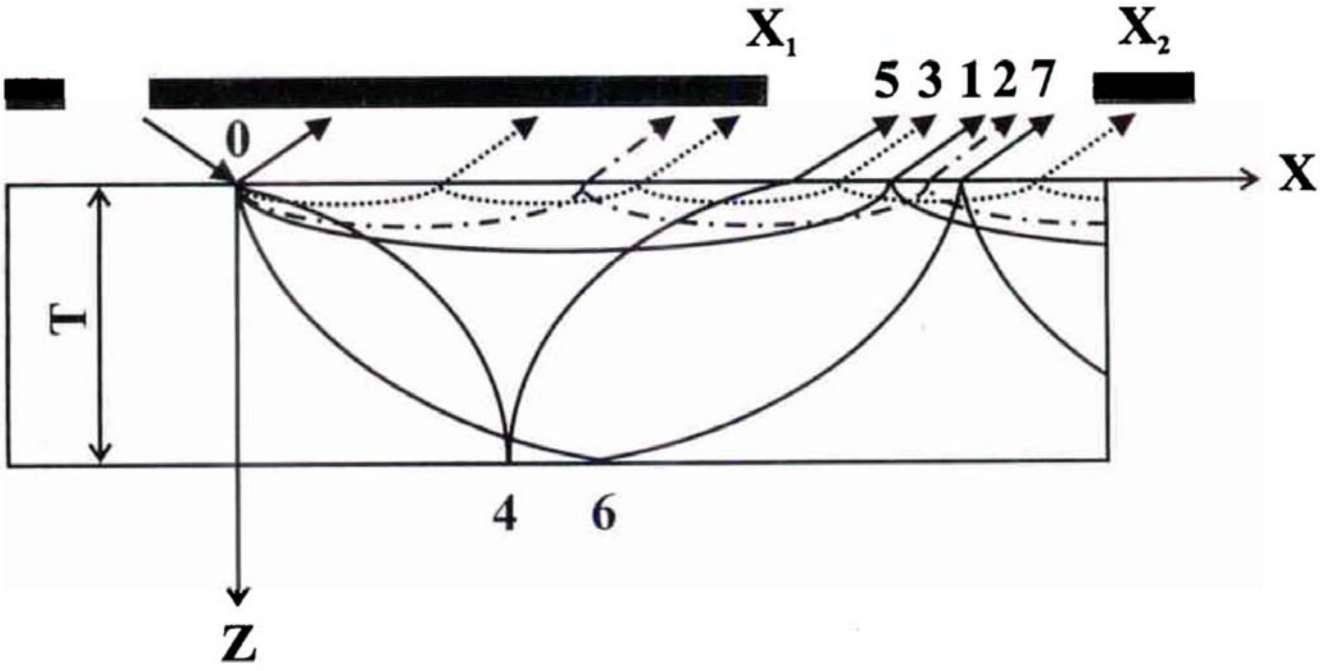






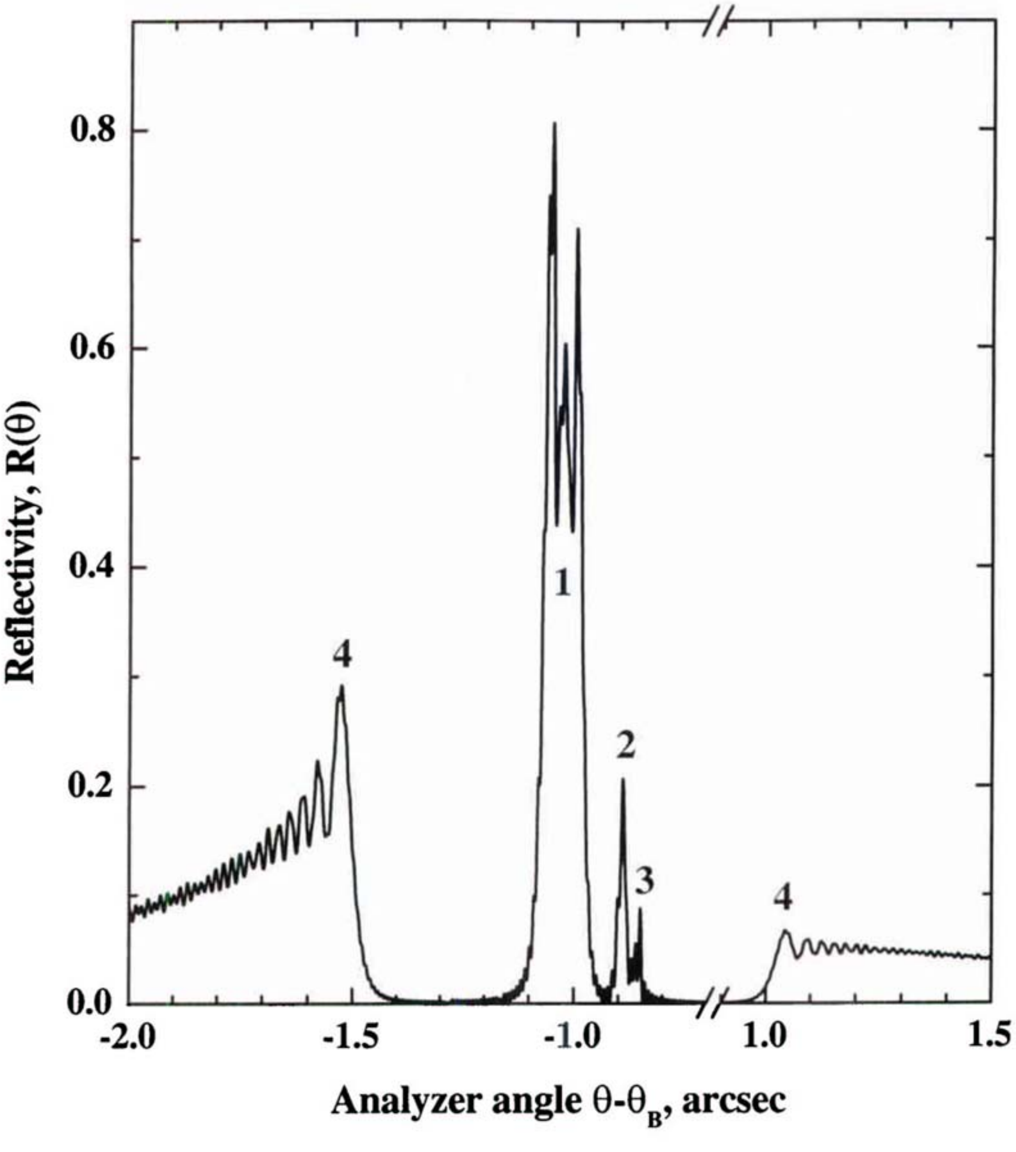
BACK-FACE (0-4-5) & GARLAND (0-1, 0-2, 0-3) TRAJECTORIES INSIDE A LIGHTLY DEFORMED CRYSTAL SHOULD BE CONSIDERED TO EXPLAIN THE OBSERVED DYNAMICAL DIFFRACTION EFFECTS

M. Agamalian *et al*, Phys. Rev. B, (2001), 64, 161402(R)



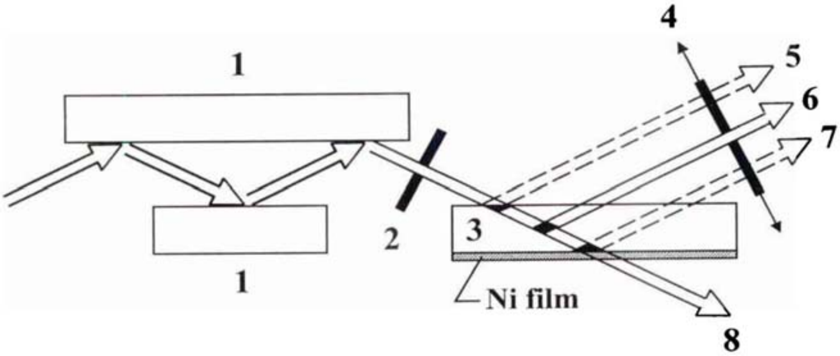
THE CALCULATED BACK-FACE (4-4) & GARLAND (1, 2, 3) REFLECTIONS FROM A LIGHTLY DEFORMED CRYSTAL

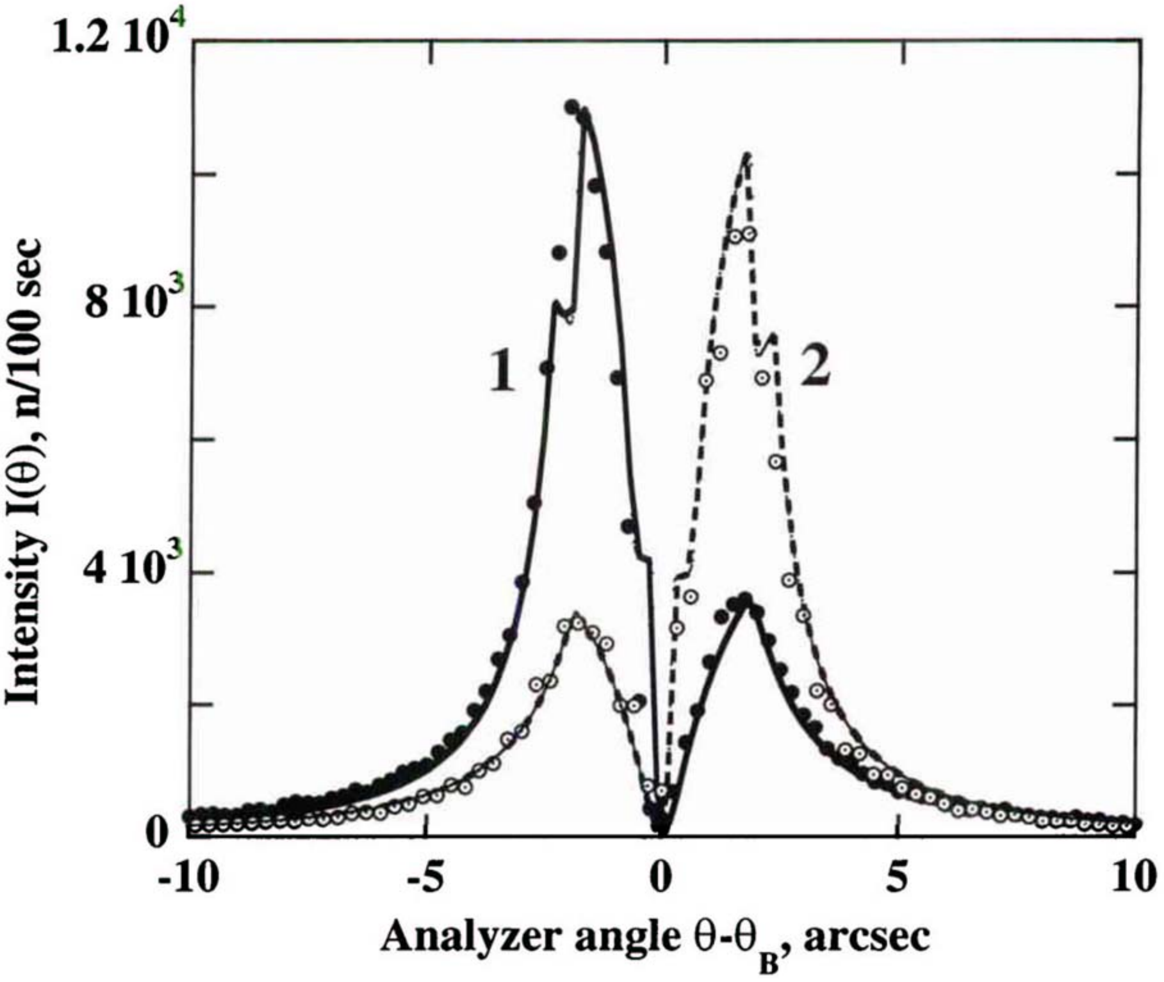
M. Agamalian *et al*, Phys. Rev. B, (2001), 64, 161402(R)



THE EXPERIMENTAL (7) & THEORETICAL BFRC OBTAINED FOR THE Si CRYSTAL, WHICH IS LIGHTLY DEFORMED WITH THE DEPOSITED THIN Ni FILM

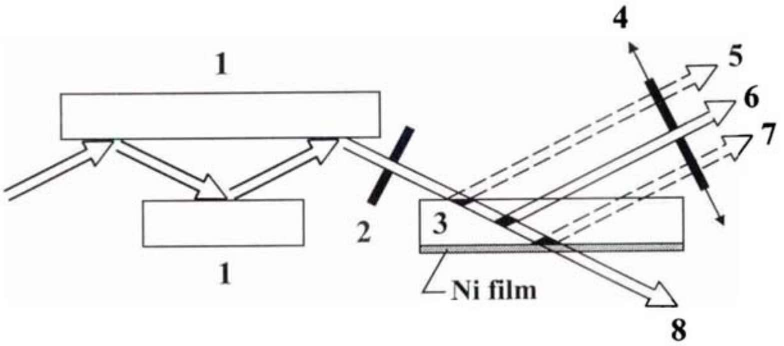
M. Agamalian *et al*, Phys. Rev. B, (2001), 64, 161402(R)

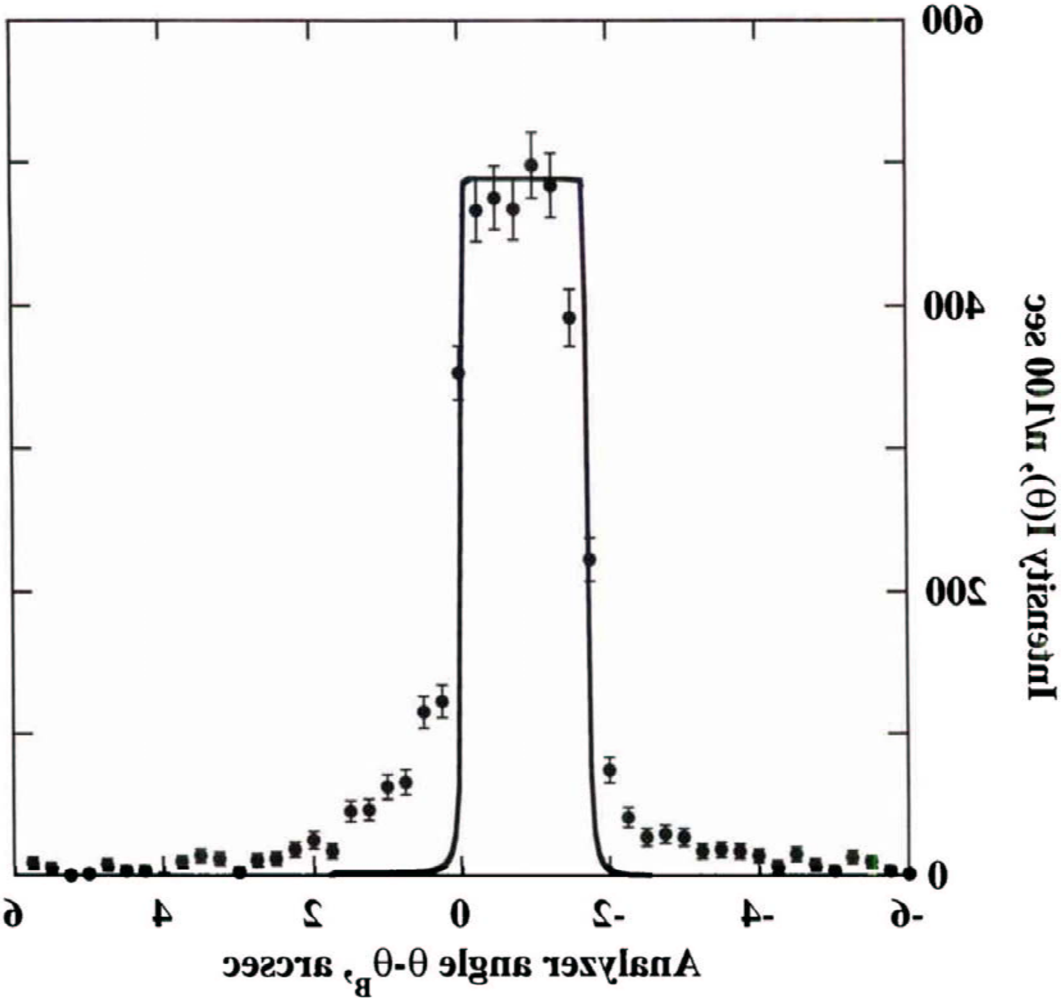




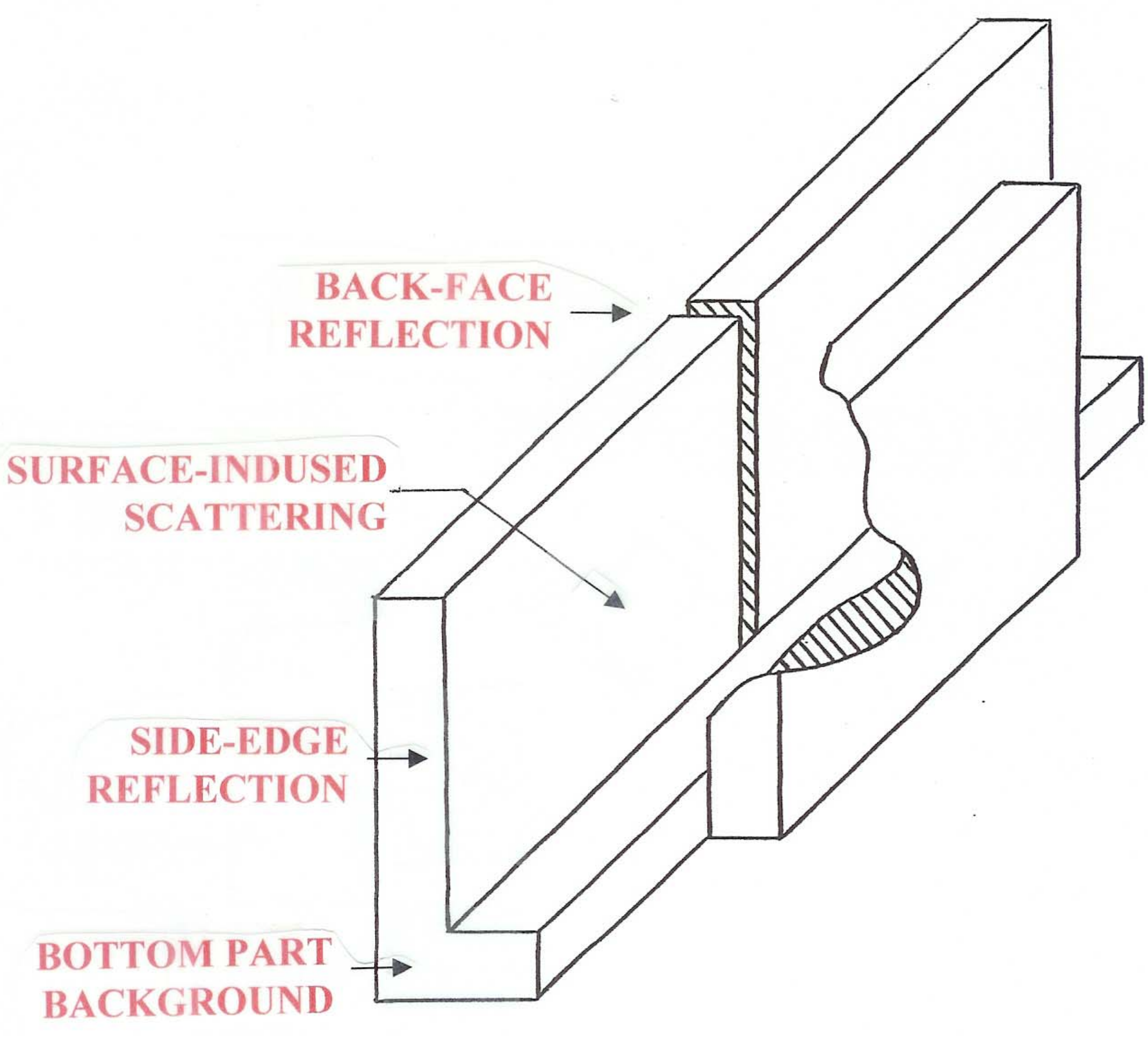
THE EXPERIMENTAL (6) & THEORETICAL GARLAND REFLECTIONS OBTAINED FOR A LIGHTLY DEFORMED Si CRYSTAL

M. Agamalian *et al*, Phys. Rev. B, (2001), 64, 161402(R)



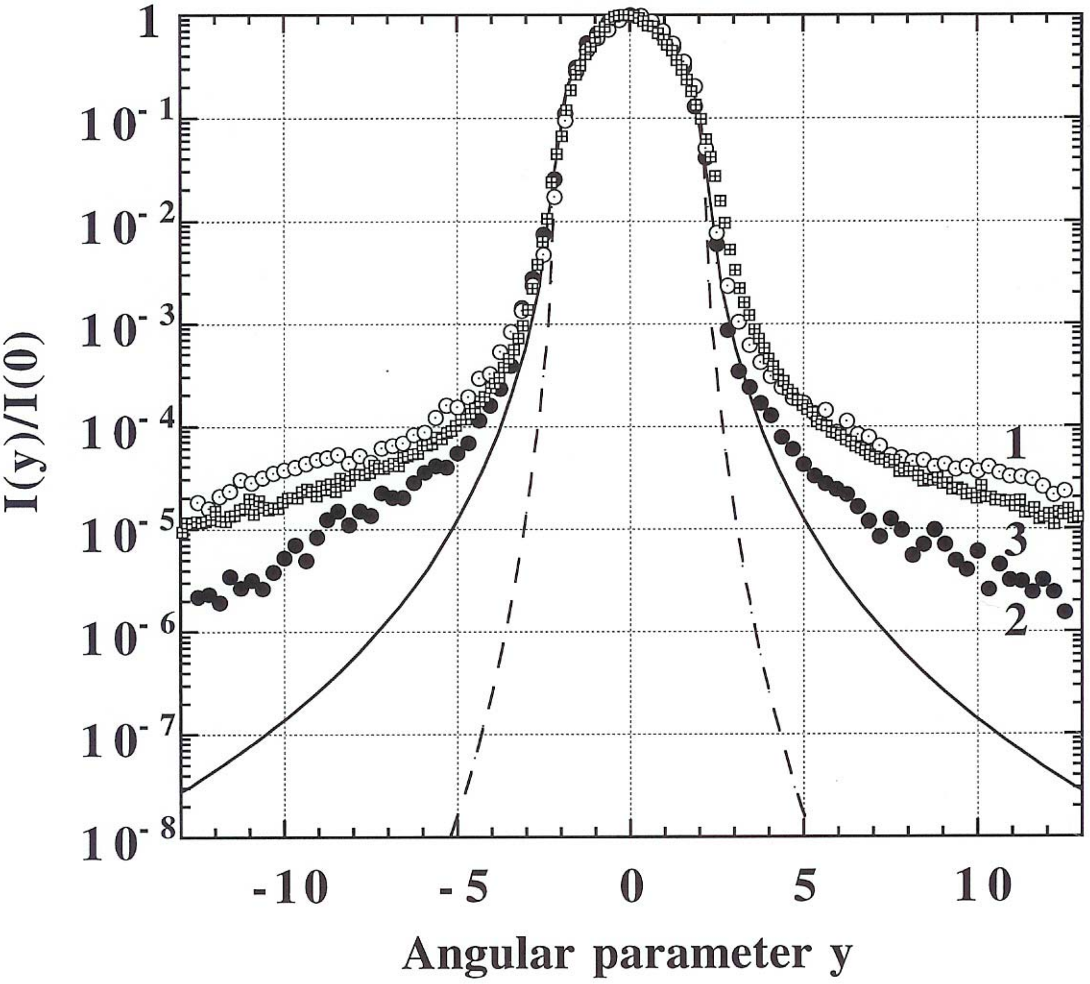


PARASITIS SCATTERING FROM A TRIPLE-BOUNCE Si CRYSTAL

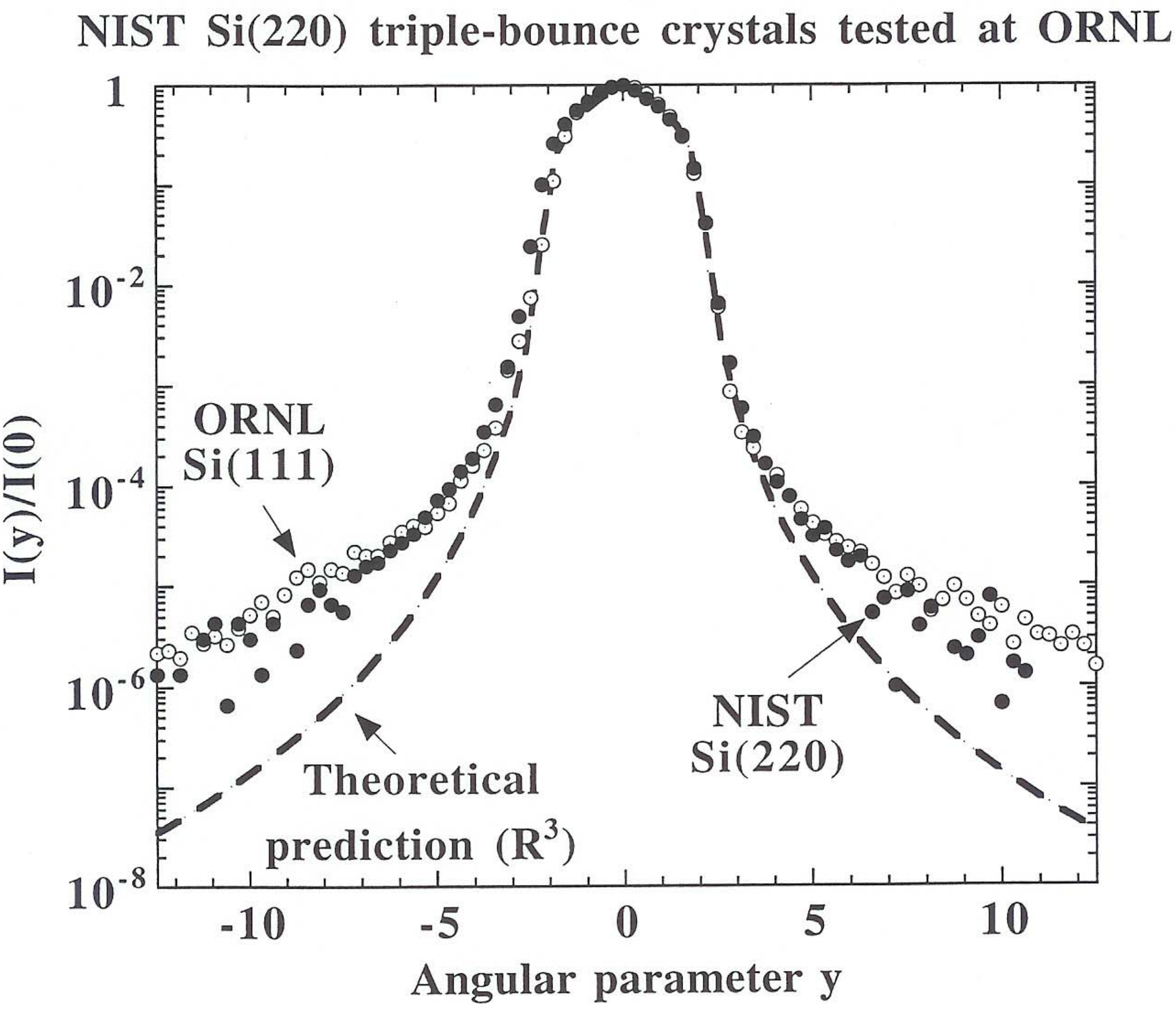


X-RAY (2) AND NEUTRON (1, 3) ROCKING CURVES AFTER BLOCKING THE SINGLE-BOUNCE BACK-FACE REFLECTION (1) AND REDUCING THE SURFACE-INDUCED SCATTERING (2)

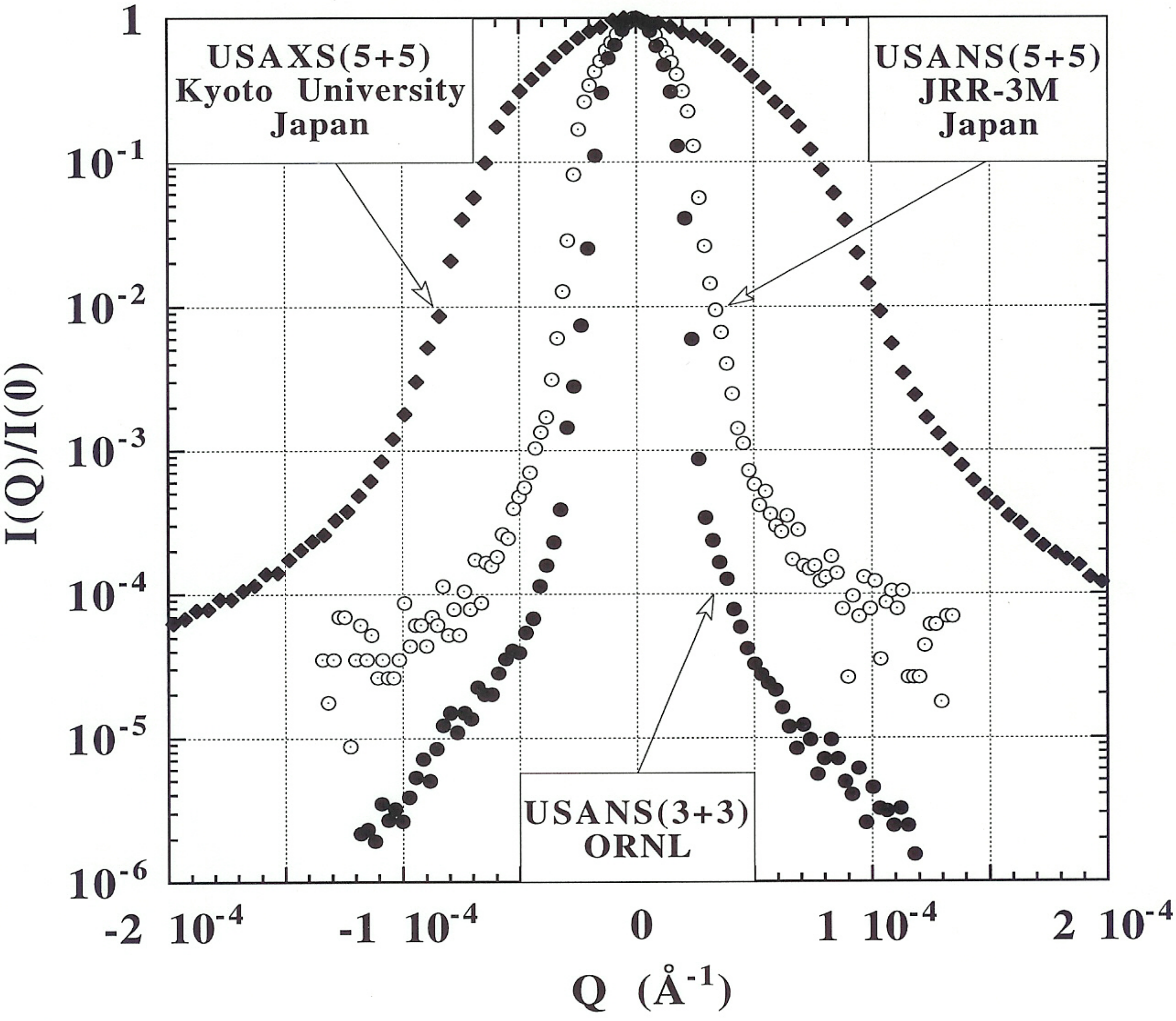
M. Agamalian *et al*, J. Appl. Cryst., (1998), 31, 235



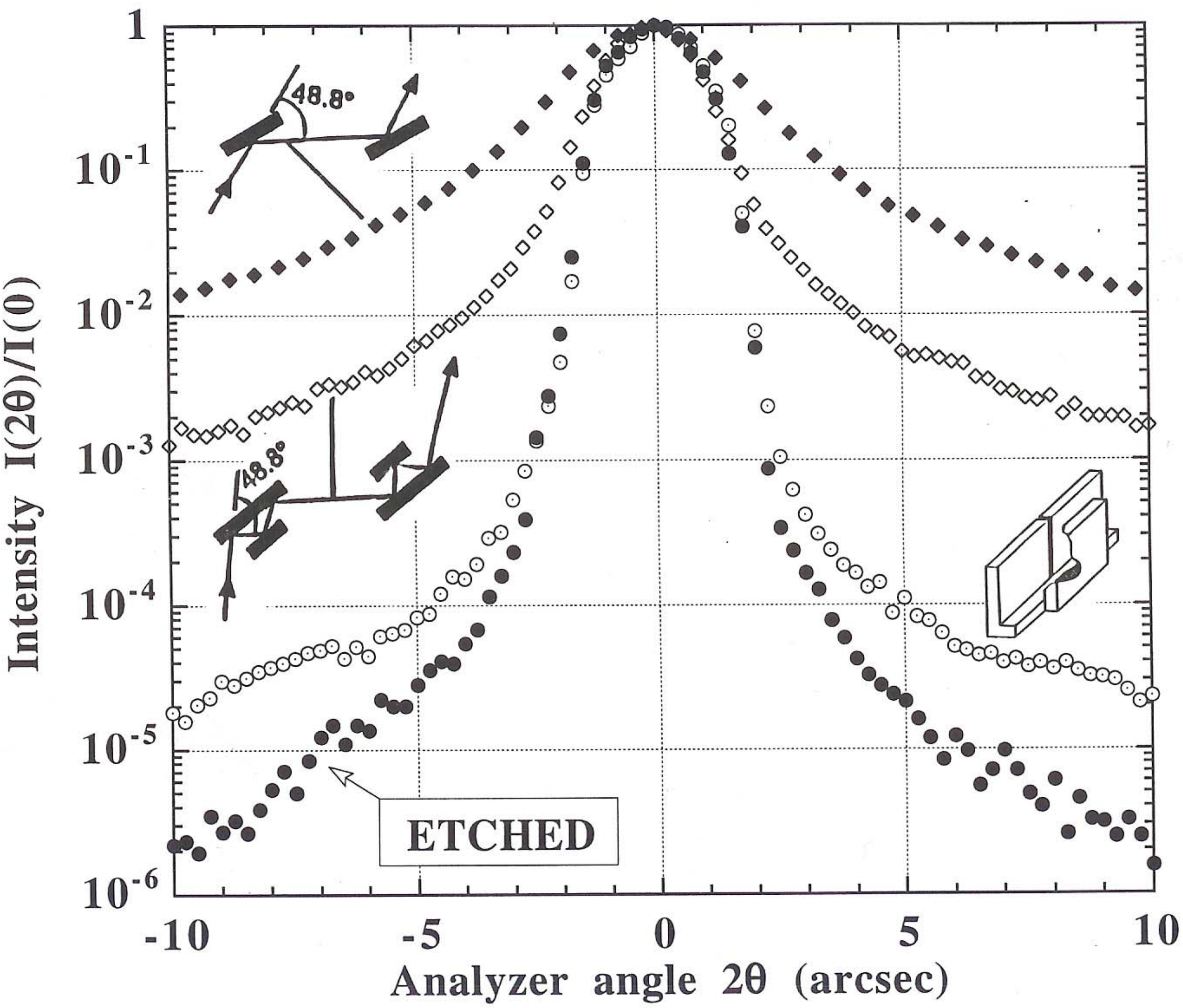
“10^{-6} QUALITY” ROCKING CURVES WERE OBTAINED ON Si(111) and Si(220) CRYSTALS



COMPARISON OF THE USAXS AND USANS ROCKING CURVES IN Q-SPACE

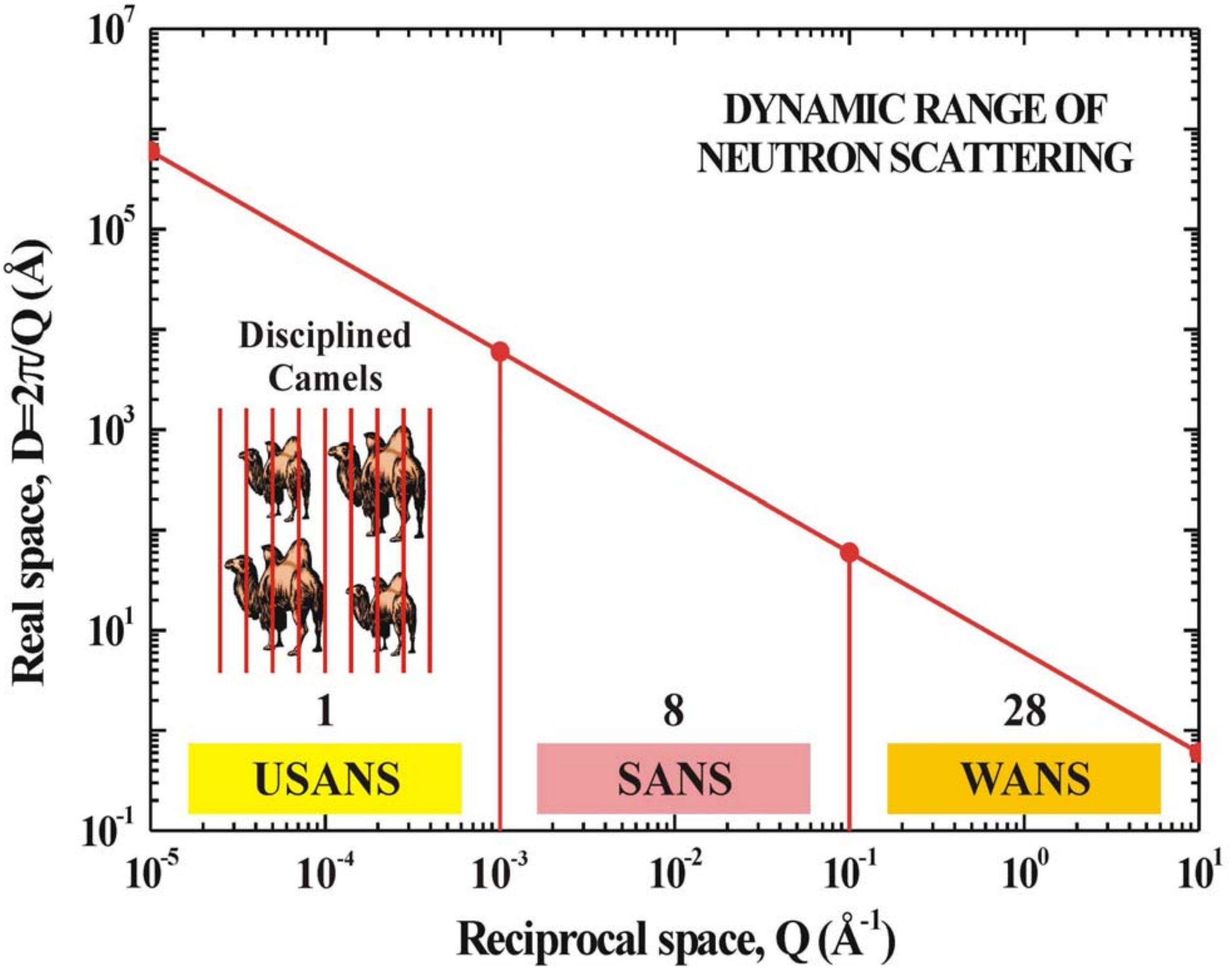


EVOLUTION OF THE USANS ROCKING CURVES



IN 1996 ORNL SECURITY BUSTED CAMELS AND PUT THEM BEHIND CADMIUM BARS…

AND THE μm MILLENIUM BEGAN



THE SIGNAL-TO-NOISE RATIO OF USANS INSTRUMENTS HAS BEEN INCREASED BY THREE ORDERS OF MAGNITUDE THAT LEADS TO NEW SCIENCE

MICROMETRIC SCALE NEUTRON DIFFRACTION STRUCTURAL ANALYSIS

> *Whenever you invent a method ten or a hundred times better than the existing ones, you can be sure that this will lead to new science.*
>
> H. Meier-Leibnitz

INSTRUMENT PERFORMANCE

THE USANS INSTRUMENT AT NIST IS TAKEN AS AN EXAPLE

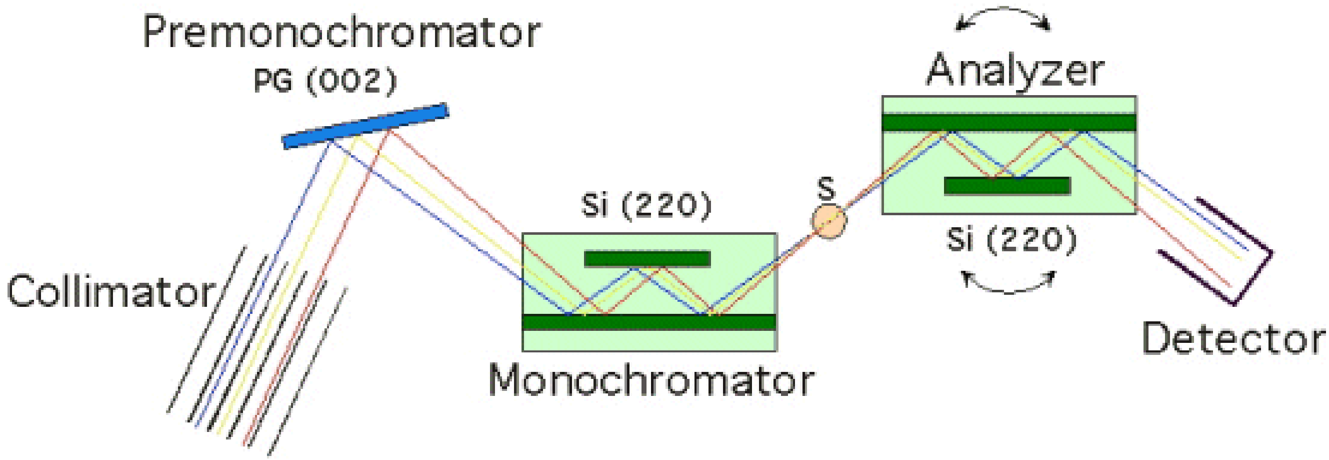
Dynamical Q-range: $2*10^{-5}$ Å^{-1} to ~ 0.01 Å^{-1}

Dynamical I-range: six orders of magnitude

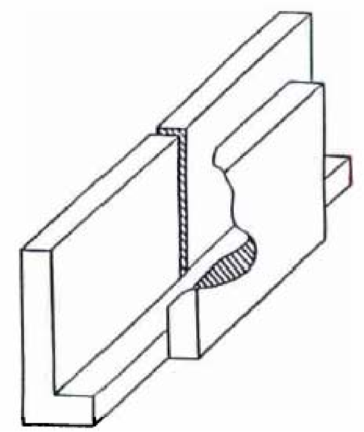
Q-resolution: $Q_{min} \approx 4\pi\delta\theta_D/\lambda \sim 2*10^{-5}$ Å^{-1}

Flux at Sample: <3,000 n/cm^{2}sec

Max Sample Size: 4 cm wide x 5 cm high

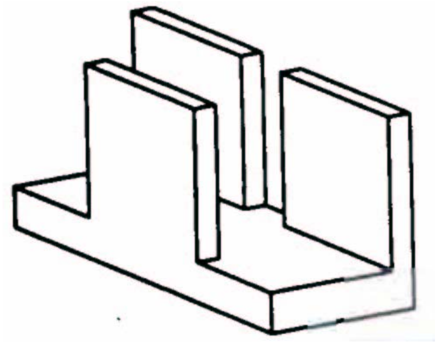


DIFFERENT DESIGN OF CHANNEL-CUT CRYSTALS



ORIGINAL “MEMORIAL” DESIGN
OF THE TRIPLE-BOUNCE
CHANNEL-CUT CRYSTAL WITH
AN ADDITIONAL GROOVE FOR
CADMIUM ABSORBER (ORNL)

CADMIUM-FREE DESIGN OF THE
TRIPLE-BOUNCE CHANNEL-CUT
CRYSTAL WITHOUT
THE “CAMEL CAVE” (ILL)



STRESS-FREE DESIGN OF THE
TRIPLE-BOUNCE CHANNEL-CUT
CRYSTAL (NIST)

EVOLUTION OF THE INTENSITY AT SAMPLE POSITION

ORNL “MEMORIAL” USANS: ~ 800 n/cm^{2}sec

NIST USANS: ~ 3000 n/cm^{2}sec (focus)

ILL USANS: ~ 5000 n/cm^{2}sec

PROPOSED ORNL USANS
(Monte-Carlo simulation): ~ 40000 n/cm^{2}sec (focus)

THE NEXT GENERATION OF USANS:
A "TIME-OF-FLYING" CAMEL