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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

ELASTIC NEUTRON DIFFRACTION ANALYSIS OF A POWDER SAMPLE OF UC

by

M. BONOMO (CNEN) and R. COLELLA (Euratom)

1967

Joint Nuclear Research Center
Ispra Establishment - Italy

Chemistry Department
Solid State Physics

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SUMMARY

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KEYWORDS

URANIUM CARBIDES
POWDERS
NEUTRON BEAMS
DIFFRACTION
NEUTRON SPECTROMETERS

ELASTIC NEUTRON DIFFRACTION ANALYSIS OF A POWDER SAMPLE OF UC(+)

Uranium monocarbide is a new compound of nuclear interest; its structure is cubic of the NaCl type, with partly covalent and partly metallic bonds (FROST, 1963).

The Debye-Scherrer diffraction pattern of a powder specimen of UC has been obtained at room temperature by means of a neutron triple axis spectrometer with the aim of gaining information about the thermal vibration amplitudes of the atoms in the UC lattice. Neutrons of wavelength 1.001 Å, corresponding to 82 meV, were monochromatized by a Pb single crystal set for the (220) reflection. The UC powder had been sieved to a 0.3 mm mesh in a purified argon atmosphere, to avoid oxydation, and the sample powder was contained in a 160x60x10 mm^3 aluminium box hermetically sealed under argon atmosphere, the thickness of the two side walls being 0.2 mm. The parallel-sided slab of UC powder was placed on the second axis of the spectrometer, in the symmetrical transmission position, so to intercept the whole of the neutron beam, and the elastic component of the diffracted beam was selected by an Al monochromator set for the (111) reflection. It has been recently shown (BUTT and O'CONNOR, 1967), by means of the Mössbauer effect, that some discrepancies exist for the Debye temperatures of Al and KCl when only the elastically scattered 14.4 Kev gamma radiation is considered, with respect to the values obtained by measuring the total diffracted radiation. The use of a third axis, where an analyzer single crystal is placed, allows only neutrons having a definite amount of energy to reach the counter, so it is possible, within some resolution limits, to select only neutrons being elastically scattered by the crystal, thus avoiding that the Bragg peak include the thermal inelastic contribution. The absorption correction was taken into account by means of a factor $e^{-\mu t/\cos\vartheta}$ where: μ = linear absorption coefficient; t = thickness of specimen; ϑ = scattering angle and

(+)Manuscript received on September 5, 1967.

the value of the product μt was measured with the counter in the zero position by the direct measurement of the reduction of the intensity of the incident beam when the powder slab was inserted perpendicularly; the ratio between transmitted and incident intensity was found equal to 0.528.

The experimental results are plotted in figs. 1 and 2. Fig. 1 refers to a conventional diffraction experiment; in this case the analyzer crystal was eliminated, and the counter set at zero position with respect to the third axis. The counter arm proceeded by steps of 3 minutes every 7.10^5 monitor counts, the counting time being about 160 sec. and the number of pulses giving the area of each peak being never less than 7.10^4. The 2ϑ interval 0°–115° was continuously scanned and since the average half-width of the diffraction peaks was of the order of 1°30', an overlapping of the peaks was avoided even at large angles. The contribution of the aluminum container was evaluated by repeating over some angular regions the diffraction pattern without powder; a correction was introduced where the diffraction peaks of Al were appreciable, the absorption of the UC powder being taken into account. Fig. 2 reports data obtained by means of elastic neutron diffraction; in this case the 3-minute steps corresponded to monitor counts of 5.10^6 pulses, which required a time of about 19 minutes for each step; the integrated intensities of the various peaks were never less than 15.000 pulses. The aluminum contribution was evaluated for a limited number of peaks, and found negligible in all cases. The resolution was better than in the case of conventional diffraction, as shown elsewhere (CAGLIOTI and TOCCHETTI, 1965a).

The background to be subtracted, in each case, was calculated by considering the average over a number of points on the wings of the diffraction peaks, far away from the center, where the intensity was uniform. The vertical lines through experimental points in figs. 1 and 2 correspond to errors of the order of $\pm 2\%$.

As a first approximation, the mean square displacements of uranium and carbon atoms have been supposed to be equal, according to experimental results on cubic crystals quoted by Lonsdale (1948)(°). This result has a theoretical foundation, as pointed out by Blackman (1956) who showed that, at temperatures higher than Θ, the mean square displacements of different atoms in a crystal do not depend on the individual masses, but only on the interatomic forces.

Θ is the characteristic temperature which, in the case of UC, has been measured by means of X-ray diffraction and found to be of the order of 265°K (COLELLA, DRAGONE and MERLINI, 1967).
In the frame of the assumption of a unique temperature factor, the plot of the logarithm of the integrated intensities vs. $(\sin\vartheta/\lambda)^2$, after proper normalization against angular, absorption and multiplicity factors, is a straight line, its negative slope being proportional to the common value of the mean square displacement of atoms(°°). The straight lines of figs. 1 and 2 have been fitted by means of the least squares method, and the values of the mean square displacements found to be: 0.0245 $Å^2$ in the case of conventional diffraction and 0.0887 $Å^2$ in the case of elastic diffraction. It is difficult to explain the scattering of the experimental points from the straight lines; every intensity has been measured with a statistical accuracy better than 1%, and no other source of erratic errors was believed to be present.
A possibility could be the occurrence of simultaneous reflections, whioh have a strong influence on neutron diffraction intensities (BORGONOVI and CAGLIOTI, 1962; MOON and SHULL, 1964). Although the departure of the experimental points from straight lines in

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(°) See also BARNEA and POST (1966).

(°°) The plot of log I vs $(\sin\vartheta/\lambda)^2$ would be approximately linear even if the two temperature factors were different by a factor of two.

figs. 1 and 2 does not seem to exhibit a definite character, the probable errors (WORTHING and GEFFNER, 1943) which affect the values of the measured mean square displacements are similar; 0.00084 $Å^2$ for conventional diffraction and 0.0099 $Å^2$ for elastic diffraction.

The value of the mean square displacement determined by elastic diffraction, 0.0887 $Å^2$, bears a much better agreement with the value determined by X-ray diffraction on single crystals: 0.0798 $Å^2$, where the inelastic effects, though present, play a minor role (BUTT and O'CONNOR, 1967). The discrepancy between the results from conventional and elastic diffraction could be attributed to the presence of inelastic scattering, which, in conventional diffraction, is a considerable fraction of the measured intensity at high values of $\sin\vartheta/\lambda$, thus decreasing the slope of the plot log I vs $(\sin\vartheta/\lambda)^2$. The energy resolution Δ E of the Al (111) analyzer has been calculated (CAGLIOTI and TOCCHETTI, 1965b) for an intermediate value of 2ϑ and found equal to 0.497 meV, to be compared with the maximum phonon energy which can be exchanged with neutrons: 22.8 meV, calculated on the basis of the relation $\Theta = h\nu_{max}/K$ valid for a Debye solid.

It is concluded that, in spite of the scattering of the experimental points from the straight lines given by the Weinstock-Debye theory, the difference between the slopes obtained in conventional and elastic diffraction is undoubtly outside of the experimental errors, and that a large inelastic contribution was probably present in the conventional diffraction experiment.

Bibliography

BARNEA, Z. and POST, B.: Acta Cryst. 21, 181 (1966).

BLACKMAN, M.: Acta Cryst. 9, 734 (1956).

BORGONOVI, G. and CAGLIOTI, G.: Nuovo Cimento 24, 1174 (1962).

BUTT, N.M. and O'CONNOR, D.A.: Proc. Phys. Soc. 90, 247 (1967).

CAGLIOTI, G. and TOCCHETTI, D.: Nucl. Instr. & Methods 32, 181 (1965a).

CAGLIOTI, G. and TOCCHETTI, D.: Proc. I.A.E.A. - Inelastic Scattering of Neutrons - Vienna, vol. II, p. 505 (1965b).

COLELLA, R., DRAGONE, D. and MERLINI, A.: to be published (1967).

FROST, B.R.C.: Jour. Nucl. Mat. 10, 263 (1963).

LONSDALE, K.: Acta Cryst. 1, 142 (1948).

MOON, R.M. and SHULL, C.G.: Acta Cryst. 17, 805 (1964).

WORTHING, A.G. and GEFFNER, J.: "Treatment of Experimental Data" - New York, John-Wiley & Sons, Inc. (1943), p. 249.

Figure Captions

Fig. 1 - Logarithm of integrated intensities versus $(\sin\vartheta/\lambda)^2$. Conventional diffraction.

Fig. 2 - Logarithm of integrated intensities versus $(\sin\vartheta/\lambda)^2$. Elastic diffraction.

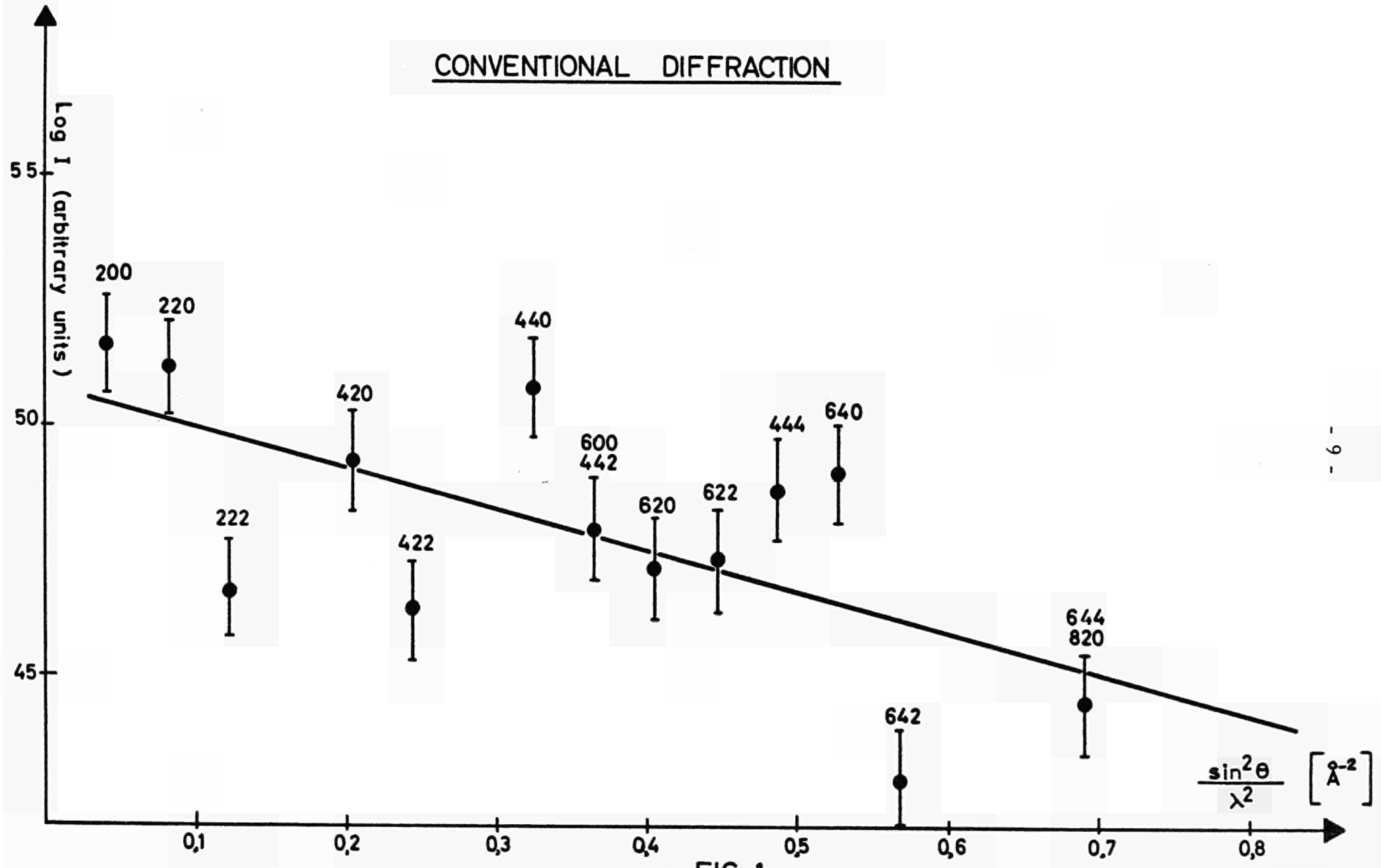
CONVENTIONAL DIFFRACTION
Log I (arbitrary units)
55
50
45
200
220
420
440
600
442
222
422
620
622
444
640
644
820
642
0,1
0,2
0,3
0,4
0,5
0,6
0,7
0,8
sin²θ / λ²
[Å⁻²]

FIG. 1

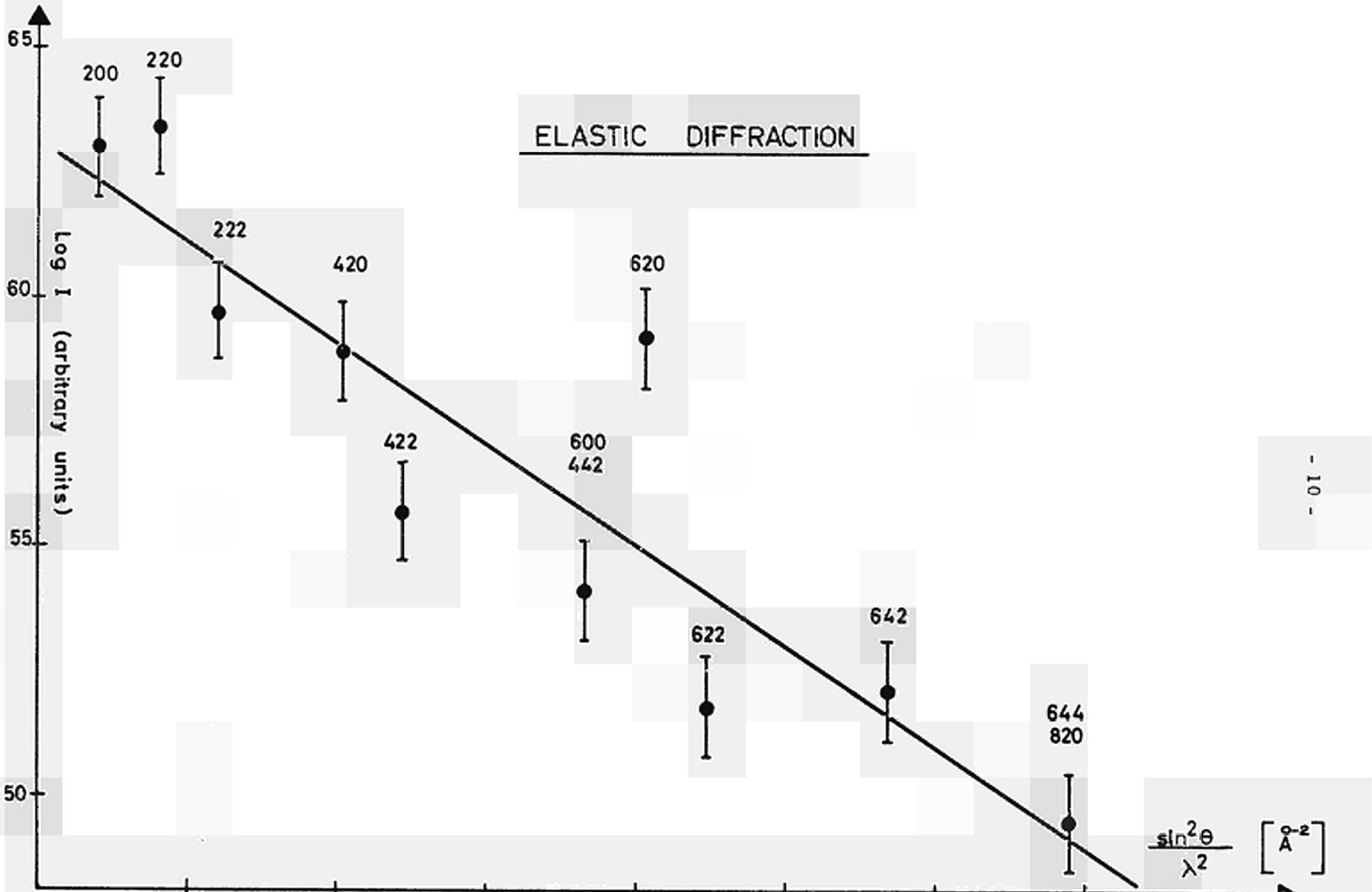
ELASTIC DIFFRACTION
Log I (arbitrary units)
65
60
55
50
200
220
222
420
422
600
442
620
622
642
644
820
0,1
0,2
0,3
0,4
0,5
0,6
0,7
sin²θ / λ²
[Å⁻²]

FIG. 2

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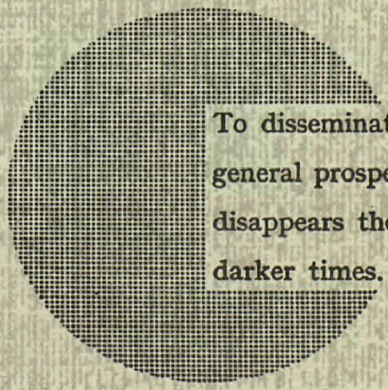

To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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