A γ Compton Experiment with an Annular 241Am Source: Momentum Resolution and Reliability Measurements

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Abstract

A γ Compton experiment with an annular 241Am γ-source is described. The distribution of the primary intensity in the plane of the sample as well as the scattering angle distribution are evaluated by a Monte Carlo calculation to determine (together with the energy resolution) the total momentum resolution of the experiment. In order to probe the reliability of the experimental arrangement and of the data processing, experimental Compton profiles of water from samples of different thicknesses are compared with the results of an IUCr project.

1. Introduction

The measurement of the energy distribution of inelastically scattered photons provides the most direct way to get information about the distribution of electrons within momentum space. Since the distribution in momentum space is much more sensitive to variations of the electron structure due to chemical binding than the distribution in real space, Compton measurements have gained interest during the last ten years (for a recent review see Compton Scattering, 1977).

Measurements on substances with higher atomic number by conventional X-ray sources such as Mo Kα or Ag Kα combined with a crystal spectrometer become less effective because of the low ratio of Compton scattering to photoabsorption cross-section at these primary energies. Thus the combination of a higher energy γ-source with energy analysis of the scattered radiation by a solid-state detector can provide better efficiency. A 241Am source with the 59.5 keV γ-line combines long life and low cost. However, because of strong self absorption the photon flux at the surface of a 241Am source is limited to 4 x 10^{14} photons/(m^2 sr). In order to hold the scattering angle distribution as narrow as possible the effective emitting area of the source must be limited, which results in a limitation of the primary intensity. By using an annular source together with an appropriate scattering geometry one can enlarge the effective emitting area of the source appreciably (Weyrich, 1975; Bonse, Schröder & Schülke, 1977; Kramer, Krusius, Schröder & Schülke, 1977).

In the following we describe the lay-out of a Compton experiment with an annular 241Am source. We also report the results of a Monte Carlo calculation of the scattering angle distribution which was performed with the geometric parameters of the actual experiment. The aim of such a calculation is to find out the influence of the scattering angle distribution on the total momentum resolution and to see how large the spread of the scattering vector direction with respect to a fixed direction of the scattering sample really is. The Monte Carlo calculation allows the question to be answered, whether a 'directional' Compton profile obtained with an annular source is still a 'defined' quantity.

Finally, the experimental set-up and the whole procedure of data evaluation is tested by comparing measurements of Compton profiles on water with those obtained in the course of a project of the IUCr: Standardization of Compton Profile Measurements (Williams, 1976).

2. Experimental procedure

2.1 Apparatus

A section through the scattering chamber is shown in Fig. 1. The 1 Ci annular 241Am source (1) is located on a conical lead holder (2). With the beam shutter (3) open, the primary γ-rays hit the sample (5) which is mounted on a goniometer head (4). The radiation scattered inelastically at θ = 175° can pass into an 8 mm wide annular tunnel in the Pb shielding (6) and finally exit through the 0.4 mm Be window (7). The photons are detected by an intrinsic Ge detector (PAR 105, 5 mm thick Ge crystal with 100 mm^2 active area). The lead

![Fig. 1. Section through scattering chamber with annular source. (1) 241Am source; (2) lead holder; (3) beam shutter; (4) goniometer head; (5) sample; (6) tunnel in Pb shielding; (7) Be window; (8) dead end tube; (9) tube end.](https://example.com/fig1.png)

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shielding of the chamber has a thickness of 50 to 120 mm in all directions which, even with the extreme backward scattering geometry, lowers sufficiently the background of high-energy quanta. Photons not scattered by the sample hit the walls of the 1 m long dead-end tube (8), whereas the detector only sees the tube end (9). Thus it can be guaranteed that, as far as single scattering events are concerned, only photons scattered from the sample and not from the shielding walls or the goniometer head reach the detector. In order to permit in situ alignment of crystalline samples, the mounting base (11) of the goniometer head can be detached from the rest of the scattering chamber and can be screwed to an X-ray orientation goniometer.

If required, the scattering angle distribution may be narrowed further by positioning a lead collimator ring in the primary beam around the exit beam tube in front of the ring source. To prevent air scattering the apparatus is evacuated to ~1 Pa.

The signals of the solid-state detector are amplified in a spectroscopic amplifier and collected by a 1024 channel MCA with an energy spread of 40-50 eV per channel. The stability of the system is controlled by an on-line computer to an accuracy of better than half a channel over a measuring cycle typically of 24 h duration.

2.2 Monte Carlo simulation of the scattering angle distribution

As already mentioned in the introduction, the use of an annular source in a Compton experiment raises at least the following three questions. What is the shape of the intensity distribution in the plane of the sample? What is the angle-of-incidence distribution on the sample measured against the centrosymmetric axis of the scattering chamber? What is the actual distribution of scattering angles?

These questions can be answered quite precisely by a Monte Carlo (MC) simulation of the scattering process with the constraints of the geometry of the apparatus. Defining an elementary source point within an infinitely small sector of the 4 mm wide ring source and assuming the beam confining apertures as completely absorbing, allows the calculation of the intensity distribution by variation of the radial position of the source point and the beam direction by commonly used MC techniques. The resulting intensity distribution in the plane of the sample is shown in Fig. 2. The overall distribution of intensity incident from the ring source in the sample plane is obtained by performing the integration

\[ J(r) = \int_0^{2\pi} i(r, \phi) \, d\phi, \]

where \( i(r, \phi) \) is the intensity distribution as shown in Fig. 2 and \( \phi \) denotes the rotation about the \( z \) axis (see Fig. 1).

The resulting radial distribution is given in Fig. 3. \( r_{\text{det}} \) denotes the radius of acceptance of the detector, \( r_{90\%} \) the radii which enclose 90% of the total intensity in the plane of the sample with and without the collimator ring, respectively. It can be seen that only 2-5% of the primary quanta (passing the aperture system) are outside the range of the detector acceptance. Samples with diameters of 16-18 mm can utilize more than 90% of the available intensity in the sample plane.

The MC simulation calculates also the distribution of the angles of incidence of the primary quanta measured against the \( z \) axis of the scattering chamber (see Fig. 1) and thus against the sample orientation. The result is shown in Fig. 4(a). The average angle is about 5°, implying that with the annular source and the detecting system the scattering vector varies on a cone of mean aperture of about 2.5°. Sample orientations in Compton scattering experiments are usually determined to an accuracy of about 1°. Hence most directional Compton profile experiments can also be performed with the annular source arrangement.

Another important question for the interpretation of the measured profiles is the distribution of the scattering angles. Assuming the scattering cross-section to be constant over the relevant high-angle range and weighting the obtainable scattering angles from a specific point on the sample by the size of the visible detector area gives a sufficient simulation of the scattering process. Consequently, such a calculation

![Fig. 2. Distribution of intensity from a sector of the annular source in the plane of the sample without (left) and with (right) additional collimator ring.](image)

![Fig. 3. Radial intensity distribution in the plane of the sample with (---) and without (——) additional collimator ring, obtained by rotating the distribution of Fig. 2 around the \( z \) axis. \( r_{90\%} \) encloses 90% of the total intensity. \( r_{\text{det}} \) gives the maximum acceptance of the detector.](image)
involves no description of the scattering event itself, but
gives the distribution of scattering angles by a 'photon-
splitting' method. The result for about 40 000 'scattered'
photons is shown in Fig. 4(b). While the scattering angle
according to the geometric construction is 175°, the
average angle in the calculated distribution is slightly
shifted to 174.5° with a full width at half maximum
(FWHM) of 4.4 without and 3.0° with the collimator
ring installed.

2.3 Momentum resolution
The total momentum resolution of a Compton
experiment can be represented as the sum of the
variance of the detector resolution function and of an
effective scattering angle distribution (Williams, 1976):

$$\sigma_{\text{tot}}^2 = \sigma_{\text{det}}^2 + \sigma_{\text{ang}}^2$$

(2)
The first term is determined by the half-width of the
nearly Gaussian response function of the Ge detector to
a strictly monochromatic energy distribution ($\delta$
function). By measuring several $\gamma$- and X-ray lines in the
range 25 to 60 keV the half-width of the resolution
function at an energy corresponding to $p_z = 0$ was found
to be 325 eV.

With an uncertainty of about half a channel (25 eV)
in the stability control of the MCA and the amplifier, one
obtains $\sigma_{\text{det}} = 0.54$ at. units. Using the angular
distribution from our MC calculation, from

$$p_z (\text{at. units}) = -137 \frac{\omega_1 - \omega_2 - \omega_1 \omega_2 (1 - \cos \theta) / 511}{(\omega_1^2 + \omega_2^2 - 2 \omega_1 \omega_2 \cos \theta)^{1.2}}$$

(3)
with $\omega_1$ and $\omega_2$ the energy of the primary and scattered
photon (in keV), respectively, and the scattering angle $\theta$
obtained by MC calculations, we find

$$\frac{\partial p_z}{\partial \theta} \Big|_{p_z = 0} = 0.018 \text{ at. units} / ^\circ,$$

and for $\Delta \theta = 3$ with and 4.4° without collimator, we get
$\sigma_{\text{ang}} = 0.053$ and 0.078 at. units, respectively. Therefore,
for the special arrangement with nearly 180° back-
scattering geometry the influence of the finite width of
the scattering angle distribution in the total momentum
resolution can be neglected. Consequently, in the case of
crystalline or disordered samples, the width of the
scattering angle distribution could be enlarged appre-
ciably to enhance the photon flux at the scatterer. On
the other hand, in the case of single crystals the
scattering vector should be restricted to a cone of an
aperture not larger than 2–3°. Hence with single-crystal
experiments further enlargement of the scattering angle
distribution width is impossible.

3. Compton profile of water
As a test for our experimental arrangement, our
measurements of water Compton profiles should be
compared with those of an IUCr project (Williams,
1976).

The measurements were performed on samples with
thicknesses of 2, 4, 8, 16, and 32 mm. Going from the 32
mm to the 2 mm sample the signal to noise ratio at the
profile centre changed from 1500:1 to 230:1.

The data processing included the following steps:
(1) Background subtraction by fitting a linear
background.
(2) Energy calibration by means of the primary $\gamma$-line
and the Pb K$_\alpha_1$ and K$_\alpha_2$ fluorescence lines.
(3) Recursive method (Paatero, Manninen &
Paakkari, 1974) to eliminate the influence of the flat tail
of the detector resolution function.
(4) Transformation of the energy to a momentum
scale.
(5) Correction according to the energy-dependent
scattering cross-section (Eisenberger & Reed, 1974),
detector efficiency and sample absorption.
(6) Normalization from $p_z = 0$ to 4 at. units to the
respective integral of the theoretical 'near HF'
Compton profile of H$_2$O by Tanner & Epstein (1974).
(7) Deconvolution by a least-squares procedure
(Paatero, Manninen & Paakkari, 1974) assuming a
Gaussian total momentum resolution function with a
FWHM of 0.56 at. units.
(8) Correction for multiple scattering by a zero
thickness extrapolation technique as used by Weyrich
(1975).

Table 1 shows the results of our measurements on the
2 mm sample and the profiles after extrapolation to zero
thickness together with the mean value from the
following three experiments of the IUCr project: Mo
K$_\alpha$, $t = 3$ mm; $\gamma$-Te, $t = 3$ mm; $\gamma$-Am, $t = 1$ mm. The three
experiments are given for comparison since all
corrections to the raw data are done in nearly the same
manner as in our experiment. We claim very good
agreement of our zero thickness result with these
experiments within the accuracy which has been
obtained with absolute Compton profile measurements
up till now (Williams, 1976). The theoretical NHF
profile of Tanner & Epstein (1974) is also given in the
table.
Table 1. Comparison of experimental and theoretical deconvoluted water Compton profiles


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<tr>
<th>$P_x$</th>
<th>This work $t = 2.0$ mm</th>
<th>$t = 0$</th>
<th>IUCr project</th>
<th>Theoretical</th>
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4. Conclusions

The comparison of the experimental results from Compton profile measurements on water with those of the IUCr project together with the Monte Carlo calculations of the scattering angle distribution have shown that a Compton experiment using an annular $\gamma$-source and an extreme back-scattering geometry can give equal accuracy and total momentum resolution as a 'point'-source experiment with the advantage of a much higher utilization of the source activity.

This is not only true for polycrystalline or disordered samples but to some extent also for Compton scattering on single crystals as could be shown by considering the scattering vector spread connected with the back-scattering geometry used.

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References


