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A CURVED CRYSTAL SPECTROMETER FOR MEASUREMENTS OF SMALL K X-RAY ENERGY SHIFTS

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ABSTRACT

A curved crystal spectrometer of the DuMond type is described. It permits the measurement of the energy difference between K X-ray lines from two different sources that are mounted on top of each other at a close distance in a special source holder so that the reflections overlap in angle. The instrument has been used in a series of investigations of new and numerically very small effects influencing the energies of K X-rays from elements with $Z > 30$. For source strengths of the order of 1 Ci and for counting periods of the order of a day or less the precision obtained was as good as $4 \cdot 10^{-4}$ of the natural line width or, in units of the total energy $3 \cdot 10^{-7}$.

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1. INTRODUCTION

The energies of atomic K X-rays have small contributions that reflect the details of atomic and nuclear structure. Apart from the nuclear size and the chemical state of the atom^{1,2)}, additional mechanisms³⁻⁵⁾ like the 1s magnetic hyperfine structure, a dynamic effect connected with the excitation mechanism, and an effect associated with the difference in electronic structures between atoms occurring as mother and daughter in an electron capture decay process influence the K X-ray energies. The detection of these effects requires a very high precision, typically of the order of 10^{-3} of the natural line width or 10^{-6} of the energy. This precision can at the present stage of the experimental development be attained through diffraction techniques only.

The preferred technique in the earlier work^{1,2)} has been to use the Cauchois geometry⁶⁾ with an extended source and a slit detector. This method requires very strong samples with a weight of 0.1 - 10 grams and it is well suited for measuring shifts in energies between different sources since the effective source area can be defined by a fixed diaphragm. The DuMond geometry⁷⁾, on the other hand, uses a line source and a large detector. It offers an attractive two to three orders of magnitude in transmission over the Cauchois geometry and allows the use of correspondingly weaker sources for similar counting rates. Thus the DuMond geometry is the method of choice in all cases where the amount of source material is limited to ≤ 0.1 grams, when enriched isotopes should be used or where small sources can be produced e.g. at electromagnetic isotope separators. The source position at a DuMond spectrometer is as critical as the detector slit position of the Cauchois spectrometer : for the experiment discussed here, the effective source position must be constant to within 1 μm .

The DuMond geometry has already been employed for precise comparison measurements by Van Eijk et al.⁸⁾ who used a defining slit in front of the source to fix its effective position. In the following we describe a technique based on a novel source design and data-taking strategy. This technique, which has been used in our experiments³⁻⁵⁾, offers a precision comparable to what has been obtained previously and in addition a higher sensitivity, as can be seen from studies⁵⁾ involving the weak $K\beta_{2\mu}$ and $K\alpha$ lines.

2. EXPERIMENTAL TECHNIQUE

The essential new feature of our spectrometer is that the line source is split in two, one above and one below the mid-plane perpendicular to the rotation axis of the crystal whose Bragg angle θ is changed by means of a sine screw. A moving shutter in front of the two sources allows the data taking to alternate between them (quasi-simultaneous measurement of radiations from the two sources)

at a fixed angular setting of the instrument. The influence of the uncertainty in the sine screw setting is in this way strongly reduced and the fast alternation of the shutter position also goes a long way towards eliminating the effects of instabilities and drift. Still, the angular distance $\delta\theta_0$ under which the two sources are seen from the spectrometer crystal is critical if only one reflection is measured. This source distance $\delta\theta_0$ adds to the true angular distance $\delta\theta_E$ of the Bragg reflections, so that the total measured shift would be $\delta\theta = \delta\theta_0 + \delta\theta_E$. Here $\delta\theta_E$ is due to the energy shift δE of the X-ray lines with energies E_1 and E_2 . With $E_1 \approx E \approx E_2$ and $\delta E = E_1 - E_2$ one finds from Bragg's law

$$\delta\theta_E = - \frac{\tan \theta}{E} \cdot \delta E. \quad (1)$$

Whereas the sign of $\delta\theta_E$ depends on the sign of θ according to Eq. (1), the sign and magnitude of $\delta\theta_0$ is independent of θ . The influence of $\delta\theta_0$ can therefore be eliminated easily by a measurement of the reflections of the X-rays from both sources 1 and 2 under positive (+) and negative(-) Bragg angles. One then obtains $\delta\theta(+) = \theta_1(+) - \theta_2(+) = \delta\theta_0 + \delta\theta_E$ and $\delta\theta(-) = \theta_1(-) - \theta_2(-) = \delta\theta_0 - \delta\theta_E$ so that

$$\delta\theta_E = \frac{1}{2} (\delta\theta(+) - \delta\theta(-)). \quad (2)$$

It is clear that this strategy does not entail any loss in statistical efficiency. It is comparable in accuracy to precision interferometry⁹⁾ and safer and no excessive mechanical accuracy is required for the crystal bearing¹⁰⁾ as long as the sources 1 and 2 are adjusted so well above each other that their X-ray line reflections overlap well.

The description given here assumes that the sources are symmetrically placed with respect to the mid-plane perpendicular to the crystal plane. If this is not the case, an additional correction enters, which will be discussed below.

In the following the apparatus is described in more detail. The first two sections deal with the two main mechanical parts, the crystal, drive and detector shown in Fig. 1 and the source holder shown in greater detail in Fig. 2. A third section briefly discusses some aspects of the source preparation.

2.1 Mechanical construction of the spectrometer

The CERN-Jülich DuMond type crystal spectrometer is depicted in Fig. 1. It consists of a quartz crystal of dimensions 100 mm × 100 mm × 4 mm which is cut and oriented in such a way that the reflection occurs from the 110 plane. The crystal is bent by imprisonment between two steel blocks to a focal length of 464 cm. The useful part of the crystal is defined by a 5.0 cm diameter window in the steel blocks.

The crystal clamping blocks (marked (1) in Fig. 1) are fixed to a plate (2) which allows the necessary adjustments of the crystal and which is mounted on the outer part of the crystal bearing (3). This carries also the arm (4) which serves to rotate the crystal and two lead counter-weights (5). The central part of the crystal bearing consists of two very precise shoulder ball bearings on the upper and lower ends of a pivot onto which the main support is rigidly fixed. A spring pressing the inner rings of the bearings against each other minimizes the play of the crystal bearing.

The crystal is rotated by means of a twin sine screw (6) which is driven by a disc motor in conjunction with a gear (7). The motor runs at high speed as long as the crystal angle θ differs from the preselected angle θ_{Bragg} by more than ~ 15 arc sec. Its speed is then reduced and for $\theta \approx \theta_{\text{Bragg}}$, the final actions of the motor are achieved by pulses from the electronic system controlling the motor current. The photons fulfilling the energy condition for Bragg reflection in the quartz at the selected angle pass a Soller slit collimator (8) with a transmission of 78%. This collimator effectively absorbs the intense direct beam at all angles except those corresponding to first order reflection of photons with energies above 1 MeV. A NaI(Tl) scintillator with a diameter of 7.6 cm and a thickness of 6 mm is used for the detection of the low-energy photons. The detector shielding (9) consists of lead with a thickness of 10 cm. The detector is moved around the spectrometer axis, which is defined by the vertical passing through the center of the quartz crystal. The sliding of the detector part takes place on two sections of two-fold wire ball bearings (10), one shorter section carrying the detector-shielding and a larger section (11) extending over the whole carrier which is resting on the main support (12) and which can be adjusted so that the centre of the arcs formed by the bearings falls on the spectrometer axis. The detection unit is moved by a motor (13) with a tooth-wheel drive along a chain attached to the carrier at its outer face. The fine adjustment is achieved with the aid of an excenter drive (14). The motor for the coarse movement and that of the excenter are also controlled by the electronic system of the spectrometer. The detector is kept in the orientation of the reflected X-ray beam by means of a laser (15) beam which is reflected in a mirror (16) on top of the crystal into a differential photocell (17) mounted on the lead shield of the detector. The scanning of a preselected energy region is controlled by an automatic electronic system.

The instrument allows measurements to be performed in an angular range of $\pm 12.5^\circ$ which corresponds to X-ray energies larger than 23 keV in second-order reflection. X-rays with lower energy can be recorded in first order reflection but absorption, mainly in the crystal, sets a practical lower limit at about 20 keV.

2.2 Source arrangement

The sources for the spectrometer (Figs. 2 and 3) consist of 10 mm cubic aluminum blocks with a small extension with dimensions 3 mm $\times$ 4 mm $\times$ 4 mm which carries the radioactive sample or the fluorescence excitation material on its inner face. The shape has been chosen so that the source can be mounted either in the upper or in the lower position in the source holder. This arrangement is important for the elimination of the up-down asymmetry, to be discussed below. The sources are mounted very closely one above the other in a way so that the projection of the active area is seen by the spectrometer as a line source. The sources can be adjusted individually in the direction of the dispersion by means of micrometer screws. In front of the sources is placed a 4 mm thick uranium plate, the shutter, with two openings so that only one source can be seen from the spectrometer at any given time. The shutter is driven by means of two electromagnets which can be easily operated in such a way that the opening time for the individual sources is optimized according to the source strengths. The shutter movements are controlled by an optical system which activates two gates so that the detected X-rays are counted separately for each source. For each measuring point the scaler contents, angular position, and measuring time are stored. The source arrangement is enclosed in a lead vessel with 5 cm thick walls. The complete unit can be moved in the focal direction, around the vertical axis and along the vertical axis for alignment. The adjustments can be controlled by a laser.

2.3 Source preparation

2.3.1 ISOLDE produced sources

Radioactive nuclei produced in a target by means of the 600 MeV proton beam of the CERN synchrocyclotron were mass-separated in the ISOLDE electromagnetic isotope separator^{11,12}). Target and ion-source arrangements¹³) are available so that chemical and mass separated beams of a number of elements can be obtained. The radioactive sources are collected by allowing a monoisotopic beam to impinge directly onto the aluminum blocks. Two different collector positions are available. In the collector chamber of the separator up to five different masses can be collected simultaneously (Fig. 3). The aluminum blocks are in this case mounted onto a small carriage so that corrections for changes in the mass positions can be made during the collection. The size of a sample collected in this position is 4 mm². A source size of 0.5 mm² can be obtained by collection in one of the external beam lines of the separator. The collector blocks are connected to a precision electrometer so that the current of the ion beam can be monitored and the position optimized.

2.3.2 Reactor produced sources

Other sources have been prepared by neutron activation. In some cases isotopically enriched metal foils (size 3 mm $\times$ 4 mm $\times$ 0.1 mm) were glued onto the aluminum blocks under pressure in order to provide a planar activity distribution. In other cases a thin layer of the enriched material is evaporated onto the aluminum source block before activation in a nuclear reactor.

2.3.3 Photoionization sources

In cases where the reference source is obtained by fluorescence excitation, a metallic foil of the element of interest is glued to the aluminum block. The foil is illuminated by a 40 Ci ^{169}Yb metal source, enclosed in an aluminum housing positioned at a distance of 0.4 mm. In order to increase the fluorescence yield, a thorium plate is inserted in the aluminum block behind the metal foil. The direct radiation from the ^{169}Yb source is prevented from reaching the other source and the spectrometer by means of uranium plates, 1 mm and 4 mm thick, respectively (see Fig. 2).

2.4 Source positioning and spectrometer calibration

When the sources have been put into the source holder, a first adjustment of them can be made by means of the micrometer screws so that they are centered with respect to the chopper slit. Then their mutual position is checked by scanning across a strong reflection. A final adjustment is then made again with the micrometer screws so that maximum overlap is achieved.

Since the distance between crystal and source is kept constant during the measurement, the focussing condition is only fulfilled for one value of the Bragg angle. In order to find the correct focal length for the line in question, the source holder is adjusted in this direction until the minimum width is achieved.

If the horizontal symmetry plane of the source arrangement does not coincide with that of the crystal, the sources will see the reflecting lattice planes under somewhat different angles, i.e. the reflected photons are registered at different angles too, thus introducing a systematic error^{14,15}). To investigate this effect, we used two identical ^{169}Yb sources and measured the strongest gamma lines in different orders of reflection. From the measured line shifts the displacement of the source height could be calculated and corrected accordingly. Instead of determining the instrumental asymmetry in this way one can also eliminate it by carrying out an additional line-shift measurement with the sources 1 and 2 interchanged. Unless symmetry of the source arrangement is then established, data taken at other Bragg angles Θ have to be corrected accordingly, or the additional measurement has to be done whenever Θ is changed.

3. MEASUREMENTS

3.1 Method of measurement

According to the Bragg law $E = nhc/2d \sin \theta$ the energy E of a reflected photon is inversely proportional to the sine of the reflection angle θ . With the sine screw we do not measure θ but directly the quantity $x = \text{const.} \cdot \sin \theta$. The small difference in energy, δE , can thus be obtained from the measured shifts δx of the line position as $\delta E/E \approx -(\delta x - \delta x_0)/x$ because $\delta x_E = \delta x - \delta x_0$, where δx , δx_0 and δx_E correspond to the angular shifts $\delta \theta$, $\delta \theta_0$ and $\delta \theta_E$ mentioned in section 2. To eliminate the remaining contribution δx_0 to the shift resulting from the difference of the zero positions of the sources, the reflections are measured at positive and negative Bragg angles, so that the above relation reduces to $(E_1 - E_2)/E = \frac{1}{2} \frac{(x_{1+} - x_{2+}) - (x_{1-} - x_{2-})}{x}$ where the indices 1^+ , 1^- and 2^+ , 2^- label the lines from sources 1 or 2 measured under positive or negative Bragg angles as outlined in Section 2.

In each run between 15 and 20 equidistant points were taken across the reflection both at positive and negative Bragg angles with a range chosen as 1.1 - 1.6 times the line width (FWHM). In order to follow an optimum measuring strategy the counting time per point (t_1 or t_2 for source 1 or source 2, respectively) was constant and distributed between the two sources so that the ratio t_1/t_2 equals roughly the square root of the intensity ratio I_2/I_1 . (A general discussion of measuring strategies have recently been given by Hansen¹⁶.) To average over possible drifts, both sources were measured for many shutter cycles at each angular setting and for each line 10-20 runs were performed.

3.2 Data analysis

Owing to the smallness of the expected shifts (of the order of 10^{-2} - 10^{-3} of the FWHM), a very careful analysis of the data and of possible sources of systematic errors is essential. One of the delicate points is that the line profiles cannot be assumed to be the same for both sources. The analysis therefore must be performed in a way that will make systematic errors arising from this cancel. Experience has shown that this is possible and the uncertainty on our results³⁻⁵) is, in fact, mainly due to counting statistics.

We have used four different methods of analysis, which give approximately the same answers :

(i) A fit to the shape of the line where a set of parameters a_i in a function

$$y = \frac{a_1}{1 + \left(\frac{x-a_0}{a_3}\right)^2 a_2 + \left(\frac{x-a_0}{a_3}\right)^4 a_4 + \left(\frac{x-a_0}{a_3}\right)^6 a_6}$$

were determined. The parameter a_0 , of course, gives the line position. The parameters a_1 , a_2 , a_3 , a_4 and a_6 are the same for the reflections under positive and negative Bragg angle.

(ii) A method where the slope at a given point is used to calculate the shift from the difference of the intensities of the normalized lines. This method can be applied to any pair of measured points and the net result is obtained as a weighted average.

(iii) The area between the normalized reflections can also be used to determine the shift. The integration is done numerically according to Simpson's rule using all of the measured points. This method has the advantage of being insensitive to background fluctuations and is in the first order of approximation independent of the line shape and differences in line shape between the two sources.

(iv) The method finally adopted represents a development of statistical techniques for finding "robust" or distribution-free estimates. Since it has already been discussed in detail¹⁶⁾, we mention it only briefly here. The location parameter for a line is estimated from the centre of gravity of a line segment to which a bias correction is applied. This correction is typically a factor of 2. This method of estimation can be shown to be almost 100% efficient in the statistical sense.

3.3 Examples of experimental results

As a first check of the instrument a measurement was performed on two identical sources of 2 Ci ^{169}Yb . The width (FWHM) of the reflection of the 63 keV gamma line was 5 arc sec which is an upper limit of the instrumental resolution. The analysis of 30 successive scans of the 63 keV line showed that the local reproducibility is of the order of 0.2 arc sec. A typical X-ray spectrum is depicted in Fig. 4

In the series of experiments with the spectrometer three new effects that cause shifts of the K X-rays have been identified :

- a) Hyperfine shifts caused by a selection rule in electron capture beta decay³⁾,
- b) Shifts from unresolved satellite lines from outer-shell excitations in photoionization⁴⁾, and
- c) Shifts caused by the difference in electron configuration of an atom after electron capture decay as compared with the normal atom⁵⁾.

All of these effects must be considered whenever X-rays are used as energy standards.

As examples of the precision achieved in these three experiments, Table 1 gives the results for $K_{\alpha 1}$ lines of three different sources. The quoted errors include the uncertainty of the asymmetry correction.

4. CONCLUDING REMARKS

The experimental techniques described here are a powerful tool for the study of the mechanisms governing the energies of K X-rays. Our most recent experiments extend the studies of ref. 5) to the 5d elements and show^{17,18)} that one or more additional effects of a nature not yet clarified must play a rôle.

The basic principle of our method restricts it to cases where the reflections of lines from different sources can be brought to overlap. We have recently pointed out¹⁹⁾ that the principle can be extended to cases in which the lines differ in energy but overlap when measured in different orders n of Bragg reflection, in other words where one has a relation

$$E_1/n_1 - E_2/n_2 = \Delta \cong 0.$$

This technique has served¹⁹⁾ to measure very accurate energy ratios for X-rays from light and heavy elements.

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Table 1
Energy shifts for $K\alpha_1$ X-rays

Element	Source 1	Source 2	Energy keV	$E_1 - E_2$ meV ²
$^{45}_{Rh}$	$^{103}_{Rh}$, PI a)	$^{103}_{Pd}$, EC a)	20.22	222 ± 10
$^{54}_{Xe}$	$^{131}_{Cs}$, EC	$^{132}_{Cs}$, EC	29.78	$112 \pm 11^b)$
$^{65}_{Tb}$	$^{159}_{Tb}$, PI	$^{159}_{Dy}$, EC	44.48	-560 ± 17
$^{67}_{Ho}$	$^{165}_{Ho}$, PI	$^{165}_{Er}$, EC	47.55	$505 \pm 22^c)$
$^{79}_{Au}$	$^{197}_{Au}$, PI	$^{197}_{Hg}$, EC	68.81	$568 \pm 36^d)$

a) The excitation modes are EC : electron-capture beta decay;
PI : photoionization.

b) Ref. 3)

c) Ref. 5)

d) Ref. 4)

FIGURE CAPTIONS

Fig. 1 : Schematic drawing of the crystal spectrometer in the position of zero Bragg angle. The source arrangement is placed at a distance of 4.6 m from the crystal. The numbers refer to the text of section 2.1.

Fig. 2 : Sketch of the source arrangement with one radioactive source (source 1) and a metal foil (source 2) excited by radiation from a strong ^{169}Yb source. The gamma-rays that have sufficient energy to penetrate the metal of source 2 (and the Al support) will usually excite photofluorescence in the thorium foil or will be partly backscattered. In this way the efficiency of the photoionization is increased by about 30%. A thin uranium plate between the sources (not shown in this figure) shields source 1 against the ^{169}Yb source. The U shield strongly absorbs radiation from the Yb source to the crystal spectrometer. The radiation from source 2 passes the slit in the U shield.

Fig. 3 : The left part of the figure shows a source holder after an irradiation in the separator chamber of the ISOLDE isotope separator. The right part shows the trolley used for the collection of several sources simultaneously.

Fig. 4 : Spectrum of ^{189}Ir EC decay in the Os K X-ray energy range. The scan covers the energy region between the $K_{\alpha 1}$ line and the $K\beta$ line of Os in second order reflection. The width of the 69.6 keV gamma line is a measure of the good resolution of the instrument and indicates that the widths of the X-ray lines are essentially determined by their natural line widths. The peak at "72.4" keV is the first order reflection of the 36.2 keV γ line.

Fig. 1

Fig. 2

Fig. 3

Fig. 4