MEASUREMENT OF ELECTRONIC ISOTOPE SHIFTS WITH THE CURVED CRYSTAL SPECTROMETER AT ISOLDE

G.I. Borchert, O.W.B. Schult
Institut für Kernphysik, Kernforschungsanlage Jülich, 5170 Jülich, F.R. Germany

P.G. Hansen *, B. Jonson **, H.L. Ravn
CERN, Geneva, Switzerland

Abstract

The electronic isotope shifts of the X-rays following the electron capture of neutron-deficient nuclei have been investigated with a curved crystal spectrometer. The isotopes $^{127}$Xe, $^{129}$Xe, $^{131}$Xe and $^{133}$Xe have been studied. The measurements yield a small shift for the isotopes $^{127}$Xe, $^{129}$Xe and $^{133}$Xe, whereas a significant effect is observed at $^{131}$Xe. The result could indicate a change of the m.s. radius between the ground state and excited states.

1. Introduction

The binding energies of the K electrons depend primarily on the nuclear charge, its density distribution and the configuration of the electron cloud. A change of one of these properties leads to a change of the transition energy between the corresponding atomic levels referred to as isotope shift, isomer shift and chemical shift, respectively. Therefore, the study of X-ray energies is a useful tool to get information about the wave functions of inner shell electrons and the nuclear m.s. radius. The first studies of these effects have been carried out by Sumbarev and Bormann, who used curved crystal spectrometers of the Gouchois type to detect X-rays excited in samples of stable isotopes by photoionization. In the present work we attempt to extend studies of this type to K-X-rays following electron-capture. Such experiments seem to offer possibilities of investigating the nuclear volume effect not only over a wide range in A but also of extremely short-lived excited levels.

2. The Experiment

As the expected shifts of the X-ray transitions are only of the order of $0.1 - 1.0\%$ of the natural line width, very precise measurements have to be performed using well suited sources. The high beam intensities of the improved mass separator ISOLDE-2 at the CERN synchrocyclotron has made it possible to obtain very strong monoisotopic sources up to 1 Ci of sufficiently small size, so that a curved crystal spectrometer in Babinet geometry can be used. The use of the sources is combined with the high resolution of the diffractometer.

The instrument is equipped with a 4 mm thick quartz crystal which is reflecting the photons at the (110) plane (d = 2.5 A) with high reflectivity through a window of 5 cm diameter. The crystal is bent to a focal length of 4.64 m between clamping blocks. The angular setting is accomplished with a sine screw, the position of which is controlled through an electronic regulation device by means of an angle decoder. One step unit corresponds to 0.1".

The maximum angular range is ± 150°. The detector is a 76 mm diameter x 6 mm NaI scintillation counter, which is shielded against the unfocused beam by means of a Boller slit collimator. An optical regulation unit, using a laser beam keeps it in the correct position with respect to the diffracted X-ray beam. To achieve the high relative accuracy the instrument is equipped with a chopper system which allows to study 2 different isotopes relative to each other at the same time without any movement of the sources or the crystal. In this way systematic errors can be minimized. The beam of radioactivity was in favourable cases exceeding $10^{10}$ atoms/s (see the paper of H. Ravn). The radioactivity was implanted on spots with 2 mm diameter on metal foils placed in the collector chamber of the separator.

The monoisotopic sources were cut out from the end plate strip, inserted into the source holder and adjusted to minimum line width. The resolution of the spectrometer was in first order as small as 11 eV at 30 keV compared with the natural line width of the Xe $K\beta$ Röntgenlines of Xe: $\tau = 17$ eV ($E = 29.8$ keV). In order to minimize systematic errors each measuring run comprises the following steps. The measurement was started with the crystal set at an angle near the reflection position of the X-ray of interest. The reflection was then recorded point by point. Counts were accumulated at each position for a preset time. By means of the chopper and a suitable gate X-rays from the two sources were recorded in different counters. After about 20 steps the same procedure was used for the measurement of the reflection at the negative Bragg angle.

Each measurement consists of 10 - 20 of such runs. The experimental data were punched on paper tape for further reduction at a FEU 15 computer.

Owing to the smallness of the shift of the reflections a very careful analysis of the experimental data was necessary. Therefore, all the data were analysed in 3 independent ways:

1) Each pair of reflections at positive and negative Bragg angles was fitted by an expression of the form

$$Z = A/(1 + B(\sin \theta / \lambda)^2) + C(\sin \theta / \lambda)^4 + D(\sin \theta / \lambda)^6 + U$$

The centroid position $\bar{\theta}$ and the amplitude $A$ are allowed to vary independently for positive and negative reflections. The difference $\bar{\theta}_1 - \bar{\theta}_2$ of the centroid distances yielded the shift in angular units. The conversion to energy was obtained with the help of the Bragg angle $\bar{\theta}$. A typical pair of reflections is plotted in fig. 1. In this case the line width of the two sources is a little bit different. The position of one source is shifted with respect to that of the other by 0.25°. The calculated difference of the centroid distances amounts to 0.14°.

2) The distance of the reflections can be calculated from the difference of the normalized intensities at the same position by means of the slope. This is done for each pair and addition of the total number of points per reflections.

3) The difference of the area between the normalized reflections is a measure for the unknown shift, which thus can be deduced from it.

* On leave from the Institute of Physics, University of Aarhus, Denmark.

** Visitor from the Department of Physics, Chalmers University of Technology, Gothenburg, Sweden.
Fig. 1: Reflections at positive and negative Bragg angles of the Kα1 lines of $^{131}$Xe (dots) and of $^{129}$Xe (crosses). The corresponding fit curves are the full line and the dashed one, respectively.

The weighted means from the 10–20 runs of one measurement were then calculated applying each of the three methods. The resulting three numbers were averaged to obtain the final result.

A total of 8 independent experiments have been performed for the study of the isotopes $^{127}$Xe, $^{129}$Xe, $^{131}$Xe and $^{132}$Xe. The experimental data are listed in Table 1. The runs with the sources on different backings were performed in order to investigate a possible influence of chemical effects on the measured shifts. As can be seen from the individual values there is no evidence for such an influence.

Table 1: Experimental results of the Kα1 X-ray shift measurement. The upper line shows the combination in which the different isotopes have been studied. The next three lines contain the results of the individual measurements: $\delta \varepsilon = E - E_0$. In the case of the $^{129}$Xe – $^{131}$Xe study one measurement was performed with both Ca sources on aluminium backing and two measurements with the use of copper as backing. The material of the corresponding backing is denoted in front of the result. In the following line the mean values $\delta \varepsilon$ are listed. The errors quoted here are not pure statistical ones but include the scattering of the individual data and an estimate for possible systematical uncertainties. If one considers only the lowest nuclear moments, $\delta \langle r^2 \rangle$ can be calculated from $\delta \varepsilon$ within a few percent accuracy with the help of the relation $\delta \langle r^2 \rangle / \delta \varepsilon = -g_0^2 / g_1$:

$\delta \varepsilon = E - E_0 = C_1 \delta \langle r^2 \rangle = C_1 (\langle r^2 \rangle_0 - \langle r^2 \rangle)$

The values are given in the next line. The last line contains $\delta \langle r^2 \rangle$ values calculated with the use of a simple uniform charge model.

$\begin{array}{cccccc}
^{127}\text{Xe} & ^{129}\text{Xe} & ^{131}\text{Xe} & ^{132}\text{Xe} \\
\delta \varepsilon_{\text{exp}} \text{ fm}^2 & -4.15 & -7.5 & -6.9 & -6.3 \\
E_\text{meV} & 103 & 8 & 62 & 15 \\
\delta \langle r^2 \rangle_{\text{exp}} \text{ fm}^2 & -0.039 \pm 0.116 & -0.558 \pm 0.116 & 0.343 \pm 0.116 & -0.504 \pm 0.116 \\
\delta \langle r^2 \rangle_{\text{th}} \text{ fm}^2 & 0.141 & 0.139 & 0.076 & \\
\end{array}$

According to a simple uniform charged model the following estimate holds for the $\delta \langle r^2 \rangle$:

$\delta \langle r^2 \rangle = \langle r^2 \rangle_0 - \langle r^2 \rangle = R^2 (0.4 A - \frac{A}{2} + \frac{1.4}{A}) (\langle \rho^2 \rangle_0 - \langle \rho^2 \rangle)$

$R = 1.2 \times A^{1/3}$ fm

The symbols have their usual meaning. The index 0 denotes the heavier isotope. The E(22) values of the even isotopes allow an estimate of the deformation parameter $\beta$ which is linearly interpolated in order to obtain an estimate for the odd nuclei. For a comparison, these values are listed in the last line of Table 1. The experimental results and the model estimates are also shown in Fig. 2. In addition, the results from the optical measurements of Fischer\(^{2}\) and Hellwig\(^{3}\) are included. The optical data and the model estimates show a rather smooth behaviour that can be approximated by two straight lines with different slopes. On the contrary, the results of the present experiment show a significant discontinuity at the mass number 131. The jump amounts to about 6 times the uncertainty and is well established by the repetition of the experiments (see Table 1) and the cross-over measurement ($^{127}$Xe – $^{133}$Xe).

Fig. 2: Results of the X-ray shift measurement compared with optical data\(^{4,6}\) and the prediction of the simple uniform charge model. The ordinate is $\delta \langle r^2 \rangle / \delta \varepsilon$ and the prediction of the simple uniform charge model.

The effect most likely represents a volume shift. A chemical shift related to the backing and to radiation damage caused by the ion implantation cannot be entirely excluded, but seems less likely in view of the consistent results obtained (Table 1) in different runs and with different backings. The internal consistency of the data is confirmed by the $^{129}$Xe – $^{131}$Xe cross-check, which is entirely in agreement with the other data.

The optical data\(^{5,6}\) allow us to exclude an isotope shift as an explanation, and it is therefore necessary to consider the excited states populated in the $\beta$ decay. Figure 3 shows the corresponding decay schemes containing only the main levels of these Xe isotopes. The situation is especially simple for the decay schemes of $^{131}$Xe and $^{132}$Xe. In the former case the total decay intensity populates the ground state of $^{131}$Xe, so that the X-rays following the electron capture should reflect the ground state radius. In
and transitions to several excited states. In addition, for the low-lying states the internal conversion becomes a competing process, which causes Röntgen transitions corresponding to the ground state mass radius. The approximate intensities of the different contributions are given in Table 2. It is tempting to surmise that the large mass radius observed for the two very similar nuclei again reflects a larger charge radius in the excited levels.

<table>
<thead>
<tr>
<th>State</th>
<th>A = 127</th>
<th>A = 129</th>
<th>A = 131</th>
<th>A = 132</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 keV</td>
<td>20 %</td>
<td>52 %</td>
<td>100 %</td>
<td>0 %</td>
</tr>
<tr>
<td>411</td>
<td>70 %</td>
<td>44 %</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>590</td>
<td>10 %</td>
<td>4 %</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>668</td>
<td>-</td>
<td>-</td>
<td>100 %</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 2: Fractions of K-X-ray intensity emitted from the nuclei A while they are in different states of excitation.

At this point it only remains to discuss the magnitude of the presumed volume effect reported in this work (0.5 fm²) in relation to similar effects observed elsewhere in nuclear physics. To start with one may take the 3⁻ vibrational level in doubly-magic 208Pb, a nucleus expected to be very stiff and for which a theoretical estimate⁶,⁹ gives δ(r²) = 0.05 fm². The isomer shifts in the 2⁺ levels of rotational nuclei, on the other hand, are very much smaller except for the transitional regions, where they approach 0.1 fm². Finally, the volume shifts⁶) associated with the onset of permanent deformations are at the scale of the shift observed in the present work. The similar effect for two different states in the same nucleus would thus seem to indicate that the time average of δ² for the 2⁺ level in 132Xe is close to the value for a permanently deformed nucleus.

Acknowledgement

We are grateful to Prof. Dr. J. Specht for a discussion about isomer shifts.

References

1) O.I. Sumbayev, Proc. Int. Conf. on Inner Shell Ionization Phenomena Atlanta 72 and earlier papers.
4) F. Boehm, Proc. Int. Conf. on Inner Shell Ionization Phenomena Atlanta 72
5) W. Fischer et al. Z. Physik 270, 113 (1974)