Tests of internal-conversion theory with precise γ- and x-ray spectroscopy: Selection of new cases

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After having completed eight precise measurements of Internal Conversion Coefficients (ICCs) at the Cyclotron Institute, we find consistent agreement with the ICCs that are calculated with full account being taken of the atomic vacancy, or hole, left behind by the conversion electron [1]. Before 2004, when we published our first measurement, calculations that ignored the hole were accepted and the code BrIcc [2], which was based on those calculations, was in standard use for the Evaluated Nuclear Structure Data File (ENSDF), the nuclear data base maintained by the US National Nuclear Data Center (NNDC). However, based on our early results, since 2004 the NNDC has adopted the calculations that include the hole and BrIcc has been modified accordingly. Our supplementary measurements have continued to support this decision. This is a rather rare example – perhaps insufficiently popularized – of basic university research serving not only basic science but also broader societal applications.

When we started the series of measurements our interest was to tackle a controversial conclusion coming from an important survey work [3], which compared ICC calculated and measured values. The authors found that the 'no-hole' calculations showed better agreement with the measured values. Yet the time for the converted electron to leave the atom can easily be calculated and shown to be much shorter that the known lifetime of a K-shell vacancy (10^{-18} s $<<$ $10^{-15} - 10^{-17}$ s). In order to clarify this situation, we chose to measure or re-measure the ICCs of transitions for which the relative difference between 'hole' and 'no hole' calculations is of several percent. With our experimental precision of ~1% we could convincingly discriminate between the two types of calculation in such cases. Our first measurement was of the most favorable case, that of the 80.2-keV $M4$ transition in ^{192m}Ir, where there is an 11% difference between 'hole' and 'no hole' calculated values. Our result agreed completely with the 'hole' value and disagreed by more than 10 standard deviations from the 'no hole' one.

This result also suggested that the problem with the survey result [3] may have lain simply with the overall lack of precision and quality of the experimental set of ICCs available at the time. Indeed, of the 100 experimental cases used, only five had a precision of < 2%, while most were in the range of 5%. In the series of measurements that we have continued since then, we have more than doubled the number of measured ICCs with ~1% precision. In some cases, we corrected previous ICC results; in others we measured the ICCs precisely for the first time. Overall we have chosen transitions that spanned a range of Z and A values, γ-ray energies and multipolarities so as to investigate the efficacy of the 'hole' theory under a variety of conditions. So far [1] we have completed measurements of eight transitions in the following nuclei: ^{191}Ir, ^{134}Cs, ^{137}Ba, ^{139}La, ^{197}Pt, ^{119}Sn, ^{127}Te and ^{111}Cd (the last two are still to be published). In all cases, the results support the ICCs calculated with the atomic vacancy included.

In looking to the future we have recently completed a systematic investigation of what cases remain that we can usefully measure with our experimental technique, which depends on there being a single predominant transition that converts in the K shell, allowing us to determine α_K from the ratio of the K x-ray intensity to the corresponding γ-ray intensity, both being measured in the same well-calibrated HPGe detector. The following is an outline of the procedures that we followed:

1. As main guidance for our search, we used Fig. 1, which displays the difference Δ_K between α_K(hole) and α_K(no hole) expressed as a percent and plotted as a function of the kinetic energy of the conversion-electron energy E_K for four multipolarities and four Z values. Evidently the most interesting cases – the ones with the highest Δ_K – have to have the highest multipolarities (E3, M4) and the lowest values of E_k.
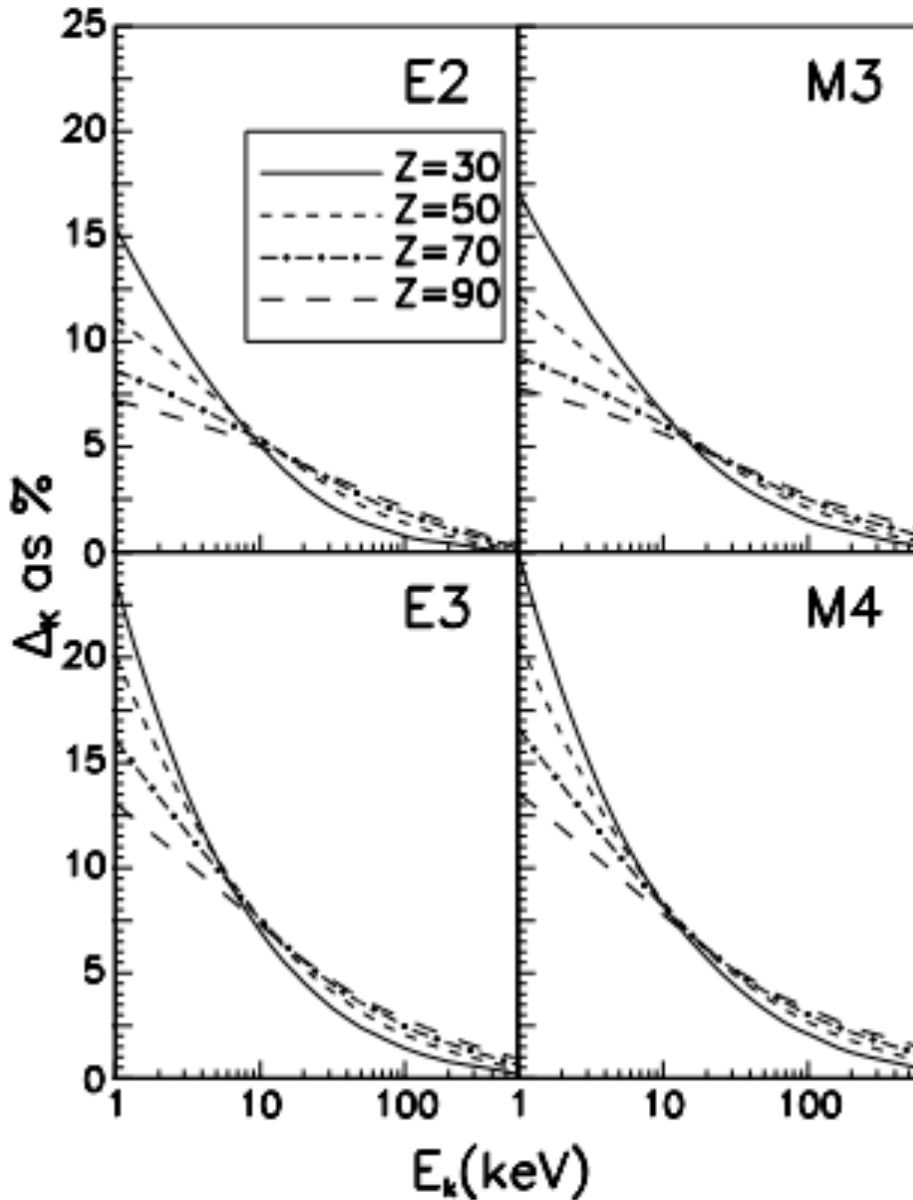
FIG. 1. The difference Δ_K between α_K('hole') and α_K('no hole') plotted as function of the kinetic energy of conversion electron E_K

2. To achieve sufficient counting rate, we depend upon neutron activation of our sources with the TRIGA reactor at Texas A&M University. Consequently we are limited to relatively long-lived nuclides, $T_{1/2} > 0.5$ h since it takes at least an hour to extract an activated source and transport it to our detector laboratory. With this condition we effectively exclude low multipolarity, $M1$ and $E2$, transitions.

3. With these conditions set we conducted a search with *NuDat*, the NNDC nuclear-data web tool based on the ENSDF database, and found a preliminary set of α_K candidates: 14 $E3$, 9 $M3$, 2 $E4$, and 8 $M4$ transitions, a total of 33 all having Δ_K of between 5 and 13%.

4. Of these, we excluded the nuclides whose production cross sections were too small to ensure enough counting statistics for the peaks of interest.

5. Also from the production point of view, we explicitly filtered out cases in which contaminant activities could play a seriously detrimental role.

6. We also excluded those nuclides in which other strongly converted transitions would contribute to the K x-ray spectrum.

7. We separately examined the 100 experimental α_K cases taken into consideration in the previously mentioned survey [3] looking for cases that disagreed with both 'hole' and 'no hole' calculations or otherwise showed indications of atypical or abnormal behavior.

8. Of the promising cases that were thus identified, four more had to be excluded for a specific reason:

- ^{58m}Co was eliminated because its 7-8 keV K x rays are too soft to be measured with our HPGe detector; we could use a Si detector but that would necessitate very thorough new efficiency calibration work.
- ^{93m}Nb, while a very good candidate ($\Delta_K = 8\%$, single 30-keV γ ray, clean and easy to measure), is commercially unavailable and we cannot produce it locally.
- ^{130m}I and ^{144m}Pr are also good candidates but have $t_{1/2}$ of 12 min and 17 min respectively and could perhaps be measured close to the reactor used to produce them. However because of the critical role our HPGe detector plays in our precision β-decay measurements we prefer not to move it away from the Cyclotron Institute.

9. Finally the cases that we can measure at this time are:

- ^{125m}Te: 109.2-keV, $M4$ γ ray, whose measured $\alpha_K = 167(7)$ in ref. [3] disagrees with both calculations, α_K(hole) = 185 and α_K(no hole) = 179. It can easily be produced by thermal-neutron activation from the enriched ^{124}Te isotope.
- ^{103m}Rh: 39.8-keV, $E3$ γ ray from ^{103}Ru β^- decay populated by thermal neutron activation of ^{102}Ru. ^{103m}Rh can also be populated from ^{103}Pd ec decay but the enriched ^{102}Pd isotope needed to produce ^{103}Pd is far too expensive. The most precise α_K values previously measured, 127(6) [4], and 138(5) [5] are of no help in distinguishing between the two calculations, α_K(no hole) = 127 and α_K(hole) = 135.

In conclusion, all the six cases, ^{58m}Co, ^{93m}Nb, ^{130m}I , ^{144m}Pr, ^{125m}Te, and ^{103m}Rh, are approachable by our method but the first four offer significant challenges. For now, we plan to measure the last two, ^{125m}Te, and ^{103m}Rh.

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