Tests of internal-conversion theory and efficiency calibration with precise γ- and x-ray spectroscopy: $^{134}Cs^m$, ^{137}Ba, ^{139}La

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Our program of precision measurements of internal conversion coefficients (ICC) continued with the completion of the $^{134}Cs^m$ and ^{137}Ba cases, and the measurement of a new case, ^{139}La. This program was motivated by a comprehensive study published in 2002 by Raman *et al.* [1], in which various methods for calculating ICC's were reviewed and the results from each were compared with one hundred selected transitions whose experimentally determined ICC's were claimed to 6% precision or better. Of the various calculations examined, the best agreement with experiment was obtained by the "Relativistic Dirac-Fock" approach; surprisingly, though, the data showed a slight preference for a version of that calculation which completely ignored the presence of the atomic hole created by the conversion process. Since simple physical considerations show that the typical time for an electron to leave the atom is less than $\sim 10^{-18}$ s, while the K-shell filling time is at least an order of magnitude longer (10^{-17} - 10^{-15} s depending on Z) [2], one should expect the presence of the hole to have a non-negligible impact on the wave function of the outgoing electron, at least in cases where the transition energy is just above the atomic-shell binding energy and thus the electron energy is low.

Even so, it was decided at the time to adopt the calculations that appeared to agree best with experiment even though they incorporated a "non-physical" assumption, and the most recent published ICC tables [3], as well as the Evaluated Nuclear Structure Data Files (ENSDF) maintained by the National Nuclear Data Center (NNDC) at Brookhaven, used the "no hole" approximation. Being readily available and pre-evaluated, ENSDF in particular is intensively used by scientists and technologists, usually without any further critical judgment. Consequently, it is clearly important that the validity of the ICC calculations used throughout ENSDF be firmly established since significant differences in calculated ICC coefficients can arise depending on whether the atomic hole is included or not. As was originally pointed out by Raman *et al.* [1], there are cases where differences of up to 10% can be expected.

Three years ago we reported a precise measurement of the K-shell conversion coefficient for the 80.2 keV, M4 transition in $^{193}Ir^m$ [4], a case originally suggested by Raman *et al.* [1] as providing the most sensitive test of the importance of the atomic hole. Our measurement, α_K=103.0(8), showed unequivocal agreement with the calculation that includes the "hole," α_K=103.5(1), and disagreement with the "no-hole" result, α_K=92.0(3). Based on our result, NNDC changed its policy and adopted the ICC values calculated with the atomic hole included; the consequent change in the ENSDF data files has had considerable impact on the nuclear-data users' community.

There remains a serious question. The survey by Raman *et al.* [1] included ICCs from 100 transitions with quoted uncertainties spanning more than an order of magnitude, from 0.5% to more than 6%. Although the authors found the average difference between experiment and the Relativistic Dirac-Fock theory to be ~1%, it is clear that their outcome was dominated by a few very precise measurements, some of which were old and of dubious merit. A few of these ostensibly precise measurements in fact disagreed significantly with both the "hole" and "no-hole" version of the theory. Can we thus rely on even the best calculated ICCs to ±1%?

To begin to answer this question, we have measured two cases that disagreed significantly with both types of calculations in the Raman *et al.* survey [1]: the 127.5-keV, E3 transition in $^{134}Cs^{m}$ and the 661.7-keV, M4 transition in ^{137}Ba. We used the same measurement technique as we had used previously in the $^{193}Ir^{m}$ experiment [4], where we determined the intensity ratio of conversion K X-rays to the γ-ray for a single transition and used the precisely known fluorescence yield [5] to obtain the α_K ICC value at <1% precision. The key to this type of measurement is our HPGe detector, the efficiency calibration of which is known to high precision [6,7]. However, the $^{134}Cs^{m}$ and ^{137}Ba decays produced K X-rays with energies below 50 keV, the lowest energy for which our detector efficiency is well known. As a result, we were forced initially to restrict our test to the ratio of the α_K values for the two transitions.

Since this experiment was described in our last progress report [8], we have solved the problem of scattered radiation, which strongly affects the 30-37-keV K x-ray peaks from cesium and barium. Actually in this energy range the (lower energy) scattered radiation is not entirely separated from the K X-rays peaks themselves. With our energy resolution of ~1 keV, the scattered continua extend only 3-4 keV below the x-ray energy and only a small fraction of it can be observed as a tail to the left side of the K_α-group peak, while it is totally hidden for the K_β group. The effect is strongly dependent on the source design and surrounding materials of the source-detector assembly. Fortunately our sources were very similar and most of the effects of scattering could be expected to cancel out in the ratio.

Nevertheless, we carefully studied the correction using the ^{137}Cs source, which was conveniently long lived and free of contaminants. We compared the observed low-energy tail of the K_α peak with Monte-Carlo calculations using the Cyltran code [9], the same one we used so successfully for our efficiency calibration. We also modeled the source configuration and included a cylinder of air, 1 meter in diameter and 1 meter long, in front of the detector face and surrounding the source, which was located at 151 mm from the face. The comparison of experiment with calculation is shown in the upper part of Fig. 1; the agreement is quite reasonable. From the Monte Carlo calculation we could then determine that the scattered continuum continues to increase as it approaches the central peak energy (where it is hidden by the finite width of the observed peak). The shape of the simulated spectrum, especially the increase of scattered photons as the central energy is approached, was qualitatively checked by a measurement of the same source with a 30-mm^2 Si(Li) detector, which has higher resolution than our HPGe detector. The result is shown in the lower part of Fig. 1, where the continuing increase in scattering is clearly evident. (Since we did not have a model for the Si(Li) detector, we did not attempt a Monte Carlo calculation for it.)

From the Monte Carlo calculation we determined the ratio of observed-to-hidden scattering tail for each source and used the results to calculate the total effect of scattering in each case based on the observed low-energy tail. The result was 2.1(3)% for ^{137}Ba, and 2.8(4)% for $^{134}Cs^{m}$ respectively, which gave a 0.8(3)% correction on the α_K ratio of the transitions studied here.

With this – and other – corrections in hand we determined the ratio of ICCs to be $\alpha_K(^{134}Cs^m)/\alpha_K(^{137}Ba)$ = 30.01(15). This result agrees well with 29.96, the Dirac-Fock ICC calculation in which the hole is accounted for in the "frozen orbital" approximation, and disagrees with the "no hole" calculation by more than three standard deviations. Our result also disagrees with the experimental ratio, 28.8(5), obtained from the data in Raman *et al.* [1], a result that was inconsistent with both calculations. The results of this study have been published [10].

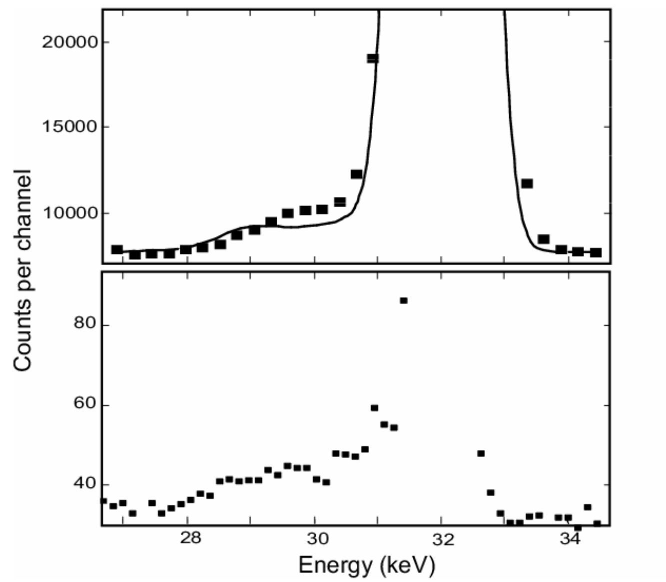

Figure 1. The top panel shows an expanded region of the HPGe spectrum at the base of the barium K_α x-ray peak. The solid squares are the measured data from the ^{137}Cs source; the curve is the result of a Monte Carlo simulation. The bottom panel gives the same region of the ^{137}Cs spectrum as measured with a Si(Li) detector; the ordinate scale has been adjusted so that both panels display approximately the same fraction of the total K x-ray peak area.

The ^{139}La experiment, which was also completed during the year reported here, was undertaken as a means of turning the $^{134}Cs^m$-^{137}Ba α_K-ratio measurement into independent determinations of $\alpha_K(^{134}Cs^m)$ and $\alpha_K(^{137}Ba)$. The calculated α_K value for the 165.9-keV M1 transition in ^{139}La is essentially independent of whether the "hole" or "no hole" assumption is used: the results differ by less than 0.2%. Thus we can use this reliable result, together with the measured ratio of K x rays to 165.9-keV γ rays, to determine our detector's efficiency at the K x-ray energy. Since the K x rays of all three elements, Cs, Ba and La, are separated by only 1.2-1.3 keV of one another, this same calibration, with minor adjustments can be applied, to the earlier measurement of Cs and Ba. This measurement has the added benefit that it extends the range of our HPGe detector's precision efficiency-calibration from 50 keV, its previous limit, down to ~20 keV.

One of the main difficulties of this measurement came from the short-lived ($t_{1/2}$ = 83 m) ^{139}Ba precursor of ^{139}La. Its short half-life precluded our processing the source after neutron activation so we were forced to activate an enriched ^{138}Ba sample that was already placed between thin mylar foils. Much care had to be taken to minimize impurities that could cause competing radioactivities. We prepared two sources of ^{138}Ba from thin uniform layers, ~1 μm thick, of barium nitrate placed on 12-μm-thick mylar, and covered by 4-μm-thick mylar. In order to reduce the contamination coming from impurities in mylar, the diameter of the whole source assembly was reduced to only 12 mm. These sources were almost absorption free: the correction, including both the source material and the covering mylar, was <0.2%.

The sources were activated for 1.8 h and 1 h respectively, at a flux of $\sim 7\times 10^{12}$ n/cm^{2}s at the Nuclear Science Center Triga reactor at Texas A&M. Even though our enriched isotope was claimed to be pure, we found $^{152}Eu^m$ ($t_{1/2}$ = 9.3 h) and ^{153}Sm ($t_{1/2}$ = 46.5 h), both of which have huge activation cross sections, 3300 b for ^{151}Eu, and 206 b for ^{152}Sm, compared to 0.36 b for ^{138}Ba. These activities affected lanthanum's K_β peaks at the level of several percent, depending on the time after activation. For this reason, we collected 8-9 spectra from each source over several days, long after ^{139}La had decayed away, and used the measured decay curves to extract the contribution of both contaminants. The analysis is in progress.

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