Further Tests of Internal-Conversion Theory with Precise γ- and x-Ray Spectroscopy: the Decays of $^{134}Cs^{m}$, ^{137}Ba

N. Nica, J.C. Hardy, V.E. Iacob, H.I. Park, J. Goodwin, W.E. Rockwell, and M.B. Trzhaskovskaya[1]
[1]*Petersburg Nuclear Physics Institute,Gatchina 188300, Russia*

Internal conversion plays an important role in the assignment of spins, parities and radiation intensities, as well as in the building of level and decay schemes. After more than fifty years of theory and experiment the overall agreement between calculated internal-conversion coefficients (ICC's) and measured ones has now reached ~1%. This rather optimistic conclusion emerged from 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 5% 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 ~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.

Two 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.

At Texas A&M we have continued to make precise ICC measurements with the goal of further testing and possibly refining the ICC calculations. We have begun by re-examining those cases in the Raman *et al.* survey [1] that disagree significantly with both types of calculations. We seek to determine

whether these cases signal further problems with the theory or are simply experimental aberrations. We report here the re-measurement of two such transitions, in $^{134}Cs^{m}$ and ^{137}Ba.

Our method is to determine the K-shell conversion coefficient, α_K, for a single transition by measuring the peak areas of its K x-rays and γ ray (N_K, N_γ) as determined in a single well-calibrated HPGe detector. We obtain α_K from the formula $\alpha_K\omega_K = N_K/N_\gamma \times \varepsilon_\gamma/\varepsilon_K$, where ε_K and ε_γ are the detector efficiencies and ω_K is the fluorescence yield. We take ω_K from reference [5], which presents a global fit to experimental data and quotes a precision of better than 0.5% for the ω_K values tabulated for Cs and Ba. (We have previously verified experimentally the table's ω_K value for iridium [6].) Our biggest asset in these measurements is the very well known efficiency of our HPGe detector. It has been determined to a precision of 0.15% (relative) and 0.2% (absolute) for energies between 50 and 1400 keV [7,8].

The K x-ray energies for Cs and Ba lie in the 30-35 keV range, however, where our efficiency is not so precisely known since any photon groups available for calibration below 50 keV are themselves x-rays whose quoted intensities depend on ICC calculations. For this reason, in the present test we focus on the ratio of the α_K's for the two transitions studied. Since the K x-rays for Cs and Ba are within 1.3 keV in energy, the detector efficiencies, ε_K, for the two transitions virtually cancel out in the ratio, removing the effects of any imprecision in their individual values.

Our method is only applicable to level schemes in which a single transition converts in the K shell, a demand well satisfied in this study. The ^{137}Cs β^- decay is followed by a single 661.7-keV transition in the ^{137}Ba daughter, and the $^{134}Cs^{m}$ isomer decays predominantly (99%) by a 128-keV transition, which is followed by an 11-keV transition that is well below the K-shell binding energy.

In the case of 30-year ^{137}Cs we took advantage of a 100% pure and attenuation-free (virtually coverless) source purchased from Isotopes Products Laboratory. The $^{134}Cs^{m}$ case was more challenging since it is rather short lived (2.9 h) and we had to prepare our own source. We used 99.999% pure CsCl and $CsNO_3$ (other chemicals were also tested and rejected) and deposited 0.10-0.15-μm uniform layers on thin mylar. Calculations were done to determine the optimum thickness and to study the influence of thickness variations. Because the chemicals are hygroscopic, we used vacuum evaporation and manipulation in a dry nitrogen atmosphere. We then checked the layers with a microscope. The prepared sources were irradiated in the TRIGA reactor at the Texas A&M Nuclear Science Center by a thermal neutron flux of $\sim 7\times10^{12}$ n/cm^{2}s. The sources were sealed with thin kapton tape after activation.

Spectra were recorded from these $^{134}Cs^{m}$ sources, as well as from the long-lived ^{137}Cs source. The $^{134}Cs^{m}$ sources were also studied long after their original irradiation in order to look for impurities. In addition, spectra from ^{109}Cd, ^{133}Cs, ^{137}Ba, and ^{241}Am sources were taken to help us characterize backscattering and other effects. In all, about 80 spectra were acquired in a total of about 1000 h. The best three spectra of $^{134}Cs^{m}$, and two of ^{137}Cs were used to extract the ratio of K x-rays to γ-ray areas.

The effort and care invested in preparation of the $^{134}Cs^{m}$ source paid dividends. We identified only very weak impurities (0.3-0.9%) that affected the Cs K x-rays; these were easily corrected for. Only small corrections were needed as well for source attenuation (0.1%), K x-rays from the weak 139-keV cross-over transition (0.8%), and random summing (< 0.5%). The correction for the Voigt-shape of the x-ray peaks was done by simulation and found to be small (0.1%) and equal for both Cs and Ba x-rays, thus canceling out in ratio. Our biggest problem turned out to be x-ray backscattering, which led to "tails" on

the low-energy side of the x-ray peaks. After careful study, we applied a 1.1% correction for this effect to the ratio of Cs to Ba K x-rays.

Our result for the ratio $\alpha_K(^{134}Cs)/\ \alpha_K(^{137}Ba)$ is shown in the table. It agrees well with the Relativistic Dirac-Fock theory (with or without the inclusion of the atomic hole) and disagrees with the previous experimental result quoted by Raman *et al.*, which disagreed with all calculations. We can conclude that the apparent conflict between theory and experiment in these cases was not due to a failure of the theory but rather was caused by experimental inadequacies.

Table I. Our result for the ratio $\alpha_K(^{134}Cs)/\ \alpha_K(^{137}Ba)$ compared with several theoretical calculations and with previous experiments as surveyed in Ref. [1].

	α_K ratio	Uncertainty
This experiment	**30.02**	**0.27**
hole(frozen orbital)	29.96	
hole(SCF)	29.88	
no hole	29.52	
Experiment (*Raman et al.*[1])	**28.82**	**0.51**

[1] S. Raman, C.W. Nestor, Jr., A Ichihara, and M.B. Trzhaskovskaya, Phys. Rev. **C66**, 044312 (2002)

[2] J.C. Hardy, N. Nica, V.E. Iacob, M.B. Trzhaskovskaya and R.G. Helmer, Appl. Radiat. Isot. (To be published).

[3] I.M. Band, M.B. Trzhaskovskaya, C.W. Nestor, Jr., P. Tikkanen, and S. Raman, At. Data Nucl. Data Tables **81**, 1 (2002)

[4] N. Nica, J.C. Hardy, V.E. Iacob, S. Raman, C.W. Nestor Jr., and M.B. Trzhaskovskaya, Phys. Rev. C**70**, 054305 (2004)

[5] E. Schönfeld and H. Janssen, Nucl. Instrum. Methods Phys. Res. **A369**, 527 (1996)

[6] N. Nica, J.C. Hardy, V.E. Iacob, J.R. Montague, and M.B. Trzhaskovskaya, Phys. Rev. C **71**, 054320 (2005).

[7] J.C. Hardy, V.E. Iacob, M. Sanchez-Vega, R.T. Effinger, P. Lipnik, V.E. Mayes, D.K. Willis, and R.G. Helmer, Appl. Radiat. Isot. **56**, 65 (2002).

[8] R. G. Helmer, J. C. Hardy, V. E. Iacob, M. Sanchez-Vega, R.G. Neilson, and J. Nelson, Nucl. Instrum. Methods Phys. Res. **A511**, 360 (2003).