Test of internal-conversion theory with a measurement in ¹¹¹Cd

TEXAS A&M PROGRAM TO MEASURE ICC N. NICA

Internal Conversion Coefficients (ICC):

- Big impact on quality of nuclear science
- Central for NSDD-USNDP and other nuclear data programs
- Intensely studied by theory and experiment
- Important result for nuclear data communities:

The calculations including the atomic vacancy are now standard!

Internal Conversion:

- Basic science:
 - Construction of nuclear level schemes
 - Total transition probabilities
 - Assignment of spins and parities
- Applications:
 - Medical
 - Pharmaceutical
 - Environmental
- Hundreds of measured values: 5-10% precision
- 2002Ra45 study:
 - Systematic discrepancy: theory up to 2-3% larger than experiment (for high multipolatities)
 - No benchmark experimental ICC values!

2002RA45 survey ICC's theories and measurements

- Theory: RHFS and RDF (1989Ba84) comparison
 Exchange interaction, Finite size of nucleus, *Hole treatment*
- Experiment:

```
100 E2, M3, E3, M4, E5 ICC values, 0.5%-6% precision, very few <1% precision!
```

• Conclusions, Δ (exp:theory)%:

No hole: +0.19(26)% BEST!

(bound and continuum states - SCF of neutral atom)

Hole-SCF: -0.94(24)%

(continuum - SCF of ion + hole (full relaxation of ion orbitals))

Hole-FO: -1.18(24)%

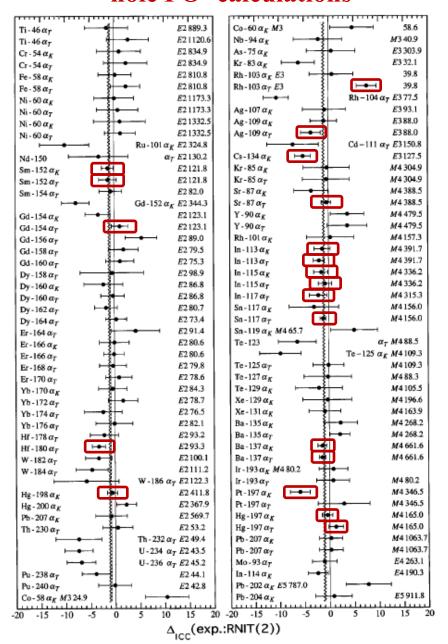
(continuum - ion field from bound wave functions of neutral atom

(commuum - ton field from bound wave functions of neutral atom (no relaxation of ion orbitals))

PHYSICAL ARGUMENT

K-shell filling time vs. time to leave atom $\sim 10^{-15} - 10^{-17} \text{ s} \gg \sim 10^{-18} \text{ s}$

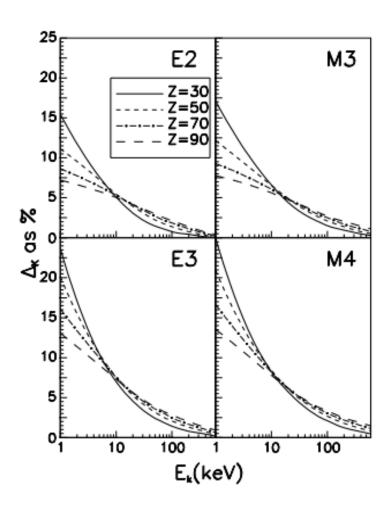
2002Ra45: 100 $\alpha_K(exp)$ cases compared with 'hole FO' calculations



2003 we started a program at Texas A&M University to measure benchmark ICC values, with the goal to provide a more precise and reliable data set, one particularly geared to addressing the atomic-vacancy issue.

- 1. Re-measure 'main set' of 100 cases
 - 2. Measure other cases
 - 3. Abnormal results
 - 4. High uncertainties
 - 5. Theory: $\Delta_K = 4-10\%$
- 6. Experiment: about 1-2% precision
 - 7. Well spread on nuclear chart

The difference Δ_K between α_K ('hole') and α_K ('no hole') (relative to α_K ('hole')) as function of kinetic energy of converted electron E_K



KX to γ rays ratio method

- Single-transition level scheme (or dominated by a strong transition)
- Sources for n_{th} activation
 - Small selfabsorption (< 0.1%)
 - **■ Dead time** (< 5%)
 - Statistics (> 10^6 for γ or x-rays)
 - High spectrum purity
 - Minimize activation time (0.5 h)
- Impurity analysis essentially based on ENSDF
 - Trace and correct impurity to 0.01% level
 - Use decay-curve analysis
 - Especially important for the K X-ray region
- Voigt-shape (Lorentzian) correction for X-rays
 - Done by simulation spectra, analyzed as the real spectra
- Coincidence summing correction (including angular correlation)

^{111m}Cd 150.8 keV, E3 transition

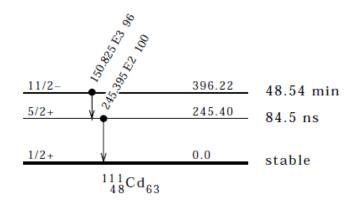
Suggested by 1984Ba84

- $\alpha(K)\exp = 1.29 \ 11 \ (1987Ne05), \%unc=8.5$
- $\alpha(K)_{hole_FO} = 1.450$, $\alpha(K)_{no_hole} = 1.425$, $\Delta_K = 1.7\%$

¹¹¹Cd IT Decay (48.54 min)

Decay Scheme

Intensities: $I(\gamma + ce)$ per 100 decays by this branch %IT = 100



- Texas A&M precision ICC measurements by KX to γ rays ratio method:
- α_{K150} measurement:

$$\alpha_{K150} = \frac{N_K}{N_{\gamma 150}} \cdot \frac{\mathcal{E}_{\gamma 150}}{\mathcal{E}_{K236}} \cdot \frac{1}{\omega_K} - \alpha_{K245} \cdot \frac{N_{\gamma 245}}{N_{\gamma 150}} \cdot \frac{\mathcal{E}_{\gamma 150}}{\mathcal{E}_{\gamma 245}}$$

• Extended for α_{T150} measurement:

$$(1+\alpha_{T150})\cdot\frac{N_{\gamma 150}}{\varepsilon_{\gamma 150}} = (1+\alpha_{T245})\cdot\frac{N_{\gamma 245}}{\varepsilon_{\gamma 245}}$$

- $\circ N_K$, N_γ measured from only one K-shell converted transition
- $\circ \omega_K = 0.842(4)$ from 1999SCZX (compilation and fit)
- o from $\alpha_{K245} = 0.05326(17)$, $\alpha_{T245} = 0.06351(17)$ (http://bricc.anu.edu.au, **2008KI07**)

Detection efficiency

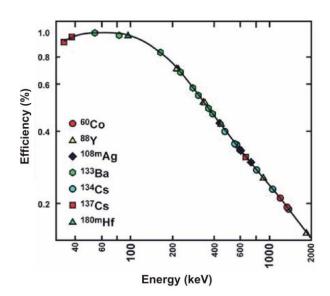
- Very precise detection efficiency for ORTEC γ-X 280-cm³
 coaxial HPGe at standard distance of 151 mm:
 - **0.2%**, 50-1400 keV (2002HA61, 2003HE28)
 - 0.4%, 1.4-3.5 MeV (2004HE34)
 - 1% , 10-50 keV (KX rays domain)

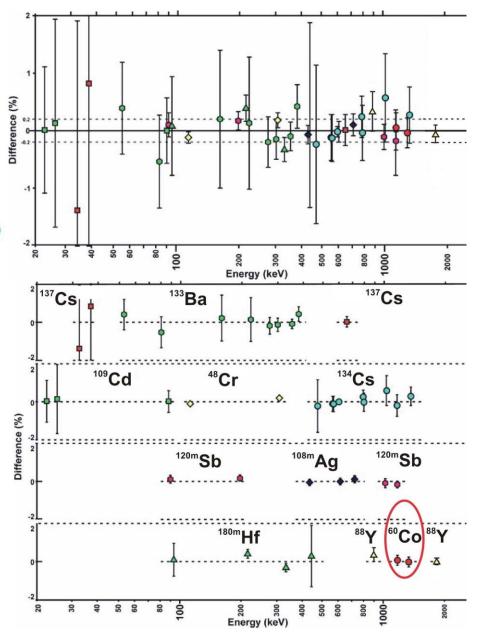
DETECTOR EFFICIENCY 50 keV < E $_{\gamma}$ < 1.4 MeV

Coaxial 280-cc n-type Ge detector:

- Measured absolute efficiency (⁶⁰Co source from PTB with activity known to + 0.1%)
- Measured relative efficiency (9 sources)
- Calculated efficiencies with Monte Carlo (Integrated Tiger Series CYLTRAN code)

0.2% uncertainty for the interval 50-1400 keV





Effect of scattering on the photo-peak efficiency (2007NI04)

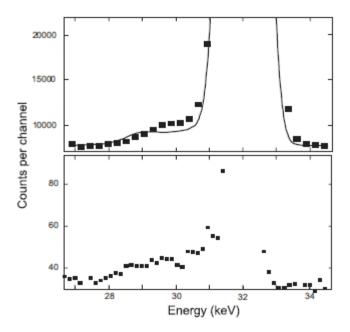
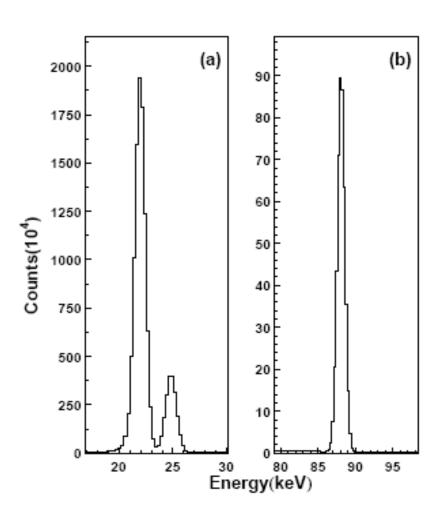


FIG. 5. The top panel shows an expanded region of the HPGe spectrum at the base of the barium K_{α} x-ray peak. The solid squares are the measured data from the 137 Cs source; the curve is the result of a Monte Carlo simulation. The bottom panel gives the same region of the 137 Cs spectrum as measured 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.

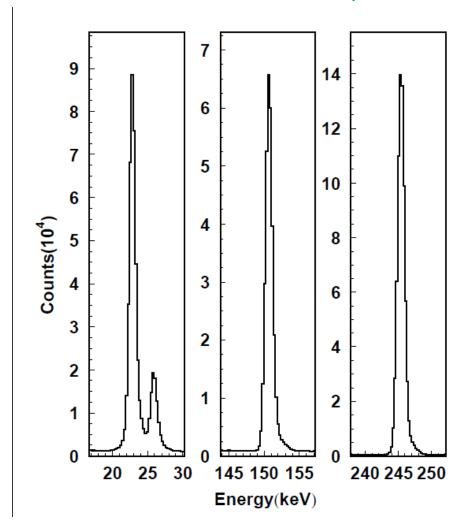
¹⁰⁹Cd Efficiency Calibration

22.6-keV AgKx & 88.0-keV E3 γ regions



111mCd Regions of interest

23.6-keV CdKx & 150.8-keV γ & 245.4-keV γ



Adopted efficiency:

$$\left(\frac{\mathcal{E}_{\gamma 150}}{\mathcal{E}_{K236}}\right)_{^{11\,\text{in}}Cd} = \left(\frac{\mathcal{E}_{\gamma 880}}{\mathcal{E}_{K226}}\right)_{^{109}\!\!A\,g,\text{exp}} \times \left(\frac{\mathcal{E}_{\gamma 150}}{\mathcal{E}_{\gamma 880}}\right)_{Cyltran} \times \left(\frac{\mathcal{E}_{K226}}{\mathcal{E}_{K236}}\right)_{Cyltran}$$

$$\frac{\mathcal{E}_{\gamma 150}}{\mathcal{E}_{K236}} = 0.917(7) \qquad \frac{\mathcal{E}_{\gamma 150}}{\mathcal{E}_{\gamma 245}} = 1.312314$$

^{111m}Cd 150.8 keV, E3 transition - ICCs measurement

- ¹¹⁰Cd 95.88% enriched (from 12.5% natural abundance), thin metal powder from *Trace Sciences International*.
- Impurities: ¹⁰⁶Cd (<0.01%), ¹⁰⁸Cd (<0.02%), ¹¹⁴Cd (0.84%), ¹¹⁶Cd (<0.16%).
- Backing: 10 µm Al foil (99.999%) from GoodFellow.
- Samples prepared by electro deposition by Dr. C. Folden III and Drd. T. Werke
- Samples: CdO, Φ=1.7 cm on 10 μm Al backing
 - S1: 0.99(2) mg, 0.53(1) µm
 - S2: 1.08(2) mg, 0.58(1) μm
- Neutron activation at Triga reactor @ TAMU, on both S1 and S2:
 - $\Phi = 7.5 \times 10^{12} \text{ n/(cm}^2\text{s})$
 - $\alpha_{th} = 0.14(5) b$
 - Samples activated 2 h (2 months apart)
 - Measured for 3 weeks

Impurity analysis

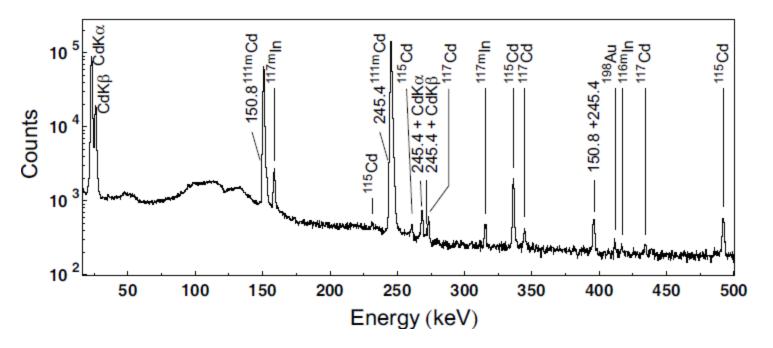


Table 1. The contributions of identified impurities that affect the cadmium *K* x-ray peaks.

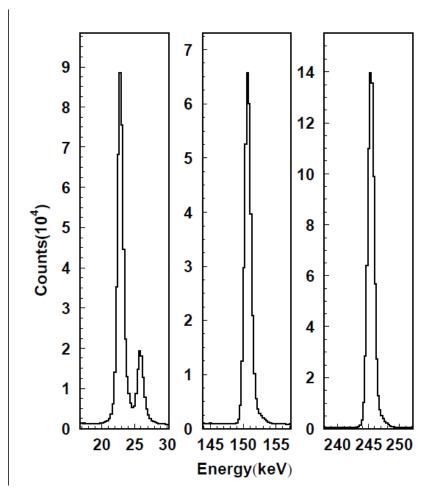
Source	Contaminant	Contribution to spectrum (%)		
		S 1	S2	
¹¹⁵ Cd	In K x rays	1.81(8)	2.71(6)	
¹¹⁷ Cd	In K x rays	0.113(12)	0.125(9)	
117 In	Sn K x rays	0.18(3)	0.21(3)	
117m In	In+Sn K x rays	0.603(9)	0.484(5)	
116m In	In K x rays	0.010(2)	0.006(1)	

Data analysis

Table 2. The total number of counts (or areas of the peaks) for 111m Cd K x rays and the 150.8- and 245.4-keV γ rays, followed by corrections and the corrected area-ratios information required to extract the value of α_{K150} .

Quantity	Value			
	S 1	S2		
$\operatorname{Cd}(K_{\alpha}+K_{\beta})$ x rays				
Total counts	$1.979(6) \times 10^{5}$	$4.695(9) \times 10^5$		
Impurities	$-5.39(14)\times10^3$	$-1.66(3)\times10^4$		
Lorentzian correction	+0.12(2)%	+0.12(2)%		
Summing correction	+0.99(6)%	+0.99(6)%		
Attenuation correction	+0.27(2)%	+0.29(2)%		
Corrected counts, N_K	$1.952(6) \times 10^5$	$4.593(10)\times10^5$		
¹¹¹ Cd 150.8-keV γ ray				
Total counts	$1.303(11)\times10^{5}$	$3.064(25)\times10^5$		
Summing correction	+1.29(6)%	+1.29(6)%		
Corrected counts, $N_{\gamma 150}$	$1.320(12)\times10^5$	$3.104(25)\times10^5$		
¹¹¹ Cd 245.4-keV γ ray				
Total counts	$3.024(22)\times10^5$	$7.082(45)\times10^5$		
Summing correction	+0.86(3)%	+0.86(3)%		
Corrected counts, $N_{\gamma 245}$	$3.050(22)\times10^5$	$7.143(45) \times 10^5$		
$N_K/N_{\gamma 150}$	1.479(14)	1.480(12)		
$N_{\gamma 245}/N_{\gamma 150}$	2.311(27)	2.301(24)		

$$\frac{N_K}{N_{\gamma 150}} = 1.479(10) \qquad \frac{N_{\gamma 245}}{N_{\gamma 150}} = 2.395(7)$$



Results

Model	α_K	$\Delta(\%)$	$lpha_T$	$\Delta(\%)$
Experiment	1.449(18)		2.217(26)	
Theory:				
No vacancy	1.425(1)	+1.7(12)	2.257(1)	-1.8(12)
Vacancy, FO	1.451(1)	-0.1(12)	2.284(1)	-2.9(12)

α_T discrepancy:

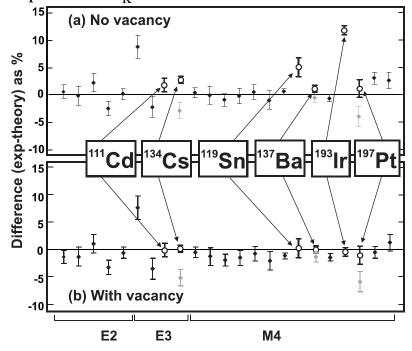
- \triangleright 150.8-keV γ is hindered by 10⁴ relative to its Weiskopf estimate.
- ➤ "Penetration" effects associated with the change from point-like to realistic finite-sized nucleus for transition electromagnetic potentials.
- ➤ ICCs can thus become dependent on nuclear structure details and nuclear transition dynamics.
- ➤ Under particular conditions it is possible for the penetration to be "hidden" by a cancellation that can affect one atomic shell but not the others (e.g. 1974KR01, for M1 transitions).
- \triangleright Hypothetically this could also explain why in our case α_K looks to be unaffected by penetration effects, while α_T is significantly reduced.
- Pespite the lack of direct support, penetration remains the best explanation for the discrepancy we observe here between experiment and theory for α_{T} .

Current status of precision ICC measurements

Table 3. Comparison of the six measured α_K values with Dirac-Fock theoretical calculations

Parent	Transition	Measured α_K	Calculate	Calculated α_K	
	Energy(keV)	N	o vacancy	Vacancy	
197m Pt	346.5(2)	4.23(7)	4.191	4.276	
193m Ir	80.22(2)	103.0(8)	92.0	103.3	
137m Ba	661.659(3)	0.0915(5)	0.09068	0.09148	
^{134m}Cs	127.502(3)	2.742(15)	2.677	2.741	
119m Sn	65.66(1)	1610(27)	1544	1618	
111m Cd	150.853(15)	1.449(18)	1.425	1.451	
χ^2			219	0.68	

Percentage difference between measured and calculated ICCs for the two Dirac-Fock calculations. The measured values include our published α_K values



References

- [5] J. C. Hardy, N. Nica, V. E. Iacob, S. Miller, M. Maguire and M. B. Trzhaskovskaya Appl. Rad and Isot. 87, 87 (2014).
- [6] N. Nica, J. C. Hardy, V. E. Iacob, S. Raman, C. W. Nestor Jr., and M. B. Trzhaskovskaya, Phys. Rev. C 70, 05430 (2004).
- [7] N. Nica, J. C. Hardy, V. E. Iacob, J. R. Montague, and M. B. Trzhaskovskaya, Phys. Rev. C 71, 054320 (2005).
- [8] N. Nica, J. C. Hardy, V. E. Iacob, W. E. Rockwell, and M. B. Trzhaskovskaya, Phys. Rev. C 75, 024308 (2007).
- [9] N. Nica, J. C. Hardy, V. E. Iacob, C. Balonek, and M.B. Trzhaskovskaya, Phys. Rev. C 77, 034306 (2008).
- [10] J.C. Hardy, N. Nica, V.E. Iacob, C. Balonek and M.B.Trzhaskovskaya, Appl. Rad. Isot. 66, 701 (2008).
- [11] N. Nica, J. C. Hardy, V. E. Iacob, J. Goodwin, C. Balonek, M. Hernberg, J. Nolan and M. B.Trzhaskovskaya, Phys. Rev. C 80, 064314 (2009).
- [12] N. Nica, J. C. Hardy, V. E. Iacob, M. Bencomo, V. Horvat, H.I. Park, M. Maguire, S. Miller and M. B.Trzhaskovskaya, Phys. Rev. C 89, 014303 (2014).
- [13] N. Nica, J. C. Hardy, V. E. Iacob, T.A. Werke, C.M.Folden, L.Pineda, and M.B.Trzhaskovskaya Phys.Rev. C 93, 034305 (2016).