Tests of internal-conversion theory with precise $\gamma$- and x-ray spectroscopy:
The case of $^{111m}$Cd

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Internal conversion is an important component of most nuclear decay schemes. In order to balance decay schemes correctly, one needs to know the internal conversion contribution to each transition as expressed by its internal conversion coefficient (ICC). Nevertheless, ICCs are only rarely measured; instead they are taken from tabulations. As a result, calculated ICCs are essential input to every decay scheme, except those for the lightest nuclei. Unfortunately, over the decades, tabulated ICC values have differed significantly from one calculation to another by a few percent. Although for many applications such differences can be tolerated, transitions used in critical calibrations require very precise and accurate ICC values, precision that has simply been impossible to guarantee at the one-percent level or below.

In order to correct for this deficiency one can only seek guidance from measured ICCs that have sufficient precision to distinguish among the various calculations. However, as recently as a decade ago, when a survey of measured ICCs was made by Raman et al. [1], there were only five published ICC values with precision of the order of ±1%, not enough to make any definitive conclusion possible. At that time, one aspect of the ICC calculations remained a particular concern. The final-state electron wave function must be calculated in a field that adequately represents the remaining atom. But should that representation include the atomic vacancy created by the conversion process? Some calculations included it and some did not.

Thus the problem of measuring ICCs at the ±1% precision level became critical and, with our very precisely efficiency-calibrated HPGe detector [2], we found ourselves in a position to be able to address it. Consequently, over the past decade we have been measuring a series of ICCs [3] covering a wide range of atomic numbers, $50 \leq Z \leq 78$. So far, all these results have indicated that the atomic vacancy should be taken into account in the calculations. With $Z = 48$, the new case reported here – the 150.8-keV $E3$ transition in $^{111}$Cd – extends the lower end of that range.

The total intensity of an electromagnetic transition is split between $\gamma$-ray emission and electron conversion, which can take place in several atomic shells and subshells, and is followed by the corresponding x rays. If only $K$-shell conversion is considered, then one can use the following formula to determine the $K$-shell conversion coefficient, $\alpha_K$:

$$\alpha_K \omega_K = \frac{N_K}{N_\gamma} \cdot \frac{\varepsilon_\gamma}{\varepsilon_K}$$  \hspace{1cm} (1)

where $\omega_K$ is the fluorescence yield, which we take from Ref. [4]; $N_K$ and $N_\gamma$ are the respective peak areas of the $K$ x rays and the $\gamma$ ray; and $\varepsilon_K$ and $\varepsilon_\gamma$ are the corresponding detector absolute efficiencies.

The 150.8-keV, $E3$ transition in $^{111}$Cd depopulates the 396.3-keV, 49-min isomeric state and is followed by a 245.4-keV $E2$ transition to the stable ground state. Naturally the Cd $K$ x-ray group is the
sum of $K$ x rays following the electron conversion of both transitions. However, the $K$ x-ray contribution from the 245.4-keV transition is relatively small and can be reliably subtracted along with even smaller contributions from other Cd isotopes to yield a clean $N_K$ number for Eq. (1).

Two samples were prepared by electro-deposition of 1 mg of 96%-enriched $^{110}$Cd oxide on 10-µm-thick Al foil, 99.999% pure. The CdO layer was 1.7-cm in diameter and about 0.55 µm thick. The 396.3-keV isomeric state in $^{111}$Cd was populated by thermal neutron activation at the Nuclear Science Center TRIGA reactor of Texas A&M University at a fluency of $7.5 \times 10^{12}$ neutron/(cm$^2$s). The samples were activated at different dates for about 2 h, cooled down for 2 h, and measured for 3 weeks. A total of four spectra were acquired, from which $\alpha K(150.8)$ was extracted, the final result being the average of the four values. Since the 150.8-keV transition is in cascade with the $E2$ 245.4-keV transition, as a byproduct we could also obtain the total conversion coefficient $\alpha$ of the former transition by measuring the intensity of the latter since the calculated total ICC value of the $E2$ transition is insensitive to the treatment of the vacancy.

As described in Ref. [2], $\varepsilon_\gamma$ for a 150.3-keV $\gamma$ ray in our detector is known to $\pm 0.15\%$ relative precision; however a special investigation was required in order to get a precise value for $\varepsilon_K$ at 23.6 keV, the average energy of the cadmium $K$ x ray group. We accomplished this by measuring a well-known calibration source, $^{109}$Cd, which decays by electron capture to $^{109}$Ag, followed by a unique 88.0-keV, $M4$ $\gamma$ transition. The $K$ x rays of silver, following both the electron capture decay and the electron conversion of the 88.0-keV transition, form a prominent $K$ x-ray group situated at 22.1-keV ($K_\alpha$) and 25.0-keV ($K_\beta$). The $K$ x-rays, together with the 88.0 keV $\gamma$ ray, were then used with a formula similar to eq. (1) to deduce $\varepsilon_K$ at the silver $K$ x-ray energies based on the calculated $\alpha_K$ value for the 88.0-keV transition in $^{109}$Ag. This $\alpha_K$ value is relatively insensitive to the treatment of the atomic vacancy so we could use the mean value of the calculated 'hole' and 'no hole' $\alpha_K$ values with an uncertainty chosen to encompass both. Then by a short interpolation from the silver $K$ x-ray energies to those of cadmium, we determined the $\varepsilon_K$ value for cadmium $K$ x-rays with a total uncertainty of the order of $\pm 1\%$.

From our analysis, we found that the contribution to the cadmium $K$ x rays coming from the electron conversion of the 245.4-keV transition in $^{111}$Cd was 11.2% of the contribution from the 150.8-keV transition. In addition, transitions in $^{115}$Cd, $^{117}$Cd, $^{117}$In and $^{116}$In were identified as contributing to the $K$ x-ray group, with a total contribution of 3 – 7 % depending on the time after activation.

Our preliminary result, $\alpha_K(150.8) = 1.454(20)$, is in best agreement with the theoretical calculation incorporating the vacancy, $\alpha_K(\text{hole}) = 1.450$, which is in conformity with all our previous results [3]. For comparison, the 'no hole' result is $\alpha_K(\text{no hole}) = 1.425$. For the total conversion coefficient of the same transition, our preliminary result is $\alpha(150.8) = 2.241(30)$, which can be compared with $\alpha(\text{no hole}) = 2.26$ and $\alpha(\text{hole}) = 2.28$. Both our results disagree with less-precise previous measurements [5], $\alpha_K(150.8) = 1.29(11)$ and $\alpha(150.8) = 1.98(5)$, which were significantly lower than both types of calculations.