Tests of internal-conversion theory with precise γ- and x-ray spectroscopy: the case of $^{119}Sn^{m}$

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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 ten years 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 [3,4] covering a wide range of atomic numbers. So far, all these results have indicated that the atomic vacancy should be taken into account in the calculations. The new case reported here, a 65.7-keV M4 transition in ^{119}Sn, extends our measurements to lower Z than any case we have yet studied.

For an isolated electromagnetic transition that converts in the atomic *K* shell, the observation of a *K* x ray is a signal that an electron conversion has taken place; whereas a γ ray indicates that no conversion has taken place. If both x rays and γ rays are recorded in a measurement, then the value of α_K is given by

$$\alpha_K \omega_K = \frac{N_K}{N_\gamma} \cdot \frac{\varepsilon_\gamma}{\varepsilon_K} \quad , \qquad (1)$$

where ω_K is the fluorescence yield that we take from Ref. [5]; N_K and N_γ are the respective peak areas of the K x rays and the γ ray; and ε_K and ε_γ are the corresponding detector absolute efficiencies. As described

in Ref. [2] ε_γ for a 65.7-keV γ ray in our detector is known to ±0.15% relative precision; however a special investigation was required in order to get a precise value for ε_K at the ~25-keV energy of the Sn K x rays.

As described in our previous progress report [6], two impurities in the source affected the 65.7-keV γ ray: ^{182}Ta and ^{75}Se both produced γ rays that contributed about 68% of a broad group that appeared at 66 keV. Since ^{118}Sn has a very low thermal-neutron capture cross section of about 10 mb for the production of ^{119}Sn^m, and the 65.7-keV transition from that isomer has a total ICC of about 5000, the emission of 65.7-keV γ rays is so much hindered that even very weak impurities with high activation cross sections can compete strongly if, unfortunately, they emit γ rays at nearly the same energy. Conveniently though the halflives of ^{182}Ta (120 d) and ^{75}Se (115 d) are significantly shorter than that of ^{119}Sn^m (293 d). We simply re-measured the source 19 months after it had first been activated, after which time the two contaminant activities had decreased by a factor of 7 or 8 compared to ^{119}Sn^m. In that spectrum the summed contribution of ^{75}Se and ^{182}Ta to the 66-keV group was only 7.6%, an amount easily corrected for. The already small contribution to the K x-rays from other tin isotopes was also reduced, to about 1%. Finally, since almost all other impurities were also shorter lived than ^{119}Sn^m, there were no intense γ rays left in the spectrum at energies above 65.7 keV, so the Compton background was reduced drastically at 66 keV, which consequently greatly improved the peak-to-background ratio for the peak whose intensity we needed to determine.

Some of the photons from a radioactive source scatter from nearby materials – including air – in the vicinity of the detector setup and, entering the detector, they form a continuum in the energy spectrum extending to lower energy from the peak created by the unscattered photons. For photons above ~50 keV this continuum is rather flat and extends to energies well below the corresponding peak so, by extrapolation, its contribution to the area of the peak can easily be determined and removed. However, for peaks with energies as low as 25.2 keV and 28.6 keV, the energies of the tin K_α and K_β x rays respectively, the continuum is more like a shelf that extends only 2-3 keV below the peak energy. At our energy resolution of ~1 keV in this region, an important part of the continuum gets "hidden" in, and potentially counted together with, the peak itself. The number of counts in the "hidden" continuum is very dependent not only on the source-detector geometry, but also on the details of its neighborhood. For this reason it is impossible to define a universal efficiency calibration with useful precision below ~50 keV. Rather, one must examine each geometry as a special case, which must be calibrated based on its specific properties.

We followed two different approaches to this part of the analysis. In the first, described fully in our paper on ^{134}Cs and ^{137}Ba [4], we employed Monte Carlo calculations with the CYLTRAN code – the same code used in our calibration procedures [2] – to simulate the scattering "shelf"; then we scaled up the result to match the small component of the shelf visible in the data; and finally used that scaled-up result to determine the component of the shelf contained within the peak.

Our second approach was to measure a calibration source, ^{109}Cd, which decays by electron capture to ^{109}Ag followed by the emission of a unique 88.0-keV M4 γ transition. The K x rays of silver, following both the electron capture decay and the electron conversion of the 88.0-keV transition, form prominent x-ray groups situated at 22.1 keV (K_α) and 25.0 keV (K_β). The K x-rays together with the 88.0-

keV γ ray can be used with a formula similar to eq. (1) to deduce ε_K at silver K x-rays energies if we use calculated α_K values for 88.0-keV transition in ^{109}Ag. By taking the mean value of calculated 'hole' and 'no hole' α_K values with an uncertainty encompassing both, we obtain an α_K value that is independent of the treatment of the atomic vacancy. Then by short interpolation from silver K x-ray energies to tin K x-ray energies, we can arrive at ε_K values for tin with a total uncertainty of about ±1%. This result and the CYLTRAN-based value are in good agreement (within 0.3%), so we can confidently use the combined result for ε_K in Eq. (1) to extract the α_K value for the 65.7-keV γ transition from ^{119}Sn^m.

Our still preliminary value, α_K = 1610(27), is in good agreement with the 'hole' calculation, which predict 1618 in the "frozen orbital" approximation, and in disagreement with the 'no hole' calculation, which predicts 1544. The current status of our series of measurements is given in Fig. 1, where it can be seen that our new result confirms and strengthens our conclusion that the atomic vacancy created by the internal conversion process must be taken into account when calculating ICCs.

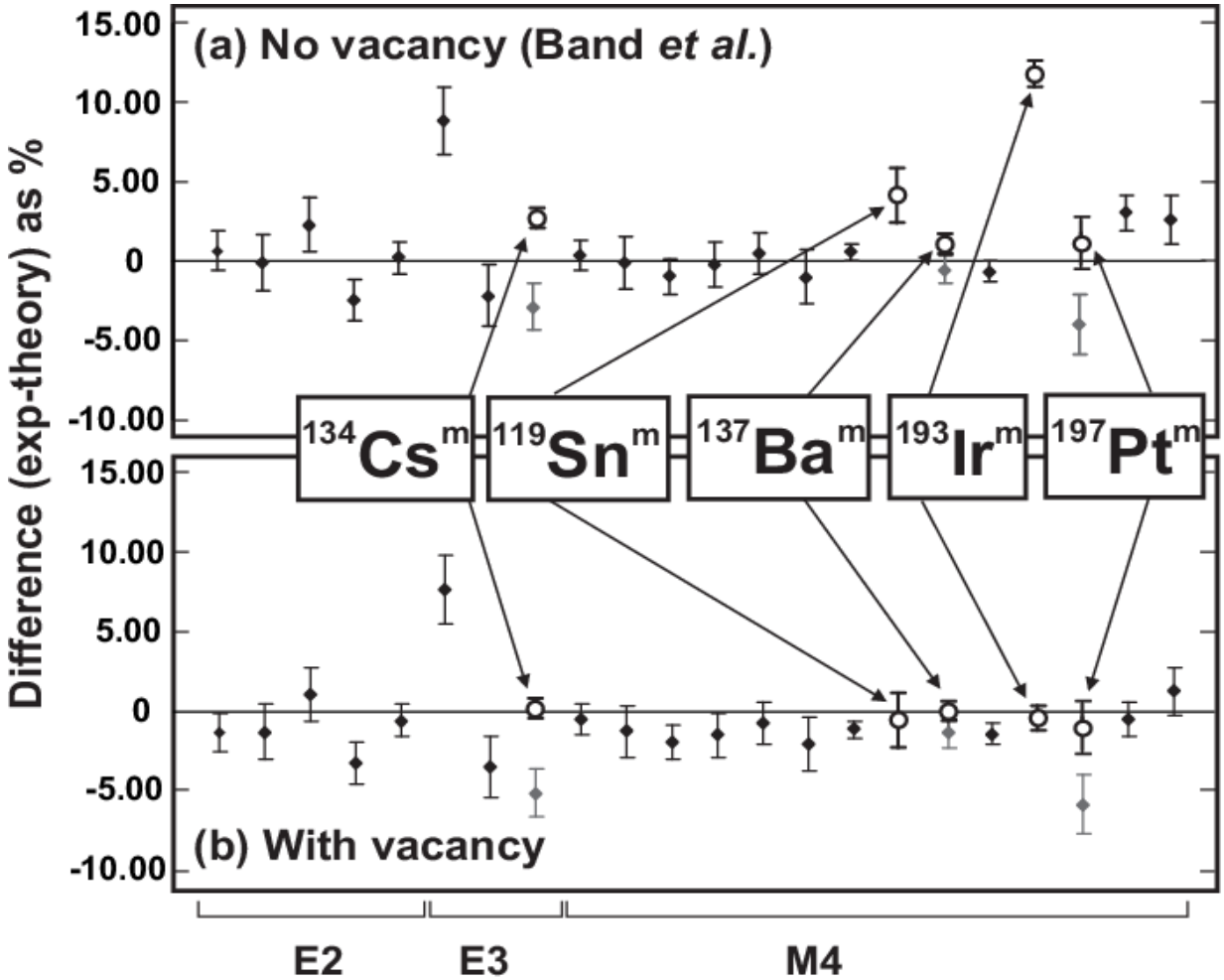

FIG. 1. Percentage differences between the measured and calculated ICCs for two Dirac-Fock calculations: one (a) is without the atomic vacancy and the other (b) is with it included in the ``frozen orbital'' approximation. The points shown as solid diamonds in both plots correspond to the twenty cases listed in Ref. [1] with better than 2% precision; as indicated at the bottom, five are for *E*2 transitions, three for *E*3, and the remainder are for *M*4 transitions. The points shown as open circles correspond to our five more-recently measured α_K values. For the cases of ^{134}Cs^m, ^{137}Ba and ^{197}Pt, the earlier Raman values are shown in grey; for ^{119}Sn^m and ^{193}Ir^m there were no earlier values with sub-2% precision.

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