Available online at www.sciencedirect.com
ScienceDirect
Applied Radiation and Isotopes 64 (2006) 1392–1395
Applied Radiation and Isotopes
www.elsevier.com/locate/apradiso

Test of internal-conversion theory with precise γ- and X-ray spectroscopy

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Abstract

Precise measurements of K-shell internal conversion coefficients (ICCs) are being used to determine the best method for calculating these coefficients. A recent result for the M4 transition from $^{193}Ir^{m}$ has been refined, demonstrating conclusively that the atomic vacancy created by the ejected electron must be properly accounted for in the calculation of ICCs. Measurements of additional cases are discussed.

Keywords: Internal conversion coefficients; Theory; $^{193}Ir^{m}$ M4 transition

1. Introduction

Even though internal-conversion coefficients (ICCs) have played an essential role in nuclear decay schemes for more than 50 yr, discrepancies still remain between calculated and experimental ICCs at the percent level and, in certain cases, this discrepancy can range up to as much as 10%. In many applications, a few-percent ambiguity in a calculated ICC has little impact; in others, it can impose a crucial limitation. For example, the precise efficiency calibration of a γ-ray detector (e.g. Helmer et al., 2003) depends in part on the use of γ-ray cascades, in which the transition intensities are known to be identical (i.e. there is no side feeding). Such transitions frequently involve the emission of conversion electrons as well as γ radiation, so the precision with which the relative intensities of the γ rays can be specified depends directly on how precisely the ICCs can be calculated. With detector efficiencies now being required at the sub-percent level (e.g. Hardy et al., 2003), the demands on calculated ICCs can be of the same order. Consequently, we have embarked on a program of measurements designed to test the accuracy of calculated ICCs.

In the calculation of an ICC, the final-state electron wave function must be calculated in a field that adequately represents the residual atom. It seems physically reasonable that this residual atom should include a vacancy in the atomic sub-shell from which the electron was emitted, since the filling time for a K-shell vacancy (10^{-15}–10^{-17} s) is appreciably longer than the time it takes for an electron to leave the atom (less than $\sim 10^{-18}$ s). Nevertheless, over the years, the calculations used to produce ICC tables have not always incorporated the hole in the atomic shell and, indeed, the most recent tables by Band et al. (2002) were computed with the hole deliberately ignored. The authors based this choice on a survey by Raman et al. (2002), which concluded that ignoring the hole led to better overall agreement with experiment. We report here on new measurements that refute this conclusion and we propose further experiments aimed at refining and improving ICC calculations.

2. Experimental method

Our experiments all utilize a HPGe detector that has been calibrated to 0.15% precision in *relative* efficiency

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doi:10.1016/j.apradiso.2006.02.093

(0.20% in *absolute*) from 50 to 1400 keV. The calibration procedure for this detector has been well documented in a series of publications (Hardy et al., 2002; Helmer et al., 2003, 2004). We focus on the measurement of K-shell ICCs and require a decay scheme in which only one observed transition converts significantly in the atomic K shell. We then measure a spectrum over an energy region that spans the K X-ray peaks and the γ-ray peak from that transition. Under these circumstances, the ratio of the total number of K X-rays observed (N_K) relative to the total number of γ rays (N_γ) relates to the K-shell internal conversion coefficient (α_K) by the simple relationship

$$\alpha_K \omega_K = (N_K/N_\gamma)(\varepsilon_\gamma/\varepsilon_K), \quad (1)$$

where ω_K is the fluorescence yield for that particular element and ε_γ and ε_K are the detector efficiencies for the γ rays and X-rays, respectively. Since fluorescence yields have been reviewed recently (Schönfeld and Janssen, 1996) and are considered to be rather precisely determined—typically to better than 0.5%—we can extract α_K directly from the measured peak areas in our detector to a precision of better than 1%. The survey of Raman et al. (2002) shows that, up until that date, only four transitions had experimental ICCs quoted to such sub-percent precision.

3. Case of $^{193}Ir^m$

Our first measurement of a K-shell conversion coefficient was for the 80.2 keV M4 transition from the 10.5 day $11/2^-$ isomer in ^{193}Ir. Since this is the only transition from the isomer and it directly populates the ground state, it is ideally suited to our measurement technique and, furthermore, its calculated α_K value depends very strongly on how the calculation handles the atomic hole: if the hole is ignored, the α_K is calculated to be 11% less than if the hole is included. In fact, this case is the most sensitive one known for testing this effect.

We made the measurement (Nica et al., 2004) with a highly purified source of $^{193}Ir^m$. Careful analysis of the γ-ray spectrum from its decay, recorded between 10 keV and 2 MeV, showed a number of weak impurities but none with an activity level above 0.5% that of $^{193}Ir^m$. Only one impurity, ^{192}Ir, and the X-rays from fluorescence of the tantalum source-backing material contributed in any way to the K X-ray peaks of iridium; no impurity made any significant contribution to the 80.2 keV γ-ray peak. After the few-percent correction for X-ray impurities and the inclusion of the efficiency ratio, $\varepsilon_\gamma/\varepsilon_K$, which was very close to 1, the measured peak ratio yielded the result $\alpha_K\omega_K = 98.7(6)$. Taking the fluorescence yield, $\omega_K = 0.958(4)$, from Schönfeld and Janssen (1996), we obtained the final result $\alpha_K = 103.0(8)$. This value strongly disagrees with the no-hole calculation of the ICC and agrees well with a calculation that incorporates the atomic hole.

The potential significance of this result led us to perform an ancillary experiment (Nica et al., 2005) aimed at confirming the value for the fluorescence yield of iridium that was used in our analysis. Although Schönfeld and Janssen (1996) assign a 0.4% uncertainty to their ω_K value for iridium, their result actually comes from a semi-empirical fit (as a function of Z) to existing data, which are quite sparse for $Z > 63$. In particular, there was no existing measurement at all for iridium ($Z = 77$), or indeed for any elements with $73 < Z < 80$ (Hubbell et al., 1994). To obtain the value of ω_K for iridium experimentally, we measured the ratio of K X-rays to 129.4 keV γ rays from the decay of the second excited state in ^{191}Ir. This (M1 + E2) transition (with a well-known mixing ratio) was observed following the β decay of ^{191}Os and is the only one in this decay that gives rise to significant numbers of K X-rays. Once again, the contribution of contaminants to the K X-ray or 129.4 keV γ-ray peaks was very small (<0.3%) and, from the measured peak ratio, we obtained the result $\alpha_K\omega_K = 2.044(11)$ via Eq. (1). In this case, the calculated ICC is virtually independent of the treatment of the atomic hole so we could simply use its value to extract ω_K from this result. We obtained $\omega_K = 0.954(9)$, which is in complete agreement with the value 0.958(4) obtained by Schönfeld and Janssen (1996) from their semi-empirical fit. This confirmed the result we had obtained in Nica et al. (2004) for the K-shell ICC of the 80.2 keV transition from $^{193}Ir^m$.

In cases like this ^{193}Ir transition, where the electron energy is only a few keV, it is important to have a precise value for the transition energy as well as for the atomic binding energy since the calculated value of α_K is a strong function of the energy difference. In Nica et al. (2004) we took the transition energy from Lindner et al. (1987) to be 80.22(2) keV. However, there is a more recent and precise measurement of the transition energy (Drissi, 1997), which was calibrated relative to the K_α X-rays of iridium and to some ^{192}Ir decay lines. A complication remains, though: the references used by Drissi for the X-ray energies (Bearden and Burr, 1967) and the ^{192}Ir decay lines (Kessler et al., 1978) have both been supplanted by more recent references, Deslattes et al. (2003) and Helmer and van der Leun (2000) respectively, so these must be taken into account before a final value for the 80.2 keV transition can be established. A careful examination of all four of these calibration references shows that the new ^{192}Ir energies act to reduce Drissi's value for the ^{193}Ir transition by 6 ppm, while the new X-ray energies act to increase the value by 6 ppm. We therefore accept Drissi's value as it stands and adopt 80.236(7) keV as the energy of our transition of interest in ^{193}Ir. For the K-shell binding energy in iridium, we use 76.112(1) keV from Deslattes et al. (2003), which is the same value we adopted in Nica et al. (2004).

With these values for the relevant energies, we have calculated α_K for the M4 transition in ^{193}Ir using three different assumptions concerning the atomic hole; our results appear in Table 1 together with Δ, the percentage difference between each calculation and the experimental value of 103.0(8). See Nica et al. (2004) for a brief description of each approximation. It is clear from the