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Improved calculations of electron capture transitions for decay data and radionuclide metrology

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HIGHLIGHTS

- An improved modelling for the calculation of electron capture is detailed.
- Overlap and exchange effects are corrected using Bahcall's and Vatai's approaches.
- Shake-up, shake-off and inner hole effects are approximately corrected.
- This modelling was compared with measurements and two other codes.
- The decay scheme of ^{40}K was updated using this modelling.

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ABSTRACT

Electron capture properties are crucial to establish the decay schemes of numerous radionuclides. The present modelling aims at improving the theoretical estimates of these decays, which are needed when no measurement is available. Allowed and forbidden unique transitions are calculated on the basis of precise relativistic wave functions of the atomic electrons, determined in previous work. In this context, correcting for atomic effects is of high importance. The two common approaches from Bahcall and Vatai to correct for the overlap and exchange effects have been extended to every subshell in a unified formulation, with the electron occupation precisely taken into account. The shake-up and shake-off effects, which create secondary vacancies, and the influence of the hole due to the capture process, have been considered. Uncertainties are also estimated. Relative capture probabilities and their ratios, including capture-to-positron ratios, have been found to be in good agreement with a selection of precise measurements. This modelling was then applied to the third forbidden unique transition of ^{40}K decay, with an update of the recommended values for the branching ratios and the total decay half-life.

1. Introduction

As the Designated Laboratory for France in charge of ionizing radiation metrology, the National Laboratory Henri Becquerel (LNE-LNHB) coordinates the evaluation of decay data within the international Decay Data Evaluation Project (DDEP) (Kellett and Bersillon, 2017), which aims at improving the fundamental knowledge of radionuclide decay schemes. The properties of electron capture transitions, such as capture probabilities and capture-to-positron probability ratios, are often pivotal information when establishing these decay schemes.

The capture process is very similar to beta decay, but instead of emitting an electron and an antineutrino, the nucleus absorbs an atomic electron and emits only a neutrino. Consequently, a ground-state to ground-state transition can only be detected through the subsequent atomic rearrangement, which makes precise measurements difficult to perform for low atomic number (Z) radionuclides. The theoretical calculations inherently depend on the precision of the atomic electron wave functions, for which a relativistic treatment is mandatory for high Z. Besides, as the nuclear structure component only acts as a constant factor in allowed transitions, ratios of electron capture probabilities are only sensitive to atomic properties and were therefore often measured to explore different atomic effects. The most complete survey of experimental and theoretical electron capture transitions was published 40 years ago (Bambynek et al., 1977).

The study of electron capture transitions is of importance in radionuclide metrology, fundamental physics and for many applications such as nuclear astrophysics and cosmochronology, absolute geochronology, nuclear medicine and nuclear energy. Regarding radionuclide metrology, the modelling of the light emission in activity measurements carried out by the Liquid Scintillation Counting (LSC) technique is sensitive to the number of emitted particles at low energy (Broda et al., 2007). For radionuclides decaying by electron capture, the LSC

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technique requires a precise modelling of the atomic rearrangement with a specific treatment for each of the different subshells. Hence, the capture probability is required for every subshell as this process creates the initial vacancies.

Investigating the ENSDF (Evaluated Nuclear Structure Data File) database (NNDC, 2017), which includes every known radionuclide, more than 35,000 beta or electron capture transitions can be found in more than 1500 radionuclides, split into 35% beta minus, 28% beta plus and 37% electron capture. It should be noted that decay data evaluations must rely on theoretical predictions when experimental data are missing. Any calculation code has thus to provide precise estimates in a wide range of cases. In modern decay data evaluations, the LogFT (2001) and EC-capture (Schönfeld, 1998) programs are commonly used for determining capture probabilities. However, these codes are limited by their models and the capture probabilities can only be obtained for the inner atomic shells.

The LogFT program calculates allowed, first and second forbidden unique transitions and also treats the beta plus component, if any (LogFT, 2001). The fraction per electron capture decay is determined for the K, L, and M_+ shells, where M_+ denotes the cumulative probability of the higher shells. It is noteworthy that these capture probabilities are not relative but absolute, that is to say $P_K + P_{L_1}+...\neq 1$. In fact, $\sum_i P_i = I_\varepsilon/(I_\varepsilon + I_{\beta^+})$, where I_ε is the absolute intensity of the electron capture branch and I_{β^+} is the absolute intensity of the competing beta plus branch. The probability ratios P_{L_2}/P_{L_1}, P_{L_3}/P_{L_1}, $P_\varepsilon/P_{\beta^+}$ and P_K/P_{β^+} are also determined. Uncertainties are propagated from the input parameters and an arbitrary modelling uncertainty of 1% is added for the total capture-to-positron ratio $P_\varepsilon/P_{\beta^+}$. Any final uncertainty less than 0.1% is not given by the LogFT program. The calculation assumes closed shells and is based on tabulated values of the radial densities at the nuclear surface of the electron wave functions, corrected for atomic overlap and exchange effects (Martin and Blichert-Toft, 1970). The atomic wave function parameters were determined using a relativistic Hartree-Fock-Slater self-consistent approach and a realistic Fermi-Dirac distribution for the nuclear charge density (Lu et al., 1971).

The EC-capture program calculates relative capture probabilities for allowed transitions and claims to calculate first forbidden non-unique ones (Schönfeld, 1998). However, they are actually treated as allowed, since the nuclear structure is not taken into account. Relative probabilities for K, L, M, N and O shells are given with uncertainty propagation. The calculation assumes closed shells and is based on tabulated ratios of the bound wave functions evaluated at the nucleus. The wave functions are from a relativistic modelling and the ratios are those given in Bambynek et al. (1977) for a small set of atomic numbers. The overlap and exchange correction is based on different models for low and high Z using the tabulated values given in Bambynek et al. (1977), again for a small set of atomic numbers. Interpolation was used by the authors to complete each table for all Z.

Due to these limitations, information is missing for the user. To improve the situation, the modelling developed in this work relies on relativistic bound wave functions of the atomic electrons calculated specifically for the radionuclides involved in the considered transition. Probabilities and their ratios are thus accessible for any subshell. This physical modelling is described in Section 2, for which good agreement with the existing codes can be expected for allowed transitions. A comparison with experimental data is shown in Section 3 for a selection of allowed transitions. Forbidden unique transitions have also been considered, but only very few measurements are available in the literature. Finally, this modelling was applied to the third forbidden unique transition that occurs in the ^{40}K decay in order to update the decay scheme with the deduced branching ratios, which in turns modifies the evaluation of the total half-life.

2. A consistent formalism for electron captures

By its very nature, electron capture decay highly depends on the atomic structure of the radionuclides involved. Precise theoretical predictions require precise bound state wave functions and it is well known that a relativistic modelling is mandatory for atomic numbers $Z \gtrsim 50$. A bound state is labelled by a set of two quantum numbers (n, κ) and the notation $x = (n, \kappa)$ is introduced to simplify the notations where no ambiguity is possible. A wave function is symbolised by $|(n, \kappa)\rangle$ or $|(n, \kappa)'\rangle$ in the parent or daughter atom respectively. Atomic energy, radial dependency, occupation number N_x and Coulomb amplitude β_x are necessary for each bound wave function. In the present work, the relativistic electron bound wave functions determined in Mougeot and Bisch (2014) for the precise calculation of the exchange effect in beta decays have been used.

The constants used throughout this work are α, the fine structure constant; m_e, the electron rest mass; c, the speed of light; $\hbar$, the reduced Planck's constant; and G_β, the Fermi constant. Natural units $\hbar = m_e = c = 1$ are applied in the present modelling. The nuclear radius is the usual one, $R = 1.2A^{1/3}$ fm, with A the mass number of the radionuclide.

Electron capture is an isobaric process which can compete with beta plus decay

$$\begin{aligned}(\varepsilon):&\quad {}^A_Z X + e^- \longrightarrow {}^A_{Z-1}Y + \nu_e\\ (\beta^+):&\quad {}^A_Z X \longrightarrow {}^A_{Z-1}Y + e^+ + \nu_e\end{aligned} \tag{1}$$

The available energy for the transition is determined from the Q-value and the energies of the initial and final nuclear states as $E_{\max} = Q+E_i - E_f$, and the total normalized energy is $W_0 = (1+E_{\max}/m_e)-2$. The recoil energy of the nucleus is neglected, as the largest known recoil occurs in ^{7}Be decay and is of 57 eV. A beta plus transition is only possible if $E_{\max}\geq 2m_e$ and the maximum energy of the beta spectrum is then $E_{\beta^+} = E_{\max}-2m_e$. The energy E_x of an orbital is by definition negative and $W_x = 1-|E_x|/m_e$ is the corresponding total energy. The momentum of the neutrino particle, assumed to be massless, is $q_x = W_0 + W_x$ and the momentum of the captured electron is $p_x = \sqrt{1-W_x^2}$.

2.1. Transition probabilities

Unlike beta decay, electron capture is a two-body process where particle energies are well-defined. However, the weak interaction acts in a very similar manner and a symmetry exists between both formalisms. A decay occurs from an initial nuclear state of the parent nucleus with spin and parity (J_i, π_i) to a final nuclear state (J_f, π_f) of the daughter nucleus. Transitions are classified as for beta decay on account of spin change $|\Delta J| = |J_i - J_f|$ and parity change $\pi_i\pi_f$. The formalism with spherical symmetry from Behrens and Bühring (1982) was followed throughout this work.

The total decay rate of a given transition is given by

$$\lambda_\varepsilon = \frac{\ln 2}{t_\varepsilon} = \frac{G_\beta^3}{2\pi^3}\sum_x n_x C_x f_x \tag{2}$$

where the partial half-life is defined from the total half-life and the branching ratio as $t_\varepsilon = T_{1/2}/P_\varepsilon$. The degeneracy of the orbital is $2k_x$, where $k_x = |\kappa|$, and n_x is the relative occupation number defined as $n_x = N_x/2k_x$ from the number of electrons N_x in the orbital. The quantity C_x plays a role similar to the shape factor in beta decay and the nuclear matrix elements ${}^A F^0_{L,L-1,1}$ are also, to a first approximation, independent of the lepton energy for allowed and forbidden unique transitions. Its expression can be found in Bambynek et al. (1977)

$$C_x = \frac{(2L-2)!!}{(2L-1)!!}\left[{}^A F^0_{L,L-1,1}\right]^2 R^{2(L-1)}\frac{p_x^{2(k_x-1)}q_x^{2(L-k_x)}}{(2k_x-1)![2(L-k_x)+1]!} \tag{3}$$

with $L = 1$ if $\Delta J = 0$ for an allowed transition and $L = \Delta J$ for any $(L-1)$th forbidden unique transition. The sum in Eq. (2) is limited by the L value such that $k_x = 1, 2, ...L$. Finally, the quantity f_x corresponds to the integrated Fermi function in beta decay and is defined as