Subthreshold internal conversion to bound states in highly ionized ^{125}Te ions

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A new mode of internal conversion decay, in which the converted electron is excited to a bound orbital instead of a continuum orbital, is discussed. General theoretical results are presented for the relation between bound internal conversion and continuum internal conversion of the nucleus. It is shown that the transition rate for internal conversion decay is continuous across the energy threshold between continuum final states and bound final states. Theoretical predictions for decay to bound states of ^{125}Te are consistent with experimental data on internal conversion in highly charged ions of this nuclide.

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I. INTRODUCTION

Many papers have focused on the search for ways of affecting the rates of elementary processes in particles and nuclei by applying electromagnetic fields [1,2]. To date, only very minor effects have been found except in certain cases where the frequency of the externally applied electromagnetic field is in resonance with eigenfrequencies of the system [2–4]. More substantial effects can be produced in certain cases where nuclear processes depend directly on the configuration of the electron shells. Such a situation occurs in the cases of internal conversion, electron or muon capture, and β decay.

New possibilites arise using beams of highly charged ions. One of the most spectacular examples of a dependence of a nuclear lifetime on the state of the electron shell has been studied recently [5,6]. It was shown that the lifetime of the first nuclear excited state $3/2^+$ in ^{125}Te can be increased by up to 640% by stripping the electron shells. This nuclear state decays by an $M1$ transition emitting a photon of energy 35.49 keV and has a lifetime of 1.49 ns in the neutral atom. The very large effect on the lifetime is due to an increase of the ionization potential of the K shell. As the atom is ionized a charge state can be reached for which the K shell binding energy is greater than the nuclear transition energy, at which point one would expect that the normal internal conversion process would stop. A significant increase of the lifetime was in fact observed, but for a critical charge state $q=47$ which is higher than the value of $q=45$ predicted on the grounds of simple energetics. It was suggested [6] that when the binding energy becomes larger than the transition energy, internal conversion can still occur by decay to appropriate bound final states. This led us to investigate the behavior of the internal conversion decay rate when the transition energy crosses the threshold from continuum final states to bound final states. It is shown herein that the rate for internal conversion remains constant across this threshold. For ^{125}Te the ion with $q=45$ can decay by excitation of the $1s$ electron to states with $n\simeq 20$. As the ionic state and transition energy increase further the conversion becomes strongly resonant. It is argued that the experimental results for ^{125}Te can be explained in terms of subthreshold discrete conversion, thus providing the first evidence for this new mode of internal conversion also known as bound internal conversion [6].

II. PHYSICAL BACKGROUND

A. Conversion to discrete levels

A Feynman graph of the process of decay to final bound states is shown in Fig. 1. The nuclear energy is transferred to the $1s$ electron which is promoted to a high-lying $n\kappa$ orbital in an intermediate state of the atom leaving a hole in the $1s$ shell. The high-lying orbital is specified by n, the principal quantum number and κ, the relativistic quantum number. Closely related transitions to bound final states resulting from applied electromagnetic fields have also been discussed under the headings of discrete or resonant internal conversion [1–4].

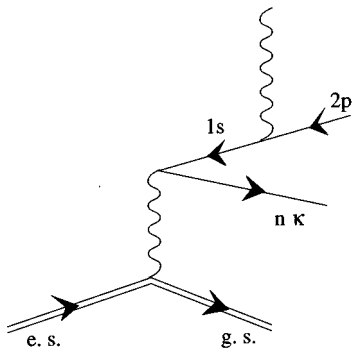

FIG. 1. Feynman diagram representing the bound internal conversion of a nuclear transition from an excited state (e.s.) to the ground state (g.s.). Double lines specify the nuclear transition. The atom is left with the converted electron in the $n\kappa$ orbital where n is the principal quantum number and κ the relativistic quantum number. The hole formed is filled by a $2p$ electron. Arrows pointing backwards in time on electron paths specify electronic hole states.

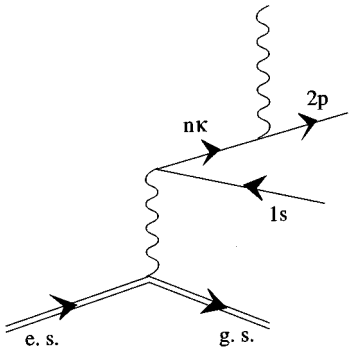

FIG. 2. Feynman diagram of a nuclear deexcitation through the electronic bridge. The atom is left with the converted electron in the $2p$ orbital and a hole in the $1s$ orbital.

A necessary condition for decay to occur to bound final states is that the electronic excitation energy be close to the energy of the nuclear transition. Energy matching can then occur since the final electron state resulting from the decay has a certain width, primarily due to the radiative decay of the hole state as well as to the slower Auger decay process. The initial state also has an associated width as a result of the internal conversion process itself, though in most cases this will be significantly less than the radiative width of the excited state. For K shell conversion of γ rays in atoms or ions possessing p electrons for example, the radiative width of the excited state is dominated by emission of a $2p \rightarrow 1s$ x ray. For ^{125}Te, such decay gives a hole state width of the excited ns level which is of the order of 5 eV for states containing one or more $2p$ electrons, as discussed below. Energy conservation is reflected in the energy of the emitted x ray, which is shifted by the resonance defect $\omega_\gamma - \omega_{n\kappa}$ where ω_γ is the transition energy, and $\omega_{n\kappa}$ is the energy difference between the states with an electron in the $1s$ and $n\kappa$ orbitals. The detection of the x ray energy shift in an experiment would give a stringent direct proof of the mechanism proposed in the present paper. A related energy shift was detected recently in the case of subthreshold Auger decay [7].

It is interesting to note that the deexcitation of the nuclear state can occur through simultaneous emission of a photon and the promotion of a $1s$ electron to a $2p_{1/2}$ orbital via a virtual ns state. This process, shown in Fig. 2, corresponds to an example of the well-known electronic bridge [8]. However, the process will have negligible probability, compared to the discrete transitions discussed above for ^{125}Te, because the width for the $ns \rightarrow 2p$ radiative transition is significantly smaller than that for a $2p \rightarrow 1s$ radiative transition.

We define the conversion factor $R^{n\kappa}$ as the ratio of the rate for internal conversion to this bound state to the rate for radiative decay of the nuclear state. $R^{n\kappa}$ is given by [1,2,9]

$$R^{n\kappa} = \alpha^{n\kappa} \frac{1}{2\pi} \frac{\Gamma}{(\omega_\gamma - \omega_{n\kappa})^2 + (\Gamma/2)^2}, \tag{1}$$

where Γ is the total width of the hole state produced and $\omega_{n\kappa}$ is the electronic excitation energy in the hole state produced by the decay. The quantity $\alpha^{n\kappa}$ is the analog of the internal conversion coefficient in the case of decay to discrete final states and is given by the same expression as for continuum decay [11] except that the wave function of the converted continuum electron, normalized on the energy scale, is replaced by the bound wave function for the $n\kappa$ orbital. For internal conversion of magnetic multipole radiation of multipolarity L, $\alpha^{n\kappa}$ is given by

$$\alpha^{n\kappa}(ML) = \pi\alpha\omega \frac{N_i(2j+1)(\kappa+\kappa_i)^2}{L(L+1)} \times \begin{pmatrix} j_i & j & L \\ \frac{1}{2} & -\frac{1}{2} & 0 \end{pmatrix}^2 |\mathscr{R}_{n\kappa}(ML)|^2, \tag{2}$$

where j is the total angular momenta of the final state orbital and j_i and κ_i are the total angular momentum and relativistic quantum number for the initial state subshell in which internal conversion occurs and which has an occupation number N_i. $\mathscr{R}_{n\kappa}(ML)$ is the radial matrix element, which is given by

$$\mathscr{R}_{n\kappa}(ML) = \int_0^\infty [g_{n\kappa}(r) f_i(r) + f_{n\kappa}(r) g_i(r)] X_L(\omega r) r^2 dr, \tag{3}$$

where $g_i(r)$ and $f_i(r)$ are the large and small components of the Dirac radial wave functions for the orbital in which conversion occurs. The quantity X_L, the potential associated with the nuclear currents, depends on the nuclear model. In the present calculations, the model of surface nuclear currents has been adopted [10,11] for which

$$X_L = \begin{cases} h_L^{(1)}(\omega r) \text{ for } r > R_0, \\ j_L(\omega r) \dfrac{h_L^{(1)}(\omega R_0)}{j_L(\omega R_0)} \text{ for } r \leqslant R_0, \end{cases} \tag{4}$$

where R_0 is the nuclear radius and the quantity $h_L^{(1)}(\omega r)$ is a spherical Hankel function of the first kind. It should be noted that, because of the different normalization of the bound and continuum wave functions, the quantity $\alpha^{n\kappa}$ in Eq. (2) has the units of energy. Since the Breit-Wigner factor in Eq. (1) has the units of inverse energy, the bound internal conversion coefficient, $R^{n\kappa}$, is dimensionless, as is required of a probability.

B. Average conversion probability

If the width Γ is less than the mean distance between electronic levels for which $\omega_\gamma \simeq \omega_{n\kappa}$, then Eq. (1) has a sharp resonance character involving a single $n\kappa$ orbital. If, on the other hand, the converted electron is in a bound state which is near the continuum threshold, the electron states may then be closely spaced, in which case the condition

$$\rho_{n\kappa}\Gamma \gg 1 \tag{5}$$

is fulfilled, where $\rho_{n\kappa}$ is the density of electronic states defined by

$$\rho_{n\kappa} = \frac{1}{\epsilon_{(n+1)\kappa} - \epsilon_{n\kappa}}, \tag{6}$$

and $\epsilon_{n\kappa}$ is the energy of the state associated with the electron in the $n\kappa$ orbital. If Eq. (5) is satisfied, we can define an average internal conversion coefficient by

$$R_{av}^{n\kappa}(M1)=\int_0^\infty \alpha^{n\kappa}(M1)\frac{1}{2\pi}\frac{\Gamma}{(\omega_\gamma-\omega_{n\kappa})^2+(\Gamma/2)^2}\rho_{n\kappa}d\epsilon_{n\kappa}, \tag{7}$$

where the integral is assumed to be taken over the energy interval $\Delta E \gg \Gamma$.

We now consider the general features of the $M1$ resonant conversion when condition (5) is fulfilled. Expressing the small components of the electron wave function appearing in Eq. (3) in terms of the large components for a vanishing electron binding energy and noting that, for the regions of r that contribute significantly to the radial integral in Eq. (3), the Coulomb potential is negligible compared to the electron rest mass, one can derive the following expression for $\alpha^{ns}(M1)$:

$$\alpha^{ns}(M1)=\frac{\alpha\pi}{6m_e^2\omega_\gamma^3}N_K|\psi_{1s}(0)\psi_{ns}(0)|^2, \tag{8}$$

where α is the fine-structure constant and $\psi_{ns}(r)$ is the nonrelativistic radial wave function for the ns orbital. For convenience in this and subsequent equations we use the notation ns in superscripts and subscripts to denote the final-state label $n\kappa$ with $\kappa=-1$. If we adopt a simple nonrelativistic hydrogenic approximation for the 1s electron wave function with an effective nuclear charge Z^* then the quantity $\psi_{ns}(0)$ is given by

$$\psi_{ns}(0)=\frac{2}{n^{3/2}}(m_e\alpha Z^*)^{3/2}. \tag{9}$$

Substituting this result into Eq. (8) yields

$$\alpha^{ns}(M1)=\frac{2\alpha\pi(m_e\alpha Z^*)^3}{3m_e^2\omega_\gamma^3 n^3}N_K|\psi_{1s}(0)|^2. \tag{10}$$

From this result it is evident that $\alpha\propto n^{-3}$. However, the density of the ns electronic levels is proportional to n^3 if n is sufficiently large:

$$\rho_{ns}=\frac{m_e n^3}{(m_e\alpha Z^*)^2}. \tag{11}$$

Substituting Eqs. (10) and (11) into Eq. (7) and assuming that $\Gamma\to 0$ so that the Breit-Wigner form becomes a δ function, one obtains the following expression for the conversion factor:

$$R_{av}^{ns}(M1)=\frac{2\alpha^2\pi Z^*}{3\omega_\gamma^3}N_K|\psi_{1s}(0)|^2. \tag{12}$$

One can see that after averaging over such closely spaced final states, the conversion probability does not depend on the energy of the converted electron.

It is instructive to compare Eq. (12) with the limiting expression for the internal conversion coefficient for a continuum electron when the transition energy tends to zero. The limiting form of the nonrelativistic hydrogenic continuum electron wave function, normalized on the energy scale, when the kinetic energy tends to zero is given by [12]

$$\psi(r)=\sqrt{\frac{2m_e}{r}}J_{2l+1}(\sqrt{8m_e\alpha Z^*}), \tag{13}$$

where J_ν is the Bessel function of order ν. Using the asymptotic expansion of J_ν near the origin for $l=0$ gives $\psi(0)=2\sqrt{m_e^2\alpha Z^*}$. Inserting this result into Eq. (8) we then obtain an expression for the internal conversion coefficient that coincides exactly with Eq. (12). This shows that the internal conversion coefficient changes smoothly as the transition energy passes through the K-shell threshold.

TABLE I. Binding energies for the 1s and 2s shells and the $2p_{1/2}$ and $2p_{3/2}$ subshells in $Te^{(0)}$, Te^{44+}, Te^{45+}, and Te^{46+}.

Orbital	Binding energy (keV)			
	$Te^{(0)}$	Te^{44+}	Te^{45+}	Te^{46+}
1s	31.82	35.262	35.581	35.909
2s	4.945	7.966	8.169	8.443
$2p_{1/2}$	4.618	7.658	7.995	8.243
$2p_{3/2}$	4.347	7.489	7.717	-

III. COMPARISON WITH EXPERIMENTAL RESULTS

The experiments on ^{125}Te decay [5,6] were carried out at the accelerator GANIL using a 27 MeV/amu ^{125}Te beam. The trajectories of the Te ions in the first nuclear excited state and different charge states ranging from 45+ to 48+ were analyzed with the spectrometer SPEG [13], which provided the necessary momentum resolution $\sim 10^{-5}$. The experimental set up and the data analysis are detailed in Refs. [5,6]. The flight time of the ions from the target to the spectrometer was such that radiative decay of excited atomic states would lead to all ions being in the ground state with the exception of a few long-lived metastable states in the case of Te^{48+}. The following halflives were obtained [5]: $T_{1/2}<2$ ns for $q=45$ and 46, $T_{1/2}=(6\pm1)$ ns for $q=47$, and $T_{1/2}=(11\pm2)$ ns for $q=48$. The experimental lifetimes for $q=45$ and $q=46$ are not significantly different, which is in accordance with the analysis discussed above.

In the present paper, the values of the internal conversion coefficient (ICC) for ^{125}Te ions in various charge states for transitions to both continuum and bound states have been calculated by means of the computer package RAINE [14,15]. The energies of the atomic states have been calculated using a Dirac-Fock code [16]. The ionization potentials $I(q)$ of the K shell are $I(44)=35.262$ keV, $I(45)=35.581$ keV, $I(46)=35.909$ keV, $I(47)=36.256$ keV, and $I(48)=36.589$ keV. For comparison, the results from the GRASP Dirac-Fock code [17] are $I(44)=35.262$ keV, $I(45)=35.581$ keV, $I(46)=35.910$ keV, $I(47)=36.257$ keV, and $I(48)=36.604$ keV. The values of $\alpha^{ns}(M1)$ for internal conversion to bound states were calculated from Eqs. (2) and (4) using the Dirac-Fock wave functions. For each ion, the width of the state with the K-shell hole was estimated by extrapolating

TABLE II. Values of the internal conversion coefficient to continuum states for the $1s$ and $2s$ shells and the $2p_{1/2}$ and $2p_{3/2}$ subshells for the 35.49 keV $M1$ transition in $Te^{(0)}$, Te^{44+}, and Te^{46+}. For conversion in the $1s$ shell, the results are presented for two cases, with (A) and without (B) allowance for the $1s$ hole created.

Ion	Subshell				
	$1s$		$2s$	$2p_{1/2}$	$2p_{3/2}$
	A	B			
$Te^{(0)}$	11.59	11.66	1.411	0.113	0.028
Te^{44+}	10.78	10.80	1.481	0.121	0.031
Te^{46+}	-	-	1.519	0.126	-

from the values given in [18], according to the number of $2p_{1/2}$ and $2p_{3/2}$ electrons.

A. $^{125}Te^{44+}$

The calculated values of the ionization energies for the $1s$, $2s$, $2p_{1/2}$, and $2p_{3/2}$ shells are shown in Table I. For comparison we also show the corresponding values for the neutral atom and for the ions Te^{45+} and Te^{46+}. In Table II we report the calculated values of the internal conversion coefficents for the $1s$, $2s$, $2p_{1/2}$, and $2p_{3/2}$ shells in $Te^{(0)}$, Te^{44+}, and Te^{46+}, in each case for decay to continuum states. The possible influence of the hole formed in the K shell on the ICC, due to the change in the self-consistent mean field of the atom in the final state, has been investigated. For this purpose, the ICC were calculated both with and without allowance for the hole created. Both sets of results are presented for the case of conversion in the $1s$ shell in Table II, from which it can be seen that the difference in the ICC values does not exceed 1% in this case. For the other shells, the effect of taking the hole into account was found to decrease the ICC values by less than 0.2%. In order to consider conversion in the $2s$ shell, which is of special interest for $q>44$, the calculations were performed for several energies. The results are presented in Table III. It is remarkable that the ionization energy in this shell changes almost by a factor of two in stripping the outer shells from the neutral atom as far as the state with $q=44$, but the corresponding ICC values change at most by 5%. For this reason, one might not expect a significant change in the lifetime as electrons are stripped from the atom as long as the deexcitation of the nucleus occurs predominantly through normal internal conversion in the $1s$ or $2s$ shells.

TABLE III. Values of the $2s$ internal conversion coefficient to continuum states as a function of energy for $Te^{(0)}$ and Te^{46+}. The results are presented for two cases, with (A) and without (B) allowance for the $2s$ hole created.

Energy	$Te^{(0)}$		Te^{46+}	
(keV)	A	B	A	B
10	60.16	60.34	63.86	64.03
20	7.691	7.698	8.192	8.193
30	2.315	2.319	2.488	2.489

TABLE IV. Transition energies and values of the bound internal conversion coefficient, $R^{ns}(M1)$, for decay to the dominant bound final state ns orbitals in Te^{45+}. Also shown are values for the quantity $\alpha^{ns}(M1)$ defined in the text.

Orbital	Transition energy (eV)	$\alpha^{ns}(M1)$ (eV)	$R^{ns}(M1)$
$7s$	34968	2095.1	0.006
$8s$	35114	1395.0	0.008
$9s$	35213	970.9	0.010
$10s$	35284	705.2	0.013
$11s$	35334	526.3	0.017
$12s$	35373	399.1	0.022
$13s$	35404	308.6	0.031
$14s$	35429	240.7	0.048
$15s$	35450	188.3	0.082
$16s$	35465	146.7	0.152
$17s$	35478	113.1	0.408
$18s$	35489	85.7	3.72
$19s$	35499	63.3	1.12
$20s$	35507	45.4	0.161
$21s$	35515	31.5	0.050
$22s$	35522	21.1	0.020
$23s$	35527	13.7	0.009
$24s$	35533	8.6	0.004
$25s$	35538	5.4	0.002
Total			5.9

B. $^{125}Te^{45+}$

For $q=45$, the nuclear energy difference is slightly less than the $1s$ ionization energy. Using the calculated values of the ionization energy of the $1s$ electron quoted above and adding the transition energy ω_γ (=35492.5 eV) we find that for $q=45$ the binding energy of the electron excited in the conversion process should lie 88 eV below threshold. This value corresponds to a high $n=18$ orbit and an energy separation between two consecutive levels of approximately 10 eV. Taking the value $\Gamma=5$ eV for the $1s$ vacancy width, calculated according to the method described above, we can see that $\Gamma\rho\simeq 0.5$ and condition (5) is therefore not too far from being satisfied. From Eq. (7) $R_{av}(M1)$ is calculated to be 8.8. Values of $\alpha^{ns}(M1)$ and $R^{ns}(M1)$ are shown in Table IV for values of n ranging between 7 and 25. As one can see from the table, summing over all ns orbitals with $7\leqslant n\leqslant 25$ yields a total bound internal conversion coefficient of 5.9. Clearly the actual value of $R^{ns}(M1)$ is strongly dependent on the exact energy of the final state. Even the small differences in the binding energies obtained with different Dirac-Fock codes that were noted above are sufficiently large to modify the theoretical values of $R^{ns}(M1)$. If the binding energies were to change by 3.5 eV, exact resonance on the $18s$ level could occur yielding a maximum value of $R^{18s}(M1)$ of 10.9.

C. $^{125}Te^{46+}$

For $q=46$, the electron is excited to a state that is more deeply bound than in the above case with $q=45$. In Table V, the calculated transition energies for $q=46$ are listed for

TABLE V. Transition energies and values of the bound internal conversion coefficient, $R^{ns}(M1)$, for decay to the dominant bound final state ns orbitals in Te^{46+}. Also shown are values for the quantity $\alpha^{ns}(M1)$ defined in the text.

Orbital	Transition energy (eV)	$\alpha^{ns}(M1)$ (eV)	$R^{ns}(M1)$
$5s$	34658	6110	0.006
$6s$	35052	3500	0.012
$7s$	35287	2180	0.033
$8s$	35439	1450	0.320
$9s$	35542	1010	0.262
$10s$	35616	733	0.031
$11s$	35670	533	0.011
Total			0.67

various $1s \rightarrow ns$ transitions. The results suggest that the energy of the dominant virtual state formed is close to the energy of the $9s$ level. In this case the energy separation between two consecutive levels is approximately 100 eV. The condition (5) is far from being satisfied and the averaging procedure is no longer valid. The calculated $R^{ns}(M1)$ values for each transition are given in Table V. It is clear that the $9s$ level gives the dominant $R^{ns}(M1)$ value and again this depends strongly on the energy of the converted electron. Summing over all ns orbitals with $5 \leqslant n \leqslant 11$ yields a total bound internal conversion coefficient of 1.5. If the energy of the electronic transition were to change by 23 eV, a maximum value of $R^{9s}(M1) = 161$, corresponding to exact resonance, would be reached.

D. $^{125}Te^{47+}$

For $q=47$, condition (5) is violated even more strongly than in the case above of $q=46$. An averaging procedure is therefore certainly not appropriate. However, the basic picture of discrete internal conversion is similar to that for the case of Te^{46+}. The dominant final state is found to be $1s^1 2s^2 2p^1 7s^1$ for which $R^{7s}(M1)=0.16$. Summing over values of $R^{ns}(M1)$ for the final states $1s^1 2s^2 2p^1 ns^1$ with $5 \leqslant n \leqslant 11$ yields a total value of 0.2. An energy shift of 66 eV would give exact resonance on the $7s$ level with $R^{7s}(M1)=760$.

E. $^{125}Te^{48+}$

For decay of the $1s^2 2s^2$ state, the absence of a $2p$ electron leads to a drastic diminution of the width for radiative decay of the excited state by at least three orders of magnitude. Internal conversion to bound final states is therefore predicted to have negligible probability. It should be noted that in the experiment the beam is such that a very short time after the target interaction, only 59% of the ions in the beam are in the $1s^2 2s^2\ {}^1S_o$ ground state and 41% are in the metastable $1s^2 2s^1 2p^1\ {}^3P_o$ state. In this situation the nuclear lifetime depends not only on the charge state but also on the electronic configuration. Taking into account only the conversion in the L shell, the calculated lifetimes are found to be $T_{1/2} = 12.1$ ns in the ${}^3P_o$ state and $T_{1/2} = 8.7$ ns in the ${}^1S_o$ state. The experiment was not able to resolve the two configurations but the observed lifetime of 11 ± 2 ns is in agreement with an average lifetime (10.1 ns) for L-shell conversion in the two configurations and is therefore consistent with the absence of internal conversion decay in the $1s$ shell by excitation to bound or continuum final states.

IV. CONCLUDING REMARKS

The theory of bound internal conversion that is also valid in the case of overlapping resonances is formulated. It is shown that the internal conversion decay rate changes smoothly as the K-shell threshold moves from a value below the transition energy to a value above this energy. It is remarkable that the conversion rate in the K shell turns out to depend on the availability of electrons in the $2p$ shell. The observed experimental data is consistent with the general features of the bound internal conversion process discussed above. Further theoretical investigations including precise calculations of the transition energies, and taking account of the effect of configuration mixing on the energies and level densities, are desirable.

It is important to note also that, in principle, the deexcitation of the nucleus by excitation of an electronic state and the subsequent decay of the electronic hole state produced are intimately connected and a simple two-step picture may be overly simplistic. Indeed, the relation between the one-step and two-step processes has been discussed recently in the context of the related subthreshold Auger decay process [7,19–21]. In this article we have restricted the discussion to the two-step case since our main aim is to highlight the possibility of subthreshold internal conversion decay and to illustrate its basic features.

From the experimental point of view, the detection of the characteristic x rays following bound internal conversion decay, and measurement of their energies, is highly desirable.

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