Charge State Blocking of K-Shell Internal Conversion in ^{125}Te

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We have studied the atomic charge state dependence of the nuclear lifetime of the 35.5-keV first excited state of ^{125}Te. For the 47^+ and 48^+ ions, 300% and 640% increases, respectively, of the half-life were found with respect to the neutral-atom value (1.49 ns). These unusually large effects are due to the energetic blocking of K-shell internal conversion as the charge state increases past 47^+.

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The influence of the electronic environment on nuclear decay processes has been investigated for many years [1]. Electron capture, β decay, and internal conversion (IC), all of which involve atomic configuration changes between the initial and final nuclear states, have been used for this purpose [2]. A common feature of these processes is their sensitivity to the electronic wave function close to the nucleus. In particular, one can see from calculations of Band, Sliv, and Trzhaskovskaya [3] that the internal conversion coefficient (ICC), which in neutral atoms ($\alpha^{(0)}$) is proportional to the electronic density $\rho(r)$ in the nuclear region [4], in an ion of charge state Q is given by the (nonrelativistic) scaling relation

$$\alpha^{(Q)} = \alpha^{(0)} \lim_{r\to 0} \rho^{(Q)}(r)/\rho^{(0)}(r)\,. \qquad (1)$$

Here, the superscripts (0) and (Q) refer to the neutral atom and to the ion of charge state Q, respectively. Any change in the configuration of the outer electronic shells modifies the screening and thus the inner electronic wave functions. The resulting alteration of the electronic density near the nucleus, and hence of the ICC, is generally small, even for s electrons. This has been confirmed experimentally by Ulrikson *et al.* [5] who found a negligible difference of the K-shell ICC of the 89-keV, $M1$ transition between the ^{197}Au^{10+} ion and the neutral atom. Recently, Phillips *et al.* have measured the ICC of the 14.4-keV transition in various ^{57}Fe ions [6]. Except for the H-like ion, where the hyperfine interaction plays an important role, the charge state dependence of the ICC in the range $Q = 19$–23 follows the scaling law (1) and leads to a variation of the ICC of $\sim$20% over this range.

In this Letter, we report a 300%–640% increase of the first excited-state lifetime of ^{125}Te due to a large change in the ICC with increasing Q. This strong Q dependence of the ICC is in contradiction with Eq. (1), because it is due to the energetic blocking of K-shell IC beyond a critical $Q = Q_c$. Since in the neutral ^{125}Te atom, K-shell IC is the dominant process, if it can no longer occur, there is a drastic reduction of the ICC above Q_c.

In ^{125}Te, the 35.49-keV first excited nuclear state ($3/2^+$) decays to the $1/2^+$ ground state by an $M1$ transition with a neutral-atom ICC of 13.91, of which K-shell IC contributes 11.99 [7]. The K-shell binding energy ranges from $E_K^0 = 31.81$ keV for the neutral atom to $E_K^{51+} = 38.18$ keV for the H-like ion [8]. If $E_K^{Q_c}$ exceeds 35.49 keV, the total ICC should drop to <1.9. The expected lengthening of the nuclear half-life $T_{1/2}$ can be obtained from

$$T_{1/2}^{(0)} = \hbar \ln 2/[\Gamma_\gamma^{(0)}(1 + \alpha_K^{(0)} + \alpha_L^{(0)} + \alpha_M^{(0)})], \qquad (2)$$

$$T_{1/2}^{(Q)} = \hbar \ln 2[\Gamma_\gamma^{(0)}(1 + \alpha_K^{(Q)} + \alpha_L^{(Q)})]. \qquad (3)$$

In the neutral atom, N and higher shell IC are negligible [4]. Equation (3) assumes that in the ion only K- and L-shell electrons are present. The partial gamma-ray width $\Gamma_\gamma^{(0)}$ is given by $\hbar \ln 2/\Gamma_\gamma^{(0)} = 22.2$ ns [9]. Above $Q = Q_c$, one expects $\alpha_K^{(Q)} = 0$.

The experimental procedure follows that of Phillips *et al.* [6]. The IC decay of the nuclear state increases the ionic charge state by one unit and the change of Q modifies the ion trajectory inside a dispersive flat-field magnetic spectrometer. The nuclear decay time is inferred from the spatial distribution of the ions at the spectrometer exit.

A beam of 27-MeV/amu ^{125}Te^{38+} ions was produced at the accelerator GANIL. The beam impinged on a 1-mg/cm^2 thick ^{232}Th target mounted at a distance $d =$ 11 cm from the entrance of the magnetic spectrometer SPEG [10]. The ^{125}Te ions exiting from the target had an approximately Gaussian charge state distribution with $\overline{Q} \simeq 47$ and $\sigma_Q \simeq 1.6$, in agreement with calculations [11]. Scattered ^{125}Te ions with $Q = 45$–49 were all collimated to angles $\theta = 38$–46 mrad with respect to the incident beam direction. Approximately 0.1% of the nuclei were thereby Coulomb excited to higher states. Since the nuclear grazing angle for this collision is ~105 mrad, nuclear reactions were essentially avoided. The feeding probabilities of the first excited state from Coulomb excited higher levels (with half-lives $\leq 10^{-9}$ s) were evaluated from known branching ratios and $B(E2)$ values [12,13].

During the ~1.5 ns (independent of Q) required to travel the distance d, K-shell vacancies created in Te ions by stripping in the ^{232}Th target have enough time to be filled. For example, in B-like Te, the $(1s2s^2 2p^2)^4 P_{1/2}$ electronic state has a half-life of $<10^{-11}$ s [14]. Except for ions with some particular L-shell metastable configurations [6], Te ions in the different charge states enter the magnetic field region in the atomic ground state. Hence, any change of Q in the spectrometer signals an IC process with electron emission. Auger processes following K-shell IC have a small probability to occur in Te ions. In B-like Te with a configuration $1s2s^2 2p^2$ the calculated fluorescence yield is 86% [14]. For all relevant charge states with $Q \geq Q_c$, a double-charge change is then completely negligible.

Beyond the exit of SPEG magnet, the X, Y positions of the ^{125}Te ions in a plane perpendicular to the optical axis (Z) were measured event by event in two identical transmission detectors, separated by ~2 m. Each detector consisted of a parallel-plate section giving a fast trigger signal and a drift chamber. At ~10 cm from the second detector, ions were stopped in an ionization chamber with a 2% resolution to determine their total energy. For a given magnetic rigidity and geometry, the data so obtained were sufficient to reconstruct each ion trajectory.

For this purpose, a ray-tracing program TURTLE$^+$ [15] was written. In Fig. 1(a), the spatial distribution along X

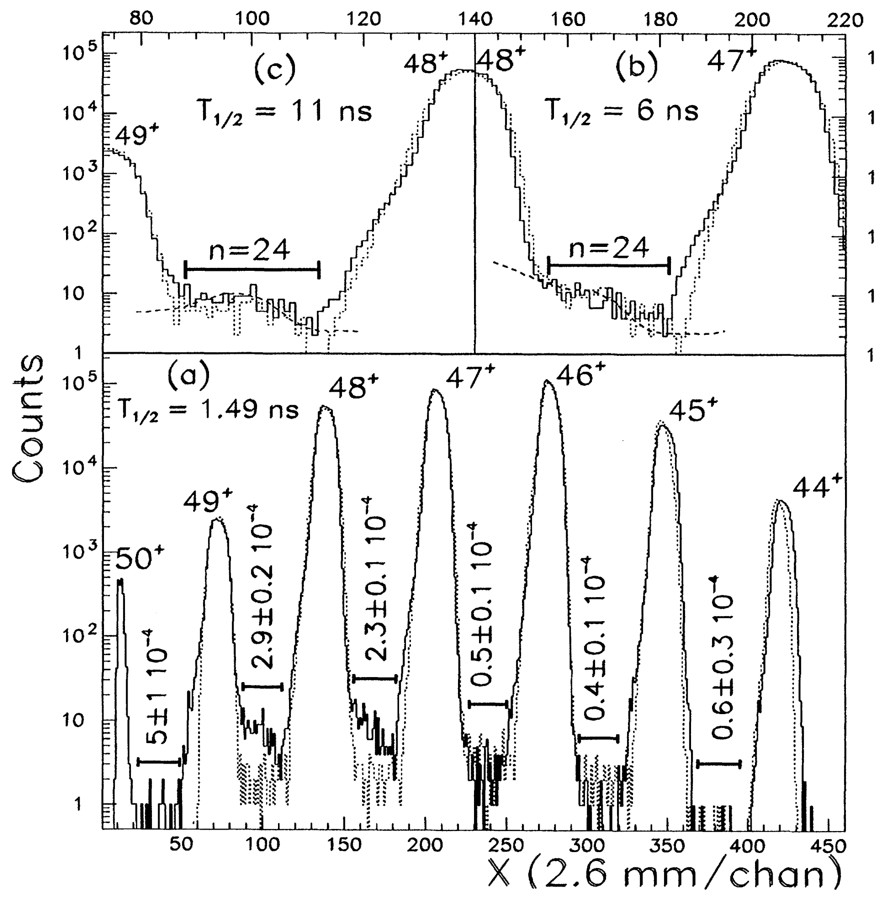

FIG. 1. (a) Experimental spatial spectrum of ^{125}Te ions. A simulation (dotted) is shown only for one Q-independent half-life $T_{1/2} = 1.5$ ns. The numbers give the ratios of the total number of interpeak counts in each selected interval indicated by the horizontal line to the counts in the corresponding peak on the right. Separately measured background ratios (see text) for $Q = 46$, 47, and 48 are $(0.4 \pm 0.1) \times 10^{-4}$, $(0.4 \pm 0.1) \times 10^{-4}$, and $(0.5 \pm 0.1) \times 10^{-4}$, respectively. (b) Expanded interpeak spectra and low- and high-statistics simulations (dotted and dashed) for $T_{1/2} = 6$ ns for $Q = 47$. (c) Same as (b), but for $T_{1/2} = 11$ ns, $Q = 48$.

in the second drift chamber is shown for Q between 44 and 49. The calculated positions, shapes, and intensities of the main peaks are in very good agreement with experiment under different conditions of magnetic rigidity, scattering angle, and incident angles and positions in the spectrometer. Hence, we are sure of the Q assignment of each peak. Since the distance between adjacent peaks is ~160 mm, only three peaks could be measured at a given magnetic rigidity. By adjusting the latter, overlapping spectra could be obtained, from which the composite figure was constructed.

Each main peak Q in the distribution corresponds to ^{125}Te ions which have kept their initial charge Q when entering the spectrometer, as well as those few ions which have changed their initial charge $Q - 1$ to Q during the flight time over the distance d as a result of IC decay, before entering the spectrometer. Each peak shape is asymmetric because of the θ dependence of the elastic scattering cross section within the $\Delta\theta$ interval fixed by the collimating slits. Energy loss and straggling in the target broaden the left-hand ("high-Q") side of each peak. IC decay within the spectrometer of an ion with initial charge Q brings additional counts under the peak $Q + 1$ if $T_{1/2}^{(Q)}$ is short (respectively, Q, if long) compared to the relevant flight time in the first dipole of SPEG (24 ns). If $T_{1/2}^{(Q)}$ is of the order of the flight time, counts appear between the peaks $Q + 1$ and Q. The latter case occurs for $Q = 47$ and 48 [Figs. 1(b) and 1(c)]. The case of a short half-life is illustrated by $Q = 44$–46. The interpeak background was determined by measurement at $\theta < 10$ mrad where Coulomb excitation is negligible (see caption of Fig. 1).

Simulations of ionic trajectories with different half-lives $T_{1/2}$ were made with TURTLE$^+$. One such simulated spectrum is show in Fig. 1(a) for the assumed value of $T_{1/2} = 1.5$ ns independent of Q. We also show part of the simulated spectra with $T_{1/2} = 6$ ns for the charge $Q = 47$ in Fig. 1(b) and $T_{1/2} = 11$ ns for the charge $Q = 48$ in Fig. 1(c). These simulations include the trajectories of the unperturbed ions and take into account the energy distribution of the ^{125}Te ions which reproduces the interpeak background.

We made a quantitative comparison between experiment and simulation over each interval of n channels, indicated by horizontal lines in Fig. 1, χ^2/n as a function of $T_{1/2}$. Because for a given initial number of parent ^{125}Te ions (before IC decay) only a very limited number of daughter ions (after IC decay) reach a given channel in an interpeak interval, the simulations were also started with a large number of daughter ions. The resulting spectra, such as those shown by dashed curves in Figs. 1(b) and 1(c), were appropriately normalized to the simulations starting with parent ions. The distributions of χ^2/n as a function of $T_{1/2}$ so obtained are shown in Fig. 2 for $Q = 46$–48. The number of interpeak counts is a double valued function of $T_{1/2}$, but large values of $T_{1/2}$ can be rejected on physical grounds. One can also obtain an approximate estimate of $T_{1/2}$ by comparing the total number of interpeak counts in a given channel interval with the results of simulations for various values of $T_{1/2}$.

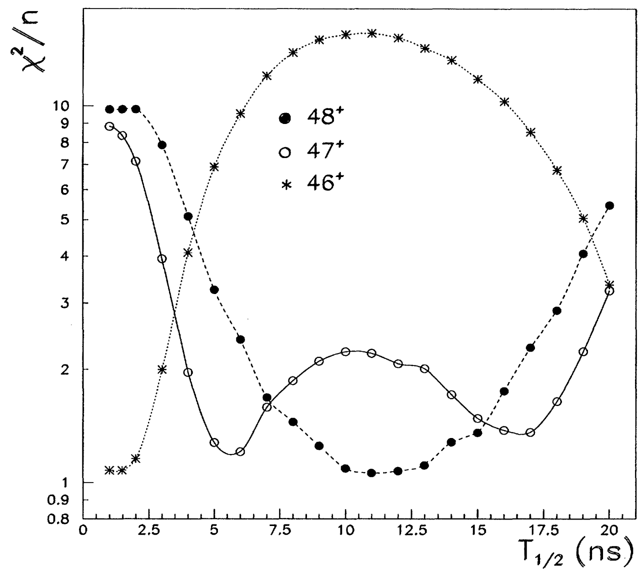

FIG. 2. Distribution of χ^2/n between simulation and experimental data in the interpeak regions for $Q = 46$–48 as a function of the assumed $T_{1/2}$.

The results are summarized in Table I, with 1 standard deviation uncertainties. We note the drastic change of $T_{1/2}$ when the charge state of the ^{125}Te ions changes from $Q = 46$ to $Q = 47$. This change is the first known evidence of K-shell IC blocking. For $Q \geq 47$, nuclear deexcitation can convert only on L-shell electrons, despite the presence of K electrons.

There are no ICC tabulated for ionic elements. Using new near-threshold L-shell ICC calculations with Dirac-Fock wave functions for the continuum and bound electrons [19], we obtained the calculated $T_{1/2}$ values shown in Table I. For $Q \geq 47$, there is general agreement between experiment and calculation.

A major discrepancy occurs between the experimental and calculated $T_{1/2}$ values for $Q = 45, 46$. The calculated E_B values of Refs. [8,16] exceed the 35.49-keV threshold value already for $Q \geq 44$, at which point K-shell IC should be blocked. This should lengthen $T_{1/2}$ significantly above the neutral-atom value. Two possible sources of error occur to us to explain this discrepancy: the threshold energy and the ionic K-shell binding energy. (i) The threshold energy is known within 0.5 eV from photoionization measurements [20]. (ii) The calculations of E_B^K [8,16] are believed to be accurate to 30–50 eV.

One process has occurred to us which could remove the discrepancy qualitatively: once E_B^K exceeds 35.49 keV,

TABLE I. Characteristics of the 35.49-keV first excited state of ^{125}Te for various ionic charge states. Boldface K-shell binding energies E_B^K exceed 35.49 keV.

Charge		$T_{1/2}$ (ns)		E_B^K (keV)	
state	Config.	Exp.	Calc.	Ref. [8]	Ref. [16]
0	(neutral)	1.49[a]	1.49	31.81[b]	31.81[b]
44	$(2s^2 2p^4)$		1.5		35.27
45	$(2s^2 2p^3)$	<2[c,e]	8.0	**35.61**	**35.58**
46	$(2s^2 2p^2)$	<2[e]	8.3	**35.91**	**35.90**
47	$(2s^2 2p)$	6 ± 1	8.5	**36.26**	**36.23**
48	$(2s^2)$	11 ± 2	10.3[d]	**36.60**	**36.57**

[a]Experimental value, used as basis for values calculated from Eqs. (2) and (3).
[b]From Ref. [17]
[c]Obtained by the less accurate integral interpeak count method (see text).
[d]Mean value, considering the presence of ~56% configuration $(2s2p)\,^3P_0$, which has a half-life of $\sim 10^{-3}$ s [18].
[e]Confidence level on the lower limit <2 ns is 68%.

K-shell IC can still occur, but the electron, rather than being ejected to the continuum, ends up in an *unoccupied bound state* of the ion. We call this process *bound internal conversion* (BIC) by analogy with bound β decay [2].

BIC and IC are a form of virtual photoexcitation and photoionization, respectively, with a similar smooth transition from below to above threshold, as the real processes. Taking into account the natural $1s$-level width of ~15 eV in neutral Te, BIC might be appreciable to states a few hundred eV below E_B^K. According to this hypothesis, the value of $\alpha_K^{(Q)}$ in Eq. (3) rather than being zero for $Q \geq Q_c$, would be finite for one, or at most two, Q values above threshold, thereby reducing the experimental half-life.

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