Charge-State Dependence of Nuclear Lifetimes

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Measurements of conversion rates are reported for the first time in one- and two-electron systems, for the 14.4-keV nuclear state in ^{57}Fe. The total half-life of this state deduced for the two-electron ion is 100 ± 5 ns, and that deduced for the total spin $F=1$ state of the one electron ion is 79 ± 6 ns.

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Internal conversion (IC), which can play a major role in determining the lifetimes of low-lying nuclear levels, is usually observed in neutral or near-neutral atoms with closed inner electron shells. (See Ref. 1 for a discussion of perturbations of nuclear decay rates caused by modifications of valence electron wave functions in neutral atoms.) No measurements of the conversion process exist for few-electron systems. Such experiments became possible through the availability of highly stripped heavy-ion beams from accelerators with very good beam qualities.

This Letter reports the first observation of K-shell conversion rates and nuclear lifetimes in one- and two-electron ions. These are simple systems with inner-shell electron wave functions that are modified because of the absence of outer electrons. The properties of such systems are of potential interest to nuclear astrophysics, to studies of nuclear properties, and in atomic physics where they may provide sensitive and straightforward tests of the ability of relativistic Hartree-Fock theory[2] to describe inner-shell electron properties. This is especially true of magnetic dipole IC, for which the conversion coefficient is a good measure[3] of the electron density at the nucleus where electron wave functions are most sensitive to relativistic effects.

The first excited state of ^{57}Fe at $E_x = 14.4$ keV, has a half-life of 98.1(3) ns in the neutral atom,[4] a total internal conversion coefficient $\alpha_T = 8.18(11)$, and the predicted[3] ratio $\alpha_K/\alpha_T = 0.91$. From these data one obtains a γ-decay probability per unit time $\lambda_\gamma = 7.7 \times 10^5$ s^{-1}, and a K-conversion decay probability per unit time in the neutral atom $\lambda_K = 5.73 \times 10^6$ s^{-1}. The deexcitation of the 14.4-keV level is predominantly a magnetic dipole transition, the ratio of the electric quadrupole to the magnetic dipole intensity being[4] $\sim 5 \times 10^{-6}$. Several short-lived higher excited states in ^{57}Fe are connected to the ground state by strong electric quadrupole matrix elements, and also deexcite with large branching to the 14.4-keV level. Multiple Coulomb excitation of these higher levels can thus produce significant population of the 14.4-keV state in a beam of ^{57}Fe scattered from a gold target. Coulomb excitation was used in the present experiments to produce excited ^{57}Fe ions. The IC decay of the 14.4-keV level in different ionic charge states was detected by measuring the trajectories of the ions after charge changes in a magnetic field.

Gold targets, about 400 μg/cm^2, were bombarded with pulsed beams of ^{57}Fe at an energy of 346 MeV, produced in the ATLAS accelerator at Argonne National Laboratory. Ions scattered at laboratory angles θ_L were accepted in an Enge split-pole magnetic spectrograph[5] through a rectangular aperture subtending a solid angle of 0.25 msr and an angular width $\Delta\theta_L \sim 0.36°$ at the target. Ions leave the target and enter the spectrometer in a distribution of charge states q centered around $\bar{q} \sim 22.5$.

After entering the magnetic field, IC decays of the 14.4-keV state may change the ionic charge by $+1$ or $+2$ depending on whether a K vacancy generated by the nuclear decay is filled with the emission of a K x ray or an Auger electron. After emerging from the field of the spectrometer the ions were detected in the focal plane with a position-sensitive gas detector which measured total energy, position, and time of flight.[6] Placing narrow windows on two-dimensional spectra of both energy versus position and time of flight versus position, one-dimensional spectra containing the charge-changing processes were produced.

Figure 1(a) shows a position spectrum at $\theta_L = 7.5°$, where the scattered ^{57}Fe ions have only a small fraction of nuclei in the excited state. A pattern of peaks is seen corresponding to ions which traverse the magnetic field without changing charge. The few events in between the main peaks in Fig. 1(a) are estimated to arise mainly from 14.4-keV level decays. Figure 1(b) shows a position spectrum at $\theta_L = 40°$. Between the main peaks are the events corresponding to $\Delta q = +1$ or $+2$ charge-changing processes after nuclear decay in the magnet. Near the main peaks are small subsidiary peaks which arise from charge changes in the nearly-field-free gap between the two poles of the spectrometer. In order to show that the events between the main peaks are caused by charge-changing processes, Monte Carlo calculations

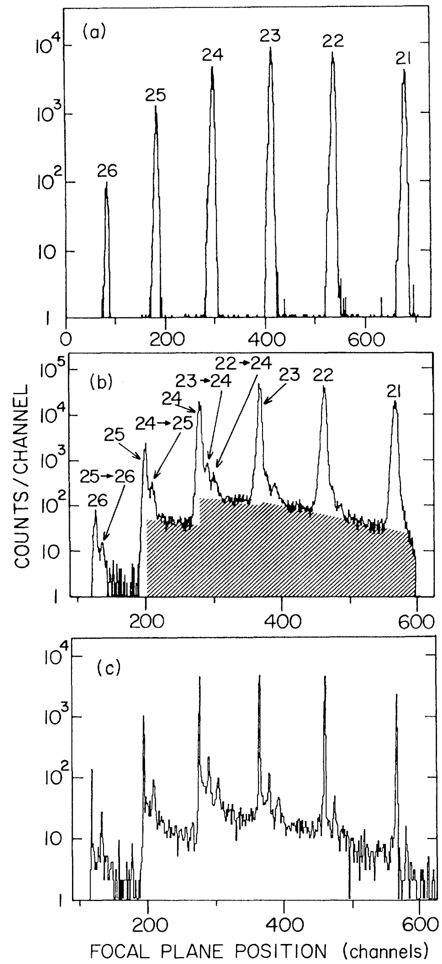

FIG. 1 (a) Position spectrum observed in the focal plane of the spectrometer for $\theta_L = 7.5°$ and a bombarding energy of 346 MeV. The strong peaks are labeled according to charge state q. (b) Position spectrum observed in the focal plane of the spectrometer for $\theta_L = 40°$. The events between the strong peaks (see the shaded area) are attributed to charge-changing processes after nuclear decay in the magnet. The small subsidiary peaks arise from charge changes (as labeled in the figure) in the nearly-field-free gap between the two poles of the spectrograph. (c) Monte Carlo simulation of a position spectrum using the ion-optical code RAYTRACE including charge-changing processes. See text for details.

with the ion-optical code RAYTRACE[7] were performed. The code was modified to include charge changes $\Delta q = +1$ or $+2$ within the magnetic field of the spectrograph. The results are shown in Fig. 1(c). In these calculations a (charge-state-independent) half-life of 98 ns was used together with fluorescent yields from Ref. 8 and a charge-state distribution as given in Ref. 9. The calculations describe the qualitative features of the structure in the measured energy spectra.

The angular distribution for the total yields in between the main peaks, assumed to arise from decays of excited ^{57}Fe ions, does follow that calculated for Coulomb excitation. Contributions from charge-exchange processes with the residual gas in the magnet are less than 10^{-5} of the elastic peaks as measured with ^{48}Ti and ^{58}Ni beams,[10] i.e., with nuclei which have no low-lying nuclear states.

The subsidiary peaks, since there is no ambiguity as to which initial charge q they came from, can be used to determine the average K-shell fluorescence yields in the electronic configurations existing in the gap for ions of charge q before K-vacancy production.[8] Here we concentrate on IC decay rates in $q=24$ and 25 ions. There is no subsidiary peak with $\Delta q = +2$ near the $q=25$ unchanged peak. This is evidence that the K shell in the $q=23$ ion is filled by the time the ions reach the magnet gap, thus prohibiting Auger emission from the two-electron ion produced after K-shell IC decay of the 14.4-keV state within the gap. All of the events between the unchanged $q=24$ and 25 peaks can therefore be attributed, with negligible error, to decays of the 14.4-keV level in ions initially with $q=24$, and all of the events between the $q=25$ and 26 peaks to ions with initial $q=25$. There are no long-lived electron states[11] in single-electron ^{57}Fe ions, and the only level in the two-electron ion with a lifetime longer than ~ 2 ns is the $(1s)^1(2s)^1\ {}^3S_1$. We assume in the following that a few ns after leaving the target the $q=24$ and 25 ions have the electrons in the $1s$ orbit.

At time $t=0$, just after leaving the gold foil, the number of ions in charge state q is N_q^0 and the fraction of nuclei in the 14.4-keV level in charge state q is $f_q^0 = f^0$, assumed to be independent of q. All ions take a time $t_1 \sim 18$ ns to reach the magnet entrance, at which time the number of ions in charge state q is N_q^1 and the fraction f_q^1 of nuclei in the excited state now depends on q by the following:

$$f_q^0 = f^0 N_q^0 \exp(-\lambda_q t_1)/N_q^1 , \tag{1}$$

and

$$N_q^1 = N_q^0 - f^0 N_q^0 R_q (\rho_{q,q+1} + \rho_{q,q+2}) + f^0 N_{q-1}^0 R_{q-1} \rho_{q-1,q} + f^0 N_{q-2}^0 R_{q-2} \rho_{q-2,q} , \tag{2}$$

where $R_q = 1 - \exp(-\lambda_q t_1)$ is the fraction of ions initially in charge state q which undergoes a nuclear decay on the way to the magnet entrance. λ_q is the total decay probability per unit time of the 14.4-keV state in ions of charge q. $\rho_{q,q+1}$ ($\rho_{q,q+2}$) is the fraction of ions in which the nucleus has decayed with initial charge q and final charge $q+1$ ($q+2$). For the range of q seen in these experiments

$$\rho_{q,q+1} = \frac{\lambda_{Kq}}{\lambda_q} \omega_{Kq} + \frac{\lambda_{Lq}}{\lambda_q} \omega_{Lq} , \qquad \rho_{q,q+2} = \frac{\lambda_{Kq}}{\lambda_q}(1 - \omega_{Kq}) , \tag{3}$$

with λ_{Kq} (λ_{Lq}) the transition probability per unit time for K- (L-) shell IC decay in ions of charge q, and ω_{Kq} (ω_{Lq}) the K- (L-) shell fluorescence yield for an ion, initially of charge q, in which a K (L) vacancy is produced. The parameters λ_q, $\rho_{q,q+1}$, etc. are averages over the electron configurations existing from $t=0$ to t_1. The total number of decays within the magnet for which ions with initial charge q increase their charge is given by

$$\Delta N_q = f_q^1 N_q^1 [1-\exp(-\lambda_q \Delta t_q)] \times (\rho_{q,q+1}+\rho_{q,q+2}), \quad (4)$$

where Δt_q is the time spent by an ion of charge q in the spectrometer, and the parameters λ_q, $\rho_{q,q+1}$, etc. are averages over the times $t=t_1$ to $t_1+\Delta t_q$. If we assume that the various parameters (λ_q,etc.) of Eqs. (1), (2), and (4) are the same then we may use Eq. (1) to rewrite (4) in the form

$$\Delta N_q = f^0 N_q^0 \exp(-\lambda_q t)[1-\exp(-\lambda_q \Delta t_q)] \times (\rho_{q,q+1}+\rho_{q,q+2}). \quad (5)$$

For the $q=25$ ion in the electronic ground state the K-shell IC decay rate of the 14.4-keV level depends very strongly on the total angular momentum F. The rate should be about 2 orders of magnitude larger[12] for $F=1$ than for $F=2$ (the spins possible from coupling the nuclear spin of $\frac{3}{2}$ to a single K electron). There are thus two different lifetimes for nuclear decay in the $q=25$ ion, and two sets of parameters λ_{25} and $\rho_{25,26}$. The modifications of the equations for $q=25$ consist of rewriting them in a straightforward way in terms of the two components of N_{25}. It is amusing to note that the transition probability for $F=1$ ions is actually larger than for $q=24$ ions since for a filled K shell the $F=1$ component contributes with a weight of $\frac{3}{8}$. Assuming a statistical population of atomic spins just after exit from the target, and neglecting IC in the $F=2$ ions, the equations can be solved for λ_{Kq} for both $q=24$ and 25.

Before discussing the analysis of the data using these equations, two matters need to be considered. On exit from the foil, a small fraction of the ^{57}Fe ions is in the 136.5-keV state[4] which has a half-life of 8.69(25) ns, and decays predominantly to the 14.4-keV level. The equations given above do not include this effect. The second matter is whether all of the events observed at $\theta_L=40°$ are ^{57}Fe ions. In order to estimate the fraction of recorded events due to quasielastic nucleon transfer processes, runs were taken at $\theta_L=50°$, 55°, 60°, and 65°, near the nuclear grazing angle. At these angles transfers are seen and can be recognized in the two-dimensional energy versus position and time of flight versus position spectra. The cross sections extrapolated forward to $\theta_L=40°$ show that the fraction of quasielastic events is less than 1% in the data used.

The fraction f^0 was estimated by using experimental values of $\sum \Delta N_q$ and $\sum N_q^0$ in Eq. (5) with a calculated average value for Δt_q and average values estimated from the neutral-atom data for λ_q and $\rho_{q,q+1}+\rho_{q,q+2}$. The data are dominated by $q=22$ and 23 for which RAYTRACE calculations give $\Delta t_{22}=62$ ns and $\Delta t_{23}=59$ ns, respectively. Spectra from two experimental runs with slightly different ionic charge-state distributions in the scattered beam (due to carbon buildup on the exit side of the gold target) were analyzed. The values of f^0 extracted for the two runs were 0.250(3) and 0.249(3). For $q=24$ and 25, spectra such as those shown in Fig. 1(b) gave ΔN_q and N_q^0 directly. These numbers were used with the above equations to obtain the values for λ_{K24} and λ_{K25} ($F=1$).

Table I presents the first results for IC rates and changes in lifetimes in one- and two-electron ions. Previous attempts[13] to observe IC rates in stripped ions have been restricted to mixtures of charge states in which only small fractions of electrons were removed. The present results are not yet accurate enough to probe changes in

TABLE I. Half-lives (in ns) and K-shell IC decay rates in one- and two-electron ^{57}Fe ions[a].

q	0	24	25 ($F=1$)	25 ($F=2$)
λ_{Kq} (10^6 s^{-1})	5.73(2)	6.16($\pm$0.27, $\pm$0.15)	8.03($\pm$0.54, $\pm$0.30)[b]	...
$T_{1/2}$(obs)[c]	98.1(3)	100.0($\pm$4.0, $\pm$2.3)	78.8($\pm$5.2, $\pm$2.9)	...
$T_{1/2}$(calc)[d]	98.1(3)	106.7(3)	82.4(4)	901(12)

[a] The errors quoted for the decay rates are statistical errors, followed by estimated systematic errors, primarily reflecting uncertainty in the assumptions made about decay processes of the ions on the way from target to magnet. The combined errors are quoted in the abstract.
[b] The K-shell IC decay rate for $F=1$ is estimated assuming statistical population of spin states in the total spin $F=1$ and 2 states for $q=25$.
[c] $T_{1/2}$(obs) for $q=24$ and 25 are calculated using the λ_{Kq} in row 2 with the constant value of $7.7(1)\times10^5$ s^{-1} for λ_γ.
[d] $T_{1/2}$(calc) are calculations from the neutral-atom decay rates with no modification assumed for K-shell electron wave functions.

K-shell wave functions compared to neutral atoms (expected[12] at the 2% level), but do show that no unforeseen large effects are influencing the IC process in few-electron ions. The ratio $\lambda_{K25}(F=1)/\lambda_{K24}$ is consistent with the simple estimate of $\frac{4}{3}$ made for constant K-shell wave functions, and also with the value deduced from calculations[14] of the one- and two-electron states. The type of measurement described here is capable of high accuracy. This class of experiments, of interest to both atomic and nuclear physics and with possible astrophysical implications, is likely to receive increased attention at the new generation of heavy-ion storage rings.[15] There, in addition to electromagnetic processes in highly charged ions, the study of modifications to weak nuclear decays is also likely to become feasible.

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