Nuclear Lifetimes in the Region of 10^{-16} sec Measured by a New Technique

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A new technique exploiting proton–x-ray coincidences has been used to measure 10^{-16}-sec lifetimes of proton-unstable levels in ^{69}As produced in the electron-capture decay of ^{69}Se. The results are compared with statistical-model calculations.

Measurements of the lifetimes of nuclear states have long provided a valuable method for scrutinizing theoretical models of the nucleus. Calculated transition matrix elements, and thus the predicted decay rates, are often characteristic of the assumptions underlying a particular class of model, so the systematic measurement of such lifetimes may make more than trivial comments on the general features of nuclear structure.

The conventional methods for measuring lifetimes directly—delayed-coincidence and Doppler-shift techniques—cover a wide range of times greater than about 10^{-15} sec.[1] But for shorter lifetimes, only the blocking technique, which has so far been restricted to a few favorable materials, is available for making direct measurements.

We wish to report the first direct determination of nuclear lifetimes in the 10^{-16}-sec region using a new technique that involves comparison of the decay time of a nuclear state with the filling time of a vacancy in the atomic K shell. Any nucleus (with atomic number Z) that decays by electron capture to excited states in the daughter $(Z-1)$ produces simultaneously a vacancy in an atomic shell. If those excited states are unstable to proton emission, then the energy of the x ray emitted with the filling of the atomic vacancy will depend upon whether the proton has already been emitted (in which case the x ray would be characteristic of a $Z-2$ element) or not (a $Z-1$ element). If the nuclear and atomic lifetimes are comparable, then the $K\alpha$ x rays observed in coincidence with protons will lie in two peaks whose relative intensities uniquely relate one lifetime with the other.

The lifetimes for the K-shell vacancies are reasonably well known both experimentally[2] and theoretically,[3] ranging from $\tau \sim 2\times10^{-15}$ sec for carbon down to $\tau \sim 6\times10^{-18}$ sec for uranium. Thus, the range of nuclear lifetimes available in principle through this technique abuts the region conventionally accessible and extends it downward by more than two orders of magnitude.

So far, data have been obtained relating to the lifetimes of levels in ^{65}Ga, ^{69}As, ^{73}Br, and ^{77}Rb, but in this report we shall concentrate on ^{69}As, for which the experimental results and analysis are most complete. The decay sequence under study was

$$^{69}_{34}\mathrm{Se}_{35} \xrightarrow{\mathrm{EC}(\beta^+)} {}^{69}_{33}\mathrm{As}^*_{36} \xrightarrow{p} {}^{68}_{32}\mathrm{Ge}_{36}\,.$$

The delayed proton precursor ^{69}Se was produced with the reaction ^{40}Ca(^{32}S, $2pn$)^{69}Se using a 100-MeV ^{32}S beam from the Chalk River upgraded MP tandem Van de Graaff. The 1.2-mg/cm^2 natural calcium target was mounted on a vertical shaft with its surface inclined at 15° with respect to the beam direction. Following a 40-sec bombardment, the beam was interrupted by magnetic deflection and the target lowered in ~2 sec from the irradiation position to the counting position 25 cm below, where three detector systems were arranged to observe the target in extremely close geometry.

A surface-barrier counter telescope for proton detection was mounted ~1.5 mm from the front of the target, a 200-mm^2 intrinsic Ge x-ray detector was placed with its entrance window ~5 mm from the back of the target, and an 86-cm^3 Ge(Li) γ-ray counter was ~2 cm away, immediately behind the telescope. The telescope subtended 2.5 sr and was composed of a 50 mm$^2\times$11 μm thick ΔE transmission counter and a 300 mm$^2\times$300 μm thick E counter. Its energy resolution and calibration were determined by use of an ^{241}Am α source and in a separate experiment in which the well-known[4] delayed proton groups from ^{25}Si were produced in the reaction ^{24}Mg(^{3}He, $2n$)^{25}Si at 33-MeV bombarding energy.

During the counting period, singles events for protons, x rays, and γ rays were routed separately into four sequential spectra for 15 sec each. In addition, data for p-x, p-γ, and x-γ coincidences were stored event by event on magnetic tape, for subsequent playback with selected gating conditions.

The spectrum of delayed protons observed following the decay of ^{69}Se is shown in Fig. 1(a) together with a simplified decay scheme. The half-life was measured to be 27.4 ± 0.2 sec. A complex scheme for the low-energy excited states of ^{69}As has also been derived from the β-delayed γ rays, and β-decay branching ratios for ^{69}Se were determined. The electron-capture-decay energy $Q_{ec} = 6795 \pm 60$ keV for ^{69}Se is the result of averaging two independent but concordant results, one a direct measurement (using a plastic scintillator) of the end point of positrons in coincidence with individual γ rays, the other an indirect determination from the measured ratio of electron capture to positron emission. The proton separation energy of ^{69}As, $B_p = 3395 \pm 50$ keV, follows from a β^+ end-point measurement for ^{69}As, combined with the known masses[5] of ^{68}Ge and ^{69}Ge. The details of these measurements, only peripherally related to the present discussion, will be presented in a future publication by the authors.

The spectrum of x rays recorded in coincidence with all protons is shown as the histogram in Fig. 2. The x-ray "standard" peak shapes and energies for each relevant element were established using coincidences with specific known γ rays recorded at the same time. These x-ray peaks, which are associated either with electron capture or internal conversion, have much better statistics than the p-x data and are plotted, renormalized, as smooth curves in Fig. 2. Utilizing the standard curves, the x-ray peak-intensity ratio was determined as a function of the coincident proton energy and the results are plotted in Fig. 1(b). On the scale at the right of the figure is the average lifetime that would correspond to the measured ratio if all observed protons fed the same final state in ^{68}Ge and originated from states of one spin only in ^{69}As. In practice, these lifetimes must be regarded as averages over all states with spins that allow them to be populated in the β decay of ^{69}Se.

The proton energy spectrum, $I_p(E_p)$, is determined by two factors: (i) the intensity of β-decay branches (including electron capture) from ^{69}Se to ^{69}As, denoted $I_\beta(E_\beta)$; and (ii) the branching

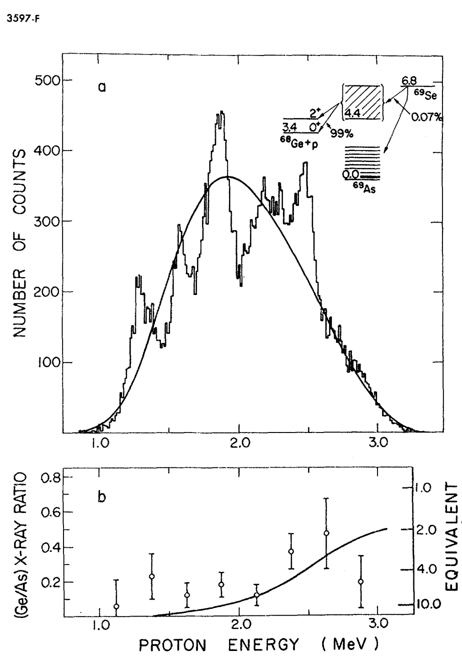

FIG. 1. (a) Spectrum of protons observed following the decay of ^{69}Se; the experimental resolution (full width at half-maximum) was ~ 90 keV. A simplified decay scheme is also shown; energies are given in MeV relative to the ^{69}As ground state. (b) Ratio of Ge x rays (measured in coincidence with protons) relative to those from As, plotted as a function of coincident proton energy. The smooth curves in (a) and (b) are the results of calculations described in the text.

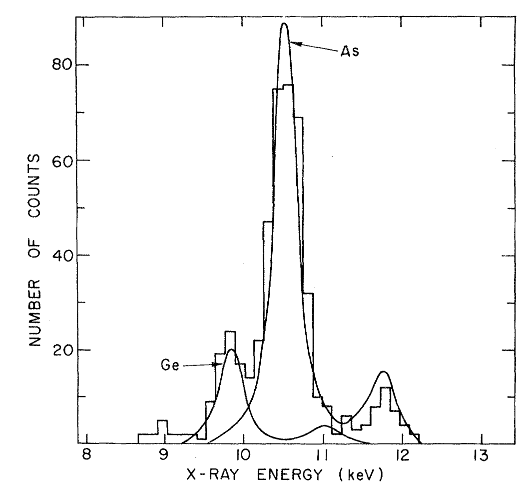

FIG. 2. The histogram gives the spectrum of x rays observed in coincidence with all delayed protons. The smooth curves are x rays measured simultaneously, with the same detector, in coincidence with specific known γ rays; they are normalized in height only to fit the histogram.

ratios for subsequent particle emission to states in ^{68}Ge. Thus[4]

$$I_p(E_p) \propto \sum_{if} \frac{\Gamma_p^{if}}{\Gamma^i} I_\beta(E_\beta), \qquad (1)$$

where $\Gamma^i = \sum_f \Gamma_p^{if} + \Gamma_\gamma^i$. Here Γ_p^{if} is the partial width for proton emission from state i in ^{69}As to state f in ^{68}Ge, Γ_γ^i is the γ-decay width of state i, and the sum extends over all pairs of states i and f between which protons of energy E_p can be emitted. Since the values of Γ_p^{if} and I_β both scatter with an assumed Porter-Thomas distribution, it is actually the average value $\langle I_p(E_p)\rangle$ that must be calculated from Eq. (1).

Qualitatively, though, the proton spectrum of Fig. 1(a) may easily be understood from Eq. (1). The low-energy part of the spectrum reflects the increasing magnitude of Γ_p relative to Γ_γ while at higher energies, where $\Gamma_p \gg \Gamma_\gamma$, it is the β-decay properties that predominate. Thus, while the relative magnitudes of Γ_p and Γ_γ can be inferred experimentally from the proton spectrum, the measurement of the x-ray ratio [Fig. 1(b)] is essential to a determination of the absolute widths.

Detailed calculations of the delayed-proton-decay properties have been made. The β intensity I_β was derived assuming allowed decay and a Gaussian strength function.[6] The level density ρ in ^{69}As was calculated using the formulas of Gilbert and Cameron[7] who combined a Fermi-gas expression at high excitations with a constant temperature representation at low. The partial γ-decay widths Γ_γ were calculated assuming $E1$ radiation with a Lorentzian strength function,[8] while the proton widths were derived from the formula

$$\Gamma_p = T(2\pi\rho)^{-1}, \qquad (2)$$

where T is the total optical-model transmission coefficient for protons.[9] The properties calculated were the spectrum shape, the total β branching ratio to proton-emitting states in ^{69}As, the proton-branching ratio to states in ^{68}Ge, and the coincident x-ray ratios. The latter were calculated by using $\tau = 2.9\times10^{-16}$ sec as the K-vacancy lifetime[2,3] and taking account of the nonexponential decay of a group of states with Porter-Thomas distributed widths.

An examination of the spins of neighboring odd-mass nuclei indicates $\frac{1}{2}^-$, $\frac{3}{2}^-$, or $\frac{5}{2}^-$ to be the most probable spin for ^{69}Se, and indeed calculations performed with all other parameters fixed at the values used in the cited references yielded reasonable results for all three spin values, while higher spins produced order-of-magnitude discrepancies with the measured results. This spin preference is also consistent with γ-ray results.[10] The calculations were then repeated for the lower spins, requiring detailed agreement with the measured lifetimes (i.e., x-ray ratios) and the low-energy portion of the proton spectrum. To do this, two parameters were allowed to vary: the level-density parameter[7] a and the absolute magnitude of Γ_γ (but not its energy dependence). Best results for the energy spectrum and lifetimes are shown in Fig. 1 (all three spins yielded similar curves) with the corresponding branching ratios and parameters listed in Table I. There appears to be a slight preference for $J^\pi = \frac{3}{2}^-$, but all results are probably within the expected accuracy of such statistical model calculations.

The level density (and thus parameter a) is related through Eq. (2) to the measured level lifetimes, $\tau(=\hbar/\Gamma)$. For comparison purposes, then, it is most convenient to regard the value of a as an embodiment of the lifetime measurements

TABLE I. Some properties of the ^{69}Se decay.

Evaluated[b] assuming ^{69}Se spin of	B_p[a] (%)	B_{p_1}/B_p[a]	Derived quantities a (MeV^{-1})	Γ_γ (eV)
$\frac{1}{2}^-$	0.11	0.006	11.7	0.25
$\frac{3}{2}^-$	0.06	0.013	11.7	0.25
$\frac{5}{2}^-$	0.03	0.038	11.2	0.25
Measured	0.07 ± 0.01	0.14 ± 0.007		

[a]The branching ratios (B) are given for all proton emission (p) and for population of the ^{68}Ge first excited state (p_1).

[b]The calculated results correspond to the fitted curves shown in Figs. 1(a) and 1(b).

even though it does depend to some extent on the assumed spin for ^{69}Se. Our derived values of a for ^{69}As compare favorably with the prescriptions of Ref. 7 and with results similarly derived from blocking measurements[11] on ^{71}As and ^{73}As: in all cases $a \sim 11$ MeV^{-1}. By comparison, the width Γ_γ, quoted in Table I for an excitation energy of 4.75 MeV (the region in which the proton spectrum is most sensitive to Γ_γ), is higher by a factor of 5 than the simple $E1$ predictions, but such a value is still consistent with known γ widths in the same mass region.

Evidently, the calculated energy dependence of the proton spectrum and level lifetimes is in reasonable agreement with the average behavior of the data (Fig. 1) although there may be an indication of significant upward fluctuations from the average at low energy particularly in the lifetime data. This suggests strongly favored proton decay in this region, a possibility that needs to be studied in future experiments with improved counting statistics. It is such features of this and other proton-emitting nuclei that are now made accessible through the new p-x technique of lifetime measurement.

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Intracavity Raman Scattering from Molecular Beams: Direct Determination of Local Properties in an Expanding Jet Beam

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An intracavity crossed laser-beam, molecular-jet-beam system has been developed for studying the properties of expanding jet beams by means of light scattering. Raman scattering has been used to measure the local rotational temperature and absolute density of monomers in a jet beam of CO_2 as a function of axial distance from the nozzle. A well-defined rotational temperature has been observed. This temperature drops rapidly for distances of a few nozzle diameters in agreement with isentropic theory and thereafter remains roughly constant.

We have developed a crossed laser-beam, molecular-jet-beam system for studies of the latter by means of light scattering. The jet beam intersects the sharply focused laser beam within the laser cavity providing a high-sensitivity, high-resolution system which does not disturb beam properties. The rotational and vibrational temperatures and absolute monomer density of a molecular jet beam can be measured directly as a function of position relative to the nozzle with an axial resolution of $\simeq 10$ μm. In addition the method is potentially capable of observing the