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Hindi, M.M. Sur, B. Wedding, K.L. et al.

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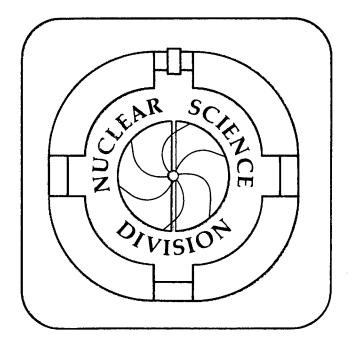


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December 1992



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β+ Decay and Cosmic-Ray Half-Life of ⁹¹Nb

M.M. Hindi, (1), Bhaskar Sur, (2,3), Kristin L. Wedding, (2)†, D.W. Bardayan (1), K.R. Czerwinski, (2)‡, M.T.F. da Cruz, (2)§, D.C. Hoffman, (2), R.-M. Larimer (2), K.T. Lesko, (2), and E.B. Norman, (2)

(1)Physics Department, Tennessee Technological University, Cookeville, TN 38505 (2)Nuclear Science Division, Lawrence Berkeley Laboratory, Berkeley, CA 94720 (3) Physics Department, Queens University, Kingston, Ontario, Canada K7L 9N6

Nuclear Science Division, Lawrence Berkeley Laboratory University of California, Berkeley, California 94720, USA

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β^+ decay and cosmic-ray half-life of ⁹¹Nb

M. M. Hindi,⁽¹⁾ Bhaskar Sur,^{(2,3)*} Kristin L. Wedding,^{(2)†} D. W. Bardayan,⁽¹⁾ K. R. Czerwinski,^{(2)‡} M. T. F. da Cruz,^{(2)§} D. C. Hoffman,⁽²⁾ R.-M. Larimer,⁽²⁾ K. T. Lesko,⁽²⁾ and Eric B. Norman,⁽²⁾

(1) Physics Department, Tennessee Technological University, Cookeville, TN 38505

(2) Nuclear Science Division, Lawrence Berkeley Laboratory, Berkeley, California 94720

(3) Physics Department, Queen's University, Kingston, Ontario, Canada K7L 9N6
(January 8, 1993)

Abstract

In the laboratory, ⁹¹Nb decays by electron capture (EC) with a 680-year half-life. However, as a high energy cosmic ray, it would be stripped of its atomic electrons and would be able to undergo only β^+ decay. We produced and chemically purified a sample of ⁹¹Nb and observed its decay with an array of Ge and NaI detectors. By following the β^+ annihilation radiation, we were able to determine the β^+ branching ratios of both the 105-keV, 61-day isomer and the ground state of ⁹¹Nb. The ground state branch is $(7.7\pm0.8)\times10^{-3}\%$, leading to a β^+ partial half-life (hence a cosmic-ray half-life) of $(8.8\pm1.9)\times10^6$ years. Such a value of the half-life makes ⁹¹Nb a good candidate for determining the confinement time of this secondary component of the cosmic rays.

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I. INTRODUCTION

In the laboratory, ⁹¹Nb decays by electron capture (EC) with a 680-year half-life [1] (Fig. 1). However, as a high energy cosmic ray, it would be stripped of its atomic electrons and would be able to undergo only (the energetically allowed, but previously unobserved) β^+ decay. ⁹¹Nb can be produced in the cosmic rays through spallation of Mo and heavier elements by interstellar hydrogen [2]. Hence the cosmic-ray half-life of ⁹¹Nb depends on its β^+ decay rate. If that half-life turns out to be long (\sim millions of years) then ⁹¹Nb could serve as another cosmic-ray chronometer and as a probe of models of the interstellar medium and of the propagation of this secondary component of cosmic-rays. Since ⁹¹Nb decays by a second-forbidden non-unique transition, the EC-to- β^+ ratio is nuclear-structure dependent [3] and the β^+ partial half-life cannot be deduced in a simple way from the known EC half-life. Therefore a direct measurement of the β^+ branching ratio is required for a reliable determination of the partial half-life.

The β^+ decay rate of ⁹¹Nb has relevance for cosmic-ray clocks in an indirect way, as well. Drach and Salamon [4] had suggested the use of elemental Tc as a cosmic-ray clock. They used the average value of log ft for all well-established second-forbidden non-unique transitions as their best guess for the ft value for the β^+ decay of ⁹⁵Tc. Since the relevant β^+ transitions in ⁹¹Nb and ⁹⁵Tc are very similar (both are $\frac{9}{2}^+ \to \frac{5}{2}^+$ second forbidden non-unique transitions involving primarily the transformation of a $1g_{9/2}$ proton to a $2d_{5/2}$ neutron [5]), there ought to be a close connection between the ft values of the two transitions. Hence knowledge of the ft value for the β^+ decay of ⁹¹Nb should help in constraining $t_{1/2}(\beta^+)$ for ⁹⁵Tc. Motivated by the above considerations, we conducted an experiment to measure the β^+ decay branch of ⁹¹Nb.

II. EXPERIMENTAL PROCEDURE

A. Source preparation

The ⁹¹Nb source was produced via the ⁸⁹Y(α ,2n)⁹¹Nb reaction. A natural yttrium foil, 0.25-mm thick, was bombarded for 17.5 hours by a 4.5 μ A beam of 35 MeV α particles from Lawrence Berkeley Laboratory's 88-Inch Cyclotron. The above bombarding energy was specifically chosen to maximize the yield of the ⁹¹Nb ground state relative to the lower spin ^{91m}Nb isomer.

Initial examination of the irradiated foil showed several impurities; therefore, a radio-chemical separation of the niobium was performed using aqueous-organic phase separation followed by an ion-exchange separation. The beam spot was cut out of the target and dissolved in 5 ml of 12 M HCl. After the yttrium completely dissolved, 10 ml of 1 M tri-isooctylamine (TIOA) in benzene was added to the solution, allowed to mix for 5 minutes and then centrifuged for 1 minute. This separated the organic phase (containing the Nb) from the aqueous phase (in which the Y remained). In addition to the Nb, the organic phase contained a ⁶⁵Zn impurity. The ⁶⁵Zn was removed from the organic solution by mixing it with 6 M HF in a 1:3 ratio and centrifuging the mixture. The ⁶⁵Zn extracted into the aqueous phase and the Nb remained in the organic phase. Further examination of the organic phase showed the presence of a ¹⁸³Re impurity. This was removed by adding 2 M HCl to the 1 M TIOA in a 1:3 organic:aqueous ratio and centrifuging the mixture. The Nb extracted into the aqueous phase, while the ¹⁸³Re remained in the organic phase. Another volume of 2 M HCl was added to the aqueous solution and the separation procedure repeated two more times.

The final purification of the Nb solution was performed using a column separation with 1:1 TIOA on Matrex as the resin. After rinsing the column with 15 ml of 6 M HCl to remove any residual Zr or Y, the Nb was removed from the column with 10 ml of 2 M HCl. The Nb solution was evaporated on a piece of cellophane tape which was then sealed with a similar

piece to form a suitable counting sample. The measurements to extract the activity of the source commenced 210 days after the source bombardment. The dominant activities at the start of these measurements were 156 nCi of 91 mNb and 5.6 nCi of 91 Nb, corresponding to 4.38×10^{10} and 6.4×10^{12} nuclei, respectively. There was no evidence for 92 mNb, which has a β^+ branch of 0.06% and a half-life of 10.15 d [6].

Two types of measurements were conducted on the source: one to measure its positron decay rate and the other to measure its K-electron capture rate. The details of these measurements are given below.

B. Positron decay rate measurements

The β^+ activity of the source was determined by measuring the coincidence count rate of the back-to-back 511-keV annihilation photons. A schematic view of the detector arrangement is shown in Fig. 2. The coincidences were recorded using two intrinsic Ge (Gamma-X) detectors placed face to face, with the source sandwiched in between. Each detector had a 52-mm diameter, 57-mm depth and a 3-mm window-to-endcap distance. Both detectors had 0.5-mm-thick Be windows. The Ge detectors were positioned at the center of an 8.25-cm hole in a 30-cm \times 30-cm NaI detector. The NaI detector served as an anti-Compton and anti-coincidence shield.

A standard circuit was used to generate coincidence signals. The energy and fast timing signals from each detector were digitized and recorded event by event on magnetic tape for later analysis.

In addition to the sought-after positron decay of 91 Nb, positrons could result from the β^+ decay of the 91m Nb isomer and from pair production by the 1205-keV photon, which is produced when the isomer decays to the 1205-keV level of 91 Zr (Fig. 1). To determine the contribution of the 91m Nb isomer to the measured 511-511 coincidence rate, we repeated our measurements four times over a period of 84 days (1.38 isomer half-lives). The durations of the measurements were 2.70, 3.06, 1.73, and 4.87 days. The background activity was

measured after each run by removing the source and collecting data for a few days using the same setup as in the niobium runs. Figure 3(a) shows a background-subtracted singles spectrum and figure 3(b) shows a 511-keV-gated spectrum.

Since the isomer gave the dominant contribution to the initial rates, we decided to perform another set of measurements 250 days later (4 isomer half-lives) at which point the contribution of the isomer was expected to be small. This measurement, and one conducted shortly thereafter, gave results that are about one standard deviation lower than the first measurement. To ascertain that the difference between the first measurement and those conducted 8 months later is simply statistical, and not due to a real decrease in the β^+ rate (which would possibly indicate that the 680-year ⁹¹Nb is not the dominant source of the observed positrons), we conducted another set of measurements 587 days after the first set. One of the above four sets of measurements was conducted at Tennessee Technological University (TTU), and the other three were conducted at the Lawrence Berkeley Laboratory (LBL). For the measurement at TTU a NaI shield was not used and smaller Ge detectors were employed. For one of the LBL measurements a 7.6 cm \times 15.2 cm NaI detector replaced one of the Ge detectors.

The efficiency of the systems for detecting back-to-back 511-keV photons was measured using calibrated sources of ⁶⁵Zn and ²²Na. Due to the close geometry of our setups, the ²²Na measurements are complicated by the substantial summing effects. Therefore, in addition to the efficiency measurements that rely on the absolute strength of the source, we conducted measurements of the efficiency using the ²²Na source by comparing the singles rate of the 1275-keV photon detected in a third detector, positioned far enough from the source for summing effects to be negligible, with the rate of 1275-keV photons detected in the third detector in coincidence with the two back-to-back 511-keV photons registered in the Ge detectors. The efficiency for detecting 511-511 photopeak coincidences ranged from 1% to 4.3%, for the various sets of measurements.

C. Electron capture rate measurements

The EC decay rate of the source was determined by measuring the yield of Zr x rays. A setup similar to the one used for determining the 511-511 coincidence rate was employed, with one of the coaxial Ge detectors replaced by a planar Ge detector with an area of 1000 mm² and thickness 13 mm, and a 0.1-mm-thick Be window.

Since $Zr \times rays$ can also result from EC decay of the isomer (Fig. 1), the $Zr \times ray$ count rate was measured three times over a period of 34 days to determine the fraction arising from the decay of the isomer. The durations of the measurements were 3.71, 4.59, and 1.51 days. After each measurement the source was removed and the background measured using the same setup for approximately one day. The x-ray count rate was also measured after some of the 511-511 measurements mentioned above. Figure 4(a) shows the x-ray region of the Nb source spectrum for the first measurement (t = 0), and figure 4(b) shows the same region for a measurement conducted 587 days later. The figure shows the decrease of the yield of Nb x rays, which result from the internal conversion of the 61-day 105-keV isomer, compared to the $Zr \times rays$, which result mostly from the EC decay of the 680-year ⁹¹Nb ground state.

The absolute efficiency of the x ray detector for Zr x rays was determined by comparing the number of coincidences between the 1205-keV γ ray and Zr K x rays to the number of singles 1205-keV γ rays observed in the Gamma-X detector. The fraction of K captures was taken to be 0.848 [3].

We also measured the absolute intensity of the x rays and the 105-keV and 1205-keV γ rays arising from the decay of the isomer in separate singles runs with the source at a distance of \approx 15 cm from the detector. For these singles runs the efficiency of the detector was obtained using calibrated 57 Co, 109 Cd, 133 Ba, and 60 Co sources.

III. DATA ANALYSIS AND RESULTS

A. Positron decay rate

To our knowledge, there are no long-lived isotopes that undergo only ground state-to-ground state β^+ decay. Therefore, contaminants in the source can be identified by their characteristic deexcitation γ rays. The spectra were searched for γ rays from all known β^+ emitters. Small amounts of ²²Na (0.85 ± 0.09 pCi at t=0), and ¹⁵²Eu (2.1 ± 0.2 pCi at t=0) were found. (The contribution of these has been taken into account in the results we report). The upper limit on the contribution of all other β^+ emitters is 5% of the final ⁹¹Nb β^+ decay rate. In addition to the positron emitting contaminants mentioned above, small amounts of ⁹⁵Nb, and ¹⁷⁵Hf were observed in the first set of measurements [Fig. 3(a)].

The positron decay rates of the ⁹¹Nb ground state and the ^{91m}Nb isomer were deduced from the measured 511-511 coincidence rates and the measured efficiency for 511-511 coincidences. The contribution of the isomer to the measured β^+ rate was determined by fitting the first four 511-511 coincidence rate measurements with the sum of two exponentials, one with the lifetime of the ground state $(t_{1/2} = 680 \pm 130 \text{ y})$ [1] and the other with the lifetime of the isomer $(t_{1/2} = 60.86 \pm 0.22 \text{ d})$ [7]. (The small [$\approx 0.5\%$] increase in the number of ⁹¹Nb nuclei due to isomer decay was ignored.) Figure 5 shows the 511-511 coincidence rate, the resulting fit, and the contributions of the isomer and the ground state to the fit. Table I shows the resulting ground state β^+ decay rate for this set of measurements (t = 0), and for the later measurements.

As mentioned earlier, the isomer contributes positrons both through its β^+ -decay to the Zr ground state and through pair production by the 1205-keV γ ray following its EC decay to the 1205-keV state. The number of 511-511 coincidences arising from pair production by the 1205-keV γ ray was obtained from the number of 511 \otimes (511 + K) sum coincidences, where K denotes the K x ray that follows EC to the excited state. Explicitly, the number of 511-511 coincidences, $N(511 \otimes 511)$, due to pair production (e^+e^-) by the 1205-keV state

is related to the number of isomer decays N_m by

$$N(511 \otimes 511) = N_{m} BR_{1205} f_{e^+e^-} \epsilon_{511-511}, \tag{1}$$

where BR_{1205} is the isomer decay branch to the 1205-keV state, $f_{e^+e^-}$ is the pair production probability of the 1205-keV γ ray, and $\epsilon_{511-511}$ is the efficiency for detecting the 511-511 coincidences. The number of sum coincidences $N[(511) \otimes (511 + K)]$, where the K x ray following K capture to the 1205-keV state is summed with one of the 511's in the same detector, is given by

$$N[(511) \otimes (511 + K)] = N_m BR_{1205} f_{e^+e^-} \epsilon_{511-511} P_K \omega_K \epsilon_K.$$
 (2)

In the above expression P_K is the fraction of captures from the K shell, ω_K is the fluorescence yield of the Zr K hole, and ϵ_K is the efficiency of the detector for K x rays. From Eqs. (1) and (2) one can then obtain the number of 511-511 coincidences due to pair production by the 1205-keV γ ray as

$$N(511 \otimes 511) = \frac{N[(511) \otimes (511 + K)]}{(P_K \omega_K \epsilon_K)}.$$
 (3)

The denominator was obtained from the ratio of the 1205 + K sum peak to the total yield of 1205-keV γ rays in the singles spectrum (Fig. 3):

$$N(1205 + K) = N(1205) (P_K \omega_K \epsilon_K).$$
 (4)

From the above equations we find that 511-511 coincidences due to pair production by the 1205-keV γ ray account for $(6\pm2)\%$ of the 511-511 coincidences attributed to the isomer, with the remaining $(94\pm2)\%$ then being due to the β^+ decay of the isomer to the Zr ground state.

B. Electron capture rates

The EC decay rates of the Nb isomer and ground state were deduced from the measured yield of the Zr $K\alpha$ x rays. To extract the contribution of the isomer to the measured Zr

x-ray yield we again fitted the yield of Zr x rays as a function of time with two exponentials, one with the lifetime of the ^{91m}Nb isomer and the other with the lifetime of the ground state.

This analysis gave us the absolute K capture decay rates of both the ground state and the isomer. To determine the EC decay rate of the Nb ground state we also need to know P_K , the fraction of K capture decays. For the accuracies desired for this experiment it was sufficient to rely on the theoretically calculated value of $P_K = 0.871$ (Ref. [3]). Table I summarizes our results for the decay of 91 Nb.

For the isomer, EC decay can occur both to the Zr ground state and to the Zr first excited state at 1205 keV (Fig. 1). We determined the fraction of x rays contributed by decay to the excited state from the number of Nb x rays observed in coincidence with the 1205-keV γ detected in the NaI shield; the shield efficiency for detecting a 1205-keV signal was determined to be 0.87 ± 0.05 from calibrations using a 60 Co source.

To determine the total EC decay rates of the isomer we also need to know P_K for each branch. We again estimated these from the tables of Ref. [3] to be 0.848 for decay to the 1205-keV state and 0.868 for decay to the ground state.

The decay of the isomer is dominated by its internal conversion (IC) branch. Our measurement of the intensities of 105-keV γ rays and of Nb K x rays allows us to determine only the K-shell IC rate. To determine absolute decay branches for the isomer we need the total IC decay rate. Since our measurement for the K-shell internal conversion coefficient, $\alpha(K) = 118 \pm 4$, agrees with the value 115 ± 5 given by Ref. [7], we make use of their value of $\alpha(K)/\alpha(L+\cdots) = 2.1 \pm 0.1$ to obtain our total IC rate. With the total IC decay rate thus determined, we convert our measured decay rates to decay branches. The results are given in Table II

C. Results and Discussion

From the measurements of the positron and EC decay rates we deduce a β^+ branch of $(7.7 \pm 0.8) \times 10^{-3}\%$ for ⁹¹Nb. When combined with the 680 ± 130 year half-life [1], this

gives a β^+ partial half-life of $(8.8 \pm 1.9) \times 10^6$ years and a corresponding log ft value of 11.9, where the f value was obtained from the tables of Ref. [8] using an allowed shape. We note here that for a stripped ⁹¹Nb there are two small and somewhat offsetting corrections to the measured laboratory partial half-life: first there is an 11% increase in the β^+ half-life for the stripped ⁹¹Nb ion due to the lack of screening by the atomic electrons. Second there is a 7% decrease in the half-life because the Q-value for the stripped-ion decay is increased by 3.6 keV (the difference in the binding energies for the electrons in Nb and Zr) compared to the laboratory Q-value. The net result is a 4% increase in the (rest frame) β^+ half-life for cosmic-ray ⁹¹Nb compared to the measured laboratory partial half-life.

The resulting β^+ half-life is comparable to current estimates of the confinement time of cosmic rays [2]. However, several problems must be overcome before ⁹¹Nb can be used as a cosmic-ray chronometer. First of all, in the cosmic rays there will be present three niobium isotopes: ^{91,92,93}Nb. ⁹³Nb is stable, is produced in stars via the s- and r-neutron capture processes, and will be injected into the cosmic rays. ^{91,92}Nb are expected to be present in the cosmic rays only as products of spallation reactions of molybdenum and heavier elements on interstellar hydrogen. ⁹²Nb is an electron-capture-only nuclide which will become essentially stable as a bare cosmic-ray nucleus. As we have shown, cosmic-ray ⁹¹Nb will have a half-life of approximately 9×10^6 years. Thus, in order to determine the age of these medium weight cosmic-ray nuclei, one will need to measure: 1) the isotopic composition of cosmic-ray niobium, and 2) the relative ⁹¹Nb and ⁹²Nb production cross sections from proton-induced spallation reactions on abundant heavier elements. Efforts in this second area are now underway in our (LBL) laboratory.

From our measured branch for the β^+ decay of the ^{91m}Nb isomer we get $\log ft = 9.45$, where the f value was obtained from the tables of Ref. [8]. This $\log ft$ value is equal to that of the analogous $\frac{1}{2}^- \to \frac{5}{2}^+ \beta^+$ decay of the ^{95m}Tc isomer. If we assume then that the $\log ft$ value for the β^+ decay of the ⁹⁵Tc ground state is also equal to that of the ⁹¹Nb ground state, we get $t_{1/2}(\beta^+) = 8 \times 10^4$ y, which is somewhat smaller than the best guess estimate of 2×10^5 y made by Ref. [4], but still within their best guess range of 10^4 –4 × 10^6 y.

ACKNOWLEDGMENTS

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- * Present address: AECL Research, Chalk River Laboratories, Chalk River, Ontario K0J 1J0, Canada.
- [†] Present address: Department of Medical Physics, University of Wisconsin, Madison, WI 53706.
- [‡] Present address: Institut für Radiochemie, Technische Universitat München, D-8046 Garching, FRG.
- § Permenant address: Physics Institute, University of São Paulo, São Paulo, Brazil.
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TABLES

TABLE I. Decay rates of the 91Nb ground state.

Time of measurement (days)	Total rate	β^+ rate (10^{-2} Bq)
	(Bq)	
0	270 ± 30	2.2 ± 0.6
250	_	1.4 ± 0.5
302	200 ± 11	1.8 ± 0.4
587	203 ± 10	1.5 ± 0.2
Weighted average:	206 ± 7	1.59 ± 0.16
β^+ branch:	$(7.7 \pm 0.8) \times 10^{-3}\%$	

TABLE II. Decay of the 91m Nb isomer.

Decay mode	Decay branch (%) 1.92 ± 0.08
Electron capture to ⁹¹ Zr(1205 keV)	
Electron capture to ⁹¹ Zr(ground state)	1.3 ± 0.4
β^+ to $^{91}{ m Zr}({ m ground state})$	$(2.4 \pm 0.5) \times 10^{-3}$
Radiative decay to ⁹¹ Nb(ground state)	0.55 ± 0.03
Internal conversion decay to ⁹¹ Nb(ground state)	96.2 ± 0.4

FIGURES

- FIG. 1. Decay schemes of ⁹¹Nb and ^{91m}Nb. Level energies are in keV.
- FIG. 2. Schematic view of the experimental setup for detecting back-to-back 511-keV photons.
- FIG. 3. (a) Background-subtracted singles spectrum of the 91,91m Nb source. The spectrum is shifted upwards by 100 counts for display purposes. (b) Compton-suppressed coincidence spectrum gated by the 511-keV line. Both spectra are from the first set of measurements (t=0).
- FIG. 4. (a) The x ray region of the spectrum of the 91,91m Nb source, recorded using a planar Ge x ray detector at t = 0. (b) The same region as in (a), recorded 587 days later. The counts in both spectra are raw counts, i.e., they have not been corrected for differences in efficiency and collection time.
- FIG. 5. The 511-511 coincidence counting rate as a function of time. The solid curve is a fit using the sum of two decay curves, one with the half-life of the isomer (61 d, dash-two-dotted curve), and one with the half-life of the ground state, (680 y, dashed curve).

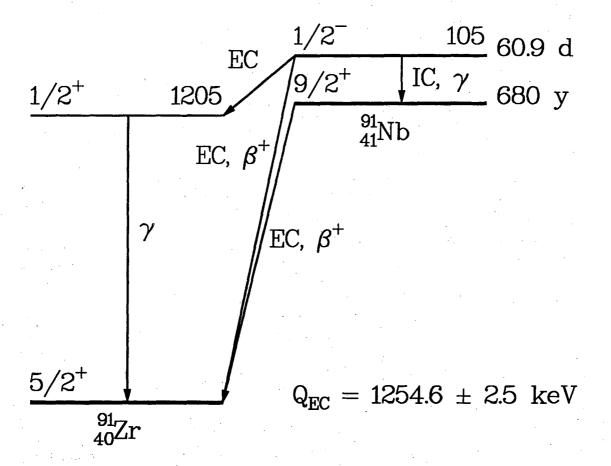


Figure 1

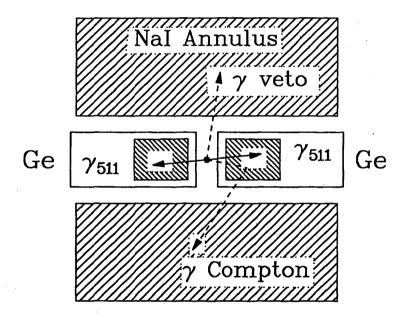


Figure 2

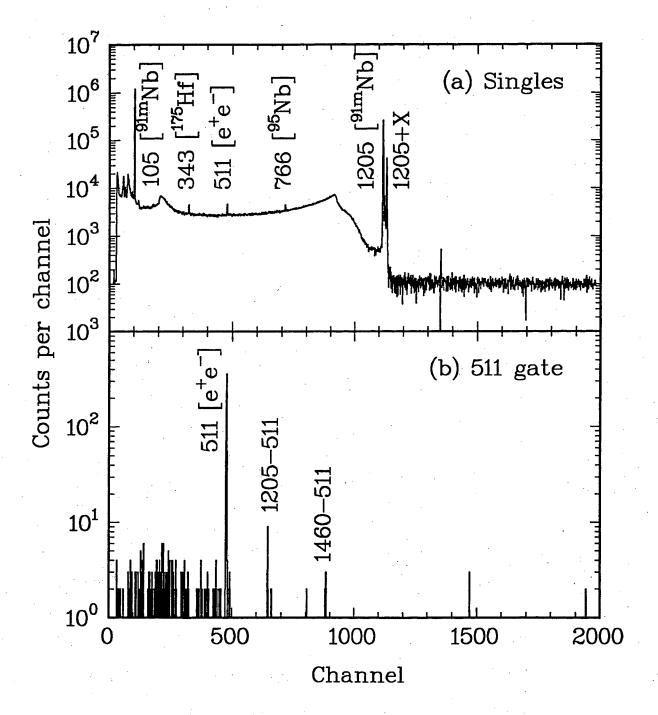


Figure 3

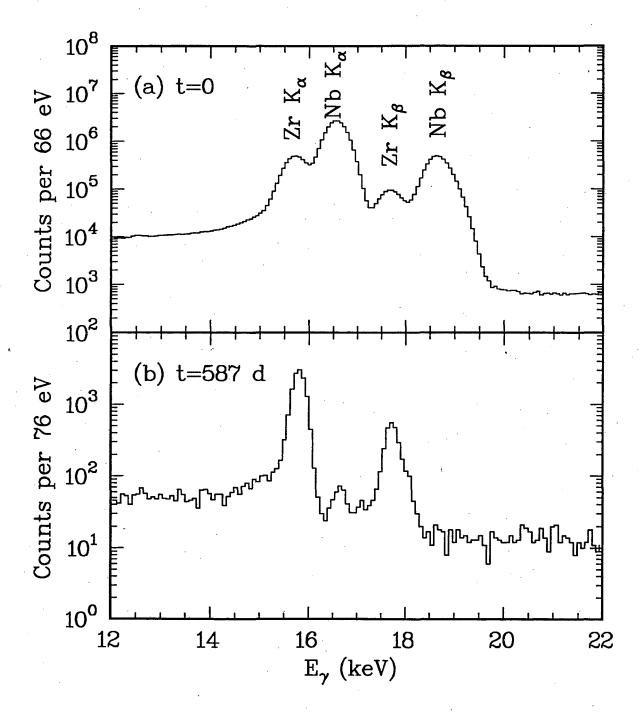


Figure 4

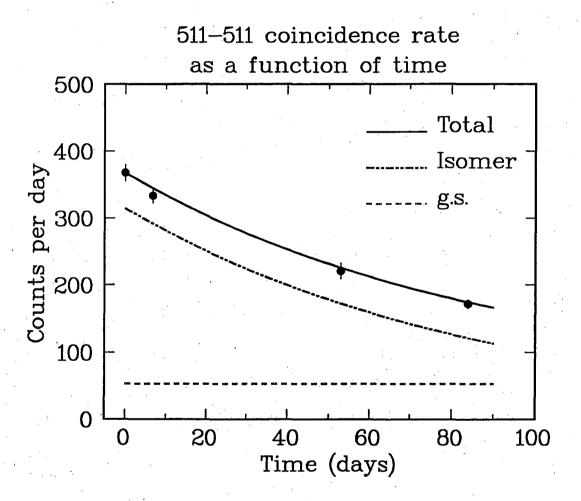


Figure 5

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