

Half-Life of Molecular Tritium and the Axial-Vector Interaction in Tritium β Decay

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The half-life of tritium has been measured by monitoring the decay of the x-ray bremsstrahlung emitted by gaseous mixtures of $^3\text{H}_2$ and Xe. The result, 12.31 ± 0.03 yr, when combined with the end-point energy measured on the same sources, yields the product $(G_A/G_V)\langle\sigma\rangle = 2.098 \pm 0.003$, where $\langle\sigma\rangle$ is the Gamow-Teller matrix element. Combined with a recent precise calculation of the matrix element, this value suggests that the renormalization of the axial-vector interaction can be entirely accounted for by nuclear effects in the triton. The deduced value of G_A/G_V constrains the neutron lifetime to lie in the range of 897 ± 3 s.

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The axial-vector interaction in tritium β decay is smaller than the corresponding interaction in neutron decay. One interpretation of this intriguing result is that the strength of the interaction is reduced in nuclear matter, i.e., the free-nucleon G_A value is quenched [1,2]. An alternative possibility is that the matrix element between initial and final nuclear states is smaller because of the presence of Δ isobars and meson exchange currents in the triton [3]. To distinguish between these two alternatives, several conditions must be met. The measured quantities relevant to the decay, namely, the end-point energy W_0 and the half-life $t_{1/2}$, must both be determined with a precision of a few tenths of a percent. This is facilitated if both quantities are measured simultaneously on tritons in a well-characterized atomic or molecular environment, and if the corrections for atomic and/or molecular excitations in that environment are calculable with a comparable level of precision. Our measurements on molecular tritium using a new bremsstrahlung technique are the first to fulfill these criteria. On the theoretical side, the nuclear matrix element describing the decay has recently been calculated [4] with an uncertainty that the authors estimate as close to 0.2%. In light of these developments, it may now be possible to make a meaningful distinction between the two interpretations of the triton β decay.

The measurement of W_0 is essential in order to calculate the phase-space factor

$$f = \int_1^{W_0} F(Z, W) p W (W_0 - W)^2 dW, \quad (1)$$

where $F(Z, W)$ is the Fermi function and p the electron momentum. The product of f and $t_{1/2}$ is related to the vector and axial-vector matrix elements through the expression [5]

$$(1 + \delta_R) t_{1/2} = \frac{K / (G_V')^2}{f_V \langle 1 \rangle^2 (1 - \delta_c) + f_A R_e^2 \langle \sigma \rangle^2}, \quad (2)$$

where $\delta_R = (1.906 \pm 0.001)\%$ is a radiative correction, G_V' is the experimental value of the Fermi coupling constant, $\delta_c = (0.06 \pm 0.01)\%$ is an isospin-impurity correction, and $R_e = G_A/G_V'$ is the ratio of axial-vector-to-vector coupling constants. The phase-space factors for

vector and axial-vector interactions differ slightly. $\langle 1 \rangle$ and $\langle \sigma \rangle$ are the Fermi and Gamow-Teller matrix elements. K is a constant.

Atomic effects influence the value of f in important ways [6]. First, excitation of the daughter atom reduces the energy available for the emitted electron and antineutrino. Second, roughly 1% of the decays result in K shell creation, in which the β particle materializes in the K shell of the ^3He daughter rather than in the continuum. This process, which is the inverse of K capture, clearly depends on the existence of vacancies in the K shell of the daughter, which in turn depends on the electronic environment of the decaying triton. Previous half-life determinations are collected in Table I. The measurements disagree, in some cases by many times the uncertainty. This may be attributable to the very different atomic environments in which the tritium is decaying. In the calorimetric half-life determination in titanium tritide [7], for example, the phase diagram for the titanium-hydrogen system shows that two phases with different atomic concentrations of hydrogen coexist in the temperature range in which the measurements were made. Part of the calorimetry measurement involves high-pressure $^3\text{H}_2$ gas. Here the solubility of tritium in the walls of the container makes the electronic environment of the tritium uncertain. In the recent lithium fission experiment [8], the interstitial site of the stopped triton fragment is not known. The accuracy of the very precise He collection measurement is difficult to estimate since the measured decay curve consists of only two points.

One of us (B.B.) [9] has calculated the modification of the f value due to bound-state decay, atomic excitation, screening, and exchange for the four species $^3\text{H}^+$, ^3H , $^3\text{H}_2$, and $^3\text{H}^-$. It was found that the f value, and consequently the half-life, could vary by as much as 1%. From this analysis, it is clear that one can minimize the uncertainty in the $ft_{1/2}$ product by combining measurements of the end-point energy and the half-life made on the same species in order to reduce the dependence on theory [6]. Such a combination of measurements was reported for ^3H atoms implanted into a lithium-drifted silicon [Si(Li)] detector [10]. However, as discussed below, a recent

TABLE I. Precision measurements of tritium half-life in different atomic environments. The calorimetry result is based on the five determinations listed immediately below it.

Half-life (yr)	Method	Source	Reference
12.262 ± 0.004	He collection	Low-pressure $^3\text{H}_2$ gas	[19]
12.250 ± 0.027	He collection	Lithium tritide	[20]
12.346 ± 0.002	Calorimetry		[7]
12.3452 ± 0.0019		Titanium tritide	
12.3479 ± 0.0030		Titanium tritide	
12.3452 ± 0.0084		Titanium tritide	
12.3583 ± 0.0057		High-pressure $^3\text{H}_2$ gas	
12.194 ± 0.056		High-pressure $^3\text{H}_2$ gas	
12.43 ± 0.05	Counting	Standard water sample	[21]
12.32 ± 0.03	Counting	Tritium implanted in Si(Li)	[10]
12.38 ± 0.03	He collection	Lithium fission	[8]
12.31 ± 0.03	Bremsstrahlung	Tritium-xenon mixture	Present work

analysis of tritium implantation in silicon [11] has shown that the electronic environment of the decaying tritium is not that of the free atom. Molecular tritium, on the other hand, is particularly attractive because the effect of molecular excitation on the extraction of the nuclear mass difference from the measured end point has been thoroughly investigated [12].

In our experiment, a Si(Li) detector recorded the x-ray bremsstrahlung emitted by gaseous tritium molecules mixed with xenon gas contained in impermeable glass bulbs. The end point of the x-ray spectrum is identical to the end point of the β decay spectrum, while an analysis of the decrease of the total counting rate with time yielded the half-life. A detailed discussion of the x-ray spectrum and the extraction of the end-point energy is given in the preceding Letter [13]. In this paper, we report the half-life and its implication for the axial-vector interaction.

The bulbs used in the half-life determination contained tritium-xenon mixtures in the ratios 70:30 and 50:50, and are described in the preceding Letter [13]. The tritium activity was 2.8 and 1.9 Ci, respectively. Based on the permeation rate of the Corning 1720 glass, less than 0.2 ppm of the tritium will have leaked out after one half-life. Upon receipt from Amersham, the bulbs were tested for leakage with a sensitive tritium sniffer calibrated with a $3.7\text{-}\mu\text{Ci } ^{133}\text{Xe}$ source. An upper limit on the leakage rate of $0.5\ \mu\text{Ci}$ per half-life was determined for each bulb.

The xenon served two important functions. First, it enhanced the bremsstrahlung rate. As shown in the preceding Letter, the ratio of external bremsstrahlung (EB) to internal bremsstrahlung (IB) is given by $1.2Z_{\text{eff}}f(k)/g(k)$, where f and g are spectral functions for EB and IB, respectively, and Z_{eff} for mixtures was given in Eq. (6). For our initial gas compositions, Z_{eff} for the two tritium-xenon mixtures was 51.8 and 53.0, respectively, which significantly increased the EB rate. The second function served by the xenon concerns the change in Z_{eff} with time. With the compositions used in our experiment, the change in Z_{eff} is small and calculable. For

example, over a 2-yr period, Z_{eff} changes by 0.2% if initially $N_{^3\text{H}_2} = N_{\text{Xe}}$.

The x-ray spectrum was recorded by a 3-mm-thick, 5-mm-diam Si(Li) detector. Its efficiency is near unity for the glass-filtered x rays that extended from 5 to 18.6 keV. The efficiency was checked daily by automatically positioning a $10\text{-}\mu\text{Ci } ^{241}\text{Am}$ source in front of the detector. The Np L x rays extend from 11.9 to 17.8 keV. Temperature drifts that could affect the detector and amplifier counting rates or a long-term deterioration in the detector efficiency were corrected by normalizing to the ^{241}Am rate. The average decline in detector efficiency, corrected for decay in the Am calibration source, was 0.2%/yr. However, superimposed on this decline were variations of 0.6% over a period of 1 yr. Short-term stability over a 10-day period was typically 0.1% or better.

Positioning of the bulbs and of the calibration source was controlled by an SC/MP microprocessor that stepped a SLO-SYN motor coupled to a precision lead screw. Reproducibility was 10^{-4} in. or better. The entire apparatus including the detector was rigidly mounted to a 1.6-cm-thick aluminum jig plate. The source holders, the plate on which they were mounted to the sample changer, and the sample changer all had holes for stainless-steel pins that facilitated repositioning to 10^{-3} in. following disassembly.

A typical x-ray spectrum is shown in the preceding Letter. The fit is not required for the half-life measurement; it serves to confirm the accuracy of the data analysis, particularly the preponderance of EB, and the absence of significant systematic error. Since the entire spectrum was summed, short-term drifts, typically of the order of 1 eV, changed the integrated rate by less than 1 ppm. Pileup events formed a well-defined peak of their own at roughly twice the energy of the main peak, but with 0.03% of its intensity. The subtraction of pileup events contributed an uncertainty of 3 ppm in the counting rate. With 5 cm of lead shielding, the integrated background counting rate for this small-volume detector was 1.4×10^{-3} Hz, while initial data acquisition rates

were 1100 and 700 Hz for the two bulbs, respectively.

Raw counting rates were corrected for dead-time losses and for inefficiencies produced by the pileup rejection circuitry of the Canberra 2020 amplifier coupled to a Canberra 35⁺ multichannel analyzer. Together, these amounted to 0.18% at a counting rate of 1000 Hz, decreasing to 0.08% at 500 Hz.

Data collection began in July 1985. The deviations of the experimental counting rate from that predicted by the fitted half-life are shown in Fig. 1 for the higher-counting bulb. The χ^2/N_{DF} for this fit is 1.02 ($N_{DF}=16$). Each point represents an average of ten runs. A typical run consisted of 82000 seconds of data from one of the tritium sources, followed by 4000 seconds of Am calibration x rays. With our automated data taking procedure, runs of hundreds of consecutive days could be carried out. In the later stages of the experiment, the samples with tritium-xenon mixture were intermittently removed to allow for the introduction of the other tritium samples described in the preceding Letter. The length of a set of runs was also reduced to typically 10–30 days. This is the reason for the gaps appearing in Fig. 1 in 1987 and in 1989.

The Am calibration source had a counting rate of 600 Hz. The expected statistical uncertainty in each data point, for both Am and tritium counting, was of the order of 0.03%. Most sets of runs, however, showed fluctuations that were twice as large that were attributable to temperature changes following a sample change and to microphonic noise from the SLO-SYN motor. An additional uncertainty of 0.06% was assigned to these systematic fluctuations. In addition, as noted above, the detector efficiency showed long-term variations as large

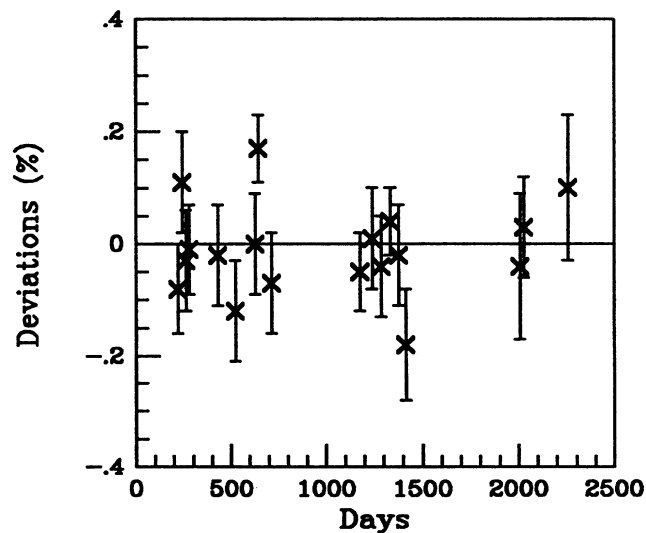


FIG. 1. Deviations (in %) of the experimental counting rates from those predicted using a half-life of 12.31 yr. The data points are for the 70% $^3\text{H}_2$ and 30% Xe mixture.

as 0.6% which are corrected for by normalizing to the Am rate. However, since these variations are large and not thoroughly understood, an additional systematic uncertainty amounting to 10% of the variation for each individual data point has been allowed. All three uncertainties due to statistics, systematic fluctuations, and long-term systematic variations were added in quadrature.

Half-lives deduced from the two bulbs are 12.31 ± 0.03 and 12.32 ± 0.06 yr, for a weighted average of 12.31 ± 0.03 yr. These values have been corrected for the half-life of the ^{241}Am source, 432.7 ± 0.6 yr, used to calibrate the detector efficiency. They also include corrections of 0.08% and 0.06%, respectively, for x rays emitted as IB and for the fraction of EB x rays emitted in the glass walls of the bulbs. Decay data from the other three bulbs used in the end-point measurement was insufficient for a half-life determination. These bulbs have been studied for only 3 yr. Their initial counting rates were an order of magnitude lower than those of the tritium-xenon mixtures.

The end-point energy for molecular tritium reported in the preceding Letter implies a nuclear mass difference for $^3\text{H}-^3\text{He}$ of $18530 \pm 4 \text{ eV} + m_e c^2$. The nuclear mass difference as determined from the weighted average of the $^3\text{H}-^3\text{He}$ atomic mass difference of earlier measurements [11] given in the preceding Letter is $18529 \pm 2 \text{ eV} + m_e c^2$, which differs insignificantly from the bremsstrahlung result. With the latter as the available energy, the phase-space factors in Eq. (2) are $f_\nu = 2.8433 \times 10^{-6}$ and $f_A = 2.8584 \times 10^{-6}$. These were scaled from the values reported in Ref. [10] for a slightly lower nuclear mass difference. Both values include a screening correction for the decay of a free atom, which is of the order of -0.22% [9]. On the other hand, the change in the f value for $^3\text{H}_2$ due to the atomic effects described earlier amount to 0.32% [9]. Thus both f factors should be increased by $(0.54 \pm 0.03)\%$. The numerator on the right-hand side of Eq. (2) has been revised recently based on a reexamination of eight pure Fermi transitions [14]. Its value is $6137.2 \pm 3.6 \text{ s}$. The product $R_e \langle \sigma \rangle$, as measured in this experiment, is 2.098 ± 0.003 , in good agreement with the tritium implantation value, 2.094 ± 0.004 .

To a certain extent, this agreement is fortuitous. A reevaluation of the end-point energy in the tritium implantation experiment found that chemical effects increase the end-point energy by 10 eV [11]. This would increase the phase-space factors, and lower the product. However, the importance of the chemical shifts calls into question the accuracy of using the correction factors for the free atom in evaluating the phase-space factors for implanted tritium, e.g., the K shell creation probability may change in the $\text{Si}(\text{Li})$.

Spurred by the implantation result, a recent calculation [4] showed that for a wide variety of nucleon-nucleon interactions, the matrix element $\langle \sigma \rangle$ is relatively insensitive to the presence of Δ isobars and to exchange currents.

This calculation confirms an early expectation [15] that an increase in the D -state admixture to the wave function suppresses the isobar contribution, such that the sum of their contributions to the matrix element is essentially constant. The authors considered the Argonne AV14, Paris, super soft-core, Bonn, and Reid soft-core potentials. The calculated matrix elements were $0.962\sqrt{3}$, $0.962\sqrt{3}$, $0.960\sqrt{3}$, $0.961\sqrt{3}$, and $0.961\sqrt{3}$, respectively. Their recommended value is $\sqrt{3}(0.962 \pm 0.002)$. When this is combined with the $R_e \langle \sigma \rangle$ product, as measured in this experiment, a value $R_e = 1.259 \pm 0.002$ is found, where the error reflects the uncertainty in experimental quantities only.

The very good agreement between this result and the bare nucleon value, $R_e = 1.261 \pm 0.004$, derived from angular correlation constants [16] in neutron decay, suggests that the axial-vector coupling constant is not quenched in the triton. The agreement is not as good with the value of R_e derived from the most recent measurements of the neutron lifetime, $R_e = 1.267 \pm 0.007$ [16]. Values of R_e derived from angular correlations are much more self-consistent than those based on neutron lifetime determinations. In addition, the most recent neutron lifetime measurements have tended toward somewhat lower values, with a mean value of 889.1 ± 1.8 s [17]. It is interesting to see what constraints the tritium β decay can place on the neutron lifetime, which is related to the R_e by

$$(f\tau)_n = \frac{K/(G_V')^2}{1 + 3R_e^2},$$

where $K/(G_V')^2$ is the same as in Eq. (2), and $f_n = 1.71465 \pm 0.00015$ [18]. This analysis has the virtue that the dependence on the vector coupling constant drops out. Using our measured value of R_e and its uncertainty, we find that τ_n should lie in the range 897 ± 3 s, roughly

2σ higher than the latest mean value.

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