

Helium-isotope mass-spectrometric method for studying tritium beta decay (idea, experiment, nuclear and molecular physics applications)

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Abstract. Experimental data on the variation of tritium nucleus beta decay constant caused by the interaction of the resulting beta-electron with orbital electrons and shell vacancies are reviewed for free atomic tritium and molecular tritium and used to obtain the half-life of atomic tritium $(T_{1/2})_a = (12.264 \pm 0.018)$ y, the half-life of the free triton $(T_{1/2})_t = (12.238 \pm 0.020)$ y, the axial-vector-to-vector weak-interaction coupling constant ratio $(G_A/G_V)_t = -1.2646 \pm 0.0035$ for beta decay of the triton, and an independent estimate of the free neutron lifetime $\tau_n = (890.3 \pm 3.9_{\text{stat}} \pm 1.4_{\text{syst}})$ s.

1. Introduction

Studies of the beta decay of atomic nuclei have played an important role in forming the main ideas of the physics of nucleus and elementary particles. The theory of a purely electromagnetic structure of atoms and their nuclei, which prevailed up to the 1930s, recognized only three elementary particles: the electron, the proton, and the photon. It was assumed that in beta decay of a nucleus only electrons with certain energy could appear (so that the law of energy conservation for the entire system was satisfied). However, experiments showed that beta decay exhibits a broad continuous spectrum of beta-electrons with energies ranging

from zero to the limiting energy E_{max} , which is determined by the mass defect. To resolve this contradiction, Wolfgang Pauli suggested in 1930 that a certain neutral particle is produced together with the beta-electron and that the sum of energies of each pair consisting of an electron and this neutral particle produced as result of the beta decay of a nucleus is constant [1]. Enrico Fermi called this particle a neutrino and built a neat theory of beta decay, in which he included the concept of the intranuclear weak interaction, which incorporated the neutron, discovered by Sir James Chadwick, and the idea that three particles are produced in the course of beta-minus decay: the proton, the electron, and the neutrino [2]. Fermi was the first to calculate the coupling constant of the weak vector interaction, whose modern more exact value is

$$\frac{G_F}{(\hbar c)^3} = 1.16639(1) \times 10^{-5} \text{ GeV}^{-2}$$

for leptonic processes [3], and

$$\frac{G_V}{(\hbar c)^3} = 1.1513(10) \times 10^{-5} \text{ GeV}^{-2}$$

for hadron – lepton processes [4].

The three-nucleon beta-active nucleus of tritium (the triton) is a very convenient object for verifying through experiments the theoretical concepts concerning the intranuclear interaction and the role that strong interaction in the form of internucleon meson exchange plays in beta decay. However, so far all attempts to analyze the theoretical models and to calculate their parameters using data on the beta decay of tritium have failed because, for one thing, the values of half-lives and the limiting energies of the beta spectrum found in experiments were determined by the type of tritium chemical compound used in these experiments. This made it

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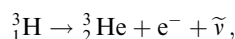
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impossible to bring into agreement the experimental values of half-lives and limiting energies of the beta spectrum and hindered the exact calculation of the reduced (comparative) half-life of the free triton, a fundamental characteristic of beta transitions, used in calculating coupling constants.

The results presented in the sections below help us to tackle the problems of finding correctly the temporal characteristics of the beta decay of the tritium nucleus. The findings on free-triton decay are used to determine the axial-vector-to-vector weak-interaction coupling constant ratio and to make an independent estimate of the free neutron lifetime. We also show that our method of measuring variations in the half-life of the triton introduced into various hydrogen-containing molecules can be used to study the electronic structure of atomic and molecular systems.

2. Helium-isotope mass-spectrometric method for measuring the half-life of the triton in gaseous substances incorporating tritium atoms

The relationship describing the beta decay of the tritium nucleus, viz.



implies that the half-life of this nucleus can be found by measuring the accumulation of the stable isotope ${}^3\text{He}$ confined to a closed space together with tritium. By adding the stable isotope ${}^4\text{He}$ to the volume containing tritium one can register the accumulation of radiogenic helium-3 isotope by measuring the changes in the ratio between ${}^3\text{He}$ and ${}^4\text{He}$ isotope concentrations. The helium-isotope technique for measuring the triton half-life is based on calculations of the parameters in the decay exponential function, which is governed by the equation

$$N = N_0[1 - \exp(-\lambda\Delta t)], \quad (2.1)$$

where N_0 is the initial number of decaying nuclei, N is the number of nuclei that have decayed by time Δt (or the number of daughter nuclei produced by that time), and λ is the decay constant related to the half-life $T_{1/2}$ and the average lifetime τ of the beta-active nucleus through the following formula

$$\lambda = \frac{1}{\tau} = \frac{\ln 2}{T_{1/2}}.$$

Equation (2.1) suggests that in a mixture of gases containing a gaseous compound of tritium and ${}^4\text{He}$, the ratio between ${}^3\text{He}$ and ${}^4\text{He}$ isotope concentrations at the instants of time t_1 and t_2 is determined by the expressions

$$\left(\frac{{}^3\text{He}}{{}^4\text{He}}\right)_{1,2} = \frac{n_0}{{}^4\text{He}} \{1 - \exp[-\lambda(t_{1,2} - t_0)]\}, \quad (2.2)$$

where $n_0/{}^4\text{He}$ is the ratio of concentrations of tritium and helium-4 at some instant of time t_0 .

To conduct an experiment on determining the half-life, the gas mixture prepared at the point in time t_0 is ‘packaged’ into several ampoules without violation of the ratio $n_0/{}^4\text{He}$. These ampoules are then stored in liquid nitrogen to reduce the diffusion of the gases to the ampoule walls. At the instants of time t_1 and t_2 , the gas from the selected ampoule is cleared from the component containing tritium and the ratios $({}^3\text{He}/{}^4\text{He})_{1,2}$ are measured with a double-beam isotope mass

spectrometer. A particular expression that follows from Eqn (2.2) is given by

$$\frac{({}^3\text{He}/{}^4\text{He})_1}{{}^3\text{He}/{}^4\text{He}_2} = \frac{1 - \exp[-\lambda(t_1 - t_0)]}{1 - \exp[-\lambda(t_2 - t_0)]}. \quad (2.3)$$

Thus, by comparing the ratios of the helium-isotope concentrations at instants of time t_1 and t_2 , one can determine the value of λ [5]. Since the sensitivity in different channels of the double-beam mass spectrometer usually differs and may vary with time, the samples used in the exact measurements of $({}^3\text{He}/{}^4\text{He})_{1,2}$ consisted of a calibration mixture with a ratio $({}^3\text{He}/{}^4\text{He})_{\text{cal}}$ close to the expected value of the helium-isotope ratios in the samples. Here, the true values of $({}^3\text{He}/{}^4\text{He})_{1,2}$ can be expressed in terms of the measured values of helium-isotope ratios in the samples, $[({}^3\text{He}/{}^4\text{He})_{1,2}]_{\text{meas}}$, and in the calibration mixture, $\{[({}^3\text{He}/{}^4\text{He})_{\text{cal}}]_{1,2}\}_{\text{meas}}$, in the following manner:

$$\left(\frac{{}^3\text{He}}{{}^4\text{He}}\right)_{1,2} = \left[\left(\frac{{}^3\text{He}}{{}^4\text{He}}\right)_{1,2}\right]_{\text{meas}} \frac{({}^3\text{He}/{}^4\text{He})_{\text{cal}}}{\{[({}^3\text{He}/{}^4\text{He})_{\text{cal}}]_{1,2}\}_{\text{meas}}},$$

with the result that

$$\frac{({}^3\text{He}/{}^4\text{He})_1}{{}^3\text{He}/{}^4\text{He}_2} = \frac{({}^3\text{He}/{}^4\text{He})_{1\text{meas}}}{({}^3\text{He}/{}^4\text{He})_{2\text{meas}}} \frac{[({}^3\text{He}/{}^4\text{He})_{\text{cal}}]_{2\text{meas}}}{[({}^3\text{He}/{}^4\text{He})_{\text{cal}}]_{1\text{meas}}}. \quad (2.4)$$

From Eqns (2.3) and (2.4) it follows that there is no need to know the exact value of $({}^3\text{He}/{}^4\text{He})_{\text{cal}}$ in order to find λ ; what is important is to ensure that the composition of the calibration mixture does not vary with time. It is also obvious that when using the helium-isotope mass-spectrometric method for finding λ one needs only to measure the ratio of the respective output currents of the detectors of the spectrometer; there is no need to measure the absolute values of such quantities as the volumes of the containers with the gas mixtures or the temperature and pressure in these containers. These features of the helium-isotope technique explain how it is possible to achieve with its help a considerable increase in accuracy and reliability of measurements of the decay constant. The material in the sections that follow will show that this played the leading role in obtaining a number of important results in the studies of beta decay processes.

Researchers at the Mass Spectrometry Laboratory of the A F Ioffe Physico-Technical Institute, RAS (St.-Petersburg, Russia) have developed special mass-spectrometric instrumentation and techniques for precise measurements of helium-isotope ratios [6, 7]. The maximum metrological possibilities of these measurement procedures are as follows: the minimum measured isotope ratio $({}^3\text{He}/{}^4\text{He})_{\text{min}} = 10^{-11}$ [8], and the attained accuracy of measuring ${}^3\text{He}/{}^4\text{He}$ correspond to an error of about 0.01% when the amount of ${}^3\text{He}$ in the sample is of order $10^{12} - 10^{13}$ atoms and the value of the ratio ${}^3\text{He}/{}^4\text{He}$ is approximately 0.1–0.001 [9]. Such characteristics of the instrumentation have made it possible to find the value of λ by the helium-isotope technique with an error of order 0.1% for different tritium-containing compounds. The special features of the design of double-beam mass spectrometers intended for use in precise measurements of ${}^3\text{He}/{}^4\text{He}$ are described in monograph [10], and some details of the design of such spectrometers are brought up in Sections 3 and 4.

3. Measuring the half-life for molecular tritium

The above technique has been used to measure the half-life $(T_{1/2})_m$ for molecular tritium. The need to do this arose from the fact that the results of well-known experiments on determining the tritium half-life were found to differ by more than 2%, with the indicated errors of some of these experiments amounting to about 0.03%. Despite the fact that the measurements were carried out with different tritium-containing compounds $^3\text{H}_2$ [11–13], $^3\text{H}_2\text{O}$ [14], Li^3H [15, 16], and Ti^3H [17], such discrepancies cannot be explained by the effect of differences in the electron environment of tritium nuclei in the samples under investigation. This is an indication of a certain unaccounted systematic error in the results obtained, an error caused, among other things, by the need to measure absolute values of physical quantities. In the helium-isotope method, the only quantity whose absolute value requires measuring is the ^3He accumulation time, but with the time of exposure being one year or longer the error in λ , caused by an uncertainty in exposure time readings that amounts to about several dozen seconds, will be, as it is easy to show, more than 1000 times smaller than the overall error in the result.

The gas mixture used in the real experiments consisted of 49% deuterium (^2H), 1% protium (H), and 50% tritium, while the calibration mixture consisted of helium isotopes with a ratio $(^3\text{He}/^4\text{He})_{\text{cal}} = 1.7 \times 10^{-2}$. The possible sources of the systematic error, caused by various procedures employed in producing tritium-containing samples and the calibration mixture, packaging of gas mixtures into ampoules, and sealing and storing of the ampoules, were studied in special experiments in which the solubility of hydrogen and helium isotopes in glass was measured at liquid-nitrogen and room temperatures and at the melting point. The hydrogen component (tritium, deuterium, and a small amount of protium) was separated from the measured helium isotopes during the interaction of the mixture with a heated palladium membrane. The degree of removal of tritium from the samples was verified by using a specially designed low-background radiometric instrument based on a proportional internal-filled counter with a minimum detected tritium activity of roughly 0.002 Bq (i.e., about 10^6 tritium atoms in the counter's volume) [6].

Naturally, the accuracy of the final result could be affected most significantly by the presence of multiplet peaks at the third and fourth masses in mass-spectrometric measurements of helium-isotope ratios. This problem was solved by using a specially developed two-stage magnetic resonance mass spectrometer (MRMS) [10] with a record-breaking resolution: the background ('tail') of the $^4\text{He}^+$ peak at the $^3\text{He}^+$ peak in the mass spectrum amounts to $10^{-8}\%$ of the amplitude of the $^4\text{He}^+$ peak, and the background of the doublet peak (HHH^+ , $^2\text{HH}^+$) at the $^3\text{He}^+$ peak amounts to $10^{-3}\%$ of the amplitude of the (HHH^+ , $^2\text{HH}^+$) peak. To reduce the concentration of hydrogen in the ion source, a getter titanium pump was connected to the mass-spectrometer chamber, with the equilibrium content of the hydrogen compounds HHH and ^2HH in the chamber reduced to roughly 10^8 molecules. The use of semitransparent collectors (grids) allowed precise measurement of the ratio $^3\text{He}/^4\text{He}$ in the first (static) stage of the MRMS without loss of resolution in the second (dynamic) stage, which made it possible to constantly monitor the level of the multiplet hydrogen peaks at the third mass.

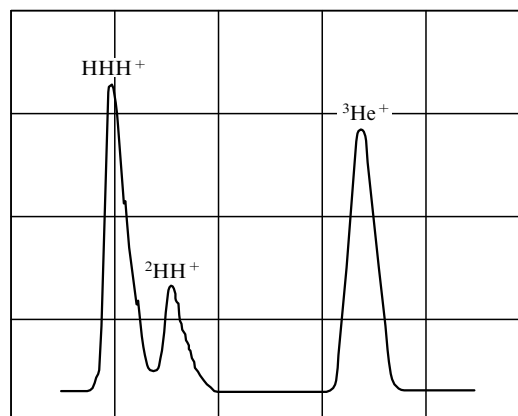


Figure 1. Mass spectrogram of a multiplet with a mass number equal to 3, recorded for a sample with 10^{12} atoms of helium-3 (the height of the ^3He peak has been reduced by a factor of 10^4 by scaling).

Figure 1 displays the spectrum of the (HHH^+ , $^2\text{HH}^+$, $^3\text{He}^+$) multiplet recorded when approximately 10^{12} ^3He atoms were fed into the MRMS chamber.

In the 586 days that tritium-containing mixtures were exposed, 116 relative measurements of the ratio $^3\text{He}/^4\text{He}$ in the samples were made. The result for the triton half-life in molecular tritium was [18]

$$(T_{1/2})_m = 12.279 \pm 0.033 \text{ y}$$

with the solar year duration assumed to be equal to 365.25 days. In 1991, Budick et al. [19] obtained the following value of the triton half-life from their studies of the decay exponential function done by recording the bremsstrahlung of beta electrons in mixtures of molecular tritium and noble gases:

$$(T_{1/2})_m = 12.31 \pm 0.03 \text{ y},$$

which is in good agreement, to within experimental errors, with the result of the present work. Below we use the weighted mean value of the above results for $(T_{1/2})_m$, i.e., we will assume that

$$(T_{1/2})_m = 12.296 \pm 0.017 \text{ y}.$$

4. Measuring the difference in half-lives for molecular and atomic tritium

According to Fermi's theory of beta decay, the probability of emitting electrons by nuclei and the spectrum of these electrons depend on the structure of the electron phase space in the atomic–molecular system, the space which is accessible to the beta-electron being formed. Here, the experimental values of the parameters of the beta transition, namely the decay constant and the limiting energy of the beta-electron spectrum, prove to be sensitive to the state of the electron surroundings of the nucleus (what is known as the chemical shift of λ and E_{max}). Tritium-containing atomic–molecular systems are the most promising from the point of view of solving experimental and theoretical problems in studies of chemical shifts in beta decay. Since the excess of mass of the second neutron in the tritium nucleus is close to the Coulomb energy of the second proton in the helium nucleus, the energy

released in the beta decay of the triton is relatively low (~ 18.6 keV). In this case a significant fraction of beta-electrons is produced with wavelengths characteristic of atomic electron systems, which leads to substantial changes in the beta spectrum and makes the chemical shifts of λ and E_{\max} measurable. When the limiting energy is measured, the shifts in E_{\max} are masked by the unavoidable changes in the energy of a beta-electron on its way to the detector, which significantly complicates the identification of the observed changes in E_{\max} . Such difficulties do not appear in measuring the chemical shifts of the decay constant, since the decay rate is determined solely by the density of vacancies for the virtual beta-electron and, obviously, is independent of the fate of the real electron that has left the circumnuclear space. In this section we give the results of the first experiment on measuring the difference in the decay constants for atomic (λ_a) and molecular (λ_m) tritium:

$$\Delta\lambda = \lambda_a - \lambda_m.$$

As in the process of determining the absolute value of the half-life for molecular tritium, at some instant of time a mixture of molecular tritium and ^4He was produced. Two ampoules contained approximately equal amounts of this mixture, then in one ampoule a gaseous radio-frequency discharge was ignited. This led to dissociation of the tritium molecules. Since the exposure time Δt in which $\Delta\lambda$ was measured amounted to several hours, we can assume with a high accuracy that

$$1 - \exp(-\lambda\Delta t) = \lambda\Delta t.$$

In this case, the equation of the helium-isotope measuring method for determining $\Delta\lambda$ for a pair consisting of an ^3H atom and an $^3\text{H}_2$ molecule assumes the form [20]

$$\frac{(^3\text{He}/^4\text{He})_a}{(^3\text{He}/^4\text{He})_m} = \frac{\Delta t_a}{\Delta t_m} \left(p\varepsilon \frac{\Delta\lambda}{\lambda_m} + 1 \right). \quad (4.1)$$

Here, $(^3\text{He}/^4\text{He})_{a,m}$ are the values of the helium-isotope ratios in the samples with atomic and molecular tritium, $\Delta t_a/\Delta t_m$ is the ratio of the exposure times for the atomic and molecular samples, the dimensionless factor $p < 1$ takes into account the accumulation of helium-3 in the ‘atomic’ sample with the decay rate of the triton in the tritium molecule, which is due to the distinction between Δt_a and the discharge burning time, ε is the degree of dissociation of molecular hydrogen in the discharge, and the value of the decay constant λ_m for molecular tritium corresponds to the above averaged value $(T_{1/2})_m$.

The key problem that emerges when an rf discharge is used to atomize molecular tritium is that the free atoms produced as a result of dissociation of molecules by electron impact clearly have a non-Maxwellian energy distribution. In accordance with the Franck–Condon principle, a hydrogen molecule passes with the highest probability to the first excited state (a state with repulsive forces), absorbing roughly 13.5 or 9.5 eV, while the dissociation energy is only 4.5 eV. Thus, the kinetic energy of the recoil atoms after molecular disintegration is roughly 9 or 5 eV and is distributed among the hydrogen atoms H , ^2H , and ^3H according to the laws of energy and momentum conservation in the center-of-mass system. This leads to rapid adsorption of the recoil atoms on the walls of the discharge cell.

Computer simulation with protium molecules showed that after roughly 15 min of direct molecular dissociation by electron impact the pressure in the ampoule drops to such an extent that the discharge is terminated. The reduction in the energy of the atoms during the dissociation of the molecules was achieved through dissociation by collisions of the second kind in the interaction of the hydrogen molecules with mercury atoms excited to the levels 6^3P_1 and 6^3P_0 . The mercury introduced into the ampoule before the experiment was evaporated and excited after high-frequency voltage was applied to the outer ring electrodes. Control experiments showed that during the dissociation of molecules in the mercury discharge the concentration of hydrogen atoms in the discharge cell did not change during exposure.

The degree of dissociation of the molecules was found by studying the flow of atoms from the discharge cell as a function of the discharge power. Hydrogen atoms were fed to the mass-spectrometer source with electron impact along a fluoroplastic antirecombination track through a capillary (Fig. 2). A discharge power of 120–140 W was sufficient for reaching the horizontal plateau of the atomic-ion current [21]. In view of the possibility of the atomic-ion output current in the mass-spectrometer source becoming saturated before complete dissociation of the molecules in the discharge cell is achieved, we also employed an optical technique for measuring the degree of dissociation. The technique was based on comparing the intensity of the Balmer-series spectral lines H_α , H_β , H_γ , and H_δ identified by a monochromator and spectral bands with wavelengths of roughly 652, 580, and 410 nm from the molecular-hydrogen spectrum for two rf-discharge modes: the flow mode, in which the gas was continuously pumped through the discharge tube, and the static mode. The molecule residence time in the space between the electrodes amounted to 0.003–0.01 s in the flow mode. In this case, the hydrogen has practically no time to

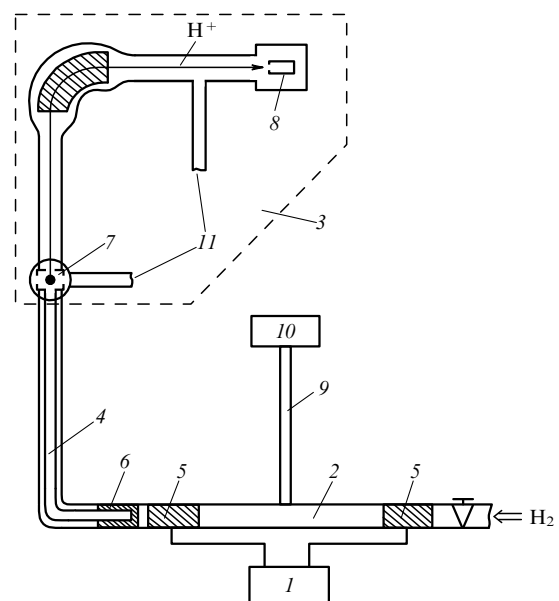


Figure 2. Schematic of the device for studying the dissociation of molecular hydrogen in an rf discharge: 1 — hf generator, 2 — discharge tube, 3 — isotope mass spectrometer, 4 — PTFE tube (through which the hydrogen atoms were fed to the source with electron impact), 5 — outer ring electrodes, 6 — capillary leak, 7 — ion source of the mass spectrometer, 8 — ion detector, 9 — optical fiber, 10 — monochromator, and 11 — vacuum fittings.

dissociate, so that molecular bands dominate in the discharge spectrum. In the static mode (i.e., without gas flow) with the same values of pressure and power dissipated in the discharge, the intensity of the lines belonging to the Balmer series increases by a factor of 10 to 15, while the intensity of the molecular spectral bands decreases by a factor of 25 or more. Comparing the coefficients of the relative variation of intensities, we can show that in the static discharge mode the degree of hydrogen dissociation exceeds 93%. The statistical weights of the ionized and excited states of hydrogen in the discharge, which were calculated with the use of estimates of the current, concentration and energy of the electrons, did not exceed $10^{-3}\%$, so that the experimental conditions guaranteed the observation of beta decay of tritium in the ground atomic and ground molecular states.

The removal of tritium from the samples was done in the process of the interaction of the hydrogen–helium mixtures with the heated palladium membrane. The relative residual activity caused by incomplete removal of tritium through the membrane was measured by a low-background proportional internal-filled counter and amounted to 0.1% of the initial specific activity of the mixture in the molecular and atomic samples. The palladium membrane penetrability for helium, the adsorption of helium by the ampoule walls, the presence of an admixture of ^3He in the ballast ^4He , and other factors determining the nonradiogenic variation of the ratio $^3\text{He}/^4\text{He}$ in mixtures were monitored by the above-mentioned magnetic resonance mass spectrometer, which has a helium-3 sensitivity of order 3×10^4 atoms [8]. The amount of ^3He produced in the samples during their exposure exceeded 2×10^{13} atoms, with the result that the possible systematic errors caused by helium leakage were recorded at a level of roughly $10^{-6}\%$.

By the end of the exposure period, the helium-isotope ratios for the atomic and molecular samples reached values of roughly 0.001. To measure these values precisely, we used a specially designed static double-beam mass spectrometer which ensured the reproducibility of relative measurements of the ratio $^3\text{He}/^4\text{He}$ at a level of 0.01% in a series of independent control experiments with a specially prepared calibration mixture. Five series of measurements were conducted in the real experiment on determining $\Delta\lambda$, and in each the helium mixtures from the ‘molecular’ and ‘atomic’ samples were alternatively fed to the chamber. As a result, the left-hand side of equation (4.1) yielded the value 0.90257 for the ratio

$$\frac{(^3\text{He}/^4\text{He})_a}{(^3\text{He}/^4\text{He})_m}$$

with a standard deviation of 0.00021. The effective exposure time intervals were as follows

$$\Delta t_m = 271.80 \text{ min}, \quad \Delta t_a = 244.90 \text{ min};$$

the discharge was sustained for 171.0 min. For the degree of dissociation ε , we adopted the value $0.95^{+0.05}_{-0.02}$, and $p = 171.0/\Delta t_a \approx 0.70$. The ratio $\Delta\lambda/\lambda_m$, which was found from equation (4.1), has a value of 0.00257 ± 0.00045 for the above-specified magnitudes of the parameters in equation (4.1). The error in the result that corresponds to one standard deviation was determined by a quadratic form including the variances of all parameters in the measurement equation. Here, the variance of the ratio $\Delta t_a/\Delta t_m$ was calculated with

allowance for a 95% positive correlation of the random quantities Δt_a and Δt_m , caused by the fact that the procedures for removing tritium from the two samples are identical. When calculating the absolute value of the difference $\Delta T_{1/2}$ of the half-lives of molecular and atomic tritium (such calculations were based on the results of measuring the relative shift of the decay constant), we allowed for the fact that

$$\frac{\Delta T_{1/2}}{(T_{1/2})_m} = \frac{\Delta\lambda}{\lambda_m} \quad \text{for} \quad \frac{\Delta\lambda}{\lambda_m} \ll 1$$

and found [22] that

$$\begin{aligned} \Delta T_{1/2} &= (T_{1/2})_m - (T_{1/2})_a = 0.0316 \pm 0.0055 \text{ y} \\ &= 11.5 \pm 2.0 \text{ d.} \end{aligned}$$

For the absolute value of the difference between the decay constants λ_a and λ_m we arrived at the result

$$\begin{aligned} \Delta\lambda &= \lambda_a - \lambda_m = \frac{\ln 2}{(T_{1/2})_m} (0.00257 \pm 0.00045) \\ &= (4.6 \pm 0.8) \times 10^{-12} \text{ s}^{-1}. \end{aligned}$$

By using the experimental data on the chemical shifts of the decay constant in compounds with a known state of the electron surroundings of the nucleus, one can represent the shift as a function of the structure of the electron phase space in the atomic–molecular system, the space which is accessible to the beta-electron being formed. For hydrogen-containing atomic–molecular systems, such representation of the chemical shift of the decay constant makes it possible to develop a diagnostic technique in which the electron state of a tritium-substituted hydrogen-containing system is identified by determining the relative difference of the decay constants of tritium for the given system and for a standard tritiated compound (e.g., the tritium molecule). Here, as equation (4.1) of the helium-isotope measuring technique suggests, the error in determining the absolute value of the decay constant has no effect on the result.

Three reaction channels are distinguished when the process of production of a beta-electron in the phase space of tritium-containing atomic–molecular systems is examined: direct production of a continuous-spectrum electron; production of a continuous-spectrum electron through substitution of an orbital electron by a beta-electron, and decay into bound states, i.e., the production of a beta-electron in one of the shells of a daughter atom or molecule. In the case of direct production of a continuous-spectrum electron in the Coulomb field of the nucleus as a result of beta decay, the effect of the electron surroundings on the magnitude of the phase–space factor (f) is described by introducing a parameter which governs the screening of the nuclear charge by orbital electrons and a parameter reflecting the changes in the observed value of the limiting energy of the beta spectrum [23].

The close attention given in recent years to the problem of the neutrino mass has stimulated calculations of the population of the excited electronic states of daughter atomic–molecular systems, when the triton incorporating into ^3H , $^3\text{H}^-$, $^3\text{H}_2$, CH_3^3H , and other hydrogen-containing compounds decays [24–27]. Theoretically, such calculations make it possible to determine, with high accuracy, the variations of the decay constant, caused by the interaction in the final state of beta-electrons from the high-energy portion of the spectrum (see Section 6) and, hence, to isolate in the

measured total value of $\Delta\lambda/\lambda_m$ the contribution resulted from the main processes in which low-energy electrons participate: the decay into bound states, and the beta-electron – orbital-electron exchange. In this respect, the tritium beta-electron diagnostics method can be compared to such a method of structural analysis as X-ray absorption near edge structure (XANES) spectroscopy [28]. Both methods examine the process of formation of low-energy electrons inside the atomic–molecular system under investigation, but in the case of the XANES method the source of electrons is not the nuclei but the K(L)-shells of heavy atoms (e.g., Fe) incorporated into the system being investigated and subjected to X-ray radiation with the photon energy exceeding the ionization energy for the K(L)-shell by an amount no greater than 50 eV. Here, the probability of beta decay and the photoionization probability prove to be very sensitive functions of the electronic state of the active atom (^3H or Fe, respectively) and its local surroundings. It is obvious, however, that the scope of problems that can be solved by the tritium method differs from that of the XANES method: in the latter it is important that the system under investigation contains fairly heavy atoms (Fe, Ni), since the photoeffect cross section on the inner electron shells of the atom depends on the atomic number, like Z^5 .

It should be emphasized that in addition to the common theoretical interpretation (based on Fermi's ideas) of the process of beta-electron formation, there is also an interesting approach to interpreting the experiments in which $\Delta\lambda$ is measured. This approach was developed in the theory of what is known as the quantum Zeno paradox. There, it was shown (first by L A Khal'fin and then by Misra and Sudarchan [29]) that the characteristic lifetime of an unstable nucleus is determined, in particular, by the fact that this nucleus is under observation (actually, by the conditions of the experimental identification of the quantum state of the system consisting of the nucleus and the emitted particle). Here, the real detector must be located at a distance smaller than the characteristic wavelength of the nuclear radiation, because only then will the interaction between the detector and the resulting particle in the virtual stage of the particle's existence affect the probability of real emission. It is obvious that only the ensemble of electrons surrounding the beta-active nucleus may serve as a detector capable of interacting with the beta-electron at the stage of the electron's formation in the circumnuclear space and affect the very probability of completion of this process. Thus, experiments on measuring the variation of the tritium decay constant as a function of the state of the chemical surroundings of tritium make it possible to verify the theory of the quantum Zeno effect and the related aspects of the theory describing the production of an electron as an elementary particle, as well as the fundamental ideas of quantum electron kinetics.

5. Determining the half-lives for atomic tritium and the free triton

The theory developed by Sherk [30] and Bahcall [31] makes it possible, at least in principle, to determine the probability of decay into bound states for arbitrary electron surroundings of the triton. However, only for the simplest systems, such as the free triton and the free tritium atom, do the results of calculations for the probability of decay into bound states, done by different researchers, agree among themselves fairly well. Tikhonov and Chukreev [32], Budick [33], and Harston

and Pyper [34] calculated the ratios of the probability of decay into bound states to the probability of decay into the continuous spectrum for the triton (t) and found them to be 1.10, 1.03, and 1.08%, respectively, i.e.,

$$\frac{\Delta\lambda_{bt}}{\lambda} = 1.07 \pm 0.04 \, \%.$$

The relative increase of the decay rate, caused by the decay into bound states in the tritium atom, was calculated in Refs [30–32, 34]. The results obtained were 0.56, 0.69, 0.66, and 0.55%, respectively, viz.

$$\frac{\Delta\lambda_{ba}}{\lambda} = 0.62 \pm 0.07 \, \%.$$

The interaction between a beta-electron and an orbital electron, which transfers the electron to a state in the continuous spectrum (here the beta-electron may remain in the bound state in the helium atom, but it can also find itself in an s-state of the continuous spectrum), must lead to an increase in the relative decay rate of the triton in the tritium atom by [34, 35]

$$\frac{\Delta\lambda_{ch}}{\lambda} \approx 0.15 \, \%.$$

For tritium-containing systems in which the phase volume accessible to the beta-electron near the triton is deformed by molecular orbitals, the results of calculations of electron exchange corrections to the beta-decay probability are only of a qualitative nature.

When decay occurs from the ground state of the tritium atom, 70.16% of the transitions are into the 1s state of the $^3\text{He}^+$ ion, 25.03% into the 2s state, 1.27% into the 3s state, etc. [26, 35]. This leads to a rise in the observed value of the limiting energy and an increase in the probability of triton decay in the atom by [34]

$$\frac{\Delta\lambda_{ex}}{\lambda} \approx 0.50 \, \%$$

compared to the probability of free-triton decay. So far, a detailed theoretical study of the screening of the nuclear charge by the orbital electrons has been conducted only when the triton decays in the ^3H atom or the $^3\text{H}^-$ ion, where the screening potential is formed by simple electron configurations: ns^1 in the $^3\text{He}^+$ ions, and $1s^2$, $2s^2$, and ns^1ms^1 in the (neutral) helium atom. For the tritium atom this results in a decrease in the decay constant by

$$\frac{\Delta\lambda_s}{\lambda} \approx 0.41 \, \%$$

compared to the value of λ for the free triton [34].

Thus, to date a total consistent set of theoretical values of corrections to the half-life that allows for four possible atomic effects in beta decay, namely, the decay into bound states, the electron exchange effect, the screening of the nuclear charge by orbital electrons, and the change in the observed value of E_{\max} , has been obtained only for the free triton and the free tritium atom. This means that the absolute value of the half-life for the free triton can be calculated only if one uses the absolute value of the half-life for the tritium atom, which must be determined through experiments. The experimental value of the half-life for the free tritium atom can be obtained from the data discussed in Sections 3 and 4 [36]:

$$(T_{1/2})_a = (T_{1/2})_m - \Delta T_{1/2} = 12.264 \pm 0.018 \, \text{y}. \quad (5.1)$$

Note that the error in Eqn (5.1) is almost entirely determined by the error in $(T_{1/2})_m$, and in finding the half-life for molecular tritium more accurately the error in determining $(T_{1/2})_a$ decreases substantially.

Since in determining $(T_{1/2})_m$ and $\Delta T_{1/2}$ we used experimental techniques that make it possible to allow for all the beta-decay reaction channels, the obtained value of $(T_{1/2})_a$ corresponds to the total probability of decay for atomic tritium (i.e., takes into account both the processes that lead to production of continuous-spectrum electrons and the decay into bound states). In this case when we go over from $(T_{1/2})_a$ to the value of the half-life $(T_{1/2})_t$ for the triton, account must be taken of all four corrections for the atomic effects showing themselves in the free tritium atom. Allowing for the fact that the magnitudes of the atomic corrections are much smaller than unity, we can find $(T_{1/2})_t$ by using the relationship

$$\begin{aligned} (T_{1/2})_a \left(1 + \frac{\Delta\lambda_{ba} + \Delta\lambda_{ch} + \Delta\lambda_{ex} - \Delta\lambda_s}{\lambda} \right) \\ = (T_{1/2})_t \left(1 + \frac{\Delta\lambda_{bt}}{\lambda} \right), \end{aligned} \quad (5.2)$$

where the second term in the parentheses on the right-hand side reflects the change in the value of $(T_{1/2})_t$ caused by the decay into bound states of the ${}^3\text{He}^+$ ion, i.e., the only possible atomic effect in the decay of the free triton. Using the above-given theoretical values of the atomic corrections, the experimental value of $(T_{1/2})_a$, and equation (5.2), we arrive at the following value for the triton half-life

$$(T_{1/2})_t = 12.238 \pm 0.020 \text{ y.}$$

Such a value of the half-life corresponds, for example, to the process in which the concentration of tritium nuclei decreases because of beta decay in a fully ionized hydrogen plasma. It should be emphasized that the value of the half-life for the triton, listed in tables of radionuclide characteristics, viz. $12.32 \pm 0.02 \text{ y}$ [37], was determined by averaging the experimental results in which the effect of the electronic structure of a tritium-containing atomic–molecular system on the decay probability had not been taken into account. Actually, this value is not a characteristic of the beta decay of the free triton, since it differs substantially (by four standard deviations) from the result obtained for the free triton in our work.

When only the effect of the Coulomb field of the nucleus on the beta spectrum is taken into account (i.e., when in calculating the phase–space factor only the standard Fermi function is used, while decay into bound states is not taken into consideration), the corresponding value of the half-life is

$$(T_{1/2})_{tc} = 12.369 \pm 0.020 \text{ y.}$$

This value of the half-life must be used, in particular, when we wish to determine the energy release due to beta decay in tritiated plasma, since in the decay into bound states the antineutrinos carry away almost all the released energy. The value of $(T_{1/2})_{tc}$ must also be utilized in calculating the reduced, or comparative, half-life $(fT_{1/2})_t$ of the triton, if one ignores the difference between the binding energies of the orbital electrons in the initial and final atomic–molecular systems, while the upper limit of the momentum interval

available to a beta-electron is taken in the form

$$c^{-1} \sqrt{E_0^2 - m_e^2 c^4},$$

where E_0 is the difference between the masses of the nuclei of tritium and helium-3 in energy units, from which the recoil energy of the helium nucleus is subtracted. Knowing the exact value of $(fT_{1/2})_t$ for the triton, we can submit independent estimates of the following quantities characterizing beta-decay processes: the (Gamow–Teller) axial-vector-to-vector (Fermi) weak-interaction coupling constant ratio, and the lifetime of the free neutron.

6. Determining the axial-vector-to-vector weak-interaction coupling constant ratio for beta decay of the triton

The nuclear difference between the masses of tritium and helium-3, determined on the basis of the weighted mean value of the results of 11 independent experiments [38, 39], is equal to $(18529 \pm 2) \text{ eV} + m_e c^2$. The recoil energy of the helium nucleus is 3.4 eV. Then, one finds

$$E_0 = (18525.6 \pm 2) \text{ eV} + m_e c^2.$$

Using a method of calculating the phase–space factor that allows for the Coulomb interaction between the beta-electron and a nucleus of finite size and mass and for processes involving virtual photons and charged particles — the products of beta decay (to within radiative corrections of order α^2) [40], for the given value of E_0 we find that

$$f = (2.894 \pm 0.006) \times 10^{-6}.$$

Then, we obtain the following result:

$$(fT_{1/2})_t = 1129.6 \pm 3 \text{ s.}$$

Note that the value of E_0 was determined either by the mass-spectroscopic methods (where the experimentally determined difference in the masses of the ${}^3\text{H}^+$ and ${}^3\text{He}^+$ ions is increased by the electron rest energy and decreased by the binding energy of the s-electron in the helium ion) or by methods based on measuring the limiting energy E_{\max} of the beta-electron spectrum for tritium, then adding $m_e c^2$ and the recoil energy of the daughter ion to it, and finally subtracting the chemical shift $\Delta E_{\max} = E_i - E_f$ from the sum, where E_i and E_f are, respectively, the initial and final (averaged with allowance for the theoretical values of the statistical weights of the final states) binding energies of the orbital electrons in the tritium-containing atomic–molecular system under investigation. Here, the weighted mean value of the difference in nuclear masses, determined on the basis of mass-spectrometric data, differs by less than 1 eV from the value of this quantity determined on the basis of the results of measuring the limiting energy E_{\max} of the beta spectrum for a number of hydrogen-containing compounds ($\text{C}_{20}\text{H}_{40}\text{O}_2$ [39], valine [41], and CH_4 [42]) in which one or several protium atoms in the CH groups were replaced by tritium atoms, and also for tritiated molecular hydrogen [38, 43]. This is an indication, in particular, that the values of the chemical shifts of the limiting energy ΔE_{\max} of the beta-electron spectrum, equal by order of magnitude to 30 eV, are determined by theoretical means with an error no larger

than 3% for tritiated molecules with covalent bonds. Since the value of the logarithmic derivative of the phase-space factor f with respect to E_{\max} equals [34]

$$\frac{\partial f / \partial E_{\max}}{f} = 1.8 \times 10^{-4} \text{ eV}^{-1},$$

an error of order 1 eV in determining E_{\max} leads to insignificant (from the point of view of the problem considered) variations of the value of f by $\sim 0.02\%$.

For allowed beta transitions, to which the beta decay of the triton belongs, $fT_{1/2}$ is related to the vector (M_V) and axial-vector (M_A) matrix elements of the Hamiltonian of the beta decay in the following way [44]:

$$fT_{1/2} = \frac{k/G_V^2}{|M_V|^2 + (G_A^2/G_V^2)|M_A|^2}, \quad (6.1)$$

where G_V and G_A are the vector and axial-vector coupling constants of the weak hadron-lepton interaction, and k is a constant factor. Since for $0^+ \rightarrow 0^+$ transitions within isospin multiplets with a total isospin $T = 1$ and its projection $T_z = 0$ in the final state we have $|M_V| = \sqrt{2}$ and $|M_A| = 0$, while for the triton beta decay we have $|M_V|_t = 1$, we can use the expressions (6.1) for the reduced half-lives of the triton and $0^+ \rightarrow 0^+$ transitions to arrive at the following relationship

$$\left[1 + \left(\frac{G_A}{G_V}\right)_t^2 |M_A|_t^2\right] (fT_{1/2})_t = 2(fT_{1/2})_{0^+ \rightarrow 0^+}. \quad (6.2)$$

The value of $(fT_{1/2})_{0^+ \rightarrow 0^+}$, determined on the basis of the data on eight purely Fermi transitions, amounts to [4]

$$3072.3 \pm 0.9_{\text{stat}} \pm 1.1_{\text{syst}} \text{ s}.$$

The magnitude of

$$|M_A|_t = \sqrt{3} (0.962 \pm 0.002)$$

was calculated by Saito et al. [45], who examined five modifications of the intranuclear potential and showed that within a broad range of shapes of the potential and structures of the wave functions for the ^3H and ^3He nuclei, the axial-vector matrix element is only slightly sensitive to the presence of Δ -isobars and exchange currents. Using equation (6.2) with the indicated values of $(fT_{1/2})_t$, $(fT_{1/2})_{0^+ \rightarrow 0^+}$, and $|M_A|_t$, we find that

$$\left(\frac{G_A}{G_V}\right)_t = -1.2646 \pm 0.0035.$$

For the mixed transition $n \rightarrow p + e^- + \bar{\nu}$, the value of the axial-vector-to-vector weak-interaction coupling constant ratio $(G_A/G_V)_n$ is determined on the basis of the results of measurements of the coefficient characterizing the asymmetry in the direction of the beta-electron escape in relation to the spin of the decaying neutron. The results of four such experiments, viz.

$$\begin{aligned} \left(\frac{G_A}{G_V}\right)_n &= -1.262 \pm 0.005 [46], \\ \left(\frac{G_A}{G_V}\right)_n &= -1.266 \pm 0.004 [47], \\ \left(\frac{G_A}{G_V}\right)_n &= -1.2594 \pm 0.0038 [48], \\ \left(\frac{G_A}{G_V}\right)_n &= -1.2686 \pm 0.0046 \pm 0.0007 [49], \end{aligned}$$

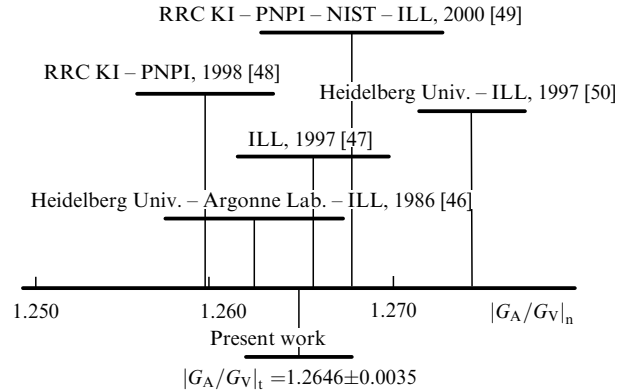


Figure 3. Results of research in determining the axial-vector-to-vector weak-interaction coupling constant ratio for the neutron and triton.

coincide within the indicated errors and possess the weighted mean value

$$\left\langle \left(\frac{G_A}{G_V}\right)_n \right\rangle = -1.2637 \pm 0.0022,$$

which is in good agreement with the value of $(G_A/G_V)_t$. This makes it possible to think of the ratio G_A/G_V as a universal constant characterizing beta processes. At the same time it must be noted that Abele et al. [50] arrived at the following value for the axial-vector-to-vector coupling constant ratio:

$$\left(\frac{G_A}{G_V}\right)_n = -1.274 \pm 0.003,$$

which differs substantially from $(G_A/G_V)_t$ (see Fig. 3). If this result is corroborated, then one can assume that the axial-vector interaction is partly suppressed in the presence of pion exchange in the triton.

7. Determining the lifetime of the free neutron

The decay of the neutron belongs (just as the decay of the tritium nucleus does) to the class of allowed transitions, i.e., the half-life $(T_{1/2})_n$ of the free neutron can be represented, via equation (6.1), as follows

$$(T_{1/2})_n = \frac{k/G_V^2}{f_n [|M_V|_n^2 + (G_A^2/G_V^2) |M_A|_n^2]}, \quad (7.1)$$

where f_n is the phase-space factor, while $(M_V)_n$ and $(M_A)_n$ are, respectively, the vector and axial-vector matrix elements of the neutron beta-decay Hamiltonian. Allowing for the fact that the values of the matrix elements of this Hamiltonian are known exactly, namely $|M_V|_n = 1$ and $|M_A|_n = \sqrt{3}$, we can express the half-life of the neutron in the form of a fraction linking equation (6.1) for the case of the beta decay of the triton with equation (7.1) as follows:

$$(T_{1/2})_n = \frac{(fT_{1/2})_t [1 + |M_A|_t^2 (G_A/G_V)_t^2]}{f_n [1 + 3(G_A/G_V)_n^2]}. \quad (7.2)$$

If we allow for the proton recoil energy, the energy of the $n \rightarrow p$ transition, which according to Ref. [3] is given by

$$m_n c^2 - m_p c^2 = (782333 \pm 6) \text{ eV} + m_e c^2,$$

assumes the value

$$(E_0)_n = 781582 \pm 6 \text{ eV};$$

here, the phase–space integral taken on the basis of a method developed by Wilkinson [51] and allowing for radiative corrections of order α and for the finiteness of the nuclear size and mass, assumes the value of $f_n = 1.71487 \pm 0.00015$.

On the assumption that the weak-interaction coupling constant ratio

$$\left(\frac{G_A}{G_V}\right)_n = \left(\frac{G_A}{G_V}\right)_t = \frac{G_A}{G_V} = -1.264 \pm 0.008$$

is of a universal nature, we find with the above values of f_n , $|M_A|_t$, and $(fT_{1/2})_t$ that

$$(T_{1/2})_n = 618.1 \pm 2.7 \text{ s},$$

which corresponds to the mean value of the lifetime of a free neutron with respect to beta decay:

$$\tau_n = 891.7 \pm 3.9 \text{ s}.$$

Since the parameter that determines the sensitivity of $(T_{1/2})_n$ to variations in G_A/G_V in Eqn (7.2) is small, viz.

$$\frac{1}{(T_{1/2})_n} \frac{d(T_{1/2})_n}{d(G_A/G_V)} = 0.018,$$

the error in determining G_A/G_V of order 0.008 leads to an indeterminacy of the value of $(T_{1/2})_n$ that is smaller than 0.1 s.

Allowance for the value of $(G_A/G_V)_n$ from Ref. [50] in calculations of the mean value of the axial-vector-to-vector weak-interaction coupling constant ratio for beta decay of the neutron leads to a situation [52] in which

$$\left\langle \frac{G_A}{G_V} \right\rangle_n = -1.2670 \pm 0.0030$$

differs from $(G_A/G_V)_t$. If we assume that this difference is significant, the value of the neutron half-life $(T_{1/2})_n$, determined by equation (7.2), is 616.1 s, i.e., one finds

$$\tau_n = 888.9 \text{ s}.$$

If we interpret the shift in the value of τ_n caused by the possible difference between $(G_A/G_V)_t$ and $(G_A/G_V)_n$ as the systematic component in the error of the method used to determine τ_n , we can propose the following interval of possible values of the neutron lifetime with respect to beta decay [53]:

$$\tau_n = 890.3 \pm 3.9_{\text{stat}} \pm 1.4_{\text{sys}} \text{ s}.$$

Figure 4 depicts the results of recent measurements of the lifetime of the free neutron, obtained by counting the number of decay acts in a calibrated beam of thermal neutrons [54, 55] and by evaluating the parameters in the decay exponential function for an ensemble of ultracold neutrons (UCN) [56–58]. The data show that the results obtained by the helium-isotope mass-spectrometric method ‘support’ large values of the neutron lifetime and, specifically, the results of beam experiments. Here, one must allow for the fact that the

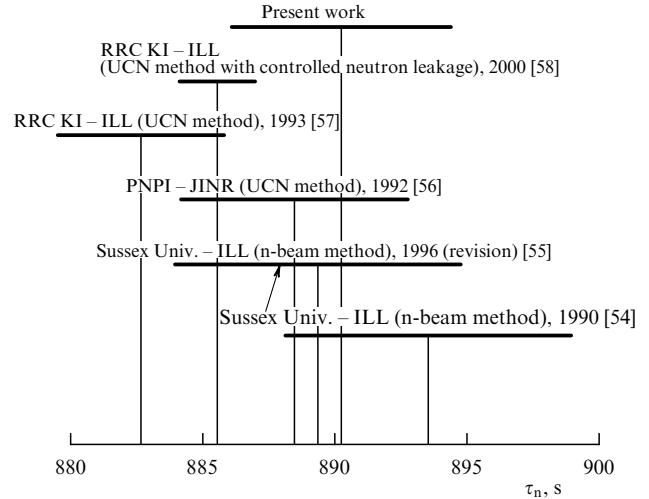


Figure 4. Results of measuring the lifetime of the free neutron in experiments performed in the last 12 years.

measurements of τ_n in UCN experiments are ‘burdened’ by a systematic error (which is difficult to pinpoint) appeared due to neutron leakage from the gravitational traps, which leads to a decrease in the observed values of τ_n . To reduce the loss of ultracold neutrons as they interact with the trap walls in the planned experiments on measuring τ_n , Ezhov et al. [59] proposed using traps with a magnetic barrier, which makes it impossible for neutrons to come into contact with the walls.

8. Conclusions

(1) A new, helium-isotope, method has been developed for measuring the decay constant of a triton being incorporated into various tritium-containing compounds. What makes this method so appealing is that with it there is no need to measure the absolute values of physical quantities (temperature, pressure, volume, and concentration of various gases), which makes it possible to achieve high accuracy and reliability of the results at relatively short exposure times of the tritium-containing compounds under investigation.

(2) A differential variant of the helium-isotope technique has been developed. The technique is used to measure the difference in the values of the half-life of a triton in various compounds. This shows that it is possible to develop a new approach to analyzing the electronic states of atomic–molecular systems, with the analysis based on using an intramolecular electron probe — the tritium nucleus. The first experimental value of the difference of half-lives for molecular and atomic tritium has been obtained.

(3) The data of the thoroughly developed theory of atomic effects that accompany the decay of a free triton and the decay of the triton in the tritium atom have been combined with the results of helium-isotope measurements to obtain values of the half-life of the free triton with and without allowance for decay into bound states, which made it possible to submit an exact estimate of the comparative half-life of the triton.

(4) The axial-vector-to-vector weak-interaction coupling constant ratio for the three-nucleon tritium nucleus has been determined for the first time. Good agreement between this finding and the value of the given ratio for the neutron suggests the ratio G_A/G_V is invariant with respect to the presence of a strong interaction of the pion-exchange type in

beta-active nuclei, which makes it possible to interpret G_A/G_V as a universal fundamental constant characterizing beta processes.

(5) The data on beta decay of the triton have been used to determine the neutron lifetime with respect to beta decay. Comparison of this value of the neutron lifetime with the results of determining τ_n through measurements of the parameters in the exponential decay function for an ensemble of ultracold neutrons and by counting the number of decay acts in a calibrated beam of thermal neutrons supports the ‘high’ values of τ_n .

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References

1. Pauli W, in *Aufsätze und Vorträge über Physik und Erkenntnistheorie* (Braunschweig: F Vieweg, 1961) [Translated into Russian, in *Teoreticheskaya Fizika XX veka* (Theoretical Physics of the Twentieth Century) (Ed. Ya A Smorodinskiĭ) (Moscow: IL, 1962) p. 386]
2. Fermi E *Collected Papers* Vol. 1 (Chicago: Univ. of Chicago Press, 1962) [Translated into Russian (Moscow: Nauka, 1971)]
3. Mohr P J, Taylor B N *J. Phys. Chem. Ref. Data* **28** 1713 (1999)
4. Gaponov Yu V, Mostovoi Yu A *Yad. Fiz.* **63** 1432 (2000) [*Phys. At. Nucl.* **63** 1356 (2000)]
5. Akulov Yu A et al. *Zh. Tekh. Fiz.* **57** 291 (1987) [*Sov. Phys. Tech. Phys.* **32** 175 (1987)]
6. Mamyrin B A et al. *Prib. Tekh. Eksp.* (2) 151 (1982)
7. Mamyrin B A et al. *Vopr. At. Nauki Tekhn. Ser. Radiats. Tekh.* (1) 30 (1986)
8. Akulov Yu A et al. *Prib. Tekh. Eksp.* (2) 173 (1985)
9. Akulov Yu A, Mamyrin B A, Shikhaliev P M *Prib. Tekh. Eksp.* (3) 114 (1995) [*Instrum. Exp. Techn.* **38** 364 (1995)]
10. Mamyrin B A, Tolstikhin I N *Izotopy Geliya v Prirode* (Helium Isotopes in Nature) (Moscow: Energoatomizdat, 1981) [Translated into English (Amsterdam: Elsevier, 1984)]
11. Jones W M *Phys. Rev.* **100** 124 (1955)
12. Eichelberger J F, Grove G R, Jones L V, Mound Laboratory Report MLM-1160 (Miamisburg, OH: Monsanto Res. Corp., 1963)
13. Jordan K C, Blanke B C, Dudley W A *J. Inorg. Nucl. Chem.* **29** 2129 (1967)
14. Unterweger M P et al. *Int. J. Appl. Radiat. Isot.* **31** 616 (1980)
15. Jones P M S *J. Nucl. Mater.* **21** 239 (1967)
16. Oliver B M, Farrar H, Bretscher M M *Int. J. Appl. Radiat. Isot.* **38** 959 (1987)
17. Rudy C R, Jordan K C, Mound Laboratory Report MLM-2458 (Miamisburg, OH: Monsanto Res. Corp., 1977)
18. Akulov Yu A et al. *Pis'ma Zh. Tekh. Fiz.* **14** 940 (1988) [*Sov. Tekh. Phys. Lett.* **14** 416 (1988)]
19. Budick B, Chen J, Lin H *Phys. Rev. Lett.* **67** 2630 (1991)
20. Akulov Yu A, Mamyrin B A, Shikhaliev P M *Pis'ma Zh. Tekh. Fiz.* **19** (18) 72 (1993) [*Tekh. Phys. Lett.* **19** 594 (1993)]
21. Akulov Yu A, Mamyrin B A, Shikhaliev P M *Zh. Tekh. Fiz.* **67** (5) 140 (1997) [*Tech. Phys.* **42** 584 (1997)]
22. Akulov Yu A, Mamyrin B A *Pis'ma Zh. Eksp. Teor. Fiz.* **68** 105 (1998) [*JETP Lett.* **68** 175 (1998)]
23. Bergkvist K-E *Phys. Scripta* **4** 23 (1971)
24. Kaplan I G, Smelov G V, Smutnĭi V N *Phys. Lett. B* **161** 389 (1985)
25. Froelich P et al. *Phys. Rev. Lett.* **71** 2871 (1993)
26. Drukarev E G *Phys. Rev. C* **54** 3277 (1996)
27. Froelich P, Saenz A *Phys. Rev. Lett.* **77** 4724 (1996)
28. Bianconi A, Marcelli A *Synchrotron Radiation Research* Vol. 1 (New York: Plenum Press, 1992)
29. Misra B, Sudarchan E C G *J. Math. Phys.* **18** 756 (1977)
30. Sherk P M *Phys. Rev.* **75** 789 (1949)
31. Bahcall J N *Phys. Rev.* **124** 495 (1961)
32. Tikhonov V N, Chukreev F E *Vopr. At. Nauki Tekhn. Ser. At. Mater.* (1) 12 (1980)
33. Budick B *Phys. Rev. Lett.* **51** 1034 (1983)
34. Harston M R, Pyper N C *Phys. Rev. A* **48** 268 (1993)
35. Haxton W S *Phys. Rev. Lett.* **55** 807 (1985)
36. Akulov Yu A, Mamyrin B A *Pis'ma Zh. Tekh. Fiz.* **26** (11) 23 (2000) [*Tech. Phys. Lett.* **26** 458 (2000)]
37. Lucas L, Unterweger M J. *Res. Natl. Inst. Stand. Technol.* **105** 541 (2000)
38. Budick B, Chen J, Lin H *Phys. Rev. Lett.* **67** 2626 (1991)
39. Kawakami H et al. *Phys. Lett. B* **256** 105 (1991)
40. Wilkinson D H, Macefield B E F *Nucl. Phys. A* **232** 58 (1974)
41. Boris S et al. *Phys. Rev. Lett.* **58** 2019 (1987)
42. Fritsch M et al. *Phys. Lett. B* **173** 485 (1986)
43. Staggs S T et al. *Phys. Rev. C* **39** 1503 (1989)
44. Blin-Stoyle R *Fundamental Interactions and the Nucleus* (Amsterdam: Elsevier, 1973) [Translated into Russian (Moscow: Mir, 1976)]
45. Saito T-Y et al. *Phys. Lett. B* **242** 12 (1990)
46. Bopp P et al. *Phys. Rev. Lett.* **56** 919 (1986)
47. Liaud P et al. *Nucl. Phys. A* **612** 53 (1997)
48. Erozhimskii B G et al. *Yad. Fiz.* **61** 572 (1998) [*Phys. At. Nucl.* **61** 507 (1998)]
49. Mostovoi Yu A et al. *Yad. Fiz.* **64** 2040 (2001) [*Phys. At. Nucl.* **64** 1955 (2001)]
50. Abele H et al. *Phys. Lett. B* **407** 212 (1997)
51. Wilkinson D H *Nucl. Phys. A* **377** 474 (1982)
52. Hagiwara K et al. (Particle Data Group) *Phys. Rev. D* **66** 010001 (2002)
53. Akulov Yu A, Mamyrin B A, in *Fizika Atomnogo Yadra i Elementarnykh Chastits. Materialy XXXV Zimnei Shkoly PIYaF* (Physics of the Atomic Nucleus and Elementary Particles. Proc. of the 35th Winter Workshop of the B P Konstantinov St. Petersburg Institute of Nuclear Physics) (St.-Petersburg: PIYaF Press, 2001) p. 81
54. Byrne J et al. *Phys. Rev. Lett.* **65** 289 (1990)
55. Byrne J *Europhys. Lett.* **33** 187 (1996)
56. Nesvizhevskii V V et al. *Zh. Eksp. Teor. Fiz.* **102** 740 (1992) [*Sov. Phys. JETP* **75** 405 (1992)]
57. Bondarenko L N et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **64** 382 (1996) [*JETP Lett.* **64** 416 (1996)]; Mampe W et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **57** 77 (1993) [*JETP Lett.* **57** 82 (1993)]
58. Arzumanov S et al. *Nucl. Instrum. Meth. A* **440** 511 (2000)
59. Ezhov V F et al., Preprint No. 2422 (St.-Petersburg: PIYaF Press, 2001)