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Application of mass spectrometry to the study of intranuclear processes

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(1) **Mamyrin B A, Akulov Yu A** (A F Ioffe Physical-Technical Institute, RAS, St.-Petersburg) “Application of mass spectrometry to the study of intranuclear processes”;

(2) **Ryazanov V V, Oboznov V A, Bol’ginov V V, Prokof’ev A S, Feofanov A K** (Institute of Solid-State Physics, RAS, Chernogolovka) “Superconductivity currents through a ferromagnet. Phase inversion in structures with Josephson π -junctions”.

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Application of mass spectrometry to the study of intranuclear processes

B A Mamyrin, Yu A Akulov

1. Introduction

The subject of consideration in our report is the interaction between the β -electron generated by an atomic nucleus and the electron ensemble of the atomic-molecular system surrounding the β -active nucleus. According to the Fermi theory of β -decay, the probability of electron emission by nuclei and the electron spectrum depend on the structure of the electron phase space in the atomic-molecular system, the space accessible to the β -electron being formed. At the same time, the experimental values of basic β -decay parameters, in particular, the disintegration constant λ , turn out to be sensitive to the state of the electron environment of the nucleus (the so-called chemical shift effect of the nuclear characteristics). The consideration of a tritium-bearing atomic-molecular system is most promising with respect to the solution of experimental and theoretical problems in the area of exploration of chemical shifts in the β -decay because the energy released in the β -decay of a tritium nucleus (a triton) is relatively low (${}^3\text{H}^+ \rightarrow {}^3\text{He}^{++} + e^- + \bar{\nu} + 18.6 \text{ keV}$). In this case, a significant fraction of the β -electrons are

generated with wavelengths typical of atomic electron systems, making the interaction of the β -electrons with orbital electrons and electron vacancies efficient and resulting in measurable changes of the nuclear disintegration constant.

On the basis of experimental data on the chemical shifts of the disintegration constant in compounds with a known state of the electron environment of the nucleus, the shift value can be represented as a function of electron phase space in the atomic-molecular system, which is within reach for a β -electron. Such a representation of the value of the disintegration-constant chemical shift can underlie a new method for the diagnostics of atomic-molecular systems. In this diagnostic method, the electronic state of some tritium-bearing system is identified by determining the relative difference of the disintegration constants in the system under study and some reference tritium compound with a known electronic structure (for instance, the tritium atom or molecule). In the realm of nuclear physics, determining atomic-molecular corrections to the value of the disintegration constant is required for the calculation of the ratio between the axial-vector and vector weak interaction constants in circumstances where the pion exchange shows up in the three-nucleon tritium nucleus, as well as for obtaining an independent estimate of the free-neutron lifetime from the data on the β -decay of the triton.

2. Experimental technique

According to preliminary theoretical estimates, the relative chemical shifts of the tritium disintegration constant do not exceed 1%. To precisely measure so small a variation of the decay rate, a helium isotopic technique has been developed in the Laboratory of Mass Spectrometry of the Physical-Technical Institute, RAS [1]. The essence of the technique is as follows. For two samples, each containing ${}^4\text{He}$ and some chemical compounds of tritium, a comparison is made of the growth rates of radiogenic ${}^3\text{He}$ -to- ${}^4\text{He}$ content ratios. A significant advantage of this approach is the complete elimination of absolute measurements and a substantial simplification of the inclusion of helium loss, thereby removing the main sources of systematic errors.

The experimental scheme implies the preparation of two samples of initially identical composition and the subsequent action upon one of them (say, the first one) with the aim of changing the electron environment of the tritium nucleus. In this case, the equation governing the measurement of the chemical shift $\Delta\lambda = \lambda_1 - \lambda_2$ of the disintegration constant is of the form

$$\frac{({}^3\text{He}/{}^4\text{He})_1}{({}^3\text{He}/{}^4\text{He})_2} = \frac{t_1}{t_2} \left(\frac{p\Delta\lambda}{\lambda_2} + 1 \right).$$

Here, $(^3\text{He}/^4\text{He})_{1,2}$ are the helium isotope content ratios measured by mass spectrometry in the first and second samples, while t_1 and t_2 are the times of sample exposure. The dimensionless parameter p defines the fraction of disintegrations in sample 1 that do occur with a constant λ_1 responsible for the triton disintegration with a modified electron environment. The amount of radiogenic ^3He sufficient for conducting high-precision mass-spectrometer measurements of the $^3\text{He}/^4\text{He}$ isotopic ratio is equal to $10^{12} - 10^{13}$ atoms and can be accumulated in a time period on the order of 3–5 h in a sample with an activity of $(2-3) \times 10^8$ Bq. The fact that the requisite time of exposure amounts to several hours makes it possible to take advantage of a wide variety of methods to modify the electron environment of the triton. Among them are the ionization and dissociation of the tritium-bearing compounds in a gas discharge, isotope hydrogen exchange reactions, and selective excitation of electrons by specific-energy photons.

The helium isotopic technique was applied to the measurement of the relative difference of the β -decay constants for atomic and molecular tritium. In this case, two ampoules of molybdenum glass were filled with the same mixture of molecular tritium (93 Pa) and ^4He (6 Pa). After sealing the ampoules off, the energy of an RF discharge effected the dissociation of molecular tritium in one of the ampoules. The extraction of tritium from the samples prior to the measurements of helium isotope ratios took place in the course of interaction of the gas mixtures with palladium membranes. The level of residual activity in the samples was monitored with the aid of internal-filling proportional counters with a minimal detectable activity of ~ 0.002 Bq (i.e., 10^6 tritium atoms in the counter volume). The helium isotope ratios in the ‘atomic’ $(^3\text{He}/^4\text{He})_a$ and ‘molecular’ $(^3\text{He}/^4\text{He})_m$ samples, which were close to 0.001 in magnitude, were measured employing an MI-1201 modified static double-beam isotope mass spectrometer. The repeatability of relative measurements of the $^3\text{He}/^4\text{He}$ isotopic ratio corresponded to a statistical error $\delta \sim 0.02\%$ [2].

Possible systematic errors which could arise from the interaction of tritium and helium with the structural materials of the facility were estimated in a series of dedicated experiments. To measure the isotope ratios and the absolute amount of helium in these experiments, advantage was taken of a double-beam magnetic resonance mass spectrometer developed in the Physical-Technical Institute, RAS, which enabled the detection of the following minimal amounts of the helium isotopes: 3×10^4 atoms of ^3He , and $\sim 10^9$ atoms of ^4He [3].

To determine the amount of helium soluble in glass during the sealing-off, an ampoule similar to those employed in the real experiment was filled with ^4He up to a pressure of 6 Pa and sealed off. Immediately afterwards, the portion of the tube subjected to heating during the sealing-off was broken off and then degassed by way of heating to the state of complete melting in an extraction metal furnace. The ^4He content in the glass, determined in a series of these experiments, was found to equal $(2.0 \pm 0.4) \times 10^{11}$ atoms, which accounts for 0.0007% of its amount in the ampoule. The fragments of the ampoules which had contained molecular tritium at a pressure of 93 Pa were heated to the state of complete melting in the extraction quartz furnace. The heating showed that in the sealing-off the glass captured less than 0.0003% of the tritium contained in the ampoule. To measure the tritium content in the samples, use was made of

the low-background proportional counters mentioned above. Along with the helium capture from the volume by the glass, in the sealing-off there occurs a release of the atmospheric helium dissolved in the glass into the ampoule volume (the ^4He concentration in the lower atmosphere is 1.42×10^{14} atom cm^{-3} , the isotope ratio is $^3\text{He}/^4\text{He} = 1.399 \times 10^{-6}$ [3]). To determine the magnitude of this effect, several empty ampoules were sealed off, following which their content was analyzed. As a result, it was determined that $(2.7 \pm 0.3) \times 10^{10}$ atoms of ^4He and less than 10^5 atoms of ^3He are released into the ampoule volume in the sealing-off. This addition of the helium isotopes would change the $^3\text{He}/^4\text{He}$ isotope ratio in the ampoule by less than 0.0001%.

The effect of ^3He implantation into the ampoule wall due to the recoil energy (up to 3.5 eV) acquired by the ^3He nucleus in the β -decay was investigated as follows. The ampoule was filled with molecular tritium up to a pressure of 93 Pa, sealed off, and kept at liquid-nitrogen temperature (when the thermal diffusion of helium in glass is virtually absent) for 74 h. The ampoule was broken at the liquid-nitrogen temperature, and therefore the glass did not interact with ^3He at room temperature. The ^3He content in glass, measured eight minutes after breaking the ampoule, was equal to 0.0008% of its amount formed in 74 h. In the real experiment, 15 min were allocated to ampoule glass degassing after breaking. During this period, the helium released from the glass mixed with the helium which was in the gaseous phase.

To determine the shifts of isotope ratios in the ampoules, arising from the helium dissolving in glass at the gas-discharge temperature, an experiment was conducted, which repeated the real one. It was different in that measurements were made not of helium isotope ratios in the gaseous phase but of the amounts of ^3He and ^4He dissolved in glass during the time of exposure. It was determined that 0.09% of the initial ^4He and 0.07% of the radiogenic ^3He produced during the time of exposure were dissolved in glass, both in the ‘atomic’ and ‘molecular’ samples. The amount of ^3He dissolved in glass is smaller, because the effective time that it remains in the ampoule is shorter. This effect should increase the helium isotope ratio in the gaseous phase by 0.02% in each of the ampoules. In this case, however, the value of $(^3\text{He}/^4\text{He})_a/(^3\text{He}/^4\text{He})_m$ for the gaseous phase will change by less than 0.002% because the values of the $^3\text{He}/^4\text{He}$ isotope ratio measured accurate to 3% in the helium dissolved in glass were, to within this error, the same for the ‘atomic’ and ‘molecular’ samples.

The findings of these check experiments give grounds to believe that the relative change of helium isotope ratios, arising from the interaction of the gas mixtures with the structural elements of the facility, is not greater than 0.1 δ . That is why we introduced no special corrections to account for the nonradiogenic change of the helium isotope ratios in the ‘atomic’ and ‘molecular’ samples. Relying on five series of measurements of the helium isotope ratios, for the relative difference of the triton disintegration constants in atomic and molecular tritium we obtained a value of $\Delta\lambda_{am}/\lambda_m = 0.00257 \pm 0.00045$ [4]. Therefore, for the first time it has been possible to experimentally record the variation of the tritium nucleus lifetime under the changes in its electron environment. To calculate the absolute value of the difference $\Delta\lambda_{am}$ between the disintegration constants, as well as the difference between the decay periods ($T_{1/2} = \ln 2/\lambda$) for molecular and atomic tritium, namely

$(\Delta T_{1/2})_{\text{ma}} = (T_{1/2})_{\text{m}} - (T_{1/2})_{\text{a}}$, use was made of the value of $(T_{1/2})_{\text{m}} = (12.296 \pm 0.017) \text{ y}$ — the weighted mean of two most recently published estimates of the decay period (half-life) for molecular tritium, which are in close agreement. One of these estimates of the decay period was obtained by the helium isotopic technique [5], and the other from the decay curve plotted by way of recording the bremsstrahlung [6]. For the absolute difference between the disintegration constants we obtain

$$\Delta\lambda_{\text{am}} = (0.00257 \pm 0.00045) \frac{\ln 2}{(T_{1/2})_{\text{m}}} \\ = (4.6 \pm 0.8) \times 10^{-12} \text{ s}^{-1}.$$

We take into account that $(\Delta T_{1/2})_{\text{ma}} / (T_{1/2})_{\text{a}} = \Delta\lambda_{\text{am}} / \lambda_{\text{m}}$ to obtain

$$(\Delta T_{1/2})_{\text{ma}} = 0.03152 \pm 0.00553 \text{ y} = 11.5 \pm 2.0 \text{ d}.$$

Carrying out experiments to determine the chemical shifts of the tritium nucleus disintegration constant in more complex compounds requires ensuring a fast — in comparison with the expected changes of the decay period — introduction of tritium into the compounds under investigation. This called for the development of a procedure for producing tritium-bearing compounds with a prescribed level of β -activity in reactions with the participation of free tritium atoms obtained under RF gas-discharge conditions to ensure a nearly 100% degree of dissociation of molecular tritium. As of now, tritiated compounds of several hydrogen halides and hydrocarbons have been produced. This allows us to undertake next-stage experiments on measuring the differences between the values of the disintegration constant for the triton incorporated into these compounds and the disintegration constant for the triton in molecular tritium.

The determination of the tritium nucleus lifetime for several known electronic states of the environment should be interpreted as the calibration of a natural electron probe having a nuclear locality but generating electrons with an atomic wavelength. A peculiarity and advantage of the technique for investigating tritium-bearing atomic-molecular systems with the aid of the β -electrons of their own nuclei are the perfect spatial correlation between the coordinates of the object of probing (the electron shell) and the radiation source (the nucleus). This enables experimental investigations not only of the energy parameters of the electron ensemble, but of its spatial configuration as well, i.e., performance of the experiments that provide answers to the questions: where are boundary orbital electrons stationed? how are chemical bonds directed relative to the nuclei? The tritium electron probe possesses a combination of properties which arise from the special features of β -decay: (i) owing to parity nonconservation in the β -decay, the momentum of β -electrons is directed primarily in opposition to the nuclear spin; (ii) the attendant antineutrino fundamentally has a definite helicity (the antineutrino spin is parallel to the momentum), the β -electron also acquiring a definite helicity in this case, and (iii) the β -electron energy spectrum unperturbed by atomic effects has an exact analytical representation in the energy range from 0 to the 18.6-keV limiting energy.

To summarize the preceding, it may be concluded that in the β -decay of the tritium nucleus in some atomic-molecular system there forms a collimated beam of polarized electrons with a known energy spectrum, i.e. the set of parameters of

the initial electron flux, required for determination of the electron structure of the system under investigation, is completely determined.

3. Determination of the constants characterizing β -processes

Until recently, attempts to determine the values of nuclear constants characterizing the β -processes from the data on tritium β -decay did not meet with success, because the experimentally obtained values of the decay period and limiting β -spectrum energy were determined by the type of tritium chemical compound employed. This gave no way of matching the experimental values of the decay period and limiting β -spectrum energy and impeded exact calculations of the reduced (comparative) decay period of a free triton — a basic characteristic of the β -transition employed in the calculation of the constants.

The problem of the influence of triton's electron environment on the value of the decay period was theoretically considered only for the simplest atomic structures: ^3H , $^3\text{H}^+$, and $^3\text{H}^-$ [7]. It was possible to determine the value of the decay period (half-life) for atomic tritium in an experiment on measuring the difference between the disintegration constants for atomic and molecular tritium: $(T_{1/2})_{\text{a}} = (T_{1/2})_{\text{m}} - (\Delta T_{1/2})_{\text{ma}} = 12.264 \pm 0.018 \text{ y}$. With the experimental value of the decay period for atomic tritium at our disposal, advantage can be taken of the available theoretical data on the atomic effects in the β -decay in the ^3H atom to find the absolute value of the decay period $(T_{1/2})_{\text{t}}$ of a free triton. In going over to the value of a triton's half-life it is necessary to consider four possible processes determining the interaction of the β -electron resided in the final state in the disintegration of the nucleus in a free tritium atom: the decay to the bound states, i.e., the production of the β -electron in one of the shells in the helium atom; the production of the electron in the continuum due to replacement of the orbital electron by the β -electron; the screening of nuclear charge by the orbital electron, and the production of excited electronic states in the $^3\text{He}^+$ ion as a result of the β -decay. The resultant value of the four corrections to the value of $(T_{1/2})_{\text{a}}$ taking into account the above atomic effects is $0.86 \pm 0.08\%$, which leads to a value of free-triton decay period $(T_{1/2})_{\text{t}} = 12.369 \pm 0.020 \text{ y}$, or $(3.9034 \pm 0.0063) \times 10^8 \text{ s}$ [8]. Such a value corresponds to the free-triton decay with direct production of β -electrons in the continuum, when the shape of the β -spectrum is affected only by the Coulomb field of the nucleus. This value of decay period should be used, in particular, in the determination of the energy release in a tritiated plasma arising from the β -decay, because virtually all released energy in the decay to the bound states is carried away by antineutrinos. Determining the triton decay period without the inclusion of decay to the bound states made it possible to employ, when calculating the reduced decay period, the difference of tritium and ^3He nuclear masses found with a high degree of accuracy. This enabled calculation of the reduced triton decay period with a small error and furnished an instrument for estimating the value of $(G_{\text{A}}/G_{\text{V}})_{\text{t}}$ — the ratio between the axial-vector and vector weak interaction constants for the β -decay of the triton — as well as of the free-neutron lifetime τ_{n} which is determined from the equations that relate the reduced decay periods of the neutron and triton to the known matrix elements of the β -decay Hamiltonian of these nuclei [9].

The critical parameter in this method of τ_n determination is the values of G_A/G_V for the triton and neutron. This ratio for the triton, obtained using the quantity $(T_{1/2})_t$, was found to be $(G_A/G_V)_t = -1.2646 \pm 0.0035$. For the mixed super-allowed $n \rightarrow p$ transition, the value of $(G_A/G_V)_n$ is determined from the measurement data for the coefficient which characterizes the asymmetry of β -electron escape relative to the spin of the decaying neutron. The results of four such experiments [10–13] have the weighted mean $\langle(G_A/G_V)_n\rangle_4 = -1.2637 \pm 0.0022$, which agrees well with the value of $(G_A/G_V)_t$, allowing the ratio G_A/G_V to be considered as a universal fundamental constant characterizing the β -processes. Assuming that the values of G_A/G_V for the triton and neutron are equal, we arrive at the following estimate of the neutron lifetime: $\tau_n = 891.7 \pm 3.9$ s. At the same time, it is pertinent to note that Abele et al. [14] obtained a value of $(G_A/G_V)_n = -1.274 \pm 0.003$ for the ratio between the axial-vector and vector coupling constants for the β -decay of the neutron. This value is significantly different from $(G_A/G_V)_t$. The inclusion of this result when determining the weighted mean ratio G_A/G_V for the neutron gives $\langle(G_A/G_V)_n\rangle_5 = -1.2670 \pm 0.0030$ [15]. The difference between $(G_A/G_V)_t$ and $\langle(G_A/G_V)_n\rangle_5$ may be testimony to a partial suppression of the axial-vector interaction in the presence of pion exchange in the triton. Under this assumption we obtain $\tau_n = 888.9$ s. Therefore, the summary result can be represented in the form $\tau_n = (890.3 \pm 3.9_{\text{stat}} \pm 1.4_{\text{syst}})$ s, where the third term defines the systematic error arising from the uncertainty as to the effect of strong interaction in the form of pion exchange on the weak interaction in the β -decay [16].

4. Conclusions

The main source of errors in the method of determining τ_n by way of measuring the decay exponent for an ensemble of ultracold neutrons is the free-neutron leakage from gravitational traps. The approach to τ_n determination proposed in our work is void of this source of systematic errors. Acquisition of trustworthy information on the lifetime of the free neutron is important both for theoretical and applied neutron physics. This permits us to develop methods for measuring the concentration of free neutrons in matter reliant on the detection of neutron decay products — that is, methods which do not necessitate a detailed knowledge of the neutron spectrum and the structure of neutron cross sections. The exact knowledge of the neutron decay period (like of the triton decay period) is also required in cosmology — this is demanded when calculating the parameters which characterize the transformation of matter in the universe: the n/p and $^3\text{H}/^3\text{He}$ ratios, as well as the cosmological constants depending on the substance density.

The concept of developing, on the basis of the effect of the chemical shift of the triton disintegration constant, a new technique for the diagnostics of the electronic states of hydrogen-bearing atomic-molecular systems by way of replacing the hydrogen atom with tritium holds promise in the area of molecular physics. The fact that hydrogen is present in many substances as the main component or an admixture makes it possible to introduce tritium into the composition of different structures, thus placing the electron probe directly in the atomic-molecular system under investigation. In doing this, one can count on successfully applying the tritium β -electron diagnostic technique to the solution of a number of physicochemical problems — from the problems

of the shape of hybrid orbitals or the distribution of an unpaired electron in radicals to the problems of charge distribution between a sorbent and an adsorbent, the form of hydrocarbon chemisorption, and the physical nature of hydrogen bonds.

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Superconducting currents through a ferromagnet. Phase inversion in structures with Josephson π -junctions

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1. Introduction

The relationship between a superconducting current I_s and a phase difference φ in superconducting weak links (Josephson junctions) is described by a 2π -periodic function; in the simplest case of a tunnel barrier or a barrier of a dirty normal metal, one finds $I_s = I_c \sin \varphi$, where I_c is the Josephson-junction critical current. Recent experiments have shown that it is possible to realize Josephson structures (π -junctions) with a phase difference π in the ground state, i.e., with an anomalous current — phase relation and a dependence of the weak link energy on the phase difference, which are shifted by a half-period (Fig. 1):

$$I_s = -I_c \sin \varphi, \quad E = E_J(1 + \cos \varphi). \quad (1)$$

Although the term ' π -junction' was introduced in Ref. [1] for superconducting tunnel junctions with magnetic impurities in the tunnel barrier, the superconducting weak links with a