Effect of molecular structure on the β spectrum and the problem of determining the rest mass of the neutrino

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The effect of the molecular structure of the source of the shape of the β spectrum is taken into account in a consistent manner. The probabilities for excitation of β decay of different classes of tritium-containing molecules are calculated. The energy characteristics of the β spectra of these molecules are calculated, including those of the working substance of the β source of the Institute of Theoretical and Experimental Physics group. The many-channel nature of the β decay in molecules and the influence of this feature on the shape of the β spectrum and the problem of determining the neutrino mass are discussed.

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1. INTRODUCTION

The first theoretical studies of the action of β decay of a nucleus on the electron shell of an atom were made by Migdal and Fainberg.[1,2] Theoretical investigations of the restructuring of the electron shell of atoms in β decay were subsequently persued by many workers, see the review by Batkin and Smirnov.[3]

In the case of β decay of a nucleus contained in a molecule, the physical situation is more complicated. The β decay can cause both electron and vibrational-rotational excitations, as well as dissociation of the molecule. In this case, only β decay of the simplest molecule HT was investigated in detail[4,7]; the most accurate calculation was made by Wolniewicz.[7] The influence of β decay of tritium on the excitation of electron shells of the molecule OHT, NH_2T, and CH_3T was investigated in Ref. 8.

Interest in the investigation of the restructuring of the electron shell of the molecule in β decay of one of the nuclei was recently stimulated by the timely problem of determining the neutrino rest mass. The reason is that the measurement of the form of the β spectrum near its high-energy edge still remains the most sensitive method of determining the neutrino rest mass. Experiments of this type are carried out with tritium sources, since tritium has the lowest β-electron end-point energy, $E_{\beta,max} = 18.6$ keV, so that the maximum energy resolution can be obtained.

In their first paper, Hanna and Pontecorvo[9] obtained an upper bound of the order of 1 keV for the neutrino rest mass, a value corresponding to the energy resolution in their method. The entire subsequent progress in the experimental research is due to improvement of the resolution of the measuring instruments. To this day, the best success in this direction was reached by Bergkvist.[10,11] With a magnetic β spectrometer having at the end-point of the β spectrum of tritium a resolution ~ 50 eV, the upper limit obtained for the neutrino mass was ~ 55 kV with 90% confidence level. Bergkvist was the first to reach resolution energies comparable in magnitude with the energies of the excited helium daughter ions obtained via β decay, and pointed out the need for taking into account the influence of these excitations on

the form of the β spectrum and on the estimate of the neutrino mass.

In 1980, data reduction of a set of experiments performed at the Institute of Theoretical and Experimental Physics (ITEP), a lower bound was first obtained on the neutrino rest mass.[12,13] The β-electron source was a doubly tritiated amino-acid - valine ($C_5H_{11}NO_2$). The measurements were made with a unique iron-free spectrometer constructed by Tret'yakov,[14] having an energy resolution ~ 45 eV and a background level 15 times smaller than in the Bergkvist spectrometer. Since the real excitation spectrum of the β source was not known to the authors of Refs. 12 and 13, they reduced the experimental data for two cases:

a) spectrum with one final state, or the absence of excitations in the source; the 99% confidence interval for the neutrino mass was found to be

$$14 \leqslant m_\nu \leqslant 26 \text{ eV};$$

b) atomic tritium in the Bergkvist two-level approximation[10]; the interval obtained was

$$24 \leqslant m_\nu \leqslant 46 \text{ eV}.$$

These results indicate that the neutrino mass obtained by reducing the experimental data depends substantially on allowance for the redistribution of the energy of the β decay among the electron degrees of freedom of the β source. By the same token this raised the question of correct allowance for the influence of the chemical structure of the source on the β spectrum.

Following the publication of the papers by ITEP group[12,13] a number of attempts were reported[15-17] to take into account the influence of the outflow of the β-decay energy into the electron channel on the neutrino rest mass obtained by reducing the experimental β-spectrum data. Since the authors of all the cited papers[15-17] have assumed that the spectrum of the working medium used in the experiments[12,13] cannot be calculated, they started out with models. In Refs. 15 and 16 the source model was atomic tritium. In Ref. 15 it was shown that exact allowance for the continuous spectrum has practically no influence on the obtained neutrino rest mass. Allowance for the molecular surround-

ing was reduced in Ref. 16 to a change in the parameter of the screening of the atomic $1s$ function and it was concluded that an exact molecular calculation is essential for final deductions. The authors of Ref. 17 compared the reduction of the experimental data of Refs. 12 and 13 in the atomic-model approximation, on the one hand, with the reduction for the HT molecule, on the other, and reached the conclusion that the atomic model overestimates the obtained value of the mass. We note in this connection that our similar[17] approach, but with the LiT rather than the HT molecule, did not confirm this overestimate. Reduction by the ITEP group of the experimental data, using the valine spectrum calculated by us, pointed to an opposite tendency.

In the present paper we determined consistently the influence of the molecular structure of the source on the shape of the β spectrum, and calculated the probability of excitation of various molecules containing tritium nuclei that undergo β decay. We discuss the multichannel character of the β decay in the molecules and the influence of this circumstance on the confidence interval for the neutrino rest mass. Preliminary results were published by us in Refs. 18 and 19.

2. ALLOWANCE FOR THE INFLUENCE OF THE MOLECULAR STRUCTURE ON THE FORM OF THE β SPECTRUM

The method proposed by Fermi[20] to calculate the β decay of a nucleus is based on a time-dependent perturbation theory, since the low value of the weak-interaction constant makes it possible to restrict oneself to first order in the theory and to use the so-called Fermi "golden rule":

$$P_{if} = \frac{2\pi}{\hbar} |\langle f|H_\beta|i\rangle|^2 \delta(E_i - E_f).\qquad (1)$$

Here $|i\rangle$ is the state vector of the parent-nucleus nucleons, $|f\rangle$ describes the final system of the daughter nucleus and of the emitted β electron and antineutrinos, and H_β is the weak-interaction Hamiltonian.

Equation (1) is valid also in the case when the radioactive nucleus enters into the composition of the molecule. The perturbations for the molecular degrees of freedom are a jump in the charge of the nucleus that experiences the β decay and the appearance of a recoil momentum. Although these perturbations cannot be regarded as small relative to the molecular interactions, the use of perturbation theory is based on their "instantaneity," since they are produced within times much shorter ($\sim 10^{-19}$ sec) than the characteristic times of the internal motions in the molecule ($\gtrsim 10^{-15}$ sec). Cantwell[5] used Eq. (1) to describe β decay in the HT molecule. Since the weak-interaction Hamiltonian H_β acts only on the nucleon and lepton coordinates, the matrix element in (1) factors out into nuclear and molecular parts:

$$\langle f|H_\beta|i\rangle = M_{\mathrm{nucl}}(\mathbf{p}_\nu,\ \mathbf{p}_\beta) M_{\mathrm{mol}}(\mathbf{p}_\nu + \mathbf{p}_\beta).\qquad (2)$$

The nuclear matrix element determines the distribution of the recoil momenta $\mathbf{P}_{\mathrm{rec}} = \mathbf{p}_\nu + \mathbf{p}_\beta$. Its calculation is the subject of many studies, see Refs. 21–23. The object of interest to us is the molecular matrix element

$$M_{\mathrm{mol}}(\mathbf{P}_{rec}) = \langle \Psi_{\mathrm{mol}}^f|\exp[-i\mathbf{P}_{\mathrm{rec}}\mathbf{R}_k]|\Psi_{\mathrm{mol}}^i\rangle,\qquad (3)$$

where Ψ_{mol}^i and Ψ_{mol}^f are the wave functions of the initial

and final molecules and $\mathbf{R}_k$ is the radius vector of the radioactive nucleus. We assume that the molecule contains N electrons and k nuclei, and assign the last number to the radioactive nucleus. The exponential containing the recoil momentum stems from the wave functions of the leptons on going from the laboratory frame to the c.m.s. of the molecule. We note that the form of Eq. (3) for the molecular matrix element coincides with the expression obtained in Migdal's theory of instantaneous perturbations.[24]

We consider hereafter tritium-containing molecules which we denote by RT. Their β decay corresponds to the reaction

$$\mathrm{RT} \to (\mathrm{RHe})^+ + e^- + \tilde{\nu}_e,\qquad (4)$$

as a result of which are produced a β electron, an antineutrino[1] $\tilde{\nu}_e$, and a molecular complex $(\mathrm{RHe})^+$. The complex $(\mathrm{RHe})^+$ can be produced in different final stages because of the presence of electronic and electron vibrational degrees of freedom, so that the reaction (4) corresponds to a multichannel process. From the energy conservation law we obtain in the case of $0 \to n$ channel the following expression for the kinetic energy of the β electron:

$$E_\beta^{(n)} = \Delta mc^2 - m_e c^2 - E_\nu - E_{\mathrm{rec}} + [E_0(\mathrm{RT}) - E_n(\mathrm{RHe}^+)],\qquad (5)$$

where Δm is the nuclear mass defect, E_{rec} is the recoil energy, E_ν is the total neutrino energy, $E_0(\mathrm{RT})$ is the energy of the ground state of the RT molecule, and $E_n(\mathrm{RHe}^+)$ is the energy of the nth quantum state of the complex $(\mathrm{RHe})^+$. For the maximum kinetic energy of the β electron we obtain

$$E_{\beta,\mathrm{max}}^{(n)} = \Delta mc^2 - m_e c^2 - m_\nu c^2 - E_{\mathrm{rec}} + [E_0(\mathrm{RT}) - E_n(\mathrm{RHe}^+)].\qquad (6)$$

To find the energy going into the molecular degrees of freedom it is necessary to know the distribution of the probabilities of formation of the daughter molecules in different quantum states. The probability of β decay in a molecule in the channel $0 \to n$ is determined by expression (1), in which it is necessary to substitute the matrix element (2) and take into account the energy conservation law (5) for the $0 \to n$ channel:

$$P_{if}(0 \to n) = \frac{2\pi}{\hbar} |M_{\mathrm{nucl}}(\mathbf{p}_\nu, \mathbf{p}_\beta)|^2 |M_{\mathrm{mol}}(\mathbf{P}_{\mathrm{rec}})|^2$$
$$\times \delta(\Delta mc^2 - E_\beta - m_e c^2 - E_\nu - E_{\mathrm{rec}} + [E_0(\mathrm{RT}) - E_n(\mathrm{RHe}^+)]).\qquad (7)$$

In contrast to the analogous expression for the "bare" nucleus, if the nucleus is contained in a molecule, Eq. (7) contains an additional factor $|M_{\mathrm{mol}}|^2$ and an additional term in the energy conservation law, which we shall call the chemical shift in the $0 \to n$ channel and designate by ΔE_{0n}:

$$\Delta E_{0n} = E_0(\mathrm{RT}) - E_n(\mathrm{RHe}^+).\qquad (8)$$

The sign of the chemical shift can either positive or negative. To ascertain whether the electron shell draws energy from the β electron or transfers additional energy to it, it is necessary to find the chemical shift $\overline{\Delta E}$ averaged over all the channels.

We consider now the molecular matrix element. Since we are interested in the probability of the molecular excitations, it is necessary to separate in (3) the motion over the internal degrees of freedom of the molecule from the motion of its mass center. Such a separation is naturally carried out

with the aid of the Jacobi transformation.[25] For simplicity we carried out for the case of a diatomic molecule; the results do not change in the polyatomic case. The matrix element (3) goes over into

$$M_{\text{mol}}(P_{\text{rec}}) = \langle \Psi^f_{\text{mol}}(Q, \dots \xi_i, \sigma_i \dots) | \exp\left[i\frac{M_1}{M} P_{\text{rec}} Q \right]$$

$$\times \exp\left[-i\frac{m_e}{M} P_{\text{rec}} \sum_{i=1}^{N} \xi_i \right] | \Psi^i_{\text{mol}}(Q, \dots \xi_i, \sigma_i \dots) \rangle, \quad (9)$$

where M is the mass of the entire molecule and M_1 is that of the first nucleus; Q and ξ_i are the relative coordinates of the electrons, σ_i is the spin coordinate of the ith electron,

$$Q = R_1 - R_2,$$

$$\xi_i = \left(M_1 R_1 + M_2 R_2 + m_e \sum_{l=1}^{i-1} r_l \right) / [M_1 + M_2 + (i-1)m_e] - r_i,$$

$$i = 1, 2, \dots, N.$$

The first and second exponentials in (9) describe respectively the influence of the recoil on the motion of the nuclei and on the relative motion of the electrons. The small value of the ratio m_e/M makes it possible to neglect the second exponential in (9), meaning also the influence of the recoil on

the electron degrees of freedom. Obviously, this conclusion remains in force also in the case of a polyatomic molecule. Consequently, excitation of the electron degrees of freedom takes place only as a result of a jump in the charge of the radioactive nucleus.

Since the energy going into the vibrational degrees of freedom is low ($\sim 10^{-1}$–10^{-2} eV), the energy of the nth quantum state of the complex $(RHe)^+$ in the argument of the δ function (7) is determined by the energy of the electron excitation. To separate the purely electron states, we use the adiabatic approximation.[26,27] In this approximation, the wave function of the molecule can be represented in the form of the product

$$\Psi_{\text{mol}}(x, Q) = \Psi_n(x, Q)\chi_{nv}(Q - Q_0^n), \quad (10)$$

where x is the aggregate of the spatial and spin coordinates of the electrons, $\Psi_n(x, Q)$ is the electron wave function of the nth quantum state and depends on the relative coordinates Q of the nuclei as parameters, while χ_{nv} is the wave function of the nuclear motion in the nth electronic state and depends on the displacement of the nuclei relative to their equilibrium configuration Q_0^n in the nth electron state.

We substitute in (10) the matrix element (9) (we set the second exponential in the latter equal to unity):

$$M_{\text{mol}} = \left\langle \Psi_n^f(x, Q)\chi_{nv}^f(Q - Q_0^{f,n}) \left| \exp\left[i\frac{M_1}{M} P_{\text{rec}} Q \right] \right| \Psi_m^i(x, Q)\chi_{mv}^i(Q - Q_0^{i,m}) \right\rangle. \quad (11)$$

Here $Q_0^{f,n}$ is the equilibrium configuration of the nuclei in the nth electron state of the daughter molecule, $Q_0^{i,m}$ is the equilibrium configuration of the nuclei in the mth electron state of the parent molecule. To find the relative probability of the electron excitation it suffices to start not from the complete expression (7) but from the molecular matrix element (11), by summing the square of the modulus of (11) over the quantum numbers of the nuclear motion in the final state of averaging over the quantum numbers of the initial state. Using the completeness of the system of the wave functions that describe the nuclear motion, we find

$$W_{m,n} = \sum_{v,w} P_w |M_{\text{mol}}|^2 = \sum_w P_w \langle \chi_{mw}^i(Q - Q_0^{i,m}) | |\langle \Psi_n^f(x, Q) |$$

$$\times \Psi_m^i(x, Q) \rangle|^2 | \chi_{mw}^i(Q - Q_0^{i,m}) \rangle. \quad (12)$$

Recognizing that the nuclear wave function differs from zero only in the vicinity of the equilibrium configuration, while the electron wave function depends little on the nuclear coordinates, we can separate in (12) the electron matrix element, taking it at a fixed $Q = Q_0^{i,m}$; this is equivalent to the Condon approximation widely used in molecular spectroscopy. As a result we obtain for $W_{m,n}$

$$W_{mn} = |\langle \Psi_n^f(x, Q_0^{i,m}) | \Psi_m^i(x, Q_0^{i,m}) \rangle|^2. \quad (13)$$

A nontrivial result with respect to the known Migdal formula for the probability of the excitation of an atom in β decay is the corroboration, in the foregoing derivation, of the choice of the electron wave function of the daughter nucleus in (13) at equilibrium configuration of the parent function,

and the corroboration of the possibility of neglecting the influence of the recoil momentum on the quantity W_{mn}.

To find the form of the β spectrum (7) it is necessary to average over the quantum numbers of the initial stage, integrate over the momentum of the neutrino $dp_\nu = p_\nu^2 dp_\nu d\Omega_\nu$ and over the emission angles $d\Omega_\beta$ of the β electron, as well as sum over the final quantum stages of the daughter nucleus. With allowance for (12) and (13), the form of the β spectrum can be given by the expression

$$dP(p_\beta) = \frac{2\pi}{\hbar} p_\beta^2 dp_\beta \int d\Omega_\beta \int d\Omega_\nu dp_\nu p_\nu^2 |M_{\text{nucl}}(p_\beta, p_\nu)|^2 \sum_n W_{0n}$$

$$\times \delta(\Delta mc^2 - E_\beta - m_e c^2 - E_\nu - E_{\text{rec}} + \Delta E_{0n}), \quad (14)$$

where the sum over n in (14) includes also integration over the states of the continuous spectrum. To carry out the integration in (14), we neglect in the conservation law the small value of the recoil energy E_{rec} and separate in M_{nucl} the kinematic factor[28,29]:

$$M_{\text{nucl}}(p_\beta, p_\nu) = M'_{\text{nucl}}\left[1 + \alpha\frac{v_\beta}{c}\cos(\widehat{p_\nu, p_\beta}) \right]; \quad (15)$$

M'_{nucl} contains the Fermi factor $F(E_\beta)$,[29] which takes into account the influence of the molecule field on the β-electron motion. Making the change of variables

$$p_\nu^2 dp_\nu = c^{-3}(E_\nu^2 - m_\nu^2 c^4)^{1/2} E_\nu dE_\nu$$

and substituting

$$p_\beta^2 dp_\beta = c^{-2} p_\beta(E_\beta + m_e c^2) dE_\beta,$$

we obtain

$$dP(p_\beta) = \sum_n W_{0n} \{Ap_\beta(E_\beta + m_e c^2) |M'_{\text{nucl}}(m_v, E_\beta)|^2$$

$$\times [(\Delta mc^2 - E_\beta - m_e c^2 + \Delta E_{0n})^2 - m_v^2 c^4]^{1/2}$$

$$\times (\Delta mc^2 - E_\beta - m_e c^2 + \Delta E_{0n})\} dE_\beta, \quad (16)$$

where A is a numerical factor. The expression in the curly brackets differs from the β spectrum for the bare nucleus in the presence of the chemical shift ΔE_{0n} in each channel.[2] In the presence of an electron shell, the β spectrum is the weighted sum of such shifted spectra with weights equal to the probabilities of excitation of the electron shell.

Far from the end point of the β spectrum, the quantity $m_v^2 c^4$ in the square brackets of (16) can be neglected. In place of (16) we obtain

$$dP(p_\beta) = \sum_n W_{0n} [Ap_\beta(E_\beta + m_e c^2) |M'_{\text{nucl}}|^2$$

$$\times (\Delta mc^2 - E_\beta - m_e c^2 + \Delta E_{0n})^2] dE_\beta. \quad (17)$$

By elementary algebraic manipulations we can represent Eq. (17) in terms of the mean values, over all the channels, of the chemical shift $\overline{\Delta E}$ and of the squared chemical shift $\overline{\Delta E^2}$:

$$\overline{\Delta E} = \sum_{n=0}^\infty W_{0n} \Delta E_{0n}, \quad \overline{\Delta E^2} = \sum_{n=0}^\infty W_{0n} \Delta E_{0n}^2, \quad (18)$$

namely

$$dP(p_\beta) = Ap_\beta(E_\beta + m_e c^2) |M'_{\text{nucl}}|^2$$

$$\times [(\Delta mc^2 - E_\beta - m_e c^2)^2 + 2(\Delta mc^2 - E_\beta$$

$$- m_e c^2) \overline{\Delta E} + \overline{\Delta E^2}] dE_\beta. \quad (19)$$

From the energy conservation law (5) we obtain for the β-electron energy averaged over all the channels

$$\bar{E}_\beta = \sum_{n=0}^\infty W_{0n} E_\beta^{(n} = \Delta mc^2 - m_e c^2 - E_v - E_{\text{rec}} + \overline{\Delta E}. \quad (20)$$

The presence of the electron shell increases the energy of the β electron by an amount equal to the mean chemical shift $\overline{\Delta E}$. In accordance with Migdal's theory of instantaneous perturbations, the electron wave function does not manage to change within the β-decay time, therefore the average chemical shift must be calculated with the electron wave functions of the initial molecule:

$$\overline{\Delta E} = \langle \Psi_0(\text{RT}) | H(\text{RT}) - H(\text{RHe}^+) | \Psi_0(\text{RT}) \rangle$$

$$= E_0(\text{RT}) - \langle \Psi_0(\text{RT}) | H(\text{RHe}^+) | \Psi_0(\text{RT}) \rangle. \quad (21)$$

Recognizing that the average excitation energy of the daughter nucleus $\overline{\Delta E^*}(\text{RHe}^+)$ is defined as

$$\overline{\Delta E^*}(\text{RHe}^+) = \langle \Psi_0(\text{RT}) | H(\text{RHe}^+) | \Psi_0(\text{RT}) \rangle - E_0(\text{RHe}^+), \quad (22)$$

we find the connection between $\overline{\Delta E}$ and $\overline{\Delta E^*}(\text{RHe}^+)$:

$$\overline{\Delta E} = E_0(\text{RT}) - E_0(\text{RHe}^+) - \overline{\Delta E^*}(\text{RHe}^+). \quad (23)$$

Here $\overline{\Delta E^*}$ includes excitations with transition of $(\text{RHe})^+$ into a state with both a discrete and a continuous spectrum [we recall that the sum over n in (16)–(18) contains also integration over the states of the continuous spectrum], i.e.,

$$\overline{\Delta E^*}(\text{RHe}^+) = \overline{\Delta E^*}(\text{RHe}^+)_{\text{discr}} + \overline{\Delta E^*}(\text{RHe}^+)_{\text{ion}}$$

$$= \sum_{n=1}^\infty W_{0n} [E_n(\text{RHe}^+) - E_0(\text{RHe}^+)] + \int_{I_1}^\infty W_{0E_i} E_i dE_i, \quad (24)$$

where $\overline{\Delta E^*}_{\text{discr}}$ and $\overline{\Delta E^*}_{\text{ion}}$ are respectively the average energies of all the transitions into the discrete and continuous spectra, E_i is the energy of the transition into the continuous spectrum, and I_1 is the first ionization potential. We note that to find $\overline{\Delta E^*}$ it is better to use not (24) but the relation (22), inasmuch as in this case the calculation calls for knowledge of only the wave functions of the ground state. The quantity $\overline{\Delta E^*}$ determines the degree of the physical broadening of the β spectrum. In the experiment, the physical broadening should manifest itself only in the immediate vicinity of the end point of the β spectrum. Far from it, the measured resultant β-decay curve (16) is the weighted mean over the electron channels.

The influence of the electron shell on the shape of the β spectrum is conveniently investigated in the Fermi coordinates (Y, E_β), where $Y = (P_n / p_\beta^2 |M_{\text{nucl}}|^2)^{1/2}$, inasmuch as the plot of the β spectrum for each channel in terms of these coordinates is a straight line called the Kurie plot. In the case $m_v \neq 0$ the straight line should bend to the left near the end points. The resultant β spectrum (16), (17) will have a more complicated form. Figure 1 shows Kurie plots calculated by us for the LiT molecule. Although the channels with formation of $(\text{RHe})^+$ in different electron states are not distinguishable in the experiment their presence in the case of sufficiently good energy resolution and of a small background can manifest itself, as follows from Fig. 1b, in inflections on the resultant β-decay curve.
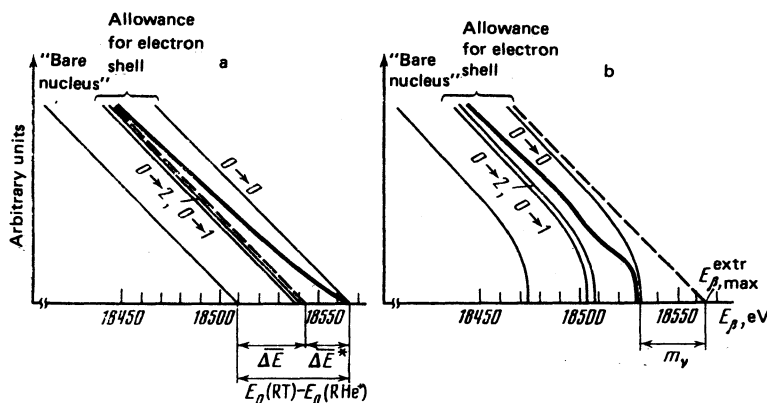

FIG. 1. β spectrum of LiT: a) $m_v = 0$; b) $m_v = 35$ eV. The thin lines are the β spectra in the electron channels $0 \to n$; the thick line is the summary spectrum; the dashed lines are the extrapolated spectrum; $Y = (P_n / p_\beta^2 |M_{\text{nucl}}|^2)^{1/2}$.

The extrapolated end point of the β spectrum is obtained by the intersection of the straight-line Kurie plot and the abscissa axis. From (6) we obtain for the $0 \to 0$ channel

$$E_{\beta,\max}^{\mathrm{extr}} = E_{\beta,\max}^{(0)} + m_\nu c^2 = \Delta mc^2 - m_e c^2 - E_{rec} + [E_0(\mathrm{RT}) - E_0(\mathrm{RHe^+})]. \tag{25}$$

This in fact is, apart from a small quantity E_{rec}, the energy E_0 of Refs. 12–14, meaning the energy released in β decay. Since the separation the channel $0 \to 0$ is so far impossible, the β-spectrum end point extrapolated in the experiment is the mean value of all the electron excitation channels. It differs from (25) by the average excitation energy $\overline{\Delta E^*}(\mathrm{RHe^+})$.

$$\overline{E_{\beta,\max}^{\mathrm{extr}}} = E_{\beta,\max}^{\mathrm{extr}} - \overline{\Delta E^*}(\mathrm{RHe^+}) = \Delta mc^2 - m_e c^2 + \overline{\Delta E} - E_{rec}. \tag{26}$$

3. CALCULATION OF THE PROBABILITY OF ELECTRON EXCITATION OF TRITIUM-CONTAINING MOLECULES IN β DECAY

Determination of the electronic wave functions

It is known that the wave functions of multielectron systems can be obtained only by approximate methods. These methods cannot be semi-empirical, inasmuch as for systems with a helium atom (we consider tritium-containing molecules that become helium-containing as a result of the β decay) there are no worked-out sets of parameters. Therefore to find the wave functions we chose the algebraic variant of the self-consistent field (SCF) method, the so-called Hartree-Fock-Rhouthaan method, abbreviated the MO LCAO method (the molecular orbitals are presented in the forms of linear combinations of atomic orbitals). The calculations of the ground state were carried out in accordance with the "Gaussian-70" program.

The virtual orbitals obtained as a result of solution of Hartree-Fock-Rhouthaan equation describe poorly the excited states, since they correspond to motion of an electron in the field of a neutral N-electron molecule.[30] To find more realistic excited orbitals we set up a program based on the Huzinaga-Arnau approach,[31] where the excited orbitals are obtained as a solution of the single-electron Schrödinger equation with a potential produced by the remaining $(N - 1)$ electrons. Since the obtained orbitals are solutions of one equation, they form automatically an orthonormalized set. This last condition is very important, for only in this case does the overlap integral in (13) determine the probability.

It is important to estimate the influence of the electron correlation, which is not accounted for in the SCF method, on the processes of restructuring of the electron shell in β decay. The published results of the calculation of the transition HT$\to$HHe$^+$ (ground state) with wave functions that take into account to different degrees the correlation of the electrons, seem quite contradictory. In a paper by Schwartz,[4] a calculation with James-Coollidge functions (the function obtained by Toy[32] was used for HHe$^+$) the value $W_{00} = 0.93$. Cantwell[5] carried out a calculation with a rough single-term James-Coollidge function and obtained $W_{00} = 0.73$. When he used a more accurate two-term function he obtained $W_{00} = 0.58$. In the most accurate calculation carried out by Wolniewicz[7] with a 64-term James-Cool-

lidge function, the result was $W_{00} = 0.572$. Such a spread in the values of W_{00}, from 0.93 to 0.572, led to an overestimate of the importance of the correlation[7] and caused a number of workers (see, e.g., Ref. 17) to assume that precision calculations of β decay in systems more complicated than HE are not possible at all.

Yet, as noted back in Ref. 33, Toy's calculation[32] contains a numerical error. Therefore Schwartz's results based on it must be disregarded. The remaining results show that refinement of the James-Coollidge function changes the value of W_{00} in the range from 0.73 to 0.572.

When discussing the influence of correlation, it must be taken into account that in the SCF method the correlation of the electrons is partially taken into account. The influence of the correlation part not taken into account in the SCF method can be smaller than in calculations with the James-Coollidge function. Indeed, calculation for HT, carried out in Ref. 8 in the MO LCAO approximation on a sufficiently long basis yielded a value of W_{00} that differed by only 1.5% from the exact Wolniewicz calculation.[7]

In the present paper we took consistent account of the influence of the electron correlation that was not taken into account in the SCF method on the probability of formation of HHe$^+$ in the ground state. To this end, the probability W_{00} was calculated with wave functions that are superpositions of different numbers of configurations that can be produced when the electrons go from a filled shell of the ground state to vacant orbitals. The basis chosen was H:$1s$, $1s'$ and He:$1s$, $1s'$, $1s''$ from the contracted, according to Dunning, gaussian functions [$2s$] for H and [$3s$] for He,[34,35] with scale factors 1 for H and 2 for He. The results of the calculations are given in Table I. The maximum error due to failure to take into account the electron correlation on a fixed basis is less than 3.5%. This is approximately the size of the error obtained in calculations on other basis. The deviation from the most accurate Wolniewicz calculations[7] is only 1.4%.

The calculation points thus to a small contribution of the electron correlation to the probability of production of the daughter molecules in an electron ground state, and consequently to the total excitation probability. This confirms by the same token the possibility of using the SCF approximation for the problem to be solved.

Derivation of the formula for the probability of electron excitation in the MO LCAO approximation

We obtain not a formula for the calculation of the probability of electronic excitation in the case when the wave

TABLE I. Influence of the electron correlation on the probability that the ion (HHe)$^+$ will remain in the electron ground state.

Number of configurations taken into account		W_{00}	Number of configurations taken into account		W_{00}
HHe+	HT		HHe+	HT	
1	1	0.6183	10	6	0.5896
3	3	0.6115	10	10	0.5875
6	3	0.6141	15	10	0.5860
10	3	0.6030			

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functions in (13) are chosen in the approximation of the molecular-orbitals method.

In the ground state of the initial molecule RT, the electron shell is closed and the total electron spin is $S = 0$. The wave function is a Slater determinant

$$\Psi_0{}^i = (N!)^{-1/2} \det |\varphi_1\alpha, \ \varphi_1\beta, \ \varphi_2\alpha, \ \varphi_2\beta, \dots, \ \varphi_{N/2}\beta|, \quad (27)$$

where φ_i are the occupied MO obtained by us in the form of a linear combination of atomic orbitals in accordance with the "Gaussian-70" program, α and β are the spin wave functions. From Eq. (13) for the transition probability it follows that excitation does not change the total electron spin, and consequently the wave functions of the excited states of the final complex $(RHe)^+$ should correspond to a spin $S = 0$. For single excitations, they are represented in the form of a sum of determinants:

$$\Psi_{k\to l}^f = (2N!)^{-1/2}\{\det|\psi_1\alpha, \ \psi_1\beta, \dots, \ \psi_k\alpha, \ \psi_l^{(k)}\ \beta, \dots, \ \psi_{N/2}\beta|$$

$$+ \det|\psi_1\alpha, \ \psi_1\beta, \dots, \ \psi_l^{(k)}\ \alpha, \ \psi_k\beta, \dots, \ \psi_{N/2}\beta|\}, \quad (28)$$

where ψ_i are the MO occupied in the ground state by the daughter molecule $(RHe)^+$; $\psi_l^{(k)}$ is the excited orbital obtained by the Huzinaga-Arnau method for a daughter molecule in which the electron is excited with a k th occupied orbital on an l th vacant one.

Since the molecular orbitals of the parent and daughter molecules are not orthogonal to each other, since they belong to spectra of different Hamiltonians, it is convenient to calculate the probability (17) with the functions (27) and (28) by using the technique developed in Ref. 36. After a number of algebraic transformations we arrive at the following expressions:

$$W_{0k\to l} = 2D^2 D_{kl}{}^2, \quad W_{00} = D^4, \quad (29)$$

where D stands for a determinant of the MO overlap integrals of the parent and daughter molecules, occupied in the ground states,

$$D = \begin{vmatrix} s_{11} & s_{12} \dots s_{1N/2} \\ s_{21} & s_{22} \dots s_{2N/2} \\ \dots \dots \dots \\ s_{N/21} & s_{N/22} \dots s_{N/2N/2} \end{vmatrix}, \quad (30)$$

$$s_{ij} = \langle \psi_i | \varphi_j \rangle. \quad (31)$$

The determinant D_{kl} is obtained from D by replacing the k th row of the overlap integrals s_{kj} by the row of the overlap integrals S_{lj}:

$$s_{ij}^{(k)} = \langle \psi_i^{(k)} | \varphi_j \rangle. \quad (32)$$

We note that the determinants D and D_{kl} do not contain any overlap integral that is larger than the others. Therefore the representation in certain papers (see, e.g., Refs. 37 and 38), of the probability of the excitation of an atom in β decay only in terms of overlap integrals that appear in calculations with non-antisymmetrized product of atomic orbitals of the independent-electron model is incorrect.

The nonorthogonality integrals in the determinants D and D_{kl} were calculated on MO obtained in the form of linear combinations of Slater atomic orbitals Φ_{ap}:

$$s_{ij} = \sum_{aa'} \sum_{pp'} \tilde{c}_{a'p'}{}^i c_{ap}{}^j \langle \tilde{\Phi}_{a'p'} | \Phi_{ap} \rangle,$$

$$s_{ij}^{(k)} = \sum_{aa'} \sum_{pp'} \tilde{c}_{a'p'}^{l(k)} c_{ap}{}^j \langle \tilde{\Phi}_{a'p'} | \Phi_{ap} \rangle, \quad (33)$$

where $\tilde{c}_{a'p'}^i$ and c^j_{ap} are coefficients determined by the "Gaussian-70" program, $c_{a'p'}^{l(k)}$ are determined by the program based on the Huzinaga-Arnau technique. The indices a and a' number the atoms, ρ and ρ' number the types of atomic orbitals, $\{\Phi_{ap}\}$ is the atomic base of the molecule RT, and $\{\tilde{\Phi}_{a'p'}\}$ is the atomic basis of the molecule $(RHe)^+$.

Results of calculation and their discussion

We calculated the probabilities of restructuring the electron shell and the energy characteristics of the spectrum for metal hydrides and for a number of organic molecules containing different functional groups. The results of the calculations are summarized in Table II. In the case of hydrides the probability of excitation was assumed only for the lithium hydride. The presented data indicate that the molecular surrounding increases the probability of electron excitation, and this effect is strongest for lithium hydride. The probability of excitation in this case is equal to the probability that the daughter molecule will remain in the ground state.

For each of the calculated molecules we obtained detailed probability distributions of excitation in different electron states. Figure 2 shows the probability distributions for the molecules C_2H_5T and LiT. For all the calculated organic molecules the probability distributions differ insignificantly from those given for C_2H_5T. The maximum probability takes place at an excitation energy 35–40 eV. This maximum corresponds to a transition of the electrons from the molecular "L-shell," produced in the ground $1s$-orbital of the He atom and the $2s$-orbital of the C atom bound with it. The probability of excitation of valence electrons is much lower. The "K-shell" electrons of the molecules are hardly excited.

Thus, the a priori representation that in the shakeup produced by the β decay it is easiest to excite the least bound valence electrons does not correspond to reality.

TABLE II. Probabilities of restructuring of electron shell and energy characteristics of β spectrum of tritium-containing molecules.

№	RT molecule	W_∞	$\sum\limits_{n\ne 0} W_{0n}$ [1]	$\overline{\Delta E}$, eV	$\overline{\Delta E^*}$, eV [2]	$\bar{E}_{\beta,\ max}^{extr}$, eV
1	T	0.7023	0.2737	27.21	13.605	18532±7
2	LiT	0.4336	0.4390	33,74	23.39	18538±10
3	BT	0.5440	–	30.28	20.62	18535±10
4	NaT	0.4010	–	32.92	25.04	18538±10
5	AlT	0.4965	–	33.84	22.09	18538±10
6	LiOT	0.6152	0.3172	29.08	18.07	18534±10
7	CH₃T	0.6160	0.3230	29.96	18.66	18535±10
8	CH₂=CHT	0.6006	0.3296	29.54	18.93	18534±10
9	CH₃–CH₂T	0.6105	0.3170	30.23	18.92	18535±10
10	CH₃–CHT–CH₃	0.6030	0.3292	30.35	19.24	18535±10
11	NH₂–CH₂T	0.6095	0,3268	29.85	18.84	18534±10
12	NH₂–C₂H₄T	0.6079	0.3270	30.00	19.09	18535±10

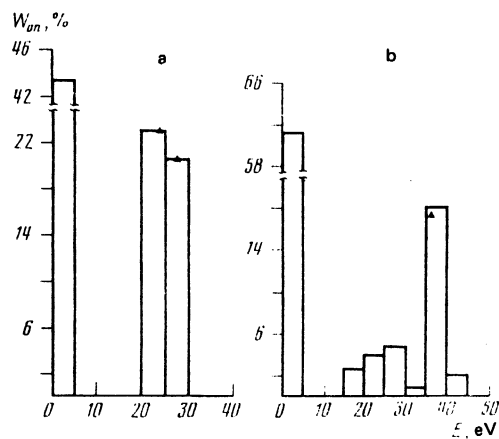[1]Only discrete excitation. [2]Average energy going both to discrete excitations and to ionization.

FIG. 2. Distribution of the summary probabilities of the electron excitations in the energy intervals $[5n, 5(n + 1)]$ eV in β decay: a) LiT; b) C_2H_5T.

The proximity of the calculated value for the different organic molecules containing the C–T bond indicates that the β decay is most influenced by the atom to which the tritium is directly bound.

These results become understandable if one analyzes the change of the electron structure of the molecule in the $RT \rightarrow (RHe)^+$ transition. Table III lists the calculation data for ethane; $n(i, He)$ characterizes the contribution made to the ith MO of the electrons of the He atoms, W_i is the probability of excitation of an electron from the ith MO. Replacement of T by He^+ lowers the energies of all the MO. The maximum lowering of the energy takes place for the orbitals of the L shell. It is most important, however, that it is precisely the orbitals of the L shell that are most sensitive to replacement of the T atom by He^+. An analysis of the population shows that helium makes it largest contribution to the MO of the L shell. Inasmuch as each MO cannot have more than two electrons, the value $n(i, He) = 1.23$ means that more than half the electron density of this orbital is carried by the helium electrons. The degree of distortion of the MO following the replacement $T \rightarrow He^+$ correlates with the probability of excitation of an electron from this orbital. Thus, the weight of the electron transitions from the seventh orbital is 20% of all the channels including the channel $0 \rightarrow 0$. This is easy to understand qualitatively. After all, it is pre-

cisely the distortion of the electron shell of $(RHe)^+$ compared with the electron shell of RT, which leads to a nonzero excitation probability (in the absence of such distortions the daughter molecule will be produced only in the ground state).

Construction of the electron-density maps shows that in the transition $RT \rightarrow (RHe)^+$ a redistribution of the electron density among the entire molecule takes place. The helium ion drags behind it the electron density from all the atoms, including also those that are far enough from it. This electron-density redistribution, however, takes place predominantly on account of the valence electrons. Decisive for the shakeup processes, as shown above, are the deeper electrons of the L shell. The latter is constructed mainly by orbitals of helium and of the atom C closest to it, and it is this which explains the low sensitivity of the calculated values to the peripheral chemical structure.

As follows from Table II, the averge chemical shift is positive for all molecules. This means that the presence of an electron shell always increases the energy of the β electron compared with the β decay of the bare nucleus. For molecules, the chemical shift $\overline{\Delta E}$ is higher than for an isolated atom T, by 2–3 eV for organic molecules and by 6.5 eV for LiT.

The presence of electron excitation channels is the cause of the physical broadening of the β spectrum. A measure of this broadening is the average excitation energy $\overline{\Delta E}^*(RHe^+)$. According to Table II, the average molecule excitation energies exceed the average excitation energy of the T atom by 7–12 eV for metal hydrides and by 5–6 eV for organic molecules. Consequently, the presence of a molecular surrounding increases the physical broadening of the β spectrum compared with the isolated T atom (concerning the experimental manifestations of the physical broadening see the end of Sec. 2).

In the last column of Table II are given the values of $\overline{E}_{\beta,\max}^{extr}$ obtained from formula (26). For the recoil energy E_{rec} we assumed a value of 3 eV, corresponding to recoil from the free tritium nucleus, inasmuch as the transformation of tritium into helium should lead for the molecules considered to an antibound state (this was shown for the CH_3T molecule in Ref. 8). The nuclear mass defect was obtained from the difference, given in Ref. 39, between the atomic masses, $[M(T) - M(^3He)]c^2 = 18\,573 \pm 7$ eV, in accordance with the relation

$$\Delta mc^2 = [M(T) - M(^3He)] + m_e c^2 - [E_0(T) - E_0(He)], \quad (34)$$

where the binding energies of the electrons in the ground states are $E_0(T) = -13.605$ eV; $E_0(He) = -78.984$ eV, whence

$$\Delta mc^2 = m_e c^2 + 18\,507 \pm 7 \text{ eV}.$$

In the case of the bare T nucleus, $E_{\beta,\max} = 18\,504 \pm 7$ eV. From the values of $\overline{E}_{\beta,\max}^{extr}$ given in Table II it follows that the presence of electron-excitation channels shifts the extrapolated end point of the β spectrum toward higher energies. For the T atom the shift is equal to 27 eV, and in the case of molecules the shift is 2–6 eV larger.

Since the accuracy with which the chemical shifts is

TABLE III. Characteristics of molecular orbitals of the molecules C_2H_5T and $C_2H_5He^+$.

iMO	C_2H_5T	$(C_2H_5He)^+$			
	E_i	E_i	$n(i, He)$	$W_i, \%$[1]	Arbitrary name of group of orbits
1	13.1	20.5	$2.2 \cdot 10^{-6}$	0.4	Valence shells
2	13.1	21.2	0.03	1.1	
3	13.7	22.4	0.02	0.8	
4	16.4	25.1	$8.5 \cdot 10^{-7}$	0.8	
5	16.4	29.0	0.12	3.5	"L-shell"
6	22.7	34.1	0.25	5.1	
7	28.0	44.1	1.23	20.0	
8	304.4	311.3	$2.7 \cdot 10^{-8}$	0	"K-shell"
9	304.4	315.5	$3.4 \cdot 10^{-5}$	0	

[1]W_i—probability of transition from ith MO to all the unoccupied MO.

calculated can be estimated at 1–3 eV, the accuracy of the presented values of $\overline{E}_{\beta,\max}^{\text{extr}}$ is determined by the accuracy with which the mass defect is measured. If it becomes possible in experiment to measure the true end point of the β spectrum, then formulas (25) and (26) can be used to calculate from them the neutrino mass m_ν.

4. CALCULATION OF THE INFLUENCE OF THE MOLECULAR STRUCTURE ON THE SPECTRUM OF THE β SOURCE OF THE ITEP GROUP

The working medium of the β source of the ITEP group[12,13] was obtained by depositing tritiated valine on a substrate by evaporation from a water-alcohol solution in vacuum. The valine layer sputtered in this manner constituted crystalline DL-valine[40] and consisted of molecules of two modifications, which differed in the location of the substitution of the tritium atom; we designate them valine I and valine II (see Fig. 3).

In the DL-valine crystal, in view of the distortion introduced by the crystal field, the equilibrium geometry of the molecules is somewhat different from the geometry of the free molecules. As a result of β decay, a helium-containing valine complex (RHe)$^+$ is produced, and this disturbs the translational symmetry of the crystal. Therefore no excitonic states are produced in β decay, and only molecular states of the complex (RHe)$^+$ are excited. Inasmuch as in molecular crystals the intermolecular interactions are much weaker than the intramolecular ones, the influence of the crystalline surrounding of the valine on the β-decay process can be neglected.

In connection with the large size of the valine molecule (19 atoms and 64 electrons), the calculations were carried out on a minimal basis of Slater atomic orbitals. The results were next corrected for the error introduced by the length of the basis. The latter was obtained in the calculations of the corresponding fragments valine I and valine II on a minimal and on a doubled basis (see Fig. 3, the fragments are enclosed in dashed boxes). The geometry of the fragments was maintained the same as in the BL-valine crystal.

The results of the calculations are summarized in Table IV and shown in Fig. 4. Comparison of the data of Tables IV and II shows that the probability of remaining in the ground state is approximately 3% higher for the valine molecules than for the related organic molecules. The reason for this is the distortion of the equilibrium geometry of the valine by the crystalline field.

The most probable excitations in β decay correspond to transitions of an electron from MO made up of $2s$ orbitals of


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 ┌─────────────────┐
 │ NH₂ — CT — COOH │        NH₂—CH —COOH
 └─────────┬───────┘              ┬
          CH                ┌─────┼─────┐
        /    \              │   / CT  \  │
     CH₃      CH₃           │ CH₃      CH₃│
                            └───────────┘
      a                          b
```
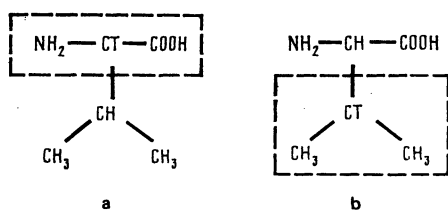
FIG. 3. Isotopic isomers of valine, calculated in the paper: a) valine I; b) valine II.

TABLE IV. Probabilities of restructuring of electron shells and energy characteristics of β spectrum of valine.

Molecule	W_{00}	$\sum_{n\neq 0} W_{0n}$[1]	$E_0(\text{RT})-E_0(\text{RHe}^+)$, eV	$\overline{\Delta E}$, eV	$\overline{\Delta E^*}$,[2] eV	$\overline{E}_{\beta,\max}^{\text{extr}}$, eV
Valine I	0.6512	0.3007	46.79	28.20	18.59	18533±10
Valine II	0.6291	0.3099	48.25	29.37	18.88	18534±10

[1]Only discrete spectrum. [2]Average energy going both to discrete excitations and to ionization.

carbon atom and the nearby orbitals of the hetero atoms, and of the $1s$ orbital of helium, to lower vacant MO, and have an energy ~ 40 eV.

The total probability of the excitations, $W_{00} + \Sigma_n W_{0n}$, amounts to 0.952 for valine I and 0.939 for valine II. From this we obtain for the ionization probability the values 0.048 and 0.061, respectively. The average energy going to ionization, E_i, can be roughly estimated by approximating the integral over the continuous spectrum with the aid of the mean-value theorem:

$$\overline{\Delta E_{\text{ion}}} \equiv \int W_{0E_i} E_i dE_i = \overline{E}_i \int W_{0E_i} dE_i. \tag{35}$$

The total probability of transitions to the continuous spectrum is obtained from the normalization condition, inasmuch as the probability of the transitions into the discrete spectrum was calculated by us. Knowledge of the energies of the discrete transitions in the molecule (RHe$^+$) and of their probabilities makes it possible to calculate also the average energy of the excitation into the discrete spectrum, $\overline{\Delta E}_{\text{disc}}^*$. As a result, $\overline{E}_i$ can be obtained from the simple formula

$$\overline{E}_i = [\overline{\Delta E^*}(\text{RHe}^+) - \overline{\Delta E}_{\text{disc}}^*(\text{RHe}^+)] / \left(1 - W_{00} - \sum_{n\neq 0} W_{0n}\right). \tag{36}$$

Calculation yields $\overline{E}_i = 1.47$ eV for valine I and 139 eV for valine II. These values are subject to a large error, since a slight error in the probability of the transitions to the discrete spectrum (of the order of several percent) alters substantially the value of the denominator in (36).

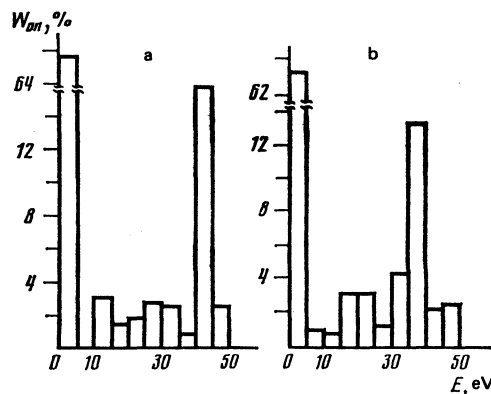The values of $\overline{E}_{\beta,\max}^{\text{extr}}$ in Table IV were calculated from

FIG. 4. Distribution of the summary probabilities of the electron excitations in the energy intervals $[5n,5(n + 1)]$ eV in β decay: a) of valine I; b) valine II.

formula (26), in which the nuclear mass defect was obtained from the atomic defect of Ref. 39 and was chosen, as in the preceding section, to be $\Delta mc^2 = m_e c^2 + 18\,507$ eV. We can proceed conversely: starting from the experimental value of the extrapolated end-point of the β spectrum, we calculate the atomic mass defect. Data of just this kind are discussed in Ref. 39. We note in this connection that Bergkvist[10] measured in his experiments $\overline{E}_{\beta,\max}^{\text{extr}}$, whereas in the experiments of ITEP group[12,13] they obtained $\overline{E}_{\beta,\max}^{\text{extr}}$, (25), which differs by an amount equal to average excitation energy $\overline{\Delta E^*}$. It is therefore incorrect to add the Bergkvist correction, 41 eV, to the value of the end point of spectrum of Refs. 12, 13, and 41, as was done in Ref. 39 (let alone the fact that the correction should be calculated for the valine molecule and not for the tritium atom). To find the atomic mass defect it is necessary to subtract from the value $E_0 = 18\,577 \pm 13$ obtained in Ref. 12 and 13 the value of $E_0(\text{RT}) - E_0(\text{RHe}^+)$, which is equal in the case of valine to 48.251 eV, and add $(E_0(\text{T}) - E_0(^3\text{He})) = 65.379$ eV. As a result we obtain $[M(\text{T}) - M(^3\text{He})]c^2 = 18\,594 \pm 13$ eV, which increases even more the deviation, noted in Ref. 39, from the value obtained by Bergkvist.[10]

The data presented in the present paper show that the β source used in Refs. 12 and 13 is not described by a single-level model. The latter means that when reducing the experimental results of Refs. 12 and 13 by the procedure described in 13, the value of the lower mass limit 14 eV should be replaced (assuming the assumptions made in 12 and 13 remain in force) by a higher value which, naturally, narrows down the confidence interval for the neutrino mass. This conclusion was confirmed in a communication by an ITEP group at the session of the Division of Nuclear Physics of the USSR Academy of Sciences (1–4 February 1982), where results of a second reduction of the experimental data of Refs. 12 and 13, with allowance for the valine excitation spectrum calculated by us, were reported.

In conclusion, the authors thank Ya. B. Zel'dovich, V. A. Lyubimov, V. Z. Nozik for helpful discussions, N. N. Vitkovskiĭ for providing the "Gaussian-70" program, and L. N. Tsygankov for help with compiling the group of programs.

[1)] For the sake of brevity we shall refer in place of the antineutrino to a neutrino, since the characteristics of interest to us are the same for them.

[2)] We emphasize that expression (16) contains the chemical shift in the N th channel, and not the excitation energy of RHe^+ in this channel, as in Eq. (6) of Ref. 13. For the problem of determining the mass of the neutrino by the least squares method, this difference in the formulas is insignificant.

[1] A. J. Midgal, J. Phys. (USSR) **4**, 449 (1941).
[2] E. Fainberg, J. Phys. (USSR), **4**, 423 (1941).
[3] N. S. Batkin and Yu. G. Smirnov, Fiz. Elem. Chastits. At. Yadra **11**, 1421 (1980) [Sov. J. Part. Nucl. **11**, 564 (1980)].
[4] H. M. Schwartz, J. Chem. Phys. **23**, 400 (1955).
[5] M. Cantwell, Phys. Rev. **101**, 1747 (1956).
[6] M. Wolfsberg, J. Chem. Phys. **24**, 24 (1956).
[7] L. Wolniewicz, J. Chem. Phys. **43**, 1087 (1965).
[8] S. Ikuta, S. Iwata, and M. Imamura, J. Chem. Phys. **66**, 4671 (1977).
[9] G. C. Hanna and B. Pontecorvo, Phys. Rev. **75**, 933 (1949).
[10] K. E. Bergkvist, Nucl. Phys. B **39**, 317 (1972); **39**, 371 (1972).
[11] K. E. Bergkvist, Physica Scripta **4**, 23 (1971).
[12] V. S. Kozik, V. A. Lyubimov, E. G. Novikov, V. Z. Kozik, and E. F. Tretyakov, Yad. Fiz. **32**, 30 (1980) [Sov. J. Nucl. Phys. **32**, 154 (1980)].
[13] V. A. Lyubimov, E. G. Novikov, V. Z. Nozik, E. F. Tret'yakov, V. S. Kozik, and N. F. Myasoedov, Zh. Eksp. Teor. Fiz. **81**, 1158 (1981) [Sov. Phys. JETP **54**, 616 (1981)].
[14] E. F. Tret'yakov, Izv. AN SSSR, Ser. Fiz. **39**, 583 (1975).
[15] M. Fukugita and R. Kubodeva, Zs. f. Phys. C **9**, 365 (1981).
[16] J. Law, Phys. Lett. **B102**, 371 (1981).
[17] C. Chengrui, H. Tschsin, and C. Shaolin, Preprint AS-ITP-81-22.
[18] I. G. Kaplan, V. N. Smutnyĭ, and G. V. Smelov, Dokl. Akad. Nauk SSSR **262**, 1127 (1982) [Sov. Phys. Dokl. **27**, 137 (1982)].
[19] I. G. Kaplan, V. N. Smutny, and G. V. Smelov, Phys. Lett. **B112**, 417 (1982).
[20] E. Fermi, Zs. f. Phys. **28**, 161 (1934).
[21] A. S. Davydov, The Theory of Atomic Nucleus [in Russian], Fizmatgiz (1958).
[22] E. L. Konopinski, The Theory of Beta Radioactivity, Oxford at the Clarendon Press (1966).
[23] R. Blyn-Stoyle, Fundamental Interaction and Atomic Nucleus [Russ. transl.], Mir. 1976.
[24] A. B. Migdal, Kachestvennye metody v kvantovoĭ teorii (Qualitative Methods in Quantum Theory), Nauka, 1975.
[25] D. I. Blokhintsev, Principles of Quantum Mechanics, Allyn and Beacon, 1949,
[26] A. S. Davydov, Quantum Mechanics, Pergamon, 1965.
[27] E. Kolos, Adv. Quant. Chem. **5**, 99 (1955).
[28] Ya. B. Zel'dovich, Usp. Fiz. Nauk **56**, 165 (1955).
[29] V. S. Dzhelepov and L. N. Zyryanov, Vliyanie elektricheskogo polya na beta-raspad, Influence of the Electric Field of the Atom on Beta Decay, AN SSSR, 1956.
[30] R. McWeeny and B. Suttcliffe, Methods of Molecular Quantum Mechanics, Academic, 1969.
[31] S. Huzinaga and C. Arnau, J. Chem. Phys. **54**, 1948 (1971).
[32] S. Toh, Proc. Phys. Math. Soc. Japan **22**, 119 (1940).
[33] H. H. Michels, J. Chem. Phys. **44**, 3384 (1966).
[34] V. F. Brattsev, V. N. Baranovskiĭ, A. I. Panin, et al. Metody rascheta elektronnoĭ struktury atomov i molekul (Methods of Calculating the Electron Structure of Atoms and Molecules), Izd. LGU, 1976.
[35] T. H. Dunning, J. Chem. Phys. **53**, 2823 (1970).
[36] I. G. Kaplan and O. B. Rodimova, Int. J. Quant. Chem. **7**, 1203 (1973).
[37] T. A. Carlson, Phys. Rev. **130**, 2361 (1963).
[38] T. A. Carlson, C. W. Nestor, and T. C. Tucker, Phys. Rev. **169**, 27 (1968).
[39] T. G. Smith, E. Koets, and A. M. Wapstra, Phys. Lett **B102**, 114 (1981).
[40] H. S. Mallikarjunan and S. T. Rao, Acta Cryst. **B25**, 296 (1969).
[41] E. F. Tret'yakov, N. F. Myasoedov, A. M. Apalikov, et al., Izv. AN SSSR, Ser. fiz. **40**, 2026 (1976).

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