

Effect of final-state interactions in allowed β decays. II. Reliability of the β -decay spectrum for T_2

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The final-state distribution of HeT^+ resulting from the β decay of molecular tritium has been investigated with particular emphasis on its reliability in connection with the interpretation of the tritium neutrino-mass experiments. This investigation concentrates on two aspects. First, the reliability within the sudden approximation is examined. This includes the extension of previously calculated final-state distributions to a larger range of energy transfer, i.e., to an excitation energy of 800 eV, the investigation of the effect of nuclear motion on the continuous electronic spectrum, and the use of improved basis sets. Second, the *molecular* transition-probability spectrum (including electronic bound and continuous states) has been calculated for the first time in a beyond sudden-impulse approximation. It is concluded that the concomitant corrections are too small to explain the problems of recent experiments difficulties to extract the neutrino mass out of the β spectrum of T_2 . [S0556-2813(97)06908-2]

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I. INTRODUCTION

Although the experimental detection of neutrinos was reported four decades ago on the basis of neutrino-induced β -decay reactions [1,2], it is still not known whether neutrinos possess a nonzero rest mass or not. The question of a possible rest mass of neutrinos is, however, of great interest in many areas of physics, like nuclear and particle physics, astrophysics, and cosmology. It is also of paramount importance for the further development of unification theories.

Because of the extreme penetrability of the neutrinos, no direct experimental detection of their rest mass is known so far. The simplest approach of an indirect measurement would be an accurate mass determination of all other particles involved in a β -decay process. However, such a measurement turned out to be not sufficiently accurate, and a better approach is provided by the measurement of the β spectrum of tritium close to its end point. As pointed out by Fermi [3], the *shape* of the β spectrum is a function of the neutrino mass (more accurately of its value squared), and a possible nonzero rest mass will show up most evidently close to the end point of the spectrum where nearly all of the energy that is released in the decay is carried away by the β electron. Actually, if the exact value of the energy release was known, already the position of the end point itself would reveal a possible rest mass of the neutrino.

If the decaying nucleus is part of an atomic or molecular system, the situation becomes more complicated, since (due to the decay process and the concomitant change of the nuclear charge of the decaying nucleus) the remaining molecular system can generally be left in any of its eigenstates with all possible excitation energies. This possibility of different energy sharings between the β electron and the remaining ion causes a distortion of the β spectrum (compared to the decay of an isolated, bare nucleus). Since it has been up to now experimentally impossible to perform a coinci-

dence measurement of the β electron and the molecular fragment(s), the molecular final-state spectrum has to be obtained from theory. Therefore, the precise evaluation of the probability distribution for the excitation of HeT^+ is of major importance for the interpretation of the experimental data, and hence for establishing reliable bounds for the neutrino mass [4,5]. In fact, the choice of T_2 as radioactive source in the more recent experiments (instead of the more complicated molecular systems used in the older experiments) is very much guided by the idea that extremely accurate quantum-chemical calculations can be performed for the two-electron systems T_2 and HeT^+ . Throughout the years, the experimental studies of β decay from tritium sources, especially molecular tritium, have consecutively narrowed the *upper bound* for the mass of the electronic antineutrino, which has been recently reported to be 7 eV [6] or even 4.5 eV [7].

However, a shadow of doubt has been cast on the interpretation of the tritium-decay experiments, since they seem to reveal an unphysical trend towards negative values of the square of the neutrino mass: $m_\nu^2 < 0$. This surprising result is possible since in these experiments the neutrino mass is extracted from the comparison of the experimental spectrum with the theoretical one on the basis of a least-squares fit, wherein the value of m_ν^2 is used as one of the free parameters. The possibility of a (of course unphysical) negative value of m_ν^2 has been included in the fit procedure in order to obtain a well-defined definition of χ^2 which specifies the goodness of the fit. If m_ν^2 is restricted to $m_\nu^2 \geq 0$, a satisfactory fit cannot be obtained. Analyzing the residual spectrum of such a fit, the deficiency seems to indicate that the theoretically predicted molecular final-state distribution carries too small probability (in certain energy ranges) compared to the experimental findings [7,8]. It should be noted, however, that the shape, intensity, and position of the experimental

excess rate seem to vary for different experiments, and even for different runs of the same experiment [8]. Nevertheless, it has been argued that the reason for the encountered problems might be due to a failure of the theoretically obtained final-state spectrum of the HeT^+ system entering the fit procedure. Therefore the reliability of this final-state spectrum will be investigated in the present work.

An extremely accurate calculation of the final-state distribution of HeT^+ within the sudden-impulse approximation has been reported in [9]. In that work the transition probabilities of the electronic bound states including their rovibrational structure have been calculated within the Born-Oppenheimer approximation with the aid of highly sophisticated quantum-chemical tools. The electronic continuum, however, was at that time treated only in an approximate way by a ‘‘simple’’ discretization ansatz. Using an approach that is related to the complex-coordinate method, the transition-probability spectrum for the electronic continuum of HeT^+ was later improved [10], but the fits of the neutrino mass remained nearly unchanged [11].

Since the extraction of the neutrino mass appears to fail, even though the theoretical spectra that are used in the fit are supposed to be highly accurate (i.e., its inaccuracy is supposed to be much smaller than imputed by the experimental excess rate), one has to search for possible reasons. From the experimental point of view there is of course a number of potential sources for problems. The experiments are comparatively complicated and a number of parameters such as the exact content of the source, the energy loss in the source and in the spectrometer, the resolution function, possible sources for background, etc., have to be known very accurately.

From the theoretical point of view the molecular final-state spectrum has been pointed out as the most prominent candidate for errors. Of course, another possible source for problems would be a failure of the Fermi theory, but up to now there seems to be no indication from any experiment that this might be the case. However, one may keep in mind that the validity of the Fermi theory of the weak interaction has been mainly concluded on the basis of the overall good agreement of the β spectra with the theoretical prediction.

Finally, the problem of determining the neutrino mass by the tritium experiments might be caused at the interface between the experimental and theoretical groups: i.e., one may ask whether the theoretical spectrum is appropriate for a given experimental situation and whether it is utilized in the appropriate way.

In this work we will concentrate only on the question of the reliability of the final-state distribution. Several aspects have been raised in recent discussions (at a topical workshop at Harvard University, 1996).

(1) The possibility of excitations of states of HeT^+ with $m \neq 0$ (especially of Π states) has so far been neglected.

(2) The final-state spectra that have been reported so far for molecular tritium did not extend beyond about 165 eV. Since the experimental groups suggest a ‘‘missing final-state contribution’’ at energies above 100 eV and since in a recent theoretical work [12] it has been suggested that a neglected or wrong high-energy tail may influence the neutrino mass extracted by fit, this tail has attracted some interest.

(3) The influence of nuclear motion has been neglected in

the treatment of the electronic continuum of final states.

(4) All final-state distributions for *molecular* tritium have been calculated so far only within the sudden-impulse approximation. The validity of this approximation has been assessed mainly on the basis of calculations of corrections to the sudden approximation for *atomic* tritium [13–18]. (It may be noted that there have been also attempts to estimate the corrections to the sudden approximation for molecules [19,20], but since in these works comparatively complex molecules were considered, a number of additional approximations had to be made already within the sudden approximation.)

(5) Even though all of the atomic calculations reported here that have investigated higher-order corrections to the sudden approximation are in agreement with respect to the conclusion that these corrections are in fact very small close to the end point of the β spectrum, the obtained results do not always agree and some confusion arose about the correctness of including or omitting certain terms. In addition, it may be erroneous to assume these results to be transferable to the molecular case, since there are obvious differences in between the atomic and molecular cases: In the atomic case one encounters spherically symmetric one-electron systems (T , He^+), while T_2 and HeT^+ are two-electron systems possessing $D_{\infty h}$ and $C_{\infty v}$ symmetry, respectively.

(6) Finally, the question of possible exchange between the β electron and one of the initially bound electrons has been raised.

In this work we will mainly address questions (2)–(5).

The energy range of the HeT^+ final-state spectrum will be extended to an excitation energy of 800 eV and its reliability will be investigated by a comparison to an atomic-type tail. In contrast to the previous work that was based on the clamped-nuclei approximation now also the influence of nuclear motion will be investigated. In order to improve on the sudden approximation, we have used a perturbative treatment where the sudden approximation can be associated with the zeroth-order treatment in perturbation theory (see [21] which we will in the following refer to as I).

Based on a general relativistic expression (arising from a perturbative treatment of the Coulombic interaction of the β electron with the remaining particles for arbitrary atomic and molecular systems) we present the first beyond sudden-approximation calculation of the excitation-probability distribution for HeT^+ . The treatment includes both the electronic bound and continuous states of HeT^+ . In order to obtain the continuous distribution on the level of the first-order perturbation theory, we have extended the method presented in [10] along the lines outlined in [22] as to extract for the first time interference terms occurring in a series expansion of the transition probability. Some of the results have been briefly summarized in [23].

II. OUTLINE OF THE THEORY

A. β spectrum

A detailed derivation of the β -decay spectrum in a beyond sudden-approximation treatment of the β decay of a nucleus

embedded in an atomic or molecular system has been given in I. We will report here mainly the final expressions and concentrate on the application to the tritium neutrino-mass experiments. In addition, the involved approximations will be discussed.

First of all, it is important to note that the ‘‘usual’’ conditions of tritium neutrino-mass experiments will be assumed, i.e., no detection of the neutrino and the final state of the molecular system, no discrimination of the direction of the β electrons, and a random orientation of the decaying

nuclei. The decaying nucleus is assumed to be one of the two nuclei in a freely moving and isolated T_2 molecule that is in its rovibronic ground state. All possible deviations from these assumptions that may be encountered in an experiment thus have to be taken into account very carefully.

According to Fermi [3] the probability of detecting a β electron within the kinetic energy interval $E_\beta^{\text{kin}} + dE_\beta^{\text{kin}}$ is given by a product of kinematic factors and a sum over the probabilities $\mathcal{P}_f(E_\beta)$ associated with different molecular excitations (see also I),

$$\frac{dw(E_\beta^{\text{kin}})}{dE_\beta^{\text{kin}}} = \sum_f \frac{32\pi^3}{c^5} p_\beta E_\beta [(\epsilon - E_{f0}^{\text{HeT}^+})^2 - m_\nu^2 c^4]^{1/2} (\epsilon - E_{f0}^{\text{HeT}^+}) \Theta(\epsilon - E_{f0}^{\text{HeT}^+}) \Theta([\epsilon - E_{f0}^{\text{HeT}^+}]^2 - m_\nu^2 c^4) \mathcal{P}_f(E_\beta) \quad (1)$$

$$= \frac{32\pi^3}{c^5} p_\beta E_\beta \int_0^\infty [(\epsilon - E)^2 - m_\nu^2 c^4]^{1/2} (\epsilon - E) \Theta(\epsilon - E - m_\nu c^2) \sum_f \mathcal{P}_f(E_\beta) \delta(E - E_{f0}^{\text{HeT}^+}) dE. \quad (2)$$

In these equations $E_\beta = E_\beta^{\text{kin}} + m_e c^2$ and $p_\beta = \sqrt{E_\beta^2/c^2 + 2m_e E_\beta}$ are the total energy and the momentum of the β electron at the detector. $\epsilon = E_{\text{max}} - E_\beta = E_{\text{max}}^{\text{kin}} - E_\beta^{\text{kin}}$ is defined as that amount of the total energy released in the β decay that is not carried away by the β electron if the neutrino has no rest mass. $E_{\text{max}} = m_{T_2} c^2 - m_{\text{HeT}} e^2 - E_{\text{rec}} - (E_0^{\text{HeT}^+} - E_0^{T_2})$ is the maximum total energy of the β electron in the case that $m_\nu = 0$ (E_{rec} being the recoil energy transferred to the molecular system and $E_0^{\text{HeT}^+}$ and $E_0^{T_2}$ being the rovibronic ground-state energies of T_2 and HeT^+ , respectively). It may be noted that in the case of a nonzero rest mass of the neutrino E_{max} is not the experimental end point of the β spectrum, since the latter is given by $E_{\text{max}}^{\text{exp}} = E_{\text{max}} - m_\nu c^2$ (where the same relation holds for the kinetic energies).

The Heaviside step functions $\Theta(x)$ arise from constraining the energy and the momentum of the neutrino to physical values in the integration over all possible momenta of the undetected neutrino, as it is discussed in I. Since $E_f^{\text{HeT}^+}$ is the energy of the final rovibronic state of the HeT^+ molecular ion, $E_{f0}^{\text{HeT}^+} = E_f^{\text{HeT}^+} - E_0^{\text{HeT}^+}$ is the excitation energy of HeT^+ . The \sum_f has to be understood as an infinite summation (integration) over all possible final states f of the *composite* system consisting out of the β electron and HeT^+ , where, however, the energy of the β electron is restricted to E_β .

Equation (2) emphasizes the fact that the β spectrum does not depend on the specific final state of HeT^+ , but only on the total probability that a certain amount of energy is left in the internal (rovibronic) degrees of freedom of HeT^+ . Thus, it is needed to evaluate the total probability connected to a certain energy value, but it is not needed to resolve energetically degenerate molecular final states which is especially suitable in the case of multiply degenerate continua.

Finally, $\mathcal{P}_f(E_\beta)$ is the squared transition amplitude describing the β decay, i.e.,

$$\mathcal{P}_f(E_\beta) = |\langle \Psi_f^{(-)} | W_i | \Phi_i \rangle|^2, \quad (3)$$

where W_i is the weak-interaction operator for the decay process, Φ_i is the complete wave function of the T_2 molecule in its rovibronic ground state, and $\Psi_f^{(-)}$ is a scattering wave function of the complete system in the final channel constrained to certain boundary conditions. These conditions include that the β particle has at the detector the energy E_β , while HeT^+ is left in the state f with the energy $E_f^{\text{HeT}^+}$ and the energy of the neutrino fulfills the energy conservation laws under these constraints. While the neutrino interacts with the remaining particles only via the weak interaction, the main problem in constructing a correct final-state wave function arises from the long-range Coulombic forces in between the β electron and the remaining ion. In the following section we will address the problem of obtaining the most difficult part in $\mathcal{P}_f(E_\beta)$, i.e., $\Psi_f^{(-)}$.

B. Final-state interactions

1. Complete final-state wave function

One way to construct the many-body scattering wave function is provided by the Lippmann-Schwinger ansatz. In this approach the final-state interaction is defined by the partitioning of the total Hamiltonian, which can be done in different ways. If the complete Hamiltonian H in the final channel is split into a sum of two operators, $H = H_0 + U_f$, the scattering wave function can be written as

$$\langle \Psi_f^{(-)} | = \langle \Phi_f | + \langle \Phi_f | U_f (E - H_0)^{-1} | + \dots, \quad (4)$$

where the wave functions Φ_f are now solutions to H_0 , again fulfilling the constraints that the β electron has in the asymptotic limit the energy E_β , etc.

In the case of the β decay the ‘‘unperturbed’’ Hamiltonian H_0 may be written as

$$H_0 = H_{\text{mol}} + T_\beta - \frac{e^2(Z_A - Z_A^{\text{eff}})}{r_{\beta A}}, \quad (5)$$

while the operator U_f is given by

$$U_f = H - H_0 = -\frac{Z_A^{\text{eff}} e^2}{r_{\beta A}} - \frac{Z_B e^2}{r_{\beta B}} + \frac{e^2}{r_{\beta 1}} + \frac{e^2}{r_{\beta 2}}, \quad (6)$$

where A and B denote the decaying and spectator nuclei, respectively. The two molecular electrons are labeled by the indices 1 and 2, while $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$. H_{mol} is the Hamiltonian describing a freely moving HeT^+ molecule and T_β is the Hamiltonian describing a free β electron. The charge of the spectator nucleus is $Z_B = 1$, but different values for Z_A^{eff} define the different partitionings of the total Hamiltonian.

The choice $Z_A^{\text{eff}} = Z_A$ leads to a complete factorization of Φ_f into the product $\psi_f^{\text{HeT}^+} \cdot \phi_\beta$ describing a noninteracting system consisting out of a HeT^+ molecule in the state \mathbf{f} and a β electron. However, even in the case of a different choice for Z_A^{eff} the wave function Φ_f can be factorized to a very good approximation, although the coupling between the β electron and all particles except the decaying nucleus still remains due to the *shared* interaction with the decaying nucleus. Within the Born-Oppenheimer approximation the factorization is complete, and taking into account the couplings between electronic and nuclear motion can be expected to retain the factorized form to a very good approximation.

2. Final-state wave function for the β electron

Arafune and Watanabe [16] have adopted $Z_A^{\text{eff}} = 0$ as is usually (but not at all necessarily) done also in treatments within the sudden approximation. In this case the wave function of the β electron resembles (in the Born-Oppenheimer approximation) the scattering solution of a He^+ ion (even in the molecular case). The wave function ϕ_β is in this case identical to a Coulomb wave corresponding to the charge $Z_A - Z_A^{\text{eff}} = 2$. Williams and Koonin [13] used, on the other hand, $Z_A^{\text{eff}} = 1$, which yields the scattering function of atomic hydrogen as wave function for the β electron; i.e., ϕ_β is in this case identical to a Coulomb wave corresponding to the charge $Z_A - Z_A^{\text{eff}} = 1$. In I we have worked out explicitly the case $Z_A^{\text{eff}} = 2$ where the wave function of the β electron is the one describing a free particle: i.e., ϕ_β is given by a plane wave.

The advantage of $Z_A^{\text{eff}} = 0$ is that in this way the zeroth-order term obtained within a perturbational treatment is identical to the (usual) sudden approximation, and most of the final-state interaction is then already contained in the zeroth-order treatment. The advantage of the choice $Z_A^{\text{eff}} = 1$ is, however, that no divergences occur in the calculation of the first-order correction term (see [13] and I). Finally, $Z_A^{\text{eff}} = 2$ has the advantage that the evaluation of the first-order correction can be done without additional approximations (or in a much simpler way), since only plane waves are involved in the evaluation of the required matrix elements, while the other choices of Z_A^{eff} require the use of Coulomb waves. However, if all terms in Eq. (4) are considered, the results obtained for different splittings should be identical. In a truncated series it is, on the other hand, important to investigate the consequences of different splittings, and this will be done in Sec. IV B 1.

3. Molecular final-state wave function and the problem of relaxation

After the discussion of the wave function of the β particle we will now concentrate on the molecular final-state wave function ψ_f^{mol} . In contrast to ϕ_β the molecular wave function ψ_f^{mol} is not affected by the different partitionings of the Hamiltonian described in Eq. (5) if the coupling between electronic and nuclear motion is neglected, i.e., if the Born-Oppenheimer approximation is used.

ψ_f^{mol} can be factorized as $\psi_{\mathbf{p}_C} \cdot \psi_f^{\text{nuc}} \cdot \psi_f^{\text{HeT}^+}$, where $\psi_{\mathbf{p}_C}$ is a plane wave describing the center-of-mass motion of HeT^+ (in terms of its momentum \mathbf{p}_C), ψ_f^{nuc} is the *nucleonic* wave function describing the final state of the nucleons of the ^3He nucleus, and $\psi_f^{\text{HeT}^+}$ is the rovibronic wave function of HeT^+ in terms of the internal coordinates. In the following we will concentrate on the rovibronic wave functions $\psi_f^{\text{HeT}^+}$.

Besides the practical problem of evaluating $\psi_f^{\text{HeT}^+}$ for all possible final states including the ones lying in the continua, there is also the question of what is really meant by a molecular ‘‘final’’ state. As will be discussed in the following, the definition of the molecular final state adopted in this work is different from the one that would be experimentally detectable if one would measure the molecular final states, and not only the energy of the β electron.

If the experiment would discriminate different molecular final states, e.g., by a coincidence measurement, possible internal relaxation channels of the HeT^+ molecule like dissociation, radiative deexcitation, etc., would have to be considered in order to obtain the true, experimentally detectable final-state distribution. A similar situation is encountered in experiments that measure the sum of the β energy and the energy released in the relaxation process as is the case in experiments where the tritium source is contained in the detector (see, e.g., [24,25]).

In the usual case of tritium neutrino-mass experiments that do not resolve the molecular final states (and where the relaxation energy is not transferred to the detector as in [24]) the problem of relaxation is more subtle, since one is only interested in the effects that will influence the single quantity that is measured, i.e., the energy of the β electron. However, as long as the β electron interacts with the remaining ion, there is a possible energy exchange. Thus the internal relaxation of HeT^+ during its interaction time with the β electron has to be taken into account. If the complete wave function $\Psi_f^{(-)}$ is constructed, the complete interaction of the β electron with the remaining system would be included. In a time-dependent picture, that would include the whole time interval from the decay up to infinity (which corresponds approximately to the time of detection). In a time-independent picture this involves the whole position space. However, this truly complete $\Psi_f^{(-)}$ would have to be a solution of a Hamiltonian that contains also the part that allows for radiative transitions. If this part is omitted, already all possible radiative relaxation channels of the excited states of HeT^+ are ignored. If the molecular part of the Hamiltonian is approximated additionally by the use of a clamped-nuclei description (setting all nuclear masses equal to infinity), also

dissociative relaxation channels, etc., are completely neglected.

On the other hand, it will be a good approximation to ignore the influence of internal relaxation processes on the β spectrum if the interaction time (or volume) of the β particle with the remaining system is small compared to the relaxation time. Since the β electron departs with a kinetic energy of 18.6 keV close to the end point of the β spectrum, it is normally assumed that the β electron really leaves the *effective* interaction range sufficiently long before the internal relaxation channels (e.g., the deexcitation of an electronically excited state of HeT^+ via fluorescence) come into action. However, if this relaxation (e.g., the fluorescence process) takes place after the interaction of HeT^+ with the β electron is terminated, the β electron will not be affected and thus its energy will remain unchanged. Therefore, such an internal molecular relaxation after the interaction time will not influence the β spectrum.

The usual measure for the time scales (defining, on the one hand, the internal relaxation processes and, on the other hand, the leaving of the interaction range by the β electron) is the ratio of the velocity of the β particle to the one of the ‘‘molecular’’ electrons. Thus, the ‘‘final’’ states considered here are in fact the states in which the HeT^+ system is left at the time when the interaction with the β electron is assumed to be terminated (at least to a sufficiently good approximation, since this interaction does not really vanish fast enough, due to the peculiarities of the Coulomb interaction and the zero rest mass of the photon).

However, this will usually not be the state of HeT^+ that would be measured in an experiment resolving the ‘‘true’’ molecular final-state distribution, and that would be obtained after (some) relaxation has occurred. (The only exception is the case where the HeT^+ ion is left in its rovibronic ground state when the β electron leaves the interaction range.) The degree of relaxation that has to be taken into account in order to predict the experimental molecular final-state distribution depends of course on the time between the decay and the measuring process of the molecular fragments.

It is important to note that the type of relaxation channels included in the treatment will be defined by the level of approximation used for the molecular Hamiltonian H_{mol} . Of course, only those channels (radiative, dissociative, etc.) that are included in H_{mol} can have an influence on the calculated β spectrum, and this influence is conveyed to the β electron via its (Coulombic) interaction with the remaining molecular ion. Since the Lippmann-Schwinger approach in Eq. (4) is a series expansion in this interaction, the effect of the relaxation channels on the β spectrum will depend on the number of expansion terms that are included in the calculation. For example, on the level of the sudden approximation where the interaction between the β electron and the molecular ion is completely neglected (i.e., if $Z_A^{\text{eff}}=2$ is used), there is no influence of the relaxation processes on the β spectrum possible.

An important consequence of this discussion is that if the effects of the interaction of the β electron with the remaining system are found to be small, then the influence of internal molecular relaxation processes on the β spectrum can be expected to be even smaller.

After having discussed $\Psi_f^{(-)}$, we will now return to the evaluation of the decay rate given in Eq. (2).

C. Born series for the transition probability

Applying now the factorization into molecular and leptonic wave functions and separating out also the center-of-mass motion of the molecule as well as the nucleonic wave function describing the motion of the nucleons within the nuclei, Eq. (2) can be written as (cf. I)

$$\frac{dw(E_\beta^{\text{kin}})}{dE_\beta^{\text{kin}}} = A f(p_\beta) \int_0^\infty [(\epsilon - E)^2 - m_\nu^2 c^4]^{1/2} (\epsilon - E) \times \Theta(\epsilon - E - m_\nu c^2) \tilde{I}(E, E_\beta) dE, \quad (7)$$

where

$$A = \frac{1}{2\pi^3 c^5} |T^{\text{weak}}|^2 \quad (8)$$

and

$$f(p_\beta) = p_\beta E_\beta F(Z_A=2, p_\beta) = p_\beta E_\beta \frac{2\pi Z_A \eta}{e^{2\pi Z_A \eta} - 1}, \quad (9)$$

with the (nonrelativistic) Fermi function $F(Z_A=2, p_\beta)$ and the Sommerfeld parameter $\eta = -e^2 m_e / p_\beta$. The matrix element $|T^{\text{weak}}|^2$ is the squared weak transition-matrix element for the β decay, averaged over final nucleonic states and the directions of the β electron. It is important to note that for tritium decay the nucleonic matrix element (and thus A) is assumed to be constant.

Finally,

$$\begin{aligned} \tilde{I}(E, E_\beta) &= \frac{I(E, E_\beta)}{F(Z_A=2, p_\beta)} \\ &= \frac{1}{F(Z_A=2, p_\beta)} \sum_f |T_{fi}(E_\beta)|^2 \delta(E - E_{f0}^{\text{HeT}^+}), \end{aligned} \quad (10)$$

is the (squared) molecular transition amplitude T_{fi} contributing to a given energy E divided by the Fermi function. The inclusion of the Fermi function facilitates the discussion of the results of a beyond sudden-approximation treatment in terms of the more familiar sudden approximation, since in the latter case the prefactor $Af(p_\beta)$ for $Z=2$ is usually adopted [cf. Eq. (7) with, e.g., Eq. (5) in [6]].

Inserting the Lippmann-Schwinger ansatz [Eq. (4)] into $I(E, E_\beta)$ [defined in Eq. (10)] gives

$$\begin{aligned} I(E, E_\beta) &= \sum_f (|T_{fi}^{(0)}|^2 + 2 \text{Re}\{T_{fi}^{(0)*} T_{fi}^{(1)}\} + |T_{fi}^{(1)}|^2 \\ &\quad + 2 \text{Re}\{T_{fi}^{(0)*} T_{fi}^{(2)}\} \dots) \delta(E - E_{f0}^{\text{HeT}^+}) \quad (11) \\ &\equiv I^{(0,0)}(E, E_\beta) + I^{(0,1)}(E, E_\beta) + I^{(1,1)}(E, E_\beta) \\ &\quad + I^{(0,2)}(E, E_\beta) \dots, \quad (12) \end{aligned}$$

where the superscripts j characterize the order of the transition-matrix element $T_{fi}^{(j)}$. Since the expansion is performed with respect to the Coulomb interaction between the β particle and the remaining ion, the amplitudes $T_{fi}^{(j)}$ are expected to be of order η^j where η is the Sommerfeld parameter that is a measure of the Coulombic interaction and has been defined before. Thus the terms $I^{(j,k)}(E, E_\beta)$ are expected to be of order $\eta^{(j+k)}$. The term $I^{(0,0)}(E, E_\beta)$ corresponds due to the pointlike nature of the weak interaction to the sudden approximation.

D. Sudden approximation

1. Influence of the recoil

The explicit form of the first term in Eq. (12) is given by (see, e.g., I)

$$I^{(0,0)}(E, E_\beta) = F(Z_A - Z_A^{\text{eff}}, p_\beta) \sum_f \delta(E - E_{f0}^{\text{HeT}^+}) \times |\langle \psi_f^{\text{HeT}^+} | e^{i\mathbf{K}\mathbf{r}_{CA}} | \psi_0^{\text{T}_2} \rangle|^2 \quad (13)$$

$$= F(Z_A - Z_A^{\text{eff}}, p_\beta) \sum_f \delta(E - E_{f0}^{\text{HeT}^+}) P_f^{(0,0)}(\mathbf{K}) \quad (14)$$

$$= F(Z_A - Z_A^{\text{eff}}, p_\beta) I_{\text{mol}}^{(0,0)}(E, \mathbf{K}), \quad (15)$$

where $\psi_f^{\text{HeT}^+}$ and $\psi_0^{\text{T}_2}$ are the complete (rovibronic) molecular wave functions of a (bound or continuous) final state f of HeT^+ and the initial (ground) state of T_2 , respectively. $\mathbf{K} = -(\mathbf{p}_\beta + \mathbf{p}_\nu)$ is the recoil momentum and C denotes the center of mass of the molecule. To evaluate Eq. (13) the approximation

$$\mathbf{K}\mathbf{r}_{CA} = \mathbf{K} \left(\mathbf{r}_{BA} + \frac{m_e}{m_B} [\mathbf{r}_{1A} + \mathbf{r}_{2A}] \right) \approx \widetilde{\mathbf{K}}\mathbf{r}_{BA}, \quad (16)$$

where

$$\widetilde{\mathbf{r}}_{BA} = \frac{m_B}{m_A + m_B + 2m_e} \mathbf{r}_{BA}, \quad (17)$$

is usually adopted. It can be shown (see, e.g., I) that the main effect of this approximation is a neglect of excitations of states of symmetry different from Σ symmetry in the case of molecular tritium (S symmetry in the case of atomic tritium) within the sudden approximation. In addition, it has been shown that this is an effect of the order $(m_e K / m_B)^2$ which is in the case of tritium approximately equal to 4.5×10^{-5} . In view of the present experimental accuracy effects of these size should be negligible.

The probabilities $P_f^{(0,0)}(\mathbf{K})$ in Eq. (14) [within the approximation given in Eq. (16)] have been calculated very accurately within the Born-Oppenheimer approximation in [26] for the five most important electronic bound states of HeT^+ . In this work we will not improve on the bound-state results within the sudden approximation; instead, we will be concerned with the transitions into the electronic continuum of states. Here, a full treatment of the rovibrational motion

within the Born-Oppenheimer approximation as has been done for the five electronic bound states in [26] would be, however, extremely complicated. Additionally, it is a non-trivial question as to how such a calculation for the combined electronic and nuclear continuum should be performed. Therefore, one may simplify Eq. (13) even further by approximating the effects of nuclear motion on the transition probability. In the following, two alternative approaches will be discussed. The main difference between them is that the first one includes the effect of nuclear motion without broadening of the spectrum with respect to the energy, while the second approximation will yield an energetically broadened spectrum.

2. Closure approximation

The first approach is based on the fact that the transition probability to a certain *electronic* state n can be correctly given (using closure) as (see [27] or I)

$$P_n^{(0,0)} = \sum_{J,v} P_f^{(0,0)}(\mathbf{K}) = \int_0^\infty S_{n0}(R) |\xi_0(R)|^2 dR. \quad (18)$$

In this equation $\sum_{J,v}$ stands for the summation over all discrete and continuous rovibrational states belonging (within the Born-Oppenheimer picture) to the electronic state n . $R = |\mathbf{r}_{BA}|$ is the internuclear distance, $\xi_0(R)/R$ the radial nuclear wave function of a T_2 molecule in its rovibronic ground state, and $S_{n0}(R)$ is the electronic overlap matrix element between the electronic wave functions ϕ_n of HeT^+ and φ_0 of T_2 depending parametrically on R ,

$$S_{n0}(R) = \langle \phi_n(R) | \varphi_0(R) \rangle. \quad (19)$$

The disadvantage of calculating $P_n^{(0,0)}$ instead of $P_f^{(0,0)}$ is that it is an integrated transition probability and thus the energy information [needed for the analysis of the neutrino-mass experiment: cf. Eq. (13)] is lost, since the probability $P_n^{(0,0)}$ can be distributed arbitrarily over all possible rovibrational states $\{J, v\}$. Even the evaluation of the correct average excitation energy would require exactly the knowledge of this distribution and requires thus a full calculation of all probabilities $P_f^{(0,0)}$. The most natural choice for associating an approximate mean excitation energy \bar{E}_{n0} to $P_n^{(0,0)}$ is probably

$$\bar{E}_{n0} = \int_0^\infty E_{n0}(R) |\xi_0(R)|^2 dR. \quad (20)$$

The evaluation of $P_n^{(0,0)}$ (and \bar{E}_{n0}) may be further simplified by the following consideration. It is always possible to express $|S_{n0}(R)|^2$ in terms of an (infinite) polynomial in R , and therefore

$$P_n^{(0,0)} = \int_0^\infty \left(\sum_{l=0}^\infty c_l R^l \right) |\xi_0(R)|^2 dR \quad (21)$$

$$= \sum_{l=0}^\infty c_l \int_0^\infty R^l |\xi_0(R)|^2 dR \quad (22)$$

$$= \sum_{l=1}^\infty c_l \bar{R}^l + c_0 \quad (23)$$

is obtained. In Eq. (23), \bar{R}^l denotes the expectation value of R^l for the rovibrational ground state of T_2 . If the matrix elements $|S_{n0}(R)|^2$ depend on R (at least to a good approximation) linearly, one finds combining Eqs. (18) and (21) the special relation

$$P_n^{(0,0)} \approx c_1 \bar{R} + c_0 = |S_{n0}(\bar{R})|^2, \quad (24)$$

i.e., it is sufficient to evaluate the electronic matrix elements at the equilibrium distance \bar{R} of the T_2 molecule. The value $\bar{R} = 1.4283a_0$ (where $a_0 = 4\pi\epsilon_0\hbar^2/m_e e^2$ is the Bohr radius) has been reported in [28].

Because of the compactness of $\xi_0(R)$, it is actually sufficient to include in the integration over R only the range $0.6a_0 \leq R \leq 2.4a_0$. It may be noted that the linear approximation has already been used in [27] for the transitions to two electronic bound states of HeH^+ . In that work the linear behavior of $|S_{n0}(R)|^2$ for these states has also been demonstrated within a small range of R close to \bar{R} .

Finally, if the potential curve for the electronic ground state of T_2 is approximated by a harmonic potential, $\bar{R} = R_0$ is obtained where R_0 is the minimum of the potential curve. An identical result is evidently obtained if a clamped-nuclei approximation is used from the beginning. The only difference is that in the harmonic approximation the zero-point vibrational energy should be considered. However, as pointed out before, the definition of the excitation energy corresponding to an electronic state becomes dubious within all calculations based on the closure approximation. This dubiousity is not contained in the clamped-nuclei approximation; however, the energies might be incorrect.

Since the transition probabilities $P_n^{(0,0)}$ connected to five electronic bound states have been calculated very accurately in [26] (including the complete treatment of the rovibrational final states), one may use these results in order to improve on the approximation given in Eq. (24). An effective value of R , i.e., R_{eff} , can be obtained by minimizing

$$f_{\text{rel}}(R) = \frac{1}{5} \sum_{j=1}^5 \left| \frac{P_j^{(0,0)} - |S_{j0}(R)|^2}{P_j^{(0,0)}} \right| \quad (25)$$

with respect to R . The improvement of using R_{eff} instead of \bar{R} should be that it partly compensates the deviation of the electronic matrix elements from linear behavior in R and the otherwise omitted influence of the final rovibrational states on the transition probabilities. When applying R_{eff} one has of course to rely on the assumption that the electronic bound-state transition probabilities are representative also for the

transitions into the electronic continuum. Additionally, such a treatment only makes sense if $f_{\text{rel}}(R_{\text{eff}})$ is found to be satisfactorily small.

There is no reason to expect that the internuclear distance R_{eff} (found for the transition probabilities) is also optimal for obtaining approximate excitation energies E_{n0} . Thus one may search for a value R'_{eff} that minimizes

$$f'_{\text{rel}}(R) = \frac{1}{5} \sum_{j=1}^5 \left| \frac{\bar{E}_{j0} - |E_{j0}(R)|^2}{\bar{E}_{j0}} \right|, \quad (26)$$

where the mean excitation energies \bar{E}_{j0} can be obtained from the energy-resolved spectra (of the five most-important bound states) given in [26]. The final spectrum could then be constructed from $|S_{n0}(R_{\text{eff}})|^2$ and $E_{n0}(R'_{\text{eff}})$.

In order to evaluate the molecular transition-probability density in the electronic continuum of states we have, however, adopted a method where the total transition-probability density at a given energy is obtained without explicitly calculating the transition probability to a specific final state (see [10]). Briefly, we evaluate

$$\bar{I}_{\text{mol}}^{(0,0)}(E, R) = \sum_n \delta(E - E_{n0}(R)) |S_{n0}(R)|^2. \quad (27)$$

Thus, within the electronic continuum of states (where final states from different branches can contribute to the probability at a given excitation energy E) it is not possible with our method to resolve these final states. Since for the analysis of the tritium neutrino-mass experiments it is not needed to resolve molecular final states that are energetically degenerated [cf. Eq. (2)], this is no problem. However, it is then obviously not possible to evaluate the transition probabilities and excitation energies at two different values of R , i.e., at R_{eff} and R'_{eff} , respectively.

In conclusion, within the closure approximation the transition probability $I_{\text{mol}}^{(0,0)}(E, \mathbf{K})$ [see Eq. (15)] is approximated as

$$\begin{aligned} I_{\text{mol}}^{(0,0)}(E, \mathbf{K}) &\approx [I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{clos}} \equiv \bar{I}_{\text{mol}}^{(0,0)}(E, \mathcal{R}) \\ &= \sum_n |S_{n0}(\mathcal{R})|^2 \delta(E - E_{n0}^{\text{HeT}^+}(\mathcal{R})), \end{aligned} \quad (28)$$

where \mathcal{R} might be R_0 , \bar{R} , or R_{eff} .

It is important to note that the name ‘‘closure approximation’’ for the approximation described by Eq. (28) might be misleading. There is no approximation involved in performing the closure leading to Eq. (18), but there is a loss of the information about the energy distribution of the probabilities.

Thus, the approximation lies in the use of $\delta(E - E_{n0}^{\text{HeT}^+})$ instead of $\delta(E - E_{j0}^{\text{HeT}^+})$ for the analysis of the tritium neutrino-mass experiment [cf. Eq. (14)]. In addition, we have extended this approximation by the assumption that the probabilities (and excitation energies) behave linearly in R which allows us to evaluate them at *one* single value of R , as is expressed by Eq. (24).

3. Reflection approximation

Another approach to include the effects of nuclear motion is known as the reflection approximation and is derived in [29]. This approach is only valid for a bound initial state and dissociative final states and is based on the analysis of the nuclear wave functions of such a dissociative state.

Since the electronic motion is much faster than the nuclear one, it may be assumed that the transition from the initial T_2 state to the final HeT^+ state occurs to a good approximation without a change of the internuclear distance. If now the final state has purely dissociative character, the radial part of its nuclear wave function will have its largest amplitude close to the classical turning point (i.e., at the Born-Oppenheimer potential curve) and then an oscillatory behavior up to infinity. The overlap of such a dissociative nuclear wave function with the bound wave function describing the rovibrational ground state of T_2 will thus be nonzero only for values of the internuclear distance R close to the classical turning point of that specific nuclear final-state wave function. Thus it may be assumed that for a given internuclear distance only one of the rovibrational states belonging to a certain Born-Oppenheimer curve will contribute to the transition probability. If the recoil operator is neglected, the rotational state will not change in the transition. Therefore, the energy of the excited rovibrational state is approximately identical to the energy of the potential curve at the considered value of R .

On the other hand, the main effect of the recoil is an excitation of higher-lying rotational states, as can be seen from the results of the explicit calculations for the dissociative, but electronically bound states of HeT^+ . In our approximation this may be considered by assuming that the final rovibrational state is not the rotational ground state, but (in the case of HeT^+) the rotational state corresponding to $J \approx 23$. This may be taken into account by simply shifting the final-state energy by $E_{\text{shift}} = E_{J=23} - E_{J=0} \approx 1.3$ eV [12]. Within this approximation one obtains

$$[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}} = \int_0^\infty \left[\sum_n \delta(E - (E_{n0}^{\text{HeT}^+}(R) + E_{\text{shift}})) \right. \\ \left. \times |S_{n0}(R)|^2 |\xi_0(R)|^2 \right] dR \quad (29)$$

$$= \int_0^\infty \bar{I}_{\text{mol}}(E - E_{\text{shift}}, R) |\xi_0(R)|^2 dR. \quad (30)$$

One may try to improve on this approximation by obtaining E_{shift} from the probability distribution that has been calculated in [26] for the most important electronically bound, but *dissociative* states using a fit procedure. It may be noted that by obtaining an overall energy shift based on the calculation in [26] has another important advantage. In this way the zero-point energy of both calculations will automatically be adapted, and thus it will be possible to consistently combine the bound-state calculation of [9] with our calculation of the electronic continuum. Equation (29) is thus changed into

$$[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}} = \int_0^\infty \left[\sum_n \delta(E + E' - E_n^{\text{HeT}^+}(R)) \right. \\ \left. \times |S_{n0}(R)|^2 |\xi_0(R)|^2 \right] dR, \quad (31)$$

where $E' = E_0^{\text{HeT}^+} - E_{\text{shift}}$. The value $E_0^{\text{HeT}^+} = -80.89724$ eV [26,9] has been used, and E_{shift} is obtained with respect to this value.

There are two main advantages of the reflection approximation compared to the result obtained by the closure approximation given in Eq. (28). First, the excitation energies are well defined and the energy broadening due to the nuclear motion is—at least approximately—taken into account, while in the closure approximation the whole probability connected to one electronic transition is put at one energy value \bar{E}_n or $E_n(\mathcal{R})$. Second, if the spectrum is integrated over E , one finds [cf. Eqs. (29) and (18)]

$$\int [I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}} dE = \sum_n \int_0^\infty |S_{n0}(R)|^2 |\xi_0(R)|^2 dR \\ = \sum_n P_n^{(0,0)}. \quad (32)$$

Evidently, within the reflection approach no approximation is made in respect to $P_n^{(0,0)}$; only its energy distribution is approximated.

The reflection approximation will, however, only work if the electronic final states of HeT^+ have purely dissociative character within that range of internuclear distances R for which $|\xi_0(R)|^2$ is significantly larger than zero. Fortunately, this is the case for all but the electronic ground state of HeT^+ , and for the latter state a complete treatment exists already. Another disadvantage of the reflection approximation is that the calculation is much more laborious than the corresponding calculation within the closure approximation.

E. Beyond the sudden approximation

1. Previous results for atomic tritium

Even if the transition probability within the sudden approximation can be calculated to a sufficient accuracy, one may wonder about the validity of this approximation. From the viewpoint of perturbation theory the largest correction to $I^{(0,0)}(E, E_\beta)$, i.e., of order η , should be expected from the second term in Eq. (12), $I^{(0,1)}(E, E_\beta)$. Williams and Koonin [13] were the first who attempted to evaluate this term [together with $I^{(1,1)}(E, E_\beta)$] for *atomic* tritium as a function of the atomic excitation energy E . However, the approximation to perform closure over the (virtual) intermediate states of He^+ yielded an overestimation of $I^{(0,1)}(E, E_\beta) + I^{(1,1)}(E, E_\beta)$ by about a factor of 10, as pointed out by Arafune and Watanabe [16]. On the basis of certain assumptions it was demonstrated analytically in that work that in the case of a *one-electron atom* the term $I^{(0,1)}(E, E_\beta)$ is only of order η^2 and proportional to $I^{(0,0)}(E, E_\beta)$. However, this proportionality is only yielded by adding two different terms that, considered separately, are not proportional to $I^{(0,0)}(E, E_\beta)$. Since the closure approxi-

mation used in [13] yielded a cancellation of one of these two terms, the proportionality to $I^{(0,0)}(E, E_\beta)$ was not found. On the other hand, since the experiments do (so far?) not detect absolute intensities, only overall corrections to the sudden approximation that effectively *redistribute* the final-state probabilities of HeT^+ (compared to the sudden approximation) are of interest for this type of experiments. (This is of course different, if one is, e.g., interested in ft values.)

In order to emphasize the redistribution compared to changes that are proportional to the sudden approximation, the results of beyond sudden-approximation calculations for tritium were usually presented after renormalizing the total distribution to the one obtained within the sudden approximation. However, this point has not always been appreciated, and thus some additional confusion arose by comparing renormalized with nonrenormalized results. The obvious reason for this misunderstanding is that in the case of experiments that analyze the total (integrated) transition probability (aiming for example for ft values), also proportional changes induced by higher-order corrections have to be included in the analysis.

It is important to note that, if the correction to the sudden approximation is found to contain a proportional and a nonproportional contribution, both terms have to be included before the renormalization is performed. However, the proportional term will contribute to a possible redistribution only via the nonproportional part and thus its effect will be determined by the magnitude of this nonproportional term (see the Appendix).

Considering the problem of calculating the corrections to the sudden approximation for arbitrary atoms, Durand and Lopez [30,31] have derived explicit expressions for these corrections within a relativistic treatment of the β particle and the neutrino, and some approximations that are mainly based on the consideration of the size of different terms. The result obtained in that work allows us to treat the atomic electrons on any level of approximation. However, their derivation is formulated for atoms, not for molecules. In addition, explicit expressions are derived only for the $L=0$ component of the partial-wave expansion of the β particle's wave function, and this wave function is assumed to be a plane wave. While taking the nonrelativistic limit of their expression, the authors were able to confirm the result given by Arafune and Watanabe for the specific case of the tritium atom. However, it may be noted that the reported numerical value is much larger than the one given by Arafune and Watanabe. It seems very probable that the value given in [31] has to be understood as being the magnitude of the overall change without renormalization to the sudden approximation.

The main outcome of all atomic works (despite obvious discrepancies in the quantitative results) is that the term $I^{(0,1)}(E, E_\beta)$ is found to be of order η^2 , while it would be expected that it should be of order η . In addition, $I^{(0,1)}(E, E_\beta)$ turned out to be purely proportional to $I^{(0,0)}(E, E_\beta)$.

The mathematical reason for the surprising result that $I^{(0,1)}(E, E_\beta)$ is of order η^2 and not η is given by the fact that according to, e.g., [31], the product $T_{n0}^{(0)*} T_{n0}^{(1)}$ reveals the structure

$$T_{n0}^{(0)*} T_{n0}^{(1)} = \langle \psi_n^{\text{He}^+} | \psi_0^{\text{T}} \rangle^* \sum_{j'} \langle \psi_n^{\text{He}^+} | D | \psi_{j'}^{\text{He}^+} \rangle \langle \psi_{j'}^{\text{He}^+} | \psi_0^{\text{T}} \rangle \quad (33)$$

$$= \langle \psi_n^{\text{He}^+} | \psi_0^{\text{T}} \rangle^* \sum_{j'} (\eta^2 \langle \psi_n^{\text{He}^+} | D_R | \psi_{j'}^{\text{He}^+} \rangle + i \eta \langle \psi_n^{\text{He}^+} | D_I | \psi_{j'}^{\text{He}^+} \rangle) \langle \psi_{j'}^{\text{He}^+} | \psi_0^{\text{T}} \rangle, \quad (34)$$

where $D = \eta^2 D_R + i \eta D_I$ is some complex operator and D_R and D_I are purely real ones. The states j' occurring in these equations are (virtual) intermediate states arising from the Born-type expansion of the final-state wave function. However, the wave functions of a one-electron system (that can be given analytically) are either purely real (bound states) or contain only an r -independent overall complex phase (the Coulombic phase shift) that can be taken out of the integration over r and cancels out, since both $\psi_n^{\text{He}^+}$ and $\psi_{j'}^{\text{He}^+}$ occur always pairwise in the form ψ times ψ^* . Therefore, one finds $\text{Re}\{T_{n0}^{(0)*} T_{n0}^{(1)}\} \propto \eta^2$ and $\text{Im}\{T_{n0}^{(0)*} T_{n0}^{(1)}\} \propto \eta$. Since only the real part of $T_{n0}^{(0)*} T_{n0}^{(1)}$ contributes to $I^{(0,1)}(E, E_\beta)$ [see Eq. (11)], one finds in the atomic case $I^{(0,1)}(E, E_\beta) \propto \eta^2$.

2. Comparison of the atomic and the molecular problem

Since changes in the final-state distribution that are only of order η^2 are expected to be too small to be of importance for the analysis of the present experimental data and especially too small for explaining the excess rates that are found in the experimental studies, the most interesting question to answer seems to be whether also for the case of molecular tritium only corrections of order η^2 occur. Since $I^{(0,1)}(E, E_\beta)$ is the only term that can be of order η , the main motivation for this work has been to investigate if the same properties, i.e., magnitude of order η^2 and proportionality to $I^{(0,0)}(E, E_\beta)$, which are found for the one-electron T atom, are also valid for the two-electron molecule T_2 . Of course, it is worth noticing that either large terms or a large number of terms of order η^2 could also yield a substantial redistribution of the final-state distribution, and therefore one may investigate also this possibility, requiring a consideration of all terms of order η^2 .

What are the main reasons to believe that $I^{(0,1)}(E, E_\beta)$ might be substantially different in the molecular case compared to the atomic case?

First, already for many-electron atoms electronic (autoionizing) resonances exist in the electronic continuum of states. These wave functions cannot just be described with the aid of a real one-electron wave function times an overall complex phase describing the Coulombic phase shift, but they are intrinsically complex. If these phases are r dependent, they cannot be taken out of the integral. Thus, the resulting complex matrix elements, i.e., the overlap matrix elements and the ones of the D_I operator, cannot be expected to possess the same phase factor. Thus both the D_R and the D_I terms in Eq. (34) could be complex and their real parts would contribute to $I^{(0,1)}(E, E_\beta)$. In this case a contribution of order η could occur. The intrinsic complexity of the wave function might be even enhanced if the case of the autoionizing state of a molecular system is considered. In addition, it

has been reported in [10] that within the sudden approximation more than 8% of the total transition probability is contained in such autoionizing states in the case of HeT^+ which is more than half of the overall probability in the continuum. (It may be also noted that in the case of the decay of atomic T, where of course no such resonances occur, the overall continuous contribution is less than 3%.)

Second, HeT^+ belongs to the symmetry group $C_{\infty v}$ while the He^+ ion belongs to the spherical symmetry group. Consider now Eq. (33) under the assumption that $D=D(\mathbf{r})$ is a one-electron operator (or a sum of one-electron operators) that has no spherical symmetry and can be expressed as a linear combination of all possible partial waves $\{L, M\}$, as is the case for the $e^{i\mathbf{k}\mathbf{r}}$ operator. For any atomic decay the product $T_{n0}^{(0)*} T_{n0}^{(1)}$ in Eq. (33) is nonzero only if all three states 0, j' , n that are involved have the same symmetry, since otherwise one or both of the overlap matrix elements S_{n0} or $S_{j'0}$ would be zero. However, if the states j' and n have the same symmetry $\{l, m\}$, only the totally symmetric component (partial wave with $L=M=0$) of the operator $D(\mathbf{r})$ can give a nonzero matrix element $\langle n|D|j'\rangle$. Thus it is sufficient for atoms to consider only the totally symmetric part of the operator D .

However, in the molecular case the situation is different. For molecular tritium—even though it possesses still comparatively high symmetry—the $^1\Sigma_g^+$ ground state of T_2 limits the states j' and n of HeT^+ to $^1\Sigma$ for nonzero overlap matrix elements S_{n0} and $S_{j'0}$. However, in the partial-wave decomposition of D all terms with $M=0$, but arbitrary value of L , can give nonzero matrix elements $\langle n|D|j'\rangle$. The consideration of only the partial wave with $L=M=0$ in the evaluation of $I^{(0,1)}(E_\beta)$ is thus not automatically justified in the molecular case. (In the case of more complex molecules than T_2 even the limitation to $M=0$ will break down.)

Third, as was already noted in [16], the commutator relation that was derived in that work and showed convincingly the proportionality of $I^{(0,1)}(E, E_\beta)$ to $I^{(0,0)}(E, E_\beta)$ is only valid for the very specific case of a one-electron atom.

In the derivation of $I^{(0,1)}(E, E_\beta)$ and $I^{(1,1)}(E, E_\beta)$ that is described in detail in I we have chosen to follow the principal concept (valid only for atoms) behind [31], since it has the advantage of transparency and generality. We have extended that concept in order to obtain explicit expressions for all partial waves (and not only the special one with $L=M=0$) and to become applicable to any molecular system (and not only to atoms). In addition, we felt it is important to discuss carefully every approximation that is included and even to suggest possible tests of the validity of these approximations. Even though the main intention was a careful investigation of the tritium β decay, the generality of the approach should allow its application to a number of related problems (like the question of ft values or neutrino-induced β decays).

3. Corrections to the sudden approximation for T_2

A full derivation of the higher-order corrections to the sudden amplitude, i.e., of $I^{(0,1)}(E, E_\beta)$ and $I^{(1,1)}(E, E_\beta)$, is given in I. Since the effect of nuclear motion and the recoil on the sudden amplitude is comparatively small (1–2 % relative changes for the electronic bound states compared to a

clamped-nuclei calculation [26]) and since there seems to be no reason to believe that this will be drastically different in the case of higher-order corrections, we will simplify the calculation by using the closure approximation for intermediate and final rovibrational states. As outlined in I, it is possible to improve on this by taking the nuclear motion (at least approximately) into account, but this seems to be appropriate only if the corrections within the closure approximation turn out to be significant.

If only the terms up to the second order with respect to the Sommerfeld parameter η are taken into account, if only the first term of a Taylor expansion of the occurring logarithmic term is considered, and if again it is assumed that the β electrons emitted in all directions or from a source containing randomly oriented molecules are detected, the interference term $P_{fi}^{(0,1)}(p_\beta) = 2 \text{Re}\{T_{fi}^{(0)*} T_{fi}^{(1)}\}$ is given by (see I)

$$P_{fi}^{(0,1)}(p_\beta) \approx F(Z_A - Z_A^{\text{eff}}, p_\beta) \{ [P_n^{(0,1)}(p_\beta)]_{\text{el}} + [P_n^{(0,1)}(p_\beta)]_{\text{nuc}} \}, \quad (35)$$

where

$$[P_n^{(0,1)}(p_\beta)]_{\text{el}} = e^2 \text{Re} \left\{ \sum_{n'} \langle \varphi_0 | \phi_n \rangle \langle \phi_n | \sum_{k=1}^{N_e} i \frac{2E_\beta}{p_\beta c^2} \ln \frac{r_{Ak}}{a_0} - \frac{\bar{E}_{n,n'}^{\text{HeT}^+} E_\beta^2}{p_\beta^2 c^4} r_{Ak} - \left(\frac{E_\beta}{p_\beta^2 c^2} + \frac{1}{E_\beta} \right) \times \frac{1}{r_{Ak}} | \phi_{n'} \rangle \langle \phi_{n'} | \varphi_0 \rangle \right\} \quad (36)$$

and

$$[P_n^{(0,1)}(p_\beta)]_{\text{nuc}} = e^2 |\langle \varphi_0 | \phi_n \rangle|^2 \left[Z_A \left(\frac{8E_\beta}{\pi p_\beta c^2} + \frac{8p_\beta}{3\pi E_\beta} \right) + Z_B \left(\frac{E_\beta}{p_\beta^2 c^2} + \frac{1}{E_\beta} \right) \langle \xi_{00}^0 | \frac{1}{R} | \xi_{00}^0 \rangle \right]. \quad (37)$$

It may be noted that Eq. (35) differs from the corresponding equation in I by the appearance of the Fermi function, but for $Z_A^{\text{eff}}=2$ (as adopted in I) one has $F(Z_A - Z_A^{\text{eff}}, p_\beta)=1$. In the cases where $Z_A^{\text{eff}} \neq 2$ there are, however, three more approximations included in the derivation of Eq. (35). First, the integration over \mathbf{r}_β , the position-space coordinate of the β electron, has been performed by approximating the Coulombic waves by plane waves. Second, in the integration over all possible virtual momenta of the β electron it has been assumed that $F(Z_A - Z_A^{\text{eff}}, p'_\beta) \approx F(Z_A - Z_A^{\text{eff}}, p_\beta)$ where p'_β is the momentum of the β electron created in a (virtual) intermediate state. Third, in this case the relation $E_\beta^2 \neq p_\beta^2 c^2 + m_e^2 c^4$ is only approximately valid, since the β electron has now also a nonzero potential energy. On the other hand, none of these three approximations have to be made for $Z_A^{\text{eff}}=2$, since then the β electron is correctly described by a plane wave. Because of the structure of $P_{fi}^{(0,1)}(p_\beta)$ in Eq. (35), one finds

$$I^{(0,1)}(E, E_\beta) = I_{\text{nuc}}^{(0,1)}(E, E_\beta) + I_{\text{el}}^{(0,1)}(E, E_\beta). \quad (38)$$

In the case of tritium it is sufficient to consider the non-relativistic limit of Eq. (38), since the kinetic end-point energy is about 18.6 keV and thus one has $E_\beta \approx m_e c^2$. Consequently, one has (using atomic units where $e = m_e = 1$ and $c \approx 137.036$)

$$\frac{E_\beta}{p_\beta c^2} \approx \frac{1}{p_\beta} = -\eta \quad \text{and} \quad \left(\frac{E_\beta}{p_\beta^2 c^2} + \frac{1}{E_\beta} \right) \approx \eta^2. \quad (39)$$

Thus, one obtains

$$I_{\text{nuc}}^{(0,1)}(E, E_\beta) \approx I^{(0,0)}(E, E_\beta) \left\langle \xi_{00}^0 \left| -Z_A^{\text{eff}} \left[\frac{8}{\pi} \eta + \frac{8}{3\pi} \frac{1}{\eta c^2} \right] + Z_B \eta^2 \frac{1}{R} \right| \xi_{00}^0 \right\rangle \quad (40)$$

and

$$I_{\text{el}}^{(0,1)}(E, E_\beta) \approx F(Z_A - Z_A^{\text{eff}}, p_\beta) \left\{ \sum_n \text{Re} \left\{ \sum_{n'} S_{n0}^*(\mathcal{R}) D_{nn'}(\mathcal{R}) S_{n'0}(\mathcal{R}) \right\} \times \delta(E - E_{n0}^{\text{HeT}^+}(\mathcal{R})), \right. \quad (41)$$

with

$$D_{nn'}(\mathcal{R}) = \langle \phi_n(\mathcal{R}) \left| \sum_{k=1}^2 \left[-\eta^2 \left(\frac{1}{r_{Ak}} - r_{Ak} E_{nn'}^{\text{HeT}^+}(\mathcal{R}) \right) - 2\eta i \ln \frac{r_{Ak}}{a_0} \right] \right| \phi_{n'}(\mathcal{R}) \rangle. \quad (42)$$

Again, \mathcal{R} is either \bar{R} , R_0 , or R_{eff} of T_2 . For consistency reasons, $I^{(0,0)}(E, E_\beta)$ in Eq. (40) should be evaluated within the approximation given in Eq. (28).

When Eq. (38) is compared with the corresponding equation for a one-electron atom, one notices two differences. First, in the electronic part [Eq. (42)] a sum has to be performed over the electrons that were initially bound in T_2 . However, the type of the matrix elements is otherwise completely identical. Second, in the nuclear part [Eq. (40)] there is now a contribution arising from the ‘‘spectator’’ nucleus B that is of course absent in an atom. Compared to the results given by Arafaune and Watanabe (or Durand and Lopez) one notices in addition the occurrence of a term that is proportional to Z_A^{eff} . Since this term depends on the decaying nucleus, it is also present in the atomic case. However, since Arafaune and Watanabe chose $Z_A^{\text{eff}} = 0$, it is (correctly) missing in their treatment. However, as will be explained later, the occurrence of this term may give some insight regarding the magnitude of the corrections to the sudden approximation.

Since the only structural difference of the matrix elements in the molecular case compared to the atomic one is the occurrence of the contribution of the term arising from the spectator nucleus, and since this contribution is obviously of order η^2 , one can already now exclude one of the three

possible reasons for obtaining a different result for $I^{(0,1)}(E, E_\beta)$ in the molecular compared to the atomic case: There will be no effect of order η caused by the non-spherical symmetry of the molecule. Thus it remains to investigate the possible effect of the resonant continuum as well as the question of strict proportionality to $I^{(0,0)}(E, E_\beta)$. This investigation will be done by calculating $I^{(0,1)}(E, E_\beta)$ explicitly. Before displaying and discussing the results, a brief overview of the computational details will be given in the next section. As will be seen, the calculation of $I_{\text{el}}^{(0,1)}(E, E_\beta)$ for final states lying in the electronic continuum will require an extension of the complex-scaling method to become applicable to the extraction of interference terms.

III. COMPUTATIONAL DETAILS

A. Calculations within the sudden approximation

In order to evaluate the molecular final-state distribution within the sudden approximation, we have used mainly the apparatus presented in [10]. Thus, for a given internuclear distance a configuration-interaction (CI) calculation in prolate-spheroidal coordinates with explicitly correlated basis functions is performed. For electronic bound states the probabilities were calculated with the aid of the program that has been written and used by Kotos *et al.* in their calculations reported in [32,9,26,33]. For the electronic continuum the method reported in [10] was used and its detailed description will be presented elsewhere. In order to check the reliability of the results of the previous work and to extend the range of the considered excitation energy, the program has further been modified to allow now for 400 basis functions.

For the T_2 ground state we have used 104 basis functions and the (R -dependent) nonlinear parameters given in [34]. The largest integer exponents of the electronic coordinates ξ_i and η_i ($i = 1, 2$) were 6, and the powers of r_{12} were limited to 0, 1, and 2. In the considered range of internuclear distances the ground-state energies obtained (in the clamped-nuclei approximation) with this basis set are $-1.174\,471\,7\,E_h$ (at $R = 1.40\,a_0$), $-0.769\,625\,9\,E_h$ (at $R = 0.60\,a_0$), and $-1.102\,415\,3\,E_h$ (at $R = 2.40\,a_0$). These values may be compared to the corresponding best available theoretical data, i.e., $-1.174\,475\,7\,E_h$ (at $R = 1.40\,a_0$), $-0.769\,635\,3\,E_h$ (at $R = 0.60\,a_0$), and $-1.102\,422\,6\,E_h$ (at $R = 2.40\,a_0$) [33].

In the case of HeT^+ the situation is more complicated, since one has to aim for one basis set that is capable of describing the whole continuum as accurate as possible, since it would be difficult to optimize the basis set for every state separately. In addition, in order to describe the continuous spectrum up to 800 eV, a reasonable number of states within this energy range is needed. Thus, there have been two reasons for extending the basis set from 200 to 400 basis functions. First, we wanted to investigate the convergence of the previous results. Second, the extension to higher excitation energies requires more states within that energy range.

We have chosen the same nonlinear parameters as in [10] (given in Table I of that work). Again we have used two sets of nonlinear parameters and thus two basis sets. However,

since the parameters of basis set 2 in [10] turned out to give in general better results, we will refer in this work to the basis set with that nonlinear parameters as basis set 1, while the nonlinear parameters corresponding to basis 1 in [10] will now define basis 2. Different from the basis set for T_2 , the nonlinear parameters of both basis sets for HeT^+ were kept unchanged for different values of R . The main difficulty was to include 200 more combinations of integer exponents without running into numerical problems due to linear dependencies. While enlarging the basis set, it was experienced that the polynomial fit that is used in order to extrapolate the Hamiltonian matrix elements into the complex plane (see [10]) is a good indication whether the basis set is close to linear dependencies or not. If this was the case, the least-squares fit immediately reported a large number of matrix elements that could not be fitted satisfactorily according to the NAG library routine E04HFF that has been used.

In order to evaluate $[I_{mol}^{(0,0)}(E, \mathbf{K})]_{refl}$ given in Eq. (29), the radial Schrödinger equation for the nuclear motion of T_2 has been solved numerically with the aid of program LEVEL6.0 [35] using the Born-Oppenheimer potential curve given in [36]. With the aid of $I_{mol}^{(0,0)}(E, R_j)$ calculated at either 45 or 55 discrete values R_j , $[I_{mol}^{(0,0)}(E, \mathbf{K})]_{refl}$ has been calculated by approximating the integral in Eq. (30) by a sum using the weights $w_j = N |\xi_0(R_j)|^2 \Delta R_j$ where N has been chosen in such a way that $\sum w_j = 1$ is fulfilled. The distribution of the R_j values ranged from $R = 0.6 a_0$ to $2.4 a_0$ with varying distances so that the range with the largest values of $|\xi_0(R)|^2$ was covered most densely. The weight factors for R_{min} and R_{max} were less than 1.0×10^{-10} , so that the contribution from R values outside the considered range can be seen to be negligibly small.

It turned out that due to the sharp structure of the resonances and due to their strong dependence on R , an extremely fine mesh would be needed in order to get rid of spurious spikes that are caused by the use of a sum instead of an integral. This problem was overcome by either binning the spectrum or smoothing it out by first binning and then performing a spline. Of course, in both cases the resolution is limited by the size of the bins, and no structure with a smaller width can be visible from such a spectrum.

B. Calculation beyond the sudden approximation

As is transparent from Eq. (38), the interference term $I^{(0,1)}(E, E_\beta)$ is a sum of a nuclear and an electronic term, i.e.,

one term that arises from the Coulombic interaction of the β electron with the nuclei and one term that arises from the interaction with the two (initially) bound electrons. The evaluation of these two terms will be discussed separately in the next two subsections.

1. Nuclear term $I_{nuc}^{(0,1)}(E, E_\beta)$

As can be seen from Eq. (40), the electronic matrix elements contained in the nuclear term are identical to the ones occurring for the sudden amplitude. Their evaluation has been already described in the last section. The remaining task is thus the evaluation of

$$D_{nuc}(Z_A^{eff}, Z_B, \eta) = \langle \xi_{00}^0 | -Z_A^{eff} \left[\frac{8}{\pi} \eta + \frac{8}{3\pi} \frac{1}{\eta c^2} \right] + Z_B \eta^2 \frac{1}{R} | \xi_{00}^0 \rangle \quad (43)$$

$$= -Z_A^{eff} \left[\frac{8}{\pi} \eta + \frac{8}{3\pi} \frac{1}{\eta c^2} \right] + Z_B \eta^2 \langle \xi_{00}^0 | \frac{1}{R} | \xi_{00}^0 \rangle \quad (44)$$

$$= -Z_A^{eff} \left[\frac{8}{\pi} \eta + \frac{8}{3\pi} \frac{1}{\eta c^2} \right] + Z_B \eta^2 \overline{R^{-1}}. \quad (45)$$

For the rovibrational ground state of T_2 the value $\overline{R^{-1}} = 0.705\,671\,3 a_0^{-1}$ has been reported in [28]. In later works the potential curve for T_2 has been slightly improved by the same authors, but it can be expected that the value given in the earlier work should still be more than accurate enough for our purposes.

2. Electronic term $I_{el}^{(0,1)}(E, E_\beta)$

Two problems have to be solved to obtain $I_{el}^{(0,1)}(E, E_\beta)$. First, in contrast to the sudden amplitude where only overlap integrals were involved, one has now to calculate three types of matrix elements that occur in Eq. (42):

$$\begin{aligned} \sum_{n'} D_{nn'} S_{n'0} = & -\eta^2 \left(\sum_{n'} \langle \phi_n | \sum_{k=1}^2 \frac{1}{r_{Ak}} | \phi_{n'} \rangle \langle \phi_{n'} | \varphi_0 \rangle + \sum_{n'} E_{nn'}^{HeT^+} \langle \phi_n | \sum_{k=1}^2 r_{Ak} | \phi_{n'} \rangle \langle \phi_{n'} | \varphi_0 \rangle \right) \\ & - 2i \eta \sum_{n'} \langle \phi_n | \sum_{k=1}^2 \ln \frac{r_{Ak}}{a_0} | \phi_{n'} \rangle \langle \phi_{n'} | \varphi_0 \rangle \end{aligned} \quad (46)$$

$$= -\eta^2 \left(\langle \phi_n | \sum_{k=1}^2 \frac{1}{r_{Ak}} | \varphi_0 \rangle + \sum_{n'} E_{nn'}^{HeT^+} \langle \phi_n | \sum_{k=1}^2 r_{Ak} | \phi_{n'} \rangle \langle \phi_{n'} | \varphi_0 \rangle \right) - 2i \eta \langle \phi_n | \sum_{k=1}^2 \ln \frac{r_{Ak}}{a_0} | \varphi_0 \rangle. \quad (47)$$

The $1/r_{Ak}$ matrix elements are of course proportional to the nuclear-attraction integrals with respect to nucleus $A=^3\text{He}$. They are therefore already contained in the computer code for evaluating the CI matrix. However, only the complete matrix of the potential energy (nuclear attraction in respect to both nuclei and electron-electron repulsion) is easily accessible. Thus a similar procedure as in [37] has been adopted. In particular, it is possible to extract the $1/r_{Ak}$ matrix elements out of the potential-energy matrices for two different charges Z_A . Of course, such procedure can only be applied if matrix elements between HeT^+ wave functions are considered. Thus, Eq. (46) instead of Eq. (47) has to be used. The correctness of the extraction procedure has been tested by evaluating these matrix elements also directly by a separate program where, however, the simplification was introduced that only basis sets with even powers of r_{12} were allowed.

Since in the prolate-spheroidal coordinate system in which the wave functions are expressed one has $r_{Ak}=0.5R(\xi_k+\eta_k)$, it is possible to reduce these matrix elements to a linear combination of overlap integrals where the integer exponents of ξ_k or η_k are changed. This is also true for the case when antisymmetrized basis functions are considered.

Finally, one is left with the integrals of the $\ln(r_{Ak}/a_0)$ operator. In this case two approaches have been tried, but in both cases the basis sets were of the simplified type; i.e., only even powers of r_{12} were allowed. In the first approach the two-dimensional integration over ξ_k and η_k ($k=1$ or 2) has been performed numerically. Alternatively, the $\ln(r_{Ak}/a_0)$ has been expanded in a Taylor series which results in an (infinite) number of products of simple one-dimensional integrals. The latter approach revealed, however, very slow convergence, so that it was only used for the purpose of checking.

While the problem of evaluating the integrals between the basis functions is of numerical nature, a more fundamental aspect had also to be considered: How can the matrix elements be evaluated in the case when the intermediate or final states are lying in the electronic continuum? In contrast to the sudden approximation where the absolute square of a matrix element was required, one has now to evaluate an interference term between two amplitudes. Even though the problem of obtaining interference terms within the complex-coordinate method has been considered in [22], no implementational details were given and so far it has not been applied.

C. Calculation of interference terms between scattering amplitudes

In order to evaluate the interference term between the zeroth-order and first-order amplitudes in the electronic continuum we will build on the same idea that was used for the

calculation of the transition probability within the sudden approximation in [10]. We may recall the following three basic ideas of that approach.

First, in the case of a *real* function $F(E)$ one has the relation

$$F(E) = \frac{1}{\pi} \text{Im} \left\{ \lim_{\epsilon \rightarrow 0} \int_a^b \frac{F(E')}{E - E' + i\epsilon} dE' \right\}. \quad (48)$$

Second, in order to obtain the transition probability, one may define $F(E)$ as

$$F(E) = |\langle \phi_E | \varphi_0 \rangle|^2 = \langle \varphi_0 | \phi_E \rangle \langle \phi_E | \varphi_0 \rangle, \quad (49)$$

which automatically fulfills the requirement that $F(E)$ be real.

Third, introducing a discrete representation of the continuous states by using a finite basis set requires the application of the complex-coordinate method in order to obtain a nonzero transition probability

$$\bar{I}_{\text{mol}}^{(0,0)}(E, \mathcal{R}) = \frac{1}{\pi} \text{Im} \left\{ \sum_n \frac{\langle \varphi_0^{\theta*} | \phi_n^\theta \rangle \langle \phi_n^{\theta*} | \varphi_0^\theta \rangle}{E - E_n^\theta} \right\}, \quad (50)$$

where ϕ_n^θ and E_n^θ are the *complex* wave functions and energy eigenvalues obtained from the dilated matrix eigenvalue problem

$$\mathbf{H}(\theta) \mathbf{c}_n^\theta = E_n^\theta \mathbf{S} \mathbf{c}_n^\theta, \quad \phi_n^\theta = \mathbf{h} \mathbf{c}_n^\theta, \quad (51)$$

within the (real) basis \mathbf{h} . In Eq. (50) we have used the notation

$$\langle \phi_n^{\theta*} | = [\mathbf{c}_n^\theta]^\text{T} \mathbf{h}^\text{T}. \quad (52)$$

In order to evaluate the interference term we may now define $F(E)$ as

$$F(E) = \text{Re} \left\{ \sum_{n'} \langle \varphi_0 | \phi_E \rangle \langle \phi_E | D | \phi_{n'} \rangle \langle \phi_{n'} | \varphi_0 \rangle \right\} \quad (53)$$

$$= \text{Re} \{ \langle \varphi_0 | \phi_E \rangle \langle \phi_E | \tilde{D} | \varphi_0 \rangle \} \quad (54)$$

$$= \frac{1}{2} (\langle \varphi_0 | \phi_E \rangle \langle \phi_E | \tilde{D} | \varphi_0 \rangle$$

$$+ \langle \varphi_0 | \tilde{D}^* | \phi_E \rangle \langle \phi_E | \varphi_0 \rangle), \quad (55)$$

with

$$\tilde{D} = \sum_{n'} D | \phi_{n'} \rangle \langle \phi_{n'} |. \quad (56)$$

[It may be noted that closure cannot be automatically performed in Eq. (56), since D contains one term that depends on $E_{n'}$.] Introducing a finite basis set and applying the complex-scaling method gives

$$I_{\text{el}}^{(0,1)}(E, E_\beta) = \frac{1}{2\pi} \text{Im} \left\{ \sum_n \sum_{n'} \frac{\langle \varphi_0^{\theta*} | \phi_n^\theta \rangle \langle \phi_n^{\theta*} | D(\theta) | \phi_{n'}^\theta \rangle \langle \phi_{n'}^{\theta*} | \varphi_0^\theta \rangle + \langle \varphi_0^{\theta*} | \phi_{n'}^\theta \rangle \langle \phi_{n'}^{\theta*} | D^*(\theta^*) | \phi_n^\theta \rangle \langle \phi_n^{\theta*} | \varphi_0^\theta \rangle}{E - E_n^\theta} \right\}, \quad (57)$$

TABLE I. Total transition probabilities (in %) connected to five electronic bound states of HeT⁺ with ¹Σ symmetry. First column: probabilities calculated at R₀=1.40 a₀. Second and third columns: probabilities calculated at the effective internuclear distance R_{eff}=1.431 a₀ for basis sets 1 and 2, respectively. Fourth column: results within the reflection approximation. Last column: transition probabilities obtained in a complete treatment of the rovibrational initial and final states (taken from [26]).

<i>n</i>	$P_n^{(0,0)}(R_0)$ (Basis 1)	$P_n^{(0,0)}(R_{\text{eff}})$ (Basis 1)	$P_n^{(0,0)}(R_{\text{eff}})$ (Basis 2)	$[P_n^{(0,0)}]_{\text{refl}}$ (Basis 1)	$P_n^{(0,0)}$ [26]
1	58.238	57.430	57.423	—	57.412
2	16.808	17.418	17.422	17.346	17.357
3	7.878	7.752	7.754	7.758	7.762
4	0.805	0.790	0.790	0.782	0.777
6	0.930	0.914	0.923	0.916	0.901

with

$$D(\theta) = D_R(\theta) + iD_I(\theta) \quad \text{and} \quad D^*(\theta^*) = D_R(\theta) - iD_I(\theta). \quad (58)$$

IV. RESULTS

A. Sudden amplitude

The discussion of the results within the sudden approximation will split in three parts: the electronic bound states, the energy range close to the ionisation threshold that is dominated by the electronic (autoionizing) resonances, and the high-energy range (above 100 eV).

1. Electronic bound states

Since the transition probabilities to the five most important electronic bound states of HeT⁺ have been calculated very accurately within the sudden approximation (including a full treatment of the rovibrational structure of the initial state and the final states) [26], those results will be used in this work to obtain R_{eff} and E_{shift} which are needed for the approximate treatment of the nuclear motion in the electronic continuum. In addition, the transition probabilities to the electronic bound states will serve as an indication for the accuracy of the adopted approximations for treating the nuclear motion.

Within the closure approximation described in Sec. II D 2 one has to evaluate first R_{eff} by minimizing $f_{\text{rel}}(R)$ [see Eq. (25)] with respect to R . In this fit procedure the five most important electronic bound states of HeT⁺ have been used. The probabilities $P_n^{(0,0)}$ were taken from [26], whereas the quantities $S_{n0}(R)$ have been calculated in this work with the aid of the basis sets described in Sec. III A. The value R_{eff}=1.431 a₀ is obtained with both basis sets. The average relative error for the five transitions [defined as $f_{\text{rel}}(R_{\text{eff}}) \times 100\%$] is 0.67% for basis set 1 and 0.89% for basis set 2. The smallness of these errors indicates that the choice of R_{eff} is (at least for the five bound states) well justified; i.e., it is possible to adopt the approximation $I_{\text{mol}}^{(0,0)}(E, \mathbf{K}) \approx \bar{I}_{\text{mol}}^{(0,0)}(E, R_{\text{eff}})$ [anticipated in Eq. (28)]. This can also be seen from Table I where $P_n^{(0,0)}$ (from [26]) and $P_n^{(0,0)}(R_{\text{eff}})$ (calculated in this work) are compared for five of the six lowest-lying ¹Σ states n of HeT⁺.

The value that has been found for R_{eff} is very close to the mean internuclear distance of the T₂ molecule in its rovibrational ground state, i.e., to $\bar{R} = 1.428 a_0$ (reported in [28]). Since the T₂ ground state has its minimum at a smaller value of R than the bound states of HeT⁺, a shift to a (slightly) larger value than 1.428 a₀ can be expected—and it has been found. The sensitivity of the results with respect to the choice of R can be estimated from the values $f_{\text{rel}}(R_0 = 1.4) \times 100\% = 2.55\%$ and $f_{\text{rel}}(\bar{R} = 1.428) \times 100\% = 0.69\%$ (both values obtained for basis set 1). Evidently, the difference between the use of R_{eff} and \bar{R} is negligible, while the use of R₀ is not a very accurate approximation, since the overlap matrix elements are sensitive to changes of the internuclear distance that are of the size 0.01 a₀. This is evident from Table I where $P_n^{(0,0)}(R_0)$ is also given. Finally, the basis-set convergence can be estimated from a comparison of the values found for $P_n^{(0,0)}(R_{\text{eff}})$ for the two basis sets (see Table I).

In order to evaluate the transition probability according to the reflection approximation [Eq. (30)], the transition probabilities $\bar{I}_{\text{mol}}^{(0,0)}(E_n, R)$ have been evaluated for the second, third, fourth, and sixth lowest-lying electronic ¹Σ states as a function of the internuclear distance ($0.6 a_0 \leq R \leq 2.4 a_0$). With these values the approximate spectrum $[I_{\text{mol}}^{(0,0)}(E_n, \mathbf{K})]_{\text{refl}}$ has been obtained using Eq. (29). The electronic ground state has not been included in this procedure, since it does not show a dissociative behavior at the R range of interest. Thus, the reflection approximation cannot be applied. The fifth electronic state is excluded, since it has not been rovibrationally resolved in [26] due to the smallness of the transition probability connected to this state.

$[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$ has been smoothed out by applying first a binning and then a spline-interpolation procedure, as has been described in Sec. II D 3. The convergence of the spectrum has been investigated by applying two different grids for R and by using different bin sizes. Since in [26] the probability density is numerically given only for a small number of energies, also that spectrum has been spline interpolated. The energy shift E_{shift} = 1.75 eV [see Eq. (31)] has been obtained by a fit procedure using the spectrum calculated within the reflection approximation and the one obtained by a full treatment of the rovibrational degrees of freedom [26].

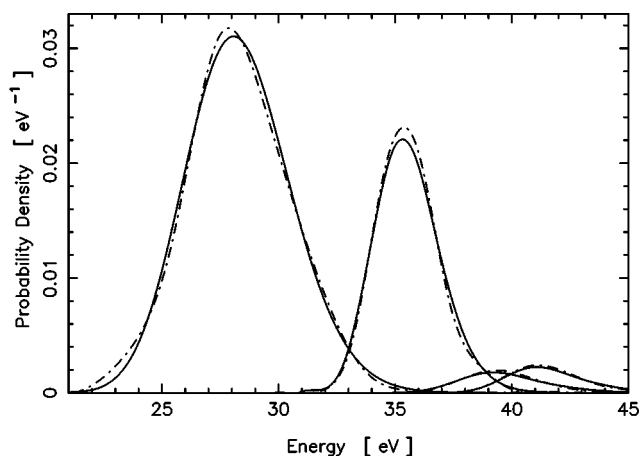


FIG. 1. Probability densities for the transitions to the second, third, fourth, and sixth $^1\Sigma$ electronic states of HeT^+ . Solid curves: rovibrationally resolved spectra as reported in [26]. Dash-dotted curves: transition probabilities within the reflection approximation (using an energy shift of 1.75 eV). (All curves are spline interpolated.)

In Fig. 1 the probability density $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$ (per eV) obtained by the reflection approximation (this work) is compared to the one that has been obtained in a full treatment of the rovibrational motion [26]. The very good agreement is obvious. Evidently, the applied energy shift E_{shift} is able to compensate most of the error that is introduced by the neglect of the recoil momentum in the zeroth-order transition amplitudes. The value that has been found for E_{shift} , i.e., 1.75 eV, is comparable to the one estimated in [12]. Note that this energy shift contains also a contribution that compensates the zero-point energy of the electronic ground state of HeT^+ missing in our calculation and automatically compensated for by E_{shift} .

Since the probabilities calculated within the reflection approximation are obtained from calculations performed at a number of different internuclear distances, the accuracy of our calculation with respect to the quality of the basis set may be estimated from the integrated probabilities $P_n^{(0,0)}$ obtained within the reflection approximation [see Eq. (32)]. In Table I these values (obtained for basis set 1) are compared with the corresponding ones given in [26]. Note that according to Eq. (32) the deviations are only due to the use of different basis sets. In this work only one set of nonlinear parameters for all states and values of R has been used, while in [26] the nonlinear parameters had been optimized for every state and every value of R separately. The good agreement that is nevertheless found demonstrates the flexibility of the basis set.

2. Low-energy range of the continuous excitation spectrum

In a previous work [10] we have reported a calculation of the transition probability in the energy range from the ionization threshold up to an excitation energy of HeT^+ of about 90 eV. In [10] the clamped-nuclei approximation has been used. Thus, $\bar{I}_{\text{mol}}^{(0,0)}(E, R_0)$ had been calculated for $R_0 = 1.4 a_0$. The energy range was chosen to display mainly the resonant part of the spectrum, since it was felt that this part had not

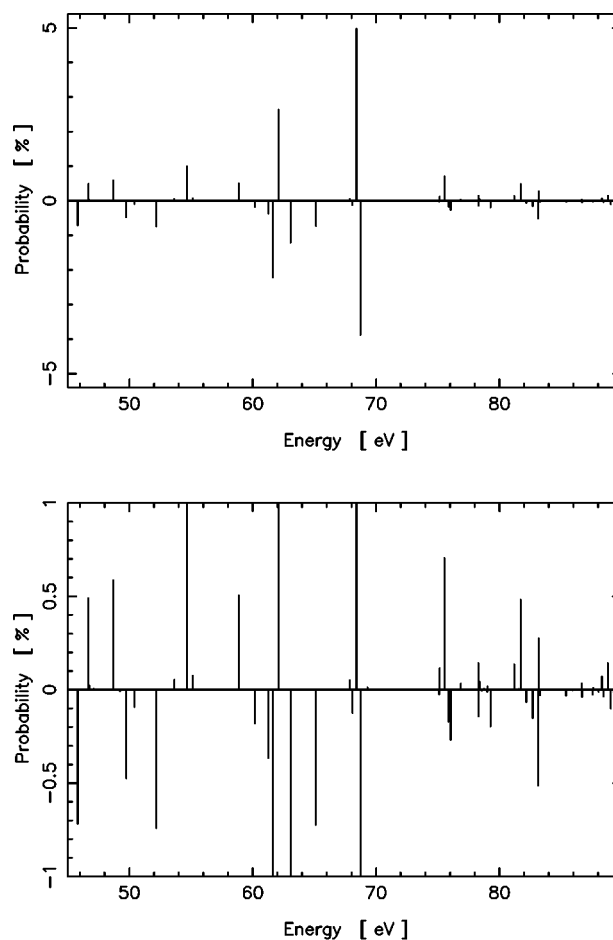


FIG. 2. Molecular final-state probabilities $P_n^{(0,0)}$ in the electronic continuum of HeT^+ calculated by a “simple” discretization approach with two basis sets (for $R = 1.40 a_0$). In order to distinguish the results for the two basis sets, $-P_n^{(0,0)}$ is plotted in the case of basis 2. The upper and lower plots differ only by the displayed range of probabilities.

been sufficiently described before. We will now improve on those previously reported results by investigating the effect of nuclear motion.

The transition probability to the electronic continuum of HeT^+ up to about 50 eV above the ionization threshold exhibits very large contributions from autoionizing states. In fact, two-thirds of the probability within this range are contained in such resonances [10]. Since the resonant contributions are so large in the molecular final-state spectrum of the β decay of T_2 , it is important to apply a method that is capable of describing the situation of a strong coupling between continuous and resonant parts. The complex-coordinate method [38–40] (see also [41–43] and references therein) has proved to be successful in handling such situations in the case of two-electron atoms [44,45]. In [10] this method had been successfully extended to diatomic molecules.

In Figs. 2 and 3 we compare the results obtained with the complex-coordinate method (for the two basis sets 1 and 2) to the ones that are obtained (with the same basis sets) with a “simple” discretization of the continuum as it has been performed in [9]. In the discretized spectra the probability connected to the continuum is distributed in a number of

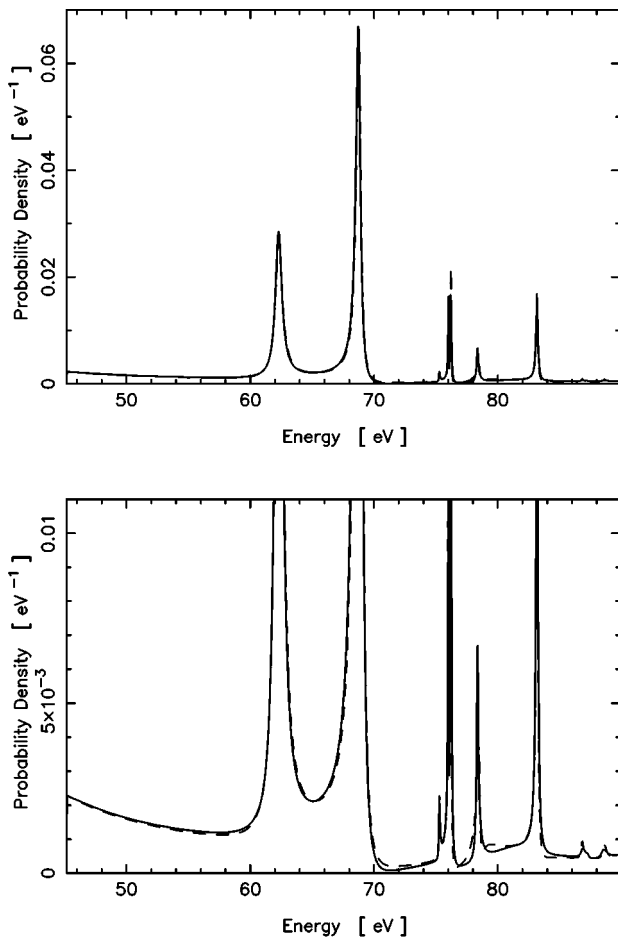


FIG. 3. Molecular probability density $\bar{I}_{\text{mol}}^{(0,0)}(E, R=1.40 a_0)$ calculated in this work with basis 1 (solid curve) and basis 2 (dashed curve). The upper and lower plots differ only by the displayed range of probabilities.

discrete lines. The total number of lines is identical with the total number of eigenvalues (in this case identical to the number of basis functions that are included in the calculation). The position and intensity of the lines are evidently basis-set dependent. Although the two lowest-lying resonances show up as dominant contributions even in the discretized spectrum, neither their position nor their intensity is in good agreement when calculated with two different basis sets. At $R=1.40 a_0$ the intensities are, e.g., 2.63% (basis 1) compared to 2.21% (basis 2) for the first resonance and 4.98% (basis 1) compared to 3.88% (basis 2) for the second resonance. The broadening of the resonances due to their finite lifetime as well as their interference with the nonresonant continuum is of course also absent in such a discretized spectrum.

The spectrum calculated within the complex-scaling method exhibits, on the other hand, a smooth background and (half-life) broadened resonances that show the interference effect with the background (*Fano* shape). It may be noted that this interference effect leads to a nearly vanishing intensity in the energy range just above the second resonance (see especially the lower plot in Fig. 3). The main advantage of the complex-coordinate method is, however, that the obtained spectrum is to a good approximation basis-set independent, provided the basis set is of sufficient quality. This is

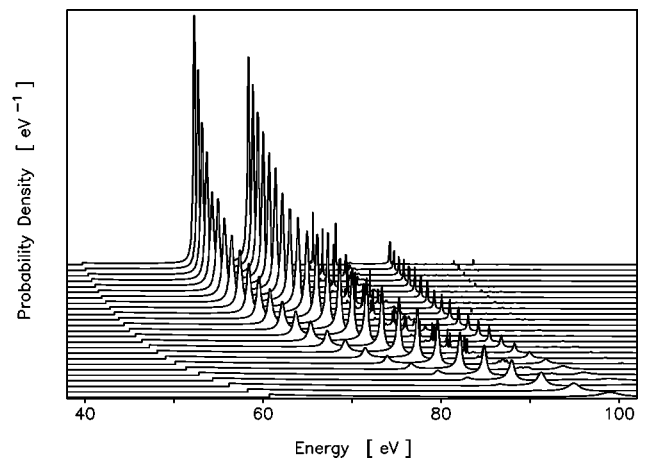


FIG. 4. Dependence of the molecular final-state distribution $\bar{I}_{\text{mol}}^{(0,0)}(E, R)$ on the internuclear distance R . Displayed are the spectra obtained for 25 values of R that are equidistantly distributed in the range $0.8 a_0 \leq R \leq 2.0 a_0$. Every spectrum is plotted as $\bar{I}_{\text{mol}}^{(0,0)}(E, R_j) + (j-1)I_{\text{off}}$ where $I_{\text{off}}=0.005 \text{ eV}^{-1}$ ($R_1=0.8 a_0$ and $R_{25}=2.0 a_0$).

evident from comparing the spectra obtained for the two basis sets employed in this work (see Fig. 3). Only minor deviations are found, which should be confronted with the differences in the discretized spectra (Fig. 2).

The enlargement of the basis set compared to the one adopted in a previous work [10] yielded an improvement in two respects. First, both sets of nonlinear parameters (basis 1 and 2) yield now very similar and stable results, while the agreement obtained for the smaller basis sets (when compared with each other) was not that good. This was the reason why in the older work different basis sets were used for different ranges of energy (cf. [10]). Second, the stability with respect to the complex-scaling angle is improved by the use of the larger basis sets. Nevertheless, it is still more accurate to obtain the final spectrum by stabilizing it in different energy ranges with respect to this scaling parameter, and this has also been done in this work.

In Fig. 4, the R dependence of $\bar{I}_{\text{mol}}^{(0,0)}(E, R)$ is displayed. The dominant effect of the variation of the internuclear distance is seen to be an overall shift of the probability distribution with respect to the energy. This illustrates the fact that (nearly) all electronic states of HeT^+ have a purely dissociative character in the considered range of internuclear distances. The steepness of the corresponding potential curves causes the energy shift relative to the ground state energy of HeT^+ . It is also apparent from Fig. 4 that not all potential curves corresponding to resonant states have the same slope, since some resonances merge together for small values of R .

A second effect of the variation of the internuclear distance is a change in the probabilities that are attributed to the electronic resonances. At large values of R the lowest-lying resonance is the most intense one, but its intensity decreases so quickly with decreasing value of R that at most values of the internuclear distance the second lowest-lying resonance acquires the largest intensity. Even though also this second resonance loses intensity with decreasing value of R , it remains the most intense one, before it fades also out.

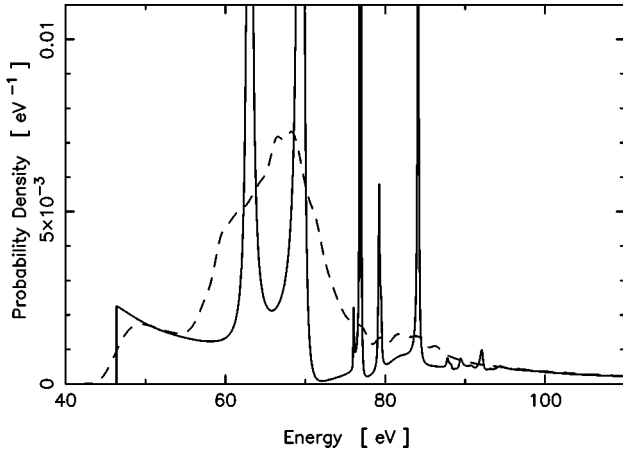


FIG. 5. Final result for the molecular probability density in the electronic continuum within the zeroth-order approximation, once calculated using the closure approximation at $R_{\text{eff}}=1.431 a_0$ (solid curve), once calculated using the reflection approximation (dashed curve). Both spectra have been shifted by $E_{\text{shift}}=1.75$ eV in order to consider the effect of the recoil (see text).

The spectrum $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$ is obtained from the R -dependent spectra, weighted with the vibrational wave function describing the rovibronic ground state of T_2 . In Fig. 5 $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$ is compared to $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{clos}}$ (calculated at R_{eff}). Both spectra have been calculated using E_{shift} in order to account for the recoil and the zero-point energy. The main effect of the nuclear motion is seen to be a considerable broadening of the resonant contributions due to the strong dependence of the resonant positions on the internuclear distance. A similar effect is the broadening of the spectrum at the ionization threshold which is due to the dissociative potential curve of the electronic ground state of HeT^{2+} . In this context it may be reminded that Fig. 5 contains solely the contribution from the electronic continuum, and its threshold is defined by the R -dependent potential curve of HeT^{2+} . The contributions of the bound states, i.e., of the Rydberg states adjacent (from below) to the continuum, are omitted in Fig. 5. This is the reason for the occurrence of a sharp threshold of the electronic continuum if the calculations are performed for a specific value of R .

3. High-energy range of the excitation spectrum

The calculations of the transition probability for the electronic continuum have so far only concentrated on the resonant part of the spectrum. Therefore, the maximum excitation energies have been 165 eV in [9] and approximately 90 eV in [46,10]. However, there are now two reasons to extend this energy range. First, the analysis of recent experimental data seems to suggest a “missing” component in the theoretical spectrum at excitation energies above 100 eV [7,8]. Second, it has been suggested that the fit procedure used for the extraction of the neutrino mass might be significantly affected by a wrong or omitted high-energy spectrum [12].

As is visible from Fig. 5, and even more transparent from Fig. 6, the effect of the nuclear motion on the transition probability fades out at molecular excitation energies of about 100 eV. Beyond that energy the differences between

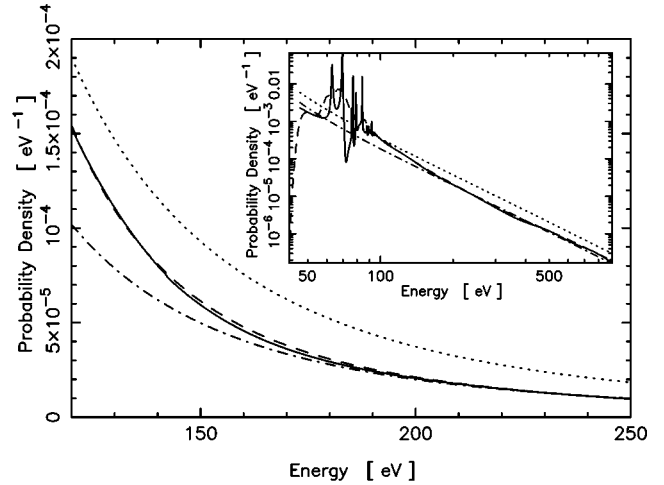


FIG. 6. High-energy part of the molecular probability density within the zeroth-order approximation. Solid curve: result of the closure approximation ($[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{clos}}$) calculated at R_{eff} . Dashed curve: result of the reflection approximation ($[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$). The remaining two curves are the atomic spectrum adapted to the molecular case, using the prefactor 3.73 as suggested in [12] (dotted curve) and using the prefactor 2.0 as suggested in this work (dash-dotted curve). In the inset the same spectra are displayed on a double-logarithmic scale covering the energy range from the ionization threshold to a molecular excitation energy of about 900 eV. (All four spectra are shifted by $E_{\text{shift}}=1.75$ eV.)

$[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$ and $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{clos}}$ can be attributed to numerical inaccuracies. While for $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{clos}}$ one spectrum was stabilized with respect to the complex-scaling angle, this stabilization procedure has been performed separately for 55 spectra (each corresponding to a given value of R) for obtaining $[I_{\text{mol}}^{(0,0)}(E, \mathbf{K})]_{\text{refl}}$. Therefore, it is to be expected that within the reflection approximation an possible “error” in the stabilization of one spectrum will be largely compensated by the other spectra.

In [12] it has been suggested to add an atomic tail to the molecular final-state spectrum given in [9], since that spectrum ends at about 165 eV. The atomic tail given in [47] was modified in two respects for the use in the molecular case.

First, the ionization energy occurring in the expression for the atomic tail has been changed to 45 eV. It may be noted that this value is not the real ionization energy of HeT^+ , but the value obtained for a vertical transition at $R \approx 1.4 a_0$. Since the approximation by a vertical transition seems to be well satisfied for the process under consideration (cf. the good agreement found for the bound-state transition probabilities calculated for R_{eff} compared to the ones that were calculated in a full treatment of the rovibrational motion), this value for the ionization energy seems to be a reasonable choice. Additionally, it may be noted that the ionization energy E_{ion} enters the atomic probability only in form of the energy difference $E - E_{\text{ion}}$. Therefore, the probability depends only weakly on E_{ion} , if $E \gg E_{\text{ion}}$. Since we are interested in the high-energy range of the spectrum, this condition should be well satisfied.

Second, an overall prefactor 3.73 has been introduced in the atomic tail. The choice of this prefactor was based on the argument that in this way the total probability will add up to

unity (within the sudden approximation), as it should do. However, such a choice assumes the probability distribution given in [9] to be exact. As has been discussed earlier, the discretization of the continuum as has been done in [9] leads to a quite arbitrary distribution of the probabilities in the electronic continuum (see Fig. 2). Assuming nevertheless the distribution to be correct to such an high accuracy, one has to note that the summed contribution (0.00138) reported in [9] for the energy range 165–200 eV would suggest a prefactor of about 3.2. Evidently, this is not consistent with the prefactor 3.73 suggested in [12].

It may be noted that the introduction of a comparatively arbitrary prefactor spoils the physical idea behind using an atomic tail, i.e., the idea that at very large excitation energies the most probable situation is the one where one of the atomic electrons is ejected with most of the energy that is released to the complete molecular system ($\text{HeT}^{2+} + e^-$). This fast escaping electron will effectively experience the remaining molecular ion (HeT^{2+}) as it would be a point charge (with charge +2). Therefore, the situation is comparable to the one where one electron escapes from a He^{2+} system, as occurs in the case of the β decay of atomic tritium. However, instead of one atomic electron that can be ejected in the case of a tritium atom, there are now two electrons in the case of T_2 . Thus, one should expect that the prefactor 2.0 should be used for the atomic tail, when applied to T_2 . In Fig. 6 we have plotted together with the explicitly calculated molecular continuous spectra also the ones predicted by an atomic tail. Excellent agreement is found between our calculation and the atomic tail with prefactor 2.0 for excitation energies above 200 eV, while the tail obtained with the prefactor suggested in [12] does not match the molecular continuum at any energy.

From the very good agreement of the molecular continuum and the appropriate atomic tail we may conclude that our method seems to be well capable of treating the continuous spectrum up to excitation energies of about 900 eV. In addition, beyond 200 eV the molecular final-state spectrum

can be very accurately approximated by the atomic tail with a prefactor of 2.0 (as suggested in this work).

B. First-order correction

1. Contribution of the decaying nucleus

Analyzing the structure of the interference term $I^{(0,1)}(E, E_\beta)$ it can be expected that its largest contribution arises from the decaying nucleus, since it is the only purely real term of order η [see Eqs. (40) and (41)]. However, this term is only present if $Z_A^{\text{eff}} \neq 0$. This is understandable when it is remembered that the value of Z_A^{eff} depends on the partitioning of the Hamiltonian [Eq. (5)]. The choice $Z_A^{\text{eff}}=0$ means that the complete Coulombic interaction between the decaying nucleus and the β particle is already contained in the zeroth-order treatment, while the choice $Z_A^{\text{eff}}=2$ means that no Coulombic interaction between these two particles is included in the zeroth-order treatment. The inclusion of this interaction in the zeroth-order treatment leads, however, to the appearance of the Fermi function in the zeroth-order transition probability which is not present otherwise. Briefly, one has

$$\mathcal{I}_{Z_A^{\text{eff}}}^{(0,0)}(E, E_\beta) = F(Z_A - Z_A^{\text{eff}}, p_\beta) I^{(0,0)}(E, E_\beta), \quad (59)$$

where $\mathcal{I}_{Z_A^{\text{eff}}}^{(0,0)}(E, E_\beta)$ is the zeroth-order probability obtained for a specific partitioning of the Hamiltonian, while $I^{(0,0)}(E, E_\beta)$ is the zeroth-order transition probability that has been derived in this work, i.e., the one for $Z_A^{\text{eff}}=2$.

We will now investigate the effect of the different (artificial) partitionings on the physical quantity $\tilde{I}(E, E_\beta)$ that enters the β spectrum [see Eq. (7)], and was defined in Eq. (10). Truncating the Born series after the interference term and including only the largest (real) contribution with respect to the Sommerfeld parameter η one finds [see Eqs. (40) and (45)]

$$\tilde{I}(E, E_\beta) \approx \frac{\mathcal{I}_{Z_A^{\text{eff}}}^{(0,0)}(E, E_\beta) + \mathcal{I}_{Z_A^{\text{eff}}}^{(0,1)}(E, E_\beta)}{F(Z_A, p_\beta)} \quad (60)$$

$$\approx \frac{\mathcal{I}_{Z_A^{\text{eff}}}^{(0,0)}(E, E_\beta) - (8Z_A^{\text{eff}}/\pi)[\eta + 1/(3\eta c^2)] \mathcal{I}_{Z_A^{\text{eff}}}^{(0,0)}(E, E_\beta)}{F(Z_A, p_\beta)} \quad (61)$$

$$\approx \mathcal{I}_{Z_A^{\text{eff}}}^{(0,0)}(E, E_\beta) \frac{1 - (8Z_A^{\text{eff}}/\pi)[\eta + 1/(3\eta c^2)]}{F(Z_A, p_\beta)} \quad (62)$$

$$\approx I^{(0,0)}(E, E_\beta) \left(1 - \frac{8Z_A^{\text{eff}}}{\pi} \left[\eta + \frac{1}{3\eta c^2} \right] \right) \frac{F(Z_A - Z_A^{\text{eff}}, p_\beta)}{F(Z_A, p_\beta)}. \quad (63)$$

TABLE II. Interference-term corrections ($P_n^{(0,1)}$) to the zeroth-order transition probabilities ($P_n^{(0,0)}$) of the six electronic lowest-lying bound states of HeT^+ with $^1\Sigma$ symmetry. All probabilities are calculated with basis 1 at $R_{\text{eff}}=1.431 a_0$. In addition, $\eta=\text{const}=-0.027\ 068$ and $Z_A^{\text{eff}}=2$ have been used.

n	$P_n^{(0,0)}$	$[P_n^{(0,1)}]_r$	$[P_n^{(0,1)}]_{1/r}$	$P_n^{(0,0)} + P_n^{(0,1)}$	$[P_n^{(0,0)} + P_n^{(0,1)}]_{\text{norm}}$
1	0.57430	3.55×10^{-4}	-7.56×10^{-4}	0.57390	0.57437
2	0.17418	-4.38×10^{-5}	-1.04×10^{-4}	0.17403	0.17417
3	0.07752	-6.05×10^{-5}	-3.18×10^{-5}	0.07743	0.07749
4	0.00790	-2.33×10^{-6}	-2.40×10^{-6}	0.00789	0.00790
5	0.00012	1.36×10^{-7}	-3.34×10^{-8}	0.00012	0.00012
6	0.00914	-9.66×10^{-6}	-2.33×10^{-6}	0.00913	0.00914

Using $\eta = -0.027$ (as it is found close to the end point of the β spectrum),

$$\tilde{I}(E, E_\beta) \approx I^{(0,0)}(E, E_\beta) \times \begin{cases} 1.000 & \text{for } Z_A^{\text{eff}}=0, \\ 0.987 & \text{for } Z_A^{\text{eff}}=1, \\ 0.967 & \text{for } Z_A^{\text{eff}}=2, \end{cases} \quad (64)$$

is obtained. Apparently, already the interference term is capable of recovering most of the effect of the interaction of the β electron with the decaying nucleus, i.e., 98.7% for $Z_A^{\text{eff}}=1$ and 96.7% for $Z_A^{\text{eff}}=2$. This result justifies the application of perturbation theory to this problem, since it demonstrates that the (large) effect of the interaction with the decaying nucleus (which is here treated equivalently with the effect of the atomic electrons and the spectator nucleus) is well taken into account in such a treatment. Additionally, it may be noted that the need for an inclusion of this effect in the evaluation of β -decay rates was recognized very early, and it is confirmed experimentally. On the other hand, if the contribution of the atomic electrons (and the spectator nucleus) were of a comparable size (i.e., of order η), this should be recognizable from experimental data. This is a strong indication that the theoretical prediction stating that the effects of the atomic electrons on the decay rate are not of the order of η , but of the order η^2 , is very reasonable, and it reflects the well-known fact that the interaction with the decaying nucleus has a much larger influence on the decay rate than any other Coulombic interaction of the β electron.

2. Contribution of the atomic electrons and the spectator nucleus

In Table II the corrections $P_n^{(0,1)}$ to the zeroth-order transition probabilities $P_n^{(0,0)}$ are given for the six lowest-lying $^1\Sigma$ states of HeT^+ . The calculation has been performed at R_{eff} and with $\eta = -0.027\ 068$. The contribution has been split into a part arising from the r matrix elements [see Eq. (42)] and the $1/r$ matrix elements [see Eqs. (40) and (42)]. Obviously, both contributions are nonproportional to the zeroth-order probability when discussed separately. However, if they are added to each other, it surprisingly turns out that they are nearly proportional to $P_n^{(0,0)}$. This can be seen from the last two columns of Table II, since after normalization of $P_n^{(0,0)} + P_n^{(0,1)}$ to the zeroth-order probability $P_n^{(0,0)}$ most of the effect of $P_n^{(0,1)}$ disappears and the normalized probabilities are nearly identical to $P_n^{(0,0)}$. (Note that this normalization has been performed using the complete spec-

trum, including the electronic continuum. Therefore, the sums of the probabilities of the first and last columns of Table II may differ.)

Inspection of the data shows that in the *molecular* case the difference which remains (even after the normalization) is of the order $0.1\eta^2$, while in the *atomic* case the normalized quantity $[P_n^{(0,0)} + P_n^{(0,1)}]_{\text{norm}}$ is *identical* to $P_n^{(0,0)}$. The latter relation has been shown analytically in [16], but was not found in [13], since in the latter work the contribution of $[P_n^{(0,1)}]_r$ had been neglected.

It should be emphasized that the normalization of $I^{(0,0)}(E, E_\beta) + I^{(0,1)}(E, E_\beta)$ to $I^{(0,0)}(E, E_\beta)$ only makes sense if a relative measurement of the β spectrum is performed, as is (at least up to now) the case in tritium neutrino-mass experiments. However, an absolute measurement would have to be compared to the un-normalized spectrum.

In Fig. 7 the corrections for the electronic continuum are shown in the form of a ratio relative to the zeroth-order probability $I^{(0,0)}(E, E_\beta)$. We have only given the separate contributions arising from the r and the $1/r$ terms, since the $\ln r$ contribution turned out to be negligibly small (or even zero). As in the case of the bound states the contributions of the r and $1/r$ matrix elements are quite different, but added together they are nearly proportional to the zeroth-order probability. This is especially apparent from Fig. 8 where (on a

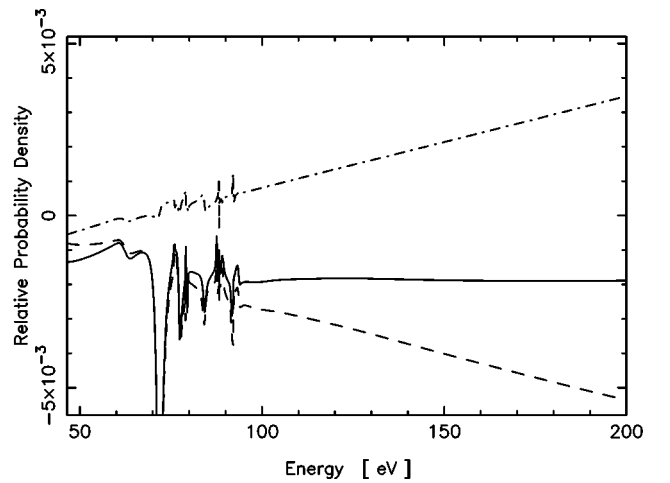


FIG. 7. Ratio of the first-order correction to the zeroth-order probability. In particular, $I_{\text{el}}^{(0,1)}(E, E_\beta)/I^{(0,0)}(E, E_\beta)$ (solid curve), $I_r^{(0,1)}(E, E_\beta)/I^{(0,0)}(E, E_\beta)$ (dashed curve), and $I_{1/r}^{(0,1)}(E, E_\beta)/I^{(0,0)}(E, E_\beta)$ (dash-dotted curve). (All spectra are shifted by $E_{\text{shift}}=1.75$ eV.)

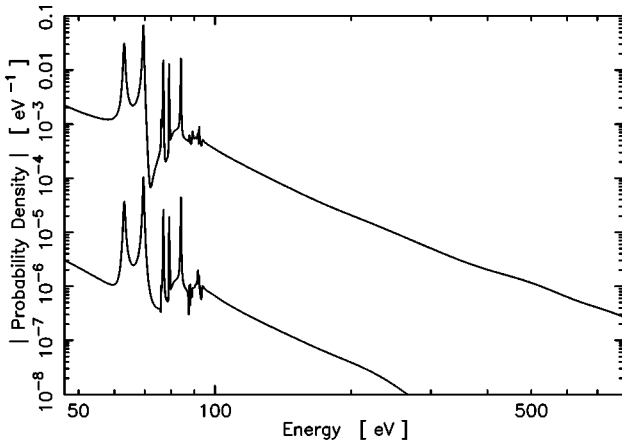


FIG. 8. Comparison of the (electronic) first-order correction $I_{el}^{(0,1)}(E, E_\beta)$ (lower curve) with the zeroth-order probability $I^{(0,0)}(E, E_\beta)$ (upper curve). (Both spectra are shifted by $E_{\text{shift}} = 1.75$ eV.)

double-logarithmic scale) the complete interference term is compared to the zeroth-order probability. Both spectra are seen to be nearly parallel to each other.

In order to investigate the degree of proportionality of $P_n^{(0,1)}$ with respect to the zeroth-order probability, Table III contains the ratio $P_n^{(0,1)}/P_n^{(0,0)}$ (in %). In the *atomic* case this ratio is identical for all electronic bound or continuous states and it is equal to $100\%(-\eta^2) \approx -0.073\ 27\%$. The difference between the atomic and molecular cases is therefore evident by comparing the entries in one *row* (i.e., the one for $R = R_{\text{eff}} = 1.431 a_0$) with that value. The ratios found in the molecular case for the different bound states are all of the same order of magnitude and quite similar to the atomic value, but not identical. The similarity (same order of magnitude and same sign) found for the different bound states indicates that the main effect of the interference term is a change of the magnitude of the transition probability, but not of its shape, as was already found by the renormalization. The only state for which a correction with an opposite sign is found is the fifth state. However, since the transition probability connected to this state is very small, this has almost no influence on the overall result.

In Table III we have also investigated the dependence of the interference-term correction on the internuclear distance in order to assure that the results found for R_{eff} can be expected to be generally valid. We report the results of calculations performed for a number of different values of R within the clamped-nuclei approximation. It may be noted that in this case the $1/R$ matrix element $\langle \xi_0 | 1/R | \xi_0 \rangle$ reduces to $1/R$, while it has been R^{-1} before [see Eq. (45)]. For most of the bound states the ratio $P_n^{(0,1)}/P_n^{(0,0)}$ turns out to be comparatively constant. However, the corrections for states 4 and 5 depend quite evidently on the internuclear distance. Still, these corrections are small, especially since the zeroth-order probabilities connected to these states are small.

For some values of R the ratios $P_n^{(0,1)}/P_n^{(0,0)}$ obtained with basis set 2 are also reported in Table III. A comparison to the corresponding results obtained with basis set 1 reveals good agreement. Therefore, it can be concluded that the results given in Tables II and III are almost basis-set indepen-

dent. The accuracy of the calculation may be estimated from the differences obtained for the two basis sets.

V. DISCUSSION

In this work the reliability of the molecular final-state distribution following the β decay of molecular tritium has been investigated in the context of its application in tritium neutrino-mass experiments.

The transition probability into the electronic continuum of HeT^+ has been reinvestigated within the zeroth order of perturbation theory. The reliability of a previous calculation [10] within the clamped-nuclei framework has been confirmed by the use of larger basis sets. The effect of the nuclear motion on the transition probability has been included for the first time for the electronic continuum. Two different approximations have been applied which were called the closure and reflection approximations, respectively. Most of the effect of the nuclear recoil which is not explicitly contained in our calculation was recovered by an effective energy shift $E_{\text{shift}} = 1.75$ eV which was obtained with the aid of a previous calculation [26] for the electronic bound states of HeT^+ that included a complete treatment of the rovibrational structure of the initial state and the final states. In addition, the introduction of E_{shift} leads to a common energy scale for the present calculation of the electronic continuum and the previous bound-state calculation in [9].

In a similar way it was possible to use the fully rovibrationally resolved spectrum for five electronic bound states given in [26] in order to obtain an effective internuclear distance R , i.e., $R_{\text{eff}} = 1.431 a_0$. This R_{eff} allows us to calculate the individual electronic transition probabilities to a very high accuracy by performing the calculation only at one single value of R . Since the transition-probability spectrum of the electronic continuum turned out to be very sensitive to the choice of the internuclear distance R (as had been found earlier also for the bound states), the change of the internuclear distance from $R_0 = 1.40 a_0$ (clamped-nuclei calculation [10]) to $R_{\text{eff}} = 1.431 a_0$ results in an obvious improvement of the calculation.

However, the application of the closure approximation leads to a loss of the energy distribution of the transition probabilities connected to a given electronic state (within the Born-Oppenheimer approximation), since in the closure approximation the whole probability of a specific electronic state is put at one certain energy value. Since the energy distribution is needed for the analysis of the tritium neutrino-mass experiments, it was felt important to improve on this approximation, and this has been done by the reflection approximation. This approximation has been chosen since the states in the electronic continuum (especially the resonant states whose probabilities exhibit the most evident dependence on the internuclear distance) are all purely dissociative within that range of R where the rovibrational ground-state wave function of T_2 is different from zero. As expected, the reflection approximation introduces a considerable broadening of the resonances. In addition, also the ionization threshold is broadened.

Beyond a molecular excitation energy of about 100 eV the differences between the closure and reflection approximations become negligible. This indicates that at large mo-

TABLE III. Dependence of the ratio of the interference term $P_n^{(0,1)}$ to the zeroth-order probability $P_n^{(0,0)}$ on the internuclear distance R . Displayed are the corresponding values (in %) for the the six electronic lowest-lying $^1\Sigma$ states of HeT^+ . All probabilities are calculated within the clamped-nuclei approximation (with $\eta = \text{const} = -0.027\ 068$ and $Z_A^{\text{eff}} = 2$). Rows denoted by an asterisk contain the results obtained for basis set 2, while all other values correspond to basis set 1.

R/a_0	$100\% \times P_n^{(0,1)}(R)/P_n^{(0,0)}(R)$					
	$n=1$	$n=2$	$n=3$	$n=4$	$n=5$	$n=6$
0.800	-0.06685	-0.09099	-0.10536	-0.08670	-0.00257	-0.11410
0.800*	-0.06635	-0.09166	-0.10579	-0.09845	0.32251	-0.13664
0.900	-0.06919	-0.09200	-0.11029	-0.08562	0.02896	-0.11950
1.000	-0.07042	-0.09161	-0.11344	-0.08263	0.04246	-0.12373
1.000*	-0.07050	-0.09164	-0.11406	-0.08695	0.04803	-0.12528
1.100	-0.07091	-0.09051	-0.11579	-0.07851	0.04983	-0.12685
1.200	-0.07093	-0.08906	-0.11752	-0.07359	0.05906	-0.12868
1.300	-0.07078	-0.08735	-0.11852	-0.06819	0.06945	-0.13037
1.400	-0.07045	-0.08572	-0.11936	-0.06243	0.08436	-0.13094
1.431	-0.07031	-0.08518	-0.11957	-0.06038	0.08723	-0.13169
1.431*	-0.06992	-0.08553	-0.11984	-0.06042	0.09119	-0.13300
1.500	-0.07001	-0.08408	-0.12010	-0.05590	0.10103	-0.13194
1.600	-0.06958	-0.08247	-0.12052	-0.04865	0.12296	-0.13352
1.700	-0.06917	-0.08112	-0.12115	-0.04127	0.15737	-0.13381
1.800	-0.06880	-0.07990	-0.12164	-0.03314	0.20033	-0.13457
1.900	-0.06852	-0.07885	-0.12215	-0.02410	0.26540	-0.13572
2.000	-0.06836	-0.07803	-0.12262	-0.01434	0.36688	-0.13699
2.000*	-0.06825	-0.07806	-0.12242	-0.01489	0.39144	-0.13641

lecular excitation energies the effects of the nuclear motion on the final-state distribution become less important. Based on this idea an atomic tail was suggested that should be able to describe the high-energy range of the spectrum to a sufficiently high accuracy. All parameters were fixed by physical considerations, while in a previously suggested tail [12] an overall prefactor was chosen by a normalization constraint. Whereas our *ab initio* molecular calculations reveal a very good agreement with the atomic tail suggested in this work for excitation energies above 200 eV, this is not the case for the previously suggested tail.

Another aspect of the reliability of the molecular final-state spectrum considered in this work is the question of whether a zeroth-order treatment is sufficient for the analysis of the tritium neutrino-mass experiments. This problem had so far never been investigated for molecular tritium, but mainly for tritium atoms. In addition, it may be noted that the different works performed for the atomic case are not in good agreement with each other. Therefore it was felt necessary to investigate this point by an explicit calculation. The main focus of our investigation was on the question of whether the interference term between the zeroth-order and first-order transition amplitudes is of order η or η^2 (where η is the Sommerfeld parameter) and whether it is proportional to the zeroth-order probability, as found for the atomic case [16]. Adopting a variable partitioning of the Hamiltonian, it was possible to show that the interference term is in fact (also for the atomic decay) of order η , as one would expect from perturbation theory. However, this contribution arises only from the decaying nucleus, recovering almost completely the Fermi function that is often already introduced in the zeroth-order treatment. Thus the partitioning adopted in

this work provides a more natural explanation of the magnitude of the interference term. In addition, it demonstrates the applicability of a perturbation-theory approach to the process under consideration, since the effect of the interaction of the β electron with the decaying nucleus (which is known to be a large effect) is recovered to about 97% with the aid of the lowest-order correction in perturbation theory (the interference term between the zeroth-order and first-order amplitudes). Therefore, it seems reasonable to assume that the (smaller) effect of the Coulombic interaction between the β electron and the remaining particles (molecular electrons and spectator nucleus) is also sufficiently well described by perturbation theory.

In contrast to the atomic case it turns out that for T_2 the interference-term correction is not strictly proportional to the zeroth-order probability, but it is almost proportional. Therefore, the shape of the β spectrum is nearly unaffected by the addition of the interference term to the zeroth-order probability, as is obvious from the renormalized results. While the magnitude of the electronic and spectator-nucleus contributions to the interference term is found to be of order η^2 for the absolute (nonrenormalized) spectrum, it is only of order $0.1\eta^2$ for the relative (renormalized) spectrum. This has been demonstrated by an explicit calculation of the interference term for the whole molecular final-state spectrum including the electronic continuum where the latter has been treated in a nondiscretized way. This calculation required an extension of the complex-coordinate method in order to evaluate interference terms between transition amplitudes, which has been implemented for the first time in this work.

The dependence of the calculated correction on the basis set and the internuclear distance has also been investigated. It

was found that the result is almost independent of both parameters, even though, e.g., two of the six electronic bound states are quite dependent on the internuclear distance, but the probability connected to these states is very small.

Since the interference term between the zeroth-order and first-order amplitudes is found to be of order η^2 also for molecular tritium (*apart* from the contribution of the *decaying* nucleus), it would be incorrect to consider only this correction, because also the pure first-order term as well as the interference term between the zeroth-order and *second-order* amplitudes is of order η^2 (see I). It has been argued that the contribution of the pure first-order probability will be (partly) canceled by the interference term between the zeroth-order and second-order amplitudes. An explicit calculation [48] for the electronic bound states of HeT^+ revealed, however, that this cancellation is not complete if transitions to *specific* final states are considered. More importantly, the remnant (after adding both contributions) is still of order η^2 and is not purely proportional to the zeroth-order probability. Therefore this term has to be taken into account if corrections of order η^2 are considered.

Additionally, it has to be noted that the partial cancellation mentioned above occurs only for the $L=0$ contribution to the pure first-order transition probability. According to the findings in I there are, however, also terms of order η^2 arising from the $L \neq 0$ contributions. In fact, these contributions will in the case of a tritium molecule add up to $N_e \eta^2 = 2 \eta^2$ (see I). In the case of a T_2 molecule decaying in its ground state with Σ symmetry all pure first-order terms corresponding to $M=0$ will lead to a transition to a HeT^+ state with Σ symmetry, whereas $M \neq 0$ will lead to a population of HeT^+ states that have no Σ symmetry. Therefore two-thirds of the total contribution of $L=1$ will result in transitions to HeT^+ states with Π symmetry (i.e., the probability of Π state population is equal to two-thirds of $0.75 \times 2 \eta^2$ and thus equal to η^2). In the same way one finds that the probability of exciting states of Δ symmetry is $0.2224 \eta^2$ [$= (4/5) \times 0.139 \times 2 \eta^2$], etc. Since within the zeroth-order treatment to a very good approximation only Σ states are populated, the correction due to the $L \neq 0$ contributions to the pure first-order term cannot be proportional to the zeroth-order probability. (As discussed in Sec. II D 1 the probability of a direct excitation of Π states within the sudden approximation is, on the other hand, only of the order 4.5×10^{-5} and thus one order of magnitude smaller than η^2 .) In conclusion, also the contributions from $L \neq 0$ to the pure first-order probability will necessarily change the shape of the β spectrum, and not only its absolute value.

Evidently, a consideration of corrections of order η^2 has to take into account the interference terms between the zeroth-order and first-order amplitudes, the pure first-order term (including the contributions from $L \neq 0$), and the interference term between the zeroth-order and second-order terms. An explicit calculation of these terms will be presented elsewhere [48].

VI. CONCLUSION

Summarizing the results the following main conclusions can be drawn from this work. First, since in the zeroth-order treatment the molecular final-state spectrum for the elec-

tronic continuum has been improved compared to previous calculations, e.g., compared to the one given in [9], this new spectrum should be used in the analysis of experimental data. Since the extraction of the (squared) neutrino mass out of the experimental data is done by a fit procedure that depends necessarily on a number of experimental parameters (resolution, scattering in the source, background, etc.), it is not possible to predict the change in the obtained neutrino mass that will arise from the use of the new continuous final-state spectrum compared with the old one. Most probably, the result will differ for different experiments. Nevertheless, the shift of the continuous spectrum due to the recoil may have some influence, since it corresponds to a shift of a probability of 14% by more than 1 eV. However, it seems to be unlikely that this effect is large enough to solve the experimental problems, where an additional component of few percent at an excitation energy of more than 100 eV has been suggested.

Second, as in the case of the electronic bound states, also the transition probability into the continuum is found to be strongly dependent on the internuclear distance. Therefore it may be useful to investigate more carefully if the assumptions employed in the theoretical investigations are really met by the experiments, since every deviation of the average internuclear distance (due to different isotopes, the formation of ions, solid-state effects, electric or magnetic fields, etc.) could significantly change the final-state distribution of HeT^+ .

Third, the largest corrections to the zeroth-order approximation are of order η^2 if the interaction with the decaying nucleus is already included in the zeroth-order treatment, as is usually done. The largest corrections (arising as expected from the interference term between the zeroth-order and first-order amplitudes) have been explicitly calculated in this work, and have been shown to be too small to explain the experimental difficulties to extract the neutrino mass from the β -decay spectrum of T_2 .

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APPENDIX: IMPORTANCE OF HIGHER-ORDER TERMS PROPORTIONAL TO THE SUDDEN AMPLITUDE

In this appendix the impact of higher-order terms that are proportional to the sudden amplitude (like the nuclear contribution) on the *shape* of the transition-probability spectrum is discussed. For simplicity we assume a discrete spectrum; the generalization to a discrete plus continuous spectrum is straightforward. The spectrum in the sudden approximation is given by the probabilities P_j^0 . The interference term between the zeroth-order and first-order transition probabilities is given by $P_j^1 = f_j + c P_j^0$ where f_j is the electronic contribution and $c P_j^0$ is the nuclear one. The absolute value of the

interference-term-corrected probability is therefore

$$P_j = P_j^0 + P_j^1 = (1+c)P_j^0 + f_j. \quad (\text{A1})$$

The renormalization of this term to the sudden probability yields

$$\begin{aligned} \tilde{P}_j &= \frac{\sum_j P_j^0}{(1+c)\sum_j P_j^0 + \sum_j f_j} [(1+c)P_j^0 + f_j] \\ &= \frac{\sum_j P_j^0}{\sum_j P_j^0 + \sum_j \tilde{f}_j} [P_j^0 + \tilde{f}_j], \end{aligned} \quad (\text{A2})$$

where $\tilde{f}_j = f_j(1+c)^{-1}$. Since $c < 1$ (which has to be valid if the perturbational approach should work at all), the expression $(1+c)^{-1}$ can be expanded in a Taylor series. This gives

$$\tilde{f}_j = f_j(1 - c + c^2 - \dots) = f_j - cf_j + c^2 f_j \dots \quad (\text{A3})$$

From this equation it is evident that the effect of the nuclear contribution on the shape of the transition-probability spectrum is by a factor η^2 smaller than the electronic contribution, independent of the size of the latter one, since c is of order η^2 .

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