The beta decay of tritium in the form of molecular T\(_2\) is the basis of sensitive experiments to measure neutrino mass. The final-state electronic, vibrational, and rotational excitations modify the beta spectrum significantly, and are obtained from theory. We report measurements of the branching ratios to specific ionization states for the isotopolog HT. Two earlier, concordant measurements gave branching ratios of HT to the bound HHe\(^+\) ion of 89.5\% and 93.2\%, in sharp disagreement with the theoretical prediction of 55-57\%, raising concerns about the theory’s reliability in neutrino mass experiments. Our result, 56.5(6)\%, is compatible with the theoretical expectation and disagrees strongly with the previous measurements.

The discovery of neutrino oscillations and mass \([1, 2]\) signals a contradiction to a prediction of the minimal standard model, and opens a window to the physics that lies beyond. Oscillations link the squares of the masses via their differences, but do not give values for the masses themselves. The recent KATRIN result from tritium beta decay gives an upper limit of 1.1 eV on each mass \([3]\). An intensive effort continues to determine the masses experimentally \([4]\). A laboratory measurement will constrain themselves. The recent KATRIN result from tritium beta decay process and help disentangle correlated parameters in cosmological models \([5]\).

The most sensitive direct method for probing the neutrino mass is by examining the beta spectrum of tritium near the end point. The shape of the beta spectrum there is affected by molecular excitations, which must be calculated with great precision in order to be confident in the additional contribution of a non-zero neutrino mass \([6]\). While these ‘final-state’ calculations can in principle be taken to arbitrary accuracy since the force is known, in practice approximations are necessary. Indeed, the first approximations are necessary. Indeed, the first taken to arbitrary accuracy since the force is known, in principle.

While these ‘final-state’ calculations can in principle be taken to arbitrary accuracy since the force is known, in practice approximations are necessary. Indeed, the first taken to arbitrary accuracy since the force is known, in principle.

One puzzling discrepancy remains. In the 1950s, two experimental studies of the molecular ions made in the beta decay of HT and T\(_2\) were carried out using mass spectrometers \([11, 12]\), and both indicated that 90 to 95\% of decays lead to the bound molecular ion HHe\(^+\) or THe\(^+\).

Theory, however, predicts the fraction to be at most 57\% (Table I). The electronic ground state of HHe\(^+\) and THe\(^+\) is bound by 2 eV, but lepton recoil can excite rotational-vibrational states that are energetically unbound. Most of the latter states are hindered from dissociation by their angular momentum, and thus quasibound.

The theory might indeed be in error, one of many hypotheses advanced \([6]\) to explain the disagreement. Strictly, the theory applies to the endpoint whereas the experiments integrate over the entire beta spectrum. Unidentified systematic errors may have affected the measurements. However, none of these explanations seems likely to accommodate such a large disagreement.

We present new measurements of the branching ratios for HT and, in a future paper, T\(_2\), with a novel time-of-flight mass spectrometer, TRIMS (Tritium Recoil Ion Mass Spectrometer). The heteronuclear parent HT allows for clear separation of various final states, and both

---

**TABLE I. Branching ratio to the bound molecular ion for HT and T\(_2\).**

| Molecule | Snell et al. (Ref. [11]) | Wexler (Ref. [12]) | Theory (Ref. [9, 10]) |
|----------|-------------------------|-------------------|------------------------|
| Quasibound | Bound |
| HT       | 0.932(19)              | 0.895(11)        | 0.02                    |
| T\(_2\)  | –                      | 0.945(6)         | 0.18                    |

The theory might indeed be in error, one of many hypotheses advanced \([6]\) to explain the disagreement. Strictly, the theory applies to the endpoint whereas the experiments integrate over the entire beta spectrum. Unidentified systematic errors may have affected the measurements. However, none of these explanations seems likely to accommodate such a large disagreement.
of the early experiments studied HT. Only a small fraction of states are quasi-bound, allowing for a more decisive test of the theory. The theoretical approach is the same for both HT and T2.

The beta decay of tritium in the form of HT gas produces the positively charged ions H+, He+, He++, and HHe+. The negative ion H− can also be produced but is expected to be rare. When T2 is also present as a contaminant, T+ and THe+ are produced. Electrons produced include the beta itself and 0, 1, or 2 shakeoff electrons.

In TRIMS, ions and electrons move under the influence of collinear, uniform magnetic and electric fields toward silicon detectors located at either end of a 20-cm-long chamber (Fig. 1). The start time is set by the arrival of an electron in the ‘beta’ detector at the anode end, and the position in the chamber where the decay occurred is determined by the energy $K_{\text{ion}}$ acquired by a positive ion on route to the ‘ion’ detector at the cathode end. The ion mass-to-charge-squared ratio is then given by

$$\frac{m}{q^2} = t^2 \frac{E^2}{2K_{\text{ion}}},$$

(1)

where $t$ is the ion’s flight time and $E$ is the uniform electric field in the chamber. Not shown, but included in the analysis, is a correction for a drift space in front of the ion detector.

The apparatus [14] at the University of Washington consists of a metal-sealed ultra-high-vacuum system with an ion pump, two SAES getter pumps with ST-101 alloy, a liquid nitrogen cold trap, and a turbomolecular pump. After baking, it reaches a base pressure of $\sim 10^{-9}$ mbar.

The decay chamber (Fig. 1) is a National Electrostatics acceleration column, made of alternating Kovar alloy and alumina rings brazed together. Shaped aluminum rings mounted to the Kovar rings and biased by a chain of 76 1-GΩ 1% resistors establish a uniform voltage gradient of 281 V/mm. Four external magnet coils produce a 0.236-T magnetic field uniform to ±0.5% throughout the decay chamber. COMSOL calculations [15] showed that the field was not significantly affected by the Kovar rings.

At the anode end is a stainless-steel disk, gold-plated to reduce emission of secondary ions, with a circular opening for the beta detector to look through. At the cathode end is an 85%-transmitting stainless-steel mesh mounted under tension on a metal ring. It is electrically isolated and held 100 V below ground to suppress secondary electrons from the ion detector, located 29 mm behind the mesh.

With the chamber isolated, commercially supplied gaseous tritium is introduced through a leak valve to a partial pressure of $\sim 3 \times 10^{-8}$ mbar in normal operation. The total pressure, mostly H2, is monitored via a spinning-rotor gauge to avoid pumping of the gas by ionization gauges, and rises $\sim 10^{-5}$ mbar/hr.

Silicon detectors with 50-mm area and 0.5-mm thickness (Canberra PD50-11-500AM), are mounted on movable re-entrant arms containing custom, low-noise, miniature preamplifiers mounted directly on the feedthrough pin for minimal connection capacitance, and cooled to $\sim 15^\circ$C with CO2 Joule-Thomson coolers. Calibrated with 241Am gammas, the beta and ion detector energy resolutions were 2.46 and 2.05 keV FWHM, respectively. By means of two translation stages, the ion detector can be moved up to 25 mm off-axis in any direction to measure radial distributions.

The detector signals, transmitted through fiber optics, are digitized with a 12-bit 250-MHz digitizer that is triggered when either signal is above a discriminator threshold. The digitizer outputs are read out with the ORCA data acquisition software [16] and translated into ROOT [17] format. To obtain the signal amplitudes, two consecutive trapezoidal filters are applied on each waveform [18, 19]. Slow-control data from devices are sent to a PostgreSQL database on a remote server.

A Monte Carlo simulation using the GEANT4 toolkit [20, 21] was developed to guide design and study the influence of various systematics. The simulated energy and timing at the detectors are smeared by the measured detector resolutions. Also incorporated in the simulation are electron backscattering and the deposited ion energy predicted by SRIM [22].

Energy lost to the dead layer and to nuclear stopping in the active layer depends on the ion type and energy, which are not known on an event-by-event basis. Energy deposition by ions was simulated with SRIM. For analysis, all ions were treated initially as He, with corrected energies in keV given by

$$K_{\text{corr}} = 2.864 + 1.507K_{\text{uncorr}} - 5.871 \times 10^{-3}K_{\text{uncorr}}^2$$
$$+ 2.895 \times 10^{-5}K_{\text{uncorr}}^3,$$

(2)

derived from raw energies $K_{\text{uncorr}}$. This correction gave satisfactory mass spectra. Once events had been grouped by charge and mass, further energy corrections could be
applied to calculate the effective fiducial ‘volumes’ (FV) for each branch, which are proportional to a selected span of ion energies, namely 20-40 keV. Ions can be backscattered and lost, effects also modeled with the aid of SRIM. The loss percentages \( b_i \) and fiducial volumes are included in Table II.

The trigger time for each waveform is obtained by fitting a Woods-Saxon (or logistic) function \[24\] to the waveform. The fit reduces the effect of electronic noise at low pulse amplitudes. The large-signal (90 keV) timing resolution is 4 ns FWHM, which is broadened by noise to 35 ns for signals at 10 keV.

During a typical 1-hr HT data run, \( T_2 \) was introduced to the desired count rate, limited to \( \sim 200 \text{ s}^{-1} \) to avoid deadtime and pileup. HT is created by exchange between the introduced \( T_2 \) and the \( \text{H}_2 \) outgassing from the walls, catalyzed by platinum-group metals present in vacuum-gauge filaments \[25\]. The gauges could be valved off from the rest of the system, allowing measurements to be taken with predominantly HT or predominantly \( T_2 \). Catalytic conversion proceeded with a time constant of 8 min to an asymptotic HT activity fraction of 95%.

The decay channels for HT are listed in Table II. Most decays lead to the single-charge ‘main’ branches, 2, 3, and 4. In cases where two positive ions and two or more electrons are produced in the final state, the different detectable configurations are listed under the primary channel responsible for producing them (e.g., 6a, 6b, etc.). These combinatorial situations are treated in analysis. Only charged-particle final states are included: for the bound-state beta decay branch, 1, Bahcall \[26\] calculates 0.69% in the case of a bare tritium ion.

Fundamentally, the branching ratio is the fraction of coincidence events with a beta that lead to a specific final channel \( i \) (see Table II). Details of the multistep extraction of branching ratios are given in \[27\].

The most significant correction arises from initial ion momentum, which causes the radial distribution in the ion-detector plane to depend on the specific decay channel. These distributions are not known \textit{a priori}: the coincidence rates are therefore measured by scanning the ion detector radially from the axis. The measured distributions are termed Raw Count Functions (RCF). Because the ion detector is of non-negligible size, a point-spread function (PSF) is calculated from the geometry in order to deconvolve the underlying radial distributions. The integrals of those distributions are the corrected quantities needed to form the branching ratios. Other corrections are noted below.

Coincidence data from TRIMS are shown in Fig. 2. Bands produced by the decay channels shown in Table II particularly from the singly charged ions with mass 1, 3 and 4 amu, may be seen. In addition, minor bands from charge-2 decays marked \( b \) and \( c \), a mass-6 band from residual \( T_2 \), marked \( f \), and secondary-emission bands to the left of time zero can be seen. The diffuse horizontal bands with ion energies \( \sim 50 \text{ keV} \) or \( < 10 \text{ keV} \) are from decays that occur in the non-accelerating regions near the detectors. Such events are filtered out via the 20-40 keV ion-energy FV cut. Some runs were affected by intermittent data-acquisition problems. Affected periods were removed in analysis.

Charge-mass spectra (Fig. 2, middle) were then generated with Eqs. 3 and 1. The effective charge is:

\[
q_{\text{eff}} = (K_{\text{corr}} + K_\beta - K^0_\beta)V^{-1},
\]

where \( K_\beta \) is the detected electron energy and \( V \) the acceleration voltage (59.7 keV). The unknown initial beta kinetic energy \( K^0_\beta \) broadens the charge distribution, but separation into charge groups is still possible. For sorting events, \( K^0_\beta \) is fixed at 3 keV.

The plane is subdivided into cells within which the events are predominantly from single decay channels. Cross-contamination between neighboring cells is corrected on the charge axis according to the Geant4 simulation and on the mass axis by the Gaussian fits to the main peaks.

In the charge-mass plane one sees groups with \( q_{\text{eff}} = 1 \) and 2, but also 1.5. The latter are from two-electron branches 6 and 7 where one ion or one electron is missed. For branch 6 with two ions and two electrons the branching ratio is quantified by treating one of the ions (either

| \( i \) | Channel | \( b_i \) (%) | FV (keV) |
|---|---|---|---|
| Zero electron (bound-state beta decay) | | |
| 1. | He + H | | |
| One electron | | |
| 2. | HeH\(^+\) + e\(^-\) | 3.2 | 20.74 |
| 3. | He\(^+\) + H + e\(^-\) | 1.1 | 20.11 |
| 4. | He + H\(^+\) + e\(^-\) | 0.3 | 18.12 |
| 5. | He\(^+\) + H\(^-\) + e\(^-\) | 1.1, 0.3 | 20.11/2 |
| Two electrons (one shakeoff) | | |
| 6. | He\(^+\) + H\(^+\) + 2e\(^-\) | a He\(^+\) + H\(^+\) + 2e\(^-\) | 1.1, 0.3 | 20.23/2 |
| 7. | He\(^+\) + H + 2e\(^-\) | b He\(^+\) + H + 1e\(^-\) | 1.1, 0.3 | 20.23/2 |
| | c He\(^+\) + 2e\(^-\) | 1.1 | 20.11 |
| | d H\(^+\) + 2e\(^-\) | 0.3 | 18.12 |
| | e He\(^+\) + 1e\(^-\) | 1.1 | 20.11 |
| | f H\(^+\) + 1e\(^-\) | 0.3 | 18.12 |
| Three electrons (two shakeoff) | | |
| 8. | He\(^+\) + H\(^+\) + 3e\(^-\) | a He\(^+\) + H + 1e\(^-\) | 1.1 | 20.11/2 |
| | b He\(^+\) + H + 2e\(^-\) | 1.1 | 20.11/2 |
| +11 combinations | | |
 FIG. 2. Raw data from HT-rich source. Top panel a), d), e), f) – the main ion bands associated with masses 1, 3, 4, and 6; b) – doubly-charged He$^{++}$ band; c) two-ion band with He$^+$, T$^+$, and H$^+$. Bands to the left of time zero are secondary emission bands. Ion energies are from Eq. 2. Bin size: (1 ns, 0.4 keV). Center panel: Derived values of charge and mass for each event. Bin size: (0.01, 0.01). Bottom panel: Mass spectrum of the main charge-1 bands after removing contributions from T$_2$ (solid green) and from two-ion detection branches 6e and 6f (hatched magenta). The mass-3 and 4 peaks are each fit with three Gaussians; the results are shown. Bin size: 0.01.

is detected, either because the other backscattered and was lost, or because near the detector edge, one or the other missed the detector, only half of those events have a valid beta. The electron-loss corrections are determined by simulation. When one ion and one electron are both lost, the event becomes indistinguishable experimentally from a main-band event. In this case, the corrections, which are scan-position dependent, are calculated from one-ion and two-ion data where both electrons are detected, coupled with electron-loss simulations.

Two-ion events in which one ion strikes the mesh and dislodges a secondary electron proved to be uniquely useful in the analysis. The time-separated pulses from the beta or shakeoff and the secondary were identified and resolved via a double Woods-Saxon fit. The relative arrival time of the other, detected, ion then organized such events into three cleanly separated groups, two from HT and one from T$_2$. These were used to determine the isotopic purity (by activity) and to obtain pure HT spectra (Fig. 2 bottom) by subtracting the determined fraction of a T$_2$-rich spectrum from an HT-rich one.

Events with only the secondary electron detected yield “beta”-ion times that are small or even negative (Fig. 2 top). These events are not to be included in the branching ratios because they lack a beta.

The branching ratios obtained are listed in Table III. The upper limits for branches 5 and 8 represent the numbers of events in the appropriate charge-mass cells, here deemed to be background rather than signal. Statistical uncertainties are listed in column 3. RCF uncertainties are additional aggregated uncertainties prior to deconvolution, and include contributions from random coincidences, T$_2$ spectrum subtraction, the cross-contamination of neighboring cells by charge and mass peaks, modeling of electron backscattering and losses, and manual scan-position setting. Point-spread function uncertainties encompass the dimensionality, scan step size, and zero offsets of the deconvolution matrix. The FV for each branch (Table II) is subject to energy calibration uncertainties. For charge-2 branches FV is half as large as for charge-1 branches (indicated by “/2” in Table II).

Misalignment of the electric and magnetic fields would cause a departure from rotational symmetry, but was measured to be negligibly small, 4.7(7) mrad. Ion backscattering corrections and small corrections for the dependence on scan position of the scan step size, magnetic field, dead-layer loss, and detector acceptance were applied and did not contribute significant uncertainties. A search for $^2$H$^+$ ions yielded 0.3(2)% of H$^+$, and representative uncertainties are listed under DT in Table III. Loss of ions to charge exchange was estimated to be less than 0.25%.

In conclusion, the mass spectroscopic measurements of [11, 12] have for 60 years been the only data on the branching ratio of HT and T$_2$ to bound and unbound
TABLE III. Branching ratios and uncertainties for decay channels of HT.

| Channel | Uncertainties (absolute %) | Branch |
|---------|-----------------------------|--------|
| 1. One electron |                            |        |
| 2. HeHeH⁺ | 0.1 0.39 0.14 0.35 0.08 0.55 | 56.51(55) |
| 3. He⁺ + H | 0.1 0.27 0.05 0.28 0.05 0.41 | 24.98(41) |
| 4. He + H⁺ | 0.09 0.4 0.17 0.09 0.01 0.45 | 5.64(45) |
| 5. He⁺⁺ + H⁻ | < 0.021 |        |
| 2. Two electrons |                            |        |
| 6. He⁺⁺ + H⁺ | from He⁺⁺ 0.19 0.41 0.09 0.15 0.02 0.49 | 11.01(49) |
| from H⁺ | 0.17 0.37 0.09 0.13 0.02 0.44 | 10.43(44) |
| 7. He⁺⁺ + H | 0.12 0.07 0.16 0.05 0.21 | 2.16(21) |
| 3. Three electrons |                            |        |
| 8. He⁺⁺⁺ + H⁺ | < 0.045 |        |

tinglin194@gmail.com

dparno@cmu.edu

rghr@uw.edu

molecular ions. The profound discrepancy of those experiments with theory would imply either a dramatic and unexpected failure of the sudden approximation [28] at relatively high beta energies, or some hindrance of dissociation by 5 orders of magnitude so that radiative decays from highly excited transient molecular states could dominate.

We have measured the branching ratios for HT over the entire beta spectrum, as did Snell et al. [11] and Wexler [12], and find strong disagreement with the results of both experiments. In contrast, our results are in accord with theory even over this full range of beta energy. The source of the disagreement is not known. It may be due to the momentum acceptance of the instruments as Wexler himself suggested [12]. Another possibility is that the secondary-emission coefficient for the molecular ions may be larger than for atomic ions, biasing the efficiency of the instruments in favor of the molecular signals.

With the present results, the last known disagreement between experiment and the final-state distribution in tritium beta decay is removed, providing support for the theoretical analysis of neutrino mass experiments such as KATRIN [3] and Project 8 [29] that make use of molecular tritium.

The authors wish to thank Alejandro Saenz and Sanshiro Enomoto for valuable discussions. This material is based upon work supported by the U.S. Department of Energy Office of Science, Office of Nuclear Physics under Award Numbers DE-FG02-97ER41020 and [sc0019304]. We further acknowledge support from the PRISMA Cluster of Excellence at the University of Mainz, the Helmholtz Alliance for Astroparticle Physics HAP funded by the Initiative and Networking Fund of the Helmholtz Association, the Institute of Experimental Particle Physics at the Karlsruhe Institute of Technology (the Research University in the Helmholtz Association), and the German Academic Exchange Service.

[1] Y. Fukuda et al. (Super-Kamiokande), Phys. Rev. Lett. 81, 1562 (1998), hep-ex/9807003.
[2] Q. R. Ahmad et al. (SNO), Phys. Rev. Lett. 87, 071301 (2001), nucl-ex/0106015.
[3] M. Aker, K. Altemüller, M. Arenz, M. Babutzka, J. Barrett, S. Bauer, M. Beck, A. Beglarian, J. Behrens, T. Bergmann, et al. (KATRIN Collaboration), Phys. Rev. Lett. 123, 221802 (2019).
[4] G. Drexlin, V. Hannen, S. Mertens, and C. Weinheimer, Adv. High Energy Phys. 2013, 293986 (2013).
[5] S. R. Choudhury and S. Choubey, J. Cosmol. Astrop. Phys. 2018, 017017 (2018), ISSN 1747-7516.
[6] L. I. Bodine, D. S. Parno, and R. G. H. Robertson, Phys. Rev. C 91, 035505 (2015), 1502.03497.
[7] R. G. H. Robertson, T. J. Bowles, G. J. Stephenson, D. L. Wark, J. F. Wilkerson, and D. A. Knapp, Phys. Rev. Lett. 67, 957 (1991).
[8] W. Stoeffl and D. J. Decman, Phys. Rev. Lett. 75, 3237 (1995).
[9] S. Jonsell, A. Saenz, and P. Froelich, Phys. Rev. C 60, 034601 (1999).
[10] A. Saenz, S. Jonsell, and P. Froelich, Phys. Rev. Lett. 84, 242 (2000).
[11] A. H. Snell, F. Pleasonton, and H. E. Leming, J. Inorg. Nucl. Chem. 5, 112 (1957).
[12] S. Wexler, J. Inorg. Nucl. Chem. 10, 8 (1959).
[13] S. Wexler and F. T. Porter, J. Chem. Phys. 50, 5428 (1969).
[14] L. I. Bodine, Ph.D. thesis, University of Washington, Seattle WA (2015).
[15] Introduction to COMSOL Multiphysics, Version: COMSOL 5.2, COMSOLAB (2015).
[16] M. H. Lowe et al., IEEE Trans. Nucl. Sci. 51, 878 (2004), see also http://orca.phys.uvic.ca/
[17] R. Brun and F. Rademakers, Nucl. Instrum. Meth. A 389, 81 (1997), see also http://root.cern.ch
[18] V. T. Jordanov and G. F. Knoll, Nucl. Instrum. Meth. A 345, 337 (1994).
[19] J. Amsbaugh et al., Nucl. Instrum. Meth. A 778, 40 (2015).
[20] S. Agostinelli et al. (GEANT4), Nucl. Instrum. Meth. A 506, 250 (2003).
[21] J. Allison et al., Nucl. Instrum. Meth. A 835, 186 (2016).
[22] P. Dondero, A. Mantero, V. Ivanchenko, S. Lotti, T. Mino, and V. Fioretti, Nucl. Instrum. Meth. B 425, 18 (2018).
[23] J. Ziegler, M. Ziegler, and J. Biersack, Nucl. Instrum. Meth. B 268, 1823 (2010).
[24] R. D. Woods and D. S. Saxton, Phys. Rev. 95, 577 (1954).
[25] P. R. Norton and P. J. Richards, Surf. Sci. 41, 293 (1974).
[26] J. N. Bahcall, Phys. Rev. 124, 495 (1961).
[27] Y.-T. Lin, Ph.D. thesis, University of Washington, Seattle WA (2019).
[28] A. B. Migdal, J. Phys. (USSR) 4, 449 (1941).
[29] D. M. Asner, R. F. Bradley, L. de Viveiros, P. J. Doe,
J. L. Fernandes, M. Fertl, et al. (Project 8), Phys. Rev. Lett. 114, 162501 (2015), 1408.5362.