First Observation of Bound-State β^- Decay

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Bound-state β^- decay was observed for the first time by storing bare ${}^{163}_{66}\mathrm{Dy}^{66+}$ ions in a heavy-ion storage ring. From the number of ${}^{163}_{67}\mathrm{Ho}^{66+}$ daughter ions, measured as a function of the storage time, a half-life of 47^{+5}_{-4} d was derived. By comparing this result with reported half-lives for electron capture (EC) from the M_1 and M_2 shells of neutral ${}^{163}_{67}\mathrm{Ho}$, bounds for both the Q_{EC} value of neutral ${}^{163}_{67}\mathrm{Ho}$ and for the electron-neutrino mass were set.

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Beta decay into *bound* electron states of the daughter atom (β_b), accompanied by the emission of a monochromatic antineutrino, was first predicted by Daudel, Jean, and Lecoin [1] in 1947 and, 14 years later, discussed in detail by Bahcall [2]. Up to now β_b decay has not been observed, although some hint of its existence was derived from the difference of the tritium lifetime in the ionic (${}^3\mathrm{H}^-$) and molecular (${}^3\mathrm{H}_2$) states [3]. For neutral atoms β_b decay is only of minor importance. It might become a strong, if not the only, decay channel for highly ionized atoms which exist, e.g., in stellar plasmas during nucleosynthesis. The astrophysical relevance of β_b decay, in particular for the *s* process and for cosmochronology, was pointed out by several authors [4–7].

It was proposed [8] to investigate β_b decay in the new heavy-ion storage ring ESR at GSI, Darmstadt, with heavy bare ions stored for extended periods of time. For instance, ${}^{163}_{66}\mathrm{Dy}$ is *stable as a neutral atom,* but as a fully stripped ${}^{163}_{66}\mathrm{Dy}^{66+}$ ion it can decay into ${}^{163}_{67}\mathrm{Ho}^{66+}$ by β_b decay into the K and L shells of the daughter atom. The ground state (g.s.) β_b decay ${}^{163}_{66}\mathrm{Dy}^{66+}$ $(I=\frac{5}{2}^-)$ $\rightarrow$ ${}^{163}_{67}\mathrm{Ho}^{66+}$ $(I=\frac{7}{2}^-)$ is an allowed Gamow-Teller (GT) transition [9]. Its Q value, which is essentially the total energy of the monochromatic antineutrino (the recoil energy amounts to only 8 meV), is given for *bare* ${}^{163}_{66}\mathrm{Dy}^{66+}$ by

$$Q^{K,L}_{\beta_b}=|B^{K,L}_{\mathrm{Ho}^{66+}}|-|\Delta B_e|-Q_{\mathrm{EC}}\,. \qquad (1)$$

Here $B^{K,L}_{\mathrm{Ho}^{66+}}$ is the electron binding energy in the K or L shell of hydrogen-like holmium, ΔB_e is the difference of the total electron binding energies in neutral holmium and dysprosium, and Q_{EC} is the Q value for electron capture in neutral ${}^{163}_{67}\mathrm{Ho}$. With $|B^K_{\mathrm{Ho}^{66+}}|=65.137$ keV [10], $\Delta B_e=12.493$ keV [11], and the directly measured Q_{EC} value of (2.3 ± 1) keV [9] it follows

$$Q^K_{\beta_b}\approx(50.3\pm1)\ \mathrm{keV} \ \text{and}\ Q^L_{\beta_b}\approx(1.7\pm1)\ \mathrm{keV}\,. \qquad (2)$$

For the experimental observation of β_b decay, up to 10^8 bare ${}^{163}\mathrm{Dy}^{66+}$ ions of 294 MeV/u were accumulated, stored, and cooled with electrons in the ESR. During the storage the β_b-decay daughters, stable hydrogen-like ${}^{163}_{67}\mathrm{Ho}^{66+}$ ions, were continuously created. Having almost the same mass-over-charge ratio (A/q) as the primary ions they were stored and cooled on the *same orbit.* The experimental procedure adapted by us to measure the number of β_b-decay daughters, ${}^{163}_{67}\mathrm{Ho}^{66+}$, was as follows (see Fig. 1): First, ${}^{163}_{66}\mathrm{Dy}^{66+}$ ions were accumulated in the ring for a typical time of 30 min. [Fig. 1(a), "accumulation"]. Then an internal argon gas jet [thickness$=6\times10^{12}$ atoms/cm^2, diameter=3 mm (FWHM)], which vertically crossed the beam, was turned on for about 500 s. By this most of the ${}^{163}_{67}\mathrm{Ho}^{66+}$ daughters pro-

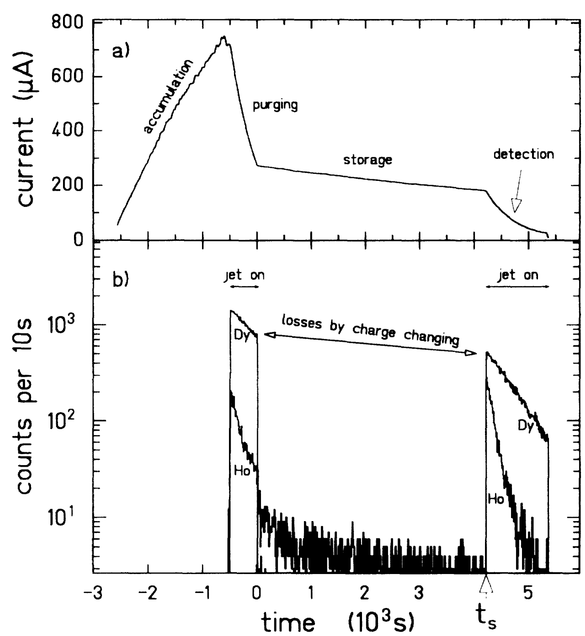

FIG. 1. (a) Current of stored bare ${}^{163}_{66}\mathrm{Dy}^{66+}$ ions in the storage ring ESR during the various stages of the experiment. (b) Rates of Dy^{65+} ions (labeled "Dy," scaled down by 2^8) and of Ho^{67+} ions ("Ho") detected in the outer and inner position-sensitive detectors, respectively, which are moved into the ring chamber after the accumulation stage. Note the strong increase of the Ho^{67+} rate by more than an order of magnitude at t_s with respect to $t=0$.

duced during accumulation were removed from the closed orbit via electron capture or electron loss ("purging"). After the gas jet was turned off, the ions—the primary $^{163}_{66}\mathrm{Dy}^{66+}$ as well as the decay products $^{163}_{67}\mathrm{Ho}^{66+}$—were stored and cooled for a variable time t_s ("storage") ranging from 10 to 85 min. The electron cooler was in operation during all stages of the experiment.

The detection and identification of these $^{163}_{67}\mathrm{Ho}^{66+}$ daughters were based on the fact that *only they* could be further ionized, thus reducing their magnetic rigidity. Therefore, in the last step ("detection") the gas jet was turned on again in order to strip off the K electron in $^{163}_{67}\mathrm{Ho}^{66+}$ and to detect the bare $^{163}_{67}\mathrm{Ho}^{67+}$ with a position-sensitive particle counter [12]. It was placed in a movable pocket behind the first dipole following the gas jet, at the inner side of the closed orbit. The Ho^{67+} ions were recorded as a narrow peak (FWHM ≈ 3 mm; see inset of Fig. 2) in this detector at the expected position. It was calibrated by stripping the electron of hydrogen-like $^{163}\mathrm{Dy}^{65+}$ ions in a complementary experiment.

A second auxiliary experiment was performed with bare ions of the neighboring isotope $^{161}\mathrm{Dy}^{66+}$ for which β_b decay and any other decay is energetically forbidden. It was used to determine the amount of $^{163}\mathrm{Ho}$ which could be produced by nuclear charge-exchange reactions in the gas jet, since these cross sections are expected to be very similar for both isotopes $^{161}\mathrm{Dy}$ and $^{163}\mathrm{Dy}$. From the experimentally determined limit of the production of $^{161}\mathrm{Ho}$ a limit $< 10^{-2}$ (with respect to β_b decay) for nuclear-reaction-induced $^{163}\mathrm{Ho}$ was deduced under the assumption that the charge-exchange cross sections are equal for $^{161}\mathrm{Ho}$ and $^{163}\mathrm{Ho}$. Therefore, this nuclear reaction in the gas jet could *not* mask the detection of $^{163}\mathrm{Ho}$ produced in β_b decay.

In Fig. 1(b) the counting rates in the inner detector (labeled "Ho") during the various stages of the experiment are shown. Also the rates in a second particle detector, placed on the *outer* side of the closed orbit, are shown ("Dy"). Mainly $^{163}\mathrm{Dy}^{65+}$ ions produced by electron *capture* of $^{163}\mathrm{Dy}^{66+}$ in the gas jet appeared there.

From rate equations which describe the ratio $N^*_{\mathrm{Ho}}(t_s)/N_{\mathrm{Dy}}(t_s)$ as a function of the storage time t_s, the β_b-decay constant in the rest frame, λ_{β_b}, can be derived,

$$\frac{N^*_{\mathrm{Ho}}(t_s)}{N_{\mathrm{Dy}}(t_s)} = \frac{\lambda_{\beta_b}}{\gamma} t_s [1 + \tfrac{1}{2}(\lambda^{\mathrm{cc}}_{\mathrm{Dy}} - \lambda^{\mathrm{cc}}_{\mathrm{Ho}}) t_s + \cdots] , \qquad (3)$$

where

$$N^*_{\mathrm{Ho}}(t_s) = N_{\mathrm{Ho}}(t_s) - N_{\mathrm{Ho}}(0)\exp(-\lambda^{\mathrm{cc}}_{\mathrm{Ho}} t_s) .$$

$N_{\mathrm{Ho}}(t_s)$ is the number of Ho^{66+} at the time t_s (end of storage), and $N_{\mathrm{Ho}}(0)$ is the number of Ho^{66+} ions at $t = 0$ (beginning of storage). γ is the Lorentz factor for the stored ions ($\gamma = 1.316$).

$\lambda^{\mathrm{cc}}_{\mathrm{Ho}}$ and $\lambda^{\mathrm{cc}}_{\mathrm{Dy}}$ are the loss factors (in the laboratory system) for Ho^{66+} and Dy^{66+}, respectively, due to charge-changing processes in the electron cooler and in the residual gas during storage. $\lambda^{\mathrm{cc}}_{\mathrm{Dy}}$ was measured on-line by monitoring the current of the stored ions by a calibrated beam transformer. $\lambda^{\mathrm{cc}}_{\mathrm{Ho}}$ was obtained by properly scaling up by 3% the corresponding loss factor found for hydrogen-like Dy^{65+} ions stored and cooled under similar conditions. As $(\lambda^{\mathrm{cc}}_{\mathrm{Dy}} - \lambda^{\mathrm{cc}}_{\mathrm{Ho}}) t_s \le 0.1$ for all storage times t_s taken, Eq. (3) basically shows a *linear* dependence of the $^{163}\mathrm{Ho}^{66+}/^{163}\mathrm{Dy}^{66+}$ ratio on t_s. The number of Dy^{66+} ions at t_s $[N_{\mathrm{Dy}}(t_s)]$ was found by setting a narrow window on the peak of the hydrogen-like $^{163}\mathrm{Dy}^{65+}$ ions in the outer detector and by fitting an exponential to the rate distribution. Similarly the number of Ho^{66+} ions at t_s $[N_{\mathrm{Ho}}(t_s)]$ was obtained from the peak of the bare $^{163}\mathrm{Ho}^{67+}$ ions in the inner detector (see Fig. 2, inset). A small but not negligible fraction of the Ho^{66+} ions, however, *captures* an electron in the gas jet and appears in the *outer* detector. Since this detector was not Z sensitive, those Ho^{65+} ions could *not* be discriminated from Dy^{65+} ions. Therefore, in order to get the number of Ho^{66+} ions stored at t_s, $N_{\mathrm{Ho}}(t_s)$, the number measured in the inner detector had to be scaled up by the ratio $(\sigma^I_{\mathrm{Ho}} + \sigma^C_{\mathrm{Ho}})/\sigma^I_{\mathrm{Ho}}$, where σ^I_{Ho} and σ^C_{Ho} denote the ionization and electron-capture cross sections of Ho^{66+} in the argon jet. This ratio $(\sigma^I_{\mathrm{Ho}} + \sigma^C_{\mathrm{Ho}})/\sigma^I_{\mathrm{Ho}} = 1.19 \pm 0.05$ was obtained by properly scaling up by 5% the corresponding result of the experiment with hydrogen-like $^{163}\mathrm{Dy}^{65+}$ ions. From this experiment the relative efficiency of the

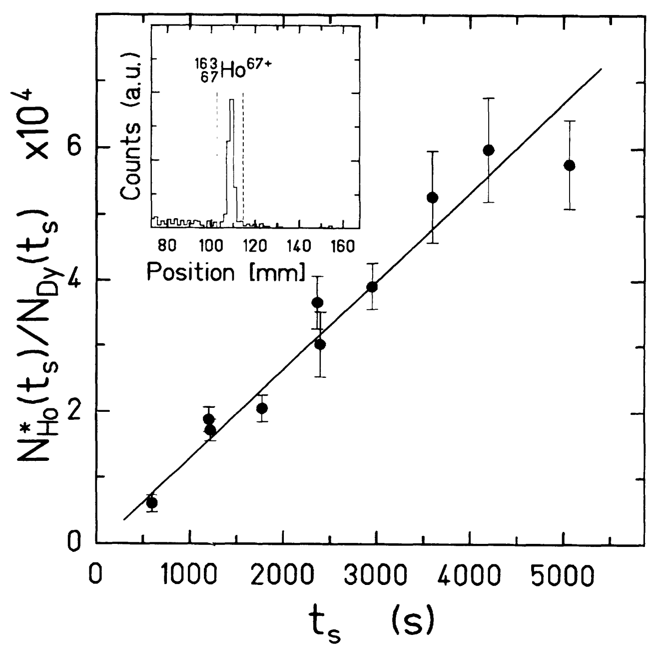

FIG. 2. Ratio of the number of β_b-decay daughter atoms $^{163}_{67}\mathrm{Ho}^{66+}$ $[N^*_{\mathrm{Ho}}(t_s)]$ to primary $^{163}_{66}\mathrm{Dy}^{66+}$ ions $[N_{\mathrm{Dy}}(t_s)]$ as a function of the storage time t_s [see Eq. (3)], together with a linear fit to the data. Inset: The peak of Ho^{67+} ions recorded by the inner particle detector. The dashed lines indicate the window used for the data analysis.

inner and outer detector was determined as $\varepsilon_i/\varepsilon_o = 1 \pm 0.03$ as well. For the evaluation of the final error we concede a systematical error of 4% in total for the scaling of the capture-to-loss ratio and for the relative detector efficiency.

By fitting a straight line to the experimental data (Fig. 2) according to Eq. (3), one gets a total β_b-decay constant in the ion rest frame of

$$\lambda_{\beta_b} = (1.72 \pm 0.1 \pm 0.07) \times 10^{-7}\,\mathrm{s}^{-1}, \quad (4)$$

where the first error is the statistical, the second the estimated systematical error. By adding both errors a total β_b decay half-life of

$$T_{1/2}^{\beta_b} = 47^{+5}_{-4}\,\mathrm{d} \quad (5)$$

for the β_b decay ${}^{163}_{66}\mathrm{Dy}^{66+} \rightarrow {}^{163}_{67}\mathrm{Ho}^{66+}$ is obtained, in excellent agreement with the value of 50 d, predicted by Takahashi [6].

Although β_b decay into the K and L shells could not be distinguished in our detection setup, the branching ratio may be calculated according to

$$\frac{\lambda_{\beta_b}^K}{\lambda_{\beta_b}^L} = \frac{(Q_{\beta_b}^K)^2}{(Q_{\beta_b}^L)^2} \frac{|\psi_K^{66+}(R)|^2}{|\psi_L^{66+}(R)|^2}. \quad (6)$$

With $Q_{\beta_b}^{K,L}$ given by Eq. (1), by using the reported value [9] of (2.3 ± 1) keV for Q_{EC} and by calculating the electron densities of the hydrogen-like daughter atoms at the nuclear surface, $|\psi_X^{66+}(R)|^2$, according to Ref. [13], one gets $\lambda_{\beta_b}^K/\lambda_{\beta_b}^L = (7^{+29}_{-4}) \times 10^3$ and, therewith,

$$\lambda_{\beta_b} \approx \lambda_{\beta_b}^K. \quad (7)$$

A further, independent confirmation of our results came from the frequency analysis of the Schottky noise which was performed on the stored ions during and after their interaction with the gas jet. In addition to the intense primary beam, a significant signal from stored ${}^{163}\mathrm{Ho}^{67+}$ ions has been observed after storage and turning on the gas jet (the particle detectors were removed in that case). This signal increased in proportion to t_s in contrast to the much smaller signals from nuclear reaction products being *not* proportional to t_s [14].

β_b decay and orbital EC are time-mirrored processes with respect to weak interaction. β_b decay of a *bare* ion into a specific electron shell on the one hand, and EC from a given electron shell of a *neutral* atom on the other hand, have transition strengths the ratio of which generally depends only on phase space, electron densities at the origin, and on statistical factors, but not on the weak interaction matrix elements. That is true in particular for the $\Delta I = 1$, $\pi_i\pi_f = +1$ allowed GT transition of ${}^{163}\mathrm{Ho}/{}^{163}\mathrm{Dy}$ where only one matrix element is involved. For instance, the ratio of β_b decay into the K shell and EC from the M_1 or M_2 shells in ${}^{163}\mathrm{Ho}/{}^{163}\mathrm{Dy}$ is given by [cf. Eqs. (2.27) and (2.28) in Ref. [15] and Eq. (3) in Ref. [16]]

$$\frac{\lambda_{\beta_b}^K}{\lambda_{\mathrm{EC}}^{M_{1,2}}} = \frac{1}{2} \frac{2I_{\mathrm{Ho}}+1}{2I_{\mathrm{Dy}}+1} \frac{q_{\bar{\nu}_e} W_{\bar{\nu}_e}^K}{q_{\nu_e} W_{\nu_e}^{M_{1,2}}} \times \frac{|\psi_{1s}^{66+}(R)|^2_{\mathrm{Ho}}}{|\psi_{M_{1,2}}^0(R)|^2_{\mathrm{Dy}}} \frac{B_{1s}^{66+}(\mathrm{Ho})}{B_{M_{1,2}}^0(\mathrm{Dy})}. \quad (8)$$

Here q_{ν_e}, W_{ν_e} are the momenta and total energies of the monochromatic neutrino and antineutrino, respectively, $|\psi_X^q(R)|^2$ are the electron densities at the nuclear surface ($R = 6.75$ fm for $A = 163$, assuming a uniform nuclear charge distribution) for the corresponding daughter atom, B_X^q are the exchange and overlap correction factors (close to unity), $I_{\mathrm{Ho}} = \frac{7}{2}, I_{\mathrm{Dy}} = \frac{5}{2}$ are the nuclear spins, and $\frac{1}{2}$ accounts for the (different) number of electrons involved in β_b decay and in EC, respectively.

Values for $\lambda_{\mathrm{EC}}^{M_1}$ as well as for $\lambda_{\mathrm{EC}}^{M_2}$ were reported in Ref. [17] from EC measurements in the M subshells of neutral ${}^{163}\mathrm{Ho}$ [$\lambda_{\mathrm{EC}}^{M_1} = (0.9740 \pm 0.0041) \times 10^{-12}\,\mathrm{s}^{-1}$, $\lambda_{\mathrm{EC}}^{M_2} = (0.0817 \pm 0.0035) \times 10^{-12}\,\mathrm{s}^{-1}$]. In Ref. [18] a total half-life for M capture in neutral ${}^{163}\mathrm{Ho}$ of $T_{1/2}^{M} = (18\,000 \pm 2000)$ yr is quoted. Scaling up this value by 8.4% in order to eliminate the M_2 contribution [15], one gets a second value for the M_1-EC probability as $\lambda_{\mathrm{EC}}^{M_1} = (1.13 \pm 0.15) \times 10^{-12}\,\mathrm{s}^{-1}$, where an estimated error of 2% for the scaling procedure was added. From these numbers an (unweighted) experimental mean value of $\langle \lambda_{\mathrm{EC}}^{M_1} \rangle = (1.05 \pm 0.075) \times 10^{-12}\,\mathrm{s}^{-1}$ follows.

Combining these results for $\lambda_{\mathrm{EC}}^{M_{1,2}}$ and for the β_b-decay probability λ_{β_b} given in Eq. (4) [setting $\lambda_{\beta_b} = \lambda_{\beta_b}^K$; see Eq. (7)], one gets from Eq. (8) two independent sets [Eqs. (11) and (12)] for the ratio of the phase-space factors of β_b decay and of $M_{1,2}$ electron capture, respectively. Thereby the electron densities at the nuclear surface were calculated with the Oxford code [19,20] and the overlap and exchange factors B_X^q were extracted from the corresponding tables in Ref. [15]. Finally, it was taken into account that the total energies W_{ν_e} of the neutrino (antineutrino) are connected with Q_{EC} by

$$W_{\bar{\nu}_e}^K \approx Q_{\beta_b}^K = 52.644 - Q_{\mathrm{EC}} = 50.597 - Q^{M_1}\ (\mathrm{keV}), \quad (9)$$

$$W_{\nu_e}^{M_{1,2}} \approx Q^{M_{1,2}} \text{ and } Q^{M_{1,2}} = Q_{\mathrm{EC}} - |B_{\mathrm{Dy}^0}^{M_{1,2}}|, \quad (10)$$

using Eq. (1), where $Q^{M_{1,2}}$ are the Q values for EC from the $M_{1,2}$ shells and where $B_{\mathrm{Dy}^0}^{M_{1,2}}$ are the binding energies in the neutral dysprosium daughter atom ($B_{\mathrm{Dy}^0}^{M_1} = 2047$ eV, $B_{\mathrm{Dy}^0}^{M_2} = 1842$ eV, Ref. [21]). Neglecting the antineutrino mass in the numerator of the following equations (the upper limit is 17 eV [22]), one gets constraints for m_{ν_e} and Q^{M_1} as

$$\frac{(50.597 - Q^{M_1})^2}{Q^{M_1}[(Q^{M_1})^2 - m_{\nu_e}^2]^{1/2}} = 6855 \pm 840 \quad (11)$$

(this work and Refs. [17,18]),

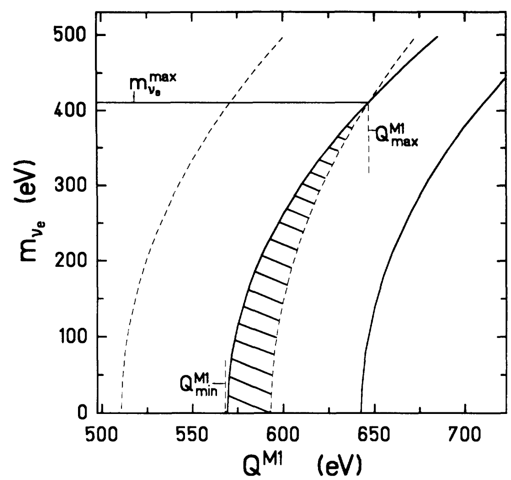

FIG. 3. Bounds for Q^{M_1}, the Q value for EC from the M_1 shell in ^{163}Ho, and for the neutrino mass m_{ν_e} from Eq. (11) (solid lines) and from Eq. (12) (dashed lines), respectively. The area allowed by Eqs. (11) and (12) is hatched. The resulting upper and lower limits for Q^{M_1} and the upper limit for m_{ν_e} (68% C.L.) are indicated. The data are from this work and from Refs. [17,18].

$$\frac{(50.597-Q^{M_1})^2}{(Q^{M_1}+0.205)[(Q^{M_1}+0.205)^2-m_{\nu_e}^2]^{1/2}} = 4412\pm 485 \qquad (12)$$

(this work and Ref. [17]). Calculated or tabulated values such as electron densities, binding energies, etc., are assumed to be without systematical errors. The unit for Q^{M_1} and m_{ν_e} in Eqs. (11) and (12) is keV.

Figure 3 shows the regions for m_{ν_e} and Q^{M_1} which lie within the bounds set by Eq. (11) (solid lines) and by Eq. (12) (dashed lines). The hatched area indicates the region in accordance with *both* Eqs. (11) and (12) and with the constraint $m_{\nu_e} \geq 0$. The resulting bounds are, on the 68% C.L.,

$$569 \leq Q^{M_1} \leq 647 \text{ eV}, \text{ or } 2616 \leq Q_{\mathrm{EC}} \leq 2694 \text{ eV},$$
$$\text{and } m_{\nu_e} \leq 410 \text{ eV}. \qquad (13)$$

These bounds sets for Q_{EC} and for m_{ν_e} are considerably more restrictive than the ones obtained in Ref. [17] from the ratio $\lambda_{\mathrm{EC}}^{M_1}/\lambda_{\mathrm{EC}}^{M_2}$ alone, although the experimental errors incorporated in Eqs. (11) and (12) are large. That is explained by the fact that the phase-space factor for β_b decay [numerators of Eqs. (11) and (12)] is very large compared to that for EC (denominators). Therefore their ratio depends very sensitively on the values of both m_{ν_e} and Q^{M_1}. The upper limit set for the *neutrino* mass m_{ν_e} in Eq. (13) is still far away from the presently smallest upper limit of 17 eV [22] set for the *antineutrino* mass. It is obvious, however, that a precise, independent measurement of $Q_{\mathrm{EC}}(^{163}\mathrm{Ho}/^{163}\mathrm{Dy})$ would lead—together with improved λ_{β_b} and λ_{EC} values—to really sensitive bounds for m_{ν_e}.

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