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April 29, 1982A LIMIT ON THE MASS OF THE ELECTRON NEUTRINO : THE CASE OF ^{163}Ho

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ABSTRACT

The partial beta-decay half-life for capture of M electrons in ^{163}Ho is $(4.0 \pm 1.2) 10^4$ y, and the ^{163}Ho - ^{163}Dy mass difference is 2.3 ± 1.0 keV. From these independent results we deduce a half-life of $(7 \pm 2) \cdot 10^3$ y and an upper limit on the electron neutrino mass of 1.3 keV.

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It was recently pointed out [1] that the shape of the internal bremsstrahlung (IB) spectrum following electron-capture beta decay (EC) may provide a new and superior approach for assessing the mass of the electron neutrino. One of the most prominent candidates for an experiment of this kind is the isotope ^{163}Ho that decays by electron capture into ^{163}Dy , and for which the half-life and Q-value data are conflicting (Q is defined, as usual, as the mass difference between the neutral ^{163}Ho and ^{163}Dy atoms). These conflicts have prompted us to perform two preliminary experiments. The first is a measurement of the M electron-capture rate in ^{163}Ho . The daughter atom's M shell x-rays and Auger electrons are observed (capture from K and L shells are energetically forbidden). The observed rate, due to a fortunate precise knowledge of nuclear physics details, can be translated into a "phase space factor", a function of Q and the electron neutrino mass m_ν . The second experiment directly determines Q in nuclear reactions, independently of m_ν . Comparison of the two experiments yields already an upper limit on m_ν that is considerably more restrictive than the one of 4.1 keV set by Beck and Daniel [2]. Our present limit is not competitive with the limits on the electron antineutrino mass [3].

The long-lived radioactive isotope ^{163}Ho was discovered by Naumann et al. [4,5] who detected radiations of about 1 keV interpreted as Auger electrons and x-rays following EC in the atomic M shell. From the absence of capture in the L shell an upper limit of 9.1 keV could be set on the $^{163}\text{Ho} - ^{163}\text{Dy}$ mass difference Q, while the direct observation [6] of capture from the M_I shell provides a lower limit at the M_I binding energy of 2.05 keV. These limits are clearly inconsistent with the Q_{EC} value of -2.9 ± 3.1 keV derived [7] from nuclear-reaction data. Via nuclear-structure arguments to be given later in the present paper, the upper limit on Q can be shown to imply a lower limit of 150 y on the ^{163}Ho half-life, inconsistent with the value of 33 ± 23 y found by Hopke et al. [5]. In the experiments reported here we have determined the ^{163}Ho half-life by counting M-shell radiations from carefully prepared sources containing a known number of atoms, and we have determined the Q value by a novel technique based on single-nucleon transfer reactions.

The radioactivity was produced in the ISOLDE Facility connected to the CERN synchro-cyclotron. A hot tantalum target consisting of 10 μm rolls of foils to give a target thickness of 122 g/cm^2 was bombarded with a 2.4 μA beam of 600 MeV protons, and rare-earth elements evaporating from the target were ionized by surface ionization and mass separated. The sample collected at mass 163 consisted predominantly of the elements holmium to lutecium (the yield is increased by collecting also the isobars, which decay to ^{163}Ho). The collection took place in a Faraday cup; from the integrated current the number of atoms on the 10 μm tantalum collector foil

was calculated to be $1.36 \cdot 10^{15}$. After some weeks of cooling the main contaminants in the sample (determined by gamma spectroscopy) were ^{147}Gd ($1.4 \cdot 10^{12}$ atoms collected as an oxide side band), and ^{167}Tm , ^{169}Yb (both about $4 \cdot 10^9$ atoms from "tails" in the separator). A further purification was obtained in a complex procedure based on wet chemistry. The collector foil was dissolved in the presence of 3 mg lanthanum carrier and microgram amounts of other rare-earths except holmium. For tracing purpose a small amount of short lived ^{160}Ho was added. The separation was carried out by standard precipitation steps followed by ion-exchange chromatography on an Aminex A5 column with 0.11 M alpha-hydroxyisobutyric acid as the eluant. The holmium fraction was cleaned twice again by the same procedure. From the shape of the elution curves it was estimated that each step gave a decontamination of a factor 10^3 from the heavier rare earths (such as Tm, Yb) and considerably more from the lighter ones. Thus the total decontamination offered by the combination of mass separation and chemistry was of the order $10^{14} - 10^{15}$. The yield in the chemical procedure was 44%.

Two thin sources were prepared from this stock of radioactivity, both by vacuum evaporation from a rhenium ribbon at 1800°C . The first (source A) was made before the ^{160}Ho tracer had decayed, so that the yield could be determined: the result of 49% corresponds to $1.9 \cdot 10^{13}$ atoms in the central part of a 64 cm^2 tantalum backing.

A second thin source (source B) was prepared by evaporating a sample of 14.6% of the original stock onto a $0.1 \times 20 \times 20 \text{ mm}^3$ beryllium foil. The evaporation yield was estimated in an experiment with a Sm tracer and the same geometry to be 35%, which would correspond to $3.1 \cdot 10^{13}$ atoms. A second estimate came from counting sources A and B in a $10 \times 10 \text{ cm}^2$ multi-wire proportional chamber (MWPC) filled with an argon-methane mixture; from the relative count rates one finds $4.5 \cdot 10^{13}$ atoms of ^{163}Ho . A third and entirely independent check on the contents of source B was obtained by a highly sensitive surface-analysis technique; elastic Coulomb scattering of alpha particles in back angles at relatively low energy offers a sensitive probe for surface layers and a good mass resolving power arising from the kinematic energy loss in the collision. The experiment carried out with a beam of 3.5 MeV alpha particles from the Aarhus single-stage Van De Graaff accelerator shows (fig. 1) a well-resolved peak at the holmium mass position with an intensity (integrated over the source) of $5.0 \cdot 10^{13}$ atoms. There is thus excellent agreement with the Faraday-cup and chemical-yield data. On the basis of these results we stress that a consistent scale for the number of ^{163}Ho atoms on the sources has been obtained by two entirely independent techniques. We also note that the counting rates of 1 keV M Auger electrons agree when measured from two different sources on backings with very different Z (4 and 73), so that back-scattering and self-absorption effects cannot be excessive.

Consequently we may use the M-Auger counting rates in the MWPC to get a partial M half-life. The result is 51 000 y.

Source B was further studied by internal counting in a gas-flow proportional counter. The disintegration rate in this measurement was 32.4 s^{-1} . The same counter was used for counting Dy M x-rays. For this experiment the counter was filled with an Ar-CH₄ mixture and the internal source was covered with a 2 μm mylar layer to absorb the Auger electrons. The transmission of the x-rays through the mylar was calculated to be 48%. The M fluorescence yield in dysprosium was calculated from the tables of Bambynek et al. [8] to be 0.98%. (We note that M fluorescence yields have been measured [9] in a few cases and agree well with calculations.) The measurement showed clearly a ray at about 1.3 keV, which agrees well with the M x-ray fine structure observed by Bennett et al. [6] and with an intensity expressed in terms of M disintegration rates of 27.4 s^{-1} . On the basis of the electron result of 32.4 s^{-1} together with an assumed $4.5 \cdot 10^{13}$ atoms in the source, we calculate a partial M half-life of 30 000 y.

The various measurements summarized above serve as a set of cross checks safeguarding against systematic errors. For this reason it would have little meaning to quote a best value, and we choose instead to represent the data by the single number $T_{1/2}^M = (4.0 \pm 1.2) \cdot 10^4 \text{ y}$. We now show how this number can be turned into a determination of the EC phase space factor, which for the M_I shell can be written

$$\phi(M_I) = (Q - E(M_I)) \cdot [(Q - E(M_I))^2 - m_\nu^2 c^4]^{1/2} \dots \quad (1)$$

where $E(M_I)$ is the M_I binding energy and m_ν the electron neutrino rest mass.

The transition probability in electron-capture beta decay involving a given atomic (sub-)shell x can be written [10] as a product of four terms : the strength of the weak interaction, a nuclear term, a term involving the bound atomic state x and the phase space factor. The properties of the bound atomic states being known and tabulated quantities [10], it suffices to calculate the nuclear matrix elements in order to derive the phase space factor from the transition probability. Owing to a most fortunate coincidence in the nuclear structure of ¹⁶³Ho this turns out to be possible.

The nuclei ¹⁶³Ho and ¹⁶³Dy are strongly deformed, and the states of the odd nucleon can be characterized by the Nilsson quantum numbers appropriate for the spheroidal nuclear potential. Leaving aside the details of the nuclear model which can be found in Bohr and Mottelson [11], we note that the ground states of the two nuclei have identical Nilsson quantum numbers except for the projection of total angular

momentum on the nuclear symmetry axis. This projection is $7/2$ for holmium and $5/2$ for dysprosium, so that the beta transition proceeds between members of a spin-orbit doublet. Such transitions belong to a class of relatively rare transitions referred to as "allowed, unhindered" (au), which have very large transition probabilities. A systematic study [12] shows that the transition probability for au decays has an absolute value that reflects the single-particle matrix element modified in an important way by spin-isospin polarization effects, but also that local variations as a function of N and Z are essentially dominated by the pairing correction and trivial statistical factors. We may therefore use the measured reduced transition probability for the case of ^{161}Ho to calculate the value for ^{163}Ho , simply by taking into account the pairing correction which in either case takes the form $u_p^2 \cdot u_n^2$, where the u_x^2 are the emptiness coefficients for the orbitals involved. A calculation by Bengtsson and Ragnarsson [13] gave $u_p^2 \cdot u_n^2 = 0.508$ and 0.354 for ^{163}Ho and ^{161}Ho , respectively. The known half-life and Q value of ^{161}Ho and the above-mentioned calculations allow us to determine the nuclear matrix element in the ^{163}Ho decay. We assume a 25% error in this semiempirical calculation and combine it with the observed ^{163}Ho M-shell capture rate to obtain $\phi^{1/2}(M_I) = 0.53 \pm 0.10$ keV, after correction for M_{II} capture. This corresponds to $Q = 2.58 \pm 0.10$ keV for an assumed $m_\nu = 0$. The partial half-life $T_{1/2}^M$ and the theoretically known ratios for M/N/O capture (given by electron wave functions at the origin) result in a ^{163}Ho half-life of $(7 \pm 2) \cdot 10^3$ y.

A measurement of Q without any dependence on the neutrino mass may be obtained by performing nuclear reaction experiments. Such a direct determination of the $^{163}\text{Ho} - ^{163}\text{Dy}$ mass difference was performed at the Niels Bohr Institute tandem accelerator and multi-angle magnetic spectrograph. The $^{163}\text{Dy}(\tau, t)$ reaction would directly connect the two nuclei, but it has a very small cross-section. Instead the reaction chain $^{163}\text{Dy}(d, t) ^{162}\text{Dy}(\tau, d^*) ^{163}\text{Ho}$ was used. (We use the conventional symbols τ and t for ^3He and ^3H , and use the asterisk $*$ to denote this particular reaction channel.) Reference lines were obtained by detecting elastically scattered τ and d ; the special feature of the experiment was to select the τ as singly charged ions, which provides a near-by reference line for the tritons.

The sum of the Q values in the reaction chain can now be written in terms of the kinetic energies E_3 of the outgoing particles

$$Q(d, t) + Q(\tau, d) = [E_3(t) - E_3(\tau)] + [E_3(d^*) - E_3(d)] + \Delta E_R$$

so that

$$Q = -[Q(d, t) + Q(\tau, d)] - [M(t) - M(\tau)]$$

where E_R is a term of the order of 10 keV correcting for the differences in recoil energies. The experiment measured in three angles (52.5° , 62.5° and 72.5°) and for reasons of intensity the evaluation was based on transitions to excited nuclear states with precisely known energies. The dispersion of the spectrograph could be obtained from a standard calibration or (consistently) from the positions of particle groups corresponding to known excited states.

The energies must be corrected for energy loss in the target. This was determined for the τ particles by placing a position-sensitive detector in the focal plane of the spectrograph. The particles were scattered through 92.5° from a thin gold layer covering the carbon-backed dysprosium target at 45° to the beam. The target was rotated between reflection and transmission geometry, where in the second case the τ particles traverse the C-Dy layers twice. The energy losses for deuterons and tritons were calculated from the measured τ losses.

For the singly-charged τ particle one must also correct for the energy needed to pick up an electron from the target. This energy was estimated by measuring elastically scattered τ in the 7.5° gap with alternatively the Dy and the C sides facing the detector. As the mean electron-loss path is very short, the shift between the two target positions gives the difference in pick-up energy between Dy and C, which came out to be 0.9 ± 0.1 keV, which was used as our (conservative) correction. The additional energy involved could approach the K binding energy of carbon (285 eV).

Our preliminary value of the Q of the ^{163}Ho EC decay is 2.3 ± 1.0 keV. The ultimate accuracy in the experiment (2 days) due to statistics alone would be about 0.3 keV. This value is consistent with the one of 2.58 ± 0.10 keV deduced from the EC phase-space factor (with $m_\nu c^2 = 0$). Together they imply an upper limit of 1.3 keV on the electron neutrino rest mass (see fig. 2), a limit which certainly can be improved further.

Finally, the prospects for IBEC experiments with ^{163}Ho should be considered. We recall that the predicted [1] very large enhancements of the internal bremsstrahlung arise when the Q value lies close to resonance points corresponding to the atomic s-electron binding energies. The existence of these large enhancements has just been demonstrated for the case of ^{193}Pt [14]. For a neutrino mass assumed to be negligible relative to our experimental errors, the Q value measured in electron capture differs from the $3S_{1/2}$ binding energy by a mere 500 eV, leading to a very large predicted enhancement of IBEC end-point P-capture rates. The isotope ^{163}Ho would thus be the best case known, although better ones might still be found. In addition, the element holmium offers a number of nuclear and chemical advantages already alluded to above. We are therefore going ahead with developing experiments based on

coincidence counting and calorimetry, but in view of the low energies involved, it is yet too early to try to predict whether eventually we shall be able to match or surpass the sensitivity obtained in the celebrated experiments on ^3H beta decay [3] that constrain the mass of the electron antineutrino.

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FIGURE CAPTIONS

Fig. 1 : Energy spectrum of 3.5 MeV alpha particles scattered from the centre of the foil B through 170° (see section 3.2). Surface impurities, such as holmium, appear as clearly resolved peaks, while bulk impurities give rise to a step function which reflects the energy loss of the alpha particle inside the target. The energy scale was calibrated by scattering the beam from a thin target of gold. The inset shows that the high-energy region is dominated by a peak at mass 164 ± 2 with weaker peaks around 183 and 203, presumably Ho, Ta/Re and Au/Pb, respectively. The intensity of the holmium peak, integrated over the area of the foil, gives an independent check on the number of atoms in the source.

Fig. 2 : Plot of $\phi^{1/2}(M_I)$ versus Q for different assumptions on the neutrino mass. The cross hatched area corresponds to the limits set by the two present experiments and gives an upper limit $m_{\nu_e} c^2 < 1.3$ keV.

Fig. 1

Fig. 2