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THE CAPTURE RATIO N/M IN THE EC BETA DECAY OF ^{163}Ho

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Considerable interest is attached to the nuclear properties of the isotope ^{163}Ho owing to its possible application in experiments to determine the mass of the electron neutrino. From recent experiments we now know the partial M-capture half-life [1,2] $T^{M}_{1/2}$, the physical half-life [3] $T_{1/2}$ and have a direct but inaccurate value [1] of 2.3 ± 1.0 keV for Q_{EC}, the beta decay energy. The M x-ray spectrum of ^{163}Ho has also been measured [2,4]. We wish to report the determination of the N/M capture ratio and the re-determination of $T^{M}_{1/2}$ by total-absorption spectrometry.

Internal counting of radioactivity in gaseous or solid detector systems is a widely used technique [5] for the study of electron capture processes. The novel feature here is to dope a silicon semiconductor detector with radioactive holmium. Such a detector offers a combination of good energy resolution, little noise at low energy and, because of the low energies involved in the ^{163}Ho case, essentially 100% containment of the radiations. Our long-range objective is to measure the neutrino mass through the shape of the energy spectrum near the upper end-point. The latter application is similar to the experiment with a tritium-implanted Si(Li) detector carried out by Simpson [6]; the advantage of the Ho experiment would be in the M,N peaks that would make the spectrum self-calibrating (cf. Dixon et al.[7]) and in the enhanced intensity at the end-point. Theoretical estimates of the counting rates in a neutrino experiment based on ^{163}Ho have been carried out by DeRǔjula and Lusignoli [8].

The first major obstacle to an experiment of this kind is immediately clear from Fig. 1: An adequate concentration of approximately 10^{16} Ho atoms/cm^3 exceeds the donor level of silicon by almost five orders of magnitude. Fortunately, a test with implanted stable holmium showed little change in the electrical properties of the silicon so that the electrical activity of holmium is small. The experiments were therefore continued with radioactive holmium.

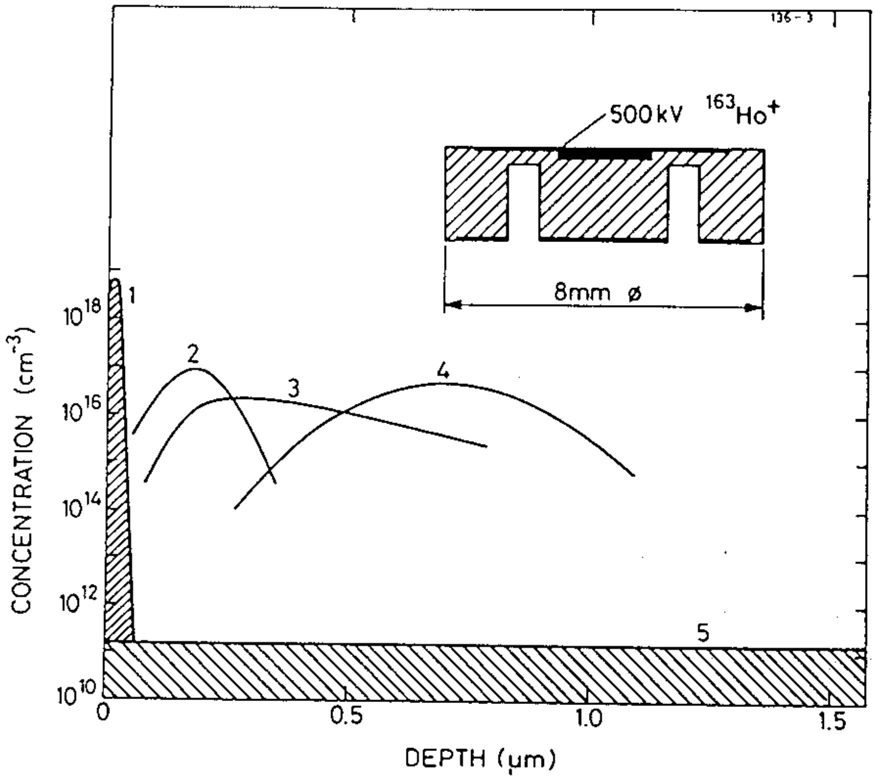

Fig. 1. The radioactivity ^{163}Ho was imbedded by ion implantation as a 3 mm dia spot in the inner part of a guard-ring detector (inset) made of intrinsic silicon. Contacts of n and p type were made by implanting boron (5 keV, curve 1) and phosphorus (50 keV) and by covering the surfaces with conducting lacquer. The curves 2 and 3 show concentrations of ^{163}Ho as a function of depth, for implantations at 500 keV without and with channeling, respectively. They are based on measurements with stable holmium. Curve 4 is the calculated distribution for a 2 MeV implantation in a random direction. The line 5 is the donor concentration of the silicon.

The ^{163}Ho was prepared by neutron irradation of ^{162}Er in the reactor at KfZ Rossendorf and was subsequently purified by repeated ion-exchange chromatography steps [9]. It was introduced into the silicon crystal (Fig.1) by means of the 600 kV and 2 MV ion-implantation units at Aarhus University. All data shown in the following are for implantations at 500 kV and along a channeling direction (i.e. a crystal axis). The use of channeling serves in two ways. The associated longer range reduces the holmium concentration, and at the same time the radiation damage to the crystal is smaller. After the contact layers had been implanted, the crystal was annealed at 300° C for 1/2 hour. It was operated at liquid nitrogen temperature.

In order to obtain a good resolution at low energy, the detection system used an integration time of 50 ns and a base-line restoration circuit. It was found that a coupling between the outer and the inner detector made high-energy events in the outer detector (from background) register as pulses with opposite polarity in the inner detector, and a small overshoot at the end of the pulse appeared near our expected N-capture events. This overshoot was suppressed by introducing a dead time of 1 ms after each count, a measure, which also helped to eliminate pulse trains from noise bursts. The live time of the detection system was monitored by a pulser.

Representative spectra are shown in Fig. 2. The energy scale was in each case determined by an external ^{137}Cs x-ray source and a precision pulser. The early results, shown in the inset, were indicative of very incomplete charge collection, but the sharp drop at the end-point and the indication of a weak peak at the correct energy of 2.05 keV, the M1 binding the energy of dysprosium, showed that ^{163}Ho was being detected. For spectroscopy purposes this result was useless but encouraging.

It turned out that a treatment of a few minutes at liquid-nitrogen temperature and with forward bias changed the characteristics of our detectors, and led to the main spectrum Fig.2. The $M_{I,II}$ peak now is clearly resolved although with a half width of ~ 380 eV as compared with 75 eV for the pulser. The peak appears below the expected value of 2.05 keV, thus indicating that incomplete charge collection still influences our results. (At the same time this illustrates how the ^{163}Ho experiment is self-calibrating with respect to energy scale and resolution). At the low-energy end of the spectrum the $N_{I,II}$ line of only 419 eV also appears.

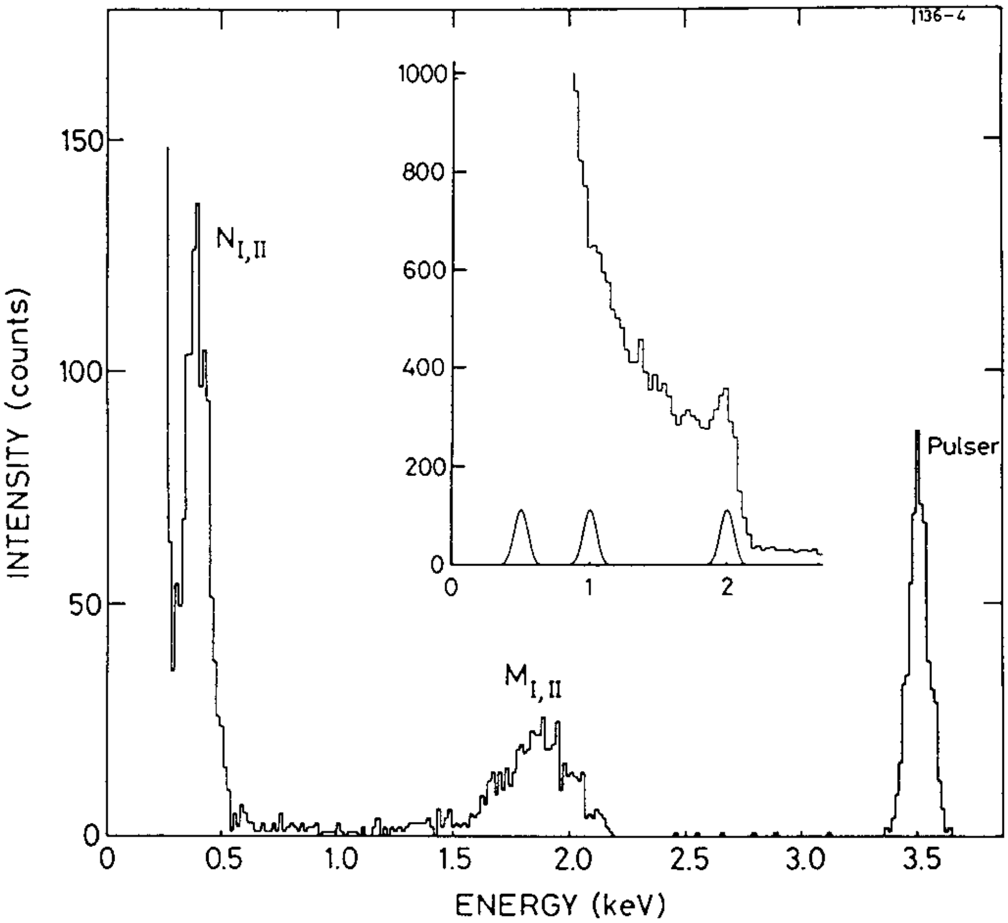

Fig. 2. The raw energy spectrum of implanted ^{163}*Ho before (inset) and after drifting with a forward bias. The pulser peak at 3.5 keV illustrates that the intrinsic electronic resolution of our system is 75 eV while the* $M_I + M_{II}$ *peak has a resolution of 380 eV. Since the calibration was carried out with the pulser and an external source it is significant that the* M_I *peak is below its correct position of 2.05 keV corresponding to* $|E(M_I)|$*. The measuring time was 15 hours.*

The amelioration of the counter brought about by forward bias was not permanent. A second spectrum taken on the day after the treatment showed the same peaks, but the M_I peak now had broadened to ~500 eV although the top part still remained in the same place. Over the following days the deterioration continued back towards spectra resembling that in the inset. The forward bias treatment could be repeated with identical results. A second counter gave similar results to Fig. 2, but in this case the noise level was too high to permit the resolution of the N line, The results of the 2 MV implantations were less satisfactory.

The experiments demonstrate that a holmium-doped silicon detector can be made and that the determination of the electron-neutrino mass by total-absorption techniques is, in principle, possible. At the present stage the technique is, however, too delicate for a direct attempt. We believe that a study of the basic problems of this detection method is essential in order to improve the resolution and longevity of the detector. We report now the new physics results that have already been obtained.

From the spectrum presented in the main part of Fig. 2 an N/M capture ratio of 2.4 ± 0.3 is obtained. The error limit represents statistical and possible systematic errors. We use this result to deduce a precise value for Q_{EC}. The lines in Fig. 2 must each represent a pair of screening doublets $M_I + M_{II}$ and $N_I + N_{II}$, with the second line ($p_{1/2}$) contributing approximately 5%. The capture rate to each sub-shell is proportional to atomic constants (tabulated by Bambynek et al. 5)) and to the neutrino phase-space factor $(Q_{EC} + E_x)^2$, where the (negative) quantity E_x is the energy of an electron in shell x of the daughter atom. A massless neutrino has been assumed. The exchange and overlap correction B(x) is poorly known; on the basis of tables 5) and some extrapolation we have obtained the estimated $B(K) = 0.993$, $B(L_I) = 1.032$, $B(M_I) = 1.080$, $B(N_I) = 1.183$ and $B(O_I) = 1.3$. The corrections for the $p_{1/2}$ states are less important and have been assumed to be unity.

From the measured value N/M = 2.4 ± 0.3 one obtains $Q_{EC} = 2.82 + 0.08/ - 0.06$ keV. In addition to the experimental error one must, however, also consider possible errors on the theoretical constants, for which there is limited experimental evidence for the M shell and essentially none for the N shell. If it is assumed, maybe optimistically, that this source of errors can be represented by a 10% error on the ratio of constants entering in the calculation, the decay energy of ^{163}Ho may be represented by the total error limits $Q_{EC} = 2.82 + 0.11/- 0.08$ keV. This analysis implies an O/N ratio of 0.204 almost independent of energy.

From monitoring of the ^{163}Ho beam during the implantation and from the measured M counting rate in the second detector a partial M-capture half-life of $T_{1/2}^{M}$ = 20 000 ± 3000 y was obtained, well below the previous measurements 40 000 ± 12 000 y (ref. 1) and 45 000 ± 15 000 y (ref. 2). It is maybe not surprising that the older results, based on absolute counting of ~1 keV ratiations from a solid source tend to underestimate the source strength. The total half-life of 4570 ± 50 y measured by Baisden et al. 3) and the shell ratios quoted above make it possible to arrive at an independent value for $T_{1/2}^{M} = T_{1/2} (1 + \frac{N}{M} (1 + \frac{O}{N}))$ = 18 000 ± 2000 y, which agrees neatly with our new measurement.

The Q_{EC} of ^{163}Ho was in our previous paper estimated in an accurate but indirect way, which combined the measured $T_{1/2}^{M}$ with a semi-empirical estimate of the nuclear transition matrix element. The new measured value of $T_{1/2}^{M}$ of 20 000 ± 3000 years leads to Q_{EC} = 2.80 ± 0.07 keV, where only the experimental error is reflected in the error limits. From this we conclude that the half-life 3) of ^{163}Ho, the N/M ratio and $T_{1/2}^{M}$ reported here and the matrix element calculated 1) by applying corrections for pairing to data from ^{161}Ho all form part of an internally consistent set. (We take the opportunity to point out that the pairing corrections were quoted in the wrong order in ref. 1. Calculated by Drs. Bengtsson and Ragnarsson, to whom our best thanks are once again due, the quantity $u_p^2 \, u_n^2$ was 0.508 for ^{161}Ho and 0.354 for ^{163}Ho. The analysis of ref. 1 remains unchanged.)

Our analysis leans heavily on calculations of M, N, O capture, processes rarely observed and very sensitive to the finer details of the atomic wave functions. (For another observation of N capture see Pengra et al. 10)). The consistency of all data, however, gives the confidence that our measured Q_{EC} = 2.82 + 0.11/- 0.08 keV is a good representation of the true value. It is low enough for ^{163}Ho to retain its place as the most interesting of the candidates for neutrino-mass experiments, but it is not as near to the M_I resonance point (2.05 keV) as one would have liked. The experiment has further demonstrated that a neutrino-mass determination by total-absorption spectrometry ("calorimetry") on ^{163}Ho is, in principle, feasible.

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