THE ^{3}H–^{3}He ATOMIC MASS DIFFERENCE FROM MASS SPECTROMETRY REQUIRES NO CORRECTION FOR THE METASTABLE $2\,^2S_{1/2}$ STATE OF ^{3}He$^+$

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Several direct measurements of the ^{3}H–^{3}He mass difference have been performed by mass spectrometric techniques. It has been suggested that, if a small fraction of the ^{3}He$^+$ ions were in the metastable excited state $2\,^2S_{1/2}$, the mass difference as determined by mass spectrometry would be shifted to a lower value than those from complementary techniques. It is shown here that, for all of the mass spectrometric determinations carried out so far, the effect of such a contamination is negligible.

An accurate value of the atomic mass difference ^{3}H–^{3}He serves as a point of reference in the determination of a possible mass for the electron neutrino from the β- decay spectrum of tritium near its end point. Accordingly there has been a resurgence in interest in precise determinations of both the β-decay spectrum [1–10] and the atomic mass difference from mass spectrometric doublets [11–15].

The data available from both β-decay studies and mass spectrometric measurements have been reviewed recently by Audi, Graham and Geiger [16] and are shown graphically in fig. 1. In order to derive the β-decay end points, various corrections have been made to account for atomic effects associated with both the parent and daughter nuclides. Simpson, Dixon and Storey recommend that earlier determinations by their groups [6,7] be superseded by the later value [10]. When this is done, and the remaining values are taken to be the corrected values of Audi et al. [16] as given in fig. 1, the weighted average of the β-decay end points is 18 588.5 ± 8.6 eV.

The recent mass spectroscopic values shown in fig. 1 are sufficiently precise that they outweigh other previous doublet measurements. We show here values derived from measurements made with the rf mass spectrometer of Smith [11,12] and three recent determinations made with the Fourier transform ion cyclotron resonance (FT–ICR) apparatus [13–15].

Amongst the mass spectrometric determinations, the FT–ICR value of Nikolaev et al. [14] was excluded by Audi et al. [16] because a systematic lowering of the doublet spacing was observed with low ^{3}He/^{3}H ratio and with long ionization times. The origin of these effects is not clear and a correction could not be estimated. This then yields a weighted average of mass spectroscopic values of 18 598.0 ± 2.3 eV. When the value of Nikolaev et al. is included, a weighted value of all mass spectroscopic values of 18 596.1 ± 3.4 eV is obtained.

In the case of the rf mass spectrometer, the mass difference in question was calculated from the values for three doublets, viz.,

$$a = {}^1H^2H - {}^3He,$$

$$b = {}^2H_2 - {}^1H^3H,$$

$$c = {}^1H_2 - {}^2H,$$

so that

$$^3H - {}^3He = a - b - c.$$

When the weighted average of the three available values [17] for "c" is used, the values given in fig. 1 are obtained [16]. The first of these two values [11] was determined without considering the possibility that, in doublet "a", an appreciable fraction of the He$^+$ in
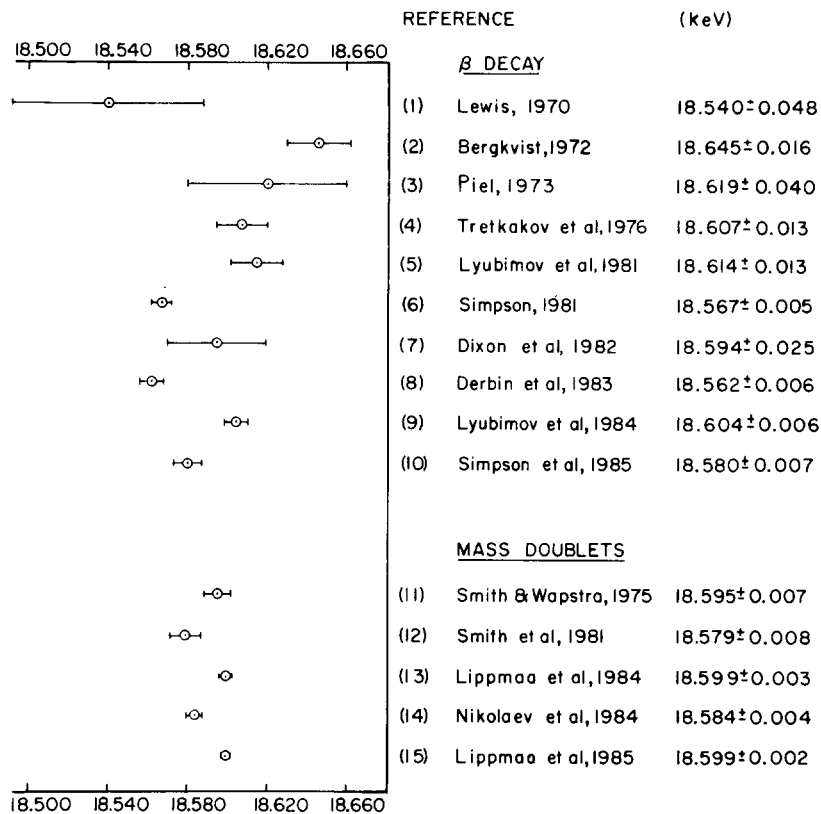
	REFERENCE	(keV)
	β DECAY	
(1)	Lewis, 1970	18.540±0.048
(2)	Bergkvist, 1972	18.645±0.016
(3)	Piel, 1973	18.619±0.040
(4)	Tretkakov et al, 1976	18.607±0.013
(5)	Lyubimov et al, 1981	18.614±0.013
(6)	Simpson, 1981	18.567±0.005
(7)	Dixon et al, 1982	18.594±0.025
(8)	Derbin et al, 1983	18.562±0.006
(9)	Lyubimov et al, 1984	18.604±0.006
(10)	Simpson et al, 1985	18.580±0.007
	MASS DOUBLETS	
(11)	Smith & Wapstra, 1975	18.595±0.007
(12)	Smith et al, 1981	18.579±0.008
(13)	Lippmaa et al, 1984	18.599±0.003
(14)	Nikolaev et al, 1984	18.584±0.004
(15)	Lippmaa et al, 1985	18.599±0.002

Fig. 1. Summary of the values for the atomic mass difference $^3H-^3He$ in keV. The corrections recommended by Audi et al. [16] for atomic effects have been included here; the uncertainties are as given in the original papers. Simpson, Dixon and Storey [10] recommend that their value replace earlier work in which they were involved [6,7].

the first doublet might be in the excited metastable $2^2S_{1/2}$ state. This possibility was raised by Bergkvist [18] in order to account for the discrepancy between his value [2] and that of Smith [11,18]. Subsequently the second value [12] was measured with this in mind; the electron energy in the ion source was maintained sufficiently low that this condition could not occur. The reservations about these measurements appear to have persisted to the present.

The concern is based on the fact that the $2^2S_{1/2}$ excited state of the isolated He$^+$ ion lies 40.8 eV above the ion ground state, and, as a result of a parity selection rule [19–21], has a relatively long lifetime of 1.92 ms. This means that the excitation energy (increased mass) is appreciable relative to the precision with which the doublet is determined, and the lifetime of the state is appreciable compared to the tran-

sit time of ions through the instrument. We will show, however, that the $2^2S_{1/2}$ state of He$^+$ does not affect the mass spectrometric determinations.

The essential point is that the presence of an electric field mixes the $2^2S_{1/2}$ state and the nearby $2^2P_{1/2}$ state via the Stark effect. Since the $2^2P_{1/2} \rightarrow 1^2S_{1/2}$ transition is dipole allowed, the mixing has the effect of quenching the $2^2S_{1/2}$ state. Magnetic fields also cause quenching via the motional electric field seen in the rest frame of the moving ion. The relation rate of the He ion in the presence of field E is [21]

$$\tau^{-1} = \tau_{2S}^{-1} + b^2 \, \tau_{2P}^{-1} , \tag{1}$$

where $\tau_{2S} = 1.92$ ms, $\tau_{2P} = 0.097$ ns and $b = 7.87 \times 10^{-5} E$, where E is expressed in V/cm.

In the rf mass spectrometer of Smith [18], as in

conventional mass spectrometers, ions are produced by electron impact and are then extracted from the source region and accelerated to an energy of 25 keV over a distance of the order of 2.5 cm. The lifetime of the excited ion state is reduced to 1.61×10^{-10} s by this accelerating field. Since the transit time for this region is approximately 40 ns, only 10^{-109} of the initial abundance of He^+ ions produced in the excited state will survive extraction. A similar degree of quenching occurs as the ions pass through the two subsequent electrostatic analysers prior to entering the region in which mass analysis takes place.

In the ion cyclotron resonance mass spectrometer the ions may be made *in situ* rather than in a separate ion source. The measurement, however, typically begins more than 1 s after ion production[15] and so, even ignoring the quenching effect of the magnetic field, less than 10^{-230} of the original excited He^+ ions will survive until the beginning of the measurement time interval. Thus the effects seen in the experiment of Nikolaev et al. [14] do not originate from this mechanism.

In all measurements reported to date, whether from FT-ICR, rf or conventional deflection mass spectrometers, the population of the excited state of He^+ is completely negligible. There will be no observable influence on the $^3H–^3He$ atomic mass difference as determined by mass spectrometry. Accordingly, *all* of the values from the Smith rf mass spectrometer [11,12] should be used.

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