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Mass of ^{6}Li and the excitation energy of its 3.56-MeV state

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The excitation energy of the second excited state of ^{6}Li has been measured to be 3562.88±0.10 keV by comparing the energy of resonance fluorescence radiation from this state to calibration lines from a ^{56}Co source. Also the Q value of the ^{6}Li$(p,\alpha)^3$He reaction has been determined relative to the ^{19}F$(p,\alpha)^{16}$O reaction, giving a result for the ground state mass excess of ^{6}Li of 14 085.5±1.1 keV, somewhat below the tabulated value of 14 087.3±0.8 keV. These measurements improve the sensitivity of experiments searching for isovector parity violation in ^{6}Li.

[NUCLEAR REACTIONS ^{6}Li$(\gamma,\gamma)^6$Li, bremsstrahlung source, E_x=3.56 MeV, measured E_x. Ge(Li) detector. ^{6}Li$(p,\alpha)^3$He, E_p=10.5 MeV, measured Q. Magnetic spectrograph.]

I. INTRODUCTION

The 0^+, $T=1$ state of ^{6}Li at 3.56 MeV is energetically able to decay into a deuteron and an alpha particle but is forbidden to do so by conservation of parity and of isospin. A measurement of the decay probability therefore gives a measure specifically of the $\Delta T=1$ part of the parity-nonconserving interaction. The ^{6}Li case has acquired new significance with the emergence of unified gauge theories of the weak and electromagnetic interactions, which imply the existence of a hadronic weak neutral current. Isovector parity violation is suppressed in conventional descriptions of charged current interactions but is not necessarily suppressed if a hadronic weak neutral current exists.[1]

This decay is now being studied by a group at the Université de Montréal[2], and by a Michigan State University–Argonne National Laboratory–Chalk River Nuclear Laboratory collaboration.[3] In these experiments and several previous ones,[4] the excitation function of the inverse reaction ^{4}He$(d,\gamma)^6$Li or ^{2}H$(\alpha,\gamma)^6$Li is investigated in the region of the 0^+, $T=1$ state. The presence of parity violation would be indicated by a weak resonance superimposed on the direct capture continuum. A limit to the experimental sensitivity is imposed by the range of energies which must be searched over in order to be certain that the resonance is included. The major contribution to this uncertainty originates in the mass difference between the 0^+, $T=1$ state of ^{6}Li and ^{4}He + ^{2}H. A direct measurement of this difference to the desired accuracy does not appear to be practical, and in this paper we describe independent measurements of the excitation energy and of the ground state mass of ^{6}Li.

It may be useful at the outset to consider what precision is needed in this program of measurements. A lower limit to the search range in the parity-violation experiment is set by the energy spread and instability of the incident beam. Measurements at Chalk River[3] have demonstrated that, at 6.24 MeV, the energy appropriate for ^{2}H$(\alpha,\gamma)^6$Li via the 3.56-MeV state, a resolution close to 2 keV can be reliably achieved, with an instability less than 1 keV. (The natural and Doppler widths are smaller.) It can be shown that the optimum search range is approximately three times the resolution, or about 6 keV. If one wishes 95% confidence that the resonance lies within the range, then the combined standard deviation in beam energy and masses must be 1.5 keV or less. A beam energy determination to 1 keV is feasible, implying a similar accuracy requirement in the resonance energy. Because of the laboratory-to-center-of-mass kinematic conversion, the precision required in the mass excesses is about 0.3 keV. Neither the ground state mass of ^{6}Li nor the excitation energy of the 3.56-MeV state is known to this accuracy. Two recent measurements[5,6] establish the excitation energy to about 0.5 keV, but there is a serious discrepancy (almost 6 keV) between these results and another of lower precision.[7]

The ground state mass of ^{6}Li has not been determined by direct mass spectroscopic techniques, and its present value (with a quoted uncertainty of

0.8 keV) is derived from nuclear reaction Q-value measurements. Although many measurements influence the ^{6}Li mass in the adjustment carried out by Wapstra and Bos[8], in practice two are dominant by virtue of their small experimental uncertainties. One is the ^{6}Li$(p,\alpha)^3$He experiment of Collins, McKenzie, and Ramm[9], and the other is a series of Q-value determinations[10] made at Notre Dame University[10] which link ^{6}Li, ^{9}Be, and ^{8}Be to ^{4}He, ^{1}H, and ^{2}H. These two determinations of the ^{6}Li mass differ by[11] 3.6(20) keV, underscoring the need for a new measurement.

II. METHODS AND RESULTS

A. Excitation energy

The technique adopted for the measurement of the excitation energy was resonant scattering of gamma rays from a ^{6}Li target and comparison of their energy with standard lines from a ^{56}Co source. The experiment was performed with the University of Illinois superconducting microtron (MUSL-2). Bremsstrahlung, produced by 7 MeV electrons, irradiated a 10 g target of metallic Li enriched to 95.7% (atom) ^{6}Li. Fluorescent radiation was observed at a nominal scattering angle of 126° by a heavily shielded 55 cm^3 Ge(Li) detector.[12] A radiation hardener consisting of 3.2 cm of Pb and 1.9 cm of Cu, in that order from the target, was interposed between the target and the detector to enhance the ratio of photopeak counts to total background for the 3.56-MeV line. A ^{56}Co source was placed between the Cu and Pb, on the line of symmetry. As total count rates were often as high as 20 000 per second, fast-logic pileup rejection and digital gain stabilization were employed.

Data were recorded in five runs with various combinations of beam current, calibration source location, amplifier gain, and analog-to-digital converter (ADC) gain. A portion of the spectrum from the fifth run is shown in Fig. 1. The ^{6}Li fluorescence line lies very near the 3548 keV transition of ^{56}Co. The energy of the latter line has been recently redetermined (for this measurement) to high accuracy by Greenwood,[13] who finds E_γ = 3547.891(34) keV. This result is both in disagreement with and substantially more precise than the energy quoted in a recent compilation[14], 3548.14(10) keV, and we have adopted Greenwood's measurement.

Peak centroids were extracted using the program SAMPO[15] and converted to energies with a linear calibration based on the ^{56}Co energies of Greenwood, Helmer, and Gehrke,[16] and of Greenwood.[13] It was anticipated that over the small energy interval between the ^{56}Co 3548-keV line and the ^{6}Li resonance line that a linear calibration would be satisfactory, and indeed it proved to fit all the lines of ^{56}Co above 1 MeV extremely well. Small-scale differential nonlinearities are presumed to average out in a series of runs taken with different ADC conversion gains and amplifier gains. The peak separations between the ^{56}Co 3458-keV line and the ^{6}Li line are summarized in Fig. 2, where the errors shown are statistical only. The χ_ν^2 of 3 indicates the presence of excess error, probably attributable to differential nonlinearity. An unweighted average gives for the doublet separation 12.619(70) keV, while an average weighted by the statistical errors gives 12.556(59) keV. The uncertainty in the former is the standard deviation of the mean of the data, and that in the latter is

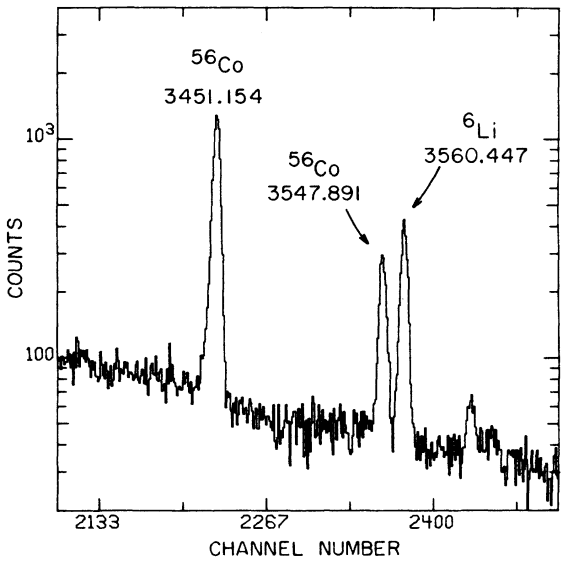

FIG. 1. Portion of γ-ray spectrum showing 3.56-MeV fluorescent line from ^{6}Li$(\gamma,\gamma)^6$Li together with calibration lines from a ^{56}Co source.

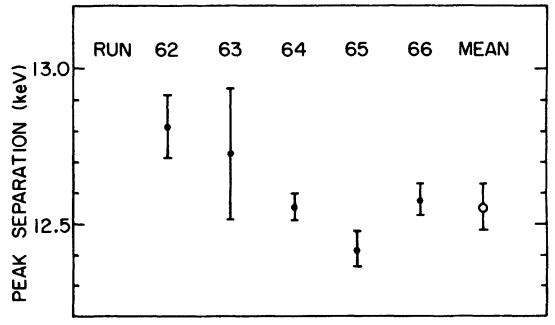

FIG. 2. Separations between ^{6}Li resonance line and the ^{56}Co 3547.891-keV calibration line, for five runs. The error bars represent statistical uncertainties only. The adopted mean, with its uncertainty, is also shown.

calculated from statistical errors increased by a factor of 2.0 to reduce χ_ν^2 to 1. We adopt the weighted average but increase the uncertainty to the larger value, i.e., 12.556(70) keV.

The possibility of a systematic error being introduced through the use of the peak-fitting program was checked by comparing its results with direct centroid determination under the assumption of a linear background. A possible systematic error of about 40 eV was inferred from this comparison.

The excitation energy of a state observed in resonance fluorescence at a scattering angle θ is greater than the measured γ-ray energy E_γ by an amount

$$(1-2\cos\theta)E_\gamma^2/2Mc^2,$$

where M is the mass of the nucleus. Determination of the angle θ is subject to a number of corrections and uncertainties. The finite solid angle subtended by the target led to a very small correction, $-0.08°$. Attenuation of the bremsstrahlung in the 1-cm thick scattering target shifts its effective center toward the production target, leading to a correction of $-0.15°$ in the scattering angle. Uncertainties in the geometrical measurement of the angle, the position of the beam spot, and the position of the Ge(Li) detector within its cryostat corresponded to 0.7°, 0.2°, and 0.7°, respectively. The corrected scattering angle is then 125.2(10) degrees. Combining the results and uncertainties, one finds for the excitation energy of the 0^+, $T=1$ state in ^{6}Li

$$E_x = 3562.88(10) \text{ keV}.$$

B. Ground state mass

A general technique for the precise determination of nuclear reaction Q values using a magnetic spectrograph has been described by Nolen, Hamilton, Kashy, and Proctor.[17] Reaction products from the reaction under investigation are recorded on nuclear emulsions simultaneously with suitably chosen calibration lines. Then, by a linearized least-squares fit, unknown parameters in the experiment, the beam energy, the reaction angle, and the spectrograph focal plane calibration can be determined. The method provides immunity to variation in the parameters which may occur during the course of the experiment, and at the same time obviates the need for detailed knowledge of the spectrograph saturation and hysteresis characteristics.

At a beam energy of 10.5 MeV and a reaction angle of about 10°, the ^{6}Li$(p,\alpha)^3$He reaction provides a good basis for a measurement of the mass of ^{6}Li. If a target of LiF is used, then the ^{19}F$(p,\alpha)^{16}$O reaction leads to excited states in ^{16}O with extremely accurately known Q values. These lines bracket the alpha particle group from ^{6}Li. Furthermore, a mixed isotope target containing both ^{6}Li and ^{7}Li provides a deuteron group from ^{7}Li$(p,d)^6$Li which shifts rapidly with beam energy, thus calibrating the beam energy effectively. Lines from ^{19}F(p,p') as well as ^{19}F(p,α) serve to establish the focal plane calibration. The reaction angle is fixed largely by the inclusion in the target backing material of a small amount of hydrogenous material (Formvar, approximately $C_5H_8O_2$) because the energy of protons scattered from hydrogen varies rapidly with angle. In addition, the ^{3}He group from the complementary reaction ^{6}Li$(p,^3\text{He})^4$He is also present on the plates, giving two (correlated) measurements of the ^{6}Li mass with each exposure.

Experiments were carried out with the Michigan State University (MSU) Cyclotron and Enge split-pole spectrograph. Proton beams of about 50 nA impinged on targets of ^{6,7}LiF evaporated onto backings consisting of 5 μg/cm^2 of carbon and 2.5 μg/cm^2 of Formvar. Two targets were used: a "thick" target of 38 μg/cm^2 LiF with an isotopic content of 96% ^{6}Li and a "thin" target of 3.1 μg/cm^2 LiF with equal isotopic contents of ^{6}Li and ^{7}Li. Reaction products entered the spectrograph through a polished brass slit 1° wide and 2° tall, and were detected in Kodak NTB-25 emulsions.

Four plate exposures were made, and each was scanned both by "hand" and with the MSU automated scanner.[18] The spectrum from the hand scan of Run 1497M is shown in Fig. 3. The numbered peaks are identified in Table I with the calibration energies assumed for them.[19] The discontinuities in the spectrum (for example, near peaks 8 and 11) are artifacts caused by incomplete scanning of un-

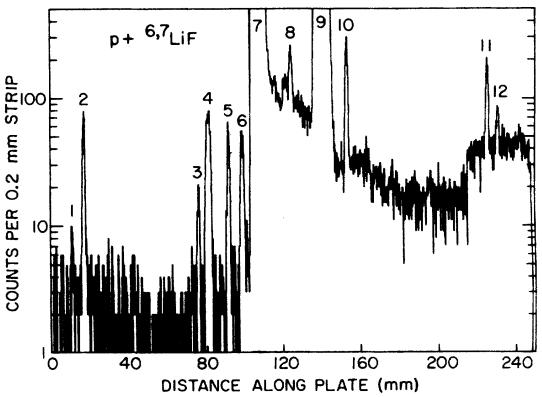

FIG. 3. Plate spectrum from proton bombardment of thin ^{6,7}LiF target. The numbered peaks are identified in Table I and the experimental parameters may be found in Table II.

TABLE I. Calibrations used in $^6Li(p,\alpha)$ experiment.

Peak number	Reaction	Final state [b]
1 [a]	$^{19}F(p,\alpha)^{16}O$	6.050
2	$^{19}F(p,\alpha)^{16}O$	6.130 43(5)
3	$^{19}F(p,\alpha)^{16}O$	6.917 1(6)
4	$^7Li(p,d)^6Li$	0.0
5	$^{19}F(p,\alpha)^{16}O$	7.116 85(14)
6	$^6Li(p,\alpha)^3He$	0.0
7 [a]	Elastic scatt.	0.0
8	$^{19}F(p,p')^{19}F$	0.197 24(19)
9	$^1H(p,p)^1H$	0.0
10	$^7Li(p,p')^7Li$	0.477 611(12)
11	$^6Li(p,^3He)^4He$	0.0
12	$^{19}F(p,p')^{19}F$	1.345 67(13)

[a] Not used as calibration.
[b] Data taken from Ref. 19.

interesting regions. The energy resolution obtained was 15–20 keV.

Peak centroids were calculated after subtraction of a linear background and were input into the least-squares fitting code DOALL.[20] The hydrogen peaks, broad and in some cases too intense to scan reliably near the centers, were analyzed using the well-defined edges as described elsewhere.[21] The parameters adjusted in the fit, weighted by combined statistical and calibration errors in the standard peaks, were the beam energy, reaction angle, and three coefficients in the focal plane calibration. The $^6Li(p,\alpha)$ and $^6Li(p,^3He)$ Q values were not constrained to be equal. On the 12° exposure (1498T), the $^6Li(p,\alpha)$ peak is unresolved from elastic protons and has therefore not been analyzed.

Since several different reactions are used in this technique, not all of them can be kinematically in focus at the same time. A compromise setting of the plate position which corresponded to the $^{19}F(p,\alpha)^{16}O$ focal plane was used. Centroid shifts for other groups were calculated to be negligible, with the exception of $^6Li(p,\alpha)^3He$, where the derived Q value had to be increased by 0.79 keV. The effect is large both because of the rapid variation of cross section with angle, +30% per degree at 11°, and the large kinematic shift of the $^6Li(p,\alpha)$ focal plane. A 0.30-keV uncertainty has been assigned to this correction.

The results are listed in Table II separately for the hand-scanned and machine-scanned spectra, along with relevant run parameters. The quantity ΔQ in the table is the experimental $^6Li(p,\alpha)^3He$ Q value minus the Q value 4020.0 keV, calculated from the 1977 mass table. The two scanning techniques are subject to different uncertainties. Human scanners may inadvertently omit or repeat frames, experience fatigue, and have difficulty with intense peaks. The automatic scanner, on the other hand, does not fatigue and has a smooth saturation characteristic but may misidentify blemishes on the plate as tracks or may reject valid tracks. A comparison of the two sets of results shows significant scatter at each point but overall averages in good agreement. The scatter may be represented as uniform, random, 1.3-keV uncertainties in each determination of the Q value (a figure which also, of course, includes much of the subjective error in spectrum analysis).

An important ingredient in the Q-value determination is the thickness and composition of the targets. The thickness of the thick target was measured to a precision of about 10% from the energy loss of 5.48-MeV alpha particles, and the thin one was then calibrated by comparing yields for the $^{19}F(p,\alpha)$ reaction. Although beam currents were relatively low in this experiment, other workers have reported composition changes in LiF targets after extensive bombardment. Spear, Switkowski, Kennedy, and Heggie[22] found significant Li enrichment at the surface during bombardment with 1.2-MeV alpha particles, an effect we may refer to as "differentiation." We have calculated the effect on our results of complete differentiation (i.e., segregation into separate layers of Li and F)

TABLE II. Summary of $^6Li(p,\alpha)^3He$ and $^6Li(p,^3He)^4He$ measurements.

Run	Target	Charge (μC)	Beam energy (MeV)	Lab angle (degrees)	Particle detected	ΔQ, keV Hand scan	ΔQ, keV Auto scan	Statistical uncertainty
1497T	Thick	200	10.499	10.16	α	−1.49	−1.46	1.4
					^3He	−3.75	−0.81	1.5
1497M	Thin	500	10.501	11.16	α	−0.60	−2.65	1.6
					^3He	−5.42	−3.03	1.7
1497B	Thick	200	10.503	11.15	α	−2.14	−2.52	1.4
					^3He	−1.85	+0.15	1.4
1498T	Thick	190	10.501	12.21	^3He	+1.50	−1.18	1.3

and have then taken 10% of this effect as an estimate of its magnitude. This estimate is expected to be conservative, because the data of Spear *et al.*, taken over much more intense bombardments, correspond to approximately a 7% differentiation effect for targets of our thickness. Contributions to the Q-value uncertainty of approximately 0.4 keV from differentiation effects and 1.0 keV from thickness uncertainties are introduced for the thick target. For the thin target, these effects are negligible.

Analysis of the uncertainties is rendered complex by the presence of correlations. Each of the seven determinations of the Q value depends on (a subgroup of) 50 statistically independent quantities, namely 10 peak centroids for each plate exposure, target thickness, the degree of target differentiation, the kinematic focus effect for the ^{6}Li(p,α) group, and scanning uncertainties. First order derivatives of the seven Q values with respect to the input quantities were calculated by finite difference techniques, and were used to set up a variance-covariance matrix in the usual way. Appropriate weights for combining the seven Q values are the sum of elements of rows of the inverse matrix, and the overall uncertainty in the average can then be obtained straightforwardly via propagation of the input uncertainties. The result is

$$\Delta Q = -1.8(11) \text{ keV},$$

which is equivalent to a ^{6}Li$(p,\alpha)^3$He Q value of 4018.2(11) keV.

Some indication of the relative importance of the individual sources of error may be obtained by setting each equal to zero in turn and evaluating the variance due to the remaining error sources. The square root of the difference between that variance and the actual variance then reflects the contribution of each error source. These contributions are centroid uncertainties, 0.80 keV; target thickness, 0.67 keV; target differentiation, 0.26 keV; kinematic focus, 0.10 keV; scanning, 0.65 keV. Of course, in the presence of correlations and the consequent readjustment of the weight factors, the total variance is not equal to the sum of the squares of the error contributions just listed.

III. DISCUSSION AND CONCLUSION

A comparison of the present measurement of the excitation energy of the 0^+, $T=1$ state with previous ones is shown in Fig. 4. As can be seen, there is good agreement with early work (summarized by Ajzenberg-Selove),[19] with the ^{6}Li(p,p') measurement of Nolen and Barker,[6] and with the ^{9}Be(p,α) experiment of Kim *et al.*[5] There is evidence that the result of Wessels *et al.*[7] should be rejected. The excitation energy is now sufficiently well known that it no longer constrains the parity violation experiments in their present form.

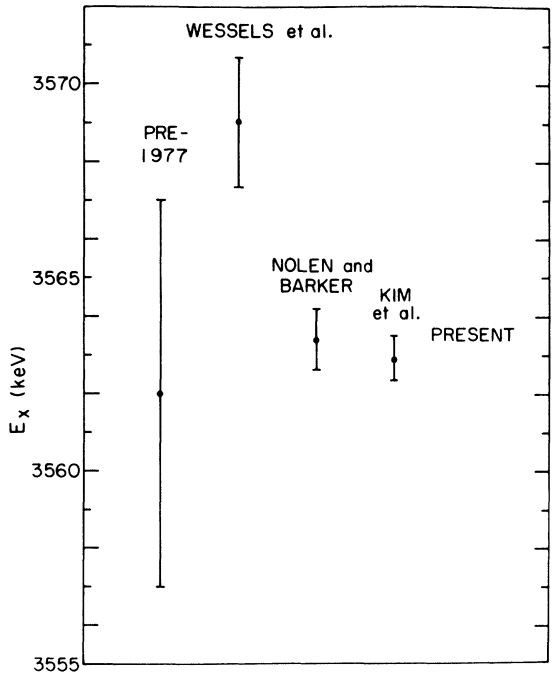

FIG. 4. Summary of measurements of excitation energy of 0^+, $T=1$ state of ^{6}Li. Sources of the previous data are, from left to right, Refs. 18, 7, 6, and 5.

Our result for the ^{6}Li$(p,\alpha)^3$He Q value, 4018.2(11) keV, may be compared directly with three previous measurements of this reaction, that of Sperduto and Buechner[23] at MIT, 4025(6) keV, Williamson *et al.*[24] at the University of Wisconsin, 4021(5) keV, and Collins, McKenzie, and Ramm[9] at Birmingham University, 4023(2) keV. There is reasonable agreement with the first two, but perhaps not with the last. The Q-value measurements for ^{9}Be$(p,d)^8$Be and ^{9}Be$(p,\alpha)^6$Li made at Notre Dame[10] may be combined to give a ^{6}Li$(p,\alpha)^3$He Q value of 4019.25(90) keV, in good agreement with the present result. Recently, Koets, Kramer, and Nonhebel[25] have performed an exploratory measurement of the D_3^+-^{6}Li$^+$ doublet in the Smith mass spectrometer at Delft.[26] While a number of possible sources of systematic error remain to be checked, they find a ^{6}Li mass excess 2.25 keV less than the 1977 mass table result, with an uncertainty of about 1.2 keV. The six independent results are displayed in Fig. 5, identified by laboratory. A weighted average of all six results yields $Q=4019.00(57)$, and a normalized χ_ν^2 of 1.5, while an average omitting the Birmingham re-

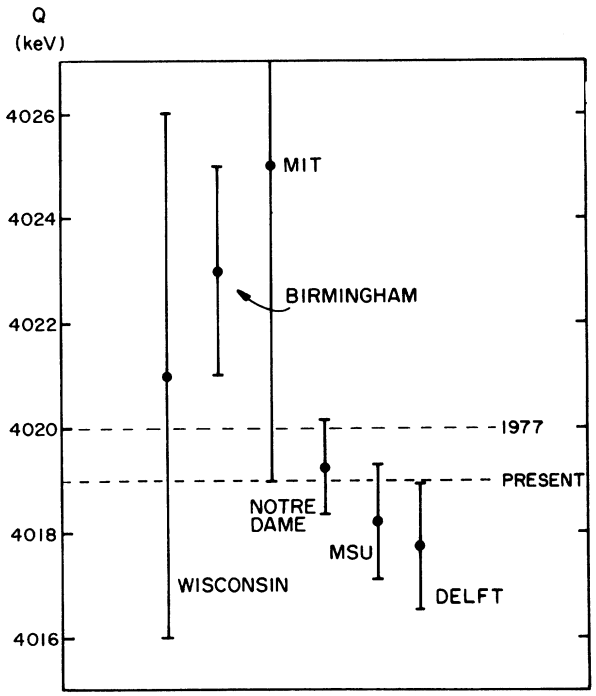

FIG. 5. Summary of existing data on the mass of ^{6}Li expressed in terms of the ^{6}Li$(p,\alpha)^3$He Q value.

sult gives $Q = 4018.64(60)$, with $\chi_\nu^2 = 0.6$. Although a complete reevaluation of the mass of ^{6}Li must await both confirmation of the Delft measurement and a new mass adjustment, the ^{6}Li$(p,\alpha)^3$He Q value can be converted to an interim ^{6}Li mass excess of 14086.2(6) keV, taking all six data points. The hoped-for precision of 0.3 keV in the mass of ^{6}Li has not been achieved, but the new results nevertheless reduce the search interval needed in the ^{2}H$(\alpha,\gamma)^6$Li parity-violation experiments by one-third. More significantly, the possibility of a gross error in the ^{6}Li mass seems now to be quite remote. The resonance is now expected to occur at a laboratory ^{4}He^{++} kinetic energy of 6238.4(18) keV, about 3 keV below the value based on the previously accepted ^{6}Li mass.

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