Precise Determination of the Mass Difference ^{76}Ge-^{76}Se and a Derived Upper Limit on the Mass for the Electron Neutrino

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Recent improvements to both the instrumentation and data analysis techniques associated with the Manitoba II high-resolution mass spectrometer have led to greater precision and accuracy. The mass difference between ^{76}Ge and ^{76}Se has been redetermined to be 2038.56(32) keV. When this new Q value is used to reexamine recent spectra reported by groups searching for the characteristic sharp peak expected for neutrinoless double-beta decay, no evidence for the occurrence of such a decay is found. New upper limits can be derived for the electron-neutrino mass.

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In 1984 and 1985, Ellis *et al.* reported a set of mass differences [1,2], determined with the Manitoba II instrument, that overdetermined the mass difference between ^{76}Ge and ^{76}Se. This mass difference is of great interest to the groups using Ge crystal detectors to search for the existence of neutrinoless double-beta decays [$\beta\beta(0\nu)$] in the decay of ^{76}Ge [3]. Should $\beta\beta(0\nu)$ decay be observed, it would indicate the presence of physics beyond the standard electroweak model and the minimal SU(5) particle model.

The signature of the $\beta\beta(0\nu)$ decay would be a sharp peak at the Q value in a spectrum of decay-product energies detected by the Ge crystal. There are two possible branches for this decay mode. The ^{76}Ge can decay from the ground state to the ground state of ^{76}Se (0^+-0^+) or it may decay from the ground state to the first excited state of ^{76}Se (0^+-2^+). For reasons described below, we are concerned only with the decay of ^{76}Ge by the 0^+-0^+ $\beta\beta(0\nu)$ branch in this work.

The Q value for the double-beta decay of ^{76}Ge given by Ellis *et al.* was 2040.71(52) keV [2]. Preliminary data from various groups using hyperpure Ge detectors in this energy region were compiled and discussed by Avignone *et al.* [3]. The presence of a peak about 4 keV above the Q value of Ellis *et al.* was noted and discussed. This was a worrisome development, as an incorrect Q value could have led to an erroneous conclusion regarding the presence of $\beta\beta(0\nu)$. On the basis of the data at the time, however, there was no evidence for $\beta\beta(0\nu)$ decay. Newer spectra, which will be discussed below, do not contain this anomalous peak.

Recently, some new measurements have cast doubt on the Q value determined by Ellis *et al.* [2]. Specifically, new (n,γ) Q values in Ge and Se [4–7] give values for double neutron separation energies (S_{2n}) that are not in agreement with values reported by Ellis *et al.* [2]. These new S_{2n} values differ by 3–6 keV from the values given by Ellis *et al.* In light of these new data and some recent instrumental improvements, we decided to undertake a remeasurement of the most important mass doublets related to the ^{76}Ge-^{76}Se mass difference.

The Manitoba II high-resolution mass spectrometer has been described extensively elsewhere [8]. This instrument can utilize two different methods in determining the coincidence of the two ion peaks, the "visual null" method and the "computer-assisted" technique. Both techniques have already been described extensively [9, 10]. It is important to note that the computer matching technique retains a permanent record of the raw data, allowing one to examine mass spectra for the presence of contaminants after the fact. This cannot be done with the visual technique as no raw data record is retained with this method. In the present work, only computer matching has been used.

Computer-assisted matches utilize eight different matching configurations to reduce the likelihood of systematic effects. These eight values are then averaged. Recent improvements to the instrument [11–13] have produced higher levels of precision for a single run than have been observed previously. Also significant for this work is the fact that the computer matching method has a higher signal-to-noise ratio than the visual technique, providing better sensitivity for the detection of contaminant peaks.

The doublets studied involve peaks in the mass spectra of Ge and Se chlorides. These solid sample materials were either placed directly in the ionization region of the source or vaporized and introduced through a heated gas inlet.

In the work of Ellis *et al.* [1,2] it was appreciated that the presence of unresolved contaminant peaks originating from dimers of the source material itself, viz. $Se_2Cl_2^{2+}$, could potentially bias the observed values in an unpredictable way. It was not known, however, that these contaminant peaks would be formed in a manner that was extremely sensitive to the operating conditions of the ion source. Moreover, the presence of such contaminants, even at levels that are imperceptible when using the real-time display, will bias results by amounts that exceed the stated errors for the mass difference of primary interest.

Very small levels of unresolved contaminant peaks can result in significant bias to the value for the mass

TABLE I. New values for doublets measured in this work are the input values for *A*, *B*, *C*, *D*, *E*, and *F*. The input value for *g* was taken from the output data of the 1986 atomic mass predictions, Ref. [14] (see text). Output values are the results of a standard least-squares evaluation to derive "best" values. Because doublet *F* is not involved in the least-squares adjustment, no output or χ^2 value exists.

Code	Doublet	Input (μu)	Output (μu)	χ^2
A	^{76}Ge-^{76}Se	2188.60(42)	2188.48(34)	0.08575
B	^{78}Se^{35}Cl-^{76}Ge^{37}Cl	1143.57(72)	1143.82(45)	0.11891
C	^{78}Se^{35}Cl-^{76}Se^{37}Cl	1044.58(45)	1044.66(39)	0.03061
D	^{76}Se^{35}Cl-^{74}Ge^{37}Cl	986.30(65)	986.17(42)	0.04019
E	^{76}Ge^{35}Cl-^{74}Ge^{37}Cl	3174.61(41)	3174.65(36)	0.00801
F	^{74}Ge^{35}Cl-^{72}Ge^{37}Cl	2052.01(26)	...	...
g	^{78}Se^{35}Cl$_2$-^{74}Ge^{37}Cl$_2$	2030.40(218)	2030.83(57)	0.03862

difference under study. For example, a contaminant-ion peak of only $\frac{1}{40}$ the size of a peak of interest, but close enough to be unresolvable, can shift the mass difference by 3–6 keV in this mass region. If the peak of interest is assumed to have 200 counts at maximum, the contaminant would have only 5 counts maximum. This can be observed by ion counting in the computer-assisted matching system used on Manitoba II. Such a contaminant would not be observable if the visual matching technique were used.

Unfortunately, all of the measurements reported by Ellis *et al.* [1,2] were made by the visual null technique of peak matching. In retrospect, it is not possible to determine whether these measurements were affected by contaminant ions, although we now believe that this was likely. The fact that production of dimers is highly sensitive to source temperature, coupled with the fact that amounts of contaminant, which are virtually invisible when visual null matching is used, can bias the results, prompts us to recommend that the values of Ellis *et al.* be replaced with the new ones presented here.

In the case of doublets whose members are chemically identical, such as ^{78}Se^{35}Cl$^+$-^{76}Se^{37}Cl$^+$, the related dimer contaminants (e.g., ^{78}Se^{76}Se^{35}Cl^{37}Cl^{2+}) may also be present. It was found that formation of these dimers was highly dependent upon source temperature, their prevalence increasing dramatically with increasing temperatures. Accordingly, the ion source was kept as cool as possible, by running at low filament currents and by reducing the power dissipated in the ion source. In this manner, dimer production was greatly reduced. Further, the absence of such dimers was confirmed by searching the mass spectrum in the neighborhood of the two doublet peaks for a clear signature of ions corresponding to the suspected contaminant. For the example of ^{78}Se^{35}Cl$^+$-^{76}Se^{37}Cl$^+$, the presence of dimers may be monitored by a search for ^{78}Se^{76}Se^{35}Cl^{37}Cl^{2+}, which will occur exactly between the peaks of interest. If none of that species is seen, it is then known that no other dimer of the same chemical form is present. For all measurements reported in this work, no contaminant peaks were seen which exceeded background levels. Typically the maximum number of counts in a peak would be 200, while background is usually less than 2 counts. This means that the maximum shift in the measured mass difference produced by such contaminants would be less than 0.2 keV.

For chemically dissimilar doublets, such as ^{78}Se^{35}Cl$^+$-^{76}Ge^{37}Cl$^+$, a different approach was necessary. In this case, the masses of all possible dimers were calculated. The instrumental resolving powers required to separate these contaminants from the ions of interest were estimated. All data were then accumulated, with the instrument operating with at least this resolving power. In cases where the dimer could not be separated sufficiently for direct observation, a related, more readily observable dimer was sought, so that the presence of the contaminant could be monitored. No such correction was required, as no contaminants were noted in any of the data accumulated and used in this work.

The doublets studied in this work, and the new values for these, are given in Table I. The mass differences are also shown schematically in Fig. 1. As is evident from the figure, the new data for *A*, *B*, *C*, *D*, and *E* overdetermine the desired mass differences. Accordingly, the method of least squares may be used to derive "best" values for these. This has been done, and the results are given as the "output" in the table.

The doublet labeled *g* in Fig. 1 is the mass difference ^{78}Se^{35}Cl$_2$-^{74}Ge^{37}Cl$_2$, taken from the 1986 mass table [14]. Because it carries a relatively large uncertainty, *g* serves as a weak constraint on this calculation. The value for *g* is also in good agreement with the new data and has little effect on the output values, although it accounts for much of the value of the total χ^2. The values of χ^2 in Table I show that the set of new data has a high internal consistency. The new datum *F* is not a part of the overdetermined set, and thus is not affected by the least-squares calculation.

The recent precise (n,γ) *Q* values yielding S_{2n} measurements in Ge and Se [4–7] are in excellent agreement with S_{2n} values derived from the new mass differences (see Table II).

Our mass difference for ^{76}Ge-^{76}Se, 2188.48(34) μu,

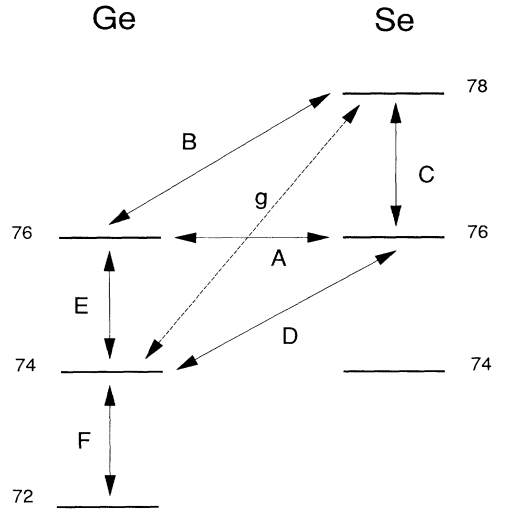

FIG. 1. Schematic diagram of mass differences measured in this work. The solid arrows represent differences using single chloride doublets (e.g., $^{78}Se^{35}Cl$-$^{76}Se^{37}Cl$) while the dashed arrow indicates a value taken from the output data of the 1986 atomic mass predictions, Ref. [14].

may be combined with the conversion factor 1 u $=931.49432(28)$ MeV, as given by Cohen and Taylor [15], to yield an energy of 2038.56(32) keV for the Q value of the 0^+-0^+ $\beta\beta(0\nu)$ decay.

It is interesting to examine the implications of this new Q value for two recent decay spectra, in order to establish limits on the half-life for the decay and on the neutrino mass. Accordingly, the spectra given by Vasenko *et al.* [16] and Caldwell *et al.* [17] were examined using a maximum-likelihood analysis, as suggested by Avignone *et al.* [3].

The spectrum given by Vasenko *et al.* [16] has a mean background of $m=3.58\pm0.28$ counts/keV. Using an enriched detector, their value for Nt was 1.64×10^{25} yr. The result of the analysis is a most probable number of counts of -14.2 with a width of the likelihood function of 4.7. Using the most conservative approach of Avignone *et al.*, we set his value c equal to the width and derive a half-life of

$$T^{0\nu}_{1/2}(0^+\text{-}0^+)\geq 2.4\times10^{24}\ \text{yr}\ (68\%\ \text{C.L.})\,. \qquad (1)$$

Similarly, the new data of Caldwell *et al.* [17] have been examined using the same technique. This detector has a value for Nt of 1.34×10^{25} yr. In this case, background is $m=19.8\pm0.5$ counts/keV. The result here is a most probable number of counts of -63.4 with a width of the likelihood function of 10.58. The lifetime is then calculated to be

$$T^{0\nu}_{1/2}(0^+\text{-}0^+)\geq 8.8\times10^{23}\ \text{yr}\ (68\%\ \text{C.L.})\,. \qquad (2)$$

As stated previously, the existence of neutrinoless double-beta decay would indicate that the electron neutrino is a Majorana particle. This decay may proceed if either the Majorana neutrino has mass or if the weak force contains right-handed terms. We may insert the preceding half-life limits for the $\beta\beta(0\nu)$ decay of ^{76}Ge into the expressions derived by Tomoda and Faessler [18] and Muto, Bender, and Klapdor [19]. These expressions lead to limits for the Majorana mass of the electron neutrino. The limits obtained neglect the possibility of right-handed coupling terms in the weak force. The theoretical expressions are of the form

$$\langle m_{\nu_e}\rangle \leq \{m_e^2(T^{0\nu}_{1/2})^{-1}/C^{(0)}_{mm}\}^{1/2}\,, \qquad (3)$$

where $T^{0\nu}_{1/2}$ is the half-life limit in years, $C^{(0)}_{mm}$ is a calculated value in $(\text{yr})^{-1}$, and m_e is the mass of the electron. All derived mass limits are to the 68%-confidence limit, and are given in Table III.

The limits which these two experiments place on the Majorana mass of the electron neutrino are among the lowest such values, and are consistent with other current estimates and limits. Kirsten *et al.*, for example, report $\langle m_{\nu_e}\rangle < 0.44$ eV [20], derived from measurements made on tellurium ores and examining the double-beta decay of ^{128}Te. One should note that the technique used to derive this limit is fundamentally different from that of direct observation used in the above Ge detector experiments.

As well, many other experiments have established limits on the mass of the electron nuetrino, irrespective of its Dirac or Majorana character. Among these works are those of Bowles *et al.*, reporting a limit derived from ^3H decay of $m_{\nu_e} < 13.4$ eV [21]. Both Bahcall and Glashow

TABLE II. S_{2n} values derived from the new mass-difference measurements in this work, from the mass-difference measurements of Ellis *et al.*, Ref. [2], and from new (n,γ) measurements.

Nuclide	S_{2n} (keV) This work	Ellis *et al.*	(n,γ)
^{74}Ge	16979.22(26)	16983.21(61)	16978.5(13) [a]
^{78}Se	17917.56(37)	17919.50(79)	17916.7(3) [b]
^{76}Ge	15933.53(34)	15939.74(69)	15933.9(10) [c]

[a]References [4,7].
[b]References [5,6].
[c]Reference [4].

TABLE III. Half-life limits for the 0^+-0^+ $\beta\beta(0\nu)$ decay of ^{76}Ge and associated theoretical limits for the Majorana mass of the electron neutrino. Both half-life and mass limits are to the 68% C.L.

Experiment	$T^{0\nu}_{1/2}$ (yr)	$\langle m_{\nu_e}\rangle$ limit [a] (eV)	$\langle m_{\nu_e}\rangle$ limit [b] (eV)
Vasenko *et al.* [16]	2.4×10^{24}	0.95	0.99
Caldwell *et al.* [17]	8.8×10^{23}	1.57	1.63

[a]Tomoda and Faessler [18].
[b]Muto, Bender, and Klapdor [19].

($m_{\nu_e} < 11$ eV) [22] and Kolb, Stebbins, and Turner ($m_{\nu_e} < 20$ eV) [23] give upper limits for the mass based on data from SN 1987A.

We should also note that the new value for the ^{76}Ge-to-^{76}Se difference should allow analysis of data related to the 0^+-2^+ $\beta\beta(0\nu)$ decay of ^{76}Ge. There have been several groups publishing data concerning this decay path, including Avignone *et al.* [24], Busto *et al.* [25] (Frejus Tunnel Collaboration), and Caldwell *et al.* [26]. The available data have poorer statistics than that for the 0^+-0^+ decay experiments and have recently revealed a potential problem.

The works of Busto *et al.* [25] and Morales *et al.* [27] have indicated the presence of a pair of peaks in their spectrum near the energy of interest. The first of these peaks may be ignored as it falls at an energy which precludes the events from being due to double-beta decay. The second peak occurs at 1480 keV, which, according to our new Q value, is the correct energy for the appearance of the 0^+-2^+ mode of $\beta\beta(0\nu)$ decay. However, Busto *et al.* have positively linked the presence of this peak to the decay of ^{214}Bi, a background contaminant. The possibility of such contamination must be considered by all groups doing this experiment before the data can be treated in an unambiguous fashion.

When our new Q value for the neutrinoless double-beta decay of ^{76}Ge, 2038.56(32) keV, is used to examine the most recent decay spectra of ^{76}Ge, there appears to be no evidence for either right-handed coupling terms in the weak force or a Majorana electron neutrino having a nonzero rest mass. It is anticipated that this new Q value will aid further analysis when more statistically significant data become available from the ultralow-background Ge detector experiments.

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