

Q value and half-life of double-electron capture in ^{184}Os

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^{184}Os has been excluded as a promising candidate for the search of neutrinoless double-electron capture. High-precision mass measurements with the Penning-trap mass spectrometer TRIGA-TRAP result in a marginal resonant enhancement with $\Delta = -8.89(58)$ keV excess energy to the 1322.152(22) keV 0^+ excited state in ^{184}W . State-of-the-art energy density functional calculations are applied for the evaluation of the nuclear matrix elements to the excited states predicting a strong suppression due to the large deformation of mother and daughter states. The half-life of the transition exceeds $T_{1/2}(^{184}\text{Os}) \geq 1.3 \times 10^{29}$ y for an effective neutrino mass of 1 eV.

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The observation of neutrinoless double-beta transitions would reveal physics beyond the Standard Model, as it would establish neutrinos to be Majorana particles, which implies a violation of the lepton number conservation. Experiments searching for these transitions have focused on the detection of neutrinoless double-beta decay ($0\nu\beta\beta$) rather than neutrinoless double-electron capture ($0\nu\epsilon\epsilon$). One reason among others is in general the significantly shorter half-life of the $0\nu\beta\beta$ process. However, in the case of neutrinoless double-electron capture, the transition is expected to be resonantly enhanced if the initial and the final state of the transition are degenerate in energy [1–3].

In this work, we investigate neutrinoless double-electron capture in ^{184}Os to various nuclear states. In particular, the transition to the nuclear excited 0^+ state with an energy of 1322.152(22) keV by a capture of two K-electrons was tagged as partially resonantly enhanced [1]. The capture of two K-electrons, the low spin of the nuclear excited state, and the large nuclear radius could result in a reasonably short half-life compared to other transitions even without being fully resonantly enhanced [1].

The capture rate of a $0\nu\epsilon\epsilon$ transition is given by [1, 3]

$$\lambda_{\epsilon\epsilon} = \frac{(\lambda G_F |V_{ud}|)^4}{(4\pi R)^2} |m_{\epsilon\epsilon}|^2 |M^{\epsilon\epsilon}|^2 P_{\epsilon\epsilon} \left(\frac{\Gamma_{2h}}{\Delta^2 + \Gamma_{2h}^2/4} \right), \quad (1)$$

where $\lambda = g_A/g_V = 1.2701(25)$ [4] is the ratio of the axial vector and the vector coupling constant, G_F the Fermi coupling constant, $|V_{ud}|$ the ud element of the

Cabibbo-Kobayashi-Maskawa (CKM) matrix, R the nuclear radius, $|m_{\epsilon\epsilon}|$ the effective electron neutrino mass, $|M^{\epsilon\epsilon}|$ the nuclear matrix element (NME) of the transition that consists dominantly of Fermi and Gamow-Teller components [5], $P_{\epsilon\epsilon} = |\Psi_{h1}|^2 |\Psi_{h2}|^2$ the absolute square of the two electron wave functions at the nucleus, Γ_{2h} the width of the two-electron hole state in the daughter nucleus, and Δ the excess energy. In order to estimate the capture rate, we have performed calculations of the NME by state-of-the-art energy density functional (EDF) calculations. These methods have been successfully applied to nuclear structure calculations (see ref. [6] for a review) and they have been recently implemented in calculations of neutrinoless double-beta decay and double-electron capture NMEs [5, 7]. In order to derive $\Delta = Q_{\epsilon\epsilon} - E_\gamma - B_{2h}$, we have experimentally determined the double-electron capture Q value $Q_{\epsilon\epsilon}$, which is the difference of the atomic masses of the mother and daughter nuclides. E_γ denotes the nuclear excitation energy of the final state, and B_{2h} is the excitation energy of the two-electron hole state in the daughter nucleus. Typically, the uncertainty of Δ is limited by the uncertainty of $Q_{\epsilon\epsilon}$. In the case of double-electron capture in ^{184}Os , the Q value of $Q_{\epsilon\epsilon} = 1451.2(1.6)$ keV [8] was only obtained indirectly from the masses of the other Os and W isotopes via (n,γ) -reactions. Q values taken from mass values in Ref. [8] can be rather inaccurate as the measurements in the references [9, 10] have revealed. Thus, for all potentially resonantly enhanced transitions, their Q values must be measured directly with Penning traps, which are presently the tool of choice [11].

The resonance condition of several promising transitions have recently been investigated with Penning traps [9, 10, 12–16], and a strong resonant enhancement was found for two nuclides, ^{152}Gd [12] and ^{156}Dy [13].

The half-life for double-electron capture in ^{184}Os has recently been estimated in ref. [1] using an empirical value for the NME, which corresponds to the maximum value obtained for medium-mass nuclei assuming spherical shapes for initial and final states [17]. However, mid-shell nuclei like ^{184}Os and ^{184}W are known to be significantly deformed [18]. As NMEs have been shown to be very sensitive to deformation [5, 7], we have calculated the NME using a method that consistently describes the deformation of the nuclear states involved. We have further recalculated the electron wave functions and atomic excitation energies in the daughter nuclide. Hence, we report about improvements of three crucial input parameters to the resonance enhancement factor, which allows us to check if the ^{184}Os and ^{184}W pair is a viable candidate to observe neutrinoless double-electron capture.

The Q -value measurement reported here was performed with the double-Penning-trap mass spectrometer TRIGA-TRAP [19]. The Q value is determined by measuring the ratio r of the cyclotron frequencies $\nu_c = qB/(2\pi M)$ of $^{184}\text{Os}^+$ to $^{184}\text{W}^+$ ions stored in a Penning trap with a magnetic field B :

$$Q_{\epsilon\epsilon}/c^2 = (M(^{184}\text{Os}) - m_e) \left(1 - \frac{\nu_c(^{184}\text{Os}^+)}{\nu_c(^{184}\text{W}^+)} \right), \quad (2)$$

where m_e denotes the electron mass. This method provides a direct measurement of the Q value. Note that the absolute atomic masses M of these two nuclides cannot be derived from such a measurement. However, the absolute masses of the two nuclides were determined within this work by measuring the ratio $r' = \nu_c(\text{C}_{15}^+)/\nu_c$ of the cyclotron frequencies of the carbon cluster ion $^{12}\text{C}_{15}^+$ and the investigated nuclide, in order to obtain the mass values via $M = (M_{\text{C}_{15}} - m_e)r' + m_e$.

A laser ablation ion source as described in ref. [20] was used to produce ^{184}Os and ^{184}W ions from solid samples of osmium and tungsten. Due to the low natural abundance of ^{184}Os (0.02%), an enriched sample with 1.5% abundance of ^{184}Os was used for the ion production. The ion pulses from the ion source were captured in the cylindrical Penning trap, called purification trap. There, the captured ion pulse was cleaned from unwanted ion species using a mass-selective buffer-gas cooling technique [21] with mass resolving power of about 3×10^4 . By using two different targets for ^{184}W and ^{184}Os , the presence of isobaric contaminants in the purification trap was avoided. A cooled monoisotopic ion bunch was transferred into the hyperbolic Penning trap, called precision trap, where the actual mass measurement took place. The cyclotron frequency of the stored ions was measured via the time-of-flight ion-cyclotron-resonance (TOF-ICR) method with a Ramsey excitation scheme [22] using two

excitation pulses of 200 ms and a waiting time of 1600 ms in between. A typical cyclotron resonance for $^{184}\text{Os}^+$ is shown in Fig. 1. The cyclotron frequency has been obtained from a fit of the theoretical line shape [23] to the experimental data.

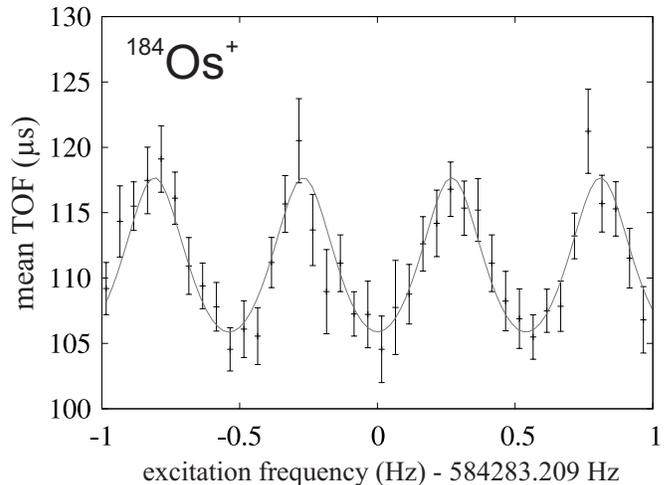


FIG. 1. Time-of-flight ion cyclotron resonance of $^{184}\text{Os}^+$ with about 450 ions. The data points show the mean time-of-flight values as function of the excitation frequency. The solid line is a fit of the theoretical line shape [23] to the data points.

In order to determine the Q value according to Eq. (2), the cyclotron-frequency ratio r of $^{184}\text{Os}^+$ to $^{184}\text{W}^+$ was determined in alternating measurements of the individual cyclotron frequencies (see Fig. 2). In the evaluation procedure for cyclotron frequency ratios, systematic uncertainties as described in detail in ref. [24] were considered. The systematic shift of the frequency ratio depending on the mass difference Δm between the two ion species used in the measurement is negligible ($\varepsilon_m/r < 10^{-11}$) in case of nuclides with the same mass number A , as in a Q -value measurement of a $0\nu\epsilon\epsilon$ process. For the measurement of the individual masses, this mass-dependent effect with a magnitude of $\varepsilon_m = 2.2(0.2) \times 10^{-9} \Delta m/\text{u}$ was considered. It arises from a possible slight misalignment between the electric and magnetic fields of the Penning trap [24].

The result of the cyclotron-frequency ratio measurement of $^{184}\text{Os}^+$ to $^{184}\text{W}^+$ and the Q value derived with

TABLE I. Results of the mass measurements. For each ion of interest the reference ion species is $^{12}\text{C}_{15}^+$. The cyclotron-frequency ratio r' , the mass excess $\text{ME} = M - A \text{ u}$, where u is the unified atomic mass unit, and the mass excess ME_{lit} listed in Ref. [8] are given.

Ion	r'	ME (keV)	ME_{lit} (keV)
$^{184}\text{Os}^+$	1.0219583764(67)	-44251.47(1.13)	-44256.60 (1.34)
$^{184}\text{W}^+$	1.0219496960(55)	-45705.40(0.94)	-45707.54 (0.87)
$^{12}\text{C}_{16}^+$	1.0666668659(70)	-0.62(1.25)	0 (0)

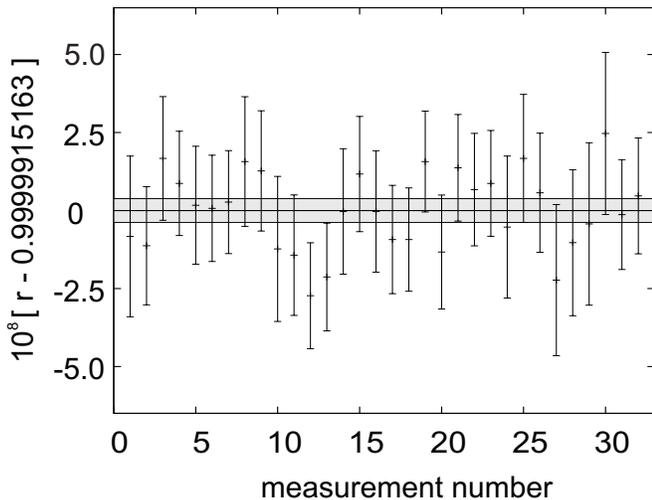


FIG. 2. The data points show the 32 cyclotron-frequency ratio measurements recorded to determine the double-electron capture Q value of ^{184}Os . The thick line represents the weighted-mean value and the gray area its 1σ -uncertainty.

Eq. (2) are $0.9999915163(38)$ and $1453.68(0.58)$ keV, respectively. The uncertainty of the literature Q value was improved by a factor of 3. A slight deviation below 1.5σ from our result to the value in Ref. [8] ($Q_{e\epsilon} = 1451.2(1.6)$ keV) was found.

The atomic mass excess values obtained from the cyclotron-frequency ratio measurements in this work are listed in Tab. I. Our measurement is a direct comparison to the atomic mass standard, unlike the values from Ref. [8], which show a deviation of $5.13(1.75)$ keV and $2.14(1.28)$ keV for ^{184}Os and ^{184}W , respectively. The Q value extracted from our values $\text{ME}(^{184}\text{Os})$ and $\text{ME}(^{184}\text{W})$ listed in Tab. I is $1453.93(1.47)$ keV and agrees well with our direct Q -value measurement.

In order to determine the excess energy Δ to the excited 0^+ state with $1322.152(22)$ keV energy in ^{184}W [25], the excitation energy of the two-electron hole state B_{2h} and its width Γ_{2h} have been calculated. The value of B_{2h} was obtained by applying the Dirac-Fock method [26], where frequency-dependent Breit interaction, quantum electrodynamics (QED) and correlation corrections were included. The calculations were performed for the Fermi model of the nuclear charge distribution (with $r_{\text{RMS}}=5.3670$ fm for ^{184}W [27]), resulting in $B_{2h}=140.418(12)$ keV. The value for the width of the autoionizing state of ^{184}W with two K-shell holes Γ_{2h} is approximated by using the double value of the width of the single K-level hole [1]. The single K-level hole width is taken from Ref. [28]. We estimate the uncertainty of Γ_{2h} by using the most conservative value in Ref. [28] and increase it by 2 eV to account for the uncertainty due to the hole-correlation effect suppressed approximately by a factor $1/Z$. The contribution of the width of the excited nuclear state has been neglected. This approach

resulted in $\Gamma_{2h} = 80(10)$ eV. Prior to this work, Δ was $-11.3(1.6)$ keV using Ref. [8] for the two mass values. The uncertainty given is obtained by adding the individual uncertainties of 1.3 keV and 0.9 keV of ^{184}Os and ^{184}W , respectively. Using our measured value, Δ becomes $-8.89(58)$ keV.

The evaluation of the NME with the EDF approach was performed with the Gogny D1M functional [29]. In our EDF approach the various initial and final nuclear states are given as superpositions of particle number and angular momentum projected Hartree-Fock-Bogoliubov states $|\Psi_{i/f}^I(\beta)\rangle$ with different quadrupole deformation β ($\beta < 0$, $\beta = 0$ and $\beta > 0$ represent oblate, spherical and prolate shapes respectively) [7]:

$$|I_{i/f}^{+\sigma}\rangle = \sum_{\beta} g_{i/f}^{I\sigma}(\beta) |\Psi_{i/f}^I(\beta)\rangle, \quad (3)$$

where $g_{i/f}^{I\sigma}(\beta)$ are the coefficients of the linear combination which are obtained by solving the corresponding Hill-Wheeler-Griffin (HWG) equations and σ labels the different states for a given angular momentum I [31]. From these coefficients, the so-called collective wave functions - the amplitude of the states for a given deformation - can be extracted [31]. Fig. 3 shows these amplitudes in the three lowest 0^+ states in ^{184}W and the ground state in ^{184}Os computed using the Gogny D1M functional. All these states are well prolate deformed. The two ground states have a similar distribution around $\beta \sim 0.25$; the first excited 0^+ state of ^{184}W correspond to a vibration of the deformed ground state while the 0_3^+ state has a larger prolate deformation $\beta' \sim 0.35$. These amplitudes will affect significantly the final value of the NMEs as we will analyze below. In Table II we show the excitation energy of the first excited 2^+ state $E(2_1^+)$ and its quadrupole transition probability $B(E2)$ to the ground state. We find larger excitation energies of the first 2_1^+ state and reduced quadrupole transition probabilities compared to the experimental values. Tab. II lists also the energies of the two lowest excited 0^+ states in ^{184}W . While the excitation energy of the 0_2^+ state has a fair agreement with data, the 0_3^+ excitation energy is too high, pointing to the need of including additional degrees of freedom. For example, the inclusion of pairing vibrations coupled to the quadrupole degree of freedom would lower the energies of the 0_2^+ , 0_3^+ states, bringing them closer to the experimental values. However, the quadrupole deformation of these states would not be very much affected [30].

The NMEs from the ^{184}Os ground state to the three different 0^+ states in ^{184}W are obtained in this framework by folding the deformation dependent wave functions with the Fermi and Gamow-Teller matrix elements expressed in the deformed basis [7]:

$$M^{\epsilon\epsilon}(0_{\sigma}^+) = \langle 0_{184\text{W}}^{+\sigma} | \hat{M}_{F/GT}^{\epsilon\epsilon} | 0_{184\text{Os}}^{+\sigma=1} \rangle, \quad (4)$$

	$E(0_2^+)$	$E(0_3^+)$	$E(2_1^+)[^{184}\text{Os}]$	$E(2_1^+)[^{184}\text{W}]$	$B(E2)[^{184}\text{Os}]$	$B(E2)[^{184}\text{W}]$	$M^{\epsilon\epsilon}(0_1^+)$	$M^{\epsilon\epsilon}(0_2^+)$	$M^{\epsilon\epsilon}(0_3^+)$	$M^{\epsilon\epsilon}(\text{Sph.})$
Expt. [32]	1.002	1.322	0.120	0.111	99.6	119.8	–	–	–	–
EDF D1M	1.135	3.169	0.172	0.155	135.0	146.6	0.631	0.504	0.163	8.177

TABLE II. Excitation energies (in MeV) of the first two 0^+ states for ^{184}W and of the 2_1^+ state and its quadrupole transition probability (in Weisskopf units) for ^{184}Os and ^{184}W are compared with EDF calculations based on the Gogny D1M functional. The values of the NMEs, $M^{\epsilon\epsilon}$, to the three 0^+ states in ^{184}W and the NME obtained assuming spherical shapes ($\beta = 0$) are also included. The experimental resonant state is written in boldface.

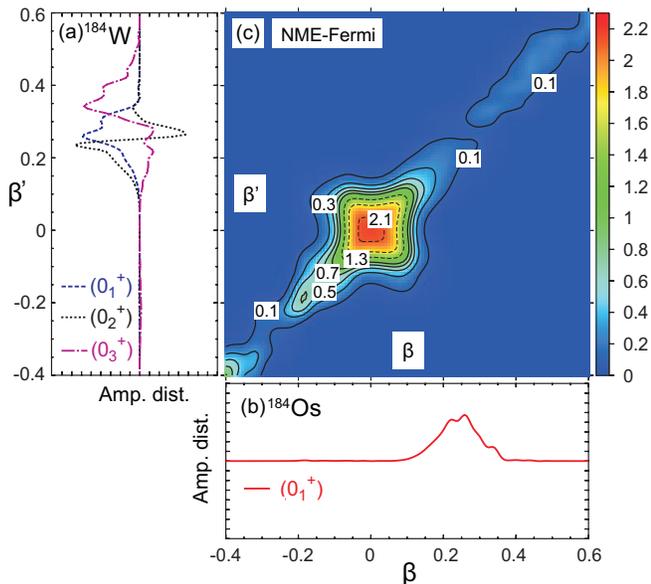


FIG. 3. (Color online) Deformation amplitudes for the three first 0^+ excited states of ^{184}W (a) and the ground state of ^{184}Os (b) calculated with EDF Gogny D1M. (c) Intensity of the Fermi component of the $0\nu\epsilon\epsilon$ NME as a function of the quadrupole deformations β and β' of the ^{184}Os and ^{184}W nuclei, respectively.

where $\hat{M}_{F/GT}^{\epsilon\epsilon}$ are two-body Fermi and Gamow-Teller transition operators. The results are listed in Table II. We obtain that the Gamow-Teller component contributes about 75% to the NME. For comparison we have also listed the NME obtained if we hypothetically assume spherical shapes for the initial and final nuclear states (i.e. $\beta = \beta' = 0$), obtaining values close to those of ref. [1]. However, if the deformation of the nuclear states is properly accounted for, the NMEs are reduced by about an order of magnitude for the ground state transitions and even more for the transitions to the excited states. To understand this behavior we have plotted in Fig. 3(c) the Fermi component of the NME as a function of the quadrupole deformations of the ^{184}Os (β) and ^{184}W (β') nuclei (the Gamow-Teller component shows the same general behavior):

$$M_{F/GT}^{\epsilon\epsilon}(\beta, \beta') =$$

$$\frac{\langle \Psi_{^{184}\text{W}}^{I=0}(\beta') | \hat{M}_{F/GT}^{\epsilon\epsilon} | \Psi_{^{184}\text{Os}}^{I=0}(\beta) \rangle}{\sqrt{\langle \Psi_{^{184}\text{W}}^{I=0}(\beta') | \Psi_{^{184}\text{W}}^{I=0}(\beta') \rangle \langle \Psi_{^{184}\text{Os}}^{I=0}(\beta) | \Psi_{^{184}\text{Os}}^{I=0}(\beta) \rangle}}. \quad (5)$$

We observe two general trends: a) for identical deformations of initial and final states (i.e. $\beta = \beta'$) the NME decreases with increasing quadrupole deformation; b) for fixed deformation β of the initial state the NME decreases with increasing difference between the deformations of initial and final states. We find significant and similar quadrupole deformations for both ground states, consistent with the large $B(E2)$ values observed experimentally. This explains why our NME values are so much reduced compared to the assumed value in ref. [1]. Although the first excited 0^+ state in ^{184}W has a similar deformation as the ground state, the corresponding NME is smaller. This is caused by the fact that the wave function of the first excited state has to be orthogonal to the one of the ground state implying a nodal structure which leads to cancellations when calculating the NME. In fact, we could interpret this state as a vibrational excitation of the ground state. The second excited 0^+ state is predicted by our calculation to have a noticeably larger deformation than the ground state. This results in a smaller NME than for similar deformations as obtained for the ground state due to the trends observed in Fig. 3(c). Even if our calculation fails to predict the correct deformation of the 0_3^+ state, the NME to this state should be smaller than the one to the ground state. If its deformation is different than the one of the ground state, the general trend b) predicts a smaller NME. If the deformation is similar, there should be strong cancellations in the NME caused by the orthogonality to the two lower 0^+ states. In this way, the NME for the capture to the ground state can be interpreted as an upper limit.

The half-life of the resonant neutrinoless double-electron capture in ^{184}Os is related to Eq. (1) by $T_{1/2} = \ln 2 / \lambda_{\epsilon\epsilon}$. Using $R = 5.382$ fm from [27], and $P_{\epsilon\epsilon} = 1.19617 \times 10^{33}$ eV⁶ (in natural units) determined by the one-configuration Dirac-Fock method for an extended nucleus [26] and the average of the two values for $M^{\epsilon\epsilon}(0_3^+)$ in Tab. II, we obtain $T_{1/2} = 2.0 \times 10^{30}$ y for an effective neutrino mass of 1 eV. If we take the ground state NME value as an upper limit (see above), we find a lower limit for the half-life of $T_{1/2} = 1.3 \times 10^{29}$ y.

In conclusion, we have investigated the resonance condition and the NME of double-electron capture in ^{184}Os

to three 0^+ states in ^{184}W . The Q value was determined in a direct measurement of the mass difference of ^{184}Os to ^{184}W to be 1453.68(0.58) keV with a factor of 2.7 smaller uncertainty than previously reported. We have computed the NME in a microscopic approach that consistently considers deformation degrees of freedom and found that initial and final nuclear states are significantly deformed. This deformation results in a suppression of the NME by a factor of about 10 compared to a calculation that is restricted to the spherical configuration. Combining our various improvements, a lower limit of the half-life for the $0\nu\epsilon\epsilon$ process of 1.3×10^{29} y to the excited 0^+ state with 1322.152(22) keV excitation energy in ^{184}W , normalized to an effective electron neutrino mass of 1 eV, has been obtained. Thus, ^{184}Os can be excluded as a promising candidate for the search for neutrinoless double-electron capture.

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