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# Double resonant enhancement in the neutrinoless double-electron capture of ${ }^{190} \mathrm{Pt}$ 

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#### Abstract

Background: The observation of neutrinoless double-beta transitions would indicate physics beyond the standard model as the lepton number conservation is violated. For a complete degeneracy in the energy of the initial and final states, the neutrinoless double-electron capture is resonantly enhanced. This shortens the half-life to similar orders of magnitude as the neutrinoless double-beta decay and expands the set of nuclei for the search of neutrinoless double-beta transitions as the observation of either process would be equally likely. Purpose: In order to clearly identify transitions that are resonantly enhanced, among other parameters the total energy of the decay, $Q_{\varepsilon \varepsilon}$, needs to be measured very precisely. Of the twelve initially identified candidates, the last remaining decay without a precise $Q_{\varepsilon \varepsilon}$ was ${ }^{190} \mathrm{Pt}(0 \nu \varepsilon \varepsilon){ }^{190}$ Os. Method: The $Q_{\varepsilon \varepsilon}$ value was determined with the Penning trap mass spectrometer LEBIT by measuring the ratio of the cyclotron frequencies of ${ }^{190} \mathrm{Pt}^{+}$and ${ }^{190} \mathrm{Os}^{+}$in a 9.4 T superconducting magnet. Result: The $Q_{\varepsilon \varepsilon}$ value was determined to be $1401.57(47) \mathrm{keV}$ with an uncertainty reduction of an order of magnitude compared to its previously known value. The absolute value is shifted by $17.17(623) \mathrm{keV}$ relative to the previously accepted one. Furthermore, the mass value of ${ }^{190} \mathrm{Pt}$ was found to be shifted by more than three standard deviations. In addition we improved the mass values for ${ }^{186,190} \mathrm{Os},{ }^{194} \mathrm{Pt}$. Conclusion: Transitions to the two nuclear excited states of ${ }^{190} \mathrm{Os}$ with $1326.9(5) \mathrm{keV}$ and 1387.00(2) keV energy were identified to be resonantly enhanced within a $1 \sigma$ uncertainty. The significantly reduced uncertainty of $Q_{\varepsilon \varepsilon}$ confirmed the potential for a resonantly enhanced transition.


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## I. INTRODUCTION

One of several effects that could indicate physics beyond the standard model is lepton number violation. This could be confirmed with the observation of neutrinoless double-beta decay $(0 \nu \beta \beta)$ or the related neutrinoless double-electron capture ( $0 \nu \varepsilon \varepsilon$ ), which would demonstrate the Majorana nature of the neutrino. Several internationally funded collaborations are currently investigating the $0 \nu \beta \beta$ decay as its half-life is expected to be several orders of magnitude shorter than the $0 \nu \varepsilon \varepsilon$ decay half-life. However, in the special case that initial and final states of the decay are degenerate in energy, a resonant enhancement is expected which could lead to an increase of the decay rate by a factor of up to $10^{12}[1,2]$. Then, the electron capture is the favored transition as no background is present from the $2 \nu \varepsilon \varepsilon[3]$ and the mere detection of the de-excitation associated with the decay would be a proof for the existence of the $0 \nu \varepsilon \varepsilon$ branch [4]. The rate of the $0 \nu \varepsilon \varepsilon$ decay is given by

$$
\begin{equation*}
\lambda=\left|V_{\varepsilon \varepsilon}\right|^{2} \frac{\Gamma}{\Delta^{2}+\Gamma^{2} / 4}=\left|V_{\varepsilon \varepsilon}\right|^{2} F \tag{1}
\end{equation*}
$$

[^0]with the transition amplitude between the two atoms, $V_{\varepsilon \varepsilon}$ [4]. $\Gamma$ denotes the width of the two-electron hole state in the excited daughter atom and $\Delta=Q_{\varepsilon \varepsilon}-B_{2 \mathrm{~h}}-E_{\gamma}$ the degeneracy parameter [2]. The fraction in Eq. (1) provides a measure for the elevated decay rate and is called the resonance parameter, $F$. It is typically normalized to the decay rate to the ground state of the daughter nuclide to give the enhancement factor, $E F$.
The degeneracy parameter is determined by the $Q_{\varepsilon \varepsilon}$ value, which is the difference of the ground state masses of mother and daughter atom, $\mathrm{E}_{\gamma}$, the energy of the excited state in the daughter nucleus, and $B_{2 \mathrm{~h}}$, the binding energy of the double-electron hole in the daughter atom. Typically $\Delta$ is large as the difference of $Q_{\varepsilon \varepsilon}-E_{\gamma}$ is usually on the order of hundreds of keV . The decay rate is only significantly elevated when $\Delta$ is of the order of 100 eV or smaller.
Feasible candidates for a resonantly enhanced neutrinoless double-electron capture have been identified [4], but in all cases the uncertainty on the $Q_{\varepsilon \varepsilon}$ value, calculated from the masses published in the Atomic Mass Evaluation (AME) 2003 [5], was too large to unambiguously confirm resonant enhancement $[3,4]$. Most of the masses used for the calculation of $Q_{\varepsilon \varepsilon}$ were deduced from $(n, \gamma)$ reactions and some of them were proven to be significantly inaccurate $[6,7]$. Therefore, several Penning trap
experiments around the world started to measure $Q_{\varepsilon \varepsilon}$ values directly [6-18]. Up to now, resonant enhancement was found in two of the candidates, ${ }^{152} \mathrm{Gd}$ and ${ }^{156} \mathrm{Dy}$, with an enhancement of up to $7.7 \times 10^{8}$ for the latter [12, 13].
Among the list of potential resonantly enhanced doubleelectron captures, only the $Q_{\varepsilon \varepsilon}$ value of ${ }^{190} \mathrm{Pt}$ remained with an uncertainty of more than 1 keV . The decay to the 1382.4 keV excited state in ${ }^{190} \mathrm{Os}$ [19] with even parity and suggested spin of 0,1 or 2 was expected to lead to resonant enhancement with the capture of two electrons from the $L$ and $M$ orbitals. Due to the large uncertainty in $Q_{\varepsilon \varepsilon}$ of 6.2 keV [20], five different combinations were candidates for a resonant enhancement [4]. Therefore, a more precise measurement on $Q_{\varepsilon \varepsilon}$ is required to determine which of those could lead to resonant enhancement. In this work the precise measurement of $Q_{\varepsilon \varepsilon}$ is discussed in the context of resonant enhancement of $0 \nu \varepsilon \varepsilon$ decays.

## II. EXPERIMENT DESCRIPTION

The $Q_{\varepsilon \varepsilon}$ value determination of the ${ }^{190} \mathrm{Pt}$ decay and all related measurements were performed at the Low-Energy Beam and Ion Trap (LEBIT) [21] located at the National Superconducting Cyclotron Laboratory (NSCL). An overview of all components of the LEBIT experiment used for this work is shown in Fig. 1. Singly charged Pt , Os, and ${ }^{12} \mathrm{C}_{16}$ ions were produced by a laser ablation ion source [22], employing a pulsed, frequencydoubled Nd:YAG laser and a rotatable target holder on which targets made of naturally composed platinum and osmium, and Sigradur ${ }^{\circledR}$, a glassy carbon produced by HTW Hochtemperatur Werkstoffe GmbH, were mounted side by side. The holder was rotated with a computer controlled stepper motor to ensure the ion production of only the desired material. Subsequently, the ions were captured in a radio frequency quadrupole (RFQ) cooler and buncher [23] for accumulation and cooling. After being ejected from the buncher a fast kicker located in the beam line between the buncher and the Penning trap was used as time-of-flight gate to mass select ions with the desired mass number. A typical spectrum showing the separation of the ions can also be found in reference [23].
The hyperbolic Penning trap [24] of the LEBIT facility is located inside a 9.4 T superconducting magnet where three-dimensional confinement of ions is realized with a strong homogeneous magnetic field, $\vec{B}$, superimposed on a weak electrostatic quadrupole field, $\vec{E}$. This results in an ion motion characterized by the axial, modified cyclotron and magnetron eigenmotions with frequencies $\nu_{z}, \nu_{+}$and $\nu_{-}$respectively [25]. The radius of the latter motion was a priori enlarged by forcing the ions to enter the trap off-axis with a four-fold segmented Lorentz steerer [26]. Following its capture, the ion ensemble was additionally purified removing contaminants that could be produced in the ion source. This was done by a dipo-


FIG. 1. Schematic overview of the LEBIT components that were used in these measurements.


FIG. 2. One of the ${ }^{190} \mathrm{Pt}^{+} 400-1200-400 \mathrm{~ms}$ Ramsey time-offlight resonances, composed of 2716 ions, used in the determination of $Q_{\varepsilon \varepsilon}^{\text {direct }}$. The solid line is the fit of the theoretical line shape to the data.
lar excitation of the mass dependent modified cyclotron motion to radii that are large enough to prevent the ions from interfering with the ions of interest.
In the actual measurement using the time-of-flight ion-cyclotron-resonance technique (TOF-ICR) [27, 28], the conversion from magnetron into modified cyclotron motion was probed with a quadrupolar rf field around the side-band $\nu_{+}+\nu_{-}=\nu_{c}$. The free cyclotron frequency of a singly charged ion,

$$
\begin{equation*}
\nu_{c}=\frac{1}{2 \pi} \frac{q}{m_{\mathrm{ion}}} B \tag{2}
\end{equation*}
$$

is directly linked to its charge-to-mass ratio $q / m_{\text {ion }}$ with charge, $q$, and mass, $m_{\text {ion }}$, and also depends on the magnetic field strength, $B$. At LEBIT, the so-called Ramsey technique is applied using a two-pulse excitation separated by a waiting time for a gain in precision compared to the continuous rf excitation of the ion motion [29-31]. Figure 2 features a ${ }^{190} \mathrm{Pt}^{+}$resonance obtained in the current measurements.

In addition to the cyclotron frequency measurements of the ions of interest, the magnetic field strength has to be determined precisely. Thus, cyclotron frequency measurements are performed with a reference ion prior and

TABLE I. Overview of the individual measurements and the deduced results. The indirect $Q_{\varepsilon \varepsilon}$ value is calculated from the frequency ratios in line 2 and 3 , the final $Q_{\varepsilon \varepsilon}$ value in line 6 is the weighted average of the two lines above. $\delta$ is the subtraction of the respective AME2012 values from the values in column 4 or 5 .

| Ion | Reference | $r$ | ME (keV) | $Q(\mathrm{keV})$ | $\delta(\mathrm{keV})$ |
| :--- | :---: | :---: | :---: | :---: | :---: |
| ${ }^{194} \mathrm{Pt}^{+}$ | ${ }^{12} \mathrm{C}_{16}^{+}$ | $1.0102223437(35)$ | $-34759.38(62)$ |  | $3.22(109)$ |
| ${ }^{190} \mathrm{Pt}^{+}$ | ${ }^{194} \mathrm{Pt}^{+}$ | $0.9793633262(35)$ | $-37305.46(88)$ |  | $19.54(606)$ |
| ${ }^{190} \mathrm{Os}^{+}$ | ${ }^{194} \mathrm{Pt}^{+}$ | $0.9793555644(35)$ | $-38707.81(88)$ |  | $1.59(182)$ |
|  |  |  |  | $Q_{\varepsilon \varepsilon}^{\text {indirect }}=1402.35(90)$ |  |
| ${ }^{190} \mathrm{Pt}^{+}$ | ${ }^{190} \mathrm{Os}^{+}$ | $1.0000079192(31)$ |  | $Q_{\varepsilon \varepsilon \varepsilon}^{\text {direct }}=1401.26(56)$ |  |
|  |  |  | $Q_{\varepsilon \varepsilon \varepsilon}^{\text {verage }}=1401.57(47)$ | $17.17(623)$ |  |
| ${ }^{190} \mathrm{Pt}^{+}$ |  |  |  |  |  |

subsequent to the measurement of the ion of interest (see all used ion pairs in Table I). The magnetic field is obtained by interpolating the frequency to the time of the measurement of the ion of interest. Then, the mass of the neutral atom can be derived with Eq. (2) as

$$
\begin{equation*}
m=r\left(m_{\mathrm{ref}}-m_{\mathrm{e}}+\frac{b_{\mathrm{ref}}}{c^{2}}\right)+m_{\mathrm{e}}-\frac{b}{c^{2}} \tag{3}
\end{equation*}
$$

where $r=\nu_{c, \text { ref }} / \nu_{c}$ denotes the frequency ratio of reference ion and ion of interest, $\left(m_{\text {ref }}-m_{\mathrm{e}}\right)$ denotes the mass of the reference ion, $b_{\text {ref }}$ and $b$ denote the first ionization potential of the reference ion and the ion of interest, $m_{\mathrm{e}}$ the electron mass and $c$ the speed of light. The $Q$ value can then be calculated from Eq. (3) as:

$$
\begin{align*}
Q_{\varepsilon \varepsilon}^{\text {indirect }} & =\left(r_{1}-r_{2}\right)\left(m_{\mathrm{ref}} c^{2}-m_{\mathrm{e}} c^{2}+b_{\mathrm{ref}}\right)  \tag{4}\\
& -b_{1}+b_{2}
\end{align*}
$$

For the $Q$ value of the ${ }^{190} \mathrm{Pt}$ double-electron capture $r_{1}=\nu_{c}\left({ }^{194} \mathrm{Pt}^{+}\right) / \nu_{c}\left({ }^{190} \mathrm{Pt}^{+}\right)$and $r_{2}=$ $\nu_{c}\left({ }^{194} \mathrm{Pt}^{+}\right) / \nu_{c}\left({ }^{190} \mathrm{Os}^{+}\right)$are independently measured frequency ratios of the reference ion ${ }^{194} \mathrm{Pt}^{+}$and the respective ion of interest. $b_{1}$ and $b_{2}$ denote the ionization potentials of ${ }^{190} \mathrm{Pt}$ and ${ }^{190} \mathrm{Os}$. The $Q_{\varepsilon \varepsilon}$ value can also be determined directly by a measurement of the frequency ratio of mother and daughter nuclide. It ultimately yields a better precision with the same number of measurements. This reduces Eq. (4) to

$$
\begin{align*}
Q_{\varepsilon \varepsilon}^{\text {direct }} & =(r-1)\left(m_{\mathrm{ref}} c^{2}-m_{\mathrm{e}} c^{2}+b_{\mathrm{ref}}\right) \\
& +b_{\mathrm{ref}}-b \tag{5}
\end{align*}
$$

The nuclides ${ }^{190} \mathrm{Os}$ and ${ }^{190,194} \mathrm{Pt}$ were linked to the atomic mass standard, ${ }^{12} \mathrm{C}$, by the measurement of the frequency ratio $\nu_{c}\left({ }^{12} \mathrm{C}_{16}^{+}\right) / \nu_{c}\left({ }^{194} \mathrm{Pt}^{+}\right)$with the carbon cluster ${ }^{12} \mathrm{C}_{16}^{+}$as mass reference.
In order to avoid systematic differences the measurement conditions were kept the same for all ions. However, systematic errors in the mass determination, for example due to trap imperfections, need to be considered if the masses of the ion of interest and the reference ion differ significantly [32]. At LEBIT, the shift of the frequency ratio, $r$, between singly charged ions of mass difference $\Delta m$, has been determined to be $2.0 \times 10^{-10} / \mathrm{u} \times \Delta m$,
which is also added in quadrature to the statistical uncertainty [33]. Furthermore, non-linear magnetic field changes during the cyclotron frequency measurement of the desired ion and between those of the reference ion can lead to errors in the frequency determination. The effect of these fluctuations on $r$ was studied with ${ }^{39} \mathrm{~K}^{+}$ ions over a time span of 22 hours using the technique described in [34]. The additional uncertainty was determined to be $1.2(6) \times 10^{-10}$ per hour. As about one hour was required to measure the cyclotron frequency with a statistical uncertainty which was more than two orders of magnitude larger, this insignificant contribution to the total uncertainty was not considered in the evaluation. The presence of isobaric contaminants in the trap during the measurement also can lead to frequency shifts [35]. This effect was minimized by the purification steps discussed above and by restricting the total number of ions in the trap. Therefore, only events with five or fewer ions were analyzed. This corresponds to fewer than nine ions in the trap at the same time as our MCP detection efficiency was $63 \%$ [36].

## III. RESULTS AND DISCUSSION

All measurements presented in the following were carried out with alternating TOF-ICR measurements of a reference ion and the ion of interest using a 400-1200400 ms Ramsey excitation scheme.
For the mass and $Q$ value measurements at the level of precision obtained in this work atomic and molecular binding energies need to be considered. The ionization potentials of Pt , $b_{\mathrm{Pt}}=8.95868(11) \mathrm{eV}$ [37], Os, $b_{\mathrm{Os}}=8.43823(20) \mathrm{eV}[38]$, and ${ }^{12} \mathrm{C}_{16}, b_{\mathrm{C}_{16}}=8(1) \mathrm{eV}$ [39], were used for the calculation of $Q$ values and mass excesses. In the case of $b_{\mathrm{C}_{16}}$ the uncertainty was inflated to 1 eV due to significant deviations of [39] and [40] on the determination of the first ionization potential of carbon clusters up to $\mathrm{C}_{15}$. For ${ }^{12} \mathrm{C}_{16}$ also the molecular binding energy of $97.3(27) \mathrm{eV}$ must be considered. This value is the mean of the binding energies published in [41] and [42] with their difference used as the uncertainty.
The results of the different frequency ratios determined in this work and derived mass excess and $Q$ values are summarized in Table I.

TABLE II. Parameters of the resonantly enhanced neutrinoless double-electron captures in ${ }^{190} \mathrm{Pt}$ to the nuclear excited state in ${ }^{190}$ Os characterized by their excitation energies, $E_{\gamma}$ and spin, $I_{f}^{\pi}$. Values for the enhancement factor, $E F$, for the transitions that are degenerate in energy within their uncertainties are given together with the values $E F^{\text {min }}$ for a $1 \sigma$ larger degeneracy parameter and $E F^{\text {max }}$ for perfect resonance.

| $E_{\gamma}(\mathrm{keV})$ | $I_{f}^{\pi}$ | Orbitals $B_{2 h}(\mathrm{keV})$ | $\Gamma(\mathrm{eV})$ | $\Delta(\mathrm{keV})$ | $E F^{\text {min }}<E F<E F^{\text {max }}$ |  |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: |
| $1326.9(5)$ | 1,2 | $K N_{1}$ | 74.53 | 52 | $0.14(69)$ | $1.3 \times 10^{6}<E F=4.5 \times 10^{7}<1.4 \times 10^{9}$ |
|  |  | $K N_{2}$ | 74.42 | 51 | $0.25(69)$ | $1.0 \times 10^{6}<E F=1.4 \times 10^{7}<1.4 \times 10^{9}$ |
|  |  | $K N_{3}$ | 74.33 | 49 | $0.34(69)$ | $8.2 \times 10^{5}<E F=7.6 \times 10^{6}<1.4 \times 10^{9}$ |
| $1382.4(2)$ | $(0,1,2)^{+}$ | $K N_{4}$ | 74.16 | 49 | $0.51(69)$ | $6.0 \times 10^{5}<E F=3.4 \times 10^{6}<1.5 \times 10^{9}$ |
|  | $L_{1} M_{1}$ | 16.02 | 22 | $3.15(51)$ |  |  |
| $1387.00(2)$ | $3_{3} L_{3}$ | 21.74 | 10 | $-2.57(51)$ |  |  |
|  |  | $L_{2} M_{5}$ | 14.34 | 7 | $0.23(47)$ | $2.5 \times 10^{5}<E F=2.3 \times 10^{6}<1.0 \times 10^{10}$ |
|  | $L_{3} M_{3}$ | 13.33 | 13 | $1.24(47)$ |  |  |

## A. Establishing a link to the mass standard via ${ }^{194} \mathbf{P t}$

The link to the mass standard ${ }^{12} \mathrm{C}$ of all measured nuclides was established by alternating TOF-ICR measurements of ${ }^{12} \mathrm{C}_{16}^{+}$and ${ }^{194} \mathrm{Pt}^{+}$. In total 19 frequency ratios were recorded with a weighted average of $r=$ $1.0102223437(35)$. The Birge ratio [43] of $0.88(11)$ for this measurement indicates a negligible overestimation of the individual statistical uncertainties. The systematic shift due to the difference in the mass-to-charge ratio between the two ion species was accounted for by correcting the frequency ratio by $4.0 \times 10^{-10}$.
Using the previously introduced ionization potentials, Eq. (3) yields the mass excess $\mathrm{ME}\left({ }^{194} \mathrm{Pt}\right)=$ $-34759.38(62) \mathrm{keV}$, which deviates by 3.22 keV , corresponding to more than $5 \sigma$, from the mass excess published in the last Atomic Mass Evaluation (AME2012) [20], $\mathrm{ME}_{\mathrm{AME}}=-34762.6(9) \mathrm{keV}$. Therefore, the new mass value is used in all further calculations.
Beyond the change of the ${ }^{194} \mathrm{Pt}$ mass, this measurement has a direct impact on the masses of all heavier platinum isotopes up to ${ }^{197} \mathrm{Pt}$, which are linked through $(\mathrm{n}, \gamma)$ reactions. This discrepancy could be caused by the link of this chain to ${ }^{197} \mathrm{Au}$ via the energy measurement of the ${ }^{197} \mathrm{Pt} \beta^{-}$decay.

## B. Resonantly enhanced transitions in agreement with the $Q_{\varepsilon \varepsilon}$ value of ${ }^{190} \mathrm{Pt}$

For the direct determination of the $Q_{\varepsilon \varepsilon}$ value of ${ }^{190} \mathrm{Pt}$ 19 frequency ratios of the cyclotron frequencies of ${ }^{190} \mathrm{Pt}^{+}$ and ${ }^{190} \mathrm{Os}^{+}$were recorded. Their weighted average is $r=1.0000079192(31)$ with a Birge ratio of $0.86(11)$. It does not require any corrections for mass dependent shifts as both nuclides form an isobaric doublet. Furthermore, all systematic effects arising from imperfections in the electric or magnetic field are negligible in this case. With Eq. (5) $Q_{\varepsilon \varepsilon}^{\text {direct }}=1401.26(56) \mathrm{keV}$ is determined. In order to confirm this result, the $Q_{\varepsilon \varepsilon}$ value was determined independently with the indirect method us-


FIG. 3. Decay scheme of the neutrinoless double-electron capture in ${ }^{190} \mathrm{Pt}$. The marked transitions energetically allow resonant enhancement within the limits of uncertainty.
ing the frequency ratios for ${ }^{194} \mathrm{Pt}^{+}$and ${ }^{190} \mathrm{Pt}^{+}$and ${ }^{194} \mathrm{Pt}^{+}$and ${ }^{190} \mathrm{Os}^{+}$in Eq. (4). For this purpose $r_{1}=$ $0.9793633262(35)$ was calculated from 18 measured frequency ratios with a Birge ratio of $0.99(11)$ and $r_{2}=$ $0.9793555644(35)$ from 17 measured frequency ratios with a Birge ratio of $1.21(12)$. Both frequency ratios are already corrected by $-7.8 \times 10^{-10}$ to account for systematic mass-dependent frequency shifts. Furthermore, the uncertainty of $r_{2}$ was inflated by multiplication with the Birge ratio of the respective distribution. Equation (4) yields then $Q_{\varepsilon \varepsilon}^{\text {indirect }}=1402.35(90) \mathrm{keV}$.
The results of both measurements agree within their uncertainties. Therefore, their error weighted average is calculated to $Q_{\varepsilon \varepsilon}^{\mathrm{av}}=1401.57(47) \mathrm{keV}$ which is more than an order of magnitude more accurate and 17.17 keV higher than $Q_{\varepsilon \varepsilon}^{\mathrm{AME} 2012}=1384.4(62) \mathrm{keV}$, published in the most recent AME [20].
Based on our new $Q_{\varepsilon \varepsilon}$ value three nuclear excited states of the daughter nucleus ${ }^{190}$ Os with energies that would allow resonant enhancement were identified from the data published in [19]. The exited states with the atomic orbitals of the captured ions being closest to the resonant condition are presented in Table II and put in context of the $0 \nu \varepsilon \varepsilon$ decay scheme in Fig. 3. For spin and parity the notation from [19] was adopted. If more than one spin or no parity values are shown, the actual values are
unknown. Nevertheless, only transitions that are possible within the presently known parameters were selected. With the two-electron hole energy, $B_{2 h}$, from [44], and the sum of the electron level widths [45], the enhancement relative to the non-resonant $K K$ capture was calculated using Eq. (1). Due to the large shift in $Q_{\varepsilon \varepsilon}$ the transitions that were considered to be resonantly enhanced [4] are now found to be non-resonant.
Five transitions to two different nuclear excited states were found to be degenerate in energy within the limits of uncertainty. However, the transition from the $0^{+}$ground state of ${ }^{190} \mathrm{Pt}$ to the $E_{\gamma}=1387.00(2) \mathrm{keV}$ nuclear excited state with $I_{f}^{\pi}=3^{-}$is three-fold forbidden. Overall, the transitions to the $E_{\gamma}=1326.9(5) \mathrm{keV}$ state are the most promising ones as the close lying binding energies of the $N$ orbitals increase the likelihood for a resonant enhancement. Based on our new $Q_{\varepsilon \varepsilon}$ value the enhancement ranges from $6.0 \times 10^{5}$ to $1.5 \times 10^{9}$ relative to the ground state for the transition to the $E_{\gamma}=1326.9(5) \mathrm{keV}$ excited state or even $1.0 \times 10^{10}$ for the $E_{\gamma}=1387.00(2) \mathrm{keV}$ excited state.
The total uncertainty of $\Delta$ is in general dominated by $Q_{\varepsilon \varepsilon}$, only for transitions to the $E_{\gamma}=1326.9(5) \mathrm{keV}$ excited state the uncertainty of the excitation energy contributes at the same magnitude. Additional reductions of $\Delta$, originating from the Coulomb interaction of the electrons, are expected to be about 0.04 keV for a $K N$ electron pair and about 0.1 keV for an $L M$ electron pair for heavy atoms [4].
At the present level of precision ${ }^{190} \mathrm{Pt}$ is, together with ${ }^{152} \mathrm{Gd}$ and ${ }^{156} \mathrm{Dy}$, the most promising candidate for a $0 \nu \varepsilon \varepsilon$ measurement. While one transition in the ${ }^{156}$ Dy decay was found to be almost perfectly resonant, the capture of two electrons from outer shells $\left(M_{1} N_{3}\right)$ reduces the nuclear matrix element significantly [12]. In case of ${ }^{152} \mathrm{Gd}$ the resonance could not be confirmed with $\Delta=0.91(18) \mathrm{keV}$, but the nuclear matrix element is significantly higher due to the electron capture from the inner orbitals $\left(K L_{1}\right)$ [13]. For ${ }^{190} \mathrm{Pt}$, however, the situation is fortunate as several close-lying transitions can even enable a double resonance.

## C. Shift of the mass values for ${ }^{190} \mathrm{Pt}$ and ${ }^{190} \mathrm{Os}$ and their consequences

Beyond the identification of so-far unconsidered resonant double-electron transitions in the ${ }^{190} \mathrm{Pt}$ decay, the shift in $Q_{\varepsilon \varepsilon}$ indicates that the mass values of ${ }^{190} \mathrm{Pt}$ and ${ }^{190} \mathrm{Os}$ published in [20] are incorrect. Therefore, their mass excesses were deduced with Eq. (3) and the frequency ratios that were used for the determination of the $Q_{\varepsilon \varepsilon}^{\text {indirect }}$ value. The results, $\operatorname{ME}\left({ }^{190} \mathrm{Pt}\right)=-37305.46(88) \mathrm{keV}$ and ME ( $\left.{ }^{190} \mathrm{Os}\right)=-38707.81(88) \mathrm{keV}$, deviate from the AME2012 values, $\mathrm{ME}_{\mathrm{AME}}\left({ }^{190} \mathrm{Pt}\right)=-37325(6) \mathrm{keV}$ and $\mathrm{ME}_{\mathrm{AME}}\left({ }^{190} \mathrm{Os}\right)=-38709.4(16) \mathrm{keV}$, by 19.54 keV and 1.59 keV , respectively. While a significant shift in
the ${ }^{190} \mathrm{Pt}$ mass excess was expected from the large shift in $Q_{\varepsilon \varepsilon}$, the additional shift in the ${ }^{190}$ Os mass excess suggests that the mass values in all osmium isotopes from ${ }^{186}$ Os to ${ }^{191}$ Os are too small as they are all linked via ( $\mathrm{n}, \gamma$ ) reactions.
In order to resolve if the shift for the other Os isotopes is similar or not, the frequency ratio of ${ }^{190} \mathrm{Pt}^{+}$and ${ }^{186} \mathrm{Os}^{+}, \quad r=1.0215436473(47)$, was measured and the mass difference determined. Subtracting the mass value of the $\alpha$ particle yields the $Q$ value of the ${ }^{190} \mathrm{Pt} \alpha$ decay of $3268.40(82) \mathrm{keV}$. A shift of $15.92(624) \mathrm{keV}$ is observed to the $Q_{\alpha}$ value from [20], which is similar to the shift in $Q_{\varepsilon \varepsilon}$. Via $\operatorname{ME}\left({ }^{186} \mathrm{Os}\right)=\operatorname{ME}\left({ }^{190} \mathrm{Pt}\right)-\mathrm{ME}\left({ }^{4} \mathrm{He}\right)-Q_{\alpha}$ the mass excess of ${ }^{186} \mathrm{Os}$ is determined to be -42998.8 (12) keV, shifted by $3.6(19) \mathrm{keV}$ with respect to the AME2012. This strongly indicates a systematic mass shift of all stable osmium isotopes linked with (n, $\gamma$ ) reactions. The mass values of the stable isotopes from ${ }^{186}$ Os to ${ }^{190}$ Os are determined by the energies of the $\left.{ }^{186} \operatorname{Re}\left(\beta^{-}\right)\right)^{186} \mathrm{Os}$ and ${ }^{187} \operatorname{Re}\left(\beta^{-}\right){ }^{187}$ Os decays. As a shift is present in all these mass values, at least one of the $\beta$ decay energy measurements is expected to be incorrect.

## IV. CONCLUSION

Using Penning trap mass spectrometry the $Q_{\varepsilon \varepsilon}$ value of the ${ }^{190} \mathrm{Pt}$ double electron capture was measured to be $1401.57(47) \mathrm{keV}$ and its uncertainty reduced by more than an order of magnitude. Further, we discovered that $Q_{\varepsilon \varepsilon}$ is $17.17(623) \mathrm{keV}$ higher than the one deduced from the AME2012, which was mainly due to a $3-\sigma$ shift of the ${ }^{190} \mathrm{Pt}$ mass value. Due to this shift, transitions to two different nuclear excited states in ${ }^{190}$ Os were found to allow resonant enhancement within their uncertainties while previously suggested transitions were excluded. However, the present precision is not high enough to unambiguously confirm resonant enhancement. For this purpose the uncertainty of $Q_{\varepsilon \varepsilon}$ needs to be reduced by at least one more order of magnitude. Furthermore, the uncertainty of the $E_{\gamma}=1326.9(5) \mathrm{keV}$ excited state needs to be reduced as a transition to this state is presently the closest one to resonance.

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