Resonant Enhancement of Neutrinoless Double-Electron Capture in ^{152}Gd

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In the search for the nuclide with the largest probability for neutrinoless double-electron capture, we have determined the $Q_{\epsilon\epsilon}$ value between the ground states of ^{152}Gd and ^{152}Sm by Penning-trap mass-ratio measurements. The new $Q_{\epsilon\epsilon}$ value of 55.70(18) keV results in a half-life of 10^{26} yr for a 1 eV neutrino mass. With this smallest half-life among known $0\nu\epsilon\epsilon$ transitions, ^{152}Gd is a promising candidate for the search for neutrinoless double-electron capture.

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The discovery of neutrino oscillations has proven neutrinos are massive particles. However, these findings cannot answer the question of whether a neutrino is a Dirac or a Majorana particle. An answer to this long-standing question lies in a search for neutrinoless double-beta transformations, i.e., double-beta decay ($\beta^-\beta^-$), double-electron capture ($\epsilon\epsilon$), and a mixture of positron-electron decay ($\beta^+\epsilon$), which are second-order weak interaction processes with very small transition probabilities. These processes can be either accompanied by an emission of two neutrinos or can be neutrinoless. An observation of the latter, not unambiguously discovered yet in spite of persistent attempts in the last decades [1], would prove that the neutrino is a Majorana particle. Furthermore, a measurement of the half-life of this process would allow a determination of the effective Majorana neutrino mass $m_{\beta\beta}$ [2].

Since the prediction of $\beta^-\beta^-$ decay in 1935 [3], eleven nuclides have been experimentally identified as two-electron emitters with half-lives between 10^{18} and 2.5×10^{24} yr [1]. During this time, $\epsilon\epsilon$ capture was no longer pursued due to its much longer expected half-lives. However, based on the theory of $\epsilon\epsilon$ capture by Winter [4], Bernabeu *et al.* [5] pointed to a possible resonant enhancement of neutrinoless $\epsilon\epsilon$ capture if the initial and excited final states of the system are degenerate in energy. Sujkowski and Wycech [6] showed, using a perturbative approach for a detailed theoretical description of neutrinoless $\epsilon\epsilon$ capture, that its probability can be competitive with neutrinoless double-beta decay. The degeneracy parameter Δ can be expressed as $\Delta = Q_{\epsilon\epsilon} - B_{2h} - E_\gamma$ and enters into the denominator of the $\epsilon\epsilon$-capture rate [5]:

$$\lambda_{\epsilon\epsilon} = |V_{\epsilon\epsilon}|^2\frac{\Gamma_{2h}}{\Delta^2+\Gamma_{2h}^2/4} = |V_{\epsilon\epsilon}|^2 R, \quad (1)$$

where $Q_{\epsilon\epsilon}$ is the difference between the initial and final atomic masses, B_{2h} is the energy of the double-electron hole in the atomic shell of the daughter nuclide, E_γ is the excitation energy of the daughter nuclide, Γ_{2h} is the sum of the widths of the double-electron hole and the nuclear excited state in the daughter nuclide, R is the resonance enhancement factor. Equation (1) can be obtained in the second order of perturbation theory for the standard β-decay Hamiltonian with massive Majorana neutrinos. The value $V_{\epsilon\epsilon}$ has the meaning of the transition amplitude between two atoms with violation of total lepton number [7,8]. In the total decay amplitude, $V_{\epsilon\epsilon}$ factorizes when the resonance conditions are satisfied. $V_{\epsilon\epsilon}$ is proportional to $m_{\beta\beta}$, the wave functions of the captured electrons averaged over the volume of nucleus, and the nuclear matrix element $M^{0\nu}$. In the case of capture of two s-orbital electrons,

$$V_{\varepsilon\varepsilon} = m_{\beta\beta}\frac{\sqrt{2}g_A^2G_\beta^2}{(4\pi)^2R_{\mathrm{nucl}}}\bar{f}_a\bar{f}_bM^{0\nu}. \quad (2)$$

Here, $G_\beta = G_F\cos\theta_C$, θ_C is the Cabibbo angle, g_A is the axial-vector nucleon coupling constant, R_{nucl} is nuclear radius, $\bar{f}_{a,b}$ is the averaged upper bispinor component of the $n_{a,b}s_{1/2}$ electron, $n_{a,b}$ is the principal quantum number. The explicit form of $M^{0\nu}$ can be found in [2].

TABLE I. Parameters of three double-electron $0^+ \rightarrow 0^+$ transitions of primary interest. $Q_{\epsilon\epsilon}$ is the difference between the initial and final atomic masses, B is a sum of the binding energies for separate orbits [10], B_{2h} for ^{152}Gd is the double-electron hole binding energy, calculated in this work, E_γ is the excitation energy of the daughter nuclide, and Δ is the degeneracy parameter. All data are given in keV. Old $Q_{\epsilon\epsilon}$ values are taken from [11].

Transformation	$Q_{\epsilon\epsilon}$ (old)	$E = B + E_\gamma$	Orbitals	$\Delta = Q_{\epsilon\epsilon}(\text{old}) - E$	$Q_{\epsilon\epsilon}$ (new)	$\Delta = Q_{\epsilon\epsilon}(\text{new}) - E$
$^{112}\text{Sn} \rightarrow {}^{112}\text{Cd}$	1919.5(4.8)	1901.7	KL_1	17.8(4.8)	1919.82(16)[a]	18.12(16)
		1924.4	KK	−4.9(4.8)		−4.58(16)
$^{152}\text{Gd} \rightarrow {}^{152}\text{Sm}$	54.6(3.5)	$B_{2h} = 54.79$	KL_1	−0.19(3.50)	55.70(18)[b]	0.91(18)
$^{164}\text{Er} \rightarrow {}^{164}\text{Dy}$	23.3(3.9)	18.09	L_1L_1	5.21(3.90)		

[a]From Ref. [12]
[b]This work

If the degeneracy parameter Δ is comparable to or smaller than Γ_{2h}, this will lead to a resonant enhancement of the $\epsilon\epsilon$-capture transition probability by several orders of magnitude and, hence, can give rise to realistic experiments searching for neutrinoless $\epsilon\epsilon$ capture. From a variety of nuclides with double-beta transformations 12 nuclides can undergo pure $\epsilon\epsilon$ capture and 22 nuclides $\beta^+\epsilon$ decay [9]. However, there are only three transitions with a change of the spin by less than two units and, hence, not suppressed, for which the degeneracy conditions were expected to hold at the level of a 1σ experimental error (Table I). The nuclide ^{112}Sn undergoes mixed $\beta^+\epsilon$ and $\epsilon\epsilon$ decays, whereas ^{152}Gd and ^{164}Er are nuclides with pure $\epsilon\epsilon$, $0^+ \rightarrow 0^+$ transitions between nuclear ground states. As can be seen from Table I, the mass differences $Q_{\epsilon\epsilon}$ evaluated in [11] were not precise enough to decide whether the resonance conditions are fulfilled (see column 4 of Table I).

Tremendous progress in Penning-trap mass spectrometry in the last decade [13,14] finally allowed sufficiently precise $Q_{\epsilon\epsilon}$ measurements for, e.g., the $^{112}\text{Sn} \rightarrow {}^{112}\text{Cd}$ [12] and for $^{74}\text{Se} \rightarrow {}^{74}\text{Ge}$ [15,16] transitions. The latter is a $0^+ \rightarrow 2^+$ transition and, hence, as shown in [15], is very unlikely. The new, much more precise results for ^{112}Sn are presented in columns 6 and 7 of Table I. They show the breakdown of the resonance condition for ^{112}Sn from possible nuclides for a search for neutrinoless $\epsilon\epsilon$ capture. The question whether the remaining candidates from Table I, ^{152}Gd and ^{164}Er, are suitable for a search of the neutrinoless process was still open.

In this Letter we report on a determination of the $Q_{\epsilon\epsilon}$ value of ^{152}Gd performed with SHIPTRAP [17] by a precise direct measurement of the cyclotron-frequency ratio of singly charged ions of ^{152}Sm and ^{152}Gd, $f_c(^{152}\text{Sm}^+)/f_c(^{152}\text{Gd}^+)$, with a time-of-flight ion cyclotron resonance technique [18]. The cyclotron frequency f_c of an ion with mass m and charge q stored in a magnetic field B is given by $f_c = qB/(2\pi m)$. A detailed description of cyclotron-frequency measurements with SHIPTRAP can be found in [19]. Ions were created by use of a laser ablation ion source [20]. Samples of Sm with natural abundances and of Gd (in the chemical form of oxide and with ^{152}Gd enriched to 38%) were deposited onto two stainless steel plates serving as targets. These were fixed to a rotary mechanical feedthrough with stepper motor. This allowed an alternate laser irradiation of the samples. Ions created in the ion source were guided into the preparation trap for cooling and centering. The cooled bunch of singly charged ions of ^{152}Sm or ^{152}Gd was then injected into the measurement trap. Here, a precise measurement of the cyclotron frequency f_c was performed. The $^{152}\text{Gd}^+$ and $^{152}\text{Sm}^+$ cyclotron frequencies were measured alternately for three days with a Ramsey-type excitation [21,22] with two 250 ms fringes separated by a waiting time of 1.5 s (Fig. 1, inset). Each of the 78 measurements lasted approximately 20 min and contained in total between 500 and 700 ions. In the measurement trap equal starting conditions were ensured for Gd and Sm ions by the same ion production mechanism, their similar masses, abundances in the samples and similar chemical properties. Along with the identical f_c-measurement

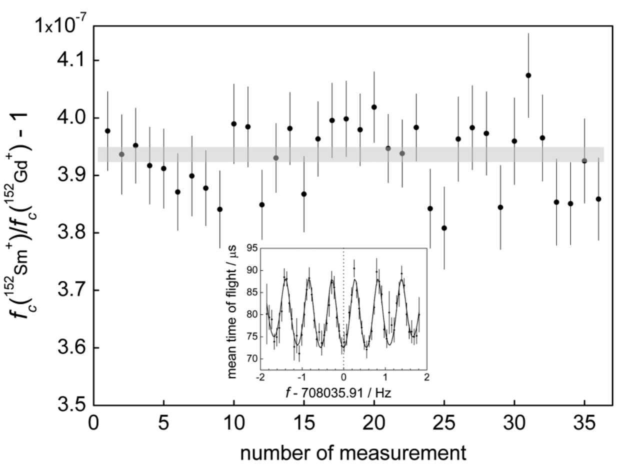

FIG. 1. Cyclotron-frequency ratios $f_c(^{152}\text{Sm}^+)/f_c(^{152}\text{Gd}^+)$ measured over three days with up to 5 detected ions. The error bars of the individual measurements are the statistical uncertainties taking into account magnetic field fluctuations. The grey shaded band represents the total uncertainty of the averaged frequency ratio. The inset displays a typical time-of-flight ion cyclotron resonance of $^{152}\text{Sm}^+$ with a Ramsey excitation pattern 250 ms–1.5 s–250 ms. The solid line is a fit of the expected line shape to the data points [21].

TABLE II. Parameters of double-electron capture of ^{152}Gd, measured or calculated in this paper: cyclotron-frequency ratio $f_c(^{152}\mathrm{Sm}^+)/f_c(^{152}\mathrm{Gd}^+)$, transition energy $Q_{\epsilon\epsilon}$, binding energy of the double hole B_{2h} in the atomic shell of the daughter nuclide ^{152}Sm, degeneracy parameter Δ, and total width of the transition final state Γ_{2h}.

$[f_c(^{152}\mathrm{Sm}^+)/f_c(^{152}\mathrm{Gd}^+) - 1]$	$3.9363(126) \times 10^{-7}$
$Q_{\epsilon\epsilon}$/keV	55.70(18)
B_{2h}/keV	54.794(9)
Δ/keV	0.91(18)
Γ_{2h}/eV	24.8(2.5)

procedure applied to $^{152}\mathrm{Sm}^+$ and $^{152}\mathrm{Gd}^+$ this will lead to equal shifts of the measured frequencies due to static imperfections of the measurement trap and, thus, will not affect the frequencies ratio [23]. Although the mean ion production rate was kept on average constant, temporal fluctuations of this rate from measurement to measurement could not be avoided. To investigate the possibility of a systematic frequency shift due to ion-ion interactions, the data were divided into 5 sets according to the number of detected ions. The data with more than 5 ions/cycle were not used in the analysis. A thorough analysis of the sets did not reveal any correlations between the frequency ratios and the number of ions. The drift of the cyclotron frequency in time did not exceed a few tens of mHz over 1 d due to an implementation of the stabilization of the temperature in the magnet bore as well as of the pressure in the liquid-helium cryostat [24]. The data analysis was performed along the lines of [25]. In Fig. 1 the cyclotron-frequency ratios $f_c(^{152}\mathrm{Sm}^+)/f_c(^{152}\mathrm{Gd}^+)$ measured with up to 5 detected ions/cycle are shown. The final frequency ratio with its uncertainty is given in Table II. A detailed description of the elaborate analysis of the experimental data of our experiment will be published elsewhere.

From the frequency ratio the $Q_{\epsilon\epsilon}$ value of ^{152}Gd is given by

$$Q_{\epsilon\epsilon} = [M(^{152}\mathrm{Sm}) - m_e]\left[\frac{f_c(^{152}\mathrm{Sm}^+)}{f_c(^{152}\mathrm{Gd}^+)} - 1\right], \quad (3)$$

where $M(^{152}\mathrm{Sm})$ and m_e are masses of neutral ^{152}Sm and the electron, respectively. The difference of the ionization energies of ^{152}Gd and ^{152}Sm is smaller than 1 eV and, hence, neglected. The final value for $Q_{\epsilon\epsilon}$ is given in Table II.

Since for ^{152}Gd the expected degeneracy parameter is less than 1 keV, an accurate calculation of the double-electron hole binding energy B_{2h} and its corresponding width Γ_{2h} is necessary. The energy B_{2h}—a difference between the energy of the $4f^7 5d^1$ configuration with $1s$ and $2s$ holes and the ground state energy ($4f^6$ configuration) of neutral ^{152}Sm—was calculated with the Dirac-Fock method [26] including the Breit (electron-correlation) and quantum electrodynamics (QED) corrections. The nuclear charge

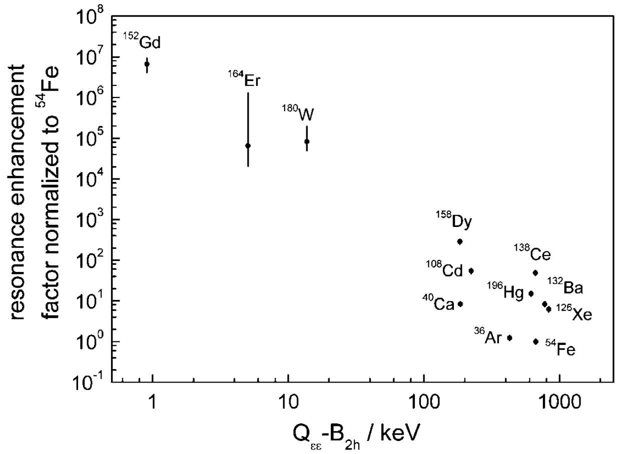

FIG. 2. Resonance enhancement factors R for pure double-electron capture transitions between ground states of the mother and daughter nuclides relative to the R value of ^{54}Fe.

distribution was taken into account within the Fermi model with the rms nuclear radius $\langle r^2\rangle^{1/2} = 5.0842$ fm [27]. The initial and final configurations of ^{152}Sm include 3106 and 295 atomic terms, respectively. We used the approximation in which the energy is averaged over all atomic terms of the nonrelativistic valence configuration. The validity of this approximation was previously demonstrated in calculations of binding energies, isotopic and chemical shifts of x-ray lines with large natural widths [28]. The uncertainty of this calculation is of the order of 10 eV mainly due to a rough estimate of the QED screening effect. The obtained value of B_{2h} is given in Table II. The width of the autoionizing state of ^{152}Sm with $1s$ and $2s$ holes is taken as the sum of the widths of the K and L_1 levels, and is equal to 24.8(2.5) eV [29]. By using these data the parameter Δ for $\epsilon\epsilon$ capture in ^{152}Gd was determined to be 0.91(18) keV.

This value is, by far, the lowest for all nuclides which can undergo $\beta^+\epsilon$ or $\epsilon\epsilon$ capture. Figure 2 shows a comparison of the resonance enhancement factor R [see Eq. (1)] for all known pure $\epsilon\epsilon$-capture transitions between ground states of the mother and daughter nuclides relative to the nuclide ^{54}Fe with the smallest R value. As can be seen from Fig. 2 the resonance enhancement for ^{152}Gd is a factor of 6×10^6 larger compared to the definitely nonresonant case of ^{54}Fe, and, thus, the largest one ever determined.

In order to determine the probability of $\epsilon\epsilon$ capture $\lambda_{\epsilon\epsilon}$ in ^{152}Gd, its nuclear matrix element $M(^{152}\mathrm{Gd})$ was calculated. This calculation is based on the quasiparticle random phase approximation [2]. The single particle energies were obtained with a Coulomb-corrected Woods-Saxon potential. Two-body G-matrix elements were derived from the charge dependent Bonn (CD-Bonn) one-boson exchange potential within the Brueckner theory. The pairing interactions were adjusted to fit the empirical pairing gaps. The particle-particle and particle-hole channels of the G-matrix interaction of the nuclear Hamiltonian H were renormalized by introducing the parameters g_{pp} and g_{ph}, respectively. The calculations were carried out for $g_{ph} = 1.0$. The particle-particle strength parameter g_{pp} of the QRPA was fixed by the assumption that the matrix element of the $2\nu\epsilon\epsilon$ process is within the range $(0, 0.1)$ MeV^{-1}. This assumption is based on the fact that the matrix element $M^{2\nu}_{GT}$ for $\beta\beta$-decay nuclei in this mass region (^{128,130}Te, ^{136}Xe and ^{150}Nd) does not exceed the above range. We note that only ^{130}Te and ^{150}Nd have a directly measured two-neutrino decay rate [30] and that there is a possibility to determine the $2\nu\varepsilon\varepsilon$ matrix element more accurately, if the corresponding Gamow-Teller transition strengths will be measured. Under these assumptions the value of the $0\nu\epsilon\epsilon$ $M(^{152}\text{Gd})$ ranges from 7.0 to 7.4. The calculation performed within the spherical QRPA approach is a good approximation only if the deformations β_2 of initial and final nuclei are comparable [31], as it is the case for ^{152}Gd and ^{152}Sm ($\beta_2(^{152}\text{Gd}) = 0.21$ and $\beta_2(^{152}\text{Sm}) = 0.24$) [32]. For the calculation of the probability of $\epsilon\epsilon$ capture $\lambda_{\epsilon\epsilon}$ we take $M(^{152}\text{Gd}) = 7.0$.

Finally, by substituting the calculated numerical values for the parameters in Eq. (1) and (2), the half-life of the neutrinoless double-electron capture in ^{152}Gd can be expressed as

$$T^{0\nu}_{1/2} \approx 10^{26} \left| \frac{1\ \text{eV}}{m_{\beta\beta}} \right|^2 \text{yr}, \quad (4)$$

where the effective neutrino mass $m_{\beta\beta}$ is given in eV. For the same value of $m_{\beta\beta}$ this value is about 2 orders of magnitude smaller than half-lives of various nuclides used in experiments searching for neutrinoless $\beta\beta$ decay. Nevertheless, an experimental search for neutrinoless double-electron capture in ^{152}Gd is feasible. Such an experiment could be based on a measurement of the full energy of the atomic deexcitations by calorimeters. The existence of a single monoenergetic peak in a calorimetric spectrum with the energy equal to the binding energy of the double-electron hole in the daughter nuclide would clearly indicate that the neutrino is a Majorana particle. A crucial point that makes this experiment superior to all experiments with $\beta\beta$-decay nuclides is the absence of a physical background from the competing two-neutrino $\epsilon\epsilon$ capture due to its very small phase space.

In conclusion, we have determined the $Q_{\epsilon\epsilon}$ value of the double electron $0^+ \to 0^+$ transition between ground states of ^{152}Gd and ^{152}Sm to be 55.70(18) keV by direct measurements of the mass-ratio of $^{152}\text{Sm}^+$ and $^{152}\text{Gd}^+$. This $Q_{\epsilon\epsilon}$ value is very close to the precisely calculated binding energy B_{2h} of two-electron hole KL_1 in the daugther nuclide ^{152}Sm. The degeneracy parameter, $\Delta(^{152}\text{Gd}) = Q_{\epsilon\epsilon} - B_{2h} = 0.91(18)$ keV, is the smallest one among all the known double-electron capture transitions, and results in a substantial resonant enhancement of neutrinoless double-electron capture in ^{152}Gd. The calculated half-life on the level of 10^{26} yr is the smallest among known $0\nu\epsilon\epsilon$-transitions and, thus, ^{152}Gd is a promising nuclide for the search for this fundamental process.

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