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Half-life of ^{184}Re populated by photodisintegration reaction with Laser Compton scattering γ-rays at NewSUBARU

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Abstract. We report a half-life of the ground state of ^{184}Re populated by the ^{185}Re(γ,n)^{184}Re reaction with the Laser Compton scattering γ-rays generated through relativistic engineering. The γ rays are provided at the electron storage ring NewSUBARU. The half-life of the 3^- ground state of ^{184}Re, 38.0 ± 0.5 d, has been taken as a recommended value, which was measured using deuteron-induced reactions. However, this half-life may include a possible contribution from the 8^+ isomer ($T_{1/2}$ = 169 ± 8 d) of ^{184}Re since the presence of the isomer was not known at that time. In contrast, the (γ,n) reaction has the advantage to selectively populate the ground state because this reaction does not bring large angular momentum. The measured half-life of 35.4 ± 0.7 d is shorter than the previous half-life by about 7%. This difference is crucial for applications using the activation method.

1 Introduction

The rare-earth element Re has two stable isotopes ^{185}Re (37.4%) and ^{187}Re (62.6%). Their nuclear data such as the half-life are important for the nuclear physics and astrophysics, for example photodisintegration nucleosynthesis in supernovae [1–6] or ^{187}Re-^{187}Os nuclear cosmochronometer [7–10]. ^{184}Re has a β-unstable isomer with a half-life of 169 d. The (γ,n) production ratio of this isomer to the ground state of ^{184}Re was measured to study the level density and decay property in viewpoint of a statistical-model [11, 12]. An activation method has been used for measurements of nuclear reaction cross sections on Re in these studies [3, 7, 8, 10–12]. The half-life $T_{1/2}$ of the ground state of the populated nucleus is crucial for the activation method since the evaluated cross section is proportional to $T_{1/2}$ of the populated nucleus.

The half-life of the 3^- ground state of ^{184}Re, $T_{1/2}$ = 38.0 ± 0.5 d, was reported by measuring the decay half-life of ^{184}Re in 1962 [13]. This half-life has been prevalently taken as the recommended value. We would like to stress that a β-unstable isomer with $J^\pi = 8^+$ ($T_{1/2}$ = 169 ± 8 d) in ^{184}Re was found in 1963 [14] after the work in 1962 [13]. Therefore, the recommended half-life reported in 1962 has been determined without taking into consideration any contamination effect from the decay of the isomer. In the experiment in 1962, ^{184}Re was populated by a deuteron-induced reaction which brings large angular momentum enough to populate the 8^+ isomer. These historical facts indicate that the true half-life of ^{184}Re is shorter than the recommended value $T_{1/2}$ = 38.0 d. The recommended value has affected cross section results measured with the activation method over the last 40 years because the activation method based on the recommended value provides a cross section larger than the true value. Therefore, one should measure the more precise half-life of the ^{184}Re ground state produced selectively. The (γ,n) reactions have an advantage that the total angular momentum transferred to populated states is lower than those of particle-induced reactions. The spin and parity of the ground state of ^{185}Re is $J^\pi = 5/2^+$, and thus the 3^- ground state of ^{184}Re is expected to be populated with little contamination of the 8^+ isomer by the ^{185}Re(γ,n)^{184}Re reaction.

The progress of the relativistic engineering (for example see ref. [15]) provides a new γ-ray source with a MeV energy range [16–18]. These γ-rays are generated by Compton scattering of relativistic electrons by laser photons. The National Institute of Advanced Industrial Science and Technology [16] and Duke Free Electron Laser Laboratory at Duke University [18] have provided the Laser Compton scattering (LCS) γ-rays in the MeV energy range. Recently, a new LCS γ-ray source was installed at an electron storage ring NewSUBARU [19] in SPring-8. We measure the half-life of the ground state of ^{184}Re selectively populated by (γ,n) reactions with the LCS γ-rays at NewSUBARU.

2 Experiment

A Nd:YVO$_4$ laser system and a nuclear experiment room with a heavy shield locate at the outside of the electron storage ring NewSUBARU [20] is shown in figure 1. The collision of the relativistic electrons and the laser photons occurs at the inside the storage ring and creates a high energy γ-ray. Its energy depends on the angle between the direction of the incident electrons and the generated γ-rays. The diameter

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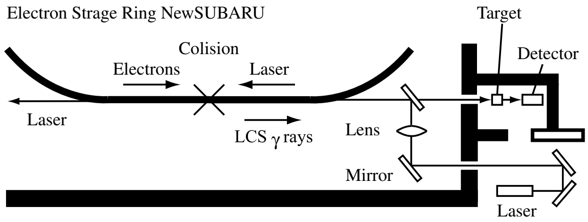

Fig. 1. A schematic view of Laser Compton scattering system at NewSUBARU. The laser is focused by the single lens with f = 5000 cm and the distance between the lens and the collision point is approximately 15 m. The target position is located at about 20 m from the collision point.

of the LCS γ-rays without a collimator is approximately 20 mm at the target position, which is far from the collision point by about 20 m. In the present experiment, we used a Q-switch Nd:YVO_4 laser system that provided laser photons with a wavelength of 1064 nm at 100 kHz. The laser power was 4 W and the estimated γ-ray flux was $0.5 \sim 1.5 \times 10^6$ photons/s with an energy range from 3.3 MeV to 16.7 MeV. This maximum energy of the LCS γ-rays was higher than the peak energy of the giant dipole resonance (GDR), and thus ^{185}Re was effectively transmuted to ^{184}Re via the GDR excitation.

We used natural Re metallic foils. The three stacked Re foils were irradiated by the LCS γ-rays for nine hours. The individual Re foil had a thickness of 0.2 mm and a size of 25 mm × 25 mm. The irradiated targets were cooled for a period of 23 days to reduce the background from short-lived radioactivities such as ^{186}Re ($T_{1/2}$ = 90.64 h) and to obtain the stability of the electronics system. To evaluate the half-life of ^{184}Re, time dependence of γ-ray intensities from the activities was measured for a period of 83 days. The γ-rays followed by β-decay were measured by a HPGe detector with lead shields. The efficiency of the HPGe detector was larger than 70% relative to a 3" × 3" NaI detector. The efficiency was calibrated by standard sources of ^{133}Ba and ^{60}Co and simulated by a Monte Calro code EGS4. The energy resolution was 2.1 keV at 1.3 MeV. The three Re foils were located on a plain in the front of the HPGe detector. The measurement system was almost stable and the peaks of the γ-rays shifted by only one or two channel/s relative to 3000 ~ 4000 channels during the measurement of 83 days. The data were recorded at a multi channel analyzer and the deadtime was lower than 0.03% because of the low decay rate of the samples.

3 Results and discussion

3.1 Population ratio of the isomer to the ground state of ^{184}Re

Before the evaluation of the half-life from the observed three γ-rays, we discuss the contribution of the isomer populated via the ^{185}Re(γ,n)^{184}Re reaction since the half-life of the isomer is different from that of the ground state. If the isomer is populated in the nuclear reactions, the decay curve of the radioactivity has two component. Figure 2 shows a partial level scheme. The 3^- ground state of ^{184}Re decays mainly to 2^+ and 3^+ excited states in ^{184}W. In contrast the 8^+ isomer deexcites through two decay paths of internal γ-decay and β-decay. The isomer decays to the ground state of ^{184}Re by γ-decay cascades, whose branching ratio is about 75.4%. The ground state of ^{184}Re subsequently decays to ^{184}W. Therefore most γ-rays including the three γ-rays measured in this experiment can be observed in both the decay of the ground state and the isomer, and the half-lives of these γ-rays may both be affected by the decay. In contrast, the decay γ-rays from a 7^- excited state in ^{184}W were observed in only the decay of the isomer because the 7^- excited state is not populated by the decay of the ground state of ^{184}Re (see fig. 2). Therefore, the intensities of the decay γ-rays, for example 216.5, 536.7, 920.9 keV, are of importance to evaluate the population ratio of the isomer in ^{184}Re. In fact these γ-rays were clearly observed from ^{184}Re^m produced by the ^{185}Re(n,2n)^{184}Re^m reaction with neutrons provided from a nuclear reactor in our previous experiment [7].

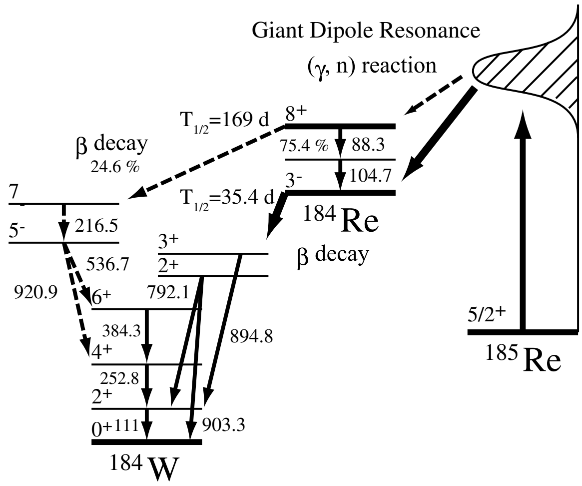

Fig. 2. A schematic view of the ^{185}Re(γ,n)^{184}Re reaction and relevant partial level scheme. The unstable isotope ^{184}Re has a ground state with $J^\pi = 3^-$ and a β-unstable isomer with $J^\pi = 8^+$. The ground state is dominantly populated from the $5/2^+$ ground state of ^{185}Re by the ^{185}Re(γ,n)^{184}Re reaction. The isomer in ^{184}Re decays to the ground state of ^{184}Re through γ-decay cascades and a 7^- excited state in ^{184}W via β-decay.

We cannot observe these γ-rays in the present experiment (see fig. 3). We take an example of the 920.9 keV γ-ray, which cannot be observed as shown in figure 4. This fact indicates that the population ratio of the isomer is negligibly small in the present (γ,n) reaction. We estimate the upper limit of the population ratio of the isomer by taking into account the statistical error of the background around the three γ rays and we obtain the upper limit ~3%. Our experimental result is consistent with previously measured ratios, 1.9% measured by using bremsstrahlung photons whose endpoint energy is 22 MeV [12] and 0.45 ~ 4.5% in an energy range of 10 ~ 20 MeV [11].

We perform a Hauser-Feshbach statistical-model calculation. We use a combination of OPTMAN [22] and GNASH [23] codes. The OPTMAN is a coupled-channel code which gives transmission coefficients to be used in the statistical-model calculation. The optical potential was adjusted to reproduce the cross sections, total and proton and neutron scattering

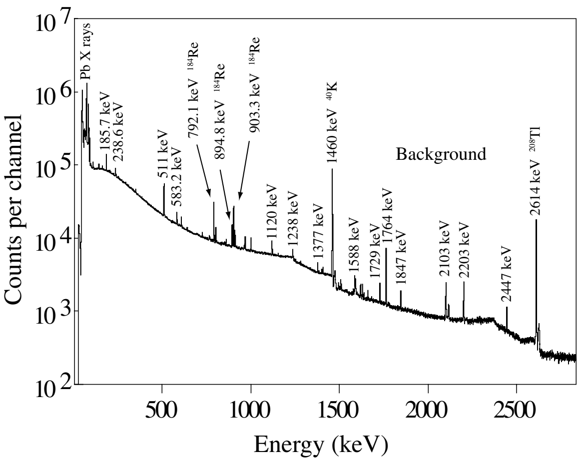

Fig. 3. Summed γ-ray spectrum measured by a HPGe detector for 83 days. The three γ rays of the ^{184}Re decay are clearly observed. The strong γ-rays originate from the background activities.

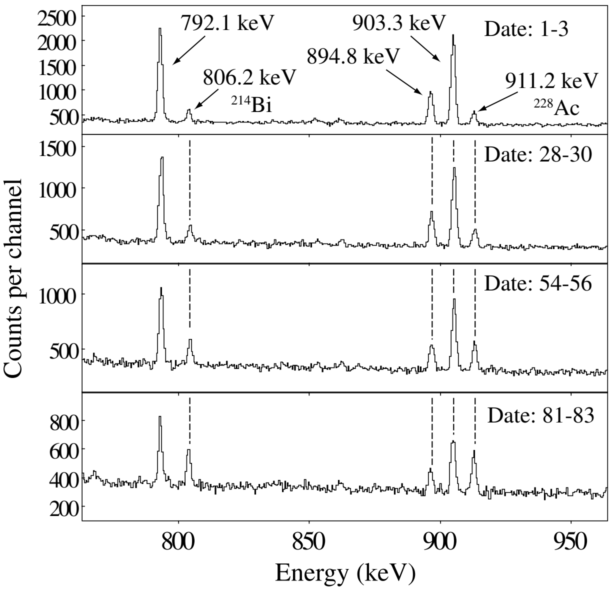

Fig. 4. Measured γ-ray spectra around the three γ-rays of 792.1, 903.3 and 911.2 keV from β-decay of ^{184}Re. It is noted that the two γ-rays of 806.2 and 911.2 keV originate from ^{214}Bi and ^{228}Ac, respectively, and their intensities are constant.

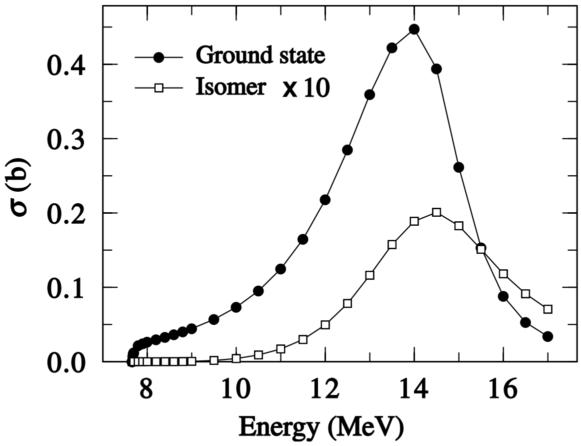

Fig. 5. Calculated (γ,n) reaction cross sections by using a Hauser-Feshbach statistical-model. The circles and squares are the partial cross sections to the ground state and the isomer, respectively.

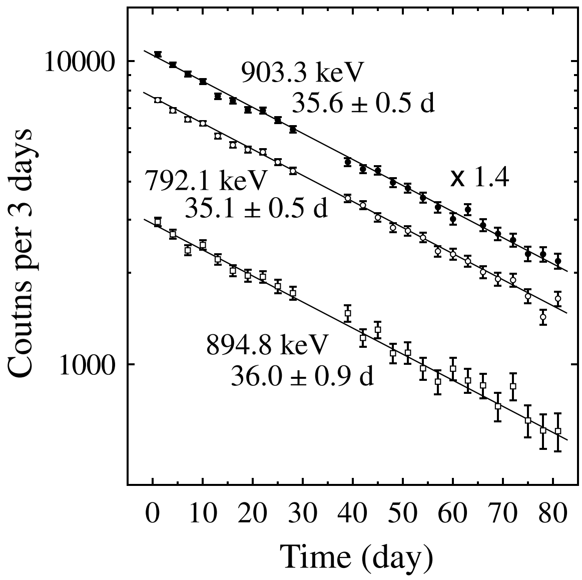

Fig. 6. Decay curve of yields of three γ-rays followed by β-decay of ^{184}Re. Each yield is a peak count in the individual γ-ray spectrum, which is measured for the period of three days. The filled circles, open circles and square are the data for 903.3, 792.1 and 894.8 keV γ-rays, respectively. The half-lives are identical within the uncertainty.

ones, of ^{181}Ta. The GDR parameters of D'Arigo [24] was used in the statistical-model calculation with GNASH. The phenomenological level density formula of Gilbert-Cameron [25], with Ignatyuk's shell correction [26], is used. A similar calculation was carried out for the population ratio of the isomer to the ground state via the ^{185}Re(n,γ)^{186}Re reaction [7]. We present cross sections of the ^{185}Re(γ,n)^{184}Re reaction and the ^{185}Re(γ,n)^{184}Re$_m$ reaction as a function of the incident γ-ray energy (see fig. 5). The cross section to the isomer is lower than that to the ground state by a magnitude of 1 ~ 3 orders. The present calculated result supports the observed experimental fact.

3.2 Half-life of the ground state of ^{184}Re

Three γ-rays of 792.1, 894.8 and 903.3 keV are clearly observed in the measured spectra (see fig. 4). The half-life was evaluated from the decay curves of the three γ-rays. The fraction of the ground state is larger than 97% in ^{184}Re activities and the decay curves are well fitted by a straight line as shown in figure 6. The individual spectrum is recorded for a period of three days. The lack of data in the period of 31 ~ 38 days is due to the maintenance power cut. We obtain the half-life of the individual γ-ray by using χ^2 fitting and the results are 35.1 ± 0.5, 36.0 ± 0.9 and 35.6 ± 0.5 d for 792.1, 894.8 and 903.3 keV γ-rays, respectively. These three half-lives are identical within the uncertainty. Finally we obtain the half-life of 35.4 ± 0.7 d as the average value of these three γ-rays.

Historically, the measurement of the half-life, 38 ± 1 d, was reported in 1960 [27]. The most precise half-life, 38.0 ± 0.5 d, was reported in 1962 [13] and this was widely taken as the recommended value. In these two studies, Re activities were prepared by using deuteron-induced reactions. After these studies, the isomer with a half-life of 169 d was found by a measurement of decay of activities populated by the neutron-induced reactions in a nuclear reactor. Therefore radioactive samples in the two historical studies [13,27] may include the isomer, but the effect of the isomer was not taken into account. After the discovery of the isomer, two half-lives, 33 ± 3 [14] and 34 ± 5 [28], were reported but their uncertainties are so large that the value of 38.0 ± 0.5 d is within the uncertainty range. In these two measurements, the ^{185}Re(n,2n)^{184}Re reaction [14] and a α+^{186}W reaction with an incident energy of 42 MeV [28] were used and both the ground state and the isomer were populated. It is noted that the half-life measured in the present experiment is consistent with these previous values [14,28] within the uncertainty.

4 Conclusion

We report a half-life of the ^{184}Re ground state populated via the ^{185}Re(γ,n)^{184}Re reaction generated by Laser Compton scattering at electron storage ring NewSUBARU. The ground state of ^{184}Re is dominantly populated in this reaction. The result of the statistical-model calculation is consistent with the present observation. The measured half-life is 35.4 ± 0.7 d. This is about 7% shorter than a recommended value $T_{1/2}$ = 38.0 ± 0.5 d, which was reported in 1962 before a discovery of an isomer with $J^{\pi} = 8^{+}$ in ^{184}Re. The present result provides essential information for applications using an activation method because the cross section should be smaller by about 7% than that based on the previous value. It is inferred that measured half-lives of some unstable nuclei near the β-stability line have never been robust. The LCS γ-rays are useful for a precise half-life measurement.

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