

Half-life of ^{184}Re populated by the (γ, n) reaction from laser Compton scattering γ rays at the electron storage ring NewSUBARU

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We report a half-life of the ground state of ^{184}Re populated by the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction from laser Compton scattering γ rays generated through relativistic engineering. The γ rays are provided at the electron storage ring NewSUBARU. The previous experiment using deuteron-induced reactions has yielded a recommended half-life of the 3^- ground state of ^{184}Re of 38.0 ± 0.5 d, including a possible contribution from the 8^+ isomer ($T_{1/2} = 169 \pm 8$ d) of ^{184}Re since the presence of the isomer was not known at that time. In contrast, the (γ, n) reaction has an 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.

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I. INTRODUCTION

The rare-earth element Re has two stable isotopes: ^{185}Re (37.4%) and ^{187}Re (62.6%). The (γ, n) reactions on these isotopes are important for the following reasons. First, systematic (γ, n) reaction cross sections are crucial for understanding nucleosynthesis by photodisintegration reactions in supernova explosions (γ process or p process) [1–5]. For this reason, the (γ, n) reaction cross sections were measured in the rare-earth region [6–8]. Second, ^{187}Re is known as a nuclear cosmochronometer for the rapid-neutron capture process in supernovae (r process) [9–12]. Therefore, neutron capture cross sections on the Re isotopes are important in using this chronometer. Since measurements of the (n, γ) reactions on unstable isotopes are extremely difficult to make, they are studied by using inverse (γ, n) reactions with photon beams [10–12]. Third, ^{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 from the viewpoint of the statistical model [13,14]. An activation method has been used for measurements of nuclear reaction cross sections in these studies [6,7,9,10,12–14]. 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 , $T_{1/2} = 38.0 \pm 0.5$ d, was reported by measuring the decay half-life of ^{184}Re activated with a deuteron-induced reaction in 1962 [15]. This half-life has been prevalently taken as the recommended value.

Here we would like to point out that a β -unstable isomer with $J^\pi = 8^+$ ($T_{1/2} = 169 \pm 8$ d) in ^{184}Re was found in 1963 [16] after the work in 1962 [15]. Therefore, the most precise half-life reported in 1962 has been determined without taking into consideration any contamination effect from the decay of the isomer. The deuteron-induced reaction generally brings angular momentum large enough to populate the 8^+ isomer. Thus the true half-life of ^{184}Re is shorter than the recommended value $T_{1/2} = 38.0$ d. This recommended value have affected cross-section results measured with the activation method over the past 40 years because the activation method based on the recommended value provides a cross section larger than the true value. Therefore, it is of importance to 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 $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction (see Fig. 1).

Progress in relativistic engineering (for example see Ref. [17]) provides a new γ -ray source with an MeV energy range [18–20]. These γ rays are generated by Compton scattering of relativistic electrons by laser photons. The Duke Free Electron Laser Laboratory at Duke University [20] and the National Institute of Advanced Industrial Science and Technology [18] have provided the laser Compton scattering (LCS) γ rays in the MeV energy range and they have been widely used for applications with photon-induced reactions [10,11,21,22]. Recently, a new LCS γ -ray source was installed at the electron storage ring NewSUBARU [23] in SPring-8. In this paper, we report measurements of γ rays followed by β decay of the ground state of ^{184}Re selectively populated by (γ, n) reactions with the LCS γ rays at NewSUBARU.

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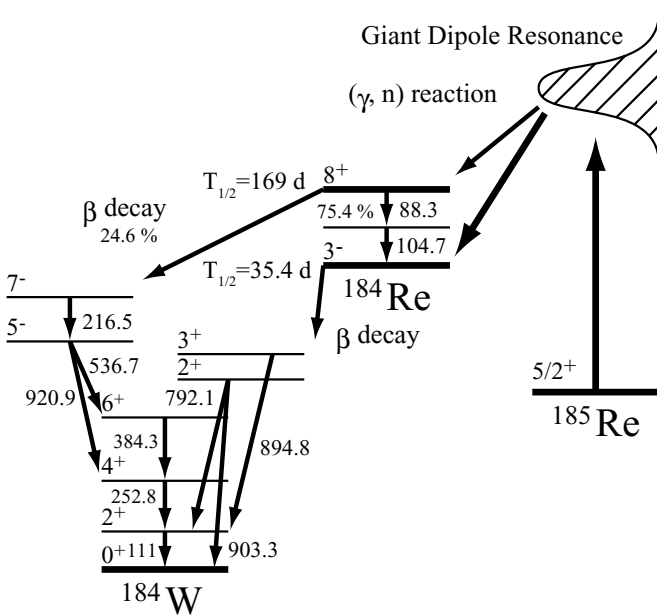


FIG. 1. A schematic view of the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction and partial level scheme. An unstable isotope ^{184}Re has a ground state with $J^\pi = 3^-$ and a β -unstable isomer with $J^\pi = 8^+$. The ground state is dominantly populated from the $5/2^+$ ground state of ^{185}Re by the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction. The isomer in ^{184}Re decays to the ground state of ^{184}Re through γ -decay cascades and an 7^- excited state in ^{184}W via β decay.

We present the upper limit of the integrated cross-section ratio of the isomer to the ground state and the half-life of the ground state of ^{184}Re .

II. EXPERIMENT

Figure 2 shows a laser system and a γ -ray irradiation room with a heavy shield located at the outside of the electron storage ring NewSUBARU [24]. The collision of the relativistic electrons and the laser photons creates a high-energy γ ray, whose energy depends on an angle between the direction of the incident electrons and the generated γ rays. Figure 3 shows a typical energy spectrum of the LCS γ rays. The energy distribution of the LCS γ rays is determined in the basic QED process. The γ -ray intensity is relatively strong at high energies

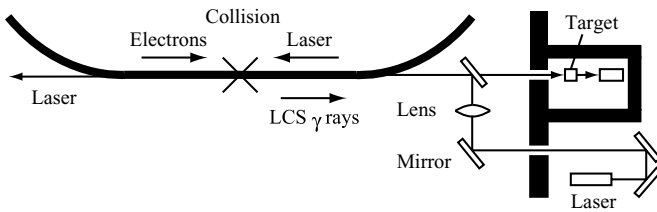


FIG. 2. A schematic view of the laser Compton scattering system at NewSUBARU. The laser is focused by the single lens with $f = 5000$ cm and the distance between the lens and the collision point is approximately 15 m. The target position is located at about 20 m from the collision point.

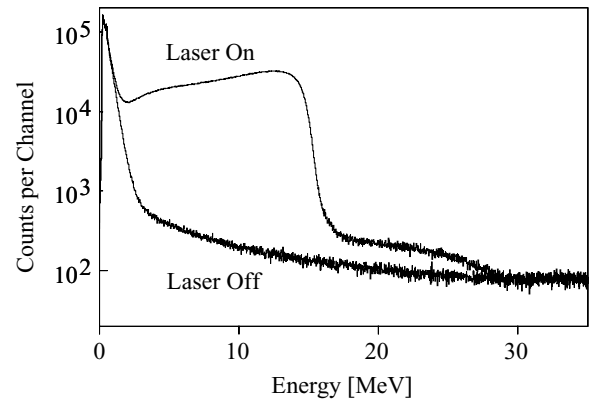


FIG. 3. A typical LCS γ -ray spectrum. The CW Nd:YVO₄ laser provides photons at 1064 nm. The electron current is limited to 10 mA to obtain a clear single peak. The γ rays are measured by a GSO detector with a size of $76 \times 76 \times 180$ mm. A single γ ray and a pile-up γ ray are observed.

and decreases suddenly above the maximum flux energy. The diameter of the LCS γ rays is about 20 mm without a collimator at the target position, which is located at about 20 m from the collision point. The electron storage ring NewSUBARU [24] can store electrons with an energy of 978 MeV up to 230 mA in a top-up mode. The 198 electron bunches circulate in the storage ring at a frequency of 2.5 MHz. An interval time of the electron bunches is about 2 ns.

In the present experiment, we used a Q-switch Nd:YVO₄ laser system that provided laser photons with a wavelength of 1064 nm at 100 kHz. A single laser pulse with a pulse length of 10 ns had a chance to collide with four or five electron bunches in the collision region. The laser power was 4 W and the estimated γ -ray flux was $(0.5-1.5) \times 10^6$ photons/s with an energy range from 3.3 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. No extra collimators were used to obtain the maximum flux in an energy range of 10–16 MeV. Three stacked natural Re metallic foils were irradiated by the LCS γ rays for 9 h. The individual Re foil had a thickness of 0.2 mm and a size of 25×25 mm. The irradiated targets were cooled for a period of 23 d to reduce the background from short-lived radioactivities such as ^{186}Re ($T_{1/2} = 90.64$ h) and to stabilize the electronics system. To evaluate the half-life of ^{184}Re , the time dependence of the γ -ray intensities from the activities was measured for a period of 83 d. The γ rays followed by β decay were measured by an HPGe detector with lead shielding. The three Re foils were located on a plane in the front of the HPGe detector. The efficiency of the HPGe detector was greater than 70% relative to a 3×3 in. NaI detector. The efficiency was calibrated by standard sources of ^{133}Ba and ^{60}Co and simulated by the Monte Carlo code EGS4. The energy resolution was 2.1 keV at 1.3 MeV. The measurement system was almost stable and the peaks of the γ rays shifted by only one or two channels relative to 3000–4000 channels during the measurement of 83 d. The data were recorded by a multichannel analyzer and the dead

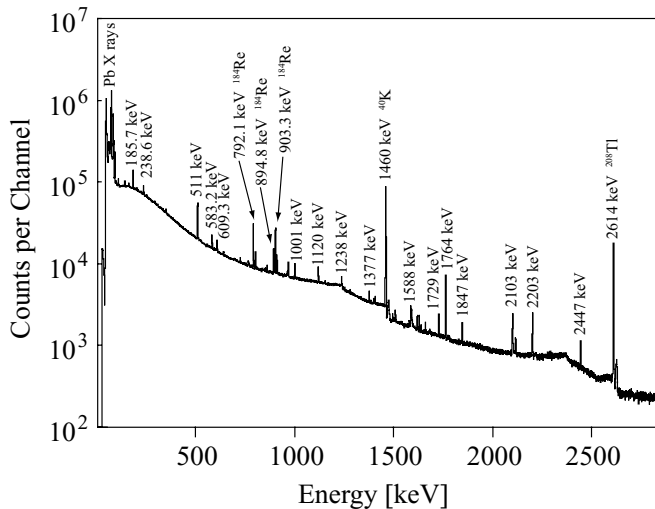


FIG. 4. Summed γ -ray spectrum measured by an HPGe detector for 83 d. The three γ rays of the ^{184}Re decay are clearly observed. The strong γ rays originate from the background activities.

time was less than 0.03% because of the low decay rate of the samples.

Figure 4 shows a sum spectrum for 83 d. The strong peaks are γ rays from the background ^{40}K and ^{208}Tl . Many small peaks originate from the decay chain of the actinides. The element Re has two isotopes, $^{185,187}\text{Re}$. Four radioactivities, $^{184}\text{Re}_{\text{gs}}$ ($T_{1/2} = 38$ d), $^{184}\text{Re}_m$ ($T_{1/2} = 169$ d), $^{186}\text{Re}_{\text{gs}}$ ($T_{1/2} = 90.64$ h), and $^{186}\text{Re}_m$ ($T_{1/2} = 2.0 \times 10^5$ y), are dominantly produced by the (γ, n) reactions. The γ rays of $^{186}\text{Re}_{\text{gs}}$ are not observed since $^{186}\text{Re}_{\text{gs}}$ almost decays out during the cooling time. The population of $^{186}\text{Re}_m$ is expected to be smaller than

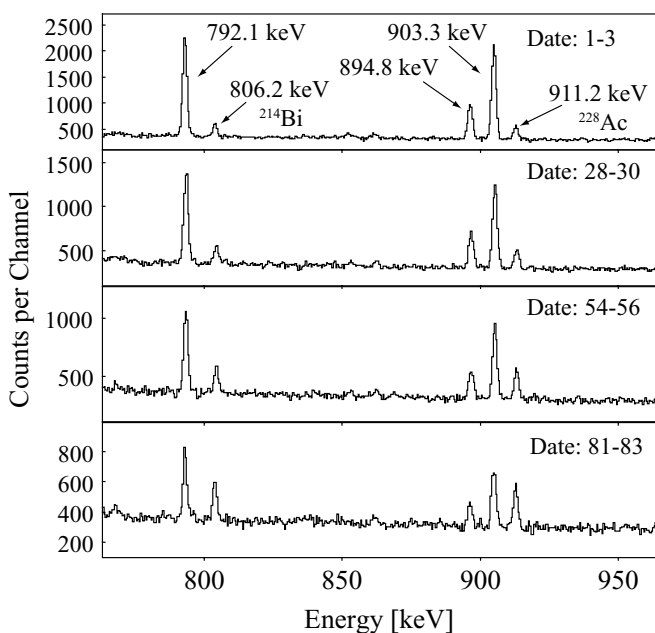


FIG. 5. Measured γ -ray spectra around the three γ rays of 792.1, 894.8, and 903.3 keV from the β decay of ^{184}Re . Note that the two γ rays of 806.2 and 911.2 keV originate from the background ^{214}Bi and ^{228}Ac , respectively, and their intensities are almost constant.

the other activities and the decay rate of $^{186}\text{Re}_m$ is low. Thus γ rays of $^{186}\text{Re}_m$ are also not observed. In contrast, we observe clearly three strong γ rays followed by the β decay of ^{184}Re , whose energies are 792.1, 894.8, and 903.3 keV (see Fig. 5), although their intensities are lower than those of some background γ rays.

III. RESULTS AND DISCUSSION

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

We evaluate an integrated population ratio of the isomer to the ground state via the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction (see Fig. 1). The population ratio is useful for verifying the level density and strength of the γ decay from the viewpoint of the statistical model. In addition, the evaluation of the population ratio is important for the evaluation of the half-life of the ground state of ^{184}Re since the half-life of the isomer is different from that of the ground state. The excited states in ^{184}Re were measured by in-beam γ -ray spectroscopy [25]. The decay scheme of the ground state and the isomer are well known from γ - γ coincidence experiments followed by the β decay of ^{184}Re [26,27]. Figure 1 shows a partial level scheme. The 3^- ground state decays mainly to 2^+ and 3^+ excited states in ^{184}W . In contrast the 8^+ isomer de-excites 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 the decay of both the ground state and the isomer, and the half-lives of these γ rays may be affected by both decays. The isomer populates a 7^- excited state in ^{184}W via β decay. The 7^- state decays to the ground state of ^{184}W through γ -decay cascades. It is noted that the 7^- state decays mainly to a 5^- state at 1285 keV and these two states are not populated by β decay of the ground state of ^{184}Re . Thus the internal decay γ rays of 83.3 and 104.7 keV and γ rays from the 7^- state, for example 216.5, 536.7, and 920.9 keV, are observed only in the isomer decay. In fact these γ rays were clearly observed from $^{184}\text{Re}_m$ produced by the $^{185}\text{Re}(n, 2n)^{184}\text{Re}_m$ reaction with neutrons provided from a nuclear reactor in our previous experiment [9]. We can estimate the population ratio of the isomer to the ground state by using measurements of intensities of these γ rays.

Here we would like to stress that the γ rays from the isomer are not observed in the present measurement; for example, the 920.9-keV γ ray cannot be observed, as shown in Fig. 5. 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 of the isomer by taking into account the statistical error of the background around the five γ rays and we obtain an upper limit of $\sim 3\%$. Our experimental result is consistent with previously measured ratios: 1.9% measured by using bremsstrahlung photons whose endpoint energy is 22 MeV [14] and 0.45%–4.5% in an energy range of 10–20 MeV [13].

To verify the upper limit of the population ratio of the isomer in this experiment, we perform a Hauser-Feshbach statistical model calculation. We use a combination of OPTMAN [28] and

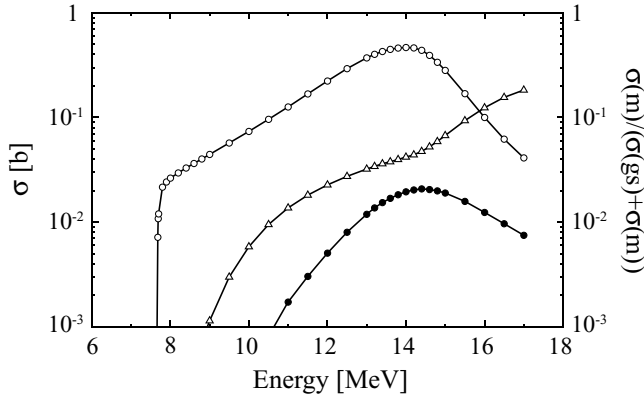


FIG. 6. Calculated (γ, n) reaction cross sections by using a Hauser-Feshbach statistical model calculation. The open and filled circles are the total cross section and the partial cross section to the isomer, respectively. The triangle is the cross-section ratio, $\sigma(m)/[\sigma(m) + \sigma(gs)]$.

GNASH [29] codes. OPTMAN is a coupled-channel code that gives transmission coefficients to be used in the statistical-model calculation. The optical potential was adjusted to reproduce the total and proton and neutron scattering cross sections of ^{181}Ta . The GDR parameters of D'Arigo [30] were used in the statistical model calculation with GNASH. The phenomenological level density formula of Gilbert-Cameron [31], with Ignatyuk's shell correction [32], is used. A similar calculation was carried out for the population ratio of the isomer to the ground state via the $^{185}\text{Re}(n, \gamma)^{186}\text{Re}$ reaction [9]. In Fig. 6, we plot cross sections of the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction and the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}_m$ reaction as a function of the incident γ -ray energy. We also present the cross-section ratio of these two cross sections, $\sigma(m)/[\sigma(m) + \sigma(gs)]$. The cross section to the isomer is lower than that to the ground state by 1–3 orders of magnitude. The ratios of the isomer population to the total are in a range of 0.01%–4% for $E_\gamma = 7.7$ –14 MeV. This calculated result supports our experimental result.

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

We here evaluate the half-life of the ground state. The fraction of the ground state is larger than 97% in ^{184}Re activities. Three γ rays of 792.1, 894.8, and 903.3 keV are clearly observed in the measured spectra (see Fig. 5). We plot yields of these γ rays as a function of time in Fig. 7. The individual spectrum is recorded for a period of 3 d. The lack of data in the period of 31–38 d is due to a maintenance power cut. We obtain the half-life of the individual γ ray by using χ -square 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 [33]. The most precise half-life, 38.0 ± 0.5 d, was reported in 1962 [15] and this was widely taken as

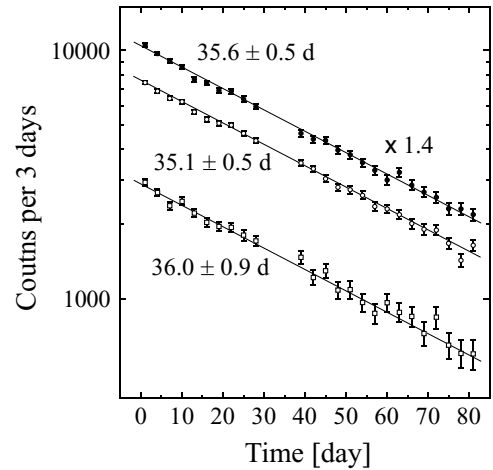


FIG. 7. Decay curve of yields of three γ rays followed by the β decay of ^{184}Re . Each yield is a peak count in the individual γ -ray spectrum, which is measured for the period of 3 d. The filled circles, open circles, and square are the data for 903.3-, 792.1-, and 894.8-keV γ rays, respectively. The half-lives are nearly identical within uncertainty.

the recommended value. In these two studies, Re activities were prepared by using deuteron-induced reactions. After these studies, an isomer with a half-life of 169 d was found by measuring decay activities populated by neutron-induced reactions in a nuclear reactor. Therefore radioactive samples in the two historical studies [15,33] may include this isomer, but the effect of the isomer was not taken into account. After the discovery of the isomer, two half-lives, 33 ± 3 [16] and 34 ± 5 [34], 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 $^{185}\text{Re}(n, 2n)^{184}\text{Re}$ reaction [16] and a $\alpha + ^{186}\text{W}$ reaction with an incident energy of 42 MeV [34] 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 [16,34] within the uncertainty.

As an alternative method that selectively produces the ground state with low spin, a successive β -decay method can be taken (for example see Ref. [35]). A parent nucleus of an odd-odd nucleus such as ^{184}Re in a β -decay chain is an even-even nucleus whose ground state is $J^\pi = 0^+$. The even-even nucleus populates strongly the low-spin ground state of the odd-odd nucleus via β decay. However, both isobars, ^{184}Os and ^{184}W , are stable isotopes in this case and this method cannot be applied to ^{184}Re . We therefore conclude that a measurement with a photon beam such as that provided by the LCS γ rays is practically the best method for this study.

IV. CONCLUSION

We report a half-life of the ^{184}Re ground state populated via the $^{185}\text{Re}(\gamma, n)^{184}\text{Re}$ reaction with relativistic engineering γ rays generated by laser Compton scattering at the electron

storage ring NewSUBARU. The ground state of ^{184}Re is dominantly populated in this reaction and we estimate the upper limit of $\sim 3\%$ as the population ratio of the isomer to the total. Our theoretical calculation result is consistent with this experimental result. The measured half-life is 35.4 ± 0.7 d. This is about 7% shorter than the recommended value $T_{1/2} = 38.0 \pm 0.5$, which was reported in 1962 before discovery of an isomer with $J^\pi = 8^+$ in ^{184}Re . Our 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. This experiment indicates that measured half-lives of all unstable nuclei near the β stability line are not robust and that the LCS γ rays are

useful for a precise measurement even if a high-spin isomer exists in a nucleus of interest.

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