Nuclear excitation of ^{176}Lu by positron annihilation

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An experimental study on the nuclear excitation process of ^{176}Lu by positron annihilation has been made. After irradiation of a natural lutecium foil by positrons from ^{64}Cu, the 88.35-keV γ rays from $^{176}Lu^m$ were observed by the Ge(Li) detector. The cross section of the annihilation-excitation process has been evaluated to be $(9.0 \pm 3.2) \times 10^{-22}$ cm^2. The result is compared with the theoretical value in the two-step model and possible reasons for the discrepancy between theory and experiment are discussed.

[NUCLEAR REACTIONS ^{176}Lu; evaluated excitation cross section by positron annihilation with K-shell electrons.]

I. INTRODUCTION

Nuclear excitation by positron annihilation is one of the annihilation modes when a positron annihilates with an electron strongly bound to the nucleus. This annihilation process was first predicted by Present and Chen in 1951.[1] They considered the possibility that this process was described by a two-step model: Positron annihilation with emission of a photon converging on the nucleus takes place and then the nucleus is excited by absorbing this photon in a similar way to the photonuclear excitation. In this two-step model, the cross section for the annihilation-excitation process of ^{115}In to the principal activation level was estimated to be $\sim 10^{-26}$ cm^2.

The first experimental evidence of this annihilation process was established by the present authors for a ^{115}In nucleus irradiated by positrons from ^{22}Na in 1972,[2] and a more refined experiment was performed for the same nuclide by using a much stronger ^{64}Cu positron source.[3] Our experiments showed that the cross sections for nuclear excitation of ^{115}In by positron annihilation are $(3.9 \pm 1.4) \times 10^{-24}$ cm^2 for the 1078-keV excitation level and $(1.4 \pm 0.5) \times 10^{-22}$ cm^2 for the 1464-keV level. Recently we have investigated the annihilation-excitation process for ^{111}Cd.[4] Only the upper limit of the cross section was estimated to be 8.6×10^{-25} cm^2 because of poor counting statistics.

Vishnevskii *et al.*[5] performed experiments similar to ours for In and Cd targets in 1979. They determined the cross sections of the annihilation-excitation process for ^{113}In and ^{115}In to be $(1.9 \pm 1.0) \times 10^{-24}$ cm^2 and $(4.8 \pm 2.1) \times 10^{-24}$ cm^2, respectively. The cross section for ^{115}In is in good agreement with our result for the 1078-keV level. For ^{111}Cd, however, they did not estimate the cross section, because the excitation level of ^{111}Cd was not clear.

The probability for the nuclear excitation process by positrons from radioactive sources is small and it is, in general, very difficult to observe this process except for a few nuclides. Recently we have shown[6] that the cross section of the photoexcitation process for ^{176}Lu is much larger than that for ^{115}In. Such a large cross section encourages us to study the nuclear excitation process by positron annihilation for this nuclide. In the present paper, the nuclear excitation of ^{176}Lu by positron annihilation has been investigated. This process was confirmed by observing the isomeric activity of ^{176}Lu induced by positron annihilation.

In contrast to the cases of ^{111}Cd and ^{115}In, for ^{176}Lu there is little information on excitation levels which can be excited by photons and cascade down to the isomeric state. The energies and spins of the excited levels in ^{176}Lu were studied by Wasson and Chrien[7] from the γ-ray spectrum emitted by neutron-capture reaction, ^{175}Lu(n, γ) ^{176}Lu. On the other hand, as shown by us,[6] the cross section of the photoexcitation for ^{176}Lu is one order of magnitude larger than that for ^{115}In. On the basis of this fact, it seems to be a reasonable assumption that the multipolarity for the upward transition is not higher than $M2$. Considering that the ground state has spin 7^-, it can be assumed that the spin of the excited level is 5 or 6. In the vicinity of the 1-MeV energy region, several energy levels are reported, but only three of them, 992.0-, 1046.1-, and 1083.4-keV levels, have spin 5 and there are no levels with spin 6. However from these data, it is difficult to determine the energy level excited by positron annihilation. The lowest excited level produced by positron annihilation is 1078 keV in ^{115}In[3] and 1330 keV in ^{111}Cd.[4] Considering these energies, we assume that only the 1083-keV level in ^{176}Lu can be excited by positron annihilation, because this level has the energy nearest to the excited levels in other nuclides.

The isomeric state of ^{176}Lu (127 keV, 3.68 h, 1^-) does not decay to its ground state,[8] but feeds to the excited level (60.4%) and to the ground state (39.6%) of ^{176}Hf by β decay.[8] From the excited level of ^{176}Hf, the 88.35-keV γ ray is emitted. The ground state of ^{176}Lu also decays to the excited level of ^{176}Hf with subsequent emission of the 88.35-keV γ ray. This 88.35-keV γ ray is regarded as the main source of the natural background in the present experiment. The partial level diagram and transitions relevant to the present work are shown in Fig. 1.

II. EXPERIMENTAL

The experiments were undertaken to detect the 88.35-keV γ rays of ^{176}Hf from a natural lutecium foil irradiated by positrons from ^{64}Cu. The positron source was produced by thermal neutron irradiation of a 20-mm diam × 241-mg/cm^2 thick natural copper disk in the Kyoto University Research Reactor (KUR). The total intensity of this ^{64}Cu source is about 17 Ci.

A natural lutecium foil, 20-mm diam × 523-mg/cm^2 thick, was irradiated by positrons just above the Cu source for about 11 h. Between the positron source and the lutecium target, a thin rubber hydrochloride film was inserted to protect the target from radioactive contamination.

After positron irradiation, the 88.35-keV γ rays were measured by the 75-cm^3 Ge(Li) detector for 20 000 s. The detection efficiency of this detector was determined to be $(6.9 \pm 0.1) \times 10^{-2}$ using the standard source ^{109}Cd (88.037 keV).[8] More details of the experimental procedures have been described in our previous papers.[3,4] The number of positrons impinging on the target and the energy distribution of the incident positrons were also estimated by the same method as the previous works.[3,4]

Figure 2 shows (a) the natural 88.35-keV γ-ray spectrum due to the ground-state decay of ^{176}Lu and (b) the observed γ-ray spectrum from the positron-irradiated lutecium foil. This spectrum was obtained as a sum of 10 experimental runs and the contribution of the natural 88.35-keV γ rays from ^{176}Lu is subtracted. In the observed spectrum, we can discern the peak around the expected energy region corresponding to the annihilation-excitation process to be studied.

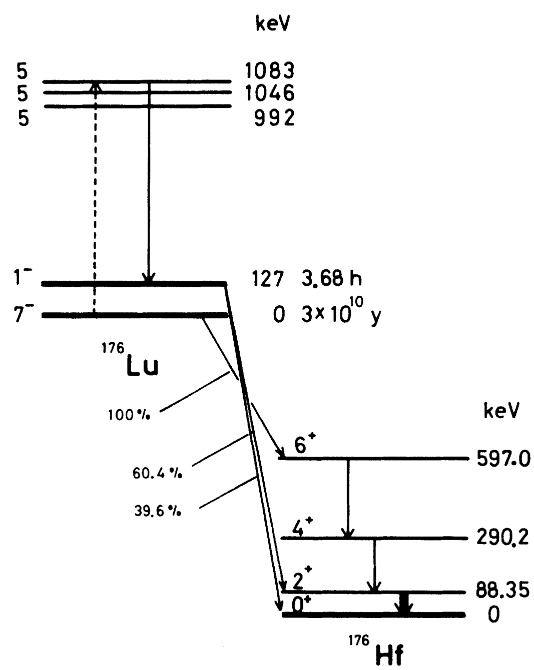

FIG. 1. A partial level diagram of ^{176}Lu, showing only levels and transitions relevant to the present work. Data are taken from Ref. 8.

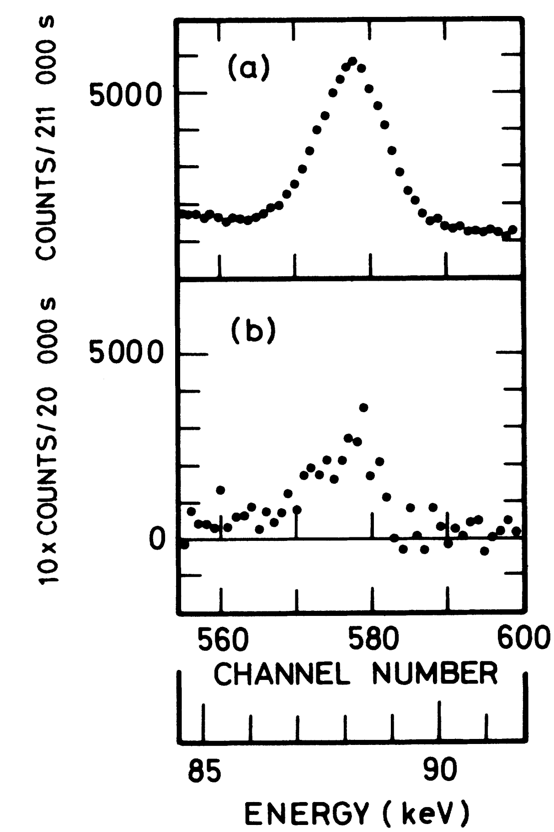

FIG. 2. (a) A profile of the 88.35-keV γ rays emitted from natural lutecium foil due to the ground-state decay of ^{176}Lu, and (b) the observed spectrum of the γ rays from natural lutecium foils irradiated by positrons from the ^{64}Cu source (contribution from the natural radioactivity is subtracted).

III. RESULTS AND DISCUSSION

A. Effective cross section

Before the cross section for the annihilation-excitation process is evaluated, contributions from competing processes should be examined carefully. Since the nucleus ^{176}Lu cannot be excited by Coulomb excitation of the incident positron because of its insufficient energy, possible competing processes are those due to photonuclear excitation. The source of photons which can excite the nucleus are considered to be the following four: viz., (1) photons from the two-quantum annihilation in flight, (2) photons from the single-quantum annihilation, (3) 1346-keV photons of ^{64}Ni emitted from the ^{64}Cu source, and (4) bremsstrahlung radiation of conversion electrons from the 1346-keV state of ^{64}Ni. For the 1083.4-keV level of ^{176}Lu, all four cases were carefully estimated in a manner similar to the previous works[3,4] and it was concluded that they were negligible.

From the observed γ activities resulting from nuclear excitation, we have attempted to evaluate the effective cross section for annihilation-excitation of ^{176}Lu. The effective cross section is defined as the cross section for the total number of positrons incident on the target. Assuming the partial width of the excited level for the transition to the isomeric state is equal to its total width, the effective cross section can be given by

$$\sigma_{eff}=\frac{C_\gamma(1+\alpha)(\lambda_2-\lambda_1)\exp(\lambda_2 t_s)}{N_0 n_p \epsilon R[1-\exp(-\lambda_2 t_m)][\exp(-\lambda_1 t_r)-\exp(-\lambda_2 t_r)]}\ . \tag{1}$$

The symbols in the expression are C_γ, the observed counts of photons from the isomeric state during the measuring period t_m; N_0, the number of target atoms per unit area (natural isotopic abundance of ^{176}Lu is 2.61%)[8] (4.70×10^{19}); n_p, the number of positrons impinging on the target foil per unit time at the beginning of irradiation; ϵ, the overall detection efficiency of the Ge(Li) detector [$(6.9\pm0.1)\times10^{-2}$]; α, the total internal conversion coefficient of the 88.35-keV transition of ^{176}Hf (5.80)[8]; t_r, the period of irradiation by positrons (11.25 h); t_s, the time elapsed between the end of positron irradiation and the start of measurement; λ_1, the decay constant of ^{64}Cu (1.52×10^{-5} s^{-1})[9]; λ_2, the decay constant of $^{176}Lu^m$ (5.23×10^{-5} s^{-1})[8]; and R, the branching ratio of the transition from the 127-keV isomeric state of ^{176}Lu to the 88.35-keV excited level of ^{176}Hf (0.604).[8]

As shown in Fig. 1, the 88.35-keV γ ray is emitted also from the ground state of ^{176}Lu. In order to evaluate the number of the 88.35-keV γ rays due to the annihilation-excitation process of ^{176}Lu, considerable care has been taken to subtract the contribution of the natural 88.35-keV γ rays. Before positron irradiation, the 88.35-keV γ rays emitted from the target were measured until good statistics were attained. The observed γ-ray spectrum was fitted to a Gaussian plus linear background by the method of nonlinear function minimization,[10] and the photopeak area of the natural 88.35-keV γ rays was determined.

Similar procedures were carried out for the positron-irradiated Lu foils, and the difference between photopeak areas before and after positron irradiation was estimated as the induced activity due to the annihilation-excitation process.

Inserting the experimental values of C_γ, n_p, and the numerical values of other factors into Eq. (1), we have obtained the effective cross section to be $\sigma_{eff}=(2.6\pm0.9)\times10^{-29}$ cm^2, as an average of 20 experimental runs. The large error is ascribed to the presence of the natural 88.35-keV γ rays.

B. Cross section

The cross section for the annihilation-excitation process, σ, is related to the effective cross section by the expression

$$\sigma_{eff}=n\sigma\Gamma_{iso}/\Gamma\ , \tag{2}$$

where Γ_{iso} and Γ are the partial level width for the transition to the isomeric state and the total level width of the excitation level, respectively, and n is the ratio of the number of positrons in the target foil within the resonance level Γ to the total number of incident positrons.

In order to evaluate the cross section σ, the total level width and the branching ratio to the isomeric state Γ_{iso}/Γ should be known. To the author's knowledge, however, accurate measurements of the level width of ^{176}Lu have not yet been reported. Therefore, we have estimated the cross section under some assumptions described below.

According to the measurements of Chertok and Booth,[11] the value of the partial width for the direct transition to the ground state $g\Gamma_0$ is $(1.7\pm1.1)\times10^{-3}$ eV for the 1330-keV level of ^{111}Cd and $(2.8\pm0.8)\times10^{-4}$ eV for the 1078-keV level of ^{115}In, where the factor g is $(2J_1+1)/(2J_0+1)$ and J_1 and J_0 are the spins of the excited level and the ground state, respectively. Considering their results, it seems to be a good approximation that the value of the level width $g\Gamma_0$ lies between 10^{-4} and 10^{-3} eV. In the present work, we assume $g\Gamma_0=1\times10^{-3}$ eV, and the cross section σ is calculated using this value.

Veres *et al.*[12] performed photoexcitation experiments on ^{176}Lu. They obtained the partial level width of $g\Gamma_0\Gamma_{iso}/\Gamma = (3 \sim 6)\times 10^{-5}$ eV. As the second assumption, we set the value of $g\Gamma_0\Gamma_{iso}/\Gamma$ to be 4.5×10^{-5} eV. Then the ratio Γ_{iso}/Γ is determined to be 0.045 for $g\Gamma_0 = 1\times 10^{-3}$ eV. Assuming that the total level width Γ is expressed as a sum of the partial widths Γ_0 and Γ_{iso}, the value of Γ is calculated to be 1.4×10^{-3} eV.

The factor n is related to the positron energy distribution inside the target material. Since the main contribution to the nuclear excitation process comes from positron annihilation with the K-shell electrons, the kinetic energy of the positron which can excite the nucleus is given by

$$E_p = W - 2m_0c^2 + B_K\,, \tag{3}$$

where W is the energy of the excited level, m_0 is the electron rest mass, and B_K is the K-shell binding energy. The number of positrons having this energy inside the target was estimated by the same method as the previous works,[3,4] and the value of n was found to be 6.4×10^{-7}.

Combining the effective cross section σ_{eff} with the factor n and the ratio Γ_{iso}/Γ, we have evaluated the cross section for nuclear excitation of ^{176}Lu by positron annihilation to be $(9.0\pm 3.2)\times 10^{-22}$ cm^2.

Theoretical estimation for the cross section has been performed by the two-step model proposed by Present and Chen.[1] In their model, the cross section for the annihilation-excitation process is expressed as

$$\sigma = \sigma_{aq}\,[\sigma_{ph}/(2l+1)\pi k^{-2}]\,, \tag{4}$$

where σ_{aq} is the cross section for the positron annihilation with emission of a spherical wave of 2^l-pole radiation, σ_{ph} is the cross section for the nuclear photoexcitation by this photon, and k is the propagation number of the photon.

Since the spin and parity of the ground state of ^{176}Lu is 7^- and the spin of the 1083.4-keV excitation level is 5, the multipolarity of the upward transition to the excitation level is $E2$ or $M2$. The large value of the cross section for photoexcitation measured by us[6] suggests that the transition is $E2$. The cross section σ_{aq} for the $E2$ transition can be written[3]

$$\sigma_{aq} = \tfrac{8}{9}\pi\,\alpha^4 Z^3(\xi+)^{-11/2}(\xi-1)^{1/2} \times(\xi^2+\xi+3)\,, \tag{5}$$

in relativistic units ($\hbar = m_0 = c = 1$). Here α is the find structure constant, Z is the atomic number of the target material, and ξ is $(E_p + m_0c^2)/m_0c^2$. The cross section for photonuclear excitation of ^{176}Lu was measured by us[6] and found to be $(3.9\pm 2.8)\times 10^{-20}$ for $g\Gamma_0 = 1\times 10^{-3}$ eV. Inserting these values into Eq. (4), the theoretical value of the cross section for the annihilation-excitation process is 1.2×10^{-24} cm^2.

In the present work, nuclear excitation of ^{176}Lu by positron annihilation has been observed by measuring 88.35-keV γ rays of ^{176}Hf fed from the isomeric state ^{176}Lu^m. The cross section for the annihilation-excitation process has been estimated to be $(9.0\pm 3.2)\times 10^{-22}$ cm^2 for the 1083.4-keV excited level on the assumption that the partial width $g\Gamma_0$ is 1×10^{-3} eV. This value is larger than the cross sections for other nuclides, ^{111}Cd ($\sim 8.6\times 10^{-25}$ cm^2) and ^{115}In [$(3.9\pm 1.4)\times 10^{-24}$ cm^2 for the 1078-keV level]. This fact is in agreement with the expectation deduced from the large photoexcitation cross section of ^{176}Lu.[6]

Theoretical studies show that the cross section for the annihilation-excitation process in the two-step model is 1.2×10^{-24} cm^2 for the $E2$ transition. Our experimental value is about 750 times as large as the theoretical one. As has been pointed out in our previous works,[3,4] however, the two-step model proposed by Present and Chen[1] is a simplified model and this process should be treated theoretically in a one-step approach.

The large discrepancy between the measured and calculated values of the cross section may be partially due to estimation of the positron distribution inside the target material. As has been discussed in the previous works,[3,4] we have neglected the contribution from the backscattering effect of positrons in the target. This leads to underestimation of the factor n. If the backscattering effect is taken into consideration, the number of positrons which can excite the nucleus will be larger and the cross section becomes smaller.

Moreover, we have made many assumptions on the nuclear properties of the excited level of ^{176}Lu. Especially, the cross section σ is sensitive to the value of the total width Γ. We have calculated the value of Γ, assuming that $g\Gamma_0 = 1\times 10^{-3}$ eV and Γ is equal to $\Gamma_0 + \Gamma_{iso}$. When $g\Gamma_0$ is one order of magnitude smaller, the value of σ becomes about one half of the present value. We hope to perform more experiments on this nuclide and obtain more accurate information on the excited levels and their nuclear properties.

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