Nuclear excitation of ^{115}In by positron annihilation with K-shell electrons

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When a positron annihilates with a K-shell electron, the excess energy liberated may be given to the nucleus involved. In the case when this energy is just the right energy for any excited state within its level width of a fraction of an electron volt, nuclear excitation can be expected, and if the excited level cascades down to an isomeric state with appreciable lifetime we can confirm the occurrence of this annihilation mode by observing γ transitions or conversion electrons from this isomeric level. In the present work we observed γ rays from $^{115}In^m$ after irradiation of natural indium foils by positrons from the β^+ decay of ^{64}Cu. Using the observed induced γ activity of $^{115}In^m$ and assuming this phenomenon to be the two-step process, we have evaluated the cross sections of nuclear excitation by positron annihilation for 1078- and 1464-keV levels of ^{115}In; $(3.9 \pm 1.4) \times 10^{-24}$ cm^2 and $(1.4 \pm 0.5) \times 10^{-22}$ cm^2, respectively.

[NUCLEAR REACTIONS ^{115}In; evaluated excitation cross sections for 1078- and 1464-keV levels by positron annihilation with K-shell electrons.]

I. INTRODUCTION

When a fast positron collides with an atomic electron strongly bound to the nucleus, there have been known several annihilation processes, single-quantum annihilation,[1] radiationless annihilation,[2] two-quantum annihilation,[3] and nuclear excitation by annihilation. The last mode of positron annihilation was first discussed by Present and Chen[4] in 1951. They have calculated the cross section for excitation of the ^{115}In nucleus to the principal activation level to be ~10^{-26} cm^2 for an incident positron with the total energy $E_+ = 1.10 m_0 c^2$. This process can be considered as occurring in two steps: The single-quantum annihilation takes place with one of the K-shell electrons, and the emitted photon is absorbed by the nucleus in the same atom in the similar way to the nuclear resonance absorption of γ rays.

In this process the energy liberated in annihilation and used to excite the nucleus can be expressed simply by

$$W = E_p + 2m_0 c^2 - B_K , \quad (1)$$

where E_p is the kinetic energy of an incident positron, m_0 is the electron rest mass, and B_K is the binding energy of the K-shell electron in the target atom.

The first experimental evidence of this process was established for ^{115}In by the present authors[5] in 1972. Instead of the nuclear radiation emitted from the excited state produced by annihilation, we attempted to detect the induced activity of the metastable state. We could observe the conversion electrons from $^{115}In^m$, first excited level (4.4 h, 336 keV, $\frac{1}{2}^-$), using natural indium foils and a ^{22}Na positron source. The experimental cross section was estimated roughly to be of the order of 10^{-25} cm^2.

It is the purpose of the present work to improve our previous experiment and to estimate a more accurate cross section for this process. For ^{115}In many excited levels have been observed, but only two of them, 1078 and 1464 keV, are possible to be excited by annihilation and known to cascade down to the 336-keV isomeric level $^{115}In^m$,[6] as shown in Fig. 1. In the present work excitation of these two levels by the process to be studied has been confirmed by measuring γ transition ($M4$) from the 4.4-h isomeric level to the ground state. It is favorable to detect γ rays, because the primary source of large errors in the previous work is ascribed to the background due to the natural β activity in the indium foil. Furthermore, the energy loss and self-absorption effects in the indium foil, estimation of which is very difficult for electrons, are negligible in the case of γ rays.

II. EXPERIMENTAL

As the positron source, pure copper discs of 20-mm diam × 241 mg/cm^2 were used, each of which contained about 10-Ci ^{64}Cu produced by the research reactor of Kyoto University (KUR). This nuclide is known to decay to the ground state of ^{64}Ni through β^+ emission and electron capture as well as to that of ^{64}Zn by β^- disintegration. The maximum energies of the β^+ and β^- transitions are reported to be 655.3 and 578.0 keV, respectively.[7]

Natural indium foils of 20-mm diam × 396 mg/cm^2 were placed on the ^{64}Cu source and irradiated by positrons for about 10.5 h. In order to avoid the radioactive contamination induced on the indium foil by direct contact of the source, as well as to distort the positron energy spectrum slightly so as to get a higher intensity of positrons favorable for the present annihilation mode, a 22-mg/cm^2 thick aluminum foil was inserted between the indium foil and the positron source. Since the half-life of ^{64}Cu is 12.71 h,[8] we could repeat such irradiation runs six times using a rather strong copper source. After the irradiation, the 336-keV γ rays from ^{115}In^m were measured by a 50-cm^3 Ge(Li) detector for 4.5 h.

The accurate number of positrons incident on the surface of the indium foils during irradiation were estimated as follows: We prepared separately a copper source of much weaker intensity but of the same size as those used in the actual experiment. The total number of positrons from this source covered with a 22-mg/cm^2 thick aluminum foil was measured by the use of a 2π gas-flow counter. For this purpose, a known β^--β^+ branching ratio of 39.6/19.3 was used,[7] because the β^+ and β^- transitions have similar maximum energies. And then the absolute intensity of this weak source was determined by measuring the 1346-keV γ rays using a NaI(Tl) detector. A branching ratio of the electron-capture decay of ^{64}Cu feeding to the 1346-keV level of ^{64}Ni was assumed to be 0.6%.[7] The effective positron emission ratio of the positron source was obtained as a ratio of the number of positrons emitted from the weak copper source to the absolute intensity of this source.

The intensity of the strong copper source just before each run of irradiation was estimated by comparing the intensity of the 1346-keV γ rays measured with the NaI(Tl) detector under the fixed geometrical arrangement with that of the weak source. Multiplying the source intensity by the effective positron emission ratio obtained above, the number of positrons incident on the indium foil at the starting point of each run was determined.

Since the annihilation process to be studied has a resonance character, only the positrons with the definite kinetic energy corresponding to Eq. (1) for each excited level can excite the ^{115}In nucleus by annihilation. In order to estimate the number of positrons in the resonance energy region, we must know the energy spectrum of positrons impinging on the indium foil. For this purpose, we prepared the ^{64}Cu source of the same thickness as the strong source in the similar manner. The momentum spectrum of β^+ particles from this copper source covered with a 22-mg/cm^2 aluminum foil was measured by a sector-type double-focusing β-ray spectrometer.

Since the isomeric level concerned, ^{115}In^m, can be also excited by the (γ, γ') reaction, we observed

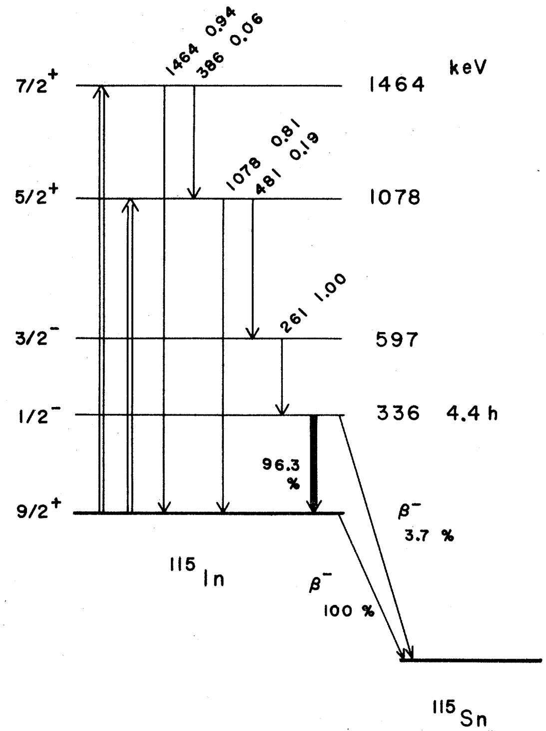

FIG. 1. A part of level diagram of ^{115}In, showing only levels and transitions relevant to the present work. Data are taken from the work of Raman and Kim (Ref. 6).

γ-ray spectrum of a natural indium foil after irradiation in a 700-Ci ^{60}Co irradiation facility, as shown in Fig. 2(a). By this observation the γ-ray transition energy and half-life of ^{115}In^m have been determined as to be 336.9 ± 0.8 keV and 4.4 ± 0.2 h, respectively. In Fig. 2(b) is shown the same γ-ray spectrum induced by the positron annihilation process to be studied. The spectrum was obtained as a sum of the experimental results of typical nine runs.

III. RESULTS AND DISCUSSION

A. Competing processes

The possibility of competing processes which may excite the ^{115}In nucleus from its ground state to higher excited levels cascading down to the 336-keV isomeric level must be examined carefully. In the present case using a ^{64}Cu positron source, from the energetics consideration only four processes of photoexcitation may contribute to the formation of the isomeric levels involved, viz.: (a) photons from the two-quantum annihilation of positrons in flight, (b) photons from single-quantum annihilation of positrons, (c) 1346-keV photons of ^{64}Ni from the source, and (d) bremsstrahlung of conversion electrons from the 1346-keV level of ^{64}Ni. Possible contributions from cases (a) and (b) could be estimated roughly by a consideration similar to that presented in the previous work[5] with the ^{22}Na positron source and found to be less than 0.2% of the observed γ activity. The 1346-keV level of ^{64}Ni is excited through electron capture of ^{64}Cu, but the branching ratio of this transition is only 0.6%.[7] This means that photons which can excite the 1078-keV level of ^{115}In are very meager, because they are produced only by the Compton scattering of the 1346-keV γ photons in the source and the indium foil. Taking account of these facts, photoexcitation due to case (c) is concluded to be negligible. The contribution from case (d) is also quite negligible, much smaller than that from case (c), because the conversion coefficient of the 1346-keV $E2$ transition is only 1.3×10^{-4}.[9]

B. Effective cross section

Using the observed γ activity of ^{115}In^m induced by the positron annihilation process, we have attempted to evaluate the total effective cross section for annihilation-excitation of ^{115}In. The effective cross section is defined as the cross section for the total number of positrons impinging on the indium target. Assuming the ratio of the partial width of each of two excited levels for the transition to ^{115}In^m to its total width to be unity, the total effective cross section for formation of ^{115m}In can be given by the following expression:

$$\sigma_{\text{eff}} = \frac{C_\gamma(1+\alpha)(\lambda_2-\lambda_1)\,e^{\lambda_2 t_s}}{N_0 n_p R\epsilon(1-e^{-\lambda_2 t_m})(e^{-\lambda_1 t_r}-e^{-\lambda_2 t_r})}\,. \tag{2}$$

The symbols in the expression are C_γ = the observed counts of the 336-keV photons during t_m, N_0 = the number of ^{115}In atoms in the natural indium foil per unit area (natural isotopic abundance of this nuclide is 95.67%[10]) (1.99×10^{21}), ϵ = the overall detection efficiency of the Ge(Li) detector for the 336-keV photons from the indium foil [$(4.1\pm0.1)\times10^{-2}$], n_p = the number of positrons incident on the indium foil per unit time at the beginning of irradiation, α = the conversion coefficient of the isomeric 336-keV transition (1.15 ± 0.08),[11] t_r = the period of irradiation by positrons (10.5 h), t_m = the period of measurement of the 336-keV photons (4.5 h), t_s = the time elapsed between the end of positron irradiation and the start of measurement of the 336-keV photons (180 s), λ_1 = the decay constant of ^{64}Cu (1.52×10^{-5} s^{-1}) ($T_{1/2}$ = 12.71 h),[7] λ_2 = the decay constant of ^{115}In^m [$(4.4\pm0.2)\times10^{-5}$ s^{-1}] [$T_{1/2} = (4.4\pm0.2)$ h, the present work], and R = the branching ratio of the 336-keV isomeric transition to the ground state of ^{115}In, provided 3.7% is adopted[11] for the β^- branching to the ground state of ^{115}Sn (0.963).

Using the experimental values of C_γ and n_p obtained for each run of the experiment and adopting the numerical values of other factors in Eq. (2) mentioned above, the effective cross section was evaluated for positrons from the ^{64}Cu source as $\sigma_{\text{eff}} = (1.9\pm0.5)\times10^{-31}$ cm^2, an averaged value of results from 32 experimental runs. The uncertainty is ascribed to those in some factors in Eq. (2) and the poor statistics of measurements of the 336-keV γ rays.

C. Cross sections

The cross sections for excitation of the 1078- and 1464-keV levels, σ_1 and σ_2, were evaluated from the total effective cross section σ_{eff}. According to the definition of σ_{eff}, we have

$$\sigma_{\text{eff}} = (\Gamma_i/\Gamma)_1 n_1\sigma_1 + (\Gamma_i/\Gamma)_2 n_2\sigma_2\,, \tag{3}$$

where Γ_i is the partial width from the excited resonance level to the deexcited 336-keV level and Γ is the total width of the excited level, and n_1 and n_2 are the ratios of the number of positrons in the target foil within the intervals of the resonance level widths, Γ, of the 1078- and 1464-keV levels to the total number of the incident positrons, respectively.

The ratio Γ_i/Γ is determined from the experimental value of Γ_0/Γ, where Γ_0 is the partial width for the direct γ-ray transition from the excited level to the ground state. Using the values of $\Gamma_0/\Gamma = 0.808\pm0.016$ for the 1078-keV level and $\Gamma_0/\Gamma = 0.937\pm0.015$ for the 1464-keV level[12] and taking into account the level scheme in Fig. 1, we obtain $(\Gamma_i/\Gamma)_1 = 0.19$ and $(\Gamma_i/\Gamma)_2 = 0.011$.

In order to estimate σ_1 and σ_2 from Eq. (3), we need some assumptions on the ratio σ_1/σ_2. In the present work this ratio was estimated theoretically, based on the assumption that the annihilation-excitation phenomenon is a two-step process: Annihilation of positrons with the K-shell electron and photoexcitation of the nucleus by the photon emitted in the former process. According to Present and Chen,[4] the cross section can be written

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

where σ_{aq} is the positron annihilation cross section with the K-shell electron with emission of a spherical wave of the 2^l-pole radiation converging on the nucleus, σ_{ph} is the cross section for the nuclear photoexcitation by this photon, and k is the propagation number of the photon. Following Present and Chen,[4] we calculated the σ_{aq} values for emission of the photons corresponding to the 1078-keV ($E2$) level and the 1464-keV ($M1$) level. The analytical expressions for σ_{aq} are given in the Appendix. Provided the binding energy $B_K = 27.94$ keV,[13] the kinetic energies of positrons corresponding to the ex-

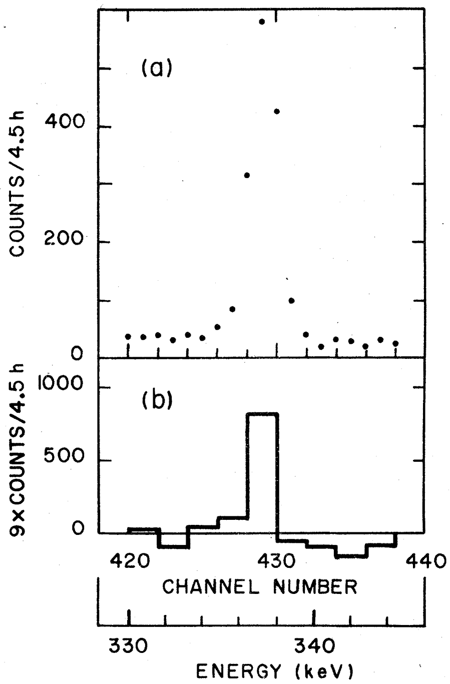

FIG. 2. Observed spectra of γ rays from a natural indium foil. (a) After irradiation by γ rays in a 700-Ci ^{60}Co irradiation facility, by (γ, γ') reaction. (b) After irradiation by positrons from a ^{64}Cu source, by the positron annihilation process.

cited levels of 1078 and 1464 keV are determined from Eq. (1) to be 83.9 and 470 keV, respectively.

On the other hand, the nuclear photoexcitation cross section, σ_{ph}, for these two excited levels are estimated from the experimental values of the integrated cross sections for photoexcitation of the isomeric state via the higher-excited levels, $\int \sigma_{iso}(E)\,dE$. Using the Breit-Wigner single-level formula for $\sigma_{iso}(E)$, we find

$$\int \sigma_{iso}(E)\,dE = \frac{\pi}{2}\Gamma_i \sigma_{ph} \,. \tag{5}$$

Chertok and Booth[14] measured the $\int \sigma_{iso} dE$ and $g\Gamma_0$ values for the excited states in ^{115}In by (γ, γ') and (e, e') reactions. The factor g is $(2J_1+1)/(2J_0+1)$, where J_1 and J_0 are the spins of the excited level and the ground state, respectively. From their results, $\int \sigma_{iso} dE = (7.1 \pm 2.3)\times 10^{-26}$ cm^2 eV and $g\Gamma_0 = (2.8 \pm 0.8)\times 10^{-4}$ eV for the 1078-keV level, and $\int \sigma_{iso} dE \doteq (2.5^{+1.5}_{-1.0})\times 10^{-25}$ cm^2 eV and $g\Gamma_0 = (1.8 \pm 0.6)\times 10^{-3}$ eV for the 1464-keV level. Using these numerical values and the Γ_0/Γ values of Dietrich *et al.*,[12] we obtain $\sigma_{ph} = (4.1 \pm 1.7)\times 10^{-22}$ cm^2 and $\Gamma = (5.8 \pm 1.6)\times 10^{-4}$ eV for the 1078-keV level, and $\sigma_{ph} = (6.1^{+4.5}_{-3.3})\times 10^{-21}$ cm^2 and $\Gamma = (2.4 \pm 0.9)\times 10^{-3}$ eV for the 1464-keV level.

Inserting the numerical values thus obtained into Eq. (4), the theoretical cross sections were estimated to be $\sigma_1 = 3.7\times 10^{-27}$ cm^2 and $\sigma_2 = 1.3\times 10^{-25}$ cm^2. The ratio of the cross sections between two levels was evaluated to be $\sigma_1/\sigma_2 = 0.028$.

Since the effect of backscattering of positrons inside the target material cannot be evaluated, it is impossible to estimate the rigorous energy spectrum at any point in the target foil theoretically or experimentally. However, if the energy spectrum at the plane lying at the depth t from the target surface is assumed to be the same as that of positrons passing through the layer of thickness t, we can calculate the energy spectrum of positrons by applying the Blunck-Leisegang theory.[15] As the energy spectrum of the incident positrons on the surface of the indium target, we used the spectrum measured by the β-ray spectrometer previously mentioned. In the present work, the shapes of energy spectra of positrons at ten different depths t_i were calculated, i.e., for $t_i = id/10$ $(i = 1, 2, \ldots, 10)$ where d is the thickness of the indium foil, 396 mg/cm^2. On the other hand the number-transmission coefficients of positrons for indium foil of thickness t_i were measured separately by means of a 2π gas-flow counter. Combining these calculated and measured values and using the values of Γ estimated above, we could evaluate $n_1 = (1.9 \pm 0.5)\times 10^{-7}$ and $n_2 = (3.1 \pm 1.1)\times 10^{-8}$.

From the values of n, Γ_i/Γ, and σ_1/σ_2 together with the measured value of σ_{eff}, the cross sections for the annihilation-excitation process have been obtained from Eq. (3) as

$$\begin{aligned} \sigma_1 &= (3.9 \pm 1.4)\times 10^{-24}\ \text{cm}^2 \\ &\quad \text{for the 1078-keV level,} \\ \sigma_2 &= (1.4 \pm 0.5)\times 10^{-22}\ \text{cm}^2 \\ &\quad \text{for the 1464-keV level.} \end{aligned} \tag{6}$$

D. Concluding remarks

In the present work, we have observed the annihilation-excitation process of positrons by measuring γ rays from the 336-keV isomeric state of ^{115}In. The cross sections for nuclear excitation of the 1078- and 1464-keV levels of ^{115}In were found to be in the order of 10^{-24} cm^2 and 10^{-22} cm^2, respectively. The value for the 1078-keV level is in agreement with the result of the previous experiment obtained from conversion-electron measurement. However, these values are larger than the calculated values in the two-step model by a factor of 10^3.

The possible reasons for large difference between the measured and calculated values are considered to be the following four: First, the theoretical values have been calculated by the very rough two-step approach. In principle, this annihilation mode should be treated theoretically as a one-step process. Moreover, it should be noted that the theoretical values were estimated by using the measured values of σ_{ph}.

Second, we used the Blunck-Leisegang theory to estimate the positron spectrum in the indium foil. This is based on the assumption that the backscattering effect of positrons in the target is negligible. If we take into account this effect, the number of positrons available to the annihilation-excitation process, n_1 and n_2, becomes larger and the measured cross sections, σ_1 and σ_2, decrease.

Third, the experimental cross sections were estimated from the measured value σ_{eff} by the use of the theoretical σ_1/σ_2 ratio. The values of σ_1 and σ_2 are sensitive to the value of σ_1/σ_2. The present value was obtained from the two-step theory for the annihilation-excitation process. When we choose a different value for this ratio, the experimental cross sections, especially σ_2, change considerably.

Finally, in order to estimate both theoretical and experimental cross sections, we used many nuclear parameters and the photoactivation cross sections for the excited levels of $^{115}In^m$. The number of experiments for these values is not large, and experimental errors are very large. The experimental results scatter each other.

Reflecting on the present work, a rigorous theoretical study of this annihilation mode is required. It is also hoped to perform more accurate measurements of nuclear properties of the excited states of ^{115}In, such as Γ and Γ_i/Γ values, and of the photoactivation cross section $\int \sigma_{iso} dE$.

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APPENDIX

The cross section σ_{aq} for annihilation of the positron with a K-shell electron, with emission of a spherical wave converging on the nucleus can be written in relativistic units ($\hbar = m_0 = c = 1$) by[4]

$$\sigma_{aq} = 2\alpha \frac{E_+}{p_+} \sum \left| \int \psi_f^* (V + \vec{\alpha} \cdot \vec{A}) \psi_i \, d\tau \right|^2, \qquad \text{(A1)}$$

where α is the fine-structure constant, $\vec{\alpha}$ is the Dirac matrix, $\sum$ denotes the average over the initial states and sums over final states, ψ_i is the wave function of the incident positron, ψ_f that of the K-shell electron, and p_+ and E_+ are the momentum and the total energy of the incident positron. The normalization is taken to be one photon passing through a sphere about the nucleus per unit time.

The radiation field is represented by the scalar V and vector potentials $\vec{A}$. According to Rose,[16]

$$\vec{A} = [2/\pi l(l+1)]^{1/2} h_{l-1}^{(1)}(kr)(r\,\mathrm{grad} + l\vec{r}/r) Y_l^m, \qquad \text{(A2a)}$$

$$V = i[2l/\pi(l+1)]^{1/2} h_l^{(1)}(kr) Y_l^m, \qquad \text{(A2b)}$$

for the electric 2^l multipole field, and

$$\vec{A} = -[2\pi/l(l+1)]^{1/2} h_l^{(1)}(kr)(i\vec{r}\times\mathrm{grad}) Y_l^m, \qquad \text{(A2c)}$$

$$V = 0, \qquad \text{(A2d)}$$

for the magnetic 2^l multipole field, where $h_l^{(1)}(x)$ is the spherical Hankel function of the first kind and Y_l^m is a normalized spherical harmonics. These potentials correspond to the normalization that the number of photons emitted per second is $1/\pi^2 k$.

Using the same wave functions as those of Present and Chen[4] and following their methods, σ_{aq} can be written in the following form:

$$\sigma_{aq} = \tfrac{8}{9}\pi\alpha^4 Z^3 (\xi+1)^{-11/2} (\xi-1)^{1/2} (\xi^2+\xi+3) \qquad \text{(A3a)}$$

for $E2$ transition, and

$$\sigma_{aq} = \pi\alpha^4 Z^3 (\xi+1)^{-7/2} (\xi-1)^{1/2}, \qquad \text{(A3b)}$$

for $M1$ transition, where ξ is E_p/m_0c^2.

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