Nuclear excitation of ^{111}Cd by positron annihilation

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(Received 19 July 1979)

Nuclear excitation of ^{111}Cd by positron annihilation has been investigated. The experimental evidence for this process was searched by observing the 245-keV γ rays from $^{111}Cd^m$ after irradiation of natural cadmium foils by positrons from the β^+ decay of ^{64}Cu. For the excited level of 1330 keV in ^{111}Cd, the experimental cross section of the annihilation-excitation process has been estimated to be less than 8.6×10^{-25} cm^2. This upper limit is larger than the simple theoretical estimate. Further improvements of theoretical and experimental studies are discussed.

[NUCLEAR REACTIONS ^{111}Cd; evaluated excitation cross section for 1330-keV level by positron annihilation.]

I. INTRODUCTION

Among various modes of positron annihilation, there exists an annihilation process without emission of radiation. When the positron annihilates with the atomic electron strongly bound to the nucleus, the excess energy liberated in annihilation is given up to the nucleus, resulting in its excitation. This process was first predicted by Present and Chen in 1951.[1] According to their simple estimate, the total cross section for the annihilation process to excite ^{115}In to its principal activation level is in the order of 10^{-26} cm^2.

Neglecting the very small recoil energy of the nucleus, the energy liberated in annihilation and used to excite the nucleus is expressed by the simple relation

$$W=E_p+2m_0c^2-B_K\,, \tag{1}$$

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 annihilation mode was established for ^{115}In by the present authors[2] in 1972. Recently more accurate experimental study was performed by us[3] and the cross sections of nuclear excitation by positron annihilation for 1078- and 1464-keV levels of ^{115}In were evaluated to be $(3.9\pm1.4)\times10^{-24}$ cm^2 and $(1.4\pm0.5)\times10^{-22}$ cm^2, respectively. Up to the present, no experimental study has been reported for other nuclides.

In the present paper, we report the nuclear excitation of ^{111}Cd by positron annihilation. The experiment has been made by observing the γ rays from the isomeric state $^{111}Cd^m$ after irradiation of cadmium foils by positrons emitted from a ^{64}Cu source. For ^{111}Cd, many excited levels have been observed, but only three of them, 740-, 1120-, and 1330-keV levels, are known to be excited by photons and have branches which decay to the ground state via the 396-keV isomeric state.[4] Of these three levels, the excitation probabilities to the 740- and 1120-keV levels by photon irradiation are more than three orders of magnitude smaller than that to the 1330-keV level.[4] In the present work, only the 1330-keV level is considered to be excited by positron annihilation.

The energy-level diagram of ^{111}Cd is shown in Fig. 1. The nuclear transitions relevant to the present experiment are indicated by the arrows. The isomeric state $^{111}Cd^m$ has a half-life of 48.7 min and decays to the ground state by emission of two γ rays, 151 and 245 keV, in cascade.[5] The nuclear excitation process to be studied has been observed by the use of 245-keV γ rays, since for the preceding transition of 151 keV the internal conversion coefficient ($\alpha_K=1.53$[5]) is much larger than for the 245-keV transition ($\alpha_K=0.0525$[5]).

II. EXPERIMENTAL

The experimental method is similar to that of our previous experiment for ^{115}In and has already been described in detail earlier.[3] Experimental conditions, however, are briefly presented here.

The positron source was produced by thermal neutron irradiation of a 20-mm diam $\times$ 241-mg/cm^2 thick copper disk in the Kyoto University Research Reactor (KUR). The intensity of this ^{64}Cu source is about 20 Ci.

Natural cadmium foils of 20-mm diam and 435-mg/cm^2 thickness were placed just above the positron source and irradiated by positrons for 3.5 h. This period was chosen so as to get maximum induced activity of ^{111}Cd. The target foil was covered with a thin rubber-hydrochloride film to protect

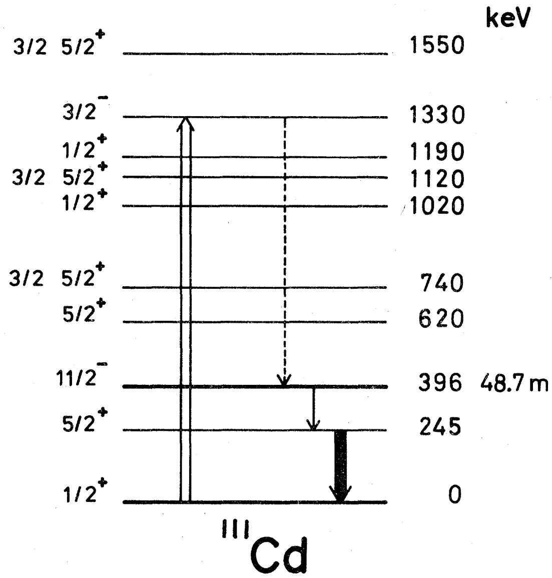

FIG. 1. A part of level diagram of ^{111}Cd. The transitions relevant to the present work are indicated by the arrows. The energies of three levels 740, 1120, and 1330 keV possible to be excited by photons are taken from Boivin, Cauchois, and Heno (Ref. 4). Other data are taken from Raman and Kim (Ref. 5).

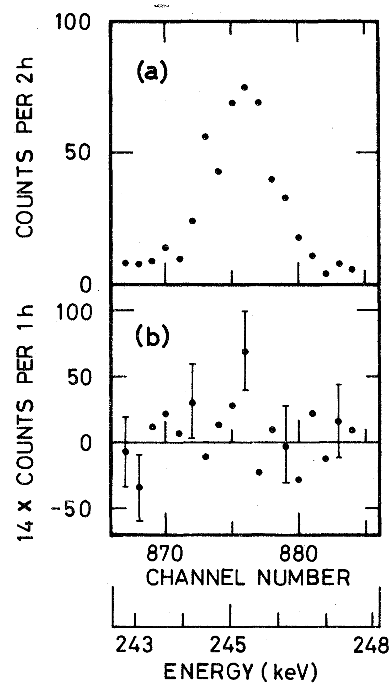

FIG. 2. Observed spectra of γ rays from natural cadmium foils. (a) After irradiation by γ rays in a 700-Ci ^{60}Co irradiation facility, by (γ, γ') reaction. (b) After irradiation by positrons from a 20-Ci ^{64}Cu source, by the positron annihilation process.

radioactive contamination by direct contact to the positron source. After positron irradiation, the 245-keV γ rays from ^{111}Cd^m were measured by a 75-cm^3 Ge(Li) detector for 1 h. The actual number of positrons incident on the target foil was determined in a similar manner to the previous work.[3]

Since this annihilation process has a resonance character, nuclear excitation can take place only for positrons with definite kinetic energy corresponding to Eq. (1). In order to estimate the cross section for this process, we need the number of positrons within the resonance width. For this purpose, the momentum spectrum of β^+ positrons was measured with a sector-type double-focusing β-ray spectrometer by the use of much weaker-intensity ^{64}Cu source of the same thickness as that of the 20-Ci source.

To estimate the reasonable profile of γ-ray spectrum from ^{111}Cd^m, a natural cadmium foil was irradiated in the 700-Ci ^{60}Co irradiation facility of our Institute[6] and photons from the isomeric level ^{111}Cd^m produced by (γ, γ') reaction were observed with the same measuring system as that used in the actual experiment. The result is shown in Fig. 2(a). From this observation the energy of γ rays and the half-life of the isomeric state have been determined to be 245.3 ± 0.4 keV and 49.7 ± 4.3 min, respectively.

III. RESULTS AND DISCUSSION

A. Effective cross section

The observed γ-ray spectrum from cadmium foils irradiated by positrons is shown in Fig. 2(b). This spectrum was obtained as a sum of the experimental results of typical 14 runs. The poor statistics make it difficult to discern distinctive features of a peak. Nevertheless, there is undoubtedly an excess of counts above the background around the peak position to be expected. Moreover, at the γ-ray energy of the isomeric transition in ^{111}Cd, there is a suggestion of a peak. However, considering the poor statistics of the present experiment, we cannot accept this as a definite observation of the isomeric transition. The counting rate under the peak was therefore used to set an upper limit of the process to be studied.

In order to deduce the upper limit of the cross section for nuclear excitation by positron annihilation, we must carefully examine the possibility of the competing processes which may excite the ^{111}Cd nucleus from the ground state to higher excited levels cascading down to the 396-keV isomeric state. As discussed in a previous paper,[3] the nuclear excitation by photons from the following four processes may contribute to the formation of this isomeric state, 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 positron source, and (4) bremsstrahlung of conversion electrons from the 1346-keV state of ^{64}Ni. For the 1330-keV excited level of ^{111}Cd, all of these possibilities were carefully estimated in a similar way to the previous cases of ^{115}In,[2,3] and concluded to be negligible.

Using the observed γ activity of ^{111}Cd^m induced by positron annihilation, the effective cross section for isomer production of ^{111}Cd by the annihilation-excitation process has been evaluated. This cross section is defined as the cross section for the total number of positrons impinging on the target. In the first approximation, we assume that the partial width of the excited level for transition to the isomeric state is equal to its total width. Then the effective cross section can be expressed as

$$\sigma_{eff} = \frac{C_\gamma (1+\alpha)(\lambda_2 - \lambda_1)\exp(\lambda_2 t_s)}{N_0 n_p \epsilon [1-\exp(-\lambda_2 t_m)][\exp(-\lambda_1 t_r) - \exp(-\lambda_2 t_r)]} . \tag{2}$$

The symbols in the expression are as follows: C_γ = the observed counts of the 245-keV photons during the measuring period t_m, N_0 = the number of ^{111}Cd atoms in the natural cadmium foil per unit area (natural isotropic abundance of this nuclide[5] is 12.75%) [2.97×10^{20}], ϵ = the overall detection efficiency of the Ge(Li) detector for the 245-keV photons from the cadmium foil [$(4.8 \pm 0.1) \times 10^{-2}$], n_p = the number of positrons impinging on the cadmium foil per unit time at the beginning of irradiation, α = the total internal conversion coefficient[7] of the 245-keV transition of ^{111}Cd (0.0619), t_r = the period of irradiation by positrons (3.5 h), t_s = the time elapsed between the end of positron irradiation and the start of measurement of the 245-keV photons (180 s), λ_1 = the decay constant of ^{64}Cu [$1.52 \times 10^{-5} s^{-1}$],[8] and λ_2 = the decay constant of ^{111}Cd^m [$(2.32 \pm 0.20) \times 10^{-4} s^{-1}$, corresponding to $T_{1/2} = (49.7 \pm 4.3)$ min, the present work].

Inserting the experimental values of C_γ, n_p, and the numerical values of other factors into Eq. (2), the effective cross section was evaluated to be $\sigma_{eff} = (4.8 \pm 2.8) \times 10^{-32}$ cm^2, as an average value of 16 experimental runs. The error contains the uncertainty due to the counting statistics and the errors of all the factors in Eq. (2). The large uncertainty is ascribed mainly to the poor statistics in measurements of the induced activity. Although the observed value is larger than the estimated uncertainty, this value was considered to be an upper limit of the effective cross section because we could not discern a definite peak, as described above.

B. Cross section

The relationship between the effective cross section and the cross section for annihilation-excitation is given by the expression

$$\sigma_{eff} = n\sigma\Gamma_{iso}/\Gamma , \tag{3}$$

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

The ratio Γ_{iso}/Γ was determined by the relation between the integral cross section for the isomer production by photoexcitation $\int \sigma_{iso}(E)dE$ and the partial width for the direct transition to the ground state of the excited level Γ_0:

$$\int \sigma_{iso}(E)dE = \frac{\lambda^2}{4} g\Gamma_0 \frac{\Gamma_{iso}}{\Gamma} . \tag{4}$$

Here λ is the wavelength of the photon with the resonance energy and 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. Using the values of our previous experimental results,[9] $\int \sigma_{iso}(E)dE = (3.5 \pm 0.4) \times 10^{-25}$ cm^2 eV, and $g\Gamma_0 = (1.7 \pm 1.1) \times 10^{-3}$ eV, obtained by Chertok and Booth,[10] the ratio Γ_{iso}/Γ was found to be 0.099 $\pm$ 0.065.

The total width of the 1330-keV level, Γ, was calculated from the values of $g\Gamma_0$ and Γ_0/Γ based on the following two assumptions: (1) The 1330-keV transition to the ground state is pure $E1(J_1 = \frac{3}{2})$, and (2) the total width of the excited level is equal to the sum of the partial width for the direct transition to the ground state Γ_0 and that for the transition to the isomeric state Γ_{iso}. Boivin *et al.*[4] concluded that the multipolarity for the upward transition to the 1330-keV level is neither $M1$ nor $E2$. Considering the large value of the photoexcitation cross section to this level, it is not reasonable to assume the multipolarity higher than $E2$. From the latter assumption, the branching ratio of the direct transition to the ground state Γ_0/Γ is expressed as $(1-\Gamma_{iso}/\Gamma)$, and the total level width is determined to be $\Gamma = (9.4 \pm 6.1) \times 10^{-4}$ eV.

The factor n was evaluated by the same way as that discussed in the previous paper.[3] Because of difficulty in estimation of the positron distribution in the target, the positron spectrum at the arbitrary point in the target was approximated by the spectrum of positrons passing through the thin layer whose thickness corresponds to the distance from the target surface. In this approximation, the effect of backscattering of positrons in the target material is neglected. Using the measured spectrum, described in Sec. II, as the energy spectrum of the incident positrons, the factor n was calculated to be 4.9×10^{-8}.

Combining the effective cross section σ_{eff} determined above with the values of n and Γ_{iso}/Γ, the upper limit of the cross section for nuclear excitation of ^{111}Cd by positron annihilation has been evaluted to be 8.6×10^{-25} cm^2 by the use of Eq. (3).

According to the two-step model of Present and Chen,[1] the cross section of the annihilation-excitation process is given by

$$\sigma=\sigma_{aq}[\sigma_{ph}/(2l+1)\pi k^{-2}], \qquad (5)$$

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

Provided that the multipolarity of the upward transition from the ground state to the excited level is $E1$, the cross section σ_{aq} is given by

$$\sigma_{aq}=\pi\alpha^2 Z^3\left(\frac{e^2}{m_0c^2}\right)^2(\xi^2+2\xi+3)\times(\xi+1)^{-9/2}(\xi-1)^{-1/2}, \qquad (6)$$

where $\xi=(E_p+m_0c^2)/m_0c^2$. From this equation, σ_{aq} is calculated to be 2.0×10^{-25} cm^2 for the 1330-keV level in ^{111}Cd. On the other hand, the photoexcitation cross section σ_{ph} measured by us[9] is $(2.4\pm1.6)\times10^{-22}$ cm^2.[11] Inserting these values into Eq. (5), the theoretical value of the cross section for annihilation-excitation was estimated to be 2.4×10^{-26} cm^2 in the case of pure $E1$ transition.

IV. CONCLUDING REMARKS

In the present work, nuclear excitation of ^{111}Cd by positron annihilation has been studied by observing γ rays from the isomeric state. The upper limit of the cross section for this annihilation process has been estimated to be in the order of 10^{-25} cm^2. This value is about 36 times larger than the two-step model proposed by Present and Chen.[1] However, their theory is based on rough assumptions, and we have already pointed out in our previous paper[3] that this annihilation mode should be treated theoretically in a one-step model. Recently Grechukhin and Soldatov[12] calculated the cross section for the nuclear excitation by positron annihilation by the use of Weisskopf single-particle nuclear transition matrix element. The cross section for ^{115}In is found to be ~10^{-25} cm^2 for the case of $E1$ transition. It is hoped to perform similar calculations for ^{111}Cd.

Experimentally our counting statistics were poor. In order to estimate the experimental cross section and compare with the theoretical value, it is necessary to measure the induced activity in good statistics. For this purpose, the experiments with much stronger positron sources and enriched ^{111}Cd targets should be performed. Furthermore, there is a problem in estimating the positron spectrum in the target foil. Our method is based on a rough assumption, as has been discussed already in our earlier work.[3] The value of n estimated by our method may be smaller than more rigorous values and this fact leads to the larger value of σ.

Finally we have made many assumptions on the nuclear properties of the excited level of ^{111}Cd, such as energy, spin, multipolarity of the upward transition from the ground state, and branching ratio to the isomeric state. In order to compare the theory and experiment, more accurate data for these properties are needed.

ACKNOWLEDGMENTS

A part of this work was performed in the Research Reactor Institute of Kyoto University. The authors would like to thank Dr. M. Koyama and R. Matsushita for their kind help and valuable advice in preparation of the ^{64}Cu source. Thanks are also due to Dr. Y. Kawase for the use of the β-ray spectrometer and to R. Katano for cooperation in measurements.

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