Nuclear Excitation by Positron Annihilation

Takeshi Mukoyama and Sakae Shimizu
Institute for Chemical Research and Radioisotope Research Center, Kyoto University, Kyoto, Japan
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A new mode of positron annihilation has been investigated experimentally. In this annihilation process without emission of radiation, the excess energy liberated is given to the nucleus involved. The experimental evidence for this process was established by observing the conversion electrons from ^{115m}In, first excited level (335 keV, $\frac{1}{2}^-$), after irradiation of natural indium foils by positrons from the β^+ decay of ^{22}Na. Using the experimental data obtained, we have attempted to evaluate the cross section for positrons having just the energy corresponding to that necessary to excite ^{115}In from the ground state to an excited level which may decay to the isomeric level concerned. The experimental value of the cross section is found to be $\sim 10^{-24}$ cm^2, while the theoretical value estimated by Present and Chen is $\sim 10^{-26}$ cm^2. Possible reasons for this difference are discussed.

I. INTRODUCTION

When a positron collides with an electron strongly bound to the nucleus, in addition to the well-known single-quantum annihilation, two modes of annihilation without emission of radiation can be expected, where the excess energy liberated is given either (1) to another shell electron in the same atom or (2) to the nucleus involved. In the former case (1), a shell electron is ejected from the atom concerned, instead of a photon being radiated. The first experimental evidence of this process, called radiationless annihilation, was established by the present authors[1] in 1965. However, the latter mode of annihilation (2) has not heretofore been observed. This is the reverse of monoenergetic positron emission, which has been observed in the decay of ^{206}Bi, ^{205}Bi, and ^{152}Eu,[2] and it can be interpreted as a special case of single-quantum annihilation where a positron with kinetic energy insufficient to excite or disintegrate a nucleus by collision annihilates with a K-shell electron of the atom with subsequent excitation of its nucleus, as shown by a diagram in Fig. 1. Denoting the kinetic energy of an incident positron by E_p, the energy liberated and used to excite the nucleus can be expressed by the following simple relation:

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

where m_0 is the electron rest mass and B_K is the binding energy of the K-shell electron in the target atom. When W is just the energy difference between an excited level and the ground state of the nucleus, the nuclear excitation by positron annihilation can take place. This mode of annihilation was first predicted by Present and Chen[3] in 1951. They have calculated the cross section for the excitation of the ^{115}In nucleus to the principal activation state to be $\sim 10^{-26}$ cm^2 for an incident positron with total energy $E_+ = 1.10m_0c^2$.

In this paper we report the experimental evidence for this mode of positron annihilation. It has been established by observing the conversion electrons from ^{115m}In, first excited level (335 keV, $\frac{1}{2}^-$), after irradiation of natural indium foils by positrons from a ^{22}Na source. The energy-level diagram of ^{115}In up to 1450 keV is presented in Fig. 2, showing transitions relevant to the present experiment. For this nucleus five excited levels have been observed in the energy region concerned, but only two of them, 1078 and 1450 keV, are known to cascade down to the 335-keV isomeric level.[4] The probability for the process to be studied for positrons from a ^{22}Na source is very small; nevertheless, the observation was possible because of the following advantageous conditions: The half-life of the 335-keV isomeric level is rather long (4.5 h); the total conversion coefficient of an $M4$ isomeric transition from this level to the ground state is nearly unity; and the natural isotopic abundance of ^{115}In is large (95.7%).[5]

II. EXPERIMENT

Taking account of the resonance character of process, positrons from the β^+ decay of ^{22}Na were used. A positron source of rectangular form (14 mm $\times$ 7 mm) containing about 7-mCi ^{22}Na dispersed into Al_2O_3 ceramics was sealed in a welded stainless-steel capsule covered by a thin Mylar film. Natural indium foils of 25 mm diam $\times$ 292 mg/cm^2 were placed at a distance of 2 mm above this source and irradiated by positrons for more than 30 h, and then the induced activity was measured. Measurements of the energy spectrum of the electrons emitted from the foil with a coincidence-type low-background β-ray spectrometer[6] were started immediately after irradiation and continued for 4 h.

This spectrometer consists of a plastic scintillator with a hollow in its lower face, viewed by a photomultiplier tube and a disk-shaped Geiger-Müller (GM) tube with thin windows on both faces. The GM tube is mounted in a hollow cut in the end of the scintillator, and the scintillation pulses gated by the GM counter pulses are recorded with a multichannel analyzer; viz., the GM tube acts as a "gate" of the scintillation spectrometer for electrons from a sample placed under the GM tube. The energy resolution of this system was 19.0% for the 624-keV conversion electrons from ^{137}Cs, and the natural background observed was only 0.7 counts/min in the energy range from 35 keV to 1.8 MeV.

In order to evaluate the background contributions from the natural activity of ^{115}In and other natural radiations, the indium foil was measured for 24 h prior to irradiation. Stability of the whole measuring system was checked using the 624-keV conversion line from ^{137}Cs before and after each experimental and background run. Such a sequence of measurements was repeated 20 times. The relation of energy versus channel number of the multichannel analyzer used was calibrated using conversion-electron lines of several nuclides after a response correction was made in a manner similar to that applied by Jacobs *et al.*[7]

An additional experiment to determine the end-point energy of the very weak natural β^- decay of the ground state of ^{115}In was performed, since a main contribution to the background comes from this natural activity. Using a thin natural indium foil of about 8.0-mg/cm^2 thickness, this energy could be determined to be 495 ± 20 keV from the β-ray energy spectrum observed by means of the measuring system described above. This value is in good agreement with 480 ± 30 keV reported by Watt and Glover,[8] but deviates considerably from 625 ± 70 keV given by Beard and Kelly.[9]

The observed spectrum of emitted electrons, representing the difference in the counting rates of the indium foil with and without positron irradiation, is shown in Fig. 3. To insure that the observed spectrum can be ascribed only to the conversion electrons from the expected isomeric transition, the same foil was irradiated by γ rays in a 2000-Ci ^{60}Co irradiation facility,[10] since the isomeric level concerned can be excited by the (γ, γ') reaction. Comparing the spectra obtained, the observed peak, shown in Fig. 3, was confirmed to be due to the internal-conversion electrons from ^{115m}In.

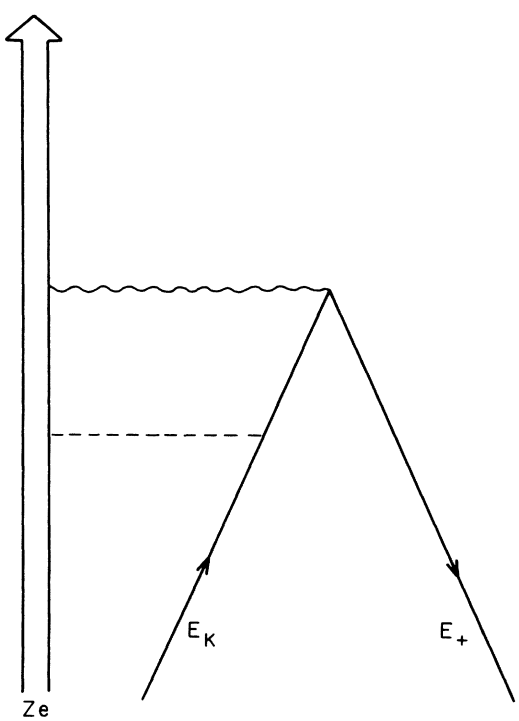

FIG. 1. Diagram for nuclear excitation by positron annihilation. E_+ and E_K are the total energies (including rest mass) of the incident positron and the K-shell electron, respectively.

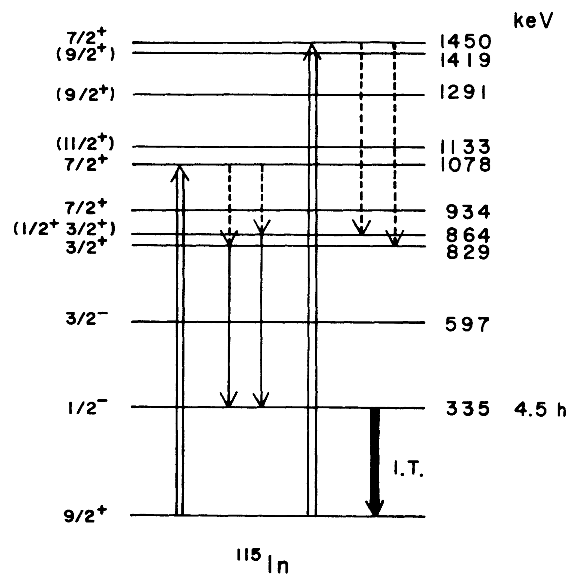

FIG. 2. Energy-level diagram of ^{115}In up to 1450 keV, showing transitions relevent to the present experiment. This diagram is taken from the work of Chertok and Johnson (Ref. 4).

Another experiment to confirm formation of this isomeric level was performed by pursuing the decay behavior of the induced activity in the foil. The indium foils irradiated by positrons were measured by the use of a building-block-type low-background β-ray counter. This system consists of three gas-flow GM counters operated in the anticoincidence mode. The natural background observed was 1.5 counts/min. The decay was followed for 5 h after irradiation. For this period counts were accumulated every 50 min. In order to get good statistics, such a run of measurements was repeated 32 times. From the least-squares fitting of the observed data, the half-life of the induced activity was estimated to be 5.0 ± 3.6 h. Although the experimental error is rather large because of low counting statistics, this value is in agreement with the value of 4.50 h so far reported.[11]

III. DISCUSSION

A. Competing Processes

We must now examine the possibility of competing processes which may excite the ^{115}In nucleus from its ground state to higher excited levels cascading down to the 335-keV isomeric level, viz.:
(1) photoexcitation by:
(a) photons from the ordinary two-quantum annihilation of positrons,
(b) photons from the two-quantum annihilation of positrons in flight,
(c) photons from the single-quantum annihilation of positrons,
(d) the 1.27-MeV photons from ^{22}Na,
(e) natural γ rays from surroundings;
(2) electroexcitation by:
(f) incident positrons,
(g) secondary electrons produced by photons (a)–(e) in the foil,
(h) conversion electrons from ^{22}Na,
(i) emitted shell electrons from the radiationless annihilation of positrons.

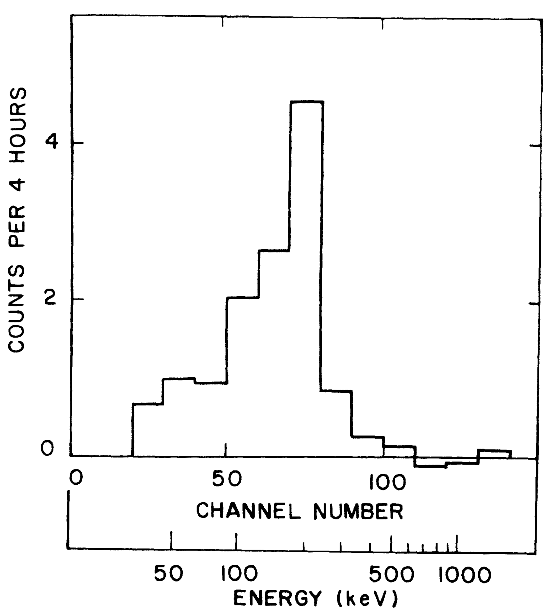

FIG. 3. Observed spectrum of electrons ejected from a natural indium foil after irradiation by positrons from ^{22}Na.

Since the direct formation of the isomeric level by the photo- and electro-excitation is highly forbidden, contributions from cases (a) and (f) can be excluded. For ^{115}In, excepting the 335-keV level, no excited level has been found below 500 keV. Possible contributions from (b) and (c) were estimated on the assumption that all the positrons have the annihilation cross section evaluated appropriately, and both positrons and produced photons traverse a distance corresponding to the full thickness of the foil. Using Bethe's formula[12] for the cross section and energy distribution of emitted photons for annihilation in flight, and taking into account that no excited level cascading to the isomeric level concerned has been found in the energy region below 1078 keV, the minimum kinetic energy of positrons necessary for emission of photons with energies larger than 1078 keV could be estimated to be 390 keV. Thus, for positrons with kinetic energies between 390 keV and the end-point energy (544 keV) of β^+ decay of ^{22}Na, cross sections for the two-quantum annihilation in flight were estimated and the maximum value was adopted. On the other hand, the maximum value of the cross section for the single-quantum annihilation in indium calculated by Johnson, Buss, and Carroll[13] was adopted. Combining these cross sections with the experimental cross section for photoactivation of indium measured by Ikeda and Yoshihara,[14] contributions from cases (b) and (c) were found to be less than 1% of the observed activity. In order to estimate a possible contribution from the case (d), the similar experiment was carried out with the indium foil irradiated by γ rays from a 5-mCi ^{60}Co source placed just above the foil. From the negative result it was concluded that case (d) is not responsible for the observed activity. It is understood that case (e) can be excluded, as the effect of the natural γ rays from surroundings, if it exists, is canceled in the present experiment. Since case (g) is a secondary process, a contribution from this origin can be concluded to be much smaller than those from (b)–(e). Because of a very small conversion coefficient $[(6.77 \pm 0.45) \times 10^{-6}]$ of the 1.27-MeV transition in ^{22}Ne,[15] case (h) can be neglected. Case (i) can be neglected because of its very small probability in comparison with that of the case (c).

B. Cross Section

Using the observed electron spectrum, we have attempted to evaluate the effective cross section for the proposed annihilation process, neglecting a contribution from ^{113}In of low isotopic abundance (only 4.3%). In the present experiment, contributions to the observed activity from two excited states, 1078 and 1450 keV, cannot be distinguished from each other. In a first approximation, assuming the ratio of the partial width of each of these excited levels for the transition to the isomeric level concerned to its total width to be unity, the total effective cross section for these two levels can be expressed as

$$\sigma_{\text{eff}} = \frac{(1+\alpha)CR}{n_F \epsilon a N}, \qquad (2)$$

where R is the induced activity per unit time extrapolated to the end of irradiation, n_F is the effective number of indium atoms in the foil per unit area, ϵ is the detection efficiency of the spectrometer for the K-shell conversion electrons concerned, a is the isotopic abundance of ^{115}In, α is the K-shell conversion coefficient for the isomeric transition, N is the number of positrons incident on the indium foil per unit time, and C is a correction factor for the effect of finite foil thickness.

The value of N was estimated by comparing the intensity of the 511-keV annihilation radiation from the ^{22}Na source covered with a lead plate with that from a thin weak ^{22}Na source with a known intensity covered with the same lead plate, using a 7.5-cm-diam×7.5-cm NaI(Tl) crystal. The detection efficiency ϵ was determined by interpolation using the values for conversion electrons from some nuclides: $\epsilon = 0.24$. The values of a and α are known to be 0.957 and 0.906, respectively.[5] The correction factor C can be defined as a ratio of the number of electrons without the effect of finite thickness of the target to that obtained taking account of the effect. For estimation of this factor, we made assumptions as follows: (1) The factor for the 1450-keV level is the same as that for the 1078-keV level; (2) the positron spectra inside the foil can be given by those of positrons after passing through thin foils of various thicknesses corresponding to distances from the surface; and (3) the number of conversion electrons escaping from the surface of the foil can be obtained by a solution of a transport equation for electrons with a semi-infinite boundary condition. Using the solutions of heat-conduction equations,[16] a solution of the transport equation in the P_2-approximation[17] was corrected for energy dependence of scattering cross section of electrons, as well as for the electron-source distribution estimated from the positron spectra calculated with the Blunck-Leisegang theory.[18] The factor C was thus evaluated to be 1.3. Adopting these numerical values in the right-hand side of Eq. (2), the effective cross section was evaluated for positrons from ^{22}Na as $\sigma_{\text{eff}} = (6 \pm 3)\times 10^{-32}$ cm^2. The estimated uncertainty is ascribed mainly to the poor statistics of measurements of the induced activity.

Using this effective cross section of 6×10^{-32} cm^2, we have also attempted to evaluate the cross section for positrons with the kinetic energy defined by Eq. (1). In this case, for simplicity, we neglected a contribution from the 1450-keV level. The cross section can be given by

$$\sigma = \sigma_{\text{eff}}/n . \qquad (3)$$

Here n is the ratio of the number of incident positrons in an interval of the resonance level width corresponding to the 1078-keV level to the total number.

Assuming the positron spectrum to be the same form as measured by Wenninger, Stiewe, and Leutz[19] and the resonance level width of the state to be 4×10^{-3} eV as estimated by Guth,[20] n was found to be of the order of 10^{-8}. Thus the cross section σ was estimated from Eq. (3) to be of the order of 10^{-24} cm^2. This value is much larger than that predicted by Present and Chen[3] (~10^{-26} cm^2). However, their theoretical value was estimated very roughly and our experimental value was derived based on many assumptions, as mentioned above. In our evaluated value, errors are introduced by neglecting the contribution from the 1450-keV level and by assuming incident-positron spectrum. Recent results of resonance-fluorescence measurements of ^{115}In suggest that the 1450-keV level would have a larger level width than the 1078-keV level.[21] Owing to the scattering effect in the source, our positron spectrum would be considerably different from that measured by Wenninger, Stiewe, and Leutz,[19] especially in the low-energy region. Considering these facts, the experimental cross section σ should be improved; it would be smaller, of the order of 10^{-25} cm^2.

C. Concluding Remarks

From the work reported in this paper, we can say that experimental evidence of nuclear excitation by positron annihilation – a new mode of positron annihilation – has been obtained with natural indium. However, reflecting on the present work, it is hoped that a further experimental study can be made using more refined methods, a stronger positron source, and other nuclides such as ^{107}Ag, ^{111}Cd, ^{103}Rh, ^{87}Sr, and ^{176}Lu. For ^{115}In it would be useful to make use of a low-background γ-ray spectrometer, because a main source of background in the present measurement is due to electrons from the natural activity of this nuclide. Detailed knowledge of nuclear properties of the excited levels of ^{115}In nucleus, such as the level scheme, nuclear spin and parity of levels, multipolarity of upward transitions, and level widths, as well as a more elaborate theoretical treatment of this annihilation mode would be required to interpret this experiment.

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