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PHYSICAL REVIEW C VOLUME 7, NUMBER 4 APRIL 1973

Pair Production in Alpha Decay*

A. Ljubičić† and B. A. Logan

Nuclear Physics Group, University of Ottawa-Carleton University, Physics Department, University of Ottawa, Ottawa, Ontario, Canada
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Electron-positron pairs have been observed in the α decay of ^{241}Am. A result of $(3.1 \pm 0.6) \times 10^{-9}$ pairs per α decay was obtained. It is believed that the pairs are due to internal pair production occurring in the α-decay process. An approximate calculation, which assumes this production mechanism, is in reasonable agreement with the experimental result.

I. INTRODUCTION

Experimental and theoretical investigations of weak interactions in nuclei have indicated the presence of various electromagnetic phenomena which modify the basic decay process. In the conventional perturbation treatment of weak interactions these phenomena are attributed to higher-order terms. These higher-order terms accompanying β decay and shell-electron capture are: (i) internal bremsstrahlung,[1] (ii) ionization and excitation of the electron cloud,[2] and (iii) internal pair production.[3-7] These higher-order effects are much less intense than the first-order processes. For example in the case of internal bremsstrahlung there is about one photon produced per 10^2 decays, and the internal ionization process only occurs about once per 10^4 decays. These two processes have been found in a considerable number of radioactive nuclei and the agreement between the experimental results and the theoretical calculations is reasonably good.

The first estimate of the contribution of the internal pair production (IPP) process in β decay was made by Arley and Møller[3] and, independently, by Tisza.[4] Huang[5] made more sophisticated calculations and evaluated the probabilities of IPP in an allowed β decay for three transition energies for an assumed nuclear charge of $Z=16$. More recently, Richards and Rose[6] have extended these calculations to first-forbidden unique β transitions. It was found that the probability of IPP varies approximately as W_0^4 at higher values of the transition energy W_0. The only experimental results, those of Greenberg and Deutsch,[7] are in good agreement with the theoretical predictions.

It is of interest to investigate the IPP process in α decay as this effect can give additional information on the higher-order phenomena accompanying nuclear transformations. In this work the IPP process in the α decay of ^{241}Am has been investigated and evidence for pair production has been obtained. A preliminary report of this work has been given.[8]

The electron-positron pair in an α-decay process can be produced either by an intermediate virtual state of the nucleus or by the emitted α particle. The energy available is shared between the three particles in a continuous way, satisfying the energy condition

$$W_0 = Mc^2 + 2mc^2 + E_\alpha + E_{e^-} + E_{e^+} . \quad (1)$$

E_α, E_{e^-}, and E_{e^+} are the kinetic energies of the α particle, electron, and positron. W_0 is the transition energy, Mc^2 is the rest mass of the α particle, and mc^2 the rest mass of the electron. E_α, E_{e^-}, and E_{e^+} can take any value between zero energy and the maximum energy $W_0 - 2mc^2 - Mc^2$. The kinetic energy of the recoiling nucleus is neglected. The possible excitations of the lower-energy levels of the daughter nucleus are not considered in Eq. (1).

Unfortunately no theoretical predictions of IPP in α decay are available at present. In order to obtain a rough estimate of the probability for this process we have considered the mechanism for which the electron-positron pair is produced by the emitted α particle. This process can be visualized with a classical picture. The α particles are accelerated in the Coulomb field of the daughter nucleus and emit bremsstrahlung radiation. Photons with energies above $2mc^2$ can produce electron-positron pairs in the nuclear region. In this approximation the probability of IPP, $T_{\alpha e^- e^+}(E)$ can be calculated by multiplying the probability $T_{\alpha\gamma}(E)$ of the internal bremsstrahlung process by the internal pair formation coefficient $\Gamma_{e^- e^+}(E)$. If the relative probabilities are considered this can be expressed as

$$\frac{T_{\alpha e^- e^+}(E)}{T_\alpha} = \frac{T_{\alpha\gamma}(E)}{T_\alpha}\,\Gamma_{e^- e^+}(E), \qquad (2)$$

where T_α represents the probability for the α-particle decay process and E is the total energy of the pair.

The internal bremsstrahlung process has been studied extensively in β decay[1] and in charged meson production.[9] However, no special attention has been paid to the α-decay process and for $T_{\alpha\gamma}(E)/T_\alpha$ we have used the expression obtained for the internal bremsstrahlung for zero-spin mesons being produced with zero orbital angular momentum,[9] modified to allow for the difference between the masses of the α particle and the meson.

The internal pair formation coefficients given by Rose[10] have been used and it has been assumed that electric dipole radiation is predominant in the bremsstrahlung process. When all the possible emission angles are included and Eq. (2) is integrated over the energy range compatible with pair production a ratio $T_{\alpha e^- e^+}/T_\alpha = 1.2 \times 10^{-9}$ is obtained.

II. EXPERIMENTAL PROCEDURE

A diagram of the experimental arrangement is given in Fig. 1. ^{241}Am was chosen as the source. The α decay of ^{241}Am has been investigated thoroughly[11] and only low-energy levels of the residual nucleus ^{237}Np are excited in the decay process. Annihilation radiation was observed when ^{241}Am sources were mounted between a 4.5-cm-diam$\times$5-cm-thick NaI(Tl) scintillation counter and a 40-cm^3 Ge(Li) detector. A coincidence between the counters was used to gate the Ge(Li) counter spectrum which was accumulated in a pulse-height analysis system. The detection efficiency of the system was measured with a calibrated ^{22}Na source. Only the annihilation radiation full-energy peaks were used in the analysis and the random coincidence rate was only about 4% of the true coincidence rate. Two sources A and B were investigated. Source A was obtained over four years ago and had an activity of 230 μCi. Source B was obtained three years ago and had an activity of 160 μCi. The sources were obtained from different manufacturers. Both of the sources consisted of $^{241}AmO_2$ deposited onto platinum backings and the oxide was covered with a thin evaporated gold layer. Mechanical support for the sources was given by a stainless steel backing. The α particles did not have a direct path to the stainless steel.

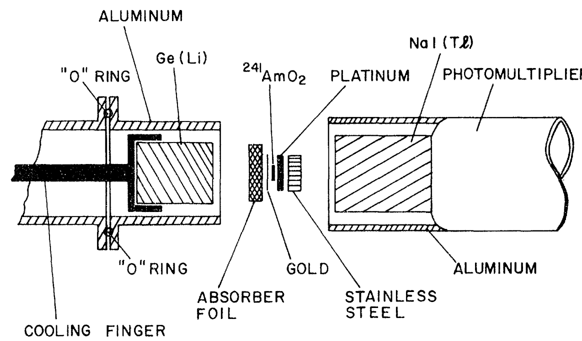

FIG. 1. A simplified diagram of the experimental arrangement.

In the case of source A, measurements were made with both lead and vanadium foils to stop the α particles emerging from the source. A lead foil was used to stop the α particles from source B. The vanadium and lead absorber foils were sufficiently thick so that any positrons emerging from the source would annihilate in them. Positrons emitted in other directions would annihilate in the platinum or stainless steel of the source holders. The α particles stopped in either the absorber foil or its platinum backing.

High-energy photons, originating from cosmic rays and background radioactivity from surrounding material, can interact via pair production in the region between the two counters. The resulting radiation from positron annihilation is an important source of background and special care was taken to estimate the contribution of this. Background corrections were made by subtracting the spectra obtained when the source was removed and a simulated source backing and the relevant absorber foil were placed between the counters. Measurements with a given experimental arrangement typically required a time interval of one week. The maximum background rate in the annihilation peak energy region was obtained with the lead absorber foil in position, and in this case was about half the rate obtained when the source was present.

The number of electron-positron pairs per α-particle decay are given in Table I for each source-absorber combination; $T_{\alpha e^- e^+}$ and T_α represent the probabilities for the IPP process and the α-particle decay process, respectively. Background contributions have been subtracted.

III. CONTRIBUTION OF OTHER POSSIBLE SOURCES OF ANNIHILATION RADIATION

In addition to the high background radiation there are other possible sources of annihilation radiation which can contribute to the measured rate. These other sources have been investigated and are discussed separately in this section.

A. External Pair Production by High-Energy Photons Produced by the α Particles

The probability of α particles producing external bremsstrahlung has been computed assuming an electric dipole production mechanism.[12] The intensity of external bremsstrahlung in the energy regions corresponding to possible pair production was found to be several orders of magnitude smaller than the observed rate. Internal bremsstrahlung associated with the α-particle emission is another possible source of high-energy photons. A rough estimate indicates that this process is expected to be more intense than the external bremsstrahlung.

High-energy photons can also be produced in nuclear reactions initiated by the α particles with the nuclei of platinum, lead, vanadium, and oxygen. In the case of platinum, lead, and vanadium, although some (α, n) and (α, p) reactions are possible, the only reaction of appreciable intensity which is possible is the (α, α') reaction via a Coulomb-excitation mechanism. Data on Coulomb excitation by α particles in the appropriate energy region are available[12,13] and the theoretical cross sections are well known. The experimental results, supplemented by theoretical calculation, indicate that the rate of excitation of nuclear levels with energies above $2mc^2$ is well below the observed rate of pair production.

TABLE I. The values obtained for the number of electron-positron pairs produced per α decay of ^{241}Am. The uncertainties are statistical standard deviations.

Source-absorber combination	$T_{\alpha e^- e^+}/T_\alpha$ (units of 10^{-9})
Source A with a lead absorber	3.4 ± 1.1
Source A with a vanadium absorber	2.9 ± 0.9
Source B with a lead absorber	2.9 ± 1.0

In the case of oxygen the reactions $^{18}O(\alpha, n)^{21}Ne$ and $^{17}O(\alpha, n)^{20}Ne$ are energetically possible. Some data are available in the former case[14] and indicate that the reaction rate in the source can be of the same order as the observed pair production rate. No data are available for the $^{17}O(\alpha, n)^{20}Ne$ reaction. However, the abundance of the ^{17}O isotope is only 18% of that of ^{18}O. The Ge(Li) counter direct spectrum was investigated for possible γ rays from the decay of excited levels of the neon isotopes. The intensities of the possible γ-ray transitions with energies above $2mc^2$ was found to be below a level corresponding to 6×10^{-9} per α decay. Any annihilation radiation due to pair production by these γ rays is well below that necessary to explain the observed production rate.

High-energy photons from the effects discussed above can create annihilation radiation in the source region. Although it is believed that the contributions of these effects to the observed rate are small, an experimental check was made by using the two different absorber foils (lead and vanadium) when source A was being investigated. The probability of pair production in the source vicinity depends strongly on the atomic numbers of the elements in this region. Taking into account the amount and nature of the material between the two detectors we have estimated that the external pair production rate would differ by a factor of 5 for the two experimental arrangements. The results obtained with the different foils are in good agreement and this shows that the contribution of the external pair production, associated with the α particles, is small compared to the observed production rate.

B. Positrons Produced by (α, n) Reactions

The only intense (α, n) reactions possible in our experimental arrangement are the $^{18}O(\alpha, n)^{21}Ne$ and $^{17}O(\alpha, n)^{20}Ne$ reactions. As internal pair formation is much less probable than the probability of γ-ray emission,[15] the negative results of the search for the neon γ rays discussed above show that this is not a serious contribution to the observed pair production rate.

Except for reactions with impurity nuclei none of the (α, p) or (α, n) reactions possible in this experimental arrangement produce nuclei which subsequently decay by positron emission. Assuming the possible impurity levels in the sources, as given by the manufacturers, any positron emission by nuclear reactions with impurity nuclei is several orders of magnitude below the observed rate. The good agreement between the measurements with the different sources is additional evidence that nuclear reactions with impurities are not important.

C. Radioactive Impurities in the Source

There is a possibility that the observed pairs are due to some radioactive impurities which decay by positron emission. As the sources were made by (n, γ) reactions any zinc impurity would be dangerous as the positron emitter ^{65}Zn can be produced by the (n, γ) process. A search was made for the 1114-keV γ ray which follows electron capture in ^{65}Zn. The upper limit on the possible abundance of ^{65}Zn corresponds to a positron emission of less than 3×10^{-10} per α particle.

The Ge(Li) detector direct spectra showed no evidence for γ rays associated with other well known long-lived radio nuclides which decay by positron emission. This investigation and the good agreement between the results obtained with the different sources are good evidence for radioactive impurities not being important.

D. Excitations of the Higher-Energy Levels of ^{237}Np

α particles leaving ^{241}Am can be emitted into the real higher-energy levels of ^{237}Np. If the excited level is higher than $2mc^2$ the electron-positron pair can be produced either internally in the process of deexcitation, or externally after the γ ray has been emitted. The latter mechanism has been discussed in Sec. III A, and it has been shown that its contribution is small compared to the observed rate. It is possible to make a rough estimate of the former process. Baranov, Kulakov, and Shatinsky[16] investigated the particle spectra down to energies corresponding to excitations of ^{237}Np above $2mc^2$. In the energy region corresponding to excitations of ^{237}Np above $2mc^2$ they placed an upper limit for possible excitations of 2×10^{-7} of the intensity of the main decay to the 59.5-keV level of ^{237}Np. This experimental result is in agreement with theoretical considerations as the probability of emission of an α particle decreases very quickly with decrease of the α-particle energy. For ^{241}Am a reduction of the decay probability in excess of 10^6 is expected if the energy of the emitted α particle is decreased by 1 MeV.[17] The probability of pair production is equal to the product of the probability of an α particle being emitted to a nuclear state of excitation E and the internal pair formation coefficient $\Gamma_{e^-e^+}(E)$. For energies just above 1 MeV, $\Gamma_{e^-e^+}$ is of the order of 10^{-4}. This means that the intensity of the created pairs is expected to be less than 10^{-10} pairs per α particle of the main group. As the excitation energy increases the pair production rate will be even lower as the α-decay probability decreases faster than the internal pair formation coefficient increases.

IV. CONCLUSIONS

Evidence for pair production in the α-particle decay process has been obtained. Several possible pair production mechanisms have been investigated and the results indicate that the observed pairs are mainly produced by a IPP process accompanying α decay. If all the results from Table I are combined, a value of $T_{\alpha e^-e^+}/T_\alpha = (3.1\pm0.6)\times10^{-9}$ is obtained. However, it is possible that some external effects or impurity contributions have been overlooked or underestimated. In this case an upper limit of about 4×10^{-9} to the IPP process should be accepted.

A rough estimate of the IPP process in α decay is given in Sec. I. The calculated value is in reasonable agreement with our experimental results. In our approximation only an electric dipole interaction has been considered while in reality other electromagnetic interactions are also possible. We have also neglected the mechanisms for which the pair creation process proceeds via nuclear intermediate states. A more detailed calculation which includes these other processes would be of considerable interest.

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PHYSICAL REVIEW C VOLUME 7, NUMBER 4 APRIL 1973

Atomic Masses of ^{232}Th, ^{235}U, and ^{238}U and a Mass Table for the Heavy Isotopes*

Rodney A. Britten† and Walter H. Johnson, Jr.
School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455
(Received 21 April 1972)

This paper reports the results of the authors' mass spectroscopic measurements on the masses of the isotopes ^{232}Th, ^{235}U, and ^{238}U. Notice is taken of recently reported mass-difference measurements and Q-value measurements, and a least-squares process is used to construct a new mass table for the heavy isotopes. This table differs from Wapstra's 1967 table in two major respects: (1) For most isotopes with $A > 220$ the new mass value is lower than the old mass value by 20–25 μu, or roughly $1\frac{1}{2}$ times the quoted error in the 1967 table. (2) As a result of the new measurements, the uncertainty in our knowledge of the masses of most of the heavy isotopes has been reduced by very roughly a factor of 3.

I. INTRODUCTION

Until recently the heaviest isotopes for which direct, high precision mass measurements were available were isotopes of bismuth and lead. In this paper the authors will present the results of their measurements on ^{232}Th, ^{235}U, and ^{238}U, the isotopes which head the natural-radioactive-decay series.

The authors' measurements on these isotopes were made using a 16-in. magnetic radius double-focusing Nier-Johnson mass spectrometer and the error-signal doublet-peak matching technique. The Minnesota mass-measuring instruments[1-3] and the error signal technique[4] have been described in some detail elsewhere. No further description will be given here. The equipment, when tuned, produced a full-width at half-maximum (FWHM) resolution of 150 000 to 200 000 for these measurements.

Two major problems were encountered in this work. One had to do with sample introduction, and the other was a calibration problem. The calibration problem arose from the fact that the measured values of known mass doublets were consistently too high by about 20 ppm. This consistent discrepancy was discovered by measuring three different types of doublets. Measured values of doublets of the type C_mH_{n+1}-C_mH_n were compared with the accepted value of the hydrogen mass. Measured values of $U^{35}Cl_{m_2}{}^{37}Cl_{n_2}$-$U^{35}Cl_{m_1}{}^{37}Cl_{n_1}$ type doublets were compared with the accepted value of the ^{37}Cl-^{35}Cl mass difference. Finally the sum of the measured values for the C_9H_{10}-$\frac{1}{2}{}^{235}U$ and $\frac{1}{2}{}^{235}U$-C_9H_9 doublets was compared with the accepted hydrogen mass value. In all three cases the measured values were found to be too high by about 20 ppm. This calibration problem for the Minnesota instrument was ultimately resolved by applying a 20 ± 3-ppm correction to all doublet measurements. Other mass measurement laboratories have also had to apply corrections of this sort.

This calibration problem is somewhat similar to a problem encountered by Hudson[5] in 1969 when he used the Minnesota instrument to make measurements on the light rare-earth isotopes. He