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Positron–*K*-Electron Annihilation in ^{180m}Ta Atoms

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Abstract—The excitation cross sections of ^{180m}Ta are measured for the first time via positron–*K*-electron annihilation with a limit energy of 0.653 MeV. Effective cross sections σ^{eff} (^{180g}Ta) are found to be $(3.9 \pm 0.8) \times 10^{-29}$ cm^2. The differential cross section is estimated from the effective cross section and agrees qualitatively with theoretical calculations for *E*1 transitions.

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INTRODUCTION

One possible process in the scattering of a positron beam on atoms is the annihilation of positrons by atomic electrons. One-photon annihilation is prohibited for free particles by the laws of energy and pulse conservation, so the most likely process is the emission of two γ-quanta. If an electron is coupled in an atom, we would then expect one-quantum annihilation, in which cross-section σ for heavy metals can be as great as 10^{-24} cm^2. In addition to one- and two-photon annihilation, so-called photon-free annihilation, in which the energy of an annihilating pair is directly transferred to the core or electron shell of an atom, is also possible.

The conditions of resonance in photon-free annihilation can be satisfied by positrons with energy $E_{\beta+}^{\text{res}} = E_i - 2mc^2 + E_K$, where E_i is the energy of excitation of the *i*-th state and E_K is the bonding energy of *K* electrons.

In photon-free annihilation, the electron shell + core system transitions to a discrete excited state with finite width $\Gamma = \Gamma_i + \Gamma_a$, where Γ_i is the width of core excitation and Γ_a is that of the excited hole state in an electron shell.

Photon-free annihilation has been considered in a number of theoretical and experimental works. Some of these were reviewed in [1]. In recent years, studies have been more theoretical in nature [2, 3], and there is still notable divergence between the experimental and theoretical values of the process's cross section.

The probability of core excitation in photon-free annihilation is quite low, so studies of this process require the use of high-intensity positron beams. Two ways of obtaining positrons are currently the ones most widely used: the recharge reaction (e^-, e^+) in the core field and the radioactive decay of neutron-deficient nuclei. For actual electron currents below 100 mA with energy $E_e \leq 5$ MeV, the density of the positron flux in recharge reactions is less than ~10^7 cm^{-2} s^{-1} even with the birth of pairs in the field of a heavy nucleus. The yield of positrons in radioactive decay can be much higher and depends on the number of radioactive nuclei produced on state-of-the-art setups. The activities achieved on accelerators (e.g., cyclotrons and electrostatic generators) are on the order of 1 Ci (3.7×10^{10} Bq), while those on reactors can be as high as 10^3–10^4 Ci.

In reactions with heat neutrons, we can obtain only four β^+-active sources with characteristics suitable for our experiments: ^{22}Na ($T_{1/2} = 2.6$ years), ^{64}Cu ($T_{1/2} = 12.7$ h), ^{106}Cd ($T_{1/2} = 6.9$ h) and ^{152}Eu ($T_{1/2} = 9.3$ h). In ^{106}Cd and ^{152}Eu nuclei, however, the probability of β^+ decay is 10^{-2}–10^{-4} relative to the total probability of decay, and the typical attainable activities of ^{22}Na are less than 10 mCi; this is not enough to obtain the strong positron flows needed for our experiments. The best source of positrons is thus ^{64}Cu, which can be obtained by irradiating copper of natural isotope composition with neutrons produced in a reactor. The density of this source's positron flow can be as high as 10^{12} cm^{-2} s^{-1}. In addition, the probability of the annihilation excitation of target nuclei can be increased by enlarging the area of contact between the positron source and the irradiated targets. The observed effect can therefore be 2–4 orders of magnitude higher. Since the maximum positron energy in the decay of ^{64}Cu is 653 keV, photon-free annihilation excites only those levels with energies ≤1.6 MeV.

The most reliable experimental data have been obtained by exciting ^{115}In with positrons from the

radioactive decay of ^{64}Cu with a boundary energy $E_{\beta+} = 653$ keV [1]. The structure of the low-lying states in ^{115}In is such that the ^{115}In isomer at these positron energies can be occupied only through the $E2$-transition, while γ-radiation in positron annihilation mainly has $E1$-multipolarity. It is therefore of great interest to obtain experimental data on photon-free annihilation through the $E1$-channel.

One candidate for such studies is ^{180m}Ta. This nucleus exists due to the low-lying isomer state with an energy of 75 keV and $I^{\pi} = 9$, while the ground state is unstable with ($T_{1/2} = 8.1$ h, $I^{\pi} = 1^{+}$) (see Fig. 1). In addition, ^{180m}Ta is a rare naturally quasi-stable, high-spin isotope with a relative content of only 0.012%.

The mechanism of this element's formation in astrophysical processes has never been described using state-of-the-art models. Studies of photon-free annihilation on tantalum are of great practical and fundamental interest from this viewpoint as well.

Studies of ^{180m}Ta excitation in the (γ, γ′)-reaction with braking γ-quanta show the energy range of 1.1–2.0 MeV contains levels of activation that can also arise upon photon-free annihilation [4].

The aim of this work was to study the excitation of ^{180m}Ta in the photon-free annihilation of positrons by irradiating natural tantalum targets with positrons from the decay of ^{64}Cu ($T_{1/2} = 12.7$ h) with boundary energies of 653 and 578 keV for positrons and electrons, respectively.

EXPERIMENTAL

Using the conventional procedure described in [1], targets of tantalum foil 50 μm thick were subjected to radiation in a multilayer assembly with irradiated copper foil. The copper and tantalum foils were alternated to increase the effective surface area of irradiation. The copper samples were 0.3 mm thick. This was due to both the mean free path and the positron distribution measured earlier at the output of an activated measured copper slab [1]. Our ^{64}Cu was activated on a BBP-10M reactor, and the layers were assembled in hot chambers. A total of four tantalum foils were irradiated in the assembly. The positron flow was $(4.0 \pm 0.1) \times 10^{10}$ β^{+} s^{-1} cm^{-2}.

Positron irradiation of the target induced the photoexcitement of tantalum by γ-quanta with an energy of 1345 keV of ^{64}Cu and by γ-quanta of the one-quantum annihilation of positrons. To assess the photoexcitation of tantalum, positrons were cut off with a lead absorber150 μm thick, and irradiation was then performed in the same multilayer geometry using new tantalum foils. Targets were irradiated with positrons for 8 h and photoexcited for 16 h. The one-quantum annihilation spectrum of γ-quanta on lead atoms was more pronounced than the one in the combined γ-quantum–positron irradiation in the main experiment, since the one-quantum annihilation cross-section is ~Z^5.

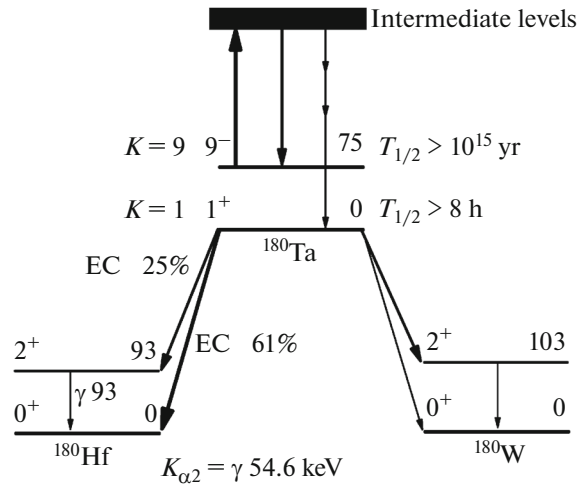

Fig. 1. Energy levels in ^{180m}Ta.

The activity of ^{180g}Ta was measured on the γ-line with an energy of 93.3 keV (Fig. 1).

Measurements were made with two exposures lasting eight hours on an anti-Compton spectrometer equipped with a Ge detector that had an efficiency of γ-beam registration 20% higher than when using a 3" × 3' NaI(Tl)-detector. Suppression of the Compton background at 80–100 keV was 90%.

The γ-spectra in this range always contain inseparable background γ-transitions with energies of 92.4 and 92.8 keV. However, the resolution of our spectrometer allowed us to distinguish the contribution from these transitions reliably. We also monitored the intensity of γ-transitions with energies of 93.3 keV according to the half-life period. The intensity of γ-transitions with energies of 93.3 keV was calculated in two ways. The count rate of the background γ-transition with 92 keV was measured with a high degree of accuracy, and the contribution from the background radiation was then subtracted from the total sum. It was found that N(93.3 keV) = 82 ± 29 counts. Mixed γ-lines of 93.3 keV were processed using two Gaussians, allowing the determination of half-widths and transition energies. In this case, we obtained N(93.3 keV) = 98 ± 15 counts. In background measurements with a lead absorber, the intensity of the γ-line with 93.3 keV was no greater than 17 counts (Fig. 2). The intensity of the background peak with 92 keV was 143 ± 17 counts with Pb and 144 ± 22 with positrons.

Under our conditions, we obtained a broad positron energy distribution formed by the β^{+} source and the conditions for a positron beam passing through a target. The experiment thus allowed us to determine

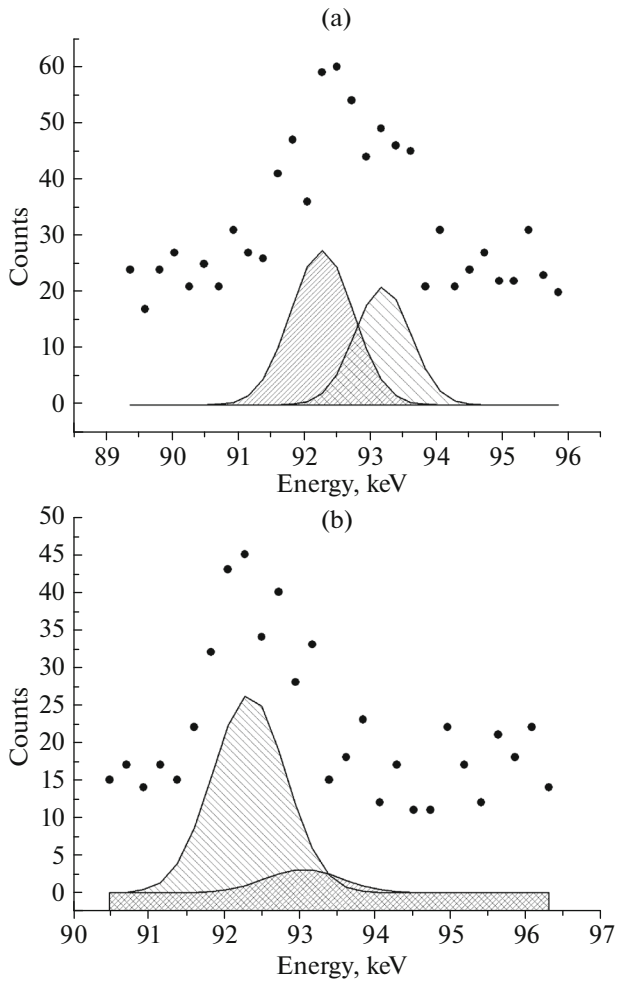

Fig. 2. Experimental spectra of ^{180g}Ta at energies of 92–94 keV upon irradiation with (a) positrons and (b) γ-quanta.

only the effective cross section of proton-free annihilation σ^{eff} [1]:

$$\sigma_{\text{eff}} = \frac{N(\lambda_2 - \lambda_1)\,e^{\lambda_2 t_3}}{N_a f \varphi (1 - e^{-\lambda_2 t_3})(e^{-\lambda_1 t_1} - e^{-\lambda_2 t_1})}, \quad (1)$$

where λ_1 and λ_2 (s^{-1}) are constants of positron source decay and the ground state of ^{180g}Ta, respectively; t_1, t_2, and t_3 (s) are the periods of irradiation, measuring, and the delay between the end of irradiation and the beginning of measurement, respectively; N_a is the total number of target nuclei in a layer thickness equal to the mean free path of a positron with maximum energy ($N_a = 3 \times 10^{17}$); f is the spectrometer's efficiency of recording, measured using calibration sources of ^{182}Ta obtained from tantalum foils with the same dimensions as in the experiments with positrons ($f = 0.092$); and φ ($cm^{-2}\,s^{-1}$) is the density of the positron flow at the start of irradiation.

In light of the obtained data and background measurements, formula (1) showed that $\sigma^{\text{eff}} = (3.9 \pm 0.8) \times 10^{-29}$ cm^2 at $E_{\text{bond}} = 0.653$ MeV.

RESULTS AND DISCUSSION

To compare the experimental photon-free annihilation cross section and the values determined theoretically, we must calculate the differential section of photon-free annihilation from data on σ^{eff} using the formula $\sigma^{\text{ph-free}} = \sigma^{\text{eff}}/n_i P_i$, where n_i is the number of resonance positrons that participate in the excitation of the i-th state, P_i is the probability of ^{180g}Ta being occupied at the excited i-th state. Any experimental estimate of the number of positrons participating in the excitation of the i-th state (n_i) must consider the energy losses of positrons in a thick target, i.e. one that completely absorbs all positrons with the limit energy of 653 keV.

In analyzing the passing of positrons through a thick target, it was assumed that positrons lose their energy through two channels: ionization losses and bremsstrahlung radiation.

Their ratio was $P_{\text{brem}}/P_{\text{ion}} = ZE/800$, where Z is the charge, and E is the energy in MeV, so the contribution from ionization losses was 95% for tantalum [5].

Studying the interaction between incident positrons and atomic electrons of the target shows that the energy transferred to atoms in one collision is very low [6]. Excitation is more likely than direct ionization even for very high primary electron energies. The full loss of energy upon passing through a layer with thickness x is thus due to a very high number of small energy losses. To describe such processes, we introduce average energy of excitation I for atomic electrons: $I = kZ$. Experimental data were obtained earlier for a number of elements, tungsten in particular, for which $I = 680$ eV.

Based on this mechanism, positrons passing through a substance lose their energy with a step equal to I. When irradiating a thick target, all positrons with energies above $E_{\beta+}^{\text{res}}$ will reduce their energy with step I and inevitably reach $E_{\beta+}^{\text{res}}$; i.e., in photon-free annihilation, the wide positron beam with a boundary energy of 653 keV shrinks to a narrow one with width I. This means we must use expression $n_i = \Gamma/I$ to calculate parameter n_i.

For excited states at energies of 1 MeV $\Gamma_i < 10^{-3}$ eV, $\Gamma_a = 35$ eV; i.e., $\Gamma = 35$ eV, I(Ta) = 670 eV. It therefore follows that $n_i = 0.05$.

Considering the distribution of positron energy in the irradiated copper sample and the data on levels of activation, the excitation of ^{180g}Ta can proceed through the states with energies of 1.22 and 1.43 MeV. The share of positrons leaving the state with an energy

of 1.22 MeV is 95%; that of positrons leaving the state with an energy of 1.43 MeV, 20%. The probabilities of ^{180g}Ta being occupied upon the decay of these levels are $P = (0.103 \pm 0.008)$ and (0.126 ± 0.022), respectively [7]. The mean value of $P = 0.1$ was used for evaluation. Allowing for magnitude n_i, we find the total cross section of photon-free annihilation to be $\sigma^{\text{ph-free}} = (0.8 \pm 0.3) \times 10^{-26}$ cm^2.

There are no experimental data on the reduced probability of $E1$ transitions, so reduced one-particle probabilities of transition were used in this work for theoretical estimates. Using the expression for $\sigma^{\text{ph-free}}$ from [8], we found that

$$\sigma^{\text{ph-free}}\ (1.22\ \text{MeV}) = 4 \times 10^{-26}\ \text{cm}^2;$$
$$\sigma^{\text{ph-free}}\ (1.43\ \text{MeV}) = 6 \times 10^{-26}\ \text{cm}^2.$$

When we consider the positron energy distribution, $\sigma^{\text{ph-free}} = 5 \times 10^{-26}$ cm^2.

We can see the discrepancy is much smaller than the one observed in [1], where this value was three orders of magnitude. In addition, the experimental $\sigma^{\text{ph-free}}$ values in [1] were overestimated relative to the theoretical ones, without any physical explanation.

A possible reason for the overestimated theoretical data is in this case the probabilities of $E1$-transitions usually being inhibited, compared to one-particle estimates.

CONCLUSIONS

Cross sections of the excitation of ^{180m}Ta were measured for the first time via the photon-free annihilation of positrons with a boundary energy of 0.653 MeV. The effective cross section was found to be $\sigma^{\text{eff}}(^{180g}\text{Ta}) = (3.9 \pm 0.8) \times 10^{-29}$ cm^2 and was used to calculate the differential $\sigma^{\text{ph-free}}$ cross section. The latter coincided qualitatively with the theoretical values for $E1$ transitions. Our results will be used in further studies of nuclei decay processes in positron annihilation that arise along with pronounced neutron emission under the corresponding conditions. This will in turn allow us to investigate neutron emission from oriented nuclei.

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