Observation of double electron-positron pair production by γ rays reexamined

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An experiment was conducted to observe triple- and quadruple-escape peaks, at a photon energy equal to 6.128 MeV, in the spectra recorded with a high-purity Ge detector working in coincidence with six bismuth germanate detectors. The peak intensities may be explained having recourse to only the bremsstrahlung cascade process of consecutive electron-positron pair creation; i.e., the contribution of simultaneous double pair formation (and other cascade effects) is much smaller. The experimental peak areas are in reasonably good agreement with those predicted by Monte Carlo simulations done with the general-purpose radiation-transport code PENELOPE.

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The detection of high-energy γ rays is becoming an increasingly important issue in many nuclear and high-energy physics experiments (see, e.g., Refs. [1,2]), requiring better knowledge of the detector response in the 1–14 MeV energy region. The detectors with the best energy resolution are high-purity Ge crystals (HPGe). At energies above 1.022 MeV a conspicuous secondary detection effect appears. This effect is a consequence of the annihilation, after slowing down, of a positron generated by pair production. The escape of one or two $m_ec^2 = 511$ keV annihilation photons from the sensitive volume of the Ge crystal originates the well-known single- and double-escape peaks [3], denoted in what follows as SE and DE, respectively.

In the course of our study of HPGe detector response, which included comparing experimental with simulated spectra, the presence of additional triple- and quadruple-escape peaks (TE and QE hereafter) was predicted by Monte Carlo simulations performed with the radiation-transport code MCNP5 [4] but were not seen in a preliminary experiment [5]. These peaks are mainly due to the production of an extra electron-positron pair by a bremsstrahlung photon from the first electron-positron pair formed. When reviewing the literature to ascertain whether this phenomenon was new, we became aware that the QE peak had been found, albeit barely visible, by Wilkinson and Alburger during their search for double electron-positron pair creation at 6.128 MeV photon energy [6]. These authors concluded that the observed peak was most likely due to two consecutive pair formation, i.e., a cascade effect, and not to the higher-order quantum electrodynamics process where two pairs are created simultaneously, and they placed an upper limit for it. In a subsequent work, carried out by Robertson, Kennett, and Prestwich [7] with a different experimental setup and a slightly higher photon energy (6.6 MeV), a positive result was reported, yielding for the ratio of double to single electron-positron pair production cross sections a value of $\sigma_{\pi\pi}/\sigma_\pi \approx (1.82 \pm 0.58) \times 10^{-5}$.

We report here the clear observation of the TE and QE peaks in an experiment similar to that of Wilkinson and Alburger [6], using the 6.128 MeV photons from the ^{19}F$(p, \alpha\gamma)^{16}$O nuclear reaction. While we confirm that no simultaneous double pair formation process is needed to explain the recorded peaks, an increase in the experimental precision could require the inclusion of this process to explain their intensity, in light of the results given by Robertson, Kennett, and Prestwich [7].

Targets consisting of 150 μg/cm^2 CaF_2 evaporated on a 0.1-mm-thick Cu backing and coated by 300 μg/cm^2 of Au were irradiated with 1.378 MeV protons (current equal to 200 nA) at the LAMFI-USP Tandem Accelerator Laboratory of the Instituto de Física, Universidade de São Paulo. The 6.128 MeV photons of the aforementioned reaction were detected with a reverse-electrode closed-end coaxial HPGe 75.5 mm in diameter and 62.5 mm in length. A graded absorber made of Al, Cu, Cd, and Pb was employed to prevent detection of high-energy electrons and positrons from the decay of the 0^+ 6.05 MeV excited state of ^{16}O by internal pair formation and very small components of two-photon emission and internal conversion [8]. The annihilation photons were observed with an annular detector formed by six independent bismuth germanate (BGO) crystals, normally used to suppress Compton events in the HPGe. Reference [9] describes the electronics setup used for that purpose, which was adopted here, but inputting the BGO segments veto signals in a coincidence module with 200 ns resolving time, which provided a hardware trigger signal when at least two segments fired simultaneously with the HPGe detector. A collimator designed to reduce detection of primary reaction photons by the BGOs, without affecting the HPGe detection efficiency, was used. The recorded event consisted of the energy deposited in each BGO, the time difference between the HPGe and every BGO, and the HPGe energy and pile-up signal. The irradiation time was 3 days, and the count rate at 6.128 MeV was about 150 counts/s. More details on the experimental arrangement will be given in a forthcoming publication [10].

Time resolution varied in the interval from 15 to 25 ns, and we set time gates within 60 to 80 ns, according with the resolution of the specific BGO detector. The event multiplicity n was determined as the number of BGOs in the event that passed the time requirement, and chance coincidences were negligible for the TE and QE events. All detectors were calibrated in energy using the observed SE, DE, and annihilation γ-ray peaks. The coincidence conditions chosen for the n-dimensional annihilation peak were

$$\sum_{i=1}^{6}{}' \frac{(E_i - m_e c^2)^2}{\sigma_i^2} \leqslant \chi^2_{\alpha,n}, \quad (1)$$

where E_i and σ_i are the observed energy and width for the ith BGO detector, respectively, and the values of σ_i range from 49 to 62 keV, determined in the BGO spectra in the $n = 2$ coincidence-fold gated by the DE peak in the HPGe spectrum. The prime in the summation symbol indicates that the sum must be restricted to the n terms that passed the time gate. The parameter $\chi^2_{\alpha,n}$ can be related to the fraction α of events in the multidimensional peak included in the gate. Because the BGO peak shape can be well approximated by a Gaussian, the fraction α of the peak area (for $n = 2$), volume ($n = 3$), or hyper-volume ($n = 4$) can be evaluated from the cumulative distribution function of the χ^2 statistics for n degrees of freedom, where $\chi^2_{\alpha,n}$ is the critical value for the α percentage point. This method is similar to that used for high-fold γ-ray coincidence events [11].

Figure 1 displays the regions of interest in the coincidence spectra with either three or four annihilation quanta, where both the TE and QE peaks are visible. These peaks are barely seen in $n = 2$ and the singles spectra. Table I gives the corresponding ratios of peak areas. The repeated occurrence of peaks in the spectra in coincidence with two, three, and four annihilation photons at the energies where the escapes are awaited warrants their identification as escape peaks, although not in all cases they stand well out of the background owing to the small signal-to-background ratio and, for the singles and two- and threefold gated spectra, to the fast variation with energy of the continuum component of the background.

Although we show only results for $\alpha = 95\%$, the peaks were seen in gates with $\alpha = 75\%$ and 50%, having areas proportional to the gated fraction of the n-dimensional annihilation peak area, and superimposed to smaller continuum components, as expected, making more certain that the detected peaks are really escape peaks; however, the relative uncertainties were similar to those reported, and we choose to present the results corresponding to the highest absolute counting statistics.

Monte Carlo simulations were carried to help understand the origin of the observed TE and QE peaks, i.e., to estimate whether the double electron-positron pair creation channel could make a significant contribution or if the second pair was created mainly via bremsstrahlung photons. To this end, the coupled electron/photon transport code PENELOPE was employed [12]. The detailed description of the simulations will be given in Ref. [10]; therefore, here only a brief account is provided. In the context of the present work it is nevertheless worth mentioning that, like most general-purpose codes, PENELOPE includes neither the double electron-positron pair creation process nor the production of a pair by an electron or positron. PENELOPE adopts a modified Bethe–Heitler differential cross section to simulate ordinary pair production, whereas the energy distribution of bremsstrahlung photons is evaluated from Seltzer and Berger's tables of differential cross sections [13].

The geometry of the HPGe detector and the hexagonal array of six BGOs around it was modeled following faithfully the data supplied by the manufacturers. The collimator, absorber plates, and dissipating material along the photon beam path were also included in the geometrical representation. On the other hand, the steering main program *penmain* was modified to incorporate the experimental energy resolutions σ_i of the BGOs and to implement various coincidence requirements to store the spectrum in the HPGe. The number of simulated primary photons in each run was at least 10^9. The peak areas extracted from the simulated coincidence spectra were divided by the area of the DE peak in the simulated coincidence spectrum with two 511 keV photons (i.e., with $n = 2$), and the ratios are listed in Table I, where the quoted uncertainties in the Monte Carlo calculation results correspond to the statistical component, reported as one standard deviation.

In Table I, it is seen that the experimental and simulated QE/DE ratios are in reasonable agreement. The underestimation of the TE/DE ratio by the simulation for the results with $n = 3$ is not significant, because the difference with the experimental value is about 2 standard deviations. Moreover, the experimental and simulated ratio SE/DE, which relates to the commonly observed and well-known secondary detection effect, are in disagreement when the small uncertainty bars are considered. Usually, a fine tuning of the simulated efficiencies demands changes in the internal detector dimensions, because there is a lack of information on internal parameters of the HPGe detector (e.g., the dimensions of dead layers), not always in agreement with the manufacturer's specifications [14]; such a plan is beyond the scope of the present work. Therefore, systematic uncertainties of the same order of the error in the SE/DE ratio, about 30%, should be added to the quoted statistical uncertainties, as is explained below.

Considering that the TE peak is the result of either a DE of annihilation photons from the positron in the first created pair followed by a SE from the second created pair or a SE from the first pair followed by a DE from the second, when calculating its ratio to the DE peak intensity the errors concerning the DE part of the process tend to cancel out; hence, the expected systematic uncertainty in this ratio will come basically from the SE/DE ratio. Also, any underestimation of the photon escape probability in the simulation of the DE process will be enhanced when calculating the QE intensity, because all four annihilation photons escape the detector. However, the DE intensity in the denominator of the QE/DE ratio should cancel out that enhancement; therefore the error in this ratio should not be bigger than that in the SE/DE ratio. It is worth mentioning that our simulation results are compatible with those obtained using MCNPX and disagree with the results of MCNP5, which predicted a much higher TE peak, previously reported by Maidana *et al.* [5]; as explained in that work, discrepant results from MCNPX and MCNP5 codes were obtained using the same input parameters.

Furthermore, a simulation for a 15 cm^3 Ge detector, modeled as a cylinder with thickness equal to the diameter, was done to compare with the experimental singles spectrum of Wilkinson and Alburger [6] where the QE peak was observed. The simulated QE/DE ratio was $(1.47 \pm 0.08) \times 10^{-4}$, in agreement with the measured value of $(1.8 \pm 0.5) \times 10^{-4}$

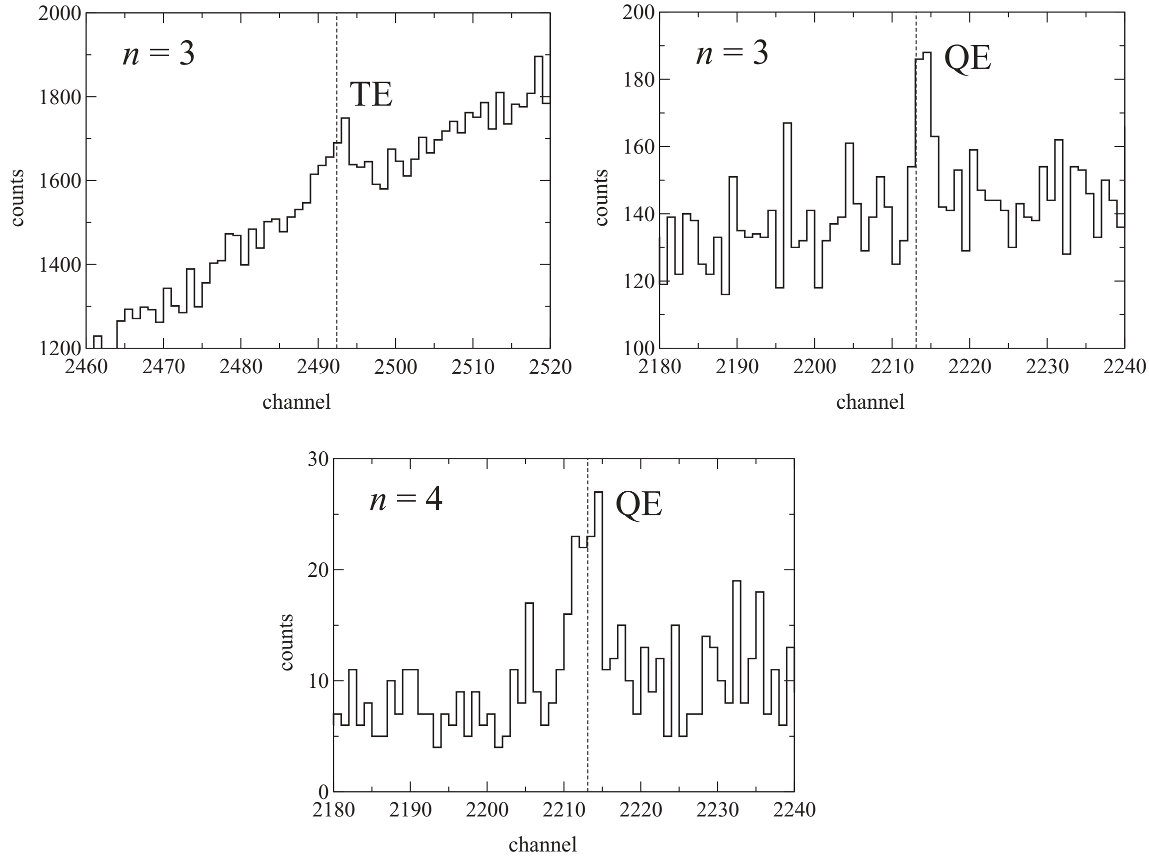

FIG. 1. Measured coincidence spectra with $n = 3$ and 4, both with $\alpha = 95\%$. The regions of interest corresponding to the TE and QE peaks are shown. The vertical dashed lines indicate the expected peak positions. The energy dispersion is 1.83 keV/channel.

and a theoretical estimate by the same authors that yielded 1.6×10^{-4}. In turn, the simulated TE/DE ratio was $(1.00 \pm 0.10) \times 10^{-4}$. This low value may explain why the TE peak could not be seen in that experiment.

If a better accordance between the experimental and simulated detector response is found, it could be possible to gather quantitative information on the double pair creation process with this type of experiment, because the uncertainties in the estimate of the sequential double pair formation, which is the main cause of the observed peaks, can now be well estimated by applying Monte Carlo methods. As already pointed out by Wilkinson and Alburger [6], the use of small detectors would facilitate its observation, because electrons and positrons have smaller chances to emit a bremsstrahlung photon (with energy above $2m_ec^2$) that subsequently interacts within the detector's active volume, thus reducing the contribution of TE and QE peaks from the ordinary cascade process. To examine this possibility, we simulated the spectrum of 6 MeV γ rays obtained with a disc-shaped HPGe detector, 2.5 cm in diameter and 1 cm thick, whose calculated QE and DE peak intensities per primary photon and neglecting double pair formation, QE_π and DE_π, respectively, are

$$QE_\pi = (5.6 \pm 1.1) \times 10^{-10} \text{ events/photon}$$
$$DE_\pi = 5.8 \times 10^{-6} \text{ events/photon}.$$

TABLE I. Experimental (exp) and simulated (MC) ratios between peak areas in the spectra acquired in coincidence with different multiplicities and the DE peak recorded with $n = 2$, with the coincidence gate parameter $\chi^2_{\alpha,n}$ set to accept $\alpha = 95\%$ of the events in the n-dimensional annihilation peak, and the ratios in the singles spectra in the two last columns. The quoted uncertainties correspond to one standard deviation.

Ratio	$n = 4$		$n = 3$		$n = 2$		Singles	
	exp	MC	exp	MC	exp	MC	exp	MC
SE/DE							2.00 ± 0.03	2.68 ± 0.01
TE/DE $\times 10^4$			2.5 ± 0.6	1.4 ± 0.1	2.6 ± 1.1	2.3 ± 0.1	2.6 ± 1.5	3.5 ± 1.2
QE/DE $\times 10^5$	1.5 ± 0.4	1.5 ± 0.1	2.7 ± 1.4	3.1 ± 0.3				

The spectrum of 6 MeV photons in the HPGe above the DE peak has outstanding structures that enable one to evaluate the number of events originated by pair interaction, $N_\pi = (13 \pm 2) \times 10^{-6}$ events/photon, and inelastic scattering, $N_{\mathrm{Compton}} = (7 \pm 2) \times 10^{-6}$ events/photon. The intensity of the DE peak can be related to the probability p that an annihilation γ ray escapes the detector active volume without interacting as

$$\mathrm{DE}_\pi \cong N_\pi p^2, \tag{2}$$

and the contribution of the double pair formation to the QE peak can be expressed as

$$\mathrm{QE}_{\pi\pi} \cong N_{\pi\pi} p^4, \tag{3}$$

where $N_{\pi\pi}$ is the number of events generated by the double pair formation inside the detector. From the equations above, we deduce

$$\mathrm{QE}_{\pi\pi} \cong \frac{N_{\pi\pi}}{N_\pi}\frac{(\mathrm{DE}_\pi)^2}{N_\pi}.$$

Assuming that containment of the created charged particles within the detector is similar for both processes (see Appendix B of Ref. [6]), $N_{\pi\pi}/N_\pi$ can be estimated as the cross-section ratio $\sigma_{\pi\pi}/\sigma_\pi$ measured by Robertson, Kennett, and Prestwich [7]. Replacing the numerical values, we estimate

$$\mathrm{QE}_{\pi\pi} \cong (0.5 \pm 0.2) \times 10^{-10} \text{ events/photon}.$$

Hence, roughly 10% of an experimentally observed QE peak in this detector will be due to the double pair formation in a single interaction, which means that separating the "normal" sequential process from the single interaction $\pi\pi$ formation will require that both the experiment and the simulation results have relative precisions better than 2%; therefore, a single direct measurement with this detector will likely be ineffective. A more refined experiment, using detectors with thicknesses in the range 4 to 8 mm to take advantage of the greater containment of $\pi\pi$ events and disentangling the two processes components by their different dependence on detector thickness, is more likely to be successful.

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