Single-Quantum Annihilation of Positrons with Shell-Bound Atomic Electrons

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The single-quantum annihilation of positrons has been studied experimentally with a positron beam and a thin lead target, at energies 1 MeV and higher. Spectral peaks corresponding to the *K*, *L*, and *M* shells have been resolved and observed distinctly for the first time. The shell ratios *L*/*K* and *M*/*K* have been determined. An analysis of the *L* peak has yielded the (*L*II+*L*III)/*L* ratio. The first measurements of the directional distributions of the annihilation quanta of the three individual electron shells are also reported. The results are in agreement with theory. They also point out the potential for applying the phenomenon to the development of a tunable, highly directional gamma-ray source.

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Despite the fundamental nature of the single-quantum annihilation (SQA) of positrons as an electrodynamic process of atomic physics [1–3], experimental investigations of the phenomenon have been scarce. The existing experimental data are based on measurements made with positrons from radioactive sources, with energy selection made by magnetic spectroscopy and photon detection by NaI(Tl) scintillator-photomultiplier assemblies coupled with light pipes [4–8]. The resulting data are meager and the predictions of the most up-to-date refined theoretical calculations [9–12] have remained largely untested.

The availability of intense positron beams and large-efficiency, high-resolution gamma-ray detectors has now made more accurate experiments on the phenomenon feasible, and we have executed such work utilizing the Brookhaven National Laboratory Dynamitron. The Brookhaven Dynamitron provides a positron beam of intensity 4×10^5 e^+/s at an energy between 0.5 and 3.0 MeV, with an angular divergence less than 1° and an energy spread below 0.2%. Our experiment was done predominantly at 1 MeV, and the accelerator was therefore tuned initially to this energy. The beam was focused to a fixed point on the tube axis throughout the experiment, which served as the target position. The diameter of the beam was determined to be 1.5 mm full width at tenth maximum (FWTM) at the focal point. The beam was terminated in a 1-cm-thick polyethylene stopper, mounted 0.6 cm downstream from the target. The wall of the beam tube was lined inside with a cylinder of polyethylene, also 1 cm thick, in the region around the target (Fig. 1), so that stray positrons backscattered from the target or the stopper could not be incident on the steel walls of the tube. Since the SQA cross section is known to depend on the atomic number of the target as Z^5, practically no SQA photons were expected to be produced from the stopper or polyethylene lining. The lead target we employed in our experiments had a thickness of 3.52 mg/cm^2 and diameter of 25 mm. The median beam intensity was 3.3×10^5 e^+/s and the rate was monitored periodically by direct counting with an upstream plastic scintillation detector by having the beam provisionally deflected onto it. It was determined quantitatively also by integrating the counts in the photopeak corresponding to the 511-keV radiation resulting from the two-quanta annihilation of positrons at rest, occurring in the beam stopper. In a later part of the experiment, the polyethylene stopper was replaced by a 1-cm-thick plastic scintillator, which served also as a downstream beam monitor. All three means of determining the beam intensity gave consistent results independently, when appropriate corrections were applied.

The photon spectrum was recorded with a HPGe detector of 45% relative efficiency. The natural radiation background was reduced by over an order of magnitude by having the detector surrounded by a 5.1-cm-thick lead-shield assembly, with only an acceptance window (0.160 sr) left out. The detector was also lined over with a 0.30-mm-thick tungsten sheet, inside the lead shield, which reduced the detection of the low-energy photons originating from the lead shield to insignificant rates. The net background level was studied with care, without and with the beam, separately, for distinguishing the roles

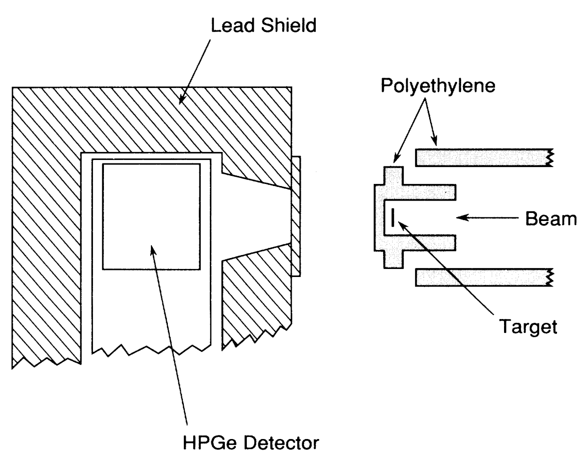

FIG. 1. The target-detector geometry for forward-direction measurement.

of natural radiation and beam-induced processes. In the control experiments for the study of beam-related noise, the lead target could be replaced with a plastic foil. The maximum contribution to the whole photon spectrum recorded in the experimental runs with the beam on could be attributed to the 511-keV radiation, which could produce signals in the region of interest (around 2 MeV for 1-MeV positron energy) to pileup effects only. Other likely contributors to undesired counts in the region of SQA photopeaks were bremsstrahlung and two-quanta annihilation in flight produced primarily in the polyethylene stopper. The former could yield photons of energy up to 1.0 MeV, and the latter, photons between 0.30 and 1.72 MeV. The cross section for two-quanta annihilation in flight was estimated to be 6.5 times larger in lead at 1-MeV positron energy [13] than for the single-quantum process, but since the atomic nucleus would play no role in momentum conservation in the two-quanta process, the photons of a pair were emitted with an angular divergence very much larger than 26°, the maximum that would permit both to be detected and to contribute to the SQA peaks. Overall, the piling up of pulses from various single photons accounted for the majority (60%) of the background level in the SQA region. Pileup was brought down to moderate levels by having the acceptance window of the detector covered with lead sheets of total thickness 9.73 mm. The integral count rate was reduced by a factor of 5, to 1.3 kHz. (However, the potential SQA detection rate was also reduced by a factor of 2.) It was seen from our detailed observations that the total background in the region of interest was linear, devoid of any observable peak (Fig. 2) which facilitated the procedure of background correction.

In accordance with the well-known equation for SQA photon energy,

$$E_\gamma = W^+ + W^- \quad (1)$$

(the two terms on the right representing the total energies of the incident positron and the bound electron), the K-shell SQA peak for lead was formed at 1.941 MeV with 1.007-MeV positrons. The L peak had a 72-keV shift from the K line towards higher energy, and M was shifted further by 12 keV. Since the net width (FWHM) of each of the lines was very nearly equal to 4.0 keV (arising out of contributions from the finite energy resolution of the detector, 2.5 keV, and the net energy spread of positrons in the target, 4.0 keV), the K, L, and M photopeaks were clearly resolved (Fig. 3). The centroids of the peaks were determined from the spectrum, and since the expected shifts were precisely known from atomic spectroscopic data, the observed shifts could yield the subshell ratios. We could determine the $(L\text{II}+L\text{III})/L$ ratio in this way, but statistical accuracies were not good enough to resolve the LII and LIII contributions or to determine the substructure of the M shell. Statistical limitations also did not permit a clear extraction of subshell ratios from linewidths.

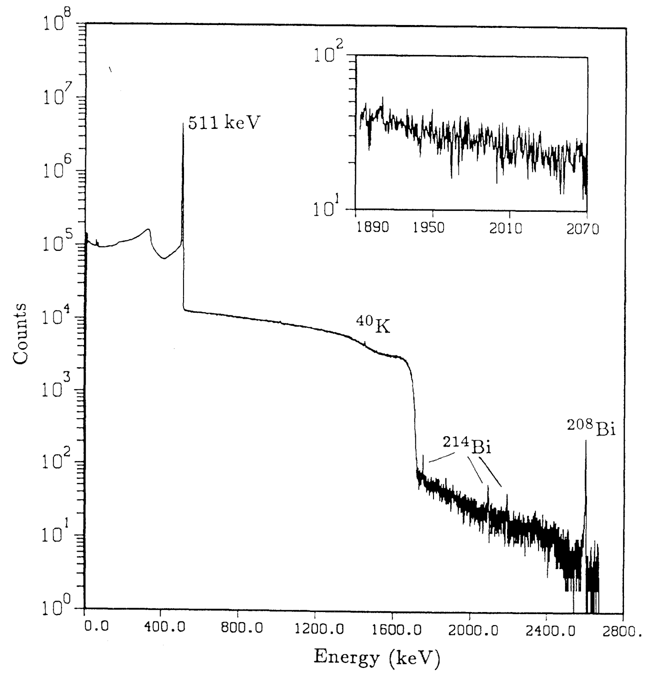

FIG. 2. The background spectrum with positron beam incident on a plastic target. The 511-keV peak and the natural radiation peaks (which are individually identified as originating from radioactive heavy elements and ^{40}K) are clearly visible. Inset: The region of interest with expansion.

The determination of the absolute values of the differential cross sections from the K, L, and M peaks required a number of corrections. An estimated 0.6% of the incident positrons were backscattered by the lead target [14] without participating in the SQA process. Reduction of the SQA photon intensity by the lead absorber covering the acceptance window of the detector was a very significant correction, but could be done with a 3% uncertainty. The *de facto* photopeak efficiency of the detector was determined experimentally with standardized gamma-ray sources positioned in front of the acceptance window of the detector-shield system at the same distance as the target. This measurement was expected to have an accuracy better than 5%. A 5% uncertainty was attributed to the determination of beam intensities. A major correction to the directional anisotropy of the annihilation radiation would be required to account for the attenuation owing to the finite size of the detector (acceptance ~13°) and the straggling of the positrons within the target (average, 8.7° at the point of annihilation). However, in view of the nonavailability of an accurate and independently proven directional correlation function valid for the present case, we opt to present the measured differential cross sections without the attenuation correction.

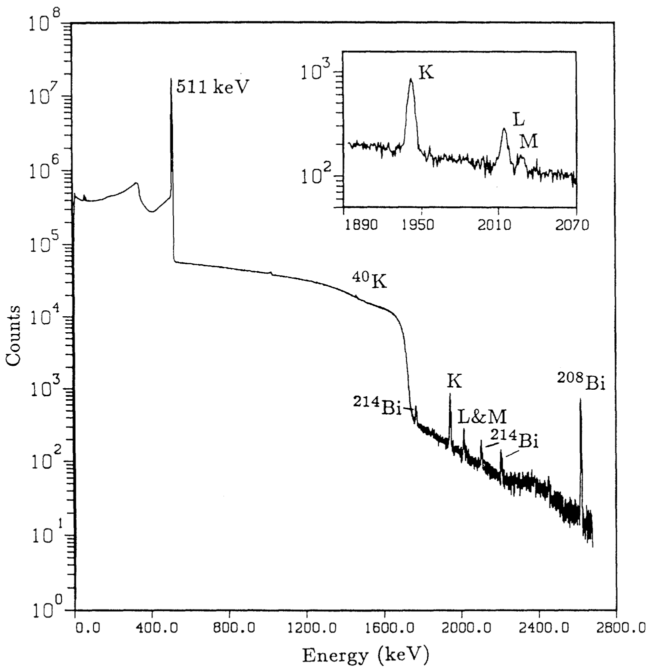

FIG. 3. Spectrum obtained over 87 h with the 3.52-mg/cm^2 lead target for 1.007-MeV positron energy. The expanded SQA region is seen in the inset.

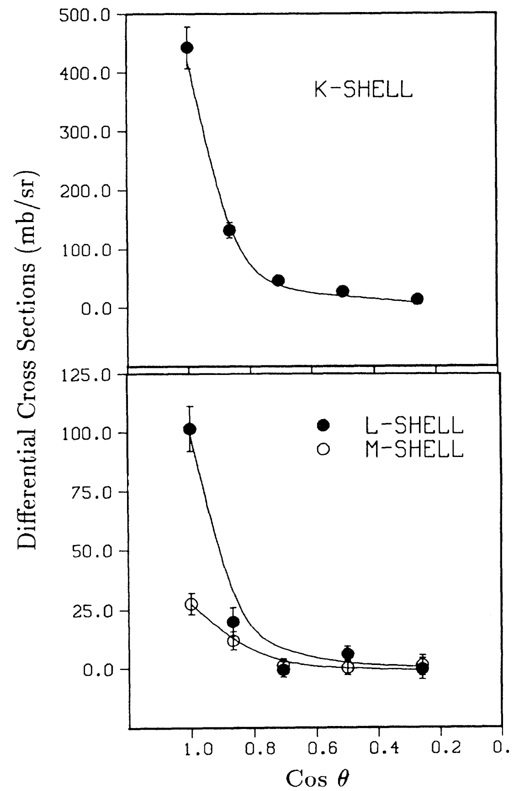

FIG. 4. The measured SQA differential cross sections for 1 MeV. The errors marked allow for a 8% systematic component. The statistical uncertainty is the least (1.4%) for the K-shell 0° data. For data points with no error indicators, the error margin is narrower than the symbol size.

The total cross section for an individual atomic shell may, in principle, be evaluated from the values of the measured differential cross sections. Our measurements at 1 MeV, however, extend only up to 75° (Fig. 4). The integral gives the total K cross section for this range to be 360 mb. All available theoretical data [10] and our own experimental results indicate that angles over 75° may contribute only ~5% to the total. We therefore include a 5% increase, and, with a 50% error margin allowed over this, quote for the integral K cross section 378 ± 32 mb. According to theoretical estimates [9,10], the K-shell cross section for 1.007-MeV positrons is 397 mb, with the screening correction not applied. The latter is expected not to exceed +1% at this energy [12]. Thus, our experimental value is in agreement with the theoretical prediction. This result may be compared with the empirical findings of lower accuracy reported earlier [4–8]. Cross sections for positron energies 1 MeV and lower were obtained under different experimental conditions with targets that were significantly thicker and were found to be in agreement with or smaller than the values predicted theoretically.

Since every SQA spectral measurement of our current work manifested the K, L, and M annihilation peaks distinctly, the shell ratios could be determined with relatively little systematic error from the peak areas (with minor corrections). For each of the three shells studied, the differential cross sections drop with angle very sharply, as expected from theory for the case of high positron energy and high atomic number of the target (Fig. 4). Although the statistical uncertainties are considerable, these ratios are in agreement with the general predictions of the theory.

As an extension of our experiment, we also made measurements of the differential cross sections for 1.5 and 2.0 MeV at 0° and 30° (Table I). The results showed that the cross section at 0° increases appreciably with energy in this range. This can be understood, however, in spite of a reduction in integral cross section expected from theoretical considerations, because of the increasingly forward-peaking nature of the SQA photon emission with increasing positron energy; the cross section drops more sharply with angle (see data for 30°) at greater energy.

While corroborating with theoretical expectations regarding the major features of the SQA phenomenon, the current work has revealed that single-quantum annihilation of positrons could provide a tunable source of monoenergetic gamma rays, of energy 1 MeV and up, that is highly directional, and of easily variable energy. This tunable gamma-ray source has other special merits, narrow bandwidth and high relative yield. With a 2-mg/cm^2 uranium target, the SQA photons produced could have an

TABLE I. The measured K-shell differential cross sections (not correlated for the attentuation of directional anisotropy) and shell ratios for 0° and 30°. The L/K experimental ratios for all angles of measurement are *consistent* with the value $L/K=0.2$ for theoretical integral cross sections, but owing to experimental uncertainties the ratios for angles larger than 30° are not useful for comparison. No theoretical value is now available for the M/K ratio. The centroid analysis gives $(LII+LIII)/L=0.42\pm0.15$ for 1 MeV at 0°, in agreement with the theoretical value, 0.3.

Energy (MeV)		K (mb/sr)	L/K ratio	M/K ratio
1.007	0°	442+36	0.23 ± 0.1	0.06+0.01
	30°	132 ± 13	0.15 ± 0.05	0.09 ± 0.04
1.482	0°	495 ± 54	0.22 ± 0.03	0.07 ± 0.02
	30°	83 ± 16	0.20 ± 0.10	0.02 ± 0.07
2.014	0°	545 ± 61	0.25 ± 0.03	0.03 ± 0.02
	30°	61 ± 14	0.11 ± 0.09	-0.15 ± 0.08

energy width not exceeding 2 keV, and a photon-production rate, for the forward direction, of 7×10^{-7}/sr per positron at 1-MeV energy, about 2-MeV photon energy. At higher positron beam energies, the integral cross section may drop [9], but as our data (Table I) show, the differential cross section at 0° could actually increase. Thus, forward photon yields of the order of 10^{-6}/srkeV per positron could be expected at beam energies over 10 MeV. This yield is higher than other tunable gamma-ray sources under development by orders of magnitude [15]. Positron beams [16] with intensities $\sim10^{10}$ e^+/s engaged in SQA work could produce large photon fluxes that could find applications in research and other areas. Yet, another relevant feature of the SQA photon beam lies in its polarizability, which could be especially useful in research. (Since the positrons that produce the photons in this case could be polarized, the photons themselves could be polarized.) Investigations along these directions, including SQA production for higher positron energies and varied targets and polarization of the photon beam, are to be undertaken by us as a follow-up effort.

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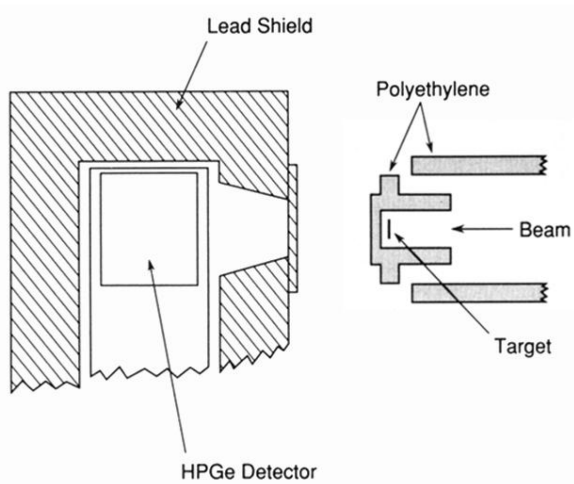
[1] E. Fermi and G. E. Uhlenbeck, Phys. Rev. **44**, 510 (1933).
[2] H. J. Bhabha and H. R. Hulme, Proc. R. Soc. London A **146**, 723 (1934).
[3] H. A. Bethe, Proc. R. Soc. London A **150**, 136 (1935).
[4] L. Sodickson, W. Bowman, J. Stephenson, and R. Weinstein, Phys. Rev. **124**, 1851 (1961).
[5] H. Langhoff, H. Weigmann, and A. Flammersfeld, Nucl. Phys. **41**, 575 (1963).
[6] H. Weigmann, H. Hanson, and A. Flammersfeld, Nucl. Phys. **45**, 555 (1963).
[7] H. Mazaki, M. Nishi, and S. Shimuzu, Phys. Rev. **171**, 408 (1968).
[8] T. Mukoyama, H. Mazaki, and S. Shimuzu, Phys. Rev. A **20**, 82 (1979).
[9] W. R. Johnson, D. J. Buss, and C. O. Carroll, Phys. Rev. **135**, A1232 (1964).
[10] W. R. Johnson, Phys. Rev. **159**, 61 (1967).
[11] C. V. Sheth and N. V. V. J. Swamy, Phys. Rev. **167**, 319 (1968).
[12] K. W. Broda and W. R. Johnson, Phys. Rev. **6**, 1693 (1972).
[13] H. A. Bethe and H. H. Wills, Proc. R. Soc. London A **150**, 129 (1934).
[14] G. Knop and W. Paul, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy,* edited by K. Siegbahn (North-Holland, Amsterdam, 1965), Chap. I.
[15] S. Lindenstruth *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **300**, 293 (1991).
[16] See, for details, Helv. Phys. Acta **63**, 377–470 (1990).



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