Single-quantum annihilation of positrons with K-shell electrons

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Reevaluations of the total K-shell cross sections of single-quantum annihilation of positrons have been made by taking into account the L-shell contribution. Z dependence of the cross section was studied for five high-Z elements, $Z = 50$, 73, 79, 82, and 92, and for 300-keV positrons. Energy dependence has also been investigated for the last two elements in the energy region of 250–400 keV. Comparisons with theoretical predictions including the screening effect are also made.

I. INTRODUCTION

The predominant process of positron annihilation is the two-quantum annihilation where positrons almost at rest annihilate with electrons in the target material. Owing to the conservation of momentum, the annihilation process of the electron and positron should emit at least two photons. In the field of the atomic nucleus, however, positrons in flight have a probability of annihilating with atomic (core) electrons, resulting in the single-quantum annihilation (SQA), where the nucleus shares in the momentum conservation. The first theoretical prediction of this mode of annihilation was proposed by Fermi and Uhlenbeck[1] in 1933. In the following years, several other theoretical works were also reported.[2-5] Johnson *et al.*[6] calculated the total cross sections of SQA for several high-Z elements using relativistic Coulomb wave functions for both the K-shell electron and the incident positron. Their results showed good agreement with those previously reported by Jaeger and Hulme.[5]

The first experimental study of this phenomenon was carried out by Meric[7] in 1950, but because of the poor experimental techniques available in those days, only rough qualitative conclusions could be achieved. More refined measurements of the total cross section of SQA have been performed by Sodickson *et al.*,[8] Flammersfeld *et al.*,[9,10] Mazaki *et al.*,[11] and Friedrich.[12] To the author's knowledge, no newly performed experiment on SQA has been reported thereafter. The results of the first two groups show good agreement as to the energy dependence, and both support the calculations with relativistic Coulomb wave functions. But with respect to the Z dependence, their results lead to the different conclusions, i.e., the exponents of Z in the total cross sections are, respectively, ~5 for the 400-keV incident positrons and 3.2–3.8 for the (760–1100)-keV positrons. Friedrich measured only the energy dependence of the SQA cross sections for iodine.

The third experiment,[11] which has been performed in our group, reveals that the exponent of Z for the 300-keV incident positrons is 4.93 ± 0.31, in agreement with calculations by Johnson *et al.*[6] The energy dependence of the total cross section, measured for $_{82}$Pb and $_{92}$U in the positron energy region from 250 to 400 keV, has also been found to be in fairly good agreement with theoretical values calculated using the relativistic Coulomb wave functions for both the K-shell electron and the incident positron.

It should be noted that in the results of the Göttingen group[9,10] the angular distribution of the emitted high-energy photons with respect to the direction of the incident positron beam was considered to be the same as that of electrons in photoelectric effect, while Sodickson *et al.*[8] assumed it to be isotropic. Johnson[13] calculated the angular distributions of emitted photons using relativistic Coulomb wave functions and showed that the angular distribution peaks sharply in the forward direction. This behavior is different from the angular distribution of photoelectrons and is also in conflict with that obtained from the Born approximation.[2] Our previous paper[11] is the only work which used this exact angular distribution to analyze the experimental data.

All the experimental studies mentioned above concern only the K-shell electrons in data analysis. However, Sheth and Swamy[14] have pointed out theoretically that the contribution of $2s_{1/2}$ bound electrons to the SQA cross section cannot be ignored. They found that the $2s_{1/2}$-shell contribution appears to be roughly 16% of the K-shell cross section. More recently, Broda and Johnson[15] have investigated theoretically the SQA by taking into consideration the screening effect of orbital electrons. They have found that the screening corrections to the Coulomb K-shell cross sections are sizable for large atomic numbers and low positron energies, and that the ratio of L- to K-shell total cross sections is significant for heavy atoms.

Based upon the development in the SQA theories, it seems to be meaningful to reevaluate our previous experimental data which have been treated by neglecting the higher-shell contributions and the screening effect. In this paper, we report the total K-shell SQA cross sections obtained by reevaluating our previous experimental data. The Z dependence of the cross sections is studied with five high-Z elements, $Z=50$, 73, 79, 82, and 92, for 300-keV positrons, and energy dependence is investigated for the last two elements in the energy region of 250–400 keV. In the present data analysis, the L-shell contribution is taken into account, and comparisons with the theoretical predictions including the screening effect are also made.

II. THEORETICAL CALCULATIONS

The exact relativistic calculations of the SQA cross sections for K-shell electrons have been performed by Johnson *et al.*[6] for seven heavy elements from $Z=47$ to 90. In their calculations wave functions in a point-Coulomb field were used for both the K-shell electron and the incident positron. However, Broda and Johnson[15] estimated the K- and L-shell SQA cross sections using wave functions based on the self-consistent relativistic Hartree-Fock-Slater (HFS) atomic model, and pointed out the importance of the screening effect for high-Z elements. Since their calculations are lengthy, the results are reported only for the limited numbers of the incident positron energies and the target elements, and it is difficult to compare with our experimental results.

In the present work, we calculate relativistically the total SQA cross sections for K shell using the normalization screening theory of Pratt and Tseng.[16] In pair production and the atomic photoelectric effect, the processes are characterized by distances which are small on an atomic scale, but large on a nuclear scale. In this case, wave functions in the atomic potential have a point-Coulomb shape and the screening effect can be taken into account by the change in normalizations of the electron wave functions in bound and continuum states. The calculated values for the pair production[17] and the atomic photoeffect[18] in this model agree well with those obtained from the exact wave functions. Tseng and Pratt[19] also applied their model to the case of SQA and found good agreement with the results of Broda and Johnson.[15]

According to the normalization screening theory, the total K-shell SQA cross section including the screening effect is expressed as[19]

$$\sigma_s(E_{+s}) = \Xi^2(p_+^2/p_{+s}^2)\sigma_c(E_+)\,, \tag{1}$$

where Ξ is the ratio of screened to point-Coulomb normalizations for K-shell wave functions, $\sigma_c(E_+)$ is the point-Coulomb SQA cross section, and $E_{+s} = E_+ + \epsilon_s - \epsilon_c$. Here E_+ is the energy of the incident positron, ϵ_c and ϵ_s are the K-shell binding energies for the point-Coulomb and screened potentials, and p_+ and p_{+s} are the momenta corresponding to E_+ and E_{+s}, respectively.

The formula for $\sigma_c(E_+)$ is given by Johnson *et al.*,[6] while the values of Ξ^2 for the HFS potential are taken from calculations by Pratt and Tseng.[16,19] For ϵ_s, we used the values of Carlson *et al.*[20] calculated in the HFS potential. All the calculations have been made on the FACOM M-190 computer in the Data Processing Center of Kyoto University.

III. DATA ANALYSIS

The details of our experimental apparatus and procedures have been described in our previous paper.[11] Here we confine ourselves to describe the procedure to estimate the intensity of the SQA photons from the measured γ-ray spectrum. In the previous paper, we estimated the most reasonable

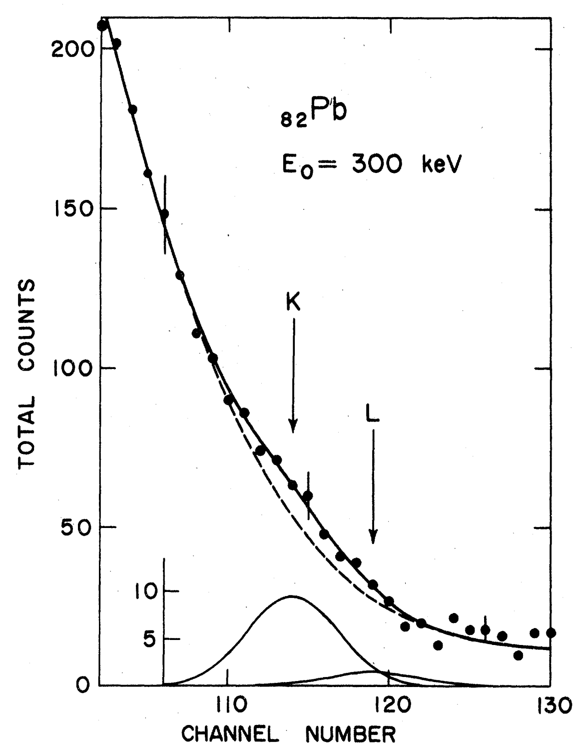

FIG. 1. Observed SQA peaks for the 300-keV incident positrons onto the lead target. The solid curve shows the best least-squares fit to the data. The dashed curve represents the background part. The expected positions of K- and L-shell SQA photons are indicated by the arrows. In the lower part, the K- and L-shell SQA peaks obtained by the least-squares fitting are shown.

profiles of the K-shell SQA peak by considering the energy distribution of positrons in the target. However, theoretical predictions indicate that L-shell contribution is significant for heavy elements.[14,15]

As described in Ref. 11, the γ-ray spectra were observed in coincidence with x rays. But, since the x-ray detector is attached with the curved Lucite light-guide pipe, its energy resolution is so poor that it is impossible to separate the photopeak of K x rays from that of L x rays. In addition, the energy resolution of the γ-ray detector is also poor because of long Lucite light-guide pipe between the NaI(Tl) crystal and the photomultiplier, and the counting statistics is not so good due to low cross section of the process to be studied. For these reasons, the SQA peak obtained in our experiment contains the contribution from L shell as well as that from K shell.

According to the theoretical prediction,[15] the contribution from L_2 and L_3 shells is small, less than 6%. Consequently, we can assume that the L-shell contribution comes mainly from L_1 shell. The SQA peaks are expected to appear at the energy position corresponding to

$$E_\gamma = 2m_0c^2 + E_p - E_{B'}, \tag{2}$$

where m_0 is the electron rest mass, E_p is the kinetic energy of the positron, and E_B is the binding energy of the K- or L_1-shell electron in the target atom. In the present work, the measured γ-ray spectrum was fitted by the least-squares method to two Gaussian peaks with polynomial background.

For Al, we could find no SQA peak. In order to estimate the shape of the background function, we fitted the γ-ray spectrum of the Al target to polynomials and found that a cubic function gives the best fit. It should be noted that the shape of the background is different from element to element. This is because the slowing-down spectrum of positrons in the target depends on the target element. However, we consider that the background shapes do not change so drastically and use the cubic function as background for all the elements, Sn, Ta, Au, Pb, and U.

The measured γ-ray energy spectra were fitted to two Gaussians plus cubic background by the nonlinear function minimization method of Powell.[21] The typical result thus obtained is shown in Fig. 1 for Pb and for 300-keV positrons. The solid line represents the fitted result and the background is indicated by the dashed curve. In the lower part of the figure, two Gaussian peaks corresponding to K- and L-shell SQA photons are separately shown.

TABLE I. Comparison of the measured SQA cross sections for K shell with the calculated ones ($\times 10^{-24}$ cm^2).

Z	Element	Energy (keV)	Theoretical	Experimental
50	Sn	300	0.0624	0.055 ± 0.036
73	Ta	300	0.410	0.43 ± 0.12
79	Au	300	0.610	0.61 ± 0.19
82	Pb	250	0.748	0.82 ± 0.22
		300	0.734	0.77 ± 0.15
		400	0.674	0.73 ± 0.44
92	U	250	1.297	1.27 ± 0.23
		300	1.296	1.46 ± 0.34
		400	1.208	1.12 ± 0.44

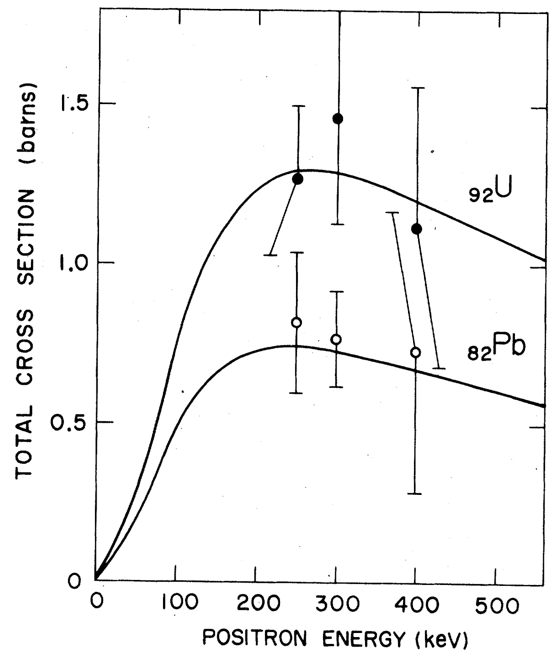

FIG. 2. Energy dependence of the total K-shell SQA cross sections for the lead and uranium targets. The solid curves show the theoretical predictions.

IV. RESULTS AND DISCUSSION

From the observed SQA peak described above, we have attempted to evaluate the total SQA cross section for K shell. The cross section σ is given by the following expression[22]:

$$\sigma = N_\gamma / N_p N \epsilon_\gamma \epsilon_x C_a C_p \omega_K . \tag{3}$$

The symbols in the expression are as follows: N_γ is the number of the observed K-shell SQA photons per unit time, N_p is the number of positrons incident on the target per unit time, N is the effective number of atoms in the target per unit area, ϵ_γ is the detection efficiency of the γ-ray detector for the SQA photons, ϵ_x is the detection efficiency of the x-ray detector, C_a is the cor-

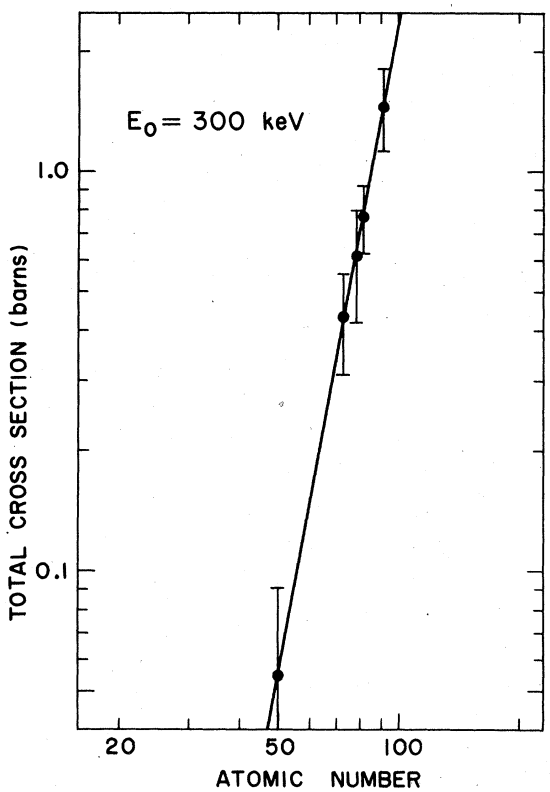

FIG. 3. Total K-shell SQA cross sections as a function of atomic number Z for positrons with the kinetic energy of 300 keV. The straight line represents the best fit of the experimental data.

rection factor for the loss of true coincidences by chance anticoincidences, C_p is the correction factor for the effect of the finite target thickness for the incident positrons, and ω_K is the K-shell fluorescence yield.

As shown in Fig. 1, N_γ was estimated from the area under the K-shell SQA peak. The values of K-shell fluorescence yield are taken from the table of Bambynek *et al.*[23] The other factors were discussed and tabulated in Ref. 11. It should be worth noting that the detection efficiency of the γ-ray detector for the SQA photons, ϵ_γ, was estimated by taking into account the exact angular distribution of the photons calculated by Johnson.[13]

By inserting numerical values of these factors into the right-hand side of Eq. (3), we obtained the values of the total K-shell SQA cross section for target elements studied. The results obtained are listed in Table I and are compared with the calculated values according to the normalization screening theory. The experimental error is mainly ascribed to the uncertainty in determination of the area under the SQA peak by the least-squares method. The reason for such a large error is evidently from low counting statistics of the SQA photons due to a small cross section and a high background from the two-quantum annihilation in flight. Errors in the present work are larger than those in our previous work. This is because N_γ in the present work was determined by nonlinear least squares fitting from the observed γ-ray spectrum including background, while in the previous work the background was estimated experimentally and the peak area was obtained from the shape of the most reasonable profile.

Although the experimental errors are large, it is clear from Table I that the measured cross sections are in good agreement with the theoretical values. Figure 2 shows the SQA cross sections for Pb and U plotted as a function of the kinetic energy of incident positrons. Theoretical calculations of the cross sections have been made in the energy range of 0–600 keV for these elements and the results are also given by solid lines in Fig. 2. It is seen from the figure that the energy dependence of the K-shell SQA cross section agrees well with the theoretical predictions.

In the simple Born approximation, the K-shell SQA cross section is proportional to Z^5. In order to study Z dependence of the cross section, the measured values for the incident positron energy of 300 keV are plotted in Fig. 3 against the atomic number Z. As can be seen from the figure, the measured values are almost on a straight line. The exponent of Z in the total SQA cross section was determined by least-squares fitting the experimental values to a straight line, and was found to be $\nu = 5.3 \pm 0.1$ at the positron energy of 300 keV. This value is in agreement with our previous values,[11] $\nu = 4.93 \pm 0.31$, and the value of Sodickson *et al.*[8] at 400 keV, $\nu = \sim 5$.

Theoretical estimates of the Z dependence have also been made in the normalization screening model. For this purpose, the SQA cross sections for even-Z elements from $Z = 50$ to 92 have been computed for a positron energy of 300 keV. The exponent of Z was evaluated in the same manner as described above and was found to be $\nu = 4.98$. Similar calculations have been made for the positron energy of 100, 200, 400, and 500 keV. The corresponding values of ν are 4.19, 4.83, 4.98, and 4.92, respectively. This result indicates that ν is almost constant in the energy region less than 500 keV.

The positrons incident on the target with the kinetic energy E_p suffer the energy loss during the course of travelling the target before annihilation. If we take into account this fact, the measured cross section does not correspond to E_p, but to $E_p - \langle \Delta E \rangle$, where $\langle \Delta E \rangle$ is the most probable energy loss of the incident positron in the target. However, since the values of $\langle \Delta E \rangle$ are small compared with E_p (Table III in Ref. 11) and the change in the SQA cross section is small in our energy region, we can accept the experimental cross sections as those for positrons with a kinetic energy of E_p.

Computer analysis permits us to separate the peak of L-shell SQA photons in the γ-ray energy spectrum, as seen in Fig. 1. However, since the counting statistics are very poor due to the smallness of the L-shell SQA cross section, it is difficult to estimate the cross section from our γ-ray spectrum.

In conclusion, we have estimated the total K-shell SQA cross sections for several heavy elements in the positron energy region between 250 and 400 keV by reevaluating our previous experimental data. The results are compared with the calculated values for the screened Coulomb potential and good agreement has been found. Energy dependence and Z dependence are also in agreement with the theoretical predictions. Further experimental studies with a high-resolution x-ray detector, as well as with a much stronger positron source, are anticipated. It would be interesting to perform elaborate experiments for the L-shell SQA process. The experimental study on the angular distribution of the emitted photons in the SQA process is also of great interest.

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