Ab initio QED Treatment of the Two-Photon Annihilation of Positrons with Bound Electrons

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The process of a positron—bound-electron annihilation with simultaneous emission of two photons is investigated theoretically. A fully relativistic formalism based on an *ab initio* QED description of the process is worked out. The developed approach is applied to evaluate the annihilation of a positron with K-shell electrons of a silver atom, for which a strong contradiction between theory and experiment was previously stated. The results obtained here resolve this longstanding disagreement and, moreover, demonstrate a sizable difference with approaches so far used for calculations of the positron—bound-electron annihilation process, namely, Lee's and the impulse approximations.

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Since the first observation of positrons [1], investigations of their interaction with atoms, molecules, and solids are of unaltered interest (see, e.g., Refs. [2–5] and the review [6]). Extensive investigations of the positron annihilation processes gave rise to numerous applications ranging from astrophysical searches [7–9] and positron-induced Auger-electron spectroscopy [10,11] to studies of the defects in metals and semiconductors [12,13], the dynamics of catalysis [11], and positron-emission tomography [14,15]. In particular, the angular distribution of the photon pairs from annihilation defines the spatial resolution of the defect analysis and tomography. Therefore, a quantitative understanding of the positron—bound-electron annihilation is highly required by the ongoing growth of studies considering positron-matter interaction as well as by upcoming positron facilities of a new generation, e.g., at the Lawrence Livermore National Laboratory [16–19] and the ELI-NP Research Centre [20,21].

The positron—bound-electron annihilation can proceed with the emission of one, two, or even more photons. More often than not, two-quantum annihilation dominates over other channels. This process, however, has not yet been described rigorously within the framework of QED and with a proper account of the interaction with a nucleus. So far the calculations of the positron—bound-electron two-quantum annihilation were just based on two approximations: Lee's approach [22] for ultraslow (thermalized) and the impulse approximation for ultrafast positrons. For slow positrons, the dominant contribution to the overall annihilation cross section with atomic targets arises from the *nonrelativistic* valence and outer shell electrons. These processes can be well described in the framework of Lee's nonrelativistic approximation [22]. On the basis of this approximation the theoretical approach which shows a remarkable agreement with related experimental studies was developed [23–26]. For ultrafast positrons, in contrast, the impulse approximation can be applied, in which all particles are assumed to be free and where the active electron is represented by a stationary wave packet of superimposed plane wave states. In this approximation, the annihilation process is based on the formulas which were derived almost a century ago by Dirac [27] and Tamm [28]. However, these two approximations cannot be applied to the annihilation of positrons with inner-shell electrons and for collision energies, at which the interaction with a nucleus plays a significant role. As an example, we refer to the experiment where the two-quantum annihilation of 300 keV positrons with K-shell electrons of silver was measured [29], and for which the theoretical cross sections by Gorshkov and co-workers [30,31] differ by more than an order of magnitude. This example demonstrates that the theory is not yet well developed in the region where the kinetic energy of a positron is commensurate with its rest mass.

Here, we develop a fully relativistic formalism based on the *ab initio* QED description of the two-quantum annihilation of positrons with bound electrons. In this formalism, positron- and electron-nucleus interaction is treated nonperturbatively. As the first application, we use the developed approach for the description of the two-quantum annihilation of 300 keV positrons with K-shell electrons of silver which was studied experimentally in Ref. [29]. In this experiment, the double differential angular cross section (DDACS) was measured for photons which are emitted under 30° and −100° with respect to the incident positron direction. Our DDACS of $22(1)$ mbarn/ sr^2 is in excellent agreement with the experimental value $15.4(12.8)$ mbarn/sr^2, and which resolves a longstanding disagreement between theory and experiment. Additionally, we compare the results of the developed exact approach with ones obtained within the Lee's and impulse approximations as well as with result of Ref. [30] and discuss possible reasons of the discrepancies.

The differential cross section for the two-quantum annihilation of a positron with a bound electron in the relativistic units $\hbar = 1$, $c = 1$, $m = 1$ is given by [32,33]

$$\frac{d\sigma}{d\mathbf{k}_1 d\mathbf{k}_2} = 4\alpha^2 \frac{(2\pi)^6}{v_i} |\tau|^2 \delta(E_a + \varepsilon_i - \omega_1 - \omega_2), \quad (1)$$

where α is the fine structure constant, ε_i and v_i are the energy and velocity of the positron, respectively, E_a is the energy of the active electron, and τ is the amplitude whose explicit form will be specified below. In the present Letter, we will consider only the double differential angular cross section defined by

$$\frac{d\sigma}{d\Omega_1 d\Omega_2} = \int d\omega_1 d\omega_2 \omega_1^2 \omega_2^2 \frac{d\sigma}{d\mathbf{k}_1 d\mathbf{k}_2}. \quad (2)$$

This cross section is assumed to be averaged over the angular momentum and spin projections of the electron and positron, respectively, and summed over the polarizations of the emitted photons. The solid angles of the emitted photons $\Omega_{1,2}$ are defined by the azimuthal $\varphi_{1,2}$ and polar $\theta_{1,2}$ angles (see Fig. 1). Here the $x - z$ plane is spanned by the momenta of the incoming positron $\mathbf{p}_i$ and one of the emitted photons $\mathbf{k}_1$ with the z axis fixed along the direction of $\mathbf{p}_i$. Here, we utilize the independent-particle approximation, in which the positron and the active electron move in an effective (Coulomb and screening) potential created by the nucleus and all the other electrons. The screening potential is induced by the Hartree charge density of these remaining electrons. Based on our prior analysis for the Rayleigh scattering of high-energetic photons [34], we expect that the independent particle approximation stays valid for the processes involving inner-shell electrons of heavy systems, where the correlation effects are suppressed by a factor $1/Z$ (Z is the nuclear charge number).

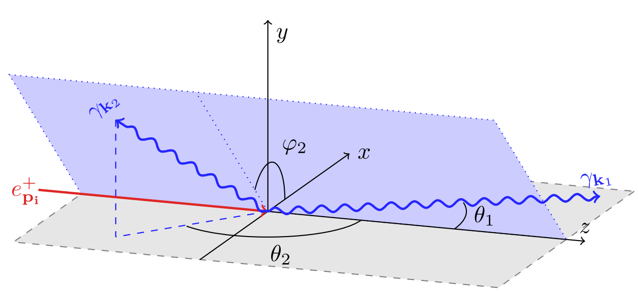

FIG. 1. Geometry (in the ion rest frame) of the positron ($e^+_{\mathbf{p}_i}$)-bound-electron annihilation with the emission of two photons $\gamma_{\mathbf{k}_1}$ and $\gamma_{\mathbf{k}_2}$.

The amplitude of the two-quantum annihilation of the positron with the electron in the bound a state is given by two Feynman diagrams shown in Fig. 2, which corresponds to the following expression [32,33]:

$$\tau = -\sum_n \left(\frac{\langle(-p_i\mu_i)|\boldsymbol{\alpha}\cdot\mathbf{A}^*_{\mathbf{k}_2\lambda_2}|n\rangle\langle n|\boldsymbol{\alpha}\cdot\mathbf{A}^*_{\mathbf{k}_1\lambda_1}|a\rangle}{E_a - \omega_1 - E_n(1 - i0)} + \frac{\langle(-p_i\mu_i)|\boldsymbol{\alpha}\cdot\mathbf{A}^*_{\mathbf{k}_1\lambda_1}|n\rangle\langle n|\boldsymbol{\alpha}\cdot\mathbf{A}^*_{\mathbf{k}_2\lambda_2}|a\rangle}{E_a - \omega_2 - E_n(1 - i0)} \right). \quad (3)$$

Here $\sum_n$ implies the complete summation over the whole spectrum, including the integration over the positive and negative continuum parts, μ_i is the helicity of the incoming positron, $\boldsymbol{\alpha}$ is the vector of Dirac matrices, and the wave function of the plane wave photon with the polarization λ is given by

$$\mathbf{A}_{\mathbf{k}\lambda} \equiv \mathbf{A}_{\mathbf{k}\lambda}(\mathbf{r}) = \frac{\boldsymbol{\epsilon}_\lambda e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{2\omega(2\pi)^3}}. \quad (4)$$

The amplitude [Eq. (3)] determines the differential cross section [Eq. (1)] uniquely and, thus, describes the two-quantum annihilation process completely. Let us turn to the details of the calculation of this amplitude.

The infinite summation $\sum_n$ in Eq. (3) is replaced by a sum over a quasicomplete set of the Dirac equation solutions. These solutions are obtained by using the dual-kinetic-balance finite basis set method [35] with basis functions constructed from B splines [36,37]. Such an approach yields the wave functions of the quasistates n, including the bound state a, but it can barely be applied for constructing the wave function of a positron with a given

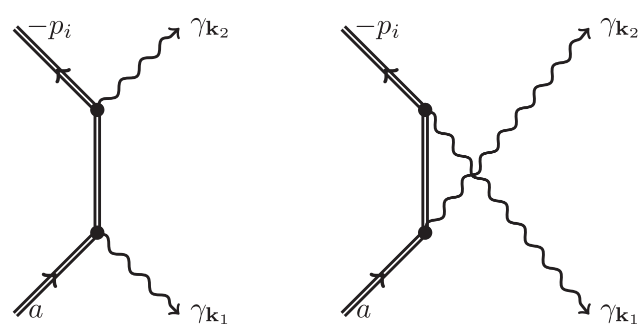

FIG. 2. Feynman diagrams for the two-photon annihilation of the positron $e^+_{\mathbf{p}_i}$ with the bound electron in the a state. The double lines indicate the electron-positron propagators and wave functions in the external field of the nucleus and remaining electrons, while the wavy lines represent the emitted photons, $\gamma_{\mathbf{k}_1}$ and $\gamma_{\mathbf{k}_2}$.

energy. The incoming positron with the four-momentum p_i and the helicity μ_i is treated as an outgoing electron with the four-momentum $-p_i$ and the helicity μ_i [38,39]. The explicit form of the wave function of such a particle can be found, e.g., in Refs. [40,41]. The numerical construction of this wave function is performed with the use of the modified RADIAL package [42]. We note that the constructed wave functions of the incoming positron, quasistates n, and initial bound state a take into account the interaction with the effective (Coulomb and screening) potential to all orders. To calculate the matrix elements, we utilize the well-known multipole expansion technique. As a result, one gets the infinite multipole summations over the photon and positron multipoles, which are further restricted by analyzing the convergence property. More details of the developed method will be presented in a forthcoming publication.

As the first application of our *ab initio* approach, we determine the most probable direction for the emission of (one of the) photons in the process of the two-quantum annihilation of the 300 keV positron with the K-shell electrons of a silver ($Z = 47$) atom and compare it with one studied in the experiment [29]. For this purpose, we explore how DDACS [Eq. (2)] varies with the angle θ_2 in the range between 75° and 180° for the emission angles θ_1 and φ_2 being fixed as in Ref. [29], namely, $\theta_1 = 30°$ and $\varphi_2 = 180°$. However, note that in the original experiment [29] the emission angle $\theta_2 = 100°$ and was kept fixed. The dependence of the DDACS on the angle θ_2 is represented in Fig. 3, which also shows the convergence of the DDACS with respect to the number of the photon multipoles $L_{\max}$ that need to be taken into account in the expansion of the photons wave function. About 30 multipoles are sufficient to obtain well-converged differential cross sections, giving rise to 60 partial waves in the decomposition of the positron wave function. We performed all computations both, in length and velocity gauges, and obtained perfect agreement as seen from Fig. 3. Moreover, the differential cross sections differ by less than 1% if other than the Hartree screening potential is applied.

We can also compare our *ab initio* QED results with those from the Lee's and impulse approximations. In Lee's approximation [22], which has been widely used for the description of the two-quantum annihilation of slow (thermalized) positrons [23–26], (i) the Dirac-Coulomb propagator is replaced by a free electron one, (ii) the binding energy of the initial electron a and the kinetic energy of the incoming positron are assumed to be much smaller than the electron rest mass, and (iii) the Dirac electron and positron wave functions are replaced by the corresponding two-component Schrödinger-Pauli wave functions. Making use of these assumptions in Eq. (3), one can obtain the expression for the two-quantum annihilation amplitude [22]

$$\tau^{(\mathrm{Lee})} = \frac{i}{2}\langle(-\mathbf{p}_i\mu_i)^{(\mathrm{SP})}|(\mathbf{k}_1 - \mathbf{k}_2)\cdot[\mathbf{A}^*_{\mathbf{k}_2\lambda_2}\times\mathbf{A}^*_{\mathbf{k}_1\lambda_1}]|a^{(\mathrm{SP})}\rangle. \quad (5)$$

Here $|(-\mathbf{p}_i\mu_i)^{(\mathrm{SP})}\rangle$ and $|a^{(\mathrm{SP})}\rangle$ refer to the Schrödinger-Pauli wave functions [33] of the positron and electron, respectively. Figure 4 compares the DDACS from this approximation with our *ab initio* results and shows that Lee's approximation overestimates the DDACS by an order of magnitude when compared with the rigorous QED prediction. This discrepancy mainly arises from the large (300 keV) kinetic energy of the positron and the importance of the binding and the relativistic effects for the inner-shell electrons of a silver atom. Let us mention that for low-energy

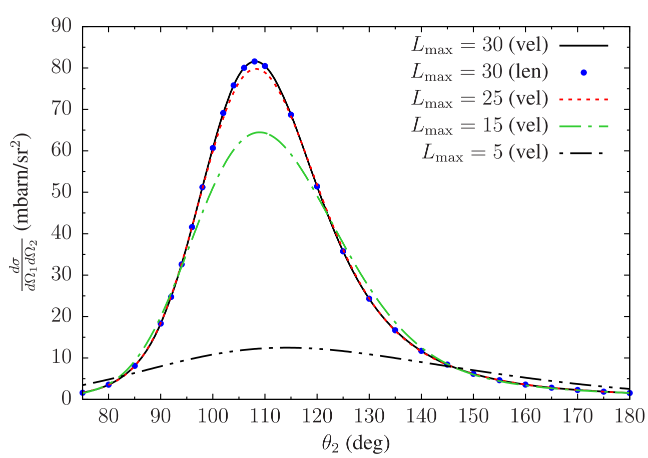

FIG. 3. Double differential angular cross section [Eq. (2)] for the two-quantum annihilation of the 300 keV positron with the K-shell electrons of a silver atom for different numbers of the photon multipoles $L_{\max}$ taken into account. The emission angles $\theta_1 = 30°$ and $\varphi_2 = 180°$.

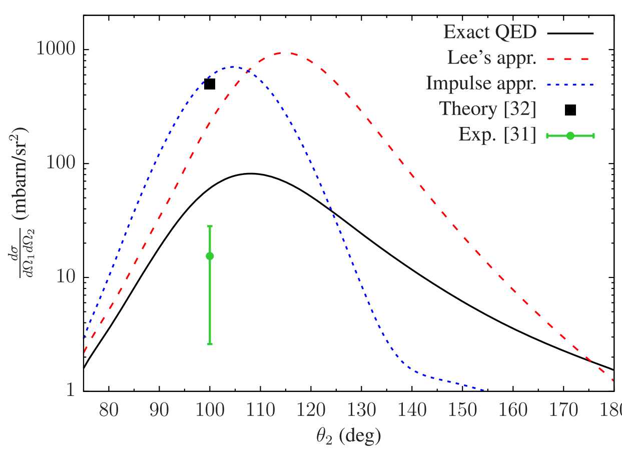

FIG. 4. Double differential angular cross section [Eq. (2)] for the two-quantum annihilation of the 300 keV positron with the K-shell electrons of a silver atom. The calculations performed within the exact approach, Lee's, and impulse approximations are represented by the black solid, red dashed, and blue dotted lines, respectively. The theoretical result from Ref. [30] is shown by a black square, and the experimental value [29] is depicted by a green circle with error bars. The logarithmic scale is chosen for the y axis.

positrons and light systems this approximation agrees well with the more exact calculations. As an example, the total cross section for the two-quantum annihilation of 5 keV positron with $1s$ electron of the H-like argon ($Z = 18$) equals 0.38 barn in Lee's approximation which agrees with the rigorous QED value 0.37 barn.

In the relativistic impulse approximation (IA), it is assumed that the interaction with a nucleus can be neglected for high-energetic positrons and that the process can be viewed as a free positron annihilation with a stationary wave packet of superimposed plane wave electron states. Following the derivation which has been previously described in detail for the Compton scattering [43,44], one can obtain the DDACS for the two-quantum annihilation in the IA

$$\frac{d\sigma^{(\mathrm{IA})}}{d\Omega_1 d\Omega_2} = \frac{1}{v_i} \int d\mathbf{p} \rho_a(\mathbf{p}) \frac{dW_{\mathrm{free}}}{d\Omega_1 d\Omega_2}(\mathbf{p}). \tag{6}$$

Here $\rho_a(\mathbf{p})$ is the momentum distribution of the initial bound-electron a state and $dW_{\mathrm{free}}/d\Omega_1 d\Omega_2$ is the double differential angular probability for the two-quantum annihilation of a free positron and a free electron with the momentum $\mathbf{p}$ [32]. The DDACS being calculated in the IA is compared with other calculations in Fig. 4. From this figure, it is seen that the IA, like Lee's approximation, *overestimates* the DDACS by an order of magnitude. This can be understood by the neglected interaction between the positron and the nucleus and, hence, an (unphysically) increased overlap of the positron and electron densities. This, in turn, leads to the growth of the cross section.

Finally, we compare the obtained results with the previous theoretical predictions by Gorshkov and co-workers [30], which is displayed in Fig. 4 by a black square. These authors started from the free-particle approximation for the two-quantum annihilation and have evaluated the corrections of the first order in the interaction with the nucleus. This approach corresponds to the expansion in powers of αZ and $\alpha Z/v$ which in the case under investigation approximately equal 0.34 and 0.44, respectively. The significant deviation from the exact treatment, however, indicates that such a perturbation expansion fails to describe the DDACS of the considered process.

Figure 4 compares the different theoretical predictions for the DDACS with the experimental value [29]. First, let us note the extra factor 2 in the denominator of Eq. (2) in Ref. [29]. This factor should appear if the contributions of the same quantum states are accounted twice, which does not apply for the DDACS. Therefore, here and below the results from Ref. [29] are multiplied by a factor 2. From Fig. 4 it is seen that all approximate theoretical results, including that of Ref. [30], are by an order of magnitude away from the experimental value, and quite in contrast to our rigorous QED treatment that provides the prediction which is rather close to the experimental result.

TABLE I. DDACS (second column) and detector-averaged DDACS (third column) in mbarn/sr^2 for the two-quantum annihilation of the 300 keV positron with the K-shell electrons of a silver atom. The experimental value from Ref. [29] is multiplied by a factor 2 (see the text for details).

Approach	DDACS	Averaged DDACS
Theory [30]	500	
Lee's appr.	230	134
Impulse appr.	582	72
Exact QED	61(1)	22(1)
Experiment [29]		15.4(12.8)

However, the *direct* comparison of the calculated DDACS with the experimental value might not be fully justified in Fig. 4. This is caused by the fact that the measured value just represents an detector-averaged DDACS. In the experiment [29], the detectors had a 50 mm diameter windows and were placed 8 cm away from the target. The size of the detectors correspond to a solid angle $\Omega_1 = \Omega_2 = 0.286$ sr. To allow for a quantitative comparison with the experiment, we evaluate the detector-averaged DDACS within the exact QED approach as well as within the Lee's and impulse approximations. In the case of the exact calculation, 20 photon multipoles were taken into account. The integration over the polar and azimuthal photon emission angles is performed by a four- and six-point Gauss-Legendre quadrature. For the averaged DDACS, we estimate a relative uncertainty of about 5% because of neglected high photon multiples and an uncertainty in the angular integration. In Table I we compare the DDACS and detector-averaged DDACS calculated within the exact approach, Lee's and impulse approximations with the experimental value [29]. From the table, one can see that the averaging of the DDACS strongly decreases because the planar geometry and detector position at $\approx 180°$ just refers to the maximum of the cross section. Any deviation from this geometry leads to the drop of the DDACS. From Table I, one can also see that the results of *ab initio* QED approach are in excellent agreement with the experimental value.

In conclusion, a fully relativistic QED description of the two-quantum annihilation of a positron with a bound electron is presented for the very first time. This novel approach has been applied for the annihilation of 300 keV positrons with the K-shell electrons of silver. Our result for the double differential angular cross section is in excellent agreement with the experimental value [29] and, thus, resolves a longstanding disagreement between theory and experiment. It was also shown that none of the approaches so far used for the calculation of the positron—bound-electron two-quantum annihilation can be applied in this case. We believe that the exact approach developed here can be extended to many other cases of the positron annihilation and, thus, can allow one to establish more precise validity criteria for the so far employed approximations as well as to help in the interpretation of the experimental data in various applications of the positron annihilation processes.

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