Search for Two-Quantum Annihilation of Positrons in Flight with K-Shell Electrons

Tsuneto Nagatomo,* Yasuyuki Nakayama, Keizo Morimoto, and Sakae Shimizu
Institute for Chemical Research and Radioisotope Research Center, Kyoto University, Kyoto, Japan
(Received 15 January 1974)

The two-quantum annihilation of positrons in flight with the K-shell electrons of silver has been investigated using 300-keV positrons. The experimental result $(7.7 \pm 6.4) \times 10^{-27}$ cm^2/sr^2 has been obtained for the double-differential angular cross section of this process, at 30 and $-100°$ for each annihilation photon with respect to the incident positron direction.

There exists a small probability that positrons having nonzero kinetic energies annihilate with *free electrons* emitting two photons (a process we denote by F-TQA).[1-4] On the other hand, when these fast positrons annihilate with *strongly bound electrons* the phenomena appear as single-quantum annihilation,[5] radiationless annihilation,[6] and nuclear excitation by annihilation.[7] Under these circumstances, we can expect a certain probability that the fast positrons annihilate with K-shell electrons emitting two photons (K-TQA). In this case the energies of the emitted photons are distributed continuously. However, neglecting the recoil energy of the nucleus, the sum of the energies of the two photons, $E_{\gamma 1}+E_{\gamma 2}$, can be given by the simple relation

$$E_{\gamma 1}+E_{\gamma 2}=2m_0c^2+E_p-B_K, \quad (1)$$

where m_0c^2 is the rest energy of the electron, E_p the kinetic energy of the incident positron, and B_K the binding energy of the K-shell electron in the target atom.

No experimental nor theoretical study on this annihilation process has so far been reported. However, since the effect of repulsion of incident positrons by the atomic nucleus may be a main factor which causes a difference between the total cross sections for K-TQA and F-TQA, when the kinetic energy of the incident positrons is high enough compared to the binding energy of the K-shell electron, the upper limit of the atomic total cross section for K-TQA can be taken as the electronic total cross section for two-quantum annihilation of a positron in flight with a free electron at rest. This value can be estimated easily according to the calculations by Bethe[1] and is of the order of 10^{-25} cm^2. The optimum condition for observing the phenomenon is the case that one photon is emitted at 0° with respect to the direction of an incident positron; the other photon is emitted at from 123 to 180° for a 300-keV positron.[1]

We have performed measurements to search for K-TQA, using a medium-Z element, Ag (B_K = 26 keV), as a target and a monoenergetic positron beam of 300 keV obtained by the use of a double-focusing β-ray spectrometer[8] with 5-mCi ^{22}Na as a positron source. As a target we used a silver foil of 5×5 cm^2 in size and 38.0 mg/cm^2 in thickness, smaller than the range of 300-keV positrons. The energy spread of 300-keV positrons on this target was estimated to be within ± 3 keV using a surface-barrier detector. A Lucite plate of 2 mm thickness was attached directly to the back of the Ag target in order to absorb the positrons passing through the target. Two γ rays emitted in this process were detected, through the vacuum chamber window of 10-mm-thick polyvinyl chloride, by 50-mm-diam by 50-mm-high NaI(Tl) crystals placed about 8 cm from the center of the target, outside the vacuum chamber. These detectors had to be arranged in the confined space around the chamber of the target assembly so as to avoid detecting a pair of 511-keV γ rays originating from the ordinary annihilation process. Considering the rather large effective solid angles subtended by the crystals and the optimum detector arrangement for observing F-TQA mentioned above, in the present experiment the angle between the axes of the detectors was chosen to be 130°, as shown in Fig. 1. A 50-mm-diam by 1-mm-thick NaI(Tl) crystal was placed just below the target to detect the 22-keV Ag K x rays accompanying K-TQA. In order to absorb the electrons and positrons scattered in the target, as well as the incident positrons which may come directly to the x-ray crystal, the crystal was covered with a 2-mm-thick Lucite cap.

The process to be studied was identified by a triple coincidence among two γ-ray detectors (of which the discriminator thresholds were set at 350 keV) and the x-ray detector (of which the channel width was set for selecting the 22-keV

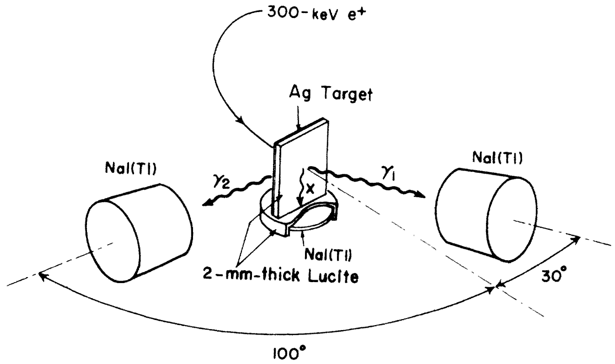

FIG. 1. Schematic diagram of the target-detector system.

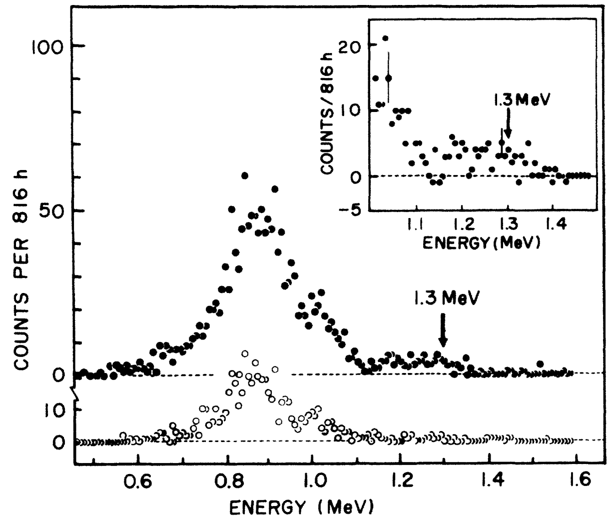

FIG. 2. The observed sum spectrum due to K-TQA for 300-keV positrons, for the two γ rays emitted at 30 and $-100°$ with respect to the incident positron direction. The closed circles show a spectrum for Ag, and the open circles show a background spectrum. The inset is the net spectrum obtained by subtracting the background spectrum from the Ag spectrum.

x rays). Since the sum of the energies of two annihilation γ rays can be defined as given by Eq. (1), the pulses from each γ-ray detector were added in a linear summing circuit and the outputs from this circuit were analyzed with a multichannel pulse-height analyzer which was gated by pulses from the triple-coincidence circuit.

The measurements of gated-sum spectra were performed for the Ag target and for background. The background measurement was performed in such a manner that the Lucite side of the target-Lucite system was faced to the positron beam. Since coincidence counting rates were very small, data were accumulated for 816 h in each measurement, which consisted of seventeen 48-h runs. The functions of all electronic circuits were checked every 48 h at adequate points in the electronic system.

The gated-sum spectrum obtained for Ag is shown by closed circles in Fig. 2. An apparent peak in the lower energy region is mainly caused by the unavoidable coincidences due to 511-keV annihilation γ rays in connection with the discriminator threshold applied for each γ-ray channel. The gated-sum spectrum of the background measurement is also shown by the open circles in the figure. In the inset of the figure is shown the net spectrum obtained by subtracting the spectrum for the background measurement from that for the Ag measurement. In the present work, considering the fact that the kinetic energies of the positrons after passing through the target foil were found to be less than 280 keV, we are interested in the positron energy region of 280–300 keV, which corresponds to the sum-energy region of annihilation γ rays ranging from 1.28 to 1.30 MeV (exactly, 1.276–1.296 MeV). But, in the range of about 1.1–1.4 MeV, we can see a broad distribution due to the positron energy loss in the target and the γ-ray detector resolutions.

In this net spectrum, there exist possible contributions which could not be removed by subtracting the background measurement. They are caused by the following processes. (1) The incident positrons after giving rise to the K-shell ionization annihilate with free electrons emitting two photons; the positrons have energies between 254 and 274 keV. (2) The γ rays emitted by the single-quantum annihilation process with K-shell electrons are scattered at one γ-ray NaI(Tl) crystal to the other crystal. (3) There are random triple coincidences among two γ rays emitted by F-TQA and Ag K x rays produced by other positrons. All other contributions[9] which may cause false and random coincidences could be removed by subtracting the background measurement.

The K-shell ionization cross section by the incident 300-keV positrons for Ag was measured by a separate experiment and found to be about 5×10^{-23} cm^2, which is in agreement with the value obtained by other workers.[10] The fraction of the scattered positrons having energies in the range 254–274 keV is estimated to be roughly less than 0.1 by referring to other works.[11-13] The probability for the annihilation in flight of these positrons with free electrons in the target is estimated to be about 10^{-3} by simple calculations according to the work of Bethe.[1] In addition, taking into account the possible angle between the direction of two γ rays due to F-TQA and the detection efficiencies of the three detectors, a contribution from case (1) was found to be less than 4% of the observed counts in the energy region concerned in the spectrum. In case (2), since the γ rays scattered by one detector to the other have energies less than 300 keV in the present experimental geometry, these γ rays would be rejected by the discriminator setting at 350 keV in the γ-ray channels. The other possible case (3) was found to be negligibly small by separate measurements, i.e., by breaking the triple-coincidence timing.

We have attempted to evaluate the double-differential angular cross section for this annihilation process, which can be expressed by

$$\frac{d^2\sigma}{d\Omega^2}=\frac{4\pi N_c}{2N_p N_a(\epsilon\Omega)_1(\epsilon\Omega)_2(\epsilon\Omega)_x\omega_K}, \qquad (2)$$

where N_c is the observed counting rate due to K-TQA in the energy region of interest; N_p the effective number of incident positrons in the target per unit time; N_a the number of Ag atoms in the target per unit area; $(\epsilon\Omega)_1$, $(\epsilon\Omega)_2$, and $(\epsilon\Omega)_x$ the weighted averages of the overall detector efficiencies, including the geometrical factors, for each of the two γ rays and for the x rays, respectively; and ω_K the K-shell fluorescence yield of Ag. A factor 2 comes from the two types of combinations for detecting two γ rays.

Reflecting the fact that all positrons after passing through the target foil were found to have energies less than 280 keV, N_p was taken as the number of positrons in the energy range from 280 to 300 keV in the target, half the number of incident positrons. The number of incident positrons was measured by a plastic detector of the same size as the target. The value of N_p thus obtained is 4.0×10^3/sec. Considering this condition and taking account of the energy resolutions of the NaI(Tl) detectors, we deduced the value of N_c using the counting data between 1.28 and 1.38 MeV in the observed spectrum: $N_c = 6.8\times10^{-6}$/sec. N_a was $2.1\times10^{20}/\mathrm{cm}^2$. The evaluations of $(\epsilon\Omega)_1$ and $(\epsilon\Omega)_2$ were made experimentally *in situ* using 662-keV γ rays from a calibrated ^{137}Cs source placed at 25 subunits on the target area, based on the assumption that γ rays from K-TQA have an average energy of $(E_{\gamma1}+E_{\gamma2})/2 = 650$ keV. The measured values of $(\epsilon\Omega)_1$ and $(\epsilon\Omega)_2$ thus obtained were 6.5×10^{-2} sr and 5.8×10^{-2} sr, respectively. $(\epsilon\Omega)_x/4\pi$ was also estimated experimentally to be 16.2% for 22-keV x rays by correcting the value measured for Ba K x rays. The fluorescence yield ω_K of silver is 0.834.[14]

Adopting these values, the double-differential angular cross section of the K-TQA process was evaluated for Ag and for the 300-keV positron under the condition that one γ ray is emitted at an angle 30° to the incident positron direction while the other is at 100° to the opposite side. The value we obtained experimentally is $(7.7\pm6.4)\times10^{-27}\ \mathrm{cm}^2/\mathrm{sr}^2$. The experimental error quoted here is due to a statistical error in N_c and systematic errors in evaluating other factors in Eq. (2).

The angular distribution of the two γ rays in this process is not known. But, if we assume that the total cross section for K-TQA can be given by multiplying the differential cross section obtained here by $4\pi\times2\pi$, we derive roughly the total cross section $6\times10^{-25}\ \mathrm{cm}^2$. An upper limit for it can be taken as that of F-TQA, of the order of $10^{-25}\ \mathrm{cm}^2$, as described before. This agreement on the order of magnitude between these values may suggest that the binding effect of the Ag nucleus on the K-TQA process is small for incident positrons of 300 keV.

Experimental evidence for the two-quantum annihilation of positrons in flight with K-shell electrons has been demonstrated for the first time by the present work. It is noted that further measurements with a Pb foil are now in progress to get a clue on the Z dependence. The angular correlation between two γ rays, energy distribution of γ rays, and energy and Z dependences of the process should be studied by the improved experiment. A theoretical study is also desired to clarify details of the process.

The financial support extended from Hitachi, Ltd. for installing the double-focusing β-ray spectrometer used in the present work is gratefully acknowledged.

---

*Present address: Department of Physics, Nara University of Education, Nara, Japan.

[1]H. A. Bethe, Proc. Roy. Soc., Ser. A 150, 129 (1935).

[2]J. B. Gerhart, B. C. Carlson, and R. Sherr, Phys. Rev. 94, 917 (1954).

[3]H. W. Kendall and M. Deutsch, Phys. Rev. 101, 20 (1956).

[4]P. Kilian, Z. Phys. 164, 416 (1961).

[5]See, for example, H. Mazaki, M. Nishi, and S. Shimizu, Phys. Rev. 171, 408 (1968); H. Friedrich, Z. Phys. 246, 407 (1971), and references cited therein.

[6]S. Shimizu, T. Mukoyama, and Y. Nakayama, Phys. Rev. 173, 405 (1968).

[7]T. Mukoyama and S. Shimizu, Phys. Rev. C 5, 95 (1972).

[8]H. Yamamoto, K. Takumi, and H. Ikegami, Nucl. Instrum. Methods 65, 253 (1968).

[9]E.g., two γ rays are emitted by F-TQA and either of these γ rays causes the K-shell ionization of Ag. Two γ rays are emitted by F-TQA and at the same time an ordinary-annihilation 511-keV photon passes through the x-ray crystal and loses about 22 keV of its energy. It is also noted here that I K x rays (28 keV) originating in either of two γ-ray crystals are completely absorbed in the polyvinyl chloride window (10 mm thick) of the vacuum chamber. Therefore, they could not contribute to the background noise.

[10]See, for example, H. Hansen, H. Weigmann, and A. Flammersfeld, Nucl. Phys. 58, 241 (1964).

[11]J. N. Das, Nuovo Cimento 12B, 197 (1972).

[12]J. W. Cooper and H. Kolbemstvedt, Phys. Rev. A 5, 677 (1972).

[13]C. A. Quarles and J. D. Faulk, Phys. Rev. Lett. 31, 859 (1973).

[14]W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freud, H. Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, Rev. Mod. Phys. 44, 716 (1972).

Microscopic Theory of Flow Alignment in Nematic Liquid Crystals

Dieter Forster*

The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637

(Received 1 April 1974)

A microscopic theory of the flow alignment in nematic liquid crystals is presented. The calculation is based on a thermodynamic sum rule derived earlier. The temperature dependence of the flow-alignment angle is obtained in terms of the nematic order parameter $S(T)$. The theory favors stable flow alignment, but the orientational instability found by Gähwiller cannot be ruled out.

The hydrodynamic theory of nematic liquid crystals[1-3] predicts that under stationary shear flow, the director, or averaged molecular orientation, stabilizes at an angle θ relative to the direction of flow which is given by

$$\cos(2\theta) = -\gamma_1/\gamma_2 = 1/\lambda, \qquad (1)$$

where γ_1 and $-\gamma_2$ are usually interpreted as two counteracting viscous torque coefficients. In isotropic molecular liquids there is a weak flow alignment at an angle of 45° relative to the direction of flow.[4] Strong alignment, at an angle $\theta(T)$ which is much smaller than 45° and has a marked temperature dependence, is a phenomenon specific to liquid crystals, and it is therefore of particular interest.

In this Letter I present a simple calculation from first principles of the temperature-dependent flow-alignment angle $\theta(T)$. To my knowledge this is the first microscopic calculation of a hydrodynamic coefficient for nematics. The result reported here agrees with the earlier model calculations by Helfrich[5] at low temperature when the nematic order is perfect and the molecules are assumed to be rigid ellipsoids. The present theory is in good, if qualitative, agreement with recent measurements of $\theta(T)$ by Meiboom and Hewitt[6] which were performed on three nematic substances, PAA (p-azoxyanisole), MBBA {N-[p-methoxybenzylidine]-p-butylaniline}, and HBAB (p-n-hexyloxybenzylidine-p'-aminobenzonitrile). At variance with these experiments are measurements by Gähwiller[7] who reported that for HBAB flow alignment does not occur below 91.8°C. While the present theory does not conclusively rule out this possibility, which amounts to $\lambda < 1$ in Eq. (1), it makes it unlikely.

In the rederivation of nematic hydrodynamics given by Forster *et al.*,[2,3] γ_1^{-1} appears as a rotational relaxation coefficient, as it does in the original Leslie-Ericksen theory. However, my rederivation insists that γ_2 is not an independent dissipative coefficient. Rather, the ratio $\lambda = -\gamma_2/\gamma_1$ enters the theory as an independent reactive coefficient which characterizes the reversible response of the director field to symmetric local stress. One might expect, therefore, that λ can be computed by equilibrium statistical mechanics.

This is true at least for the dominant part of λ.