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80 Years of Positron Annihilation Radiation

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Short history of the investigations of annihilation radiation is presented, from early experiments with ThC″ to the introduction of two main methods: angular correlation of annihilation quanta and positron lifetime spectroscopy.

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Although positron was discovered by Carl D. Anderson [1] in 1932, the radiation appearing at the annihilation of positron and electron was observed much earlier. It was found in the processes of interaction of high energy gamma quanta with matter. In those years the most popular source of "hard" gamma rays was ThC″, i.e. natural isotope ^{208}Tl decaying by β^- process to double magic nucleus ^{208}Pb. De-excitation of the 3^- state of that nucleus produces the gamma quanta of energy 2.6 MeV, that is sufficient to effectively produce e^+e^- pairs.

In 1930, Chinese student in CalTech (USA), C.Y. Chao [2] investigated the scattering of ThC″ radiation by aluminium and lead. For Al the scattered radiation followed the Klein–Nishina formula, but for Pb additional scattered rays were observed. Their energies (determined by absorption method) and intensity were independent of scattering angle (in the range from 22.5° to 135°), thus not related to scattering processes. Chao determined the wavelength of that additional radiation as 22.5 X.U. (i.e. 0.0225 Å), which corresponds to the quantum energy of 540 keV. The presence of additional component in scattered radiation of ThC″ was also noticed by Lisa Meitner and Hupfeld [3]. In 1932, a similar experiment was performed by Gray and Tarrant [4], who found, among the scattered radiation, the quanta of energy close to 0.5 MeV and 1 MeV.

Although the work by Gray and Tarrant was not as precise as that by Chao (no study of angular distribution), Chao's paper is rather forgotten while the one published by Gray and Tarrant awakened a great interest. After the discovery of e^+e^- pair creation [5–7] the results of Gray and Tarrant, explained by authors themselves as the effect of nuclear excitation, were now ascribed to positron "recombination" with an electron. In 1933, Blackett and Occhialini [6] stated that the Gray and Tarrant's results *"may be connected with the formation of positive electrons and their disappearance [...] The reemitted radiation has an energy of the same order as that to be expected for the annihilation spectrum"*. The same authors noted that positrons *"will die according to a probability law"* (exponential decay) and estimated their mean lifetime in water as 3.6×10^{-10} s. According to Oppenheimer and Plesset [8] the pairs should lose all of their kinetic energy and near the end of range should combine with an electron *"with the radiation of two quanta of about half million volts"*. They describe the origin of 1 MeV component seen by Gray and Tarrant as *"not clear"*. Fermi and Uhlenbeck [9] have performed the calculations of the probability of one-quantum annihilation with strongly bound electron in lead as about 0.3% comparing to two-quantum process. They also estimated the relative probability of annihilation in flight, *"the upper limit for 1 MeV positrons in lead is only 5 percent, and the actual value is probably less"*. Thus, the 1 MeV component seen in the Gray and Tarrant experiment was rather an artefact.

The progress in research of positron annihilation processes depended on availability of positron sources. The ThC″ was not a practical solution. In 1934, for the first time the artificial radioactive nuclides were produced when Irène Curie and F. Joliot [10] obtained three positron emitters: ^{13}N, ^{27}Si, ^{30}P in the (α,n) reactions. The same year Cockroft et al.[11] produced ^{11}C using the accelerated deuterons. All these nuclides were very short-lived, nevertheless, they became convenient sources of positrons.

In the works by Chao, Meitner or Gray and Tarrant only the total radiation intensity was measured by ionization chamber and the energies of quanta were determined by absorption method. However, it is also important to have a possibility of counting individual particles. In the case of charged particles, the most primitive method is to use the cloud chamber. This way, Anderson and Neddermeyer [7] observed the tracks of pairs from ThC″ source; Curie and Joliot, using a chamber in magnetic field, were able to measure the momenta of positrons from the decay of their radionuclides. The positron energy spectra were found identical (i.e. continuous) as those observed in the natural β^- decay. The counter of individual particles was introduced by Rutherford and Geiger [12] to register α particles. It was later modified by Geiger [13] who made it sensitive also to β particles and finally got the form of Geiger–Müller (GM) counter [14]. Particle counters were not popular due to the need of visual observation of signals (scintillations, sparks, stirs of electrometer thread, ticks in earphones) and manual recording. Automatic counting of pulses from the Geiger counters was realized by Kovarik [15] who used a vacuum tube amplifier actuating the relay and telephonic pulse counter. Invention of coincidence circuit by Rossi [16] made possible to trigger the decompression of cloud chamber when a charged particle passed two Geiger–Müller counters above and below the chamber. This method was used already by Blackett and Occhialini [6].

There is no problem with counting positrons, however, the annihilation quanta cannot be registered directly, but via the production of photoelectrons or recoil electrons in the Compton scattering. As the working medium in GM counter is gas (some electrons can be produced also in the walls of counter), thus the efficiency of registration of gamma quanta is very low. In spite of that low efficiency, Klemperer managed to get more data about the annihilation radiation [17]. A sample of borax, bombarded by deuterons producing active ^{11}C, was placed between two semicylindrical counters. The coincident signals from two-quantum annihilation were registered, the absorption of these quanta by the set of foils allowed to determine their energies. It was found that there were no quanta of energies over 0.5 MeV, "other mentioned processes, if they exist at all, must be extremely rare".

The effectiveness of registration of gamma quanta in Klemperer's experiment was only 0.4%. A slight improvement of efficiency could be achieved by using the pairs of GM counters connected in parallel, placed one behind the other. This way, Alikhanian et al. [18] obtained the efficiency of 1.25%. In their experiment, the positron source ^{30}P (with half-life of 2.5 min only) was used. It was possible to turn the axis connecting the source and the one pair of counters by 90° and state that the angle between the directions of annihilation quanta is in the limits 150°–180°, and the energy of annihilating pairs is less than 80 keV. Broad limits of angular distribution resulted from the small (only 3.5 cm) distance between the source and the counter.

Increasing the distance between the source and the counter to 15 cm Beringer and Montgomery [19] have found that deviation of annihilation quanta from colinearity does not exceed ±3° and the energy of annihilating pair is less than 1 keV. In this experiment the positron source was ^{64}Cu (half-life of 13 h), obtained in the (d,p) reaction on a cyclotron, but now it can be produced also in a nuclear reactor by neutron absorption. Twelve years after Alikhanian et al. [18], Vlasov and Tsirelson [20] were able to put the upper limit of the energy of annihilating pair as 80 eV by using two sets of five GM counters in a row. With the ^{64}Cu source and the distance source — first counter equal to 52 cm, the counting rate in the peak of distribution was 160 pulses per hour.

Beside the question how narrow is the angular distribution (i.e. momentum) of annihilating pairs, other interesting problem was polarization of annihilation quanta. Wheeler [21] noticed that the polarization planes of two quanta should be perpendicular to each other (detailed theory was given by Yang [22]). Experimental check was done by Bleuler and Bradt [23]. At scattering of radiation at an angle of 90°, the maximum intensity appears in the plane perpendicular to the polarization direction. The annihilation quanta were scattered by aluminium blocks, and those scattered by 90° angle were registered by GM counters. The number of coincidences between them was determined for parallel and perpendicular axes of counters. The ratio of counting rates for these two orientations was found to be about 2, as expected. It is worth to mention that the counting rate was one coincidence per 7 min.

Slow progress in the study of annihilation radiation was determined by low registration efficiency of gamma quanta by gas-filled counters. In spite of that it was possible to determine the energy of annihilation quanta by their diffraction. Owing to focusing the radiation diffracted on a bent crystal (Rowland type spectrometer) one could measure precisely not only the energy of quanta, but also observe the Doppler broadening of 511 keV line. In such an experiment performed by DuMond et al. [24] the average energy of pairs annihilating in copper was estimated as 16 eV. The ^{64}Cu source was of enormous activity of 2.5 Ci; practical application of the Doppler technique became possible 25 years later, with the advent of germanium detectors.

Revolutionary changes appeared in 1948–49 with the introduction of scintillation counters. Solid scintillators, in particular those containing heavy elements, allowed to obtain high counting rates of coincident events (proportional to the square of efficiency). In the years 1949–1951, an eruption of e^+ annihilation experiments appeared. Already in 1949 De Benedetti et al. [25] were able to show the deviation from colinearity for the pairs annihilating in gold. Their experimental arrangement consisted of ^{64}Cu source and two counters with angular resolution of 4 mrad, placed 120 cm from the sample. Rough description of outer electron momentum distribution in various media was given soon by Warren and Griffiths [26]. By improving the resolution it was possible to determine in detail the momentum distribution of annihilating pairs (i.e. the momenta of atomic electrons), as it was done by Lang et al. [27]; the slits in front of counters limited the angle of observation to 1.2 mrad. For the first time, the ^{22}Na nuclide was applied as a source of positrons; now a standard source. Additional advantage of using the ^{22}Na consists in emission of 1276 keV gamma quantum being the signal of positron birth (the half-life of excited state in ^{22}Ne daughter nuclide is 3 ps only).

Application of scintillation counters widened largely the field of research: the three-quantum annihilation was observed [28, 29], the e^-e^+ bound state (positronium) discovered [30], polarization of quanta re-measured [31]. Duration of scintillation in some crystals (or polymers) could have been reduced to bare 1 ns, that made possible to start with positron lifetime measurements. After 40 years, the estimate of positron lifetime in water, as calculated by Blackett and Occhialini [6], could have been checked experimentally. In 1952, Bell et al. invented the delayed coincidence spectrometer [32], and next year the same authors measured the positron lifetime spectrum [33]. Already in their first experiment they noticed the sensitivity of e^+ lifetime to the presence of defects.

The works presented above opened the way to two fundamental techniques of positron spectroscopy: angular distributions and lifetime spectroscopy, being successfully used up to now.

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