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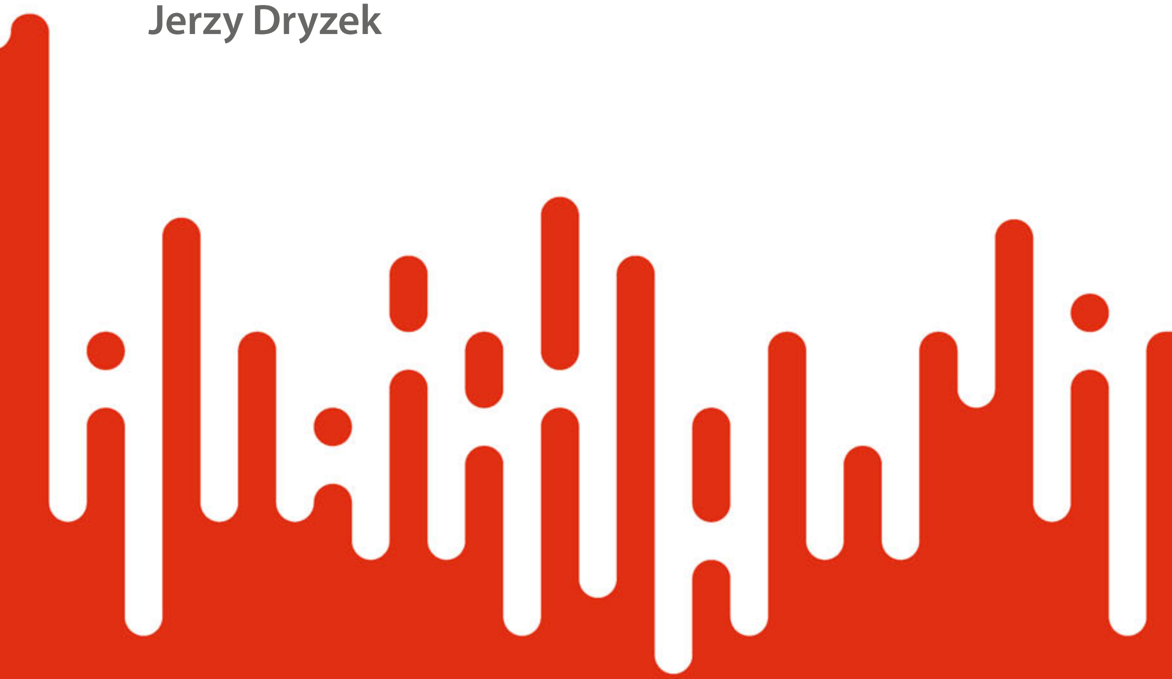
Jerzy Dryzek



Positron Profilometry

Probing Material Depths for Enhanced Understanding

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Chapter 1
Introduction

The structure of matter at the atomic and molecular level is intensively studied with the use of several dozen or perhaps even several hundred research methods. The use of such a large number of them reveals various aspects of the state of matter and the multiple relationships between them. For this purpose, processes occurring at the level of atomic nuclei are used, as is the case with, for example, nuclear magnetic resonance or Mössbauer spectroscopy, at the level of electron orbits, e.g., spectroscopic techniques.

Because matter at the level of atoms and molecules reveals itself as a complex, granular structure of electric charges; hence, the interaction with the electromagnetic wave field is special here. Photons as particles are exceptionally suitable for such research, they are easy to produce, today practically with a huge flux and in a very wide range of energies, and most importantly, they are easy to detect. The quantum nature of phenomena at the atomic level favors the resonant interaction of photons with matter, which greatly improves detection. Not only photons are used here, but other elementary particles are also used.

Neutrons without electric charge, huge fluxes of which can be produced in nuclear reactors, penetrate matter almost freely, but their magnetic moment reveals, for example, the location of local magnetic fields. Muon possesses both the electric charge and the magnetic moment. This favors their trapping as well as interactions on a spin with any environment. Muons originated in nuclear reactions in proton accelerators.

Positron is another particle that is successfully used to study matter at the atomic level. It is easy to obtain, because it is, for example, a product of very common radioactive decay, i.e., beta plus decays. The positron, being an antielectron, annihilates with the electron, resulting in the emission of mainly two photons, which carry information about the electronic state in the annihilation place.

The discovery of a positron, a positively charged electron, by Anderson in 1932 in the background of cosmic rays was not the culmination of the search for a particle that Dirac theoretically predicted two years earlier [1–3]. It was a chance observation of traces in Wilson's chamber of a particle with a mass close to the mass of an electron,

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whose energy of about 62 MeV was enough to pass through a 6-mm-thick sheet of lead and bend its flight path in an external magnetic field. This discovery was almost immediately confirmed by Blackett and Occhialini and other cosmic ray researchers of the time, who additionally observed traces of the formation of a positron–electron pair [4, 5]. This marked the beginning of a series of other important discoveries.

One of the most important is the discovery in 1950 year by DeBenedetti et al. that the positron in matter undergoes thermalization, so in the process of annihilation, by observing the momentum of the annihilating photons, the momentum of the annihilating electron can be determined [6]. This opened the possibility of studying the momentum states of the electronic structure in the matter. The discovery of positron trapping at open volume defects in the crystal structure was another impetus for the development of this technique, as it became a very sensitive tool for their detection [7]. In molecular structures, the formation of a bound state of positron and electron called positronium has been found [8]. Linking the lifetime of this state with the properties of this structure gave wide opportunities for studying molecular materials, including polymers [9, 10]. The discovery of the negative work function of the positron of some metals has enabled the construction of small accelerators that allow the implantation of positrons at nanometer to micrometer depths from the entrance surface [11–13]. These are just some of the important discoveries.

The book points to yet another potential application of positron techniques, i.e., the study of defect profiles near the surface. The book is intended as an introduction for students and people who want to learn the basics of annihilation spectroscopy to use them in their research. This chapter and Chap. 2 serve this purpose. In the following chapters, the author focuses on the use of the method in defect profile studies. It aims to popularize the so-called positron profilometry. It contains, to a large extent, the results of the author's research from the last dozen or so years, illustrated with many examples. The author shares with the reader many of his comments and thoughts on this spectroscopy.

The book can also be of use to specialists in the field of annihilation spectroscopy. What was difficult to fit in the basic descriptions, the author placed in several supplements, which is a short compendium of knowledge, without going into too much detail. There are tables with data, but also useful formulas and relationships.

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Chapter 2
Positron Annihilation Techniques

Abstract Basic facts about positron annihilation in matter will be presented. The sources of positrons used in laboratories have been reviewed. Next, a description of the experimental techniques commonly used in positron annihilation spectroscopy and the measured characteristics used to describe the experimental results are presented.

2.1 Physical Outline

For the use of positrons in studies of condensed matter, three important facts are essential. Firstly, a positron is a stable antiparticle of electrons, but after meeting an electron, both can annihilate, transforming their masses, most often into two energetic photons with a strictly defined energy of 511 keV, Fig. 2.1. This happens when the spins of the annihilating particles are opposite, but when aligned in parallel, three photons with a continuous spectrum of energy appear; this process is marginal. The second is that energetic positron entering matter, due to its positive charge, loses energy very quickly, approaching the thermal energy, which is about three orders lower to the energy of surrounding electrons. The cross-section for the annihilation process is approximately proportional to the reciprocal value of the positron velocity, so the annihilation during slowing down is marginal and this is a third favorable circumstance. But there are others too.

A positively charged positron repels itself from the nucleus, thus avoiding the atomic core and preferring interstitial regions. The density of electrons is not uniform in the matter; there are regions where only high-momentum electrons are present, e.g., in the atomic core, but in the interstitial regions, their momentum is much lower. The angular correlation of the two annihilation photons allows a fairly precise distinction to be made between these differences. But the interstitial regions are also not uniform, and they also show local differences in electron density and hence electron momentum. This can be detected by positron techniques. Hence, the crystal imperfections, incl. vacancies, their clusters, dislocations, grain boundaries, and additives or impurities can be detected at very low concentrations unattainable by other techniques. In molecular solids or polymers, a positron may form a bound state with an

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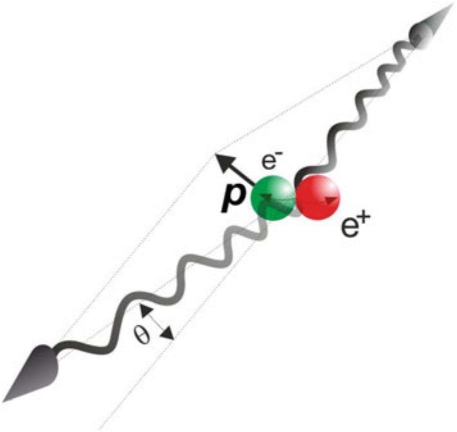

Fig. 2.1 Diagram of two quanta annihilation. The momentum of the positron is negligible compared to that of the electron, which contributes to the deviation of the two photons from collinearity. A measure of this deviation is the angle θ which depends on the projection of the momentum of the positron–electron pair $\boldsymbol{p} : \theta = p_{\mathrm{T}}/m_{\mathrm{e}}c$ where m_{e} is the rest mass of an electron and c is the speed of light and p_{T} is the projection of the momentum of the annihilating pair $\boldsymbol{p}$ in the direction perpendicular to a photon emission

electron called positronium (Ps) [1]. The properties of this hydrogen-like state, an atom without a nucleus, reveal many interesting and useful features of these solids. In small accelerators, positrons may have a reduced kinetic energy before entering matter, so that they can penetrate the regions close to the surface or even on the surface. All these fortunate circumstances make a positron a powerful probe for the study of various aspects of the matter.

2.2 Positron Sources

The positron, the antiparticle of the electron does not occur naturally in nature, but it accompanies several physical processes, including β^+ decay and pair production. They are applied for its production. More than two hundred isotopes decay via positron emission during decay, however the most popular in use are: ^{22}Na, Fig. 2.2, ^{68}Ge/^{68}Ga, Fig. 2.3, ^{64}Cu, Fig. 2.4.

The ^{22}Na isotope is produced in an accelerator [2]. The 15 MeV Deuterons beam irradiates a water-cooled high-purity magnesium target and induces the nuclear reaction: ^{24}Mg(D,α)^{22}Na. Then the nuclide is radiochemically extracted. Its three properties make this isotope readily used in laboratories. Its half-life is 2.62 years, the endpoint of the energy spectrum of the emitted positrons is low, i.e., 0.545 MeV, and after a short period of about 3.6 ps (1 ps $= 10^{-12}$ s), a gamma photon of 1.275 MeV is emitted from the ^{22}Ne nucleus, which in practice indicates the birth of a positron.

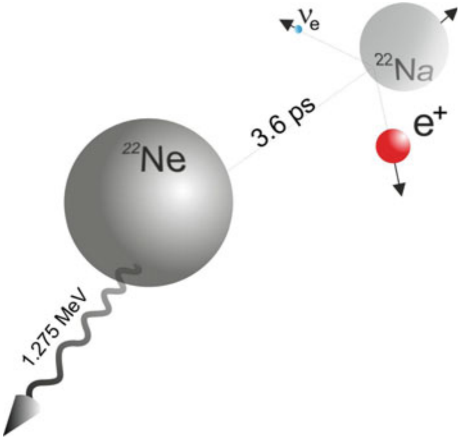

Fig. 2.2 Decay diagram of the ^{22}Na isotope. After a positron and electron neutrino emission, the nucleus is transformed into a meta-stable ^{22}Ne nucleus, which after about 3.6 ps emits 1.275 MeV gamma photon and it becomes stable

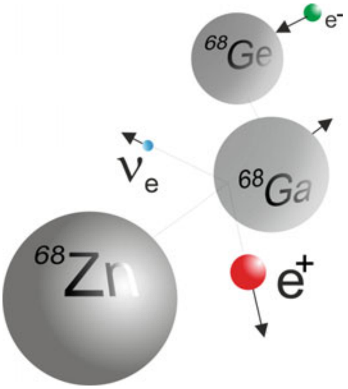

Fig. 2.3 Decay scheme of ^{68}Ge/^{68}Ga-generator. The emission of a positron from the ^{68}Ga nucleus does not accompany a gamma photon emitted from the ^{68}Zn nucleus

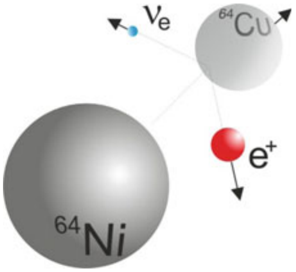

Fig. 2.4 Decay scheme of ^{64}Cu isotope

The decay channel presented in Fig. 2.2 is realized with 90% efficiency; the rest is electron capture. All this makes this isotope very useful in laboratory practice.

The second most used source of positrons is the ^{68}Ge/^{68}Ga-generator. The ^{68}Ge isotope with a half-life of 271 days decays 100% by electron capture into the parent ^{68}Ga nucleus. The latter, after a half-life of 68 min, is transformed in the β^+ decay to the stable ^{68}Zn isotope. The endpoint of the emitted positrons is approximately 1.897 MeV, and the efficiency of this decay channel is approximately 88%. The nuclear reaction of ^{69}Ga(p,2n)^{68}Ge or ^{66}Zn(α,2n)^{68}Ge is used for the production of the generator. The accelerator protons or α particles of energy about dozen of MeV hit the target of a gallium metal or zinc. The popularity of this generator rises from the fact that the production of positrons is not accompanied by gamma photons, and one can say that only positrons result with almost no gamma background. This generator due to this and a short half-time is mainly used in nuclear medicine in the diagnostic procedures in positron emission tomography.

The production of the third ^{64}Cu isotope takes place in a nuclear reactor. Neutron capture by ^{63}Cu, abundant around 69.1% in copper, leads to the appearance of this isotope. Its half-life is short by about 12.8 h and is only 17.4% efficient in the production of positrons. However, the activity obtained can be very high due to the high flux of neutrons. To use this isotope as a positron source, one needs a nuclear reactor nearby. Table 2.1 lists these and some other isotopes used in the production of positrons and used in medicine or science, and in Appendix A.1, the decay modes of the isotopes are depicted as well.

The use of isotopes in laboratories requires environmental protection, but in such a way that positrons may penetrate a sample. For example, ^{22}Na is supplied as chloride which is highly hygroscopic and can easily contaminate the environment. For this reason, the chloride liquid evaporates to a crystalline form on a thin Kapton foil and then closes with the same foil. The edges are taped together to form a closed envelope, see Fig. 2.5a. The thickness of the foil is about 7 μm, which allows the energetic positrons to pass to the outside, but the crystals with the isotope are inside. The envelope is sandwiched by identical samples, which are the target for emitted in full solid angle positrons, and then placed in front of gamma detectors of a spectrometer, Fig. 2.5b. Such prepared sources can be used many times. The thickness of the foil is critical; it must not be too thick as the positron can annihilate in it too, but not

Table 2.1 Selected isotopes as positron sources, their crucial parameters, and nuclear reactions for their production

Isotope	Efficiency of β^+ decay (%)	Half-life	Endpoint (MeV)	Production mode
^{11}C	99	20.3 min	0.96	^{11}B(p,n)^{11}C
^{13}N	100	9.96 min	1.20	^{16}O(p,α)^{13}N
^{15}O	100	2.04 min	1.74	^{15}N(p,n)^{15}O
^{18}F	97	109.7 min	0.635	^{18}O(p,n)^{18}F
^{22}Na	90.6	2.62 y	0.545	^{24}Mg(D,α)^{22}Na
^{44}Ti/^{44}Sc	94.0	60 y	1.47	^{45}Sc(p,2n)^{44}Ti
^{48}V	49.9	15.97 d	0.698	^{48}Ti(p,n)^{48}V
^{58}Co	15.0	71.3 d	0.475	^{58}Ni(n,p)^{58}Co
^{64}Cu	17.4	12.8 h	0.656	^{63}Cu(n)^{64}Cu
^{68}Ge/^{68}Ga	88.0	275 d	1.88	^{69}Ga(p,2n)^{68}Ge