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Monte Carlo simulation of the positron implantation profiles in the layered samples

Jerzy Dryzek,
Paweł Horodek

Abstract. Theoretical studies of the positron implantation profiles in the layered samples are presented. Simulations performed using a GEANT4 tool kit revealed accumulation of positrons in denser layer embedded by less dens environment. This effect is significant for implantation profiles of slow positrons formed in a beam. Nevertheless, it is also present in conventional experiments, where positrons are emitted from radioactive nuclei. In some cases the diffusion process, which follows the implantation and thermalization processes, can smear this effect. However, defects on the interfaces or differences in the positron affinity can sustain it.

Key words: positron implantation • implantation profile • GEANT4 tool kit

J. Dryzek[✉]
The Henryk Niewodniczański Institute
of Nuclear Physics,
Polish Academy of Sciences,
152 Radzikowskiego Str., 31-342 Kraków, Poland
and Institute of Physics,
University of Zielona Góra,
4a Prof. Z. Szafrana Str., 65-516 Zielona Góra, Poland,
Tel.: +48 12 662 8370, Fax: +48 12 662 8458,
E-mail: jerzy.dryzek@ifj.edu.pl

P. Horodek
The Henryk Niewodniczański Institute
of Nuclear Physics,
Polish Academy of Sciences,
152 Radzikowskiego Str., 31-342 Kraków, Poland

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Introduction

Interaction of energetic positrons which enter condensed matter can be divided into three stages: implantation, thermalization, diffusion and finally annihilation with electrons with emission of two energetic photons. On close inspection, definite boundaries between the three processes cannot be determined. The success of the positron annihilation spectroscopy (PAS) in studies of condensed matter arises from the fact that processes associated with the implantation and thermalization do not affect the measured annihilation characteristics. Only annihilation of the positron at thermal energy with an electron contributes to these characteristics. That conclusion is generally valid for the conventional PAS, where the positrons emitted from the nuclei in beta plus decay are injected directly into a sample. Nevertheless, that is not the case in the slow positron defect spectroscopy called variable energy positron annihilation spectroscopy (VEPAS). The information on the space distribution of the implanted positrons is necessary for finding the fraction of positrons which diffuse back to the entrance and finally annihilate there. From that, we can conclude about the positron diffusion process and interactions with defects close to the surface [3]. In this paper we intend to consider the implantation profile when the sample exhibits layered structure. For that structure, the implantation profile may play a significant role in measured annihilation characteristics. For calculations of the profile, we used the tool kit GEANT4 (G4) dedicated to the Monte Carlo (MC) simulations of trajectory of energetic particles in condensed matter.

The G4 is a new-generation tool kit for the MC simulations, intended for a wide range of applications; these include high energy physics, space and cosmic ray simulations, nuclear and radiation analysis, and heavy ion and medical applications [1]. The G4 offers also a great possibility of geometrical shapes of a target where the particles are injected. This opens new possibilities for studies of particles' fates including positrons. In our recent publication we have shown that the tool kit G4 is also effective in simulation of positron implantation profiles required in VEPAS [4].

Theoretical results for monoenergetic positron beam

In our previous paper the reader can find the details of the simulation procedures performed using the G4 codes together with a brief description of the tool kit [4]. Therefore, we skip this and focus on obtained results. Nevertheless, it is necessary to remind the definition of the implantation profile which is the main topic of the paper. The profile is defined as a probability of finding thermal positrons at the end of the thermalization process, initially implanted with energy E, at the depth between z and $z + dz$ from the entrance of the sample. In our calculations we traced the trajectory of 10^6 positrons generated by the G4 codes to the annihilation, i.e. when energy of positrons reached the energy ca. 1 keV and the coordinates of annihilation spots were stored and analyzed. In this section we considered positrons which were implanted perpendicularly to the entrance surface.

Our considerations are based on the model of the stack which consists of two overlayers, i.e. Al and Sn on an Al substrate (Al/Sn/Al). The thickness of the Al and Sn layers are equal to 25 nm. Such a stack was studied experimentally using the VEPAS with the positron kinetic energy equal to 6 keV [5]. Figure 1a presents the generated profile using G4 for such a case. For comparison, we also included the profile in Al without the Sn layer also obtained using G4. In this case the well-known Makhovian profile with the flat maximum at depth equal to 0.19 μm is recognized. After inserting the Sn layer between the Al substrate and Al layer, the profile is apparently changed. Major feature is the sharp maximum in the Sn layer. In order to describe the obtained result qualitatively we notice that the average positron range in matter is defined as follows:

$$\lambda = \int_0^{E_{max}} dE \left(-\frac{dE}{ds} \right)^{-1} \quad (1)$$

and it depends on the stopping power: dE/ds which is a function of the density ρ, kinetic energy and atomic number and other parameters. Taking into account the stopping power form proposed by Batra [2], we can obtain: λ = 57 nm and 45 nm for Al and Sn, respectively. Thus positrons have bigger chance to stop in the Sn layer than in Al. Integrating the number of positrons localized in the Sn layer we obtain that the relative number of positrons located in this layer is equal to 20.4%, but in the corresponding region in the profile for Al only, it is equal to 9.8%. An increase by a factor of ca. two is due to the differences in the values of the stopping power in both materials. These causes the accumulation of positrons in denser layer, i.e. a Sn layer. Such an effect is always present, according to our calculations, for other layered systems. We propose to call it the accumulation effect of energetic positrons. We can notice the accumulation of positrons near the interface in the Al layer that is due to backscattering of positrons from the Sn layer.

In order to convince readers that the accumulation effect is not caused only by the density increase of one layer with respect to another one, in the inset of Fig. 1a we repeated the dependency from Fig. 1a for layered structure, but on the X axis we changed the scale as follows:

$$u = \begin{cases} \rho_{Al} z & \text{for } z < d_{Al} \\ \rho_{Sn}(z - d_{Al}) + \rho_{Al} d_{Al} & \text{for } d_{Al} \le z < d_{Sn} + d_{Al} \\ \rho_{Al}(z - d_{Al} + d_{Sn}) + \rho_{Sn} d_{Sn} + \rho_{Al} d_{Al} & \text{for } z \ge d_{Sn} + d_{Al} \end{cases} \quad (2)$$

where z is the depth in micrometers and u is the corrected depth, but expressed in units of (g/cm^2), $d_{Sn,Al}$, and $\rho_{Sn,Al}$ are the thicknesses and densities of Sn and Al layers. We noticed that the main features of the

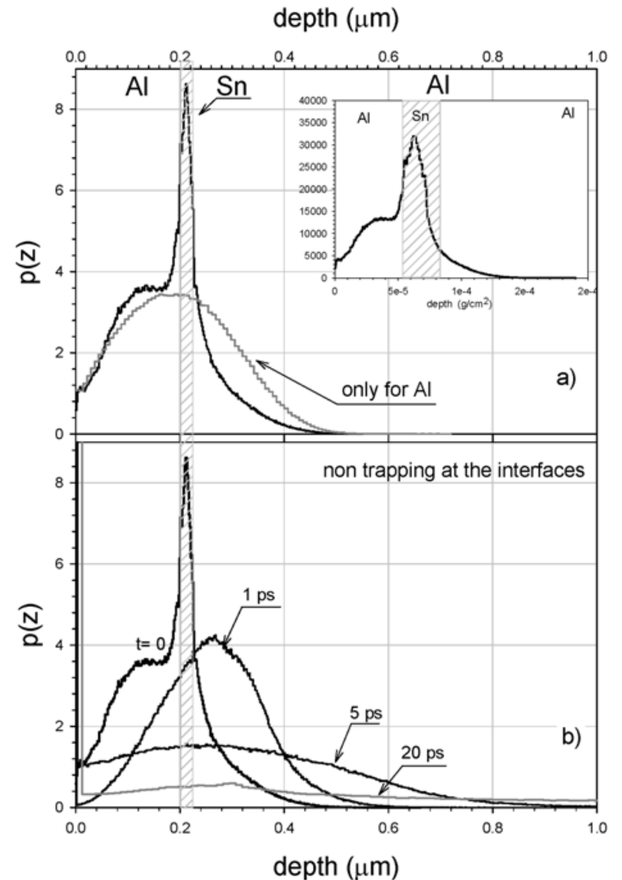

Fig. 1. The implantation profile of positrons implanted perpendicularly to the surface of the stack which consists of 200 nm thick Al layer, 25 nm thick Sn layer and Al substrate (in the right). The energy of the positrons was equal to 6 keV. The Sn layer is marked as the hatched region. In (a) the implantation profile obtained for positrons implanted only in Al sample is added. In (b) the profiles of positrons after the time of 1, 5 and 20 ps of random walk after implantation are depicted. In this case positrons were moved freely across the interfaces. The surface under all profiles was normalized to the unity.

accumulation effect are well recognized in the implantation profile expressed in the new units, but the profile in the new scale does not reproduce the typical Makhovian profile which describes the profile in the uniform sample, e.g. Al in Fig. 1a.

One should emphasize that the profile presented in Fig. 1a results only from the implantation and thermalization processes, when the positrons are slowed down to the energy below 1 keV. We know that after that they begin the diffusion or random walk and this process is not taken into consideration in the G4 codes. It seems that this blurs the characteristic features present in Fig. 1a. To show this we used another code where only the random walk was simulated. The initial position of positrons for this code was taken as the final position obtained from the G4. We assumed that this profile corresponds to the starting condition at time equal to zero. In Fig. 1b we depicted the results of the simulation of the random walk with the thermal energies only, after the time equal to 1, 5 and 20 ps from the time equal to zero. In this simulation we assumed that positrons can move freely across the interfaces and the diffusion coefficient is equal to 1.6 cm^2/s in both materials, i.e. Sn and Al. Now, the Sn layer is invisible for thermally moving positrons. It is well visible that after 5 ps the main features of the implantation profile disappear. Note, that after 1 ps a thermalized positron traverses an average distance of about 30 nm and this is the main reason that the sharp maximum vanishes quickly. We argue that the accumulation effect noticed above is spread by the diffusion process.

However, it is known that the interface as a border can contain open volume defects which effectively localize positrons. Then, the energetic positrons initially localized in the Sn layer during the implantation process cannot move as thermalized out from the layer. Thus, the defects or other traps at the interface could sustain the accumulation effect. In Fig. 2a we present the calculated profile assuming that the interface is 2 nm wide and can perfectly localize positrons when they cross it. Thus, the interface is a perfect sink. Indeed, the number of positrons at the interface increases significantly and additionally they do not leave the Sn layer. This figure convinces us that the accumulation effect can be observed in VEPAS experiments in such a case when the interface is a sink for positrons. There is also another effect which should be considered. It is well known that positron has different affinity to different metals which arises from the difference in contact potential between two metals. According to the theoretical calculations, the affinity to Al is equal to 4.41 eV and to Sn – 7.60 eV [6]. Thus, the potential barrier at the interface is equal to 3.6 eV and thermalized positrons of energy equal to 25 meV cannot move from Sn to Al layer, but can jump from Al to the Sn layer. This process can be simulated as a perfect reflection at the barriers when positron are in the Sn layer. In Fig. 2b the results of the simulations are presented. They are similar to the results from Fig. 2a, i.e. the accumulation effect induced by the implantation process is sustained. Since the reflection takes place close to the interface, thus the increased number of positrons close to the interface is observed in Fig. 2b. We can argue that both processes, i.e. trapping at the interfaces and reflection at the contact potential barriers can make visible the accumulation effect despite the diffusion process.

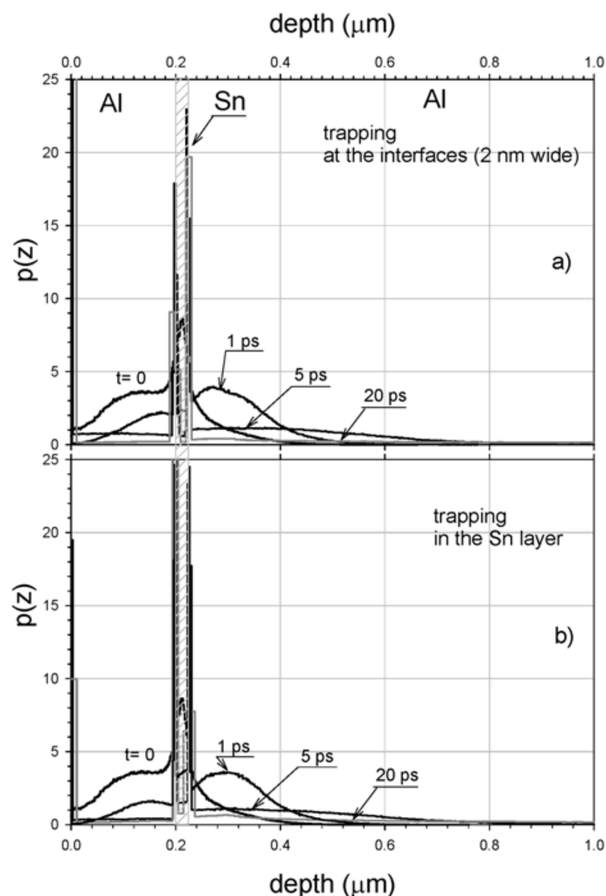

Fig. 2. The implantation profile of positrons implanted perpendicularly to the surface of the stack which consists of 200 nm thick Al layer, 25 nm thick Sn layer and Al substrate (in the right). The energy of the positrons was equal to 6 keV. The Sn layer is marked as the hatched region. In (a) the profiles of positrons after the time of 1, 5 and 20 ps of random walk after implantation are depicted, now the interfaces are the perfect sinks for positrons. In (b) similarly like in (a) but now positrons are reflecting when crossing the interface from Sn to Al. The surface under all profiles was normalized to the unity.

Theoretical results for positrons emitted from ^{22}Na

The accumulation effect can be present also in conventional experiments with positrons emitted from the isotopes. We modelled the stack which consists of a thick Al plate, a positron source enveloped in a kapton foil 7 μm thick, an Al foil of 40 μm thick, an Ag foil 40 μm thick and a thick Al plate. In this case positrons are emitted in a full solid angle randomly, and during implantation then can pass through the Ag foil, whose density is three times as high as that of Al foil. Thus, the major condition for the accumulation effect is fulfilled. The implantation profile generated by the G4 for such a stack is presented in Fig. 3. In simulations we assumed that positrons have a continuous energy spectrum which corresponds to the spectrum of beta plus positrons emitted from ^{22}Na isotope. In the left part, the obtained profile corresponds to the implantation profile in uniform solid. However, in the

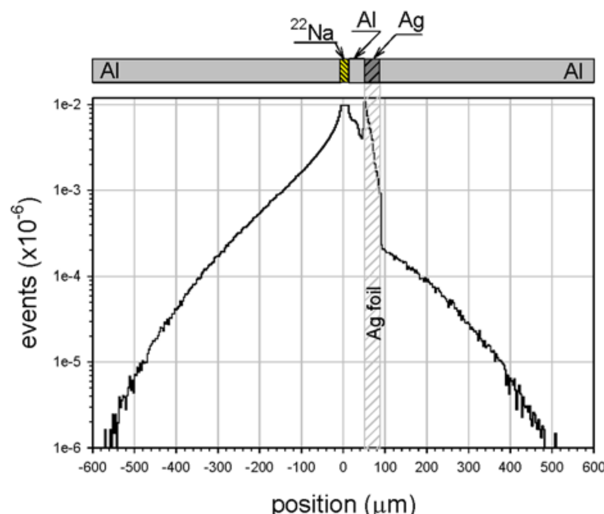

Fig. 3. Implantation profile for positrons emitted from ^{22}Na nuclei into the stack which consists of Al plate, 40 μm thick Al foils, 40 μm thick Ag foil and Al plate. The Ag foil is marked as the hatched region.

right part the apparent increase in the region of Ag foil is visible. This is the accumulation effect noticed above. The relative number of positrons in the Ag foil region to all positrons is equal to 16.4%. If we would replace this region by the Al foil we get only 10% of positrons in this region. The accumulation effect is much lower in this case than in the Al\Sn\Al system illuminated by the 6 keV positron beam. This is well understood because in the conventional experiment only a small fraction of positrons reaches the Ag foil because they are emitted in solid angle. It seems that, as it was mentioned above, the sharp maximum at the interface has the same origin as discussed above. Now, the Ag foil is 40 μm thick and the diffusion process cannot spread the maximum in this foil. The average diffusion length is defined as follows: $\langle L \rangle = \sqrt{6D\tau_{\text{bulk}}}$, where τ_{bulk} is the bulk lifetime, equal to 130 ps in Ag, and D_+ is the diffusion coefficient equal to 1 cm^2/s. The value of the average diffusion length equal to 280 nm is much lower than the thickness of the Ag foil. Certainly, the defects at the interface and differences in the affinity are not so important in this case.

In order to show the accumulation effect we performed the positron lifetime measurements in the stack described above. Well annealed foil of Al and Ag were used, the positron lifetime in Al foil was equal to 165±1 ps and in Ag 138±1 ps and these values correspond to the values reported already as values of positron lifetime in bulk. Our experimental stack consists of the Al plate 2 mm thick, ^{22}Na positron source enveloped in the 7 μm thick kapton foil, the 40 μm thick Al foil, the 40 μm thick Ag foil and the Al plate 2 mm thick. The positron lifetime spectrum contained 3×10^6 counts. Two lifetime components were resolved in the spectrum equal to the values mentioned above. The intensity of the component which corresponds to the positrons annihilating in Ag foil was equal to 12.2±0.2%. That is less than 16.4% mentioned above but more than 10%. It suggests the accumulation effect in the Ag foil, but it seems that the G4 codes may overestimate it. More experimental studies in this matter are necessary to determine the status of the effect.

The presence of layers either generally participates in other phases or in the regions whose densities are higher than the surrounding host in the sample and causes an increase of the number of positrons localized there above the volume fraction of those regions. This effect is caused by the reduction of average positron range in comparison to the range in a less dense host. At this stage it is difficult to give a simple mathematical formula which allows predicting properly the fraction of positrons localized in the regions, because this effect essentially depends on their volume and geometry. The presence of other effects is not excluded. One can also mention that our observation of positrons is based on the detection of annihilation photons. Such photons emitted from denser regions are absorbed more effectively than photons emitted from the host. Thus, photon absorption can deplete the accumulation effect. In experiments this effect should be taken into account.

Conclusions

The simulation of positron implantation profile in a simple layered system Al\Sn\Al revealed a significant increase of the number of positrons in the region of Sn layer. Such an accumulation of energetic positrons seems to be a general tendency in other non-uniform systems where the denser regions are embedded in a less dense region. This effect arises from the differences in stopping power. The diffusion process with thermal energies which follows the implantation can smear this effect but the presence of the open volume defects and the differences in affinities can sustain it. The accumulation effect, significant in the positron beam experiment but also in conventional measurements with the use of isotope sources, can increase the fraction of positrons in denser regions above their volume fraction. The measurements of the positron lifetime in the layered stack, which consist of Ag foil 40 μm thick embedded in Al points out the increase of the positron annihilating fraction in the Ag foil according the G4 simulations.

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