

Research progress at the Slow Positron Facility in the Institute of Materials Structure Science, KEK

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Abstract. Recent results at the Slow Positron Facility (SPF), Institute of Materials Structure Science (IMSS), KEK are reported. Studies using the total-reflection high-energy positron diffraction (TRHEPD) station revealed the structures of rutile-TiO₂(110) (1×2), graphene on Cu (111) and Co (0001), and germanene on Al (111). First observations of the shape resonance in the Ps⁻ photodetachment process were made using the positronium negative ion (Ps⁻) station. Experiments using the positronium time-of-flight (Ps-TOF) station showed significant enhancement of the Ps formation efficiency and the energy loss in the Ps formation-emission process. A pulse-stretching section has been implemented, which stretches the positron pulse width from 1.2 μs up to almost 20 ms.

1. Introduction

At the Slow Positron Facility (SPF), Institute of Materials Structure Science (IMSS), High Energy Accelerator Research Organization (KEK), pulsed positrons generated through bremsstrahlung pair production at the beam dump of an electron linac are moderated using W foils which have a negative positron work function (-3 eV). Prior to transport, they are accelerated to the desired energy to form an energy-tunable monenergetic positron beam, i.e. a slow positron beam [1].

The intensity of the beam is 5×10^7 e⁺/s in the accelerator long pulse mode (electron pulse width 1.2 μs) and 5×10^6 e⁺/s in the variable short pulse mode (1-12 ns). Figure 1 shows the present layout of the beam line branches in the facility. The connections of the stations to the beam line branches have been exchanged from the original arrangement a few years ago [1]. The currently used stations are for total-reflection high-energy positron diffraction (TRHEPD) on branch SPF-A3, for the positronium negative ion (Ps⁻) related experiments on SPF-B1 and for the positronium time-of-flight (Ps-TOF) measurements on SPF-B2. A station for low energy positron diffraction (LEPD) is under construction on SPF-A4.



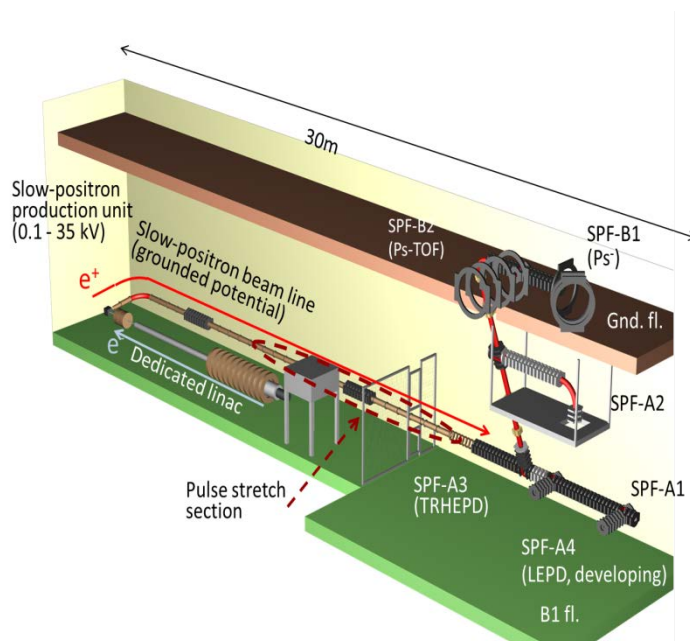


Figure 1. Layout of the stations at SPF as of March, 2016. SPF-A3, SPF-B1, SPF-B2 are for TRHEPD, Ps^- spectroscopy and Ps -TOF, respectively. A LEPD stations is under construction on SPF-A4. A pulse stretching section has been added to the beam line.

2. TRHEPD and its applications

The properties of materials are determined by the kinds of atoms composing them and their precise positions. Thus knowledge of the structure of materials is critical in discerning their properties. In bulk materials research, X-ray diffraction is acknowledged as the standard technique used to determine the structure of the crystals of new materials, proteins, *etc.*

In surface research, however, a technique for the definitive determination of the topmost- and near-surface atomic configurations is yet to be established. This has resulted in a number of studies where possible atomic geometries consistent with the observed characteristics are proposed; as a consequence, sometimes many different models are proposed by various authors.

Recently, the TRHEPD technique, formerly called RHEPD [2-4], is developing to be a standard method for surface structure determination. Its surface sensitivity originates from the inelastic scattering, as is the case for electron diffractions (LEED and RHEED), i.e., once a particle undergoes inelastic scattering, it loses coherence to contribute to the diffraction pattern. In addition, TRHEPD has another feature which gives its unique in its surface sensitivity, i.e., the electrostatic potential inside every solid is positive and thus the surface is repulsive to a positron. As shown in figure 2, a positron directed at a surface with a glancing angle smaller than a certain critical angle, θ_c , whose value depends on the material and the energy of the incident positron, is totally reflected. In this condition, the diffraction pattern depends only on the atoms on the topmost surface [5]. It is also possible to get information on the immediate subsurface by increasing the glancing angle across θ_c . In this condition, the positron is refracted into the crystal in the direction toward the surface, in contrast to the electron which is refracted away from the surface and penetrates deeper. TRHEPD is the only technique in which the Bragg diffraction condition and the total reflection condition happen to be simultaneously satisfied in a considerable angular range of measurement. Furthermore, since TRHEPD is based on the self-interference of an elastically scattered positron, just as the other diffraction techniques, it is very sensitive not only to the positions of the atoms but also to the kinds of atoms.

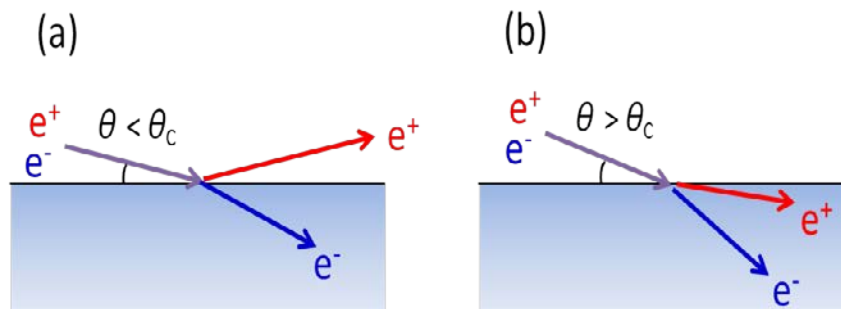


Figure 2. When the incident glancing angle, θ , is smaller than a critical angle θ_c , positrons are totally reflected (a). When $\theta > \theta_c$, positrons are refracted into the crystal in the direction towards the surface (b). The electrons are always refracted into the crystal in the direction away from the surface.

To achieve useful data in diffraction experiments, brightness enhancement of the beam is important. The positron beam for TRHEPD is transported with an energy of 15 keV, guided by a magnetic field. It is released into a magnetic-field free region and then focussed onto a spot of diameter smaller than 1 mm on a 100 nm-thick W foil remoderator, kept at an electrostatic potential of 10 kV[6]. The positrons thermalize in the foil and are emitted from the back of the foil as a quasi-point source with a kinetic energy of 3 eV. The electrostatic potential of the foil then accelerates the positrons to 10 keV as required for the TRHEPD experiments. This process reduces the normalized emittance by two orders of magnitude, while the intensity is reduced by one order of magnitude owing to annihilation inside the foil. The brightness, *i.e.* intensity/ (normalized emittance)², is thus enhanced by three orders of magnitude. The spot positron source is analogous to a W filament point electron source.

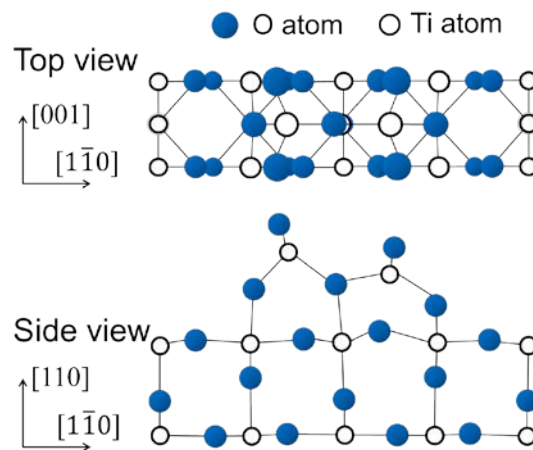


Figure 3. The top view and the side view of the structure of rutile-TiO₂ (110) (1×2) surface determined by TRHEPD(experiment) [7] and USPEX (density functional theory) [11]

Recently, TRHEPD has been used in the determination of the structure of the rutile-TiO₂(110) (1×2) surface [7], which had been under debate over the past 35 years [8]. TiO₂ is used in a variety of applications including photo-catalysts, metal-nanoparticle catalyst supports, gas sensors and corrosion-protective coating materials [9]. Knowledge of the surface structure, where the catalytic processes occur, is crucial for studying the fundamental basis of the reactivity and reaction mechanisms of solid

catalysts. The rocking curves of the 00-spot obtained from the TRHEPD patterns were compared to the curves for various models calculated based on a dynamical diffraction theory, followed by further adjustment of the atomic positions to get a better agreement. The experimental curve matched the curve for the model consisting of Ti_2O_3 composition, originally suggested by Onishi and Iwasawa [10], but with a further modification of atomic positions as shown in figure 3. This was found to be close to the model proposed by Wang et al. [11] after a global search made by changing both the chemical composition of the surface and the atomic positions with the USPEX code.

Graphene is attracting increasing attention with focus on its features such as high carrier mobility, high thermal conductivity and robust mechanical properties [12]. Theoretical calculations demonstrated that the nature of the spacing between graphene and a metal substrate can be classified into two groups, depending on the interaction between them: weak interaction with simple and noble metal substrates; and strong interaction with transition metal substrates [13-15]. Thus, the experimental determination of the spacing between graphene and the substrate is crucial to elucidate the origin of the electronic properties of graphene. Although the spacing had been extensively investigated theoretically, reports on its experimental determination were limited. For example, the spacing between graphene and the Cu(111) substrate was yet to be confirmed experimentally, whereas a scanning tunneling microscopy study showed that the spacing between graphene and Co(0001) was in the range 1.5 to 2.2 Å [16].

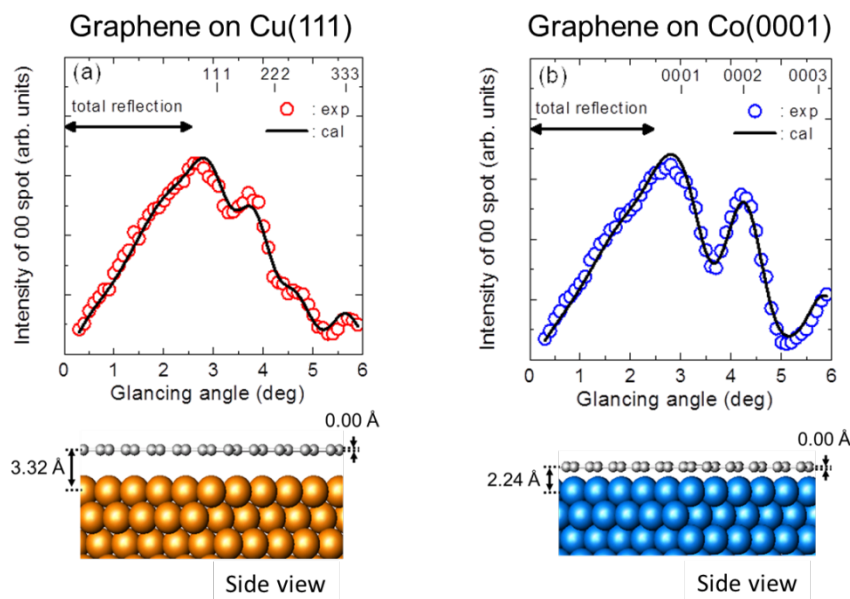


Figure 4. The TRHEPD rocking curve analysis revealed that the spacing between graphene and the Cu surface (left) is much larger than that between graphene and the Co surface (right), as predicted by theory.

As demonstrated in the study of silicene on a Ag(111) substrate [17], TRHEPD is most suitable for studying the possible buckling of a single atomic layer and its distance from the substrate surface. Rocking curve analysis based on the dynamical diffraction theory shows that the spacing between graphene and a Cu(111) substrate is 3.34 Å and that between graphene and a Co(0001) substrate is 2.06 Å (figure 4) [18]. The former is close to the interlayer spacing in graphite, experimentally verifying a weak interaction with a noble metal substrate, whereas the latter small value verifies a strong interaction with a transition metal substrate. The study also confirmed that graphene on these substrate metals do not have buckling, in agreement with what had been expected.

Recently the structure of germanene on Al(111) was measured and a structure which was not expected from the previous theoretical and experimental studies was revealed [19].

3. Observation of a resonance in the photodetachment process of Ps negative ions (Ps^-)

Positronium (Ps) negative ions, Ps^- ($e^-e^+e^-$), are efficiently produced from positrons which are injected into a Na-coated W surface and come back to the surface after thermalization [20]. The ions are accelerated to a desired energy by an electrostatic field, followed by the removal of one of the electrons to make an energy-tunable, neutral Ps beam [21]. Since the constituents of the ion are solely point-like leptons with the same mass, Ps^- provides an excellent testing ground for the three-body problem in quantum mechanics. While theoretical works on its energy levels and dynamics *etc.*, have been extensively performed, experimental investigations of its characteristics had been hampered by the small ion yield and short annihilation lifetime.

Theoretical studies suggest that Ps^- does not have an excited state but only a ground state where the two electrons have opposite spins [22, 23]. However, quasi-bound states (resonances) are predicted in the vicinities of the formation thresholds of the excited states of Ps (of principal quantum number $n \geq 2$) [24]. Photodetachment via this state is possible and the interaction of this process with the direct detachment process gives characteristic structures on the cross section.

Recently, a laser spectroscopic study of Ps^- was performed and a shape resonance near the Ps ($n=2$) formation threshold was observed [25]. The width of the positron pulse using the short pulse mode at KEK, 12 ns, is suitable for investigating the interaction with a high-intensity pulsed laser beam of similar width. A positron beam with a kinetic energy of 4.2 keV and a repetition of 50 Hz was transported to the measurement chamber on branch SPF-B1. It was deflected by 45° along a curved magnetic field, and impacted onto a W target coated with 0.3 monolayer of Na. Some positrons diffusing back to the surface after thermalization in the bulk form Ps^- ions, which are then emitted spontaneously with a small kinetic energy. The proportion of Ps^- ions to the total incident positron flux is reported to be as high as 2% [20].

The Ps^- ions, accelerated by an electrostatic potential, were intersected by a laser beam from a tunable dye laser in the electric field-free region between two grids kept at the same electrostatic potential. Neutral Ps atoms were formed both by the direct photodetachment process and via the shape resonance. *Para*- Ps and *ortho*- Ps are formed but the *para*- Ps self-annihilates quickly (125 ps). Only *ortho*- Ps travels to the MCP, which was placed at a distance of 0.88 m from the Ps^- formation target, and thus detected. The charged particles were removed by the curved magnetic field.

Figure 5 shows the Ps count rate measured as a function of the laser wavelength for the acceleration voltages of $V=3400$ V and 1500 V. Peaks corresponding to the resonance were clearly observed in both cases. The solid lines are fitted model profiles in the laboratory frame, with the Doppler-broadening and the longitudinal Doppler-effect taken into account. From this data, the resonance energy in the rest frame of the ions was deduced to be $5.437(1)$ eV.

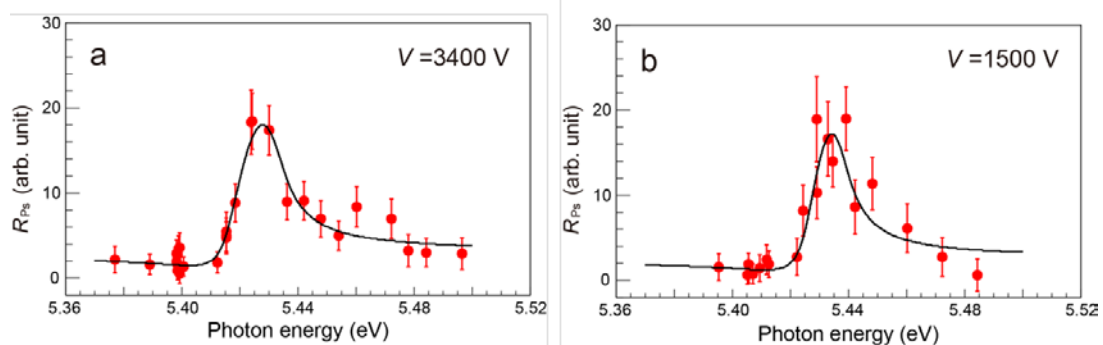


Figure 5. Shape resonance of the Ps^- photodetachment at different acceleration voltage V [25].

4. Ps time-of-flight (Ps-TOF) spectroscopy from alkali-metal-coated W surfaces

The mechanism of large enhancement of the Ps^- production from alkali-metal-coated W surfaces is not yet fully understood. Changes in the work functions of the electron and the positron upon coating may be relevant. If this is the main cause of the enhancement, it may be possible that the coating does not have an effect on the Ps formation probability since Ps consists of one electron and one positron, whereas Ps^- consists of one positron and two electrons.

In order to check this idea, Ps-TOF measurements were performed. Positrons of an energy of 4.5 keV and a width of 12 ns (electron linac short pulse mode) were transported to the Ps-TOF measurement chamber on SPF-B2. The beam was collimated by an aluminum plate with an aperture of 6 mm in diameter before entering the chamber.

One of the γ -rays emitted when *ortho*-Ps self-annihilates in flight was detected by a scintillation counter behind a lead slit set at a fixed distance from the sample. The annihilation γ -rays from the target were also recorded and used to determine time 0 of the TOF spectra.

As shown in figure 6, Ps emission was remarkably enhanced by the addition of sub-monolayers of various alkali metals [26, 27]. It was also revealed that the low energy tail of the spectra has a component with a specific energy loss which varied with the alkali metal [27]

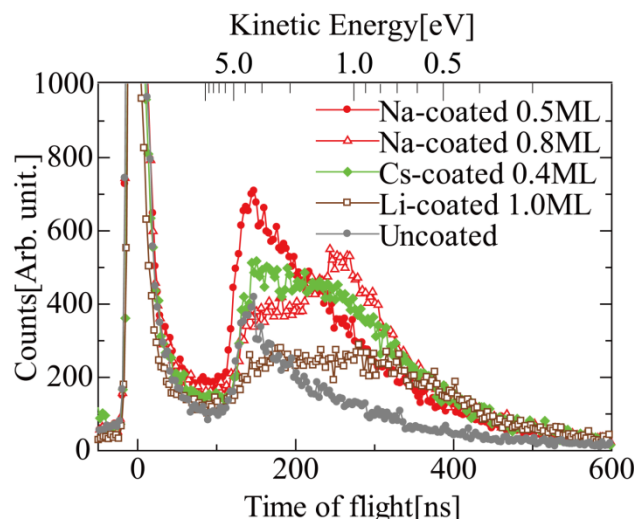


Figure 6. Ps-TOF spectra of W surfaces coated with submonolayers of alkali metal as indicated.

5. Positron beam pulse-stretching section

A pulse-stretching section has been installed in the beam line, which stretches the slow positron pulse of a width of 1.2 μs (in the long pulse mode) to a variable width of up to almost 20 ms at a beam energy of 5 keV. The purpose of this system is twofold. Firstly, the stretched beam will be used for the position-sensitive delay-line detector of the LEPD system, for future spectroscopic studies of the 511-keV annihilation γ -rays such as coincidence Doppler broadening (CDB) spectroscopy and angular correlation of annihilation radiation (ACAR), and for the construction of a short pulse producing system for positron lifetime spectroscopy (PALS). Secondly, it provides the know-how to improve a similar system for PALS experiments at the National Institute of Advanced Industrial Science and Technology (AIST). There, after stretching the original pulse of width 1-4 μs [28], the beam is chopped and pulsed to have a ~ 100 -ps width, which is suitable for PALS [29, 30]. The efficiency of the system will be improved if we have a pulse-stretching section with low intensity loss and with almost constant outlet beam energy.

The system at KEK-SPF is essentially a Penning-Malmberg trap as is the one at AIST [28]. The entrance electrode voltage, normally kept at 5.5 kV, is temporarily lowered to 4.5 kV to accept a 1.2

μs wide positron pulse with an energy of 4.8 ± 0.05 keV into a 6 m long trapping electrode. The voltage of the entrance electrode is then raised back up to 5.5 keV before the positrons reflected at the exit electrode, kept at 5.0 kV, return. The voltage of the trapping electrode is then increased gradually, letting the positrons spill over the exit electrode, which is kept at a constant voltage of 5.0 kV. By adjusting the sweeping speed of the trapping electrode voltage, we obtain a beam of controllable pulse width of up to almost 20 ms with a fixed energy of 5.0 keV, operated at 50 Hz synchronized with the linac operation.

The performance of the pulse-stretching section was examined by letting the positrons annihilate on a closed gate valve downstream. The annihilation γ -rays were detected by a plastic scintillator mounted on a photomultiplier tube. Its anode signals were recorded by a digital oscilloscope as shown in figure 7 and counted. It was confirmed that the 50 Hz positron pulses were stretched up to ~ 20 ms at the same frequency with an efficiency of 70%. This technique will be transferred to the AIST PALS system.

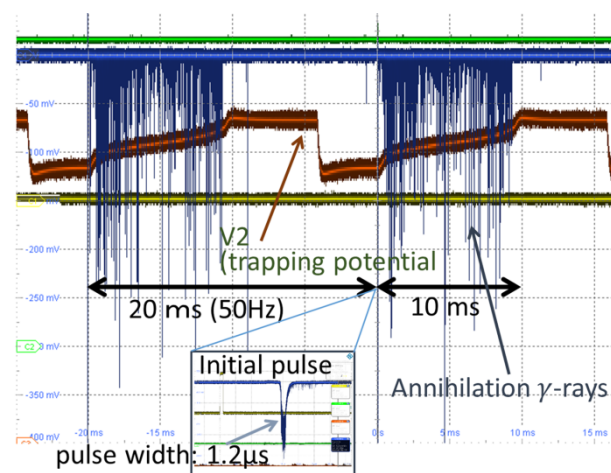


Figure 7. Performance of the pulse-stretching section. The original positron pulse of $1.2 \mu\text{s}$ width (inset at the bottom) is stretched to a width of 10 ms. Stretching up to ~ 20 ms is possible.

6. Summary

Description of the layout of the measurement stations at the Slow Positron Facility in the Institute of Materials Structure Science, KEK is updated. Recent results obtained by using the active stations have been reported. Total-reflection high-energy positron diffraction (TRHEPD) determined the structures of rutile- $\text{TiO}_2(110)$ (1×2), graphene on Cu (111) and Co (0001), and germanene on Al (111). The unique surface sensitivity of TRHEPD is attributed to the positive electrostatic potential common to all the materials. A shape resonance in the Ps^- photodetachment process was observed for the first time. Coating of a W surface with submonolayers of alkali metals, which enhances Ps^- formation, has been found to enhance the formation of neutral Ps as well. A pulse-stretching section which stretches the positron pulse width from $1.2 \mu\text{s}$ up to almost 20 ms has been added to the beam line.

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