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Time-of-Flight Measurements of Positron-Annihilation Induced Auger Electrons

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ABSTRACT

We have developed an apparatus for positron-annihilation induced Auger electron spectroscopy (PAES) with a time-of-flight (TOF) technique. We report the details of the experimental set up and an example of the TOF-PAES analysis for a $MoS_2(0001)$ surface.

I. INTRODUCTION

Auger electron spectroscopy (AES) is widely used for elemental analysis of solid surfaces. In the conventional AES measurements, the core-hole excitations are formed by the bombardment of electrons or photons with energies larger than the core-electron binding energies. The observation depth is determined by the inelastic mean free path of Auger electrons, and is about 10–30 Å. The positron-annihilation induced Auger electron spectroscopy which was developed by Weiss *et al.* [1], uses the annihilations of positrons with core electrons for creating core holes. PAES has the following advantages over the conventional AES. Firstly, the core-holes are created mainly by the positrons trapped in the image-potential well outside the surface, so that the PAES signal originates preferentially from the surface top-layer. For this reason, PAES can easily identify the elements at the surface top-layer. Secondly, the use the primary positron beams with energies less than the core-electron binding energies eliminates the large secondary-electron background around the Auger signals. Thirdly, the high signal-to-background ratio reduces radiation damages.

At the Electrotechnical Laboratory (ETL), a high-intensity slow positron beam is generated with an electron linac. Using this beam, we have developed an apparatus for PAES with a time-of-flight technique, which enables high energy-resolution and high count-rate measurements. In this paper, we show the experimental set up and the performance of the TOF-PAES apparatus, and an example of the analysis for a $MoS_2(0001)$ surface.

II. EXPERIMENTAL SET UP

A slow positron beam ($\sim10^8$ e^+/s) was generated with the electron linac [2]. A 70 MeV electron beam from the linac was incident onto a Ta converter where the bremsstrahlung γ rays generate high-energy (keV–MeV) positrons through pair productions. The positrons were then moderated with a set of thin W foils to form a slow ($\sim$6 eV) positron beam. The slow positron beam was guided to the sample chamber in an axial magnetic field ($\sim$0.01 T) with a series of solenoid coils. The positron beam generated with the linac was pulsed since the electron beam from the linac was pulsed at a pulse width of 1 μs and a repetition rate of 100 pulse/s. However, this pulsed beam cannot be used for the TOF-PAES measurements, because the pulse width is too wide and the repetition rate is too low. Thus, the $\sim\mu$s positron pulse was stretched to $\sim$10 ms with a linear storage section [2]. The quasi-continuous beam was used in the TOF-PAES measurements.

Figure 1 shows a schematic of the TOF-PAES apparatus. The apparatus consists of a pulsing system, an E×B plates deflector, a magnetic field electron paralleliser, and a time-of-flight energy analyzer [3]. An axial magnetic field of 4×10^{-3} T was applied to the whole apparatus for the beam guiding and for the parallelisation of the emitted electrons. The sample chamber was pumped by a cryo-pump to a base pressure of 2×10^{-9} Torr.

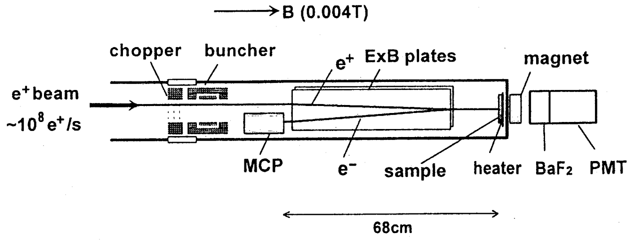

FIG. 1. Schematics of the TOF-PAES apparatus.

The quasi-continuous positron beam was pulsed with a chopper and a buncher. The chopper consists of three mesh grids. A pulsed voltage was applied to the middle grid to make a positron pulse with a pulse width of 30 ns. The buncher consists of two cylindrical tubes, as shown in fig.1. A 20 MHz sine wave potential was applied to the inner tube. The positron pulse was modulated at the gaps between the inner and outer tubes, and was compressed at the sample. The compressed positron beam was deflected with a E×B plates, and then incident onto the sample. The pulse width at the sample was monitored with a BaF_2 scintillator and photomultiplier (PMT) detector, and was 5 ns for the present set up.

An incident energy of 75 eV was used in the present measurements. At this energy, the implantation depth is shorter than the diffusion length of the thermalized positrons. Therefore, a large fraction of the positrons diffuse back to the surface, and are trapped into the image potential outside the surface. Some fraction (less than a few %) of the surface-state positrons annihilate with core electrons of the surface top-layer atoms [4], resulting in the Auger electron emission.

A Nd-Fe-B permanent magnet (0.2 T) was mounted behind the sample to reduce the angular spread of the emitted electrons and to improve the energy resolution. The electrons emitted over 2π steradians from the sample in the high (0.2 T) magnetic field are parallelised in the weaker (4×10^{-3} T) uniform field [5]. The parallelized electrons were separated from the incident positron beam with the E×B plates, and were detected with a micro-channel-plate (MCP) detector which was placed 68 cm away from the sample. The energy distributions of the electrons were determined by measuring the time interval between the trigger signal from the pulsing system and the detector signal with a time-to-amplitude converter.

III. RESULTS

To demonstrate the ability of the apparatus, we chose a layered material, MoS_2 as the target. It is known that a basal plane (0001) surface of MoS_2 is composed of S as the topmost layer and Mo as the second one [6]. The layered structure is suitable for the tests of the surface sensitivity of PAES.

Figure 2 (a) shows a time-of-flight spectrum of the positron-annihilation induced Auger electrons for MoS_2(0001). The incident positron pulse measured with the BaF_2 scintillation detector is also plotted. Figure 2 (b) shows the corresponding energy spectrum. The count rate for the S-LVV Auger peak at ~ 150 eV was ~2 counts/s, and the duration of the measurement was ~1000 s. A large component at energies below 100 eV is due to the secondary electrons excited by the primary positron beam. In the PAES spectrum, the strong S-LVV Auger peak is seen, but Auger peaks from the Mo atoms that exist 1.6 Åbelow the top layer are absent. In contrast, the electron-induced AES showed Mo-NMM Auger peaks at ~180 and 220 eV [7]. This is because PAES probes preferentially the surface top-layer whereas the electron-induced AES probes a few atomic layers beneath the surface.

In the PAES spectrum, small Auger peaks from C (KLL) and O (KLL) contaminants are seen also. It should be noted that before the measurement the sample was annealed up to ~800 °C for surface cleaning, and that the electron-induced AES spectrum taken for the same surface showed no Auger peak from the contaminants. The result demonstrates that PAES is extremely sensitive to impurities adsorbed on the surface. The high sensitivity to impurities implies that the surface-state positrons are efficiently trapped at impurities and defects on the surface [7].

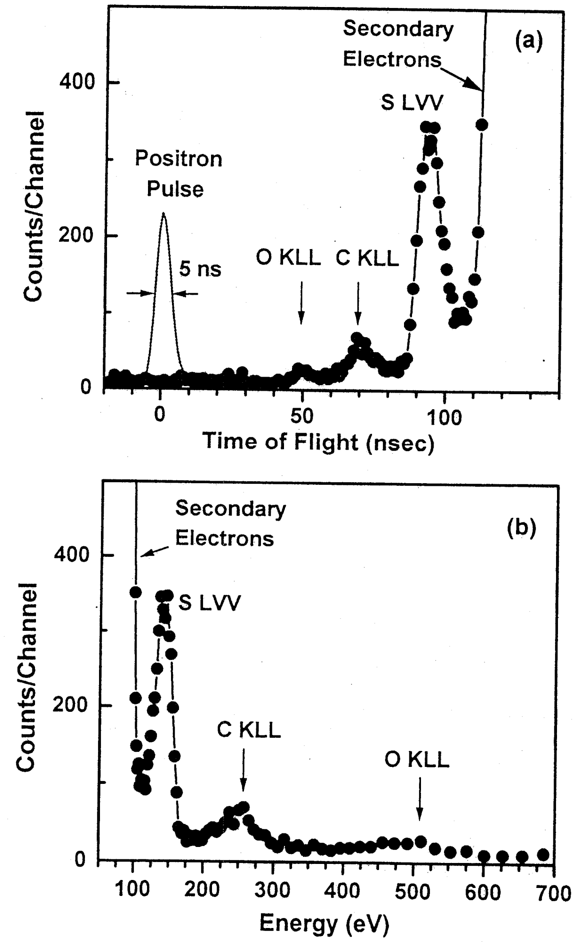

FIG. 2. (a)Time-of-flight spectrum of positron annihilation induced Auger electrons for MoS_2(0001). (b) Corresponding energy spectrum.

It is clear from fig. 2 (a) that the energy resolution of the present apparatus is determined mainly by the pulse width of the incident positrons. For the positron pulse of 5 ns width, the energy resolutions at 150 eV and 500 eV are ~20 eV and 100 eV, respectively. The resolution of the present apparatus is better than that of the previously reported apparatus [5,8], but is not good compared with the conventional AES apparatus with the CMA. The use of a shorter pulse would improve the energy resolution. For the present apparatus, however, it was not possible to obtain the positron pulse with width less than 5 nsec. This is because the magnetically guided positron beam was not monoenergetic but had an energy spread of ~2 eV. In order to overcome this problem and improve the energy resolution, we are constructing a new TOF-PAES apparatus with a longer flight distance and a more efficient pulsing system.

In summary, we developed the apparatus for PAES with the TOF technique. We demonstrated the TOF-PAES analysis for the $MoS_2(0001)$ surface, and found that PAES probes only the surface top-layer, and that PAES is very sensitive to surface impurities. Further improvements are needed to increase the energy resolution.

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