In-situ positron lifetime spectroscopy of radiation damage by simultaneous irradiation of slow-positron and ion beams

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Abstract. An in-situ positron analysis system has been developed for positron annihilation lifetime spectroscopy during ion beam irradiation. An electron linear accelerator was used to produce a slow positron beam for this system. Positron lifetime spectroscopy of SiO₂/Si and annealed pure Fe samples was successfully demonstrated during simultaneous irradiation with a 150 keV Ar⁺ beam. The lifetime spectra changed with increasing ion dose, indicating a decrease in ortho-positronium intensity (SiO₂) and the decrease in positron diffusion length (Fe).

1. Introduction
Defects induced by neutrons and ions have been important subjects to investigate radiation damage of nuclear materials and to optimize ion implantation processes of semiconductor materials. Radiation induced defects are initially formed as point defects (vacancies and interstitial atoms). Depending on material properties and irradiation temperatures, point defects diffuse, annihilate and form secondary defects [1]. To understand the kinetics of radiation-induced defects, in-situ characterization techniques have been one of the important methods. The most typical in-situ technique is transmission electron microscopy (TEM) combined with ion irradiation [2]. With this technique, the movements of nanometer-sized defects can be observed in the time range from video rates to several hundred seconds (or longer). Because of its benefit, many in-situ TEM systems have been developed.

On the other hand, positron annihilation spectroscopy is recognized as a sensitive probe for atomic-scale open-volume defects below the detection limits of TEM. Because of its sensitivity for vacancy-type defects, in-situ analysis techniques during ion beam irradiation have been developed in recent years. An in-situ positron analysis system using Doppler broadening annihilation radiation (DBAR) with ²²Na-based slow-positron and MeV ion beams was developed by Iwai et al. [3] and successfully applied for various radiation damage studies [3-5]. Positron annihilation lifetime spectroscopy (PALS) can give more direct information (i.e., vacancy size) in comparison with DBAR. Tsuchida et al. reported an in-situ PALS system based on a β⁻-γ coincidence method using high-energy positrons from a ⁶⁸Ge source [6]. Since ion-beam induced defects are formed near the surface, energy-variable slow-positron beams are preferable to get sufficient depth resolutions. Therefore, a combination of PALS and a slow positron beam was introduced in this study. In particular, an accelerator-based positron source is employed for higher positron intensity.
2. Experimental method

An electron beam at about 70 MeV from an electron linear accelerator (LINAC) was converted to a slow-positron beam through pair creation by a Ta converter and a W moderator. A pulse width and a repetition rate of the LINAC were \(~1\) µs and 10 pps, respectively. A slow positron beam was magnetically guided from the W moderator to a sample chamber. An ion accelerator with a radiofrequency (RF) ion source was used to generate ion beams, where ion species from inert gas sources can be accelerated up to 150 keV. A typical ion beam current was in the range from 1 nA to 1 µA.

Figure 1 shows a schematic view of the sample chamber and the pulse-forming system. The incident angle of the ion beam is 45º off the positron beam direction. A positron spot-size measured by a microchannel plate (MCP) was about 1 cm in diameter. The ion spot size measured by burn marks on Kapton sheets was about 1.5 cm in diameter in the case of 150 keV Ar\(^+\). We confirmed that the positron-beam spot was aligned near the center of the ion-beam spot. A sample size was approximately 1.5 cm \times 1.5 cm. Ion-beam current was measured by a Faraday cup installed on the sample holder before and after the PALS measurements.

In our previous study, a linear storage section formed a quasi-continuous positron beam to avoid detector saturation [7]. But no such storage device was used in this study. The pulse width of the electron beam was \(~1\) µs. When an extraction voltage of 5 V was applied to the W moderator, the pulse width increased to \(~20\) µs during the beam transport from the W moderator to the sample chamber. Each single positron pulse was divided and squeezed into fine pulses by chopper, prebuncher and buncher electrodes [7,8]. Annihilation gamma-rays were detected by a detector with a BaF\(_2\) scintillator and a photomultiplier tube. The detector and the chopper signals were recorded by a fast waveform digitizer. The time differences between these two signals were accumulated on a computer to obtain lifetime spectra [9]. The details of the beamline have been described elsewhere [10]. The ion beam was also chopped by deflection-type chopper electrodes. The irradiation timing of ion and positron pulses can be synchronized.

![Figure 1. Schematic diagram of the sample chamber and the pulse-forming electrodes.](image)

3. Results and discussion

A thermally grown silicon-oxide (SiO\(_2\)) layer with a thickness of 1 µm on Si was firstly measured by the in-situ analysis system with a 3 keV positron beam and a 150 keV Ar\(^+\) beam. Note that a high voltage of 3 keV was applied to the sample holder to accelerate positrons. As the high voltage for positrons is also effective for ions, the actual ion-beam energy was 153 keV. The average positron implantation depth and the ion range were calculated to be 0.10 µm and 0.11µm, respectively. All the positrons were implanted into the SiO\(_2\) layer and the positron distribution mostly overlaps the ion distribution.

Figure 2 shows the lifetime spectra obtained before the ion irradiation, during the first ion irradiation and after the ion irradiation. Total counts to take one spectrum were 50k - 100k counts and it took 7 - 14 min. The spectrum before irradiation in figure 2 shows the long tail originating from the ortho-positronium (o-Ps) component in SiO\(_2\). Following ion irradiation the tail region (i.e., the o-Ps
component) decreased because of radiation damage. The measured spectra were decomposed into two lifetime components. The second lifetimes, corresponding to the o-Ps component, were fixed to the initial value. Figure 3 shows the o-Ps intensity as a function of ion dose. The open and closed circles correspond to the spectra with (Beam ON) and without (Beam OFF) the ion irradiation, respectively. As shown in figures 2 and 3, the PALS spectra were successfully measured and the spectrum changes were monitored during the ion irradiation.

![Figure 2. Lifetime spectra of a SiO\textsubscript{2}/Si sample for 153 keV Ar\textsuperscript{+} irradiation.](image)

![Figure 3. o-Ps intensity as a function of ion dose corresponding to figure 2.](image)

A mirror-polished pure Fe (99.994%) sample was also measured by the in-situ analysis system. The sample was annealed at 700 ºC for 0.5h in a flowing H\textsubscript{2}+Ar atmosphere to remove mechanical damage. PALS measurements were performed with a 5 keV positron beam and a 150 keV Ar\textsuperscript{+} beam (The actual ion energy was 155 keV). The average positron implantation depth and the ion range were calculated to be 0.067 µm and 0.053 µm, respectively. Some of the positrons were implanted slightly deeper than the ion-induced defects but this is expected to have a negligible effect on the analysis.

Figure 4 shows the lifetime spectra obtained before the ion irradiation, during the first ion irradiation and after the ion irradiation. Total counts to take one spectrum were 100k counts and it took 25 min on average. The spectrum changes were also observed in figure 4. If the first lifetime is fixed to the Fe bulk lifetime (0.11 ns), the initial spectrum before the ion irradiation can be decomposed into two lifetime components. The obtained second lifetime was 0.364 ns. This lifetime can be ascribed to the surface oxide layer [11]. Only one lifetime component was identified in the case of the other lifetime spectra. Figure 5 shows the lifetimes as a function of ion dose. The average lifetime was plotted in the case of the initial spectrum. In the same way as figure 3, the open and closed circles correspond to the spectra with (Beam ON) and without (Beam OFF) ion irradiation, respectively. The measured lifetime decreased with increasing irradiation dose. This result can be explained as the reduction of positron diffusion length. The positron diffusion length in the same type of Fe was measured to be 0.15 µm. Before irradiation, a large amount of implanted positrons diffuse back to the surface, reflecting the surface lifetime. With increasing dose, defects induced near the surface reduce the diffusion length and the surface lifetime became less influential. On the other hand, the lifetime of the bulk region increased with increasing defect concentration. The curve in figure 5 was obtained as a mixture of these two effects.

The time resolutions of the spectra in figures 2-5 were in the range of 0.32 – 0.46 ns. The peak background ratios of the spectra were ~20. These values were worse than those of typical positron lifetime spectra. We speculate that these values were caused by (1) insufficient total counts, (2) the data handling method for the waveform digitizer, (3) detector noise, and (4) the response of the scintillator. In terms of (1), as the AIST LINAC can be operated at 100 pps, it is possible to increase total counts and reduce statistical errors. In terms of (2)-(4), by improving the control program and the detector setting, it would be possible to get better time resolutions and peak-background ratios.
4. Conclusion

An in-situ analysis system for positron annihilation lifetime spectroscopy during ion beam irradiation has been developed. The slow positron beam was directly generated by a 70 MeV electron beam from a linear accelerator. Preliminary measurements have been successfully performed for a SiO$_2$ layer on Si and annealed pure Fe with 150 keV Ar$^+$ irradiation. The ortho-positronium intensity of SiO$_2$ was observed to decrease with increasing ion dose. The lifetime of the pure Fe decreased with increasing dose, indicating the decrease in positron diffusion length. Both results demonstrated the in-situ monitoring of radiation damage.

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