Analysis of irradiated materials by intense slow positron beams

Kinomura, Atsushi

(出版者 / Publisher)
法政大学イオンビーム工学研究所
(雑誌名 / Journal or Publication Title)
PROCEEDINGS OF THE 37th SYMPOSIUM ON MATERIALS SCIENCE AND ENGINEERING RESEARCH CENTER OF ION BEAM TECHNOLOGY HOSEI UNIVERSITY (December 5, 2018)
(巻 / Volume)
37
(開始ページ / Start Page)
3
(終了ページ / End Page)
6
(発行年 / Year)
2019-02
(URL)
https://doi.org/10.15002/00030339

Analysis of irradiated materials by intense slow positron beams

Atsushi Kinomura[#]
Institute for Integrated Radiation and Nuclear Science, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan

Slow positron beams (variable-energy positron beams) are powerful analytical tools to investigate vacancy-type defects in materials with depth-dependent defect profiles and/or surface layers. In particular, it is very important for ion-irradiated materials in which defect distribution is localized from surfaces to projected ranges. In this paper, fundamental aspects and facilities of positron annihilation spectroscopy using slow positron beams are reviewed as well as analysis examples of irradiated materials such as ion-irradiated semiconductors.

I. Introduction

A positron is an anti-particle of an electron. It has the same mass and charge as the electron but its charge state is opposite (positive). Positrons tend to stay away from atomic nucleuses in solids owing to its charge state. If there are vacancies in crystalline materials or vacant spaces in amorphous materials, positrons are trapped in such locations. On the other hand, positrons annihilate with electrons (pair annihilation) and emit two gamma-rays at an energy of 511 keV for opposite directions. When the positron annihilation occurs in vacancies or vacant spaces, longer positron lifetime and less Doppler broadening are observed. Therefore, evaluation of positron lifetimes or Doppler broadening states can be used to analyze vacancies or vacant spaces in solids. Such analytical techniques are called positron annihilation spectroscopy [1-3].

Positron annihilation lifetime spectroscopy (PALS) and Doppler broadening annihilation radiation (DBAR) are major analytical methods in positron annihilation spectroscopy. Positron lifetimes increase with increasing defect sizes. Extent of Doppler broadening is characterized by S and W parameters that are defined as shape parameters calculated from 511 keV gamma-ray peaks in energy spectra. S parameters increase with increasing defect sizes and/or defect densities. Atomic-scale vacancy-type defects (e.g., single vacancy, divacancy and so on) can be detected by using these techniques. Detection limits (detectable defect sizes) of positron annihilation spectroscopy is better than those of transmission electron microscopy.

II. Slow positron beam techniques

PALS measurements are often performed with so-called ‘bulk measurement’ systems, consisting of radioisotope positron sources and gamma-ray detectors. Usually ^{22}Na radioisotopes are sandwiched with samples and emitted positrons are directly implanted into samples. Such analysis method do not need large scale facilities and many bulk measurement systems have been used. The ^{22}Na positrons have energies in the rage of 0 to 545 keV with an average energy of approximately 200 keV. Positrons implantation depths are almost uniform and hence it is suitable for analysis of bulk, neutron-irradiated and electron-irradiated samples as shown in Fig. 1(a).

However, defects formed by ion irradiation localize from surfaces to projected ranges. Such defect profiles cannot be measured by the bulk measurement method. A slow positron technique have been developed to measure depth-dependent defect profiles.

[#] akinomura@rri.kyoto-u.ac.jp

Positron with wide energy distributions are at first decelerated (thermalized) to the thermal energy (~0.026 eV) by using moderator materials (e.g., W) with negative work functions. Then, thermalized positrons are extracted and accelerated to desired energies. Such positron beams are called 'slow positron beams'. Slow positron beams can be used for thin films and surface coating/modification layers in addition to ion-irradiated materials as shown in Fig. 1(b). As thermalization efficiencies are usually not very high ($\sim 10^{-4}$), positrons generated by electron accelerators and nuclear reactors through pair-creation reactions are used to obtain intense slow positron beams.

Fig. 2 shows calculated depth profiles of mono-energetic (slow) positrons in Si. Positron depth profiles can be chosen by changing positron energies. By scanning positron energies, it is possible to investigate depth information of defects in samples. Indeed, DBAR measurements are often performed as depth dependent measurements by changing positron energies. Such data is called S-E curves where S and E mean S-parameter and positron energy, respectively.

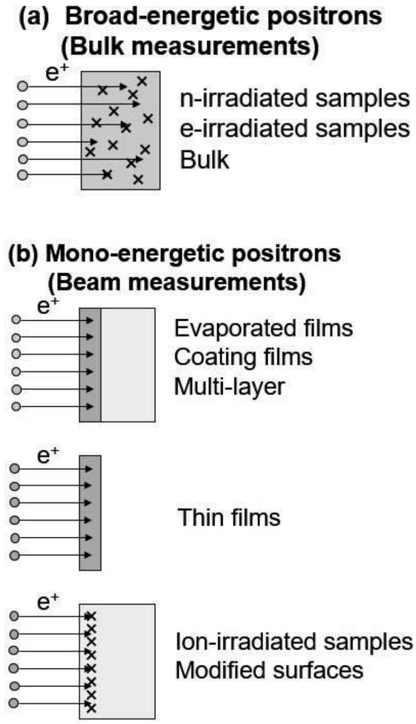

Fig. 1 Difference in bulk and beam positron measurements.

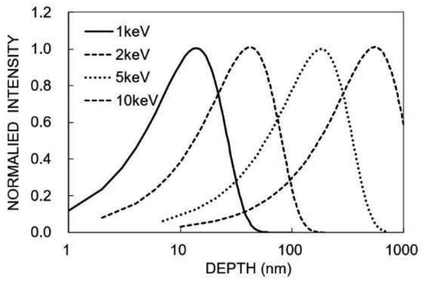

Fig. 2 Depth profiles of positrons at different energies in Si.

III. Examples of irradiated materials analysis

Typical examples of DBAR and PALS measurements can be found in many literatures (see for example, [2] and [3]). As particular examples, let us introduce the following work.

It is well known that high dose hydrogen ion (H^+) implantation in Si followed by thermal annealing forms nanovoids (nanocavities) that act as strong gettering sites for metal impurities. It is important to elucidate fundamental processes of the nanocavity formation process to optimize the gettering efficiencies. Positron annihilation spectroscopy was one of the important analytical techniques to probe nanocaivities in Si [4, 5]. In addition, Rutherford backscattering (RBS) was employed together. In this case, positron annihilation spectroscopy gives information on vacancy-type defects and RBS analysis combined with a channeling technique gives information on interstitial-type defects. Such combination of analytical techniques give unique information on measured samples. Consequently, four types of analytical results (i.e., PALS, DBAR, RBS and RBS/channeling) were compared to understand the evolution of nanocavities during temperature increase. The result supported a model on formation of cracks stabilized by hydrogen absorption during evolution of irradiation-induced damage layer.

Silicon carbide (SiC) has been expected to be a semiconductor material for high efficiency power devices. One of the important subjects is to form fine SiC/oxide interfaces. Previous studies showed that there are disordered states at the interface and they degrade device performance. It is important to investigate interface states by various analytical techniques to overcome this problem. Age-momentum correlation (AMOC) method was employed to investigate the SiC/oxide interface. AMOC is coincidence measurements combined with PALS and DBAR [6]. As one positron annihilation events emits two 511 keV gamma-rays for opposite directions, it is possible to perform coincident measurements corresponding to each gamma-ray. The results indicated that the SiC/oxide interface state was significantly different from states of the oxide layer, bulk SiC or ion irradiated SiC. Such disordered states can be attributed to the origin of the degraded performance of the SiC/oxide interface.

Depth profiles of vacancy-type defects can be calculated from S-E curves obtained from energy-dependent DBAR measurements. Such analysis was applied to investigate depth profiles of ion irradiated layers in metal samples [7, 8]. The S-E curve measurements followed by depth profile analysis showed that the ion irradiation induced damage layer existed at depths much deeper than nominal projected ranges of ion irradiation. The reason of this results was ascribed to partial channeling during ion irradiation, since metal samples are in polycrystalline states and some of the crystalline grains were aligned to the incident ion beam, inducing deeper ion penetration than the nominal projected ranges.

IV. Future plan

A slow positron beamline based on a nuclear reactor has been developed in Kyoto University [9]. Positrons can be generated through electron-positron pair creation from intense gamma-rays from a reactor core. Positron generated and thermalized with W converter and moderator are extracted and transported to a sample chamber. Semiconductor Ge and BF_2 scintillation detectors have been prepared for DBAR and PALS measurements as well as their coincidence measurements (AMOC and coincidence Doppler broadening: CDB). Currently DBAR and CDB have been ready for use. The beamline is opened for external users through the joint use program of Institute for Integrated Radiation and Nuclear Science, Kyoto University.

Usually irradiated materials are analyzed after the irradiation but simultaneous analysis during irradiation (i.e., in-situ analysis) is expected to give further and important information on defect evolution and secondary defect formation during irradiation. To date, various in-situ analysis facilities have been developed. A popular technique is a combination of transmission electron microscopy and ion beam irradiation. High voltage electron microscopy, that can introduce defects by electrons, is similar to this technique. Detection limits of positron annihilation spectroscopy is better than those of transmission electron microscopy. Therefore, an in-situ analysis using positron annihilation spectroscopy and ion irradiation can be an important tool to understand irradiation effects. Based on such motivation, several facilities have been developed in University of Tokyo, Kyoto University and Institute of Advanced

Industrial Science and Technology (AIST) [10, 11]. In particular, the AIST system aimed at detecting transient phenomena by a pulsed positron source based on an electron linear accelerator [12, 13]. The use of these systems was successfully applied to investigate transient states of samples under ion irradiation [10, 14]. It is expected to extend the research on irradiation effects by using this types of in-situ analysis systems.

V. Summary

Positron annihilation spectroscopy using slow positron beams is a powerful and unique tool for analysis of irradiated materials. In particular, it is effective for ion-irradiated materials of which defect profiles are localized near the surface. Intense positron sources using pair creation reactions are required for slow positron beams. For this purpose, a reactor-based slow positron beamline have been developed at Kyoto University. In-situ analysis techniques with simultaneous irradiation of positron and ion beams are expected to give important information for irradiated materials.

Acknowledgement

We thank many colleagues of AIST, Kyoto Univ., Tohoku Univ., KEK and Tokyo Univ. of Science for their invaluable help on conducting research and the development of the beamlines.

References

[1] P.J. Schultz and K.G. Lynn, Rev. Mod. Phys. 60 (1988) 701.
[2] P. Asoka-Kumar, K.G. Lynn and D.O. Welch, J. Appl. Phys. 76 (1994) 4935.
[3] R. Krause-Rehberg and H.S.Leipner, Positron Annihilation in Semiconductors Defects Studies (Springer, Berlin, 1998).
[4] A. Kinomura, R. Suzuki, T. Ohdaira, M. Muramatsu, C. He, N. Oshima, T. Matsumoto, H. Tanoue and Y. Horino, J. Appl. Phys. 104 (2008) 034301.
[5] A. Kinomura, R. Suzuki, T. Ohdaira, M. Muramatsu, C. He, N. Oshima, T. Matsumoto, Y. Horino, Physics Procedia 35 (2012) 151.
[6] A. Kinomura, R. Suzuki, N. Oshima, T. Ohdaira, S. Harada, M. Kato, Y. Tanaka, A. Kinoshita, and K. Fukuda, Jpn. J. Appl. Phys. 47 (2008) 8391.
[7] A. Kinomura, R. Suzuki, T. Ohdaira, N. Oshima, K. Ito, Y. Kobayashi and T. Iwai, Journal of Physics Conference Series 262 (2011) 012029.
[8] A. Kinomura, R. Suzuki, T. Ohdaira, N. Oshima, K. Ito and Y. Kobayashi, Surf. Coat. Technol. 206 (2011) 834.
[9] K. Sato, Q. Xu, T. Yoshiie, T. Sano, H. Kawabe, Y. Nagai, K. Nagumo, K. Inoue, T. Toyama, N. Oshima, A. Kinomura and Y. Shirai, Nucl. Instrum. Methods Phys. Res. B 342 (2015) 102.
[10] T. Iwai, H. Tsuchida, Nucl. Instrum. Methods Phys. Res. B285 (2012) 18.
[11] A. Kinomura, T. Iwai and H. Tsuchida, Positron Science 3 (2014) 27 (in Japanese).
[12] A. Kinomura, R. Suzuki, T. Ohdaira, N. Oshima, B.E. O'Rourke and T. Nishijima, Journal of Physics Conference Series 443 (2013) 012043.
[13] A. Kinomura, R. Suzuki, N. Oshima, B.E. O'Rourke, T. Nishijima and H. Ogawa, Rev. Sci. Instrum. 85 (2014) 123110.
[14] H. Tsuchida, S. Mizuno, H. Tsutsumi, A. Kinomura, R. Suzuki and A. Itoh, Materials Research Express 3 (2016) 055201.