Bremsstrahlung Based Positron Annihilation Spectroscopy for Material Defect Analysis

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Abstract. The Idaho Accelerator Center (IAC) has developed new techniques for Positron Annihilation Spectroscopy (PAS) using highly penetrating γ-rays to create positrons inside the material via pair production. γ-Ray induced positron annihilation spectroscopy can provide highly penetrating probes for material characterization and defect analysis. Bremsstrahlung beams from small, pulsed electron Linacs (6 MeV) have been used to bombard the materials to generate positrons, which annihilate with the material electrons emitting 511 keV radiation. We have also synchronized bremsstrahlung pulses with laser irradiation pulses to study dynamic structural changes in material. In addition, we have developed another method using (p,γ) reactions from a 2 MeV proton beam, which induce coincident γ-rays to perform positron life-time spectroscopy. We have showed the feasibility of extending PAS into thick samples and a wide variety of materials and industrial applications.

INTRODUCTION

Positron annihilation spectroscopy (PAS) has been often performed using positrons from radioactive sources or positron beams [1,2]. It has recently been shown that PAS can be performed using γ-rays as primary beam instead of positrons [3,4]. This new technique employs different types of accelerators to induce γ-rays, which create positrons inside the material via pair production. Because of the high penetrability of MeV γ-rays and the high sensitivity of positron to defects, this technique provides a uniquely sensitive highly penetrating probe for material characterization and defect measurements. It allows measurements of defects in thick materials up to tens of g/cm², a thickness that is inaccessible by conventional positron beam techniques or other nondestructive methods. This work shows that Accelerator-based Gamma ray induced PAS(AG-PAS) can be extended to a very wide field. We have conducted AG-PAS in three research areas. First, Doppler broadening spectroscopy (DBS) has been performed using bremsstrahlung beams from a 6 MeV electron Linac to measure stress/strain in thick materials. Second, positron annihilation lifetime spectroscopy (PALS) has been performed using coincident γ-rays induced from proton capture on low atomic number targets. This represents an alternative approach for positron lifetime spectroscopy, which could possibly solve some of the PALS problems associated with standard positron lifetime technique. In addition it enables PALS in thick materials. Third, we have synchronized short pulsed laser irradiation of materials with Bremsstrahlung pulses from a 30 MeV electron linac and performed Doppler broadening spectroscopy during the pulses to dynamically study structural changes in matter during and following laser excitation in a short time domain. Our goal was to evaluate the momentum distributions of electrons during such changes. These kinds of dynamic measurements may provide detailed information about structural changes in atoms and molecules and lead to better understanding of the mechanisms of defect formation and evolution.

DOPPLER BROADENING SPECTROSCOPY USING A 6 MEV ELECTRON LINAC

Doppler broadening spectroscopy was performed using bremsstrahlung beams instead of positrons to investigate the dependence of Doppler broadening on the applied stress in thick materials. A 6 MeV electron beam from a pulsed electron Linac produced bremsstrahlung on a W foil. The bremsstrahlung beam was collimated and then irradiated the specimens resulting in pair production and other electromagnetic processes. Positrons induced from pair production
annihilate with the material electrons, emitting 511 keV annihilation radiation. The experimental setup and the first measurements made on steel specimens have been explained in detail in ref. [3]. Measurements here were performed on cylindrical tensile specimens (10.16 cm long, 2.54 cm gauge length and 0.64 cm gauge diameter) of Zr-4, which had been subjected to varied amounts of tensile loading using a material-testing system machine. These tensile specimens were machined from annealed Zr-4 alloys in the longitudinal rolling direction. Each specimen was exposed to the bremsstrahlung beam and the spectrum was recorded during the pulse with a HPGe detector. The energy resolution was 1.1 keV at the 356 keV Ba-133 line and 1.3 keV at the 662 keV Cs-137 line.

Doppler broadening of the annihilation peak for each specimen has been analyzed in terms of line shape parameters S, W which are often used to describe the annihilation peak [2]. S represents the fraction of annihilation with low momentum electrons in the total peak whereas W represents the fraction of annihilation with high momentum electrons. We define t as the ratio between W and S. Figure 1 shows relative t parameter as a function of applied stress, where relative t is defined as the measured t-parameter for each specimen divided by t reference (the unstressed specimen).

The high statistical errors are due to the low counting rate associated with the pulsed beam, since the repetition rate of the electron Linac is only 200 Hz.

**PROTON CAPTURE INDUCED COINCIDENT γ-RAYS FOR LIFETIME SPECTROSCOPY**

A new technique has been developed and reported in our previous paper [4] to perform lifetime spectroscopy based on inducing coincident γ-rays from proton capture on low atomic number targets. We have used a 1-1.5 MeV proton beam from a Van deGraff accelerator to bombard a thin aluminum window mounted at the end of the beam line. The reaction $^{27}$Al(p,γ)$^{28}$Si induces coincident γ-rays of 2.8 MeV and 1.78 MeV. The positron birth was indicated by the detection of the 1.78 MeV quanta whereas the 2.8 MeV quanta bombards the material under investigation, to create positron via pair production. The positron death was indicated by the detection of one of the two 511 keV photons emitted from the positron annihilation event as illustrated in Figure 2.

Dislocation lines or any change in the spacing of the lattice planes due to applied stress act as open volume defects. They form an attractive potential that trap positrons at these sites. This leads to more contribution from annihilation with valence electrons and consequently a decrease in the line width of the 511 peak, an increase in S parameter and a decrease in W and t parameters. This effect is clearly apparent in the decrease of t parameter with stress up to the maximum stress in Figure 1. The last point represents the specimen, which was stressed up to the breaking point.
This technique differs from conventional positron lifetime spectroscopy because it yields a spectrum with single exponential positron decay from the pure free defect material with no other lifetime components. One of the biggest problems of standard lifetime technique is that the measured spectrum always contains other lifetime components due to source and surface effects.

A 20 MeV pulsed electron beam was converted to pulsed bremsstrahlung beam through a stainless steel window. The resulting photon beam was doubly collimated with 0.5" and 0.75" lead collimators. The collimated bremsstrahlung beam was incident upon a silicon target of 25mmX25 mm size and 9 mm thick. At the same time, the laser beam was transported to the silicon target through a mirror and a concave lens to expand the beam diameter to 25 mm to match the target size. The laser pulses and bremsstrahlung pulses are synchronized such that the bremsstrahlung pulse is in the center of the laser pulse. A signal from the electron gun was used to trigger the laser. A BaF$_2$ crystal coupled to a fast photomultiplier tube and a plastic scintillator coupled to another fast photomultiplier tube, were used to synchronize the laser and bremsstrahlung beam. The plastic scintillator detects the laser pulse and provides a start signal whereas the BaF$_2$ detector detects the bremsstrahlung beam and provides a stop signal. The timing spectrum was used to feedback to the laser trigger and maintain the bremsstrahlung-laser synchronization. Figure 4 represents a schematic drawing of the experimental setup. The laser source can provide three wavelengths at 1064, 532 and 266 nm. In our initial measurements, we chose the 1064 nm wavelength to allow the penetration of the laser beam throughout the thickness of the silicon target. The laser beam also contains 1% of its initial energy as green light to feed the photomultiplier tube with a signal. Both the laser and the electron accelerator were operated at 10 Hz. Each laser pulse delivers 660 mJ to the target resulting in an average power of 6.6X10$^7$ W and a power density of 1.3X10$^7$ W/cm$^2$. To measure the Doppler broadening of the annihilation radiation, a high-energy resolution HPGe detector was used to record the 511 keV photons emitted from the silicon target during the laser pulses. The measurements have been also performed on the Si target before and after laser irradiation.
From the Doppler broadening spectrum, we can extract the one-dimensional momentum distribution of the electron-positron annihilating pairs [2]. Since positrons thermalize before annihilation, the measured distribution reflects only the momentum distribution of electrons. Figures 5 and 6 show the cumulative and probability distribution function (CDF and pdf) of the electron momentum for bulk Si during laser pulses and after laser irradiation. The cumulative curve of the momentum distribution obtained during laser pulses decays slowly with increasing momentum compared to the distribution obtained after laser irradiation. This indicates more contribution from high momentum electrons during laser pulses. From their probability density functions in Figure 6, differences are clearly visible in the low momentum region ($P_L < 5 \times 10^{-3} m_0c$), which reflects the contribution from valence electrons and in the high momentum region ($P_L > 12 \times 10^{-3} m_0c$). The Doppler broadening of the 511 keV peak has been analyzed in terms of S parameter. The obtained values for the bulk Si are: $S_1 = 0.4492 \pm 0.0162$ before laser irradiation, $S_2 = 0.4463 \pm 0.0204$ during laser pulses, $S_3 = 0.4710 \pm 0.0204$ after laser irradiation. The higher S value measured after laser irradiation indicates that the laser pulses induced permanent damage in the Si lattice. This is probably due to the generation and propagation of shock waves through the lattice because of the high power short laser pulses [6]. During laser irradiation, instead of seeing an increase in S parameter and more contribution from low momentum electrons in the measured momentum distribution because of the induced damage and thermal effect, we have seen the opposite. This might be due to nonlinear effects associated with the high power laser, since the electric field of an intense laser can perturb the states of electrons and atoms [7]. Another question arising from these measurements concerns the effect of laser-induced phonon excitations on the overlap of positron wave functions with both valence and high momentum electrons. No theoretical predictions or measurements are available regarding this issue.

**CONCLUSION**

We report the first experiment to dynamically measure electron momentum distributions during structural changes in materials induced by laser excitation. Our initial bremsstrahlung laser synchronization experiments demonstrate the possibility of obtaining detailed information about the structural changes in materials during laser excitation by AG-PAS. However, to derive strong conclusions about the effect of laser excitation on the electron momentum distributions, additional experiments must be done. On the other hand, our previously reported results demonstrated the development of AG-PAS. However, we have focused in this work on the major issues of AG-PAS as a highly penetrating probe for material characterization and defects measurements by performing both Doppler broadening and positron lifetime spectroscopy on thick materials.

**REFERENCES**