Materials Analysis Using Positron Beam Lifetime Spectroscopy
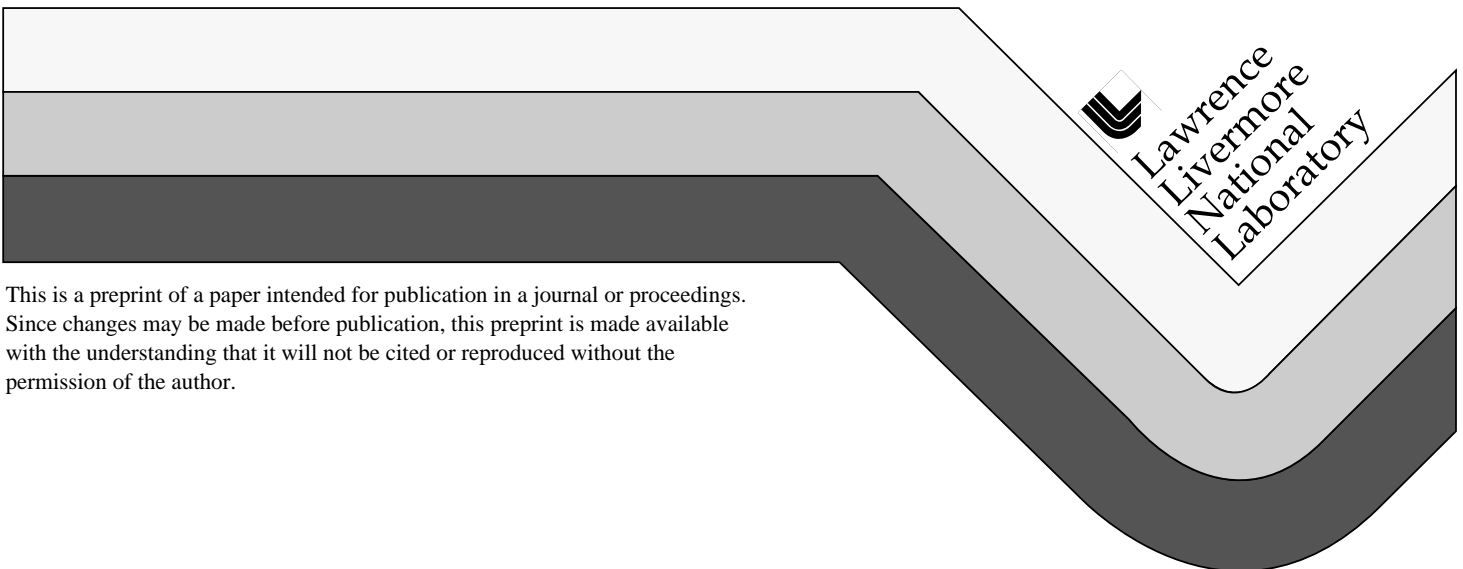
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We are using a defect analysis capabilities based on two positron beam lifetime spectrometers: the first is based on a 3 MeV electrostatic accelerator and the second on our high current linac beam. The high energy beam lifetime spectrometer is routinely used to perform positron lifetime analysis with a 3 MeV positron beam on thick sample specimens. It is being used for bulk sample analysis and analysis of samples encapsulated in controlled environments for *in situ* measurements. A second, low energy, microscopically focused, pulsed positron beam for defect analysis by positron lifetime spectroscopy is under development at the LLNL high current positron source. This beam will enable defect-specific, 3-dimensional maps of defect concentration with sub-micron location resolution. When coupled with first principles calculations of defect specific positron lifetimes it will enable new levels of defect concentration mapping and defect identification.

1. Introduction

When used as a defect spectroscopy, positron lifetime spectroscopy can provide the defect size, concentration and location for open volume defects in most solid materials. The correlation between low local electron density and larger size defects leads to longer positron lifetimes for larger defects. Using the lifetime results from positron annihilation lifetime spectroscopy and lifetime values obtained from theoretical calculations we can estimate the size and local environment of vacancies, vacancies at dislocations, and voids, and thereby identify unknown defect species in metals, semiconductors and insulators. Concentration values are determined from the fraction of positrons that trap at a defect before annihilating. The rate for trapping in defects is high even for single atomic vacancies and so positron spectroscopies are sensitive to even low levels of defect concentrations, 10^{-3} to 10^{-6}. Positron annihilation lifetimes can be measured by the difference between the time of positron entry into the sample and the later time of annihilation.

The sample volume over which positron annihilation lifetime spectroscopy can be perfromed is defined by the path of the positron. Thus by using a beam of positrons with controled energy we can measure defect ensembles over either large or small volumes.

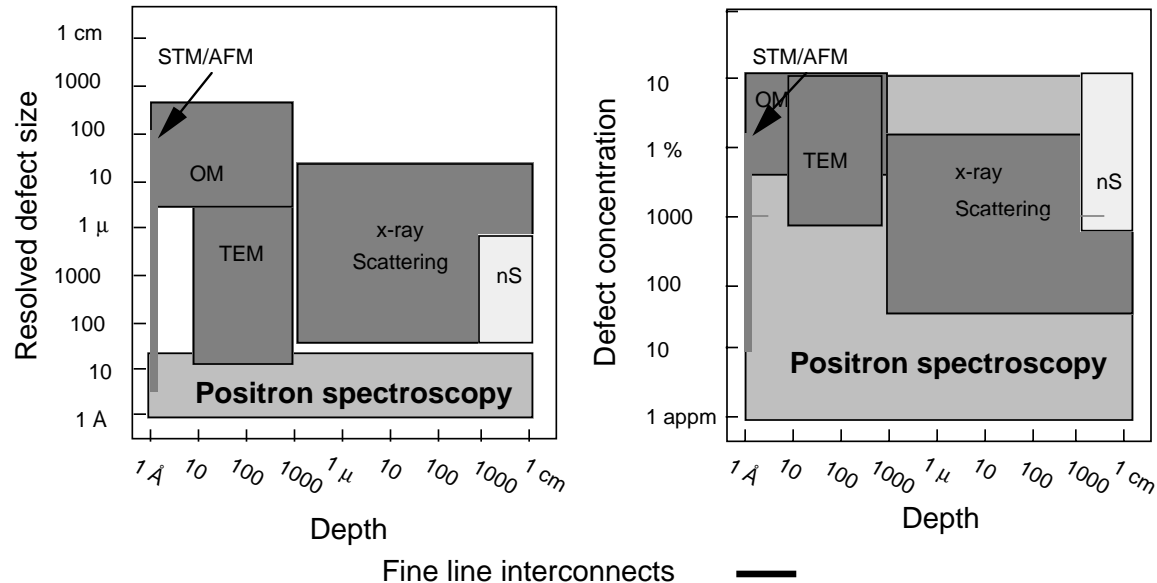Figure 1. Positron annihilation lifetime analysis fills a special niche in the group of techniques for general vacancy defect analysis, optical microscopy, OM, neutron scattering, ns, transmission electron microscopy, TEM, scanning tunneling microscopy, STM and atomic force microscopy, AFM, and x-ray scattering. Positron techniques are both highly sensitive and can resolve the size of atomic vacancies at any depth in a sample.

There are other methods for the general detection of open volume defects. Some of the best known are microscopies such as transmission electron microscopy, atomic force microscopy, scanning tunneling microscopy, and optical microscopy. There are also other defect spectroscopies such as small angle x-ray or neutron scattering. Each of these techniques has specific regions of high sensitivity and resolution. The capabilities of these techniques and positron annihilation lifetime spectroscopy are compared in figure 1. In figure 1-a each method is displayed by its ability to resolve or detect defects of some size at some sample depth. We see that positron annihilation lifetime spectroscopy is effective in providing size information at any sample depth for defect sizes below the effective resolution of other generally applied techniques. Figure 1-b shows the same comparison for defect concentration. Since defect concentrations are determined by the probability of a positron trapping during thermal diffusion, concentration values can also be determined with similar accuracy for any sample depth.

2 MeV positron beam system

We have obtained higher throughput and a significant easing of sample geometry restrictions over conventional radioactive source techniques by performing positron annihilation lifetime analysis analyses with a positron beam accelerated in our 3 MV Pelletron electrostatic accelerator. This system is modeled on one in use at Stuttgart, Germany[1]. Lifetime measurements are performed by detecting the time difference between the time of passage of the positron through a 100% efficient detector outside the sample and the annihilation gamma ray after implantation. This results in single sample measurements in a relaxed geometry with significantly higher counting rates and suppressed backgrounds in the positron annihilation lifetime spectra.

The accelerator-detector configuration is shown in figure 2. Positrons are captured from a 100 mCi source of ^{22}Na and a 2 micron W moderator foil placed in the terminal of a 3 MV Pelletron electrostatic accelerator. Both fast and thermalized positrons are focused by electrostatic lenses in the terminal and a magnetic lens after acceleration. The positron beam is focused onto a 3 mm thick plastic detector that times the implantation of each positron. The positrons are then implanted into the sample and annihilation gamma rays are detected by a BaF$_2$ annihilation gamma-ray detector. Positrons not annihilating in the sample are rejected by a plastic scintillator anti-coincidence detector. The initial positron beam contains $6 \ 10^{5}$ e^{+}/s and final counting rates are ~1000 c/s in a close geometry.
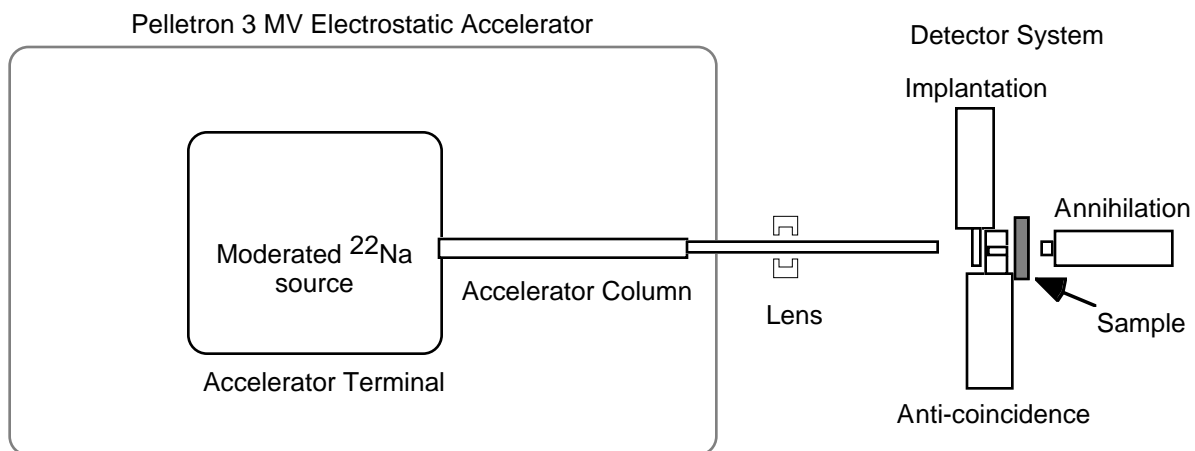

Figure 2.   Schematic diagram of the critical components of the high energy beam positron annihilation lifetime system.

At these rates, bulk average defect analysis in engineering samples of encased materials can be performed on up to 50 samples a day. Energetic, 3 MeV positrons will implant to mm depths in most materials and can penetrate thin windows to reach the sample. Thus measurements can be performed on material encased in vacuum chambers or other necessary containment. With our mm beam spot, defect analysis with this instrument provides bulk values averaged over ~0.1 cm^3 sample volume. The beam-detector geometry also allows coincident measurement of both electron momentum and electron density by separate positron spectroscopic techniques. This provides identification of gas filling in voids and a more detailed description of the local structure surrounding a vacancy defect.

3 Technical capabilities of a pulsed, positron microprobe

We have begun to install and test an intense, pulsed, sub-micron size positron beam for high spatial resolution positron annihilation lifetime spectroscopy. The instrument is designed to run for either spot analysis or in scanning mode to provide a three dimensional map of defect concentrations. By coupling our apparatus to the highest current positron beam in existence (10^{10} e$^+$s^{-1}) we will provide an intense, variable energy (0 to 50 keV), pulsed (less than 100 ps), high intensity (over 10^7 e$^+$s^{-1}) sub-micron size positron beam. Our intense beam and positron microprobe enable us to maintain high data rates while detecting and identifying the depth dependent concentration of vacancies, voids, gas filled voids and other negatively charged defects to a depth of a few microns and at typical depth and lateral resolutions of less than 0.1 micron.

To build the microprobe we are integrating the concepts of existing designs for pulsed positron beams[2,3], focused positron beams[4] and pulsed microprobes[5] into an optimized design to specifically utilize the physical layout, high beam intensity and timing characteristics of the linac positron beam. A diagram of our pulsed positron microprobe system can be seen in figure 4. The pulsed microprobe instrument is physically and logically separated into two stages: a primary stage containing the high current positron beam, a Penning trap beam stretcher, the pulsing system and initial focusing, and a secondary stage containing the final beam acceleration and microscopic focusing. The diagram in figure 4 only shows the apparatus for

the microprobe its self. Figure 3 (inset below) shows the current beam spot on a 40 mm microchannel plate in an electrostatic focus after exiting the stretcher magentic field. The beam spot size is less than 3 mm after stretching, bunching into a 20 Mhz, 1.3 ns duration pulse train.
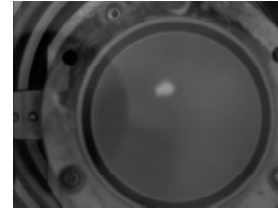

Figure 3. Focused, stretched, pulsed beam spot after exiting the stretcher magnetic field.

Two articles [6, 7] to be published soon describe the details of the microprobe operation and present level of operation.

In addition to the microprobe, the characteristics of the beam at the end of the stretcher are ideal for several experiments requiring high beam current. Such experiments include two dimensional angular correlation of annihilation radiation, positron diffraction, and positron stimulated Auger spectroscopy.

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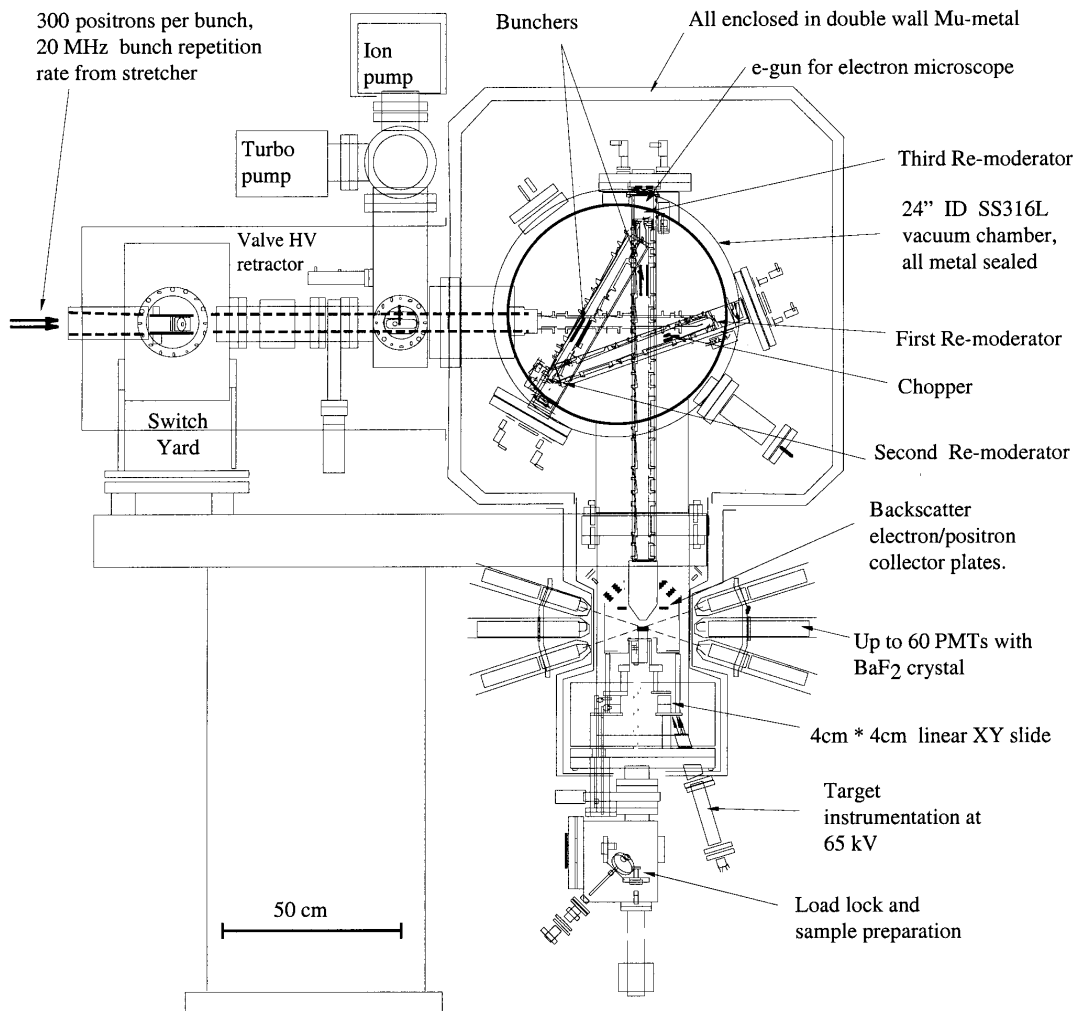
Fig 1. A side view of the LLNL positron microprobe. The stretcher/buncher system introduces beam into the microprobe from the right. The microprobe system is completely enclosed in a double walled mu-metal shell.

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