

Section VI. Facilities

Positron re-emission microscopy and its applications

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Positron re-emission microscopes (PRMs) are distinctly different from electron microscopes in the physical origin of their image contrasts. In a PRM, positrons of several keV energy are implanted into a sample and those positrons that are subsequently re-emitted at several eV are accelerated, focused and imaged. Contrast is produced by any process that affects the transport to, or re-emission from, the sample surface. After an introduction to the basic features of positron microscopy, applications of a PRM in four broad areas of research will be considered. These areas include: materials research, surface catalysis, microelectronic devices and biological systems.

1. Introduction

The first positron re-emission microscopes (PRMs) were built at the University of Michigan [1] and at Brandeis University [2]. Positron microscopes are similar to electron microscopes in much of their optical design, but they are totally different in the physical origin of their image contrasts. In a reflection-geometry PRM of the type discussed in ref. [1], positrons of several keV energy are implanted in a thick target where they thermalize and diffuse. Those that reach the sample surface and are re-emitted are subsequently accelerated, magnified and focused onto a detector such as a channel-electron multiplier array to form an image. Any process that affects the transport to, or re-emission from, the surface can produce image contrast. Such processes include defect and impurity trapping of positrons, changes in the work function of the surface, thin film overlayers and islands, and adsorbates that affect positronium formation or positron surface trapping. After a brief discussion of the essential features of a PRM and its resolution, we will focus the remainder of the paper on PRM applications in four broad research areas where we believe positron microscopy will provide novel information.

In order to understand the unique features of positron versus electron microscopy, we must first present a minimum background on the interactions of positrons in bulk matter and at surfaces. (For a recent review see ref. [3].) Being anti-electrons, positrons are identical to electrons in all respects except charge. Thus they behave in solids in ways that are identical in many respects. The thermalization processes and consequent implantation profiles of positrons and electrons are very similar, and once they are implanted in a solid (at least in metals and semiconductors), the scattering processes

that determine the motions of electrons and positrons are also similar.

A very significant feature of positrons, however, is that they are distinguishable from electrons. There is no way to follow the diffusion history of a particular electron implanted in a target – it becomes lost in the sea of identical electrons in the solid. In the case of positrons, however, it is possible to follow the history of each positron after it has thermalized but before it annihilates. The influence on positron diffusion of such material properties as internal fields, impurity and defect distributions, and spatial changes in composition such as occur in microelectronic devices and in layered structures can all in principle be measured.

A further important feature specific to positrons is their positive charge. Because of it, positrons can participate in many processes not available to electrons. They can trap at negatively charged lattice defects such as monovacancies and other open-volume defects, at impurities, and in image-potential-induced (i.e. “extrinsic”) surface states both at external surfaces and at internal surfaces bounding large open-volume defects such as voids. Moreover, because the surface-dipole contribution to the electron work function is attractive, in the case of the positron work function, this contribution is negative for many materials. Thus positrons are re-emitted into the vacuum from the surfaces of these materials, or emitted into the interior of a large defect. Finally, a positron can bind to an electron in a hydrogen-like atom called positronium. Although a positronium atom cannot exist in the interior of a metal because the electron density is too large, it can exist inside an insulator, and can be emitted from the surface of any material.

PRMs have several practical features that may be very useful in a variety of measurements in which

positron microscopy serves to complement existing electron microscopies. Targets need not be thin as required for TEM analysis – thick robust targets can be used in ultrahigh vacuum with in-situ sample preparation such as ion sputtering, annealing, thin film evaporation, chemical catalysis, and surface analysis. Since the incident positron beam energy is typically less than 10 keV, PRM analysis is virtually nondestructive and thus image analysis can be alternated with incremental changes in sample preparation. In some applications, depth profiling can be accomplished by systematically changing the positron implantation energy and hence the implantation depth. The lateral resolution of current PRMs [1,2] is limited by positron current density, or beam brightness. The ultimate, diffraction-limited resolution is expected to be about 10 Å [1]. We are currently building an ultrahigh-vacuum PRM with three remoderators for beam brightness enhancement. Assuming we can fabricate ^{58}Co positron sources of 100 Ci activity or greater [10], this PRM should have resolution of order 100 Å. Depth resolution should be comparable with the lateral resolution except under certain conditions of positrons tunneling through thin film overlayers in which case monolayer thickness resolution has been demonstrated [4] in non-imaging experiments.

2. Applications in materials research

The diffusive transport of positrons to the surface of a sample can be affected by such material properties as: impurities and defects which trap positrons; internal and applied fields which can drift positrons; and spatial changes in material composition such as are encountered in multilayer systems and with thin-film islands and adsorbates on subsurfaces. In this latter case we have demonstrated [4] monolayer sensitivity of the positron tunneling probability through thin-film overlayers. Under such tunneling conditions we can refer to the PRM as a positron tunneling microscope (PTM). We have discussed PTM in greater detail elsewhere [5]. It should straightforwardly allow nondestructive, monolayer depth resolution of thickness variations in thin films. In addition, PTM should permit imaging of local changes in the bulk chemical potential due to such phenomena as lattice strain in pseudomorphic growth or interdiffusion alloying, even in systems that present no topographical contrast [5].

The sensitivity of PRM to surface defects from ion sputtering was demonstrated in the “defect maps” of ref. [1]. Ion fluences as low as 10^{15} ions/cm² produced high contrast compared with unsputtered regions. The contrast is due to the trapping of positrons at sputter-induced, open-volume defects which reduces the effective positron diffusion length and hence the probability of a positron diffusing back to surface. For comparison, the imaging of secondary electrons produced when an



Fig. 1. A PRM image of a 25 μm thick Mo foil that was annealed for one hour at 1800 $^{\circ}\text{C}$. The area imaged is approximately 500 μm on a side; the magnification is 22 \times ; and the resolution is 2 μm . The image presumably depicts crystalline domain growth or possibly patches of impurities that have segregated at the surface.

electron beam was used instead of the positron beam produced no contrast for defect damage below an ion fluence of 10^{19} /cm². At ion fluences above 10^{19} , the contrast is topographical in origin, as sputter etching was clearly visible in a light microscope.

Another example in which the PRM displays a clear contrast where none was observed for electrons is shown in fig. 1. The target is a 25 μm thick Mo foil that has been annealed for one hour at 1800 $^{\circ}\text{C}$. We attribute the image features to be either randomly oriented crystalline domains [6], or bulk impurities that segregate on the surface in patches. The positron work function, and hence the positron re-emission rate, are known to depend on crystal orientation [3]. Impurity overlayers may simply attenuate positron re-emission from the underlying Mo. Optical microscopes reveal images similar to fig. 1, but secondary-electron images showed no significant contrast.

3. Applications in surface catalysis

The surface morphology and alloy composition of bimetallic catalysts can greatly influence the chemical activity and product selectivity of chemical reactions. Because of the difficulty of performing detailed surface analysis on high-surface-area particulates and aggregates, surface catalysis research often involves model systems consisting of metals deposited onto a single-crystal or thin-film substrate. It is in the analysis of these model catalytic systems that the PRM may

strongly complement existing surface spectroscopies, in particular TEM analysis. There are many bimetallic systems which do not lend themselves to TEM analysis. For high- Z substrates with large electron cross sections (e.g. Ru, for which Cu films activate catalytic activity) the substrate film for TEM analysis would have to be less than 100 Å thick in order to avoid overwhelming the overlayer signal. This film is too fragile for catalysis work – a major problem that leaves many questions about such systems unanswered. In other cases there is little or no TEM contrast if the substrate and overlayer have similar Z -values. Some important overlayer and adsorbate species, such as sulfur and carbon, cannot be imaged at all with the TEM. Using the reflection geometry PRM, substrate targets can be thick crystals and can easily be cycled between a reaction chamber and the microscope, since PRM analysis is virtually nondestructive. In addition, the PRM contrast mechanisms can depend much more strongly on Z than electron scattering cross sections; on islanding of the overlayer and alloying of the metals involved; and on defect concentrations at the surface.

Such contrast mechanisms seem perfectly matched to the needs of surface catalysis researchers where the major questions concerning model systems are:

- (1) What are the overlayer island density, shapes and sizes?
- (2) Has alloying occurred between substrate and overlayer, and is the alloying uniform in concentration?
- (3) Are defects in either the overlayer or the substrate involved in activating catalysis?
- (4) Where do residual adsorbates of the catalytic reaction reside on the surface?

4. Applications in solid-state electronics

Reduction of the size of circuit elements of integrated circuits is one of the keys to general advances in electronic and computer applications. The technology of integrated circuit fabrication is presently nearing the size limits of what can be achieved using traditional methods, methods based primarily on visible or near-UV light optics. Devices are nearing optical-diffraction limits (of order 1000–2000 Å) and in order to further decrease their size new techniques are necessary for device fabrication, device characterization and failure analysis. In the particular area of device characterization, scanning electron microscopy is the most common technique, and this is likely to remain true for future generations of circuits. For many particular uses in this broad field, however, the unique features of positron microscopy should provide information not easily obtained using electron techniques.

In particular, positrons are the most sensitive probe of lattice defects available, being able to detect, for

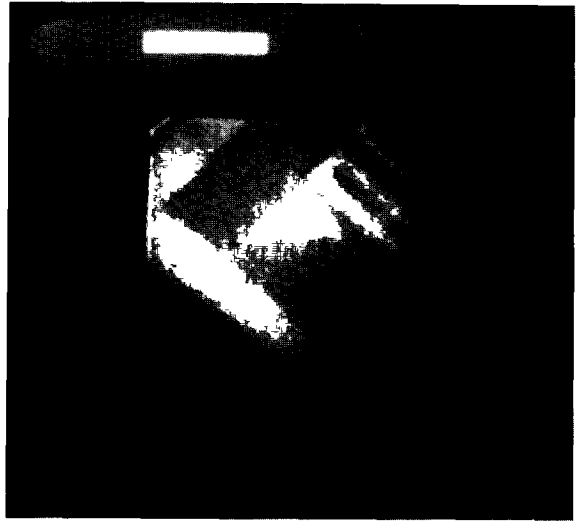


Fig. 2. A PRM image of ion-beam-implanted boron heater elements in an undoped Si substrate. Implantation-induced defects were removed by annealing. The bright areas of high positron re-emission are the B elements and interconnects. The area shown is 200 μm on a side; the magnification is 60 \times ; and the resolution is about 0.7 μm .

example, monovacancy concentrations as low as 0.1 ppm. Defect maps of a semiconductor device obtained with the positron microscope would permit the observation and analysis of damage induced, for example, by thermal stress during the manufacturing process, or by operation at elevated temperatures. This is important because heat removal from high-density devices is a significant problem, and thermal effects will increase rapidly in the higher-density devices expected in the future. Thermal problems are also significant causes of migration of metal ions from gate electrodes and interconnects, another phenomenon which should be visible in positron micrographs (see fig. 2, a demonstration that interconnects are clearly distinguished from a silicon substrate). Yet another important class of defects which can be imaged are those due to radiation damage.

Another type of PRM application involves the investigation of hot-carrier effects in devices. Since the diffusion and drift properties of positrons in a semiconductor are very similar to the corresponding properties for holes, the mapping of spatially varying hole mobilities is feasible using a PRM. To accomplish this mapping, one could introduce a positron beam with a diameter on the order of a diffusion length (about 1000 Å) onto a selected area of a sample. Positrons would then enter areas outside the small-illumination region by the processes of drift and diffusion, broadening the re-emitted beam profile. The drift and diffusion parameters of the sample could then be obtained from a deconvolution of the spatial profile of the re-emitted image.

As we have noted, the PRM can image a thick sample nondestructively. In addition, the ability to depth profile without destructive delayering is of great interest. No current electron technique can compare with positron microscopy in this regard. Examination of defect and impurity profiles near buried junctions or detection of metal diffusing away from gate electrodes in the underlying silicon are just two important practical examples that can benefit from nondestructive depth profiling.

5. Biological applications

The PRM has significant promise for the study of biological cell walls. Particular areas of interest include the high-contrast imaging of protein receptors and the imaging of surface charge distributions on cell walls. High-contrast imaging of protein receptors and imaging of surface charge distributions at resolutions below 50 Å is of considerable interest since alterations in cell surface receptors and surface charge distributions have been implicated in a variety of disorders including cancer and other major diseases. New information provided by the PRM would provide new insights into the etiology of these diseases.

The cell surfaces of higher organisms [7] are thought to consist largely of protein receptors protruding from a thin (~ 10 Å) lipid bilayer. Imaging cell surfaces is a challenge because there are no significant differences in atomic compositions of receptors possessing radically different biological functions, and because of the fragile nature of the cell membrane. Current imaging techniques are intrinsically low in contrast for cell surface receptors, with variations in contrast between proteins and lipids for unstained samples being typically less than a few per cent. For these techniques, staining agents or labeling compounds attached to site-specific antibodies are required to highlight specific cell features. Unfortunately, such techniques alter the biological structure of the sample being investigated. By comparison, the PRM may display high contrast for differentiating proteins from lipids without the use of staining agents or labeling compounds. We have already demonstrated [8] that the PRM can distinguish between different amino acids as well as between amino acids, bases and lipids without the use of staining agents or labeling compounds. This indicates that the PRM has the potential for distinguishing between lipids and proteins on the surface of cell walls. Since the state of aggregation of specific cell surface proteins greatly affects biological function, any new microscopic technique which can visualize lateral protein distribution in planar lipid/protein membranes is of great interest in biophysics.

Another application of the PRM which has no parallel in light or electron microscopies would be in the

study of surface charges on biological cell walls [9]. Most cells bear a net negative charge, which is believed to arise primarily from COO⁻ radicals linked to the membrane proteins. Because of their positive charge, positrons may bind to these negative sites, resulting in contrast in the PRM image, as has been demonstrated using the PRM to observe positron trapping in negatively charged defects [1]. The PRM may also be sensitive to positively charged sites through the mechanism of positronium breakup. Positronium diffusing through the bulk of the sample could lose its electron to a positive charge (electron transfer chemistry), resulting in a local enhancement in the positron re-emission rate. Imaging of surface charge distributions would be of considerable interest, since alterations in such distributions have been indirectly linked to alterations which occur in cells which become malignant. Observation of specific changes in surface charge distributions of particular cell surface receptors in cancer cells would be of extreme interest, since such cells could then be targeted for destruction using site-specific monoclonal or polyclonal antibodies. If resolutions below 50 Å can be achieved, it may also be possible to image ion channels on either real cell surfaces, or else ion channels reconstituted onto model biological membranes. This feature, if realized, would be a major new capability in membrane biophysics.

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