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Development of first ever scanning probe microscopy capabilities for plutonium

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ABSTRACT

Scanning probe microscopy capabilities have been developed for plutonium and its derivative compounds. Specifically, a scanning tunneling microscope and an atomic force microscope housed in an ultra-high vacuum system and an inert atmosphere glove box, respectively, were prepared for the introduction of small non-dispersible δ-Pu coupons. Experimental details, procedures, and preliminary imaging of δ-Pu coupons are presented to demonstrate the functionality of these new capabilities. These first of a kind capabilities for plutonium represent a significant step forward in the ability to characterize and understand plutonium surfaces with high spatial resolution.

1. Introduction

The birth of scanning probe microscopy (SPM) is generally associated with the invention of the scanning tunneling microscope (STM) in the early 1980s, which enabled 3-dimensional atomic-scale imaging of Si(111) [1]. Following this innovation, numerous scanning probe techniques have been and are still being developed, which allow highly localized measurements to be made for almost any kind of tip sample interaction imaginable [2]. Although SPM methods have become ever more ubiquitous with time, their application to plutonium (Pu) materials has been elusive due to the radiological and toxic hazards associated with the material, as is so often the case. This is not, however, for lack of interest or scientific merit in establishing such a capability.

The study of Pu and related compounds is critical in understanding the itinerant-to-localized crossover within the 5f-electron actinide series since this crossover occurs within the allotropes of Pu (δ-phase Pu being at the center of this crossover) [3]. STM and scanning tunneling spectroscopy (STS) techniques have already proven to be powerful tools in the study of emergent phenomena in strongly correlated electronic systems [4,5], and the experimental data provided through this capability are essential to address prevailing limitations in the current theoretical knowledge of Pu materials. [6,7]. Specifically, STM and STS capabilities for Pu would enable quasiparticle mapping of both the occupied and unoccupied states [6], as well as direct observations differentiating between $s^{\pm}$- and d-wave pairing in Pu-based superconductors provided temperatures below T_c can be reached [7].

While STM and STS techniques with atomic scale resolution enable probing of point defects and electronic structure within a material [8], atomic force microscopy (AFM) with its myriad of imaging modes [2] can provide information on surface morphology at greater length scales, as well as surface mechanical properties of materials. AFM probing of Pu and its derivative compounds has the potential to provide significant insights into the details of Pu surface aging (such as oxidation, corrosion, self-radiation damage), reactivity, and morphology along with other surface signatures (revealing its processing history). Described herein is the development of the first ever STM and AFM capabilities for Pu materials.

Abbreviations: AFM, Atomic force microscope/microscopy; Ar, Argon; HEPA, High efficiency particulate air; HOPG, Highly oriented pyrolytic graphite; Pu, Plutonium; RGA, Residual Gas Analyzer; SPM, Scanning probe microscope/microscopy; STM, Scanning tunneling microscope/microscopy; STS, Scanning tunneling spectroscopy; T_c, Critical temperature; UHV, Ultra high vacuum; VT, Variable temperature; δ-Pu (Xat% Ga-stab.), X atomic percent gallium-stabilized δ-phase Pu.

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2. Materials and methods

2.1. Scanning tunneling microscope

An Omicron variable temperature STM (VT STM) housed in an ultra-high vacuum (UHV) system was utilized to establish the Pu STM capability. The UHV-STM system is comprised of an analytical chamber with a base pressure of 3×10^{-10} Torr, a sample preparation chamber with a base pressure of 1×10^{-9} Torr, and a load lock system for introduction and removal of tips and samples. The STM resides in the analytical chamber, which is also equipped with an Omicron Auger electron spectrometer (with 4-grid SpectaLEED) used for determining sample cleanliness prior to STM imaging. Both the analytical and preparation chambers are equipped with manipulators having various sample annealing capabilities, SRS residual gas analyzers (RGA), and leak valves for backfilling and gas dosing purposes. Sputtering is only performed in the preparation chamber (using an RBD backfill ion source) in order to minimize contamination within the analytical chamber.

In order to minimize mechanical and acoustic vibrations that can interfere with atomic resolution imaging, the UHV-STM system is located in a dedicated ground floor room. During imaging all rotary pumps are turned off so that UHV is maintained by ion pumps and titanium sublimation pumps, and in addition to the vibration isolation stage of the STM itself, the entire UHV-STM system is floated on four Newport pneumatic vibration isolators. Prior to the introduction of Pu into the UHV-STM system, the ability of the STM to produce atomic resolution images was verified with and calibrated to well-known standards. Specifically, highly-oriented pyrolytic graphite (HOPG), Si(111) 7×7 reconstruction, and gold coated mica were imaged to verify atomic scale resolution in all three dimensions (Fig. 1).

Two high efficiency particulate air (HEPA) filters were added in series to the exhaust end of all rotary pumps associated with the UHV system to provide containment of Pu within the UHV-STM system and prevent worker exposure to Pu. Additionally, a transport cell was designed to enable introduction (and removal) of Pu samples into the UHV system while maintaining Pu containment. The top left image of Fig. 2 shows the transport cell used to contain Pu samples during transport. These samples are mounted in the Pu facility onto STM sample plates which are then inserted into a metal pocket welded onto the butterfly valve of the transport cell. When the butterfly valve is closed, the Pu sample is contained underneath the valve with the surface of the Pu sample facing down. The dimensions of the mounting plate relative to the cell are such that the sample cannot slide out of the pocket during transport. A port on the bottom of the transport cell (with a small internal HEPA filter) is available for pulling vacuum and backfilling with inert gas if desired. If not used, the port is sealed with a threaded plug. This port can be seen in the right image of Fig. 2.

When the transport cell arrives from the Pu facility, it is attached to a magnetic transfer arm and gate valve (show on the right side of Fig. 2), after which both the butterfly valve and gate valve are opened, exposing the Pu sample to the magnetic transfer arm without losing containment. Once the Pu sample is lifted above the gate valve by the transfer arm, both valves are closed and the

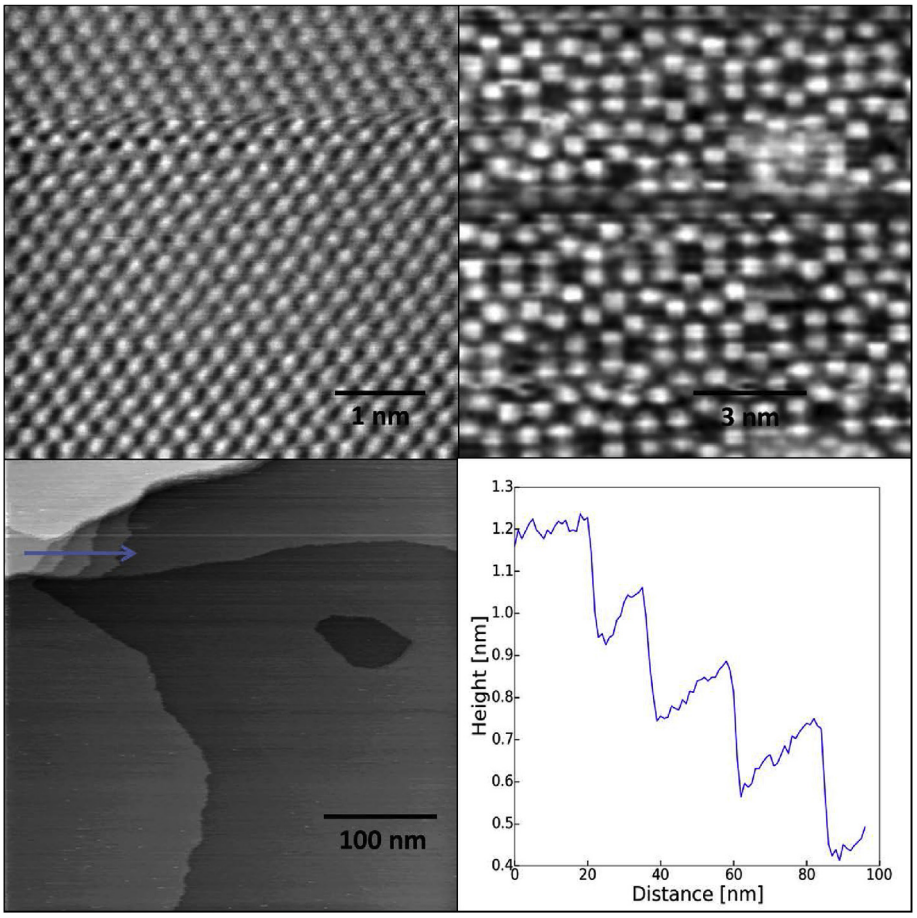

Fig. 1. STM images of well-known standards. Top left: HOPG; Top right: Si(111) 7×7 reconstruction; Bottom left: Gold coated mica. The line path shown in the bottom left image is represented on the bottom right demonstrating the expected monolayer step height along this path.