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Meeting-report

A Comprehensive Examination of Aluminum Oxide (Al_2O_3) Using Extreme and Near Ultraviolet Laser-Assisted Atom Probe Tomography

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Aluminum oxide (Al_2O_3) is a widely used insulating material, often employed as a gate dielectric or separating material in electronics, ceramics, and steels. At the interface of multilayered materials, the microstructure can be crucial to achieve the desired bulk properties but are often difficult to properly analyze with atomic precision. Laser-pulsed atom probe tomography (APT) has been a universal and powerful technique for visualizing the 3D atomic-level structures of complex materials, leading to an enhanced understanding of structure-related properties at the interface of layered assemblies. Near ultraviolet light (NUV) has traditionally been used for laser-pulsed field evaporation of aluminum oxide atom probe tomography, with literature reports of both aluminum-rich [1, 2] and aluminum-poor concentrations [3]. Due to the variations in laser pulse energy, wavelength, applied field, and other variables in APT, the experimentally observed composition may change over several percent. Despite the numerous applications of aluminum oxide, the conditions leading to accurate compositional APT analysis is still lacking, inevitably resulting in potentially inaccurate material analysis. Understanding the experimental variables which lead to measured deviations from stoichiometric concentrations of aluminum oxide and the extent of their effects will lead to better analysis in bulk systems and thus improved materials.

Laser-pulsed atom probe tomography utilizes field evaporation to trigger ionization of atoms, one layer at a time, at the surface of a sharp (tip diameter < 100 nm) sample followed by measuring the ejected ions on a position-sensitive detector (Fig. 1a), producing a 3D reconstruction of the starting material. After detection of each evaporation event, the time-of-flight spectrum is converted to a mass spectrum (Fig. 1b), allowing for atomic and isotopic investigation. After thousands to millions of laser-triggered events, the mass spectra are averaged, and an overall composition profile can be determined. Wide bandgap insulators, such as Al_2O_3, may benefit from high-energy photon-assisted field evaporation in APT due to increased absorption cross sections and photon energies exceeding the optical bandgap. Thus far, NUV has been used and supplies photon energies (< 5 eV) well below the optical bandgap of insulating materials, providing an interesting comparison to extreme ultraviolet (EUV). In our modified atom probe system, EUV photons of ~ 45 eV, pulse durations of ~ 10 fs and a repetition rate of 25 kHz are employed to assist in the field evaporation process, as described previously [4]. EUV-pulsed APT has already been used for the field evaporation of several semiconducting materials, showing the ability to vary the measured atomic compositions [5]. Additionally, changes to the base temperature of the sample may further exacerbate the apparent compositional variation, as previously demonstrated with SiO_2 [6]. With the ability to vary the EUV pulse energy over four orders of magnitude (2.5×10^{-9} J/cm^2 to 1×10^{-4} J/cm^2), several independent field evaporation conditions can be explored and compared to NUV experiments.

In this presentation, the change in composition, peak shapes, and other experimental factors due to the magnitude of the applied voltage, pulse energy, and base temperature of single crystal α-Al_2O_3 using APT will be discussed and compared using both EUV- and NUV-pulsed field evaporation. After varying the base temperature of an aluminum oxide sample at five different EUV pulse energies, it is determined that a nominal 40% Al composition can be achieved at a higher base temperature (> 150 K), using low EUV pulse energies at a constant 0.5% detection rate (Fig. 2a). This is compared to NUV APT, which shows a lower base temperature may be used to achieve similar compositional results at varying pulse energies. Peak tails are also compared between the EUV and NUV spectra as a function of pulse energy (Fig. 2b), which are detrimental to compositional analysis and typically supposed to be caused by a thermal response to NUV photon absorption. The Al^{2+} peak tail using EUV is seen to have a negligible increase due to changes in pulse energy but is confirmed to rise when the NUV pulse energy is increased, consistent with other mass peaks. Finally, the changes in apparent Al at. % composition, detection rate, % of multiple detection events, and experimental background as a function of the pulse energy are compared when using EUV- and NUV-pulsed APT (Fig. 2c). Overall, the composition and related experimental factors of laser-pulsed Al_2O_3 APT are more predictable and sensitive over a wider range of EUV pulse energies over NUV, with negligible increases in the peak thermal tails when using EUV, compared to NUV. Further comparisons will be discussed, where the experimental factors that affect the analysis of APT data may be finely tuned to provide more accurate measurements. Understanding these APT experimental factors may lead to better sample analysis, improved material quality, and higher value material characterization using APT.

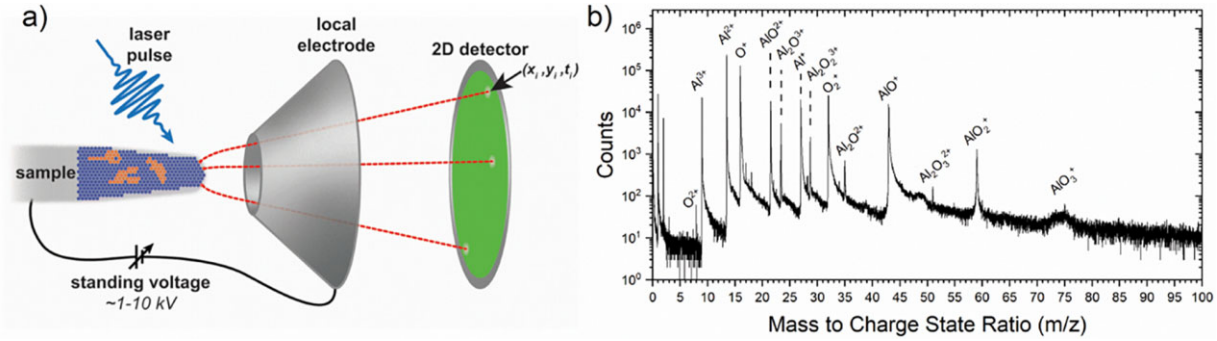

Fig. 1. a) Representation of the laser-assisted atom probe process leading to detection, adapted from [4] and b) a demonstrative mass spectrum from the evaporation of an α-Al_2O_3 tip using EUV pulses.

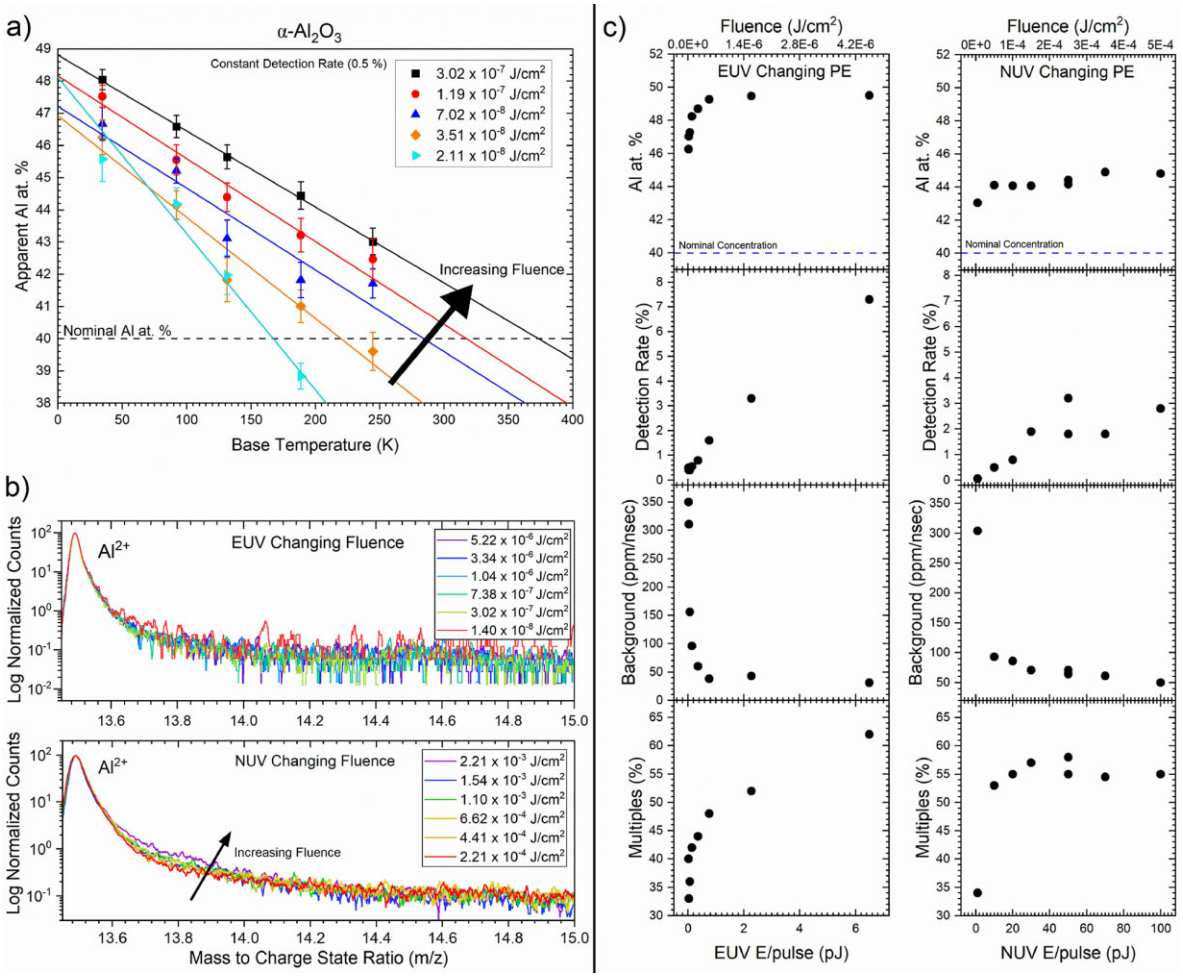

Fig. 2. Laser-pulsed APT results showing a) the change in apparent Al at. % composition with base temperature at five EUV fluence values with a best linear fit and 2σ composition error, b) the change in the Al^{2+} peak tails as a function of pulse energy for EUV (top) and NUV (bottom), and c) the change in apparent Al composition, detection rate, background counts, and multiples due to changes in EUV pulse energy (left) and NUV pulse energy (right) at a constant applied voltage.

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