

## Influence of Laser-Pulse Energy on Field Evaporation of LaAlO<sub>3</sub> in Atom Probe Tomography Analysis

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As followed by the design concept of metal-oxide-semiconductor (MOS) devices, the development of dielectric materials with high-k property becomes a crucial for reducing the equivalent oxide thickness. In order to develop the novel dielectric materials, understanding of correlations between electrical properties and chemical nature at metal-oxide interfaces has been an essential topic [1]. To identify this subject, atomic-scale investigation of bulk oxide materials through laser-pulsed atom probe tomography (APT) has been widely performed [2-7]. From the fundamental point of view, we present the influence of laser-pulse energy on APT results of lanthanum aluminum oxide (LaAlO<sub>3</sub>), termed LAO, especially mass-resolving power (MRP). The LAO is one of the potential candidates for substituting the conventional SiO<sub>2</sub>, which exhibit many advantages such as a high dielectric constant and large band-gap energy with ranging between 5.8 and 6.6 eV. Analysis were performed using a LAWATAP microscope in laser-pulsing mode of pulse energy varying with systematically 0.03~0.21 μJ at ~100 kHz pulse repetition rate, 0.002 atom/pulse detection rate, and 30 K.

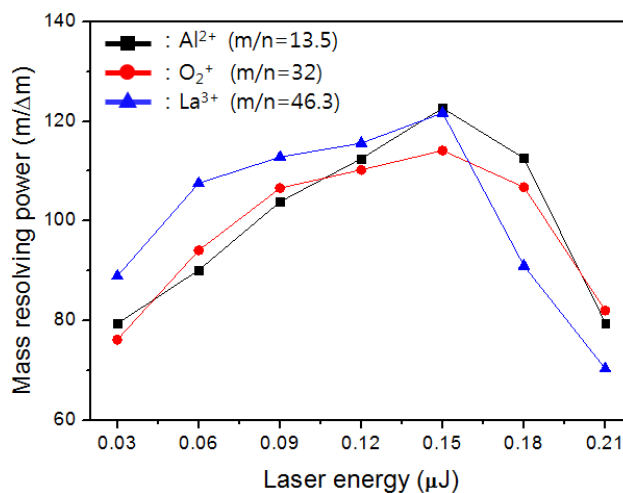
Fig. 1 shows the MRP of Al<sup>2+</sup>, O<sub>2</sub><sup>+</sup>, and La<sup>3+</sup> varying with laser-pulse energy, which is generally designated as  $m/\Delta m$  value at full-width half-maximum (FWHM). We found that the MRP values measured by APT of LAO increase from 80 up to 125, as the applied laser-pulse energy increases from 0.03μJ to 0.15μJ. Interestingly, the MRP is reduced when the laser-pulse energy of above 0.15μJ is applied. This is due to delayed field evaporation at the tip surface. It means that higher laser-pulse energy applied to the APT tips of LAO requires a longer cooling time at the tip surface during analysis, leading to delayed field evaporation of triggering the fragment ions at the tip surface under the residual elevated temperature. Indeed, the delay effects are detrimental for materials having low thermal diffusivity such as LAO (0.03~0.19 cm<sup>2</sup>/s).

Fig. 2 shows the 2-D distribution maps of time of flight for La<sup>3+</sup> ions as a function of the applied laser energy. The inhomogeneous mass to charge distribution ranging from 46.1 to 46.5 at laser pulse energy of above 0.15μJ corresponds to delayed field evaporation for ~4ns from incident laser side to the shadowed side. This reveals that uniform heating at the tip surface is important for improving MRP. In addition, silver capping onto needle-type specimens of APT tips enables an improvement in the uniform field evaporation at the surface as shown in Fig. 2(h). The distribution map obtained from laser energy of 0.21μJ in conjunction with silver capped LAO demonstrates that a prospective approach through silver capping deposited onto APT tips of oxides is beneficial for obtaining better improvement of MRP during laser-assisted APT analysis of oxides. [8,9]

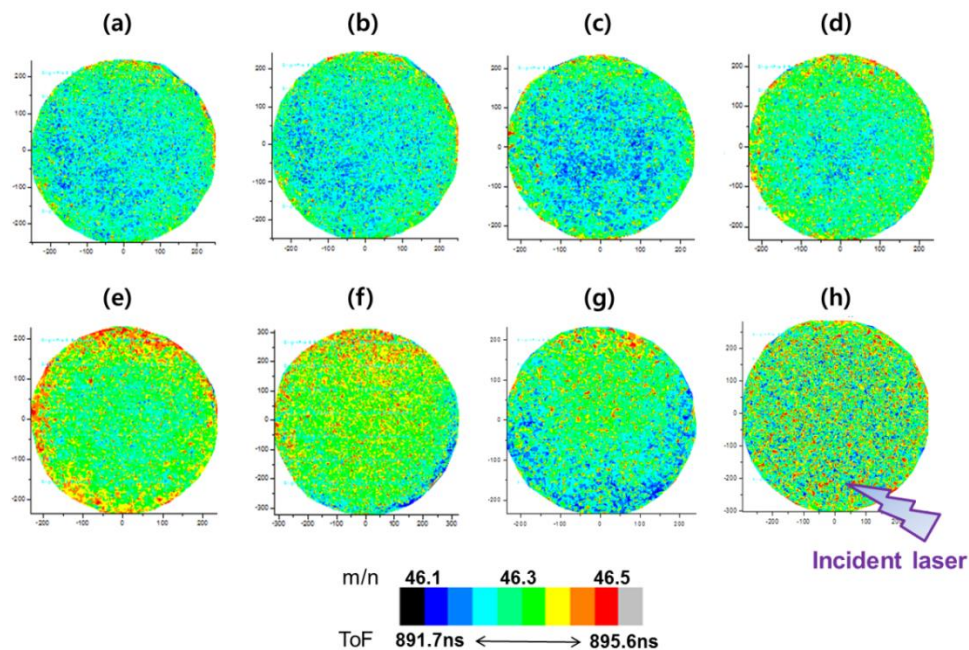
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**Figure 1.** Mass resolving power ( $m/\Delta m$ ) of  $\text{Al}^{2+}$  (black),  $\text{O}_2^+$  (red),  $\text{La}^{3+}$  (blue) as a function of laser pulse energy at specimen base temperature of 30K.



**Figure 2.** Mass to charge ( $m/n$ ) distribution maps ranging from 46.1 to 46.5 for  $\text{La}^{3+}$  ions as a function of the applied laser-pulse energy; (a)  $0.03\mu\text{J}$ , (b)  $0.06\mu\text{J}$ , (c)  $0.09\mu\text{J}$ , (d)  $0.12\mu\text{J}$ , (e)  $0.15\mu\text{J}$ , (f)  $0.18\mu\text{J}$ , (g)  $0.21\mu\text{J}$ , and (h)  $0.21\mu\text{J}$  in conjunction with Ag capping deposited onto APT tips of LAO.