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In-situ observation of phase transformation during ion beam irradiation

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In-situ observation

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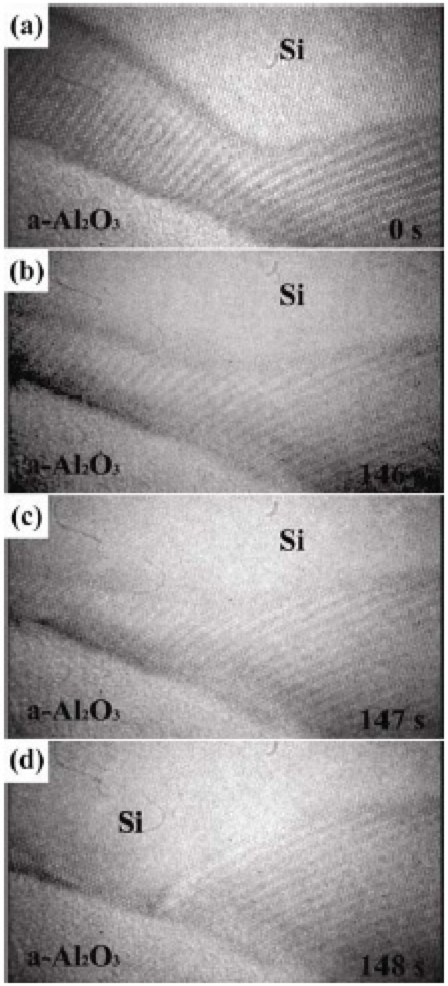
What is In-situ observation ?

1. Real-time data acquisition
   - providing a “live” image or graph

2. Keeping the exact same position during observation

3. Saving time and effort



* J.Won, Doctoral dissertation

In-situ observation during ion beam irradiation (XRD)

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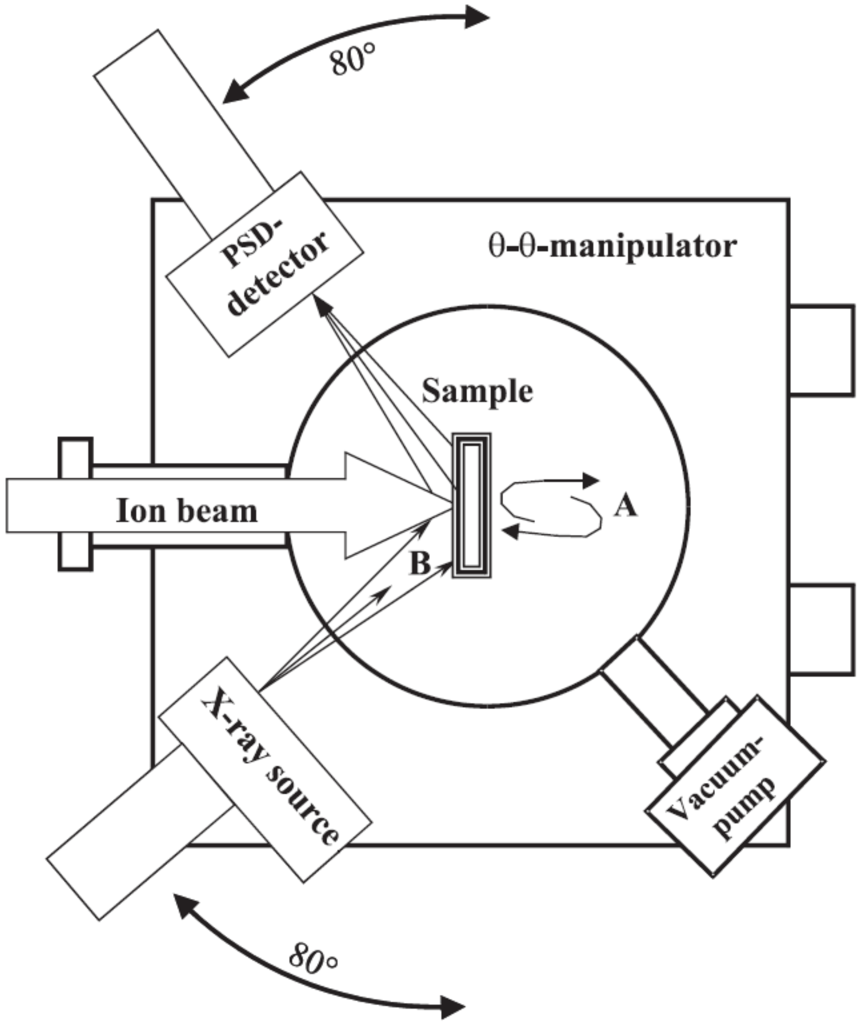
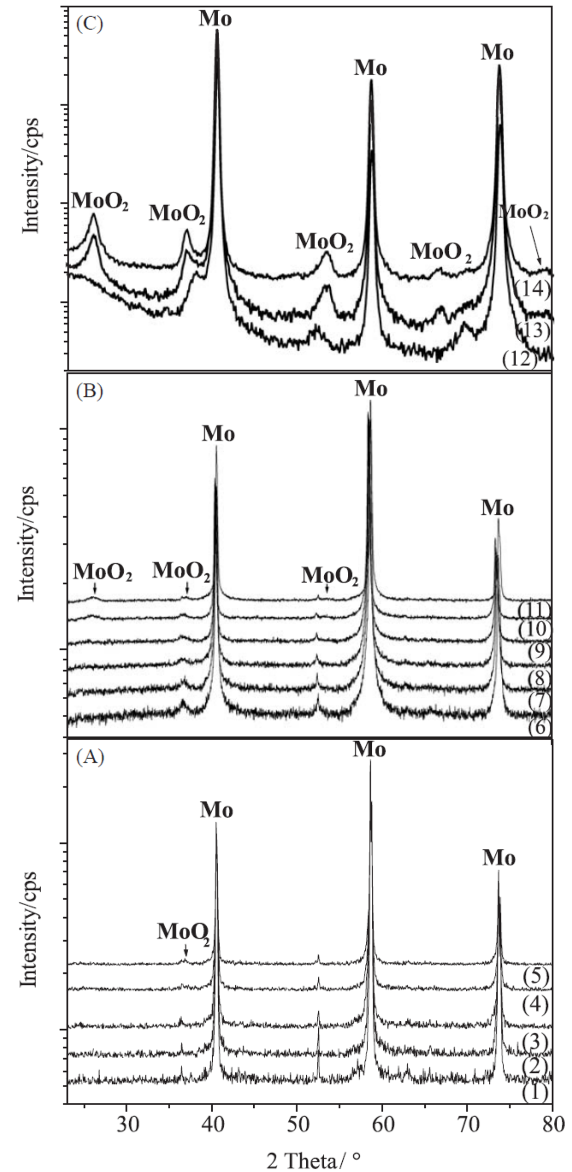
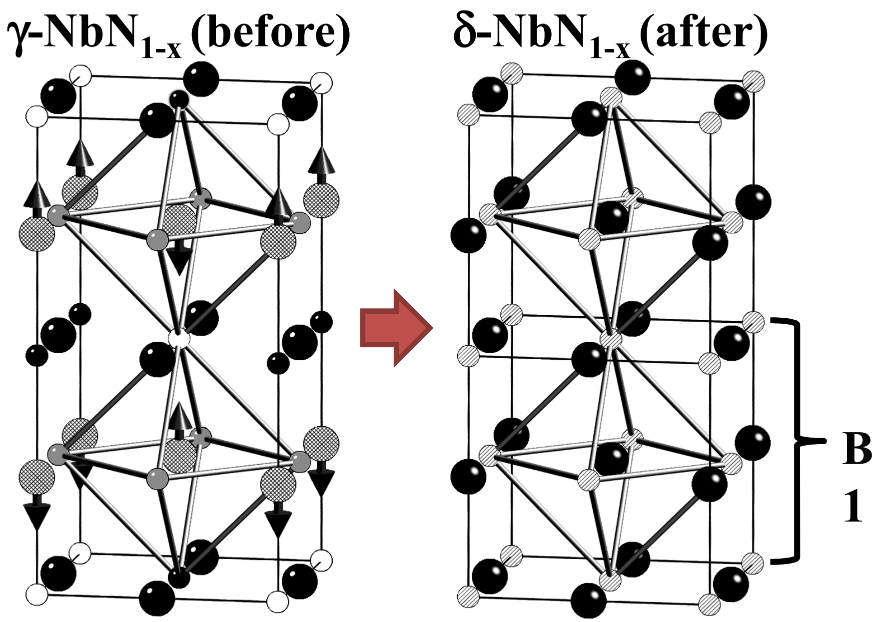
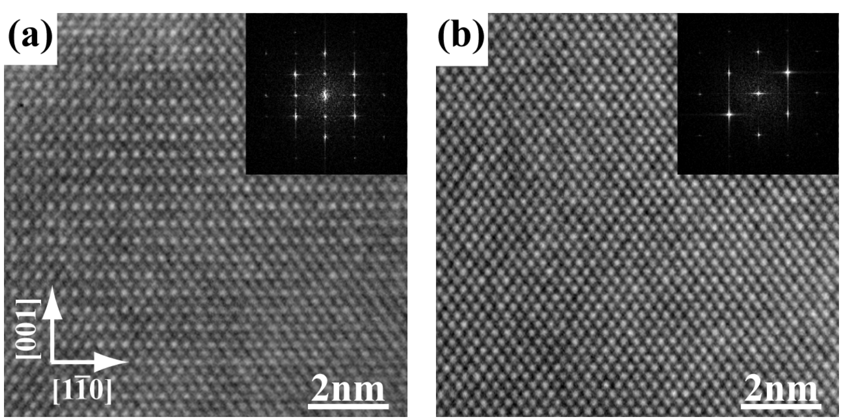


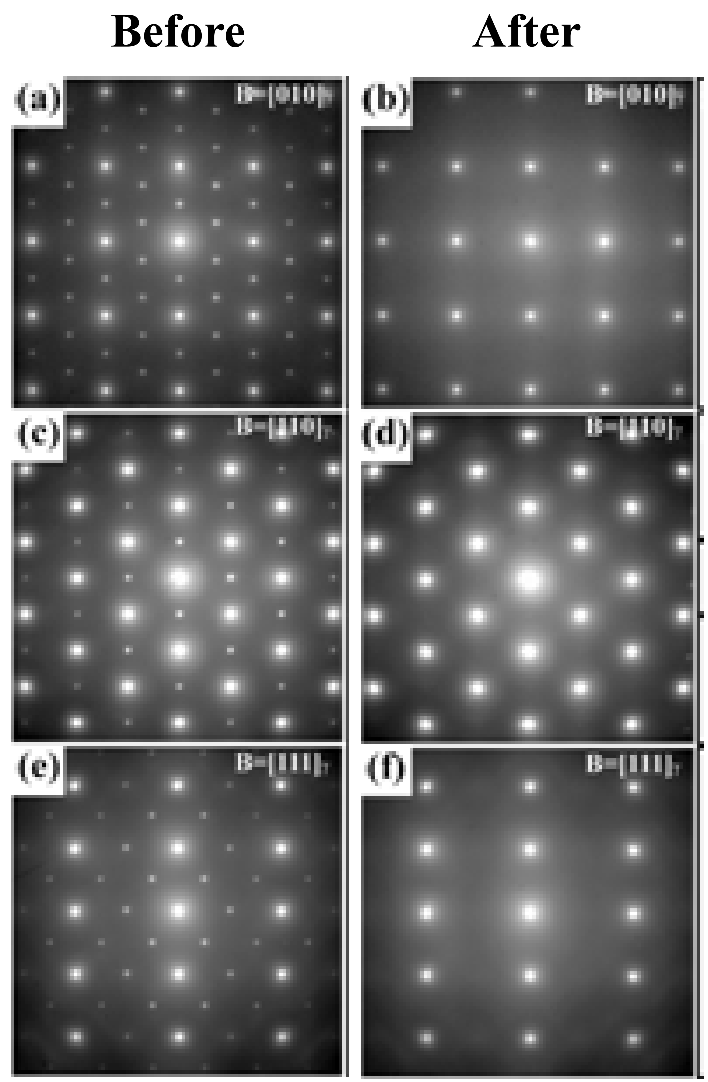


* Y. Bohne et al. Vacuum 76 (2004)

In-situ TEM observation during e⁻ beam irradiation







*J.Won et al., Scripta Materialia 60 (2009)

Reflection high-energy electron diffraction (RHEED)

- A high-energy electron beam (3-100 keV) is directed at the sample surface at a grazing angle of incidence
- It is possible to use both kinematic (elastic) and dynamic (inelastic) scattering

Advantages

► High surface sensitivity

- ability to measure very few atomic layers beneath the surface

► In-situ observation

- phosphor screen with charge-coupled device (CCD) camera
- real time sequential images and movie can be generated

	XRD	TEM	RHEED
Observation area	large	very small	large
Observation depth	deep	>100 nm	better than TEM
Instrument cost	high	high	Low
nondestructive	yes	no	yes

Experimental procedure

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- **Specimen** : $LaAlO_3$ single crystal (MTI corporation)
  - Rhombohedral at 25 ^oC and Cubic at > 435 ^oC, (100) surface normal
- **Ion implanter**: 200 kV Varian Implanter, 100 keV Ar^{++} source
- **RHEED**: Guns system from STAIB Instruments (up to 60 keV)

  Imaging system (kSA 400 analytical RHEED system)

  from k-Space associates, Inc.
- **Characterization**
  - GIXRD: Bruker AXS D8 Advance X-ray diffractometer

    (Cu-K_α and θ–2θ geometry)
  - TEM : Sampling-combination of mechanical grinding and ion milling (PIPS)

    Observation- FEI Technai F30 TEM/STEM

System configuration

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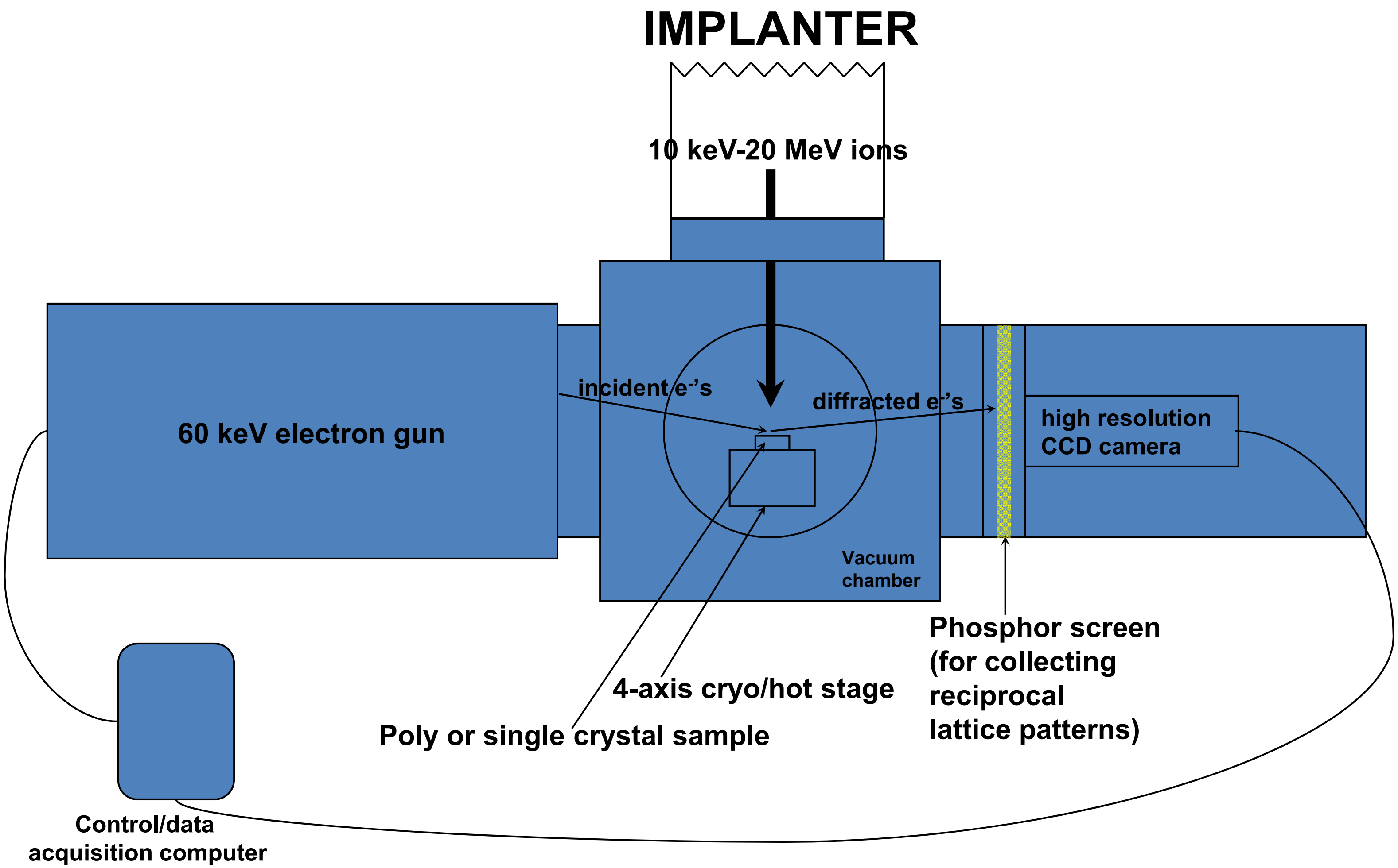

System configuration

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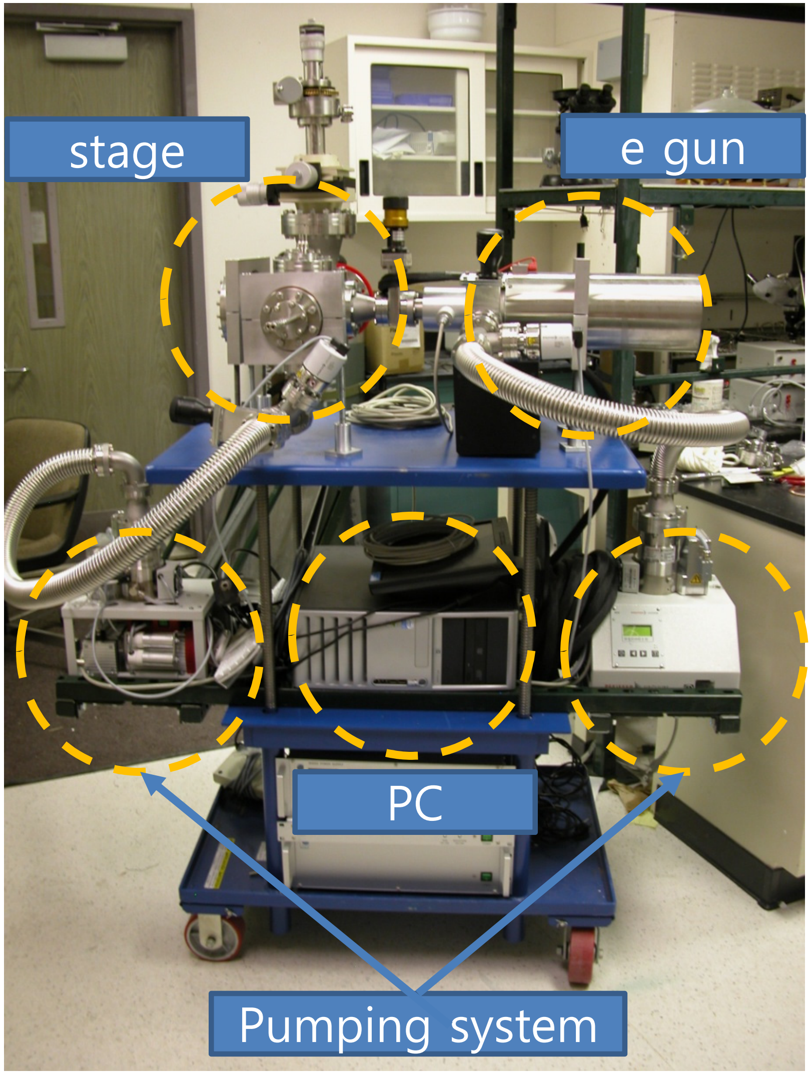
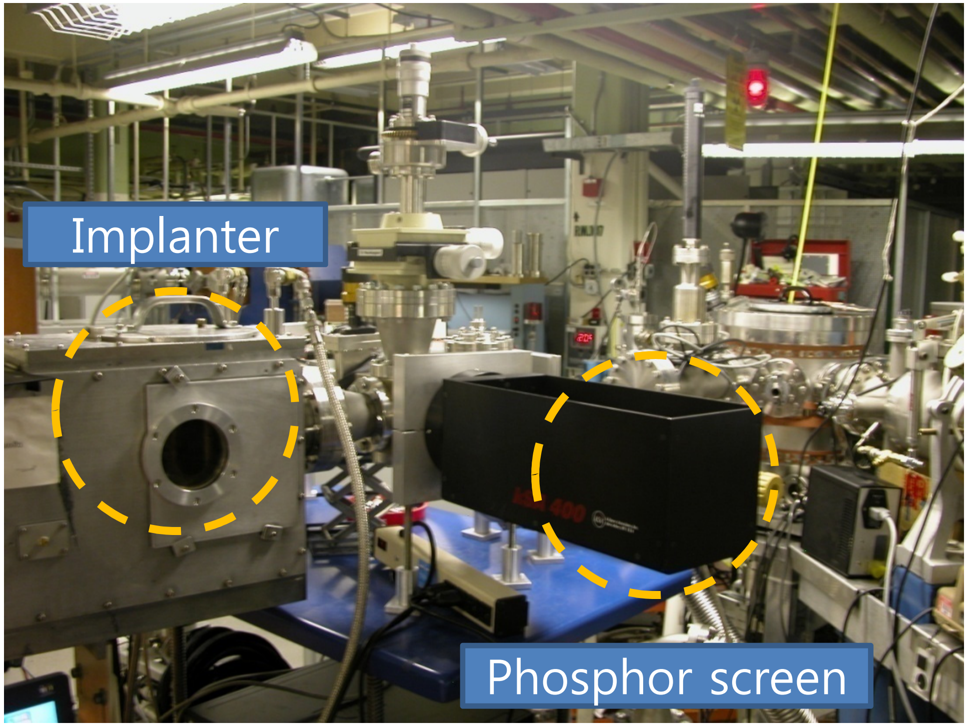


1. on implanter
2. RHEED
   - electron gun
   - sample stage
   - phosphor screen and CCD camera
   - control/data acquisition PC



Ion penetration depth calculated by SRIM2009 and GIXRD patterns from $LaAlO_3$

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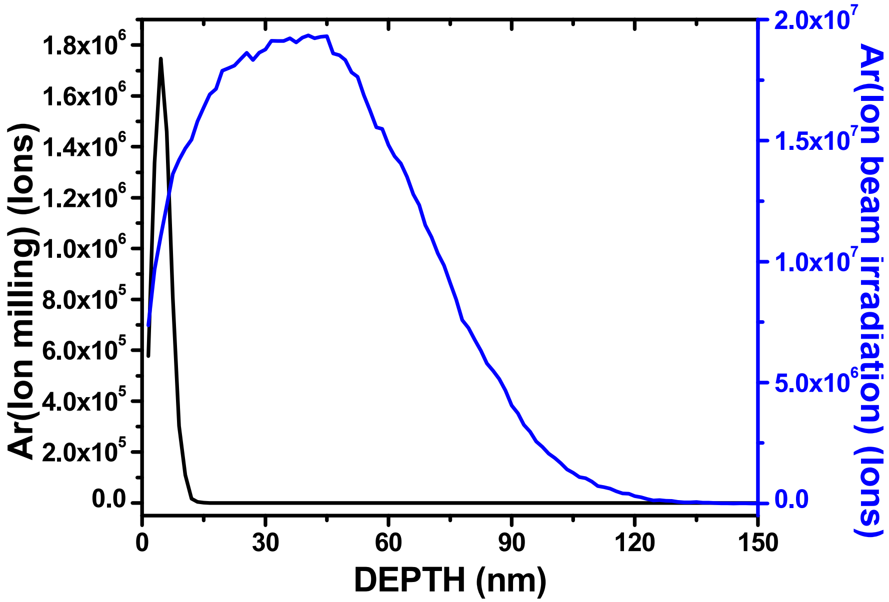
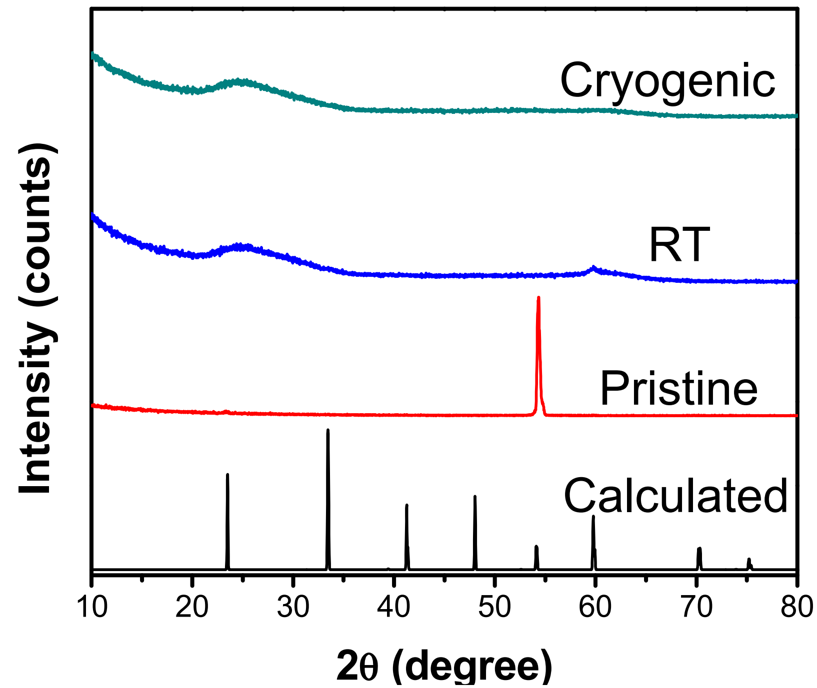
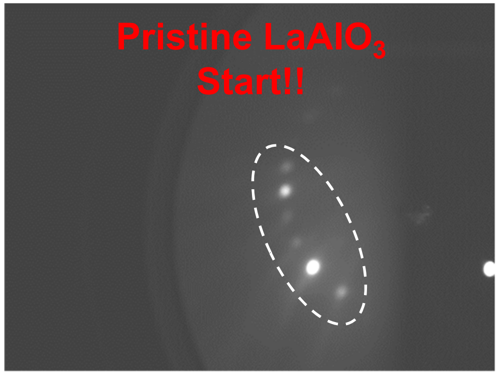




- Monte Carlo simulation show that the penetration depth is ~ 120 nm from the surface
- Calculated X-ray penetration depth is < 10 nm at a grazing angle of 0.25°
- After Ar irradiation, the $LaAlO_3$ single crystal phase near the surface is transformed into an amorphous phase

In-situ RHEED sequential images for 100keV Ar^{2+}ions irradiation of c-$LaAlO_3$ at RT



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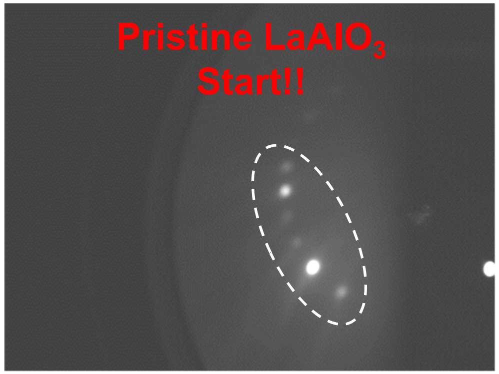

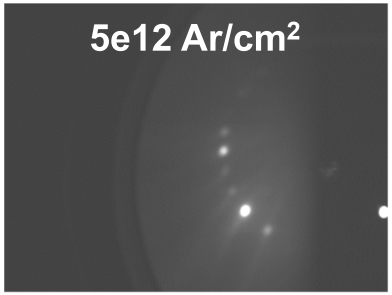

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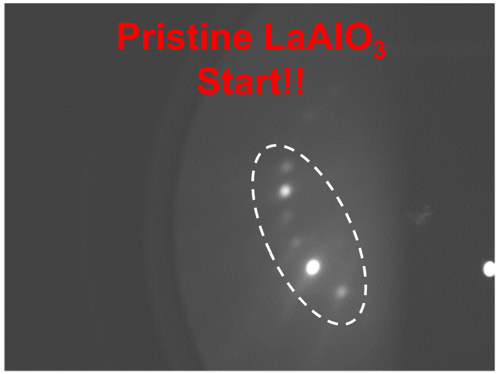

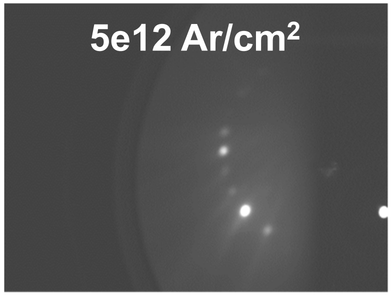

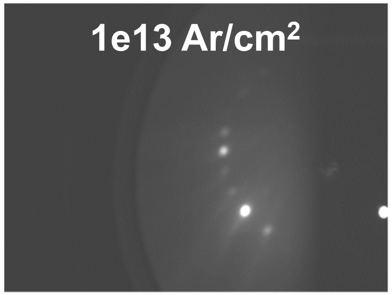

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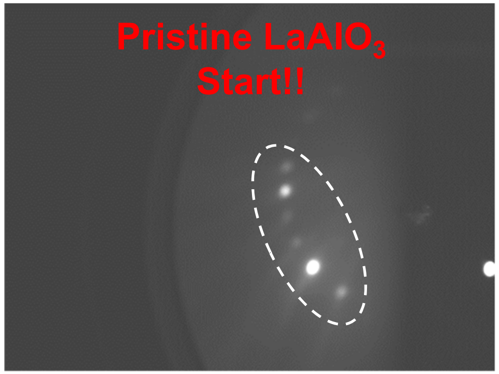

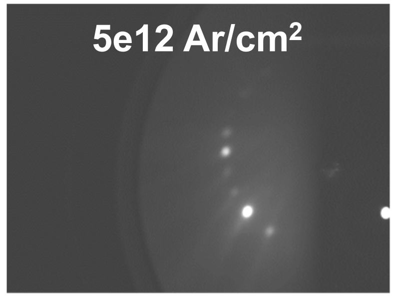

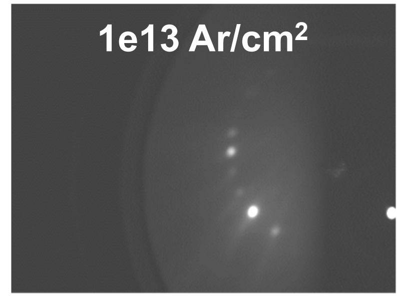

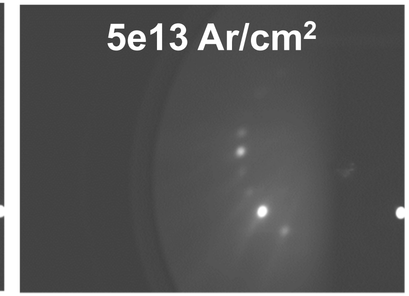

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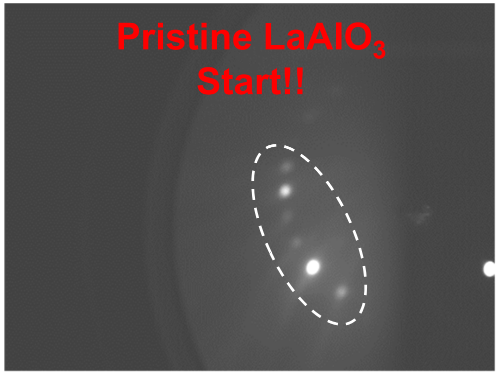

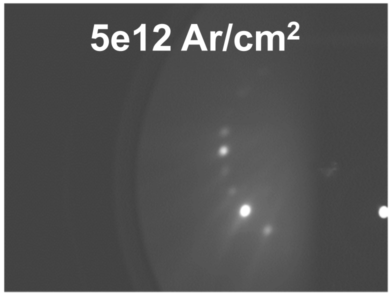

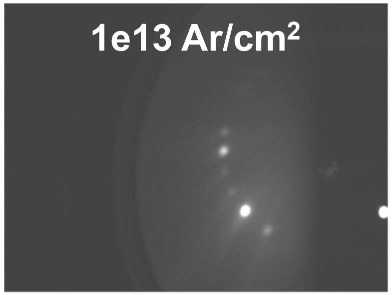

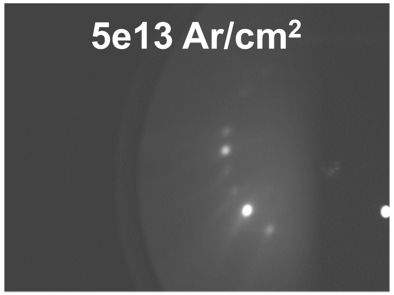

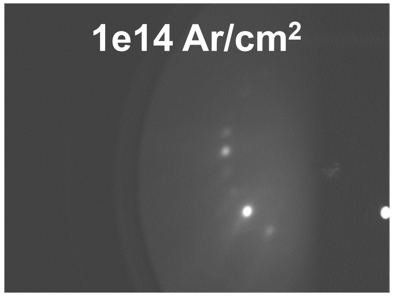

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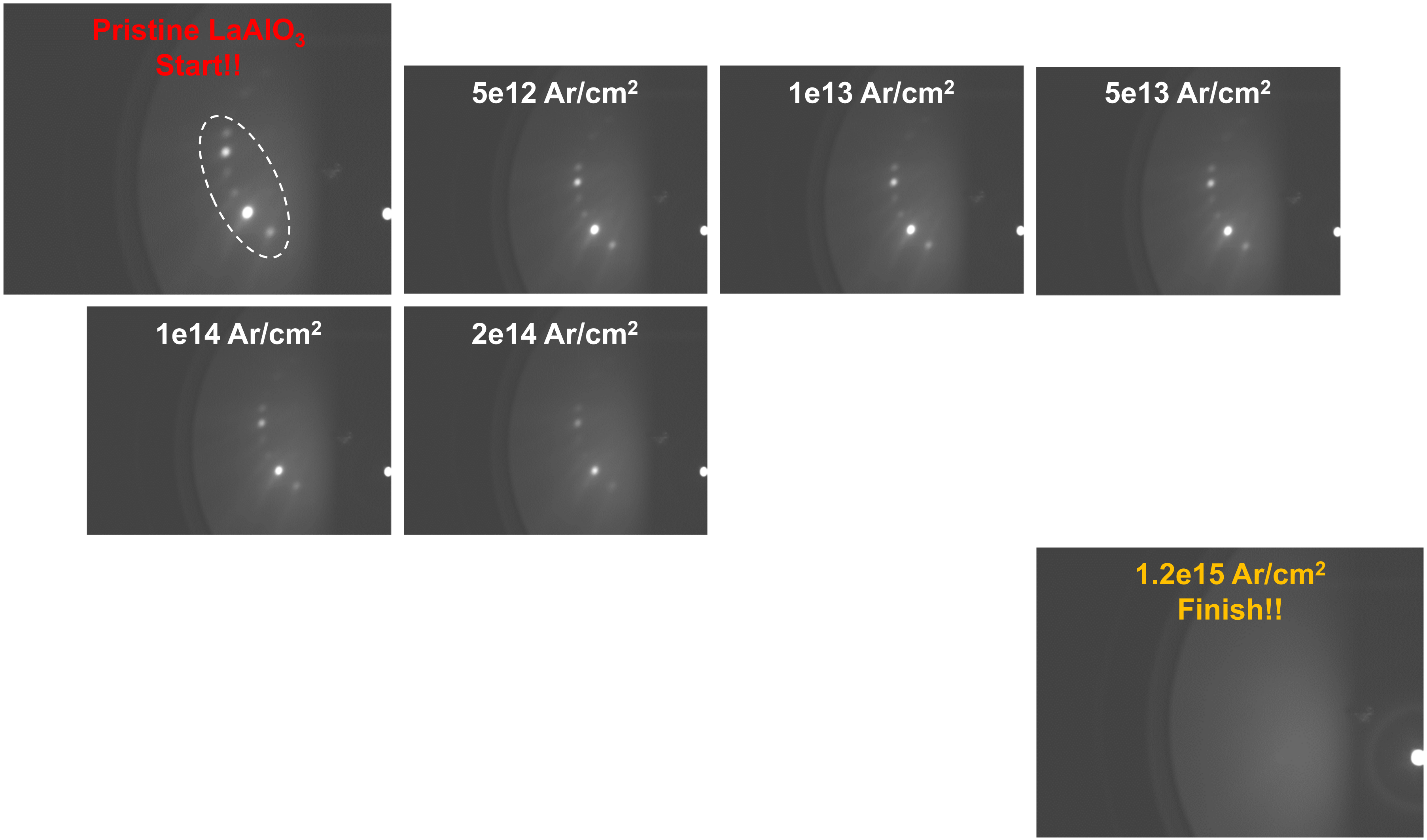

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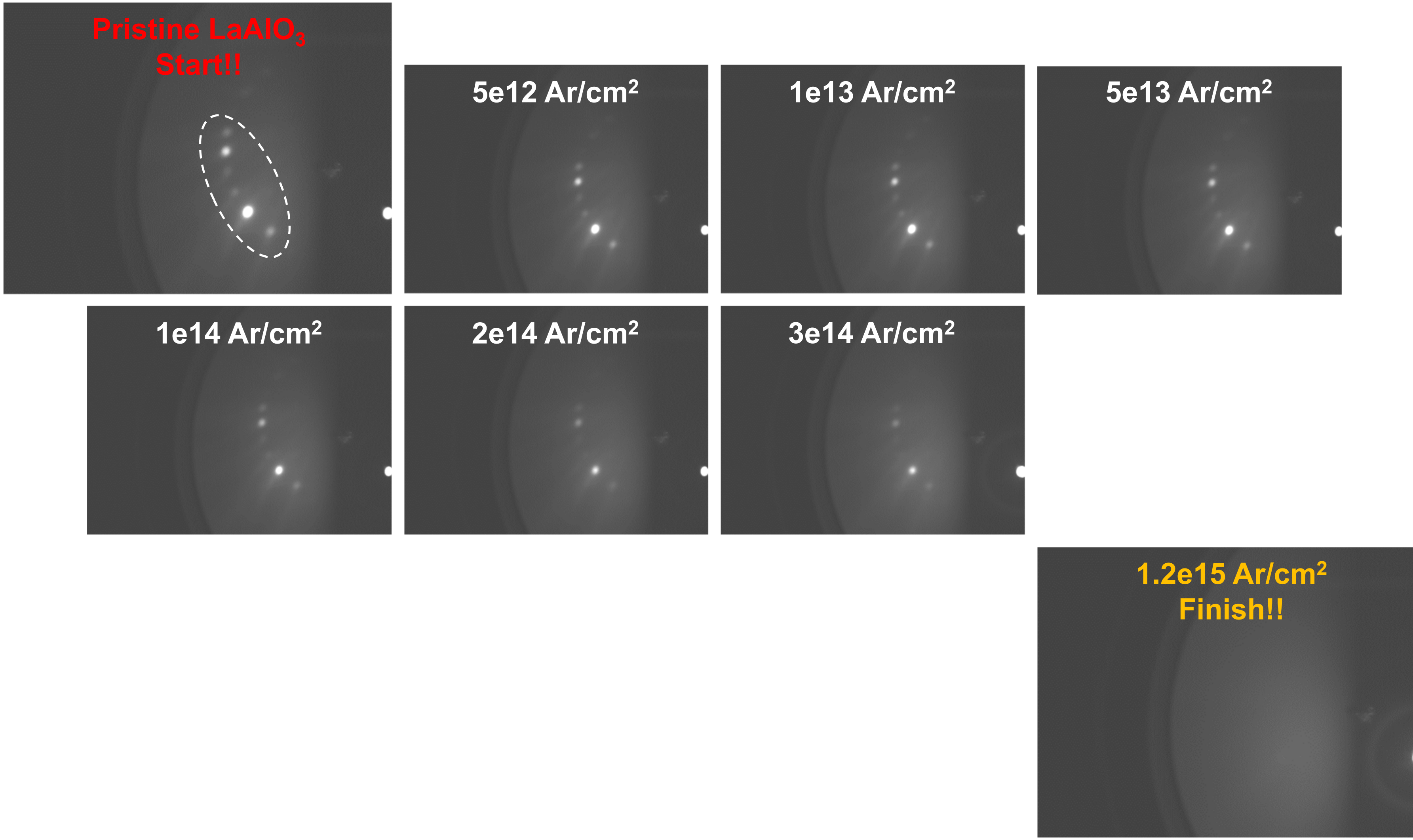

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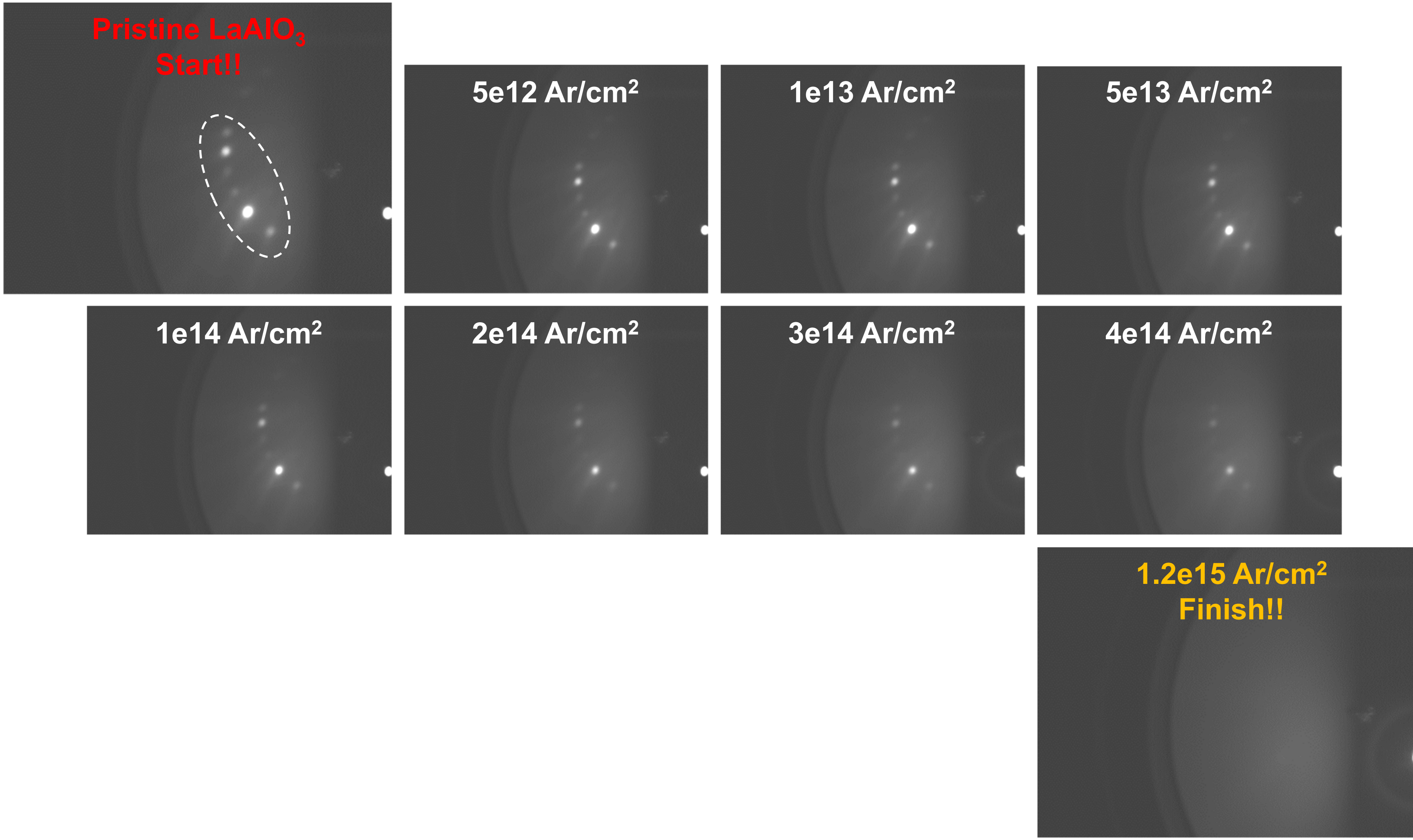

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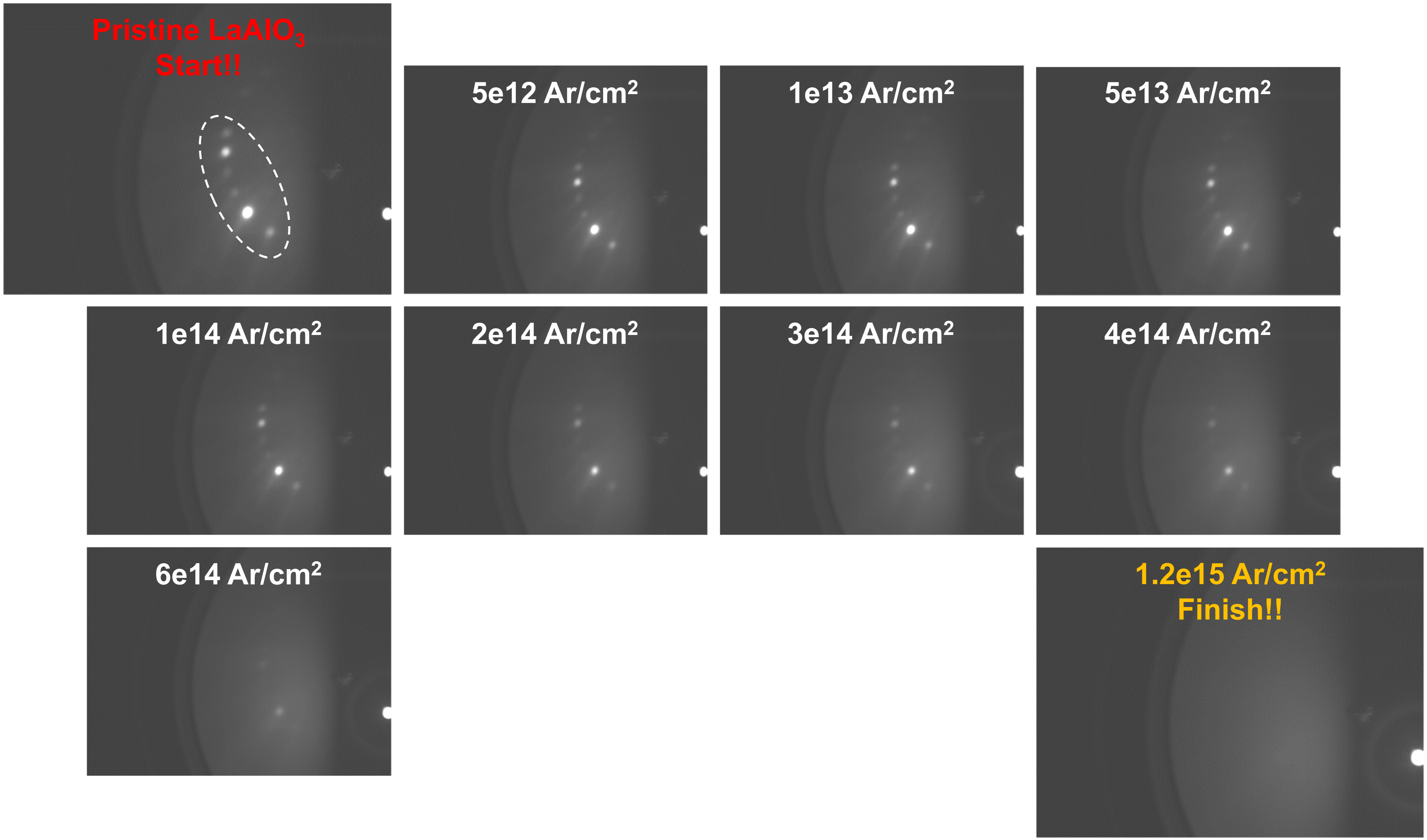

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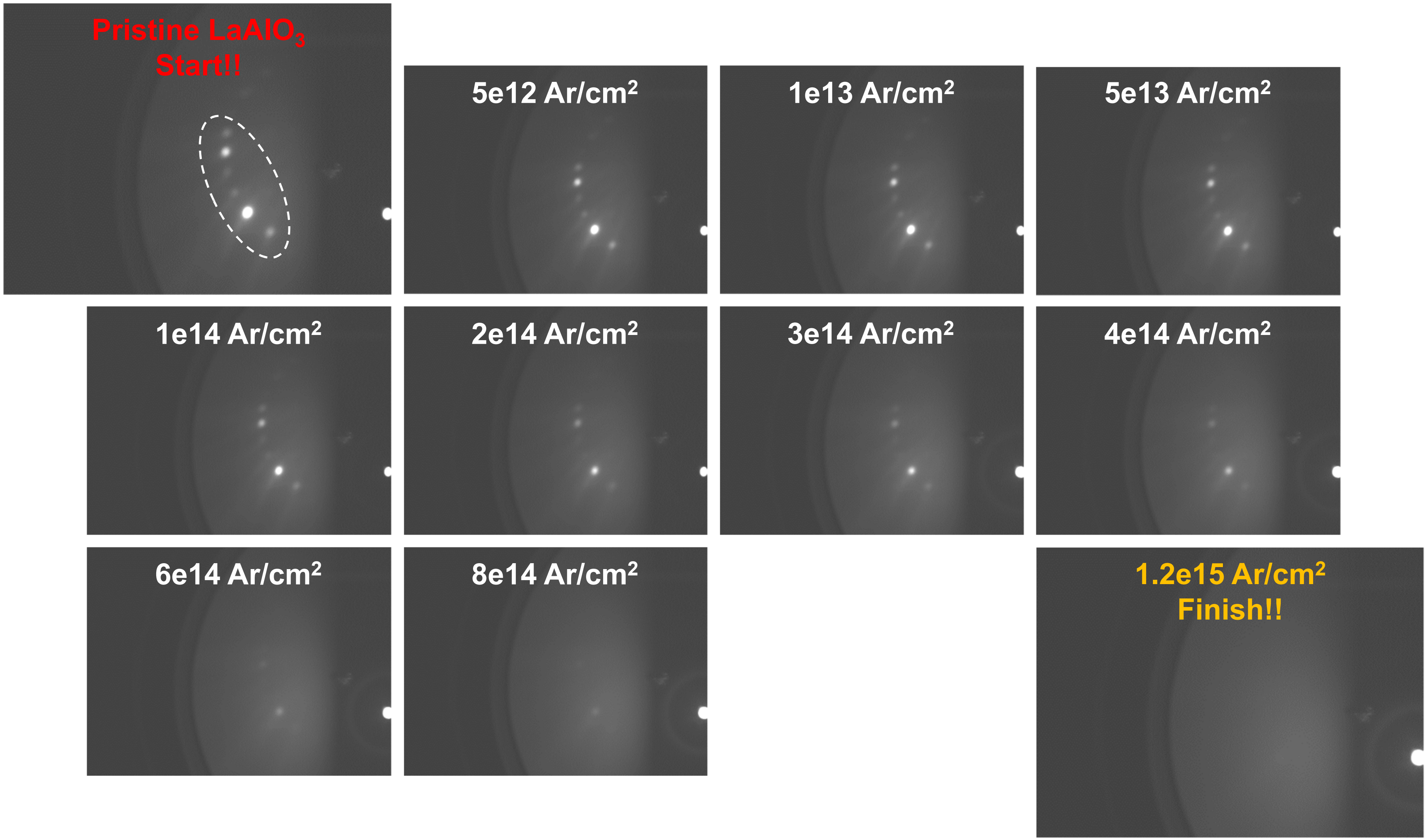

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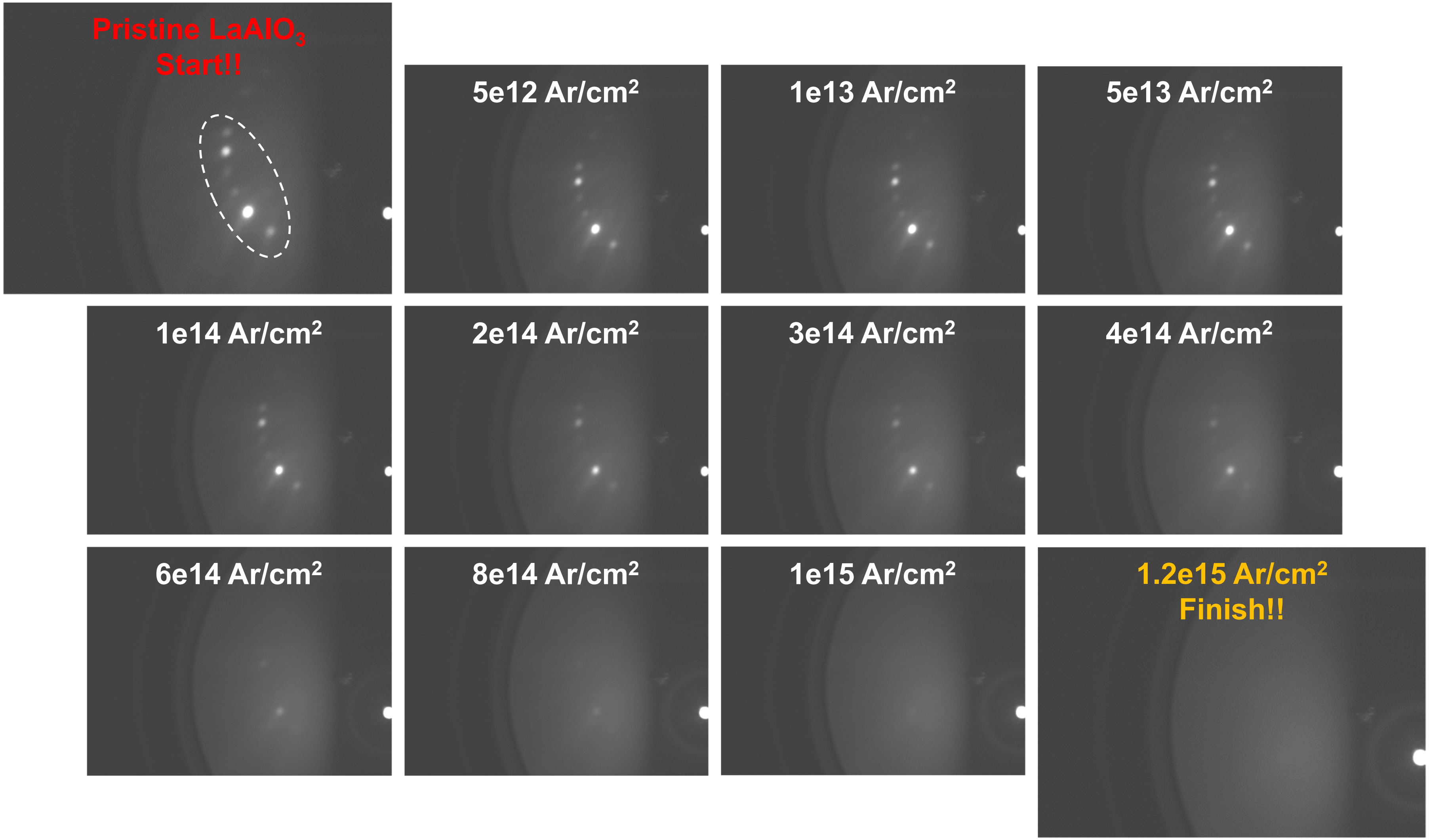

Readjusted e^- beam incident angle probe

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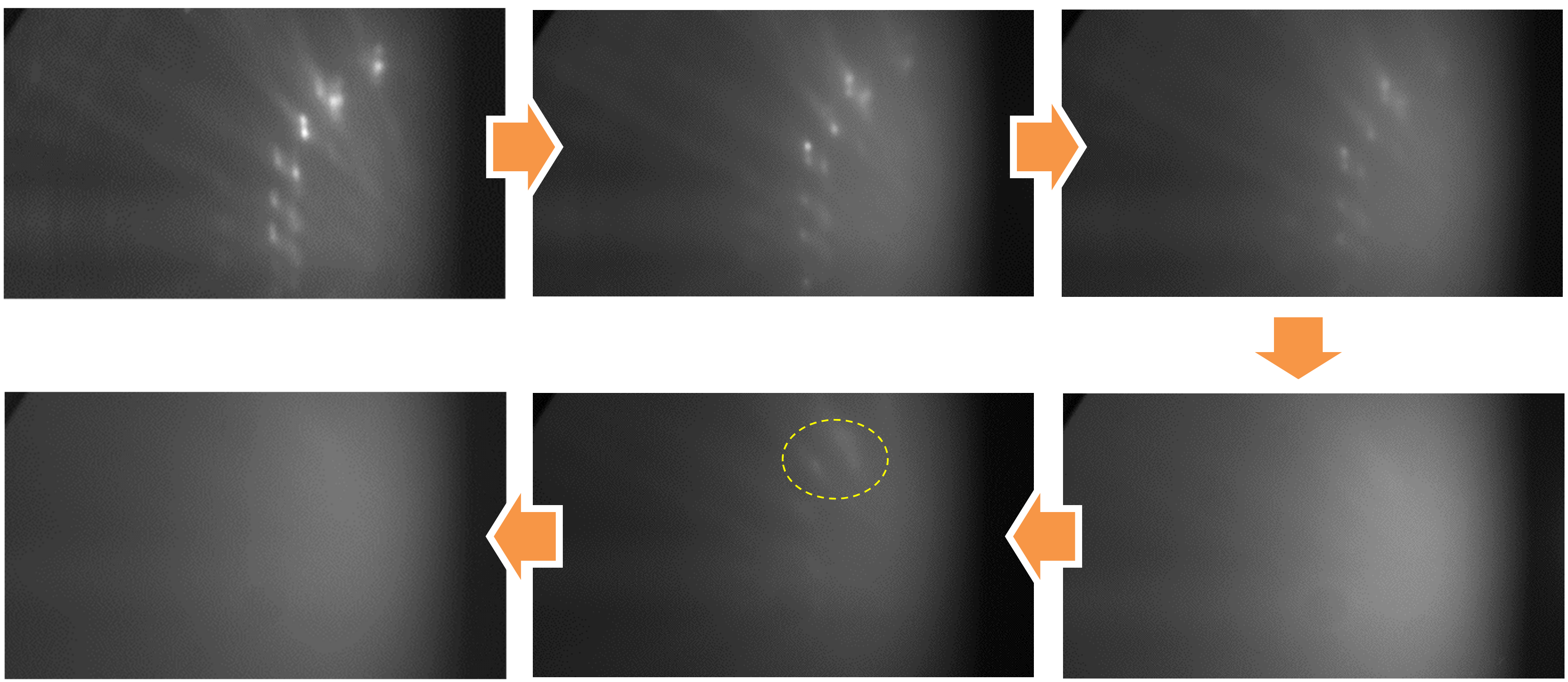
Pristine- $LaAlO_3$ | **5e13 Ar/cm^2** | **2.38e14 Ar/cm^2**

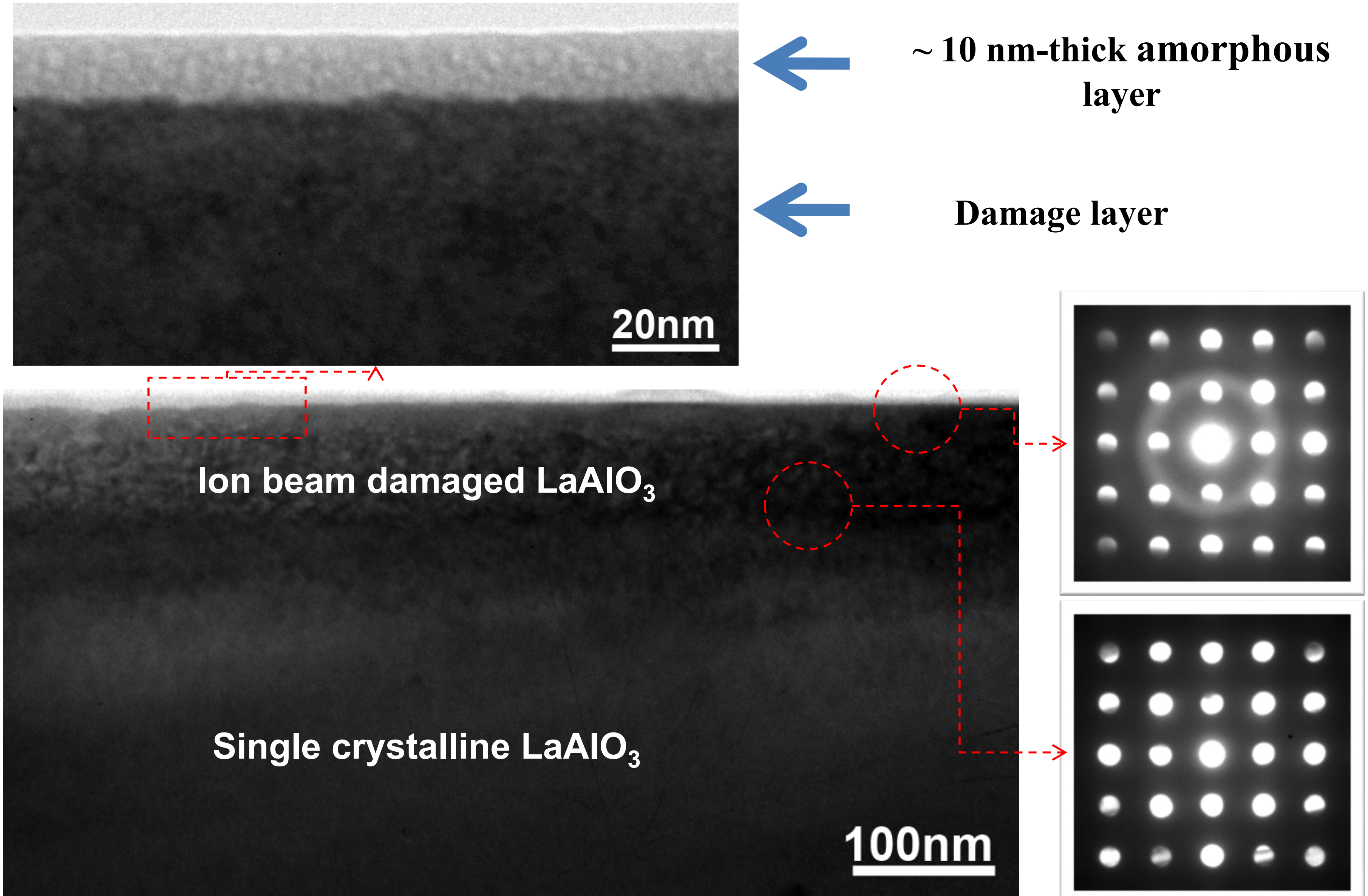


6.5e14 Ar/cm^2

Readjusted e^- incidence angle probe deeper into sample 3.5e14 Ar/cm^2

3.5e14 Ar/cm^2

TEM and MBD analysis of 100kev Ar^{2+} ions irradiated sc-$LaAlO_3$ at RT



MDP analyses of ion beam induced a-$LaAlO_3$

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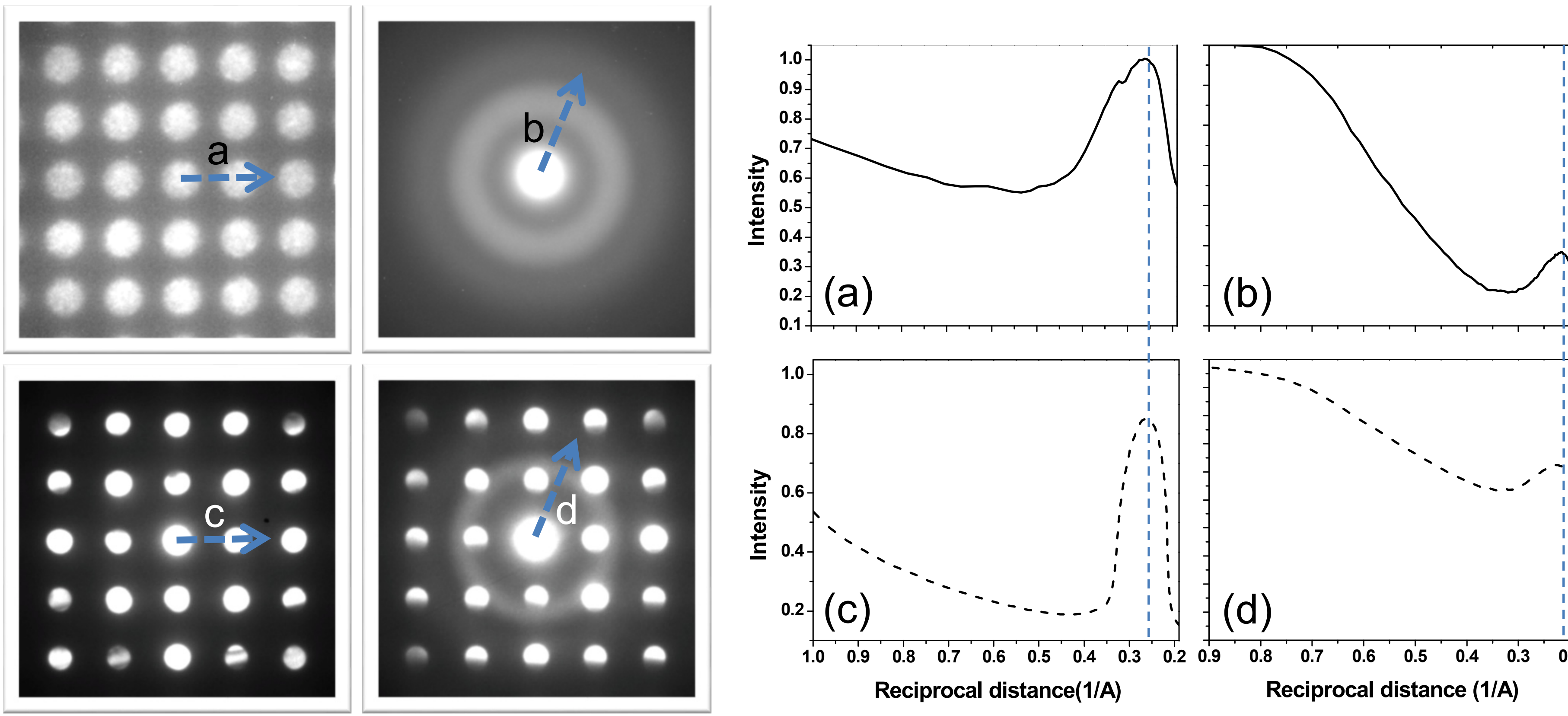
400 kV Xe irradiation

Spot spacing

Hallo spacing



100kev Ar irradiation

Calculated reciprocal lattice distances from Xe (upper) and Ar (lower) are identical → Crystalline (c) $LaAlO_3$ is transformed to an amorphous (a) phase by ion irradiation

In-situ diffraction spot intensity evolution of $LaAlO_3$ at cryo temp.

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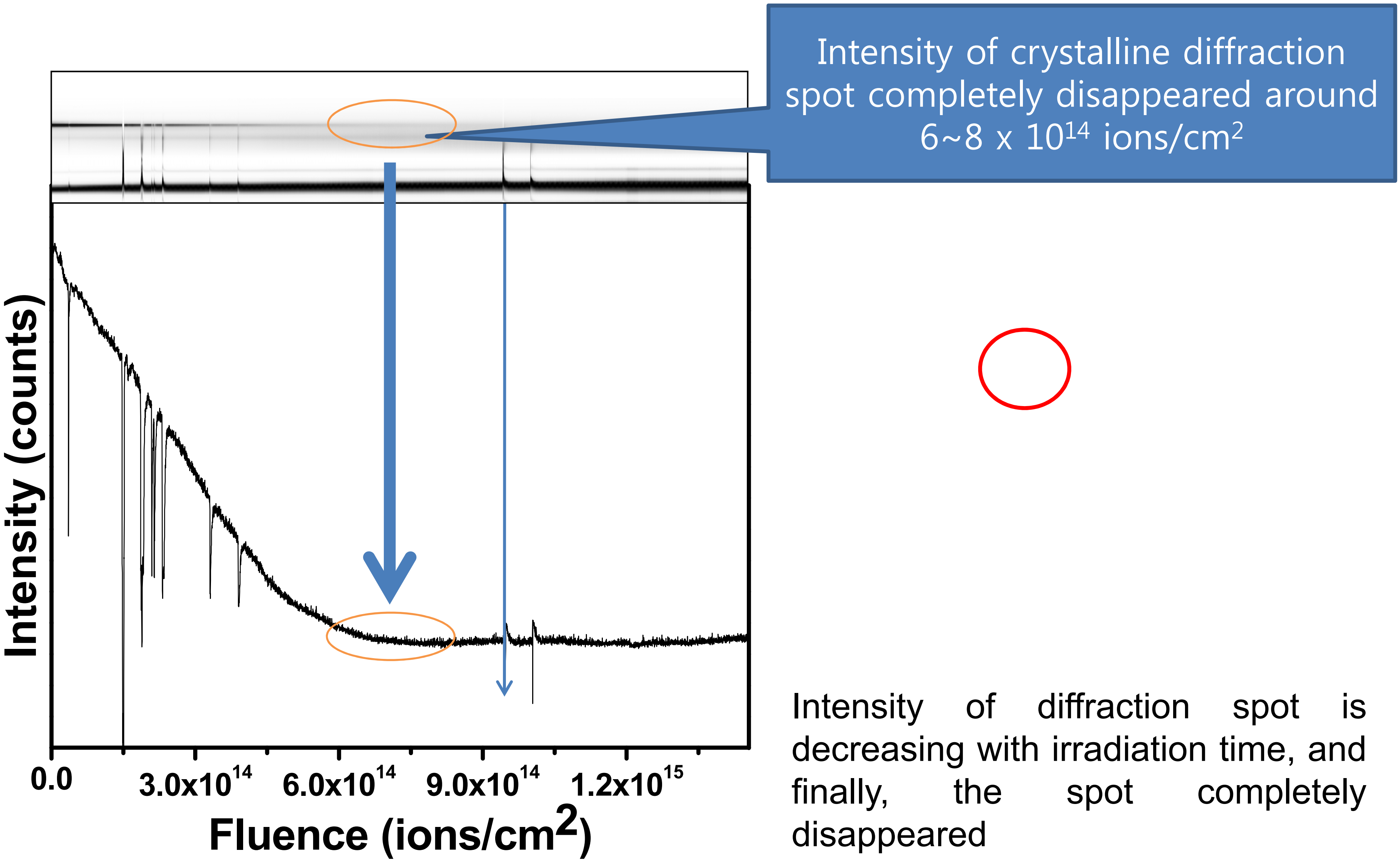

Intensity of diffraction spot is decreasing with irradiation time, and finally, the spot completely disappeared

In-situ diffraction spot intensity evolution of $LaAlO_3$ at cryo temp.

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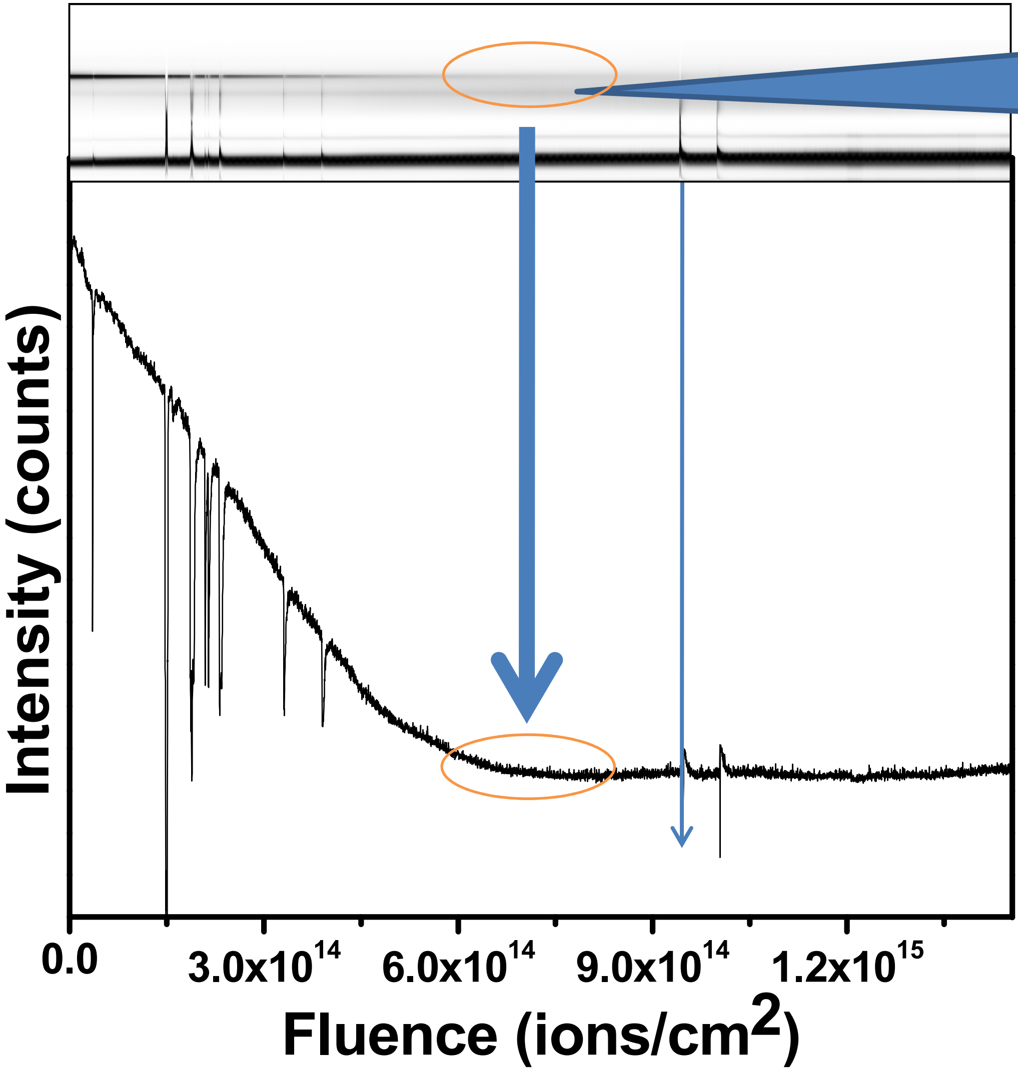
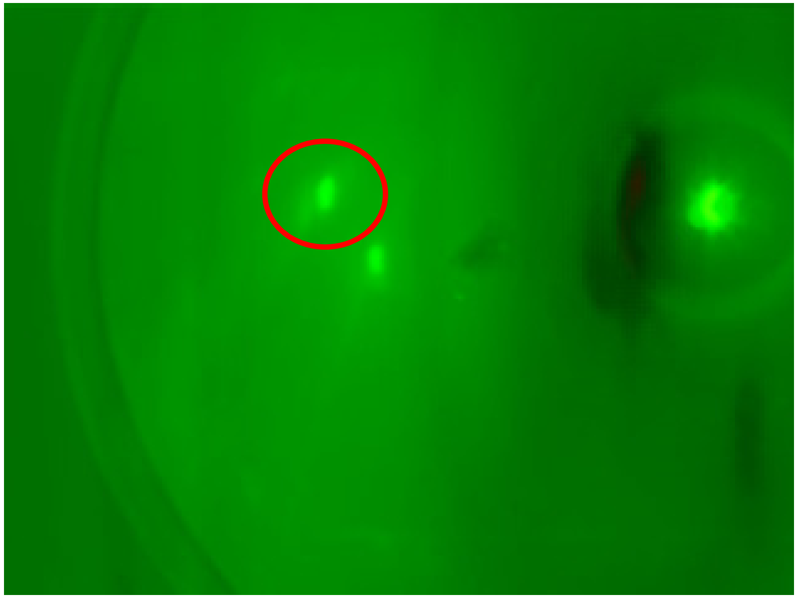
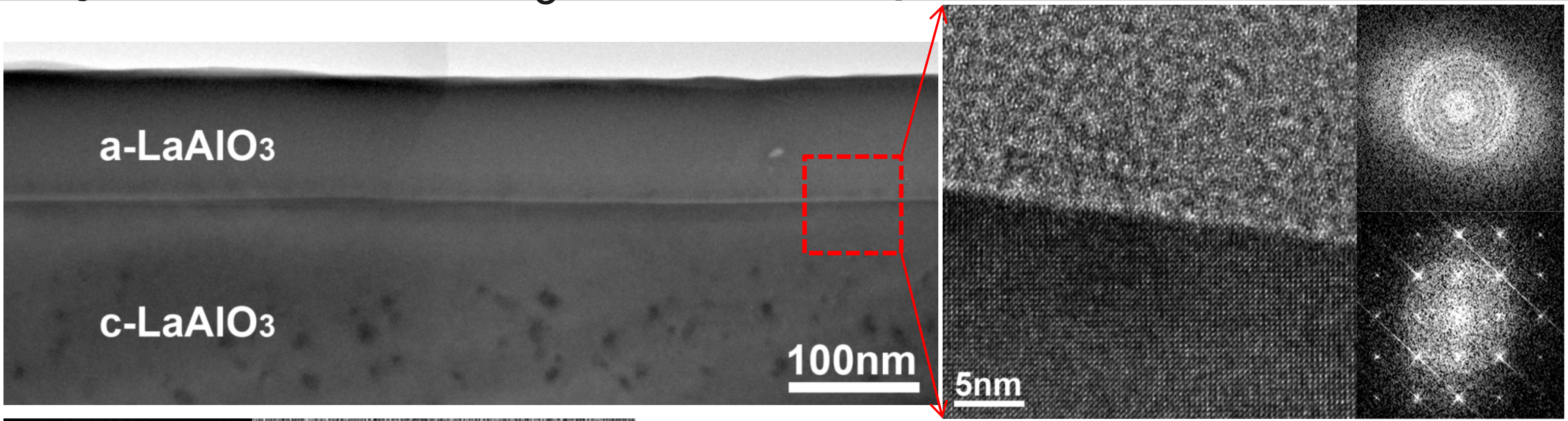
Intensity of crystalline diffraction spot completely disappeared around 6~8 x 10^{14} ions/cm^2

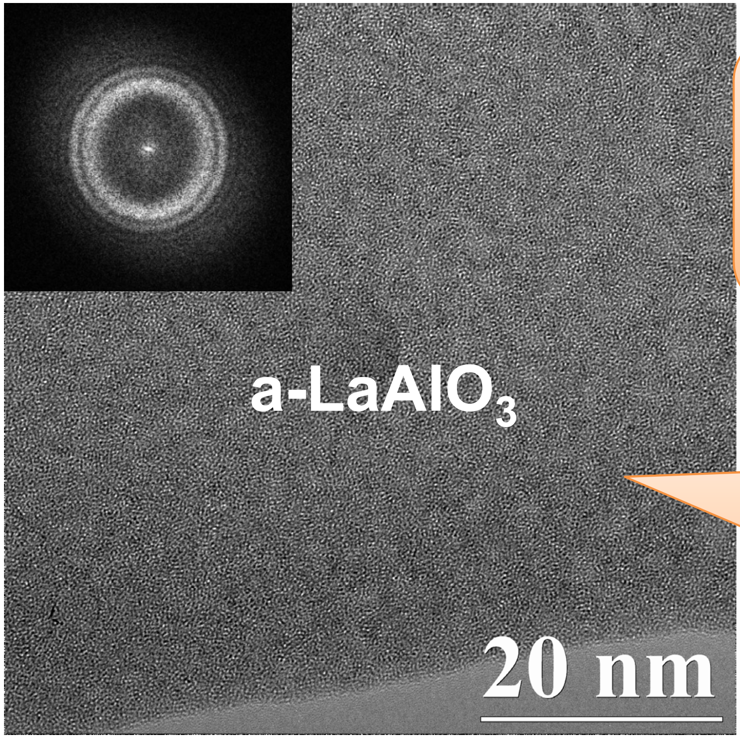




Intensity of diffraction spot is decreasing with irradiation time, and finally, the spot completely disappeared

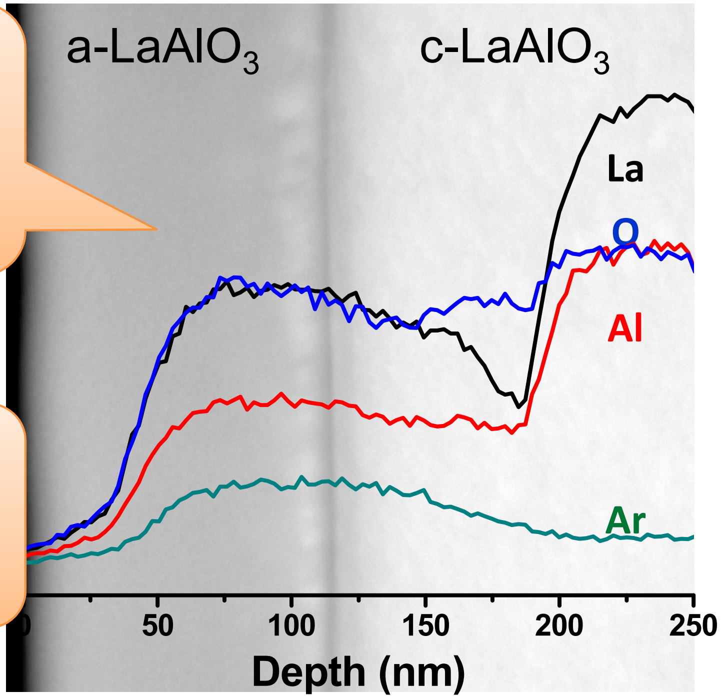
TEM/STEM-EDS observations for crystalline $LaAlO_3$ at low temp.





HAADF image with STEM-EDS

High resolution TEM image of $a\text{-}LaAlO_3$



Summary

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1. Crystalline-to-amorphous (c-a) phase transformation of $LaAlO_3$ was investigated by **combining 100 keV Ar^{++} ion irradiation with in-situ RHEED system**.

   - **RT**: around 100 nm-thick amorphous layer formed by irradiation

   - **Cryogenic temp.**: more than **100 nm-thick** amorphous layer was formed by irradiation (~ identical to damage depth from SRIM2009 calculation)

2. Measured c-to-a phase transformation time (fluence) is dependent on the **incident angle of the electron-beam**. In the case of an incident angle of 3^{o} at cryogenic temp., in-situ RHEED observations show that the phase transformation fluence is around **$6\sim8 \times 10^{14}$ ions/cm^2**

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Thank You

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