Novel Synthesis and Characterization of Indium-free Transparent Conductor

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Chemical Engineering and Materials Science
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Grant No. DMR-1420013

Upper MidWest SID Chapter Webinar
April 5th, 2018

AFOSR YIP

Transparent Conductors

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Key Component for many technologies:

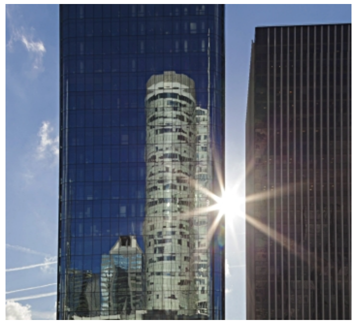
Light ⟷ Electricity

LED
10~30 Ω/☐

Smart glass/windows

Resistive touch panels
300~750 Ω/☐

TVs: 10~50 Ω/sq



Low e-windows

Capacitive touch panels 70~200 Ω/☐

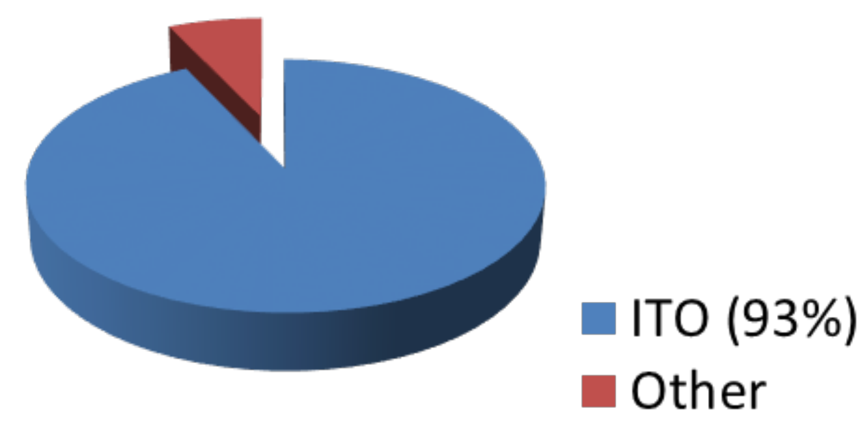
Transparent Conductors

'TCO' cations: post-transition elements:
Cd^{2+}, Zn^{2+}, In^{3+} , Ga^{3+}, Sn^{4+}

IA	IIA	IIIB	IVB	VB	VIB	VIIB	VIIIB			IB	IIB	IIIA	IVA	VA	VIA	VIIA	VIIIA
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac-Lr															

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Market Share
transparent conductors



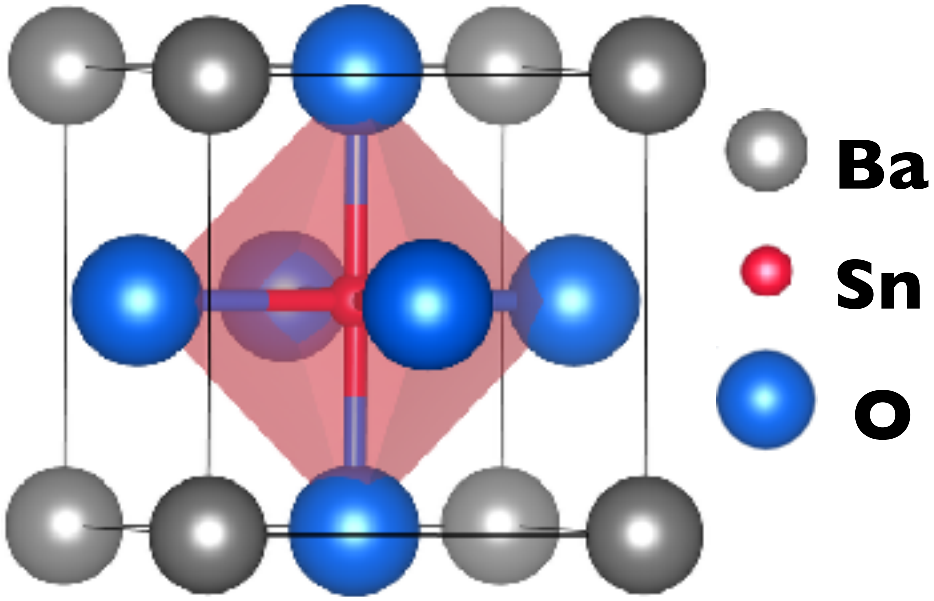
Indium is expensive !

In: $754/kg (Ag: $511/kg)

2015 United States Geological Survey

Key requirements: Wide Band gap, and high conductivity
Additionally, low cost, and low temperature synthesis are needed

Why $BaSnO_3$?



- ☑ Simple cubic (Pm-3m), a = 4.116 Å

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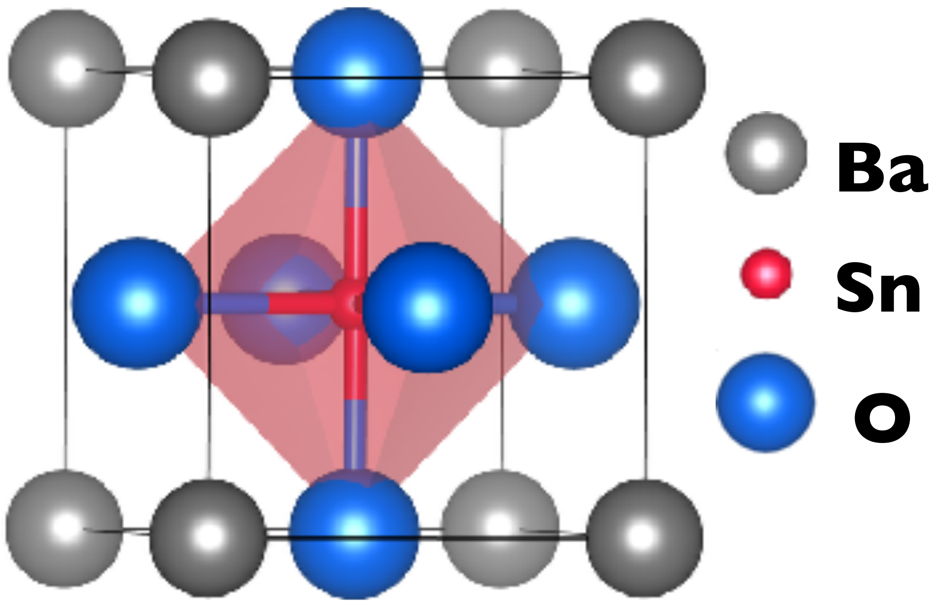

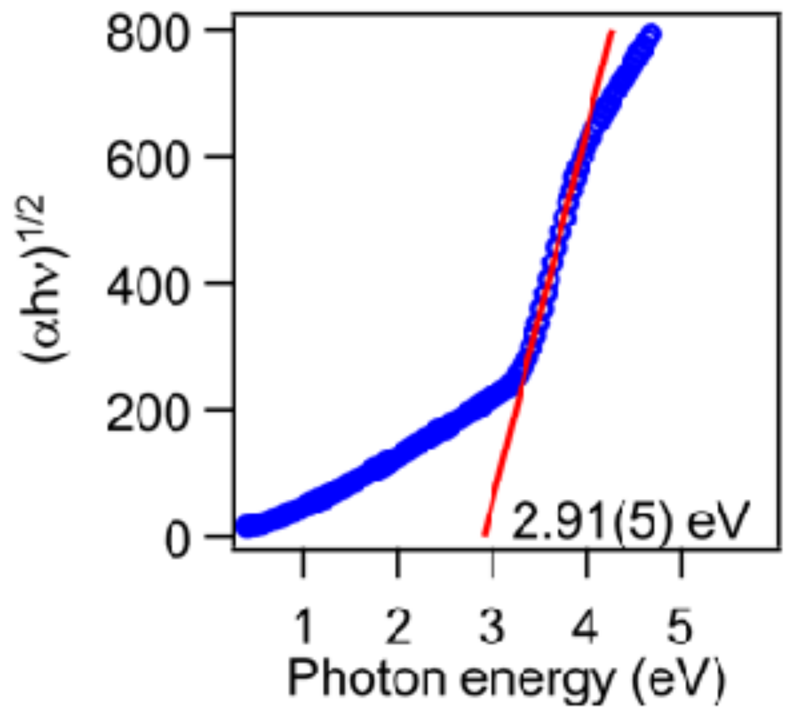

- ☑ Simple cubic (Pm-3m), a = 4.116 Å
- ☑ Band insulator with Indirect band gap, E_g = ~3 eV*,

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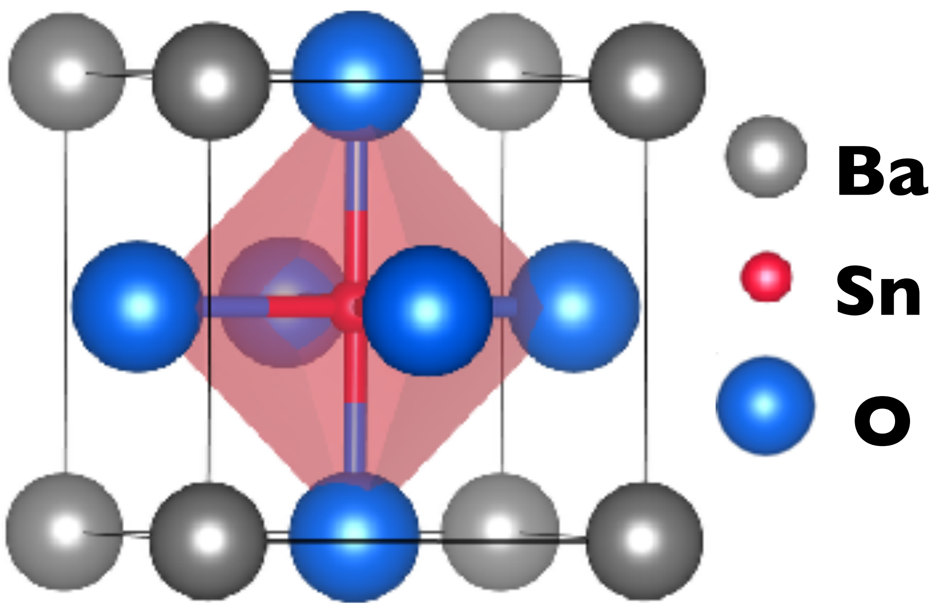

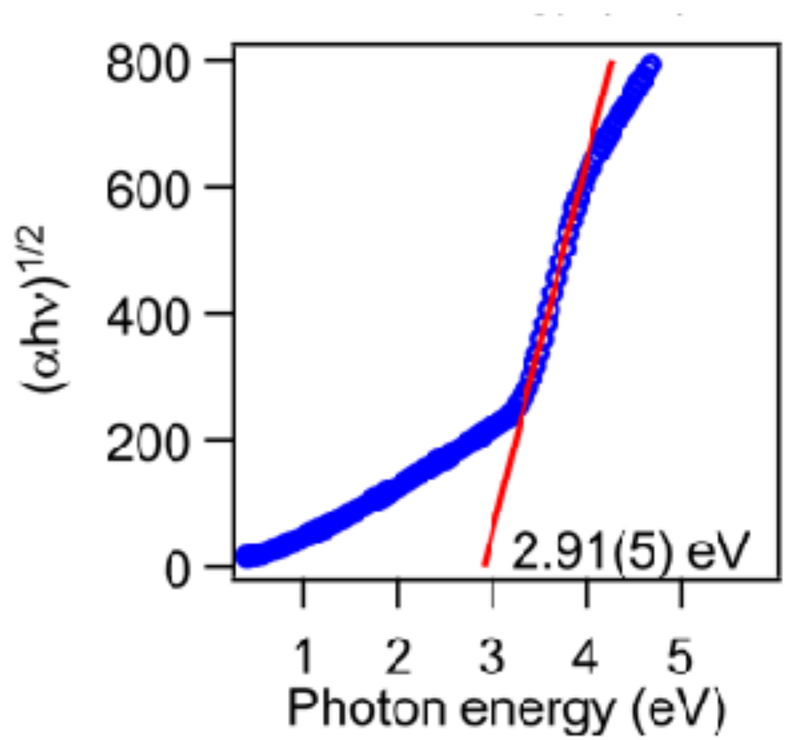

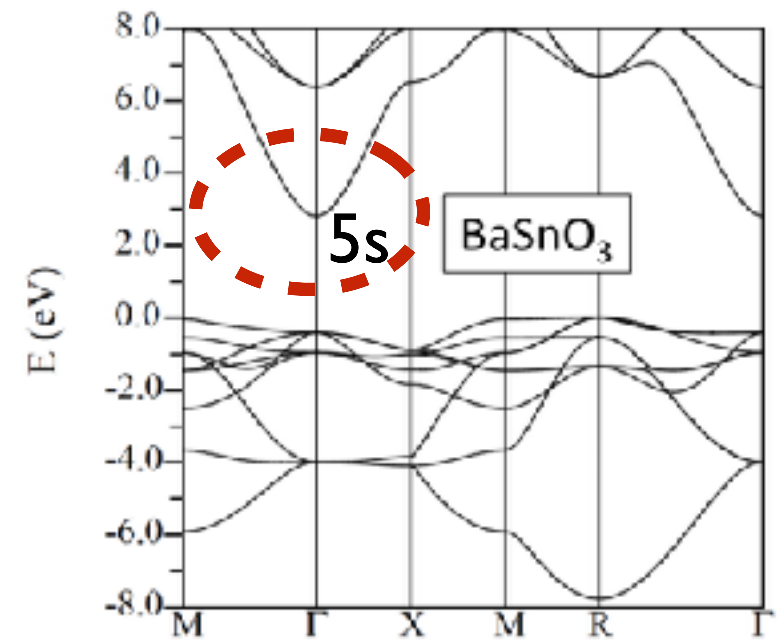

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Y. Li *et al.*, APL Mater. **3**, 011102 (2015)

D. J. Singh et. al., PRB 44, 9519 (1991)

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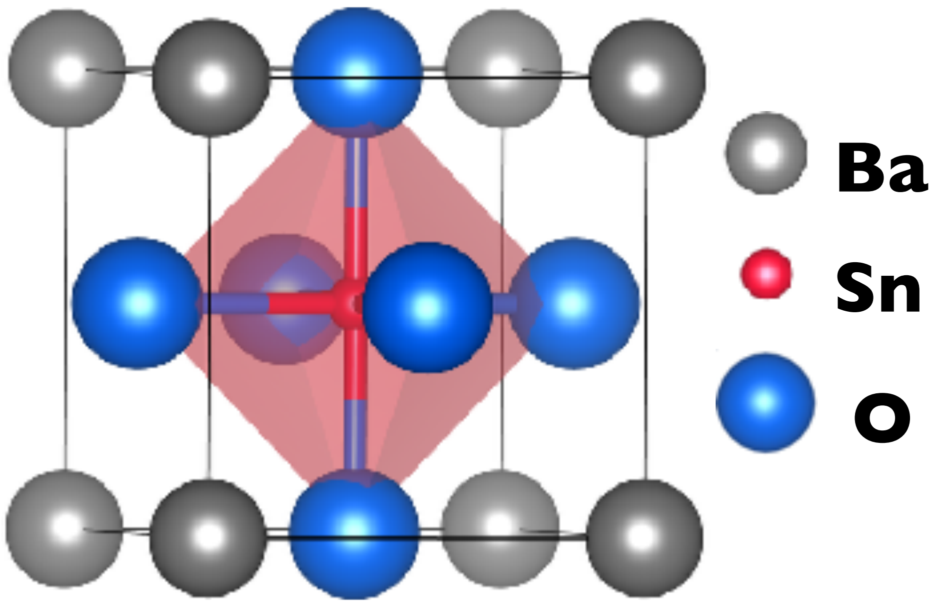

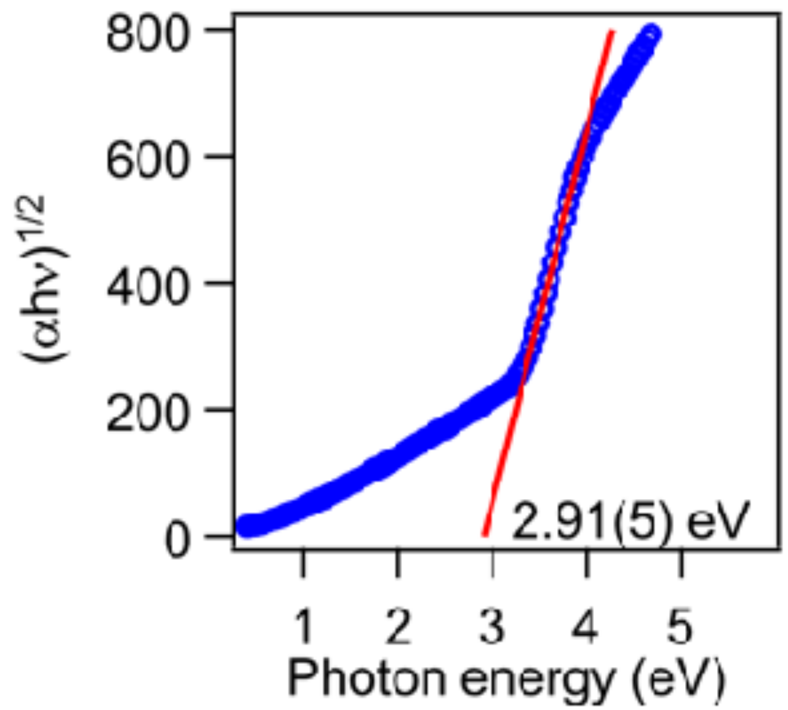

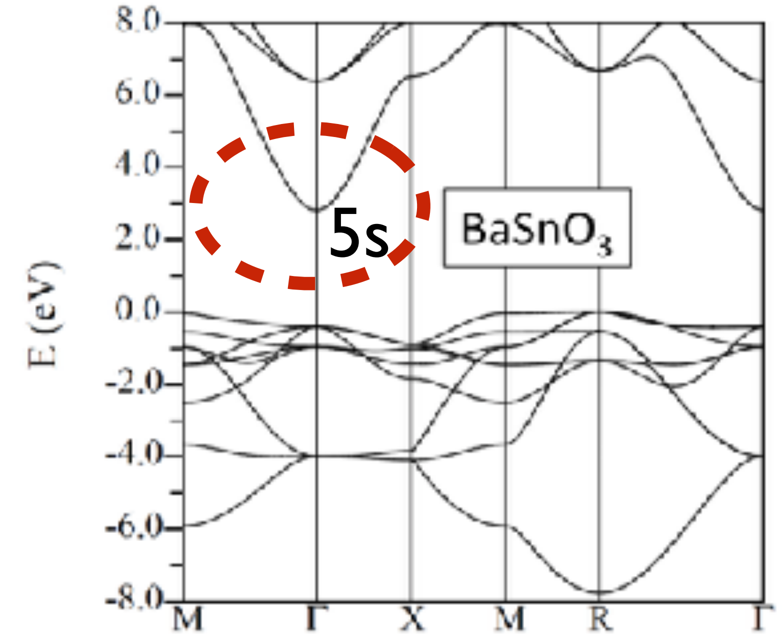

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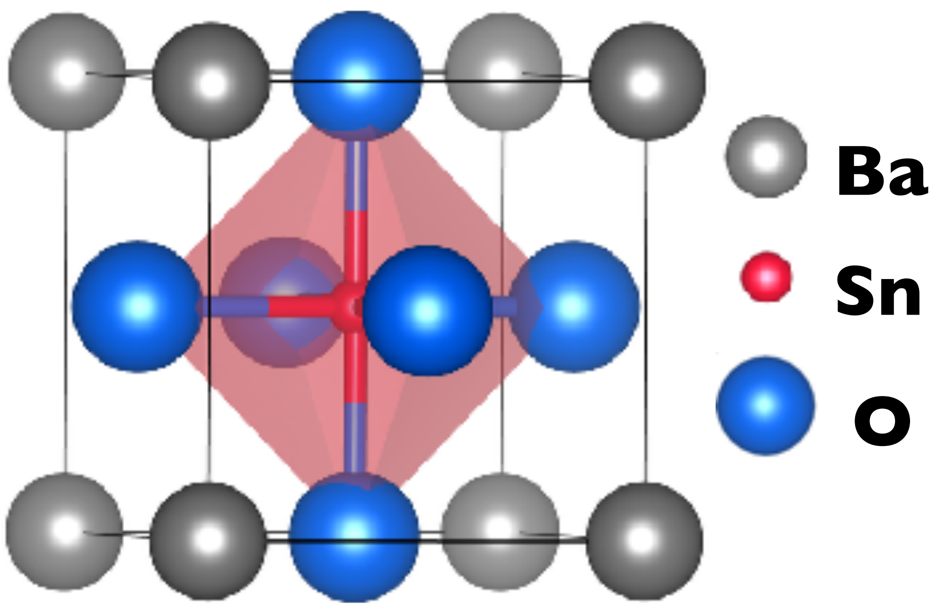

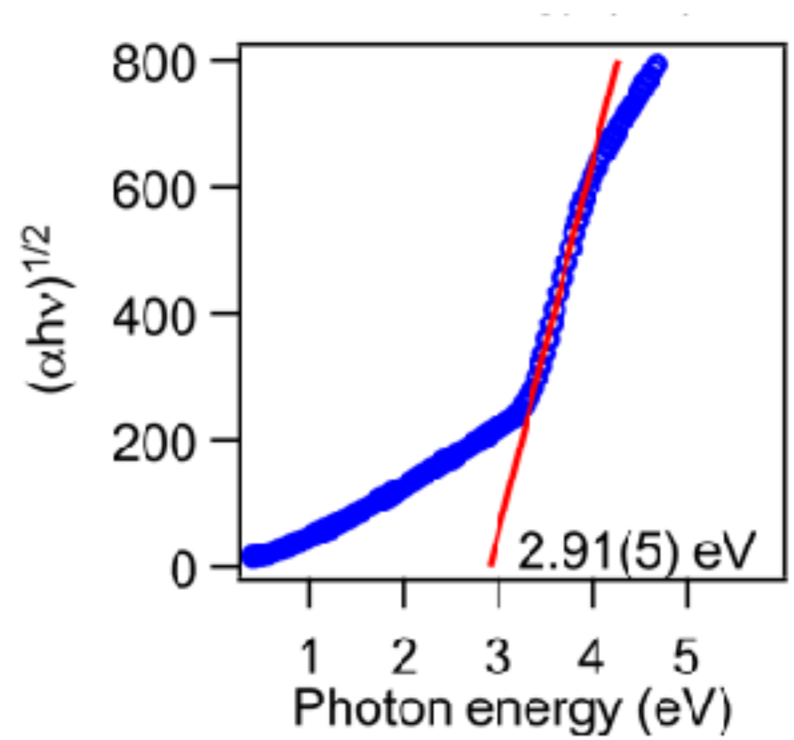

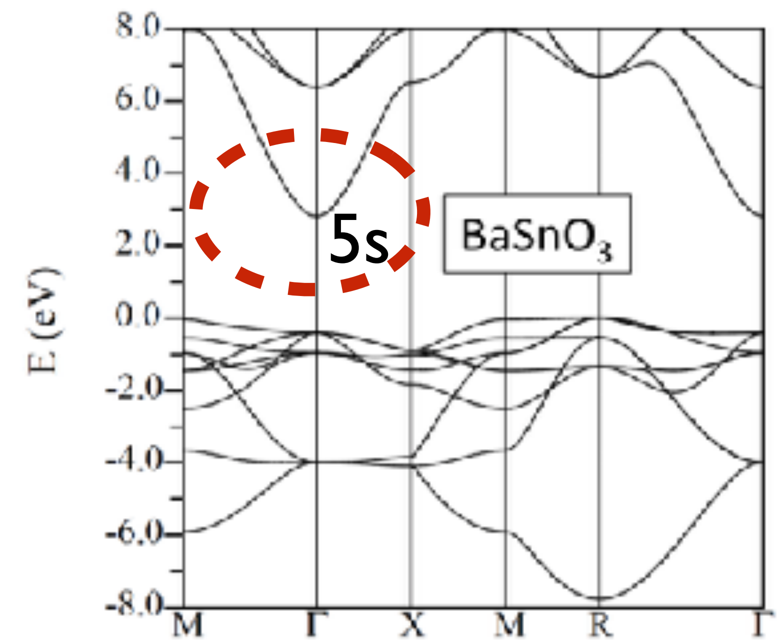

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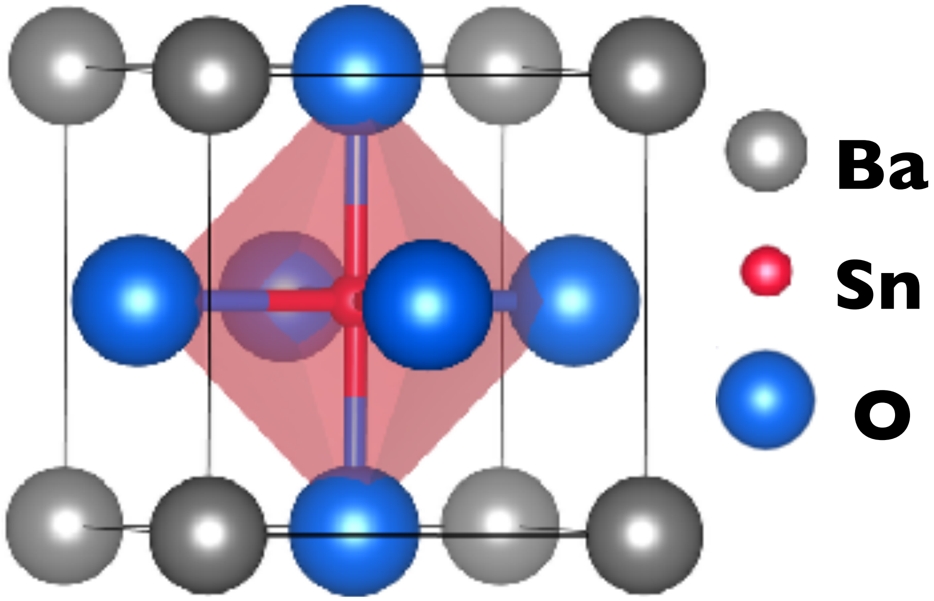

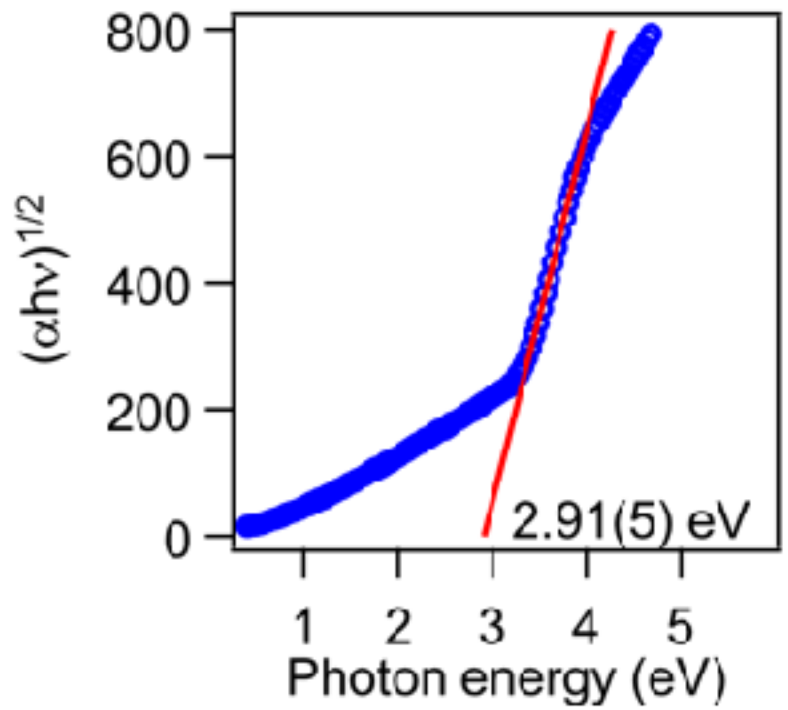

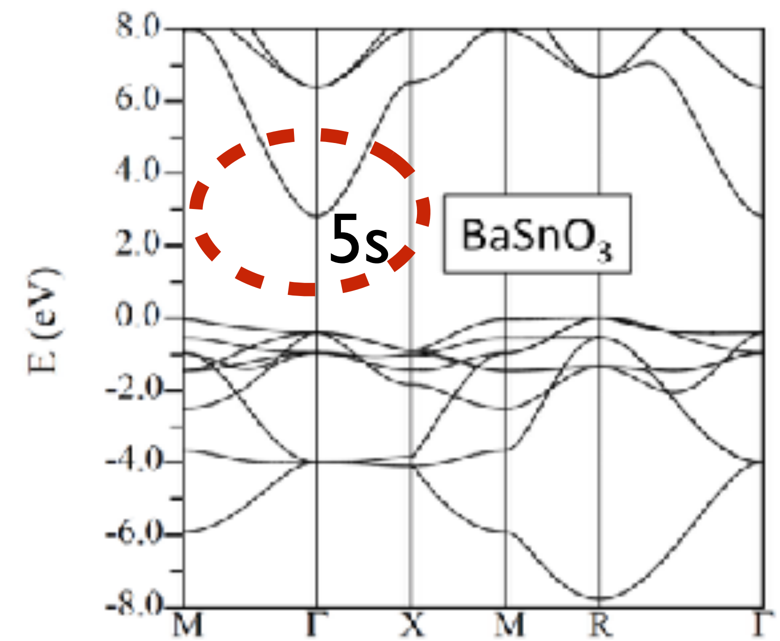

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- Potential applications: Transparent conducting oxide, power electronics, low-dimensional physics of complex oxides with high mobility structures,….

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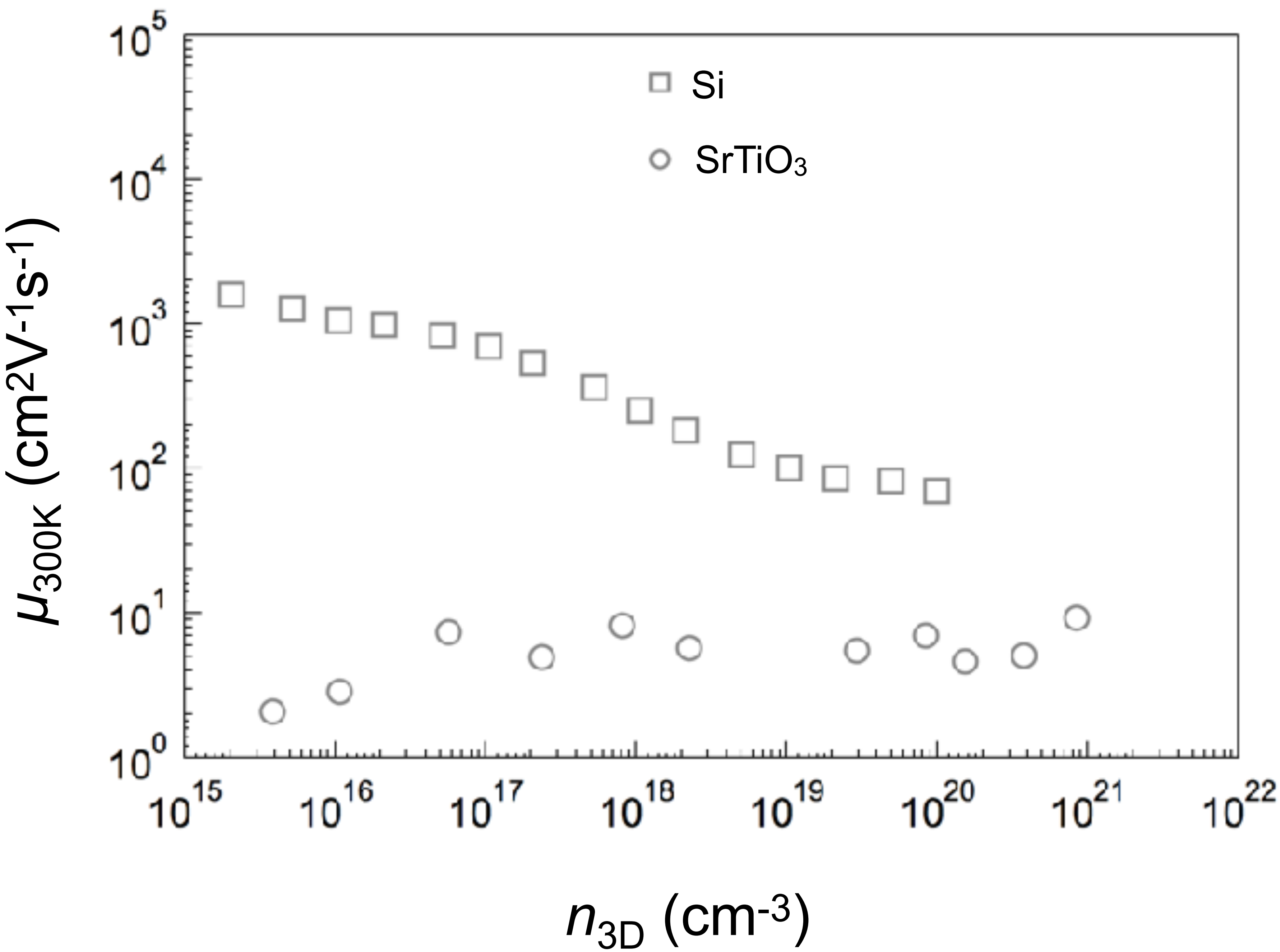
Complex Oxide as a Semiconductor



- $SrTiO_3$ has μ_{300K} = 5-10 $cm^2V^{-1}s^{-1}$

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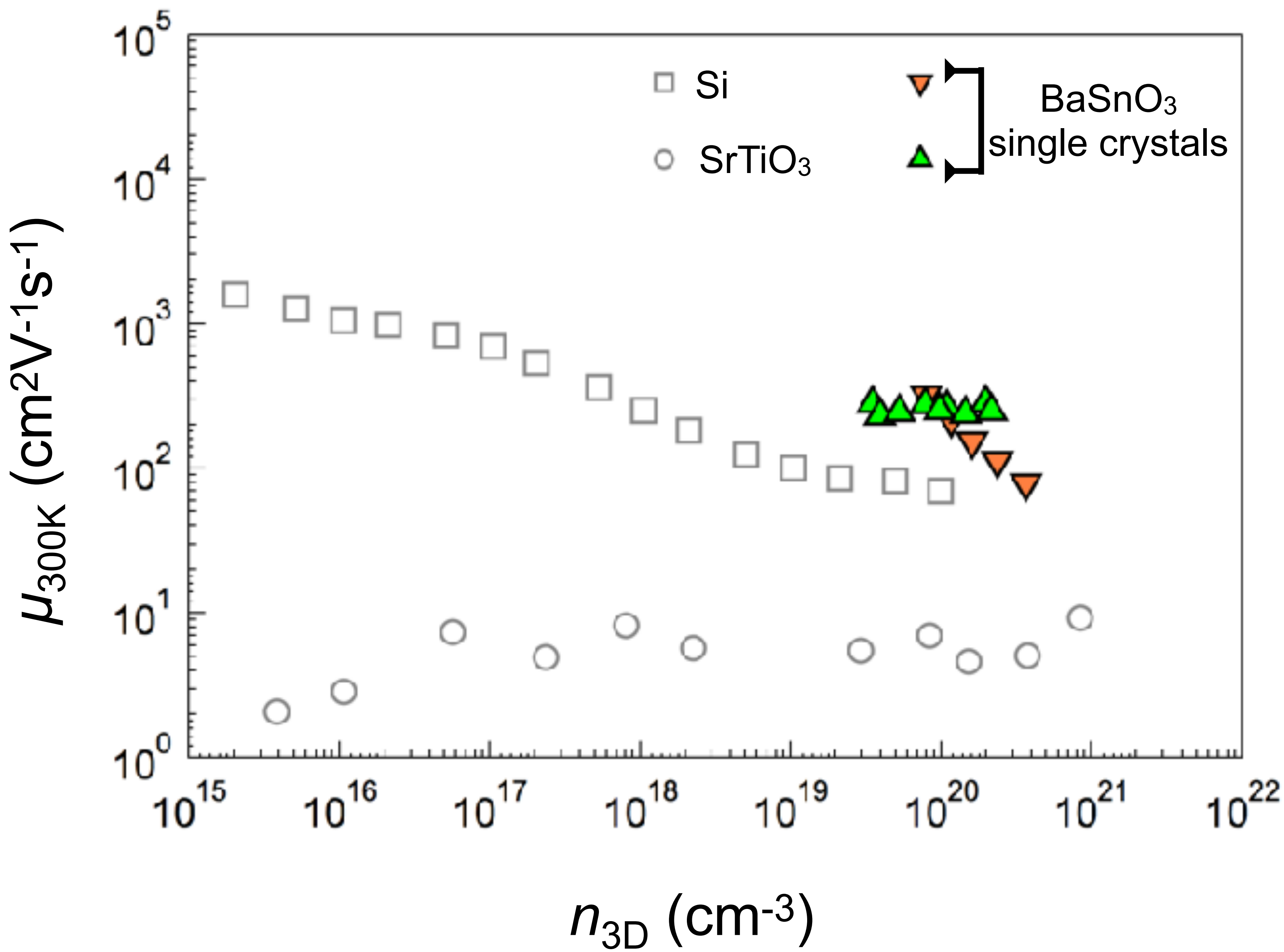
$BaSnO_3$: Mobility Comparison



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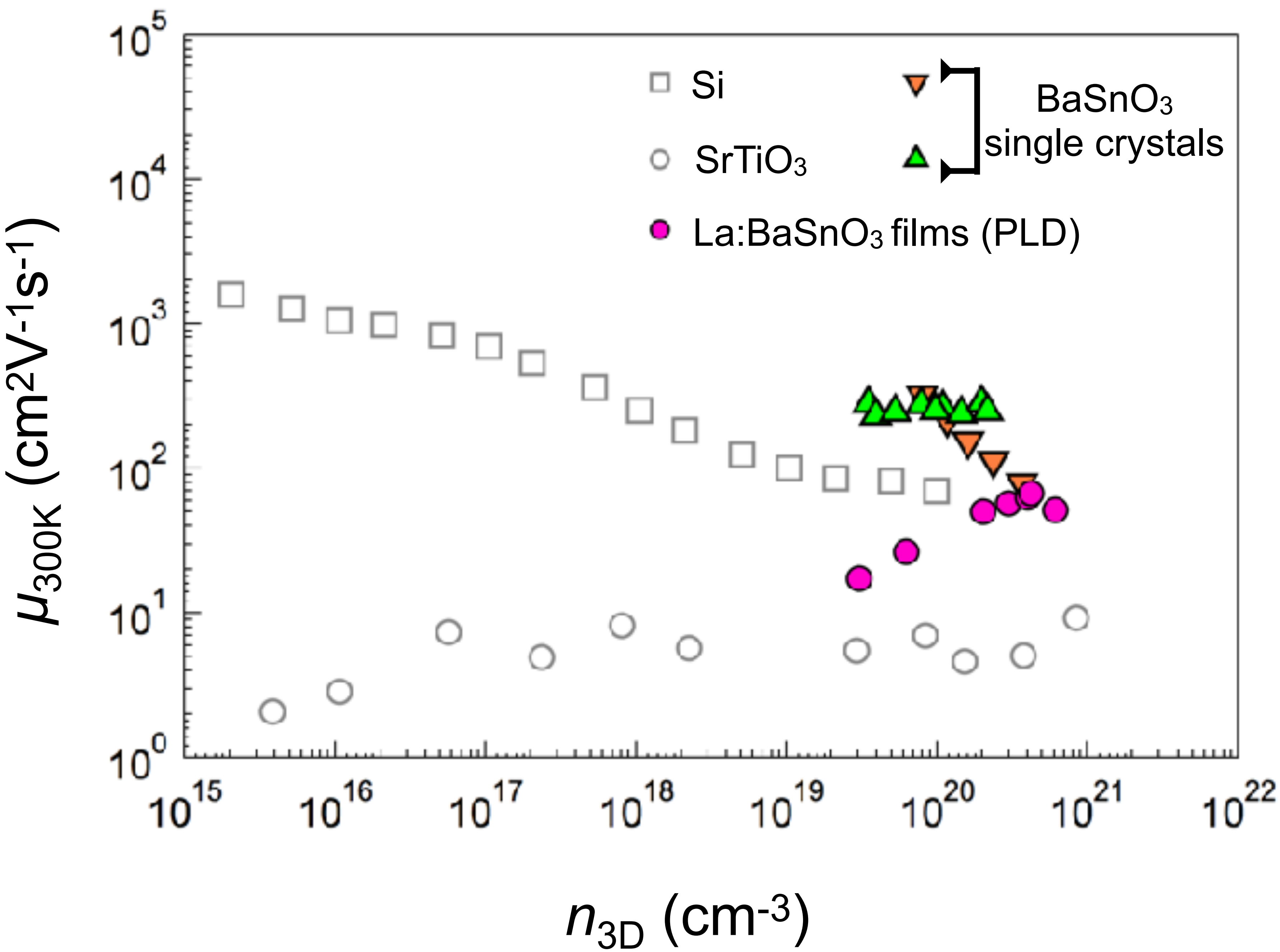

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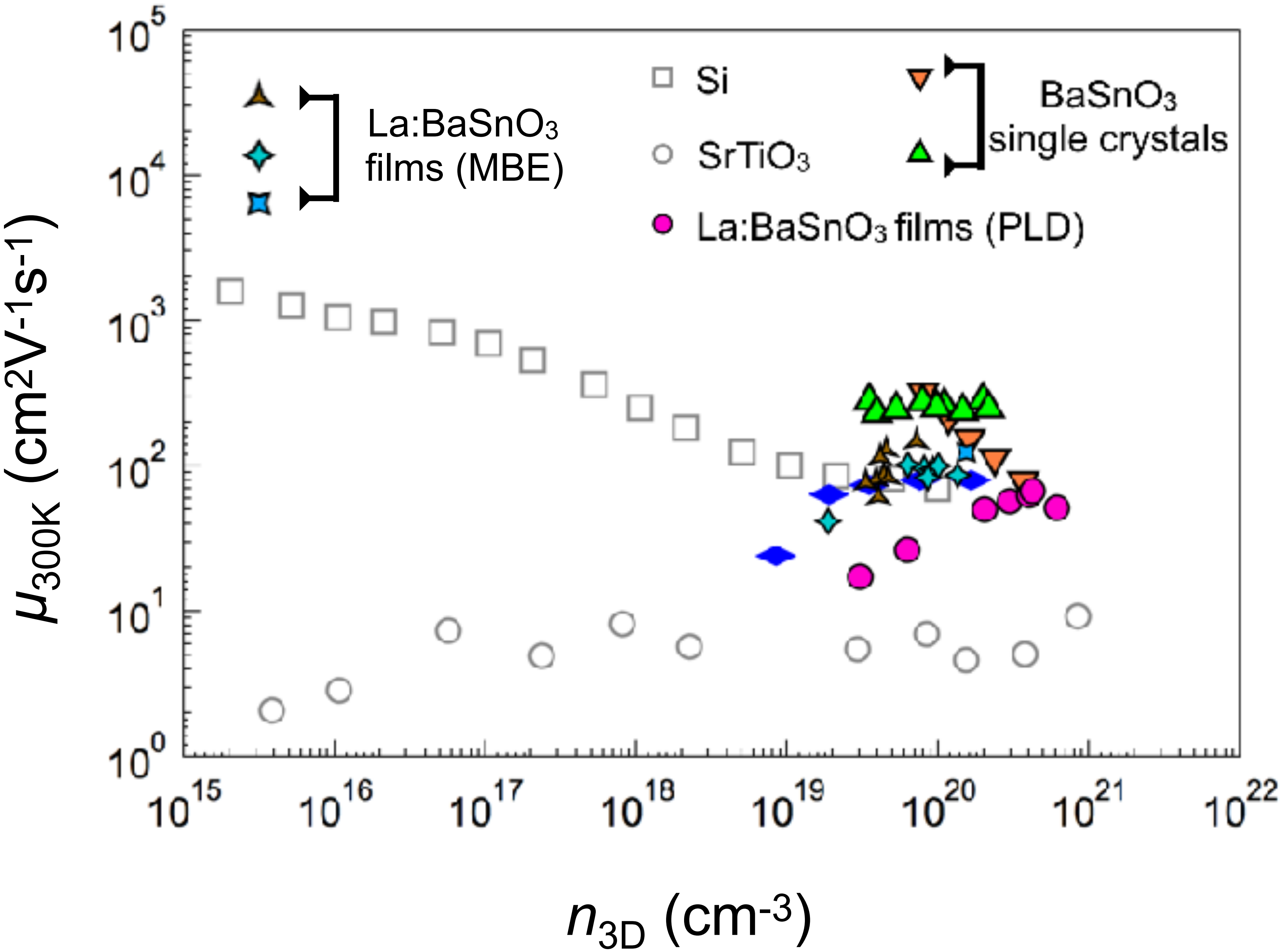

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- $BaSnO_3$ thin films: μ_{300K} = 20-100 $cm^2V^{-1}s^{-1}$(on STO (001)); 150 $cm^2V^{-1}s^{-1}$(on $PrScO_3$ (110))

C. Jacoboni *et al.*, Solid State Electron **20** (2), 77 (1977)
A. Spinelli *et al.*, Phys Rev B **81** (15) (2010)
Z. L-Higgins *et al.*, PRL **116** 027602 (2016)
S. Raghavan *et al.*, APL Mater. **4** 016106 (2016)
H.J. Kim *et al.*, Phys Rev B **86** (16) (2012)
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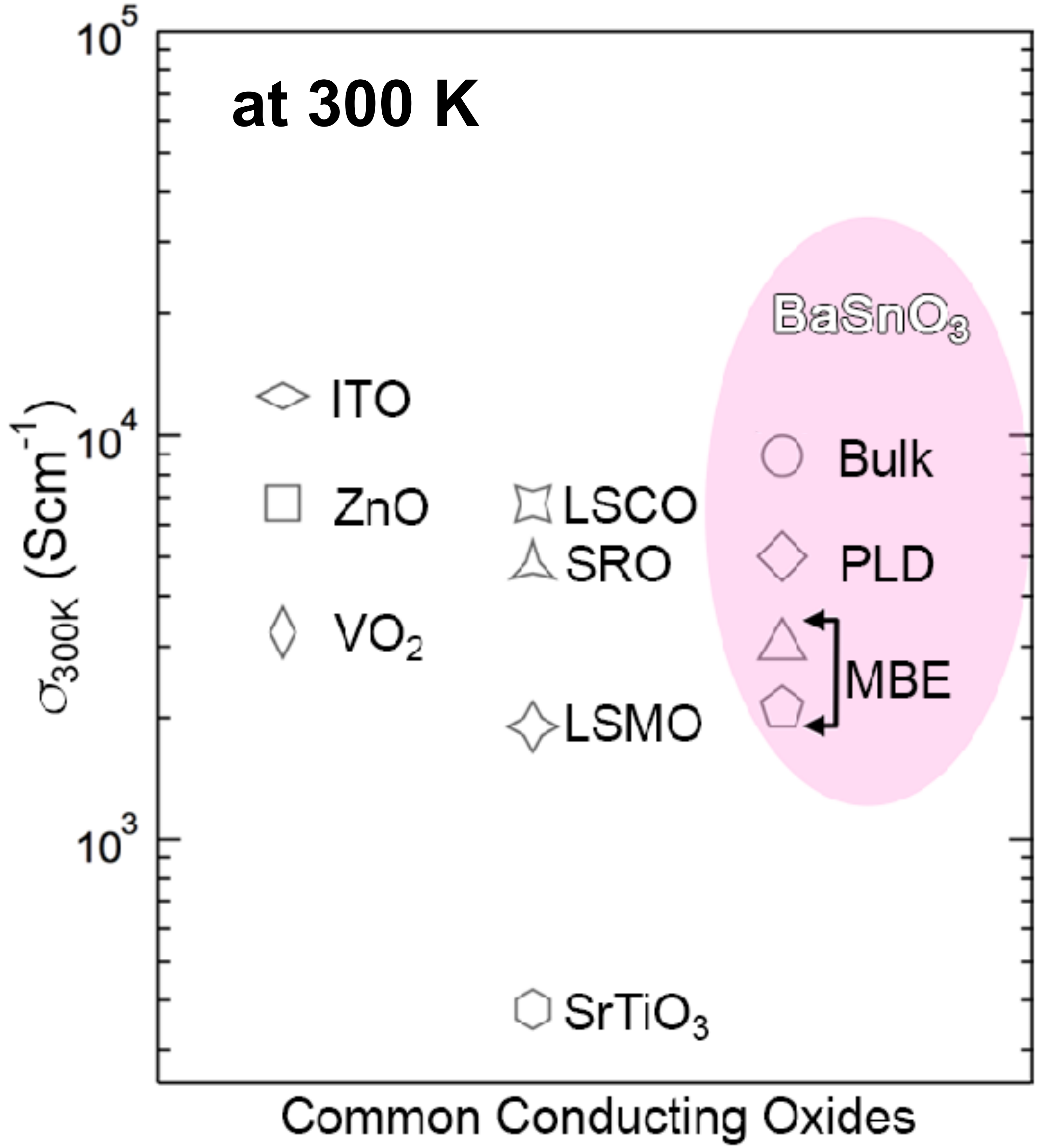
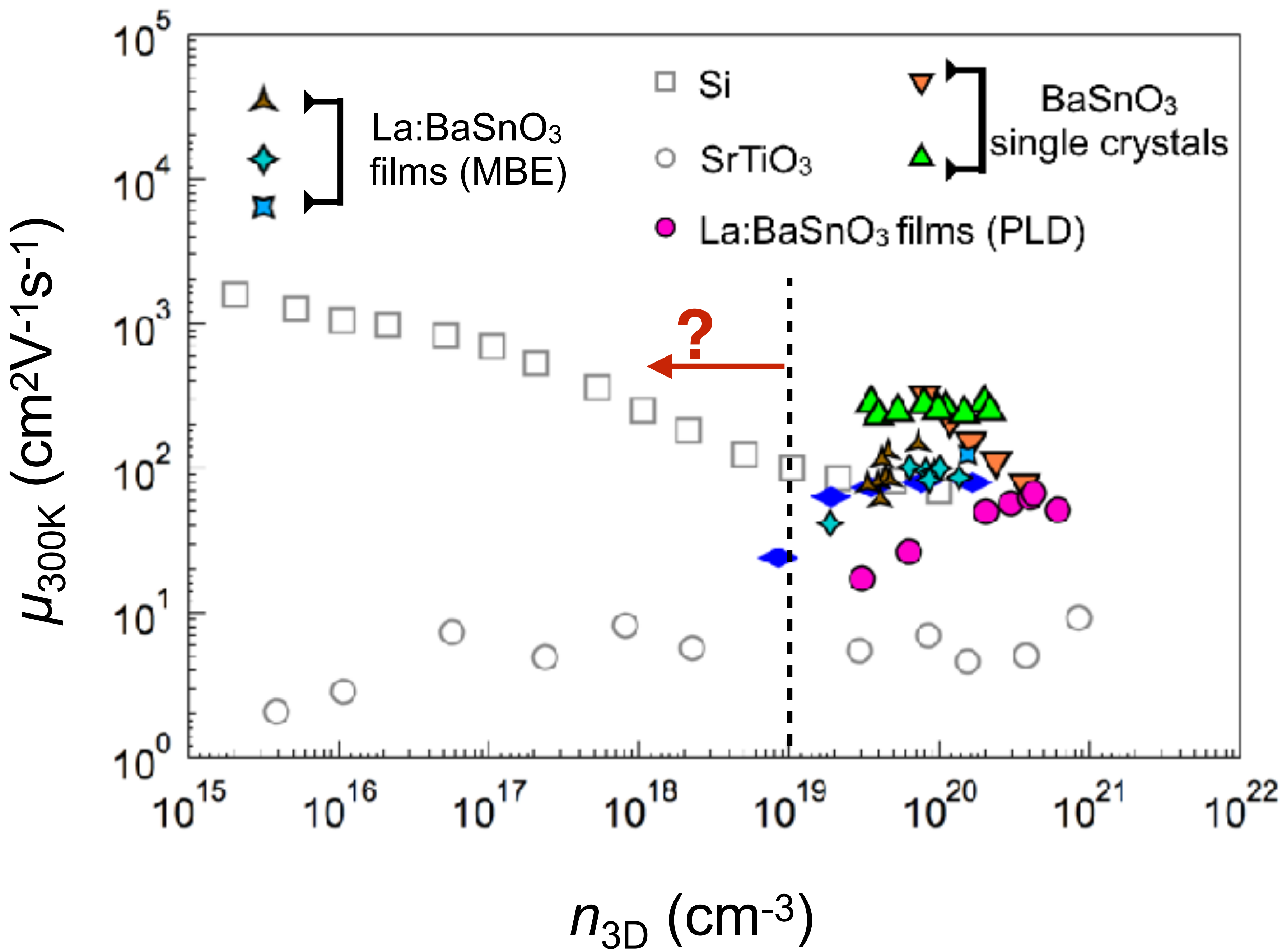
Conductivity: ITO vs $BaSnO_3$



A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

$BaSnO_3$: Scientific Questions



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$BaSnO_3$: Scientific Questions

- What limits the low doping in $BaSnO_3$?
- What limits the electron mobility in thin films?
- What is the ultimate RT conductivity in this material?
- Additional scientific questions?
  - ➡ Role of defects (point defects, stoichiometry, dislocations, etc)
  - ➡ Heterostructure engineering (modulation doping, polarization doping, etc) for 2D physics?

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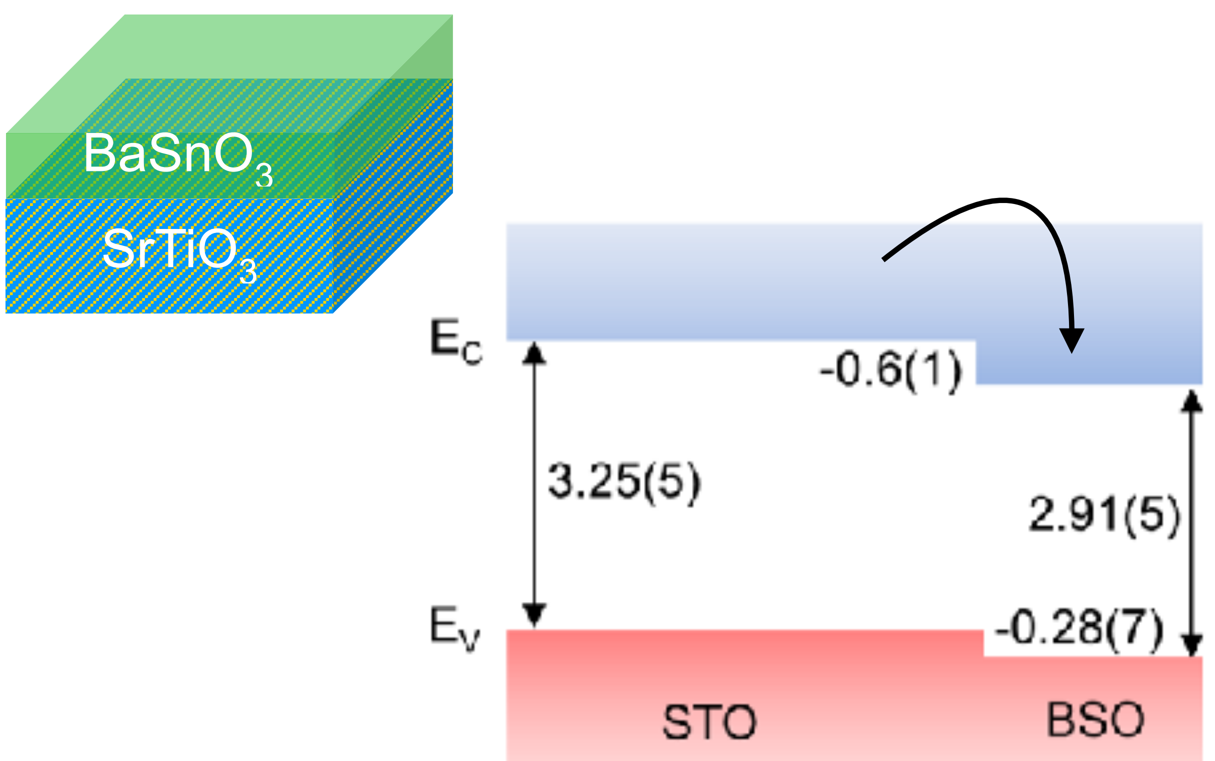
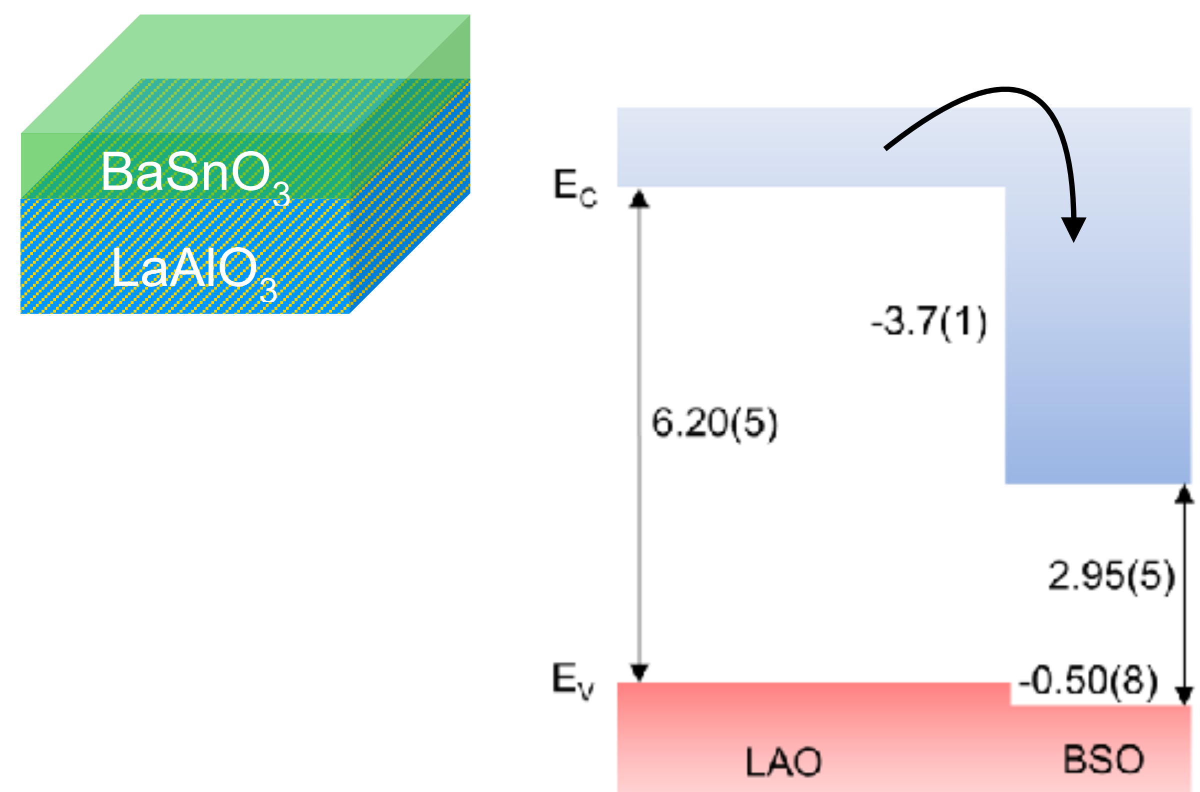




☑ Favorable band offsets for band engineered transport in $BaSnO_3$

S. A. Chambers, T. C. Kaspar, A. Prakash, G. Haugstaad, B. Jalan, *APL* 108, 152104 (2016)

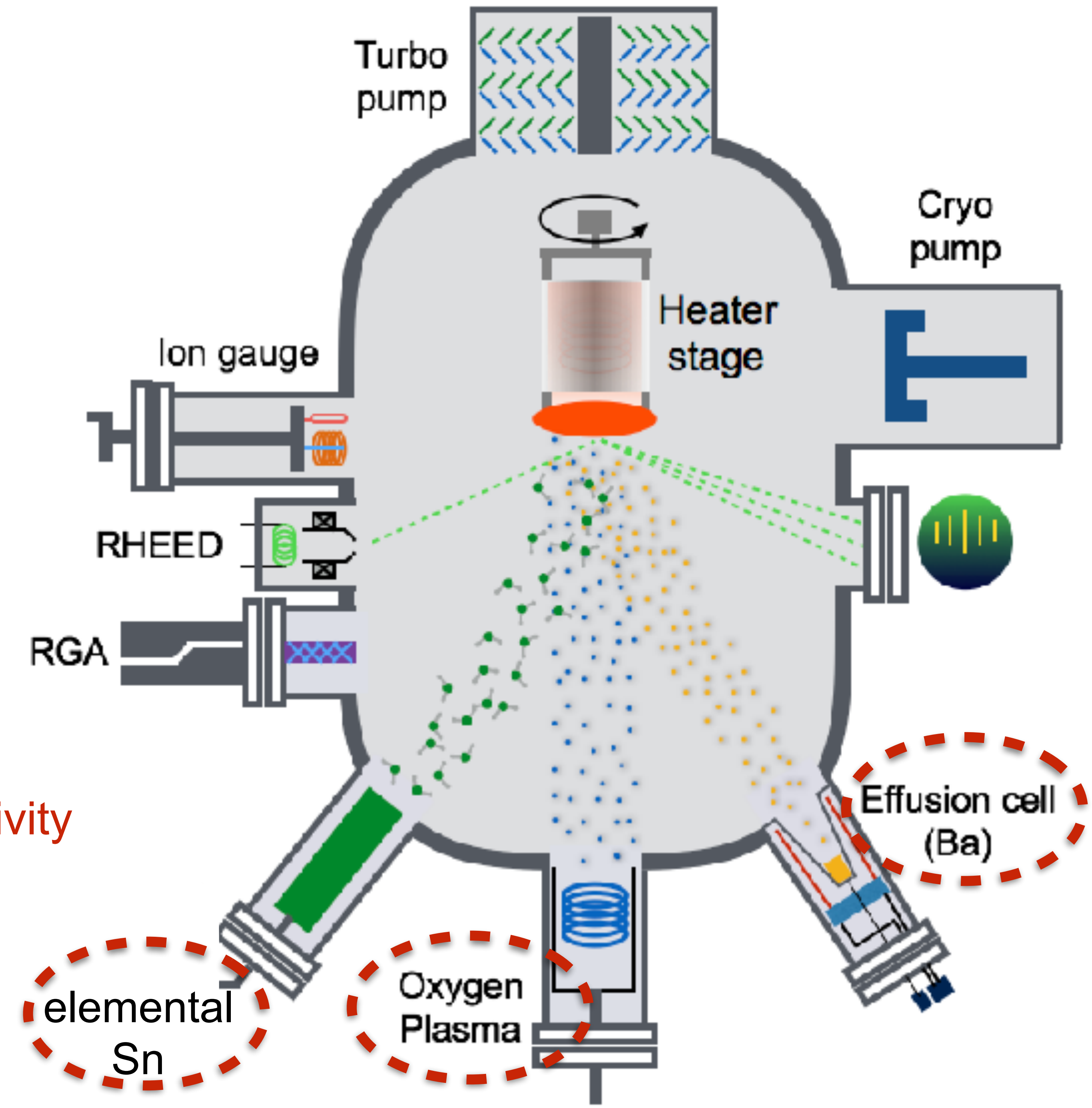
Molecular Beam Epitaxy (MBE)

Advantages:

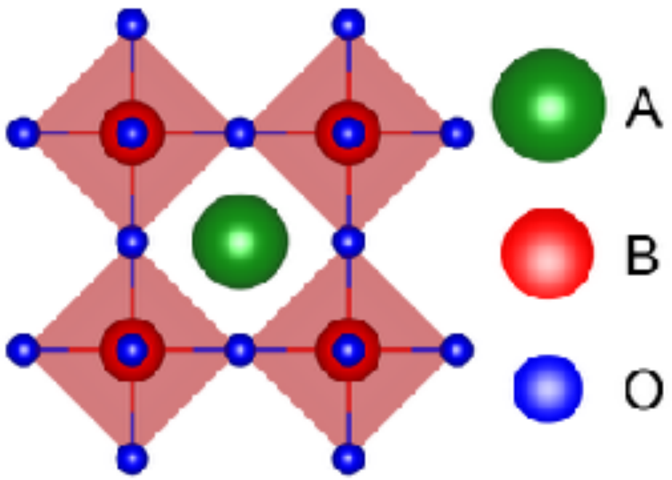
- Low-energetic deposition
- Ultra-pure source materials
- Near-monolayer control
- In-situ diagnostics
- Highest quality III-V films grown by ME

Technical Challenges (for oxide growth):

- Flux Instability in the presence of oxygen
- Stoichiometry control
- Incomplete oxidation for high electronegativity elements (Sn, Ni, W, Ir,……)



MBE: Low Oxidation Potential Elements



Common perovskite oxides

$SrTiO_3$, $BaTiO_3$, $LaTiO_3$, ..
$SrVO_3$, $LaVO_3$, ..
$SrSnO_3$, $BaSnO_3$, ..
$LaNiO_3$, $(La,Sr)CoO_3$, ..

M	$E^0_{oxidation}$ (V)	$M \Rightarrow M^{n+} + ne^-$
Ba	2.912	Easier to oxidize
Sr	2.899	
La	2.379	
Y	2.372	
Ti	1.209	
V	0.868	
Sn	-0.007	
Co	-0.453	
Ni	-0.682	Harder to oxidize

A. Prakash, J. Dewey, H. Yun, J.S. Jeong, K.A. Mkhoyan, and B. Jalan,
J. Vac. Sci. Technol. A **33**, 060608 (2015)

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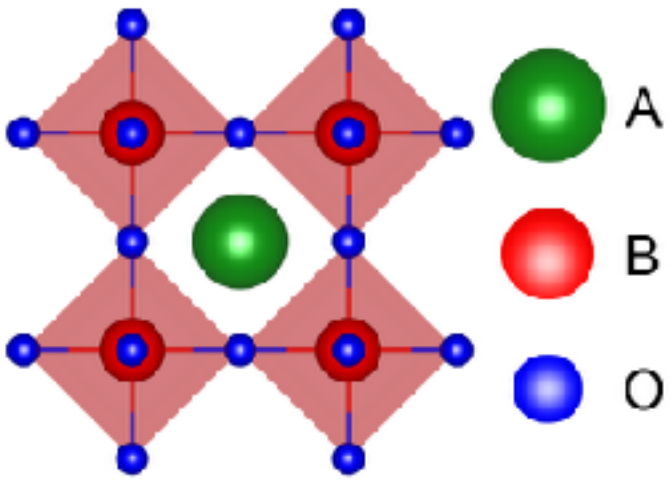

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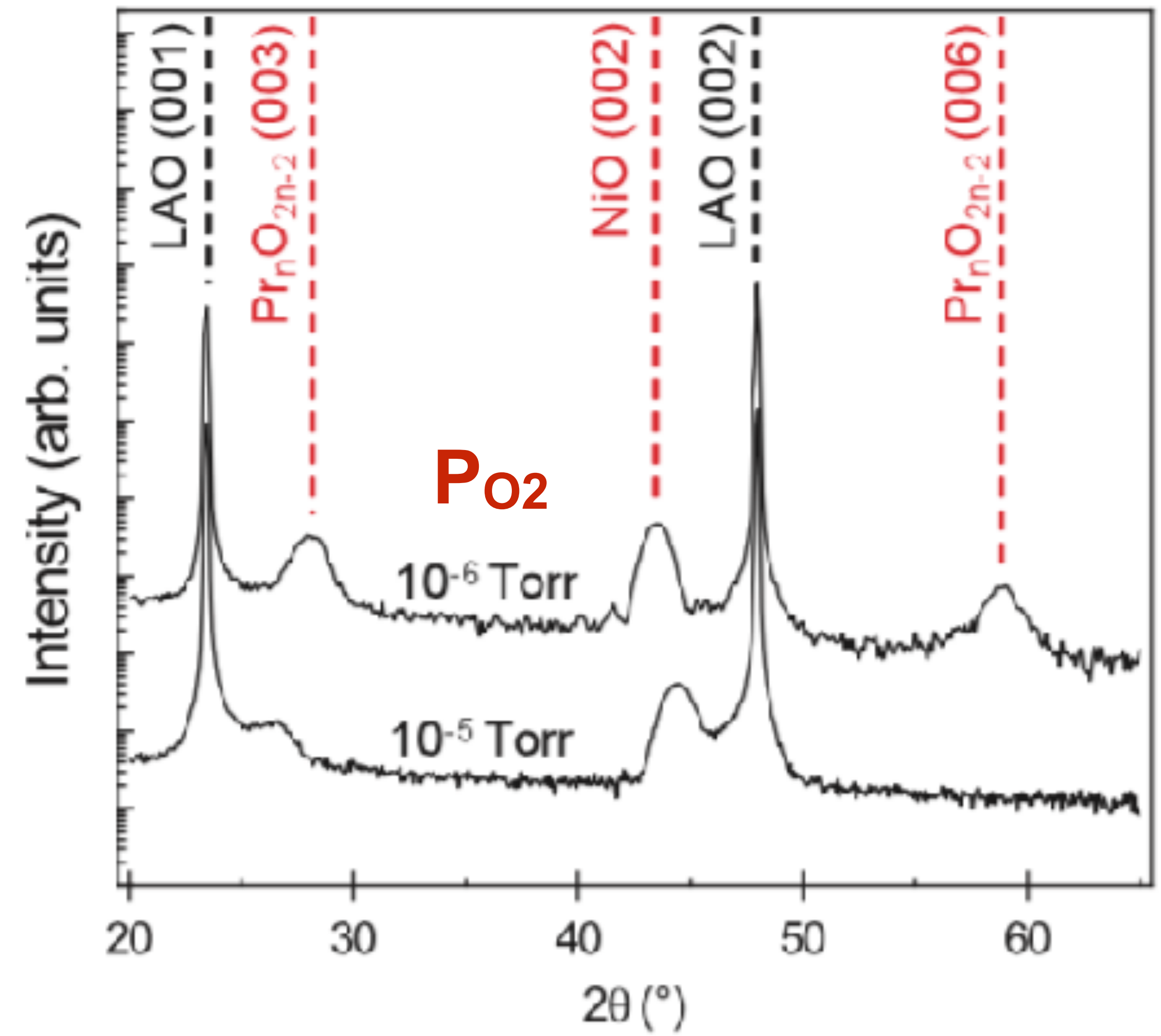
MBE Growth of $PrNi^{+3}O_3$

$Ni^{2+}O^{2-}$ phase due to incomplete oxidation



L. Feigl *et al.*, J Cryst. Growth **366**, 51 (2013)

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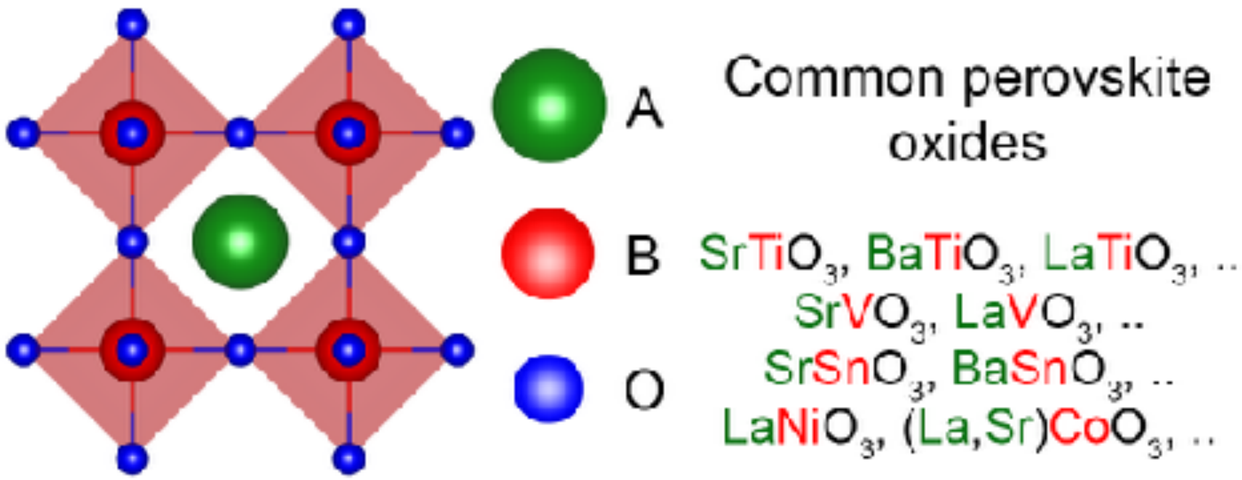

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Remedies:

- Combination of high substrate temperature and high oxygen pressure or
- Use of reactive gases like ozone..

Consequences:

- Instability of metal fluxes in presence of high oxygen, filaments oxidation..

Alternative Approach:

- MOMBE/hybrid MBE with precursor carrying oxygen, e.g. TTIP for Ti, VTIP for V; non-trivial to find oxygen containing and "MBE compatible" precursors

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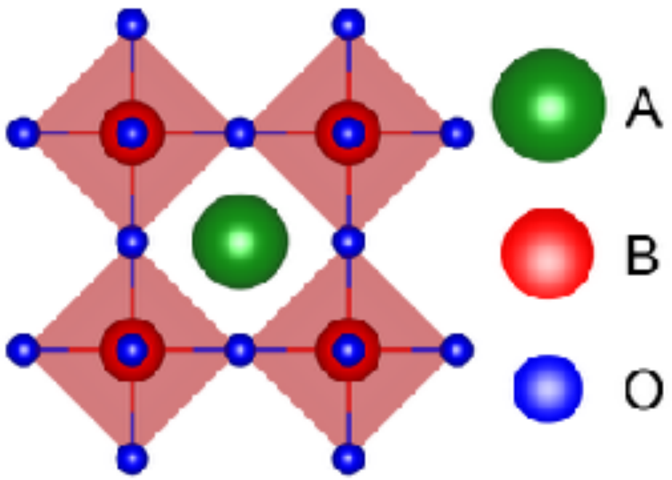

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Our Approach:

- **Instead of making oxidant more reactive, make metal itself more reactive**

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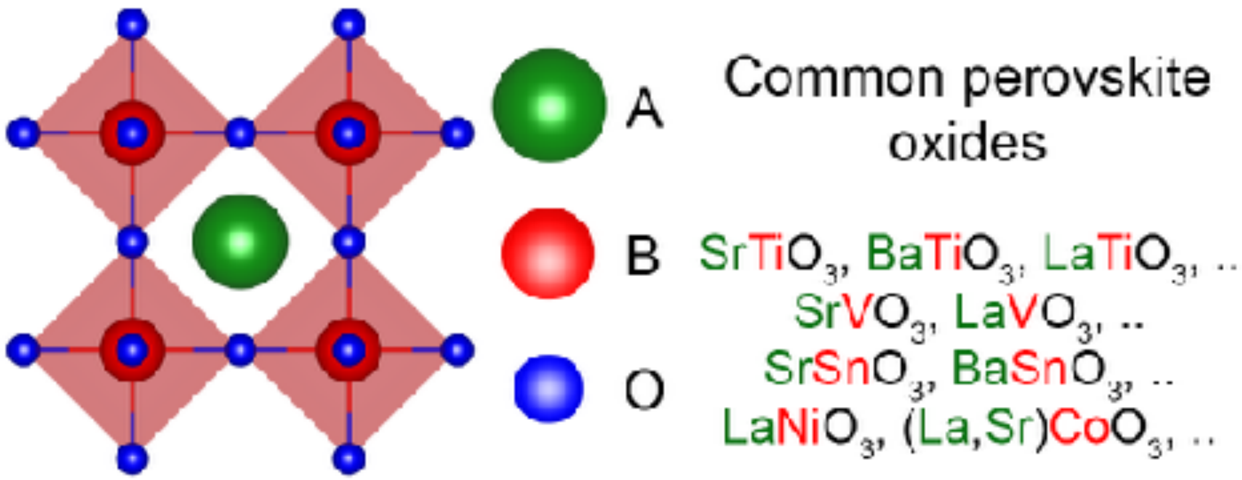

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Consequences:

- Instability of metal fluxes in presence of high oxygen, filaments oxidation..

Alternative Approach:

- MOMBE/hybrid MBE with precursor carrying oxygen, e.g. TTIP for Ti, VTIP for V; non-trivial to find oxygen containing and "MBE compatible" precursors

Our Approach:

- **Instead of making oxidant more reactive, make metal itself more reactive**

Hybrid MBE with Reactive Radicals

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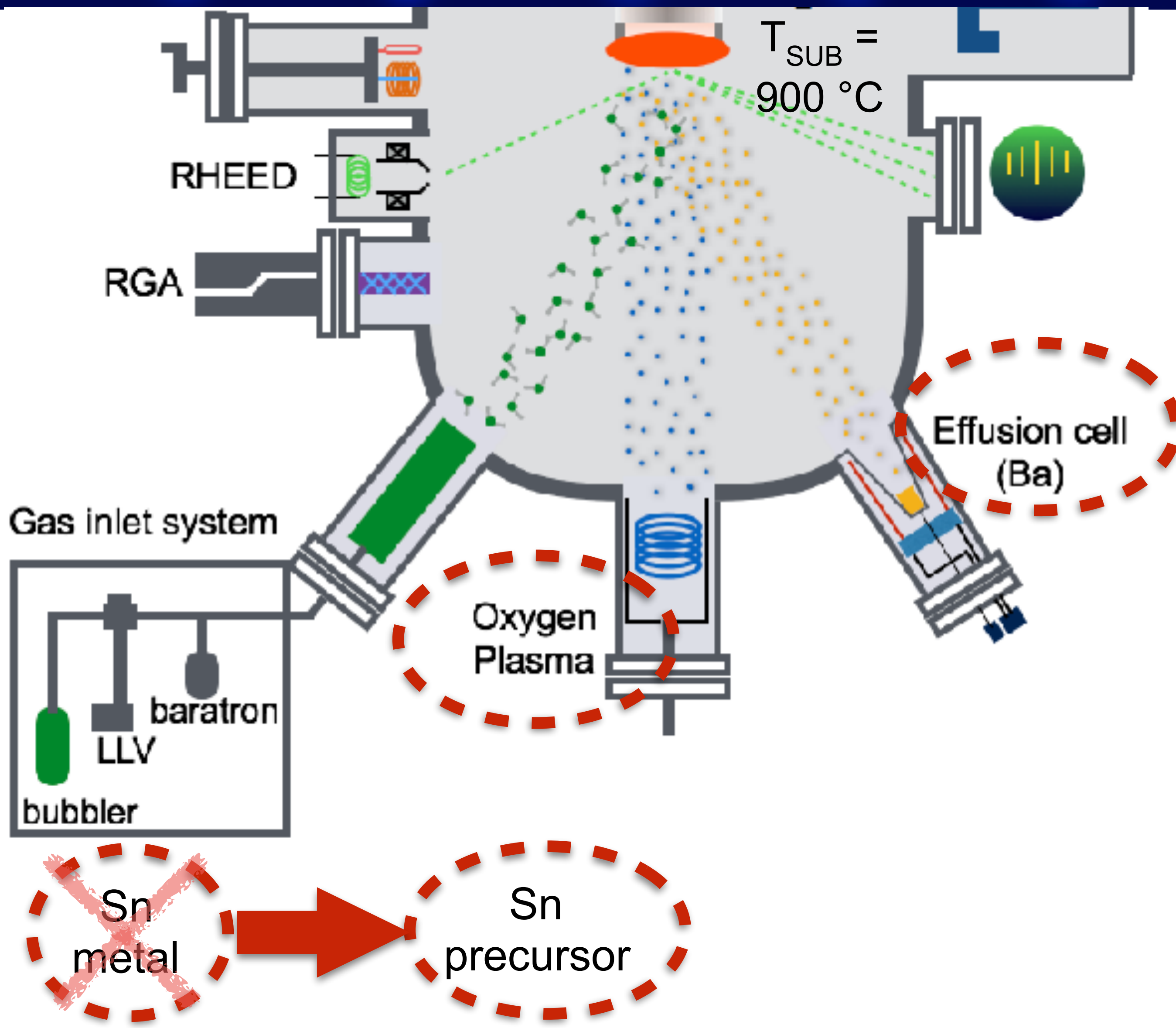

A. Prakash, J. Dewey, H. Yun, J.S. Jeong, K.A. Mkhoyan, and B. Jalan, J. Vac. Sci. Technol. A **33**, 060608 (2015)

Hybrid MBE with Reactive Radicals

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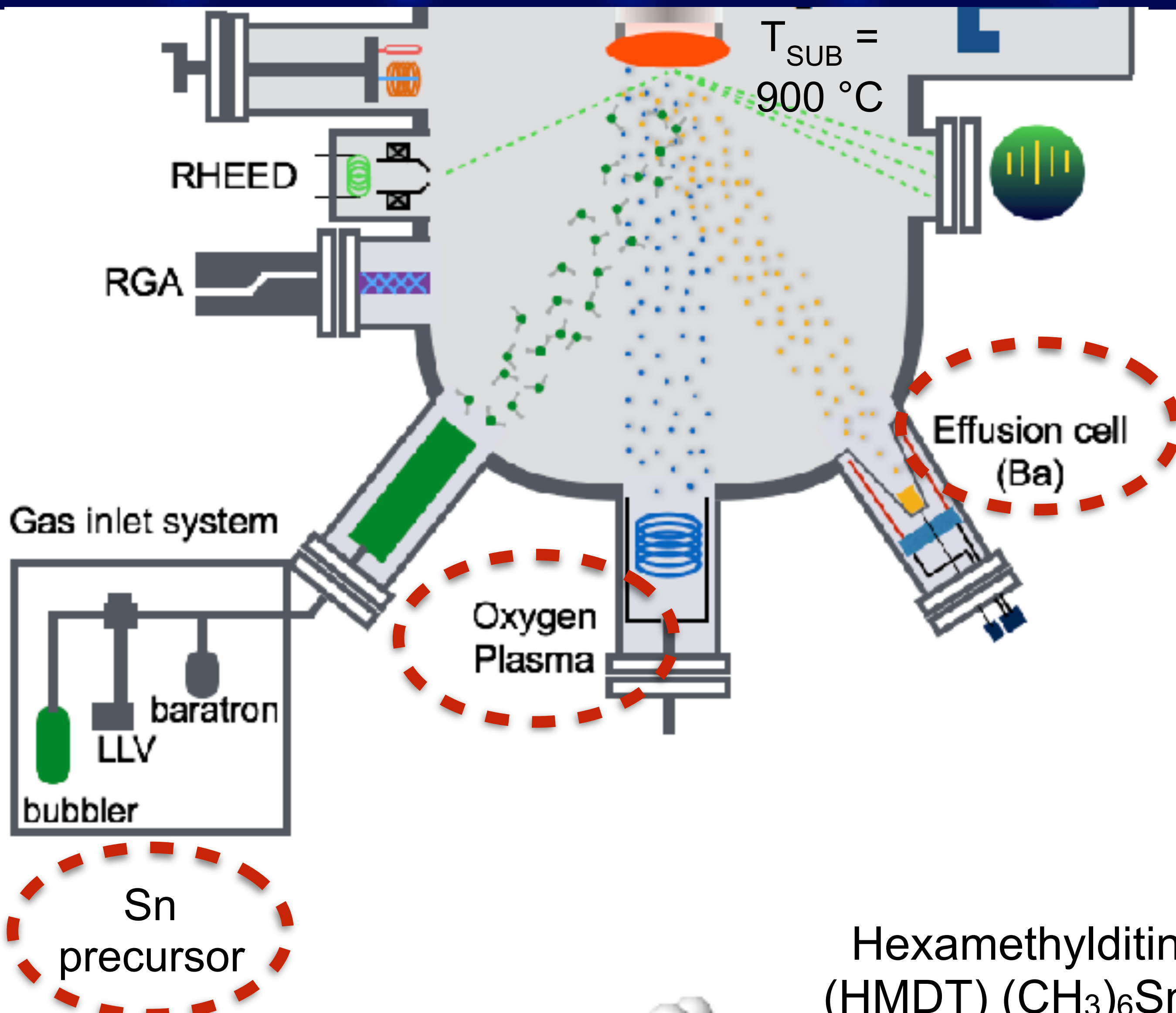

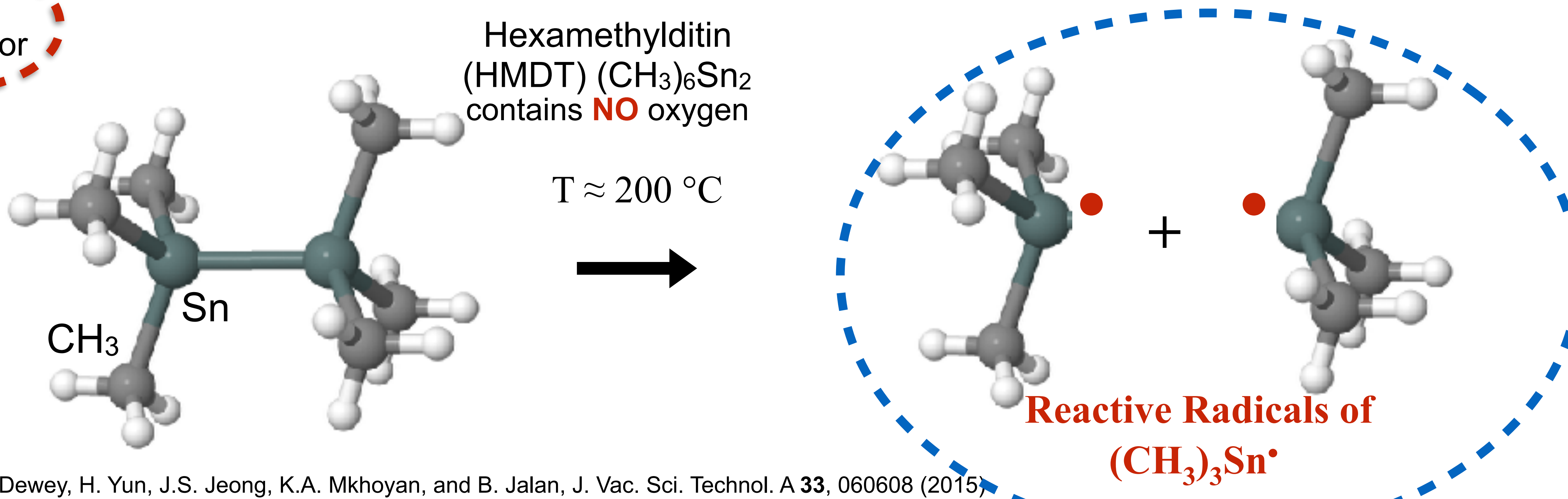

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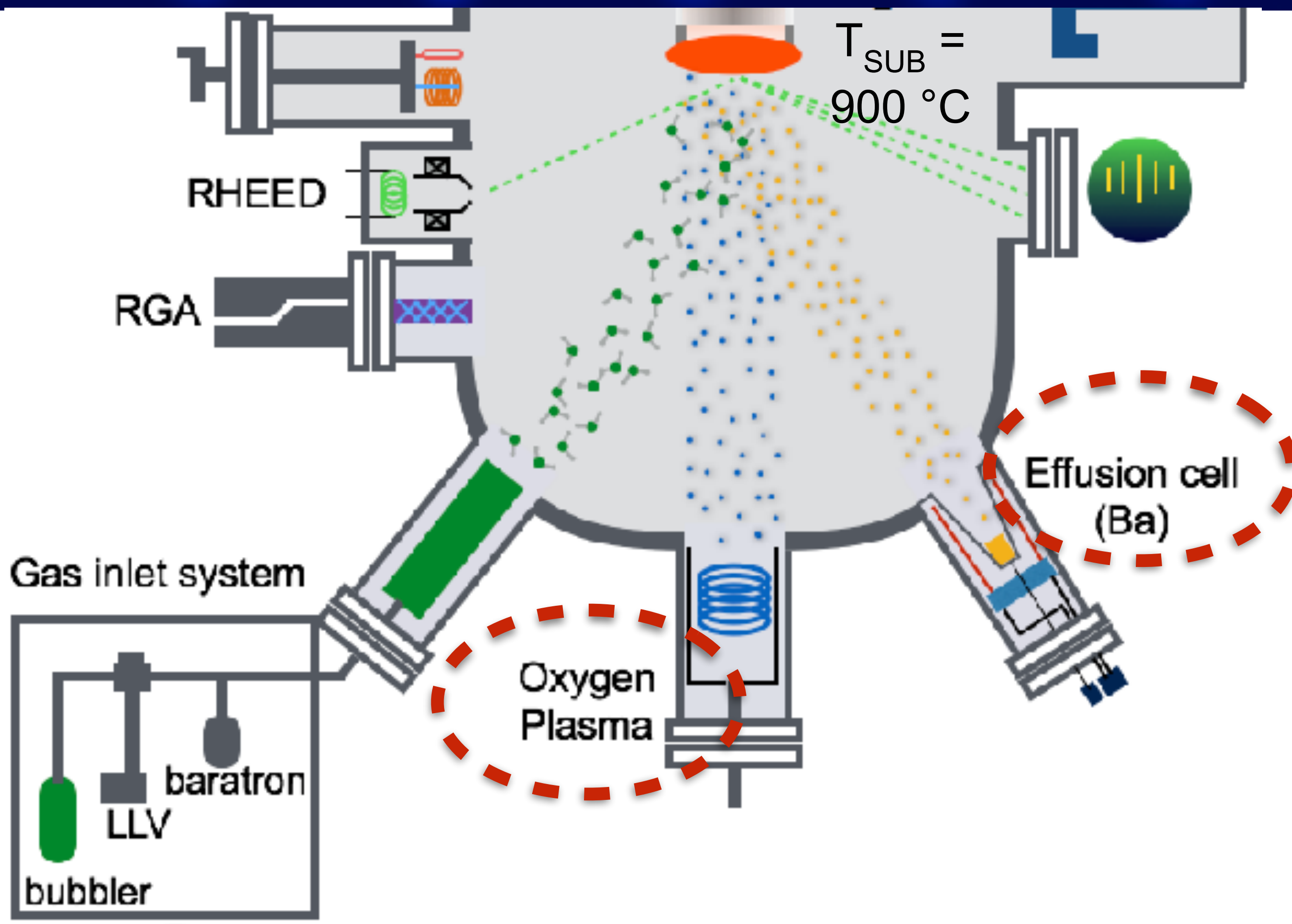
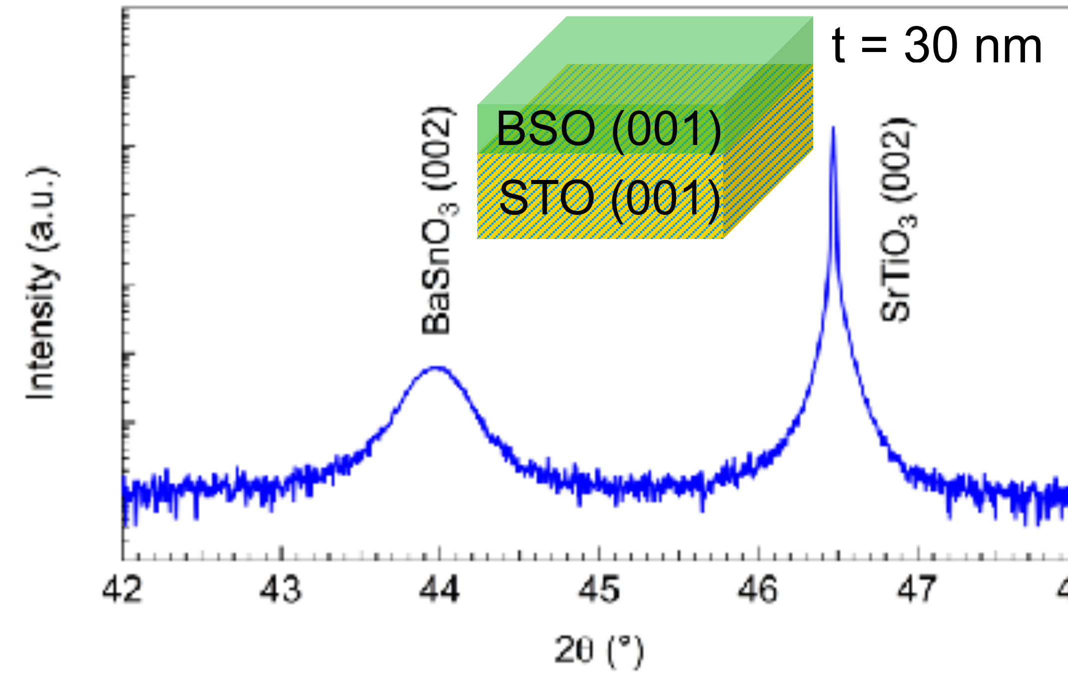


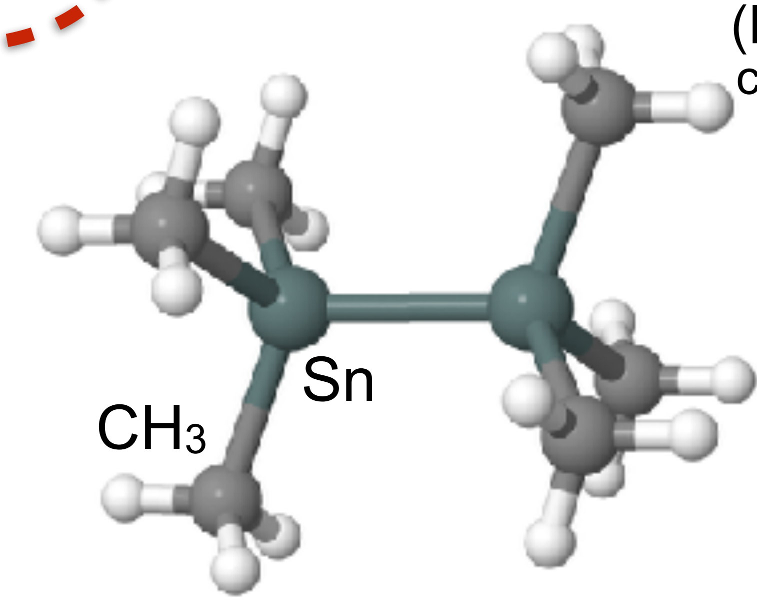
Sn precursor

Phase-pure $BaSnO_3$ even without plasma

out-of-plane lattice parameter = 4.116 Å

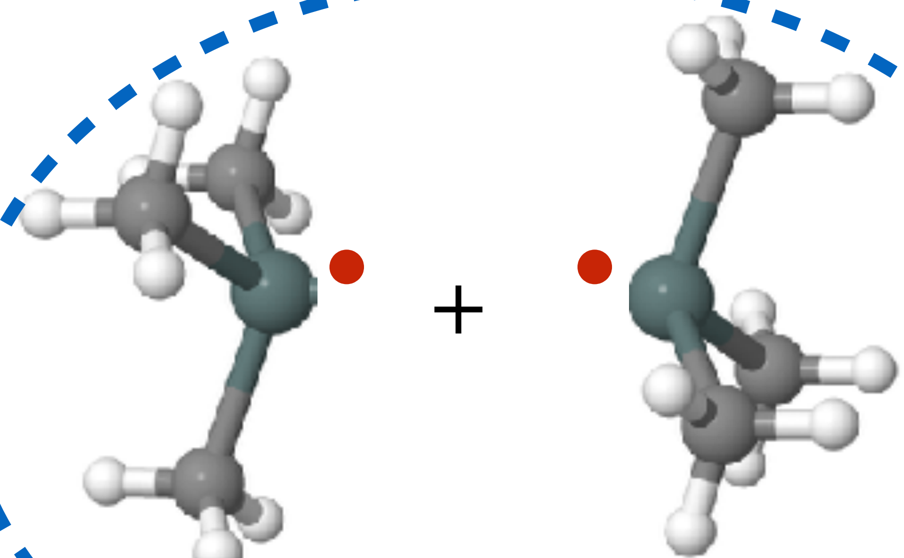


Hexamethylditin (HMDT) $(CH_3)_6Sn_2$ contains **NO** oxygen



$T \approx 200\ °C$ →

molecular oxygen ↑



Reactive Radicals of $(CH_3)_3Sn^{\bullet}$

A. Prakash, J. Dewey, H. Yun, J.S. Jeong, K.A. Mkhoyan, and B. Jalan, J. Vac. Sci. Technol. A **33**, 060608 (2015)

Hybrid MBE with Reactive Radicals

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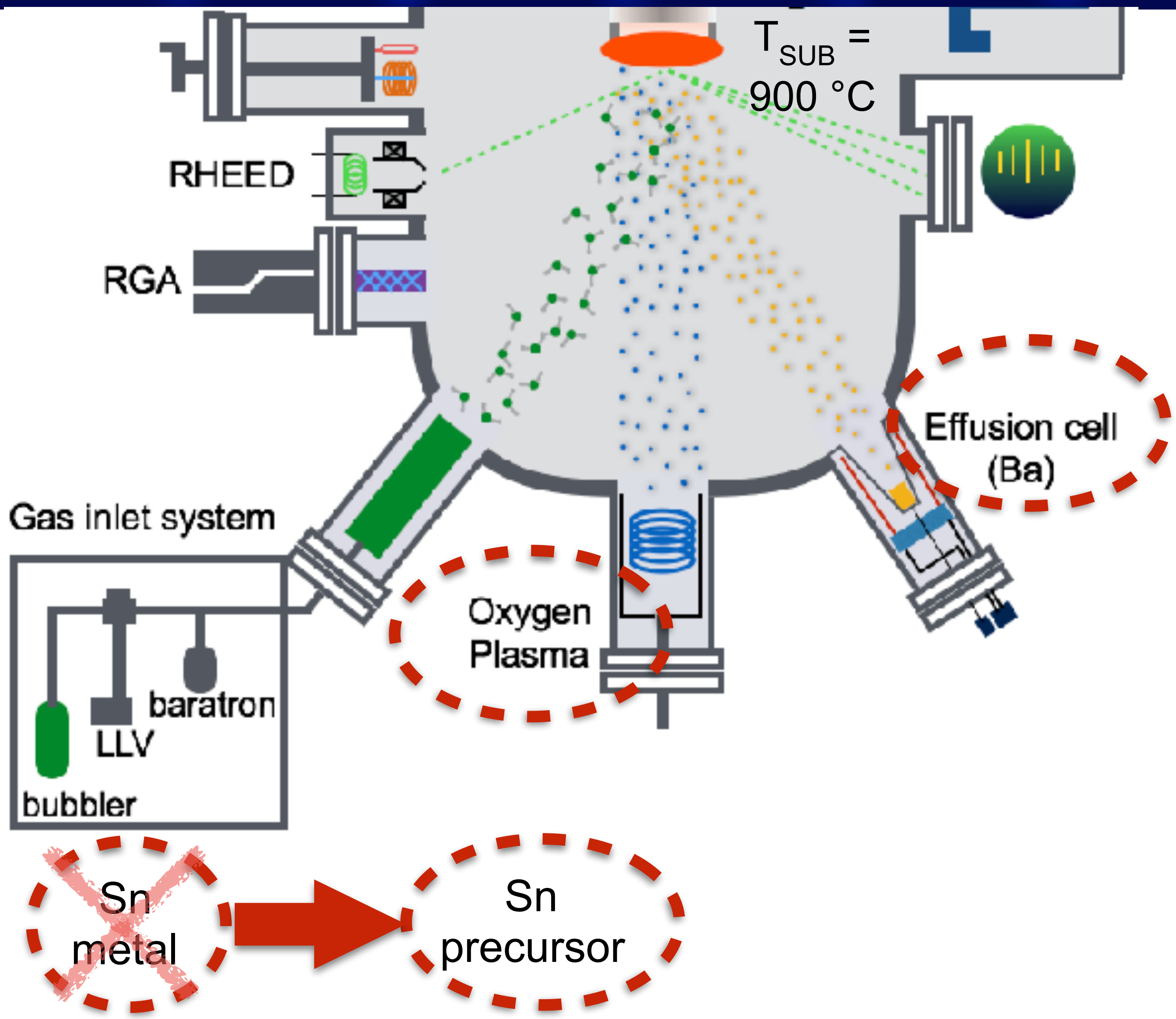
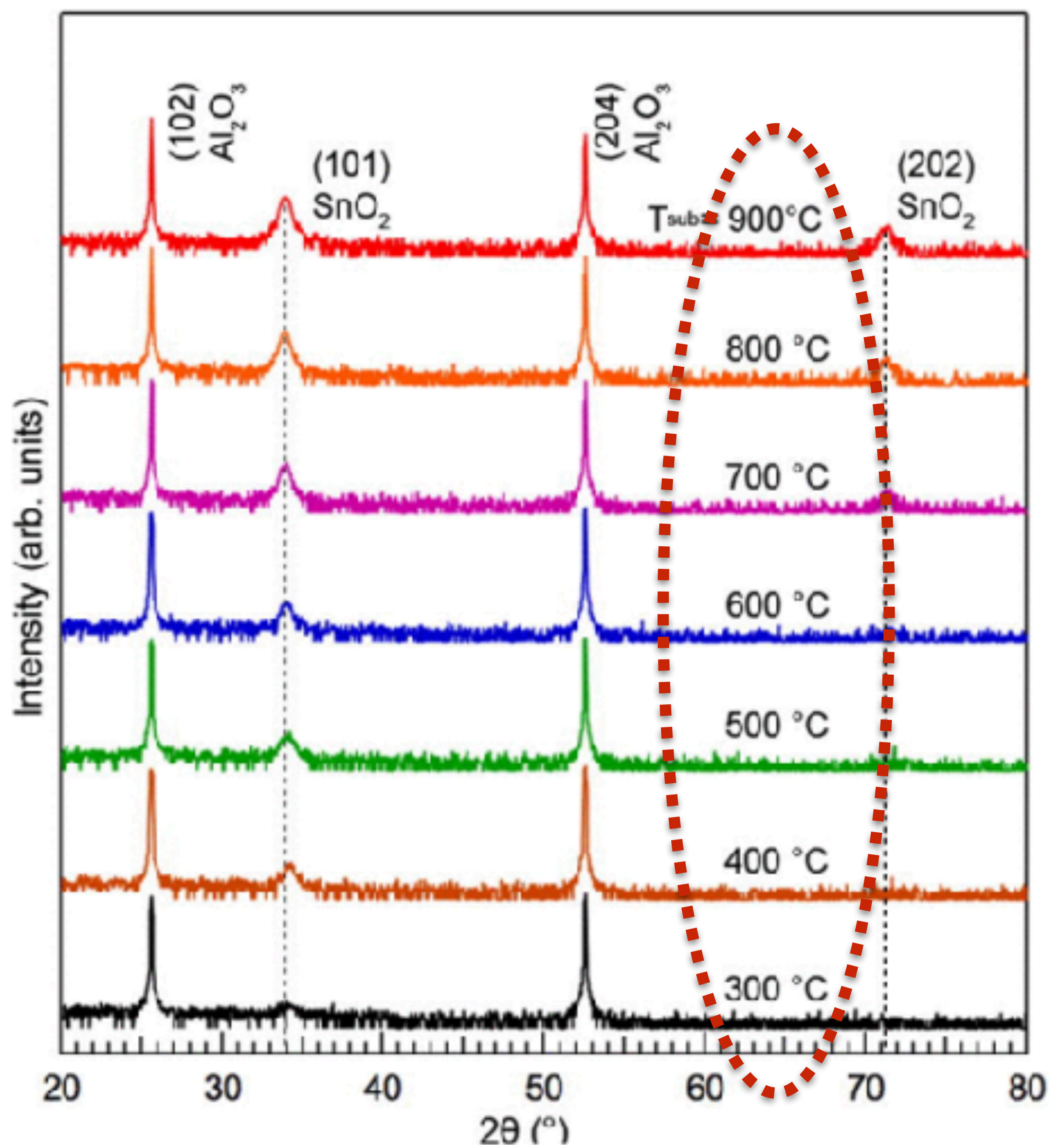


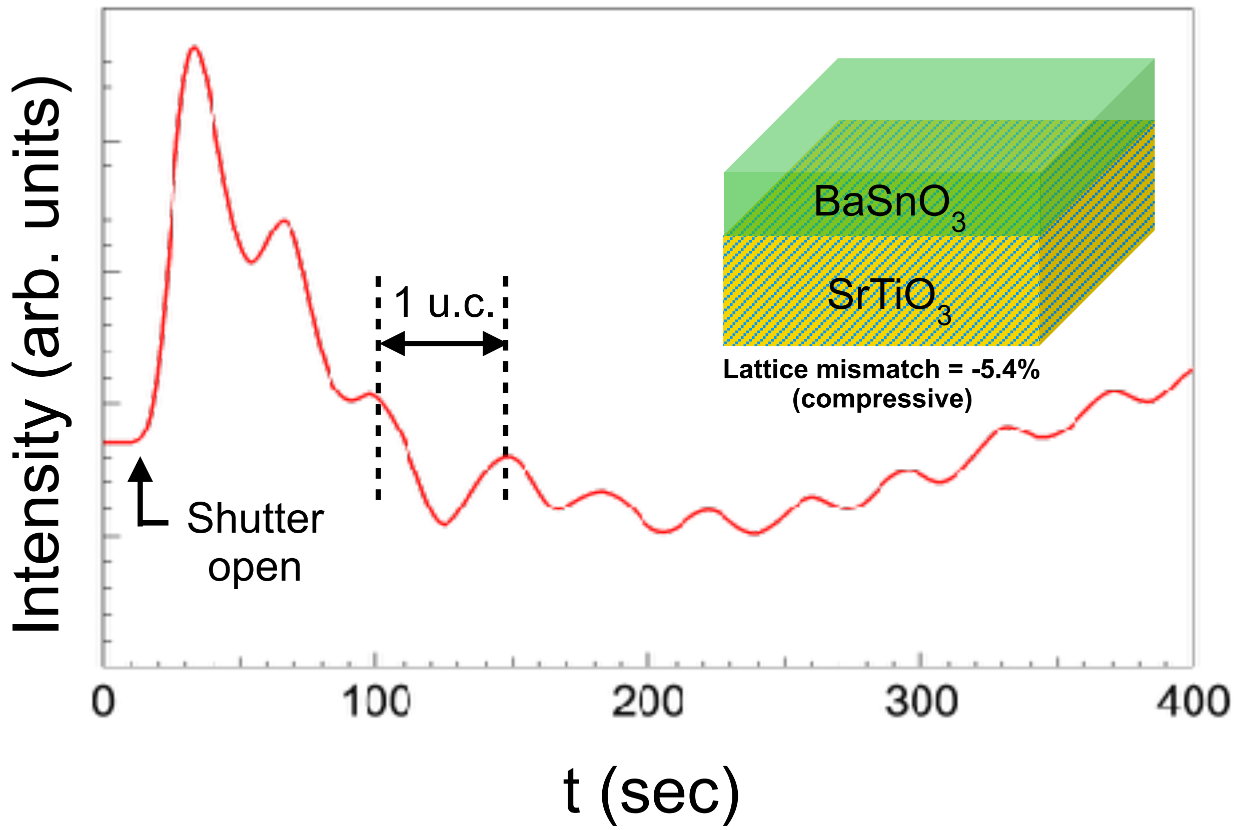
Demonstration for Low Temperature Synthesis of High Quality SnO_2 Films



T. Wang, A. Prakash, E. Warner, W. L. Gladfelter, and B. Jalan, J. Vac. Sci. Technol. A 33, 020606 (2015).

Growth Modes and Strain Relaxation

In-Situ Reflection High Energy Electron Diffraction (RHEED)



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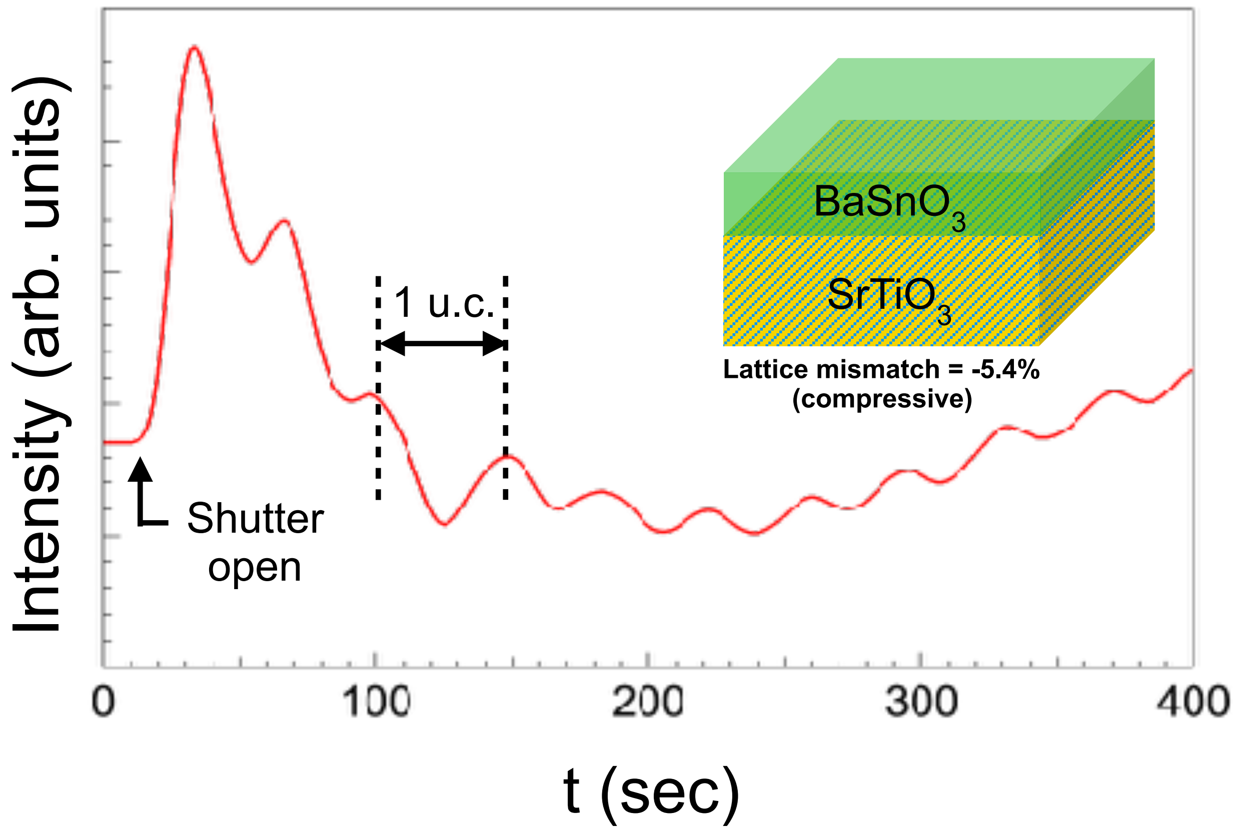
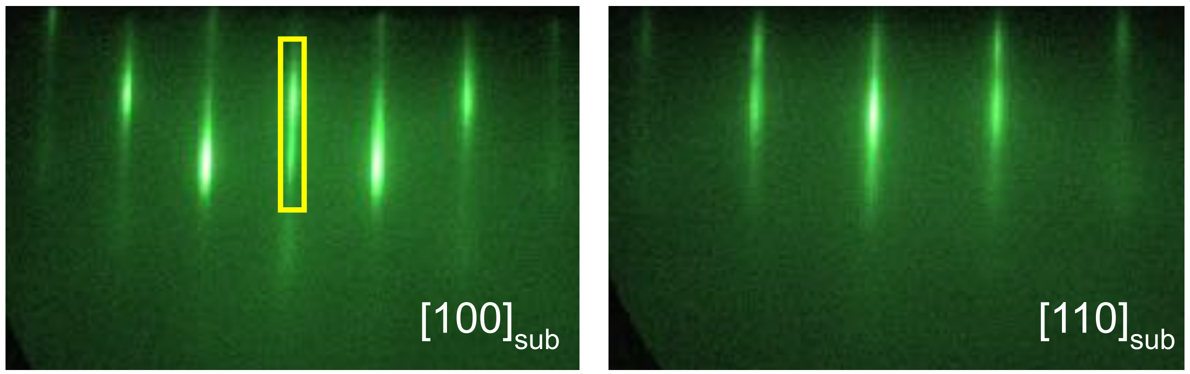
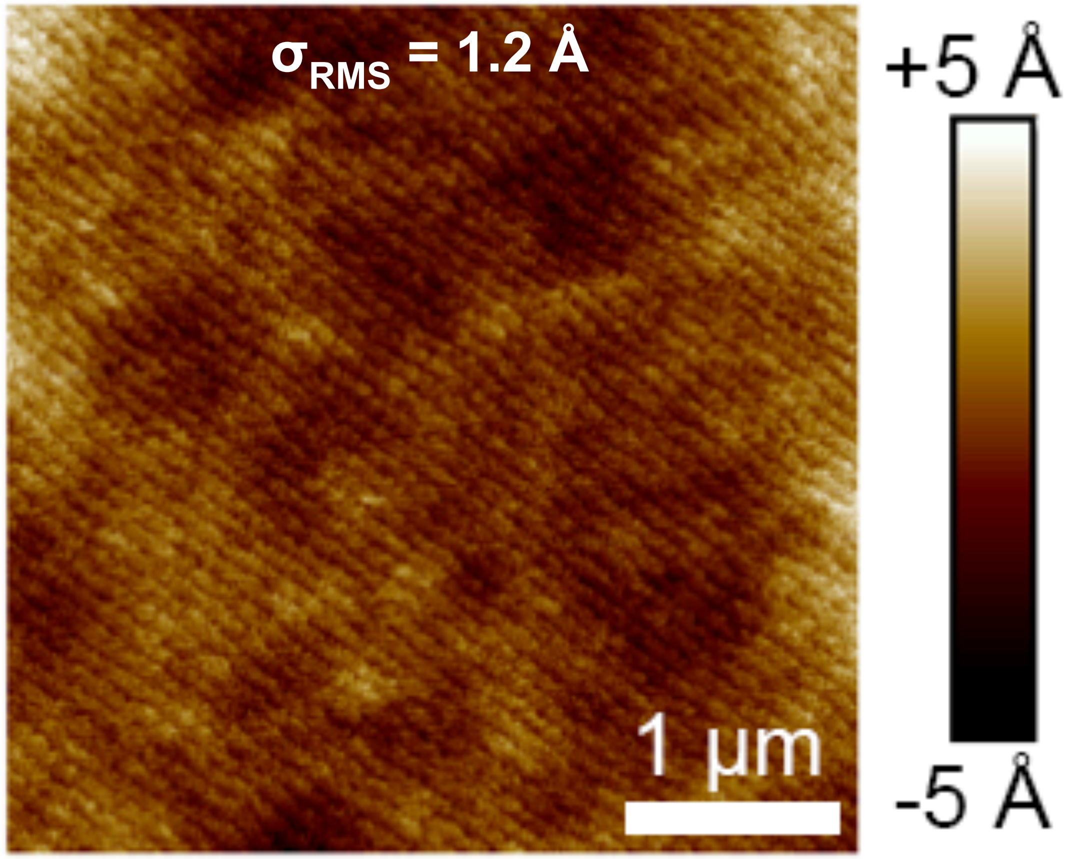


film after growth



AFM



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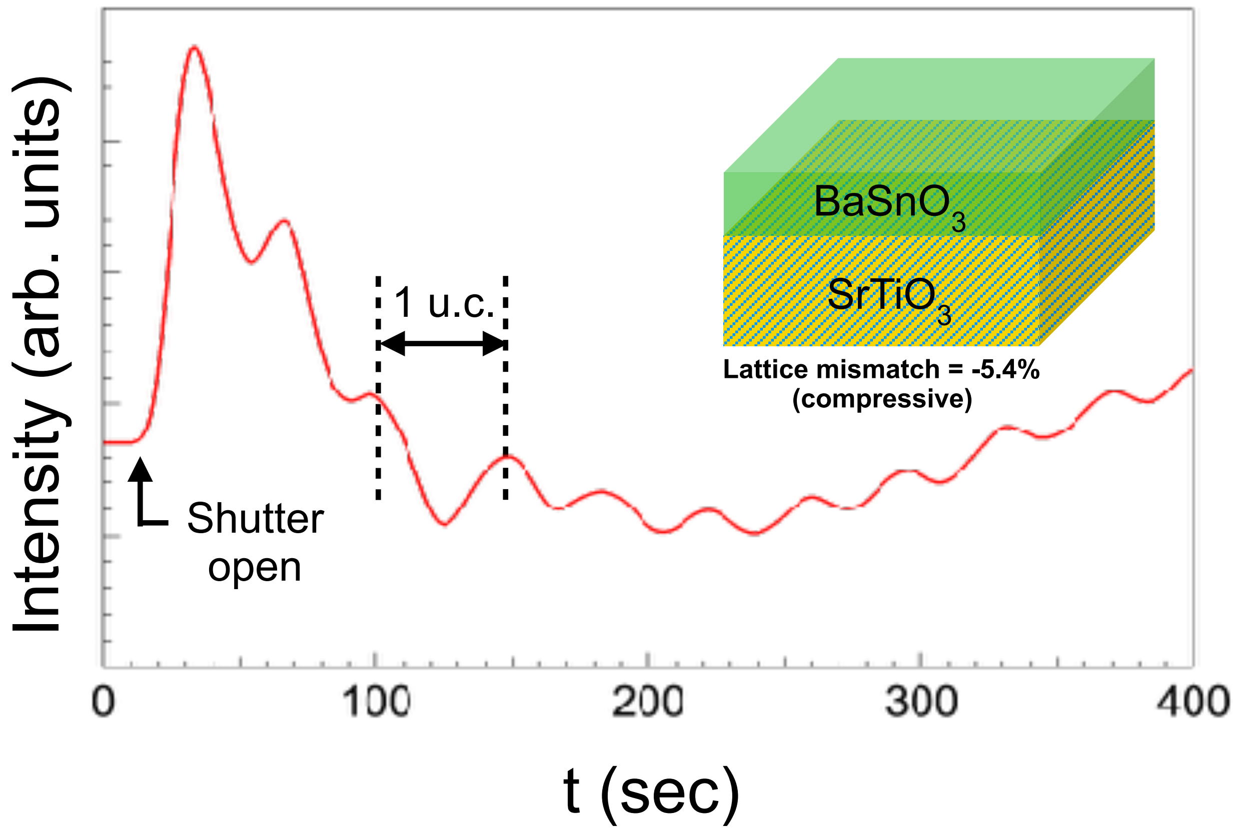
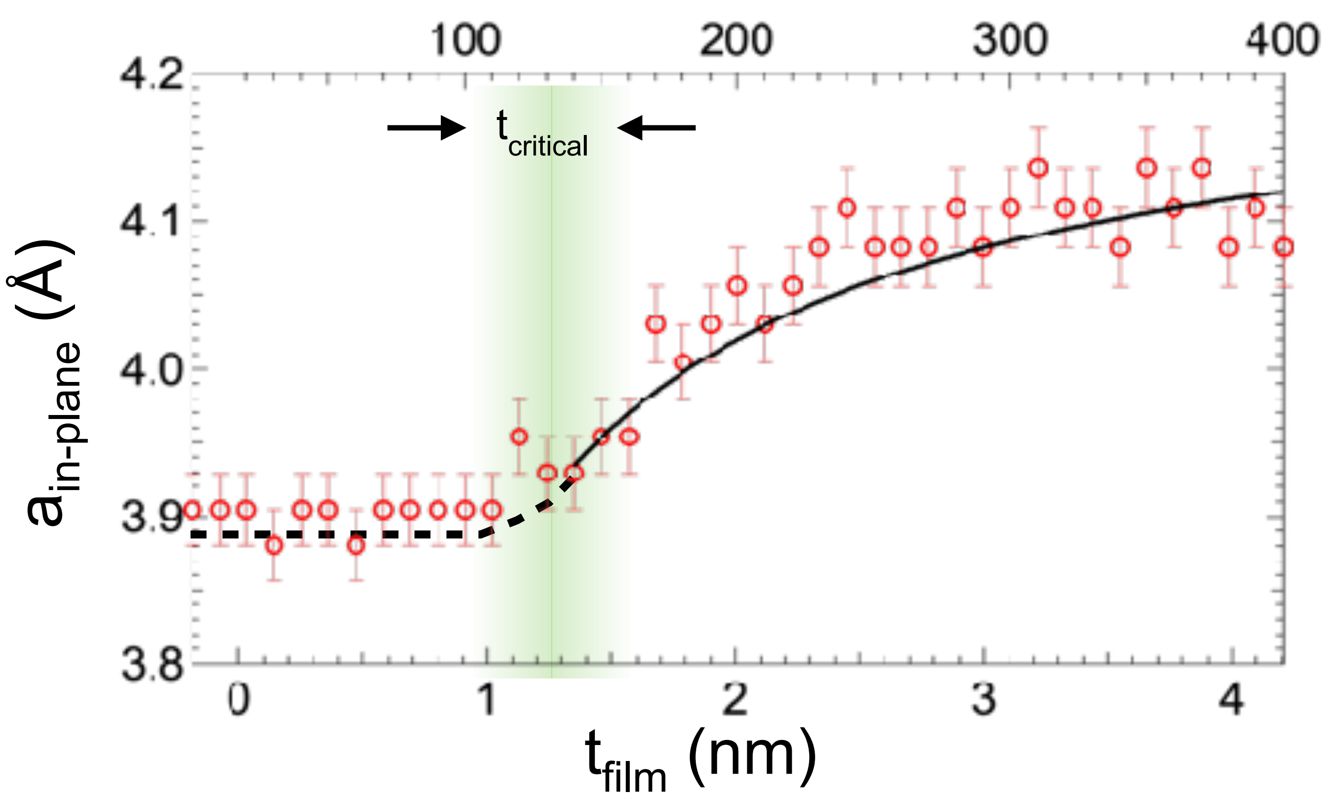
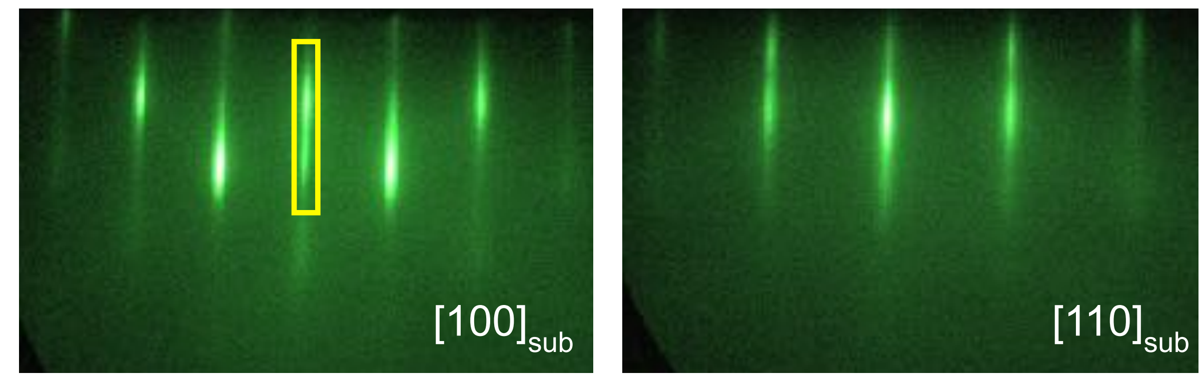
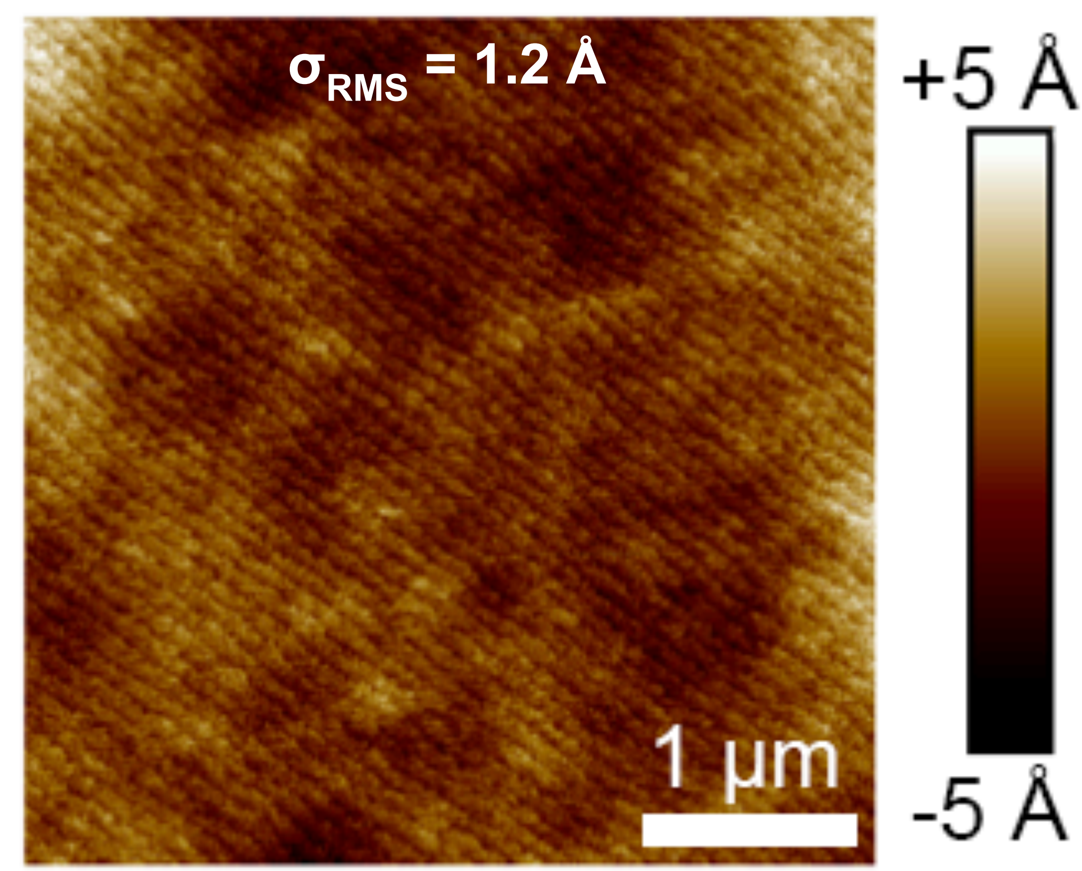




film after growth



AFM



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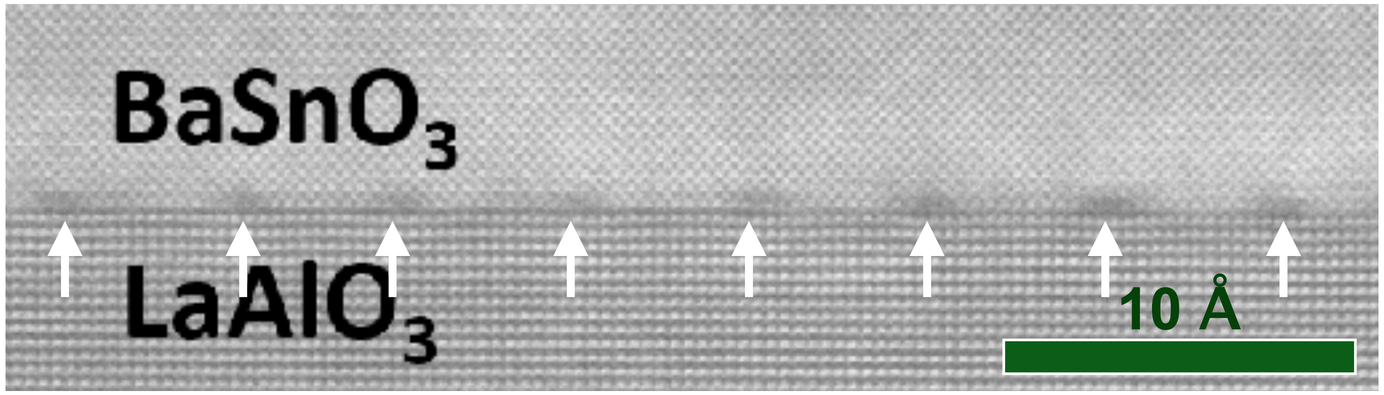
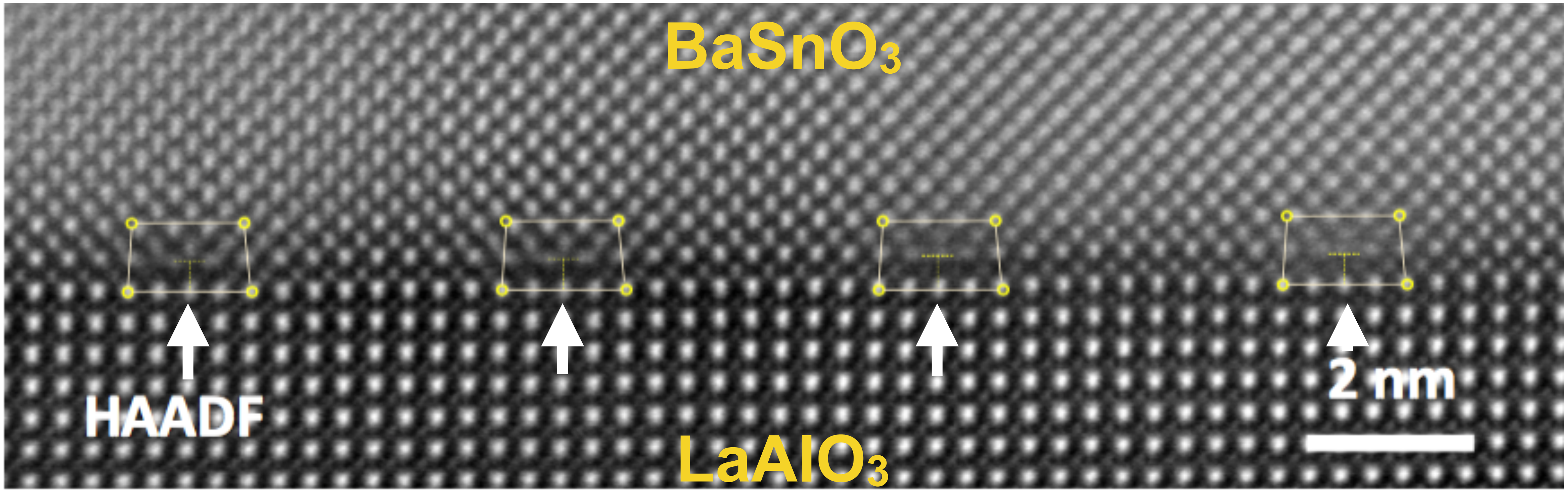
Strain Relaxation via Misfit Dislocation

HAADF Scanning TEM



Lattice mismatch = - 8%



- Strain relaxation due to misfit/threading dislocations
- Perovskite structure with cube-on-cube epitaxial relationship

STEM in collaboration with Mkhoyan Group, UMN

Strain Relaxation via Misfit Dislocation

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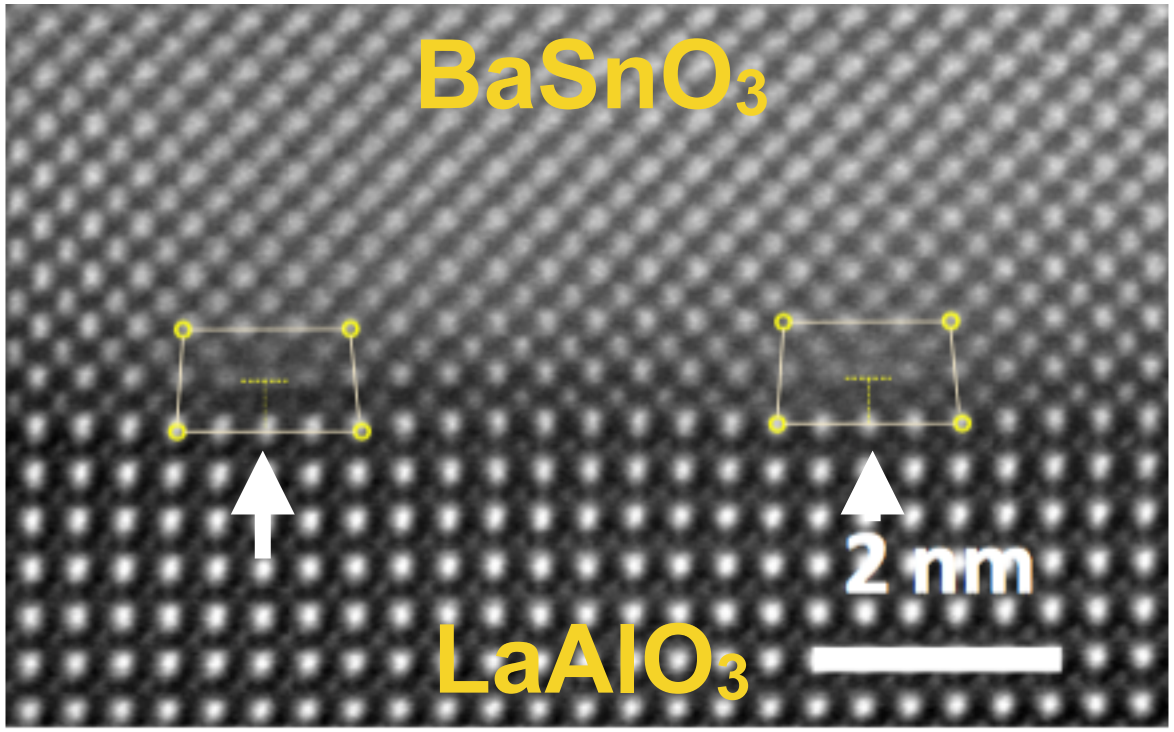
HAADF Scanning TEM



STEM in collaboration with Mkhoyan Group, UMN

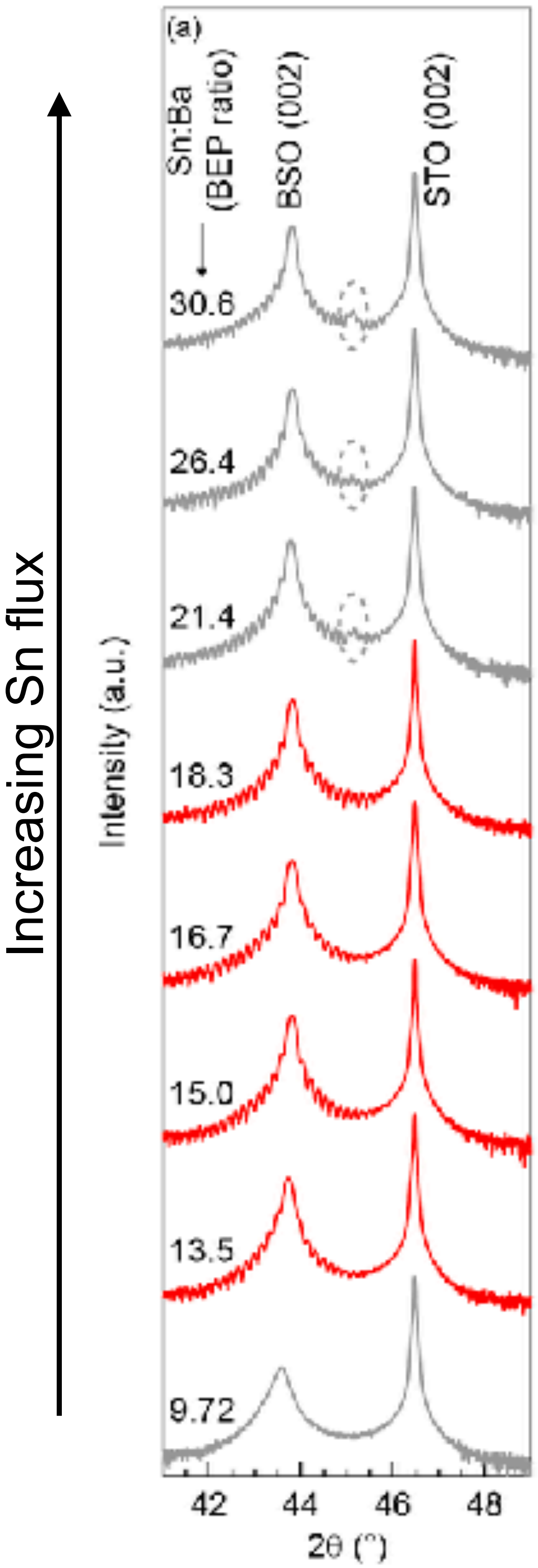
So far…

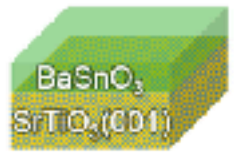
- ✓ **Phase-pure, epitaxial film on $SrTiO_3$ (001)**
- ✓ **Films grow in a layer-by-layer fashion**
- ✓ **Strain relaxation via misfit dislocation**
- ➡ **Film cation stoichiometry ??**

  I. **Lattice parameter measurements**
  II. **Rutherford backscattering spectroscopy**
  III. **Electrical transport**
  IV. **Thermal conductivity**

- ✦ Strain relaxation due to misfit/threading dislocations
- ✦ Perovskite structure with cube-on-cube epitaxial relationship

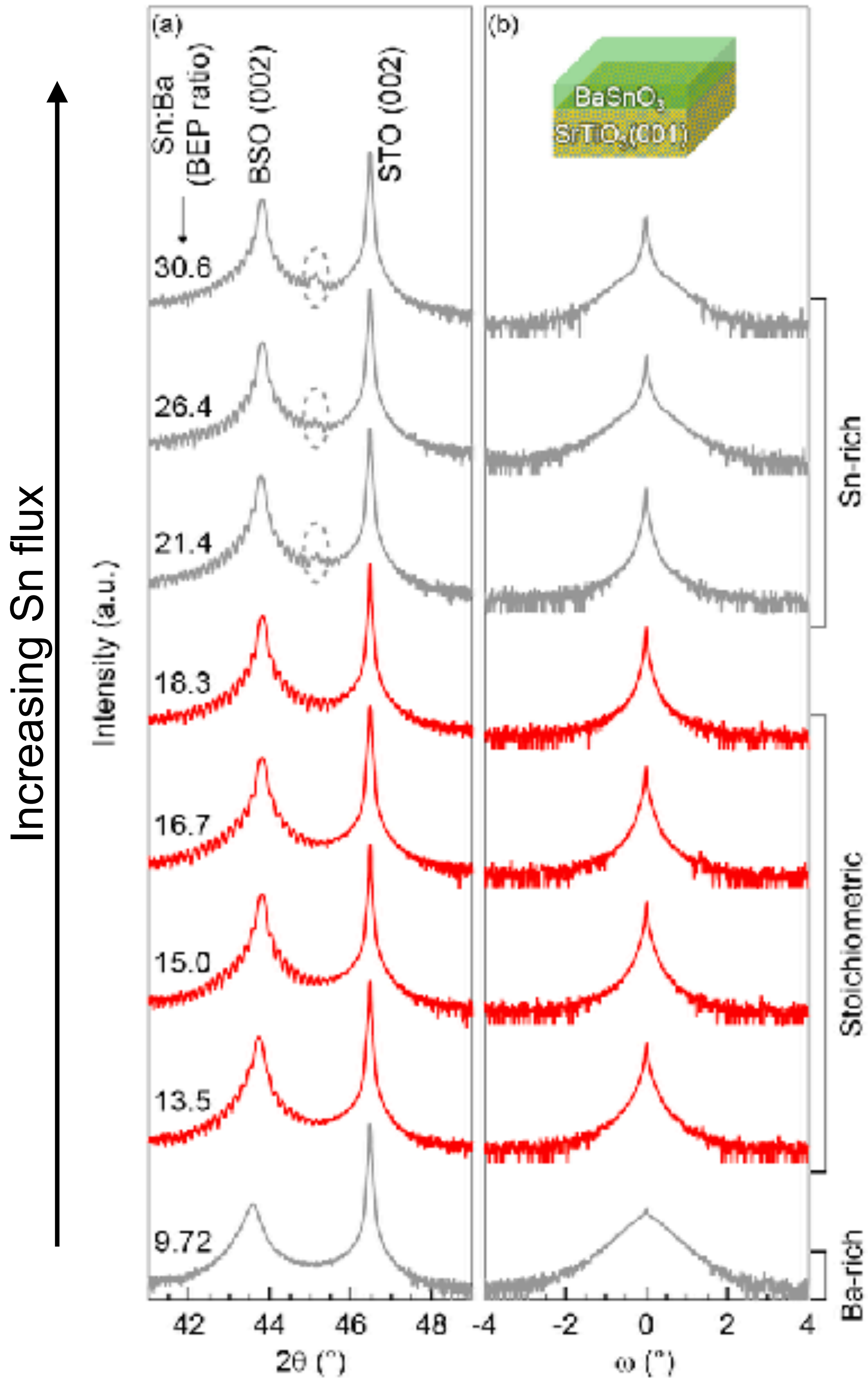
Stoichiometry Optimization: I





A. Prakash, P. Xu, X. Wu, G. Haugstad, X. Wang, and B. Jalan, *J. Mater. Chem. C* (2017) DOI: 10.1039/C7TC00190H

Stoichiometry Optimization: I



A. Prakash, P. Xu, X. Wu, G. Haugstad, X. Wang, and B. Jalan, *J. Mater. Chem. C* (2017) DOI: 10.1039/C7TC00190H

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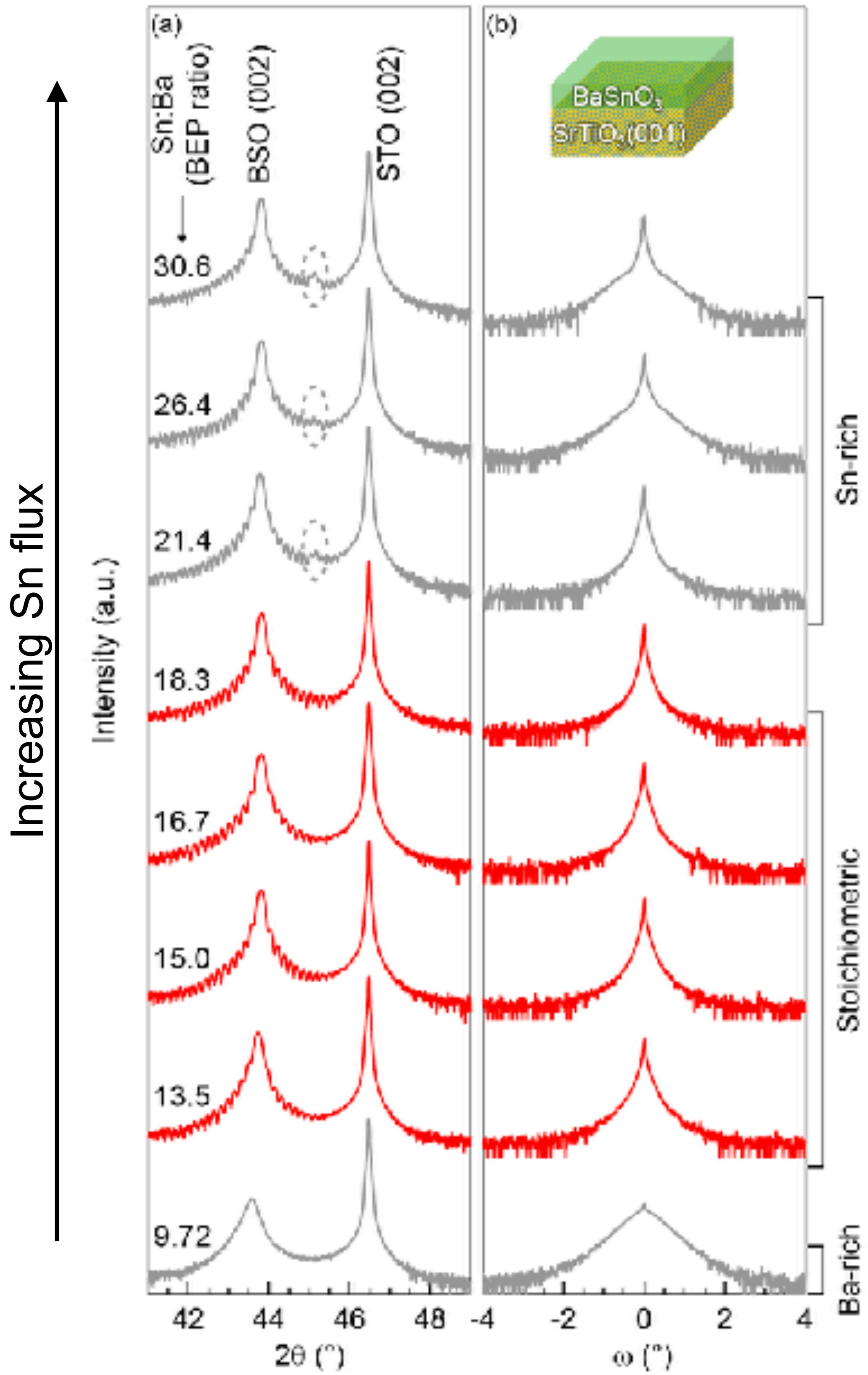
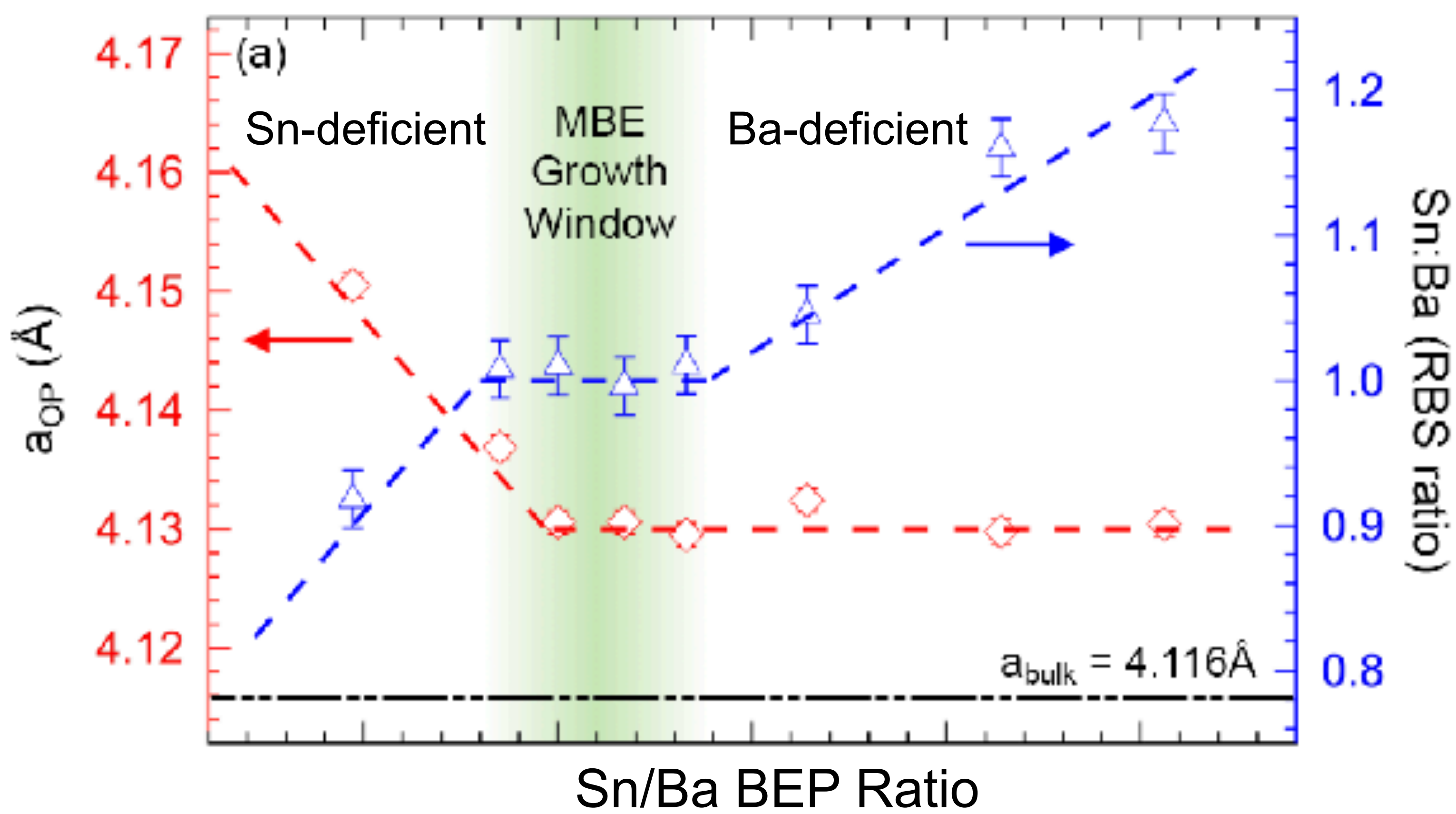


Adsorption-Controlled Growth



- Additional diffraction peak for Sn-rich films
- Lattice parameter increases for Ba-rich and remains unchang for stoichiometric and Sn-rich films.
- RBS confirms "MBE growth window" i.e. for a range of Sn:Ba flux ratio, cation stoichiometry is *self-regulating*.

A. Prakash, P. Xu, X. Wu, G. Haugstad, X. Wang, and B. Jalan, *J. Mater. Chem. C* (2017) DOI: 10.1039/C7TC00190H

Stoichiometry Optimization: II

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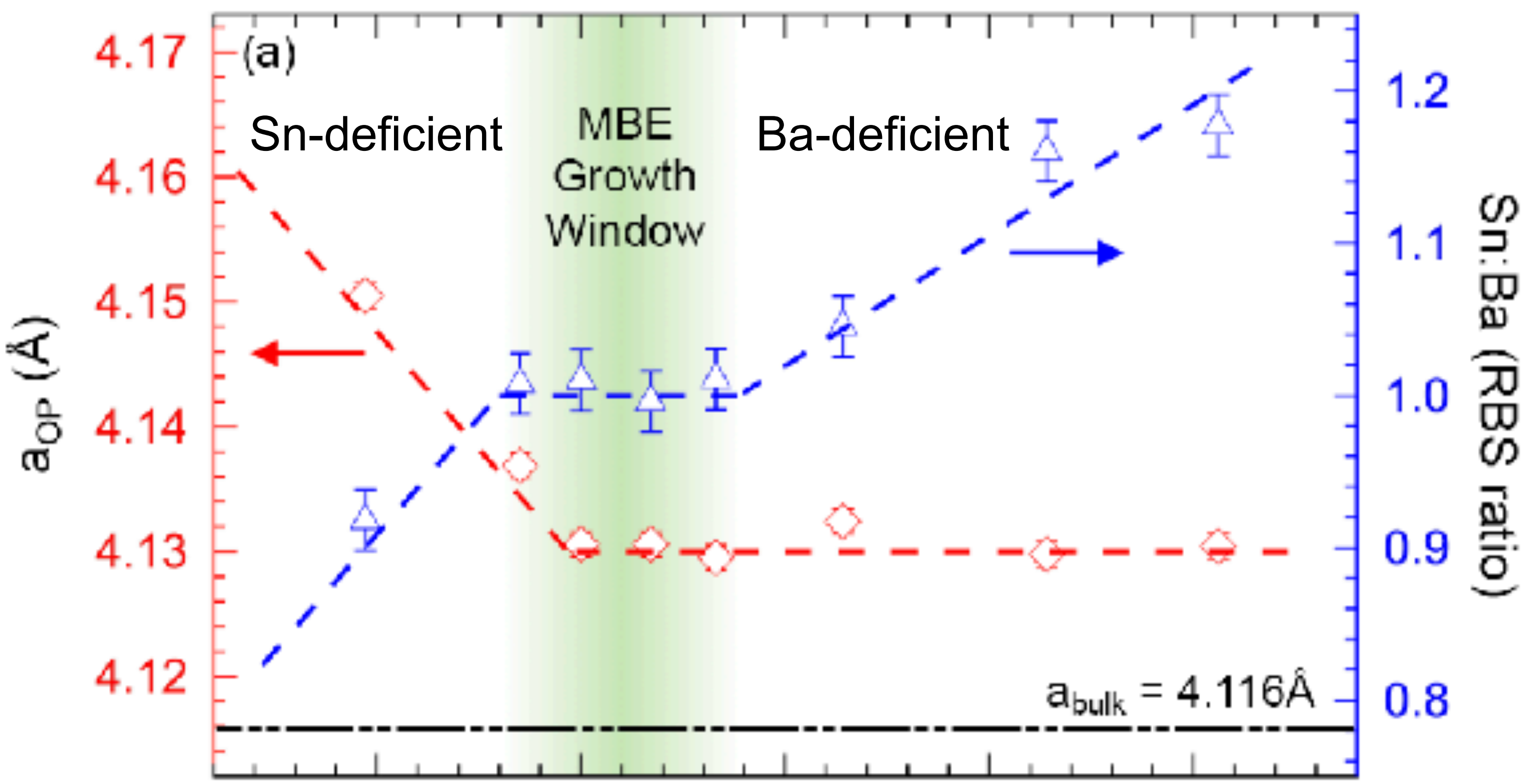
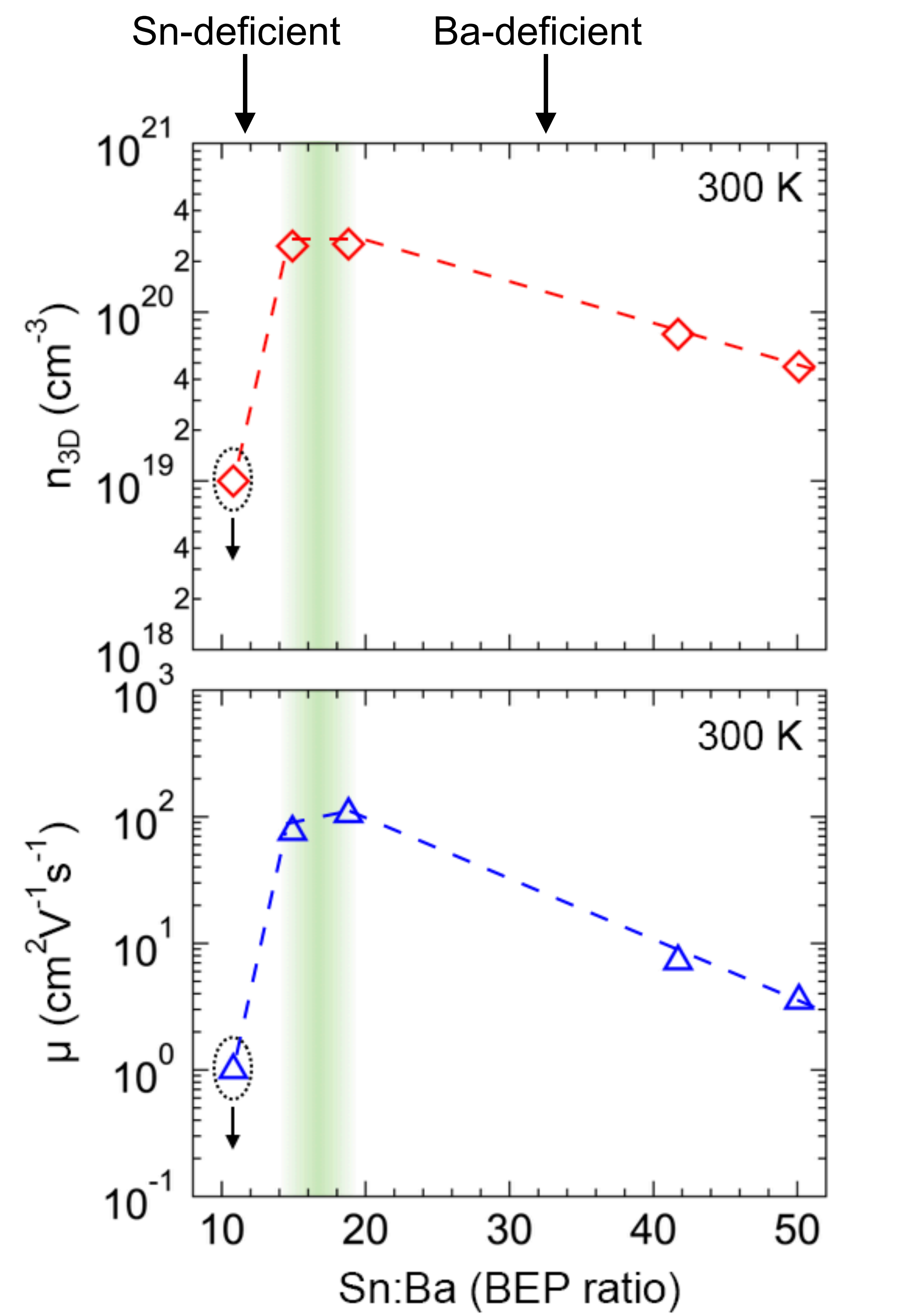




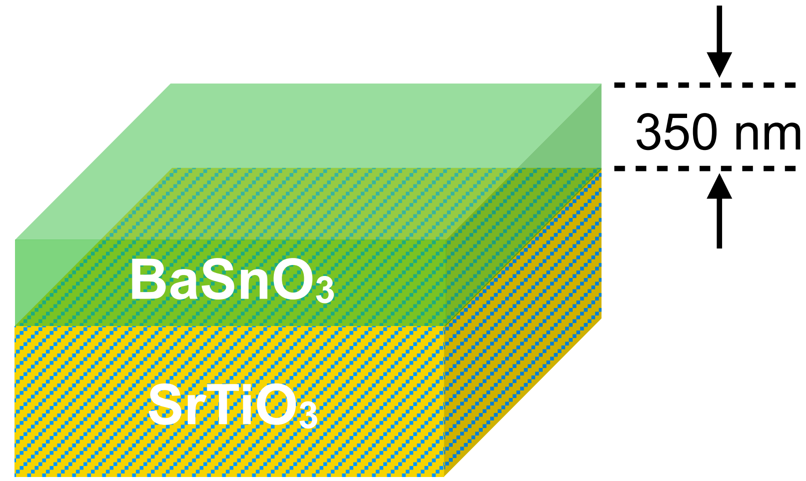
- Carrier density and mobility remains higher within the growth window.
- Both Sn- and Ba-vacancies act as acceptor-like defects

A. Prakash, P. Xu, X. Wu, G. Haugstad, X. Wang, and B. Jalan, J. Mater. Chem. C, (2017) DOI: 10.1039/c7tc00190h

Stoichiometry Optimization: III

Thermal Conductivity as a Measure of Stoichiometry

Thermal conductivity measurements in collaboration with Prof. Xiaojia Wang's group, UMN



Non-Stoichiometric

Stoichiometric

Λ (bulk single crystal)* ≈ 13.2 $Wm^{-1}K^{-1}$

*H.J. Kim *et al.*, Thermochim. Acta **585**, 16 (2014)

Stoichiometry Optimization: III

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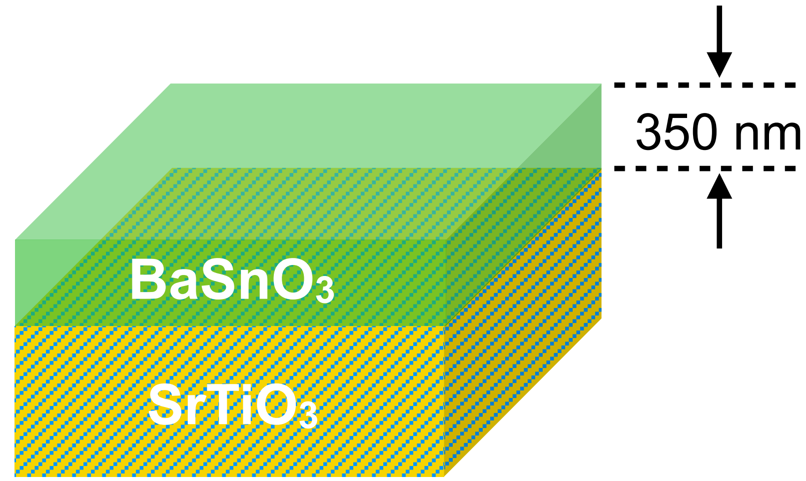

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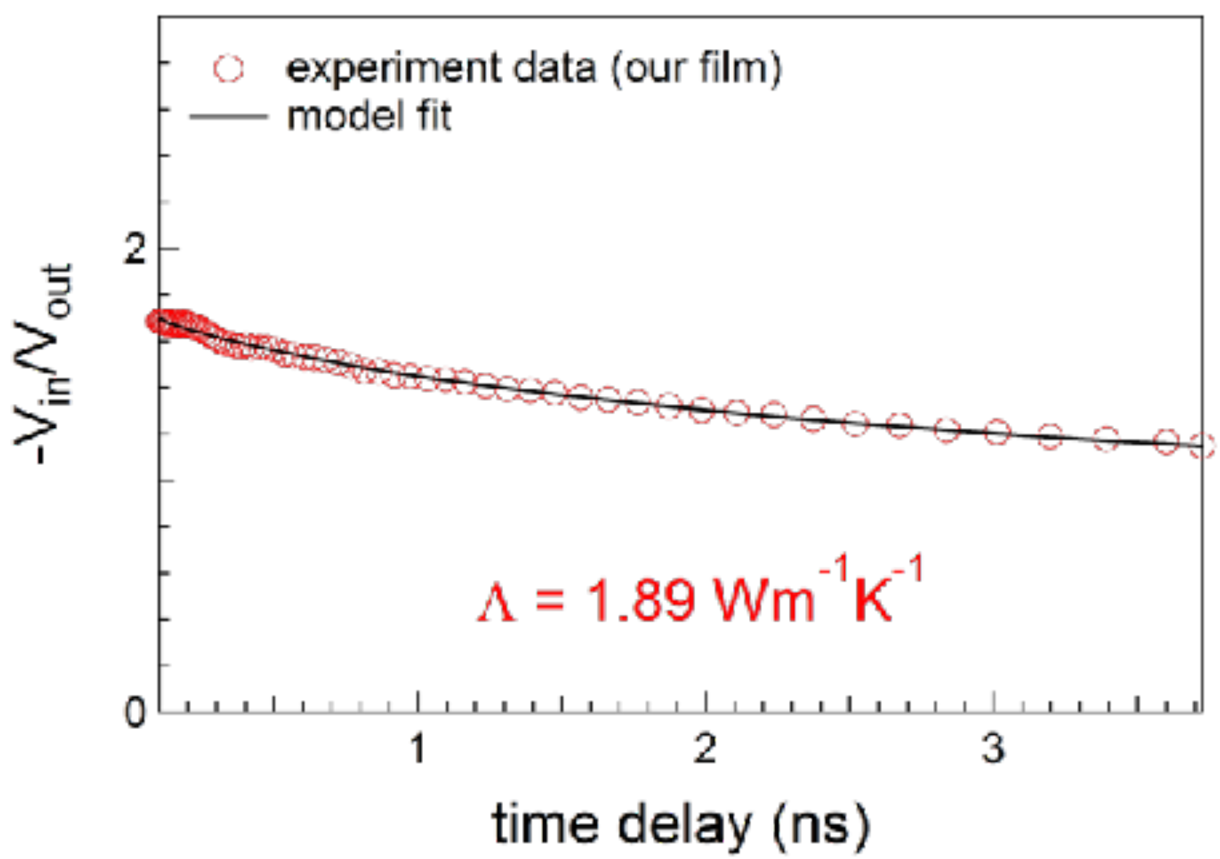

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Stoichiometry Optimization: III

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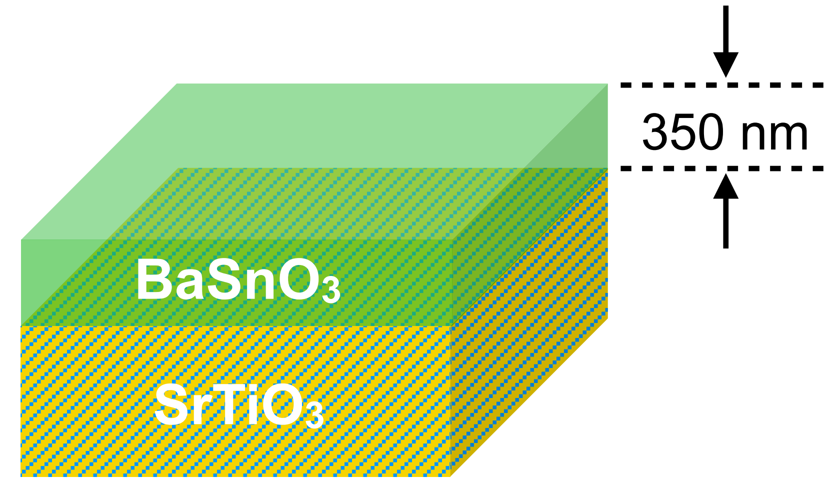

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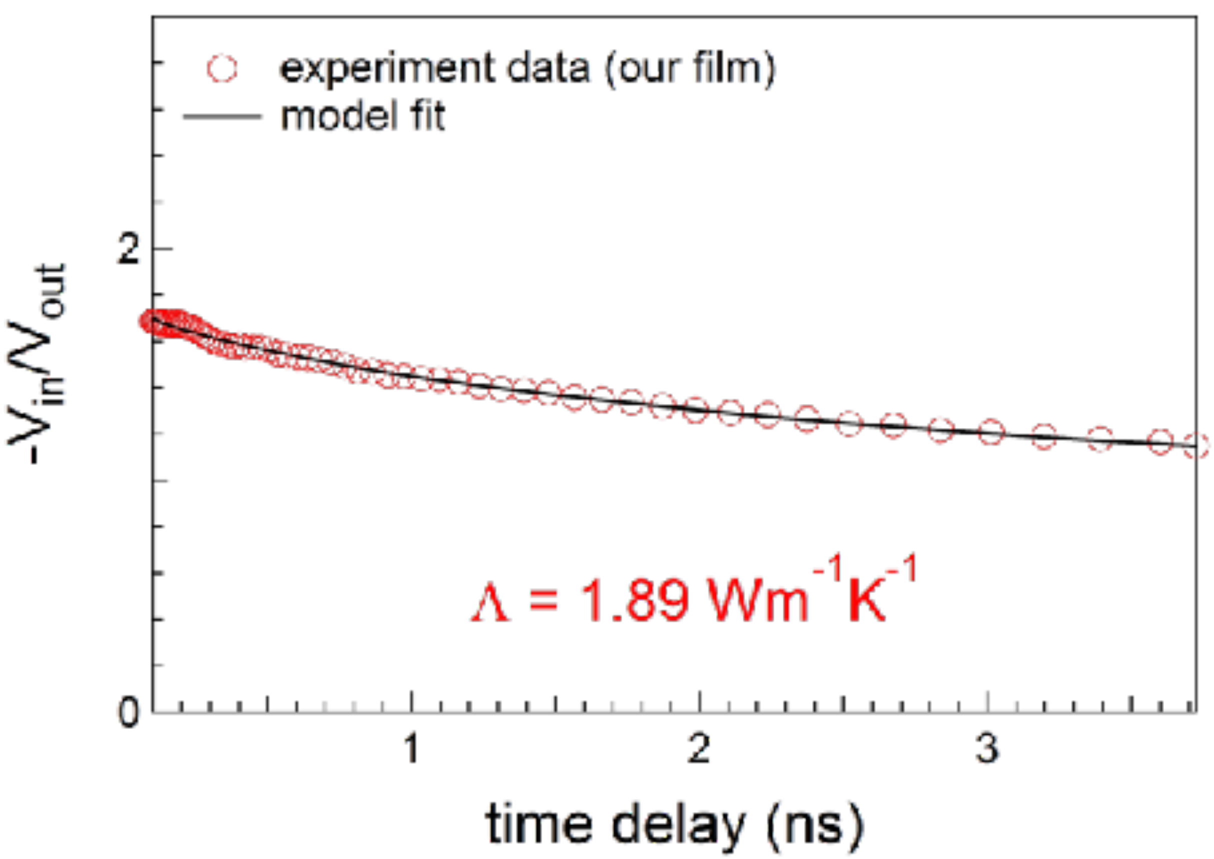
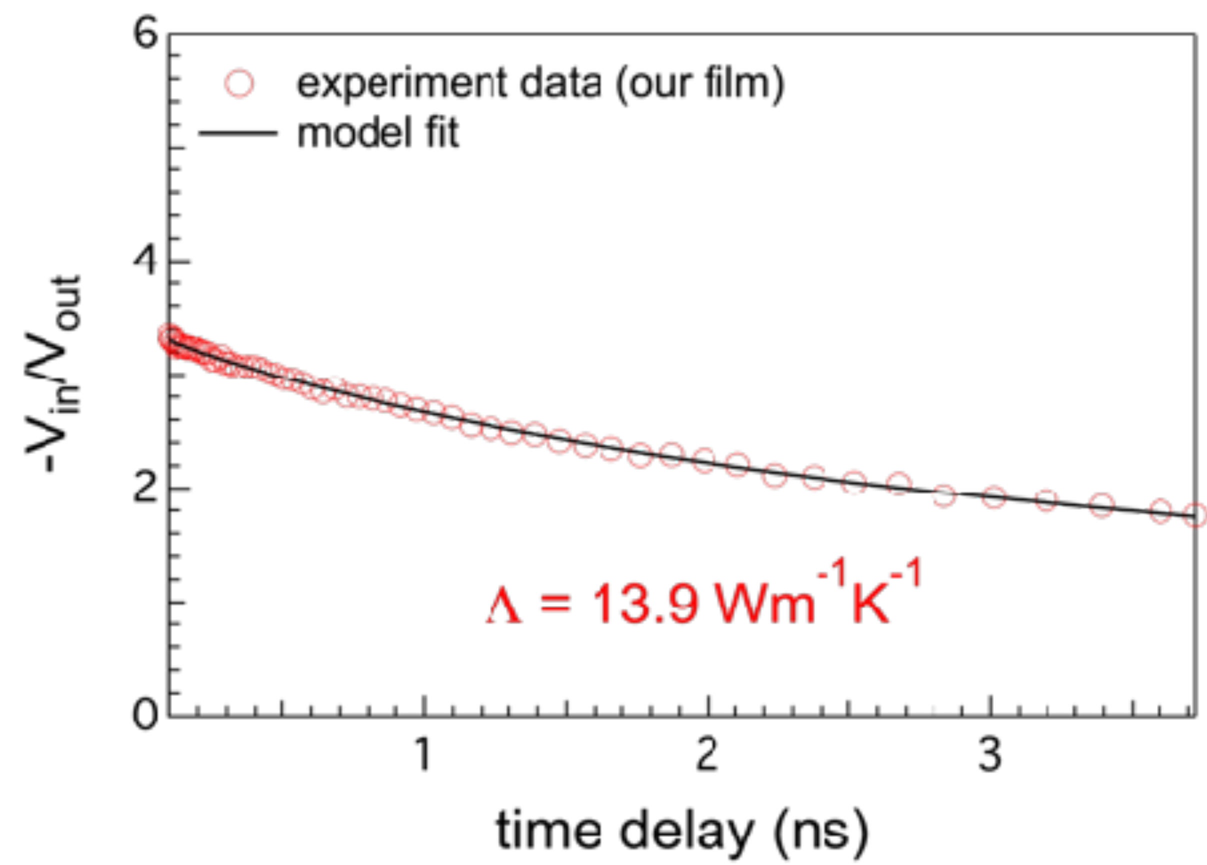


Stoichiometric



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Doping and Electronic Transport

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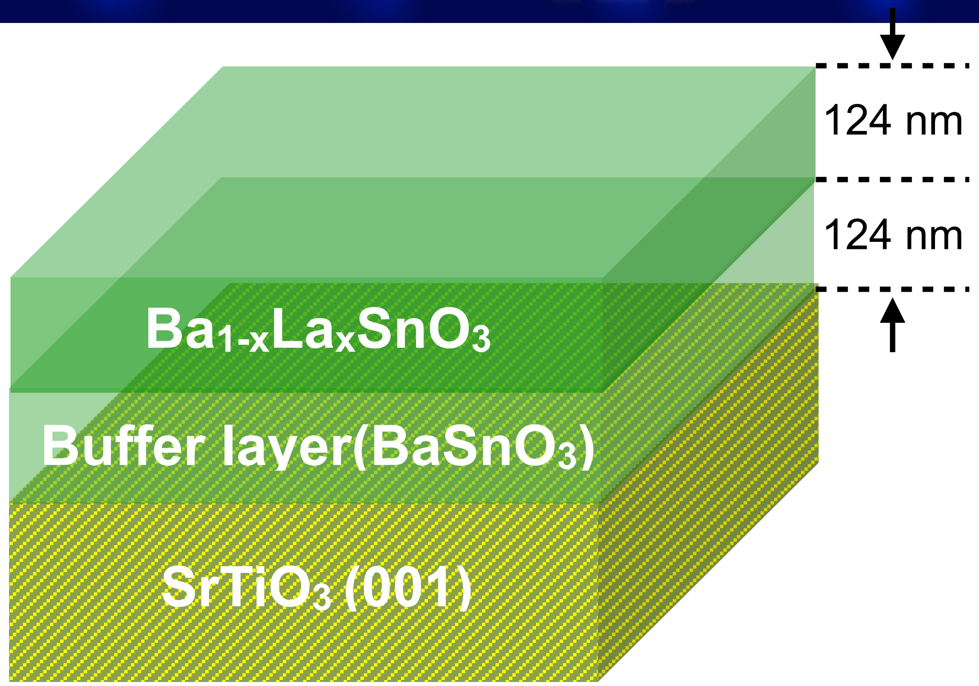

Note:
Lattice mismatch = -5.4% (compressive)

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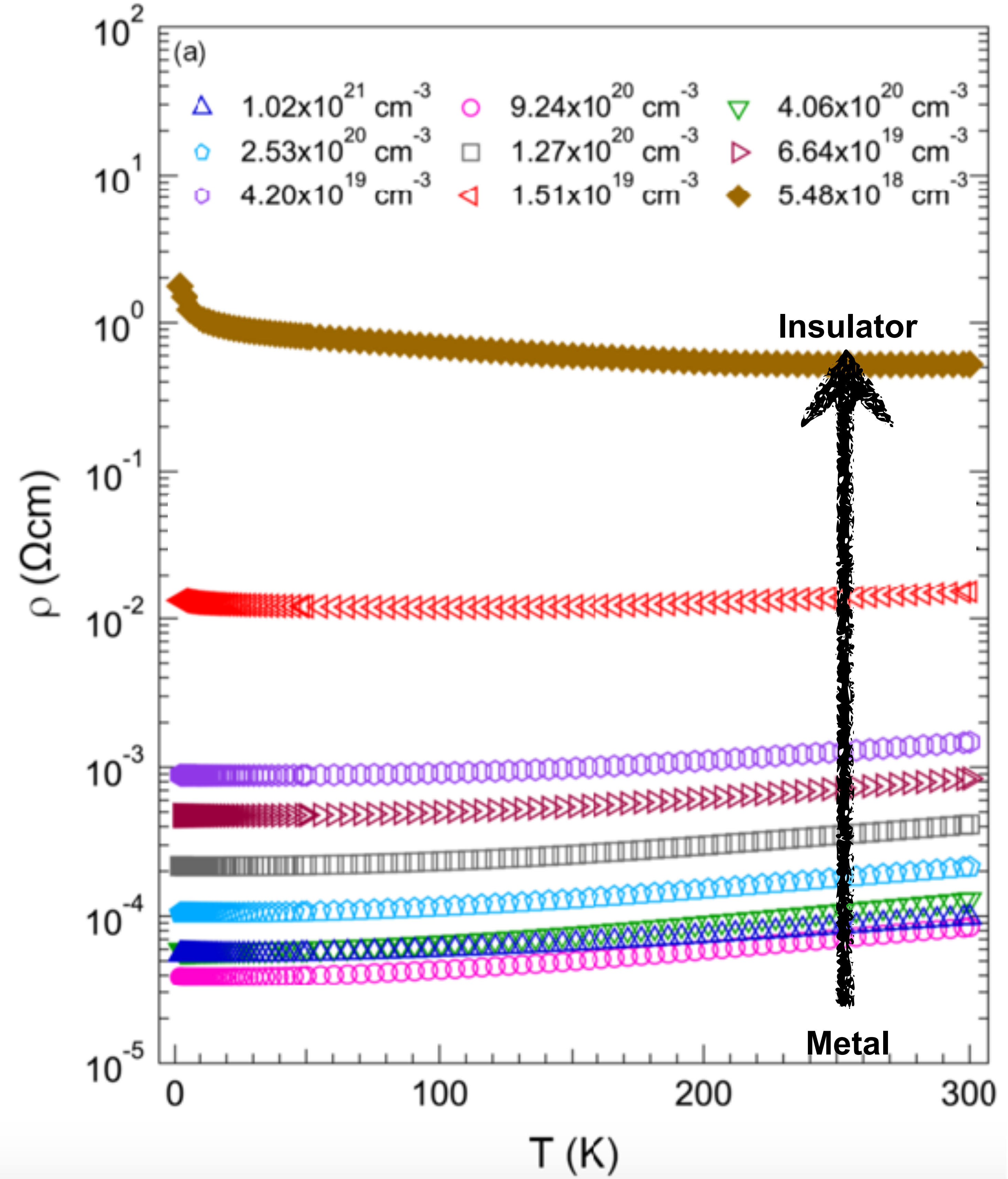

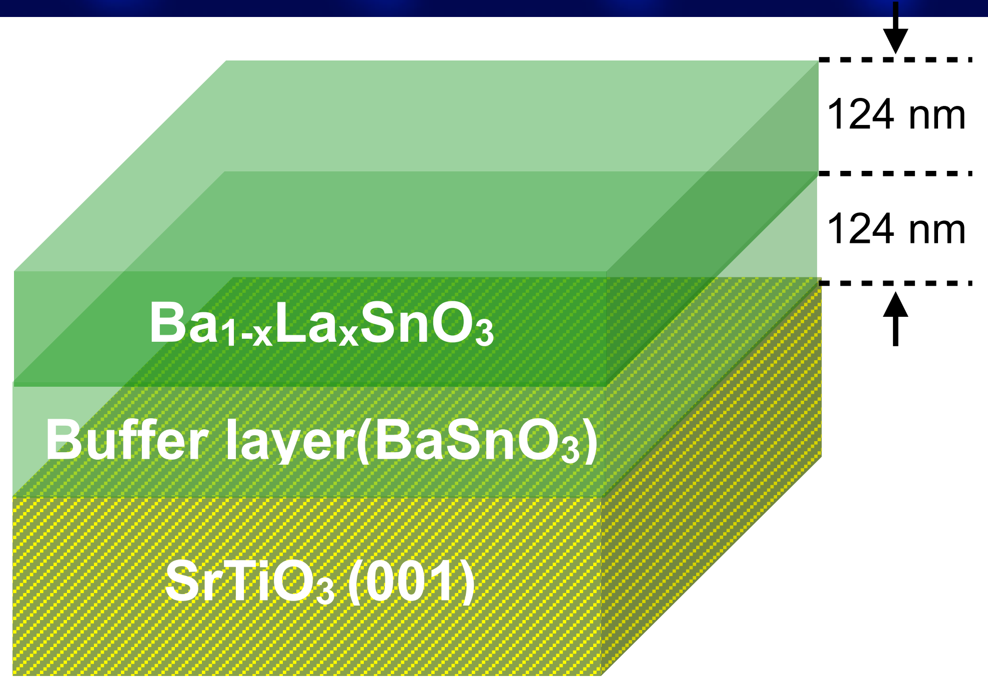

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Experimentally, M ⇒ I occurs at

$n_c = 5\times10^{18} - 1.5\times10^{19}$ cm^{-3}

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

Doping and Electronic Transport

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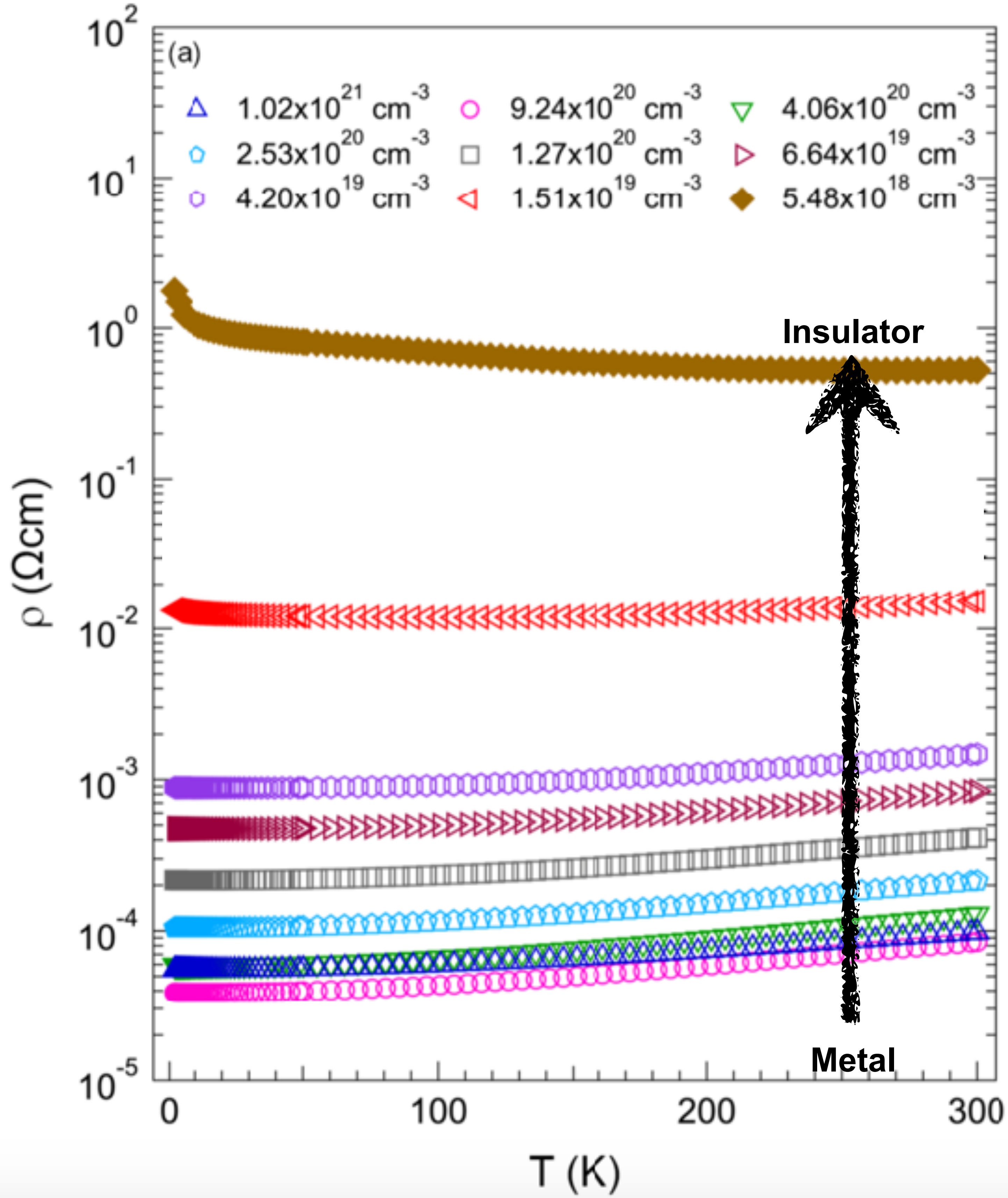

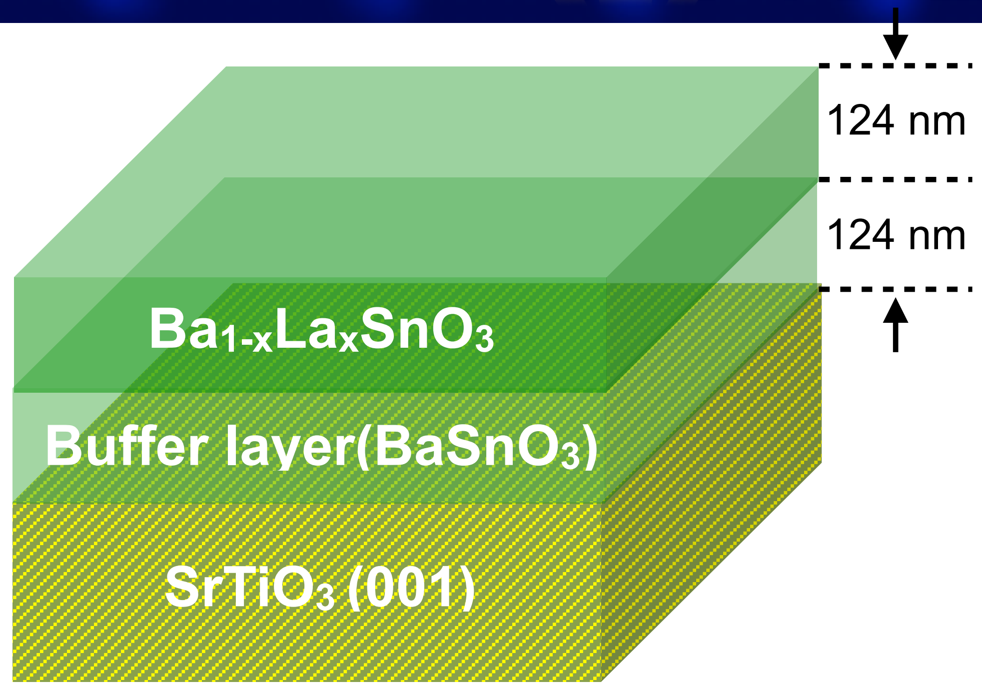

Note:
Lattice mismatch = -5.4% (compressive)

Experimentally, M ⇒ I occurs at

$n_c = 5\times10^{18} - 1.5\times10^{19}$ cm^{-3}

Calculated n_c (intrinsic M ⇒ I)

$= 10^{17} - 10^{18}$ cm^{-3}

(an order of magnitude lower)

Compensating Defects ?

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

Hall Electron Density and Mobility

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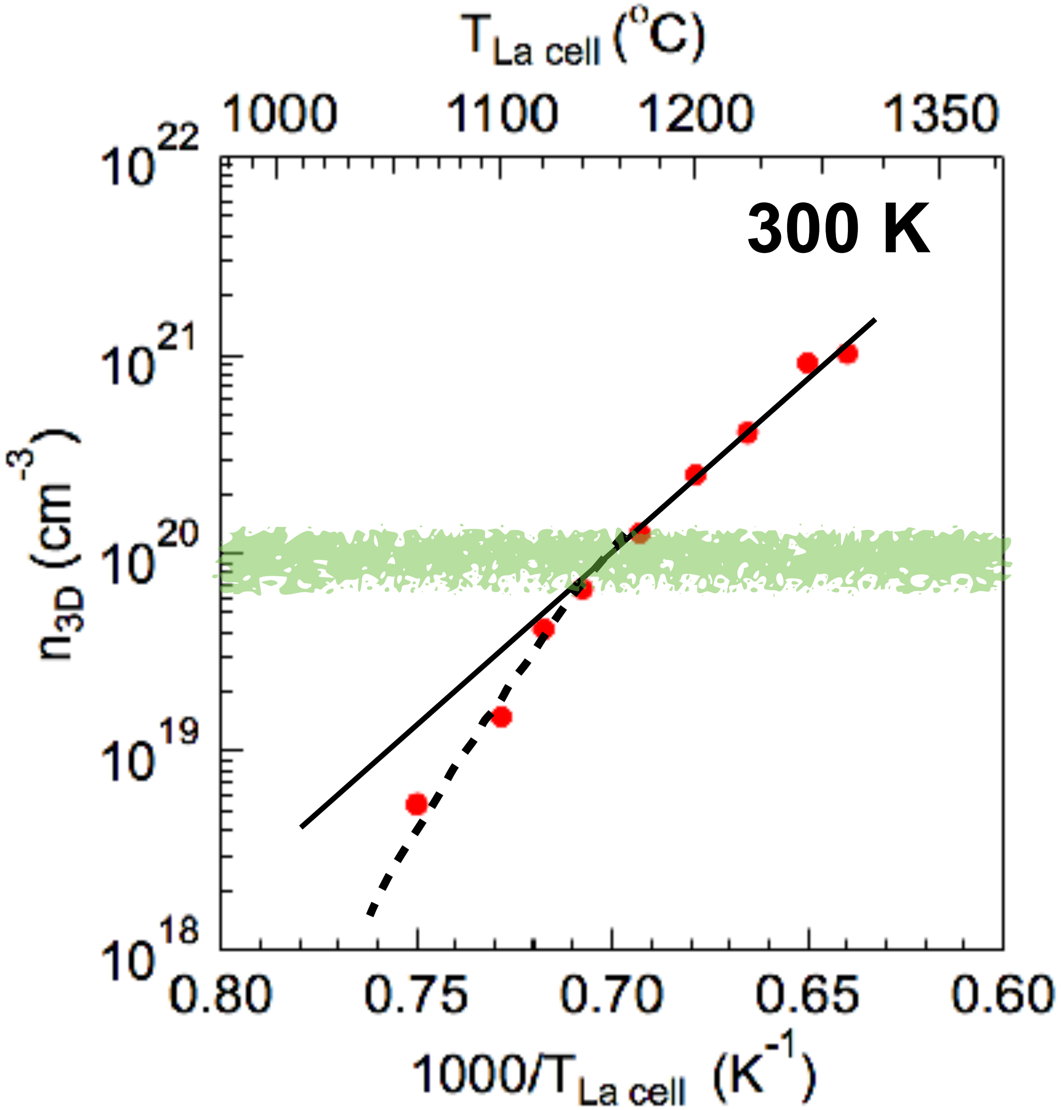

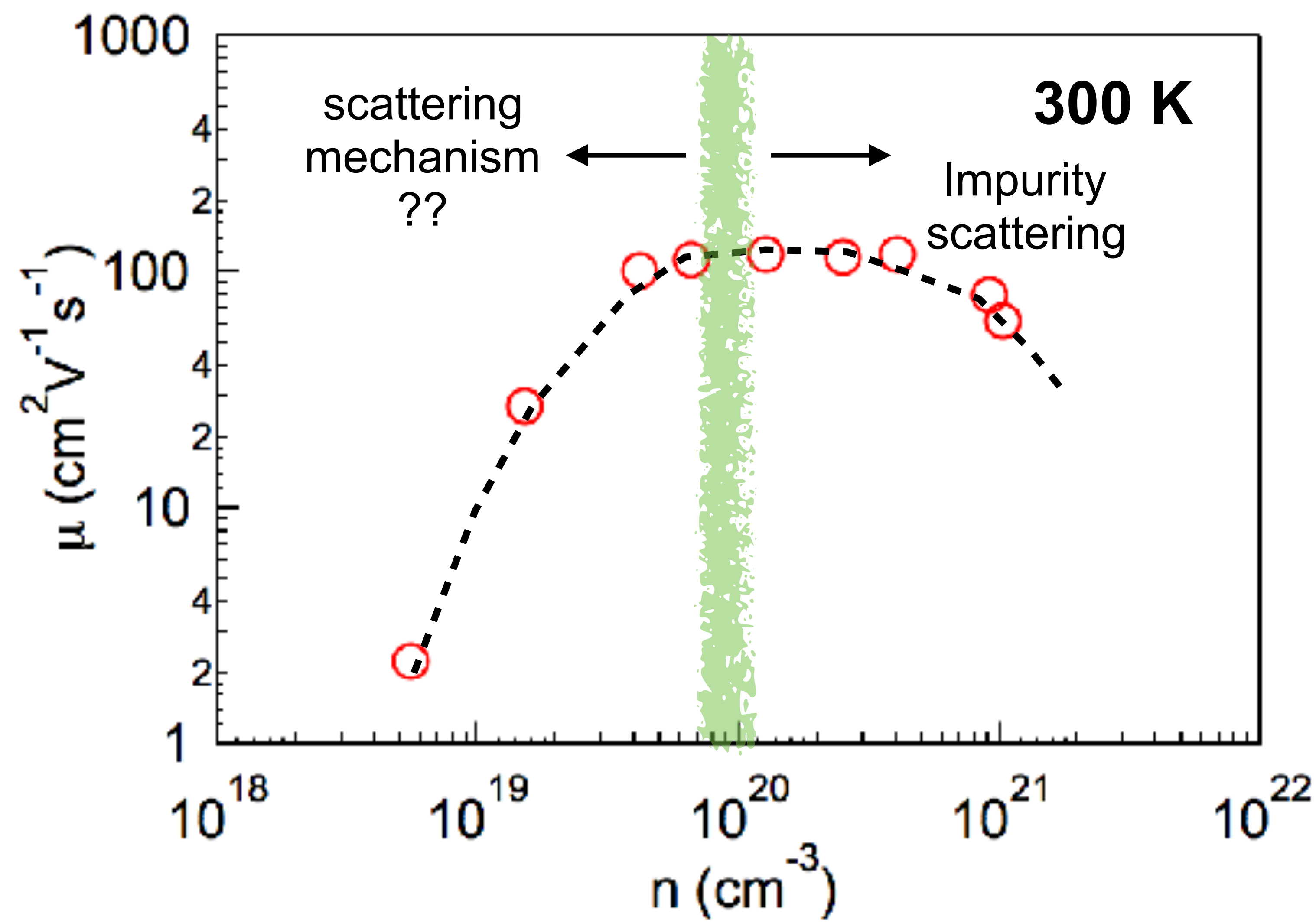

- For $n_{3d} < \sim 10^{20}$ cm^{-3}, mobility⬇ and the Hall density *deviates* from linearity (solid line)
- Indicative of scattering and compensation due to charged defects*

*J. H. You et. al., JAP **99**, 033706 (2006)

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

Hall Electron Density and Mobility

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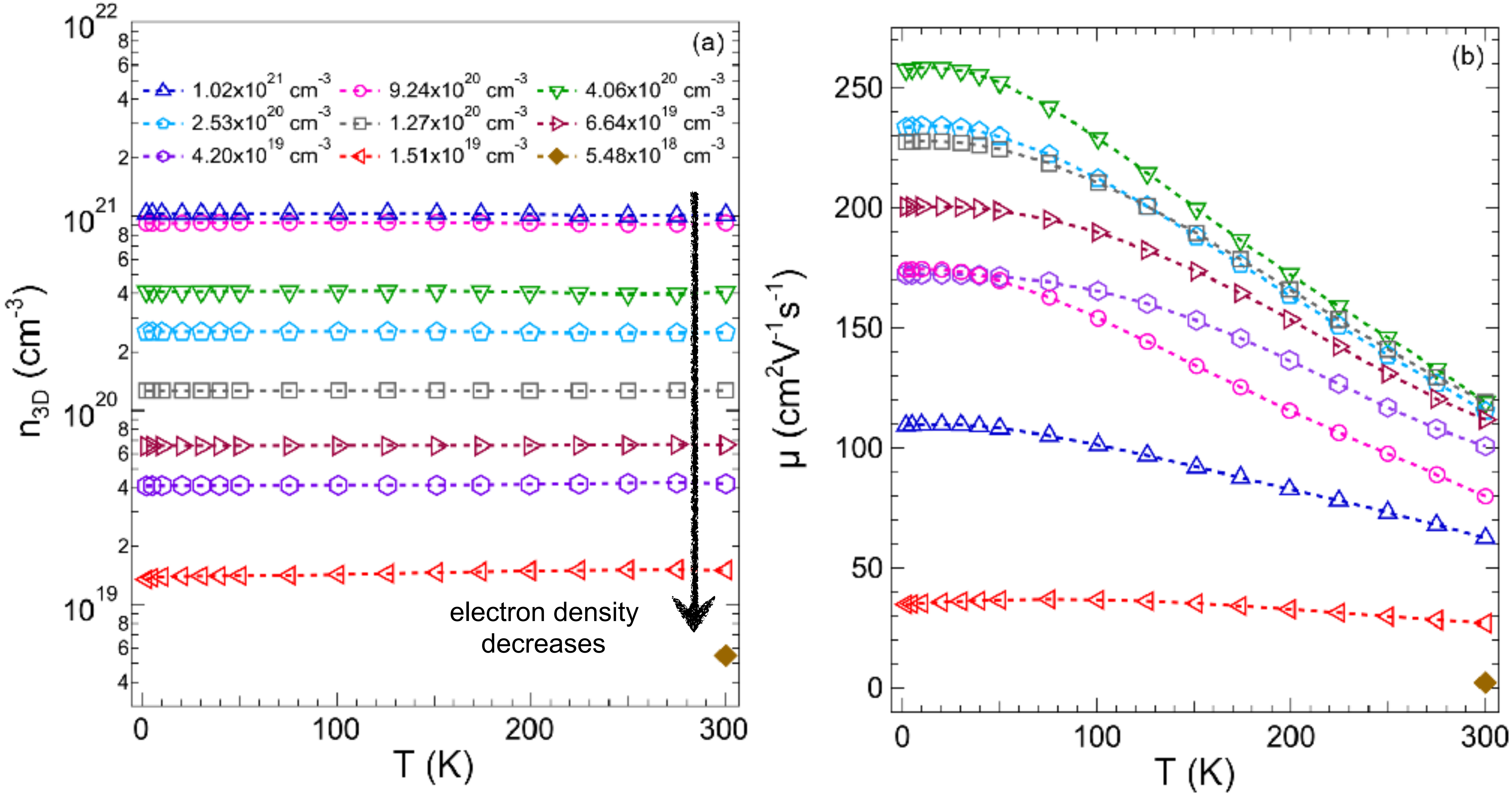
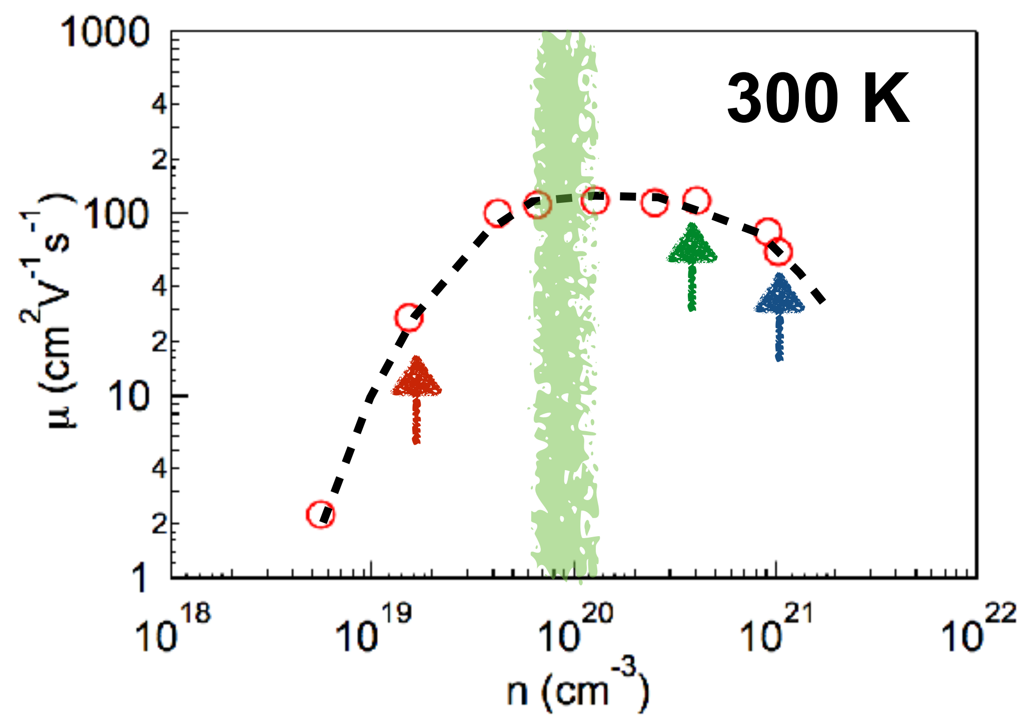
Temperature Dependent Measurements

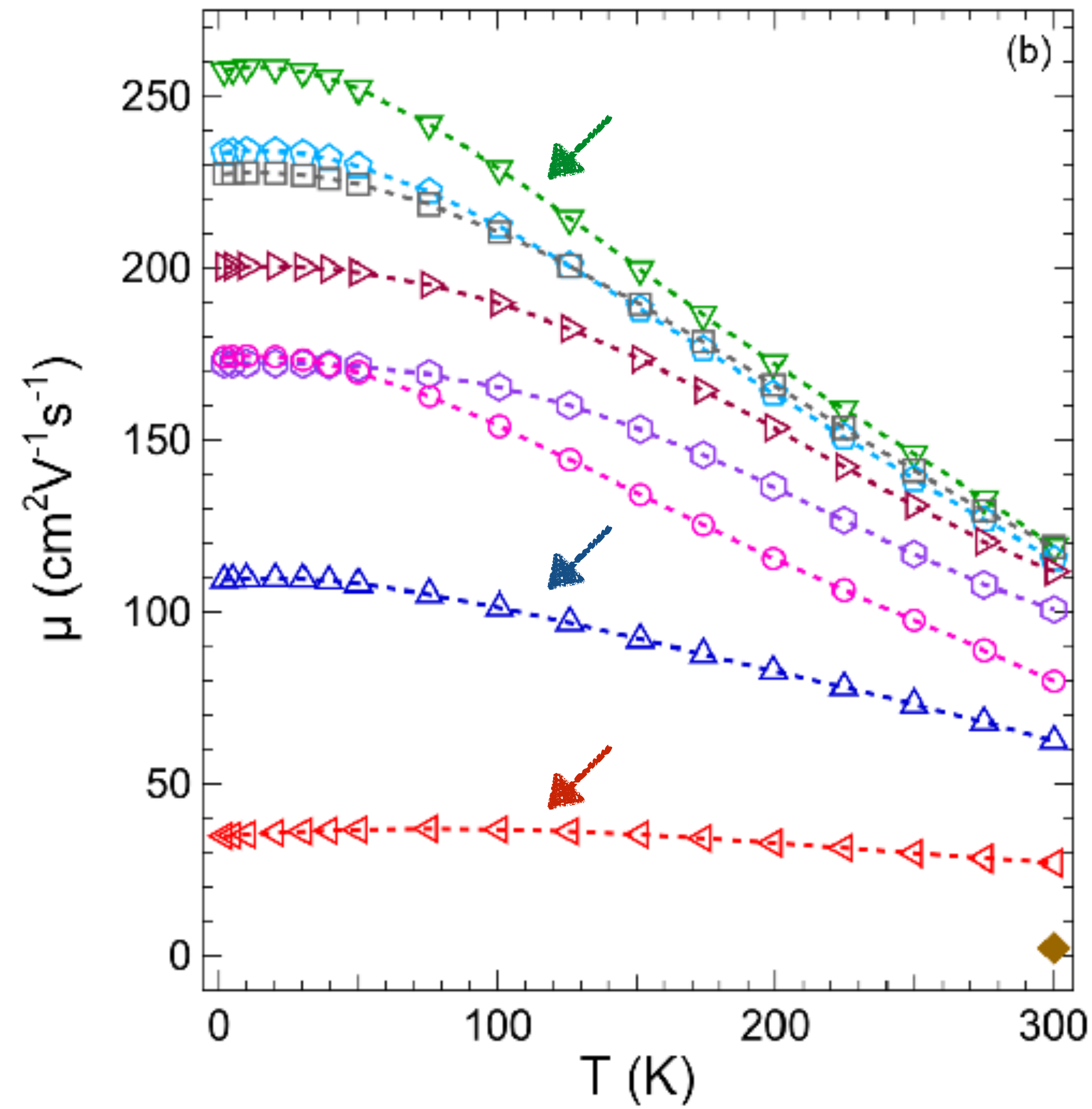


- No freeze-out over 1.8 - 300 K, a *degenerate* semiconductor
- T-dependent mobility suggests different scattering mechanisms at play

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

Mobility Limiting Scattering Mechanisms





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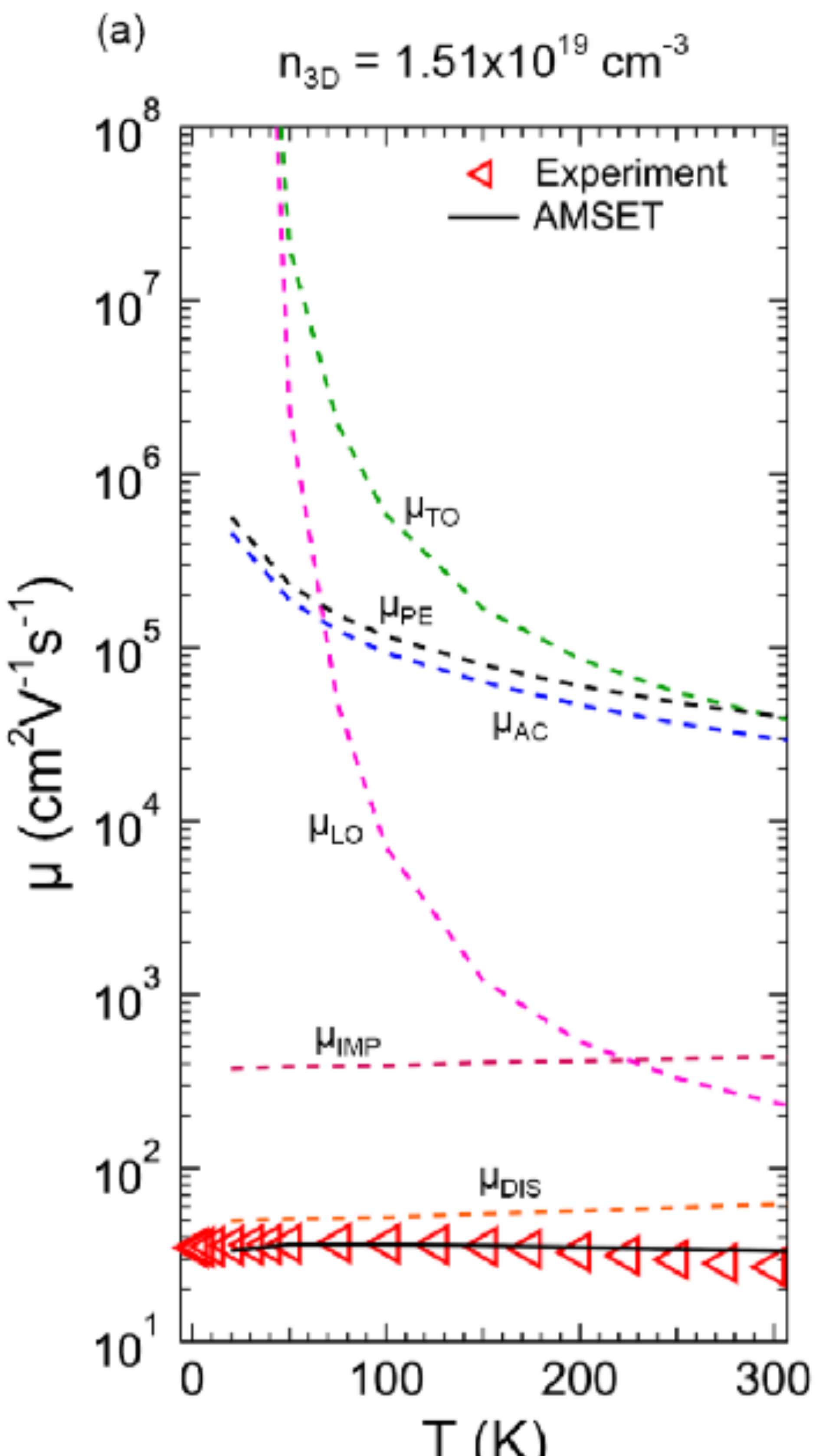
Mobility Limiting Scattering Mechanisms



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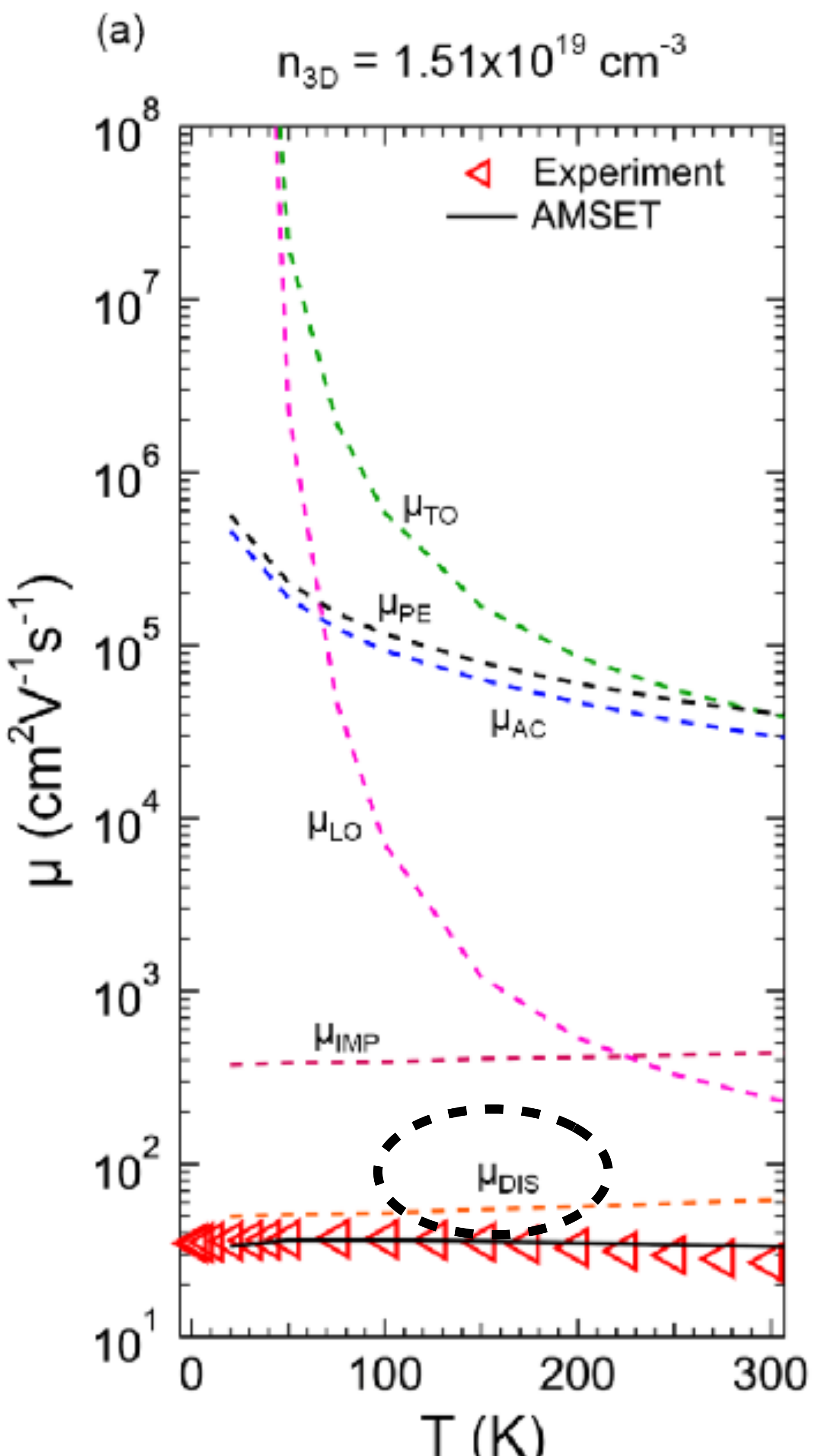
Mobility Limiting Scattering Mechanisms



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Mobility Limiting Scattering Mechanisms

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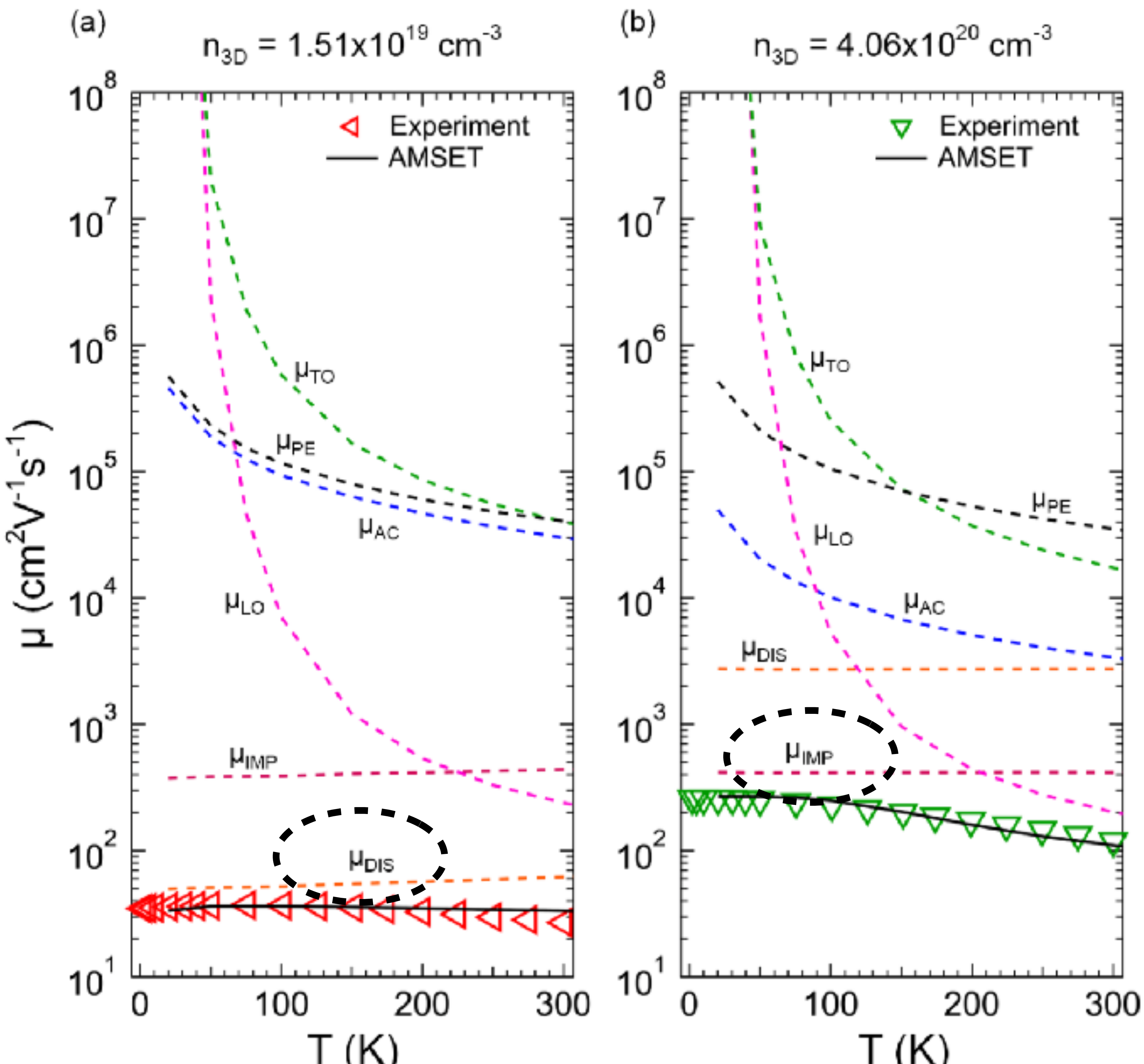

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Mobility Limiting Scattering Mechanisms

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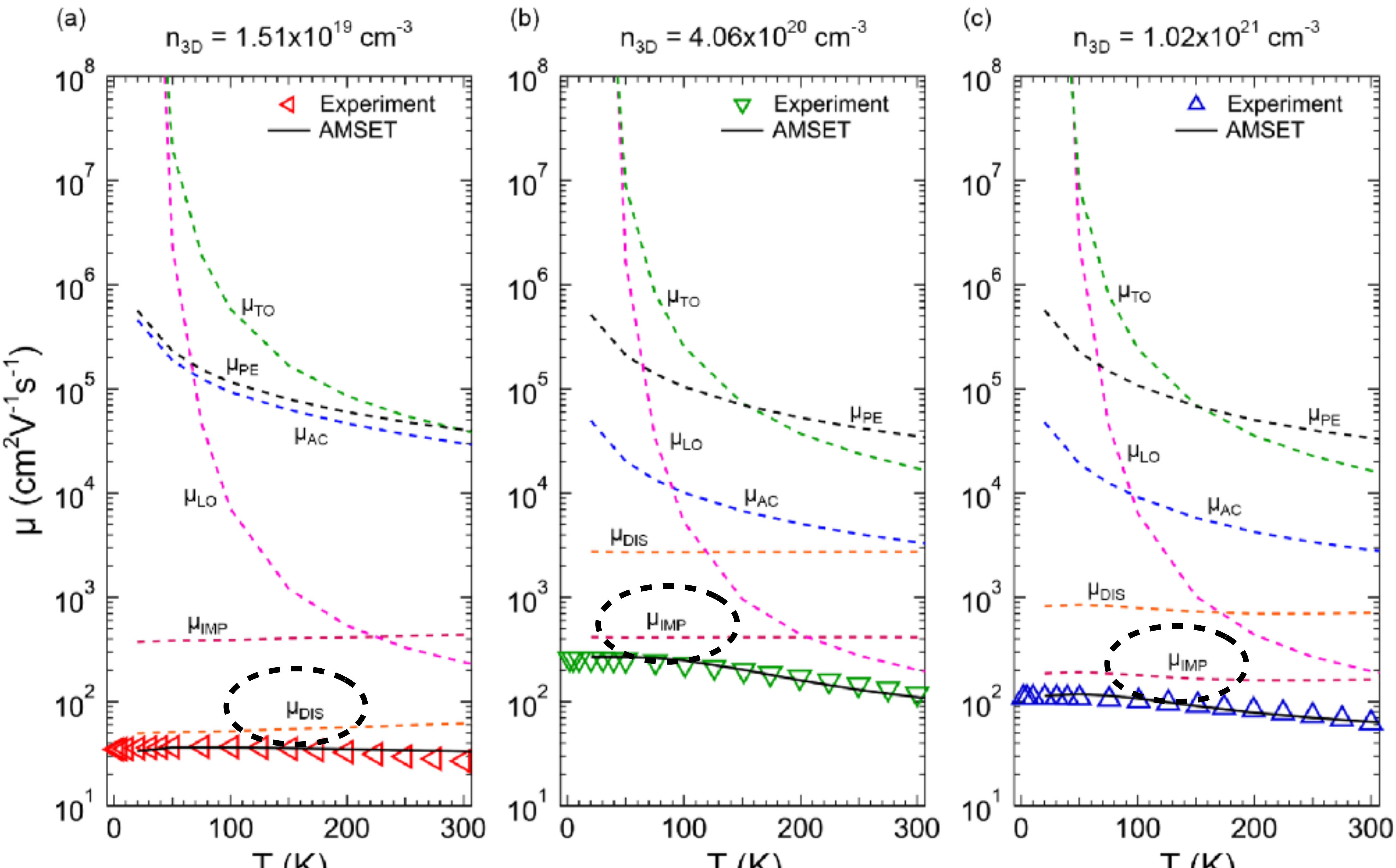

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

BaSnO$_3$ Vs GaN

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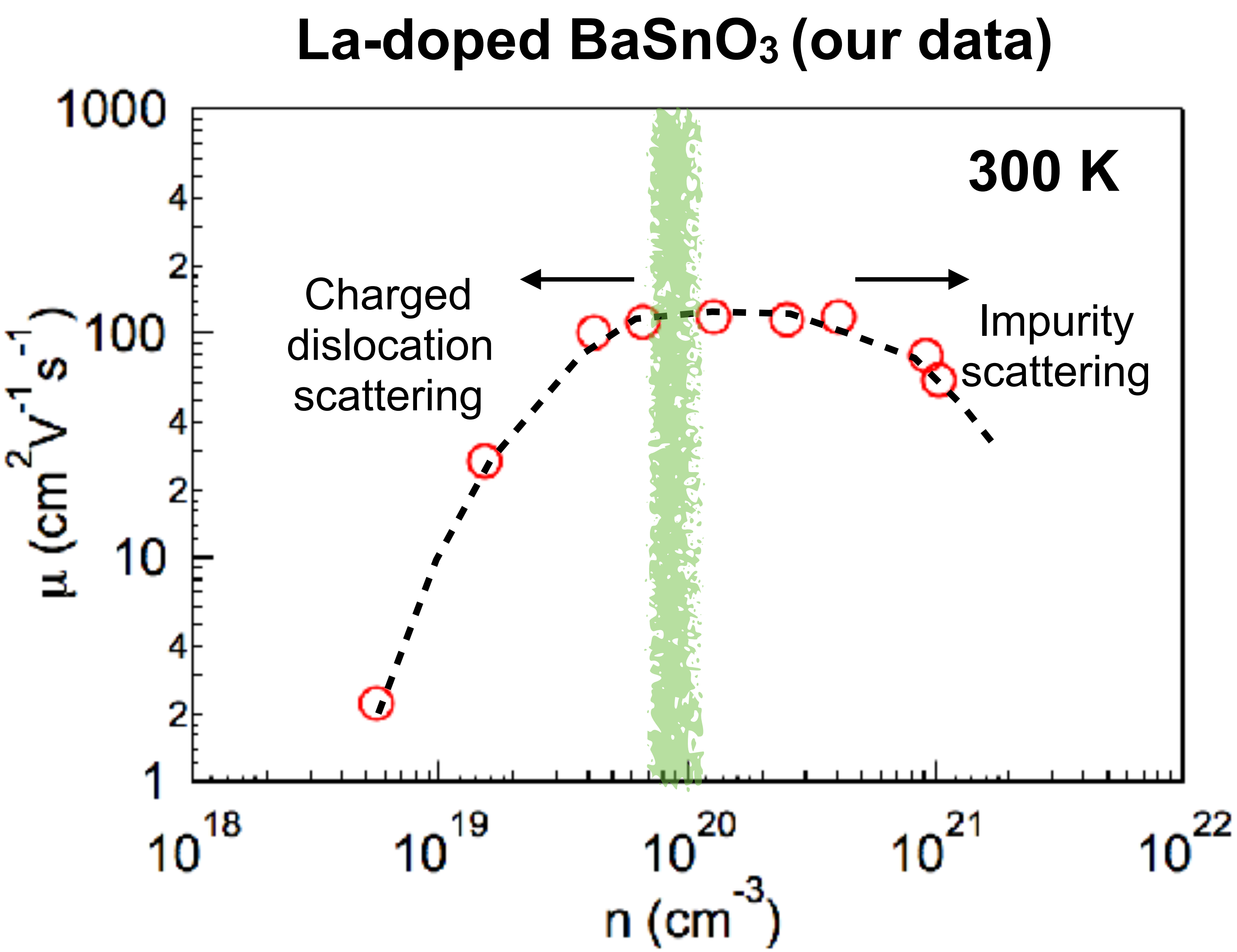

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

$BaSnO_3$ Vs GaN

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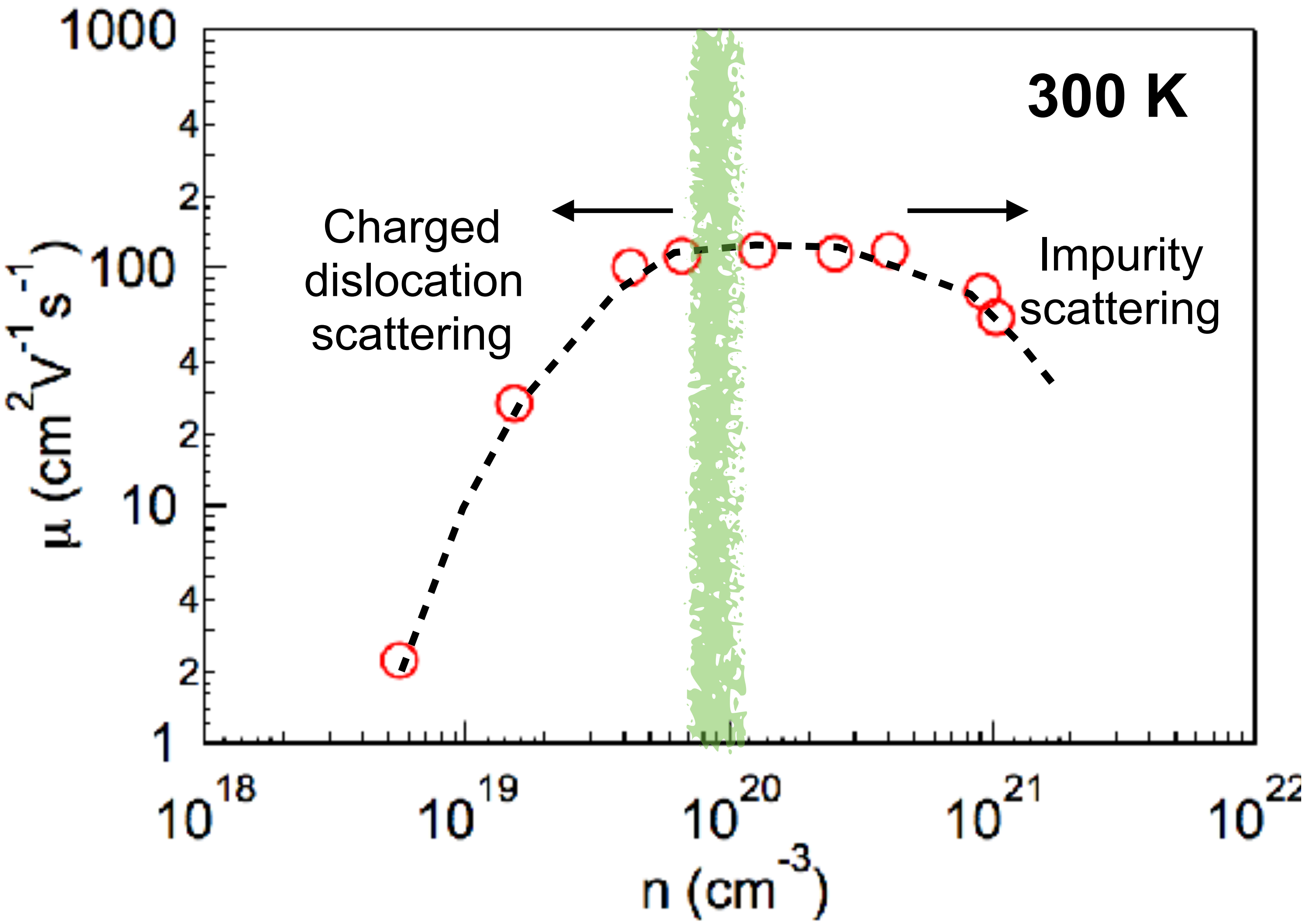
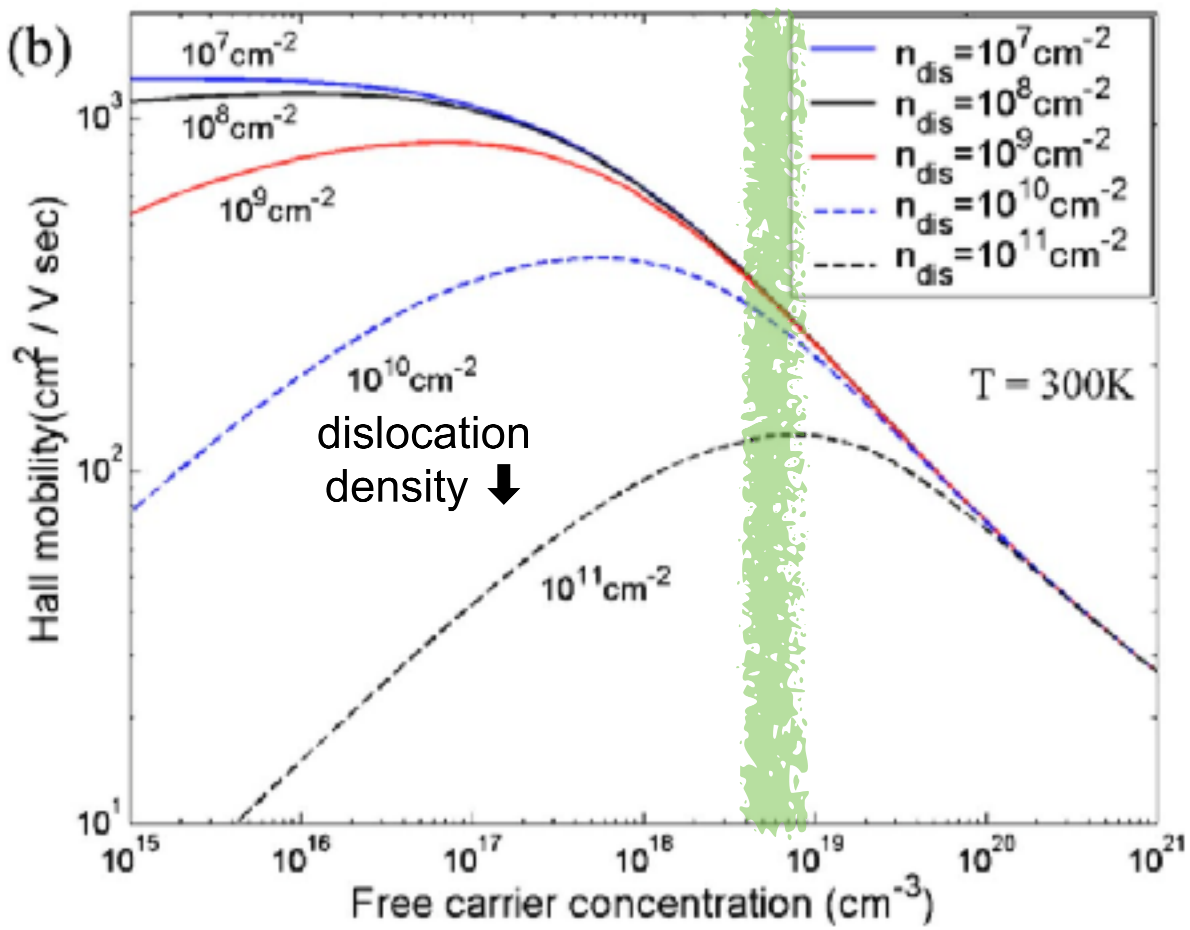
La-doped $BaSnO_3$ (our data)



n-doped GaN*



*J. H. You et. al., JAP **99**, 033706 (2006)

- Taking analogy from GaN, mobility and density in low-doped $BaSnO_3$ is limited by charged dislocations
- RT mobility of $BaSnO_3$ films grown on STO substrates ~125 cm^2/Vs
- **MUCH** room for improvements if dislocation densities are brought down

A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

$BaSnO_3$ Vs GaN

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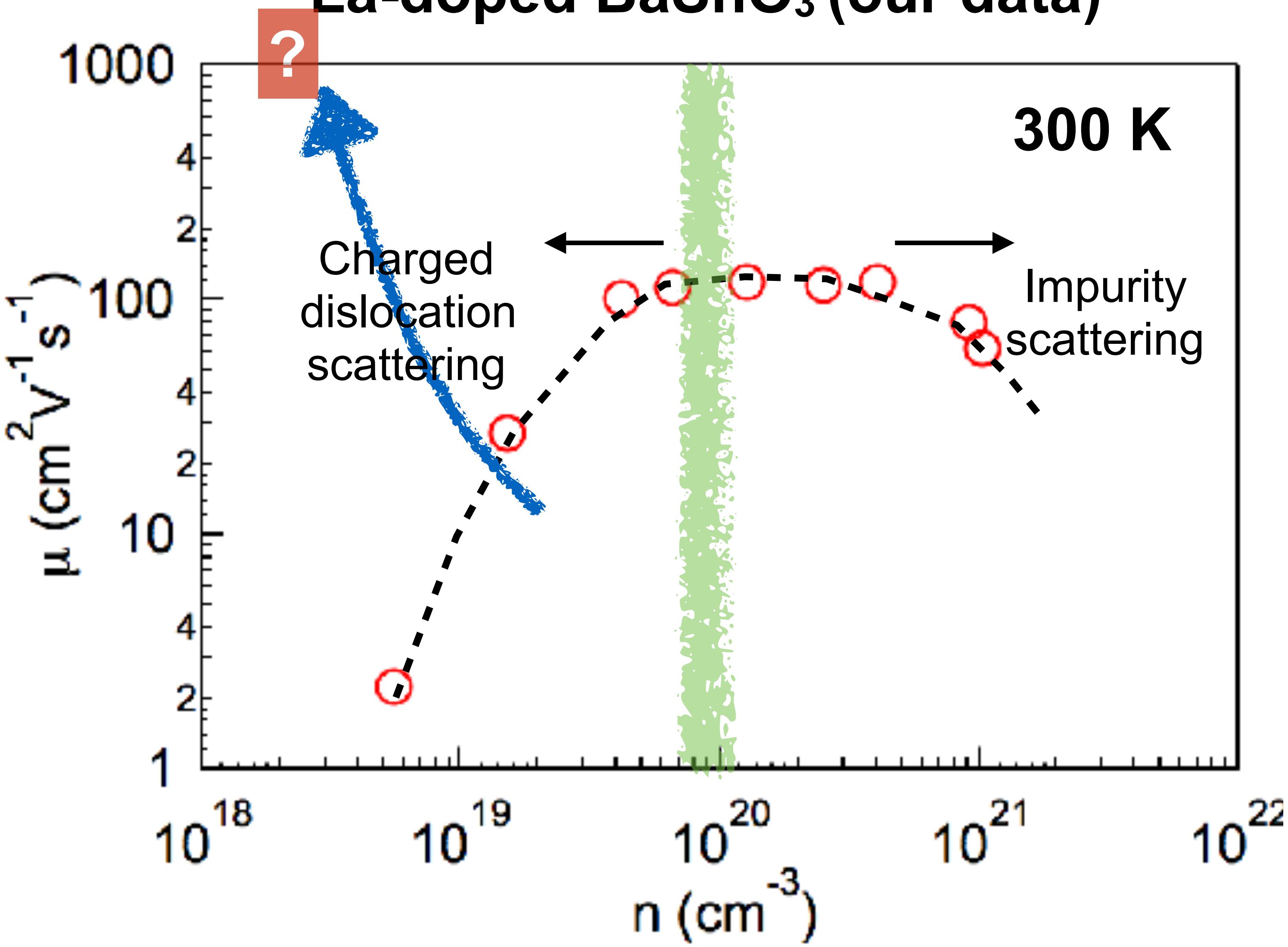

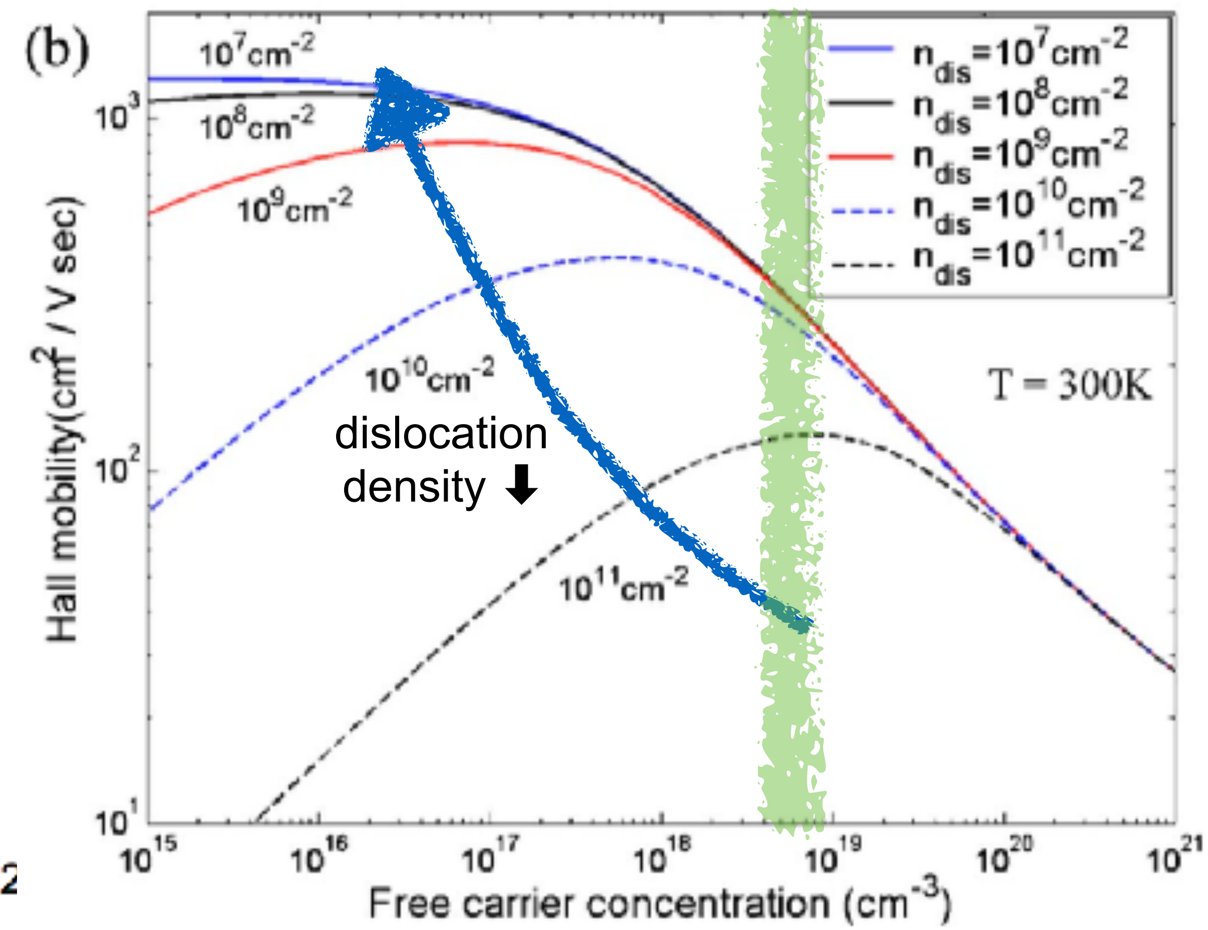

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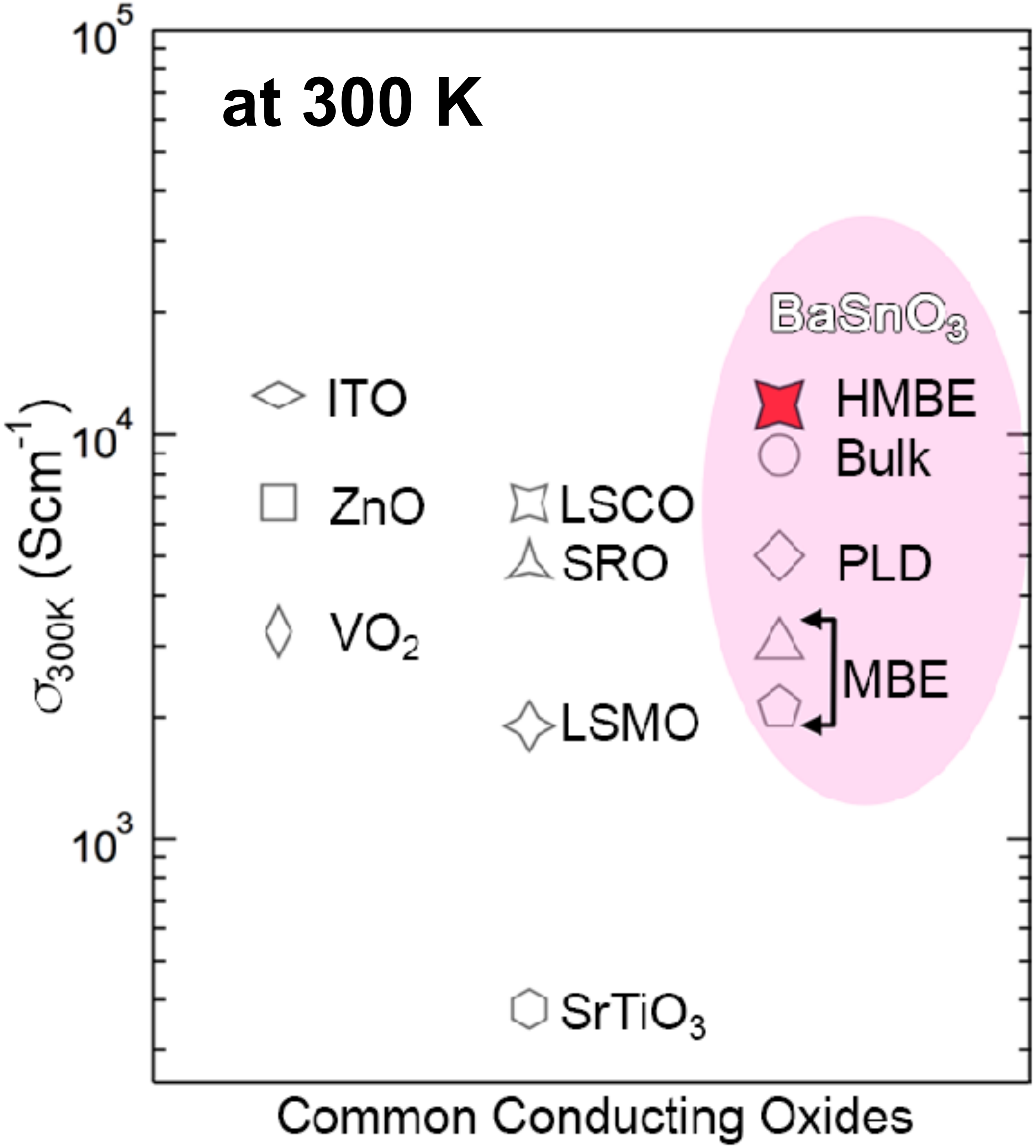
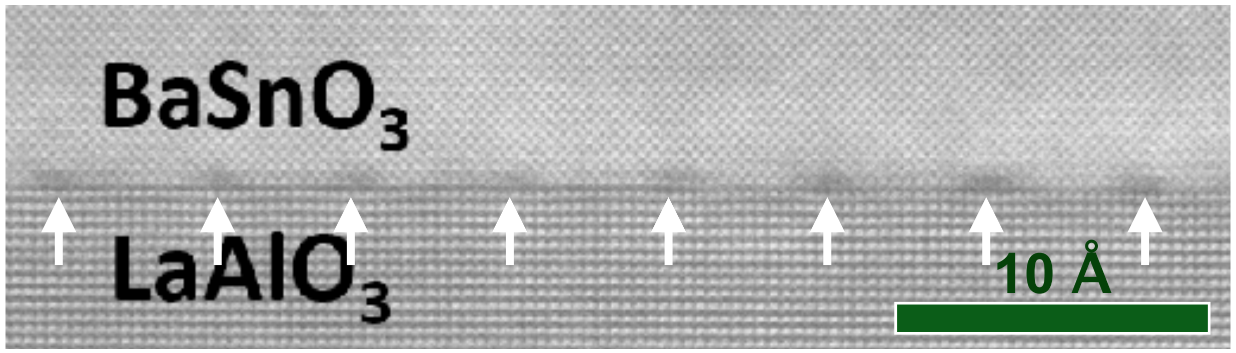
Outlook: $BaSnO_3$ as TCO

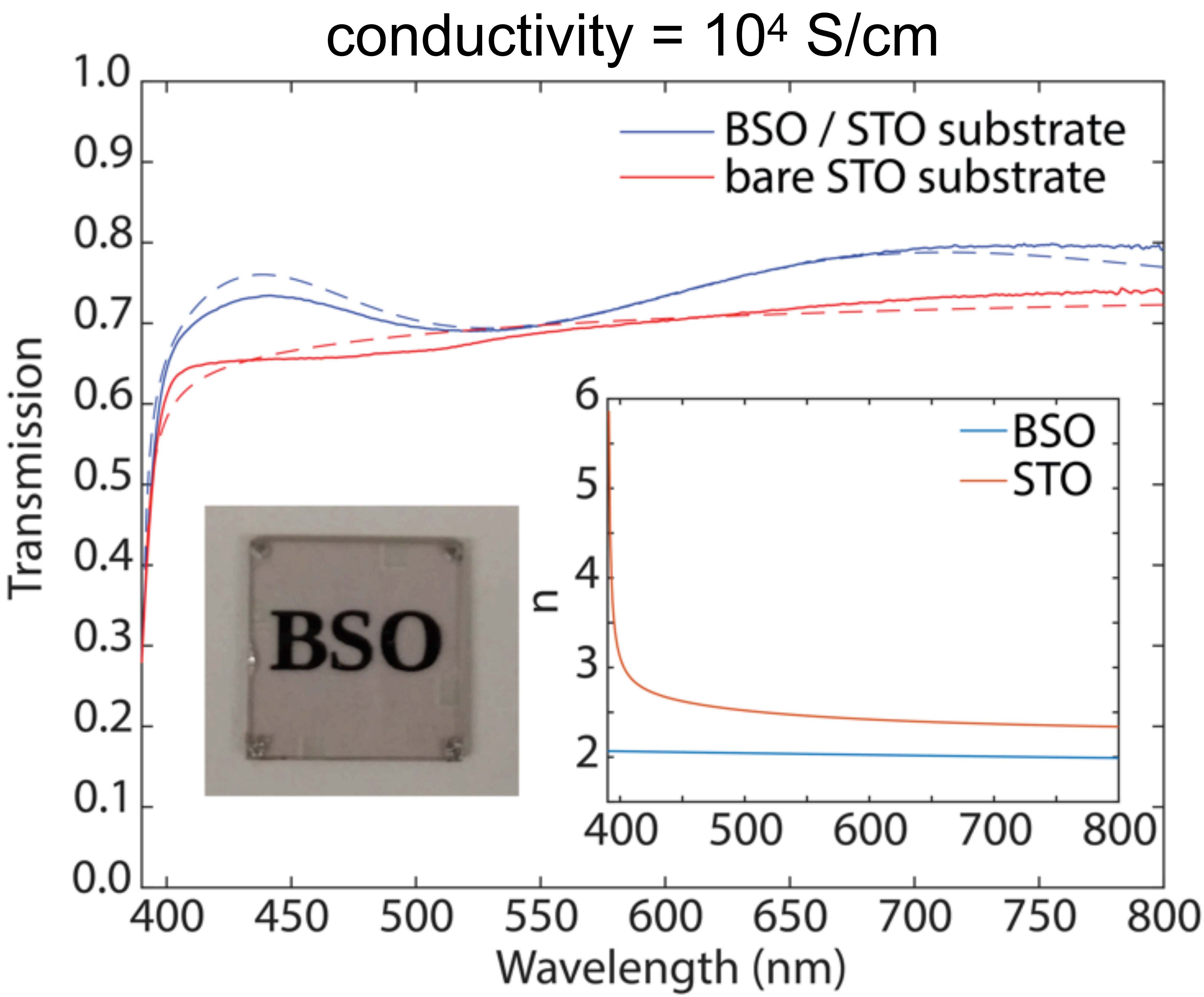


- RT conductivity comparable to that of the best ITO film.
- Much room for improvement if defect density is brought down



A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager III, C. S. Lo, and B. Jalan, *Nat. Comm. 8, 15167* (2017)

Optical Transmission



S. Arezoomandan, A. Prakash, A. Chanana, J. Yue, A. Mao, S. Blair, A. Nahata, B. Jalan, and B. Sensale-Rodriguez, Sci. Rep. 8, 3577 (2018)

Summary

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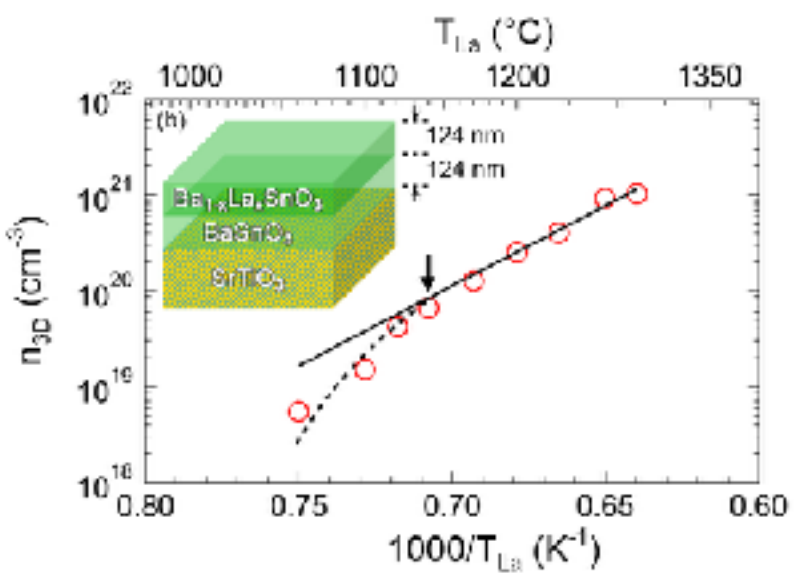
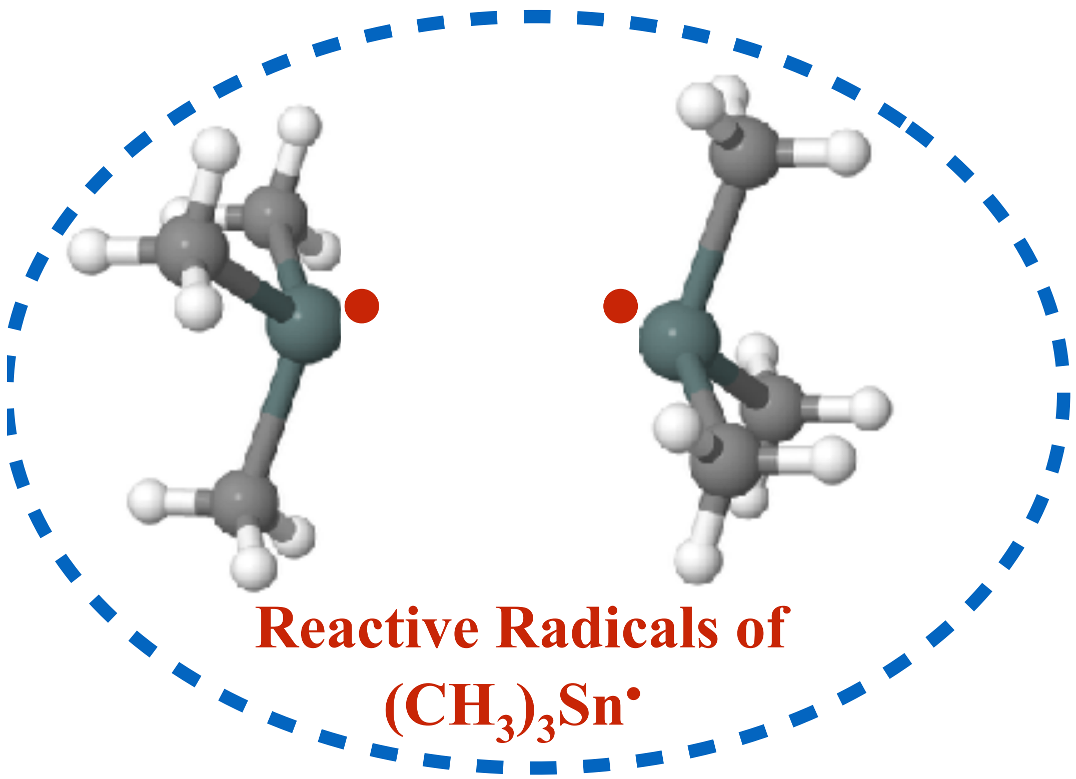
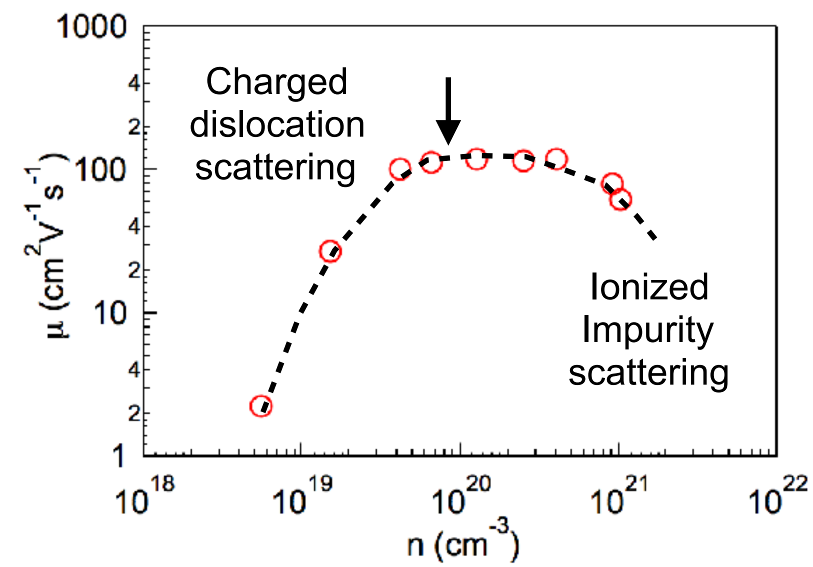
- Novel growth approach for $BaSnO_3$ using reactive radical mechanism
- Discovered MBE growth window for adsorption controlled growth
- Highest room temperature mobility of 125 $cm^2V^{-1}s^{-1}$ for $BaSnO_3$ films grown on $SrTiO_3$
- Mobility is limited by dislocation scattering at low n_{3D} while ionized impurity scattering is the dominant mechanism at high n_{3D}
- RT mobility is limited by LO phonon scattering in the intermediate doping regime
- Much room for improvement if dislocation density is reduced.
- Sheet resistance as low as **2-3 ohm/sq.** was obtained with significant room for improvement.







Novel Hybrid MBE/MOCVD growth approach using reactive radicals

Acknowledgments

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Graduate Student: Mr. Abhinav Prakash

Thank You!

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