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Sandia National Laboratories

Heavy Ion Irradiation Effects on MoS_2 Memtransistors

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Outline

Background and Motivation

- Relevant radiation environments
- Radiation tolerance of TMDs
- Why is the MoS_2 memtransistor interesting?

Heavy Ion Irradiation Effects

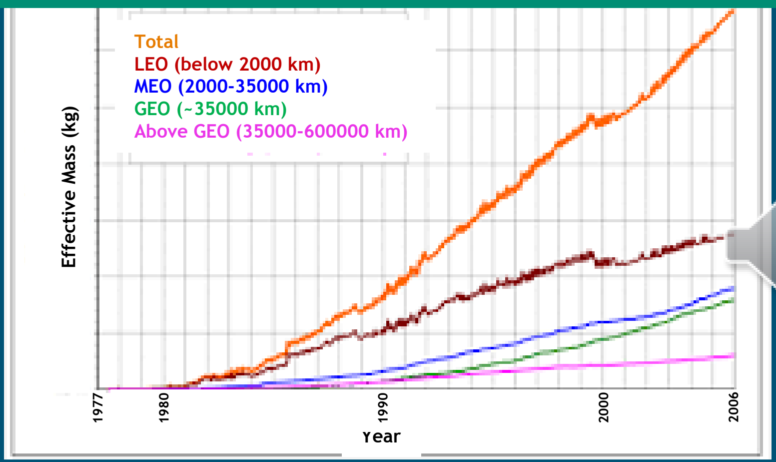
- MoS_2 physical properties vs radiation fluence
  - Chemistry
  - Defect density and morphology
- Memtransistor behavior vs radiation fluence

Background and Motivation

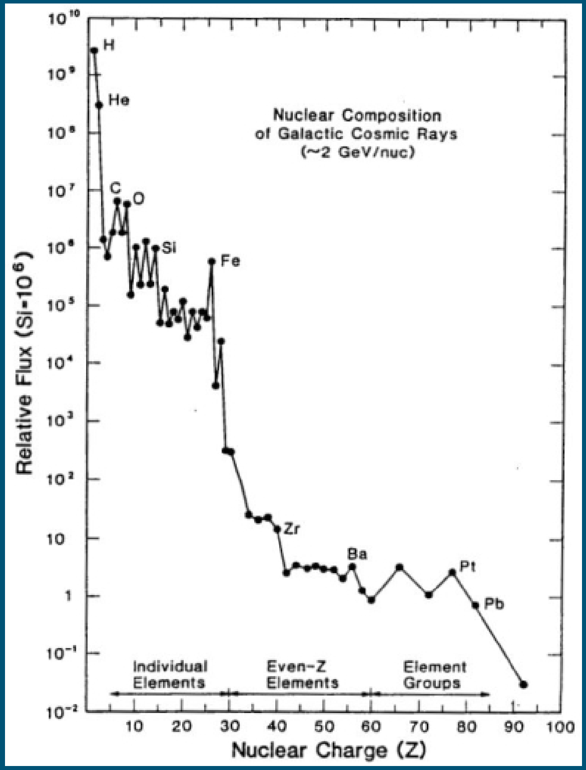
Why do we need radiation tolerant electronics now more than ever?

- Increasing satellite launch rate
- Invigorated global interest in interplanetary exploration

Monthly Effective Mass of Objects in Earth Orbit by Region

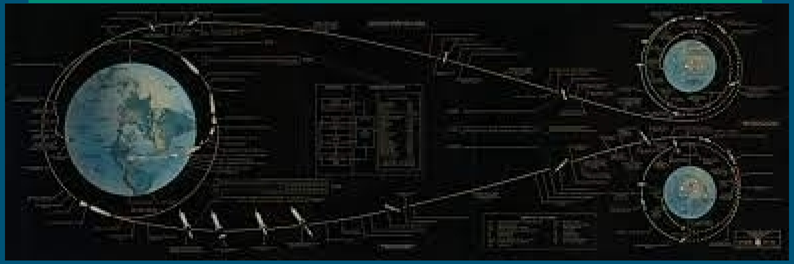


Applicable Space Environments



R. A. Mewaldt, Adv. Space. Res. 1994, 14, 10, 737-747

Spacecraft Trajectory for Manned Lunar Landing

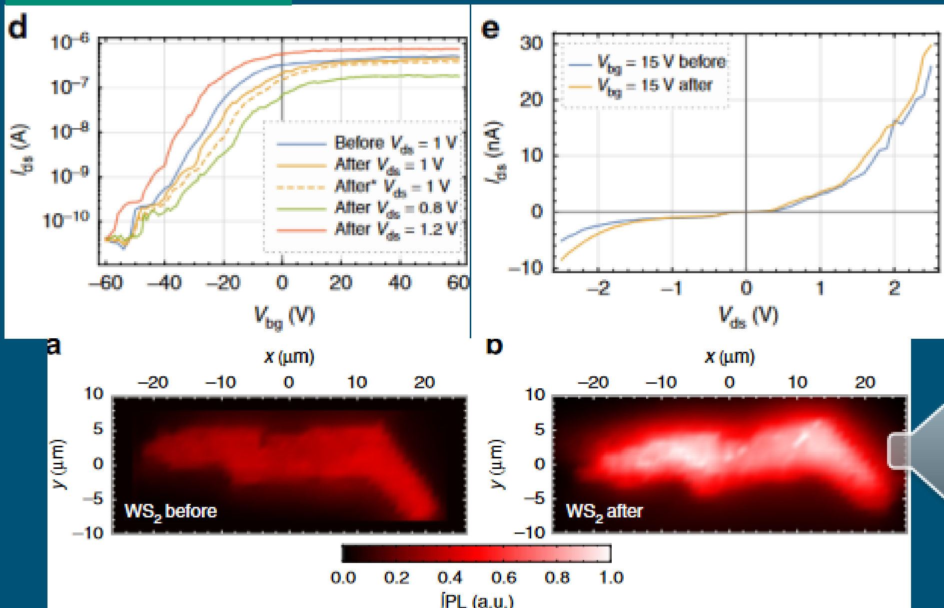


Desired properties of radiation tolerant electronics:

- Reliable, high endurance, low power, SEU-

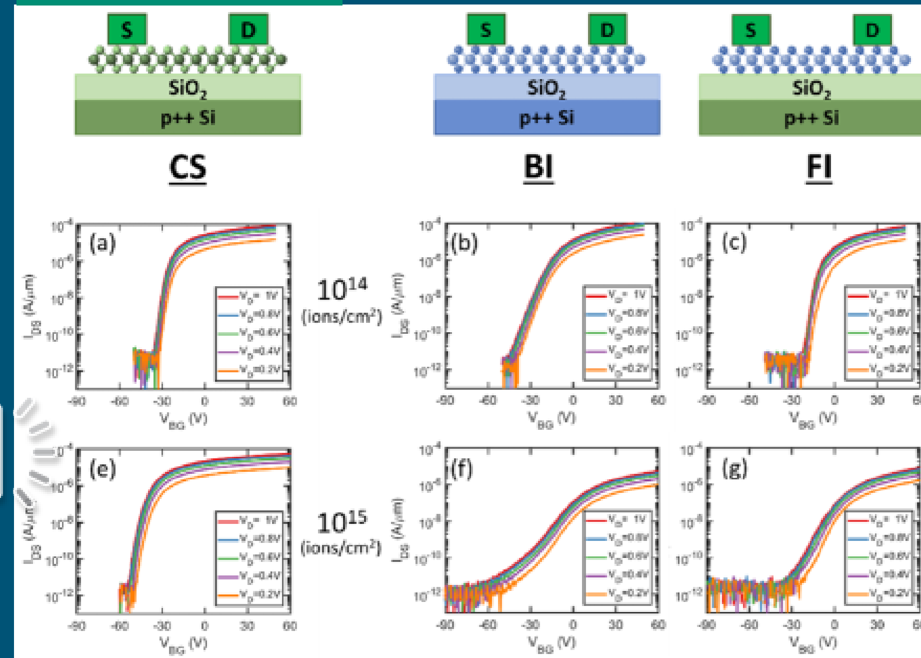
Transition Metal Dichalcogenides for Applications in Ionizing Environments

γ-Ray Irradiation



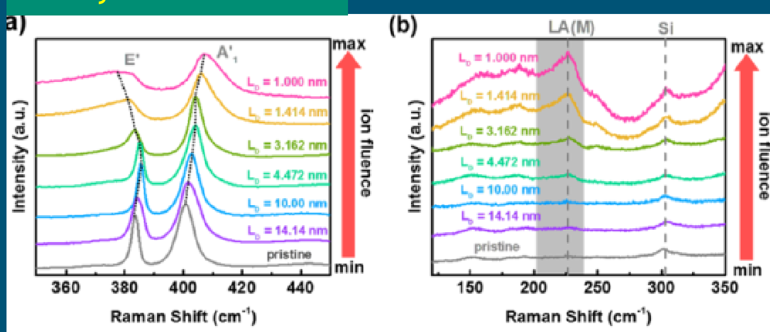
T. Vogl et al., Nat. Commun. 2019, 10, 1202

Light Ion Irradiation



A. J. Arnold et al., ACS Appl. Mater. Interfaces 2019, 11, 8391-8399

Heavy Ion Irradiation



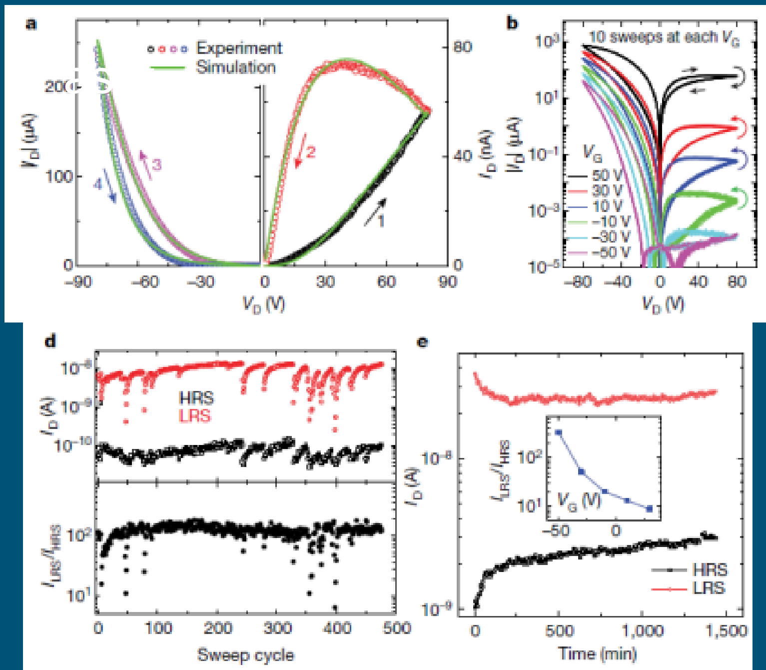
Z. He et al., ACS Appl. Mater. Interfaces 2018, 10, 42524-42533

TMDs are promising for logic applications under energetic photon and light ion irradiation

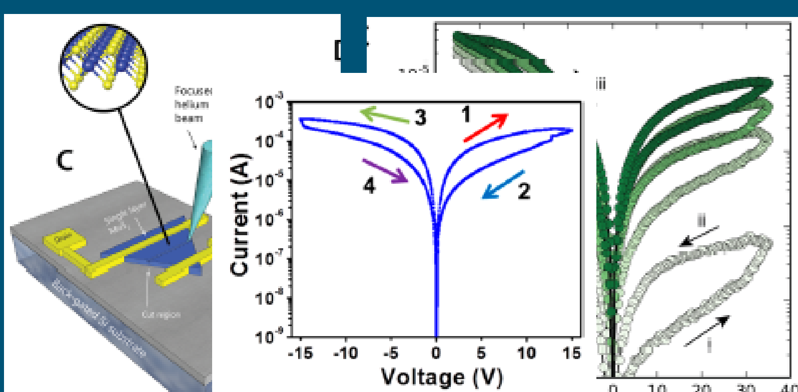
What about device performance under swift heavy ion irradiation?

Why Study the Radiation Tolerance of the MoS_2 Memtransistor

MoS_2 Memory Devices



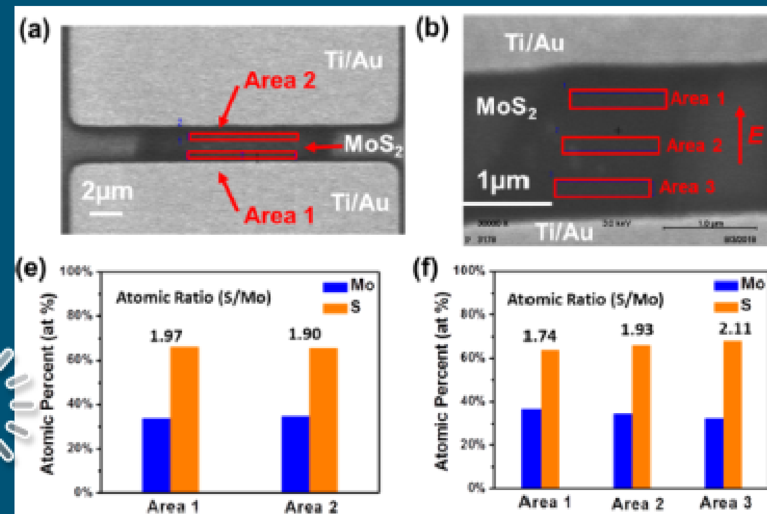
V. K. Sangwan et al., Nature 2018, 554, 500-504



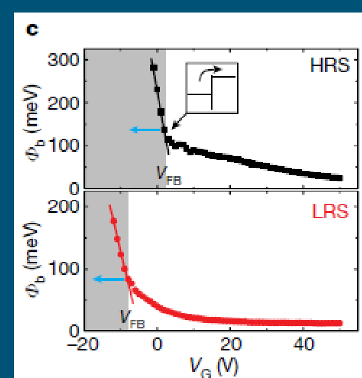
D. Li et al., ACS Nano 2018, 12, 9240-9252

J. Jadwiszczak et al., ACS Nano 2019, 13, 14262-14273

Memristive Behavior based on Defect-Mediated Switching



D. Li et al., ACS Nano 2018, 12, 9240-9252



V. K. Sangwan et al., Nature Nanotechnol. 2015, 10, 403-406

Is the defect-reliant MoS_2 memtransistor inherently radiation tolerant?

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  - Chemistry
  - Defect density and morphology
- Memtransistor behavior vs radiation fluence

Experiment Details: Ex-situ Irradiation of MoS_2 Devices and Films

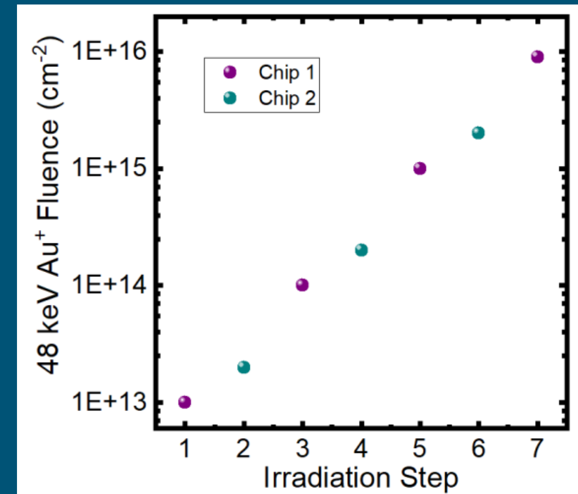
keV - MeV Au^+ used for all irradiations in this work

Device Irradiation:

- 2× array of back gated memtransistors
- Device *and* back gate irradiated

Large Area MoS_2 Irradiation

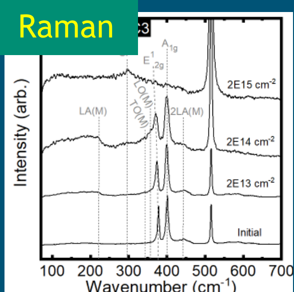
- 2× large area CVD MoS_2 on SiO_2/Si



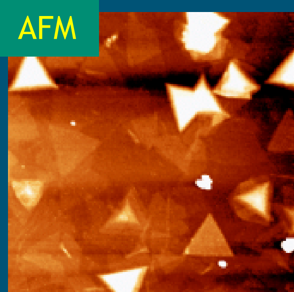
Large Area CVD MoS_2

MoS_2 Memtransistors

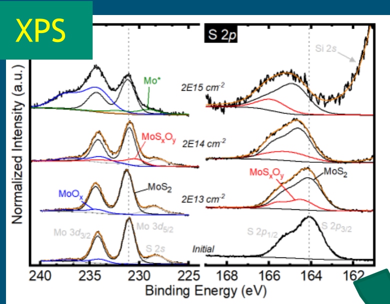
Raman



AFM

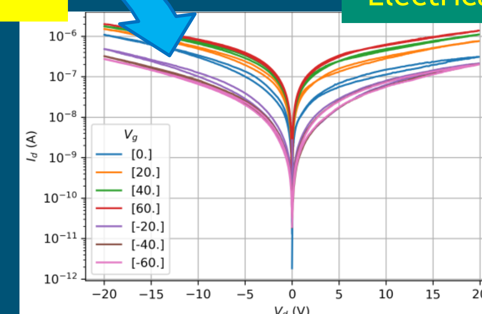


XPS



48 keV or 2.8 MeV Au^+

Electrical



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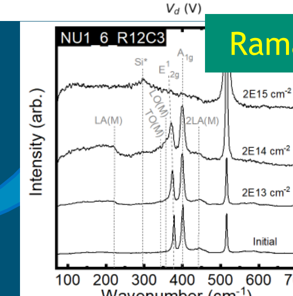

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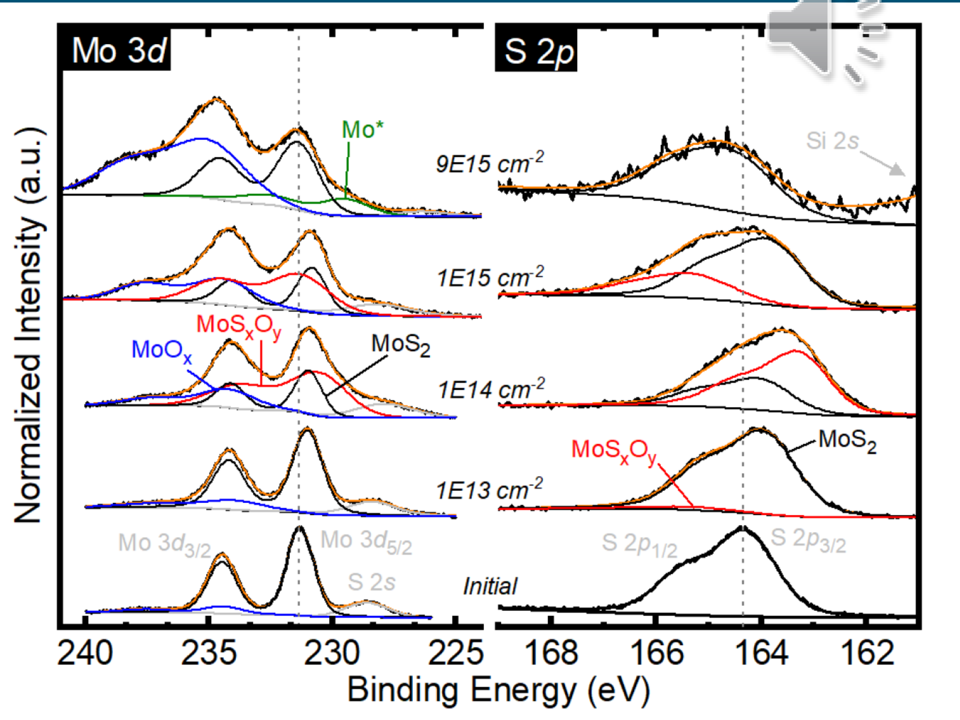
Etching and Oxidation of MoS_2 Induced by Au^+ Irradiation

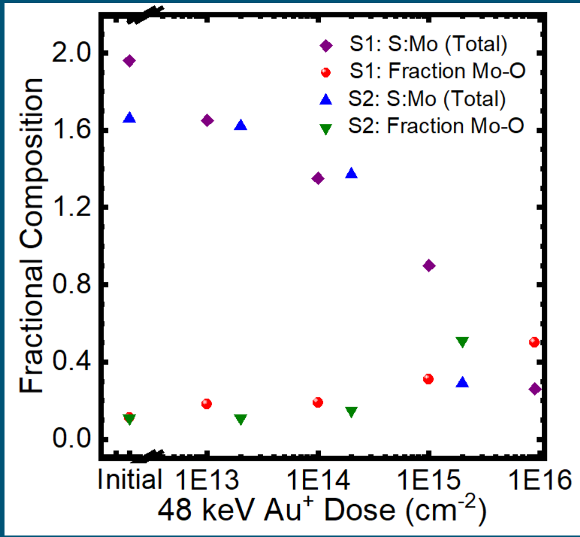
Initial MoS_2 chemistry:

- ~1.9 S:Mo ratio
- Nominal MoO_x concentration for polycrystalline CVD MoS_2

48 keV Au^+ irradiation:

- Mo, S sputtering occurs
  - 43% decrease in Mo 3*d* intensity after 2E15 cm^{-2} Au^+
  - 92% decrease in S 2*p* intensity after 2E15 cm^{-2} Au^+
- MoO_x concentration increases from 10% to 50% after 2E15 cm^{-2} Au^+





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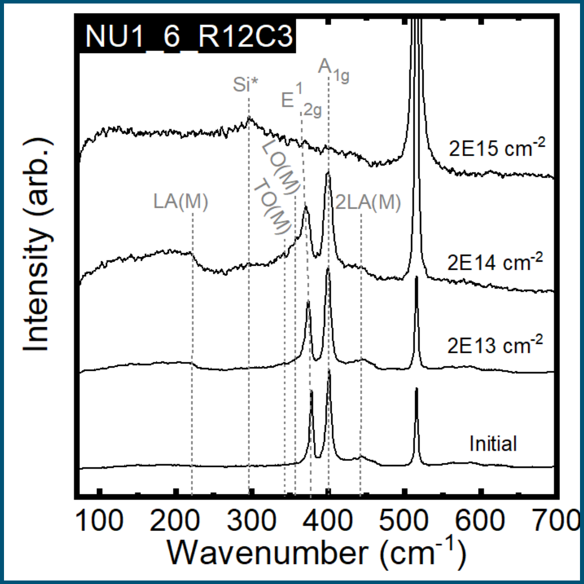
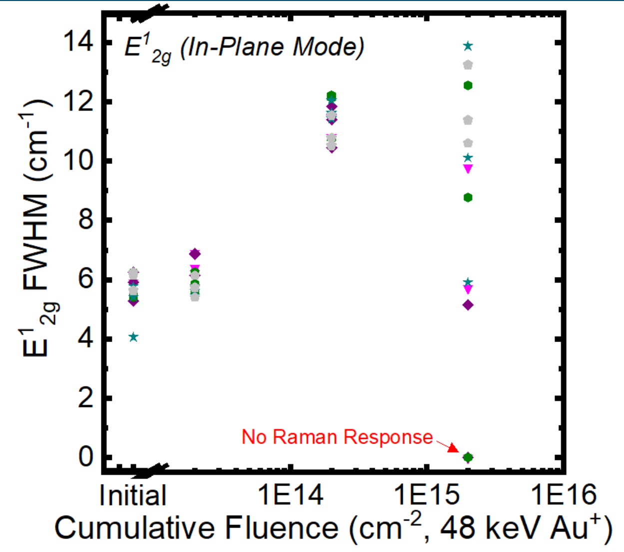
Increased Disorder and Point Defect Density in Au^+ Irradiated MoS_2

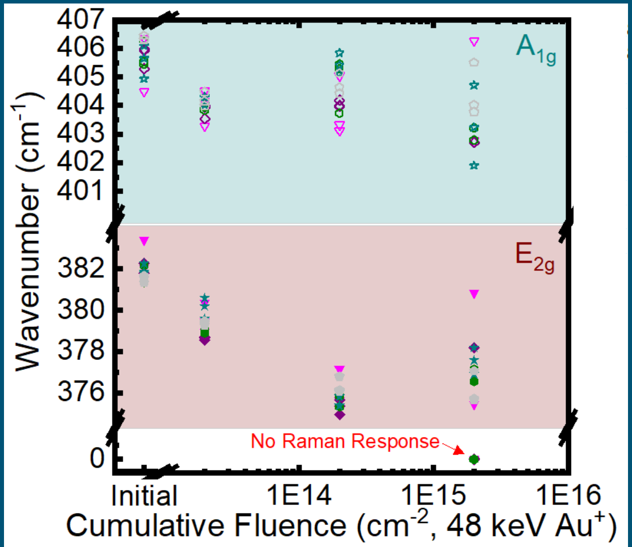
Irradiation-induced tensile strain in MoS_2: E^1_{2g} and A_{1g} red shift after 2E13 cm^{-2} Au^+

Significant increase in defect density at higher Au^+ fluences

- ~2× increase in E^1_{2g} (in-plane mode) FWHM
- Complete loss of Raman response in some MoS_2 regions







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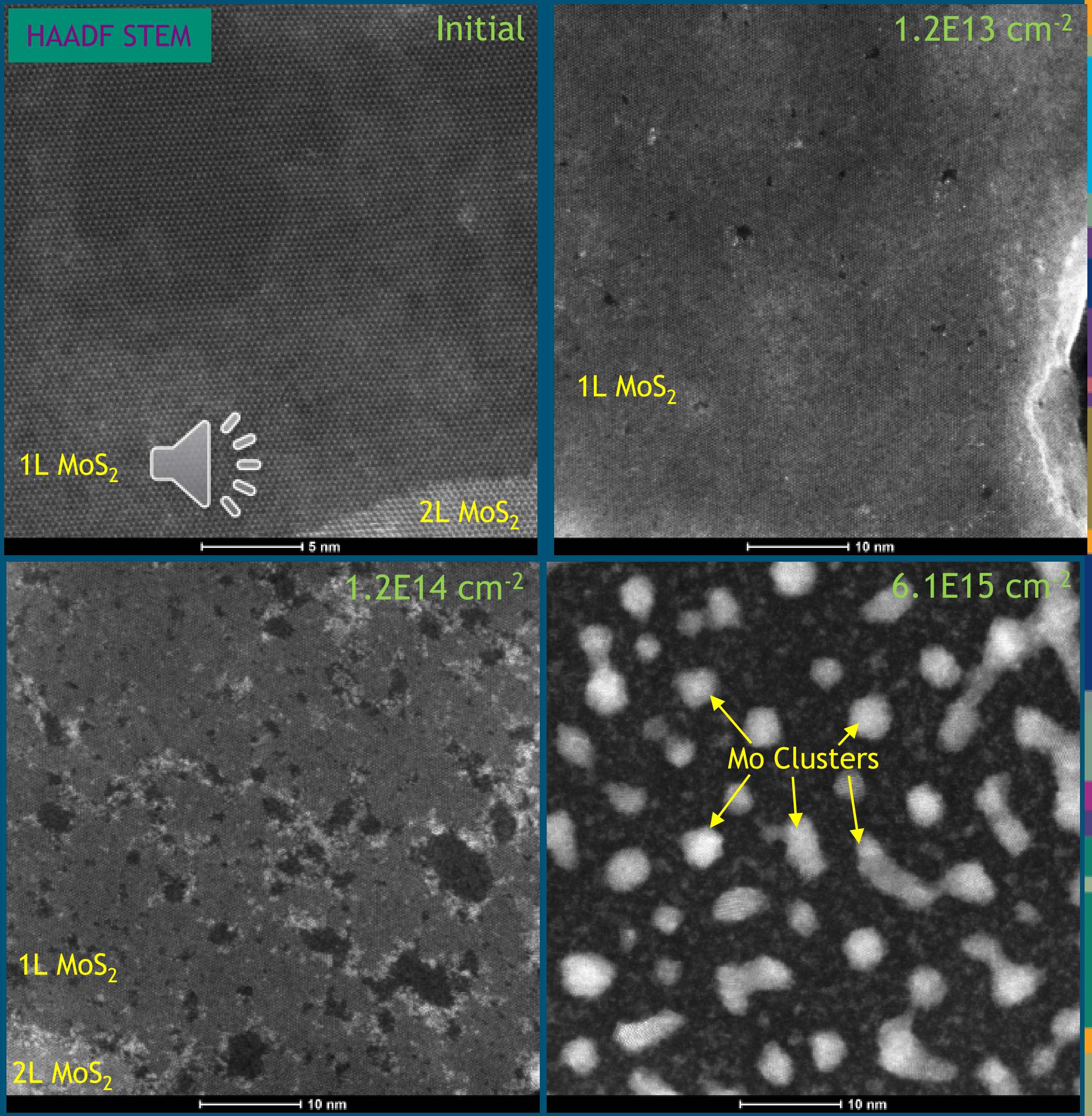
Atomic Scale Defects in MoS_2 Induced by 2.8 MeV Au^+

What we did: Irradiated CVD MoS_2 on a lacey carbon grid with 2.8 MeV Au^+

Multi-atom defects generated in MoS_2 by Au^+ irradiation

S:Mo sputtering rate ratio ≈ 4.7 under 10^4-10^8 eV Au^+ irradiation

Mo sputtering rate under Au^+ irradiation is 10^4 higher than under H^+ or α particle irradiation



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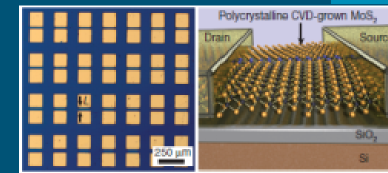
Resistance Switching Collapse in Heavy Ion Irradiated Memtransistors

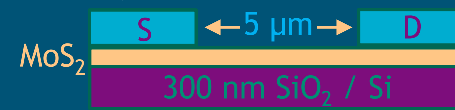
Initial memtransistor behavior: 10^1-10^2 resistance ratio, gateable I_D range over 10^5

Au^+ irradiation:

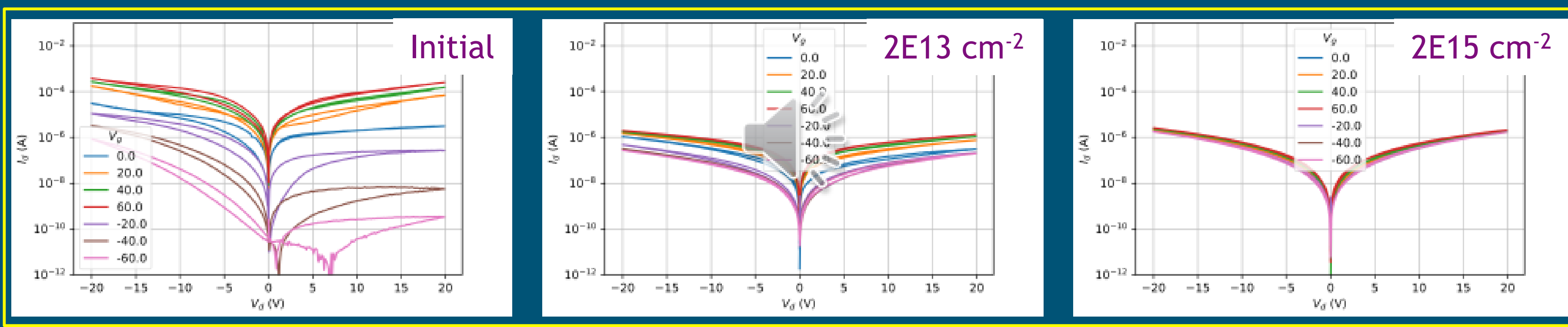
- Causes collapse of resistance switching and gateability
- I_D increase with increasing Au^+ fluence due to variable hopping conduction*

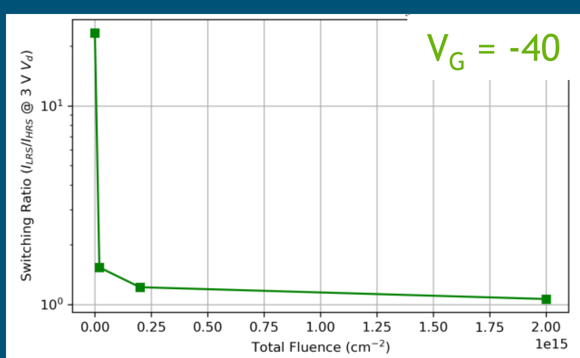


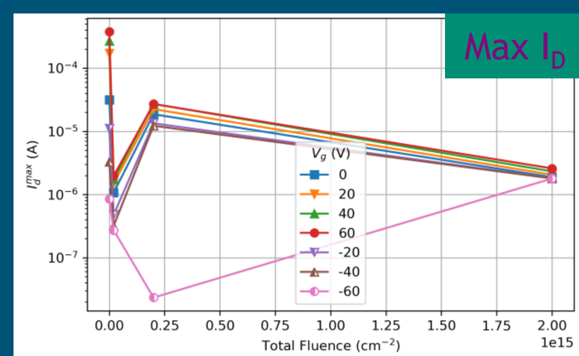
V. K. Sangwan et al., Nature 2018, 554, 500-504

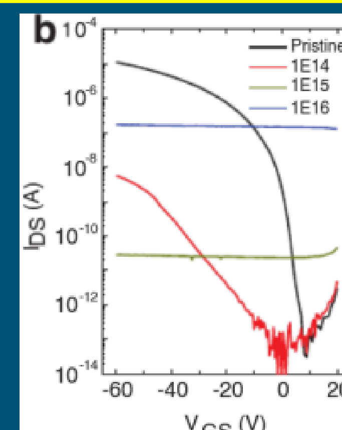


Increasing Au^+ Fluence









*M. G. Stanford et al., Adv. Funct. Mater. 2017, 27, 1702829

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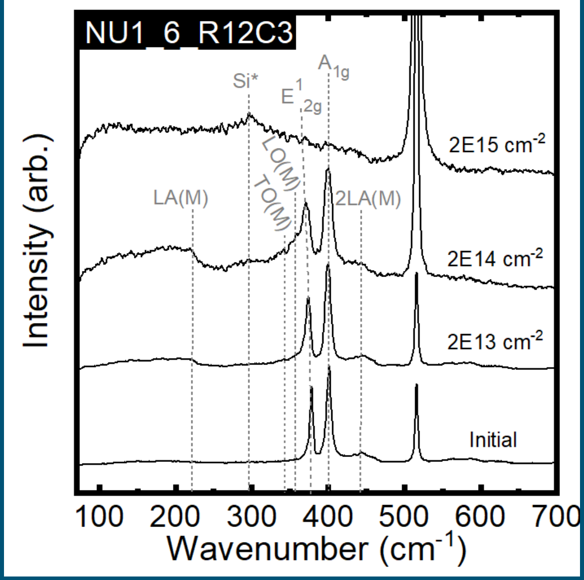
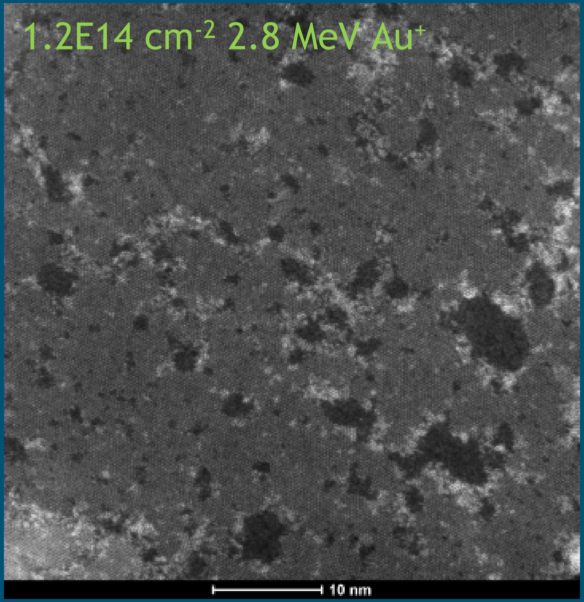
Conclusions

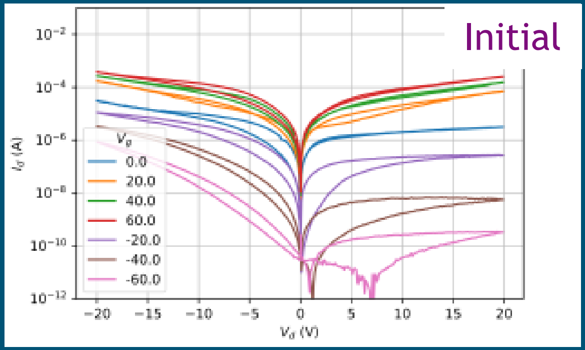
Au$^+$ irradiation in the keV – MeV energy range:

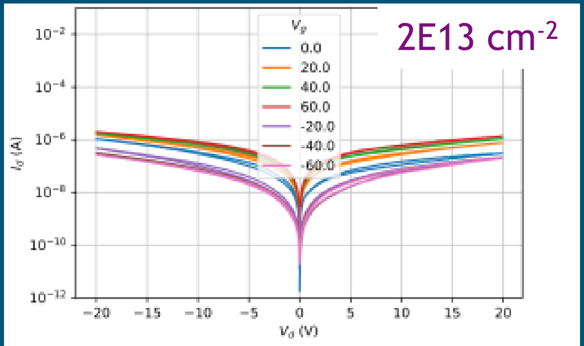
- Creates Mo and S vacancies and larger holes in 1L MoS_2
- Significant sputtering, oxidation, and reduction in S:Mo ratio occurs

Memtransistor behavior persists through an unusually high heavy ion fluence of 2E13 cm^{-2}, which is effectively 200 years worth of galactic cosmic rays*. This highlights the potential for the MoS_2 memtransistor to be highly reliable over extended missions beyond low earth orbit









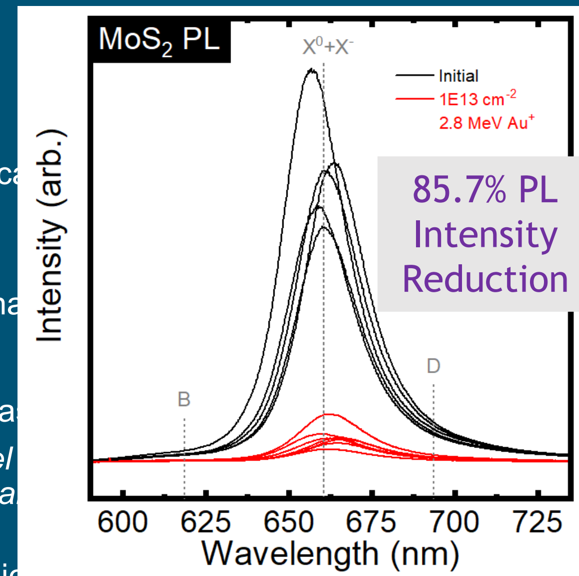
*when scaled based on active device area in MoS_2 memtransistor versus active device area in a single commercial logic device

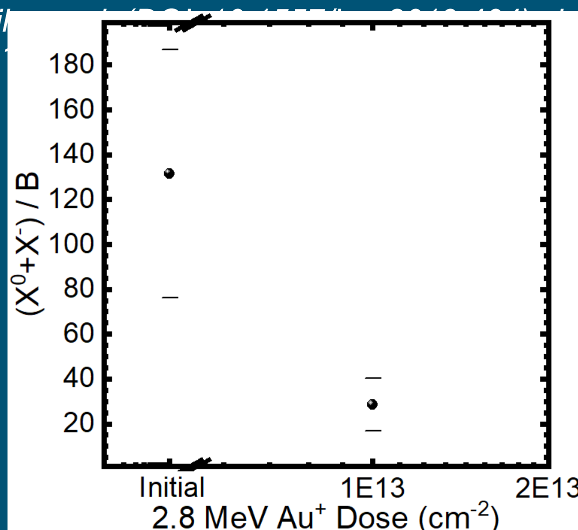
Supporting Information

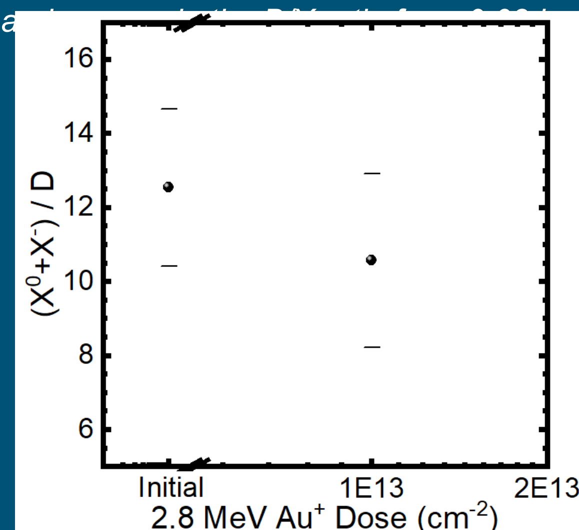
Supporting Information: Photoluminescence vs Au^+ Fluence

PL Argument:

- Nano Research 2018, 11 (8): 4123-4132
  - Increase in X peak can indicate an increase in sulfur vacancies
  - Increase in X peak often corresponds with decreased B peak, which indica
    the higher energy spin orbit valence band
  - B peak decreases with increasing sulfur vacancy concentration
  - Lower energy D peak corresponds with sulfur vacancy-induced recombina
- Our samples:
  - X peak decreases by 85.7%, B peak decreases by 46.5%, D peak decrea
  - *Nearly simultaneous decrease in X and D points to a signature of Frenkel*
    and S removal instead of preferential S vacancy creation, which could ma
    decrease in the B peak / increase in the D peak
  - Comparatively smaller decrease in B peak than X and D peaks could indic
    increasing disorder
  - Very small ~0.02-0.07 eV shift of all peaks after irradiation
  - *Other simil* ... *ows a* ... *VD* MoS_2 *to*
    0.52 after







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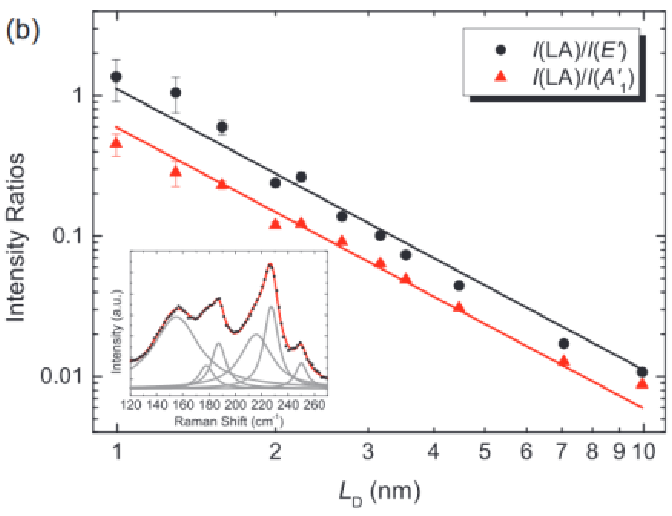
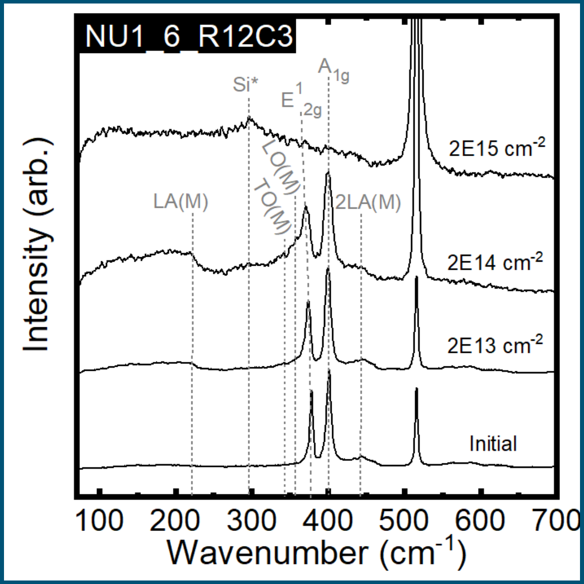
Increased Disorder and Point Defect Density in Au^+ Irradiated MoS_2

Point defect concentration in MoS_2 can be extracted using an empirical relationship based on the LA(M):A_{1g} or LA(M):E^1_{2g} intensity ratio



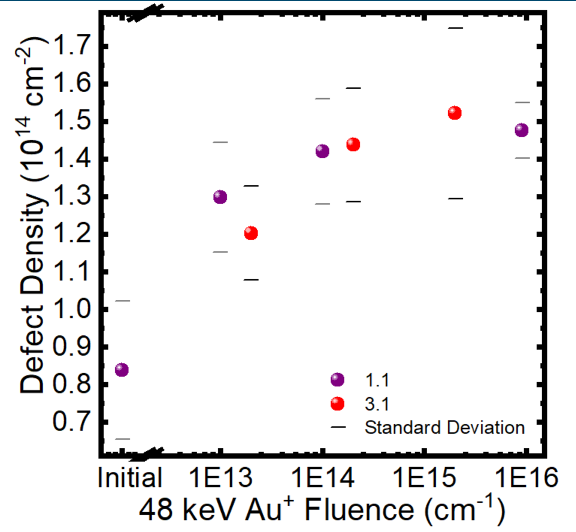
S. Mignuzzi et al., Phys. Rev. B 2015, 91, 195411

$$\frac{I(LA)}{I(A_{1g})} = \frac{C(A_{1g})}{L_D^2}$$



I(LA)/I(A_{1g}) ratio indicates 2× increase in MoS_2 point defect density after irradiating with 2E15 cm^{-2} 48 keV Au^+

The Raman-extracted defect density conflicts with XPS results, which indicate significant S sputtering and disordering



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In-Situ SAED during 2.8 MeV Au^+ Irradiation of MoS_2

In-Situ Irradiation and SAED

Initial

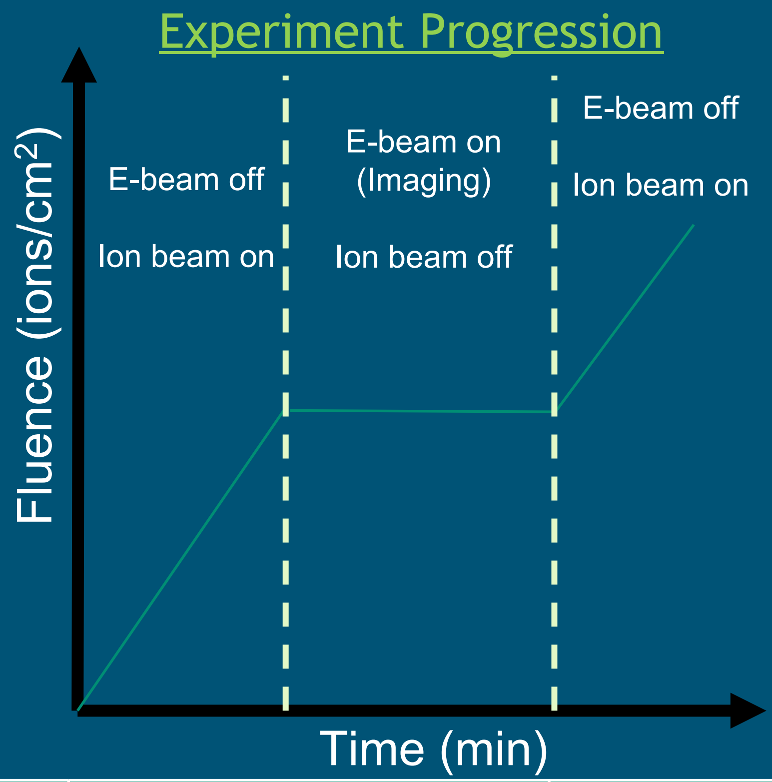
1.2E14 cm^{-2}

8.7E14 cm^{-2}

In-situ Irradiation and TEM Experiment Details

Ion beam conditions
- 2.8 MeV Au^{4+}
- 0.03 cm^2

TEM conditions
- $T_x = 30.4°$, $T_y = 0°$
- SAED taken at 60 cm
- Magnification of Images: 40kx
- Electron beam blanked during irradiation
- TEM image and SAED took ~3 minutes each step



Elapsed Time (min)	Current Start (nA)	Current End (nA)	Estimated Fluence (ions/cm²)	Set Number
10	4.0	4.1	1.25×10^{14}	1
20	2.6	2.5	2.5×10^{14}	2
30	2.6	2.8	3.74×10^{14}	3
40	2.8	2.8	4.99×10^{14}	4
70	2.8	2.8	8.74×10^{14}	5

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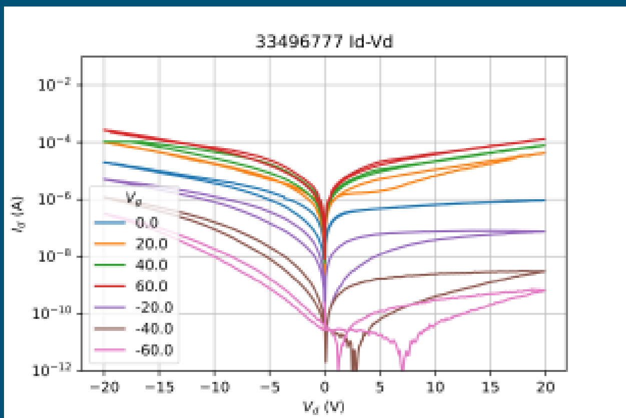
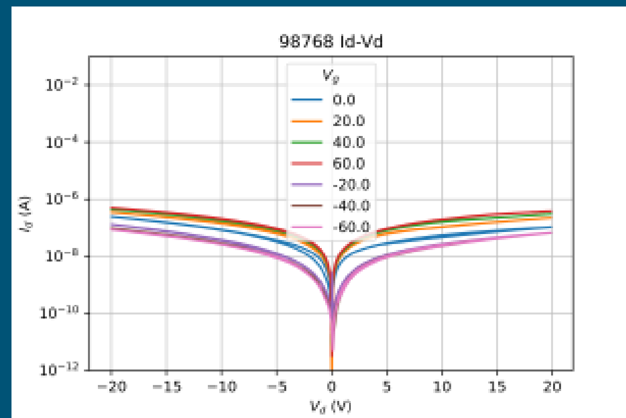
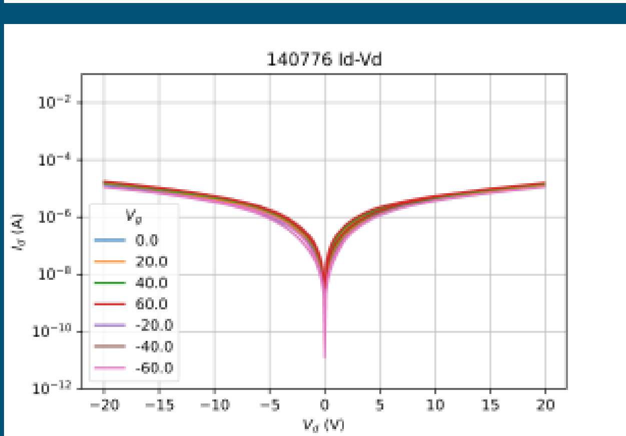
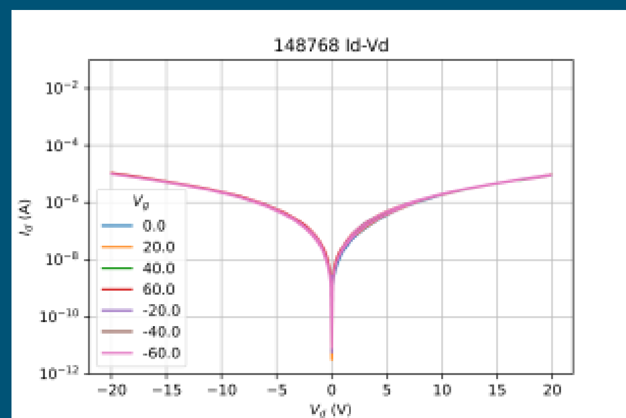
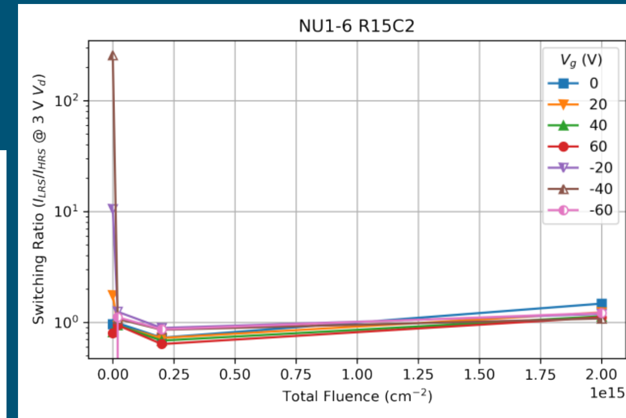
Ion Irradiation Effects on Device Performance: R15C2

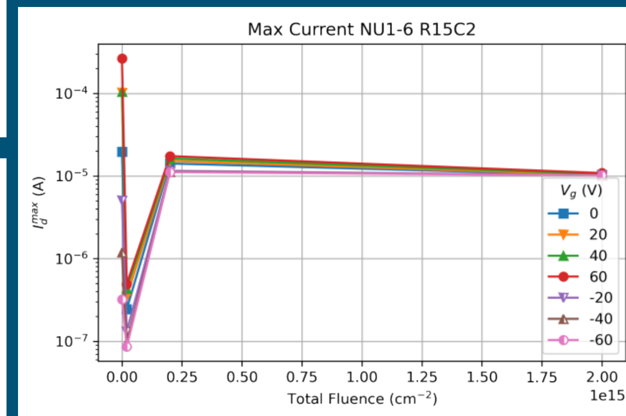


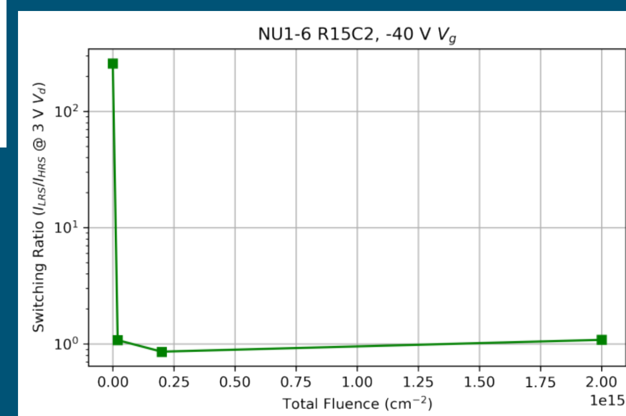




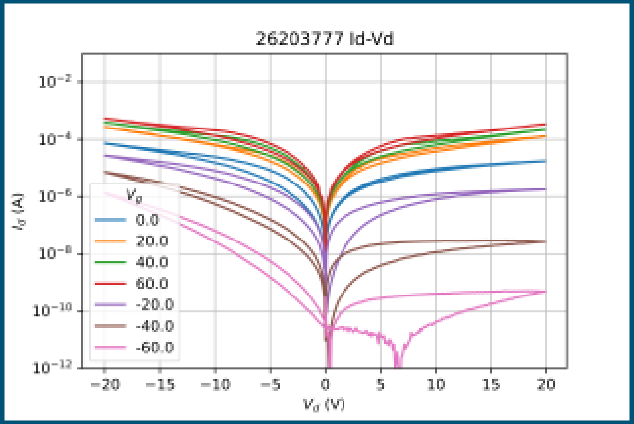


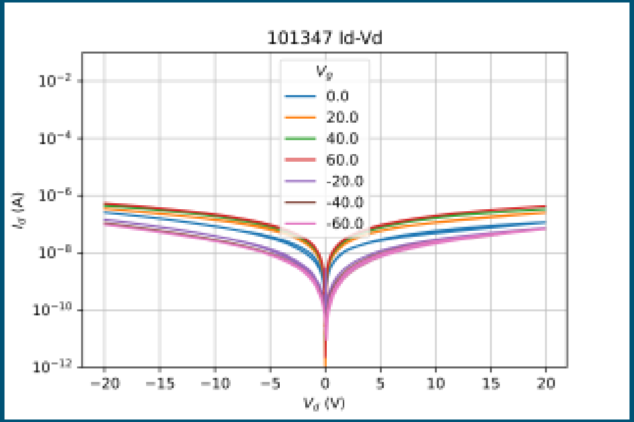


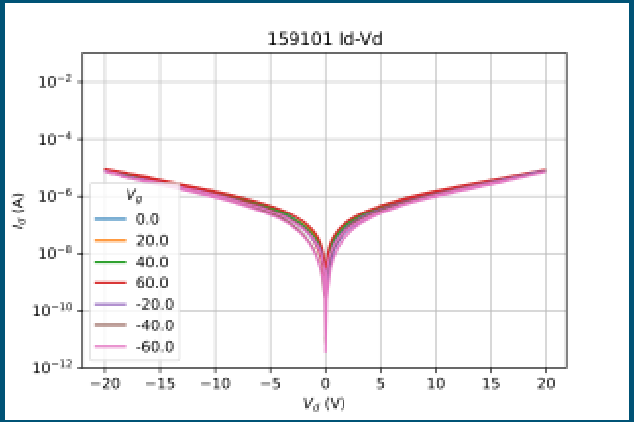


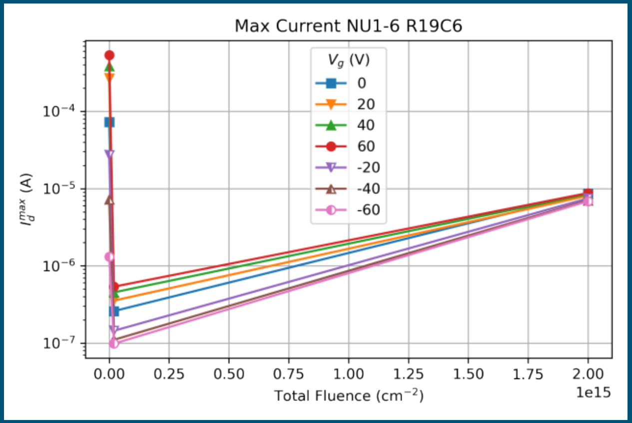


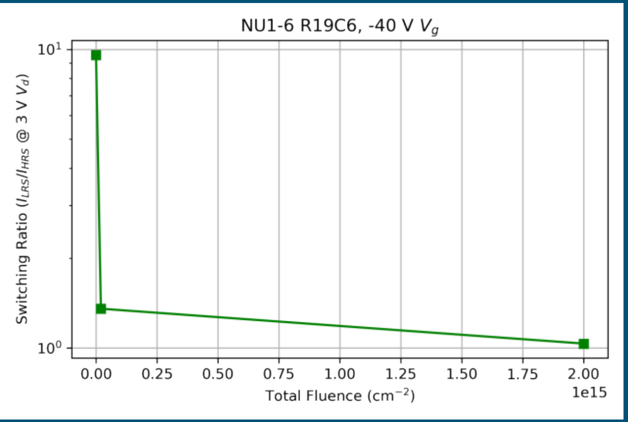
Ion Irradiation Effects on Device Performance: R19C6

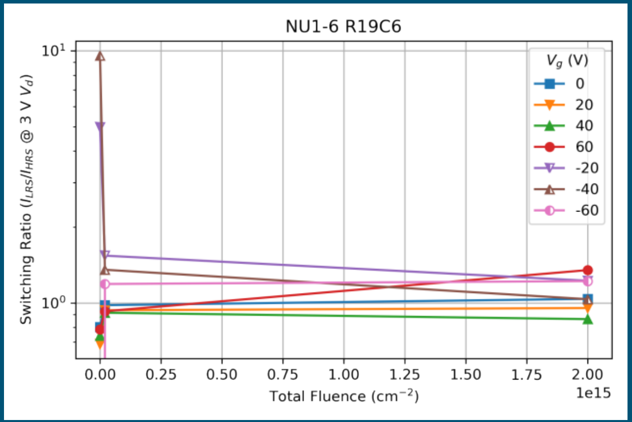












Ion Irradiation Effects on Capacitance: NU1-3

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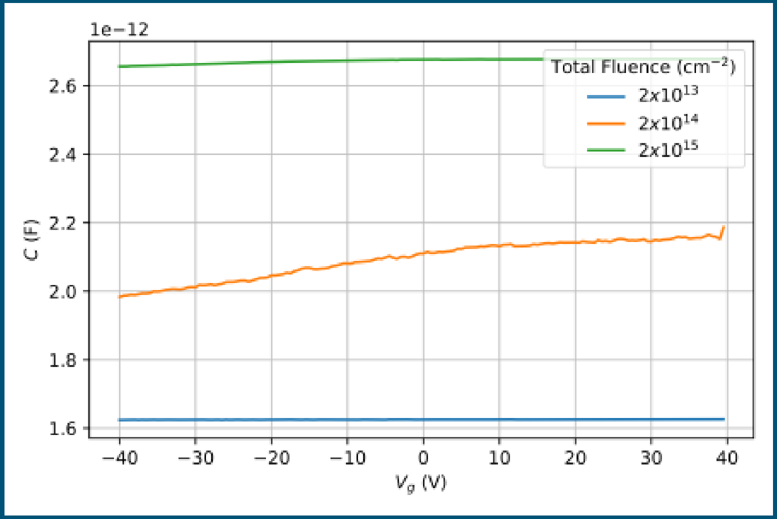

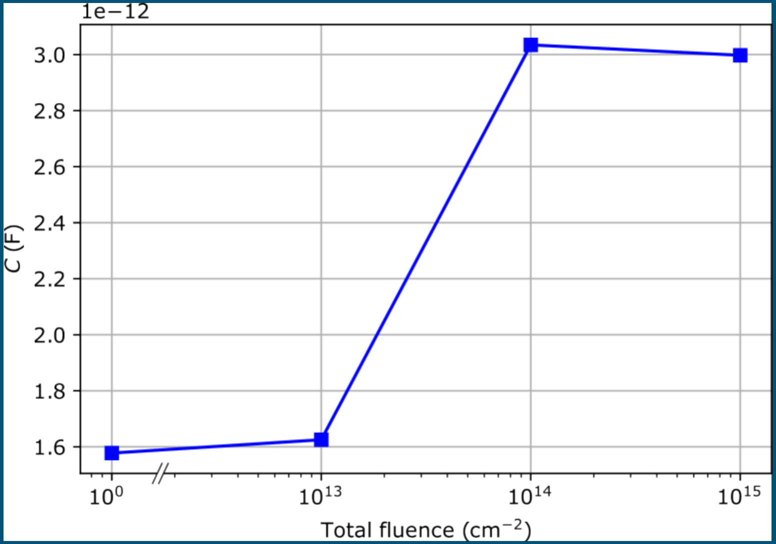

Outline

Background and Motivation

- Radiation tolerance of TMDs
- Why is the MoS_2 memtransistor interesting?

Heavy Ion Irradiation Effects

- Memtransistor behavior vs radiation fluence
- MoS_2 physical properties vs radiation fluence

Laser Irradiation Effects

- Persistent memtransistive behavior after laser irradiation
- Differences in defects created by laser and ion irradiation

Persistent Resistance Switching after Laser Irradiation: A Control Experiment

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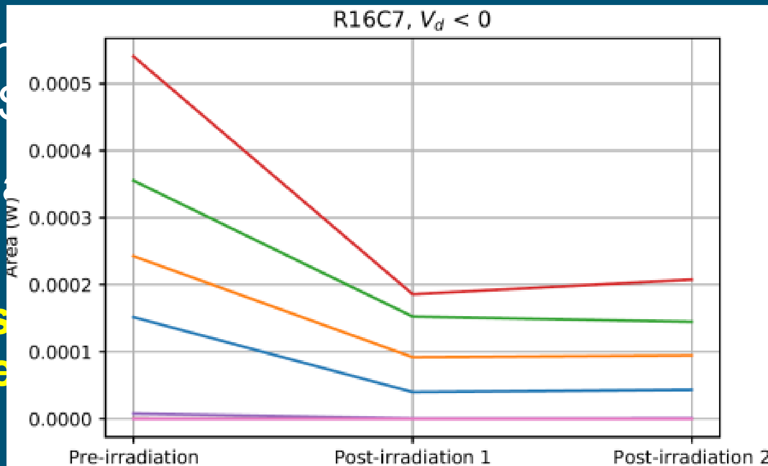
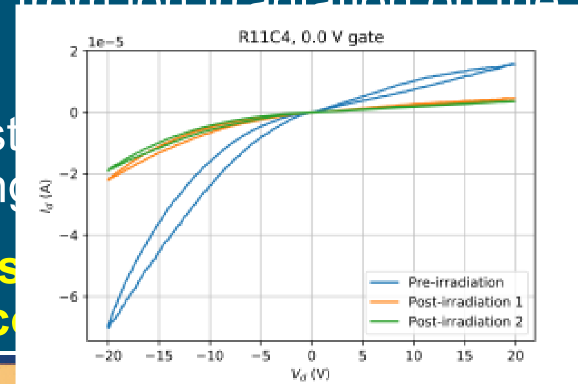
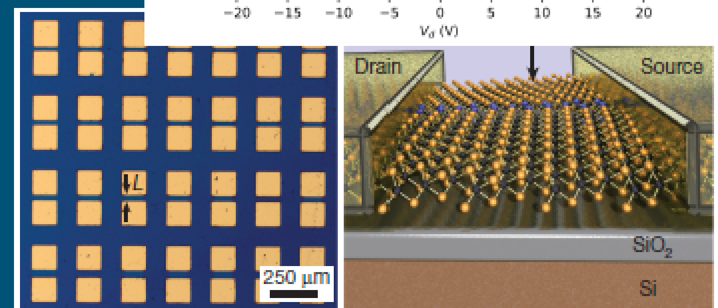






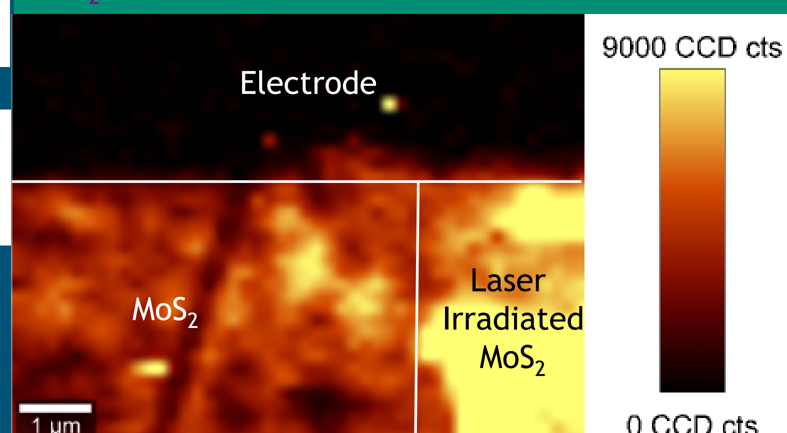
V. K. Sangwan et al., Nature 2018, 554, 500-504

Id-Vd vs Vg as a function of Au^+ fluence (one representative device)

Scatter plot: switching ratio vs Au^+ fluence

Scatter plot: Absolute HRS and LRS vs Au^+ fluence

MoS_2 Photoluminescence: Increased defect concentration and passivation



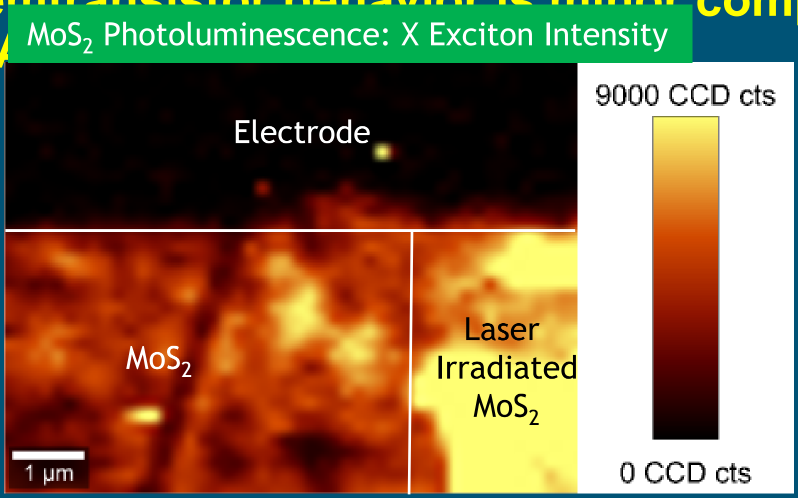
Manuscript in preparation

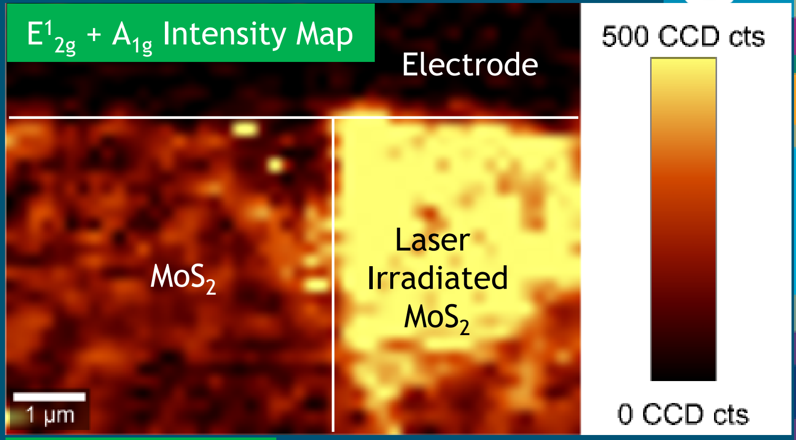
Optical Response and Defects of Laser Exposed MoS_2

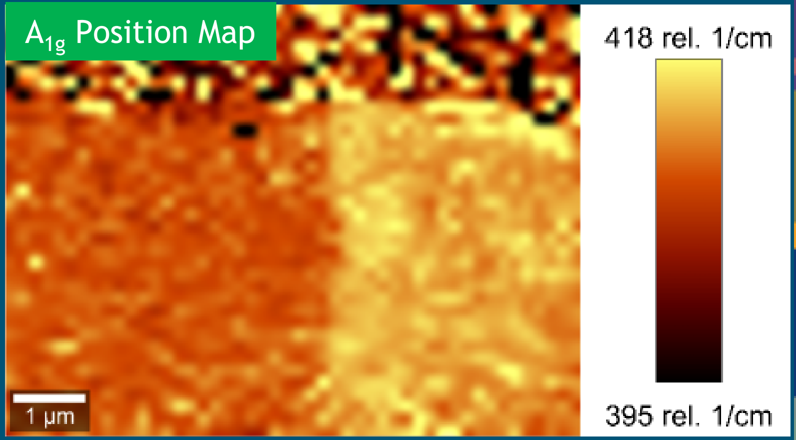
Sulfur vacancies generated by laser irradiation: blue shift and intensification of E_{2g} and A_g modes

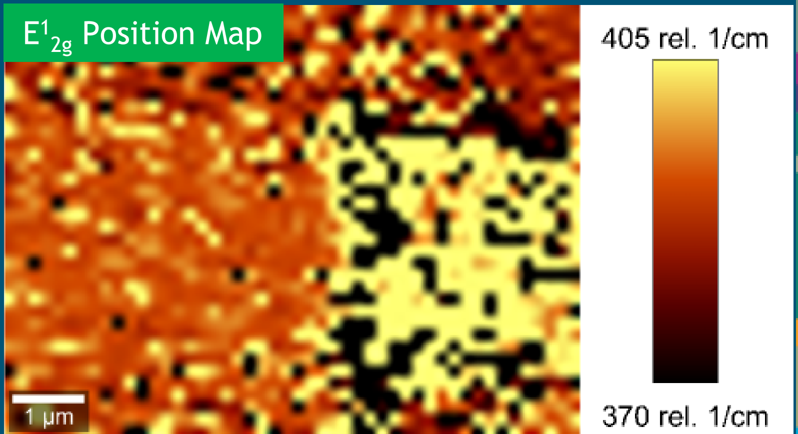
Increase in PL intensity in laser irradiated region indicates laser irradiation catalyzes vacancy passivation (consistent with previous reports)

Laser irradiation generates and passivates sulfur vacancies in MoS_2, effect on memtransistor behavior is minor compared to A









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