Ultrafast Carrier Dynamics of Exfoliated Transition Metal Dichalcogenides with Optical-Pump Terahertz-Probe Microscopy

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Atomically thin two-dimensional (2D) transition metal dichalcogenides (TMD) exhibit extraordinary physical properties similar to graphene with the distinctive feature of having an intrinsic bandgap, enabling their vast potential applications in ultrafast photonic devices and optoelectronics as semiconductors. The characterization of these materials is essential for developing such technologies and the emerging terahertz (THz) microscopy proves to be an ideal technique allowing contactless probing that reveal electrical properties of microscale structures. While the optical properties of 2D TMDs have been studied extensively, their properties with respect to near-field terahertz response are not yet as well-understood. The far-field, millimeter scale nature of standard terahertz techniques is suitable for measuring the response of bulk standard semiconductors, but a terahertz microscope further reveals the carrier dynamics of TMDs. In this study, we employ a unique, optical-pump terahertz-probe microscope to measure the response of both monolayer and bulk samples of TMD MoS_2 and WSe_2 deposited on $LiNbO_3$ crystal substrate. The thin-layer TMDs were fabricated using mechanical exfoliation, resulting in pure, high-quality single crystal samples that cannot be obtained with chemical vapor deposition methods. Using optical-pump THz-probe microscopy, we measure the decay lifetime of MoS_2 electron carriers with a temporal resolution of approximately five hundred femtoseconds in the 1 THz range to be 8.74 picoseconds and show the THz electric field response of these thin TMDs.

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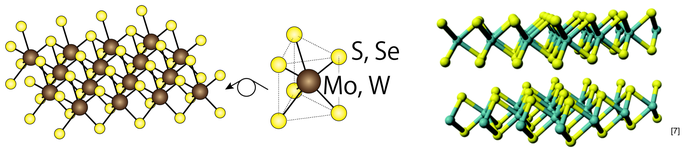
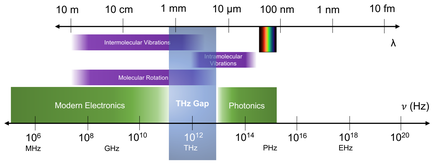
2D Materials Beyond Graphene



- TMDs are transition metal atoms covalently bonded to chalcogens (S, Se, or Te) stacked in layers by weak van der Waals interactions.
- Transition metal dichalcogenides (TMD) are atomically thin materials like graphene that are distinguished by an **intrinsic bandgap**.
- Vast potential applications in ultrafast photonic devices and **optoelectronics** as **semiconductors**, including in solar cells.

The Terahertz Frontier



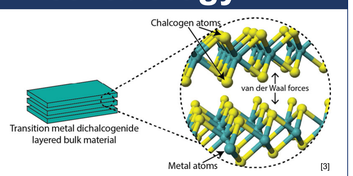
Bridges **electronics** and **photonics** where semiconductors are currently too inefficient [4].

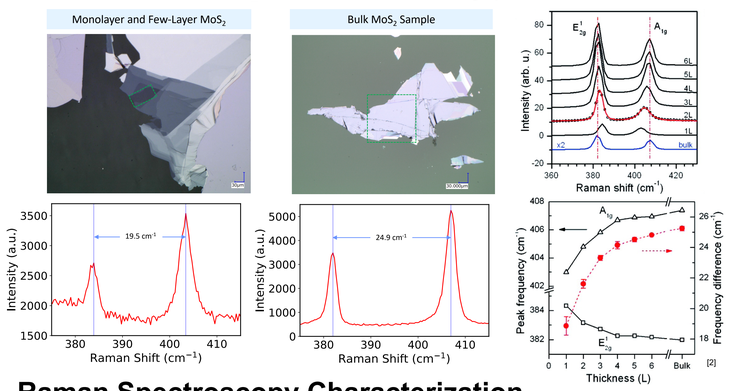
Terahertz probing enables unique access to investigating molecular interactions and **free carrier dynamics** and imaging samples normally opaque to visible and near-IR wavelengths.

TMD Fabrication Methodology

Mechanical Exfoliation

- High-quality, **single crystal** MoS_2 thin samples unobtainable with chemical vapor deposition (CVD)
- Deposited on $LiNbO_3$ (LN) elemental crystal substrate.
- Fabricated bulk MoS_2 with an indirect bandgap of 1.23 eV.



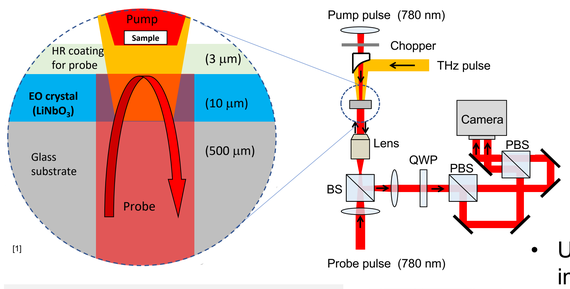


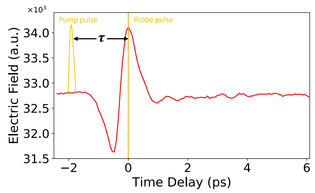
Raman Spectroscopy Characterization

- Taking Raman spectra reveals the number of layers in a sample based on molecular vibrations and Raman modes.

Near-Field Terahertz Microscopy

Spatially-Resolved Optical-Pump Terahertz-Probe Spectroscopy

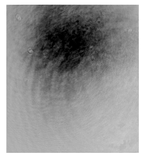




- Unique setup allows TMDs to be imaged in near-field
- Peak of THz pulse (E_{THz}) is probed
- Captured differential signal (ΔE_{THz}) between pump on and off

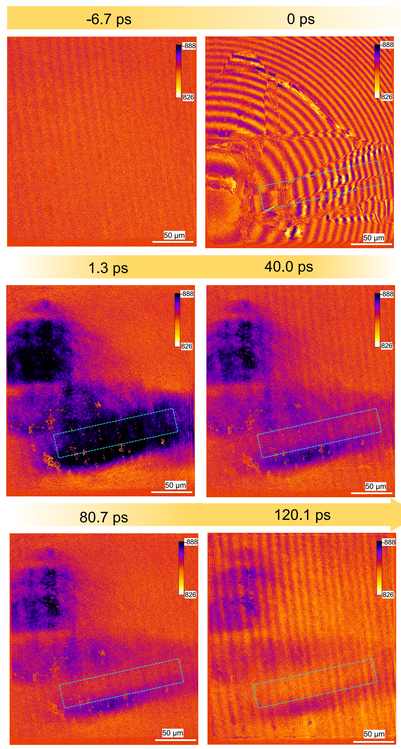
Non-destructive imaging

Spatial resolution	~ 10 μm (λ/50)
Time resolution	~ 100 fs
Pump source	~ 780 nm (1.59 eV)
Imaging dimension	~250 × 250 μm
THz pulse generated from LN crystal	

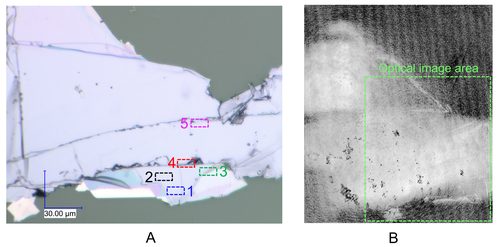


Terahertz pulse spot imaged without pump on reference LN substrate occurring at time delay of 0

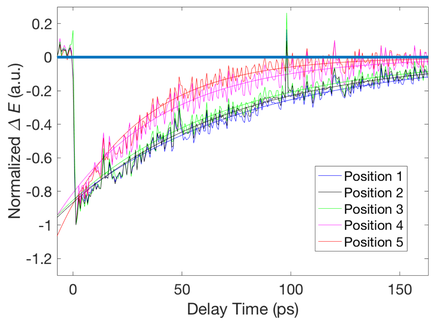
Terahertz Imaging and Free Carrier Dynamics



Nonuniform Position-Dependent Relaxation



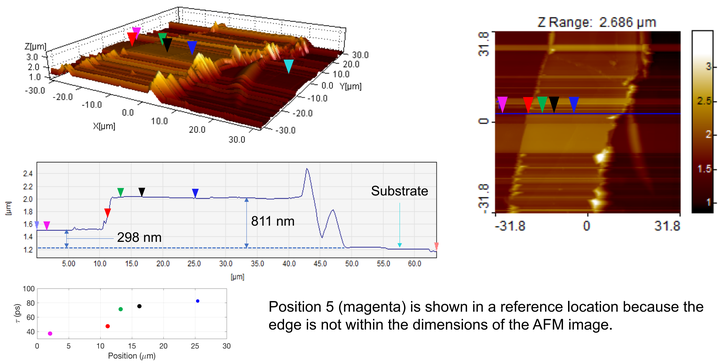
A. Optical image map of bulk MoS_2 sample, positionally annotated with the corresponding decay rates shown in the ΔE vs. delay time plot below.
B. THz image taken at ~1 ps immediately following the radial interference fringes at 0 ps, where the probe and pump pulse occur simultaneously.



Discussion

Atomic Force Microscopy (AFM)

- Determining thickness and smoothness of bulk sample.



Position 5 (magenta) is shown in a reference location because the edge is not within the dimensions of the AFM image.

- Layer thickness difference of ~510 nm observed between internal edges.
- Position 4, where an edge is located, was observed to have an especially rapid decay even when compared to the thinner region of Position 5.
- The likely combination of a thinner region and edge state may contribute to faster carrier relaxation in Position 3 [5].

Conclusions and Next Steps

- Performed near-field optical-pump terahertz-probe imaging with clear evidence of TMD sample observed.
- Demonstrated THz microscopic imaging potential for non-invasive characterization of bulk and few-layer TMDs.
- Relaxation rate of carriers may be attributed to the edge state and the thickness given a particular positional point.
- Imaging heterostructure TMDs to observe free carrier migration for photovoltaic applications.

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Acknowledgements

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