Numerical simulation of terahertz carrier dynamics in monolayer MoS_2

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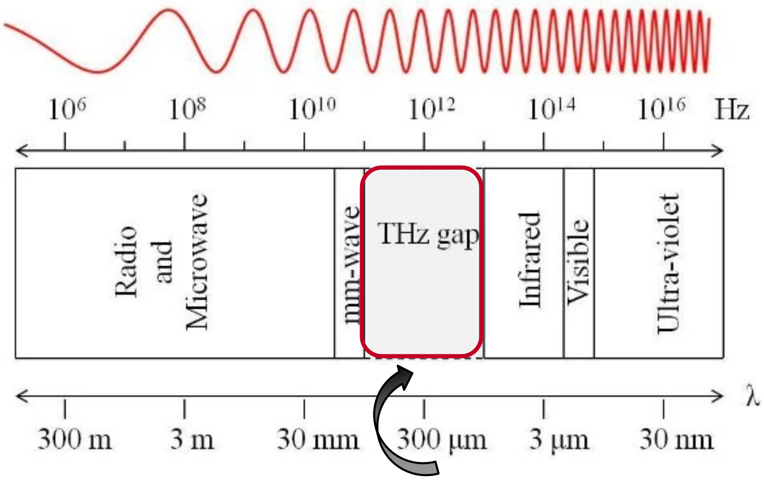
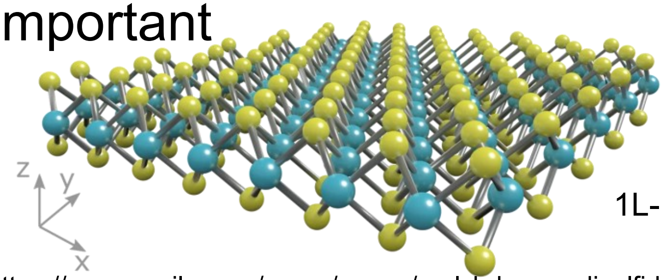
Motivation

Terahertz (THz) excitation captures the interplay between field and carrier

- Quasi-electrostatic solvers assume $\omega\tau << 1$, which becomes invalid
- Needs full electrodynamic solver
- Screening plays an important role in 2D



Most 2D semiconductors have THz scattering rate



1L-MoS_2

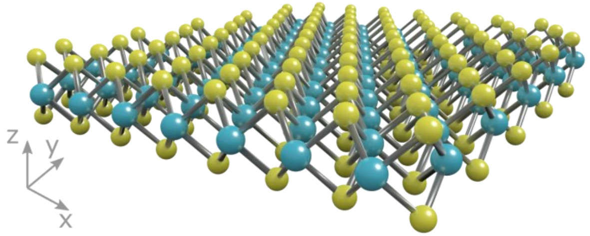
https://www.ossila.com/en-us/pages/molybdenum-disulfide-mos2

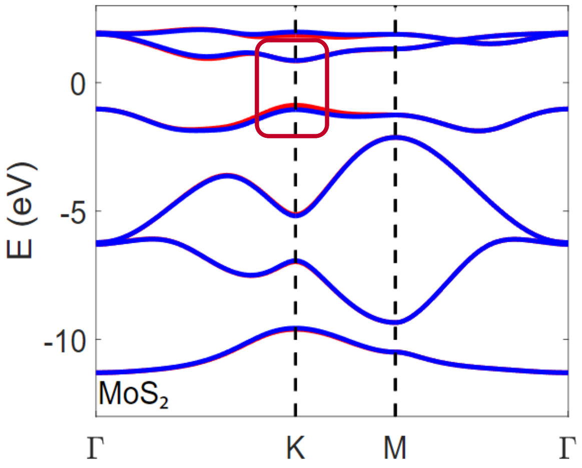
Numerical characterization of THz conductivity in 2D materials is limited

Our work

We present a multiphysics numerical tool for THz conductivity

Monolayer MoS_2 is direct band, has bandgap ~1.8 eV → no interband (optical) transitions





Band structure via 11-orbital basis tight binding method.

K-Q band separation **~200 meV**

CB K-valley m^* = $0.51m_0$

S. Mitra, L. Avazpour, and I. Knezevic, Journal of Computational Electronics, 2023. https://doi.org/10.1007/s10825-023-02023-x

Computational scheme

Ensemble Monte Carlo (EMC)

$$\frac{\partial f}{\partial t} + v\cdot\nabla_r f + \mathbf{F}\cdot\nabla_p f = \left.\frac{\partial f}{\partial t}\right|_{coll}$$

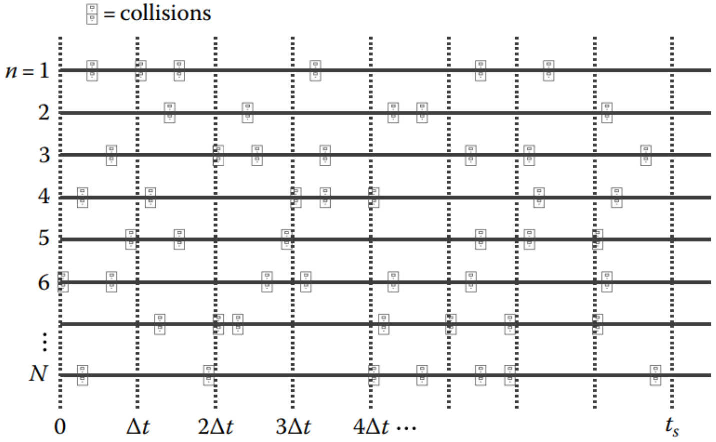
Carrier dynamics

Finite-Difference Time-Domain (FDTD)

$$\frac{\partial \mathbf{H}}{\partial t} = -\frac{1}{\mu}\nabla\times\mathbf{E} - \frac{1}{\mu}\mathbf{M}$$

$$\frac{\partial \mathbf{E}}{\partial t} = \frac{1}{\varepsilon}\nabla\times\mathbf{H} - \frac{1}{\varepsilon}\mathbf{J}$$

Electrodynamics

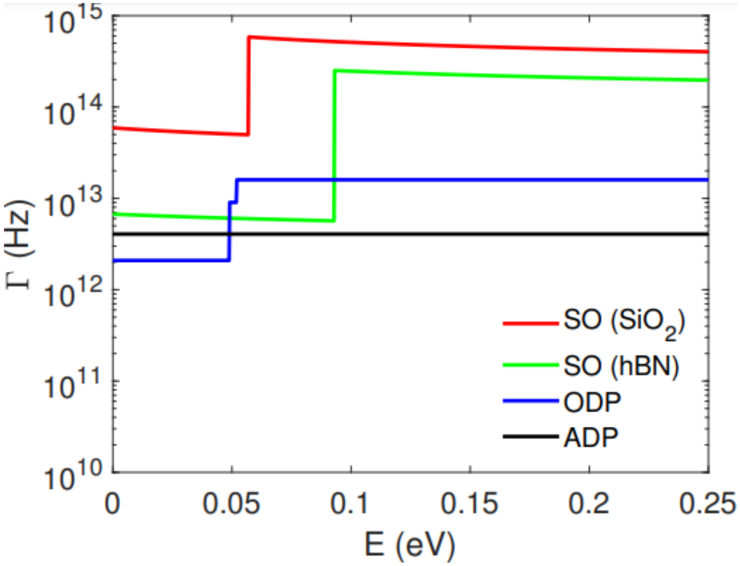


1.K. J. Willis, S. C. Hagness, and I. Knezevic, Journal of Applied Physics, vol. 110, no. 6, p. 063714, 2011.
2.N. Sule, K.J. Willis, S.C. Hagness, and I. Knezevic, J Comput Electron 12:563–571, 2013.

Computational scheme

EMC:

- Employs semiclassical scattering rates



Scattering mechanisms:

☐ Acoustic phonon

☐ Optical phonon

☐ Surface optical (SO) phonon

☐ **Ionized impurities**

Real space distribution

$$\Gamma_{\mathrm{ac}}(k) = \frac{D_{\mathrm{ac}}^2 k_B T m^*}{\hbar^3 \rho_s v_s^2}$$

$$\Gamma_{\mathrm{op}}(k) = \frac{D_{\mathrm{op}}^2 g_d m^*}{2\hbar^2 \rho_s \omega_{\mathrm{op}}}\left[N_q + (N_q + 1)\Theta(E - \hbar\omega_{\mathrm{op}})\right]$$

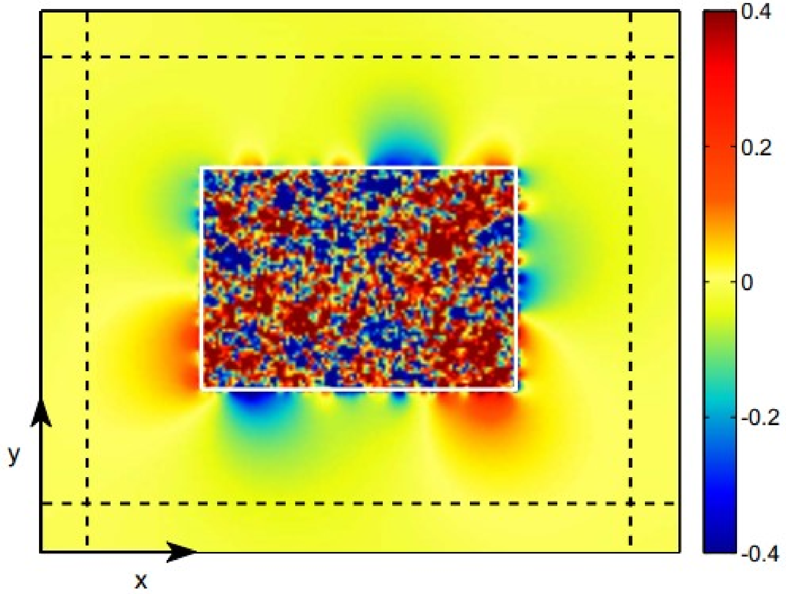
$$\Gamma_{\mathrm{so}}(k) = \frac{32\pi^3 e^2 F_v^2 m^* S}{\hbar^3 a^2}\left(N_q + \frac{1}{2} \pm \frac{1}{2}\right)\times$$

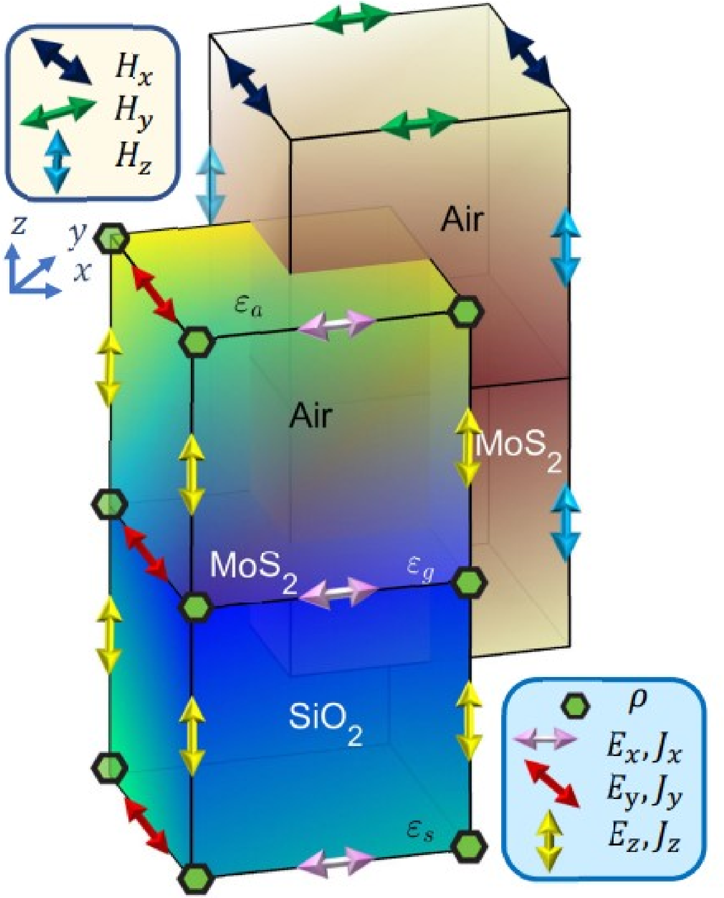
$$\int_{-\pi}^{\pi} \frac{1}{q} \frac{\sinh\left(\frac{aq}{2}\right)}{\left(4\pi^2 q + a^2 q^3\right)^2} \mathrm{d}\theta$$

Computational scheme

FDTD:

- Accurate high-frequency analyses
- Yee grid implementation
- Convolutional perfectly matched layer (CPML)
- Total-field scattered-field (TFSF) source[1]





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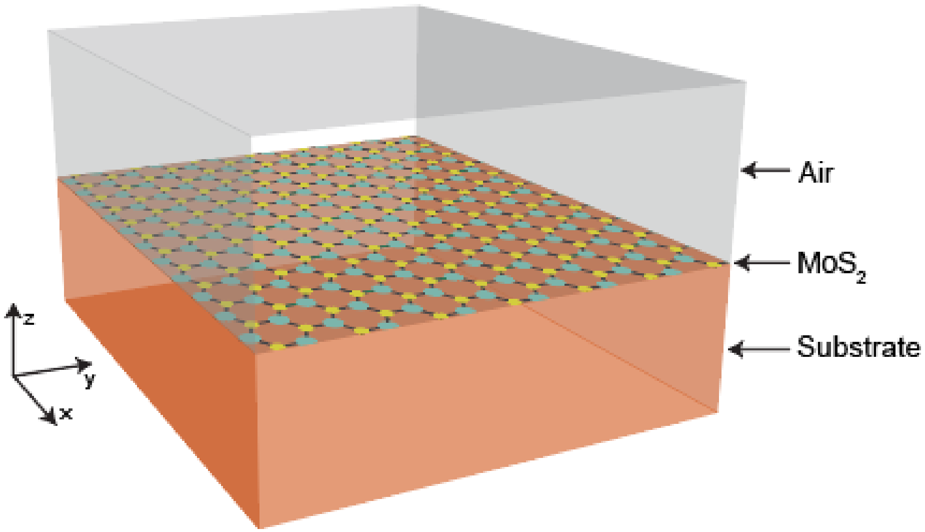
[1] K. J. Willis, S. C. Hagness, and I. Knezevic, Journal of Applied Physics, vol. 110, no. 6, p. 063714, 2011

Simulation domain

- Periodic boundary condition in xz and yz planes
- Pauli exclusion principle
- Real space distribution ions and electrons
- TFSF source E_y
- CPML boundary condition in xy planes



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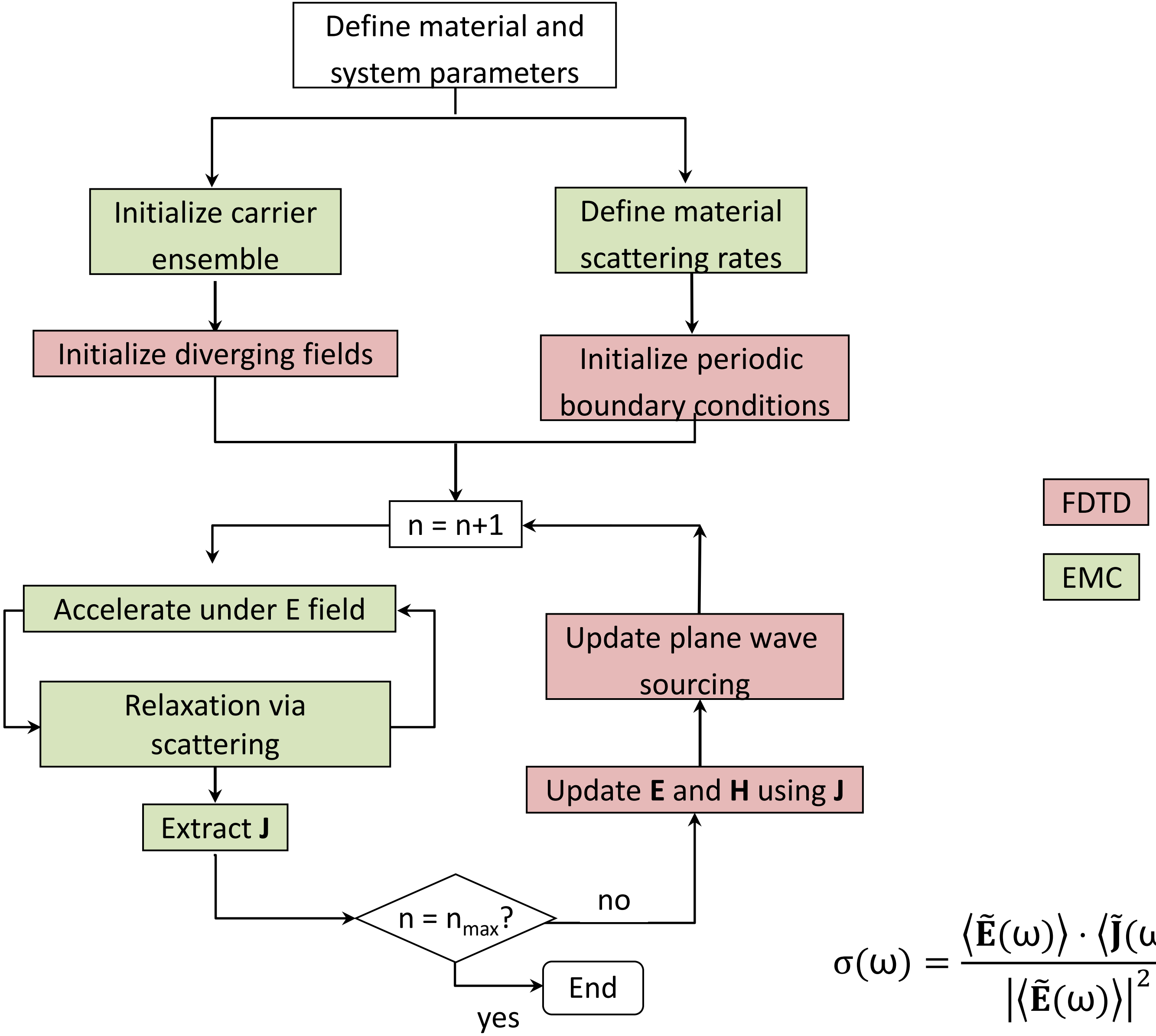
Process



Define material and system parameters

Initialize carrier ensemble

Define material scattering rates

Initialize diverging fields

Initialize periodic boundary conditions

n = n+1

Accelerate under E field

Relaxation via scattering

Extract **J**

Update plane wave sourcing

Update **E** and **H** using **J**

n = n_{max}?

no

yes

End

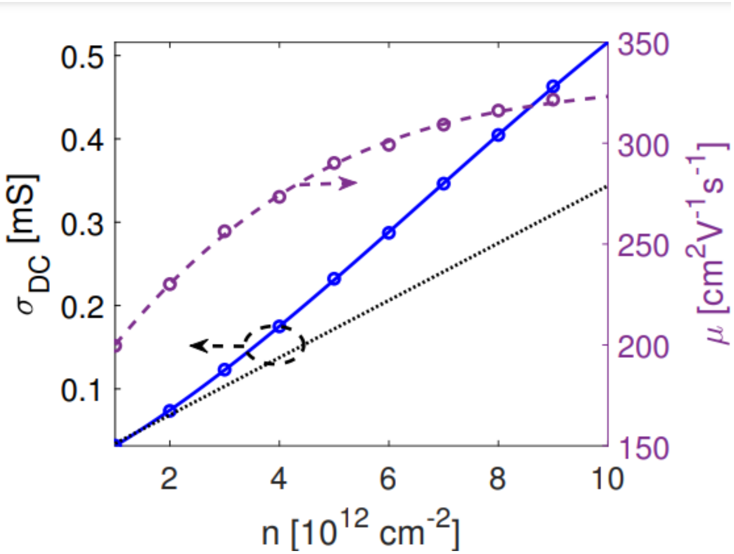
FDTD

EMC

$$\sigma(\omega) = \frac{\langle \tilde{\mathbf{E}}(\omega) \rangle \cdot \langle \tilde{\mathbf{J}}(\omega) \rangle}{\left| \langle \tilde{\mathbf{E}}(\omega) \rangle \right|^2}$$

Results

σ_{dc} vs n:



- At low doping density, transport limited by ionized impurity scattering
- At high n, screening becomes important
- Mostly carrier independent μ i.e., linear conductivity
- Almost full screening of impurities for carrier-to-impurity ratio of 10

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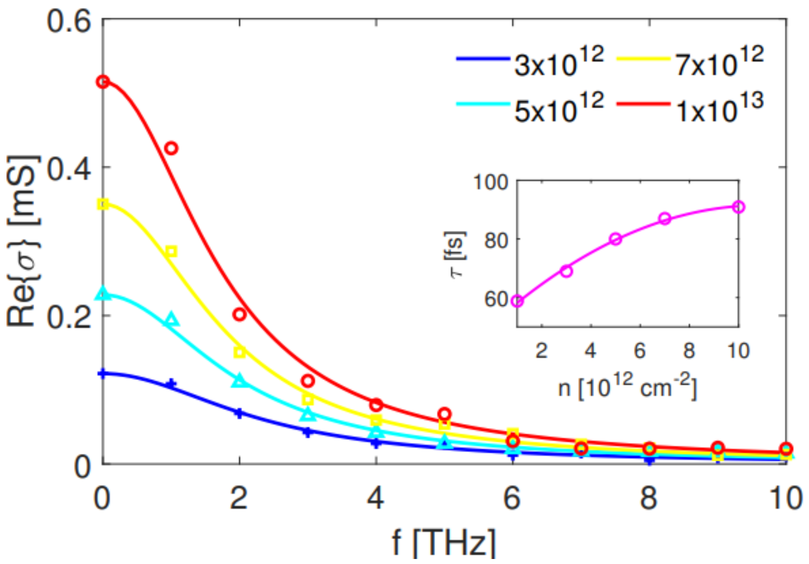
Results



- 2D material with m^* → weak E dependent scattering → Drude model

$$\sigma(\omega) = \frac{\sigma_{\mathrm{dc}}}{1 - i\omega\tau}$$

- σ_{dc} limited by impurity at low n, phonon at high n

- Effective relaxation time τ approaches impurity free value of 90 fs

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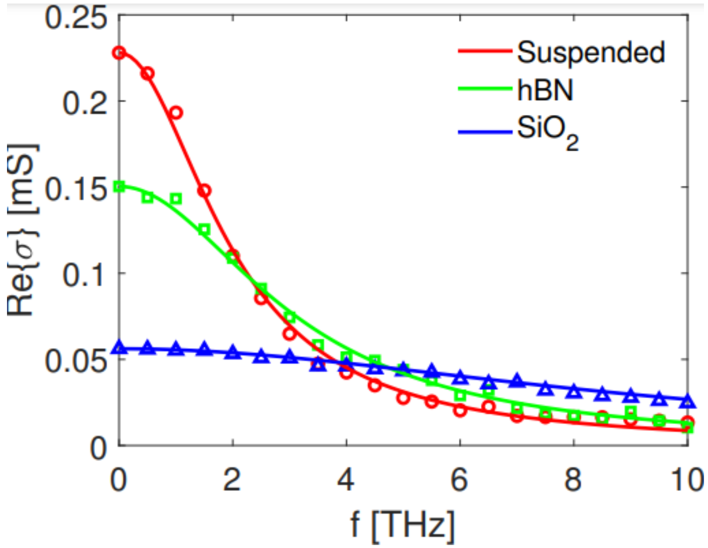
Results



- Suspended MoS_2 → No dominant scattering mechanism
- Supported MoS_2 → SO phonon scattering dominates
- Weak polar HBN substrate → higher conductivity at low THz
- However, strong polar SiO_2 → retains σ at high THz

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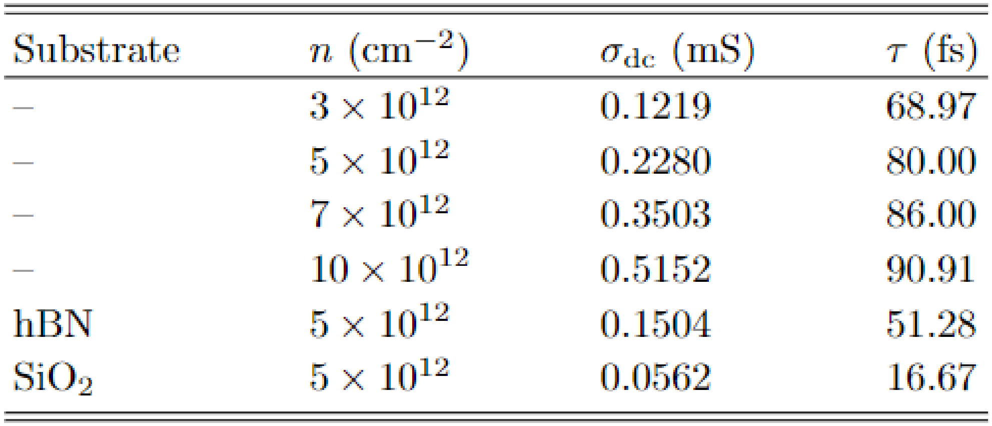
Fitting parameters



Substrate	n (cm^{-2})	σ_{dc} (mS)	τ (fs)
–	3×10^{12}	0.1219	68.97
–	5×10^{12}	0.2280	80.00
–	7×10^{12}	0.3503	86.00
–	10×10^{12}	0.5152	90.91
hBN	5×10^{12}	0.1504	51.28
SiO_2	5×10^{12}	0.0562	16.67

For suspended MoS_2,

$$\mu_{\mathrm{dc}} = -1.683n^2 + 31.59n + 172.8 \quad cm^2V^{-1}s^{-1}$$

$$\tau = -0.34n^2 + 7.4n + 51.14 \quad fs \qquad 10^{12} \leq n \leq 10^{13}cm^{-2}$$

For HBN & SiO_2 supported MoS_2, τ and μ are ~1.5 and ~5 times smaller.

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Conclusion

- EMC-FDTD numerical solver couples electrodynamics with carrier dynamics
- Tune impurity density, spatial impurity distribution, and carrier density
- Useful for extracting many body effects from experiments
- Useful for substrate selection for THz operation
- Can be readily extended to other TMDs and 2D materials

Thanks!