Numerical simulation of terahertz carrier dynamics in monolayer MoS_2

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I. Introduction

Among the rapidly advancing two-dimensional (2D) quantum materials, transition-metal dichalcogenides (TMDs) are attractive for their promising electronic properties [1], [2]. Monolayer molybdenum disulfide (MoS_2) is particularly suitable for electronic and optoelectronic applications, as it is a direct bandgap material, with additional advantages such as high on–off current ratio and speed [3]. The complex conductivity $\sigma(\omega)$, where ω is the angular frequency, is an important quantity that characterizes the electrical transport of materials under electrical and electromagnetic sources. The terahertz (THz) frequency range is particularly interesting because most 2D materials including TMDs have scattering rates in this range. The interplay between carrier transport and electromagnetic fields cannot be approximated by traditional time-dependent device simulation techniques, where electronic solvers are coupled with a quasi-electrostatic Poisson's solver because at THz frequencies, $\omega\tau \approx 1$, where τ is the effective carrier lifetime, makes the quasi-electrostatic approximation inaccurate.

In this paper, we analyze the frequency-dependent low-field THz-frequency electrical conductivity of monolayer MoS_2 both suspended and supported on different substrates [4]. Our numerical simulation tool employs the ensemble Monte Carlo (EMC) technique, widely used for diffusive carrier transport simulation, with the finite-difference-time-domain (FDTD) technique as the electromagnetic field solver. The EMC and FDTD source each other, where EMC incorporating various scattering mechanisms and updates charge and current densities based on calculated fields from FDTD, and FDTD updates fields based on the charge and current distributions from EMC. The EMC–FDTD technique was previously used to compute the THz conductivity of bulk silicon and graphene sheets [5], [6].

II. Numerical Model

Figure 1 shows the numerical simulation domain. The domain consists of a single layer of MoS_2 with air in the top half and a dielectric in the bottom half. For the suspended case, the dielectric is also air, and for the supported case, we considered commonly used SiO_2 and hBN substrates. The horizontal planes at the top and top are terminated via convolutional perfectly matched layer (CPML) boundary conditions and the vertical planes via periodic boundary conditions to simulate a large semiconductor. Carrier transport is limited by acoustic and optical phonon scattering, as well as long-range Coulomb interaction with ionized impurities present in the MoS_2 layer and the substrate. The scattering rates are calculated under the deformation potential approximation with the effective mass extracted from tight-binding calculations. The time-dependent fields and currents are Fourier transformed to obtain the conductivity using

$$\sigma(\omega) = \frac{\tilde{\mathbf{E}}(\omega) \cdot \tilde{\mathbf{J}}^*(\omega)}{\left|\tilde{\mathbf{E}}(\omega)\right|^2}, \tag{1}$$

where $\tilde{\mathbf{E}}(\omega)$ and $\tilde{\mathbf{J}}(\omega)$ are the spatially averaged steady-state electric-field and current-density phasors.

We demonstrate the application of our technique here. Figure 2 shows the real part of $\sigma(\omega)$ for suspended MoS_2 at different carrier densities. The conductivity shows Drude-type behavior. $\mathrm{Re}[\sigma(\omega)]$ increases at high carrier densities while the effective lifetime approaches the impurity-free value taking the screening of ionized impurities into account. Figure 3 shows the conductivity when the sample is placed on substrates. Remote phonon scattering lowers the conductivity. However, MoS_2 on a highly polar SiO_2 has a higher conductivity than on hBN at high frequencies and an overall flatter frequency dependence.

III. Conclusion

In conclusion, we presented a coupled EMC–FDTD technique to simulate terahertz frequency dynamics of monolayer MoS_2. This macroscopic simulation tool can be effective to understand frequency-dependent electronic transport and substrate effects on emerging 2D materials.

References

[1] G. Fiori, F. Bonaccorso, G. Iannaccone, T. Palacios, D. Neumaier, A. Seabaugh, S. K. Banerjee, and L. Colombo, "Electronics based on two-dimensional materials," *Nature Nanotechnology*, vol. 9, no. 10, pp. 768–779, Oct. 2014. [Online]. Available: https://doi.org/10.1038/nnano.2014.207

[2] D. Lembke, S. Bertolazzi, and A. Kis, "Single-layer $\mathrm{mos_2}$ electronics," *Accounts of Chemical Research*, vol. 48, no. 1, pp. 100–110, Jan. 2015. [Online]. Available: https://doi.org/10.1021/ar500274q

[3] M. Amani, M. L. Chin, A. G. Birdwell, T. P. O'Regan, S. Najmaei, Z. Liu, P. M. Ajayan, J. Lou, and M. Dubey, "Electrical performance of monolayer $\mathrm{MoS_2}$ field-effect transistors prepared by chemical vapor deposition," *Applied Physics Letters*, vol. 102, no. 19, p. 193107, May 2013. [Online]. Available: https://doi.org/10.1063/1.4804546

[4] S. Mitra, L. Avazpour, and I. Knezevic, "Terahertz conductivity of monolayer MoS_2," *Journal of Computational Electronics*, 2023, in press.

[5] K. J. Willis, S. C. Hagness, and I. Knezevic, "Terahertz conductivity of doped silicon calculated using the ensemble monte carlo/finite-difference time-domain simulation technique," *Applied Physics Letters*, vol. 96, no. 6, p. 062106, 2010. [Online]. Available: https://doi.org/10.1063/1.3308491

[6] N. Sule, K. J. Willis, S. C. Hagness, and I. Knezevic, "Terahertz-frequency electronic transport in graphene," *Physical Review B*, vol. 90, no. 4, Jul. 2014. [Online]. Available: https://doi.org/10.1103/physrevb.90.045431

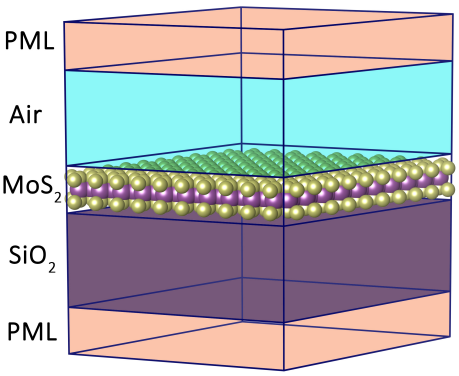

Fig. 1. Schematic of the three-dimensional (3D) simulation geometry of monolayer MoS_2 between air on top and SiO_2 substrate on bottom. The vertical boundaries have periodic boundary conditions.

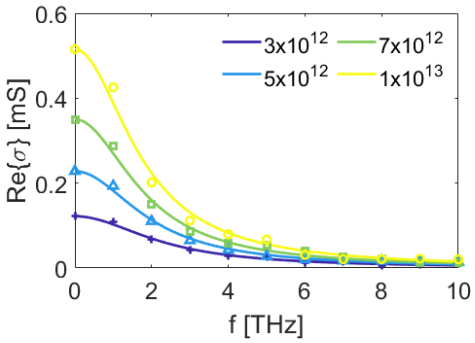

Fig. 2. Real part of $\sigma(\omega)$ as a function of terahertz frequency for different electron densities in suspended monolayer MoS_2. The impurity density throughout is 10^{12} cm^{-2}. The points are numerically calculated and the solid lines are Drude fits.

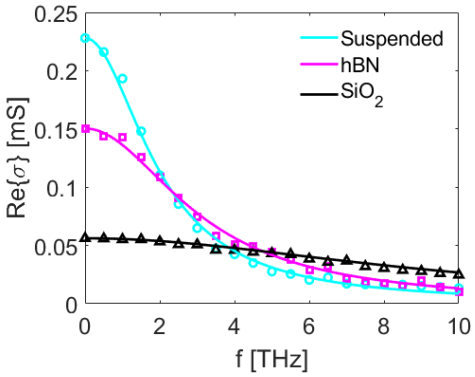

Fig. 3. Real part of $\sigma(\omega)$ versus frequency for MoS_2 that is suspended, supported on hBN, and supported on SiO_2.