Simulation of ac conductivity of monolayer MoS_2 at terahertz frequencies

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***Abstract*—We present a multiphysics numerical tool for calculating the terahertz (THz) conductivity of transition-metal dichalcogenides (TMDs). The tool combines the ensemble Monte Carlo (EMC) technique for carrier transport with a three-dimensional finite-difference-time-domain (FDTD) solver for electromagnetic fields. We use the coupled EMC–FDTD technique to calculate the frequency-dependent conductivity in the terahertz range for monolayer MoS_2, both for suspended layers and for layers supported on the commonly used silicon-dioxide substrate.**

I. Introduction

Among recent advancements in two-dimensional (2D) materials, transition-metal dichalcogenides (TMDs) are promising for electronic and optoelectronic applications. Monolayer MoS_2 is an often-studied TMD because of its direct bandgap, high current on–off ratio, and high speed [1], [2]. Though considerable work has been done in the first-principles analysis [3], [4], low-field, and high-field dc electronic transport [5], frequency-dependent transport, which plays a significant role in technologies such as photodetectors, sensors, and high-frequency devices, has not been extensively studied.

In conventional time-dependent device simulation, an electronic transport solver is usually coupled with a quasi-electrostatic Poisson's equation solver in order to incorporate field effects. However, at THz frequencies, the quasi-electrostatic approximation is no longer accurate [6]. Thus, a deeper understanding of carrier–field interaction is needed to model carrier transport under the influence of ac electromagnetic fields.

In this paper, we analyze the frequency-dependent low-field conductivity of monolayer MoS_2 in the terahertz range, where no interband transitions are possible. Our numerical simulation tool combines the ensemble Monte Carlo (EMC) technique for describing diffusive carrier transport with the finite-difference-time-domain (FDTD) technique for solving Maxwell's curl equations. FDTD is accurate in high-frequency analyses where the quasistatic approximation for electromagnetic fields fails. The EMC and FDTD solvers source each other: FDTD calculates fields from charge and current distributions, while EMC calculates charge and current densities based on microscopic scattering mechanisms and the fields from FDTD that accelerate the carriers. This multiphysics EMC–FDTD technique, combined with molecular dynamics to account for short-range interaction, was previously used to accurately model the terahertz conductivity of bulk silicon [7] and 2D graphene sheets [8].

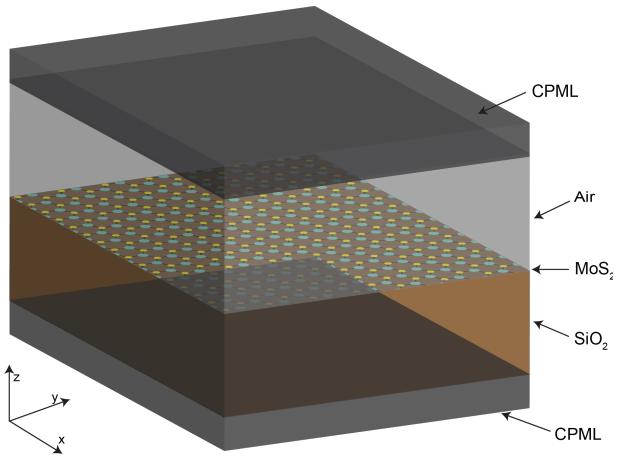

Fig. 1. Three-dimensional (3D) simulation geometry. A TEM wave is used to excite the MoS_2 layer. The four vertical boundary planes are terminated by periodic boundary conditions, while the top and bottom boundaries have convolutional perfectly matched layer (CPML) absorbing boundary conditions.

II. Numerical Model

The simulation domain is shown schematically in Fig. 1. The domain consists of a monolayer MoS_2 with air in the top half and a SiO_2 substrate in the bottom half. We also considered suspended MoS_2, where the bottom half is air. The dielectric constants are ϵ_{semi} = 7.5, ϵ_a = 1, ϵ_{sub} = 3.9. In order to simulate a very large sheet of the semiconductor, we used periodic boundary conditions in the vertical planes (the *x*–*z* and *y*–*z* planes in Fig. 1). The horizontal planes at the top and bottom are terminated via convolutional perfectly matched layer (CPML) absorbing boundary conditions.

The dominant scattering mechanisms at room temperature in monolayer MoS_2 are nonpolar acoustic phonon scattering, optical phonon scattering (both nonpolar and polar), as well as surface-optical-phonon (SO) scattering with surface phonons from the polar substrate. Moreover, there is impurity scattering coming from the material itself and from the substrate. The scattering rates are calculated using Fermi's golden rule. The coupling constants for acoustic and optical-phonon scattering are based on the parameters from first-principle calculations

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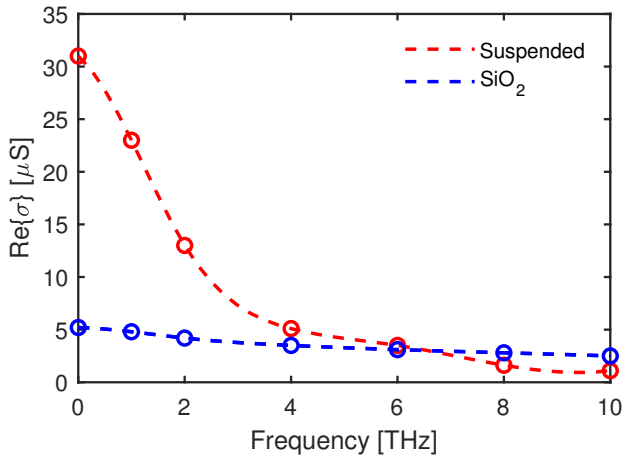

Fig. 2. Preliminary results for the real part of the complex conductivity of MoS_2 monolayers versus frequency as obtained from the combined EMC–FDTD solver. The sheet carrier density is 10^{11}cm^{-2}.

[3], [4]. The electron–SO phonon scattering rates are calculated using the interaction Hamiltonian theory [9]. In our calculations, we considered that all electrons reside in the K valleys. Although additional Q valleys are argued to be energetically relevant, the K–Q energy separation is not yet well established [10]. Moreover, at low fields (0.1 kV/cm in our simulation), election occupation of the Q valleys is likely negligible.

The complex conductivity is calculated from

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

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

III. Results

In Fig. 2, we show preliminary data for the real part of the complex conductivity as a function of frequency for suspended MoS_2 and for MoS_2 on the silicon-dioxide dielectric substrate. Static (dc) conductivity of suspended MoS_2 is 31 μS in this case, which corresponds to the electron mobility around 310 cm^2/Vs. These values are consistent with the results of analytic BTE solution and other Monte Carlo simulations [3], [4]. While the frequency-resolved electron transport in suspended MoS_2 is limited by intrinsic phonons, we observe that the substrate plays a critical role in carrier transport when SO scattering is included. At frequencies below 6 THz, this effect is quite pronounced. However, at high terahertz frequencies, the supported structure appears to exhibit higher conductivity than its suspended counterpart. The conductivity in suspended MoS_2 does not depend on frequency as strongly as it does in the supported case. While this is still preliminary data, we note that the conductivity magnitude is comparable with experimental results [11]. Including the effects of short-range interactions, carrier screening, and distribution of impurities will further improve conductivity predictions.

IV. Conclusion

In conclusion, we have developed a multiphysics EMC–FDTD numerical solver that couples electrodynamics with carrier dynamics. We have used the tool to calculate the complex conductivity of monolayer MoS_2 in the terahertz frequency range. The tool can be used to independently tune the carrier density, impurity density, as well as the real-space distribution of impurities. Numerical characterization of MoS_2 may be useful for determining the values of experimental parameters that are hard to measure directly. Furthermore, the technique can be readily extended to predict the terahertz conductivity of other promising TMDs and 2D materials.

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