Modeling the Physical Properties of Two-Dimensional Nanomaterials

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***Abstract*— In our presentation, I will review work done in enabling the development of two-dimensional nanomaterials for optoelectronic applications. The focus will be on the materials and the theories available for modeling them. I will start with a survey of the nanomaterials fabricated and predicted to exit, together with a discussion of their physical properties. This will be followed by a few examples of optoelectronic devices designed using these materials. Finally, we will present an overview of the theoretical tools developed to study electrons and phonons in these materials.**

I. 2D NANOMATERIALS

It is well-known that the first two-dimensional (2D) material was graphene [1]. The other most studied 2D material is the class of transition metal dichalcogenides (TMD), with the best known example being MoS_2. Other compound 2D materials, such as BN, have also been studied. Lesser known is the whole series of elemental 2D materials such as silicene [2], germanene, stannene, phosphorene, and borophene that have been fabricated [3].

II. OPTOELECTRONIC DEVICES

Of the above 2D materials, only phosphorene, BN and the TMD's have a band gap. Hence, they are prime candidates for optoelectronic devices. Nonetheless, the others can have a gap introduced via a variety of mechanisms such as finite-size (i.e., nanoribbons) and on a substrate.

Use as a photodetector has been the most studied optoelectronic device. An example of a study of using photonic integration to enhance the interaction of light and a 2D material is shown in Fig. 1. Other optoelectronic devices will be presented.

One of the most sophisticated codes for computing device properties ia the Atomistix Toolkit (ATK) one from Quantum Wise. An example device simulation is shown in Fig. 2. Electron transport through two-probe devices can be simulated.

III. ELECTRON THEORY

The electronic properties have been computed using both first-principles and empirical methods. First-principles methods used were invariably density-functional theory (DFT) based. These are known to be computationally intensive, particularly for modeling devices. Nonetheless, they have been used to modeled, e.g., graphene nanoribbon transistors using the ATK software. It is well-known that the electronic properties are subject to the band-gap problem, which therefore makes it difficult to really predict the optical properties.

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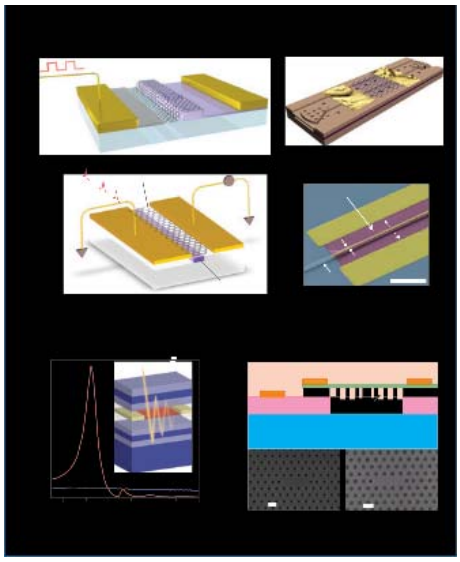

Fig. 1. Photocurrent study of a graphene device integrated with a silicon cavity. From [4].

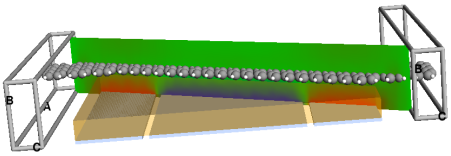

Fig. 2. A graphene nanoribbon transistor modeled using ATK software. From http://docs.quantumwise.com/manuals/Introduction.html.

For both of the above reasons, empirical techniques are much more preferable for modeling optoelectronic devices. The method of choice is the $k \cdot p$ method [5], though occasionally the tight-binding method is used. A number of applications of the theory have been made to 2D materials. The earliest was, of course, for graphene with its linear band structure near the Fermi energy. For example [6],

$$\begin{aligned}\mathcal{H}_i &= a_{61}\left(k_y\mathbf{J}_x + k_x\mathbf{J}_y\right) + a_{11}\left(k_x^2 + k_y^2\right)\\ &+ a_{62}\left[(k_y^2 - k_x^2)\mathbf{J}_x + 2k_xk_y\mathbf{J}_y\right], \qquad (1)\end{aligned}$$

where the J_i are pseudospin matrices. This has been subsequently extended to other materials such as phosphorene [7]:

$$\mathcal{H} = \mathcal{H}_i + \mathcal{H}_e, \qquad (2)$$

$$\mathcal{H}_e = \mathcal{H}^\epsilon + \mathcal{H}^E + \mathcal{H}^B + \mathcal{H}^{mix}, \qquad (3)$$

$$\mathcal{H}_i = a_1k_x^2 + a_2k_y^2 + \textstyle\sum_{i\le j} a_{ij}k_i^2k_j^2 + \ldots, \qquad (4)$$

$$\begin{aligned}\mathcal{H}^\epsilon &= \left(e_1 + e_2k_x^2 + e_3k_y^2\right)\epsilon_{xx} + \left(e_5 + e_6k_x^2 + e_7k_y^2\right)\epsilon_{yy}\\ &+ \left(e_9 + e_{10}k_x^2 + e_{11}k_y^2\right)\epsilon_{zz} + \ldots, \qquad (5)\end{aligned}$$

$$\mathcal{H}^E = \left(c_4 + c_5k_x^2 + c_6k_y^2\right) E_x^2 + \left(c_8 + c_9k_x^2 + c_{10}k_y^2\right) E_y^2 + \left(c_{12} + c_{13}k_x^2 + c_{14}k_y^2\right) E_z^2 + \ldots, \quad (6)$$

$$\mathcal{H}^B = b_4k_xB_x + b_5k_yB_y + \left(b_7 + b_8k_x^2 + b_9k_y^2\right) B_x^2 + \left(b_{11} + b_{12}k_x^2 + b_{13}k_y^2\right) B_y^2 + \left(b_{15} + b_{16}k_x^2 + b_{17}k_y^2\right) B_z^2 + \ldots, \quad (7)$$

$$\mathcal{H}^{mix} = m_2B_yE_zk_x + m_3B_zE_xk_y + \ldots \quad (8)$$

The Hamiltonian for silicene has also been derived.

IV. PHONON THEORY

Phonon spectra in 2D nanomaterials have almost exclusively been computed using DFT-based codes and are complex and prone to qualitative errors due to the various approximations such as use of functionals [8]. An alternative model is a classical continuum model and one such model has been introduced for acoustic phonons in graphene nanoribbons [9].

For example, for graphene, we get

$$c_{11}\frac{\partial^2u_x}{\partial x^2} + c_{12}\frac{\partial^2u_y}{\partial x\partial y} + \frac{1}{2}\left(c_{11} - c_{12}\right)\left(\frac{\partial^2u_x}{\partial y^2} + \frac{\partial^2u_y}{\partial x\partial y}\right) = -\rho\omega^2u_x, \quad (9)$$

$$\frac{1}{2}\left(c_{11} - c_{12}\right)\left(\frac{\partial^2u_x}{\partial x\partial y} + \frac{\partial^2u_y}{\partial x^2}\right) + c_{12}\frac{\partial^2u_x}{\partial x\partial y} + c_{11}\frac{\partial^2u_y}{\partial y^2} = -\rho\omega^2u_y, \quad (10)$$

$$44\frac{\partial^2u_z}{\partial x^2} + c_{44}\frac{\partial^2u_z}{\partial y^2} = -\rho\omega^2u_z. \quad (11)$$

where u_i are the displacements, ρ is the mass density, and ω is the vibrational (angular) frequency, and T_I and S_J denote stress and strain, respectively. Here we have used Voigt notation for tensors. The displacements u_x and u_y are in-plane displacements and u_z is the out-of-plane displacement.

These equations are modified in the MoS_2 case due to its piezoelectricity. One finds instead

$$c_{11}\frac{\partial^2u_x}{\partial x^2} + c_{12}\frac{\partial^2u_y}{\partial x\partial y} + \frac{1}{2}\left(c_{11} - c_{12}\right)\left(\frac{\partial^2u_x}{\partial y^2} + \frac{\partial^2u_y}{\partial x\partial y}\right) - e_{x1}\frac{\partial E_1}{\partial x} + e_{x1}\frac{\partial E_2}{\partial y} = -\rho\omega^2u_x, \quad (12)$$

$$\frac{1}{2}\left(c_{11} - c_{12}\right)\left(\frac{\partial^2u_x}{\partial x\partial y} + \frac{\partial^2u_y}{\partial x^2}\right) + c_{12}\frac{\partial^2u_x}{\partial x\partial y} + c_{11}\frac{\partial^2u_y}{\partial y^2} + e_{x1}\frac{\partial E_2}{\partial x} - e_{x1}\frac{\partial E_1}{\partial y} = -\rho\omega^2u_y, \quad (13)$$

$$c_{44}\frac{\partial^2u_z}{\partial x^2} + c_{44}\frac{\partial^2u_z}{\partial y^2} = -\rho\omega^2u_z. \quad (14)$$

This system of equations is only complete if we also solve for the Maxwell-Poisson equation

$$D_i = \varepsilon_{ij}E_j + e_{iI}S_I + P_{sp}. \quad (15)$$

Example acoustic phonon dispersions for graphene are given in Fig. 3. Models for silicene and phosporene have also been introduced.

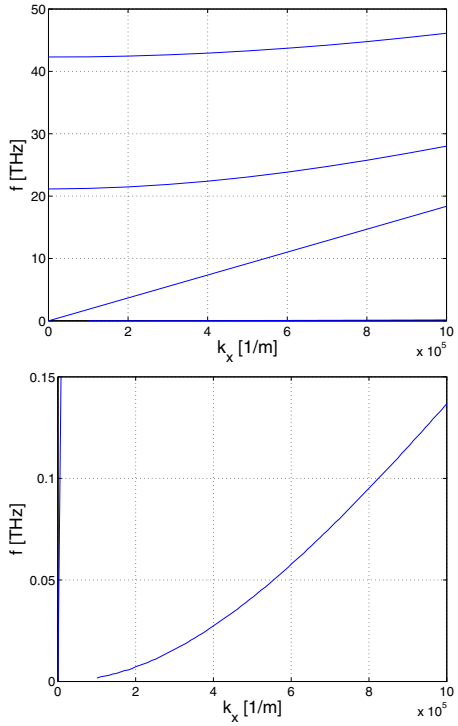

Fig. 3. Acoustic phonons of graphene.

V. SUMMARY

Empirical models of electrons and phonons in 2D materials have been developed in order to facilitate the realistic modeling of optoelectronic devices.

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