UDC 621.315.592

FIRST-PRINCIPLES MODELING OF ELECTRON-PHONON SCATTERING RATES IN HYDROGENATED GRAPHENE

V.N. Mishchanka

Belarusian State University of Informatics and Radioelectronics, Minsk, Republic of Belarus,
mishchenko@bsuir.by

Abstract: Graphene has a high mobility of charge carriers, which exceeds the mobility of charge carriers for all known materials, and is currently considering as one of the most promising materials for the creation of new semiconductor devices. The results of modeling of electron scattering rates on acoustic and optical phonons in a single layer of hydrogenated graphene C_2H_2 type without a substrate are presented. When modeling these rates, variants of both emission and absorption of phonons are considered. The obtained dependences of the charge carrier scattering rates will allow us to study the main characteristics of charge carrier transport in semiconductor structures containing different layers by modeling using the Monte Carlo method. Characteristics and parameters of graphene and its modifications can be used to create new heterostructured devices with improved output characteristics.

Keywords: graphene, phonon, modelling, semiconductor structure.

I. INTRODUCTION

Graphene is of great interest as a promising material for the development of new semiconductor devices for various frequency ranges [1-2]. The study of charge carrier transfer processes for semiconductor compounds containing graphene and other semiconductor materials layers is an urgent task, which is associated with the development of fast and powerful devices in the microwave and ultrahigh frequency ranges, as well as in the optical frequency range. The Monte Carlo statistical method is widely used to analyze semiconductor structures. One of the main features of this method is that it allows to take into account the processes of charge carriers scattering in the semiconductor and to study the operation of semiconductor devices in different operating conditions.

Quasi-analytical dependences of electron-phonon (e-ph) coupling matrices for various scattering processes have been developed. They have been used to obtain scattering rates (velocities) based on the Fermi golden rule and thus to describe the charge carrier transport properties [3, 4]. Based on the concept of deformation potential, a number of analytical expressions were proposed to estimate the scattering of electrons on optical and acoustic phonons for different materials (amount then and graphene). However, the semi-empirical expressions obtained in this way have serious limitations because of the simplifications made in their derivation. The main simplification of these expressions is related to the necessity of selecting the value of deformation potentials either from experimental measurements or from the calculated data of other theoretical approaches.

The limitations associated with the use of deformation potentials are largely removed by using density functional perturbation theory (DFPT) and an interpolation scheme using Wannier functions [5]. This approach allows us to fully determine the coupling matrix from ab initio calculations (first-principles calculations), without using empirical values of the deformation potential.

In this work, an ab initio study of scattering rates associated with electron scattering on optical and acoustic phonons in hydrogenated graphene has been carried out. Using Wannier functions, the coupling matrices of electron-phonon interaction were calculated, which are then used to model the rates (velocities) of electron scattering on acoustic and optical phonons. The obtained modeling results allow us to determine the contribution of various electron-phonon interaction components in the overall process of charge carrier scattering.

II. METHOD AND PECULIARITIES OF MODELING OF ELECTRON-PHONON SCATTERING INTENSITIES IN HYDROGENATED GRAPHENE

First-principles simulations of hydrogenated graphene (graphane) C_2H_2 type were performed with the Quantum Espresso [6] and EPW [7] software packages using the Perdew-Burke-Ernzerhof (PBE) parametrization within the local density approximation (LDA). Initially, self-consistent energy simulations were performed using the Quantum Espresso software complex with program pw.x. Then the calculation of electron-phonon dynamic matrices was performed using the program ph.x. The pseudopotentials of the Norm-conserving type [22] and the following modeling parameters were used in the Quantum Espresso software package: the cutoff energy of the wave function was 60 Ry (1 Ry ≈ 13.605 eV), the cutoff energy of the charge density and potentials was

240 Ry. The Brillouin zone (BZ) was represented using a 12 x 12 x 1 Monkhorst-Pack grid. To eliminate possible parasitic energy oscillations during the simulation, a vacuum layer of 20 Å thickness (1 Å = $1 \cdot 10^{-10}$ m) was added to the considered structure.

The program epw.x from the EPW software package [7] was used to simulate the scattering rates. This program is permitted to calculate the imaginary part of the eigenenergy

$$\mathrm{Im}\left(\sum_{n,\mathbf{k}}^{e-ph}\right) = \pi \cdot \sum_{m\nu} \int \frac{d\boldsymbol{q}'}{\Omega_{BZ}} \left|g_{mn,\nu}(\mathbf{k},\mathbf{q})\right|^2 \times$$

$$\left\{\left(n_{\boldsymbol{q}\nu} + f_{m\boldsymbol{k}+\boldsymbol{q}}\right) \times \delta\left(\omega - \left(\varepsilon_{m\boldsymbol{k}+\boldsymbol{q}} - \varepsilon_F\right) + \omega_{\boldsymbol{q}\nu}\right) + \left(n_{\boldsymbol{q}\nu} + 1 - f_{m\boldsymbol{k}+\boldsymbol{q}}\right) \times \delta\left(\omega - \left(\varepsilon_{m\boldsymbol{k}+\boldsymbol{q}} - \varepsilon_F\right) - \omega_{\boldsymbol{q}\nu}\right)\right\}, \quad (1)$$

where ω is the phonon frequency, $\varepsilon_{m\mathbf{k}+\mathbf{q}}$ is the energy for the band with number m and wavevector **k** in the Brillouin zone (Ω_{BZ}), $\omega_{\boldsymbol{q}\nu}$ is the phonon energy with mode ν and wave vector **q** in the BZ over which the integration is performed, the parameters $f_{m\boldsymbol{k}+\boldsymbol{q}}$ and $n_{\boldsymbol{q}\nu}$ are the Fermi and Bose distributions, respectively, which are estimated at a given temperature, $g_{mn,\nu}(\mathbf{k},\mathbf{q})$ is the electron-phonon interaction matrix for the bands with number n and m; ε_F is the Fermi energy, the symbol δ of the function means the necessity of performing Gaussian smoothing operations during integration.

The total scattering rates of the electron-phonon interaction with phonon absorption and emission were calculated from the imaginary part of the eigenenergy as [7, 8]

$$\tau^{-1} = 2 \cdot \mathrm{Im}\left(\sum_{n,\mathbf{k}}^{e-ph}\right), \quad (2)$$

The following values of the modeling parameters were chosen for modeling in the EPW program of the dependences of the scattering rates. So the size of grids of the form NxNx1, which corresponded to the conditional directions of coordinates x, y, z, for electrons and holes during interpolation procedures, was set by the value of the parameter N, the value of which was equal to 264. The values of other modeling parameters were taken as follows: the value of the Gaussian smoothing coefficient (parameter “dg”) - equal to 0.001 eV; the value of the parameter “fsthick”, which determines the value of the range of energies during modeling relative to the Fermi energy level - equal to 4 eV; the number of Wannier functions - equal to the value of 12. The mode of setting the parameters “auto_projection” and “scdm_proj” to value “true” was used in modeling. The value of the concentration of electrons and holes for all presented modeling results was taken as $1 \cdot 10^{13}$ cm^{-3}.

III. RESULTS OF MODELING FROM FIRST PRINCIPLES OF ELECTRON-PHONON SCATTERING RATES IN HYDROGENATED GRAPHENE

The dispersive phonon dependences of single-layer hydrogenated graphene C_2H_2 type are considered for modes of the ZA, TA, LA, ZO, TO, LO, LB, TB, LB*, TB*, ZS, ZS* type [9]. The first group of dependencies, denoted as ZA, LA, TA, represents the scattering on acoustic phonons along the conventional longitudinal and transverse directions (x, y coordinates), as well as the z coordinate orthogonal to them, respectively. The second group of dependencies, denoted as ZO, LO, TO, represents the result of scattering on optical phonons along the conventional longitudinal and transverse directions (x, y coordinates), as well as the z coordinate orthogonal to them, respectively.

Among the additional modes that appear in hydrogenated graphene (graphane) by comparison with graphene and are related to the processes of bending of the structure, two symmetric modes - longitudinal and transverse LB and TB, and two asymmetric modes LB* and TB* can be noted. In the z direction, two modes associated with stretching processes are formed. One of them is the symmetric mode ZS, and the other is the asymmetric mode ZS* [9].

The results of modeling the scattering rates for modes TO, LO from energy obtained in the EPW program using formulas (1-2) are presented in Figures 1-2 by lot of the blue and dark green dots, respectively. When modeling these rates, variants of both emission and absorption of phonons are considered.

The obtained point data sets were subjected to approximation using analytic degree functions in the data processing and plotting program ORIGIN when performing Fitting and Polinomial Fit operations in the Analysis section. When performing these operations in the ORIGIN program, analytical dependencies are obtained with minimal approximation errors.

Results of approximation of the first-principles modeling data for the ZA, TA, LA, LB, TB, LB*, TB*, ZO, TO, LO, ZS, ZS* modes are presented by the curve 1 on the Figures 3-6. The total scattering rates τ^{-1} have the dimension in s^{-1} and the energy E have dimension in eV. Curve 1 shows the approximated dependences of scattering rates on energy in the case of TO and LO mode in Figure 1 and 2, respectively.

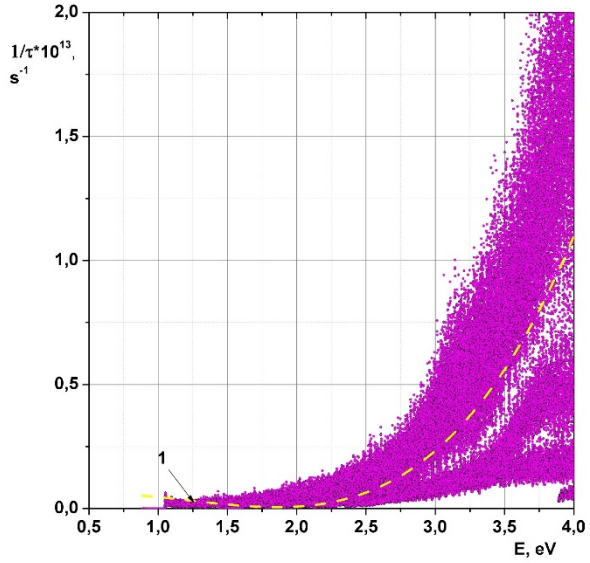

Figure 1. Dependence of the scattering rates for the TO optical mode on energy

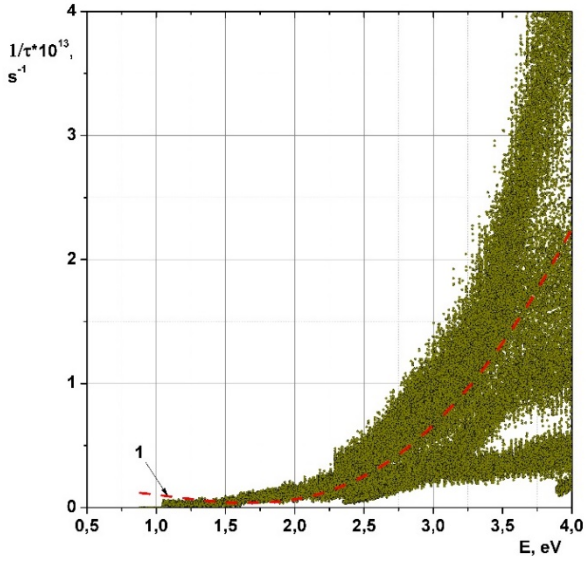

Figure 2. Dependence of scattering rates for LO optical mode on energy

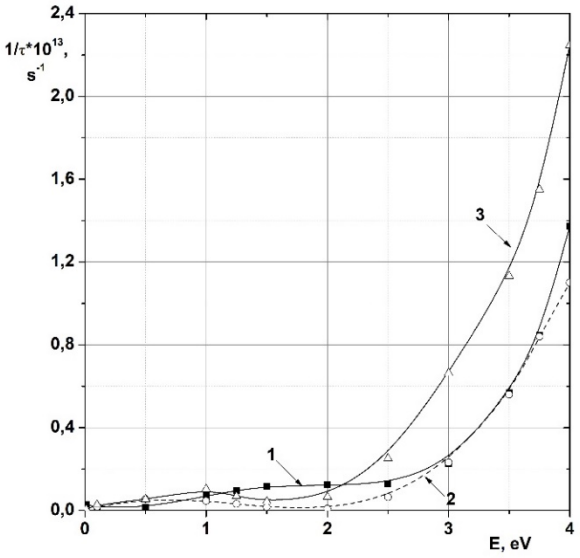

Figure 3. Dependences of scattering rates on energy in the case of ZO (curve 1), TO (curve 2) and LO (curve 3) optical mode

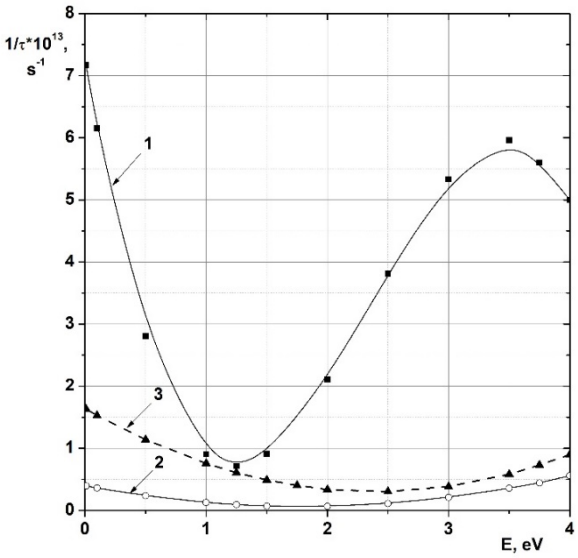

Figure 4. Dependences of scattering rates on energy in the case of ZA (curve 1), TA (curve 2) and LA (curve 3) acoustic mode

The analysis of Figures 3-6 shows that the largest scattering rates are observed for the LB mode for small energy value. ZA, ZO, LO, TO, ZS, ZS* modes have largest scattering rates at energy near the value 4 eV. The scattering rates for the other modes LB, LB*, TB, TB*, TA, LA are significantly smaller than the scattering rates for the above mentioned modes for the energy near 4 eV.

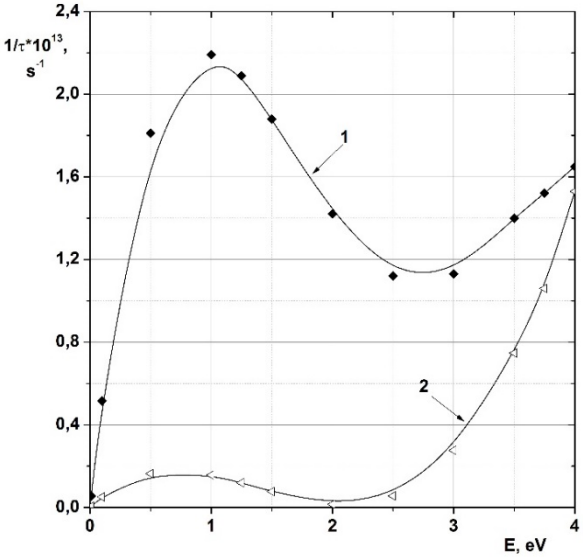

Figure 5. Dependences of scattering rates on energy in the case of ZS (curve 1) and ZS* (curve 2) mode

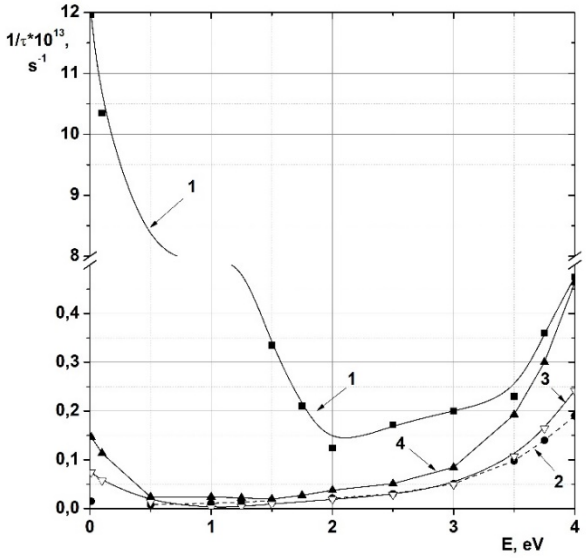

Figure 6. Dependences of scattering rates on energy in the case of LB (curve 1), TB (curve 2), LB*(curve 3), and TB* (curve 4) mode

From the presented data, we can see that the scattering rates of mode LO are higher than scattering rates of the modes TO and ZO in energy range from 2 eV up to 4 eV.

IV. CONCLUSIONS

The results of the study of electron scattering rates on phonons in a single layer of hydrogenated graphene C_2H_2 type without a substrate are presented. The electron scattering rates for modes of ZA, TA, LA, ZO, TO, LO, LB, TB, LB*, TB*, ZS, ZS* type at modeling from first principles are obtained. The presented dependences and parameters of electron scattering rates on acoustic and optical phonons in hydrogenated graphene can serve as a basis for modeling of new heterostructured devices containing graphene and other semiconductor materials.

REFERENCES

[1] Electric field effect in atomically thin carbon film / K. S. Novoselov, A. K. Geim [et al.] // Science. 2004. Vol. 306. P. 666-669.

[2] Giant intrinsic carrier mobilities in graphene and its bilayer / V. S. Morozov [et al.] // Phys. Rev. Lett. 2008. 100, P. 016602.

[3] Hess K., Advanced Theory of Semiconductor Devices / K. Hess // Wiley-IEEE Press, Piscataway, NJ, 1999.

[4] Lundstrom M., Fundamentals of Carrier Transport / M. Lundstrom // Cambridge University Press, Cambridge, UK, 2009.

[5] First-principles calculations of charge carrier mobility and conductivity in bulk semiconductors and two-dimensional materials / S. Poncé., W. Li,S. Reichardt, and F. Giustino F // Rep. Prog. Phys. 2020; V. 83: 036501.

[6] QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials / P. Giannozzi, S. Baroni., N. Bonini, M. Calandra [et al.] // J. Phys.: Condens. Matter. 2009. Vol. 21. P. 395502.

[7] EPW: Electron–phonon coupling, transport and superconducting properties using maximally localized Wannier functions / S. Poncé S., E.R. Margine, C. Verdi // Computer Physics Communications. 2016. V. 209. P. 116 – 133.

[8] Ab Initio Study of Hot Carriers in the First Picosecond after Sunlight Absorption in Silicon / M. Bernardi, D. Vigil-Fowler, J. Lischner, J. B. Neaton, and S. G. Louie // Phys. Rev. Lett. 2014. V.112, P. 257402.

[9] How to resolve a phonon-associated property into contributions of basic phonon modes / Long Cheng, Chenmu Zhang and Yuanyue Liu. // J. Phys.: Mater. 2019. 2. P. 045005.

МОДЕЛИРОВАНИЕ ИЗ ПЕРВЫХ ПРИНЦИПОВ ИНТЕНСИВНОСТЕЙ ЭЛЕКТРОННО-ФОНОННОГО РАССЕИВАНИЯ В ГИДРИРОВАННОМ ГРАФЕНЕ

Мищенко В.Н.

Белорусский государственный университет информатики и радиоэлектроники,
Минск, Республика Беларусь, mishchenko@bsuir.by

Аннотация: Графен обладает высокой подвижностью носителей заряда, превышающей подвижность носителей заряда для всех известных материалов, и в настоящее время рассматривается как один из наиболее перспективных материалов для создания новых полупроводниковых приборов. Представлены результаты моделирования интенсивностей рассеяния электронов на акустических и оптических фононах в однослойном гидрированном графене типа C_2H_2 без подложки. При моделировании этих интенсивностей рассмотрены варианты как испускания, так и поглощения фононов. Полученные зависимости интенсивностей рассеяния носителей заряда позволят исследовать основные характеристики их транспорта в полупроводниковых структурах при моделировании с использованием метода Монте-Карло. Характеристики и параметры гидрированного графена могут быть использованы для создания новых гетероструктурных приборов с улучшенными выходными характеристиками.

Ключевые слова: графен, фонон, моделирование, полупроводниковая структура.