

12,18

Particular features of the thermoelectric power factor of encapsulated structures formed by two-dimensional layers

© S.Yu. Davydov¹, O.V. Posrednik²

¹ Ioffe Institute,
St. Petersburg, Russia

² St. Petersburg State Electrotechnical University „LETI“,
St. Petersburg, Russia

E-mail: Sergei_Davydov@mail.ru

Received February 3, 2023

Revised February 3, 2023

Accepted February 19, 2023

Model approach is used to obtain expression for the thermoelectric power factor σS^2 (σ — is the static conductivity, S — is the Seebeck coefficient), characterized monoatomic 2D layer of IV group elements, placed between two slabs, formed by hexagonal 2D layers of III–V compounds. The *h*-BN/graphene/*h*-BN structure is thoroughly studied and the dependence of the σS^2 value on the chemical potential μ is examined. The analytical estimates of the function $\sigma S^2(\mu)$ are given.

Keywords: static conductivity, Seebeck coefficient, hexagonal monoatomic 2D layers.

DOI: 10.21883/PSS.2023.04.56007.15

1. Introduction

One of widely used characteristics of the thermoelectric capability of materials is the $Z = \sigma S^2 / \kappa$ parameter known as figure of merit, where σ being electric conductivity, S being thermoelectric power (or Seebeck coefficient), κ being thermal conductivity. Sometimes the dimensionless product of ZT is used for the same evaluations, where T being temperature. For a long enough time developers managed to achieve $ZT \approx 1$ for the best thermoelectrical bulk materials [1]. A significant progress became visible after the publication of study [2], where structures with quantum wells have been considered. With appearance of two-dimensional materials and structures based on them the modern stage of thermoelectricity has been started [3]. In this study we consider the σS^2 product as a thermoelectric characteristic.

The overwhelming majority of modern device structures are a set of vertically arranged microlayers and/or nanolayers of different nature [4]. This study considers hexagonal monolayers (ML) sandwiched between two covers, i.e. encapsulated monolayers (EnML). To describe the electron spectrum of EnML, a model is used [5], which is based on the adsorption approach to the problem of epitaxial monolayers (EpML) [6]. The essence of this model is reduced to the representation of EpML as a lattice of adatoms, namely: if the electronic state of an adatom is described by the Green function $g_{ad}(\omega) = (\omega - \varepsilon_a - \Sigma(\omega))^{-1}$, where ω is energy variable, ε_a is energy of the level of adsorbed atom A , $\Sigma(\omega)$ is self-energy part that describes the interaction with the substrate, then, by arranging the adatoms into

a lattice and including interaction t between the nearest neighbors, we get for EpML the Green function $G_{EpML}^{-1}(\omega, \mathbf{k}) = g_{ad}^{-1}(\omega) - t f(\mathbf{k})$, where the $f(\mathbf{k})$ periodic function corresponds to the geometry of the adatom lattice, \mathbf{k} is wave vector. The same result can be obtained if the Green function $g_{ML}(\omega, \mathbf{k}) = (\omega - \varepsilon_a - t f(\mathbf{k}))^{-1}$ is assigned to a free ML and then the interaction with substrate $\Sigma(\omega)$ is included. The purpose of this study is to determine the effect of EnML covers on the value of the σS^2 product of an encapsulated ML.

2. General relationships

According to the Mott formula [7], the Seebeck coefficient is

$$S = -\frac{1}{3} (\pi^2 k_B^2 T / e) [d \ln \sigma(\mu, T = 0) / d\mu],$$

where e is elementary charge, k_B is Boltzmann constant, T is temperature, μ is chemical potential, the problem is reduced to the determination of the $\sigma(\mu)$ dependence at zero temperature. The problem of static conductivity and Seebeck coefficient of epitaxial graphene within the framework of adsorption approach was solved for the first time in [8] for a semiconductor substrate, which density of states (DoS) was described by the Haldane–Anderson model.

When turning from EpML to the EnML problem, it is easy to understand that the corresponding Green function has the same form as $G_{EpML}(\omega, \mathbf{k})$ but $\Sigma(\omega)$ is replaced with $\Sigma(\omega) = \Sigma_1(\omega) + \Sigma_2(\omega)$, where $\Sigma_{1(2)}(\omega)$ being contribution from cover 1(2) [9]. Using the results of [8,9], it

can be shown that for μ inside the resulting band gap with boundaries of E_C^* and E_V^* defined by the following inequality

$$E_V^* = \max\{E_{V_1}, E_{V_2}\} < E_C^* = \min\{E_{C_1}^*, E_{C_2}^*\},$$

where $E_{V_{1,2}}^*$ and $E_{C_{1,2}}^*$ are edges of the valence band and the conduction band of covers 1 and 2, the reduced static conductivity σ^* : $= \sigma \pi \hbar / e^2$ at $T = 0$ has the following form

$$\sigma^* = \frac{\xi^2 F}{F^2 + 4\tilde{\mu}^2 \gamma^2} + \frac{\tilde{\mu}^2 + \gamma^2}{2\tilde{\mu} \gamma} R \equiv \sigma_1^* + \sigma_2^*,$$

$$R(\mu) = \arctan \frac{F(\mu)}{2\gamma\tilde{\mu}} + \arctan \frac{\tilde{\mu}^2 - \gamma^2}{2\gamma\tilde{\mu}}. \quad (1)$$

Here

$$F = \xi^2 + \gamma^2 - \tilde{\mu}^2, \quad \tilde{\mu} = \mu - \bar{\Lambda}(\mu), \quad \bar{\Lambda}(\mu) = \Lambda_1(\mu) + \Lambda_2(\mu),$$

where $\Lambda_{1,2} = \text{Re} \Sigma_{1,2}$, \hbar is reduced Plank constant, $\xi = \sqrt{2\pi\sqrt{3}t}$ is cutting energy for ML [10], γ is parameter of the natural attenuation of electronic states in free ML; the Dirac point energy $\varepsilon_D = \varepsilon_a$ is taken equal to zero. In the same region (E_V^*, E_C^*) derivatives $d\sigma^*/d\mu = d\sigma_1^*/d\mu + d\sigma_2^*/d\mu$ are

$$d\sigma_1^*/d\mu = -\frac{2\xi^2 \tilde{\mu} C}{F^2 + 4\gamma^2 \tilde{\mu}^2} \left(1 - 2 \frac{F(F - 2\gamma^2)}{F^2 + 4\gamma^2 \tilde{\mu}^2}\right),$$

$$d\sigma_2^*/d\mu = \frac{\tilde{\mu}^2 - \gamma^2}{2\gamma\tilde{\mu}^2} CR + \frac{\tilde{\mu}^2 + \gamma^2}{2\gamma\tilde{\mu}} \frac{dR}{d\mu},$$

$$\frac{dR}{d\mu} = -4\gamma CD, \quad C = 1 - d\bar{\Lambda}/d\mu,$$

$$D = \left(\frac{\xi^2 + \gamma^2}{F^2 + 4\gamma^2 \tilde{\mu}^2} - \frac{1}{2(\tilde{\mu}^2 + \gamma^2)} \right). \quad (2)$$

3. Graphene encapsulated by *h*-BN layers

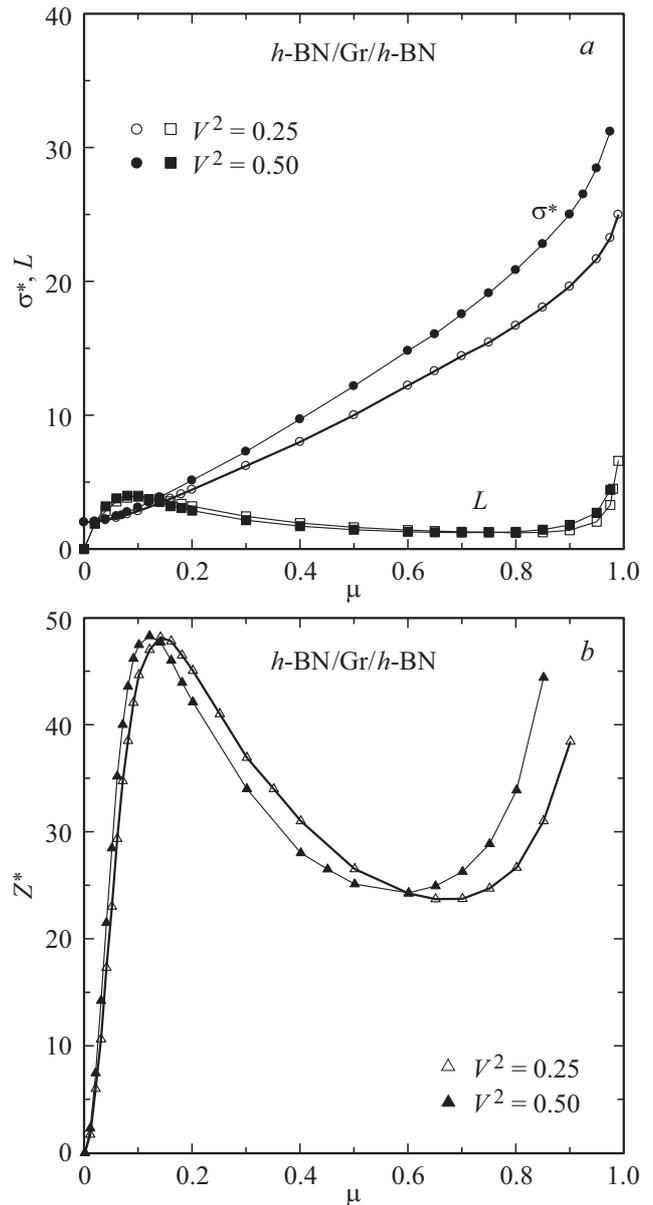
Let us consider as an example the *h*-BN/Gr/*h*-BN structure, where Gr is monolayer graphene, *h*-BN is two-dimensional hexagonal boron nitride. According to evaluations of [11], distances between the nearest neighbors, hopping parameters t and Dirac points ε_D for free Gr and *h*-BN can be considered equal. At the same time, $\sigma(\mu)$ and $S(\mu)$ functions are, respectively, symmetric and antisymmetric in relation to the $\mu = 0$ point for both the EpML and the EnML, with $\sigma(0) = \sigma_{\min}$ and $S(0) = 0$. The $\Lambda(\mu)$ shift function for graphene-like binary compounds calculated within the low-energy approximation is

$$\bar{\Lambda}(\mu) = (4V^2 \mu / \xi^2) \ln |(\Delta^2 - \mu^2) / (\xi^2 + \Delta^2 - \mu^2)| \quad [12],$$

whence it follows that $\tilde{\mu} = \mu M$, $M = 1 + \bar{\Lambda}(\mu)/\mu$,

$$C = M + 8V^2 \mu^2 / [(\Delta^2 - \mu^2)(\xi^2 + \Delta^2 - \mu^2)],$$

where 2Δ is gap width in the ML spectrum, V is matrix element of ML interaction with cover. Due to the



Dependencies of a) dimensionless static conductivity σ^* (circles) and $L = d \ln \sigma^* / d\mu$ (squares) and b) $Z^* = \sigma^* L^2$ (triangles) functions on the position of chemical potential μ inside the band gap of *h*-BN for *h*-BN/Gr/*h*-BN structures (Gr — graphene) at $\gamma = 0.1$, $V^2 = 0.25$ (light symbols) and $V^2 = 0.50$ (dark symbols). All energy parameters are given in Δ units. Only right halves of even $\sigma^*(\mu)$, $Z^*(\mu)$ functions and odd $L(\mu)$ function are shown.

fact that $\xi \approx 10$ eV and $\Delta \approx 3$ eV, we have $\Delta^2 / \xi^2 \ll 1$. Assuming $\gamma / \Delta = 0.1$, $V^2 / \Delta^2 = 0.25, 0.50$, the $\sigma^*(\mu)$, $L(\mu) = d \ln \sigma^* / d\mu$ and $Z^*(\mu) = (\sigma^*)^{-1} (d\sigma^* / d\mu)^2 \propto S^2$ dependencies shown in the figure can be derived. It can be easily shown that at $\mu \rightarrow 0$ the following is true:

$$\sigma_0^* \approx 2(1 + \mu^2 M_0^2 / \gamma^2), \quad L_0 \approx 2\mu M_0^2 / \gamma^2,$$

$$Z_0^* \approx 2\mu^2 M_0^4 / \gamma^4, \quad (3)$$

where $M_0 = M(\mu = 0)$ and $C_0 = C(\mu = 0)$, so that $M_0 = C_0$. At $\mu^2 \rightarrow \Delta^2$ the following can be written:

$$\begin{aligned}\sigma_{\pm\Delta}^* &\approx \frac{1}{2} \pi \bar{\Lambda}_\Delta / \gamma, \quad L_{\pm\Delta} \approx \pm C_\Delta / \bar{\Lambda}_\Delta, \\ Z_{\pm\Delta}^* &\approx \frac{1}{2} \pi C_\Delta^2 / (\gamma \bar{\Lambda}_\Delta),\end{aligned}\quad (4)$$

where

$$\bar{\Lambda}_\Delta \approx (4V^2 \Delta / \xi^2) \ln[\xi^2 / (\Delta^2 - \mu^2)]$$

and

$$C_\Delta \approx 4V^2 \Delta^2 / \xi^2 / (\Delta^2 - \mu^2).$$

Whence it follows that the effect of covers is taken into account by $\bar{\Lambda}$, M and C functions: with increase in V^2 / ξ^2 coupling constant the conductivity, the Seebeck coefficient and the thermoelectric factor increase.

4. On extreme points of $L(\mu)$ and $Z^*(\mu)$ functions

Now let us consider $L(\mu)$ and $Z^*(\mu)$ functions, which points of extreme $\pm\mu_{\text{ext}}^L$ and $\pm\mu_{\text{ext}}^{Z^*}$ can be determined, respectively, from the following equations: $\sigma^*(d^2\sigma^*/d\mu^2) = (d\sigma^*/d\mu)^2$ and $2(d^2\sigma^*/d\mu^2) = (d\sigma^*/d\mu)^2$. Using formulae (2), the following can be written:

$$\begin{aligned}\frac{d^2\sigma_1^*}{d\mu^2} &= -B_1^* \frac{dA_1^*}{d\mu} - A_1^* \frac{dB_1^*}{d\mu}, \\ \frac{dA_1^*}{d\mu} &= \frac{2\xi^2 C^2}{F^2 + 4\gamma^2 \tilde{\mu}^2} \left(1 + \frac{\tilde{\mu}}{C^2} \frac{dC}{d\mu} + 4\tilde{\mu}^2 \frac{F - 2\gamma^2}{F^2 + 4\gamma^2 \tilde{\mu}^2} \right), \\ \frac{dB_1^*}{d\mu} &= \frac{8C\tilde{\mu}}{F^2 + 4\gamma^2 \tilde{\mu}^2} \left(F - \gamma^2 - \frac{F(F - 2\gamma^2)^2}{F^2 + 4\gamma^2 \tilde{\mu}^2} \right),\end{aligned}\quad (5)$$

where we have assumed $d\sigma_1^*/d\mu := -A_1^* B_1^*$ and $A_1^*(B_1^*)$ is the first (the second) product term in (2);

$$\begin{aligned}\frac{d^2\sigma_2^*}{d\mu^2} &= \frac{dA_2^*}{d\mu} + \frac{dB_2^*}{d\mu}, \\ \frac{dA_2^*}{d\mu} &= \frac{\gamma C^2 R}{\tilde{\mu}^3} + \frac{\tilde{\mu}^2 - \gamma^2}{2\gamma \tilde{\mu}^2} \left(R \frac{dC}{d\mu} + C \frac{dR}{d\mu} \right), \\ \frac{dB_2^*}{d\mu} &= \frac{\tilde{\mu}^2 - \gamma^2}{2\gamma \tilde{\mu}^2} C \frac{dR}{d\mu} + \frac{\tilde{\mu}^2 + \gamma^2}{2\gamma \tilde{\mu}^2} \frac{d^2 R}{d\mu^2}, \\ \frac{d^2 R}{d\mu^2} &= -4\gamma D \frac{dC}{d\mu} - 4\gamma C \frac{dD}{d\mu}, \\ \frac{dD}{d\mu} &= \frac{4(\xi^2 + \gamma^2) C \tilde{\mu}}{(F^2 + 4\gamma^2 \tilde{\mu}^2)^2} (F - 2\gamma^2) + \frac{C \tilde{\mu}}{(\tilde{\mu}^2 + \gamma^2)^2},\end{aligned}\quad (6)$$

where $A_2^*(B_2^*)$ is the first (the second) sum term in formulae (2) for $d\sigma_2^*/d\mu$. The analysis of (5) and (6) in the extreme case of weak bond $V^2 \ll \Delta^2 \ll \xi^2$ yields the following

$$\begin{aligned}\mu_{\text{ext}}^L &\sim \mu_{\text{ext}}^{Z^*} \sim \gamma, \quad \sigma^* \sim \pi/2, \\ |L_{\text{ext}}| &\sim \frac{2}{\pi\gamma}, \quad Z_{\text{max}}^* \sim \frac{2}{\pi\gamma^2}.\end{aligned}\quad (7)$$

It follows from order estimates (7) that there is almost no effect of covers on the characteristics in question.

5. Conclusion

It is worth to note that the considered structures of monolayers of gapless graphene and *h*-BN, are not only of academic interest but also of real device interest [13–15]. Moreover, the results obtained in this study can be easily expanded on other monatomic layers of group IV elements (gapless silicene, germanene and stanene) [16] and covers of A_3B_5 type 2D compounds. The main conclusion of this study consists in that maximum values of the thermoelectric power coefficient are realized in the cases when the chemical potential is near edges of the resulting band gap of the encapsulated structure. We believe, that this conclusion is also true for monolayers with a gap in the spectrum (gapped MLs) and for dielectric and/or semiconductor two-dimensional and three-dimensional covers. In the case of metal covers the maximum thermoelectric power coefficient should be expected when the chemical potential is near the edge of gap in the monolayer spectrum (see, for example, [17]).

Conflict of interest

The authors declare that they have no conflict of interest.

References

- [1] A.M. Dehkordi, M. Zebarjadi, J. He, T.M. Tritt. *Mater. Sci. Eng. R* **97**, 1 (2015).
- [2] L.D. Hicks, M.S. Dresselhaus. *Phys. Rev. B* **47**, 19, 12727 (1993).
- [3] D. Li, Y. Gong, Y. Chen, J. Lin, Q. Khan, Y. Zhang, Y. Li, H. Zhang, H. Xie. *Nano-Micro Lett.* **12**, 1, 36 (2020).
- [4] T. Tan, X. Jiang, C. Wang, B. Yao, H. Zhang. *Adv. Sci.* **7**, 11, 2000058 (2020).
- [5] S.Yu. Davydov. *Tech. Phys. Lett.* **47**, 9, 649 (2021).
- [6] S.Yu. Davydov, O.V. Posrednik. *Phys. Solid State* **58**, 4, 647 (2016).
- [7] J.M. Ziman. *Principles of the theory of solids*. University Press (1965).
- [8] Z.Z. Alisultanov. *Low Temp. Phys.* **39**, 7, 592 (2013).
- [9] S.Yu. Davydov, O.V. Posrednik. *Semiconductors* **55**, 7, 782 (2021). DOI: 10.1134/S1063782621070071
- [10] N.M.R. Peres, F. Guinea, A.H. Castro Neto. *Phys. Rev. B* **73**, 12, 125411 (2006).
- [11] S.Yu. Davydov. *Phys. Solid State* **60**, 9, 1865 (2018).
- [12] S.Yu. Davydov. *Semiconductors* **51**, 2, 217 (2017).
- [13] A.K. Geim, I.V. Grigorieva. *Nature* **499**, 7459, 419 (2013).
- [14] J. Duan, X. Wang, X. Lai, G. Li, K. Watanabe, T. Taniguchi, M. Zebarjadi, E.Y. Andrei. *PNAS* **113**, 50, 14272 (2016).
- [15] I.V. Antonova. *Semiconductors* **50**, 1, 66 (2016).
- [16] F.B. Wiggers, A. Fleurence, K. Aoyagi, T. Yonezawa, Y. Yamada-Takamura, H. Feng, J. Zhuang, Y. Du, A.Y. Kovalgin, M.P. de Jong. *2D Materials* **6**, 3, 035001 (2019).
- [17] A.A. Varlamov, A.V. Kavokin, I.A. Luk'yanchuk, S.G. Sharapov. *Phys.-Uspekhi* **55**, 11, 1146 (2012).

Translated by Y.Alekseev