Heat capacity of nonequilibrium electron-hole plasma in graphene layers and graphene bilayers

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We analyze the statistical characteristics of the quasi-nonequilibrium two-dimensional electron-hole plasma in graphene layers (GLs) and graphene bilayers (GBLs) and evaluate their heat capacity. The GL heat capacity of the weakly pumped intrinsic or weakly doped GLs normalized by the Boltzmann constant is equal to $c_{GL} \approx 6.58$. With varying carrier temperature the intrinsic GBL carrier heat capacity $c_{GBL}$ changes from $c_{GBL} \approx 2.37$ at $T \lesssim 300$ K to $c_{GBL} \approx 6.58$ at elevated temperatures. These values are markedly different from the heat capacity of classical two-dimensional carriers with $c = 1$. The obtained results can be useful for the optimization of different GL- and GBL-based high-speed devices.

I. INTRODUCTION

The properties of the graphene layers (GLs) and graphene bilayers (GBLs), in particular, their optical characteristics, conductivity, plasmonic properties, thermal conductivity (both associated with the lattice and the carriers), heat capacity, and others have been extensively studied theoretically and experimentally \cite{1-19} (see the references therein). The contributions of the carriers in GLs and GBLs to the overall heat capacity is small in comparison with the contribution of the lattice vibrations \cite{20}. However, the electron and hole heat capacity determines the rate of the carrier heating and cooling. This heating/cooling rate affects for the ultimate high-speed performance, including the dynamic response and the modulation characteristics of the GL- and GBL-based devices using the variation of the two-dimensional electron-hole plasma (2DEHP) parameters (such as the effective carrier temperature, conductivity, transparency of the incident radiation). Such GL- and GBL-devices include the carrier bolometric detectors \cite{21, 22}, electro-optical modulators \cite{22}, fast thermal radiation emitters \cite{24, 31}, and superluminescent and lasing diodes \cite{32}. Many papers deal with the theoretical evaluation of the GL- and GBL-lattice heat capacity (see, for example, a recent review \cite{33}). However, the carrier capacity of the intrinsic quasi-nonequilibrium 2DEHP in GLs and GBLs was not addressed. The case of highly doped GLs was briefly discussed in \cite{34, 35}. In this paper, we calculate the heat capacity of the 2DEHP in the equilibrium and of the 2DEHP somewhat deviating from the equilibrium due to the radiation absorption and/or the carrier injection.

II. GENERAL RELATIONS

The dispersion relations for electrons (upper sign) and holes (lower sign) in the GLs and GBLs are presented as

$$\varepsilon_{\pm}^{\pm}_{GL} = \pm v_W p, \quad \varepsilon_{\pm}^{\pm}_{GBL} \approx \pm \frac{\gamma_1}{2} \left[ \sqrt{1 + 4v_W^2 p^2/\gamma_1^2} - 1 \right],$$

(1)

Here $\hbar$ is the Planck constant, $v_W \approx 10^8$ cm/s is the characteristic carrier velocity in GL and GBLs, $p = |p|$ is the carrier momentum, and $\gamma_1 \approx 0.4$ eV is the band parameter (the GBL hopping parameter) \cite{36, 37}. We assume that the frequent carrier-carrier collisions lead to the formation in the 2DEHP of the electron and hole systems described by quasi-Fermi energy distribution functions $f_e(\varepsilon)$ and $f_h(\varepsilon)$ with common effective temperature $T : f_e(\varepsilon) = \left[ 1 + \exp\left(\frac{\varepsilon - \mu_e}{k_B T}\right) \right]^{-1}$ and
f_h(\varepsilon) = \left[1 + \exp\left(\frac{\varepsilon - \mu_h}{k_B T}\right)\right]^{-1}, \text{ where } k_B \text{ is the Boltzmann constant, } \varepsilon \geq 0 \text{ is the carrier kinetic energy and } \mu_e \text{ and } \mu_h \text{ are the electron and hole quasi-Fermi energies, respectively. In the undoped GLs and GBLs, } \mu_e = \mu_h = 0. \text{ If, in particular, the GL (or the GBL) is doped by donors, } \mu_e > 0, \text{ while } \mu_h < 0. \text{ In the equilibrium, i.e., without optical or injection pumping and with no heating of the 2DEHP by the electric field, } \mu_e + \mu_h = 0. \text{ In this case, } \mu_e \text{ and, consequently, } \mu_h \text{ are determined by the donor sheet density } \Sigma_d. \text{ In the acceptor doped GLs (GBLs), } \mu_h > 0 \text{ and } \mu_e < 0 \text{ with } \mu_h \text{ and } \mu_e \text{ determined by the acceptor density } \Sigma_a. \text{ When the 2DEHP is off equilibrium, generally, } \mu_e + \mu_h \neq 0.

When the 2DEHP deviates from equilibrium due the irradiation or the carrier injection, the combined quasi-Fermi energy \( \mu = \mu_e + \mu_h \) can be either positive or negative (see, for example, [14]). The main mechanism, which enables tending of \( \mu \) zero, is the Auger recombination. However, in the 2DEHP under consideration, the Auger recombination-generation processes are relatively ineffective [14, 38, 39].

The net carrier (electrons and holes) densities, \( \Sigma_{GL} \) and \( \Sigma_{GBL} \), in the GL and GBL, respectively, in line with Eq. (1) are given by

\[
\Sigma_{GL} = \frac{2}{\pi h^2 v_W} \int_0^\infty d\varepsilon \left[\frac{1}{1 + \exp\left(\frac{\varepsilon - \mu_e}{k_B T}\right)} + \frac{1}{1 + \exp\left(\frac{\varepsilon - \mu_h}{k_B T}\right)}\right] \\
= \frac{2(k_B T)^2}{\pi h^2 v_W^2} \left[ F_1\left(\frac{\mu_e}{k_B T}\right) + F_1\left(\frac{\mu_h}{k_B T}\right)\right]
\]

and

\[
\Sigma_{GBL} = \frac{2}{\pi h^2 v_W} \int_0^\infty d\varepsilon (\varepsilon + \gamma_1/2) \left[\frac{1}{1 + \exp\left(\frac{\varepsilon - \mu_e}{k_B T}\right)} + \frac{1}{1 + \exp\left(\frac{\varepsilon - \mu_h}{k_B T}\right)}\right] \\
= \frac{2(k_B T)^2}{\pi h^2 v_W^2} \left[ F_1\left(\frac{\mu_e}{k_B T}\right) + F_1\left(\frac{\mu_h}{k_B T}\right)\right] + \frac{\gamma_1}{k_B T} \left[ F_0\left(\frac{\mu_e}{k_B T}\right) + \frac{\gamma_1}{k_B T} F_0\left(\frac{\mu_h}{k_B T}\right)\right].
\]

Here \( F_\zeta(y) \) is the Fermi-Dirac integral.

The carrier energy in the 2DEHP can be calculated as

\[
\mathcal{E}_{GL} = \frac{2}{\pi h^2 v_W} \frac{2(k_B T)^2}{\pi} + \frac{4 \ln 2}{\pi} \left[\frac{\mu}{k_B T}\right] \left[ F_1\left(\frac{\mu_e}{k_B T}\right) + F_1\left(\frac{\mu_h}{k_B T}\right)\right] + \frac{\gamma_1}{k_B T} \left[ F_0\left(\frac{\mu_e}{k_B T}\right) + \frac{\gamma_1}{k_B T} F_0\left(\frac{\mu_h}{k_B T}\right)\right] \frac{\mu}{k_B T}.
\]

Equations (4) and (5) result in the following expressions for the carrier thermal energy density (thermal energy per GL and GBL area) as a function of the carrier effective temperature \( T \) and the combined quasi-Fermi energy \( \mu \):

\[
\mathcal{E}_{GL} \simeq \frac{2(k_B T)^3}{\pi h^2 v_W} \left[ 3 \zeta(3) + \frac{\pi^2}{3} \left[\frac{\mu}{k_B T}\right] \frac{\mu}{k_B T} \right],
\]

\[
\mathcal{E}_{GBL} \simeq \frac{2(k_B T)^3}{\pi h^2 v_W} \left[ 3 \zeta(3) + \frac{\pi^2}{12} \left[\frac{\gamma_1}{k_B T}\right] \frac{\mu}{k_B T}\right] + \frac{\pi^2}{3} \left[\frac{\gamma_1}{k_B T}\right] \frac{\mu}{k_B T}.
\]
where $\zeta(x)$ is the Riemann zeta function: $\zeta(3) \approx 1.202$.

Considering that the 2DEHP heat capacities in GLs and GBLs (per area) are defined as $C_{GL} = dE_{GL}/dT$ and $C_{GBL} = dE_{GBL}/dT$, we obtain from Eqs. (8) and (9)

$$C_{GL} \simeq \frac{2(k_BT)^2}{\pi h^2 v_W} \left[ 9\zeta(3) + \frac{2\pi^2}{3} \frac{\mu}{k_BT} \right] \simeq \frac{6.58\pi}{3} \left( \frac{k_BT}{h v_W} \right)^2, \quad (10)$$

$$C_{GBL} \simeq \frac{2(k_BT)^2}{\pi h^2 v_W} \left[ 9\zeta(3) + \frac{\pi^2}{3} \frac{\gamma_1}{k_BT} \right] + \left( \frac{2\pi^2}{3} + \ln 2 \frac{\gamma_1}{T} \right) \frac{\mu}{k_BT} \simeq \frac{\pi}{3} \left( \frac{k_BT}{h v_W} \right)^2 \left( 6.57 + \frac{\gamma_1}{k_BT} \right), \quad (11)$$

Since at $\mu = 0$, according to Eqs. (6) and (7),

$$\Sigma_{GL} \simeq \frac{\pi}{3} \left( \frac{k_BT}{h v_W} \right)^2, \quad (12)$$

$$\Sigma_{GBL} \simeq \frac{\pi}{3} \left( \frac{k_BT}{h v_W} \right)^2 \left( 1 + \frac{6\ln 2}{\pi^2} \frac{\gamma_1}{k_BT} \right), \quad (13)$$

the pertinent heat capacitances, $c_{GL} = C_{GL}/k_B \Sigma_{GL}$ and $c_{GBL} = C_{GBL}/k_B \Sigma_{GBL}$ (normalized by $k_B$, i.e., in units of the Boltzmann constant), per one carrier are equal to

$$c_{GL} \simeq \frac{54\zeta(3)}{\pi^2} \simeq 6.58, \quad (14)$$

$$c_{GBL} \simeq \frac{\pi^2}{6\ln 2} \left( \frac{1 + \frac{54\zeta(3)}{\pi^2} \frac{k_BT}{\gamma_1}}{1 + \frac{6\ln 2}{\pi^2} \frac{k_BT}{\gamma_1}} \right) \simeq 2.37 \left( \frac{1 + 6.58\frac{k_BT}{\gamma_1}}{1 + 2.37\frac{k_BT}{\gamma_1}} \right). \quad (15)$$

**IV. COMMENTS**

At $k_BT \ll \gamma_1$ ($T \lesssim 300$ K), Eq. (13) yields $c_{GBL} \simeq (\pi^2/6\ln 2) \simeq 2.37$. When $T$ is rather high, $c_{GBL}$ increases tending to $c_{GBL} \simeq 6.58$. Figure 2 shows the temperature dependences of the energy densities $E_{GL}$ and $E_{GBL}$ and the heat capacities per one carrier $c_{GL}$ and $c_{GBL}$ calculated using Eqs. (10), (11), (14), and (15) for $\mu = 0$ (equilibrium 2DEHP) assuming $\gamma_1 = 0.4$ eV.

A noticeable deviation of $c_{GL}$ and $c_{GBL}$ from the classical value for nondegenerate 2D systems (i.e., from $c = 1$) seen from Eqs. (14) and (15) and from Fig. 2(c), is associated with the nonparabolicity of the carrier spectra in both GLs and GBLs. The nonparabolicity provides different densities of states (a linear in GLs and a linear rising from a constant at the Dirac point in GBLs), whereas the absence of the energy gap leads to a weak degeneration near the Dirac point ($f_e(0) = f_h(0) \simeq 1/2$). In particular, if we would neglect the partial degeneracy effect, we obtain $c_{GL} = 6$ and $1 < c_{GBL} = \left( 1 + 6k_BT/\gamma_1 \right)/\left( 1 + k_BT/\gamma_1 \right) < 6$, respectively.

The variation of $\mu$ with the effective carrier temperature leads to a small modification of $c_{GL}$ and $c_{GBL}$ assuming a weak deviation from equilibrium. Depending on the pumping or heating conditions, this effect can result in either increase or a decrease in $\mu$ (see, for example, [14]) and, hence, in somewhat varying of $c_{GL}$ and $c_{GBL}$.

In the case when the gapless carrier density of state is
given by a power energy dependence $\rho(\varepsilon) \propto \varepsilon^\xi$, for the heat capacity per a carrier $c_\xi$ one can obtain

$$c_\xi = (\xi + 2) \frac{\int_0^\infty dx x^{\xi+1} [1 + \exp(x - \mu/k_B T)]^{-1}}{\int_0^\infty dx x^{\xi} [1 + \exp(x - \mu/k_B T)]^{-1}}$$

$$= (\xi + 2) \frac{\mathcal{F}_\xi(\mu/k_B T)}{\mathcal{F}_\xi(\mu/k_B T)}.$$  \hspace{4em} (16)

In particular, at $\mu = 0$, Eq. (16) yields

$$c_\xi = (\xi + 2) \frac{\Gamma(\xi + 1) \sum_{\ell=0}^{\infty} \frac{1}{\xi + 1}}{\Gamma(\xi + 1) \sum_{\ell=0}^{\infty} \frac{1}{\xi + 1}}.$$  \hspace{4em} (17)

where $\Gamma(\ell)$ is the Gamma function. For GLs ($\xi = 1$) and GBLs ($\xi = 0$, $k_B T \ll \gamma_1$), from Eq. (17) we obtain

$$c_{GL} = c_1 = \frac{54}{\pi^2}$$

and $c_{GBL} \approx c_0 = \frac{\pi^2}{6 \ln 2}$, that actually coincides with Eqs. (14) and (15).

The renormalization of the carrier spectrum and the density of state energy dependence in GLs, associated with the carrier-carrier interactions (for example, \[40-43\]), affects the GL heat capacity. To estimate the role the Fermi-liquid effect in GLs associated with the inter-carrier interaction, following \[43\], in comparison with Eq. (1) we modify the carrier dispersion law in GLs as follows:

$$\varepsilon^\pm = \pm v_W p \left[ 1 + g \ln \left( \frac{Kh v_W}{\kappa} \right) \right].$$  \hspace{4em} (18)

Here $g = e^2/(8\pi \hbar v_W \kappa)$ is the dimensionless carrier-carrier interaction parameter, where $\kappa$ is the effective dielectric constant, and $K$ is the cut-off parameter \[40\] ($K \approx 0.5 \times 10^8$ cm$^{-1}$).

Considering Eq. (16), i.e., accounting for the carrier velocity renormalization, at $\mu = 0$ for the renormalized carrier density $\Sigma_{GL}$, density of the carrier energy $\mathcal{E}_{GL}$, and the carrier heat capacity per one carrier $c_{GL}$ we obtain

$$r = \frac{\Sigma_{GL}}{\Sigma_{GL}} \approx \frac{\mathcal{E}_{GL}}{\mathcal{E}_{GL}} \approx \left[ 1 + g \ln \left( \frac{Kh v_W}{\kappa} \right) \right]^{-2} \leq 1$$  \hspace{4em} (19)

and

$$c_{GL} \approx c_{GL}.$$  \hspace{4em} (20)

Setting $\kappa = 2.5$, at $T = (10 - 300)$ K, we obtain $r \approx 0.59 - 0.72$. One can see from Eq. (19) that the inclusion of the the Fermi-liquid effect results in natural lowering of the thermal carrier energy (due to a decrease in the density of states near the Dirac point), but, according to Eq. (20), this does not lead to a change in $c_{GL}$.

V. CONCLUSIONS

We calculated the heat capacity per one carrier of the quasi-equilibrium 2DEHP in GLs and GBLs and demonstrated that it can be larger from its classical values. The speed of operation (the switching time, turn-on time, and maximum modulation frequency) of the GL- and GBL-based devices, such as the bolometric photodetectors of the terahertz and infrared radiation, electro-optical modulators, fast thermal radiation emitters, and superluminescent and lasing diodes is affected by the carrier heating/cooling and is determined by the product of $c_{GL}$ or $c_{GBL}$ and the carrier energy relaxation time. Therefore, our results are important for the evaluation of the ultimate characteristics and optimization of such devices.

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