Graphene-on-silicon near-field thermophotovoltaic cell

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(Dated: August 27, 2014)

A graphene layer on top of a dielectric can dramatically influence ability of the material to radiative heat transfer. This property of graphene is used to improve the performance and reduce costs of near-field thermophotovoltaic cells. Instead of low bandgap semiconductors it is proposed to use graphene-on-silicon Schottky photovoltaic cells. One layer of graphene absorbs around 90% of incoming radiation and increases the heat transfer. This is due to excitation of plasmons in graphene, which are automatically tuned in resonance with the emitted light in the mid infrared range. The absorbed radiation excites electron-hole pairs in graphene, which are separated by the surface field induced by the Schottky barrier. For a quasi-monochromatic source the generated power is one order of magnitude larger and efficiency is on the same level as for semiconductor photovoltaic cells.

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I. INTRODUCTION

Thermophotovoltaic (TPV) generators convert radiation emitted by a heated body into electricity [1]. In these devices a hot emitter radiates the electromagnetic energy that is absorbed by a cold photovoltaic (PV) cell (collector). In the solar TPV generators [2] solar light is absorbed and then re-emitted as thermal radiation in a spectrally selective way. Any other heat source also can be used for transformation into electricity: wasted industrial heat, the heat from car engines, computer chips etc. [3, 4]. TPV systems are expected to be quiet, modular, safe, low-maintenance, and pollution-free [5].

The main challenge is to increase both efficiency and output electric power of the devices. The power is restricted by the emissivity of the black body. This restriction, however, is not applicable when the bodies are separated by a distance much smaller than the thermal wavelength [5]. In this near-field range the radiative heat transfer (RHT) can be increased by orders of magnitude [6-8]. Significant enhancement of the RHT in the near-field was demonstrated experimentally [9-14]. The highest efficiency is reached when the emitter is a nearly zero thickness Si substrate, there is no problem to couple the evanescent radiation to electrons in graphene, and the device has a simple structure. The silicon substrate is transparent in the wavelength range \( \lambda = 1.2 - 8.0 \) \( \mu m \) providing the optimal conditions for the RHT [15].

Here we propose to use graphene-on-silicon (g/Si) Schottky photodiodes as a PV cell. In such a generator the emitted radiation is resonantly absorbed in graphene where it excites electrons able to overcome the Schottky barrier, there is no p-n junction and related optical losses. For this device there is no resonance energy exchange between graphene and the semiconductor. A PV cell consisting of InSb substrate covered with graphene was discussed in [26], where hexagonal Boron Nitride (hBN) was used as the emitter. In this device the RHT is enhanced due to the resonance tuning but most of the photons are absorbed in graphene without generation of photoelectrons.

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II. HEAT TRANSFER

We consider graphene-on-silicon Schottky junction in combination with the hBN emitter that gives quasi-monochromatic radiation at 0.195 eV (see Fig. 1(a)). This specific emitter is not the point of interest as long as the photoelectrons can overcome the Schottky barrier. Moreover, a wider range of emitted frequencies is preferable. It is assumed that the bottom of the PV cell is kept at room temperature $T_s = 300 K$ but the emitter temperature $T_e$ can vary.

The energy scheme of the photodiode is shown in Fig. 1(b). Silicon of n-type is shown but p-type also can be used. Infrared radiation from the emitter is absorbed in graphene providing hot electrons that are able to overcome the Schottky barrier $\Phi_b = E_F - \chi_{Si}$, where $E_F$ is the Fermi level in graphene and $\chi_{Si}$ is the electron affinity in silicon. The surface barrier from the Si side is $V_s$ and the forward bias applied to the diode is $V$. The edges of the conduction and valence bands are $E_c$ and $E_v$, respectively, the Fermi level in Si is $E_F^{Si}$.

In contrast with the ordinary PV cells Si is transparent for the emitted radiation. The Fermi level in pristine graphene is $E_F^{0} = 4.56 \text{ eV}$ but it can be adjusted by chemical doping of graphene in a wide range $[29,31]$. The barrier height has to be $\Phi_b = 0.19 \text{ eV}$ or lower if hBN emitter is used. This height can be reached for n-doping in graphene with the relative Fermi level $E_F^g = E_F^0 - E_F = 0.32 \text{ eV}$ counted from the Dirac point. The band bending in Si is defined by the surface potential $eV_s = \Phi_b - (E_F^{Si} - E_c)$.

The photocurrent is generated by photons with the energy $\hbar\omega > \Phi_b$ absorbed in the graphene layer. The radiation of emitter absorbed in graphene $R_g$ can be calculated as

$$ R_g(T_s, d) = R^-(T_s, d) - R^+(T_s, d). $$

Here $R^-(T_s, d)$ is the heat flux from the emitter to the collector taken in the gap just above the graphene layer. This flux is calculated as the $z$-component of the averaged Pointing vector induced by the fluctuations in the emitter. The flux $R^+$ is calculated in a similar way but just below the graphene layer. These fluxes can be presented in the form

$$ R^{\pm}(T_s, d) = \int_0^{\infty} \frac{d\omega}{2\pi} \hbar \omega n_B(\omega, T_e) \Pi^{\pm}(\omega, d), $$

where $n_B(\omega, T) = (e^{\hbar \omega/T} - 1)^{-1}$ is the Bose factor (the Boltzmann constant here is $k_B = 1$). The evanescent spectral function $\Pi^-(\omega, d)$ is

$$ \Pi^-(\omega, d) = \int \frac{d^2q}{(2\pi)^2} \frac{4 \hbar \omega \Im \epsilon_\omega \hbar \omega e^{-2qd}}{|1 - r_s e^{-2qd}|^2}. $$

Here $r_s$ and $r_c$ are the reflection coefficients of the source and collector, respectively, which are functions of $\omega$ and the wave vector $q$ parallel to the plates. The result for $\Pi^+(\omega, d)$ is well known $[8,10,17]$ but the expression for $\Pi^+(\omega, d)$ has to be calculated. It can be done using the standard approach of fluctuational electrodynamics. Skipping the details, the final expression for the spectral function $\Pi_g = \Pi^- - \Pi^+$ is

$$ \Pi_g(\omega, d) = \int \frac{d^2q}{(2\pi)^2} \frac{4 \kappa \hbar \omega \Im \epsilon_\omega \hbar \omega |1 - r_s e^{-2qd}|^2}{|1 - r_s e^{-2qd}|^2}, $$

where $\epsilon_\omega(\omega, q)$ is the dielectric function of graphene and $\kappa$ is the average dielectric constant of the media above and below the graphene layer.

Graphene is responsible for the energy exchange in the system. Without graphene on top of silicon the heat transfer is negligible because Si is transparent for the emitted radiation. It is known that the effect of graphene can be evaluated with a good precision ($\sim \alpha = e^2/\hbar c$) in the non-retarded limit $c \to \infty$ $[18,32,33]$, where only p-polarized evanescent fluctuations contribute to the momentum or energy exchange between parallel plates separated by a submicrometer gap. Therefore, in Eqs. (2)-(4) it is sufficient to take into account only the contribution of p-polarized evanescent waves.

In order to calculate the total energy exchange between bodies we have to include the heat flux going to the emitter and originating from fluctuations in the collector. Normally it can be done by the substitute in Eq. (1)

$$ n_B(\omega, T_s) \to N(\omega, T_s, T_c) = n_B(\omega, T_s) - n_B(\omega, T_c) $$

because in the thermal equilibrium $T_s \to T_c$ the total RHT has to be zero $[5]$. However, the procedure is different if the collector is used as a PV element. As for semiconductors $[34,35]$ the potential difference $V$ results in non-thermal photons that have non-zero chemical potential $\mu_{ph} = eV$ but still can be described by the collector temperature $T_c$. This is true for photons with the energy above the Schottky barrier $\hbar \omega > \Phi_b$; the photons with smaller energies have $\mu_{ph} = 0$. In this case the thermal factor $N(\omega, T_s, T_c)$ is defined as

$$ N(\omega, T_s, T_c) = n_B(\omega, T_s) - n_B(\omega - \mu_{ph}/\hbar, T_c), $$

where we have to understand $\mu_{ph}$ as a discontinuous function of $\omega$

$$ \mu_{ph}(\omega) = \begin{cases} 0 & \hbar \omega < \Phi_b, \\ eV & \hbar \omega > \Phi_b. \end{cases} $$
The final result for the radiative power $P_{rad}$ per unit area is

$$P_{rad}(d, T_s, T_c) = \int_0^\infty \frac{d \omega}{2\pi} \hbar \omega N(\omega, T_s, T_c) \Pi^-(\omega, d). \quad (8)$$

It is similar to the expression used in [20, 27]. The difference is that in our case the collector emits radiation with $h\omega < \Phi_b$ due to presence of graphene, while for a semiconductor PV cell with the bandgap $E_{gap}$ the adopted approximation is that the radiation with $h\omega < E_{gap}$ is not emitted. The radiative power absorbed in the graphene layer $P_g$ can be calculated from [9] with the substitute $\Pi^- \rightarrow \Pi_g$.

### III. GENERATED POWER

Let us assume first that each photon with the energy above $\Phi_b$ which is absorbed in graphene produces one electron in the conduction band of Si. The actual responsibility of the photodiode will be discussed later. In this case the photocurrent generated in the cell [15] is

$$I_{ph}(V) = e \int_0^\infty \frac{d \omega}{2\pi} N(\omega, T_s, T_c) \Pi_g(\omega, d). \quad (9)$$

It is proportional to the number of photons with the energy above $\Phi_b$ absorbed in graphene. The generated electrical power and efficiency of the cell are defined as

$$P_{PV} = V I_{ph}(V), \quad \eta = P_{PV}/P_{rad}. \quad (10)$$

where $P_{PV}$ is similar to that used in Refs. [20, 27]. Note that $\eta$ does not include the efficiency of heating of the emitter and the efficiency of light transformation by the photodiode. The electric power is zero for $V = 0$ corresponding to the short circuit and for $V = (1 - T_c/T_s)\Phi_b/e$ corresponding to the open circuit voltage. The maximal power is realized somewhere in between these values.

The dielectric function of the emitter is described by the Drude-Lorentz model

$$\varepsilon_{hBN}(\omega) = \varepsilon_\infty \left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_L^2 - \omega^2 - i\Gamma \omega}\right) \quad (11)$$

with the parameters of hBN from [36] $\varepsilon_\infty = 4.88$, $\omega_L = 0.2$ eV, $\omega_T = 0.17$ eV, and $\Gamma = 0.66 \times 10^{-3}$ eV. The reflection coefficient of the emitter (p-polarization, non-retarded) $r_s = (\varepsilon_{hBN} - 1)/(\varepsilon_{hBN} + 1)$ has a surface phonon-polariton resonance at 0.195 eV.

The reflection coefficient of the collector $r_c$ can be presented in the form

$$r_c = \frac{\varepsilon_{Si} - 1 + 2\kappa(\varepsilon_g - 1)}{\varepsilon_{Si} + 1 + 2\kappa(\varepsilon_g - 1)}, \quad (12)$$

where $\varepsilon_{Si} \approx 11.9$ is practically a constant for Si in mid IR and $\varepsilon_g(\omega, q)$ is the dielectric function of graphene. In the limit of small relaxation frequency $\gamma \to 0$ the latter can be presented in the form

$$\varepsilon_g(\omega, q) = 1 + \frac{4\sqrt{2}e^2}{\hbar v_F q} \left(1 - \frac{\omega}{\sqrt{\omega^2 - v_F^2 q^2}}\right), \quad (13)$$

where $\alpha_g = e^2/\hbar v_F$ is the coupling constant and $v_F$ is the Fermi velocity in graphene. Equation [13] can be applied in the range $q < 2T/v_F$; finite $\gamma$ can be accounted with the substitute $\omega \to \omega + i\gamma$ and some not essential modification of $\varepsilon_g$ (see [18, 20] for details).

The spatial dispersion of $\varepsilon_g$ is an important property. It results in the plasmon frequency that depends on $q$ even in the non-retarded limit:

$$\hbar \omega_p(q) \approx (2\alpha_g \hbar v_F q \varepsilon_F)^{1/2}. \quad (14)$$

This dependence means that for a body covered with graphene one can always find a value of $q$ that gives the plasmon resonance matching the surface mode in the opposite body. When the substrate permittivity is large, graphene gives only small correction to $r_c$ and there will be no significant increase in RHT. For silicon it is rather large but even a thin native oxide ($h \sim 1$ nm) on Si influences the reflection coefficient. We take this oxide into account in our calculations and use the graphene relaxation frequency $\gamma = 33$ meV ($5 \times 10^{13}$ rad/s).

The results are shown in Fig. 2. First, using Eq. (10) the output power was maximized by varying the operating voltage of the cell $V$. The theoretical limit of this voltage $V < (1 - T_c/T_s)\Phi_b/e$ and the value found from the maximization of $P_{PV}$ at $d = 10$ nm are shown in Fig. 2a as functions of the emitter temperature. The optimal value of $V$ varies only slightly with the distance. The net radiative power $P_{rad}$ and the power absorbed in graphene $P_g$ are shown in Fig. 2b. The curves are nearly coincide because about 90% of incoming radiation is absorbed in graphene. This exceptional phenomena is the result of plasmon excitation in resonance with the radiation of the emitter. The output power of the cell is also presented in Fig. 2b. For comparison $P_{PV}$ for the InSb PV cell is shown too, which is one order of magnitude smaller. The efficiency for g/Si cell shown in Fig. 2c is somewhat smaller than that for the InSb cell. It happens because 90% of the light incoming on the g/Si cell generates photocarriers, while it was assumed that 100% of the light generates carriers in the semiconductor cell. The latter, of course, is not true because to contribute to the photocurrent the electron-hole pairs has to reach the depletion layer before recombination [37, 38]. We can conclude that the TPV element with the graphene-on-Si Schottky PV cell can outperform semiconductor PV cells if the responsivity of the Schottky photodiode will be comparable with that of low bandgap semiconductors.

The dependence of the scaled output powers $P_{PV}^* = P_{PV} \times (d/10nm)^2$ on the distance $d$ is shown in Fig. 2d for three different thicknesses of the SiO$_2$ layer on Si. All the curves are presented for $T_c = 600$ K. At the maximum the frequency of plasmon matches the resonance in
the emitter $\hbar \omega_p(q) \approx 0.195$ eV at $q \approx 1/2d$. Deviation from this relation in any direction will result in decrease of the scaled power. Graphene on a thin silicon oxide film corresponds to some intermediate situation between pure Si and pure SiO$_2$.

For low barrier height the surface electric field at high $T_s$ becomes too small for the carriers separation (the same problem exists for low bandgap semiconductors). It is therefore interesting to analyze the behavior of the g/Si cell for different barrier heights. To perform this analysis we model the emitter with the dielectric function similar to (11) but with the parameters $\varepsilon_\infty = 5$, $(\omega_L/\omega_T)^2 - 1 = 0.5$, $\Gamma/\omega_T = 5 \times 10^{-3}$, which are close to those for hBN. The resonance frequency of the reflection coefficient

$$\omega_r = \omega_T(\varepsilon_{em}(0) + 1)^{1/2}(\varepsilon_\infty + 1)^{-1/2}$$

is the varied parameter, where $\varepsilon_{em}(0)$ is the static permittivity of the emitter. It is assumed that the barrier height is somewhat smaller than $\omega_r$. Some results are shown in Fig. 3. The left panel shows that the output power is smaller for higher barrier at low $T_s$ but becomes comparable or even larger at high $T_s$ as the curves 1 and 2 demonstrate. However, when the barrier is too high $P_{PV}$ becomes smaller at all temperatures. The reason is that the value of the momentum $q$ in (14) for which $\hbar \omega_p > \Phi_b$ becomes significantly larger than $1/2d$. It reduces the radiative power due to the factor $e^{-2qd}$ and results in the decrease of $P_{PV}$. The output power produced by InAs PV cell with the bandgap 0.36 eV is shown for comparison in the same panel. The right panel shows the fraction of the radiative power absorbed in graphene for different barrier heights. This fraction is always large especially at high $T_s$. It is somewhat smaller at low temperatures and high barriers.

Dependence on the barrier height demonstrates that g/Si PV cell is able to support plasmons in the mid IR range where it is superior over the semiconductor PV cells. At shorter wavelengths $\lambda < 3.5 \mu m$ the radiative heat exchange is significantly reduced and the g/Si cell loses its advantages.

IV. DISCUSSION

Up to now we assumed that the photodetector is perfect. The typical responsivity of low bandgap semiconductors is 1 A/W. It is still larger than the reported value 0.13 A/W [25] for the g/Si Schottky photodiode. However, the main problem of graphene-based photodetectors is the coupling of light with graphene. For incoming propagating waves absorption in graphene is small $\sim \alpha$. To increase the responsivity one has to couple light with sophisticated optical structures (see [25] and references therein). These structures inevitably add losses reducing the effective responsivity. Additionally, the graphene-Si junction was not optimized. The configuration used in our scheme does not suffer from the coupling problem because the heat transfer is realized via the evanescent waves, for which we have found that 90% of incoming radiation is absorbed in a single graphene layer. Therefore, the responsivity for our configuration can be increased.
FIG. 3: (Color online). (left) The output power at $d = 10$ nm as a function of temperature for different barrier heights. The curves 1, 2, and 3 correspond to the barriers 0.18, 0.28, and 0.33 eV, respectively. The emitter resonance frequency is 0.02 eV higher in all cases. The gray curve shows the result for InAs PV cell. (right) Fraction of the incident radiation absorbed in the graphene layer for different barrier heights.

We considered the near-field TPV element having as a PV cell graphene-on-silicon Schottky photodiode. Due to presence of graphene this PV cell has resonant heat exchange with the emitter; a single layer of graphene absorbs 90% of incoming radiation that can be efficiently transformed to photocurrent. Already for quasi-monochromatic emitter the device demonstrates well advanced characteristics.

V. CONCLUSIONS

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