Theory of the giant plasmon-enhanced second-harmonic generation in graphene and semiconductor two-dimensional electron systems

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An analytical theory of the nonlinear electromagnetic response of a two-dimensional (2D) electron system in the second order in the electric field amplitude is developed. The second-order polarizability and the intensity of the second harmonic signal are calculated within the self-consistent-field approach both for semiconductor 2D electron systems and for graphene. The second harmonic generation in graphene is shown to be about 2 orders of magnitude stronger than in GaAs quantum wells at typical experimental parameters. Under the conditions of 2D plasmon resonance the second harmonic radiation intensity is further increased by several orders of magnitude.

I. INTRODUCTION

Graphene is a recently discovered[1] purely two-dimensional (2D) material consisting of a monolayer of sp^2-bonded carbon atoms arranged in a hexagonal lattice. Electrons and holes in graphene are massless Dirac fermions and this leads to a variety of interesting and unusual electrical and optical properties of this material.[2–7] It promises a lot of applications in electronics, optics, and other areas.[4–7]

It has been predicted[8] that the unusual linear energy dispersion of charge carriers should lead to a *strongly nonlinear* electromagnetic response of graphene: irradiation with electromagnetic waves should stimulate the emission of higher frequency harmonics from graphene. The theory of the nonlinear electromagnetic response of graphene has been further developed in Refs. 9–16. Experimentally the higher harmonics generation and frequency mixing effects have been observed in Refs. 17–21.

In theoretical papers,[9–16] only the normal incidence of radiation on a uniform graphene layer has been studied. The experimental demonstration[20] of third-order emission of radiation at the frequency $2\omega_1 - \omega_2$ at the bichromatic irradiation by the frequencies ω_1 and ω_2 has confirmed that graphene manifests nonlinear properties and that its third-order effective nonlinear susceptibility is much higher than that in a number of other materials.[15,16,20] However, the intensity of the emitted signal $I_{2\omega_1 \pm \omega_2} \propto I_{\omega_1}^2 I_{\omega_2}$ is proportional to the third power of the intensities of the incident waves;[20] therefore, to observe the third-order nonlinear effects one needs quite powerful sources of radiation.

Substantially stronger nonlinear effects could be expected in the second order in a radiation electric field. The second-order effects, for example, the second harmonic generation, are proportional to the second power of the incident wave intensity, $I_{2\omega} \propto I_{\omega}^2$. However, graphene is a centrosymmetric material; therefore, at the normal incidence of radiation, second-order effects are forbidden by symmetry.

The symmetry arguments do not hinder the observation of second-order effects at the oblique incidence of radiation on the 2D electron layer. If the incident wave has a wave-vector component **q** parallel to the plane of the 2D layer, one could observe much stronger second-harmonic radiation compared to the third-order effects.[20] Moreover, at the oblique incidence of radiation, one can resonantly excite the 2D plasma

waves[22–29] in the system (e.g., in the attenuated total reflection or grating coupling geometry), which would lead to resonant enhancement of higher harmonics.[30]

In this paper we theoretically study the second-order nonlinear electromagnetic response of 2D electron systems, including both graphene and conventional 2D structures with parabolic electron energy dispersion (for a recent work on the second harmonic generation in bulk solids, see, e.g., Ref. 31). We calculate the second-order polarizability $\alpha^{(2)}$ of 2D electrons (Sec. II) and show that in graphene it is *at least 1 order of magnitude larger* than in typical semiconductor structures (e.g., in GaAs/AlGaAs quantum wells). Then we calculate the self-consistent response of the system to external radiation, taking into account 2D plasmon excitation (Sec. III), and show that the intensity of the second harmonic signal can be further increased *by several orders of magnitude*. In Sec. IV we summarize our results.

II. RESPONSE EQUATIONS

A. General solution of the quantum kinetic equation

We consider a 2D electron system occupying the plane $z = 0$ and described by the Hamiltonian $\hat{H}_0$. The single-particle Schrödinger equation has the form

$$\hat{H}_0|\lambda\rangle = E_\lambda|\lambda\rangle, \tag{1}$$

where λ is a set of quantum numbers, and E_λ and $|\lambda\rangle$ are the eigenenergies and eigenfunctions of the system. Our goal is to describe the system response at frequencies corresponding to excitation of the 2D plasma waves. Since the phase velocity of 2D plasmons is typically much lower than the velocity of light, we apply the quasi-static approximation and describe the electric field acting on the electrons by the scalar potential

$$\phi(\mathbf{r},t) = \phi_{\mathbf{q}\omega}(z)e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + \text{c.c.}, \tag{2}$$

where $\mathbf{q} = (q_x, q_y)$ and c.c. means the complex conjugate. The response of the 2D gas to potential (2) is determined by the Liouville equation,

$$i\hbar\frac{\partial\hat{\rho}}{\partial t} = [\hat{H},\hat{\rho}] = [\hat{H}_0 + \hat{H}_1,\hat{\rho}], \tag{3}$$

where $\hat{\rho}$ is the density matrix and $\hat{H}_1 = -e\phi(\mathbf{r},t)$. Using Eq. (3) we now have to calculate the charge density

fluctuations,

$$\rho(\mathbf{r},t) = \rho_{\mathbf{q}\omega}e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + \rho_{2\mathbf{q},2\omega}e^{2i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + \text{c.c.}, \qquad (4)$$

in the first and second orders in the potential amplitudes $\phi_{\mathbf{q}\omega}$ and the corresponding first- and second-order polarizabilities $\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega}$ and $\alpha^{(2)}_{2\mathbf{q},2\omega;\mathbf{q}\omega,\mathbf{q}\omega}$, defined as

$$\rho_{\mathbf{q}\omega} = \alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega}\phi_{\mathbf{q}\omega}, \qquad (5)$$

$$\rho_{2\mathbf{q},2\omega} = \alpha^{(2)}_{2\mathbf{q},2\omega;\mathbf{q}\omega,\mathbf{q}\omega}\phi_{\mathbf{q}\omega}\phi_{\mathbf{q}\omega}. \qquad (6)$$

In the absence of the perturbation $\hat{H}_1$ the density matrix $\hat{\rho}_0$ satisfies the equation

$$\hat{\rho}_0|\lambda\rangle = f_\lambda|\lambda\rangle, \qquad (7)$$

where $f_\lambda = f(E_\lambda)$ is the Fermi distribution function. Expanding $\hat{\rho}$ in powers of the electric potential, $\hat{\rho} = \hat{\rho}_0 + \hat{\rho}_1 + \hat{\rho}_2 + \cdots$, and calculating the charge density fluctuations $-e\mathrm{Sp}[\delta(\mathbf{r}-\mathbf{r}_0)(\hat{\rho}_1 + \hat{\rho}_2 + \cdots)]$, we get

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} = \frac{e^2}{S}\sum_{\lambda\lambda'}\frac{f_{\lambda'} - f_\lambda}{E_{\lambda'} - E_\lambda + \hbar\omega + i0}\langle\lambda'|e^{-i\mathbf{q}\cdot\mathbf{r}}|\lambda\rangle\langle\lambda|e^{i\mathbf{q}\cdot\mathbf{r}}|\lambda'\rangle, \qquad (8)$$

$$\begin{aligned}\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega} = &-\frac{e^3}{S}\sum_{\lambda\lambda'}\frac{\langle\lambda'|e^{-i2\mathbf{q}\cdot\mathbf{r}}|\lambda\rangle}{E_{\lambda'} - E_\lambda + 2\hbar\omega + 2i0}\\ &\times\sum_{\lambda''}\langle\lambda|e^{i\mathbf{q}\cdot\mathbf{r}}|\lambda''\rangle\langle\lambda''|e^{i\mathbf{q}\cdot\mathbf{r}}|\lambda'\rangle\\ &\times\left(\frac{f_{\lambda'} - f_{\lambda''}}{E_{\lambda'} - E_{\lambda''} + \hbar\omega + i0}\right.\\ &\left. -\frac{f_{\lambda''} - f_\lambda}{E_{\lambda''} - E_\lambda + \hbar\omega + i0}\right), \qquad (9)\end{aligned}$$

where S is the sample area. The first-order polarizability, Eq. (8), is proportional to the polarization operator $\Pi(\mathbf{q},\omega)$ (see Ref. 32), $\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} = -e^2\Pi(\mathbf{q},\omega)$. For a conventional 2D electron gas (with a parabolic electron energy dispersion), the linear polarizability, Eq. (8), has been calculated in Ref. 32; for 2D electrons in graphene (with a linear energy dispersion) this has been done in Refs. 22 and 23.

We apply the general formulas, (8) and (9), to conventional 2D electron systems in semiconductor heterostructures and to graphene. In the former case the spectrum of 2D electrons is parabolic; in the latter case it is linear. In both cases the set of quantum numbers $|\lambda\rangle = |l\mathbf{k}\sigma\rangle$ consists of the sub-band index l, the wave vector $\mathbf{k}$, and the spin σ. To specify the general expressions (8) and (9), we consider the long-wavelength limit, which is quantitatively described by the conditions

$$q \ll \max\{k_F,k_T\}, \quad q \ll \omega/\max\{v_F,v_T\}, \qquad (10)$$

in semiconductor 2D electron systems and the conditions

$$q \ll \max\{k_F,k_T\}, \quad q \ll \omega/v_F, \qquad (11)$$

in graphene. Here k_F and v_F are the Fermi wave vector and Fermi velocity, respectively, and k_T and v_T are the thermal wave vector and velocity, respectively. In a gas with a parabolic dispersion, $k_F = \sqrt{2mE_F}/\hbar$, $k_T = \sqrt{2mT}/\hbar$, and

$v_{F,T} = \hbar k_{F,T}/m$, where m is the effective electron mass, T is the temperature, and the Fermi energy E_F is counted from the bottom of the parabolic band. In a 2D gas with a linear energy dispersion (in graphene), $k_F = |\mu|/\hbar v_F$ and $k_T = T/\hbar v_F$, where μ is the chemical potential counted from the Dirac point (μ can be positive and negative) and v_F is the Fermi velocity. With typical experimental parameters, conditions (10) and (11) restrict the wave vector q by values of $\sim 10^6$ cm^{-1}, which is sufficient for the description of most experiments.

Under conditions (10) and (11), the general expressions (8) and (9) can be substantially simplified and we get the first- and second-order polarizabilities of a 2D electron gas in the form

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} \approx \frac{e^2 q_\alpha q_\beta}{(\hbar\omega)^2}\frac{g_s}{S}\sum_{l\mathbf{k}}\left(-\frac{\partial f_{l\mathbf{k}}}{\partial k_\alpha}\right)\frac{\partial E_{l\mathbf{k}}}{\partial k_\beta}, \qquad (12)$$

$$\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega} \approx \frac{3e^3 q_\alpha q_\beta q_\gamma q_\delta}{2(\hbar\omega)^4}\frac{g_s}{S}\sum_{l\mathbf{k}}\frac{\partial f_{l\mathbf{k}}}{\partial k_\alpha}\frac{\partial E_{l\mathbf{k}}}{\partial k_\beta}\frac{\partial^2 E_{l\mathbf{k}}}{\partial k_\gamma\partial k_\delta}, \qquad (13)$$

where $g_s = 2$ is the spin degeneracy.

B. 2D electron gas with a parabolic energy dispersion

Let us now apply the obtained formulas, (12) and (13), to a conventional 2D electron gas with a parabolic energy dispersion. In this case there is only one energy sub-band ($l = 1$),

$$E_{l\mathbf{k}} \equiv E_{\mathbf{k}} = \frac{\hbar^2 k^2}{2m}, \qquad (14)$$

and the wave functions are plane waves. Substituting Eq. (14) into Eqs. (12) and (13) we get

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} \approx \frac{n_s e^2 q^2}{m\omega^2} \qquad (15)$$

and

$$\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega} \approx -\frac{3n_s e^3 q^4}{2m^2\omega^4}. \qquad (16)$$

In semiconductor 2D electron systems both $\alpha^{(1)}$ and $\alpha^{(2)}$ are proportional to the 2D electron gas density n_s. The formula (15) was obtained in Ref. 32.

C. 2D electron gas with a linear energy dispersion (graphene)

In graphene the spectrum of electrons can be found in a tight-binding approximation.[33] It consists of two energy bands, the wave functions are described by Bloch functions and the energy of the electrons is

$$E_{l\mathbf{k}} = (-1)^l t|\mathcal{S}_{\mathbf{k}}|, \quad l = 1,2, \qquad (17)$$

where t is the transfer integral, $\mathcal{S}_{\mathbf{k}}$ is a complex function defined as

$$\mathcal{S}_{\mathbf{k}} = 1 + e^{i\mathbf{k}\cdot\mathbf{a}_1} + e^{i\mathbf{k}\cdot\mathbf{a}_2} = 1 + 2\cos(k_x a/2)e^{i\sqrt{3}k_y a/2}, \qquad (18)$$

$\mathbf{a}_1 = a(1/2,\sqrt{3}/2)$ and $\mathbf{a}_2 = a(-1/2,\sqrt{3}/2)$ are the basis vectors of the graphene hexagonal lattice, and $a = |\mathbf{a}_1| = |\mathbf{a}_2|$ is the lattice constant. In graphene $t \approx 3$ eV and $a = 2.46$ Å.

Formulas (8) and (9) and formulas (12) and (13) are valid for the full graphene energy dispersion; that is, the integration in these formulas is performed over the whole Brillouin zone.

Under the real experimental conditions, when $|\mu| \ll t$, the main contribution to integrals (12) and (13) is given by the vicinity of two Dirac points, $\mathbf{K}_1 = (2\pi/a)(1/3, 1/\sqrt{3})$ and $\mathbf{K}_2 = (2\pi/a)(2/3, 0)$. Near these points the function $\mathcal{S}_\mathbf{k}$ vanishes, $\mathcal{S}_{\mathbf{K}_1} = \mathcal{S}_{\mathbf{K}_2} = 0$, and the energy, Eq. (17), can be approximated by linear functions,

$$E_{l\mathbf{k}} = (-1)^l \hbar v_F |\tilde{\mathbf{k}}| = (-1)^l \hbar v_F |\mathbf{k} - \mathbf{K}_v|, \quad v = 1, 2, \quad (19)$$

where v is the valley index and $\tilde{\mathbf{k}} = \mathbf{k} - \mathbf{K}_v$ is the electron wave vector counted from the corresponding Dirac points. Here the velocity parameter v_F (the Fermi velocity) is related to the transfer integral and the lattice constant $v_F = \sqrt{3}ta/2\hbar$; in graphene $v_F \approx 10^8$ cm/s. Omitting below the tilde over the wave vector $\mathbf{k}$ and calculating the derivatives,

$$\frac{\partial E_{l\mathbf{k}}}{\partial k_\alpha} = (-1)^l \hbar v_F \frac{k_\alpha}{k}, \quad \frac{\partial^2 E_{l\mathbf{k}}}{\partial k_\alpha \partial k_\beta} = (-1)^l \hbar v_F \frac{k^2 \delta_{\alpha\beta} - k_\alpha k_\beta}{k^3}, \quad (20)$$

we get the first-order polarizability in the form

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} = \frac{e^2 g_s g_v q^2 T}{2\pi \hbar^2 \omega^2} \ln\left(2 \cosh \frac{\mu}{2T}\right), \quad (21)$$

where $g_v = 2$ is the valley degeneracy. If the temperature is low compared to the chemical potential, $T \ll |\mu|$, we get, from (21), the result obtained in Refs. 22 and 23:

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} = \frac{e^2 g_s g_v q^2 |\mu|}{4\pi \hbar^2 \omega^2}, \quad T \ll |\mu|. \quad (22)$$

In the opposite case, $|\mu| \ll T$, one has

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} = \frac{e^2 g_s g_v q^2 T}{4\pi \hbar^2 \omega^2}\left(2 \ln 2 + \left(\frac{\mu}{2T}\right)^2\right), \quad |\mu| \ll T. \quad (23)$$

Now consider the second-order polarizability of graphene. Using Eqs. (13) and (20) we get

$$\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega} \approx -\frac{3e^3 g_s g_v q^4 v_F^2}{32\pi \hbar^2 \omega^4} \tanh \frac{\mu}{2T}$$

$$\equiv -\frac{3e}{8}\left(\frac{q v_F}{\omega}\right)^2 \frac{\partial \alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega}}{\partial \mu}. \quad (24)$$

The second-order graphene polarizability $\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}$ is an odd function of the chemical potential and *does not depend on the electron or hole density* at $|\mu| \gtrsim T$. This is a direct consequence of the linear energy dispersion, Eq. (19), and essentially differs from the case of a conventional 2D electron gas, Eq. (16), for which $\alpha^{(2)} \propto n_s$. The q and ω dependencies of $\alpha^{(2)}$ in the linear- and parabolic-spectrum cases are the same: $\alpha^{(2)} \propto q^4/\omega^4$.

In the graphene formulas (12) and (13), and (21) and (24), interband transitions are neglected. In the case most relevant for the experiments, $|\mu| \gg T$, interband transitions can be ignored at $\hbar\omega \ll 2|\mu|$, which agrees with the long-wavelength conditions, Eq. (11), and embraces the broad frequency range from microwave up to near-infrared frequencies. At $\hbar\omega \gtrsim 2|\mu|$ as well as at $|\mu| \lesssim T$ the interband terms in Eqs. (8) and (9) lead to a weak interband absorption,[34] which also does not substantially influence our results.

Comparing the second-order polarizability of graphene, Eq. (24), with that of a conventional (semiconductor) 2D

electron gas, Eq. (16), we get

$$\frac{\alpha^{(2)}_{\text{graphene}}}{\alpha^{(2)}_{\text{semicond}}} = \frac{(v_F^2)_{\text{graphene}}}{2(v_F^2)_{\text{semicond}}}. \quad (25)$$

Since the Fermi velocity in graphene ($\simeq 10^8$ cm/s) is substantially higher than in typical semiconductor (e.g., GaAs) structures, the nonlinear polarizability of graphene is 1–2 orders of magnitude larger than in semiconductors. For example, in GaAs/AlGaAs quantum wells with an electron density of $n_s \simeq 3 \times 10^{11}$ cm^{-2}, the Fermi velocity is $v_F^{\text{GaAs}} \simeq 2.25 \times 10^7$ cm/s, and we get

$$\frac{\alpha^{(2)}_{\text{graphene}}}{\alpha^{(2)}_{\text{GaAs}}} \approx 10. \quad (26)$$

III. SELF-CONSISTENT FIELD

Equations (5) and (6) determine the first- and second-order response of a 2D electron gas to an electric field *really acting* on the electrons. Consider now the experimentally relevant formulation of the problem when the system responds to an *external* field $\phi^{\text{ext}}_{\mathbf{q}\omega}$. Using a self-consistent field concept, we solve, first, the linear response and, then, the second-order response problem.

A. Linear response

Consider a 2D electron system under the action of an external electric potential $\phi^{\text{ext}}(\mathbf{r}, z, t) = \phi^{\text{ext}}_{\mathbf{q}\omega}(z)e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t}$. In the first order in the external field amplitude the resulting 2D charge density will also contain the $\mathbf{q}\omega$ harmonic $\rho(\mathbf{r}, t) = \rho_{\mathbf{q}\omega}e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t}$. The density fluctuation creates, in its turn, an induced potential $\phi^{\text{ind}}(\mathbf{r}, z, t) = \phi^{\text{ind}}_{\mathbf{q}\omega}e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t}$ determined by the Poisson equation,

$$\Delta\phi_{\text{ind}}(\mathbf{r}, z, t) = -4\pi\rho(\mathbf{r}, t)\delta(z), \quad (27)$$

and given by

$$\phi^{\text{ind}}_{\mathbf{q}\omega}(z = 0) \equiv \phi^{\text{ind}}_{\mathbf{q}\omega} = \frac{2\pi}{q}\rho_{\mathbf{q}\omega}. \quad (28)$$

The density $\rho_{\mathbf{q}\omega}$ is determined here by the response equation (5), in which the potential really acting on electrons should stay in the right-hand side. This is not the external field but the *total* electric field $\phi^{\text{tot}}_{\mathbf{q}\omega} = \phi^{\text{ext}}_{\mathbf{q}\omega} + \phi^{\text{ind}}_{\mathbf{q}\omega}$ produced both by the external charges and by the 2D electrons themselves. Then we get

$$\phi^{\text{ind}}_{\mathbf{q}\omega} \equiv \phi^{\text{tot}}_{\mathbf{q}\omega} - \phi^{\text{ext}}_{\mathbf{q}\omega} = \frac{2\pi}{q}\rho_{\mathbf{q}\omega} \equiv \frac{2\pi}{q}\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega}\phi^{\text{tot}}_{\mathbf{q}\omega} \quad (29)$$

and the known linear-response formula

$$\phi^{\text{tot}}_{\mathbf{q}\omega} = \frac{\phi^{\text{ext}}_{\mathbf{q}\omega}}{\epsilon(\mathbf{q}, \omega)}, \quad (30)$$

with the dielectric function

$$\epsilon(\mathbf{q}, \omega) = 1 - \frac{2\pi}{q}\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega}. \quad (31)$$

If the wave vector $\mathbf{q}$ and the frequency ω of the external wave satisfy the condition

$$\epsilon(\mathbf{q}, \omega) = 0, \quad (32)$$

one gets a resonance in Eq. (30). This resonance corresponds to the excitation of the eigen–collective modes of the system: the 2D plasmons.

Consider a 2D electron gas with a parabolic energy dispersion. Substituting the linear polarizability, Eq. (15), into Eq. (31), we get, from (32), the known spectrum of the 2D plasmon,

$$\omega^2 = \omega_p^2(q) = \frac{2\pi n_s e^2}{m} q, \tag{33}$$

first obtained in Ref. 32 (we ignore the dielectric constant of the surrounding medium; if the 2D gas is immersed in an insulator with the dielectric constant κ, e^2 should be replaced here with e^2/κ).

In the case of graphene, the spectrum of 2D plasmons follows from Eqs. (21), (31), and (32):

$$\omega^2 = \omega_p^2(q) = \frac{e^2 g_s g_v T q}{\hbar^2} \ln\left(2\cosh\frac{|\mu|}{2T}\right). \tag{34}$$

In the limit $|\mu| \gg T$ this gives the result

$$\omega_p^2(q) = \frac{e^2 g_s g_v |\mu|}{2\hbar^2} q \tag{35}$$

obtained in Refs. 22 and 23. In the opposite case, $|\mu| \ll T$, we get

$$\omega_p^2(q) = \frac{e^2 g_s g_v T \ln 2}{\hbar^2} q. \tag{36}$$

The 2D plasmon problem in the regime $\mu = 0$ has been considered in Ref. 35. The result reported in Ref. 35 differs from the correct formula, Eq. (36), by a factor of 4π.

B. Second-order self-consistent response

Let us now consider the second-order response to the external potential $\phi_{\mathbf{q}\omega}^{\text{ext}} e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t} + \text{c.c.}$ The induced and total potential will now contain the frequency harmonics $\pm(\mathbf{q}\omega)$ and $\pm 2(\mathbf{q}\omega)$. The self-consistent charge density then reads

$$\rho(\mathbf{r},t) = \alpha_{\mathbf{q}\omega;\mathbf{q}\omega}^{(1)} \phi_{\mathbf{q}\omega}^{\text{tot}} e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t} + \alpha_{2\mathbf{q}2\omega;2\mathbf{q}2\omega}^{(1)} \phi_{2\mathbf{q}2\omega}^{\text{tot}} e^{i2\mathbf{q}\cdot\mathbf{r}-i2\omega t}$$
$$+ \alpha_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}^{(2)} \phi_{\mathbf{q}\omega}^{\text{tot}} \phi_{\mathbf{q}\omega}^{\text{tot}} e^{i2\mathbf{q}\cdot\mathbf{r}-i2\omega t} + \text{c.c.}, \tag{37}$$

where the first two terms correspond to the linear response to the first and second harmonics, and the third term, to the second-order response to the first $(\mathbf{q}\omega)$ harmonic of the total potential. The complex conjugate terms describe the negative $(\mathbf{q}\omega)$ harmonics. The second-order response to $\phi_{\mathbf{q}\omega}^{\text{tot}} \phi_{-(\mathbf{q}\omega)}^{\text{tot}}$ vanishes.

The Fourier harmonics of the induced potential follow from Eq. (37) and the Poisson equation (27):

$$\phi_{\text{ind}}(\mathbf{r},t) = \frac{2\pi}{q} \alpha_{\mathbf{q}\omega;\mathbf{q}\omega}^{(1)} \phi_{\mathbf{q}\omega}^{\text{tot}} e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t}$$
$$+ \frac{2\pi}{2q} \alpha_{2\mathbf{q}2\omega;2\mathbf{q}2\omega}^{(1)} \phi_{2\mathbf{q}2\omega}^{\text{tot}} e^{i2\mathbf{q}\cdot\mathbf{r}-i2\omega t}$$
$$+ \frac{2\pi}{2q} \alpha_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}^{(2)} \phi_{\mathbf{q}\omega}^{\text{tot}} \phi_{\mathbf{q}\omega}^{\text{tot}} e^{i2\mathbf{q}\cdot\mathbf{r}-i2\omega t} + \text{c.c.}$$
$$\equiv \phi_{\mathbf{q}\omega}^{\text{ind}} e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t} + \phi_{2\mathbf{q}2\omega}^{\text{ind}} e^{i2\mathbf{q}\cdot\mathbf{r}-i2\omega t} + \text{c.c.} \tag{38}$$

Now equating the amplitudes of the first-order harmonic $(\mathbf{q}\omega)$, we get from here Eqs. (29) and (30). Equating the coefficients

at the $(2\mathbf{q},2\omega)$ harmonic and taking into account that the second harmonic component is absent in the external potential $\phi_{2\mathbf{q}2\omega}^{\text{ext}} = 0$, we get

$$\phi_{2\mathbf{q}2\omega}^{\text{ind}} \equiv \phi_{2\mathbf{q}2\omega}^{\text{tot}} = \frac{2\pi}{2q} \alpha_{2\mathbf{q}2\omega;2\mathbf{q}2\omega}^{(1)} \phi_{2\mathbf{q}2\omega}^{\text{tot}} + \frac{\pi}{q} \alpha_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}^{(2)} \phi_{\mathbf{q}\omega}^{\text{tot}} \phi_{\mathbf{q}\omega}^{\text{tot}} \tag{39}$$

and, finally,

$$\phi_{2\mathbf{q}2\omega}^{\text{tot}} = \frac{\pi}{q} \frac{\alpha_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}^{(2)}}{\epsilon(2\mathbf{q},2\omega)} \phi_{\mathbf{q}\omega}^{\text{tot}} \phi_{\mathbf{q}\omega}^{\text{tot}}$$
$$= \frac{\pi}{q} \frac{\alpha_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}^{(2)}}{\epsilon(2\mathbf{q},2\omega) [\epsilon(\mathbf{q},\omega)]^2} \phi_{\mathbf{q}\omega}^{\text{ext}} \phi_{\mathbf{q}\omega}^{\text{ext}} \tag{40}$$

(compare with Ref. 36).

Formula (40), together with (16) and (24), represents the main result of this work. One sees that the amplitude of the second harmonic potential is resonantly enhanced at frequency $\omega = \omega_p(q)$ [the second-order pole corresponding to the vanishing dielectric function $\epsilon(\mathbf{q},\omega)$] and at frequency $\omega = \omega_p(q)/\sqrt{2}$ [the first-order pole corresponding to the vanishing dielectric function $\epsilon(2\mathbf{q},2\omega)$]. These resonances lead to a huge enhancement of the second harmonic radiation intensity.

C. Estimates of the second harmonic radiation intensity

Let us estimate the intensity of the second harmonic signal. Assume that the external potential is

$$\phi^{\text{ext}}(\mathbf{r},t) = \phi_0 \cos(\mathbf{q}\cdot\mathbf{r} - \omega t), \tag{41}$$

so that $\phi_{\mathbf{q}\omega}^{\text{ext}} = \phi_0/2$. Then the total potential at frequency 2ω obtained from Eq. (40) reads

$$\phi_{2\mathbf{q}2\omega}^{\text{tot}}(\mathbf{r},t) = \frac{\pi\phi_0^2}{2q} \alpha_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}^{(2)}$$
$$\times \frac{\omega^6 \cos(2(\mathbf{q}\cdot\mathbf{r} - \omega t))}{\sqrt{\left(\omega^2 - \frac{\omega_p^2(2q)}{4}\right)^2 + \frac{\omega^2\gamma^2}{4}} \left[(\omega^2 - \omega_p^2(q))^2 + \omega^2\gamma^2\right]}, \tag{42}$$

where we have introduced the momentum scattering rate γ in the dielectric function,

$$\epsilon(\mathbf{q},\omega) = 1 - \frac{\omega_p^2(q)}{\omega(\omega + i\gamma)}, \tag{43}$$

to remove unphysical divergencies of the plasma resonances. Now introducing the intensity of the incident $I_{\mathbf{q}\omega}^{\text{ext}} \sim cE_0^2/8\pi$ and of the second harmonic wave $I_{2\mathbf{q}2\omega}^{\text{tot}} \sim cE_{\text{tot}}^2/8\pi$, where E_0 and E_{tot} are the fields corresponding to potentials (41) and (42), respectively, we get the following results.

(1) In a 2D electron gas with a parabolic electron energy dispersion (conventional semiconductor structures),

$$I_{\text{tot}}^{\text{semic}} = \frac{18\pi^3 n_s^2 e^6 q^4}{m^4 c}$$
$$\times \frac{\omega^4}{\left[(\omega^2 - \frac{\omega_p^2(q)}{2})^2 + \frac{\omega^2\gamma^2}{4}\right]\left[(\omega^2 - \omega_p^2(q))^2 + \omega^2\gamma^2\right]^2} I_{\text{ext}}^2. \tag{44}$$
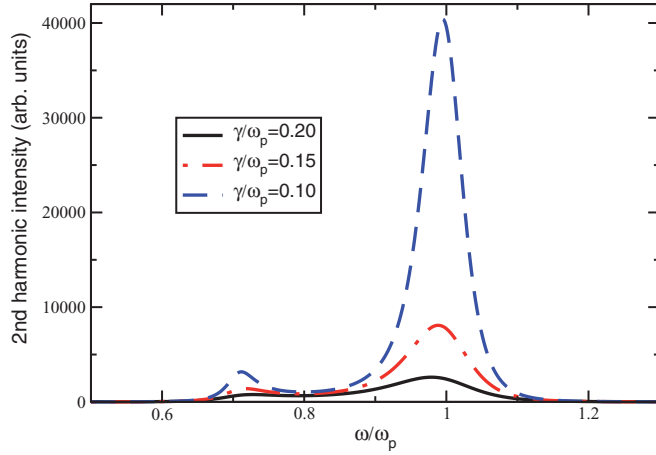
FIG. 1. (Color online) Intensity of the second harmonic radiation in semiconductor structures and in graphene as a function of the frequency $\omega/\omega_p(q)$. The parameter of the curves is $\gamma/\omega_p(q)$.

(2) In a 2D electron gas with a linear energy dispersion (graphene),

$$
I_{\text{tot}}^{\text{graphene}} = \frac{9\pi e^6 v_F^4 q^4}{8\hbar^4 c} \tanh^2\left(\frac{\mu}{2T}\right)
$$
$$
\times \frac{\omega^4}{\left[\left(\omega^2 - \frac{\omega_p^2(q)}{2}\right)^2 + \frac{\omega^2\gamma^2}{4}\right]\left[\left(\omega^2 - \omega_p^2(q)\right)^2 + \omega^2\gamma^2\right]^2} I_{\text{ext}}^2.
\tag{45}
$$

The ratio of intensities (45) and (44) is proportional to the squared ratio of the polarizabilities $\alpha^{(2)}$. For the same parameters that were used in Eq. (26), one gets

$$
\frac{I_{\text{tot}}^{\text{graphene}}}{I_{\text{tot}}^{\text{GaAs}}} \simeq \left(\frac{\alpha_{\text{graphene}}^{(2)}}{\alpha_{\text{GaAs}}^{(2)}}\right)^2 \simeq 100.
\tag{46}
$$

The frequency dependence is the same in both the graphene and the semiconductor cases and is shown in Fig. 1. The intensity-versus-frequency curve has a huge resonance at the frequency $\omega \simeq \omega_p(q)$ and a weaker one at the frequency $\omega \simeq \omega_p(q)/\sqrt{2}$. The chemical potential and temperature dependence of $I_{\text{tot}}^{\text{graphene}}$ is shown in Fig. 2.

If $\omega = \omega_p(q)$ (the main resonance maximum), the ratio $I_{\text{tot}}^{\text{graphene}}/I_{\text{ext}}$ can be presented as (at $|\mu| \gg T$)

$$
\frac{I_{\text{tot}}^{\text{graphene}}}{I_{\text{ext}}} \simeq \frac{9\pi e^6 v_F^4 q^4 I_{\text{ext}}}{2\hbar^4 c \omega_p^8(q)} \times \frac{\omega_p^4(q)}{\gamma^4}
$$
$$
\simeq \left(\frac{3E_0}{16 e k_F^2}\right)^2 \left(\frac{\omega_p(q)}{\gamma}\right)^4.
\tag{47}
$$

In this formula E_0 is the electric field of the external incident electromagnetic wave and $e k_F^2$ is the internal electric field in the 2D system (the field produced by an electron at the average interelectron distance k_F^{-1}). The ratio $E_0/e k_F^2$ in the first set of parentheses is therefore typically very low, $E_0/e k_F^2 \ll 1$. The
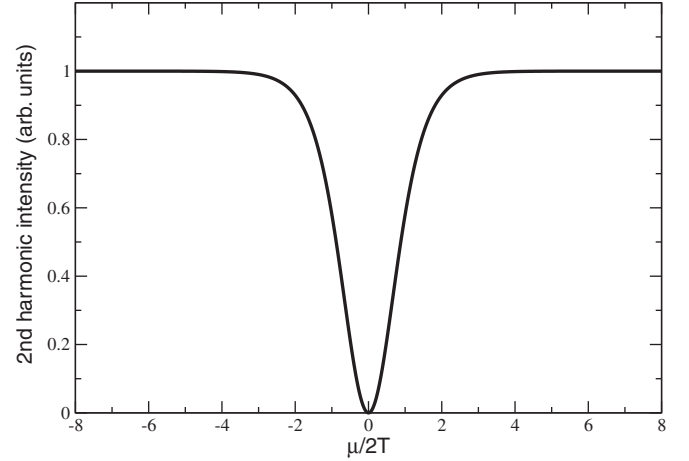

FIG. 2. Intensity of the second harmonic radiation in graphene as a function of the chemical potential and temperature $\mu/2T$.

second factor, $\omega_p(q)/\gamma$, is the quality factor of the 2D plasmon resonance, which can be very large in high-quality samples. This may, at least partly, compensate the smallness of the first factor and substantially facilitate the observation of the second harmonic generation.

IV. SUMMARY

We have presented a self-consistent analytical theory of the second harmonic generation in two-dimensional electron systems. The theory is applicable to semiconductor structures, with a parabolic, and graphene, with a linear, electron energy dispersion. We have shown that the intensity of the second harmonic is about 2 orders of magnitude higher in graphene than in typical semiconductor structures. Under conditions of 2D plasmon resonance the intensity of the second harmonic can be enhanced by several orders of magnitude.

The second harmonic generation in graphene was recently considered in Ref. 37. The theoretical approach admitted in Ref. 37 differs from the one used in this paper. In Ref. 37 scattering of electrons was taken into account (while we assume that $\omega \gg \gamma$) but plasma frequency effects leading to enhancement of the nonlinear response were not considered. Under the conditions $\omega_p \ll \omega$ and $\omega \gg \gamma$ our results coincide with those in Ref. 37, as it should be.

The frequency of 2D plasmons in graphene lies in the terahertz range.[38–40] The phenomena discussed in this paper can be used for creation of novel devices (frequency multipliers, mixers, lasers) operating in this technologically important part of the electromagnetic wave spectrum.

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