Strong enhancement of third-harmonic generation in a double layer graphene system caused by electron-hole pairing

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Abstract – A manifestation of electron-hole pairing in nonlinear electromagnetic response of a double layer graphene system is studied. It is shown that the pairing causes the appearance of a number of peaks in the frequency dependence of the intensity of the third-harmonic generation (THG). The highest peak corresponds to \( \hbar \omega = (2/3)\Delta \), where \( \omega \) is the incident wave frequency, and \( \Delta \) is the order parameter of the electron-hole pairing. The absolute value of the THG intensity in the systems with electron-hole pairing is of several orders of magnitude greater than the THG intensity in the unpaired state. It is shown that huge enhancement of the THG intensity occurs both in the double monolayer and double bilayer graphene systems.

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Introduction. – Nonlinear optical and microwave properties of graphene attract considerable attention. A strong nonlinear response of graphene can be seen from the classical equation of motion for a charged particle with the linear-in-momentum spectrum [1,2]. This equation yields the electric current that contains all odd Fourier harmonics with the amplitudes decreasing very slowly with the harmonic number. The quantum approach [3–7] predicts a resonant behavior for the third-harmonic generation in graphene. The frequency dependence of the THG intensity has the main peak at \( \hbar \omega = (2/3)\varepsilon_F \) and two minor peaks at \( \hbar \omega = \varepsilon_F \) and \( \hbar \omega = 2\varepsilon_F \), where \( \varepsilon_F \) is the Fermi energy. Nonlinear optical effects in graphene systems were observed experimentally. In [8] the coherent nonlinear optical response of single- and few-layer graphene was measured using four-wave mixing. Sharp-contrast images of graphene flakes on a dielectric substrate at combined frequencies were observed. Graphene with effective thickness 3 Å demonstrates that the nonlinear emission intensity is 10 times larger than that of a 4 nm gold film. Nonlinear susceptibility at the wavelength \( \lambda \approx 1 \mu m \) is evaluated as large as \( 10^{-7} \) esu.

In the THG experiment [9] the effective nonlinear susceptibility \( \chi^{(3)} \sim 5 \cdot 10^{-9} \) esu at the incident wavelength \( \lambda_i = 1.7 \mu m \) was registered. Strong THG in a monolayer graphene on an amorphous silica substrate was reported in [10]. It was shown that the effective nonlinear susceptibility of graphene is 4.6 times larger than that of a thick Au film (for \( \lambda_i = 789 \) nm).

It is known that the linear optical properties of graphene are also quite unusual. In a wide frequency range the absorption coefficient \( A \) of pristine graphene is determined by the fundamental constants: \( A = \pi e^2/\hbar c \) [11]. In a doped graphene the absorption is suppressed in the frequency range \( \hbar \omega < 2\varepsilon_F \) due to the reduction of the interband transitions.

Graphene double layer systems have good perspectives for achieving the electron-hole pairing. The electron-hole pairing is an analog of the Cooper pairing. It may reveal itself in the so-called counterflow superconductivity. The effect can also be understood as the superfluidity of a gas of electron-hole pairs. The possibility of electron-hole pairing in a double monolayer graphene (DMLG) system was considered in [12–15]. The pairing in DMLG in the quantum Hall regime was analyzed in [16–18]. In [19] we have considered electromagnetic properties of DMLG in the paired state and found a resonant behavior of the absorption and reflection coefficients at the photon energy equal to the excitonic energy gap in the spectrum \( \hbar \omega = 2\Delta \).

The general idea of the electron-hole pairing in double layer systems was put forward in [20,21] well before the proposals of [12–15]. The electron-hole pairing in quantum
Hall systems was predicted in [22–24]. The pairing was registered in double quantum well AlGaAs heterostructures in the quantum Hall state with the study of their transport properties [25–28]. The possibility of electron-hole pairing was also considered with reference to topological insulator heterostructures [29–32], double bilayer graphene [33], double few-layer graphene [34], transition metal dichalcogenide [35–37] and black phosphorene [38] double layers. Recently several experimental efforts to register electron-hole pairing in double layer graphene systems were made [39–42], but the results of these experiments are controversial.

The electron-hole pairing is caused by the Coulomb attraction. The bare Coulomb interaction is a strong one, but the results of these experiments are controversial.

where $j^{(3)}_{\pm} = j^{(3)}_1 \pm j^{(3)}_2$, $E_{\pm} = E_1 \pm E_2$, $j_i$ is the electric current in the layer $i$ and $E_i$ is the electric field in this layer. Equations (1) are presented in the symbolic form. In the general case they are matrix integral equations that account for the tensor nature of the conductivity and nonlocality of the response in time and space. In the case of uncoupled layers (unpaired electrons and holes) the conductivities in (1) are expressed through $\sigma^{(3)}_{\pm \pm}$, the 3rd-order single-layer conductivity: $\sigma^{(3)}_{+++} = \sigma^{(3)}_{--} = \sigma^{(3)}_{1}/4$, $\sigma^{(3)}_{++-} = \sigma^{(3)}_{-+-} = 3\sigma^{(3)}_{1}/4$.

In this study we put $E_- = 0$ due to the following reason. The ratio of the amplitudes of the fields $E_-$ and $E_+$ is evaluated as $|E_-|/|E_+|$ ≈ $q_z d/2$, where $q_z$ is the normal component of the wave vector of the incident wave, and $d$ is the interlayer distance. We consider the frequency range $\hbar \omega \lesssim \varepsilon_F$. In this range $|E_-|/|E_+| \lesssim k_F d \varepsilon_F/\varepsilon_F$, where $c$ is the velocity of light. The electron-hole pairing occurs at rather small interlayer distance $d$. In particular, for the double monolayer graphene the condition $k_F d \lesssim 0.1$ should be fulfilled [45]. Therefore, $|E_-|/|E_+| < 10^{-3}$ and the nonlinear response to the small field $E_-$ can be neglected.

Let the electric component of the incident wave be $E_x(z, t) = E_0 \cos(q_z z - \omega t)$. Then, the 3rd harmonic of the electric current is given by the expression

$$j^{(3\omega)}_x(t) = \sigma^{(3)}_{+++}(\omega, \omega, \omega) E_0^3 e^{-3i\omega t} + c.c.,$$

where $\sigma^{(3)}_{+++}(\omega, \omega, \omega)$ is the $xxx$ component of the high-frequency nonlinear conductivity tensor.

The boundary conditions determine the relation between the current (2) and the magnetic component of the generated wave. It yields

$$B_y^{(3)}(z, t) = \mp \frac{2\pi}{c} \frac{\sigma^{(3)}_{+++}(\omega, \omega, \omega) E_0^3 e^{\pm 3i\omega z - 3i\omega t} + c.c.},$$

where sign ± corresponds to different half-spaces. The boundary condition (3) assumes that the double layer system is considered as a zero-thickness conducting layer. It is equivalent to the limit $q_z d \rightarrow 0$.

The intensity of the third harmonic is given by the Poynting vector averaged over the period $T$:

$$I^{(3)} = \frac{c}{4\pi T} \int_0^T |\mathbf{E}^{(3)} \times \mathbf{B}^{(3)}| dt.$$  

Assuming, for simplicity, that the dielectric constant of the environment is $\varepsilon = 1$, we obtain

$$I^{(3)} = 4 \left( \frac{2\pi}{c} \right)^4 |4\sigma^{(3)}_{+++}(\omega, \omega, \omega)|^2 I_{inc}^3,$$

where $I_{inc}$ is the intensity of the incident wave. For two uncoupled layers ($\sigma^{(3)}_{+++} = \sigma^{(3)}_{1}/4$) eq. (5) corresponds to the quadruplicate intensity of a single layer. The factor 4 is due to the constructive interference.
Electron-hole pairing in the double layer graphene. In what follows we use the Dirac Hamiltonian that describes the electromagnetic properties of graphene in the low-energy approximation. In this approximation two valleys near Dirac points $K$ and $K'$ are considered independently and each valley has two spin components. Each Dirac component yields the same contribution into the nonlinear conductivity. Below we consider only one component and take into account the other ones by the factor 4 in the final answer.

The Hamiltonian of the DMLG system has the form

\begin{equation}
H = \sum_{i,k,\lambda} \xi_{k\lambda} c_{i,k,\lambda}^{\dagger} c_{i,k,\lambda} + \frac{1}{2S} \sum_{i,j,q} V_{ij}(q) :n_{i,q} n_{j,-q} + \frac{1}{2} \sum_{i,q} e \varphi_{i,q}(t) \hat{n}_{i,q},
\end{equation}

where $c_{i,k,\lambda}^{\dagger}$ ($c_{i,k,\lambda}$) is the creation (annihilation) operator for the electron in the layer $i$ in the state with the momentum $k$ and the subband index $\lambda = \pm 1$. This state corresponds to the energy $\xi_{k\lambda} = \varepsilon_{k\lambda} - \mu$, where $\varepsilon_{k\lambda} = \hbar v_F k$ is the electron spectrum of the monolayer graphene near the $K$ and $K'$ points, and $\mu$ is the chemical potential in the layer $i$. In what follows we imply $\mu_1 = -\mu_2 = \mu$ and neglect the difference between $\mu$ and $\varepsilon_F$. In the Hamiltonian (6) $\hat{n}_{i,q}$ is the electron density operator, $V_{ij}(q)$ is the Fourier component of the Coulomb interaction energy between electrons in the layers $i$ and $j$, $\varphi_{i,q}(t)$ is the Fourier component of the scalar potential of the external electromagnetic field, the notation $: \hat{O} :$ indicates the normal ordering of creation and annihilation operators, and $S$ is the area of the system. The explicit expression for the electron density operator reads

\begin{equation}
\hat{n}_{i,q} = \sum_{k,\lambda,\lambda'} g_{k+q,\lambda',k,\lambda} c_{i,k,\lambda}^{\dagger} c_{i,k,\lambda'},
\end{equation}

where

\begin{equation}
g_{k_1,\lambda_1;k_2,\lambda_2} = \frac{e^{\frac{i}{\hbar} (\hat{\theta}_{k_1} - \hat{\theta}_{k_2})} + \lambda_1 \lambda_2 e^{-\frac{i}{\hbar} (\hat{\theta}_{k_1} - \hat{\theta}_{k_2})}}{2}
\end{equation}

and $\hat{\theta}_{k}$ is the angle between $k$ and the $x$-axis.

In (6) we use the gauge in which the vector potential is directed normally to the layers and the in-plane electric field is given by the scalar potential $E_x = -\partial \varphi / \partial x$. Such a gauge can be used if the $x$ component ($q_x$) of the wave vector of the incident wave is nonzero. The response at normal incidence can be computed as the limit $q_x \to 0$.

The number of holes for the electron-hole pairing is given by the equation

\begin{equation}
\Delta_{k\lambda} = \frac{1}{S} \sum_{q,\lambda'} V_{12}(q) \left( 1 + \lambda' \cos(\hat{\theta}_{k+q} - \hat{\theta}_{k}) \right)
\times (\xi_{k+q,\lambda'}^{\dagger} \xi_{k+q,\lambda} - \varepsilon_{k,\lambda}^{\dagger} \varepsilon_{k,\lambda}),
\end{equation}

where the number of electrons in one layer and the number of holes in the other layer are indefinite, and, in this sense, it can be considered as the anomalous average.

The mean-field Hamiltonian has the form

\begin{equation}
H_{MF}(t) = H_0 + H_{int}(t),
\end{equation}

where

\begin{equation}
H_0 = \sum_{k,\lambda} \left[ \xi_{k\lambda} \left( c_{1,k,\lambda}^{\dagger} c_{1,k,\lambda} - c_{2,k,\lambda}^{\dagger} c_{2,k,\lambda} \right) - \left( \Delta_{k\lambda} c_{2,k,\lambda}^{\dagger} c_{1,k,\lambda} + H.c. \right) \right],
\end{equation}

\begin{equation}
H_{int}(t) = \frac{1}{2S} \sum_{q} e \varphi_{-q}(t) \hat{n}_{+,-q},
\end{equation}

$\xi_{k\lambda} = \varepsilon_{k\lambda} - \mu$, $\varphi_+ = \varphi_1 + \varphi_2$, and $\hat{n}_{+} = \hat{n}_1 + \hat{n}_2$. Due to the same reason as for $E_-$ we put $\varphi_- = \varphi_1 - \varphi_2 = 0$ in eq. (12).

It is known that in conventional superconductors the order parameter fluctuations influence the linear and nonlinear response to the external electromagnetic field [48,49]. Nevertheless, in (12) we do not account for such fluctuations. The reason is the following. In the case of the electron-hole pairing the order parameter fluctuations are coupled with the field $\varphi_-$. The field $\varphi_-$ induces the fluctuations of the anomalous average in (9) that in turn induces the variation of the difference of electron densities in the layers $n_-$. This results in a renormalization of the linear and nonlinear response to the field $\varphi_-$. On the contrary, the field $\varphi_+$ is decoupled from the amplitude and phase fluctuations of the order parameter, and the response to the field $\varphi_+$ is not modified when accounting for the order parameter fluctuations.

To proceed further we apply the $u \sim v$ transformation that diagonalizes the Hamiltonian $H_0$. The transformation reads

\begin{equation}
c_{1,k,\lambda} = u_{k\lambda} a_{\alpha,k,\lambda} + v_{k\lambda} a_{\beta,k,\lambda},
\end{equation}

\begin{equation}
c_{2,k,\lambda} = u_{k\lambda} a_{\beta,k,\lambda} - v_{k\lambda} a_{\alpha,k,\lambda},
\end{equation}

where $a_{\alpha(\beta),k,\lambda}$ are new second quantization operators that satisfy Fermi anticommutation relations. The coefficients in (13) are expressed as

\begin{equation}
u_{k\lambda} = \sqrt{\frac{1}{2} \left( 1 + \frac{\xi_{k\lambda}}{E_{k\lambda}} \right)},
\end{equation}

\begin{equation}u_{k\lambda} = \sqrt{\frac{1}{2} \left( 1 - \frac{\xi_{k\lambda}}{E_{k\lambda}} \right)},
\end{equation}

where $E_{k\lambda} = \sqrt{\varepsilon_{k\lambda}^2 + \Delta_{k\lambda}^2}$ are the eigenenergies of the Hamiltonian $H_0$.

The transformation (13) reduces (11) and (12) to the form

\begin{equation}
H_0 = \sum_{\nu} E_{\nu} a_{\nu}^{\dagger} a_{\nu},
\end{equation}

\begin{equation}
H_{int}(t) = \frac{1}{2S} \sum_{q} e \varphi_{-q}(t) \delta_{k_2,k_1} \delta_{\nu_1,\nu_2} a_{\nu_1}^{\dagger} a_{\nu_2}.
\end{equation}
The terms in the series (21) satisfy the recurrent equation

$$R_{\alpha,\kappa,\lambda,\beta,\kappa_1,\lambda_1,\beta_1,\kappa_2,\lambda_2} = g_{\kappa_1,\lambda_1,\beta_1,\kappa_2,\lambda_2}(u_{\kappa_1,\lambda_1,\beta_1} + u_{\kappa_2,\lambda_2}),$$

$$R_{0,\alpha,\kappa,\lambda,\beta,\kappa_1,\lambda_1,\beta_1,\kappa_2,\lambda_2} = g_{\kappa_1,\lambda_1,\beta_1,\kappa_2,\lambda_2}(u_{\kappa_1,\lambda_1,\beta_1} - u_{\kappa_2,\lambda_2}).$$

The DBLG system is treated analogously. The monolayer graphene spectrum is replaced with the bilayer one:

$$\varepsilon_{k\lambda}^{(b)} = \hbar^2k^2/2m_{\text{eff}},$$

and the factor $g$ is modified as

$$g_{\kappa_1,\lambda_1,\beta_1,\kappa_2,\lambda_2} = \frac{\exp(i\varepsilon_{\kappa_1} - \varepsilon_{\kappa_2}) + \lambda_1\lambda_2}{2}.$$

Nonlinear conductivity.—To compute nonlinear conductivity we use the density matrix approach [50]. The density matrix satisfies the Liouville-von Neumann equation

$$\frac{\partial \rho(t)}{\partial t} = \frac{1}{i\hbar}[H_{MF}(t), \rho(t)] - \gamma \left( \rho(t) - \rho(0) \right),$$

where $\rho(0)$ is the equilibrium density matrix, and $\gamma$ is the phenomenological relaxation rate. The equilibrium density matrix is diagonal in the basis of eigenfunctions of the Hamiltonian (15),

$$\rho(0)_{\nu,\nu'} = \delta_{\nu,\nu'} f_\nu,$$

where $f_\nu = [\exp(E_\nu/T) + 1]^{-1}$ is the Fermi-Dirac distribution function. The interaction with the external field is considered as a small perturbation and the density matrix is sought as the series

$$\rho = \rho(0) + \rho(1) + \rho(2) + \rho(3) + \ldots.$$

The terms in the series (21) satisfy the recurrent equation

$$\rho^{(i)}(t)_{\nu,\nu'} = \frac{1}{i\hbar} \int_{-\infty}^{t} dt' \left[ \left( \hat{H}_{\text{int}}(t'), \rho^{(i-1)}(t') \right)_{\nu,\nu'} \times e^{i\omega_{\nu,\nu'}(t'-t)} \right],$$

where $\omega_{\nu,\nu'} = (E_\nu - E_{\nu'})/\hbar$.

Let the scalar potential in the graphene layers be equal to $\varphi(r, t) = \varphi_0 \sin(q_x x - \omega t)$. It corresponds to the electric field of the incident wave $E_0 = \cos(q r - \omega t)$, where $E_0 = (E_0x, 0, E_0z)$, $q = (qx, 0, qz)$, and $E_0z = -qz \varphi_0$. The nonlinear in $\varphi_0$ part of electron density oscillations is given by the equation

$$n_+^{(3)}(r, t) = \text{Tr}[\rho^{(3)}(t) \hat{n}_+(r)].$$

The 3rd-harmonic term in the electron density oscillations reads

$$n_+^{(3)}(r, t) = n_0^{(3)} e^{i(3(q_x x - \omega t) + \text{c.c.})}.$$

where the explicit expression for $n_0^{(3)}$ can be obtained from eq. (23). Using the continuity equation $\partial n_+ / \partial t + \nabla j_+ = 0$ one finds

$$j_+^{(3)}(r, t) = \frac{\omega n_0^{(3)} e^{i(3(q_x x - \omega t))}}{q_x} + \text{c.c.}.$$

Equations (23)–(25) yield the following expression for the current:

$$j_+^{(3)}(r, t) = e^{i(3(q_x x - \omega t))} \sigma_+^{(3)}(q_x, q_x; \omega, \omega, \omega) E_0^3 + \text{c.c.},$$

where

$$\sigma_+^{(3)}(q_x, q_x; \omega, \omega, \omega) = -i \frac{e^4 \omega}{2 S q_x^3}$$

$$\times \sum_{j_1 j_2 j_3} \delta_{k_1 k_2 + q_x} \delta_{k_1 k_3 + q_x} \delta_{j_1 k_2 + j_2} \delta_{j_1 k_3 + j_3} R_{12} R_{23} R_{34} R_{41}$$

$$\times \left[ 1 - \frac{f_1 f_2 f_3}{(E_1 - E_3 - 2\hbar \omega - i\hbar \gamma)} - \frac{f_1 f_2 - f_3}{(E_1 - E_2 - 2\hbar \omega - i\hbar \gamma)} - \frac{f_1 f_3 - f_2}{(E_1 - E_3 - 2\hbar \omega - i\hbar \gamma)} - \frac{f_2 f_3 - f_1}{(E_2 - E_3 - 2\hbar \omega - i\hbar \gamma)} \right].$$

Results and discussion.—It was shown in [13] that in a DMLG system the order parameter $\Delta_{k,\lambda}$ is peaked at the Fermi surface and decreases far from this surface. In the DBLG system a regime with almost constant $\Delta_{k,\lambda}$ at $k \ll 4k_F$ was found in [33]. In our computations of the THG intensity we neglect the wave vector dependence of the order parameter and replace $\Delta_{k,\lambda}$ with $\Delta = E_g/2$, where $E_g$ is the energy gap. Such an approximation works well near the Fermi surface. The resonant features in the THG intensity described below are caused, in the main part, by the transitions between the electron states near the Fermi surface. This justifies the use of the approximation $\Delta_{k,\lambda} = \Delta$.

We fix the temperature and the relaxation rate as $T = 0.1 \mu$ and $\hbar \gamma = 0.001 \mu$. The ratio of the THG intensity to the incident wave intensity vs. the incident wave frequency is shown in figs. 1 and 2. Figure 1 corresponds to the DMLG system and fig. 2 to the DBLG system. The dependencies are presented for the paired state for two different values of the order parameter and for the unpaired state ($\Delta = 0$). The absolute value of the ratio $I^{(3)}/I_{inc}$ in figs. 1 and 2 is computed for $\mu = 0.01$ eV and

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Fig. 1: (Colour online) THG intensity for the double monolayer graphene in the state with the electron-hole pairing (Δ = 0.5μ, 0.2μ) and in the unpaired state (Δ = 0).

Fig. 2: (Colour online) The same as in fig. 1 for the double bilayer graphene.

\[ I_{\text{inc}} = 5 \text{ W/cm}^2. \] The dependencies presented are scaled as

\[ \frac{I^{(3)}}{I_{\text{inc}}} \propto \frac{I^2_{\text{inc}}}{\mu^8} \]

for the DMLG system, and

\[ \frac{I^{(3)}}{I_{\text{inc}}} \propto \frac{I^2_{\text{inc}}}{\mu^5} \]

for the DBLG system.

One can see that the dependencies obtained have a number of peaks. There are three peaks that correspond to the incident photon energies \( h\omega = (2/3)\Delta, h\omega = \Delta, \) and \( h\omega = 2\Delta. \) Numerically the peaks appear because the denominators in the expression for the nonlinear conductivity (27) become resonant when the incident photon energy \( h\omega \) exceeds \( E_g/3, E_g/2 \) and \( E_g. \) Physically, this means the opening of new channels of nonlinear absorption. In addition, a double peak emerges at \( \omega = (2/3)\sqrt{\mu^2 + \Delta^2}. \)

The peaks in the THG intensity are quite sharp. We connect their sharpness with the divergence of the electron density of states in the paired state. Indeed, in the normal state the sum of the density of states in the electron and the hole layers is the constant \( n = n_e + n_h = 2n_F \) in the interval \(-\mu \leq \epsilon \leq \mu, \) where \( n_F \) is the density of states at the Fermi level for an isolated monolayer (bilayer) graphene, and the energy \( \epsilon \) is counted from the Fermi level. For the paired state simple calculations yield \( n(\epsilon) = 2n_F/\sqrt{\epsilon^2 - \Delta^2} \) (in this case the density of states cannot be divided into the electron and the hole parts). This function diverges at \( \epsilon = \pm \Delta. \) The influence of pairing on the spectrum and on the density of states in a DMLG system is illustrated in fig. 3. Note that far from the Fermi level the energy spectra for the paired and the normal states approach each other. This means that the wave vector dependence of \( \Delta_{k_A} \) yields only an inessential correction of the spectrum that can be considered as another justification of the approximation \( \Delta_{k_F, A} = \Delta. \)

The dependencies that correspond to \( \Delta = 0 \) do not demonstrate any peaks. The peaks predicted in \( [3,4,7] \) emerge at lower temperature \( (T \lesssim 10^{-2} \mu). \) Under transition to the normal state the double peak at \( h\omega \approx (2/3)\sqrt{\mu^2 + \Delta^2} \) is transformed to the peak at \( h\omega = (2/3)\mu, \) and the peaks at \( h\omega = (2/3)\Delta, h\omega = \Delta, \) and \( h\omega = 2\Delta \) disappear.

One can see that the electron-hole pairing causes a huge increase of the intensity of THG in a certain frequency range. At the main peak \( h\omega = (2/3)\Delta \) the THG intensity is in 8 orders of magnitude greater than the one for the unpaired state. For the out-of-resonance frequencies the factor of THG enhancement is also very large (about \( 10^4) \). We note that the enhancement of THG is not observed at large frequencies \( (h\omega \gg \Delta) \) and at small ones \( (h\omega \ll \Delta). \) In the latter case the pairing even suppresses the THG. This can be understood from the classical picture of a
tightly bound electron-hole pair that does not respond to a static electric field if the same field is applied to the electron and the hole component of the pair.

Thus, we consider that the enhancement of the THG intensity is physically caused by the appearance of new resonant frequencies connected with the gap $E_g = 2\Delta$, and by the divergence of the density of states near the gap.

From the practical point of view the enhancement means that a strong nonlinear response in the double layer graphene with electron-hole pairing can be observed at much smaller incident wave intensity than in the system where the pairing does not occur.

In conclusion, we have shown that the electron-hole pairing in the double layer graphene system results in the strong enhancement of the nonlinear response to the electromagnetic radiation. We predict the appearance of a number of peaks in the THG intensity. The main peak corresponds to the frequency equal to one-third of the energy gap in the spectrum and the intensity of this peak exceeds by many orders the THG intensity at the same frequency in the unpaired state. The impact of the pairing is basically the same for the double monolayer and double bilayer graphene systems and we expect THG enhancement in other systems in which the electron-hole pairing may occur.

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REFERENCES

[1] Mikhailov S. A., EPL, 79 (2007) 27002.
[2] Mikhailov S. A. and Ziegler K., J. Phys.: Condens. Matter, 20 (2008) 384204.
[3] Mikhailov S. A., Phys. Rev. B, 90 (2014) 241301(R).
[4] Mikhailov S. A., Phys. Rev. B, 93 (2016) 085403.
[5] Cheng J. L., Vermeulen N. and Sipe J. E., New J. Phys., 16 (2014) 053014.
[6] Cheng J. L., Vermeulen N. and Sipe J. E., Phys. Rev. B, 91 (2015) 235320.
[7] Rostami H. and Polini M., Phys. Rev. B, 93 (2016) 161411(R).
[8] Hendry E., Hale P. J., Moger J., Savchenko A. K. and Mikhailov S. A., Phys. Rev. Lett., 105 (2010) 097401.
[9] Kumar N., Kumar J., Gerstenkorn C., Wang R., Chiu H.-Y., Shuml A. L. and Zhao H., Phys. Rev. B, 87 (2013) 121406(R).
[10] Hong S.-Y., Dadap J. I., Petrone N., Yeh P.-C., Hone J. and Osgood R. M. Jr., Phys. Rev. X, 3 (2013) 021014.
[11] Nair R. R., Blake P., Grigorenko A. N., Novoselov K. S., Booth T. J., Stauber T., Peres N. M. R. and Geim A. K., Science, 320 (2008) 1308.
[12] Lozovik Yu. E. and Sokolik A. A., Pisma Zh. Eksp. Teor. Fiz., 87 (2008) 61; JETP Lett., 87 (2008) 55.
[13] Zhang C.-H. and Joglekar Y. N., Phys. Rev. B, 77 (2008) 233405.
[14] Min H., Bistritzer R., Su J.-j. and MacDonald A. H., Phys. Rev. B, 78 (2008) 121401(R).
[15] Seradjeh B., Weber H. and Franz M., Phys. Rev. Lett., 101 (2008) 246404.
[16] Berman O. L., Lozovik Y. E. and Gumbs G., Phys. Rev. B, 77 (2008) 155433.
[17] Fil D. V. and Kravchenko L. Yu., Fiz. Nizk. Temp., 35 (2009) 904; Low Temp. Phys., 35 (2009) 712.
[18] Pikalov A. A. and Fil D. V., Nanoscale Res. Lett., 7 (2012) 145.
[19] Germash K. V. and Fil D. V., Phys. Rev. B, 93 (2016) 205436.
[20] Shevchenko S. I., Fiz. Nizk. Temp., 2 (1976) 505; Sov. J. Low Temp. Phys., 2 (1976) 251.
[21] Lozovik Yu. E. and Yudson V. I., Zh. Eksp. Teor. Fiz., 71 (1976) 738; Sov. Phys. JETP, 44 (1976) 389.
[22] Fertig H. A., Phys. Rev. B, 40 (1989) 1087.
[23] Yoshioka D. and MacDonald A. H., J. Phys. Soc. Jpn., 59 (1990) 4211.
[24] Moon K., Mori H., Yang K., Givrin S. M., MacDonald A. H., Zheng L., Yoshioka D. and Zhang S. C., Phys. Rev. B, 51 (1995) 5138.
[25] Kellogg M., Eisenstein J. P., Pfeiffer L. N. and West K. W., Phys. Rev. Lett., 93 (2004) 036801.
[26] Wiersma D. R., Lok J. G. S., Dietscsh W., von Klitzing K., Schuh D., Bichler M., Tranitz H.-P. and Wegscheider W., Phys. Rev. Lett., 93 (2004) 266805.
[27] Tuttuc E., Shayegan M. and Huse D. A., Phys. Rev. Lett., 93 (2004) 036802.
[28] Nandi D., Finck A. D. K., Eisenstein J. P., Pfeiffer L. N. and West K. W., Nature, 488 (2012) 481.
[29] Seradjeh B., Moore J. E. and Franz M., Phys. Rev. Lett., 103 (2009) 066402.
[30] Cho G. Y. and Moore J. E., Phys. Rev. B, 84 (2011) 165101.
[31] Efimkin D. K., Lozovik Yu. E. and Sokolik A. A., Phys. Rev. B, 86 (2012) 115436.
[32] Germash K. V. and Fil D. V., Phys. Rev. B, 87 (2013) 115313.
[33] Perali A., Neilson D. and Hamilton A. R., Phys. Rev. Lett., 110 (2013) 146803.
[34] Zarenia M., Perali A., Neilson D. and Peeters F. M., Sci. Rep., 4 (2014) 7319.
[35] Fogler M. M., Butov L. V. and Novoselov K. S., Nat. Commun., 5 (2014) 4555.
[36] Wu F.-C., Xue F. and MacDonald A. H., Phys. Rev. B, 92 (2015) 161211.
[37] Berman O. L. and Kezerashvili R. Ya., Phys. Rev. B, 93 (2016) 245410.
[38] Berman O. L., Gumbs G. and Kezerashvili R. Ya., Phys. Rev. B, 96 (2017) 014505.
[39] Gorbachyev R. V., Geim A. K., Katsnelson M. I., Novoselov K. S., Tudorovskiy T., Grigorieva I. V., MacDonald A. H., Morozov S. V., Watanabe K., Taniguchi T. and Ponomarenko L. A., Nat. Phys., 8 (2012) 896.
[40] Gamucci A., Spirito D., Carrega M., Karmakar B., Lombardo A., Bruna M., Pfeiffer L. N., West K.
Enhancement of third-harmonic generation caused by electron-hole pairing

W., Ferrari A. C., Polini M. and Pellegrini V., Nat. Commun., 5 (2014) 5824.

[41] Li J. I. A., Taniguchi T., Watanabe K., Hone J., Levchenko A. and Dean C. R., Phys. Rev. Lett., 117 (2016) 046802.

[42] Lee K., Xue J., Dillen D. C., Watanabe K., Taniguchi T. and Tutuc E., Phys. Rev. Lett., 117 (2016) 046803.

[43] Kharitonov M. Y. and Efetov K. B., Phys. Rev. B, 78 (2008) 241401(R).

[44] Germash K. V. and Fil D. V., Phys. Rev. B, 91 (2015) 115442.

[45] Sodemann I., Pesin D. A. and MacDonald A. H., Phys. Rev. B, 85 (2012) 195136.

[46] Lozovik Yu. E., Ogarkov S. L. and Sokolik A. A., Phys. Rev. B, 86 (2012) 045429.

[47] Volkson W., Miller R. D. and Dubois G., Chem. Rev., 110 (2010) 56.

[48] Kulik L. O., Entin-Wohlman O. and Orbach R., J. Low Temp. Phys., 43 (1981) 591.

[49] Cea T., Castellani C. and Benzatto L., Phys. Rev. B, 93 (2016) 180507(R).

[50] Boyd R. W., Nonlinear Optics, third edition (Academic Press) 2008.