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EXCITONIC ABSORPTION IN GAPPED GRAPHENE SYSTEMS

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Excitonic absorption in monolayer and bilayer graphene systems with opened energy gap in the field of laser radiation is investigated. The obtained value of excitonic binding energy in monolayer is in good agreement with the exact analytical solution. It is shown that the account of all tight binding parameters in bilayer graphene leads to an increase of exciton binding energy.

***Keywords*:** monolayer graphene, graphene bilayer, laser radiation, exciton.

Introduction. Graphene is a 2D semimetal with interesting physics associated with its unusual electronic structure and promising device applications [1,2]. Optical properties of monolayer graphene display many intriguing features such as a near constant optical conductivity and gate-dependent optical absorbance [3]. Theoretical and experimental investigations have shown that a perpendicular electric field applied to graphene bilayer may open an energy gap in the electronic spectrum, which is tunable by the gate voltage [4]. The dependence of the gap on the stacking order of consecutive graphene planes is studied in [5,6]. The interest in optical response of bilayer graphene is boosted by recent progress of terahertz radiation technology, which is another frontier research area. In order to understand the influence of Coulomb interaction and impact of many body physics in graphene systems it is essential to study electronic and optical properties of graphene systems with opened energy gap. In gapless monolayer graphene the Coulomb problem has no true bound states, but resonances [7]. The optical response of graphene with an opened energy gap between the conduction and valence bands is dominated by bound excitons [8,9]. In the work excitonic absorption in monolayer and bilayer graphene systems with opened energy gap in the field of laser radiation has been investigated.

Theory. Here interaction of electromagnetic wave with graphene systems with opened energy gap is considered. It is assumed that the laser pulse propagates in the perpendicular to graphene plane (XY), direction and the electric field of pulse lies in the graphene plane. The Hamiltonian for graphene in the second quantization formalism in the presence of electron–electron interaction is

$$H = \int \Psi^{+}(\mathbf{r},t) H_s \Psi(\mathbf{r},t) d\mathbf{r} + H_{Coul}, \tag{1}$$

where $\Psi(\mathbf{r},t)$ are the fermionic field operators, and H_{Coul} is the two particle Coulomb Hamiltonian [10]. For a graphene bilayer, the single particle Hamiltonian $H_s = H_0 + H_d$ in the

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vicinity of the **K** point is

$$H_0 = \begin{pmatrix} -\dfrac{U}{2} & v_3(p_x+ip_y) - \dfrac{(p_x+ip_y)^2}{2m} \\ v_3(p_x-ip_y) - \dfrac{(p_x+ip_y)^2}{2m} & \dfrac{U}{2} \end{pmatrix}, \qquad (2)$$

$$H_d = \begin{pmatrix} e\mathbf{r}\mathbf{E}(t) & 0 \\ 0 & e\mathbf{r}\mathbf{E}(t) \end{pmatrix}.$$

In Eqs. (2) H_0 corresponds to bilayer graphene in the field of perpendicular electric field, and H_d is the interaction Hamiltonian between a laser field $\mathbf{E}(t)$ and bilayer; v_3 is the effective velocity: $v_3 = \sqrt{3}\gamma_3 a/2\hbar$ (γ_3 describes the interaction between B atoms in the neighboring layers, a is the lattice constant); U is the gap introduced by the electric field.

We expand the fermionic field operator $\Psi(\mathbf{r}, t)$ over the free wave function $\psi_\sigma(\mathbf{p})$ of bilayer

$$\Psi(\mathbf{r},t) = \frac{1}{\sqrt{S}} \sum_{\mathbf{p},\sigma} \hat{a}_{\mathbf{p},\sigma}(t)\psi_\sigma(\mathbf{p}) \exp\left(\frac{i}{\hbar}\mathbf{p}\mathbf{r}\right), \qquad (3)$$

where annihilation operator $\hat{a}_{\mathbf{p},\sigma}(t)$ is associated with positive and negative energy solutions $\sigma = \pm 1$, S is the area of graphene layer. Denoting $\theta(\mathbf{p}) = \arctan(p_y/p_x)$ the energy spectrum of bilayer can be presented in the form

$$\varepsilon_{\mathbf{p},\sigma} = \sigma\left(U^2/4 + (v_3 p)^2 - (v_3 p^3/m)\cos 3\theta + \left(p^2/2m\right)^2\right)^{1/2}. \qquad (4)$$

The free solutions in bilayer graphene $\psi_\sigma(\mathbf{p})$ have the following form:

$$\psi_\sigma(\mathbf{p}) = \left(\frac{\varepsilon_{\mathbf{p},\sigma} + U/2}{2\varepsilon_{\mathbf{p},\sigma}}\right)^{1/2} \begin{pmatrix} 1 \\ \dfrac{1}{\varepsilon_{\mathbf{p},\sigma} + U/2}\gamma(p,\theta) \end{pmatrix}, \qquad (5)$$

where $\gamma(p,\theta) = -\dfrac{p^2}{2m}\exp(i2\theta) + v_3 p\exp(-i\theta)$. Using in Eq. (1) the expansion of the fermionic field operator over the annihilation and creation operators (see Eq. (3)) the expression for total Coulomb Hamiltonian, that consists of the 16 terms, can be obtained.

In order to find time dependent equations for the interband polarization $P_{\mathbf{k}}(t) = \langle \hat{v}^+_{\mathbf{k}} \hat{c}_{\mathbf{k}} \rangle$ one can use Heisenberg picture for an operator evolution (the operator $\hat{a}^+_{\mathbf{k},\sigma}$ in the conduction band is defined as $\hat{c}^+_{\mathbf{k}}(t)$, and the operator $\hat{a}_{\mathbf{k},\sigma}$ in the valence band as $\hat{v}_{\mathbf{k}}(t)$). Taking into account only excitonic effects, we apply the Hartree–Fock approximation to the many particle system, i.e. four field operator averages in Coulomb Hamiltonian are expressed as products of polarization and population.

In order to obtain the Coulomb Hamiltonian for gapped graphene monolayer, the same procedure described above for graphene bilayer is used. Experimentally an energy gap U can be obtained by placing graphene onto a substrate, in which the A and B atoms experience different on-site energies [11]. For the dipole matrix element

$$\mathbf{d}_{\sigma\sigma'}(\mathbf{p},\mathbf{p}') = ie\psi^+_\sigma(\mathbf{p})\psi_\sigma(\mathbf{p}') \int \frac{\mathbf{r}}{S} \exp\left(\frac{i}{\hbar}(\mathbf{p}'-\mathbf{p})\mathbf{r}\right) d\mathbf{r},$$

we obtained an expression, that is not presented here due to its long form. Note, that in contrast to the non-relativistic case, the dipole matrix element $\mathbf{d}_{\sigma\sigma'}(\mathbf{p},\mathbf{p}')$ in the bilayer depends on electron momentum.

Using Eqs. (1)–(5), one can obtain the evolution equation for the single-particle density matrix $\rho_{\sigma\sigma'}(\mathbf{p},\mathbf{p}') = \langle \hat{a}^{+}_{\mathbf{p},\sigma}\hat{a}_{\mathbf{p}',\sigma}\rangle$ in the external laser field for different values of $k_x = p_x/\hbar$, $k_y = p_y/\hbar$:

$$\begin{aligned} i\hbar\frac{\partial\rho_{1,-1}(\mathbf{k},t)}{\partial t} &= \Sigma(\mathbf{k})\rho_{1,-1}(\mathbf{k},t) + \Lambda(\mathbf{k})\left[2\rho_{1,1}(\mathbf{k},t) - 1\right], \\ i\hbar\frac{\partial\rho_{1,1}(\mathbf{k},t)}{\partial t} &= \left[\rho_{1,-1}(\mathbf{k},t)\Lambda^{*}(\mathbf{k}) - \rho_{-1,1}(\mathbf{k},t)\Lambda(\mathbf{k})\right], \end{aligned} \tag{6}$$

where for renormalized energy $\Sigma(\mathbf{k})$ and Rabi frequency $\Lambda(\mathbf{k})/\hbar$ we obtain:

$$\begin{aligned} \Sigma(\mathbf{k}) &= 2\varepsilon_{\mathbf{k}1} + \sum_{q\neq 0} V_{2D}(q)T(\varepsilon_k, U, \rho_{1,-1}(\mathbf{k}), \rho_{1,1}(\mathbf{k})), \\ \Lambda(\mathbf{k}) &= \mathbf{E}(t)\mathbf{d}(\mathbf{k}) + \sum_{q\neq 0} V_{2D}(q)P(\varepsilon_k, U, \rho_{1,-1}(\mathbf{k}), \rho_{1,1}(\mathbf{k})). \end{aligned} \tag{7}$$

In Eqs. (7) $V_{2D}(q)$ is the Fourier transform of 2D Coulomb potential, and the electric field has the form $E(t) = E_0\exp(-i\omega t)\exp\left(-(t/\tau)^2\right)$ with $\tau \gg 2\pi/\omega = T$, and analytical expressions for functions T and P are obtained. The integro-differential Eqs. (6) and (7) are solved numerically using Runge–Kutta method with time step $\Delta t < T$. As a result, one can obtain the interband polarization as a function of time and define the macroscopic polarization, i.e.

$$P(t) = \sum_{\mathbf{k}} P_{\mathbf{k}} d^{*}(\mathbf{k}) + c.c.$$

Using Fourier transform of $P(t)$, we compute the optical susceptibility and get the absorption coefficient as its imaginary part.

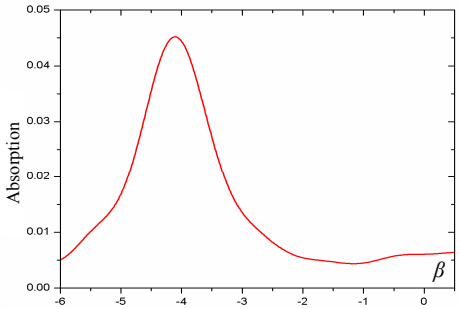

Fig. 1. Excitonic absorption in monolayer graphene as a function of the detuning β for the value of effective fine structure constant $\alpha = 0.175$.

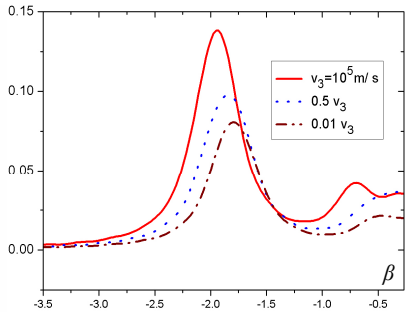

Fig. 2. Excitonic absorption in bilayer graphene as a function of the detuning β for different values of the parameter v_3.

Discussion. Fig. 1 shows excitonic absorption spectrum of gated monolayer graphene with the energy gap 500 *meV* as a function of detuning $\beta = (U - \hbar\omega)/E_R$, where $E_R = \mu e^4/2\hbar^2\chi^2$ is the effective Rydberg energy. Reduced effective electron–hole mass is introduced by the expression $\mu = U/4v_F^2$ as proposed in [9] (v_F is the Fermi velocity in monolayer graphene), so the mass is proportional to the band gap. In the contrast with the standard semiconductor case, in graphene one deals with fine structure constant $\alpha = \chi e^2/\hbar c$ and the appearance of bound states strongly depends on the value of α. For $\alpha = 0.175$ ($\chi = 12.5$) the obtained maximum of the excitonic peak at $4.12\,R^*$ is in a good agreement with the exact analytical solution for relativistic 2D hydrogen atom [12]. In Fig. 2 the excitonic absorption of gated graphene bilayer with opened energy gap 250 *meV* as a function of detuning β is shown. Numerical results are obtained for different values of effective velocity v_3. The solid curve in Fig. 2 corresponds to the value $v_3 = 0.1v_F$. We found that for a smaller value of the parameter $v_3 = 0.5\cdot 10^5\, m/s$ the exciton binding energy becomes smaller (dotted curve).

And finally, when v_3 is almost zero, i.e. when the parameter γ_3, that is responsible for the azimuthal asymmetry of energy bands [6], is not taken into account, the absorption peak decreases and exciton binding energy becomes much smaller (dash-dotted curve).

Conclusion. The excitonic sabsorption in monolayer and bilayer graphene with opened energy gap is studied. The obtained value of excitonic binding energy in monolayer is close to the exact analytical solution. It is shown that the account of azimuthal asymmetry of energy bands in bilayer graphene leads to strong increase of exciton binding energy.

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