Pis'ma v ZhETF, vol. 87, iss. 1, pp. 61 – 65 © 2008 January 10

Electron-hole pair condensation in graphene bilayer

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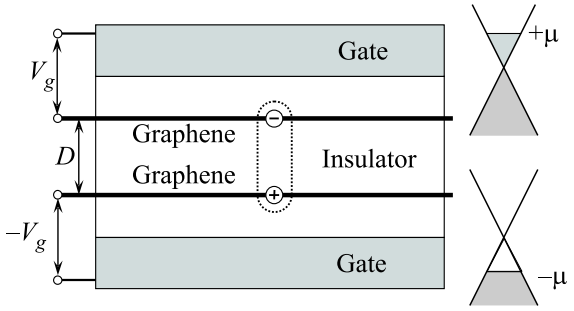
Submitted 26 November 2007

We consider the pairing of electrons and holes due to their Coulomb attraction in two parallel, independently gated graphene layers, separated by a barrier. At weak coupling, there exist the BCS-like pair-condensed state. Despite the fact that electrons and holes behave like massless Dirac fermions, the problem of BCS-like electron-hole pairing in graphene bilayer turns out to be rather similar to that in usual coupled semiconductor quantum wells. The distinctions are due to Berry phase of electronic wave functions and different screening properties. We estimate values of the gap in one-particle excitation spectrum for different interlayer distances and carrier concentrations. Influence of disorder is discussed. At large enough dielectric susceptibility of surrounding medium, the weak coupling regime holds at arbitrarily small carrier concentrations. Localized electron-hole pairs are absent in graphene, thus the behavior of the system *versus* coupling strength is cardinally different from usual BCS-BEC crossover.

PACS: 73.21.Ac, 74.78.−w, 81.05.Uw

Recent progress in experimental technology allowed a fabrication of graphene, one atomic layer separated from graphite crystal [1 – 4]. A lot of theoretical attention to graphene has emerged due to peculiar properties of its band structure, consisting in linear dispersion of electron energy near two inequivalent points of the Brillouin zone [5 – 7]. An electron wave function close to these points is well described by the two-dimensional Dirac equation for massless particles [6, 7] with the Fermi velocity $v_F \approx 10^6\,\mathrm{m/s} \approx c/300$ playing the role of effective "speed of light" [2]. Several peculiar transport phenomena in graphene has been discovered experimentally, e.g., anomalous quantum Hall effect [3] and minimal conductivity [2, 8]. Unique properties of graphene, such as unusually high mobility of charge carriers [1] and a phase coherent transport [9, 10], allow to propose it as a base of future nanoelectronic devices [11 – 13].

In the present work we consider the formation of the condensate of spatially separated electron-hole pairs in bilayer graphene structure when interlayer tunneling is negligible. A condensation of spatially separated electron-hole pairs in usual coupled semiconductor quantum wells (CQW) due to their Coulomb attraction has been proposed theoretically in [14]. A nondissipative motion of resulting pairs leads to appearance of persistent electric currents, flowing in two layers in opposite directions (contrary to 3D case, where phase fixation leads to a formation of excitonic insulator state [15]). In the present Letter we consider another physical realization of a two-dimensional electron-hole system – graphene bilayer. Its schematic setup is shown on Figure.



Schematic set-up of a system for realization of a pairing of spatially separated electron and holes in graphene bilayer. In the right: chemical potential positions in two graphene layers, adjusted by gate voltages V_g and $-V_g$

Two parallel graphene sheets are separated by a dielectric layer of thickness D, large enough to neglect tunneling between them. By applying the gate voltage V_g between graphene sheet and a gate electrode, isolated from it by dielectric layer, one can adjust the charge carrier concentration, fixing the chemical potential μ at any desired level [1] (in addition to electrical one, chemical or electrochemical [16] doping of graphene is also possible). Chemical potentials in either of graphene layers may be adjusted independently. We consider the case of equal densities, when in the top layer the chemical potential is $\mu > 0$, and charge carriers are electrons, whereas in the bottom layer the chemical potential is $-\mu < 0$, and charge carriers are holes. At weak coupling conditions, the system is unstable with respect to Bardeen-Cooper-

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Schrieffer (BCS) interlayer pairing of electrons and holes due to their Coulomb attraction.

Consider the effective Hamiltonian of the system, responsible for pairing of electrons from the top graphene layer and holes from the bottom layer. The influence of remaining part of the total Hamiltonian, corresponding to electron and hole interactions within individual graphene layers, manifests itself via screening of interlayer Coulomb interaction. The effective Hamiltonian can be presented in the form:

$$H_0 = g_s g_v \sum_{\mathbf{k}} \xi_{\mathbf{k}} (a^+_{\mathbf{k}} a_{\mathbf{k}} + b^+_{\mathbf{k}} b_{\mathbf{k}}) + \frac{g_s^2 g_v^2}{S} \times$$
$$\times \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{q}} V(\mathbf{q}) \cos\frac{\varphi_1}{2} \cos\frac{\varphi_2}{2} a^+_{\mathbf{k}_1+\mathbf{q}} b^+_{\mathbf{k}_2-\mathbf{q}} b_{\mathbf{k}_2} a_{\mathbf{k}_1}, \quad (1)$$

where $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ are destruction operators for Dirac electron and hole quasiparticles with in-plane momentum $\mathbf{k}$, $\xi_{\mathbf{k}} = \hbar v_F |\mathbf{k}| - \mu$ is a quasiparticle energy measured from the Fermi level, $V(\mathbf{q})$ is the potential of screened electron-hole interaction, $\mathbf{q}$ is a momentum transmitted via Coulomb scattering, φ_1 and φ_2 are scattering angles for electron and hole. The graphene-specific factor $\cos(\varphi_1/2)\cos(\varphi_2/2)$ results after folding over components of a spinor electron wave function. Coefficients $g_s = g_v = 2$ arise due to spin and valley degeneracy of electron states in graphene, and S is the area of a bilayer.

Within the framework of BCS approach for a pairing of electrons and holes with opposite momenta [17], the Hamiltonian (1) transforms into the form:

$$H = g_s g_v \sum_{\mathbf{k}} \xi_{\mathbf{k}} (a^+_{\mathbf{k}} a_{\mathbf{k}} + b^+_{\mathbf{k}} b_{\mathbf{k}}) + \frac{g_s^2 g_v^2}{S} \times$$
$$\times \sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q}) \frac{1+\cos\varphi}{2} a^+_{\mathbf{k}+\mathbf{q}} b^+_{-\mathbf{k}-\mathbf{q}} b_{-\mathbf{k}} a_{\mathbf{k}}, \quad (2)$$

where φ is angle between $\mathbf{k}$ and $\mathbf{k}+\mathbf{q}$, i.e. scattering angle, equal for electron and hole. Here the factor $(1+\cos\varphi)/2$ means an overlap of initial and final electron states. It originates from Berry phase of electronic wave functions in graphene and have no analogue in CQW [18].

The Hamiltonian (2) can be diagonalized by Bogolyubov transformation:

$$a_{\mathbf{k}} = u_{\mathbf{k}} \alpha_{\mathbf{k}} + v_{\mathbf{k}} \beta^+_{\mathbf{k}}, \quad b_{-\mathbf{k}} = u_{\mathbf{k}} \beta_{\mathbf{k}} - v_{\mathbf{k}} \alpha^+_{\mathbf{k}}.$$

Introducing usual notations

$$u^2_{\mathbf{k}} = \frac{1}{2}\left(1 + \frac{\xi_{\mathbf{k}}}{E_{\mathbf{k}}}\right), \quad v^2_{\mathbf{k}} = \frac{1}{2}\left(1 - \frac{\xi_{\mathbf{k}}}{E_{\mathbf{k}}}\right),$$
$$u_{\mathbf{k}} v_{\mathbf{k}} = \frac{1}{2}\frac{\Delta_{\mathbf{k}}}{E_{\mathbf{k}}}, \quad E_{\mathbf{k}} = \left(\xi^2_{\mathbf{k}} + \Delta^2_{\mathbf{k}}\right)^{1/2},$$

we derive the self-consistent gap equation

$$\Delta_{\mathbf{k}} = -\frac{g_s g_v}{4\pi^2} \int d\mathbf{q} V(\mathbf{q}) \frac{1+\cos\varphi}{2} \frac{\Delta_{\mathbf{k}+\mathbf{q}}}{2E_{\mathbf{k}+\mathbf{q}}}, \quad (3)$$

where we turned from summation to integration on $\mathbf{q}$. Note that Eq. (3) differs from the analogous one for CQW in three aspects. The first one is the additional double degeneracy g_v of electron states due to existence of two valleys. The second one is the linear dependence of $\xi_{\mathbf{k}}$ on $\mathbf{k}$ instead of a quadratic one for CQW; however, near the Fermi energy it can be linearized, hence this difference is not essential. And the last aspect is the presence of the overlap factor $(1+\cos\varphi)/2$, which suppresses backscattering in graphene. At weak coupling this factor turns into unity due to a predominance of small scattering angles in the integral in (3), but at stronger coupling it can weaken the pairing.

The main contribution to the integral in (3) comes from the region near to the Fermi energy, where the dynamically screened interlayer electron-hole interaction $V(\mathbf{q},\omega)$ is attractive. In the linear response approximation,

$$V(\mathbf{q},\omega) = \frac{-v_{\mathbf{q}} e^{-qD}}{1 - v_{\mathbf{q}}(\chi_1+\chi_2) + v^2_{\mathbf{q}}\chi_1\chi_2(1-e^{-2qD})}, \quad (4)$$

where $v_{\mathbf{q}} = 2\pi e^2/\varepsilon q$ is the bare Coulomb interaction, ε is the dielectric constant of a surrounding medium, χ_1 and χ_2 are dynamic polarizabilities within the top and bottom graphene sheets. In the case of equal densities, due to the particle-hole symmetry, polarizabilities are equal in both graphene sheets: $\chi_1 = \chi_2 = \chi$. The equation

$$1 - 2v_{\mathbf{q}}\chi(\mathbf{q},\omega) + v^2_{\mathbf{q}}\chi^2(\mathbf{q},\omega)(1-e^{-2qD}) = 0 \quad (5)$$

describes two branches $\omega_\pm(q)$ of a plasmon dispersion in the system, corresponding to in-phase and antiphase plasma oscillations [14, 19, 20].

The weak coupling (or BCS) regime takes place when the region of pairing near to the Fermi energy is narrow with respect to the Fermi energy itself. In such a case, the radial integration on $\xi \equiv \xi_{\mathbf{k}+\mathbf{q}}$ in the gap equation (3) can be decoupled from the integration on the polar angle φ. We restrict the integration on ξ to the region limited by the cutoff energy $\hbar\tilde{\omega}$, where the *dynamically* screened interaction (4) is attractive. As for the integration on φ, we perform it with the *statically* screened electron-hole attractive potential $V(\mathbf{q}) \equiv V(\mathbf{q},0)$.

Assume that the main part of the integral in (3) comes due to integrands with small q, not larger than $\tilde{q}$ by order of magnitude. Let define $\tilde{q}$ as satisfying the equation $V(\tilde{q}) = V(0)/2$. The explicit form of $V(\mathbf{q})$ is determined by the static polarizability [20–22]

$$\chi(\mathbf{q},0) = -\frac{g_s g_v \mu}{2\pi\hbar^2 v_F^2} \equiv -\frac{\varepsilon}{\pi e^2 a}, \qquad (6)$$

where a is the Thomas-Fermi screening length in graphene.

Let introduce the Fermi momentum $k_0 = \mu/\hbar v_F$ and the dimensionless parameter

$$\alpha = \frac{2\varepsilon\hbar v_F}{g_s g_v e^2} = \frac{1}{2r_s} \approx 0.23 \times \varepsilon,$$

where the dimensionless Wigner-Seitz radius r_s measures the ratio of the characteristic Coulomb energy of quantum system to its characteristic kinetic energy [20]. In 2D semiconductor system r_s increases with decreasing carrier density, but in graphene it is determined only by a dielectric constant ε of surrounding medium.

There are three characteristic distances in the system, namely a, D and the mean separation between charge carriers within each graphene layer $l \sim 1/k_0$. A behavior of the system depends on a relation between a, D and l. As can be shown, $a/l = \alpha$ and $D/l = k_0 D$. Therefore, a behavior of the system is governed by two dimensionless parameters, α and $k_0 D$.

The characteristic momentum $\tilde{q}$ can easily be found from (4) and (6) in two limiting cases: $\alpha \ll k_0 D$ and $\alpha \gg k_0 D$. In the first one, an effective momentum cutoff occurs due to the factor $\exp(-qD)$ in the numerator of (4), thus we get $\tilde{q} \approx 2/D$. In the second one, it is determined by the Thomas-Fermi screening, and we get $\tilde{q} \approx 4k_0/\alpha$. Both results can be written as

$$\tilde{q} = \min\left(\frac{4k_0}{\alpha}, \frac{2}{D}\right). \qquad (7)$$

The cutoff energy $\hbar\tilde{\omega}$ is determined by a characteristic frequency of the lower branch of plasma oscillations and can be estimated as $\hbar\tilde{\omega} = \hbar\omega_-(\tilde{q})$. To the first order in the electron-electron interaction, the dynamic polarizability at $q \to 0$ and $\omega > v_F q$ is [20, 22]

$$\chi(\mathbf{q},\omega) = \frac{g_s g_v \mu q^2}{4\pi\hbar^2\omega^2}. \qquad (8)$$

In the case $\alpha \ll k_0 D$, from (5) and (8) we find $\omega_-(q) = v_F q(k_0 D/\alpha)^{1/2}$ and $\omega_+(q) = v_F(2k_0 q/\alpha)^{1/2}$. Thus the cutoff energy is $\hbar\tilde{\omega} = 2\mu/(k_0 D\alpha)^{1/2}$. In the case $\alpha \gg k_0 D$, the approximate expression (8) is inapplicable, because the lower branch of a plasmon dispersion, formally found with it, falls into a single-particle excitation continuum $\omega < v_F q$. Actually, in this case $\omega_-(q) = v_F q$ and $\omega_+(q) = v_F(2k_0 q/\alpha)^{1/2}$, and the cutoff energy is $\hbar\tilde{\omega} = 4\mu/\alpha$.

Electron-hole Cooper pairs have a size of order of $1/\tilde{q}$ in the in-plane direction. The weak coupling requires a large pair size relative to mean separation between nearest pairs, i.e. $\tilde{q}l \ll 1$. Using (7), we conclude, that the weak coupling regime occurs when at least one of parameters α or $k_0 D$ is large with respect to unity.

Assuming that $\Delta = \text{const}$ at $|\xi| \leqslant \hbar\tilde{\omega}$ and $\Delta = 0$ at $|\xi| > \hbar\tilde{\omega}$, one can find an approximate solution of (3). We derive asymptotic expressions for the gap for different relations between α, $k_0 D$ and unity.

At $\alpha \ll k_0 D$ we get

$$\Delta = \frac{4\mu}{(k_0 D\alpha)^{1/2}} \exp\left\{-4\pi k_0 D\left(1 + \frac{k_0 D}{\alpha}\right)\right\}. \qquad (9)$$

This expression is suitable for values of α, both small and large with respect to unity.

At $\alpha \gg k_0 D$ we distinguish cases of small and intermediate interlayer distances D. In the first case, $k_0 D \ll 1 \ll \alpha$, the gap is

$$\Delta = \frac{8\mu}{\alpha} \exp\left\{-\frac{\pi\alpha}{\ln(1+\alpha/2)}\right\}. \qquad (10)$$

In the second case, $1 \ll k_0 D \ll \alpha$, the gap is

$$\Delta = \frac{8\mu}{\alpha} \exp\left\{-\frac{\pi\alpha}{\ln(\alpha/4k_0 D) - \gamma}\right\}, \qquad (11)$$

where $\gamma \approx 0.577$ is the Euler constant.

We have considered the weak coupling regime, but what can occur at stronger coupling? In CQW, at $T = 0$, on increase of a coupling strength there exist a crossover from BCS-like state to Bose-Einstein condensation (BEC) in a dilute gas of localized, non-overlapping electron-hole pairs (or excitons for quasi-equilibrium state created after laser pumping). At $T \neq 0$, both BCS-like state and gas of local pairs are in superfluid state below the temperature of Kosterlitz-Thouless transition to the normal state [14]. In graphene, there are no bound solutions for the Dirac problem of single electron in attractive potential due to the absence of a gap in the energy spectrum. Similarly, as can be shown, there are no localized electron hole-pairs in a bilayer. Therefore a behavior of graphene electron-hole bilayer on increase of the coupling strength will be cardinally different from BCS-BEC crossover in CQW. Strong coupling regime will be studied in subsequent publication. Note, that in a perpendicular magnetic field the existence of localized magnetoexcitons makes BCS-BEC crossover possible [23, 24].

Consider the weak coupling conditions more closely at $\alpha \ll 1$ or $\alpha \sim 1$. Such values of α can be realized with, e.g., commonly used SiO_2 substrate ($\varepsilon \approx 4$). In this case the coupling strength is determined only by a value of $k_0 D$. The Fermi momentum k_0 is proportional

to μ, which can be tuned from zero to maximal values of $\approx 0.3\,\mathrm{eV}$ in electrically doped graphene [1,2]. The weak coupling regime ($k_0 D \gg 1$) can be achieved with reasonable carrier concentrations at any interlayer distance $D > 100\,\text{Å}$. On the other side, by tending μ to zero, one can always achieve a strong coupling regime. Thus, the whole transition from weak to strong coupling can be realized experimentally by changing the gate voltage.

The case $\alpha \gg 1$ takes place at large values of the dielectric constant ε of surrounding medium (at least $\varepsilon > 5$), which can be achieved with, e.g., HfO_2 ($\varepsilon \approx 25$). In this case, the weak coupling regime sustains even at $\mu \to 0$, i.e. at arbitrarily small carrier concentrations, and the gap tends to zero as $\Delta \sim \mu$ according to (10). This provides a remarkable contrast with CWQ, where a strong coupling regime occurs inevitably at vanishingly small carrier concentrations.

With the typical $\mu \approx 0.1\,\mathrm{eV}$ and minimal reasonable interlayer distance $D = 50\,\text{Å}$, at $\varepsilon = 10$ (when weak coupling approximation is still reliable) the expression (10) gives $\Delta = 3 \cdot 10^{-5}\,\mathrm{eV}$, which is equivalent to the temperature $0.35\,\mathrm{K}$. The maximal value of Δ can be achieved at strong coupling, when $\alpha \sim k_0 D \sim 1$. In this case $\Delta \sim \mu$, up to hundreds of Kelvins.

Let now estimate the influence of disorder. In the case of conventional phonon-mediated superconductivity, a presence of magnetic impurities acts destructively on BCS state because of a different scattering of electrons with opposite spins, which form Cooper pairs. In our case any impurity localized in either graphene layer acts destructively on pair condensate, since it scatters only one pair constituent. The BCS-like state maintains if the mean free path λ exceeds the coherence length $l_\Delta = \hbar v_\mathrm{F}/\Delta$ (analogously to Ref. [25]). From the expression for diffusive Boltzmann conductivity [26] we derive $\lambda \approx (\mu/ev_\mathrm{F})\mu_\mathrm{c}$, where μ_c is the carrier mobility in graphene. In dirty graphene samples $\mu_\mathrm{c} \approx 1000\,\mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$ at room temperature [1] and the corresponding mean free path is $\lambda \approx 10\,\mathrm{nm}$ at $\mu = 0.1\,\mathrm{eV}$. On the contrary, for clean graphene at the temperature of liquid helium $\mu_\mathrm{c} \approx 10^6\,\mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$, so $\lambda \approx 10\,\mu\mathrm{m}$. The coherence length is $l_\Delta = 22\,\mu\mathrm{m}$ for $\Delta = 3 \cdot 10^{-5}\,\mathrm{eV}$ and $l_\Delta = 7\,\mathrm{nm}$ for $\Delta = \mu = 0.1\,\mathrm{eV}$. Therefore, in the weak coupling regime a rather weak disorder can destroy the BCS-like state, though in the strong coupling regime the BCS-like state can survive at rather strong disorder.

The onset of electron-hole pairing may be observed experimentally via drag effect peculiarities. It has been shown for CQW, that an occurrence of pair condensation leads to a sharp increase of drag resistivity [27–29]. Another possibility is an observation of Josephson-like effect (see, e.g., [30] and references therein). Also the condensation modifies an electromagnetic response of the system. In particular, an application of in-plane magnetic field leads to a formation of persistent dipolar supercurrent, which can be detected directly [14,30,31].

In CQW, the serious obstacle to an occurrence of a BCS-like electron-hole pairing is an anisotropy of the hole band [27,32]. However in graphene the particle-hole symmetry provides almost perfect matching in shape between electron and hole Fermi lines. In this connection it is interesting to discuss a possible effect of the trigonal warping in graphene on BCS-like pairing in our system. The trigonal warping breaks the isotropy of Dirac spectrum in graphene and causes a deviation of the Fermi line from a perfect circle towards a triangle-like shape [18]. The warping is negligible at low carrier concentrations, but becomes considerable at large enough μ. The triangle-like deviation has opposite orientations in two graphene valleys and therefore breaks the valley symmetry. Therefore the pairing between electrons and holes from same valleys will be prevailing, but the pairing from different valleys will also be present, if the trigonal warping is not very large. In such a situation, a two-gap state, similar to that in superconductor with overlapping bands [33], can be formed.

Two spatially separated *bilayer* graphene sheets can also be a candidate for a realization of the BCS-like pair condensation. In a perpendicular electric field an electron spectrum in bilayer graphene acquires a tunable gap and energy dispersion has a quite unusual "Mexican hat" shape [34,35]. After electrical doping a nontrivial Fermi surface in the shape of a ring will be formed [36]. In principle, the BCS-like pairing between electron and hole Fermi-rings is possible, leading to a formation of two energy gaps – inside and outside of the ring.

In conclusion, we have analyzed a possibility of the BCS-like pairing between spatially separated electrons and holes in two parallel graphene layers with negligible interlayer tunneling. At weak coupling the problem of pairing is rather similar to that in CQW, except for several graphene-specific differences, i.e. Berry phase of electronic wave functions and different screening properties. We have derived asymptotic expressions for the gap in the excitation spectrum at various characteristics of the system and estimated its numerical value under reasonable conditions. Estimations of an influence of disorder have been carried out. An appearance of the BCS-like electron-hole condensate can be observed experimentally via Coulomb drag measurements, by studying a Josephson-like effect or by probing an electromagnetic response of the system. The trigonal warping in the electron energy dispersion can lead to a formation of a two-gap state.

There is no localized electron-hole pairs in graphene bilayer due to absence of a gap in the energy spectrum. Therefore a behavior of the system on increase of the coupling strength is cardinally different from BCS-BEC crossover in CQW. Both weak and strong coupling conditions can be achieved experimentally. If a dielectric constant of surrounding medium is large enough, then the weak coupling regime sustains at arbitrarily small carrier concentrations, in contrast to a situation in CQW.

Authors are obliged to participants of the Workshop on strongly correlated systems (Institute of High Pressure Physics) for useful discussions of the results. Also the support from the Russian Foundation for Basic Research (Grant 06-02-81036-Bel-a) is acknowledged.

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