Coherent phases and collective electron phenomena in graphene

Yu E Lozovik and A A Sokolik
Institute of Spectroscopy, Russian Academy of Sciences, 142190 Troitsk, Russia
E-mail: lozovik@isan.troitsk.ru

Abstract. Unique band structure peculiarities of graphene imply that near Fermi level electrons are effectively described by two-dimensional Dirac equation for massless particles. We investigate how these peculiarities manifest in electron-hole pairing and properties of indirect magnetoexcitons in two spatially separated, independently gated graphene layers. We study electron-hole pairing in weak-coupling regime, find the gap in energy spectrum and discuss system behavior at various controlling parameters. In case of extremely strong coupling, we show that localized electron-hole pairs are absent in graphene, and thus a behavior of graphene bilayer versus coupling strength is cardinal different from crossover to local pairs in usual Fermi systems. We discuss spectroscopy of indirect magnetoexcitons in graphene and possibility of their superfluidity. The systems under consideration can reveal coherent properties, dissipationless currents and Josephson-like phenomena at room temperature.

Introduction
Graphene, being a single atomic layer separated from graphite crystal, has a two-dimensional honeycomb lattice. Its peculiar band structure implies that conduction and valence bands touch at two inequivalent degeneracy points within the Brillouin zone (two “valleys”). Near each of these points electron low-energy dynamics is effectively described by two-dimensional Dirac equation for massless particles with the Fermi velocity \( v_F \approx 10^6 \text{ m/s} \approx c/300 \) playing the role of effective “speed of light”. Within each valley, electron effective (envelope) wave function is a two-component spinor, which components represent electronic Bloch states built-up on two triangular graphene sublattices. Graphene has been fabricated recently, and Dirac-like behavior of its electrons has been confirmed experimentally [1]. Due to its fundamental novelty and large potential for applications, graphene has emerged a great interest in condensed-matter community [2, 3].

“Ultrarelativistic” nature of electrons in graphene has two important consequences: (i) electrons are chiral particles and are described by multicomponent wave function; (ii) linear dispersion law leads to peculiar relation between potential and kinetic energies of electron system. Therefore investigation of electron phenomena in graphene is of great fundamental interest. In this paper we discuss electron-hole pairing (sections 1 and 2) and magnetoexcitton formation (section 3) in two spatially separated graphene layers. The results tells on the possibility of superfluidity and existence of dissipationless electric currents in coupled graphene layers at room temperature.
1. Electron-hole pairing in graphene bilayer

We consider a system of two parallel graphene layers separated by a dielectric layer of thickness $D$, large enough to neglect interlayer tunneling. Its schematic set-up is shown in figure 1. By applying a gate voltage $V_g$ between graphene sheet and gate electrode, one can adjust a charge carrier concentration $n$, fixing the chemical potential $\mu = \hbar v_F \sqrt{\pi n}$ at any desired level [1]. Chemical potentials in both graphene layers can be adjusted independently. We consider the case of antisymmetrically biased electron-hole bilayer, when top and bottom layers are doped with electrons and holes, having the chemical potentials $\mu > 0$ and $-\mu$ respectively. This system is unstable with respect to interlayer pairing of electrons and holes due to their Coulomb attraction. In weak coupling regime, this pairing is similar to phonon-mediated electron pairing in metals described by Bardeen-Cooper-Schrieffer (BCS) theory [4]. The onset of electron-hole pairing can manifest in transport characteristics as sharp amplification of Coulomb drag effect. Other possibilities are observation of both stationary and non-stationary Josephson-like effects and studying electromagnetic response of the system.

Electron-hole pairing in usual semiconductor coupled quantum wells (CQW) was considered theoretically in [6]. Similar phenomenon can occur in 3D semimetals: a condensation of spontaneously created electron-hole pairs leads to a formation of excitonic insulator state [7] (insulating behavior arise due to fixation of order parameter phase, prohibiting any supercurrents).

To describe the pairing in graphene bilayer in a weak coupling regime, we single out a relevant part of the total Hamiltonian of the system, pairing Hamiltonian, which deals only with electrons from conduction band of the top graphene layer and with holes from valence band of the bottom layer.

Thus, we compose the pairing Hamiltonian from kinetic energies of conduction-band electrons from the top layer and valence-band holes from the bottom layer, and from potential energy of screened interaction of electrons and holes with opposite momenta. Diagonalizing this Hamiltonian by means of Bogolyubov transformation, we derive the self-consistent gap equation

$$\Delta_p = \frac{\int d\mathbf{q} (2\pi \hbar)^2 V(\mathbf{q}) \frac{1 + \cos \varphi}{2} \frac{\Delta_{p+\mathbf{q}}}{2 \sqrt{\xi_{p+\mathbf{q}}^2 + \Delta_{p+\mathbf{q}}^2}}}{\xi_p = v_F |p| - \mu}$$

Here $\xi_p = v_F |p| - \mu$ is a quasiparticle (electron or hole) energy measured from the Fermi level, $V(\mathbf{q})$ is potential energy of screened electron-hole interaction. The factor $(1 + \cos \varphi)/2$, where $\varphi$ is angle between $\mathbf{p}$ and $\mathbf{p} + \mathbf{q}$, means an overlap of initial and final electron states in Coulomb scattering. It originates from pseudospinor structure of electronic wave functions in graphene and has no analogue in CQW.

Main contribution to the integral in (1) comes from the region near the Fermi energy, where the dynamically screened interaction $V(\mathbf{q}, \omega)$ is attractive. In random phase approximation,
\( V(q, \omega) \) can be expressed through polarizabilities of each graphene layer [6, 8]. The weak coupling (or BCS) regime takes place when the width of the pairing region is narrow with respect to the Fermi sphere. Let denote the characteristic half-width of this region by \( \hbar \omega \).

The solution of (1) for \( l \)-wave pairing has the conventional BCS-like form \( \Delta = 2\hbar \omega \exp\{\frac{-1}{\mathcal{N}V_{l}}\} \), where \( \mathcal{N} = k_{0}/2\pi \hbar v_{F} \) is the density of states at the Fermi level, \( k_{0} = \mu/\hbar v_{F} \) is the Fermi wave vector, and

\[
V_{l} = \frac{1}{2\pi} \int_{0}^{2\pi} d\varphi \, \frac{1 + \cos \varphi}{2} e^{-i\varphi} V_{l} \left( 2\hbar k_{0} \left| \sin \frac{\varphi}{2} \right| \right) .
\] (2)

is \( l \)-wave harmonic of statically screened pairing potential \( V(q) \) averaged over the Fermi line. At weak coupling, \( V_{l} \) is maximal at \( l = 0 \), so \( s \)-wave pairing dominates.

Let introduce the parameter \( \alpha = \varepsilon \hbar v_{F}/2e \approx 0.23 \times \varepsilon \), where \( \varepsilon \) is a dielectric susceptibility of surrounding medium. One can show [5] that behavior of the system is governed by two dimensionless parameters, \( \alpha \) and \( k_{0}D \), describing the screening of the pairing interaction. When \( \alpha \gg k_{0}D \), an effective cutoff of this interaction occurs due to the Thomas-Fermi screening within graphene layers, and when \( \alpha \ll k_{0}D \), the cutoff is provided by large interlayer separation \( D \). If one of the parameters \( \alpha \) or \( k_{0}D \) is large with respect to unity, then the screening is strong enough, and the weak coupling regime is realized.

The cutoff energy \( \hbar \omega \) is determined by a characteristic energy of the lower (acoustic) branch of plasma oscillations in the bilayer. When \( \alpha \ll k_{0}D \), we get \( \hbar \omega = 2\mu/(k_{0}D\alpha)^{1/2} \), and when \( \alpha \gg k_{0}D \), we get \( \hbar \omega = 4\mu/\alpha \).

Calculating asymptotics of (2) at different relations between \( \alpha \), \( k_{0}D \) and unity, one can get analytical expressions for the gap [5]. For example, in the case of small interlayer separation \( (k_{0}D \ll 1) \) and large dielectric constant of surrounding medium \( (\alpha \gg 1) \), we get

\[
\Delta = \frac{8\mu}{\alpha} \exp \left\{ -\frac{1}{\mathcal{N}V_{0}} \right\} , \quad \text{where} \quad \mathcal{N}V_{0} = \frac{1}{2\pi\alpha} \left\{ \frac{\pi}{\alpha} - 1 + \sqrt{1 - \frac{4}{\alpha^{2}}} \cosh^{-1} \frac{\alpha}{2} \right\} .
\] (3)

In the case when \( \alpha \ll 1 \) or \( \alpha \sim 1 \) (this takes place for small enough \( \varepsilon \)) the coupling strength is determined only by the value \( k_{0}D \). The weak coupling regime \( (k_{0}D \gg 1) \) will take place at large enough \( D \), whereas the strong coupling \( (k_{0}D \ll 1) \) can always be recovered when \( \mu \) goes to zero. Thus the whole transition from weak to strong coupling can be traced in experiment by changing a gate voltage.

In the case \( \alpha \gg 1 \) (if \( \varepsilon \) is large enough) the weak coupling regime sustains even at \( \mu \to 0 \), i.e. at arbitrarily small carrier concentrations, and the gap tends to zero as \( \Delta \propto \mu \) according to (3). This turns out to be in essential contrast with CQW, where a strong coupling regime occurs inevitably at vanishingly small carrier concentrations.

Thus, we have considered the weak coupling regime but we have shown, that strong coupling conditions are also achievable experimentally. A treatment of strong-coupling pairing is much more complicated, than that of weak-coupling one. Note, that in CQW at \( T = 0 \) on increase of the 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 [6]. At \( T \neq 0 \), both BCS-like state and local pairs gas are in superfluid state below Kosterlitz-Thouless transition temperature. In graphene, there are no localized electron-hole pairs due to absence of gap in the band spectrum. The next section is devoted to detailed consideration of this problem.

2. The problem of single electron-hole pair in graphene

In this section we present the results of consideration of toy model — one isolated electron-hole pair in one graphene layer or in coupled graphene layers, neglecting Fermi sea presence
(i.e. \( \mu \) is negligible). The analysis of solution of eigenfunction problem for the Hamiltonian
\[ H = v_F (\sigma_x p_1 + \sigma_y p_2) + V(|r_1 - r_2|), \]
\( V \) is electron-hole interaction, shows that rest electron-hole pair in graphene bilayer is not localized: its wave function at \( r \to \infty \) behaves as a wave function of infinite motion at any screening conditions (contrary to CQW case, where excitonic-like pairs are formed at strong coupling). Therefore, a formation of dilute gas of electron-hole pairs is impossible in our system, and the overlap of neighboring pairs is essential. Hence we assume, that on increase of the coupling strength the nature of pair-condensed state will remain to be BCS-like (instead of BCS-BEC crossover in CQW), though with some modifications arising due to pseudospinor character of electron and hole wave functions. A critical temperature of such “ultrarelativistic” BCS-like state can be very large, up to room values.

3. Indirect magnetoexcitons in graphene bilayer
Absence of localized electron-hole pairs in graphene is consistent with the problem of ultrarelativistic particles, which cannot be confined by electric potential. However, electrons in graphene can be confined in magnetic field [12]. We consider here properties of localized pairs made up by electrons and holes from spatially separated graphene layers placed into perpendicular magnetic field.

A problem of electron-hole pair in magnetic field, magnetoexciton, is simplified due to existence of conserving magnetic momentum \( P \), playing the role of total momentum of the pair [13]. However, is contrast to usual two-body problem, a center-of-mass motion of electron and hole is intimately connected with their relative motion in magnetic field, leading to dispersion dependence of magnetoexciton energy on \( P \) [14]. Properties of direct magnetoexcitons in single graphene layer has been investigated in [15]. We consider indirect magnetoexcitons in graphene bilayer by analogy with magnetoexcitons in CQW [16].

Electrons in graphene layer in magnetic field \( B \) populate “ultrarelativistic” Landau levels
\[ E_n^L = (\hbar v_F/l) sgn(n) \sqrt{2|n|}, \]
where \( n = 0, \pm 1, \pm 2, \ldots \) and \( l = \sqrt{\hbar c/eB} \) is the magnetic length [17]. Electrical doping of graphene layers allows to control their Landau levels fillings. If \( n_1 \)-th Landau level of the top graphene layer is populated by a small amount of electrons, whereas \( n_2 \)-th level of the bottom layer is populated by the same amount of holes, then electrons and holes will form dilute gas of interlayer magnetoexcitons.

In the symmetrical gauge \( A = \frac{1}{2} [B \times r] \), the Hamiltonian of electron-hole pair in joint basis of their sublattices reads
\[ H = \frac{\hbar v_F \sqrt{2}}{l} \left( \begin{array}{cccc} 0 & a_2 & a_1 & 0 \\ a_2^+ & 0 & 0 & a_1 \\ a_1^+ & 0 & 0 & a_2 \\ 0 & a_1^+ & a_2^+ & 0 \end{array} \right) + V(|r_1 - r_2|), \]
where \( a_1 = (l/h)[p_{1-} - i(eB/2c)r_{1-}] \) and \( a_2 = (l/h)[p_{2+} - i(eB/2c)r_{2+}] \). \( p_{k\pm} = (p_{kx} \pm ip_{ky})/\sqrt{2} \), and analogously for \( r_{k\pm} \), \( k = 1, 2 \) (the subscript 1 refers to the electron, 2 — to the hole). The following commutation relations take place: \([a_k, a_m^+] = \delta_{km} \), \([a_k, a_m] = 0 \), \([a_k^+, a_m^+] = 0 \).

The magnetic momentum of the pair is \( P = p_1 + p_2 - (e/2c)[B \times (r_1 - r_2)] \). Its components \( P_x \) and \( P_y \) commute with each other and with \( H \). Therefore we can find solution of \( H \Psi = E \Psi \) as an eigenfunction of \( P \) (analogously to [14]):
\[ \Psi(r_1, r_2) = \frac{1}{\sqrt{S}} \exp \left\{ \frac{i}{\hbar} R \left( P + \frac{e}{2c} [B \times r] \right) \right\} \Psi'(r - r_0), \]
where \( R = (r_1 + r_2)/2 \), \( r = r_1 - r_2 \), \( r_0 = (l^2/Bh)[B \times P] \) and \( S \) is the system area.

The formula (4) introduces the unitary transformation \( \Psi' = S \Psi \), dependent parametrically on \( P \). Making corresponding transformations for operators of the form \( A' = SAS^+ \), we get
\( V_p = V(|r + r_0|), a' = a, a' = -b \), where \( a = (l/\hbar)[p_- - i(eB/2c)r_-], b = (l/\hbar)[p_+ - i(eB/2c)r_+], p = -i\hbar \nabla_r = (p_1 - p_2)/2 \). The new operators are expressed only through coordinate and momentum of relative electron-hole motion, hence we can diagonalize the transformed Hamiltonian

\[
H' = \frac{\hbar v_F \sqrt{2}}{l} \begin{pmatrix}
0 & -b & a & 0 \\
-b^+ & 0 & 0 & a \\
a^+ & 0 & 0 & -b \\
0 & a^+ & -b^+ & 0
\end{pmatrix} + V(|r + r_0|),
\]

only in the relative motion space.

Let ignore the Coulomb interaction for a moment. The bosonic commutation relations \([a, a^+] = 1\) and \([b, b^+] = 1\) allows us to construct an eigenvector of (5) corresponding to the state of magnetoexciton composed from electrons in \(n_1\)-th Landau level and holes from \(n_2\)-th level:

\[
\Psi'(|r\rangle = \Phi_{n_1,n_2}(r) = (\sqrt{2})^{\delta_{n_1,0} + \delta_{n_2,0}} \begin{pmatrix}
\text{sgn}(n_1) \text{sgn}(n_2) \phi_{|n_1|,|n_2|}(r) \\
\text{sgn}(n_1) \phi_{|n_1|,|n_2|}(r) \\
\text{sgn}(n_2) \phi_{|n_1|,|n_2|}(r) \\
\phi_{|n_1|,|n_2|}(r)
\end{pmatrix},
\]

where

\[
\phi_{|n_1|,|n_2|}(r) = \frac{(a^+)^{|n_1|} (b^+)^{|n_2|} e^{-r^2/2l^2}}{\sqrt{|n_1|!} \sqrt{|n_2|!} \sqrt{2\pi l}}
\]

is the wave function of two-dimensional harmonic oscillator \([14, 15]\).

In the case \(D \gg l\) (strong magnetic field or large interlayer separation) characteristic value of electron-hole Coulomb energy \(e^2/\varepsilon D\) is small compared to Landau levels separation \(\sim \hbar v_F / l\). Hence the interlayer Coulomb interaction \(V_p\) can be treated as a small, first-order perturbation (however, intralayer electron interactions are not small and lead to renormalization of Landau levels properties, not taken into account in the present work). The mean value of \(V'_0 = -e^2/\varepsilon \sqrt{(r + r_0)^2 + D^2}\) in magnetoexcitonic state described by \(n_1, n_2\) and \(P\), gives its energy \(E_{n_1,n_2}(P, D) = \langle \Phi_{n_1,n_2}|V'_0|\Phi_{n_1,n_2}\rangle\). This expression obeys the scaling: \(E_{n_1,n_2}(P, D) = E_0 \hat{E}_{|n_1|,|n_2|}(P/l, D/l)\), where \(E_0 \equiv -E_0(0,0) = (e^2/\varepsilon l)\sqrt{\pi/2}\), independence of \(\hat{E}\) on signs of \(n_1\) and \(n_2\) is provided by the particle-hole symmetry in graphene.

At small \(P\), the magnetoexciton energy can be represented as

\[
E_{n_1,n_2}(P, D) \approx -E_{n_1,n_2}^{(b)}(D) + \frac{P^2}{2M_{n_1,n_2}(D)},
\]

where \(E_{n_1,n_2}^{(b)}(D)\) is magnetoexciton binding energy and \(M_{n_1,n_2}(D)\) is its effective magnetic mass. At large interlayer distances, when \(D \gg l\) and \(D \gg Pl^2/\hbar\), equation (8) is reduced to the asymptotics with universal parameters

\[
E^{(b)}(D) \approx E_0 \frac{l}{D} \sqrt{\frac{2}{\pi}}, \quad M(D) \approx \frac{M_0}{2} \sqrt{\frac{\pi}{2}} \left( \frac{D}{l} \right)^3,
\]

where \(M_0 = 2\hbar^2/E_0l^2\).
Functions $E_{n_1,n_2}^{(b)}(D)$ and $M_{n_1,n_2}(D)$ can be expressed analytically at any $n_1$ and $n_2$, using (6) and (7). For example

\begin{align*}
E_{00}^{(b)}(D) &= -E_0 e^{d^2/2} F \left( \frac{d}{\sqrt{2}} \right), \\
E_{11}^{(b)}(D) &= -E_0 \left\{ \frac{d^4 - 2d^2 + 11}{16} e^{d^2/2} F \left( \frac{d}{\sqrt{2}} \right) - \frac{d(d^2 - 3)}{8\sqrt{2\pi}} \right\}, \\
E_{20}^{(b)}(D) &= -E_0 \left\{ \frac{d^4 - 6d^2 + 7}{16} e^{d^2/2} F \left( \frac{d}{\sqrt{2}} \right) - \frac{d(d^2 - 7)}{8\sqrt{2\pi}} \right\}, \\
M_{00}(D) &= M_0 \left\{ (1 + d^2) e^{d^2/2} F \left( \frac{d}{\sqrt{2}} \right) - d\sqrt{\frac{2}{\pi}} \right\}^{-1}, \\
M_{11}(D) &= M_0 \left\{ \frac{(d^2 + 1)(d^4 + 6d^2 + 7)}{16} e^{d^2/2} F \left( \frac{d}{\sqrt{2}} \right) - \frac{d(d^2 + 3)^2}{8\sqrt{2\pi}} \right\}^{-1}, \\
M_{20}(D) &= M_0 \left\{ \frac{(d^2 + 5)(d^4 - 2d^2 - 1)}{16} e^{d^2/2} F \left( \frac{d}{\sqrt{2}} \right) - \frac{d(d^4 + 2d^2 - 11)}{8\sqrt{2\pi}} \right\}^{-1},
\end{align*}

where $d = D/l$, $F(x) = (2/\sqrt{\pi}) \int_x^\infty e^{-t^2} dt$ is the complementary error function. Results of $E_{n_1,n_2}^{(b)}$ and $M_{n_1,n_2}$ calculation for certain values of $n_1$ and $n_2$ are shown in figures 2a and 2b respectively. On increase of interlayer distance $D$, binding energies decrease, whereas magnetic masses generally increase. Singularity in $M_{20}(D)$, seen in figure 2b, occurs since at small $D$ the dispersion curve $E_{20}(P,D)$ has a “roton” minimum at $P \neq 0$, vanishing at larger $D$ values.

In typical laboratory magnetic field $B = 20$ T, with SiO$_2$ surroundings ($\varepsilon \approx 4.5$), we have $E_0 \approx 0.07$ eV $\approx 800$ K. At $D = 200$ Å, (9) gives $E_{n_1,n_2}^{(b)} \approx 0.016$ eV $\approx 190$ K. Therefore, interlayer magnetoexcitons in graphene can survive at rather high temperatures. Superfluidity in a gas of interlayer magnetoexcitons in graphene is possible below Kosterlitz-Thouless temperature inversely proportional to exciton magnetic mass (see also [18]). Besides, Bose-condensation of magnetoexcitons in coupled quantum dots based on locally gated coupled graphene layers or graphene pieces is also possible.

![Figure 2](image_url)

**Figure 2.** Binding energy (a) and effective magnetic mass (b) of indirect magnetoexciton as a function of interlayer distance $D$ at: $n_1 = n_2 = 0$ (---), $n_1 = n_2 = 1$ (---), $n_1 = 2$, $n_2 = 0$ (---), $n_1 = n_2 = 3$ (---).
Weak tunneling between graphene layers leads to recombination of magnetoexciton with emission of photon. Conservation of momentum and energy allows only recombination of magnetoexcitons with $P = 0$. However, an application of additional magnetic field $B_{\parallel}$, parallel to graphene layers, effectively shifts the dispersion relation $E_{n_1,n_2}(P,D)$ by $\Delta P = eDB_{\parallel}/2c$ and thus allows to “scan” the dispersion curve [19]. The energy of emitted photon is

$$E_{\gamma} = E_{n_1}^{L} - E_{n_1-1}^{L} + V_{n_1,n_2}^{(\text{self})} + E_{n_1,n_2}(\Delta P, D),$$

where

$$V_{n_1,n_2}^{(\text{self})} = \sum_{n=-n_c}^{n_1-1} \langle \Phi_{n,n} | V_{P=0} | \Phi_{n_1,n_2} \rangle - \sum_{n=-n_c}^{n_2} \langle \Phi_{n,n} | V_{P=0} | \Phi_{n_2,n_2} \rangle$$

is a sum of electron and hole self-energies arising due to their exchange with Fermi seas of corresponding graphene layers, filled down to a cutoff Landau level $n_c$ [15]. For example, $V_{00}^{(\text{self})} = E_0$, $V_{11}^{(\text{self})} = (11/16)E_0$, $V_{20}^{(\text{self})} \approx 1.8E_0$, $V_{33}^{(\text{self})} \approx 0.5E_0$. Magnetoexcitonic spectroscopy of graphene can be a convenient tool for investigating properties of graphene Landau levels and electron interactions in graphene in magnetic field. In particular, it can be helpful in uncovering recently discovered mysteries concerning anomalies in Landau levels spacing and unusual splitting of lowest Landau level [20].

Conclusions
The results presented in the paper show that graphene and graphene-based nanostructures are perspective systems for realization of superfluidity and other coherent electron phenomena. High quality of graphene samples and feasibility to create compact layered graphene structures allow to hope that observation of coherent phenomena will be much simpler in these than in conventional quasi-two-dimensional systems. Graphene band structure peculiarities make it possible to achieve new regimes in quantum electron systems.

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