

Phases of the excitonic condensate in two-layer graphene

Yevhen F. Suprunenko, Vadim Cheianov, and Vladimir I. Fal’ko
Department of Physics, Lancaster University, Lancaster LA1 4YB, United Kingdom.
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Two graphene monolayers that are oppositely charged and placed close to each other are considered. Taking into account valley and spin degeneracy of electrons we analyze the symmetry of the excitonic insulator states in such a system and build a phase diagram that takes into account the effect of the symmetry breaking due to the external in-plane magnetic field and the carrier density imbalance between the layers.

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I. INTRODUCTION.

The excitonic insulator was predicted theoretically four decades ago in 3D semiconductors and then in spatially separated layers of electrons and holes. Since then, an excitonic insulator has been searched for in a variety of systems. The excitonic insulator is a material where the electron-hole excitonic correlations lead to the formation of a gapped state characterized by the order parameter resembling a superfluid condensate of excitons. Such a correlated state has been observed in double-quantum well semiconductor structures in quantizing magnetic fields. After the experimental discovery of graphene, it has been discussed as a possible candidate for experimental realization of the excitonic insulator state, sparking the on-going debate about the critical temperature of graphene
doing to the Zeeman energy in an in-plane magnetic field and the asymmetry between electron/hole densities in the layer 1 and 2.

Transitions between phases are found to be of the first order. These transitions are subject to the use of an in-plane magnetic field and a variation of external gate voltages, leading to different charge carriers densities in layers: the density of all electrons $n_{1e}$ in layer 1 (which corresponds to the Fermi energy $E_F^{(1)} = \hbar v_F \sqrt{\pi n_{1e}}/2$, and density of holes $n_{2h}$ in layer 2 (which corresponds to the negative Fermi energy in the layer 2, $E_F^{(2)} = -\hbar v_F \sqrt{\pi n_{2h}}/2$).

The B phase, Fig. 1(b), exists when there is no magnetic field and charge carriers densities are the same in both layers $n_{1e} = n_{2h}$ (i.e. when $E_F^{(1)} = E_F^{(2)}$). The B’ phase exists at the same condition $n_{1e} = n_{2h}$ but when an in-plane magnetic field is applied, which causes a Zeeman splitting of energies of electrons with different spin projections. The A$_1$ phase exists when a symmetry of charge carriers density is violated, e.g. $n_{1e} > n_{2h}$, and when the corresponding splitting of Fermi energies $E_{F1}^{(1)} - |E_{F2}^{(2)}|$ is equal to the Zeeman splitting due to an in-plane magnetic field.

The diversity of obtained phases is due to the high symmetry of the normal ground state, which can be broken in several different ways leading to a variety of phases possessing different symmetry groups. A well known example of the system with the diversity of phases due to various normal state symmetry breaking is liquid Helium-3. In liquid Helium-3 the symmetry of the order parameter can be changed by correspondent external parameters, leading to phase transitions.

The analysis in this paper is organised as follows. Section III describes the theoretical model of the considered system. Pairing of electrons and holes within mean field theory is introduced in the Section IV Section V pro-

FIG. 1. (a) The excitonic condensation due to an electron-hole pairing is studied in the system of two spatially separated graphene monolayers with an excess of electrons on layer 1 and a lack of electrons on layer 2. (b) The schematic phase diagram of the excitonic condensation in the system at different values of a Zeeman splitting and different values of the asymmetry between Fermi energies in layer 1 and 2, $\epsilon F = \mu B |h|$ is the Zeeman energy in an in-plane magnetic field $h$. 

\textit{a} \\
\textit{GATE 1}  \\
\textit{GATE 2}  \\
\textit{GRAPHENE}  \\
\textit{0}$ \\
\textit{Zeeman splitting, $\epsilon F$}  \\
\textit{B', A$_1$}  \\
\textit{B}  \\
\textit{E$_F^{(1)}$ - E$_F^{(2)}$}
vides symmetry analysis and the phase classification of the excitonic correlated state. Section [VI] contains detailed description of the most symmetric phases, their properties are summarized in Table [I] Results are discussed in Section [VI]

II. TWO-LAYER HAMILTONIAN

Graphene is a gapless semiconductor with the Fermi surface consisting of two distinct points, \( K_+ \) and \( K_- \), called valleys. Near these Fermi points electrons have a linear dispersion \( E(p) = \pm vp \), with a velocity \( v \approx 10^6 \text{cm/sec} \) here \( p = |p|, \ p = k - K_+ \) is the momentum of an electron relative to the Fermi point. Using external gates, one can independently tune the carrier density in each of the two graphene flakes. Neglecting tunneling, the electrons in the two layer graphene system initially can be described with the Hamiltonian \( H_{2\text{layer}} = H_{s.p.} + H_{11} + H_{22} + H_{12} \), here the single particle part of the Hamiltonian is

\[
H_{s.p.} = \sum_{l,\xi,p,s} (s\nu p - E_F^{(l)})(a_{l,\xi,p,s}^{\dagger}a_{l,\xi,p,s}) \tag{1}
\]

the operators \( a_{l,\xi,p,s}^{\dagger} \) create (annihilate) an electron on the \( l = 1, 2 \) layer on the \( s = +/− \) conduction or valence band with momentum \( p = p(\cos \phi p, \sin \phi p) \), \( E_F^{(l)} \) and \( E_F^{(s)} \) are the Fermi energies, which correspond to charge carrier densities in the layers. The index \( \xi \) denotes 4 different pairs of spin projection (\( \uparrow, \downarrow \)) and valleys \( (K_+, K_-) \). In the Hamiltonian \( H_{2\text{layer}} \) the terms \( H_{11} \) and \( H_{22} \) can be taken into account the intra-layer interaction. These terms can be ignored in the following studies, provided that one uses a screened inter-layer interaction in the term \( H_{12} \). Hence in \( H_{12} \) we keep only those terms that contribute to the BCS mean field theory absorbing other contributions into a renormalization of the velocity and the Fermi energy in the single particle part of the Hamiltonian

\[
H_{12} = -\sum_{p,p',s,s'} V(|p - p'|) \frac{1 + ss' \cos(\phi p - \phi p')}{2} \sum_{\xi,\xi'} a_{l,\xi,p,s}^{\dagger} a_{l,\xi',p',-s'} a_{l,\xi,p',s'} a_{l,\xi',p,s} \tag{2}
\]

The scattering process, described by \( H_{12} \), is shown on Fig. 2. The function \( V(q) \) denotes a screened Coulomb interaction in the static limit \( V(q) = V(q, \omega \ll q) \). The factor \( [1 + ss' \cos(\phi p - \phi p')] / 2 \) in Eq. (2) reflects chiral properties of electrons related to the sublattice composition of electronic Bloch wave functions.44–46 These chiral properties of electrons result in the suppressed backwards scattering if an electron does not change the energy band upon scattering \( (ss' = +) \), otherwise \( (ss' = -) \) the electron can not forward-scatter.47

![FIG. 2. A typical transition which is described by \( \hat{H}_{12} \) in Eq. (2). Indices \( l = 1 \) or 2, \( \xi, p, s \) denote a layer, a pair of the spin projection and valley, a momentum of an electron and a conduction \( s = + \) or valence \( s = − \) band.](image)

![FIG. 3. The excitonic electron-hole bound state in the two-layer graphene. The left hand side of the figure shows the electron's spectrum in graphene layer 1 and 2. An electron on the Fermi surface in the layer 1 is shown as a fulfilled circle. Absence of an electron on the Fermi surface in layer 2 is shown as an empty circle. Closed line around both circles represents an excitonic pairing, which is developed due to a Coulomb interaction (shown as a wavy line). The right hand side of the figure shows the coincided Fermi circles in both layers at Fermi momentum \( p_F \).](image)
ternal conditions when excitonic correlations \[ \mathbf{H} \] can be developed. Thus, although the excitonic insulator state disappears when the symmetry \( n_{1z} = n_{2z} \) is violated by external gates, we show below that excitonic correlations can be restored by the in-plane magnetic field. Based on the detailed analysis of excitonic correlations in monolayer graphene in the in-plane magnetic field, which is done by Aleiner and co-authors,\(^{22}\) we show that the excitonic insulator state can exist in various phases in the two-layer graphene system.

In order to study phases of the excitonic insulator state at different external conditions, firstly we apply the standard mean-field approximation.\(^{40}\) We assume that the product of the operators \( a_{\uparrow 1.,\downarrow \cdot}^\dagger a_{\uparrow 2.,\downarrow \cdot} \) weakly deviates from its non-vanishing ground state average. We expand the interacting part \( \hat{H}_{12} \), Eq. (2), of the Hamiltonian \( \hat{H}_{2}\text{-layer} \) up to the linear order with respect to these small deviations and neglect constant terms. The mean field Hamiltonian of the system becomes

\[
\hat{H}_{\text{mf}} = \hat{H}_{s,p} + \sum_{\mathbf{p},s,\zeta,\zeta'} [a_{1.,\zeta,s}^\dagger \Delta_{\zeta',s}(\mathbf{p}) a_{2.,\zeta',s} + \text{H.c.}],
\]

where H.c. stands for “Hermitian conjugate”, and

\[
\Delta_{\zeta',s}(\mathbf{p}) = - \sum_{\mathbf{p}',s'} F_{\zeta',s'}(\mathbf{p}') V(|\mathbf{p} - \mathbf{p}'|) \times \frac{1 + ss' \cos(\phi_p - \phi_{p'})}{2}.
\]

Quantities \( \Delta_{\zeta',s}(\mathbf{p}) \) form the matrix \( \mathbf{\Delta} \) of the order parameter. Index \( \zeta \) denotes 4 different pairs of spin projections and valleys (\( \uparrow \uparrow, \uparrow \downarrow, \downarrow \uparrow, \downarrow \downarrow \)). Thus in the spin\( \otimes \)valley space the order parameter is given by the \( 4 \times 4 \) matrix \( \mathbf{\Delta} \) with matrix elements given by (5). For brevity we omit index \( s \) and momentum \( \mathbf{p} \) in the notation for the order parameter \( \mathbf{\Delta} \).

For further analysis it is convenient to rewrite the Hamiltonian (4) as follows: \( \hat{H}_{\text{mf}} = \sum_{\zeta,\zeta',\mathbf{p},s} \Psi_{\zeta,\zeta',\mathbf{p},s}^\dagger H_{\text{mf}}(\mathbf{p},s) \Psi_{\zeta,\zeta',\mathbf{p},s} \) where \( \Psi_{\zeta,\zeta',\mathbf{p},s} = (a_{1.,\zeta,s}^\dagger, a_{2.,\zeta',s}) \) \( ^T \), and

\[
H_{\text{mf}}(\mathbf{p},s) = \begin{pmatrix}
(svp - E_F) \mathbb{1} & \mathbf{\Delta} \\
\mathbf{\Delta}^\dagger & -(svp - E_F) \mathbb{1}
\end{pmatrix}.
\]

Here all elements of the matrix \( H_{\text{mf}} \) are \( 4 \times 4 \) matrices in the spin\( \otimes \)valley space: diagonal elements have structure of the identity matrix \( \mathbb{1} \) in this space, whereas \( \mathbf{\Delta} \) is given by some \( 4 \times 4 \) matrix, whose structure is identified in this paper for each phase of the excitonic correlated state. The matrix of the order parameter \( \mathbf{\Delta} \) describes the correlations between conduction/valence electrons in the layers 1 and 2 below a critical temperature \( T_c \).

Nevertheless, the phase classification can be made regardless of the value of the transition temperature \( T_c \).

Assuming that the excitonic insulator state can be observed in two-layer graphene system, we analyze the symmetry of the mean field Hamiltonian (4) and the order parameter \( \mathbf{\Delta} \). As a result the classification of all phases of the excitonic insulating state of the two-layer graphene system is presented in the next Section, and a detailed discussion of each phase is presented in Section IV.

IV. SYMMETRY ANALYSIS OF THE CORRELATED STATE

The analysis in this section is based on the idea of breaking of the initial symmetry of the Hamiltonian by the order parameter. The initial symmetry group \( G \) of the Hamiltonian \( \hat{H}_{2}\text{-layer} \) is formed by global unitary transformations of an electronic single-particle state in the 4-component spin\( \otimes \)valley space independently in the layer 1 and 2. These transformations are represented by independent matrices \( U^{(i)} \) and \( U^{(2)} \) in layer 1 and 2 respectively. Therefore the group \( G \) is given by the direct product of corresponding unitary groups \( U_4 \)

\[
G = U^{(1)}_4 \times U^{(2)}_4.
\]

Unitary group \( U^{(l)}_4, l = 1, 2 \), consists of \( 4 \times 4 \) unitary matrices \( U^{(l)} \) which perform transformations of electron’s operators in the \( l \)-th layer as follows:

\[
a_{l.,\zeta,s} \rightarrow \sum_{\zeta'} U^{(l)}_{\zeta\zeta'} a_{l.,\zeta',s}.
\]

Thus, as it is seen from Eqs. (4) and (5), under symmetry transformations (6) the order parameter \( \mathbf{\Delta} \) transforms as:

\[
\mathbf{\Delta} \rightarrow U^{(1)}_4 \mathbf{\Delta} U^{(2)}_4.
\]

This implies that the Hamiltonian of the system is not invariant under the action of the group \( G \) any longer. However for any fixed non-zero \( \mathbf{\Delta} \) there is always some subgroup \( H \) of the group \( G \), \( H \subset G, H \neq G \), such that all transformations from the group \( H \) do not transform \( \mathbf{\Delta} \), i.e. the order parameter \( \mathbf{\Delta} \) remains invariant:

\[
U^{(1)}_H \mathbf{\Delta} U^{(2)}_H = \mathbf{\Delta}.
\]

Such transformations \( U^{(1)}_H \) in layer 1 and \( U^{(2)}_H \) in layer 2 form a symmetry group \( H \)

\[
\begin{pmatrix}
U^{(1)}_H & 0 \\
0 & U^{(2)}_H
\end{pmatrix} \in H \subset G.
\]

Only transformations from the group \( H \) leave the ground state of the excitonic insulator invariant, i.e. only these
transformations leave the mean field Hamiltonian $H^{(1)}$ and $H^{(2)}$ invariant:

$$
\left( \begin{array}{cc}
U^{(1)}_{H} & 0 \\
0 & U^{(2)}_{H} \\
\end{array} \right) H_{mf}(p, s) \left( \begin{array}{cc}
U^{(1)}_{H} & 0 \\
0 & U^{(2)}_{H} \\
\end{array} \right) = H_{mf}(p, s).
$$

(12)

Thus the symmetry group $G$ of the initial uncorrelated normal ground state of the system is broken down to the symmetry group $H$ of the ground state of the excitonic insulator.

All transformations from $G$, which are not included in $H$, form the factor-space $G/H$. These transformations change the order parameter $\Delta$, however they do not change the energy of the corresponding ground state. Therefore the manifold of all matrices $\Delta$, which can be obtained by transformations from $G/H$, form a degeneracy space of the order parameter. Consequently the manifold of the correspondent ground states form a phase of the correlated state. It is important to notice, that all these ground states within the same phase are described by the same symmetry group $H$, which is a symmetry group of the order parameter. Therefore phases of a correlated state can be classified by the symmetry group $H$ and the degeneracy space of the order parameter.

The phase classification presented in this paper is also reminiscent of the classification of the various degeneracy spaces of the order parameter in liquid Helium-3.\cite{To68,To69,To70,To71} This classification principle was used in the determination of superconducting phases in nontrivial superconductors\cite{To66} and superfluid phases in liquid Helium-3.\cite{To68,To69,To70,To71}

In order to classify phases of the excitonic insulating state we are going to classify the degeneracy spaces of order parameters with the same symmetry. For this we consider the condition (10) and use the method of a singular value decomposition.\cite{To68} It allows us to represent an arbitrary matrix $\Delta$ as a product of a unitary matrix $\tilde{V}$, a diagonal matrix $D$ with real non-negative numbers on the diagonal, and another unitary matrix $V$. Applying the singular value decomposition to the order parameter at any given values of $s$ and $p$ we obtain

$$
\Delta = \tilde{V}_s^{†}(p)D_s(p)V_s(p).
$$

(13)

However, first of all, we notice that in the considered system, the lowest ground state energy is realized when matrices $V$, $\tilde{V}$ do not depend on momentum $p$ and index $s = \pm$. The reason for this is that in such a case in the expression for the ground state energy there is a cancellation of the product of matrices $V$ and $\tilde{V}$ ($\tilde{V}$ and $\tilde{V}^{†}$) into a unit matrix. Such a cancellation leads to the maximal negative contribution to the ground state energy, and therefore the energy of the ground state achieves its minimal value. If we assume that unitary matrices in Eq. (13) depend on momentum $p$ and index $s = \pm$, then unitary matrices at different momenta $p, p′$ and different indices $s, s′$ do not cancel each other, which increases the ground state energy comparatively to the previous case. Thus, we conclude, that in order to realize the lowest energy of the ground state, matrices $\tilde{V}$ and $V$ can not depend on the momentum $p$ and index $s$. Therefore the singular value decomposition of the matrix of order parameter becomes

$$
\Delta = \tilde{V}^{†}DV,
$$

(14)

where in the right hand side of the equation (14) only matrix $D$ depends on $p$ and $s$, but for brevity we omit these indices.

Second, all transformations from the group $G$, including those from the factor-space $G/H$, do not change the diagonal elements of the matrix $D$, but change matrices $\tilde{V}, V$ into any other unitary matrices. Thus if we introduce the notations

$$
\tilde{V}^{†} = U^{(1)}_{H}^{†}, \quad V^{†} = U^{(2)}_{H},
$$

(15)

then under the transformation (8)-(9) the order parameter will transform in the following way:

$$
\Delta = \tilde{V}^{†}DV \rightarrow \Delta′ = \tilde{V}^{†}DV′.
$$

(16)

Thus under this transformation the diagonal matrix $D$ does not change. Recall that the degeneracy space of the order parameter is obtained by acting on the order parameter $\Delta$ by all transformations from the group $G$ (here transformations from subgroup $H$ will not change the order parameter while remaining transformations from factor-space $G/H$ will create the degeneracy space of the order parameter). As long as only matrices $V$ and $\tilde{V}$ are changed by transformations from $G$, we obtain that the degeneracy space of the order parameter and a phase of the correlated state are determined only by the diagonal elements of the matrix $D$.

Finally, from the condition (10) we have found that all possible degeneracy spaces of the order parameter are classified by numbers of equal and different diagonal elements in matrix $D$. In the case of physically relevant phases there are additional restrictions on the diagonal elements of matrix $D$. Thus among all possible matrices $\Delta$ only physically relevant order parameters satisfy the self-consistency equation. For the phase classification it is sufficient to consider the BCS self-consistency equation for the order parameter. Diagonalizing the self-consistency equation by unitary matrices from Eq. (14), one obtains 4 equations for diagonal elements of the matrix $D$, each equation corresponds to some value of index $ζ$. These equations have the same structure and depend on the Fermi momentum $p_F$. If the Fermi momentum $p_F$ is the same for all types of electrons (for all indices $ζ$), then these 4 self-consistency equations are identical, and apart from a trivial zero solution they have the same non-zero solution. Hence in such situation in physically relevant phases the arbitrary diagonal element in matrix $D$ can be equal either to other non-zero diagonal elements,
or be equal to a zero. The application of the in-plane magnetic field in principle changes such a description because of Zeeman splitting. However, in the case where the Fermi energy is much greater than the Zeeman energy, $E_F \gg \epsilon_Z$, the magnetic field does not change the situation essentially as long as it is possible to neglect the difference between Fermi momenta for electrons with opposite spin projections in the self-consistency equations. Therefore four self-consistency equations on four diagonal elements of the matrix $D$ become approximately identical also when relatively small in-plane magnetic field ($\epsilon_Z \ll E_F$) is applied.

The non-zero solution of these equations is given by the gap function $g_s(p)$ which at the Fermi surface ($s = \pm, |p| = p_F$) determines a gap in the single-particle excitation spectrum. Thus we conclude that in all physically relevant phases the matrix $D$ in the single particle excitation (14) of the order parameter $\Delta$ consists of zeros or non-negative diagonal elements which approximately are equal to the gap function $g_s(p)$.

Substituting the obtained result into Eq. (14), we extract the gap function as a multiplier. Thus we conclude that the order parameter $\Delta$ in all physically relevant phases has a form

$$\Delta \cong g_s(p)\bar{V}^\dagger DV.$$  \hspace{1cm} (17)

Here the matrix $D$ is a diagonal matrix with 0 or 1 on the diagonal. The representation (17) becomes approximate in the case of the applied in-plane magnetic field with the condition $\epsilon_Z \ll E_F$. The dependence of the order parameter $\Delta$ on variables $s$ and $p$ is completely given by the function $g_s(p)$.

Having the matrix of the order parameter provided, the symmetry group $H$ is found from the equation (10). For this the matrix of the order parameter is represented as a single value decomposition (14). The constant matrices $V$ and $\bar{V}$ are absorbed into matrices $U_1^{(1)}$ and $U_2^{(2)}$ of the global symmetry transformations from the symmetry group $H$. Then the equation (10) connects two unitary matrices $VV^\dagger U_1^{(1)}$ and $\bar{V}V^\dagger U_2^{(2)}$ and the diagonal matrix $D$ with 0 or 1 on the diagonal. Thus the matrices $U_1^{(1)}$ and $U_2^{(2)}$ of transformations from the symmetry group $H$
are obtained.

V. PHASES

In this section we provide a detailed description of phases of excitonic insulator state in two-layer graphene system. Results of this section are summarized in Table I.

A. The $B$ phase.

Firstly we consider the situation when there is no external magnetic field and when the charge carrier densities in layers are the same $n_{1c} = n_{2c}$. In such case the symmetry group $G$ of the two-layer Hamiltonian of the system in normal state is given in Eq. (1). Under the mentioned conditions the Fermi circle in the conduction band in layer 1 coincides with the Fermi circle in the valence band in layer 2 due to the electron-hole symmetry in graphene. Hence the non-vanishing ground state average $F$, Eq. (3), can be formed by all species of electrons. Taking into account that the ground state with the lower energy is more stable, we consider the phase when excitonic correlations are developed among all species of electrons, the most stable ground state is characterized by the matrix $D$ in Eq. (17) with equal non-zero diagonal elements, i.e. $D$ is an identity matrix. As discussed in the previous section, this conclusion follows from the consideration of the self-consistency equation on the order parameter. Thus substituting $D = I$ into the equation (17) we obtain the following structure for the order parameter in spin-valley space:

$$\Delta = g_s(p)V, \quad V \in U_4. \quad (18)$$

Such a structure of the order parameter determines the symmetry group $H$ of the ground state and the degeneracy space of the order parameter, and consequently it determines the phase of the excitonic insulator.

The symmetry group $H$ of the ground state in the considered phase can be found as a group of all unitary transformations $U^{(1)}_H$, $U^{(2)}_H$ in layers 1 and 2, which leave the order parameter invariant, Eq. (10). Solving the condition (10) with the order parameter (18) we obtain matrices $U^{(1)}_H$ and $U^{(2)}_H$ of symmetry transformations in the layer 1 and 2 correspondingly. Thus, having $U^{(1)}_H$ and $U^{(2)}_H$,

$$U^{(1)}_H = U, \quad U^{(2)}_H = V^\dagger UV, \quad (19)$$

we can express an arbitrary element of the group $H$, Eq. (11), which transforms electron operators in layers 1 and 2 according to Eq. (8). Omitting indices $\zeta, p, s$ we have:

$$\begin{pmatrix} a_1 \\ a_2 \end{pmatrix} \rightarrow \begin{pmatrix} U & 0 \\ 0 & V^\dagger UV \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}. \quad (20)$$

Here the matrix $V$ is given by the fixed matrix of the order parameter (18). The unitary matrix $U$ is present in transformations in both layers. This means that the symmetry group $H$ of the ground state in the considered phase (18) consists of the combined transformations in both layers 1 and 2. The unitary group of the combined transformations in layers 1 and 2 is denoted as $U^{(1,2)}_4$,

$$\begin{pmatrix} U & 0 \\ 0 & V^\dagger UV \end{pmatrix} \in U^{(1,2)}_4 \equiv H. \quad (21)$$

The combined transformations from the group $U^{(1,2)}_4$ can also be described in terms of generators of these transformations. For this, each element of the group is written as the exponential function of the element of the group’s algebra

$$\begin{pmatrix} U & 0 \\ 0 & V^\dagger UV \end{pmatrix} = \exp\left[i\theta \Gamma_H \right] \in H, \quad (22)$$

where $\theta$ is a vector of real variables and a vector $\Gamma_H$ consists of generators of the group $H$. For the considered phase these generators are:

$$(\Gamma_H)_m \equiv \begin{pmatrix} \lambda_m & 0 \\ 0 & V^\dagger \lambda_m V \end{pmatrix}, \quad m = 0, 1, ..., 15. \quad (23)$$

In contrast to Eq. (23) transformations, which change the order parameter and create a degeneracy space $G/H$, are described by the following generators:

$$(\Gamma_{G/H})_m \equiv \begin{pmatrix} \lambda_m & 0 \\ 0 & -V^\dagger \lambda_m V \end{pmatrix}, \quad m = 0, 1, ..., 15. \quad (24)$$
Here the 2 × 2 block matrices \((\Gamma_H)_m\) and \((\Gamma_G/H)_m\) act in the space of layers 1 and 2, the matrix \(\lambda_m\) acts on the spin-valley basis in the layer 1 and matrices \(±V\lambda_mV^\dagger\) acts on the basis \(\Phi\) in layer 2. The spin-valley basis \(\Phi\) is the same in both layers. Matrices \(\lambda_m\) are 4×4 Hermitian traceless matrices of generators of transformations from the unitary group \(U_4\). The total number of generators \(\Gamma_{G/H}\), Eq. (23), equals to the dimension of the degeneracy space \(G/H\), \(\text{dim}(G/H)\). In the B phase \(\text{dim}(G/H) = \text{dim}(H) − \text{dim}(H) = 32 − 16 = 16\).

The electron operators in the second layer can be transformed by the matrix of the order parameter: \(Va_2 \rightarrow a'_2\), see Eqs. (18) and (23). Here the 2×2 matrix of the order parameter \(\delta E\) is again given by Eq. (27). It follows that the transformation from the group \(H\), in contrast to the transformation from \(G/H\), can be represented by identical transformations in both layers. There are identical transformations act by the same matrix \(U\) on the spin-valley basis \(\Phi\) in the layer 1 and on the transformed spin-valley basis \(V\Phi\) in the layer 2, Eq. (27) Hence the matrix of the order parameter (18) defines the relative unitary rotation of the spin-valley basis \(\Phi\) in layer 2 with respect to layer 1. It signifies the relative symmetry breaking: the ground state is not invariant under unitary transformations of the basis \(\Phi\) in one layer relatively to the basis \(\Phi\) in another layer. The basis \(\Phi\) in layer 1 is "locked" relatively to the basis \(\Phi\) in layer 2 by the matrix of the order parameter which defines the relative unitary rotation of one basis with respect to another.

Because of the presence of the relative symmetry breaking by the order parameter, the phase discussed here resembles the superfluid B phase in the liquid Helium-3. In the B phase the matrix of the order parameter is not degenerate. It means that all species of charge carriers develop excitonic correlations, therefore a single particle excitation spectrum is gapped.

The external conditions \((\varepsilon = 0, n_{1c} = n_{2c})\) for the B phase can be violated by an in-plane magnetic field or by external gates. However the excitonic correlations continue to exist in the B phase until the difference of Fermi energies \(g_{+}(p_F)/v\), where \(g_{+}(p_F)\) is a gap in a single-particle excitation spectrum in the B phase. Indeed such behavior can be seen, if one creates an asymmetry between charge carriers densities in layers, which can be expressed in terms of a shift \(\delta E_F > 0\) of Fermi energies: \(E^{(1)}_F = E_F + \delta E_F\), \(E^{(2)}_F = −E_F + \delta E_F\). Substituting these values to the mean field Hamiltonian (29) and finding its eigenvalues, one obtains two branches of excitation spectrum \(\varepsilon(±)(p) = \sqrt{(s\nu_p − E_F)^2 + g^2_{+}(p)} ± \delta E_F\). At values \(\delta E_F = g_{+}(p_F)\) one of branches of excitation spectrum becomes zero at \(s = ±, p = p_F\). At this situation the excitonic pairing stops being energetically favorable and the system appears in the normal state via a first order phase transition. In similar way, when Fermi circles for charge carriers with opposite spin projection are separated by \(2g_{+}(p_F)/v\) due to an in-plane magnetic field, excitonic correlations between charge carriers on these Fermi circles vanish. This fact is schematically shown in the phase diagram Fig.1: at the borders of the B phase in the phase diagram excitonic correlations are no longer energetically stable and the excitonic insulator state transforms into either a normal state or into another phase via a first order phase transition.

B. The \(A_0, A'_1, A'_2\) phases.

In this section we consider phases under the same external conditions as in B phase, thus the symmetry group \(G\) is again given by Eq. (7). We consider phases where order parameters are characterized by the degenerate matrices of rank \(r < 4\). In such phases only a part of electron species develop excitonic correlations, therefore the single particle excitation spectrum is gapless for certain species of electrons. The matrix of the order parameter can be chosen as follows, compare with Eq. (27):

\[\Delta = g_0(p)\tilde{V}\text{Diag}(a, b, c, 0)V, \quad \tilde{V}^\dagger, V \in U_4. \quad (25)\]

Here the diagonal matrix \(\text{Diag}\) determines the order parameter in phases, which are denoted as \(A'_0, A'_1, A'_2\); numbers \((a, b, c)\) are given by \((1, 1, 1)\) in \(A'_0\) phase, \((1, 1, 0)\) in \(A'_1\) phase and \((1, 0, 0)\) in \(A'_2\) phase. Using the transformed electron operators \(V\tilde{a}_1\) in the layer 1 and \(V\tilde{a}_2\) in the layer 2, see Eqs. (18) and (23), the self-consistency equation on the order parameter becomes diagonal, and only first \(r\) out of four equations for diagonal elements will have non-zero solutions. We assume that in the self-consistency equations we can use the screened interaction among charge carriers in the system in normal state. In this case self-consistency equations in \(A'_0, A'_1, A'_2\) phases are identical to self-consistency equations in B phase, therefore their non-zero solutions are given by the same gap function \(g_0(p)\).

Substituting the order parameter in the symmetry condition (31) one obtains matrices of symmetry transformations in layer 1 and 2, for example for \(A'_1\) phase one gets

\[U^{(1)}_H = \tilde{V}\begin{pmatrix} u & 0 & 0 \\ 0 & u' & 0 \\ 0 & 0 & u'' \end{pmatrix}V, \quad U^{(2)}_H = \tilde{V}\begin{pmatrix} u & 0 & 0 \\ 0 & u' & 0 \\ 0 & 0 & u'' \end{pmatrix}V. \quad (26)\]

where

\[u \in U_2^{(1,2)}, \quad u' \in U_2^{(1)}, \quad u'' \in U_2^{(2)}. \quad (27)\]

Here, similarly to the B phase, the 2 × 2 unitary matrix \(u\) determines the combined unitary rotations of the first two components of the spin-valley basis \(V\Phi\) in layer 1 and the first two components of the spin-valley basis \(V\Phi\) in layer 2. Therefore such phase is characterized by a partial relative symmetry breaking. Remaining matrices \(u', u'' \in U_2\) determine independent unitary rotations
of the other 2 components of corresponding spin-valley basis in layers. These other 2 components correspond to quasiparticle’s states which do not contribute to the excitonic condensation, their single particle excitation spectrum is gapless. Therefore only 2 out of 4 electron’s species are involved in the excitonic condensation. Because of this, such a phase is similar to the superfluid $A_1$ phase of liquid Helium-3, where paired states with only one spin projection $S_z = +1$ are present in the condensate. The $A_1$ phase of Helium-3 exists only in an external magnetic field, in order to underline the stability of the phase in the absence of the magnetic field we denote the phase discussed here by an additional prime, therefore it is denoted as the $A_1'$ phase of the excitonic insulator. Other phases with degenerate matrices of order parameters are denoted as $A_0'$ and $A_2'$.

In phases $A_0$, $A_1$, $A_2$ the number of non-zero diagonal elements in the diagonal matrix $\text{Diag}$ determines the rank $r$ of the symmetry group of combined unitary rotations, denoted as $U^{(1,2)}_r$. Zeros in the diagonal of the matrix $\text{Diag}$ correspond to electron states which do not develop excitonic correlations, and, therefore, these states can be unitary transformed independently in each layer.

Consequently the symmetry group $H$ for $A_0$, $A_1$, $A_2$ phases can be easily identified. For example, the symmetry group $H$ for $A_1'$ phase is following:

$$H = U_2^{(1)} \times U_2^{(1,2)} \times U_2^{(2)}.$$

The dimension of the degeneracy space is calculated as follows: for $A_0'$ phase $\dim[G/H] = 32 - 1 - 9 - 1 = 21$; for $A_1'$ phase $\dim[G/H] = 32 - 3 \times 4 = 20$; for $A_2'$ phase $\dim[G/H] = 32 - 9 - 1 - 9 = 13$.\n
C. The $B'$ phase.

In this section we consider the two-layer graphene system in an in-plane magnetic field. Our analysis is based on the comprehensive study by Aleiner and co-authors of the spontaneous symmetry breaking in graphene subjected to an in-plane magnetic field. When an in-plane magnetic field is applied, the Fermi circles for quasiparticles with different spin projections become separated due to a Zeeman splitting. Such splitting changes the symmetry group $G$, Eq. (17), of the initial Hamiltonian $H_{2\text{layer}}$ toward a direct product of 4 unitary groups $U_2$,

$$G = U_2^{(1)} \times U_2^{(1,1)} \times U_2^{(21)} \times U_2^{(21)}.$$  

Each of these $U_2$ groups transforms a valley space of electrons with corresponding spin projections in one layer, e.g. $U_2^{(1)}$ transforms electrons with spin up in layer 1.

The $B'$ phase can be obtained from the $B$ phase by the application of an in-plane magnetic field. Such magnetic field should be big enough to break the excitonic correlations in the $B$ phase and to split Fermi circles. Therefore a Zeeman energy $\epsilon_Z$ should be bigger than a gap in the excitation spectrum in the $B$ phase, $\epsilon_Z > g_+(p_F)$. In such a case, due to the initial equality of charge carrier densities $n_{1e} = n_{2h}$ in the $B$ phase, the two Fermi circles in layer 1 coincide with two Fermi circles in layer 2. Thus it leads to the appearance of two different Fermi circles in the system, Fig. 5. Consequently, the electron-hole pairs, which appear on different Fermi circles, have different properties: thus such electron-hole pairs have different spin projection, +1 or −1, Fig. 5. Also due to slightly different Fermi momenta, electron-hole pairs on different Fermi circles are characterized by slightly different gap functions. Hence in the spin-valley basis $\Phi$,

$$\Phi = (\uparrow K_+, \uparrow K_-, \downarrow K_+, \downarrow K_-),$$  

the order parameter has the following structure (compare with Eq. (18))

$$\Delta = \begin{pmatrix} 0 & g_s'(p) \nu \\ g_s'(p)\bar{\nu} & 0 \end{pmatrix}. $$

FIG. 5. Excitonic correlations in the two-layer graphene system with an in-plane magnetic field $h$ in the case of equal charge densities in layers $n_{1e} = n_{2h}$. Because of a Zeeman splitting $2\epsilon_Z$ there are two Fermi circles with radiuses $p_F \pm \epsilon_Z/v$. The absent electron with a particular spin projection is considered as a quasiparticle (hole) with an opposite spin projection.
Using this approximation, the symmetry group $H$ of the order parameter can be found from the condition (10).

As a result one obtains matrices $U_{1H}^{(1)}$ and $U_{1H}^{(2)}$ of the transformations (11) from the group $H$ in the layer 1 and 2 respectively (matrices are written in the basis $|\psi\rangle$ in each layer):

$$U_{1H}^{(1)} = \left( \begin{array}{cc} u & 0 \\ 0 & \bar{u} \end{array} \right), \quad U_{1H}^{(2)} = \left( \begin{array}{cc} \bar{v}^\dagger u v & 0 \\ 0 & v^\dagger u v \end{array} \right). \quad (31)$$

The corresponding electron operators are transformed as follows:

$$a_{1,+} \rightarrow ua_{1,+}, \quad a_{2,+} \rightarrow v^\dagger u v a_{2,+}, \quad u \in U_2^{(11,2 \times 1)}.$$

$$a_{1,+} \rightarrow \bar{u} a_{1,+}, \quad a_{2,+} \rightarrow \bar{v}^\dagger u^\dagger v^\dagger a_{2,+}, \quad \bar{u} \in U_2^{(11,2 \times 2)}.$$

The unitary $2 \times 2$ matrix $u$, Eq. (32), determines a subgroup of the group $H$, which consists of combined unitary rotations of valley space of spin up in layer 1 and electrons with spin down in layer 2: $u \in U_2^{(11,2 \times 2)}$. The unitary $2 \times 2$ matrix $\bar{u}$, Eq. (33), defines another corresponding subgroup of the group $H$, $\bar{u} \in U_2^{(11,2 \times 2)} \subset H$.

Hence in the phase considered here the symmetry group $H$ of the order parameter is given by direct product of two subgroups

$$H = U_2^{(11,2 \times 2)} \times U_2^{(11,2 \times 2)}.$$  

Using expressions for groups $G$ and $H$ in the $B'$-phase, we found that the degeneracy space $G/H$ is 8-dimensional, $\dim[G/H] = 4 \times 4 - 4 - 4 = 8$. It is also defined by the structure of the order parameter (30), i.e. here the degeneracy space is determined as a space of all possible unitary $2 \times 2$ matrices $v$ and $\bar{v}$. Because of the non-degenerate matrix of the order parameter, the single particle excitation spectrum in this phase is gapped.

Similarly to the $B$ phase, the excitonic correlations in the $B'$ phase cease to exist when the external conditions $(\epsilon_Z > g_1(p_F), n_{1e} = n_{2h})$ are perturbed, i.e. when Fermi circles in different layers are separated for the energy interval which is bigger than a double value of a gap in the single-particle excitation spectrum. Thus, in particular, in the schematic phase diagram Fig. (11 b) at the border of the $B'$-phase (when a symmetry $n_{1e} = n_{2h}$ of charge carriers densities is violated) the ground state of the system transforms to an uncorrelated normal ground state via the first order phase transition.

D. The $A_1$ phase.

In contrast to $B$ and $B'$ phases, where all species of charge carriers develop excitonic correlations, in this subsection we discuss another possible realization of the excitonic insulator state in the two-layer graphene system. We show that the excitonic correlated state can exist in the presence of an in-plane magnetic field and a specially chosen asymmetry in charge carriers densities in layers.

In order to achieve a necessary external conditions, firstly we consider the two-layer system without a magnetic field and with equal charge carrier densities in layers. Under such conditions the spectrum of electrons in both layers have only one Fermi circle at the Fermi momenta $p_{F1}$ and $p_{F2}$.

By changing the external gate voltages we create asymmetry between charge carrier densities in layers: $n_{1e} > n_{2h}$. Thus the Fermi circle in the layer 1 is situated at the momentum $p_{F1} + \delta p_F$, and the Fermi circle in the layer 2 is situated at the momentum $p_{F2} - \delta p_F$. Here $\delta p_F$ is the Fermi momentum in the case $n_{1e} = n_{2h}$ (i.e. in $B$ and $B'$ phases). It is assumed that the separation between Fermi circles is big enough to prevent the development of excitonic correlations. Keeping the chosen values of densities, we switch-on an in-plane magnetic field with such a magnitude that the Zeeman energy $\epsilon_Z$ is equal to the energy shift of each Fermi surface, $\epsilon_Z = v\delta p_F$, Fig. 6. The presence of an in-plane magnetic field signifies that the symmetry group $G$ in such case is the same as in $B'$ phase.

The external conditions mentioned above lead to the situation when only two out of four Fermi circles coincide: both the Fermi circle of electrons with spin up in the layer 1 (a Fermi circle with a smaller radius in the layer 1) and the Fermi circle of electrons with spin up in the layer 2 (a Fermi circle with a bigger radius in the layer 2) are situated at the same Fermi momenta $p_F$.  

![FIG. 6. Excitonic correlations in the $A_1$ phase. Starting from a zero magnetic field the asymmetry between charge carrier densities $n_{1e} > n_{2h}$ is created. In terms of Fermi energies it means $E^{(1)}_{F1} > |E^{(2)}_{F2}|$. A magnitude of an in-plane magnetic field is chosen such that a Zeeman energy $\epsilon_Z$ satisfies the condition: $E^{(1)}_{F1} - \epsilon_Z = |E^{(2)}_{F2}| + \epsilon_Z$, where $(E^{(1)}_{F1} - \epsilon_Z)/v$ is the radius of the Fermi circle for electrons with spin up in layer 1, and $(|E^{(2)}_{F2}| + \epsilon_Z)/v$ is a radius of the Fermi circle for electrons with spin up in layer 2. Both of these Fermi circles are situated at the same Fermi momentum $p_F$. Therefore two out of four Fermi circles coincide, leading to excitonic correlations only between half of electron’s species. Using the expression for the Fermi energies $E^{(1,2)}_{F1} = \pm v\delta p_F + v\delta p_{F2}$, $p_F > \delta p_{F2}$, where $\delta p_{F} = \sqrt{(n_{1e} - \sqrt{n_{2h}})/2}$, the condition on the Zeeman energy is the following: $\epsilon_Z = v\delta p_{F}$.](chart)
Thus electron-hole pairs are formed on these two coincided Fermi circles. Notice, that a total spin projection of such an electron-hole pair is equal to a zero in contrast to electron-hole pairs with spin projections +1 or −1 in the $B'$ phase. Fermi circles for electrons with spin down in both layers do not coincide with any other Fermi surfaces. Therefore corresponding electrons and holes are in a normal state (i.e. they do not participate in excitonic correlations), their single-particle excitation spectrum is gapless.

Thus for the phase discussed here only half of all electron species in the system develop excitonic correlations. It is reflected in the order parameter, whose structure in the basis (29) in both layers is given by the following expression:

$$\Delta = g_s(p) \begin{pmatrix} v & 0 \\ 0 & 0 \end{pmatrix}. \quad (35)$$

Here the gap function $g_s(p)$ is the same as in other phases due to the same Fermi momentum $p_F$ in the self-consistency equation and due to the approximation of the interaction among charge carriers in all phases by the screened interaction among charge carriers in the system in normal state. Similarly to the phases discussed previously, the unitary $2 \times 2$ matrix $v$ in the order parameter (35) determines a relative unitary rotation of the valley space of electrons with spin up in layer 2 relatively electrons with spin up in layer 1.

Solving the condition (10) with the order parameter (35) we found that transformations from the group $H$ are represented in layer 1 and 2 by following matrices $U_H^{(1)}$ and $U_H^{(2)}$ respectively (both matrices are written in the basis (29) in each layer):

$$U_H^{(1)} = \begin{pmatrix} u & 0 \\ 0 & u' \end{pmatrix}, \quad U_H^{(2)} = \begin{pmatrix} v^\dagger uv & 0 \\ 0 & u'' \end{pmatrix}. \quad (36)$$

Here matrices $u, u', u''$ are unitary $2 \times 2$ matrices. The matrix $u$ performs a combined unitary transformation of a valley space of electrons with spin up in both layers, in addition the valley space of electrons in layer 2 are rotated by the order parameter (35), compare with Eq. (32). The valley space of electrons with spin down is transformed by the unitary matrix $u'$ in layer 1 and by the unitary matrix $u''$ in layer 2 correspondingly:

$$a_{1,\uparrow} \rightarrow ua_{1,\uparrow}, \quad a_{2,\uparrow} \rightarrow v^\dagger uv a_{2,\uparrow}, \quad u \in U_2^{(1\uparrow,2\uparrow)}, \quad (37)$$

$$a_{1,\downarrow} \rightarrow u'a_{1,\downarrow}, \quad u' \in U_2^{(1\downarrow)}, \quad (38)$$

$$a_{2,\downarrow} \rightarrow u''a_{2,\downarrow}, \quad u'' \in U_2^{(2\downarrow)}. \quad (39)$$

Thus the group $H$ consists of the direct product of 3 unitary groups,

$$H = U_2^{(1\uparrow)} \times U_2^{(1\downarrow,2\uparrow)} \times U_2^{(2\downarrow)}. \quad (40)$$

Using the expressions for initial symmetry group $G$ and Eq. (40), we find that the degeneracy space $G/H$ in this phase is 4 dimensional, $\dim[ G/H ] = 16 - 3 \times 4 = 4$. It is determined by the manifold of all possible matrices $v \in U_2$ in the structure of the order parameter (35).

The subgroups $U_2^{(1\uparrow)}$ and $U_2^{(2\downarrow)}$ are present in both groups $G$ and $H$, the appearance of the correlated state does not change them. Therefore for the phase discussed here the initial symmetry is broken only partially. According to the similarities with the superfluid $A_1$ phase of liquid Helium-3 (i.e. that the phase described here exists in magnetic field and has a partial relative symmetry breaking), the phase discussed in this subsection was denoted as $A_1$ phase.

VI. RESULTS AND DISCUSSIONS

In the present paper we consider a two-layer graphene system where external gate voltage induces a finite density of electrons in one layer and holes in another. Assuming that the transition temperature $T_c$ towards excitonic insulator is high enough so that it can be observed, we classify phases of such correlated state. In order to obtain different excitonic correlations and therefore different phases we propose to use parallel to graphene layers magnetic field and perpendicular to graphene layers electric field.

Firstly we consider the Hamiltonian of the two-layer graphene system. We recognize that the ground state is characterized by a high symmetry group - the group of unitary rotations of spin-valley space of electrons in each layer independently. Below a transition temperature $T_c$ this symmetry is reduced by a non-zero order parameter towards a symmetry of the excitonic insulating ground state, which consists of electron-hole pairs with electrons on one layer and holes on another. Following the BCS theory of superconductivity, we identify the condition for such electron-hole pairing, determine the order parameter and build a BCS-like mean-field theory of the excitonic insulator. Analyzing a symmetry breaking of the initial ground state by the order parameter, we consider a condition that mutually determines the order parameter and the corresponding symmetry group of the excitonic insulator ground state. Using a singular value decomposition of the matrix of the order parameter, for each phase of the excitonic insulator we obtain a corresponding symmetry group of the ground state, a structure of the order parameter and its degeneracy space. The results of a phase classification of the excitonic insulator are shown in Table I, the most energetically stable phases are shown in the phase diagram, Fig. 1(b), and on Figs. 4. It is important to notice that the excitonic correlations in all phases discussed in this paper origin from the coincided Fermi surfaces at approximately the same Fermi momentum $p_F$ (we use assumption $E_F \gg \epsilon_2$, where the Fermi energy $E_F$ is determined in the system without a
magnetic field and with equal densities of charge carriers in layers \( n_{e} = n_{h} \). Thus assuming that the interaction among charge carriers is the same in all phases (i.e. that the effect of excitonic correlations on the screening of the interaction can be neglected\( ^{25} \)), we obtain that the energy gap in the single particle excitation spectrum in all phases is determined by the same self-consistency equation. Therefore the transition temperature \( T_{c} \) estimated from the self-consistency equation\( ^{25} \) should be the same for all phases.

At a temperature lower than the transition temperature \( T_{c} \), transitions between phases in the phase diagram are found to be of the first order. Phases of excitonic insulator have different properties: thus the electron-hole pairs in \( B \) phase have total spin projection +1 or −1, Fig. 5 whereas in the \( A_{1} \)-phase a total spin projection of an electron-hole pair is equal to zero, Fig. 6.

According to the number of estimations of the critical temperature in the considered system, the most optimistic estimation gives values of \( T_{c} \) close to a room temperature\( ^{22} \). However this estimation\( ^{22} \) does not take screening of the Coulomb interaction into account, explaining it by the assumption of the first order phase transition in the system. Some other estimations\( ^{25–27} \) point on the improbability of observation of excitonic condensation due to extremely low transition temperature \( \lesssim 1 \text{ mK} \) (\( T_{c} \approx 10^{-7} E_{F} \)). According to Refs.\( ^{25–27} \), the reason for low transition temperature lays in the effective screening of the Coulomb interaction by a big number \( N \) of species of electrons.\( ^{25,51} \) In the considered system \( N = 8 \), which is given by product of 2 valleys, 2 spin projections and 2 layers. Such large \( N \) increases screening and makes excitonic condensation not so effective, as in the monolayer graphene\( ^{22} \) and especially in the monolayer graphene in a magnetic field\( ^{29,53} \) where \( T_{c} \) can reach value up to \( 10^{-4} E_{F} \). However, recent investigations, based on a detailed treatment of the screened Coulomb interaction\( ^{28–30,54,55} \) and on a consideration of a multi-band pairing\( ^{31,54} \) or pairing with nonzero momentum\( ^{56} \) suggest that the transition temperature \( T_{c} \) can be sufficiently big for the experimental observation of the excitonic insulator in the considered system. Together with recent experimental realization of the two-layer graphene systems\( ^{25–37} \) it provides a hope that the phase diagram of the excitonic insulator, Fig. 1(b), under favorable conditions\( ^{57–60} \) will be observed experimentally.

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