Metastable electron–electron states in double–layer graphene structures

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The prototypical exciton model of two interacting Dirac particles in graphene was analyzed in [1] and it was found that in one of the electron–hole scattering channels the total kinetic energy vanishes, resulting in a singular behaviour. We show that this singularity can be removed by extending the quasiparticle dispersion, thus breaking the symmetry between upper and lower Dirac cones. The dynamics of an electron–electron pair are then mapped onto that of a single particle with negative mass and anisotropic dispersion. We show that the interplay between dispersion and repulsive interaction can result in the formation of bound, Cooper–pair–like, metastable states in double–layered hybrid structures.

Graphene continues to receive significant attention for its numerous intriguing interaction effects and transport properties [2–5]. Recent experiments with high–quality samples suggest the existence of non–trivial correlated phases in graphene, such as excitonic condensates in non–zero magnetic fields [6]. However, the zero–field condensate predicted theoretically [7–9] has not been observed [6] [10]. This has provoked significant interest in the archetypical two–body problem [1] [11] [12].

Most interesting properties of graphene are due to the existence of Dirac cones [13] located near two points in the Brillouin zone, \( K^+ \) and \( K^- \). Each cone hosts positive and negative energy states, each with linear dispersion \( \epsilon = \pm v_F p \), akin to electron and positron states in quantum electrodynamics. (Here \( v_F = 10^6 {\text{ms}}^{-1} \) is the Fermi velocity [14] and \( p \) is the magnitude of the momentum \( \mathbf{p} = (p_x, p_y) \).) The symmetry between positive and negative cones results in the compensation of total kinetic energy for two particles with opposite momenta. Thus, the two–particle states can be divided into dispersing (spanned by states where \( E \neq 0 \)) and non–dispersing (\( E = 0 \)) sectors. States in the non–dispersing sector have momentum–independent eigenvalues, and are therefore infinitely degenerate. However the linear dispersion is only accurate at low energies; higher order terms can at times reveal important physics hidden by the conical approximation.

The effect of dispersion on the excitonic physics can be seen if one considers Dirac particles interacting via the Coulomb potential, which scales as \( U(r) \sim \frac{Ze^2}{r} \). In the conical approximation, the kinetic energy scales as \( v_F p \sim \frac{h v_F}{2} \), du to the uncertainty principle. In the case of a single particle interacting with a static charge, Dirac vacuum reconstruction occurs when the potential energy dominates over kinetic energy: \( \frac{Ze^2}{r} > \frac{h v_F}{2} \). In the case of two carriers, however, a doubling of the relative velocity effectively doubles the critical charge for which collapse is possible: \( Z_C \gtrsim 2 \) [1]. This renders the strongly–interacting regime irrelevant for electron–hole physics.

For a weaker, quadratic dispersion (\( p^2 \sim \frac{h^2}{m^2} \)), however, bound states can form for arbitrarily small interaction strength. For this reason, the previously neglected contribution of non–dispersing states [1] [11] can be important for understanding the excitonic physics of graphene. To this end, the two–body problem was re–analyzed in [12] with the inclusion of trigonal–warping terms which preserve the symmetry between the two cones but do not lift the degeneracy. In this Letter we introduce quadratic momentum terms due to next–nearest–neighbour hopping which were ignored by [12] and show that this removes the degeneracy.

In this Letter, we show that Cooper–pair–like states can be formed in the subspace of non–dispersing two–particle states. The dynamics in this sector is governed by quadratic terms in the single–particle dispersion. Two such contributions are possible: an isotropic term due to next–nearest–neighbour hopping, \( \epsilon_I \propto p^2 \), and an anisotropic term due to trigonal–warping, \( \epsilon_A \propto p^2 \sin(3\phi_p) \), where \( \phi_p \) is the polar angle in momentum space defined by \( \tan(\phi_p) = \frac{p_\perp}{p_\parallel} \). We show that, depending upon the relative magnitudes of these two terms, two regimes are possible. When the isotropic contribution dominates, bound states can be formed; otherwise it is possible to form non–dispersing quasibound states which are approximately localized to the width of the potential, but can leak into the continuum without tunnelling. We calculate the binding energies of such states numerically, for a double–layer configuration, and discuss the decay rate due to coupling to the continuum of dispersing states.

We begin by analyzing the kinetic energy of two Dirac quasiparticles in graphene. In the linear–dispersion approximation, the dynamics of the pair is governed by the dispersion arising from the relative motion of its constituent particles, \( \hat{H}_L = v_F \sigma_1 \cdot \hat{p}_1 + v_F \sigma_2 \cdot \hat{p}_2 \), where \( \hat{p}_i \) and \( \sigma_i \) are the momentum and pseudospin operators, respectively, and subscripts denote the particle number. We focus on states with zero total momentum, such that
\[ p_1 = -p_2. \] The eigenstates of \( \hat{H}_I \) are given by:

\[
\begin{align*}
|1, \phi_p \rangle &= \frac{1}{\sqrt{2}} \left[ e^{-i\phi_p} |\uparrow\uparrow\rangle + e^{i\phi_p} |\downarrow\downarrow\rangle \right], \\
|2, \phi_p \rangle &= \frac{1}{\sqrt{2}} \left[ |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \right], \\
|3, \phi_p \rangle &= \frac{1}{\sqrt{2}} e^{i\phi_p} |\downarrow\uparrow\rangle - \frac{1}{2} e^{-i\phi_p} |\uparrow\downarrow\rangle + \frac{1}{2} \left[ |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right], \\
|4, \phi_p \rangle &= \frac{1}{\sqrt{2}} e^{-i\phi_p} |\uparrow\uparrow\rangle - \frac{1}{2} e^{i\phi_p} |\downarrow\downarrow\rangle + \frac{1}{2} \left[ |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right],
\end{align*}
\]

where the arrows represent the pseudospin configuration. The corresponding eigenvalues are \( E_{1,2} = 0 \) and \( E_{3,4} = \pm 2v_Fp \). The subspace spanned by \( |1, \phi_p \rangle \) and \( |2, \phi_p \rangle \) from Eq. (1) is the non–dispersing sector. Such states are formed by electron quasiparticles in opposite cones, with the same magnitude of momentum, so that the relative velocity of the pair vanishes. Similarly, the subspace spanned by \( |3, \phi_p \rangle \) and \( |4, \phi_p \rangle \) forms the dispersing sector, in which the velocities are opposite. In the absence of interactions, all states in the non–dispersing sector are infinitely degenerate.

We notice that this degeneracy is lifted if the symmetry between the upper and lower cones is broken, e.g., by a small band curvature. We extend the kinetic energy by quadratic terms compatible with the symmetries of the honeycomb lattice (see, e.g., [15, 16]). We write the single–particle kinetic energy in the form:

\[
\hat{H}_K z,j = v_F \sigma_j \cdot p_j - \frac{p_j^2}{4m*} + \tau \mu (p_{x,j} + ip_{y,j})^2 \sigma_{+,j} + \text{h.c.}
\]

where \( j \) is the particle number, \( p_j^2 = p_{x,j}^2 + p_{y,j}^2 \), \( \sigma_{+,j} = \frac{1}{2} (\sigma_{x,j} + i\sigma_{y,j}) \), \( \tau = \pm 1 \) for an electron in the \( K \)–valley (determining the sign of the trigonal–warping) and h.c. denotes the Hermitian conjugate. The second term in Eq. (2) is invariant under all two–dimensional rotations, and arises microscopically from contributions due to the hopping of electrons from one atom to its next–nearest–neighbour, giving \( m* = \frac{\hbar^2}{2m \tau} \), where \( \tau \) is the next–nearest–neighbour hopping parameter [17].

The third term (including h.c.) is invariant under rotations by 120°. This term represents trigonal–warping, and originates from nearest–neighbour hopping, expanded to second order in momentum, so that \( \mu = \frac{3\hbar^2}{8m} \) [15, 17]. It has been shown by a variety of different approaches that \( \tau \) and \( \tau' \) have the same sign, however there is a disagreement on the precise value of \( \tau' \). \textit{Ab initio} calculations [17] give the range 0.02\( t' \leq 0.2t \), while cyclotron resonance [14], quantum capacitance [19] and polarization–resolved magnetospectroscopy [20] measurements have produced \( t' = 0.04t, 0.11t \) and 0.14t respectively. The full two–particle kinetic energy is \( \hat{H}_{1,2} = \hat{H}_I + \hat{H}_2 \), and so \( -m* \) plays the role of a two–particle reduced mass due to the \( -\frac{p^2}{2m} \) term which arises when \( \hat{H}_{1,2} \) is written explicitly, with the corresponding range of values \( 0.7 \leq \frac{m*}{m} \leq 7.5 \). This implies that the isotropic kinetic energy term is negative definite, which will be shown to be of crucial importance to the spectrum of two–particle states later in this Letter.

To examine the dynamics in the non–dispersing sector, we restrict the two–particle Hamiltonian to this subspace. We explicitly treat two distinct cases: direct pairs (when both particles are in the same valley) and indirect pairs (opposite valleys). All states \( |1, \phi_p \rangle \) and \( |2, \phi_p \rangle \) are annihilated by the operator \( (\sigma_1 - \sigma_2) \cdot p \). Calculating the matrix elements of the kinetic energy we find the effective Hamiltonian:

\[
\hat{H}_{1,2}^{\text{eff}} = \begin{bmatrix}
-\frac{p^2}{2m*} & \tau_{1,2}\mu p^2 \sin(3\phi_p) \\
\tau_{1,2}\mu p^2 \sin(3\phi_p) & -\frac{p^2}{2m*}
\end{bmatrix},
\]

where the rows and columns correspond to states \( |1, \phi_p \rangle \) and \( |2, \phi_p \rangle \) and \( \tau_{1,2} = \tau_1 + \tau_2 \) (i.e. \( \tau_{1,2} = 0 \) for indirect pairs, \( \tau_{1,2} = \pm 2 \) for direct pairs).

To understand the dynamics of pairs described by the kinetic energy terms in \( \hat{H}_{1,2} \), let us first consider the simplest case of indirect pairs, where the electrons are in opposite valleys. Due to the opposite signs of \( \tau_1 \) and \( \tau_2 \) at the two Dirac points, the contribution of trigonal–warping vanishes in this configuration, and the only remaining kinetic term is \( -\frac{p^2}{2m} \). The dynamics of the interacting pair is therefore described by the Hamiltonian

\[
\hat{H}_I = -\frac{p^2}{2m} + U(r),
\]

for states with configuration \( |2, \phi_p \rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow \rangle + |\downarrow\uparrow \rangle) \), where \( U(r) \) is the potential energy. The Hamiltonian \( \hat{H}_I \) describes the motion of a particle with negative effective mass \( -m* \) in the external potential \( U(r) \). To diagonalize it, we notice that

\[
\hat{H}_I \text{ describes the motion of a particle with a positive mass } m* \text{ in the potential } -U(r).
\]

In two–dimensional systems, an arbitrarily weak attractive potential exhibits at least one bound state at negative energies for massive particles [21]. It follows, therefore, that \( \hat{H}_I \) will exhibit positive energy bound states as \( U(r) \) is repulsive. This interesting property is a direct consequence of the negative definite kinetic energy of the pair, the dynamics of which can be compared to the motion of a hole–like state near the top of the valence band in a semiconductor: the repulsive potential due to a negatively charged impurity is perceived as an attraction due to negative band curvature. In the real space picture, two electrons with opposite momenta reside in different cones and have nearly the same velocities. The repulsive force tends to increase the momentum of one electron, and decrease the momentum of the other. Due to the negative dispersion term, this decreases the velocity of the first particle, and increases the velocity of the other, reducing the distance between them. Unlike conventional bound states, these positive eigenstates are metastable. Formation of regular (electron–hole) excitons is prohibited in this regime.

Since the effective mass \( -m* \) is only about five times larger than the free electron mass, the binding can be
quite strong. For the example of a repulsive Coulomb interaction, \( U(r) = \frac{e^2}{\epsilon_g r} \), the problem reduces to the two-dimensional hydrogen atom \([22]\). \( \epsilon_g = \epsilon_s + \frac{N_\pi \omega^2}{8\epsilon_0 r} \) is the intrinsic dielectric constant of graphene embedded in a material with dielectric constant \( \epsilon_s \) \([23]\). For single (double) layer graphene, the number of Fermion species is \( N = 4 \) \((N = 8)\). The highest energy level is given by \( E_1 = \frac{2 \pi^2 \epsilon_s}{\epsilon_g \zeta^2} \), which results in \( E_1 \sim 5\text{eV} \) and Bohr radius \( a_B \sim 0.5\text{A} \) for \( t' = 0.1t \). At such short distances, the low–energy approximation to the graphene band structure is not valid \([24]\), and our solution is not self–consistent. More importantly, the dynamics of particles at such high energies is affected by Pauli blocking due to the Dirac sea. For the bound state to be observable, the relevant phase space domain must be free from other particles. This can be achieved by, e.g., gating, if the bound state energy is well below 1eV. Two–particle states with smaller binding energies can be realized in double–layered structures where the electrons in opposite layers are separated vertically by a dielectric spacer of thickness \( d \). Hexagonal boron nitride \((\epsilon_s = 3–4)\) spacers have been experimentally shown to electrically isolate parallel graphene layers at a thickness of 4 atomic layers \((d = 1.3\text{nm}) \([25]\). This suppresses the \( \frac{1}{d} \) singularity, yielding smaller binding energies. For a rough estimate of the binding energy in the double well we approximate the potential as \( V(r) \sim \frac{1}{\epsilon_g} \sqrt{r^2 + d^2} \), where the dielectric constant is taken for \( N = 8 \). The spectrum of the resulting shallow well can be found in the harmonic approximation, assuming \( r \ll d \). The energy of the highest bound state is \( E_0 = -\hbar \omega + \frac{\epsilon_s^2}{\epsilon_0 \zeta} \), where \( \omega = \sqrt{\frac{\epsilon_0 \zeta \epsilon_s \omega}{\epsilon_g \zeta^2}} \) is the oscillation’s angular frequency near the potential maximum. For example, \( t' = 0.1t \) gives binding energy \( E_0 = 48\text{meV} \).

To analyze the case of the direct pair with anisotropic dispersion, we derive its effective Hamiltonian in momentum space. The potential energy \( V(r) \) is represented by a non–local operator proportional to it's Fourier transform \( V_{\mathbf{p},\mathbf{p}'} = \hat{V}(\mathbf{p} - \mathbf{p}') \), while the kinetic energy terms are given by Eq. \([3]\). Restricting the potential energy to the non–dispersing sector requires some care due to a non–trivial overlap between non–dispersing states with different momenta: \( \langle 1, \phi_{\mathbf{p}'} | 1, \phi_{\mathbf{p}} \rangle \). For the case of direct interactions, the \((\mathbf{p}, \mathbf{p}')\) block of the Hamiltonian matrix takes the form:

\[
\hat{H}_{\mathbf{p},\mathbf{p}'} = \delta_{\mathbf{p},\mathbf{p}'} \hat{H}_{1,2}^{\text{eff}} + \hat{V}_{\mathbf{p},\mathbf{p}'} \begin{bmatrix} \cos(\phi_{\mathbf{p}} - \phi_{\mathbf{p}'}) & 0 \\ 0 & 1 \end{bmatrix},
\]

where \( \hat{H}_{1,2}^{\text{eff}} \) is given by Eq. \([3]\). In the absence of inter–particle interaction the eigenvalues of this matrix are given by the kinetic energy terms: \( \epsilon_{g2}^{(2)} = -2\mu p^2 [\eta + \sin(3\phi_{\mathbf{p}})] \), where we have introduced the anisotropy parameter \( \eta = \frac{6t'}{\pi} \) which is not physically tunable (uncertainty in the value of \( t' \) gives a range of possible values \( 0.12 \leq \eta \leq 1.2 \)). Depending on the value of \( \eta \), the kinetic energy is either negative–definite \((\eta > 1)\) or sign–indefinite \((\eta < 1)\).

We proceed by numerically diagonalizing the Hamiltonian given by Eq. \([4]\) using the complete interlayer interaction from \([9]\). Although it is assumed that the relevant phase space domain is free of other particles to avoid the effect of Pauli blocking, we will treat the case of screening at half–filling \((\rho_f = 0)\) as a first approximation. Indeed, the dielectric contribution to the screening giving rise to \( \epsilon_g \) occurs at scales smaller than the Fermi wavelength \((\lambda_f)\), and is most important in the realistic limit of \( \lambda_f \ll d \). To visualize the resulting wavefunctions, we calculate the local density of states (LDOS), \( \nu(\epsilon, x, y) = \sum_{\mathbf{p}} \delta(\epsilon - \epsilon_{\mathbf{p}}) |\psi_{\mathbf{p}}(x, y)|^2 \), where \( x \) and \( y \) are the components of the in–plane separation and \( n \) labels the eigenstates. In the isotropic regime, \( \eta > 1 \), as is evidenced by Fig. \( 2a \) for \( \eta = 1.1 \), there is a formation of distinct, highly localized, bound states at \( \epsilon_1 = 45\text{meV} \), \( \epsilon_2 = 30\text{meV} \) and \( \epsilon_3 = 27\text{meV} \). We note that the energy of the highest bound state, \( \epsilon_1 \), is very similar to the value predicted in the harmonic approximation above. This validates the further use of such an approximation in the calculations of the transition rates that are to follow. At negative energies there is a low intensity continuum of unbound states, which are only weakly coupled to the bound states due to the symmetry of the Hamiltonian, resulting in their large lifetimes.

In the anisotropic regime \((\eta < 1)\), there are six \textit{easy} axis angles, defined by the relation \( \sin(3\phi_0) = -\eta \), along which the dispersion is effectively suppressed despite the broken conduction–valence symmetry. By concentrating the wavefunction along these axes, one constructs a state qualitatively similar to the non–dispersing solutions \( \psi(r) \propto \delta(r - r_0) \). The energies of these states (Fig. \( 2b \)) follow the profile of the interaction potential, \( \epsilon \approx U(r_{\phi}) \). Further, a negative energy state dragged into the positive continuum by the interaction potential can decay by changing its pseudospin configuration rather than by tunnelling through a barrier. This is wholly due to the sign–indefinite kinetic energy.

In our analysis so far we considered only the non–dispersing sector. Coupling to the dispersing sector could lead to the decay of the metastable states found above, which we now analyze via Fermi’s Golden Rule. Note that the decay is only allowed into positive energy states, \(|3, \phi_{\mathbf{p}}\rangle\), due to energy conservation. The coupling between these two sectors occurs via trigonal–warping and potential energy terms, due to the non–trivial overlap between states with different momenta. The relevant matrix elements of the Hamiltonian are \( H_{1,3} = -\frac{\nu_3}{\sqrt{2}} V_{\mathbf{p},\mathbf{p}'} \sin(\phi_{\mathbf{p}} - \phi_{\mathbf{p}'}) \) and \( H_{2,3} = \nu_2 \hbar p^2 \sin(3\phi_{\mathbf{p}}) \delta_{\mathbf{p},\mathbf{p}'} \), where \( H_{i,j} = \langle j, \phi_{\mathbf{p}'} | \hat{H} | i, \phi_{\mathbf{p}} \rangle \). The matrix element due to the interaction potential, \( H_{1,3} \), vanishes by symmetry if \( \psi_{\mathbf{p}}(\mathbf{p}) \) is an \( s \)–state. The
Therefore, $\Gamma = \psi$ state of the 2D harmonic oscillator, as discussed above:

$$2$$

Rule, $\Gamma = \psi$ approximate the final state wavefunctions by plane waves,

$$2$$

and inter–valley scattering for particles with velocity density of final states in the absence of spin–flipping.

$$p$$ momenta satisfy conservation laws:

$$E_i = p_i \cdot v_i$$

The smallness of $p_i$ results in a small matrix element which is proportional to $p^2$. The decay rate is given by Fermi’s Golden Rule, $\Gamma = \frac{2\pi}{\hbar} |M_{f,i}|^2 \rho_f$, where $M_{f,i} = \langle \psi_f | H | \psi_i \rangle$ is the transition matrix element and $\rho_f = \frac{|E|}{8\pi\hbar^2 v_i^2}$ is the density of final states in the absence of spin–flipping and inter–valley scattering for particles with velocity $2v_F$.[13]. To calculate the kinetic contribution, we approximate the final state wavefunctions by plane waves, $\psi_f \sim \delta(p' - p_f)$, and the initial state by the ground state of the 2D harmonic oscillator, as discussed above: $\psi_i(p) \sim \frac{1}{p_0} \exp(-\frac{p^2}{2p_0^2})$. The transition rate for $H_{2,3}$ is therefore $\Gamma = \frac{\mu^2 v_f^2 E_0}{2\hbar v_i f_{0}^2} \sim 10^{-8} \frac{E_0}{\hbar}$. This suggests only weak coupling to the continuum, justifying our consideration of the non–dispersing sector independently.

In this Letter we have analyzed the problem of an isolated electron–electron bound state, however further work remains to be done in considering the many–body effects, the simplest example of which is interaction screening. Recall that in order to prevent Pauli blocking the system must be gated so that the Fermi energy exceeds the binding energy. This would introduce a non–zero density of states and metallic screening of the interaction. If the inter–particle separation is larger than the screening radius, the binding energy would be renormalized, but the bound state would not be destroyed: for a massive particle in an arbitrarily weak potential, at least one bound state exists in two dimensions[21]. The states we considered here have positive energy and therefore do not represent energy minima, however once created they have long lifetimes. One way to create them is by coupling the aforementioned graphene structure to a superconductor. As the metastable states are akin to Cooper pairs, this would lead to a giant enhancement of the proximity effect, which has been observed in graphene recently[26–28].

In conclusion, we have studied the problem of interacting electron–electron pairs in hybrid graphene–dielectric–graphene structures. We have shown that, in the isotropic regime, the conduction–valence band asymmetry allows the formation of a new kind of Cooper–pair–like bound state in the sector spanned by eigenfunctions which are dispersionless in the conical approximation.

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