Writing Sample 2:
Bilayer Graphene

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The following writing sample is a brief analytical investigation into the superconducting properties of bilayer graphene. The central focus is to provide theoretical justification for potentially novel mechanism of superconductivity in transition metal intercalated bilayer graphene. Our approach is to propose a Hamiltonian that includes a local spin-orbit coupling term that interacts with the itinerant electrons responsible for the magnetic properties. The addition of these terms into the Hamiltonian result in characteristic features in the band structure and can be easily identified. The proposed model allows for comparison with previous models of superconductivity in bilayer graphene through numerical and experimental investigations. In summary, a technical analysis was performed on material graphene to investigate its superconducting properties. This analysis allows us to identify, characterise, and predict the properties of the material under various external conditions. By understanding the origin of the material properties we can confidently determine the constraints under which the material can be applied. This type of analysis is of significant utility for evaluating the threat that a weapon or technology may present to US interests.

I. INTRODUCTION

A Dirac material is characterized by a non-Bravais lattice structure with a real-space inversion center and a Dirac cone present in the electronic structure; produced through a coupling of the momentum and the (pseudo)-spin[13]. In general, a non-Bravais structure can be described by a lattice and a basis; a typical example being the honeycomb lattice, whose two sublattice (2SL) structures are connected through an inversion center. The electrons in the 2SL structure provide two separate modes which couple through the (pseudo)-spin, this coupling between the two electron modes of opposite spin-direction, produces a crossing (Dirac node) in the allowed energy levels. Furthermore, the coupling restricts the back-scattering of fermions by imposing an additional symmetry constraint into the system, e.g., the requirement of a simultaneous (pseudo)-spin flip.

It is typical for the existence of a Dirac node at the level crossing to result in a large reduction of the available phase space for low-energy excitations in Dirac materials[13]. In other words, the dimensionality of the set of points in momentum space where zero-energy excitations are well defined is smaller for Dirac materials relative to normal metals. This controlled reduction of phase space through the addition of symmetries is a hallmark characteristic of Dirac materials and the ability to control the formation of Dirac points in the excitation spectrum by manipulating system symmetries is largely applicable to the field of spintronics[7]. The low-energy quasi-particle excitations will determine the response of the system to external probes and the symmetries that control these excitations will, in general, be different for different materials. In essence, because the characteristic properties of a Dirac material are a direct consequence of the spectrum of Dirac quasi-particles, designing a multifunctional Dirac material requires an examination of various degrees of freedom, e.g., orbital, lattice, spin, etc.

The most common Dirac material is graphene, where the honeycomb lattice of carbon atoms provides the two sublattice structure for the preservation of the multimode crossover. In graphene the same Dirac-like spectrum as in superconducting and superfluid materials can be an intrinsic property of the band structure, ultimately stemming from the crystalline order of the material. Each unit cell of the hexagonal Bravais lattice contains two carbon atoms which can be decomposed into two sublattices A and B. The atoms in sublattice A are surrounded by three nearest-neighbor atoms in sublattice B and vice-versa. When only nearest-neighbor coupling is considered the lattice becomes bipartite, and in the vicinity of the Fermi level, all electronic states emerge from the out-of-plane carbon p_z orbitals. These form π-bonds with neighboring atoms and the resulting π-band energy levels can be described by a tight-binding Hamiltonian

$$\hat{H} = -t \sum_{<i,j>} a_i^\dagger b_j + a_j^\dagger b_i \tag{1}$$

where t is the nearest-neighbor hopping term[13]. The operators a_i and b_i will annihilate electrons in the p_z orbital of atomic carbon at lattice point i, for sublattice A and B respectively. To motivate the relation between symmetry and energy the above Hamiltonian is rewritten in momentum space[13]

$$H(\vec{k}) = \begin{pmatrix} 0 & \xi(\vec{k}) \\ \xi^*(\vec{k}) & 0 \end{pmatrix} \tag{2}$$

where $\xi(\vec{k})$ is the dispersion. The off diagonal elements result from hopping terms between atoms in sublattice A and B. The hopping amplitudes depend on the location of the surrounding atoms, and can be expressed in terms of nearest-neighbor coupling vectors $\vec{\delta_l}$, $\xi(\vec{k}) = -t \sum_l e^{i\vec{\delta_l}\cdot\vec{k}}$, where the sum on l runs over the three nearest-neighbors. The dependence on the dynamic hopping terms $\vec{\delta_l}$ in the Hamiltonian, encodes an intimate connection between the systems' inherent symmetry and lattice geometry.

To illustrate this, consider the expanded form of the dispersion relation at the points K and $K' = -K$ in the Brillouin zone,

$$\xi(\mathbf{K}) \propto e^{i\vec{\delta_1}\cdot\mathbf{K}} + e^{i\vec{\delta_2}\cdot\mathbf{K}} + e^{i\vec{\delta_2}\cdot\mathbf{K}} \tag{3}$$

where $\mathbf{K} = \pm K$. Here the three hopping amplitudes sum with phase factors $e^{i(\vec{\delta}_{1,2,3})\mathbf{K}}$ and reduce to 1, $e^{+2\pi i/3}$ and $e^{-2\pi i/3}$, which destructively interfere to zero. The energy spectrum $\epsilon(\vec{k}) = \pm|\xi(\vec{k})|$, at the degenerate points K and $K' = -K$, defines the location of the Fermi level with energies $\epsilon(\pm K) = 0$. An expansion of the energies near the Fermi surface reveals four low-energy excitations; two electron spin-degenerate cones for each of the two Brillouin points K and K'

$$H(\pm K + \vec{q}) = \hbar v_F \begin{pmatrix} 0 & q_x \pm iq_y \\ q_x \mp iq_y & 0 \end{pmatrix}; \quad \vec{q} = (q_x, q_y) \tag{4}$$

The pseudospin in the above Hamiltonian corresponds to the sublattice degree of freedom and is fundamentally different from the real electron spin. Furthermore, the Dirac particles in graphene originate from the electronic band structure and are considered charged quasiparticles. For brevity, the emergence of Dirac fermions in graphene maintains two requirements;

1. the destructive interference of the three partial hopping amplitudes at $\xi(\vec{k} = K)$ connecting each atom in sublattice A with its three nearest-neighbors in sublattice B and vice versa,

2. the absence of a Hamiltonian term proportional to σ_z.

The inversion symmetry of the sublattices,

$$I : H(\vec{k}) = \sigma_x H(-\vec{k})\sigma_x$$

and the time-reversal symmetry in the absence of a magnetic field,

$$T : H(\vec{k}) = H^*(-\vec{k})$$

couple together to produce

$$TI : H(\vec{k}) = \sigma_x H^*(\vec{k})\sigma_x,$$

which forces terms proportional to σ_z to vanish, or risk breaking the coupled symmetries[13].

II. BOSONIC DIRAC MATERIALS

Since the Dirac equation focuses on the coupling of momentum to pseudospin, one goal is to investigate the interaction of magnetic degrees of freedom in the Dirac system. A challenge for magnetic Dirac materials is the integration of the magnetic degrees of freedom in accordance with the intrinsic degrees of freedom of the system. The introduction of magnetic moments can change the overall sublattice structure, obstructing the existing symmetries. Typically, the placement of magnetic impurities into a Dirac material will remove the Dirac cone by producing a gap between the electron bands through a breaking of inversion symmetry[13]. Transition metals exhibit intrinsic magnetic properties and the introduction of transition metals into graphene has been shown to lower the Fermi energy of graphene due to the presence of d-orbital electrons[4]. A complimentary phenomena emerging from magnetic interactions is the existence of magnetic Dirac modes; bosonic excitations induced by nearest-neighbor interactions of free electron spins. These Dirac magnons emerge naturally as a consequence of the spatial magnetic structure due to the breaking of inversion symmetry in particular magnetic phases[5]. Recently, it has been shown that granular superconductors on a honeycomb lattice can realize bosonic Dirac materials (BDM); and support two component superfluid states with collective phase oscillations that exhibit Dirac points in the spectrum at Brillouin points K and K'[1].

It is also been long known that graphene exhibits superconducting states in both pure, doped and more recently, twisted bilayers[3,12]. The microscopic origins of superconductivity in Ca-intercalated bilayer graphene were shown to stem from phonon-mediated processes[10]. These results motivated an investigation into the superconducting properties of transition-metal intercalated bilayer graphene through magnetic exchange[8]. A honeycomb lattice of AA-stacked graphene sheets, intercalated with centered 3d-transition metals, was studied using Density Functional Theory (DFT)[8]. The electronic band structure showed hybridization of carbon p-bands with transition-metal (TM) d-bands and the formation of an electron pocket below the Fermi energy at the Γ-point[8]. The characteristic of d-band dipping below the Fermi level is similar to what is observed in superconducting intercalated alkaline and alkali metals in bilayer graphene[9], indicative of potential superconductivity in TM intercalted bilayer graphene. The previous summary has motivated us to consider following; magnetic pairing of electrons as a mechanism for superconductivity, freedom to distinguish between superconductivity that would occur between parallel and anti-parallel spins, and to indicate whether stronger or weaker J would favor conductivity, as potentially novel directions for future research.

In a previous analysis, Boyko et al. introduced an effective magnon model through a Heisenberg spin-lattice Hamiltonian,

$$H_s = -\sum_{<ij>} J_{ij}\mathbf{S}_i^A \cdot \mathbf{S}_j^B \tag{5}$$

where the sum is over nearest-neighbor interactions and the spins $\mathbf{S}_i^A$ and $\mathbf{S}_j^B$ correspond to sublattices A and B, respectively[2]. Considering homogeneous ferromagnatic interactions $J_{ij} = J > 0$, a Holstein-Primokoff transfor-

mation gives the Hamiltonian for the effective magnon model

$$H_{FM} = \sum_i (\epsilon_A a_i^\dagger a_i + \epsilon_B b_i^\dagger b_i) - J\sqrt{\mathbf{S}_A \mathbf{S}_B} \sum_{<ij>} (a_i^\dagger b_j + a_i b_j^\dagger) - 3JN\mathbf{S}_A\mathbf{S}_B \tag{6}$$

The first term on the right hand side of equation (6) corresponds to the on-site magnon energies, i.e., $\epsilon_{A(B)} = 3J\mathbf{S}_{B(A)} + g\mu_\mathbf{B}\mathbf{B}$; while the second term represents the coupling between the sublattices A and B. A Fourier transformation using

$$a_i = \frac{1}{\sqrt{N}} \sum_k a_k e^{ik\cdot r_i} \tag{7}$$

and

$$b_j = \frac{1}{\sqrt{N}} \sum_k b_k e^{ik\cdot r_j} \tag{8}$$

results in the momentum space representation of the magnon Hamiltonian

$$H_{FM} = \sum_i (\epsilon_A a_k^\dagger a_k + \epsilon_B b_k^\dagger b_k) + [\phi(\mathbf{k}) a_k^\dagger b_k + \phi(\mathbf{k}) a_k b_k^\dagger] - 3JN\mathbf{S}_A\mathbf{S}_B. \tag{9}$$

Here the structure factor is given in terms of the nearest-neighbor coupling vectors δ_i

$$\phi(\mathbf{k}) \;=\; -J\sqrt{\mathbf{S}_A\mathbf{S}_B} \sum_i e^{i\mathbf{k}\cdot\delta_i}$$

where for a honeycomb lattice $\delta_1 = \frac{a}{2}(\sqrt{3}, 1)$, $\delta_2 = -\frac{a}{2}(\sqrt{3}, -1)$, and $\delta_3 = -a(0, 1)$ where a is the lattice parameter[11]. The resulting eigenenergies are given by

$$E_\pm(\mathbf{k}) = \frac{1}{2}[\epsilon_A + \epsilon_B \pm \Omega(\mathbf{k})] \tag{10}$$

where $\Omega^2(\mathbf{k}) = \Delta^2 + 4|\phi(\mathbf{k})|^2$ and $\Delta = \epsilon_A - \epsilon_B = 3J(\mathbf{S}_A - \mathbf{S}_B)$. For ferromagnetic structures the lattice anisotropy is zero, i.e., $\mathbf{S}_{A(B)} = S$, which implies the difference in lattice spin energies is $\Delta = 0$. This yields a linear dispersion at the degeneracy points $\pm\mathbf{K} = \frac{2\pi}{3a}\left(\frac{\sqrt{3}}{3}, 1\right)$,

$$\Omega(\mathbf{k}) = |\phi(\mathbf{k})| \Longrightarrow \tag{11}$$

$$\phi(\mathbf{k} \pm \mathbf{K}) \approx v_J k e^{\pm i(\pi/3 - \psi)} \tag{12}$$

where $v_J = \frac{3aJS}{2}$ and $\psi = \arctan(k_y/k_x)$[11]. Building on the above analysis, Boyko et al. found the magnon band for the ferromagnetic lattice quantitatively similar to the fermionic band structure of graphene, with characteristic Dirac point degeneracy at K and K', and a linear energy-momentum dispersion relation[5]. In addition, the introduction of asymmetry in the lattice, via ferrimagnetism, was shown to open a gap at the Dirac points with a size proportional to the difference between spins in the two sublattices, $3J|S_A - S_B|$. However, if the system supports spin-orbit coupling, a non-negligible chiral next-nearest-neighbor interaction is introduced in the effective model. This chiral term stems from the linearly decaying Dzyaloshinski-Moriya interaction and can be destructive towards magnons by introducing parity-breaking contributions, potentially breaking the sublattice symmetry[5].

III. HAMILTONIAN MODEL

The electronic properties of the intercalated bilayer graphene lower the energy of the fermionic Dirac cone and produce an electron pocket below the Fermi energy at the Γ-point. This behavior indicates the potential for hybridization between the p and d bands of carbon and transition metals (TM), respectively. On the other hand, magnetic properties in compounds arise from spin and orbital angular momentum contributions of the free electrons, but to first-order, the spin-orbit coupling is typically ignored. However, in cases of low-spin complex and heavy-ion transition metals, the spin-orbit coupling cannot be ignored. This leads us to consider the following, in the presence of strong carbon p-band and TM d-band hybridization; do the magnetic properties of spin-orbit coupling preserve the unique linear dispersion found in monolayer graphene, and to what extent are bosonic Dirac modes the relevant driving mechanism? In what follows, we provide a rough sketch of the framework we intend to develop. The idea here is to construct a two term Hamiltonian whose only imposed constraint is the preservation of $z \to -z$. We propose an effective Hamiltonian for magnons and look for signature changes in the band structure caused by the spin-orbit coupling (SOC). In particular, the existence of any non-trivial topological band structure as in the Haldane model[6].

Does the crossing of the d band with the Fermi surface result from strong hybridization between TM d-bands and carbon p-bands, and is this crossing a reliable characteristic of superconductivity in TM intercalated bilayer graphene? We approach this question by studying a spin Hamiltonian for spin-orbit coupled ferromagnets on a honeycomb lattice. Since we are trying to model graphene electrons it is natural to consider the simplest electronic system for itinerant electrons on a honeycomb lattice with spin-orbit coupling, the $\mathbf{k}\cdot\mathbf{p}$ theory

$$H_e = v_F(\pm\tau_x p_x +_y p_y) \pm \Delta_{so}\tau_z\sigma_z \tag{13}$$

where the $\pm$ indicates the two valleys and the $\tau_{x,y,z}$ are Pauli matrices associated with the sublattice degrees of freedom. In addition, there should be an exchange term, that accounts for coupling between localized spins and spin density of the Fermi sea. We write this as

$$J\mathbf{S}_i \cdot \mathbf{s}(\mathbf{R_i}) \tag{14}$$

where

$$\mathbf{s}(\mathbf{R}_i) = \sum_{\mathbf{r}} \delta^{(2)}(\mathbf{r} - \mathbf{R}_i)\sigma(\mathbf{r}) \tag{15}$$

and $\sigma(\mathbf{r})$ is the vector of Pauli matrices. Expanding the exchange term to lowest order in the exchange constant gives the RKKY Hamiltonian for the localized spins

$$H_{RKKY} = J^2 \sum_{i,j} \chi_{\alpha,\beta}(\mathbf{R}_i, \mathbf{R}_j) S_i^{\alpha} S_j^{\beta} \tag{16}$$

The magnetic interactions are mediated by the itinerant electrons and we can write the proposed Hamiltonian as

$$H = H_e + J\mathbf{S}_i \cdot \mathbf{s}(\mathbf{R}_i) \tag{17}$$

The first term describing dynamics of surface electrons, while the second describes the coupling between the localized spins and the spin density of the metal at the position of a magnetic impurity. This can be thought of as deposited magnetic impurities on metals with strong spin-orbit coupling.

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