Possible high-temperature superconducting state with a $d+id$ pairing symmetry in doped graphene

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Motivated by a suggestion in our earlier work [G. Baskaran, Phys. Rev. B **65**, 212505 (2002)], we study electron correlation driven superconductivity in doped graphene where on-site correlations are believed to be of intermediate strength. Using an extensive variational Monte Carlo study of the repulsive Hubbard model and a correlated ground state wave function, we show that doped graphene supports a superconducting ground state with a $d+id$ pairing symmetry. We estimate superconductivity reaching room temperatures at an optimal doping of about 15%–20%. Our work suggests that correlations can stabilize superconductivity even in systems with intermediate coupling.

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I. INTRODUCTION

Attempts to achieve room-temperature superconductivity has lead to the discovery of "high-T_c" superconductivity in *layered* materials such as cuprates,[1,2] organic superconductors, MgB_2 (Ref. 3) and most recently, Fe pnictides[4] family. Graphene, a semimetal, is a single atom thick layer of carbon net.[5–7] A newly discovered method to cleave and isolate single or finite number of atomic layers of graphene, its mechanical robustness, and novel electrical properties has caught the attention of the scientific and nanotechnology community. Undoped graphene is a semimetal and does not superconduct at low temperatures. However, on "doping optimally," if graphene supports high-T_c superconductivity, it will make graphene even more valuable from basic science and technology points of view.

GB (Ref. 8) suggested the possibility of high temperature superconductivity in graphene based on an effective phenomenological Hamiltonian that combined band theory and Pauling's idea of resonating valance bonds (RVB).[9] The model predicted a vanishing T_c for undoped graphene, consistent with experiments. However, for doped graphene superconducting estimates of T_c's were embarrassingly high. Very recently Black–Schaffer and Doniach[10] used GB's effective Hamiltonian and studied graphitic systems and found that a superconducting state with $d+id$ symmetry to be the lowest energy state in a mean field theory. The mean field theory also predicts a rather high value of the optimal T_c. Other authors have studied possibility of superconductivity based on electron-electron and electron-phonon interactions.[7,11–14] While there is an encouraging signal for high T_c superconductivity in the phenomenological GB model, it is important to establish this possibility by the study of a more basic and realistic model. Since the motivation for GB model arose from a repulsive Hubbard model, here we directly analyze this more basic repulsive Hubbard model that describes low energy properties of graphene. We construct variational wave functions motivated by RVB physics, and perform extensive Monte Carlo study incorporating crucial correlation effects. This approach, which has proved to be especially successful in understanding the ground state of cuprates, clearly points to a superconducting ground state in doped graphene. Further support is obtained from a slave rotor analysis, which also includes correlation effects. Our estimate of superconducting T_c is of the order of room temperatures, and we also discuss experimental observability of our prediction of high temperature superconductivity in graphene.

In the following section we discuss the Hamiltonian for graphene and the possibility of superconductivity. Sec. III contains the description and results of our variational calculations, and this is followed by the slave rotor mean-field analysis in Sec. IV. We discuss our results in Sec. V.

II. GRAPHENE: HAMILTONIAN ETC

Low-energy electrical and magnetic properties of graphene are well described[15] by a tight binding Hubbard model defined on a honeycomb lattice with a single $2p_z$ orbital per carbon atom,

$$\mathcal{H}_H = -\sum_{\langle ij\rangle} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + \text{H.c.} + U\sum_i n_{i\uparrow} n_{i\downarrow}. \quad (1)$$

Here, i labels atomic sites, $c_{i\sigma}$ is an annihilation operator for an electron with spin σ at site i, $n_{i\sigma}$ is the number operator at site i of σ spin electrons, $t \approx 2.5$ eV is the hopping matrix element and $U \approx 6$ eV is the onsite Hubbard repulsion. The unique band structure of the above model leads to a "Dirac cone" type of spectrum for electron motion close to two points in the Brillouin zone, giving rise to a density of states that varies linearly with energy near zero energy (half filling).

The Hubbard U is about half the $p\pi$ free bandwidth, and this places graphene in an intermediate or weak coupling regime. Based on this one is tempted to conclude that electron correlations are not important. Nonetheless electron correlations are known to be important in finite $p\pi$ bonded planar molecular systems such as benzene, naphthane, anthracene, caronene etc, all having nearly the same value

for quantum chemical parameters t and U.[15] One of the consequences of this is that the first excited spin-0 state lies above the first excited spin-one state by more than 1 eV. There is a predictable consequence of this large singlet-triplet splitting: graphene can be viewed as an end member of a sequence of planar $p\pi$ bonded system; this has been also suggested to have a new spin-1 collective mode spectrum, as a consequence of finite U.[16]

Pauling[9] was the first to recognize dominance of singlet correlation between two neighboring $p\pi$ electrons in the ground state. He argued that doubly occupied or empty $2p_z$ orbitals (polar configurations) are less important because of electron-electron repulsion in the $2p_z$ orbital. Pauling, thus, ignored polar configurations. Once we ignore polar fluctuations (states with double occupancy) and consider a resonance among the nearest-neighbor valence bond configurations we get the well known RVB state. However, such a Hilbert space actually describes a Mott insulating state rather than a metal. Experimentally, undoped graphene is a broad band conductor, albeit with a linearly vanishing density of states at the Fermi energy.

To recover metallicity in Pauling's RVB theory, GB combined the broad band feature of $p\pi$ electrons with Pauling's real space singlet (covalent) bonding tendency and suggested[8] a low-energy phenomenological model for graphene:,

$$\mathcal{H}_{\mathrm{GB}} = -\sum_{\langle ij\rangle} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + \mathrm{H.c.} - J\sum_{\langle ij\rangle} \mathbf{b}^{\dagger}_{ij}\mathbf{b}_{ij}, \tag{2}$$

where $\mathbf{b}^{\dagger}_{ij}=\frac{1}{\sqrt{2}}(c^{\dagger}_{i\uparrow}c^{\dagger}_{j\downarrow}-c^{\dagger}_{i\downarrow}c^{\dagger}_{j\uparrow})$ creates a spin singlet on the $i-j$ bond. $J(>0)$ is a measure of singlet or valence bond correlations emphasized by Pauling, i.e., a nearest-neighbor attraction in the spin singlet channel. In the present paper we call it as a "bond singlet pairing" (BSP) pseudo potential. The parameter J was chosen as the singlet triplet splitting in a 2 site Hubbard model with the same t and U, $J=[(U^2+16t^2)^{1/2}-U]/2$. As U becomes larger than the bandwidth this psuedopotential will become the famous superexchange characteristic of a Mott insulator. As shown in,[8] this model predicts that undoped graphene is a "normal" metal. The linearly vanishing density of states at the chemical potential engenders a critical strength J_c for the BSP to obtain a finite mean field superconducting T_c. The parameter J for graphene was less than the critical value, and undoped graphene is not a superconductor despite Pauling's singlet correlations. Doped graphene has a finite density of state at the chemical potential and a superconducting ground state is possible. Black-Schaffer and Doniach[10] confirmed GB's findings in a detailed and systematic mean field theory and discovered an important result for the order parameter symmetry. They found that the lowest energy mean field solution corresponds to $d+id$ symmetry, an unconventional order parameter, rather than the extended-s solution. The value of mean field T_c obtained was an order of magnitude larger than room temperature!

Although results of the mean-field theory are encouraging, it is far from certain that the superconducting ground state is stable to quantum fluctuations. In particular, the GB Hamiltonian does not include U, which inhibits local number fluctuations. In the more basic Hubbard model, a superconducting state will suffer further quantum mechanical phase fluctuations since U inhibits local number fluctuations. The key question therefore is: does the singlet promotion arising out of the local correlation physics strong enough to resist the destruction of superconductivity arising out of quantum phase fluctuations induced by U?

III. VARIATIONAL GROUND-STATE CALCULATION

To investigate the possibility of superconductivity in doped graphene we construct a variational ground state and optimize it using variational quantum Monte Carlo (VMC).[17] The ground state we construct is motivated by the mean-field theory of the GB model,

$$\mathcal{H}^{MF}_{\mathrm{GB}} = \sum_{\boldsymbol{k}} [-t\varepsilon(\boldsymbol{k})(a^{\dagger}_{\boldsymbol{k}\sigma}b_{\boldsymbol{k}\sigma}) + \mathrm{H.c.}] - \mu_f \sum_{\boldsymbol{k}} (n^a_{\boldsymbol{k}\sigma} + n^b_{\boldsymbol{k}\sigma}) - \sum_{\boldsymbol{k}} [\Delta(\boldsymbol{k})(a^{\dagger}_{\boldsymbol{k}\uparrow}b^{\dagger}_{-\boldsymbol{k}\downarrow} - a^{\dagger}_{\boldsymbol{k}\downarrow}b^{\dagger}_{-\boldsymbol{k}\uparrow}) + \mathrm{H.c.}], \tag{3}$$

where $a_{\boldsymbol{k}\sigma}, b_{\boldsymbol{k}\sigma}$ are electron operators on the A and B sublattices, $\boldsymbol{k}$ runs over the Brillouin zone of the triangular Bravais lattice, $\boldsymbol{a}_\alpha, \alpha=1,2,3$ are vectors that go from an A site to the three nearest B sublattice sites. The free electronic dispersion is determined by the function $\varepsilon(\boldsymbol{k})=\Sigma_\alpha e^{i\boldsymbol{k}\cdot\boldsymbol{a}_\alpha}$, and the superconducting gap function $\Delta(\boldsymbol{k})=\Sigma_\alpha \Delta_\alpha e^{i\boldsymbol{k}\cdot\boldsymbol{a}_\alpha}$. The $d+id$ symmetry motivated by the meanfield solution[10] provides $\Delta_\alpha=\Delta e^{[i2\pi(\alpha-1)]/3}$ where Δ is the "gap parameter," μ_f is a "Hartree shift."

Starting from this mean field theory, we construct a BCS state $|\mathrm{BCS}\rangle$ by diagonalizing the kinetic energy part of Eq. (3) results in two bands $c^{\dagger}_{\boldsymbol{k}\sigma}$ and $d^{\dagger}_{\boldsymbol{k}\sigma}$. The superconducting pairing term splits into intra-band pairing and interband pairing. The latter being unimportant at zero temperature can be dropped giving

$$\mathcal{H}_{MF} = \sum_{\boldsymbol{k}} [E^{+}(\boldsymbol{k})c^{\dagger}_{\boldsymbol{k}\sigma}c_{\boldsymbol{k}\sigma} + E^{-}(\boldsymbol{k})d^{\dagger}_{\boldsymbol{k}\sigma}d_{\boldsymbol{k}\sigma}] + \sum_{\boldsymbol{k}} [\Delta_d(\boldsymbol{k})(c^{\dagger}_{\boldsymbol{k}\uparrow}c^{\dagger}_{-\boldsymbol{k}\downarrow} - d^{\dagger}_{\boldsymbol{k}\uparrow}d^{\dagger}_{-\boldsymbol{k}\downarrow}) + \mathrm{H.c.}]$$

where, $\Delta_d(\boldsymbol{k})=\Sigma_\alpha\Delta_\alpha\cos[\boldsymbol{k}\cdot\boldsymbol{a}_\alpha-\varphi(\boldsymbol{k})]$, $\varphi(\boldsymbol{k})=\arg\varepsilon(\boldsymbol{k})$. Thus, the variational ground state can be written as $|\mathrm{BCS}\rangle=\Pi_{\boldsymbol{k},\alpha=\pm}(u^{\alpha}_{\boldsymbol{k}}+v^{\alpha}_{\boldsymbol{k}}e^{\dagger}_{\boldsymbol{k}\alpha\uparrow}e^{\dagger}_{-\boldsymbol{k}\alpha\downarrow})|0\rangle$ where $e^{\dagger}_{\boldsymbol{k}+\sigma}=c^{\dagger}_{\boldsymbol{k}\sigma}$, $e^{\dagger}_{\boldsymbol{k}-\sigma}=d^{\dagger}_{\boldsymbol{k}\sigma}$ and the BCS coherence factors $\frac{v^{\alpha}_{\boldsymbol{k}}}{u^{\alpha}_{\boldsymbol{k}}}=-\alpha\frac{\Delta_d(\boldsymbol{k})}{E^{\alpha}_{\boldsymbol{k}}+\sqrt{(E^{\alpha}_{\boldsymbol{k}})^2+|\Delta_d(\boldsymbol{k})|^2}}$. To treat system containing fixed number N of electrons (this corresponds to a hole doping of $1-N/L$), the variational wave function is projected onto a fixed number subspace $|\mathrm{BCS}\rangle_N$.

Our candidate ground state $|\Psi\rangle$ is now a state with a Gutzwiller-Jastrow factor[18,19] g

$$|\Psi\rangle = g^{\mathcal{D}}|\mathrm{BCS}\rangle_N \tag{4}$$

where $\mathcal{D}=\Sigma_i(n^a_{i\uparrow}n^a_{i\downarrow}+n^b_{i\uparrow}n^b_{i\downarrow})$ is the operator that counts the number of doubly occupied sites. The wave function (4) with partial Gutzwiller projection has three variational parameters, the gap parameter Δ, the Hartree shift μ_f, and the Gutzwiller–Jastrow factor g. The ground-state energy $\langle\Psi|\mathcal{H}_H|\Psi\rangle$ is calculated using quantum Monte Carlo

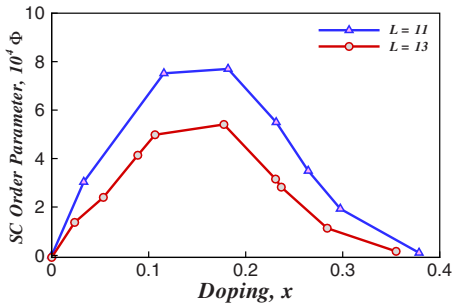

FIG. 1. (Color online) Doping dependence of superconducting order parameter Φ as obtained from VMC calculation of the Hubbard model on a honeycomb lattice for $U/t=2.4$ for periodic boxes with $L=13,11$.

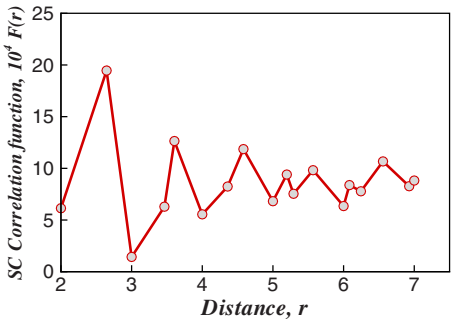

FIG. 2. (Color online) Dependence of superconducting correlation function $F(r)$ on distance r as obtained from VMC calculation of the Hubbard model on a honeycomb lattice for $U/t=2.4$, $x=0.2$.

method,[17] and is optimized with respect to the variational parameters.

We monitor superconductivity by calculating the following correlation function using the optimized wave functions:

$$F_{\alpha\beta}(\boldsymbol{R}_i-\boldsymbol{R}_j)=\langle \mathbf{b}^{\dagger}_{ia_\alpha}\mathbf{b}_{ja_\beta}\rangle, \tag{5}$$

where $\mathbf{b}^{\dagger}_{ia_\alpha}$ is the *electron* singlet operator that creates a singlet between the A site in the ith unit cell and the B site connected to it by the vector $\boldsymbol{a}_\alpha$ (this is just $\mathbf{b}^{\dagger}_{ij}$ defined earlier with a minor change of notation). The superconducting order parameter, off-diagonal long range order (ODLRO), is

$$\Phi=\lim_{|\boldsymbol{R}_i-\boldsymbol{R}_j|\to\infty} F(\boldsymbol{R}_i-\boldsymbol{R}_j). \tag{6}$$

where $F(\boldsymbol{R}_i-\boldsymbol{R}_j)=\Sigma_\alpha F_{\alpha\alpha}(\boldsymbol{R}_i-\boldsymbol{R}_j)$. All results we show in this paper are performed on lattices with 13^2 unit cells.

The superconducting order parameter Φ as a function of doping, calculated for physical parameters corresponding to graphene, obtained using the optimized wave function is shown in Fig. 1 for two different system sizes. Remarkably, a "superconducting dome," reminiscent of cuprates,[20] is obtained and is consistent with the RVB physics. The result indicates that undoped graphene had no long range superconducting order consistent with physical arguments and mean-field theory[10] of the phenomenological GB Hamiltonian. Interestingly, the present calculation suggests an "optimal doping" x of about 0.2 at which the ODLRO attains a maximum.[21] These calculations strongly suggest a superconducting ground state in doped graphene.

We now further investigate the system near optimal doping in order to estimate T_c. Figure 2 shows a plot of the order parameter function $F(r)$ as function of the separation r. The function has oscillations up to about six to seven lattice spacings and then attains a nearly constant value. From an exponential fit one can infer that the coherence length ξ of the superconductor is about six to seven lattice spacings. A crude estimate of an upper bound of transition temperature can then be obtained by using results from weak coupling BCS theory, using $k_bT_c=\frac{1}{1.764}\frac{\hbar v_F}{\pi\xi}$. *Conservative estimates give us* $k_bT_c=\frac{t}{50}$, i.e., T_c is about twice room temperature. Evidently, this is an upper bound, and an order of magnitude lower than the mean-field theory estimates of Black-Schaffer and Doniach.[10] Further improvement of our estimate of T_c becomes technically difficult. It is interesting to compare these results with those obtained in a Hubbard model on a square lattice that captures cuprate physics. In this latter case, a similar estimate of the coherence length ξ is about two to three lattice spacings;[20] however, the hopping scale is nearly a magnitude lower and the estimate of T_c is about $T_{\text{Room}}/2$. Again, this provides further support for the possibility of high temperature superconductivity in graphene. We also note that the superconducting transition in the realistic system will be a Kosterlitz–Thouless type transition.

IV. MEAN-FIELD SLAVE ROTOR ANALYSIS

It is interesting to see if the results of the VMC can be reproduced in a "simple mean-field type" analysis that accounts for the presence of U. The recently developed slave rotor[22] technique is useful to study an intermediate coupling regime that allows for number fluctuations at a site, and has been used successfully, in the context of cuprates, to study the t-J-U model.[23] We adopt a similar approach and use a t-J-U model, which is equivalent to introducing a Hubbard repulsion U in the GB Hamiltonian.

For the t-J-U model,

$$\mathcal{H}=-\sum_{ij\sigma} t_{ij}c^{\dagger}_{i\sigma}c_{j\sigma}+U\sum_i n_{i\uparrow}n_{i\downarrow}+J\sum_{\langle ij\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j, \tag{7}$$

the idea is to decompose electronic degree of freedom into spinon and charge (rotor) degrees of freedom (DOFs)—$c^{\dagger}_{i\sigma}=f^{\dagger}_{i\sigma}e^{-i\theta_i}$, where $f^{\dagger}_{i\sigma}$ creates a spin σ at site i and $e^{\pm i\theta_i}$ are the ladder operators for electron density, n. In this representation, t-J-U model takes the form,

$$\mathcal{H}_{SR}=-\sum_{ij\sigma} t_{ij}f^{\dagger}_{i\sigma}f_{j\sigma}e^{-i(\theta_i-\theta_j)}+\frac{U}{2}\sum_i n^{\theta}_i(n^{\theta}_i-1)+J\sum_{\langle ij\rangle}\boldsymbol{S}^f_i\cdot\boldsymbol{S}^f_j. \tag{8}$$

The total number of particles at any site has to be unity to remove unphysical states in the expanded Hilbert space. At mean field level, the spinon (rotor) DOF can be integrated out to give a Hamiltonian in the rotor (spinon) space. The two Hamiltonians are coupled via the kinetic energy term. They are solved self-consistently using standard techniques.[23]

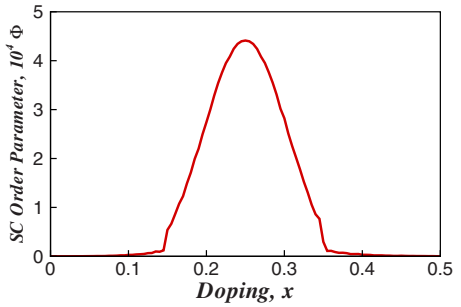

FIG. 3. (Color online) Doping dependence of superconducting order parameter as obtained from Slave rotor mean field theory of the t-J-U model.

The ODLRO calculated from the slave rotor analysis is shown in Fig. 3, and bears a remarkable qualitative resemblance with the VMC result, i.e., there is an optimal doping that produces superconductivity. This result, again, supports the possibility of high temperature superconductivity in doped graphene.

V. DISCUSSION

It is important to ask about the possibility of competing orders that could overshadow superconductivity at optimal doping that we have found. Honerkamp[24] has addressed this issue by means of a functional renormalization group study of a general Hamiltonian on the honeycomb lattice. He finds that in the regime of physical parameter corresponding to graphene, the system appears to flow toward a $d+id$ superconducting state as the temperature is lowered.

It is interesting to contrast the superconductivity in cuprates with that in graphene. In the case of cuprates (doped Mott insulators), Bloch electrons in the entire Brillouin zone are affected by the Hubbard U which is larger than the bandwidth. To this extent all electrons participate in superexchange or singlet bond formation. Consequently correlation hole development is complete, i.e., an electron at a given site with an up spin manages to avoid an electron with down spin on its site completely, at low-energy scales. Whereas, in the broad band graphene, only those Bloch electrons in the range of energy scale of U around chemical potential are affected by the on site repulsion. Since this scale is about half the bandwidth, about one half of the electrons are involved in singlet bond formation in the ground and low energy states. Consequently correlation hole development is not complete. The key point is that there is a sufficiently enhanced singlet correlation, compared to free Fermi gas, to be able to support superconductivity induced by the on site Coulomb repulsions in optimally doped graphene. This heuristic picture is supported by the variational Monte Carlo analysis that we have presented in this paper.

Our prediction of high temperature superconductivity raises some obvious questions. Intercalated graphite can be viewed as a set of doped graphene layers that have a strong 3 dimensional electronic coupling. Maximum T_c obtained in these systems is around 16 K.[25] Systems such as CaC_6 has a doping close to optimal doping that we have calculated. Why is the T_c so low? On the other hand, superconducting signals with a T_c around 60 K and higher have been reported in the past in pyrolitic graphite containing sulfur.[26,27] A closer inspection reveals that for systems like CaC_6 (i) an enhanced three dimensionality arising through the intercalant orbitals makes the effect of Hubbard U less important (effect of U for a given bandwidth progressively becomes important as we go down in dimensions) and (ii) encouragement of charge density wave order arising from the intercalant order. Sulfur doped graphite, however, gives a hope that there is a possibility of high temperature superconductivity. Our present theoretical prediction should encourage experimentalists to study graphite from superconductivity point of view systematically, along the line pioneered by Kopelevich and collaborators.[27] In the past there have been claims (unfortunately not reproducible) of Josephson-like signals in graphite and carbon based materials;[28] Again, our result should encourage revival of studies along these lines.

Simple doping of a freely hanging graphene layer by gate control to the desired optimal doping of 10%–20% is not experimentally feasible at the present moment. It will be interesting to discover experimental methods that will allow us to attain these higher doping values. A simple estimate shows that a large cohesion energy arising from the strong σ bond that stabilizes the honeycomb structure will maintain the structural integrity of graphene.

The discovery of time reversal symmetry breaking $d+id$ order[10] for the superconducting state, within our RVB mechanism is very interesting. This unconventional order parameter has its own signatures in several physical properties: (i) spontaneous currents in domain walls, (ii) chiral domain wall states, (iii) unusual vortex structure, and (iv) large magnetic fields arising from the $d=2$ angular momentum of the cooper pairs, which could be detected μSR measurements. Suggestions for experimental determination of such an order by means of Andreev conductance spectra have been made by[29]

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