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Zigzag carbon nanotubes with generic electron-electron interactions

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Abstract

A Hamiltonian is derived for a zigzag carbon nanotube with an arbitrary number of weak electron-electron charge and spin interactions, which become significant in ultraclean systems. The renormalization group and bosonization are used to determine the ground-state phase diagram. Our phase diagram contains some exotic phases which have not previously been predicted in carbon nanotubes with physically possible interaction profiles. Phases of the undoped case include Mott insulators and a variety of density wave states. In the doped case a Tomanaga-Luttinger liquid is possible.

Keywords

interactions, carbon, nanotubes, electron, generic, zigzag

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Zigzag carbon nanotubes with generic electron-electron interactions

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A Hamiltonian is derived for a zigzag carbon nanotube with an arbitrary number of weak electron-electron charge and spin interactions, which become significant in ultraclean systems. The renormalization group and bosonization are used to determine the ground-state phase diagram. Our phase diagram contains some exotic phases which have not previously been predicted in carbon nanotubes with physically possible interaction profiles. Phases of the undoped case include Mott insulators and a variety of density wave states. In the doped case a Tomanaga-Luttinger liquid is possible.

Simple tight-binding calculations show that an undoped (n,m) carbon nanotube (CNT) will be metallic provided $p=(n-m)/3$ is an integer, implying that one third of all CNT are metallic.[1] However, recent experiments on ultra-clean CNT have indicated that all CNT are Mott insulators with gaps of about 10–100 meV.[2,3] These results are supported by theoretical models of CNT which include electron-electron (e-e) charge, or Coulomb, interactions.[4] Other possible mechanisms for the creation of gaps include curvature effects and lattice distortions but the resulting gaps, if they appear at all, tend to be small and only correspond with experiments in cases where e-e interactions can be ignored.[5,6]

Many theoretical studies of e-e interactions in CNT consider just on-site, and possibly nearest-neighbor, charge interactions.[4,7–9] This is a reasonable approximation if the interactions are strongly screened by, for example, a metallic substrate or a CNT bundle.[10,11] In other cases CNT have strong long-range charge interactions[12–14] which heavily influence several physical properties.[2,3,15–18] Furthermore, e-e spin interactions are rarely discussed, despite spin effects being quite important in CNT.[19,20] In this Brief Report we construct a Hamiltonian for zigzag CNT which includes both e-e spin and charge interactions. The interaction profile is completely general and may account for an arbitrary number of interactions between electrons on any two lattice sites. However, these interactions must be weak relative to the hopping strength. We use our Hamiltonian to determine the phase diagram of both an undoped and doped zigzag CNT, and find some exotic phases, such as a *f*-density wave (FDW) and a Tomonaga-Luttinger liquid (TLL).

A zigzag CNT is constructed by rolling the graphene lattice shown Fig. 1 about the *y* axis so that the *x* axis is around the circumference. The hexagonal lattice divides into two identical triangular sublattices, *A* and *B*, with lattice constant *a*. We assume that hopping only occurs between nearest-neighbor sites but e-e interactions can exist between any two sites. The full Hamiltonian is separated into four parts $H=H_0+H_U+H_V+H_J$. The Hubbard Hamiltonian describes hopping,

$$H_0 = -t \sum_{(i,j) \in A} \sum_\sigma [c^\dagger_{ij\sigma}c_{(i-1)(j+1)\sigma} + c^\dagger_{ij\sigma}c_{(i+1)(j+1)\sigma} + \text{H.c.}]$$
$$- t_\perp \sum_{(i,j) \in A} \sum_\sigma [c^\dagger_{ij\sigma}c_{i(j-1)\sigma} + \text{H.c.}], \quad (1)$$

where σ represents the spin and H.c. is the Hermitian conjugate. The hopping strengths t and $t_\perp$ may be different when the curvature of the CNT is large but here we will assume $t=t_\perp$. For on-site interactions,

$$H_U = \sum_{(i,j) \in A,B} U^{ij} n_{ij\uparrow} n_{ij\downarrow}, \quad (2)$$

where $n_{ij\sigma}=c^\dagger_{ij\sigma}c_{ij\sigma}$. The charge interaction between different sites can be represented by the Hamiltonian

$$H_V = \frac{1}{2} \sum_{(i,j) \in A,B} \sum_{(k,l) \neq (0,0)} V^{ij}_{kl} n_{ij} n_{(i+k)(j+1)}, \quad (3)$$

where two integers (k,l) define the separation of two distinct sites, and $n_{ij}=\sum_\sigma n_{ij\sigma}$. Similarly, for spin interactions between different sites,

$$H_J = \frac{1}{2} \sum_{(i,j) \in A,B} \sum_{(k,l) \neq (0,0)} J^{ij}_{kl} \mathbf{S}_{ij} \cdot \mathbf{S}_{(i+k)(j+l)}, \quad (4)$$

with spin operators $\mathbf{S}_{ij}=\frac{1}{2}\sum_{\sigma\sigma'}c^\dagger_{ij\sigma}\boldsymbol{\tau}_{\sigma\sigma'}c_{ij\sigma'}$ and Pauli spin matrices $\boldsymbol{\tau}=(\tau_x,\tau_y,\tau_z)$. Note that in H_V and H_J we do not
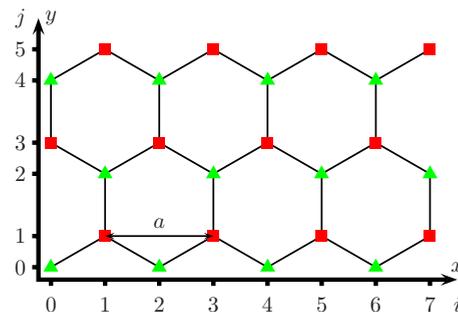

FIG. 1. (Color online) A graphene lattice with the two sublattices represented by triangles (sublattice *A*) and squares (sublattice *B*). A lattice site is represented by the set of integers (i,j).

include $(k,l)=(0,0)$ as this is already accounted for in H_U, and the factor of one half is required because we have counted each interaction twice.

The energy dispersion of an interactionless CNT can be obtained from H_0 and the Fermi momentum, or Dirac points, of a metallic $(n,0)$ zigzag CNT can be shown to be $\pm(2\pi/3a, \pm 2\pi/\sqrt{3}a)$ at or very near to half filling. The partial Fourier transform in the x direction will be dominated by the x component of the Fermi momentum and we can simplify to $c_{kl}=\frac{1}{\sqrt{n}}\Sigma_{q=\pm}e^{2\pi iqk/3}d_{ql}$, where $q=\pm$ accounts for momentum in both directions and $d_{ql\sigma}$ is a fermion operator.[7] After substituting this Fourier transform into Eq. (2) one obtains a two-leg ladder Hamiltonian with no rungs. The legs are represented by $q=\pm$ and lie in the y direction.[7,9]

After substituting the partial Fourier transform the interaction Hamiltonians become

$$H_U = \sum_j \mathcal{U}^j \Bigg[\sum_{qq'} d^\dagger_{qj\uparrow} d_{qj\uparrow} d^\dagger_{q'j\downarrow} d_{q'j\downarrow} + \sum_q d^\dagger_{qj\uparrow} d_{\bar{q}j\uparrow} d^\dagger_{\bar{q}j\downarrow} d_{qj\downarrow} \Bigg],$$

$$H_V = \sum_{jl\sigma\sigma'} \Bigg[\mathcal{V}^j_l \sum_{qq'} d^\dagger_{qj\sigma} d_{qj\sigma} d^\dagger_{q'j+l\sigma'} d_{q'j+l\sigma'} $$
$$+ \mathcal{V}'^j_l \sum_q d^\dagger_{qj\sigma} d_{\bar{q}j\sigma} d^\dagger_{\bar{q}j+l\sigma'} d_{qj+l\sigma'} \Bigg],$$

$$H_J = \sum_{jl\sigma\sigma'\alpha\alpha'} \Bigg[\mathcal{J}^j_l \sum_{qq'} d^\dagger_{qj\sigma} \boldsymbol{\tau}_{\sigma\sigma'} d_{qj\sigma'} d^\dagger_{q'j+l\alpha} \boldsymbol{\tau}_{\alpha\alpha'} d_{q'j+l\alpha'} $$
$$+ \mathcal{J}'^j_l \sum_q d^\dagger_{qj\sigma} \boldsymbol{\tau}_{\sigma\sigma'} d_{\bar{q}j\sigma'} d^\dagger_{\bar{q}j+l\alpha} \boldsymbol{\tau}_{\alpha\alpha'} d_{qj+l\alpha'} \Bigg], \quad (5)$$

where $\bar{q}=-q$ and the effective interactions are

$$\mathcal{U}^j = \sum_i U^{ij}/n^2 = U^j/n,$$

$$\mathcal{X}^j_l = \sum_{ik} X^{ij}_{kl}/2n^2 = \sum_{k=-N^\mathcal{X}_x}^{N^\mathcal{X}_x} X^j_{kl}/2n,$$

$$\mathcal{X}'^j_l = \sum_{ik} e^{4\pi ikq/3a} X^{ij}_{kl}/2n^2 = \sum_{k=-N^\mathcal{X}_x}^{N^\mathcal{X}_x} \cos(4\pi k/3a) X^j_{kl}/2n, \quad (6)$$

for $\mathcal{X}=\mathcal{V},\mathcal{J}$ and $X=V,J$. We can remove the i dependence on the interactions because for each j we have an identical interaction profile for $i=0,1,\dots,(n-1)$. In $\mathcal{X}'_l$, symmetry arguments allow us to remove the q dependence. We have introduced the integer $N^\mathcal{X}_x$ to define a hard cutoff for k. For a $(n,0)$ zigzag CNT, $N^\mathcal{X}_x \leq n$ and if $N^\mathcal{X}_x = n$ the lower limit of the sum should be changed to $-(N^\mathcal{X}_x-1)$ in order to avoid double counting.

The interacting two-leg ladder problem can be solved using a well-developed method,[21–29] although with some differences due to the absence of rungs.[23,30] The Fermi operators $d_{qj\sigma}$ are linearized about the Fermi momentum k_F by expanding in terms of chiral left-moving and right-moving fields and rapidly varying terms are discarded. One can then write the interaction Hamiltonian in terms of four-field current operators, the coefficients of which are the coupling constants which describe scattering about the lattice and ultimately determine the phase. A requirement of this method is that the effective interaction strengths are weak, $\mathcal{U}^j, \mathcal{X}^j_l, \mathcal{X}'^j_l \ll t$ but this does not imply that the original interactions U^{ij} and X^{ij}_{kl} must be weak. The effective interactions are obtained from the original interactions after rescaling by n, as can be seen in Eq. (6). Therefore, provided we assume that n is not small it is possible for the effective interactions to be weak, even if the original interactions are not.

All coupling constants may be expressed in terms of the interaction strengths. The forward-scattering coupling constants are

$$f^\rho_{12} = 2b\Bigg\{ \mathcal{U} + \sum_l \Bigg[4\mathcal{V}_l - \cos 2lbk_F\Big(2\mathcal{V}_l + \frac{3}{2}\mathcal{J}_l\Big) \Bigg] \Bigg\},$$

$$f^\sigma_{12} = 2b\Bigg\{ \mathcal{U} + \sum_l \Bigg[\cos 2lbk_F\Big(2\mathcal{V}_l - \frac{1}{2}\mathcal{J}_l\Big) - \mathcal{J}_l \Bigg] \Bigg\}, \quad (7)$$

where $q=\pm$ is relabeled as $q=1,2$, $b=a\sqrt{3}/4$ and $f_{qq}=0$, $f_{q\bar{q}}=f_{\bar{q}q}$. The backward-scattering coupling constants are

$$b^\rho_{11} = 2b\Bigg\{ \mathcal{U} + \sum_l \Bigg[4\mathcal{V}_l - \cos 2lbk_F\Big(2\mathcal{V}'_l + \frac{3}{2}\mathcal{J}'_l\Big) \Bigg] \Bigg\},$$

$$b^\rho_{12} = 2b\Bigg\{ \mathcal{U} + \sum_l \Bigg[-\cos 2lbk_F\Big(2\mathcal{V}_l + \frac{3}{2}\mathcal{J}_l\Big) + 4\mathcal{V}'_l \Bigg] \Bigg\},$$

$$b^\sigma_{11} = 2b\Bigg\{ \mathcal{U} + \sum_l \Bigg[\cos 2lbk_F\Big(2\mathcal{V}'_l - \frac{1}{2}\mathcal{J}'_l\Big) - \mathcal{J}_l \Bigg] \Bigg\},$$

$$b^\sigma_{12} = 2b\Bigg\{ \mathcal{U} + \sum_l \Bigg[\cos 2lbk_F\Big(2\mathcal{V}_l - \frac{1}{2}\mathcal{J}_l\Big) - \mathcal{J}'_l \Bigg] \Bigg\}, \quad (8)$$

with $b_{qq'}=b_{\bar{q}\bar{q}'}$. Finally, for umklapp scattering,

$$u^\rho_{11} = 2b\Bigg[\mathcal{U} + \sum_l (-1)^l (2\mathcal{V}_l - \mathcal{J}_l) \Bigg],$$

$$u^\rho_{12} = 2b\Bigg[2\mathcal{U} + \sum_l (-1)^l \Big(2\mathcal{V}_l + 2\mathcal{V}'_l - \frac{3}{2}\mathcal{J}_l - \frac{3}{2}\mathcal{J}'_l\Big) \Bigg],$$

$$u^\sigma_{12} = 2b\sum_l (-1)^l \Big(-2\mathcal{V}_l + 2V'_l - \frac{1}{2}\mathcal{J}_l + \frac{1}{2}\mathcal{J}'_l\Big), \quad (9)$$

with $u_{qq'}=u_{\bar{q}\bar{q}'}$ and $u_{qq}=0$. All sites in the same sublattice are identical and one can map between the two sublattice with $l\to -l$. Therefore, all sites have an identical interaction profile and we can drop the j superscript. We introduce the integer $N^\mathcal{X}_y$ to define a hard cutoff for l, $|l|\leq N^\mathcal{X}_y$, for $\mathcal{X}=\mathcal{V},\mathcal{J}$. Note that we cannot have any (k,l) which satisfies $|k|\leq N^\mathcal{X}_x$ and $|l|\leq N^\mathcal{X}_y$ as not all these values of (k,l) define distances between two lattice sites in the CNT lattice. The above solutions of the coupling constants are completely general and any values can be used for U, V_{kl}, and J_{kl}, provided the effective interactions are weak $U,V_{kl},J_{kl}\ll nt$. Our

solutions reduce to just on-site interactions when $N_{x,y}^{\mathcal{X}}=0$ or up to nearest-neighbor interactions when $N_{x,y}^{\mathcal{X}}=1$. By renormalizing the coupling constants and bosonizing the Hamiltonian, the Hamiltonian can be much simplified and the ground-state phase can be determined.[23]

To illustrate how a ground-state solution may be obtained we consider a special case where the interactions are defined in terms of two independent variables, V and J. We assume that all spin and charge interactions are unscreened up to $|l|=N_y^{\mathcal{X}}$ and $|k|=N_x^{\mathcal{X}}$ so that the interaction strength between any two sites is inversely proportional to the distance between them, i.e., $\mathcal{X}_{kl}=\mathcal{X}/d(k,l)$, where $d(k,l)$ is the distance represented by the integers (k,l). In using this expression we ignore not only short-range screening but also the shape of the atomic wave function, which has a significant influence on the interaction strength. However, as the main influence is the distance, our simplification is reasonable. The charge interaction V must be positive as it is repulsive. The spin interaction J can be positive or negative for antiferromagnetism and ferromagnetism, respectively. A third variable U may be defined but as the renormalization group is determined solely by V/U and J/U we simply make U constant, although it must be positive.

In the undoped, or half filled, case there are nine possible phases, four Mott phases, four density wave phases, and a TLL phase. As these phases and their associated order parameters are explained in detail elsewhere[9] we will only give a brief description here. The possible Mott phases are a D-Mott, S-Mott, D'-Mott, and S'-Mott. The S refers to symmetric s-wave pairing of chiral fields and the D refers to antisymmetric d-wave pairing. For the unprimed Mott phases the center of mass of the pairing falls, on average, between two nearest neighbor sites, while for the primed Mott phases the center of mass falls between two next-nearest neighbor sites. The density wave phases include a CDW, a p-density wave (PDW), a FDW, and a chiral current phase (CCP). The CDW has an electron density which alternates between no electron and two electrons per site. The PDW consists of nearest-neighbor dimers. A CCP allows currents to flow along the zigzags of the CNT lattice while a FDW has nonzero current flow between next-nearest neighbors. In the doped case there are only five possible phases because of the broken particle-hole symmetry. There are two "superconducting" phases, so-called because they have a nonzero order parameter which resembles a superconducting order parameter. The antisymmetric order parameter defines a d-wave superconductor, or D-SC, and the symmetric one defines a s-wave superconductor, or S-SC. There are two density wave phases, one is a combination of the undoped CDW and PDW, which for convenience we name a CDW, and the other is a combination of the undoped CCP and the FDW, which we name a CCP. The doped case may also have a TLL phase.

Figure 2 shows phase diagrams of a (12, 0) zigzag CNT at half filling and when doped but still close to half filling. For the undoped case with $N_{x,y}^{\mathcal{X}}=1$, when V/U is small the on-site repulsion U will dominate and set one electron per site. If, in addition, J/U is small the electron spins are not ordered and one always finds a D-Mott phase. Increasing V will encourage on-site pairings on alternative sites when the repulsion from the three nearest neighbors exceeds the on-site repul-
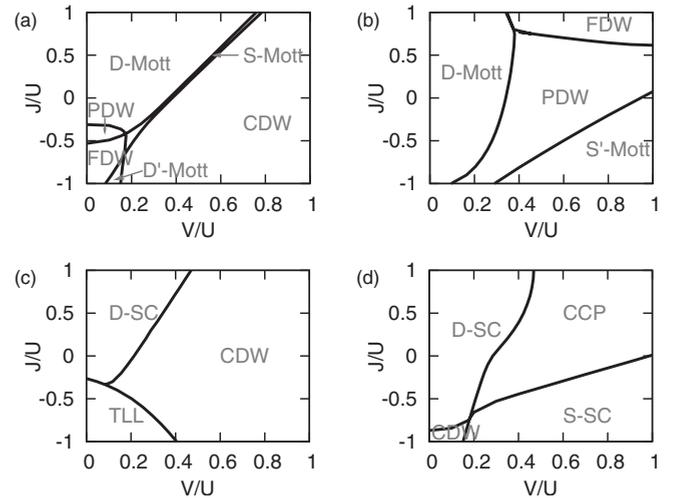

FIG. 2. Phases of a (12, 0) CNT (a) undoped, $N_{x,y}^{\mathrm{X}}=1$; (b) undoped, $N_{x,y}^{\mathrm{X}}=2$; (c) doped, $N_{x,y}^{\mathrm{X}}=1$; and (d) doped, $N_{x,y}^{\mathrm{X}}=2$.

sion, resulting in a CDW phase. Antiferromagnetic spin interactions between nearest neighbors tends to favor the D-Mott phase over the CDW phase and so the D-Mott phase has a greater range when J/U is large and positive. However, the opposite is true for ferromagnetic interactions. There is some conflict between the interactions when V/U is small and J/U is large and negative, resulting in a number of different phases.

In the doped case with $N_{x,y}^{\mathcal{X}}=1$ the D-SC and CDW phases are the analogs of the D-Mott and CDW phases in the undoped case. For small charge interactions and large ferromagnetic spin interactions a TLL phase can be found. It can be shown that the TLL may only appear when $b_{11}^{\rho}>f_{12}^{\rho}$ (Ref. 29) and from Eqs. (7) and (8) it can be shown that this constraint generally requires $J<0$. A TLL is defined by the convergence of all coupling constants as the system is renormalized. Since one cannot renormalize indefinitely it is not possible to state conclusively that the TLL phase shown in Fig. 2(c) is definitely a TLL as it may be some other phase which appears to converge after much renormalization and then suddenly diverges.[29] In either case, after considering a number of different values for $N_{x,y}^{\mathcal{X}}$ we conclude that TLL-like behavior only arises when the e-e spin interactions are ferromagnetic and when these spin interactions are approximately of the same order as the charge interactions. This gives a clear indication of the importance of e-e spin interactions in CNT.

Interactions which extend beyond nearest neighbors tend to lead to frustration and so the phase is not always obvious. When V/U and J/U are small the undoped case for $N_{x,y}^{\mathcal{X}}=2$ will be a D-Mott phase, like the simpler $N_{x,y}^{\mathcal{X}}=1$ case. As V increases a conflict arises between satisfying repulsion for on-site, nearest-neighbor and next-nearest-neighbor interactions. The PDW can satisfy some of these repulsive interactions as well as antiferromagnetic nearest-neighbor interactions and so the PDW dominates for most of $J>0$. For ferromagnetic interactions on-site pairing is more likely as it minimizes the ferromagnetic conflict between nearest neighbors and next-nearest neighbors. However, a CDW phase

would conflict with the next-nearest-neighbor repulsive interactions, so instead the phase is a S'-Mott in which the pairs are distributed randomly. In the doped case the D-SC, S-SC, and FDW phases are analogs of the undoped D-Mott, S'-Mott, and CCP phases, respectively. The CCP dominates this phase diagram, particularly for large V/U and positive J as frustration is minimized through the flow of current. Such a phase is less likely in the undoped CNT as repulsive interactions in a half-filled lattice tend to restrict current flow.

In the undoped case we find that it is N_y which essentially determines the appearance of the phase diagram. For odd N_y the phase diagram will be similar to Fig. 2(a) but if N_y is even the phase diagram will be similar to Fig. 2(b). While there may be some minor differences, for example, the S-Mott phase is larger when $N_y=3$ compared to $N_y=1$ and is shifted to the right of the phase diagram, the types of phases and their approximate positions is much the same. The differences between odd and even N_y are essentially due to the cosine term in Eqs. (7) and (8) reducing to $(-1)^l$ at half filling when $k_F=\pi/2b$.[9] In contrast, N_x has only a slight influence on the phase diagram. For example, increasing N_x with constant even N_y may cause the FDW phase region to spread to reasonably small values of J/U and V/U but the same phases are observed in all cases. The phase diagrams for the doped cases show more variety compared to the undoped cases. While they are not overly sensitive to changes in N_x, very different phase diagrams can be obtained for different N_y. The two cases shown in Figs. 2(c) and 2(d) are fairly typical examples.

In conclusion, we have derived a Hamiltonian which is applicable to CNT with generic e-e spin and charge interactions, in contrast to most theoretical studies which only consider on site and possibly nearest-neighbor charge interactions. As it is now possible to produce ultraclean CNT in which long-range interactions are not strongly screened, theoretical models that are able to deal with numerous interactions are of increasing importance. The illustrative example shows that a far richer phase diagram is possible when both charge and spin interactions are considered, for example, a TLL requires ferromagnetic spin interactions. Furthermore, we find that solutions obtained from only nearest-neighbor interactions do not necessarily produce an accurate picture.

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