

Low-energy model for doped graphene nanoribbons

J. Ferrer^{1,2,3}, A. García-Fuente^{1,2}, Y. Yang^{1,3}, S. Volosheniuk^{1,3}, and H. S. J. van der Zant^{1,3}

¹*Departamento de Física, Universidad de Oviedo, 33007 Oviedo, Spain*

²*Centro de Investigación en Nanomateriales y Nanotecnología, CSIC, 33940 El Entrego, Spain*

³*Kavli Institute of Nanoscience, Delft University of Technology, Delft, The Netherlands*



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In this article we analyze the many-body behavior of freestanding doped graphene nanoribbons where the chemical potential lies inside the bulk single-particle bands. We perform an exact mapping from both an extended and an on-site Hubbard model of the ribbons to a Kanamori model, which includes ferromagnetic exchange and pair-hopping interactions. We determine the resulting Coulomb matrix elements analytically, and identify their scaling behavior as a function of ribbon width and length. We propose a low-energy version of the Kanamori Hamiltonian to address the response of the ribbons to external fields, with a view to their use as transport channels in nanoelectronics. We find that the model and the proposed ribbon parameters can produce open-shell, high-spin, many-body states that can lead to shell- and spin-blockade responses.

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I. INTRODUCTION

Graphene was originally envisioned as the dreamed-of electronic material [1]. The vision included the realization of nanoelectronic devices such as transistors or quantum dots, frequently based on graphene nanoribbons (GNRs) [2]. However, the expected revolution was hindered by a variety of factors, among which are the difficulties in producing atomically precise GNRs [3]. Recently, atomically precise GNRs have been grown in ultrahigh vacuum chambers and transferred to substrates with metal electrodes. Transport through single- or few-nanoribbon devices has recently been measured, showing single-electron transistor and quantum dot behavior [4–7]. In understanding their behavior, it is important to realize that graphene nanoribbons are different from carbon nanotubes because $\pm\mathbf{k}$ wave vectors are mixed by boundary conditions, so valleys are not good quantum numbers. Spin splitting is expected to be absent in graphene nanoribbons except when the chemical potential is pinned around the edge-state energy window. Furthermore, both the intrinsic and Rashba spin-orbit interactions are expected to be small like in bulk graphene, which exhibits a gap of about 25 μeV [8–10].

In previous work [11,12], we diagonalized analytically the tight-binding model of GNRs with arbitrary length and width, and showed that the existence or not, magnetic nature, and energy of edge states in $N = 5, 7$, and 9 GNRs could be described by the use of an effective dimer Hubbard model. The tight-binding solution was extended to the full Hubbard model of a GNR in Ref. [13]. The Hubbard term was rewritten in the ribbon eigenstate basis, whereby a huge sum containing operator products $\hat{c}_{1\sigma}^\dagger \hat{c}_{2\sigma'}^\dagger \hat{c}_{3\sigma'} \hat{c}_{4\sigma}$ was generated. The sum was truncated by keeping only direct 1–4, 2–3 pairings,

and the Coulomb integrals were calculated analytically. Explicit analytical mean-field equations for the paramagnetic, ferromagnetic, and antiferromagnetic phases were deduced. The electronic band structure for narrow ribbons agreed with previous mean-field Hubbard model simulations [14,15]. The band structure for bulk-sized samples with edges did contain both the bulk Dirac cone as well as the edge states band at the same time, something that had not been achieved before.

In this article, we consider freestanding finite-length graphene nanoribbons (GNRs). The ribbons are doped and possibly subject to a gate voltage so that the chemical potential lies inside the bands of bulk states. We provide an exact mapping of the extended Hubbard model of GNRs [16–18] to a Kanamori Hamiltonian [19], which is written in terms of the single-particle bulk eigenstates of the ribbon. We thus show that Hubbard interactions lead not only to direct Coulomb integrals among these single-particle bulk eigenstates, but also to ferromagnetic exchange and pair-hopping terms. The pristine extended Hubbard Hamiltonian is written in a real-space site basis; in contrast the Kanamori Hamiltonian is written in the basis of single-particle eigenstates. This new Hamiltonian can be useful for the prediction or the analysis of phenomena that address given energy windows. An example is recent transport experiments of single- or few-ribbon junctions [4–7,20] that have uncovered their quantum dot response. In these low-temperature experiments, a gate voltage tunes the chemical potential, and a narrow energy window of bulk ribbon eigenstates is addressed. A detailed stability diagram featuring Coulomb diamonds and excited-state lines can be drawn by sweeping both the bias and gate voltages in those experiments.

We will discuss in this article how the new Hamiltonian allows us to address cleanly narrow energy windows of eigenstates, whose position is given by the chemical potential and whose width is determined by and can be adapted to the number of single-particle states that are half filled. The resulting low-energy Hamiltonian possesses a small enough

Fock space that can be analyzed exactly and in detail either analytically or numerically. We find that the Kanamori exchange interactions are ferromagnetic and thus produce high-spin open-shell many-body states in doped GNRs. Several theorems preclude the existence of saturated ferromagnetism for the Hubbard model [21,22], but do predict high-spin (ferromagnetic) ground states that fulfill specific conditions [22,23]. We find equivalent conditions for the high-spin ground states described below. Similarly, some studies predict mesoscopic Stoner instabilities in open quantum dots [24] as we do here.

The low-energy Hamiltonian approach is complementary to describing GNRs by density functional theory (DFT) simulations. This is because the Coulomb parameters introduced in the initial extended Hubbard model can and must be deduced from DFT [11,16–18]. On the other hand, correlated electron phenomena like Coulomb blockade or Kondo physics cannot be described correctly by DFT simulations. The reason is that DFT is a one-body theory so that even the exact functional does not describe the energy position and number of many-body quasiparticle poles [25,26]. A DFT-based analysis of Coulomb blockade phenomena delivers a wrong number and positioning of Coulomb diamonds, and of excited state lines.

We will make this article self-contained and will therefore make use of some results that have been published previously [11,13]. The layout of the article is as follows. Section II summarizes the analytical solution of the tight-binding model of graphene ribbons of arbitrary width and length [11]. Section III explains the extended Hubbard model of bulk graphene and introduces it for graphene nanoribbons. This section also shows how the extended Hubbard Hamiltonian can be mapped to a Kanomori model [19], and determines the Coulomb matrix elements. Section IV introduces an on-site Hubbard model and fits the U parameter using density functional theory (DFT) simulations. The Coulomb matrix elements are vastly simplified here, and found to obey simple relationships among them, and a remarkable size-scaling law. The section also discusses analytical mean-field solutions that were found earlier [13]. Section V uses the previous results to propose a low-energy Kanamori Hamiltonian to describe the open-shell and spin behavior of doped GNRs. The low-energy Hamiltonian is solved for the case of two and three shells, and a specific example using a $N = 9$ armchair ribbon is discussed. Section VI introduces a transfer Hamiltonian and discusses transition rates in the sequential electron tunneling regime. The section thus shows how the model can lead to shell and spin blockade in electrical transport experiments using the ribbons as channels connecting two leads. A short conclusion closes the article.

II. TIGHT-BINDING MODEL OF GRAPHENE RIBBONS

We consider nanoribbons whose lengths and widths are M_x and $M_y = \frac{N+1}{2}$, according to the convention set in Fig. 1. Unit cells are rectangles having four atoms. We measure lengths along the Cartesian X and Y axes in units of $\sqrt{3}c$ and c , respectively, where $c = 2.46 \text{ \AA}$ is graphene's lattice constant. Cell coordinates are $\mathbf{R} = (n_x, n_y)$ with $n_{x/y} = 1, 2, \dots, M_{x/y}$. Furthermore, we address for simplicity ribbons where no edge states contribute to the physics. This condition happens in both armchair and zigzag ribbons when-

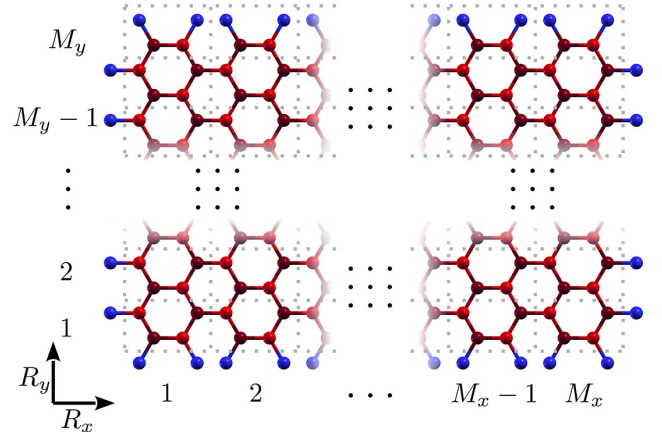


FIG. 1. Real graphene atoms are indicated by red circles. Fake atoms, whose wave-function coefficients are set to zero, are indicated by blue circles. Each unit cell is surrounded with a gray dotted box.

ever the effective doping or the gate voltage is high enough that the chemical potential lies inside the conduction or valence bands.

The tight-binding Hamiltonian is constructed by placing a Wannierized p^z orbital at each site $\mathbf{i} = \mathbf{R} + \mathbf{d}_{A/B}$ of the lattice with unit cell coordinates $\mathbf{R}$ and intracell coordinates $\mathbf{d}_{A/B}$. The corresponding states are represented by the kets $|\mathbf{i}\rangle = |\mathbf{R} + \mathbf{d}_{A/B}\rangle$. The second-quantized Hamiltonian is

$$\hat{H}^0 = -t \sum_{\langle \mathbf{i}, \mathbf{j} \rangle, \sigma} \hat{r}_{\mathbf{i}\sigma}^\dagger \hat{r}_{\mathbf{j}\sigma}, \quad (1)$$

where the sum is restricted to nearest neighbor sites. The linear operator $\hat{r}_{\mathbf{i}\sigma}^\dagger$ creates an electron with spin σ at the ribbon site $\mathbf{i}$, and $\hat{n}_{\mathbf{i}\sigma} = \hat{r}_{\mathbf{i}\sigma}^\dagger \hat{r}_{\mathbf{i}\sigma}$ is the corresponding number operator, with $\hat{n}_{\mathbf{i}} = \sum_{\sigma} \hat{n}_{\mathbf{i}\sigma}$.

The ribbon boundary conditions define the set of allowed wave vectors, that we define as $\bar{\mathbf{k}} = (\bar{k}_x, \bar{k}_y) = (\sqrt{3}c k_x, c k_y)$. Here, $\bar{k}_y = k_y = \pi \frac{m}{M_y}$ with $m = 1, \dots, M_y$. The wave vectors $\bar{k}_x = k_{m\alpha}$, with $\alpha = 1, \dots, 2M_x$, must be computed numerically and are not spaced evenly. The subset k_{m2M_x} with $k_m \geq \frac{2}{3}\pi$ corresponds to edge ribbon states [11,13] and is discarded, while all the rest are labeled bulk ribbon states and are kept. The Hamiltonian H^0 can be diagonalized analytically for any nonchiral finite-length ribbon [11]. The site operators can be decomposed in terms of bulk eigenstate operators as follows,

$$\begin{aligned} \hat{r}_{\mathbf{i}\sigma}^\dagger &= \sum_{m\alpha\tau} \hat{r}_{m\alpha\tau\sigma}^\dagger \langle \phi_{m\alpha\tau} | \mathbf{i} \rangle, \\ \langle \mathbf{i} | \phi_{m\alpha\tau} \rangle &= \sum_{R_{x,y}=1}^{M_{x,y}} \frac{2 \sin k_m (R_y - d_i)}{(\mathcal{M}_x M_y A_{m\alpha})^{1/2}} \\ &\quad \times \left(-\tau (-1)^\alpha \sin(k_{m\alpha} (R_x - d_i)) \right. \\ &\quad \left. \times \sin[k_{m\alpha} (M_x + 1 - (R_x + d_i))] \right), \end{aligned} \quad (2)$$

where $\mathcal{M}_x = 4M_x + 1$, and $\tau = \pm 1$ is a conduction and valence band index, and $d_1 = 0$ and $d_2 = 1/2$. $\{m, \alpha, \tau, \sigma\}$ is a complete set of quantum numbers in the ribbon. The eigenstates are spin degenerate, and we call the pair

$(m\alpha\tau\uparrow)$, $(m\alpha\tau\downarrow)$ a *shell* from now on. The normalization factor is

$$A_{m\alpha} = F^1(k_{m\alpha}) - \frac{\delta_{k_m, \pi}}{\mathcal{M}_x} F^2(k_{m\alpha}), \quad (4)$$

$$F^1(k) = 1 - \frac{\sin(\mathcal{M}_x k/2)}{\mathcal{M}_x \sin(k/2)}, \quad (5)$$

$$F^2(k) = 1 - \frac{\cos(\mathcal{M}_x k/2)}{\mathcal{M}_x \cos(k/2)}. \quad (6)$$

The diagonalized Hamiltonian and the single-particle eigenenergies are

$$\hat{H}^0 = \sum_{m\alpha\tau} \epsilon_{m\alpha\tau}^0 \hat{n}_{m\alpha\tau}, \quad (7)$$

$$\epsilon_{m\alpha\tau}^0 = \tau \sqrt{1 + \Delta_m^2 + 2\Delta_m \cos \frac{k_{m\alpha}}{2}}, \quad (8)$$

where $\Delta_m = 2 \cos \frac{k_m}{2}$.

III. EXTENDED HUBBARD MODEL OF GRAPHENE

A. Bulk graphene

The extended Hubbard model is constructed by adding to $\hat{H}^0$ the following interaction Hamiltonian:

$$\hat{H}^I = U^0 \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + \sum_{n, i, \delta^n} U^n \hat{n}_i \hat{n}_{i+\delta^n}. \quad (9)$$

The second sum extends to the different atomic neighbor shells n , while δ^n runs over all the atoms in the n th-neighbor shell of atom i . The on-site and extended Hubbard parameters U^0 and U^n correspond to the effective Coulomb interactions among electrons residing in the Wannierized p^z orbitals. These interactions correspond to the bare Coulomb interaction among p^z orbitals, screened by all the remaining higher-

energy orbitals, and eventually by the presence of a substrate and perhaps an overlayer. U^0 and U^n have been obtained for freestanding graphene and for bilayer graphene encapsulated or deposited on idealized metallic substrates in references [16,18]. The reported Hubbard parameters U^n for freestanding graphene can be rewritten in terms of the distance r between two sites and are very well fitted to the law [16,18]

$$U(r) = \frac{10.3}{1 + 0.6r} \text{ eV}, \quad (10)$$

where r must be given in angstroms. In addition, Ref. [17] has analyzed whether an on-site Hubbard model can describe freestanding graphene. The article finds that this is the case and reports an effective on-site $U^0 \approx 4.3$ eV, except for extreme doping cases very far away from half filling.

B. Graphene ribbons

We rewrite in this section the interaction Hamiltonian $\hat{H}^I$ in the basis of single-particle eigenstates of the ribbon. Inserting the decomposition 2 into Eq. (9), we find a large sum of terms of the type

$$\hat{r}_{m_1\alpha_1\tau_1\sigma_1}^\dagger \hat{r}_{m_2\alpha_2\tau_2\sigma_2}^\dagger \hat{r}_{m_3\alpha_3\tau_3\sigma_3} \hat{r}_{m_4\alpha_4\tau_4\sigma_4} \quad (11)$$

that are multiplied by Coulomb integrals. However, many of these integrals are zero because momentum conservation implies the condition $k_{m_1\alpha_1} + k_{m_2\alpha_2} = k_{m_3\alpha_3} + k_{m_4\alpha_4}$. The reason for the cancellation is that the wave vectors $k_{m\alpha}$ are not separated evenly for a ribbon, so the condition is not fulfilled except when $\mathbf{k}_1 = \mathbf{k}_3$ and $\mathbf{k}_2 = \mathbf{k}_4$, or when $\mathbf{k}_1 = \mathbf{k}_4$ and $\mathbf{k}_2 = \mathbf{k}_3$. In addition, the two single-particle bulk states having the same $k_{m\alpha}$ and opposite τ are effectively unentangled because they possess very different energies. As a consequence, momentum conservation allows us to rewrite the following effective Hamiltonian for bulk ribbon states; the Hamiltonian is of the Kanamori type [19]:

$$\hat{H} = \sum_a \epsilon_a^0 \hat{n}_a + \sum_a V_a \hat{n}_{a\uparrow} \hat{n}_{a\downarrow} + \sum_{a \neq b} \left(V_{ab}^n \hat{n}_a \hat{n}_b - V_{ab}^S \hat{\mathbf{S}}_a \cdot \hat{\mathbf{S}}_b + V_{ab}^\Delta \frac{\hat{\Delta}_a^\dagger \hat{\Delta}_b + \hat{\Delta}_b^\dagger \hat{\Delta}_a}{2} \right), \quad (12)$$

where a and b are shorthand for the single-particle quantum numbers $(m\alpha\tau)$ and $(m'\alpha'\tau')$. $\hat{\mathbf{S}}_a = \hat{r}_{a\sigma}^\dagger \frac{\tau_{\sigma\sigma'}}{2} \hat{r}_{a\sigma'}$ are spin operators while $\hat{\Delta}_a^\dagger = \hat{r}_{a\uparrow}^\dagger \hat{r}_{a\downarrow}^\dagger$ are pair-hopping operators. The Hamiltonian parameters possess the following explicit expressions:

$$\epsilon_a = \tau \epsilon_a^0 + V_{aa}, \quad (13)$$

$$V_a = V_{aa}^0 + V_{aa}, \quad (14)$$

$$V_{ab}^n = \frac{V_{ab}^0}{4} + V_{ab} - \frac{J_{ab}}{2}, \quad (15)$$

$$V_{ab}^S = V_{ab}^\Delta = V_{ab}^0 + 2J_{ab} V_{ab}^\Delta. \quad (16)$$

The direct and exchange matrix elements can be written as follows:

$$V_{ab} = V_{ab}^1 + V_{ab}^2 + V_{ab}^3 + \dots, \quad (17)$$

$$J_{ab} = J_{ab}^1 + J_{ab}^2 + J_{ab}^3 + \dots, \quad (18)$$

where each V_{ab}^n and J_{ab}^n derives from the Hubbard terms $U^n \hat{n}_i \hat{n}_{i+\delta^n}$ that connect sites separated by n shells. The on-site-derived interactions are

$$V_{m\alpha, m'\alpha'}^0 = 2C U^0 \sum_{i=1,2} \sum_{n_x} \sin^2(k_{m\alpha} n_x^i) \sin^2(k_{m'\alpha'} n_x^i) \times \sum_{n_y} \sin^2(k_m n_y^i) \sin^2(k_{m'} n_y^i), \quad (19)$$

$$C = \frac{16}{\mathcal{M}_x^2 M_y^2 A_{m\alpha} A_{m'\alpha'}}, \quad (20)$$

with vectors $\mathbf{n}^1 = (n_x, n_y)$ and $\mathbf{n}^2 = (n_x - 1/2, n_y - 1/2)$. The explicit expressions for the shell-dependent matrix elements depend on whether U^n connects same or opposite

sublattices. When U^n connects same-sublattice shells with neighbor vectors δ^n , we find

$$V_{m\alpha;m'\alpha'}^n = C U^n \sum_{i=1,2} \sum_{\delta^n} \sum_{n_x} \sin^2(k_{m\alpha} n_x^i) \sin^2(k_{m'\alpha'}(n_x^i + \delta_x^n)) \sum_{n_y} \sin^2(k_m n_y^i) \sin^2(k_{m'}(n_y^i + \delta_y^n)), \quad (21)$$

$$J_{m\alpha;m'\alpha'}^n = C U^n \sum_{i=1,2} \sum_{\delta^n} \sum_{n_x} \sin(k_{m\alpha} n_x^i) \sin(k_{m'\alpha'} n_x^i) \sin(k_{m\alpha}(n_x^i + \delta_x^n)) \sin(k_{m'\alpha'}(n_x^i + \delta_x^n)) \\ \times \sum_{n_y} \sin(k_m n_y^i) \sin(k_{m'} n_y^i) \sin(k_m(n_y^i + \delta_y^n)) \sin(k_{m'}(n_y^i + \delta_y^n)). \quad (22)$$

In contrast, when U^n connects opposite-sublattice shells with neighbor vectors δ^n , we find

$$V_{m\alpha;m'\alpha'}^n = C U^n \sum_{i=1,2} \sum_{\delta^n} \sum_{n_x} \sin^2(k_{m\alpha} n_x^i) \sin^2(k_{m'\alpha'}[M_x + 1 - (n_x^i + \delta_x^n)]) \sum_{n_y} \sin^2(k_m n_y^i) \sin^2(k_{m'}(n_y^i + \delta_y^n)), \quad (23)$$

$$J_{m\alpha;m'\alpha'}^n = C U^n \sum_{i=1,2} \sum_{\delta^n} \sum_{n_x} \sin(k_{m\alpha} n_x^i) \sin(k_{m'\alpha'} n_x^i) \sin(k_{m\alpha}[M_x + 1 - (n_x^i + \delta_x^n)]) \sin(k_{m'\alpha'}[M_x + 1 - (n_x^i + \delta_x^n)]) \\ \times \sum_{n_y} \sin(k_m(n_y^i)) \sin(k_{m'}(n_y^i)) \sin(k_m(n_y^i + \delta_y^1)) \sin(k_{m'}(n_y^i + \delta_y^1)). \quad (24)$$

Finally, the Hubbard parameters U^n are well fitted by Eq. (10) for freestanding bulk graphene [16,18], but are unknown for graphene ribbons. A possible strategy consists of borrowing and using the bulk parameters in Eq. (10).

IV. ON-SITE HUBBARD MODEL OF GRAPHENE RIBBONS

A. Determination of the on-site Hubbard parameter U^0

In the rest on this article we follow a second strategy, which consists of reducing the parameters of the extended Hubbard model down to an on-site renormalized interaction U^0 . This strategy was implemented for bulk graphene in Ref. [17], where the authors found a renormalized $U^0 \simeq 4.3$ eV. We attempt a similar task in this section for graphene ribbons. We make use of the fact that DFT is a mean-field theory akin in spirit to performing a Hartree decomposition of the on-site Hubbard model. We perform DFT and Hartree decompositions of the on-site Hubbard model for armchair ribbons with $M_y = 4, 5, 9$ (corresponding to $N = 7, 9, 17$) and lengths ranging from $M_x = 2$ up to infinity. We find that an on-site Hubbard $U^0 = 4.9$ eV fits the DFT bulk gap and the lowest-lying electron and hole excitations (also called HOMO-1, HOMO, LUMO, and LUMO + 1), where HOMO is the highest occupied molecular orbital, and LUMO is the lowest-lying molecular) rather closely, as demonstrated in Tables I–III. We therefore choose $U^0 = 4.9$ eV for the remainder of this article.

Notice that the matrix elements in Eqs. (13)–(18) simplify for the on-site Hubbard model and fulfill $4 V_{ab}^n = V_{ab}^S = V_{ab}^\Delta = V_{ab}^0$ so that the Hamiltonian in Eq. (12) simplifies to

$$\hat{H} = \sum_a (\epsilon_a^0 \hat{n}_a + V_{aa}^0 \hat{n}_{a\uparrow} \hat{n}_{a\downarrow}) \\ + \sum_{a \neq b} V_{ab}^0 \left(\frac{\hat{n}_a \hat{n}_b}{4} - \hat{\mathbf{S}}_a \cdot \hat{\mathbf{S}}_b + \frac{\hat{\Delta}_a^\dagger \hat{\Delta}_b + \hat{\Delta}_b^\dagger \hat{\Delta}_a}{2} \right). \quad (25)$$

A further advantage of this model is that the on-site matrix elements in Eq. (19) can be computed analytically [11,13]. This allows us to determine them even for ribbons with sizes

of the order of micrometers. The expressions are

$$V_{m\alpha;m'\alpha'}^0 = \mathcal{D} \frac{\delta_{mm'}^+ C_{m\alpha,m'\alpha'}^{B,+} + \delta_{mm'}^- C_{m\alpha,m'\alpha'}^{B,-} / \mathcal{M}_x}{\Lambda_{m\alpha}^\phi \Lambda_{m'\alpha'}^\phi}, \quad (26)$$

where

$$\mathcal{D} = \frac{U^0}{2 \mathcal{M}_x \mathcal{M}_y}, \quad (27)$$

$$\delta_{mm'}^+ = 1 + \frac{1}{2}(\delta_{m,m'} + \delta_{m,M_y} \delta_{m',M_y}), \quad (28)$$

$$\delta_{mm'}^- = \frac{1}{2} \delta_{m+m',M_y} - (\delta_{m,M_y} + \delta_{m',M_y}), \quad (29)$$

$$C_{m\alpha,m'\alpha'}^{B,+} = 2 F^1(k_{m\alpha}) + 2 F^1(k_{m'\alpha'}) - F^1(k_+) - F^1(k_-), \quad (30)$$

$$C_{m\alpha,m'\alpha'}^{B,-} = 2 F^2(k_{m\alpha}) + 2 F^2(k_{m'\alpha'}) - F^2(k_+) - F^2(k_-), \quad (31)$$

with $k_\pm = k_{m\alpha} \pm k_{m'\alpha'}$.

Upon computing numerically the matrix elements $V_{m\alpha;m'\alpha'}^0$ for ribbons of different sizes, we come to the conclusion that they collapse into three types that only depend on the ribbon

TABLE I. Energy differences E_{AB} between levels A and B, in eV $M_y = 4$, $N = 7$ armchair GNRs of different lengths M_x , as obtained from our density functional theory (DFT) and antiferromagnetic mean-field Hubbard (MFH) calculations with $U^0 = 4.9$ eV. First and second column, A = HOMO, B = LUMO. Third and fourth columns, A = HOMO-1, B = LUMO+1. The infinite ribbon DFT gap is 1.559 eV.

M_x	DFT H/L	MFH H/L	DFT H-1/L+1	MFH H-1/L+1
2	0.943	0.958	3.809	3.775
3	0.596	0.600	3.127	3.199
4	0.556	0.534	2.545	2.650
5	0.546	0.504	2.210	2.237
6	0.545	0.491	2.017	1.975
7	0.546	0.484	1.898	1.801
8	0.547	0.482	1.817	1.681
9	0.547	0.481	1.760	1.595
10	0.547	0.480	1.719	1.532

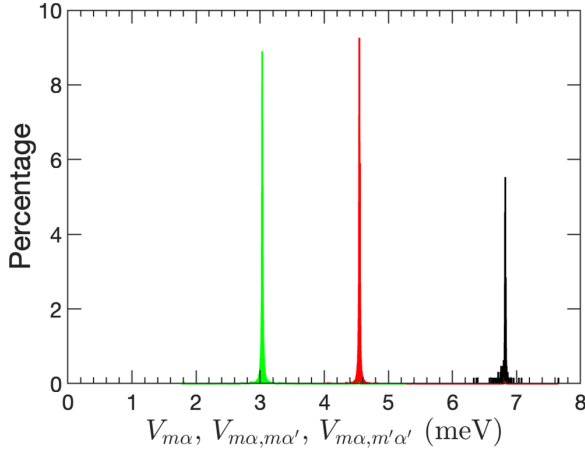


FIG. 2. Histograms of the Coulomb matrix elements of an armchair ribbon of width $M_y = 5$ ($N = 9$) and length $M_x = 80$ ($L = 34.1$ nm), as a function of energy measured in meV, for the four bands $m = 1, \dots, 4$ inter all wave vectors $\alpha = 1, \dots, 2M_x - 1$. $V_{m\alpha}$ is shown in black; the peak is placed at 6.8 meV. Intraband $V_{m\alpha, m\alpha'}$ and interband $V_{m\alpha, m'\alpha'}$ are shown in red and green, respectively. The two peaks are placed at 4.53 and 3.02 meV.

size: same-state integrals $V_{m\alpha}^0 = V_{aa}^0 = \mathcal{U}(M_x, M_y)$, same-band integrals $V_{m\alpha, m\alpha'}^0 = \mathcal{V}(M_x, M_y)$, and the rest $V_{m\alpha, m'\alpha'}^0 = \mathcal{W}(M_x, M_y)$. Consequently, only three Coulomb integrals are needed for each ribbon size. An example of this behavior for an armchair ribbon of size $M_y = 5$ ($N = 9$) and $M_x = 80$ is shown in Fig. 2. We have also found that these three Coulomb integrals follow closely the ratios $\mathcal{V} = 2/3\mathcal{U}$ and $\mathcal{W} = 2/3\mathcal{V}$, up to an accuracy of less than 1%, as can also be inferred from the same figure. As a consequence, it suffices to determine one of the three, and we choose $\mathcal{U}$. We have fitted $\mathcal{U}$ for armchair ribbons with widths and lengths ranging from $M_y = 3$ to 9 and $M_x = 3$ to 110, respectively, and have found that the size-scaling relationship

$$\mathcal{U} = \frac{3.19}{(1 + 1.156M_x)M_y} \text{ eV} \quad (32)$$

is accurate up to 1% or 2%.

B. Mean-field solution for undoped armchair ribbons

We have performed the Kanamori mapping for undoped ribbons in a previous article [13]. There, we kept both edge

TABLE II. Same as in Table I, with $M_y = 5$, $N = 9$. The infinite DFT gap is in this case 0.830 eV.

M_x	DFT H/L	MFH H/L	DFT H-1/L+1	MFH H-1/L+1
2	0.699	0.745	2.760	2.662
3	0.663	0.678	2.075	2.063
4	0.660	0.672	1.713	1.749
5	0.659	0.658	1.498	1.558
6	0.658	0.658	1.357	1.431
7	0.657	0.657	1.258	1.341
8	0.658	0.657	1.186	1.275
9	0.657	0.657	1.133	1.225
10	0.658	0.657	1.088	1.186

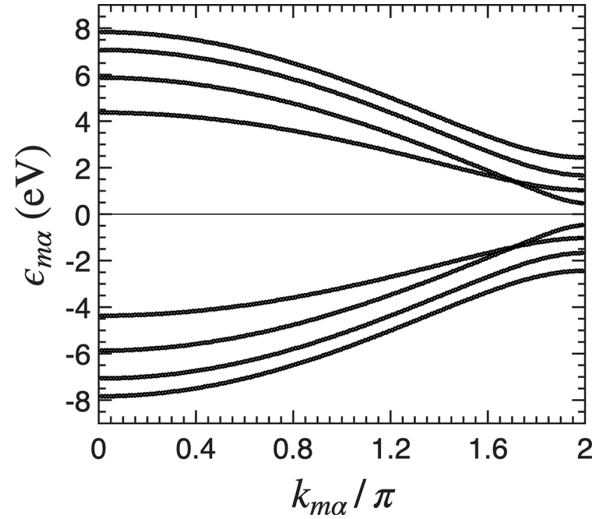


FIG. 3. Bulk eigenenergies $\epsilon_{m\alpha}^{MF}$ of the antiferromagnetic mean-field solution of an armchair ribbon of width $M_y = 5$ ($N = 9$) and length $M_x = 80$ ($L = 34.1$ nm) [13]. These eigenenergies are discrete; they resemble bands because of the close spacing of the wave vectors $k_{m\alpha}$ for this ribbon.

and bulk states but dropped the exchange and pair-hopping terms. We performed an analytical mean-field treatment of the Hamiltonian and found paramagnetic, ferromagnetic, and antiferromagnetic solutions. These solutions agreed with previous numerical calculations carried out for infinite-length ribbons, both for the bulk and for the edge bands. We also found that the three solutions delivered the same properties for bulk states, and only differed on the nature and energies of the edge states. Specifically, mean-field bulk states kept a one-to-one relationship with their noninteracting counterparts: they retained the quantum numbers $(m\alpha\tau\sigma)$ and were spin degenerate. All in all, bulk states could still be classified by *shells* labeled by $(m\alpha\tau)$. We also found that the filled bulk mean-field states undergo an energy shift,

$$\epsilon_{m\alpha}^0 \rightarrow \epsilon_{m\alpha\sigma}^{MF} = \epsilon_{m\alpha}^0 + \mathcal{U} n_{m\alpha-\sigma} + \sum_{occ} V_{m\alpha, m'\alpha'}^0 n_{m'\alpha'}, \quad (33)$$

where the sum runs over all occupied states. The energy shift is essentially the same for all bulk states, and so it amounts to a rigid shift of all the bulk bands. These mean-field bulk energies are illustrated in Fig. 3 for a $M_y = 5$ ribbon of length $M_x = 80$. The mean-field eigenenergies can be used to determine the single-particle addition energies $\Delta\epsilon_{m\alpha, m'\alpha'} = \epsilon_{m\alpha}^{MF} - \epsilon_{m'\alpha'}^{MF}$ between consecutive-energy states. Notice that the two single-particle energies can belong to the same band or two different bands. The first case leads to a uniform pattern, while the second follows a random distribution. This behavior is illustrated in Fig. 4 for the same ribbon, with $M_y = 5$ and $M_x = 80$. Panel (a) shows $\Delta\epsilon_{ab}$, where the uniform patterns belonging to same-band pairs can be appreciated; superimposed to them, the random cloud of addition energies correspond to different-band pairs. Panel (b) shows the histogram of addition energies. This histogram resembles a Wigner-Dyson random distribution, although we do not pursue this issue further due to the limited number of counts. Instead, we would like to highlight the large number

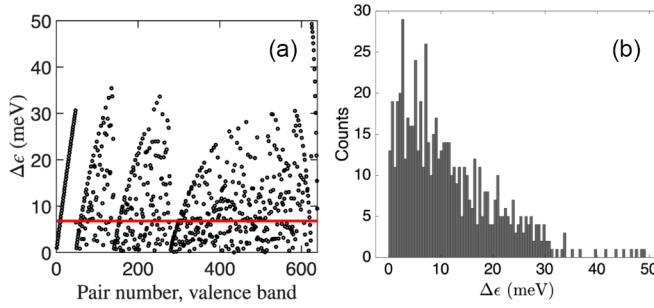


FIG. 4. (a) Mean-field single-particle energy differences $\Delta\epsilon_{ab} = \epsilon_a^{\text{MF}} - \epsilon_b^{\text{MF}}$ between consecutive-energy bulk states belonging to the valence band, all extracted from Fig. 3. The horizontal red line is placed at an energy equal to $V = 6.8$ meV, which corresponds to the Coulomb matrix element $\mathcal{U}$ of this ribbon. (b) Histogram of the same energy differences.

of counts with $\Delta\epsilon_{ab} < \mathcal{U} \approx 6.8$ meV. This means that the electronic correlations of ribbons of these widths and lengths belong to the intermediate regime, and may lead to interesting behavior beyond that expected for weak electron correlations. This intermediate regime behavior will be exploited and analyzed in the rest of this article.

V. LOW-ENERGY HAMILTONIAN

A. Hamiltonian

We remind the reader that we analyze in this article situations where the chemical potential μ lies well within the bands of bulk single-particle states. With these conditions, bulk shells with energies sufficiently below μ are filled and inert, and we call them *closed shells*. Similarly, bulk shells with energies sufficiently above μ are empty and inert, and similar words can be said for the ribbon edge shells, provided that they exist. “Inert” means here that virtual transitions towards or from those states, triggered by either quantum or thermal fluctuations, have negligible probabilities. As a consequence, we expect that only a few shells will be active for a given value of the chemical potential, and we call them *open shells*. Therefore, when a gate voltage is swept, shells are opened and closed sequentially.

We expect that a mean-field treatment should be sufficient to describe closed shells. However, shells that are close enough to the chemical potential might be open or closed depending on the delicate interplay between kinetic and Coulomb energies. We thus discard from the Kanamori Hamiltonian those closed shells whose mean-field energies are sufficiently far from the chemical potential and keep only a small number O of shells with mean-field energies around μ . We thus arrive to the following low-energy many-body Hamiltonian that addresses cleanly these O putative open shells,

$$\begin{aligned} \hat{H} = & \sum_{a \in O} \epsilon_a^{\text{MF}} \hat{n}_a + \mathcal{U} \sum_{a \in O} \hat{n}_{a\uparrow} \hat{n}_{a\downarrow} \\ & + \sum_{a,b \in O} V_{ab}^0 \left(\frac{\hat{n}_a \hat{n}_b}{4} - \hat{\mathbf{S}}_a \cdot \hat{\mathbf{S}}_b + \frac{\hat{\Delta}_a^\dagger \hat{\Delta}_b + \hat{\Delta}_b^\dagger \hat{\Delta}_a}{2} \right) \\ \epsilon_a^{\text{MF}} = & \epsilon_a^0 + \sum_{b \in \text{occ}} V_{ab}^0 n_b \end{aligned} \quad (34)$$

where the same-state, opposite-spin interaction of the open shells is kept out of the mean-field treatment. The Coulomb integrals V_{ab}^0 are equal to $\mathcal{V} = 2/3 \mathcal{U}$ or $\mathcal{W} = (2/3)^2 \mathcal{U}$ depending on whether a and b belong, or not, to the same band, respectively.

The Fock space is spanned in the single-particle number basis $\{|n_{i\sigma}\rangle = |n_{1\uparrow}, n_{1\downarrow}, \dots, n_{O\uparrow}, n_{O\downarrow}\rangle\}$ and has dimension 4^O . The many-body Hamiltonian commutes with the total number operator $\hat{N}$ and with the total spin operators $\hat{S}_T^2, \hat{S}_T^z$. As a consequence the Fock space can be classified into $2O + 1$ subspaces labeled by $N = 0, \dots, 2O$. Each of these subspaces can be further split into subspaces with given S . We find that there exists frequently more than one subspace with quantum numbers (N, S) . Therefore, an additional quantum number R needs to be added to identify unambiguously each subspace (N, S, R) . Each of these subspaces contains $2S + 1$ eigenstates. The highest-spin subspace available occurs at half-filling $N = O$ and the spin equals $S_T = O/2$. The ribbon many-body eigenstates are decomposed in the single-particle basis as

$$|N\lambda\rangle = \sum |n_{i\sigma}\rangle (n_{i\sigma} |N\lambda\rangle) = \sum |n_{i\sigma}\rangle \mathcal{R}_{n_{i\sigma}, \lambda}^N, \quad (35)$$

where λ is shorthand for the many-body quantum numbers (R, S, S^z) , and where the sum is restricted to the combinations that sum up to N . The expansion coefficients define the matrices $\mathcal{R}^N$ that diagonalize the Hamiltonian within each N subspace. The diagonalized Hamiltonian is

$$\hat{H} = \sum_{N, \lambda} E_{N, \lambda} \hat{\Gamma}_{N, \lambda}^\dagger \hat{\Gamma}_{N, \lambda}, \quad (36)$$

where the linear operator $\hat{\Gamma}_{N, \lambda}^\dagger$ creates the ribbon eigenstate (N, λ) .

The low-energy many-body Hamiltonian describes adequately the competition between open- and closed-shell behavior, beyond a mean-field treatment. This competition is driven by the interplay between $\Delta\epsilon_{ab} = \epsilon_b^{\text{MF}} - \epsilon_a^{\text{MF}}$, $\mathcal{U}$, and V_{ab}^0 . We discuss below the conditions needed for two shells a, b to be open. The exchange term in Hamiltonian equation (34) promotes that S_T is maximized if the two shells are open, because V_{ab}^0 is always positive.

The low-energy Hamiltonian can be diagonalized analytically for $O = 2$ or 3 , but becomes analytically unmanageable for larger O because of the exponential size growth of its Fock space. However, the mechanisms leading to open-shell and high-spin behavior are captured already for $O = 2$ and $O = 3$. These mechanisms are discussed in the following two sections. The conditions that will be found below for the existence of open-shell high-spin states are equivalent to similar conditions that have been established for the Hubbard model [22,23]. Those results are also akin to the mesoscopic Stoner instabilities predicted to exist in open quantum dots [24,27].

B. Two-shell model

The two-shell model illustrates how more than one shell can be open at the same time. The Hamiltonian is

$$\begin{aligned} \hat{H} = & \epsilon_1 \hat{n}_1 + \mathcal{U} \hat{n}_{1\uparrow} \hat{n}_{1\downarrow} + \epsilon_2 \hat{n}_2 + \mathcal{U} \hat{n}_{2\uparrow} \hat{n}_{2\downarrow} \\ & + V_{12} \left(\frac{\hat{n}_1 \hat{n}_2}{2} - 2 \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 + \hat{\Delta}_1^\dagger \hat{\Delta}_2 + \hat{\Delta}_2^\dagger \hat{\Delta}_1 \right), \end{aligned} \quad (37)$$

where we have dropped the superscripts MF and 0, and where we assume that $\epsilon_1 < \epsilon_2$. In addition, V_{12} can be equal to $\mathcal{V}$ or $\mathcal{W}$, depending on whether 1 and 2 belong to the same band or not. The Fock space has five subspaces labeled by $N = 0, 1, 2, 3, 4$. The highest-spin subspace $S_T = 1$ is achieved at half filling $N = 2$. The ground state for $N = 1$ corresponds to the open-shell doublet $|1, 0, 0, 0\rangle, |0, 1, 0, 0\rangle$ with energy $E_{1,1,1/2,\pm 1/2} = \epsilon_1$. The subspace $N = 2$ has two closed-shell singlets with energies and wave functions,

$$E_{2,1-2,0,0} = \epsilon_1 + \epsilon_2 + \mathcal{U} \pm \sqrt{(\Delta\epsilon_{21})^2 + V_{12}^2}, \quad (38)$$

$$|2, 1 - 2, 0, 0\rangle = A_+ |1, 1, 0, 0\rangle \pm A_- |0, 0, 1, 1\rangle, \quad (39)$$

$$A_{\pm} = \frac{1}{\sqrt{2}} \left(1 \mp \frac{\Delta\epsilon_{21}}{\sqrt{(\Delta\epsilon_{21})^2 + V_{12}^2}} \right)^{1/2}, \quad (40)$$

with $\Delta\epsilon_{21} = \epsilon_2 - \epsilon_1$. The singlet $|2, 2, 0, 0\rangle$ has a higher weight in the single-particle state $|1, 1, 0, 0\rangle$ and vice versa. The subspace also contains an open-shell triplet,

$$E_{2,1,1,1-3} = \epsilon_1 + \epsilon_2, \quad (41)$$

$$|2, 1, 1, 1\rangle = |1, 0, 1, 0\rangle, \quad (42)$$

$$|2, 1, 1, 0\rangle = \frac{|1, 0, 0, 1\rangle + |0, 1, 1, 0\rangle}{\sqrt{2}}, \quad (43)$$

$$|2, 1, 1, -1\rangle = |0, 1, 0, 1\rangle, \quad (44)$$

and an open-shell singlet,

$$E_{2,3,0,0} = \epsilon_1 + \epsilon_2 + 2V_{12}, \quad (45)$$

$$|2, 3, 0, 0\rangle = \frac{|1, 0, 0, 1\rangle - |0, 1, 1, 0\rangle}{\sqrt{2}}. \quad (46)$$

The open-shell singlet-triplet energy difference defines the exchange energy,

$$J = E_{\text{singlet}} - E_{\text{triplet}} = -2V_{12}, \quad (47)$$

where the convention is that $J > 0$ means ferromagnetic alignment.

On the other hand, the ground state is either the closed-shell singlet $|2, 2, 0, 0\rangle$ or the open-shell triplet $|2, 1, 1, 1 - 3\rangle$ depending on whether the *effective* exchange constant

$$\mathcal{J} = E'_{\text{singlet}} - E_{\text{triplet}} = \mathcal{U} - \sqrt{(\Delta\epsilon_{21})^2 + V_{12}^2} \quad (48)$$

is smaller or larger than zero, respectively. Notice that if the two shells are open at the same time, then the effective exchange is positive (e.g., ferromagnetic) and the ground state has high spin $S = 1$. The full level ordering in the $N = 2$ subspace is illustrated in Fig. 5.

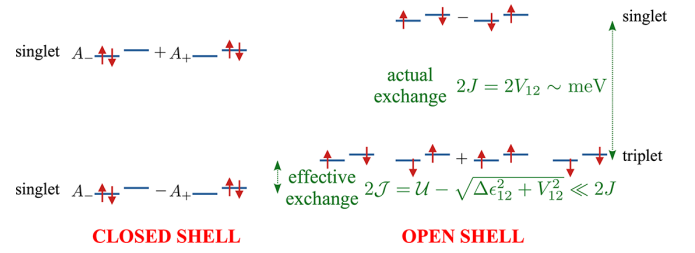


FIG. 5. Illustration of the level ordering for the $N = 2$ subspace of the two-shell model.

C. Three-shell model

The three-shell model describes how a higher-spin $S = 3/2$ can emerge in an armchair ribbon. The Hamiltonian is

$$\begin{aligned} \hat{H} = & \sum_{i=1,2,3} (\epsilon_i \hat{n}_i + \mathcal{U} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}) \\ & + V_{12} \left(\frac{\hat{n}_1 \hat{n}_2}{2} - 2 \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 + \hat{\Delta}_1^\dagger \hat{\Delta}_2 + \hat{\Delta}_2^\dagger \hat{\Delta}_1 \right) \\ & + V_{13} \left(\frac{\hat{n}_1 \hat{n}_3}{2} - 2 \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_3 + \hat{\Delta}_1^\dagger \hat{\Delta}_3 + \hat{\Delta}_3^\dagger \hat{\Delta}_1 \right) \\ & + V_{23} \left(\frac{\hat{n}_2 \hat{n}_3}{2} - 2 \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_3 + \hat{\Delta}_2^\dagger \hat{\Delta}_3 + \hat{\Delta}_3^\dagger \hat{\Delta}_2 \right), \end{aligned} \quad (49)$$

with $\epsilon_1 < \epsilon_2 < \epsilon_3$. In addition, V_{ab} can be equal to $\mathcal{V}$ or $\mathcal{W}$, depending on whether a and b belong to the same band or not. The Hamiltonian is diagonalized in detail in the Appendix. The Fock space has seven subspaces labeled by $N = 0, 1, \dots, 6$. The highest spin subspace $S_T = 3/2$ is achieved at half filling $N = 3$.

We discuss now the conditions for *shell* opening and closing for the $N = 2$ subspace. The subspace contains three triplet ($S = 1$) and six singlet eigenstates. We analyze the limit $\epsilon_3 \gg \epsilon_{1,2}$ to provide a simpler discussion. The two relevant shells are then $i = 1, 2$. We find that the lowest singlet corresponds always to a closed-shell situation, while the triplets correspond to open shells. The energy difference between the lowest-lying triplet and singlet states then tells whether the two shells are open or closed, and also defines the effective exchange constant,

$$\mathcal{J} = E_{\text{singlet}} - E_{\text{triplet}} = \mathcal{U} - \sqrt{(\Delta\epsilon_{21})^2 + V_{12}^2}. \quad (50)$$

In other words, if the two shells are open, then the effective exchange is ferromagnetic $\mathcal{J} < 0$, and vice versa.

The subspace $N = 3$ corresponds to the half-filled model. It contains 20 states that are split into eight doublets with $S = 1/2$ and a quadruplet with $S = 3/2$. Six doublets contain one fully occupied, one half-occupied, and one empty shell. The remaining two have three open shells but their eigenenergies are too large. The quadruplet corresponds to the three shells being open at the same time. The quadruplet is the ground

TABLE III. Same as in Table I, with $M_y = 9$, $N = 17$. The infinite DFT gap is 0.114 eV.

M_x	DFT H/L	MFH H/L	DFT H-1/L+1	MFH H-1/L+1
2	0.793	0.706	0.848	0.931
3	0.624	0.609	0.884	0.919
4	0.592	0.579	0.892	0.914
5	0.585	0.570	0.804	0.807
6	0.584	0.567	0.666	0.678
7	0.566	0.566	0.583	0.585
8	0.490	0.514	0.584	0.566
9	0.431	0.458	0.584	0.566
10	0.384	0.414	0.584	0.566

state if the conditions

$$\mathcal{U} + \frac{V_{ab} + V_{cb}}{2} > \sqrt{\left(\Delta\epsilon_{ac} + \frac{V_{ab} - V_{cb}}{2}\right)^2 + (V_{ac})^2} \quad (51)$$

for the three possible configurations $a \neq b \neq c = 1, 2, 3$ are fulfilled at the same time. The tightest of these conditions defines the energy difference between the lowest quadruplet and the lowest doublet, and the effective exchange constant $\mathcal{J}$. Therefore if the three shells are open, then $\mathcal{J}$ is ferromagnetic; otherwise one shell is closed, another is empty, and a third is half filled. In addition, several of the doublet states contain one full or one empty shell. The model could in these cases be reduced to a simpler two-shell model. Furthermore, this feature illustrates the mechanism by which a model containing a large number of shells folds into a simpler effective model with fewer shells.

The discussion above can be extended to larger O -shell models to determine the conditions for the existence of spin- $O/2$ states, as well as the probability of finding them in a given GNR of size (M_x, M_y) . The exponential growth of the Fock space prevents, however, the analytical determination of these conditions and probabilities. The whole discussion, however, shows strong indications that states with spin as large as 3 or even 4 may be found in GNRs.

D. Example: A $N = 9$ armchair ribbon

To provide a more explicit example, we discuss the armchair GNR with $N = 9$, $M_x = 80$ introduced in Fig. 3 in view of the three-shell model, although this discussion applies to any ribbon with similar widths and lengths, as shown in Table IV. Our estimates indicate that this freestanding ribbon has a Coulomb $\mathcal{U} \approx 6.8$ meV. Shell pairs or triples with energy differences smaller than 6.8 meV are candidates to

TABLE IV. Percentage of states fulfilling the conditions for $S = 1$ ($S = 3/2$) for the half-filled two-shell (three-shell) models in a doped armchair GNR of length M_x and width M_y . Numbers inside the parentheses indicate the percentages when an effective dielectric constant $\epsilon = 2$ is taken into account.

M_x	10	100	500
M_y			
4	22 (12) [7 (0)]	28 (14) [15 (4)]	28 (15) [17 (6)]
5	29 (14) [13 (4)]	30 (16) [19 (6)]	31 (17) [19 (7)]
9	34 (20) [19 (4)]	36 (17) [24 (8)]	36 (21) [25 (9)]
16	31 (18) [19 (3)]	39 (21) [27 (8)]	40 (22) [29 (10)]
26	34 (20) [24 (8)]	41 (23) [30 (11)]	42 (24) [31 (12)]
36	38 (19) [24 (7)]	41 (23) [30 (11)]	42 (24) [32 (12)]

show open-shell high-spin behavior, according to Eqs. 50 and 51. The two panels in Fig. 4 show that a significant fraction of shell pairs may fulfill these conditions. We have determined that the fraction of shells fulfilling the condition (50) is $P_{S=1} \approx 29\%$. Similarly, we have also found that a fraction $P_{S=3/2} \sim 17\%$ of the shells fulfills the three conditions in Eq. (51) for the existence of $S = 3/2$ states in this ribbon. We have finally computed these two fractions for GNRs of different sizes to estimate the likelihood to find high-spin open-shell behavior. The results are summarized in Table IV and demonstrate that GNRs should contain a non-negligible number of open shells. The table indicates that the chances to display double and triple open-shell behavior increase, but saturate, with the length and width of the ribbon.

Graphene ribbons are, however, experimentally addressed when they are deposited on a substrate. Furthermore, in electrical experiments, the ribbons are contacted at the two sides by metallic electrodes. To provide a phenomenological understanding of these substrate and contact effects, we have included an effective dielectric constant ϵ in our calculations, so that $U^{\text{eff}} = U^0/\epsilon$. Table IV shows in parentheses the percentages found when $\epsilon = 2$. Dielectric screening reduces the fraction of open shells because it decreases the size of the effective Coulomb matrix elements.

VI. ELECTRONIC TRANSPORT THROUGH DOPED GRAPHENE NANORIBBONS

Graphene ribbons may have use in different technologies, and we single out here their possible use as channel components in nanoelectronics [5–7,20]. Electronic transport through a ribbon connected to two electrodes $A = L, R$ is governed by the transfer Hamiltonian,

$$\hat{H}_t = \sum_A \sum_{k,i,\sigma} (\mathcal{V}_{A,k,i,\sigma} \hat{r}_{i\sigma}^\dagger \hat{c}_{A,k\sigma} + \text{H.c.}) = \sum_A \sum_{k,\sigma,\lambda,\lambda'} (\mathcal{V}_{A,k\sigma;N\lambda \rightarrow N+1\lambda'} \hat{r}_{N+1\lambda'}^\dagger \hat{r}_{N\lambda} \hat{c}_{A,k\sigma} + \text{H.c.}) \quad (52)$$

where $\hat{c}_{A,k\sigma}^\dagger$ creates an electron with spin σ and wave vector k in electrode A and $\mathcal{V}_{A,k,i\sigma}$ denotes a single-particle transfer matrix element. The matrix elements are written in the many-body eigenstate basis as follows,

$$\mathcal{V}_{A,k\sigma;N\lambda \rightarrow N+1\lambda'} = \sum_i \mathcal{V}_{A,k,i\sigma} M_{\lambda'\lambda}^N(i, \sigma), \quad (53)$$

$$M_{\lambda'\lambda}^N(i, \sigma) = \langle N + 1\lambda' | \hat{r}_{i\sigma}^\dagger | N\lambda \rangle = (\mathcal{R}^{N+1})_{\lambda'n'}^\dagger (n' | \hat{r}_{i\sigma}^\dagger | n) \mathcal{R}_{n\lambda}^N, \quad (54)$$

where we have made use of Eq. (35). The transfer rates in the single electron transfer regime (SET) are

$$\gamma_{A,N\lambda \rightarrow N+1\lambda'} = \frac{2\pi}{\hbar} g_A(\mu_A) f(E_{N+1\lambda'} - E_{N\lambda} - \mu_A) \times \sum_{i\sigma} |\mathcal{V}_{A,k_F,i,\sigma}|^2 |M_{\lambda'\lambda}^N(i, \sigma)|^2, \quad (55)$$

$$\gamma_{A,N\lambda \rightarrow N-1\lambda'} = \frac{2\pi}{\hbar} g_A(\mu_A) [1 - f(E_{N\lambda} - E_{N-1\lambda'} - \mu_A)] \times \sum_{i\sigma} |\mathcal{V}_{A,k_F,i,\sigma}|^2 |M_{\lambda'\lambda}^{N-1}(i, \sigma)|^2, \quad (56)$$

where $g_A(\mu_A)$ is the density of states at the chemical potential μ_A of the A electrode.

The above expressions for electron transfer together with the conditions for open-shell behavior found in this article lead to a clear mechanism for spin and shell blockade in the transport response of nanoribbons. The mechanism for shell blockade in the SET regime is illustrated with the following example: the transition matrix element $\langle 1, 1, 0, 0, 0, 0 | \hat{r}_{i\sigma}^\dagger | 0, 0, 1, 0, 0, 0 \rangle$ is always zero because the transition needs two electrons to change their single-particle state simultaneously. Similarly, the origin of the mechanism for spin blockade in the SET regime lies in spin conservation. For example, the transition matrix element $\langle N = 2, R, S = 0, S_z | \hat{r}_{i\sigma}^\dagger | N = 3, R', S = 3/2, S'_z \rangle$ is always zero because a single electron can raise the spin by only 1/2.

The explicit expressions for the three-shell model are

$$\gamma_{A,N\lambda \rightarrow N+1\lambda'} = \gamma_{A,1} |M_{\lambda'\lambda}^N(1)|^2 + \gamma_{A,2} |M_{\lambda'\lambda}^N(2)|^2 + \gamma_{A,3} |M_{\lambda'\lambda}^N(3)|^2, \quad (57)$$

where we have defined $\gamma_{A,i} = \frac{2\pi}{\hbar} g_A(\mu_A) |\mathcal{V}_{A,k_F,i}|^2$ and $|M_{\lambda'\lambda}^N(i)|^2 = |M_{\lambda'\lambda}^N(i, \uparrow)|^2 + |M_{\lambda'\lambda}^N(i, \downarrow)|^2$.

VII. CONCLUSIONS

In conclusion, we have addressed the electronic behavior of freestanding doped GNRs using an extended Hubbard model. We have performed an exact mapping of the model to a Kanamori Hamiltonian. We have reduced the range of the Hubbard parameters, and showed that an on-site Hubbard model with $U \approx 4.9$ eV fits well the low-lying mean-field solutions to DFT simulations of ribbons of different widths and lengths. We have determined, from the on-site model, analytical equations for the Coulomb matrix elements. We have found that those matrix elements follow a remarkable scaling law, and that they collapse into just three for each ribbon. We have then introduced an effective low-energy Hamiltonian to address the many-body behavior of ribbons where the chemical potential lies inside the bulk bands. The model allows us to unveil a robust mechanism that stabilizes open-shell high-spin states. We have estimated the percentage of cases where open-shell high-spin states are expected to exist. These percentages increase but saturate with the ribbon lengths and

widths and decrease when an effective dielectric constant is included in the calculations.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX: DIAGONALIZATION OF THE THREE-SHELL MODEL

The Hamiltonian for the three-shell model is

$$\hat{H} = \sum_{i=1,2,3} (\epsilon_i \hat{n}_i + U \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}) + V_{12} \left(\frac{\hat{n}_1 \hat{n}_2}{2} - 2 \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 + \hat{\Delta}_1^\dagger \hat{\Delta}_2 + \hat{\Delta}_2^\dagger \hat{\Delta}_1 \right) + V_{13} \left(\frac{\hat{n}_1 \hat{n}_3}{2} - 2 \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_3 + \hat{\Delta}_1^\dagger \hat{\Delta}_3 + \hat{\Delta}_3^\dagger \hat{\Delta}_1 \right) + V_{23} \left(\frac{\hat{n}_2 \hat{n}_3}{2} - 2 \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_3 + \hat{\Delta}_2^\dagger \hat{\Delta}_3 + \hat{\Delta}_3^\dagger \hat{\Delta}_2 \right). \quad (A1)$$

The states are ordered so that $\epsilon_1 < \epsilon_2 < \epsilon_3$. The Fock space $\mathcal{F}$ has dimensions $4^3 = 64$ and is spanned by the single-particle basis $\{|n_{1\uparrow}, n_{1\downarrow}, n_{2\uparrow}, n_{2\downarrow}, n_{3\uparrow}, n_{3\downarrow}\rangle\}$. The Hamiltonian commutes with the total number operator $\hat{N}$. $\hat{N}$ has eigenvalues $N = 0, 1, \dots, 6$, so that $\mathcal{F}$ is split into seven subspaces labeled by N . The Hamiltonian also commutes with total spin operator $\hat{S}^2$, so that each of those subspaces is further split into subspaces labeled by S . However, $\hat{H}$, $\hat{N}$, and $\hat{S}$ do not constitute a complete set of commuting observables because there exist for some of the subspaces more than one same- S multiplet. We label these different subspaces with an additional integer number R whenever they exist. All in all, we use the following notation to describe the Hamiltonian eigenstates: $|N, R, S, S^z\rangle$. We describe below the subspaces $N = 0, 1, 2$, and 3 , their eigenstates and their energies. Subspaces $N = 4, 5$, and 6 are easily deduced from the electron-hole symmetry of the Hamiltonian. The highest-spin subspace occurs at half filling, $N = 3$, and corresponds to a quadruplet with $S = 3/2$. We denote $\Delta\epsilon_{ab} = \epsilon_a - \epsilon_b$.

(1) Subspace $N = 0$. This trivial subspace is one-dimensional and contains only the vacuum state:

$$|0, 0, 0\rangle = |000000\rangle E = 0.$$

(2) Subspace $N = 1$. This subspace has dimensions equal to 6 and is made up of the following three doublets:

$$|1, 1, 1/2, 1/2\rangle = |100000\rangle \quad E = \epsilon_1,$$

$$|1, 1, 1/2, -1/2\rangle = |010000\rangle \quad E = \epsilon_1,$$

$$|1, 2, 1/2, 1/2\rangle = |001000\rangle \quad E = \epsilon_2,$$

$$|1, 2, 1/2, -1/2\rangle = |000100\rangle \quad E = \epsilon_2,$$

$$|1, 3, 1/2, 1/2\rangle = |000010\rangle \quad E = \epsilon_3,$$

$$|1, 3, 1/2, -1/2\rangle = |000001\rangle \quad E = \epsilon_3.$$

(3) Subspace $N = 2$. This subspace has dimensions equal to 15 and is made up of six singlets and three triplets. The Hamiltonian for the basis states $|1, 1, 0, 0, 0, 0\rangle$, $|0, 0, 1, 1, 0, 0\rangle$, and $|0, 0, 0, 0, 1, 1\rangle$ is

$$\begin{pmatrix} 2\epsilon_1 + \mathcal{U} & V_{12} & V_{13} \\ V_{12} & 2\epsilon_2 + \mathcal{U} & V_{23} \\ V_{13} & V_{23} & 2\epsilon_3 + \mathcal{U} \end{pmatrix}. \quad (\text{A2})$$

Solving the eigenvalue equation yields a complicated cubic equation, whose explicit solution delivers three eigenenergies. The corresponding three eigenstates are singlets of the form

$$|2, R, 0, 0\rangle = A_{R1}|1, 1, 0, 0, 0, 0\rangle + A_{R2}|0, 0, 1, 1, 0, 0\rangle + A_{R3}|0, 0, 0, 0, 1, 1\rangle,$$

where $R = 1, 2, 3$. The remaining three singlet eigenstates and eigenenergies are

$$|2, 4, 0, 0\rangle = \frac{|100100\rangle - |011000\rangle}{\sqrt{2}} \quad E = \epsilon_1 + \epsilon_2 + 2V_{12},$$

$$|2, 5, 0, 0\rangle = \frac{|100001\rangle - |010010\rangle}{\sqrt{2}} \quad E = \epsilon_1 + \epsilon_3 + 2V_{13},$$

$$|2, 6, 0, 0\rangle = \frac{|001001\rangle - |000110\rangle}{\sqrt{2}} \quad E = \epsilon_2 + \epsilon_3 + 2V_{23}.$$

The three triplets are

$$|2, 1, 1, 1\rangle = |101000\rangle \quad E = \epsilon_1 + \epsilon_2,$$

$$|2, 1, 1, 0\rangle = \frac{|100100\rangle + |011000\rangle}{\sqrt{2}} \quad E = \epsilon_1 + \epsilon_2,$$

$$|2, 1, 1, -1\rangle = |010100\rangle \quad E = \epsilon_1 + \epsilon_2,$$

$$|2, 2, 1, 1\rangle = |100010\rangle \quad E = \epsilon_1 + \epsilon_3,$$

$$|2, 2, 1, 0\rangle = \frac{|100001\rangle + |010010\rangle}{\sqrt{2}} \quad E = \epsilon_1 + \epsilon_3,$$

$$|2, 2, 1, -1\rangle = |010001\rangle \quad E = \epsilon_1 + \epsilon_3,$$

$$|2, 3, 1, 1\rangle = |001010\rangle \quad E = \epsilon_2 + \epsilon_3,$$

$$|2, 3, 1, 0\rangle = \frac{|000110\rangle + |001001\rangle}{\sqrt{2}} \quad E = \epsilon_2 + \epsilon_3,$$

$$|2, 3, 1, -1\rangle = |000101\rangle \quad E = \epsilon_2 + \epsilon_3.$$

The lowest-lying states are the closed-shell singlet $|2, 1, 0, 0\rangle$ and the open-shell triplet $|2, 1, 1, S^z\rangle$. The condition for the ground state to be a triplet with spin $S = 1$ can be found only numerically.

We analyze here two situations in more detail. The first highlights how effective a ferromagnetic exchange energy arises, and how a third shell may or may not enter into the calculations. The second situation highlights how a shell becomes closed.

(1) Limit $\epsilon_3 \gg \epsilon_2$. Here, the third shell is completely empty. The problem thus reduces to a simpler half-filled two-shell model. The two singlet states simplify to

$$|2, R, 0, 0\rangle = A_{R1}|1, 1, 0, 0\rangle + A_{R2}|0, 0, 1, 1\rangle,$$

where $R = 1, 2$, with eigenenergies

$$E = \epsilon_1 + \epsilon_2 + \mathcal{U} \pm \sqrt{(\Delta\epsilon_{21})^2 + V_{12}^2}.$$

The energy difference between the singlet and the triplet defines an effective exchange coupling

$$\mathcal{J} = \mathcal{U} - \sqrt{(\Delta\epsilon_{21})^2 + V_{12}^2}. \quad (\text{A3})$$

The triplet state $|2, 1, 1, S^z\rangle$ becomes the ground state if $\mathcal{J} > 0$, e.g.: if the effective exchange coupling becomes ferromagnetic.

(2) Limit $\epsilon_1 \ll \epsilon_2$. This limit corresponds to the closing on the lowest shell. Again the equations simplify so that the singlet states are

$$|2, R, 0, 0\rangle = A_{R1}|1, 1, 0, 0\rangle + A_{R2}|0, 0, 1, 1\rangle,$$

where $R = 2, 3$, and with eigenenergies

$$E = \epsilon_2 + \epsilon_3 + \mathcal{U} \pm \sqrt{(\Delta\epsilon_{32})^2 + V_{23}^2}.$$

(1) Subspace $N = 3$. This subspace has dimensions equal to 20 and is made up of eight doublets and a quadruplet. We

list first all the basis states,

$$\begin{aligned}
|A, S^z = 1/2\rangle &= |111000\rangle, & E_A &= 2\epsilon_1 + \epsilon_2 + \mathcal{U} + V_{12}, \\
|B, S^z = -1/2\rangle &= |110100\rangle, & E_B &= E_A, \\
|C, S^z = 1/2\rangle &= |001011\rangle, & E_C &= \epsilon_2 + 2\epsilon_3 + \mathcal{U} + V_{23}, \\
|D, S^z = -1/2\rangle &= |000111\rangle, & E_D &= E_C, \\
|E, S^z = 1/2\rangle &= |110010\rangle, & E_E &= 2\epsilon_1 + \epsilon_3 + \mathcal{U} + V_{13}, \\
|F, S^z = -1/2\rangle &= |110001\rangle, & E_F &= E_E, \\
|G, S^z = 1/2\rangle &= |001110\rangle, & E_G &= 2\epsilon_2 + \epsilon_3 + \mathcal{U} + V_{23}, \\
|H, S^z = -1/2\rangle &= |001101\rangle, & E_H &= E_G, \\
|I, S^z = 1/2\rangle &= |101100\rangle, & E_I &= \epsilon_1 + 2\epsilon_2 + \mathcal{U} + V_{12}, \\
|J, S^z = -1/2\rangle &= |011100\rangle, & E_J &= E_I, \\
|K, S^z = 1/2\rangle &= |100011\rangle, & E_K &= \epsilon_1 + 2\epsilon_3 + \mathcal{U} + V_{13}, \\
|L, S^z = -1/2\rangle &= |010011\rangle, & E_L &= E_K, \\
|M, S^z = 1/2\rangle &= |101001\rangle, & E_M &= \epsilon_a + V_{13} + V_{23}, \\
|N, S^z = 1/2\rangle &= |100110\rangle, & E_N &= \epsilon_a + V_{12} + V_{23}, \\
|O, S^z = 1/2\rangle &= |011010\rangle, & E_O &= \epsilon_a + V_{12} + V_{13}, \\
|P, S^z = -1/2\rangle &= |010110\rangle, & E_P &= \epsilon_a + V_{13} + V_{23}, \\
|Q, S^z = -1/2\rangle &= |011001\rangle, & E_Q &= \epsilon_a + V_{12} + V_{23}, \\
|R, S^z = -1/2\rangle &= |100101\rangle, & E_R &= \epsilon_a + V_{12} + V_{13},
\end{aligned}$$

where $\epsilon_a = \epsilon_1 + \epsilon_2 + \epsilon_3$ and $\epsilon_b = \epsilon_a + (V_{12} + V_{13} + V_{23})$. The first set has six doublets, all of them having one fully occupied, one half-occupied, and one empty shell. The first two doublets in this set are made of the states $|A\rangle$ and $|C\rangle$ for spin $S^z = +1/2$ and $|B\rangle$ and $|D\rangle$ for spin $S^z = -1/2$. They have interstate couplings $V_{AC} = V_{BD} = V_{13}$. Both Hamiltonians are identical, so we write just the first one:

$$\begin{pmatrix} E_A & V_{AB} \\ V_{AB} & E_B \end{pmatrix}. \quad (\text{A4})$$

The doublets' eigenstates and eigenenergies $|N, L, S, S^z\rangle$ are

$$\begin{aligned}
|3, 1, 1/2, 1/2\rangle &= u_+ |A\rangle - u_- |C\rangle, \\
|3, 1, 1/2, -1/2\rangle &= u_+ |B\rangle - u_- |D\rangle, \\
\epsilon_{3,1,1/2,\pm 1/2} &= \frac{E_A + E_C}{2} - \sqrt{\left(\frac{E_C - E_A}{2}\right)^2 + V_{AC}^2} \\
&= \epsilon_a + \mathcal{U} + \frac{V_{12} + V_{23}}{2} \\
&\quad - \sqrt{\left(\Delta\epsilon_{31} + \frac{V_{23} - V_{12}}{2}\right)^2 + V_{13}^2},
\end{aligned}$$

$$|3, 2, 1/2, 1/2\rangle = u_- |A\rangle + u_+ |C\rangle,$$

$$|3, 2, 1/2, -1/2\rangle = u_- |B\rangle + u_+ |D\rangle,$$

$$\begin{aligned}
\epsilon_{3,2,1/2,\pm 1/2} &= \frac{E_A + E_C}{2} + \sqrt{\left(\frac{E_C - E_A}{2}\right)^2 + V_{AC}^2} \\
&= \epsilon_a + \mathcal{U} + \frac{V_{12} + V_{23}}{2} \\
&\quad + \sqrt{\left(\Delta\epsilon_{31} + \frac{V_{23} - V_{12}}{2}\right)^2 + V_{13}^2}.
\end{aligned}$$

The next two doublets are made of the states $|E\rangle$ and $|G\rangle$ for spin $S^z = +1/2$ and $|F\rangle$ and $|H\rangle$ for spin $S^z = -1/2$. Their interstate couplings $V_{EG} = V_{FH} = V_{12}$. The Hamiltonian, eigenstates, and eigenenergies are the same as above upon the replacement $A \rightarrow E, B \rightarrow F, C \rightarrow G$, and $D \rightarrow H$, and the doublets are labeled now as $|3, 3, 1/2, \pm 1/2\rangle$ and $|3, 4, 1/2, 1/2\rangle$. Their eigenenergies are

$$\begin{aligned}
E_{3,3,1/2,\pm 1/2} &= \epsilon_a + \mathcal{U} + \frac{V_{13} + V_{23}}{2} \\
&\quad - \sqrt{\left(\Delta\epsilon_{21} + \frac{V_{23} - V_{13}}{2}\right)^2 + V_{12}^2}, \\
E_{3,4,1/2,\pm 1/2} &= \epsilon_a + \mathcal{U} + \frac{V_{13} + V_{23}}{2} \\
&\quad + \sqrt{\left(\Delta\epsilon_{21} + \frac{V_{23} - V_{13}}{2}\right)^2 + V_{12}^2}.
\end{aligned}$$

The last two doublets in this set are made of the states $|I\rangle$ and $|K\rangle$ for spin $S^z = +1/2$ and $|J\rangle$ and $|L\rangle$ for spin $S^z = -1/2$. Their interstate couplings are $V_{IK} = V_{JL} = V_{23}$. The Hamiltonian, eigenstates, and eigenenergies are again the same as above upon the replacement $A \rightarrow I, B \rightarrow J, C \rightarrow K$, and $D \rightarrow L$, and the doublets are labeled now as $|3, 5, 1/2, \pm 1/2\rangle$ and $|3, 6, 1/2, 1/2\rangle$. Their eigenenergies are

$$\begin{aligned}
E_{3,5,1/2,\pm 1/2} &= \epsilon_a + \mathcal{U} + \frac{V_{13} + V_{12}}{2} \\
&\quad - \sqrt{\left(\Delta\epsilon_{32} + \frac{V_{13} - V_{12}}{2}\right)^2 + V_{23}^2}, \\
E_{3,6,1/2,\pm 1/2} &= \epsilon_a + \mathcal{U} + \frac{V_{13} + V_{12}}{2} \\
&\quad + \sqrt{\left(\Delta\epsilon_{32} + \frac{V_{13} - V_{12}}{2}\right)^2 + V_{23}^2}.
\end{aligned}$$

The last two doublets have three open shells,

$$|3, 7, 1/2, 1/2\rangle = \frac{1}{D_+^{1/2}} [(V_{23} - V_{12} - R) |M\rangle + (V_{13} - V_{23} + R) |N\rangle + (V_{12} - V_{13}) |O\rangle],$$

$$|3, 7, 1/2, -1/2\rangle = \frac{1}{D_+^{1/2}} [(V_{23} - V_{12} - R) |P\rangle + (V_{13} - V_{23} + R) |Q\rangle + (V_{12} - V_{13}) |R\rangle],$$

$$|3, 8, 1/2, 1/2\rangle = \frac{1}{D_-^{1/2}} [(V_{23} - V_{12} + R) |M\rangle + (V_{13} - V_{23} - R) |N\rangle + (V_{12} - V_{13}) |O\rangle],$$

$$|3, 8, 1/2, -1/2\rangle = \frac{1}{D_-^{1/2}} [(V_{23} - V_{12} + R)|P\rangle + (V_{13} - V_{23} - R)|Q\rangle + (V_{12} - V_{13})|R\rangle],$$

$$R = \frac{1}{2} [(V_{12} - V_{13})^2 + (V_{12} - V_{23})^2 + (V_{13} - V_{23})^2]^{1/2},$$

$$D_{\pm} = 2R^{1/2} \left(R \pm \frac{1}{2} (V_{12} + V_{13}) - V_{23} \right)^{1/2}.$$

Their energies are

$$\epsilon_{3,7,\pm 1/2} = \epsilon_b - R,$$

$$\epsilon_{3,8,\pm 1/2} = \epsilon_b + R,$$

The quadruplet eigenstates are

$$|3, 3/2, 3/2\rangle = |1, 0, 1, 0, 1, 0\rangle,$$

$$|3, 3/2, 1/2\rangle = \frac{1}{\sqrt{3}} (|M\rangle + |N\rangle + |O\rangle),$$

$$|3, 3/2, -1/2\rangle = \frac{1}{\sqrt{3}} (|P\rangle + |Q\rangle + |R\rangle),$$

$$|3, 3/2, -3/2\rangle = |0, 1, 0, 1, 0, 1\rangle,$$

and their eigenenergy is $E = \epsilon_a$.

Inspecting the above expressions for the eigenenergies, we come to the conclusion that the two doublets having all open shells have too large eigenenergies. The quadruplet thus competes with the first set of six doublets to be the ground state. The ground state will be the quadruplet state provided that the following three conditions are met together:

$$\mathcal{U} + \frac{V_{12} + V_{23}}{2} - \sqrt{\left(\Delta\epsilon_{31} + \frac{V_{23} - V_{12}}{2}\right)^2 + V_{13}^2} > 0, \quad (\text{A5})$$

$$\mathcal{U} + \frac{V_{13} + V_{23}}{2} - \sqrt{\left(\Delta\epsilon_{21} + \frac{V_{23} - V_{13}}{2}\right)^2 + V_{12}^2} > 0, \quad (\text{A6})$$

$$\mathcal{U} + \frac{V_{13} + V_{12}}{2} - \sqrt{\left(\Delta\epsilon_{32} + \frac{V_{13} - V_{12}}{2}\right)^2 + V_{23}^2} > 0. \quad (\text{A7})$$

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