

Particle View of Many-Body Electronic Structure with Neural Network Wave Function

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In the study of electronic structure, the wave function view dominates the current research landscape and forms the theoretical foundation of modern quantum mechanics. In contrast, valence bond (VB) theory represents chemical bonds as shared electron pairs and can provide an intuitive, particle-based insight into chemical bonding. In this work, using a newly developed periodic dynamic Voronoi Metropolis sampling (PDVMS) method, we project classical many-body electronic configurations from the neural network wave function and apply VB theory to construct a complementary particle-view paradigm. The powerful neural network wave function can achieve near-exact *ab initio* solutions for the ground state of both molecular and solid systems. It allows us to definitively characterize the ground state of benzene by reassessing the competition between its two VB structures. Extending PDVMS to solids, we also predict a spin-staggered VB structure in graphene, which explains emergent magnetic properties in graphene-based nanostructures. Furthermore, this particle view provides insight into the optimization process of the neural network wave function itself. Our work thus introduces a framework for analyzing many-body electronic structure in molecules and solids, opening new avenues for investigating this complex problem and its associated exotic phenomena.

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I. INTRODUCTION

The particle view and the wave function view of electronic structure in molecules and materials form the foundation of condensed matter physics and quantum chemistry. In the prequantum mechanics era, the particle view, treating electrons as charged point particles, predominated in advancing both physics and chemistry [1–6]. However, since the introduction of de Broglie's wave-particle duality [7] and Schrödinger's wave equation [8] in the 1920s, the study of electronic structure has undergone a profound paradigm shift toward the wave function view. Numerous *ab initio* computational methods based on the wave function view, such as post-Hartree-Fock approaches [9] and density functional theory [10], have since been

widely employed to address the electronic structure of complex systems. Nevertheless, all these methods rely on necessary approximations to contend with the Hilbert space dimensionality, which grows exponentially with system size [11]. As a result, the exact determination of eigenstate wave functions remains infeasible for all but the smallest systems.

Recent research has demonstrated that combining neural network representations of many-body wave functions with quantum Monte Carlo methods achieves a favorable balance between computational efficiency and accuracy. These approaches now enable the calculation of nearly exact eigenstate wave functions for molecules and solids containing over 100 electrons [12–19]. Another advantage of the neural network is that it systematically represents the wave function in real space, thereby circumventing the basis set selection dependencies and truncation errors that affect conventional electronic structure methods.

While neural network wave functions can achieve nearly exact results, the wave function view still faces two limitations. First, the optimization of neural network parameters is often treated as a black-box process, which hinders further targeted improvements of the resulting wave function. Second, although the many-body wave function

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theoretically contains all information about a system, its high-dimensional nature makes direct human interpretation intractable. Progress therefore hinges on our ability to distill this vast information into lower-dimensional, intuitive physical pictures. A particularly successful and enduring class of these distilled descriptions is based on particle view, which captures the key chemical and physical information needed to explain diverse phenomena. In the study of topological quantum phases, for instance, the particlelike manipulation of emergent quasiparticles has become essential for the design of fault-tolerant quantum bits [20–22]. Moreover, valence bond theory, treating chemical bonds as localized electron pairs, provides an intuitive framework to understand ground-state bonding geometry through resonance structures and remains widely used in both chemistry and materials science [23–25]. Notably, in condensed matter physics, Anderson’s resonating valence bond theory, initially aimed at explaining high-temperature superconductivity, is now a central framework for understanding other exotic quantum phases, such as quantum spin liquids [26,27].

These dual challenges of restricted optimization improvability and overwhelming informational complexity have motivated the development of complementary approaches that bridge the quantum wave function formalism with the particle-based perspective [28,29]. In this work, we integrate the neural network wave function approach with the periodic dynamic Voronoi Metropolis sampling (PDVMS) method to extract classical electron configurations from nearly exact ground state, as illustrated in Fig. 1. Originally designed for molecular systems, dynamic Voronoi Metropolis sampling (DVMS) has been shown to extract profound chemical insights such as direct visualization of “curly arrows,” which depict electron reorganizations along reaction coordinates [30], and quantitative decomposition of specific bond energies [31]. Given that such a particle view can yield equally profound insights into solid materials, we extend this method to periodic systems for the first time through an improved algorithm named PDVMS. Using this integrated framework, we systematically analyze the ground-state wave function of benzene, trained toward exactness, to unveil the delicate competition between two classical valence bond structures, while also elucidating the contributions of individual determinants within the neural network wave function. Furthermore, we report the first evidence of a spin-staggered valence bond structure in graphene, offering theoretical insights that help explain recent experimental observations.

II. RESULTS AND DISCUSSION

A. Computational framework

Neural-network-based quantum Monte Carlo method, which we used in this work to obtain the ground state of molecules and solids, has achieved great success through

adopting powerful and expressive neural networks [Fig. 1(a)] as wave function ansatz [12–19]. The inherent expressivity of the neural network architecture allows the ansatz to capture the complex correlations within the many-body state, yielding significantly more accurate results than conventional trial wave functions. To treat solid systems, periodicity is incorporated by generalizing the input coordinates $\mathbf{r}$ and distances $r = |\mathbf{r}|$ to their periodic counterparts, $\tilde{\mathbf{r}}$ and $\tilde{r}$ [18]. This powerful method can provide a near-exact ground-state wave function, laying the foundation for high-accuracy extraction of the particle-view picture. The corresponding hyperparameters used for the neural networks can be found in Supplemental Material Note 1 [32].

DVMS derives electron coordinate configurations from a calculated wave function [Fig. 1(b)] [28,29]. Since same-spin electrons are indistinguishable, the many-body probability density $|\psi(\mathbf{r})|^2$ exhibits exact permutation symmetry when their spatial coordinates are swapped. Simply calculating the electron position weighted expectation $\bar{\mathbf{r}} = \mathbb{E}_{\mathbf{r} \sim |\psi|^2}(\mathbf{r})$ would average all same-spin electrons into the same point, failing to describe the electronic structure. To address this, the wave function space should be partitioned into superregions associated with permutation symmetry, commonly referred to as wave function tiles [34]. This partitioning can be achieved using Voronoi diagrams, a geometrical concept that divides space into distinct zones based on the closest distance to a given set of reference sites. Here, these sites correspond to an electronic configuration and all its permutational equivalents. Calculating the position expectation within one such tile effectively treats the electrons as distinguishable, yielding a configuration that reflects the electronic structure. As for the selection of Voronoi sites, DVMS proposes a rule: The Voronoi site should match the position expectation of its corresponding region. This is accomplished by iteratively updating the Voronoi site with the calculated position expectation until convergence (see Supplemental Material Note 3 for more details [32]). Visualizing this position expectation in three-dimensional space directly provides a classical representation of the many-body electronic structure. It is also worth noting that in this work, this framework is applied not only to molecule but also to solid, and this is achieved by extending the DVMS method to periodic systems (dubbed as PDVMS), where two critical methodological developments shall be highlighted (Fig. 2).

The first technical challenge to address is that partitioning wave function space using a traditional Voronoi diagram in periodic systems has fundamental conceptual limitations, as such partitioning fails to reflect the translational symmetry inherent of the system. To resolve this issue, we employ the periodic distance as the distance metric, expressed as

$$\tilde{l}(\mathbf{p}, \mathbf{q}) = \min_{n_1, n_2} \{|\mathbf{p} - \mathbf{q} + n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2|\}, \quad (1)$$

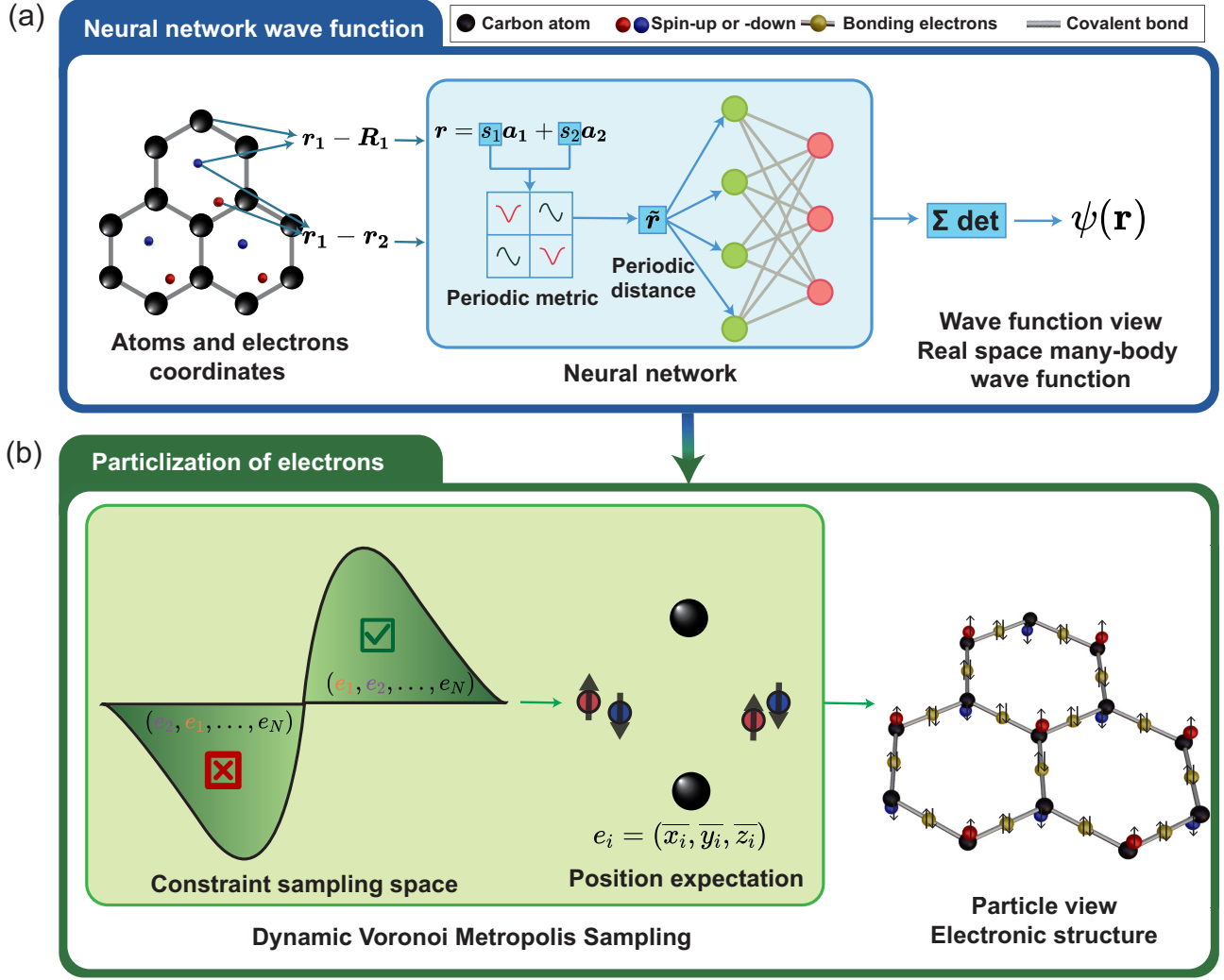


FIG. 1. Illustration of the transition from wave function view to particle view. (a) Illustration of the neural network wave function. The network takes intrinsic atomic coordinates ($\mathbf{R}_i$) in the molecule or solid and sampled electron coordinates ($\mathbf{r}_i$) as direct inputs. For periodic systems defined by lattice vectors ($\mathbf{a}_i$), periodicity is imposed on the electron-nucleus and electron-electron distances used to construct the network's input features. (b) Illustration of particulation of electrons. The DVMS method performs Monte Carlo sampling within a constrained region where electrons are distinguishable, ultimately yielding the classical position expectation of the electrons. By visualizing the electron positions and applying Lewis structure theory, the valence bond structure is obtained.

where $\mathbf{a}_1$ and $\mathbf{a}_2$ are the lattice vectors, and n_1, n_2 are integers. For a system with an N -electron unit cell, let $\mathbf{S} = (\mathbf{S}_1, \dots, \mathbf{S}_N) \in \mathbb{R}^{N \times 3}$ represent the reference configuration (site I), and the corresponding periodic Voronoi cell $\tilde{\Omega}_I$ is then defined as

$$\tilde{\Omega}_I = \{\mathbf{R} = (\mathbf{R}_1, \dots, \mathbf{R}_N) \in \mathbb{R}^{N \times 3} \mid \forall \mathcal{P}, \tilde{l}(\mathbf{R}, \mathbf{S}) \leq \tilde{l}(\mathbf{R}, \mathcal{P}\mathbf{S})\}, \quad (2)$$

where $\mathcal{P}$ denotes permutations of positions among same-spin electrons. Unlike the traditional Voronoi diagram, a cell $\tilde{\Omega}_I$ in the periodic version is no longer a single connected region but splits into infinite disconnected regions related by translational symmetry [Fig. 2(a), red

zones]. We select only one of these regions for the calculation of electron position expectations, as all others are equivalent under translational symmetry.

The second introduced method is a new scheme to calculate the center of mass (c.m.) in periodic systems. Conventionally, the c.m. of a particle ensemble is determined as a weighted average, i.e., $\text{c.m.} = [(\sum m_i \mathbf{X}_i) / \sum m_i]$, where m_i denotes the mass of particle i and $\mathbf{X}_i$ is its spatial coordinates. However, this definition faces a fundamental challenge in periodic systems due to the absence of a well-defined coordinate origin, as illustrated in the upper panel of Fig. 2(b). In the depicted coordinate system, calculating the expectation value of walker positions within a single unit cell yields a c.m. near the cell centroid (red cross), whereas the physically correct c.m. should reside at the red star position.

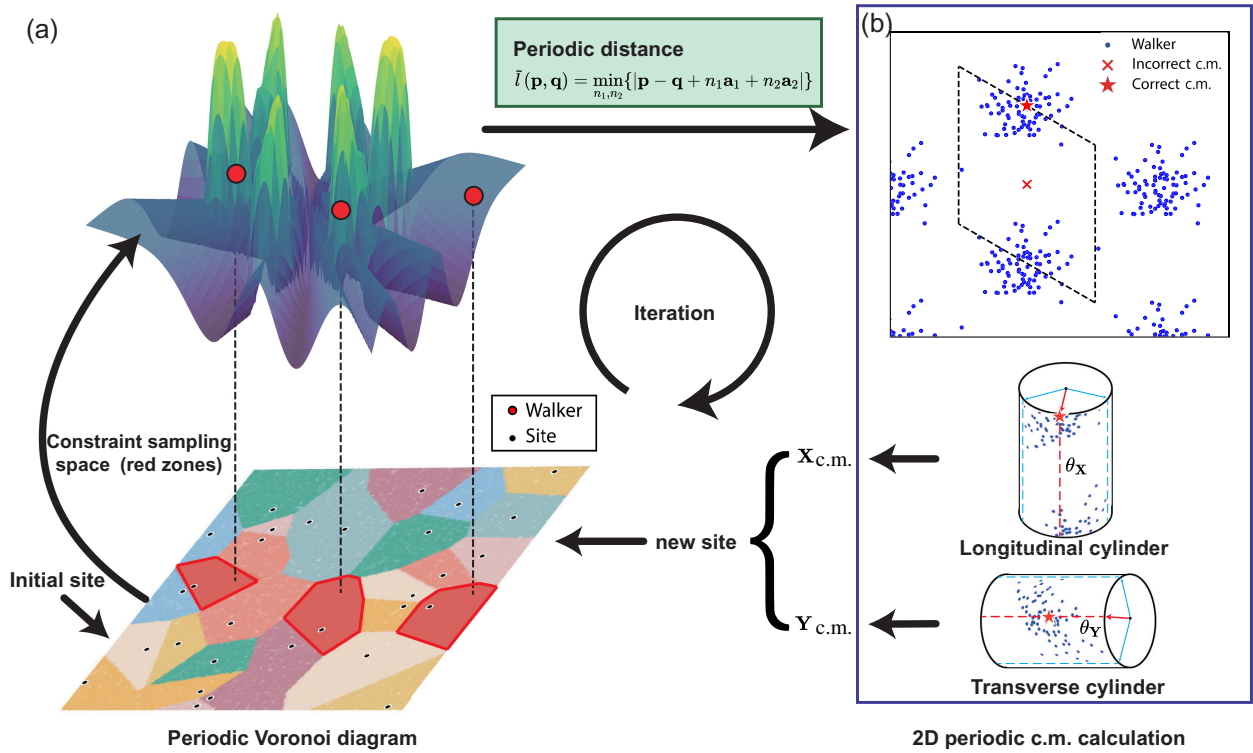


FIG. 2. The process of periodic dynamic Voronoi Metropolis sampling. (a) Illustration of the sampling process. The sampling space is partitioned by a periodic Voronoi diagram generated from the sites, with sampling constrained to specific cells (red regions). The plot illustrates the spatial probability distribution for electrons, where the xy plane represents spatial coordinates and the height along the z axis corresponds to the probability density ($|\Psi|^2$). (b) Sketch of 2D periodic c.m. calculation algorithm. For a periodic distribution of an electron's walkers (blue dots), a naive c.m. calculation can yield an incorrect result (red cross) that is sensitive to the choice of origin. To find the correct c.m. (red star), the periodic cell is mapped to a rectangle via a coordinate transformation. This rectangle is then conceptually wrapped into longitudinal and transverse cylinders. The position expectation of the walkers are inverse mapped to give $\mathbf{X}_{c.m.}$ and $\mathbf{Y}_{c.m.}$, respectively.

To eliminate dependence on the selection of a suitable coordinate origin, we adopt a robust periodic c.m. calculation [35]. Figure 2(b) illustrates the core idea using a two-dimensional system, though the method generalizes naturally to three dimensions. Without loss of generality, any lattice cell can be transformed into a rectangular cell through coordinate mapping. By rolling this rectangle along its two orthogonal periodic directions, we form two mutually perpendicular cylindrical surfaces, with periodic boundary conditions inherently satisfied along the angular coordinates of each cylinder. The c.m. is then computed separately on each cylindrical manifold, accounting for the wrapped topology. After performing the inverse coordinate mapping, we obtain $\mathbf{X}_{c.m.}$ and $\mathbf{Y}_{c.m.}$, representing the c.m. coordinates along the two original spatial dimensions (see Supplemental Material Note 2 for detailed implementation [32]).

B. Electronic structure of benzene

To demonstrate the accuracy of our approach, we first reassess the electronic structure of benzene, a subject of chemical debate for nearly two centuries. In 1865, Kekulé

proposed an influential model of a six-carbon ring with alternating single and double bonds [eclipsed structure in Fig. 3(a)] [36,37]. However, this structure suffered from symmetry breaking and failed to explain benzene's aromaticity, limitations later addressed by molecular orbital theory and Pauling's resonance theory [38,39]. Subsequently, in the 1960s, Linnett advanced Lewis structure theory by incorporating spin, proposing a "staggered" structure with spin-staggered electrons, where each C–C is effectively a bond with an order of 1.5 [Fig. 3(a)] [40]. More recently, Liu and co-workers applied the DVMS method to show that both eclipsed and staggered structures coexist, and electron correlation drives a preference for the staggered structure [29]. Using the FermiNet as our neural network wave function ansatz, we also obtain both eclipsed and staggered structures in benzene [13]. The corresponding DVMS convergence curves are shown in Fig. 3(a). DVMS analysis further reveals that each of these structures corresponds to two distinct sites related by a 60° rotation [the same color insets in Fig. 3(b)], resulting in four reasonable sites. The sixfold

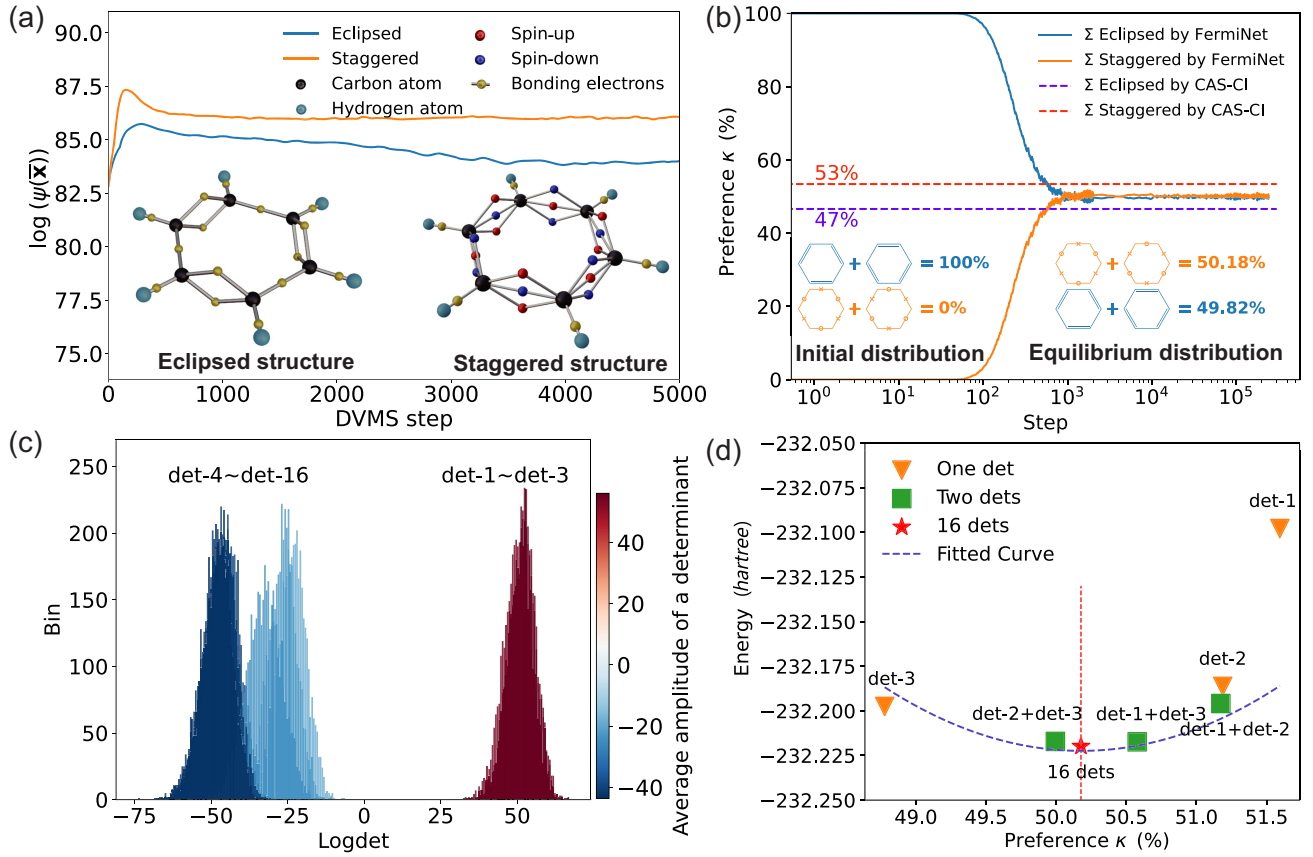


FIG. 3. Analysis of benzene's electronic structure. (a) DVMS convergence curves: wave function values corresponding to the mean site under different steps of DVMS and the converged electronic structure. Left: eclipsed structure. The Kekulé structure with alternating single and double bonds. Right: staggered structure. The opposite-spin electrons unpairing, presenting a spin-staggered structure. (b) Quantitative analysis of structural preference (κ) for staggered structure. The solid line is the convergence curve of the preference κ based on our FermiNet wave function, while the dashed line shows a previous result from a CAS-CI wave function [29]. The inset shows the four different structures that DVMS converged to. (c),(d) Using the particle view to assist in better understanding the determinants in neural network wave function. (c) Analysis of determinant contributions. Histogram of the amplitudes for each of the 16 determinants in the FermiNet wave function, labeled det-1 to det-16 in descending order of average amplitude. The distribution shows that only the first three determinants make a dominant contribution. (d) Correlation between structural preference and energy accuracy. Energy versus staggered structure preference (κ) for wave functions constructed from different combinations of the dominant determinants. Results are benchmarked against the full 16-determinant wave function (red star).

rotational symmetry of benzene is thus understood as a resonance among these four configurations.

Based on the near-exact neural network wave function, we can further reassess the structural preference in benzene. We perform Monte Carlo sampling without constraining walkers in a specific Voronoi cell, and count the number of walkers (denoted as N) within the Voronoi cells corresponding to the two structures. The preference for the staggered structure is quantified as $\kappa = N_{\text{staggered}} / (N_{\text{staggered}} + N_{\text{eclipsed}})$ [29]. In fact, benzene's structural preference is due to electron correlation between opposite-spin electrons, without which the two structures would be indistinguishable due to symmetry. However, for traditional quantum chemistry methods, accurately reflecting electron correlation to obtain an

accurate benzene wave function remains a key challenge. Thus, Liu *et al.* employed a higher-accuracy complete active space configuration interaction (CAS-CI) method and derived that the staggered structure has a 53% preference [29]. Fortunately, neural-network-based quantum Monte Carlo methods now enable near-exact solutions for benzene [13,16,17]. Using a nearly exact FermiNet wave function, our analysis yields a preference of $50.18\% \pm 0.02\%$, as shown in Fig. 3(b). While only marginally above 50%, this result qualitatively confirms the preference for the staggered structure. The robustness of this finding is demonstrated by the simulation's convergence to this value even when initialized with all walkers in the eclipsed structure. Notably, while recent breakthroughs in neural network methods have enabled the calculation of

benzene’s absolute energy to chemical accuracy [19], the quantitative results for its structural preference remain unchanged.

Beyond providing insights into benzene’s electronic structure, the structural preference can be used to investigate the optimization of different determinants within a neural network wave function. Although multiple determinants are often used to enhance performance of neural network wave function, their explicit roles remain unclear. Neural network wave functions incorporate electron correlation quite differently from traditional *ab initio* methods. While conventional Slater determinants model electron correlation via interdeterminant interactions, neural network architectures can encode all-electron interactions within a single determinant [13]. This architecture makes structural preference particularly useful for understanding neural network wave functions. In our study of benzene with a 16-determinant FermiNet, we find that only three determinants dominate after training. In Fig. 3(c), we plot the distributions of determinant amplitudes, labeling them det-1 to det-16 by descending average amplitude. Figure 3(d) analyzes energy and staggered structure preference for different wave functions constructed from individual and combined determinants (the convergence curves of preference are shown in Supplemental Material Note 5 [32]). The 16-determinant wave function serves as the benchmark for both the energy and the preference. Notably, single determinants fail to reproduce energy and staggered preference, while two-determinant mixes significantly improve both, revealing a clear correlation between energy accuracy and structural preference. This suggests that a single determinant is inadequate in representing the wave function, insufficiently capturing the correct spatial distribution of electrons. It is evident that neural network determinants exhibit different preferences for the eclipsed and staggered structures, requiring a balance to optimize wave function quality. For instance, both det-1 and det-2 favor the staggered structure, while det-3 favors the eclipsed structure. Combining det-1 and det-2 does not yield improvement, whereas combining det-1 or det-2 with det-3 leads to significantly better results. This demonstrates that benzene’s ground state is a hybrid of both structures, which the neural network represents by balancing determinants with different geometric biases. Therefore, the particle-view structural preference serves a dual role: It reflects the spatial bias of a wave function and acts as a sensitive metric for its energy calculation quality, making it a valuable tool for many-body electronic structure research.

C. Electronic structure of graphene

Having reassessed the valence bond structure of the benzene using our method, in this section we proceed to examine the electronic structure of graphene. As a two-dimensional honeycomb lattice of carbon atoms, graphene has received substantial attention since its experimental

fabrication in 2004 [41]. Its unique electronic structure gives rise to diverse properties. Recent advancements in constructing various graphene nanostructures and artificial superlattices have further enabled the exploration and tuning of emergent quantum phases [42,43]. Theoretical understanding of graphene is largely based on molecular orbital theory, which posits that sp^2 hybridization forms the backbone, with each carbon atom contributing a p_z electron to form the delocalized π bond. In contrast, the true valence bond structure of graphene has yet to be established and is often vaguely described as a mixed single-double bond network, an intuitive extension of the Kekulé structures in benzene [44–46]. This conceptual ambiguity stems from the computation and interpretation limitations mentioned earlier.

Here, we employ the DeepSolid neural network framework to calculate the accurate ground-state wave function of graphene, and then apply the newly developed PDVMS method to obtain a particle-view representation of the electrons [18]. We uncover that graphene has a spin-staggered structure, rather than a mixed single-double bond network. Figure 4(a) plots the electron positions of graphene and Fig. 4(d) depicts a sketch of its bonding structure: Each C–C bond hosts a pair of electrons forming a σ bond, and a single electron resides above or below each carbon atom in the out-of-plane direction. Analogous to the case in benzene, the true ground state emerges as a resonance of these spin-staggered structures, which collectively restores the full lattice symmetry of graphene. Additionally, using the wave function pretrained from Hartree-Fock orbitals, we also identified a Kekulé-like structure in graphene [inset in Fig. 4(b)], characterized by alternating single and double bonds, where each six-membered ring contains only two double bonds arranged in columns. With highly accurate DeepSolid wave function, our PDVMS simulation shows a clear convergence from this Kekulé-like structure to the more stable spin-staggered structure [Fig. 4(b)]. The simulation trajectory reveals that electron correlation drives this transition by dissociating the double bonds and promoting spin separation. The robustness of the spin-staggered ground state is confirmed by this convergence, i.e., when the simulation is initialized entirely within the Kekulé-like structure.

The particle view provides further insight into the optimization process of the DeepSolid wave function for graphene. Our analysis reveals that the training follows a path of initial symmetry breaking followed by eventual symmetry restoration. By examining structures at different stages of the training curve [Fig. 4(c)], we observe that the three double bonds within a unit cell gradually dissociate as the network is optimized (see Supplemental Material Note 4 for the corresponding 3D particle-view electronic structure [32]). During this process, the separated spin-up and spin-down electrons can move asymmetrically, leading to intermediate states that are not perfectly spin staggered [right-hand inset of Fig. 4(c)]. The wave function optimization ultimately converges to a perfect spin-staggered

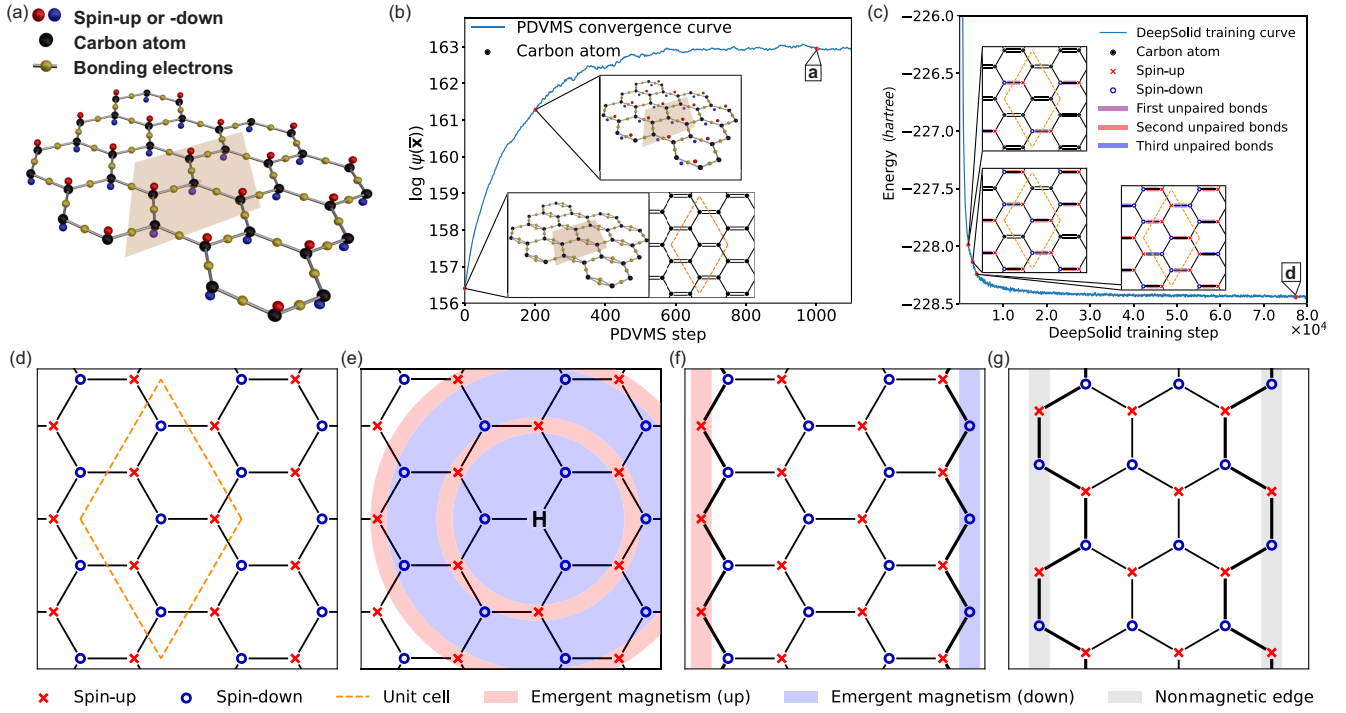


FIG. 4. Analysis of graphene's electronic structure. (a) The spin-staggered valence bond structure of graphene, as identified by converged PDVMS. The shaded region indicates the six-atom unit cell. (b) PDVMS convergence curve: wave function values corresponding to mean site under different steps of PDVMS, and the corresponding electronic structure. The process is initialized with a Kekulé-like structure containing a mixed single-double bond network (lower inset). With each iteration, the double bonds progressively unpair (upper inset), leading to the final converged structure shown in (a). (c) DeepSolid training curve: with Hartree-Fock as pretraining, it converges to graphene's near-exact ground state. The insets provide the 2D valence bond structures at different training steps, with colored background highlighting indicating the sequential unpairing of double bonds as the wave function approaches the true ground state. (d) Two-dimensional representation of the spin-staggered valence bond structure. Only the unpaired electrons localized at carbon atoms are displayed. (e) Hydrogenated graphene. Adsorbing a single hydrogen atom induces local emergent magnetism (shaded region). This magnetic distribution, deduced from the spin-staggered structure, matches experimental observations (Ref. [47]). (f),(g) Graphene nanoribbons. The edge magnetism, derived from the spin-staggered structure, is consistent with experimental observations (Ref. [48]). (f) Zigzag boundary. Electrons along the edge share the same spin, producing emergent edge magnetism (shaded regions). (g) Armchair boundary. The edge contains an equal number of opposite-spin electrons, causing magnetic moments to cancel out and resulting in a nonmagnetic edge.

structure, demonstrating that this arrangement lowers the energy due to the mutual repulsion between opposite-spin electrons. In contrast, using a more restrictive two-atom unit cell would preclude the necessary symmetry breaking step and force the system to converge incorrectly to the higher-energy Kekulé-like structure, as shown in Supplemental Material Note 5 [32].

The unique particle-view electronic structure of graphene we have identified provides a fresh perspective for interpreting a range of its properties. First of all, the presence of unpaired delocalized electrons offers an intuitive explanation of graphene's high electric conductivity, complementing the ballistic electron transport behavior based on the band-dispersion theory [49]. Second, these unpaired electrons serve as natural sites for adsorption and chemical reactions, providing *a priori* identification of reactive locations that align with the most stable conformation of graphene [50,51]. Furthermore, when a portion

of these electrons is saturated by adsorbed atoms or their distribution is altered via nanostructuring, the remaining unpaired electrons can give rise to emergent magnetism, a phenomenon documented in numerous experiments [47,48,52–54]. To illustrate this, the spin-staggered structure provides a direct real-space mechanism for hydrogenation-induced magnetism. As shown in Fig. 4(e), when a single H atom adsorbs and saturates an unpaired electron, it acts as a local perturbation that biases the resonance weights. This unbalanced resonance exposes the underlying spin-staggered structure, inducing a nanometer-scale magnetic feature [47], a phenomenon that a mixed single-double bond network completely fails to capture. Similarly, as illustrated in Figs. 4(f) and 4(g), introducing open boundaries via nanostructuring, such as in graphene nanoribbons, acts as a spatial perturbation that selectively pins specific spin-staggered resonance components. At a zigzag edge, where the boundary is dominated by a single

sublattice, the spin-staggered structure is exposed, directly leading to robust edge magnetism [48]. In contrast, an armchair edge contains an equal number of opposite-spin electrons, which causes their magnetic moments to perfectly cancel out and leaves the boundary nonmagnetic. Last but not the least, spin-staggered structure provides insight into the exotic quantum phases, such as charge density waves, magnetic orders, and topological states, that emerge in stacked and twisted graphene superlattices [42,55–57]. While these phenomena can also be explained via band-dispersion theory, the emergent magnetism rooted in the spin-staggered structure provides a natural pathway that makes time-reversal symmetry breaking possible from the particle view, whereas the single-double bond structure would face serious difficulties. In turn, the existence of these experimentally validated physical phenomena lends further credence to the spin-staggered ground-state structure identified in our work.

III. CONCLUSION

In this work, we have developed a robust, self-consistent *ab initio* framework to characterize the electronic structure of materials from the particle view. Our framework integrates the highly accurate real-space neural network wave function with an advanced sampling technique to project classical positions of electrons. The computational framework is validated with the classic example of benzene, confirming the valence bond structure predicted by Linnett [40] and offering novel insights into the roles of determinants within the neural network wave function. Applying this method to graphene, we discovered the spin-staggered electronic structure, which sheds light on the emergence of exotic quantum phases in graphene-based systems from a new perspective. Looking ahead, our framework can be utilized to investigate the complex many-body electronic structure of any periodic system, thereby fostering a particle-view understanding of materials. It also holds tremendous potential for deepening our understanding of condensed matter physics phenomena. Beyond static ground state, this framework can also be applied to investigate dynamic electronic structures under external perturbations (e.g., electric or magnetic field) or during real-time evolutions.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [58].

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