

Bilayer Graphene Quantum Dots as a Quantum Simulator of Haldane Topological Quantum Matter

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We demonstrate here that a chain of bilayer graphene quantum dots (BLGQDs) can realize topological quantum matter by effectively simulating a spin-1 chain that hosts the Haldane phase within a specific range of parameters. We describe a chain of BLGQDs with two electrons per dot using an atomistic tight-binding model combined with exact diagonalization to solve the interacting few-electron problem. Coulomb interactions and valley-mixing effects are treated within a single microscopic framework, allowing us to systematically investigate spin and valley polarization transitions as functions of interaction strength and external tuning parameters. We calculate the low energy states for single and double QDs as a function of the number of electrons, identifying regimes of highly correlated multielectron states. We confirm the presence of a spin-1 ground state for two electrons. Then, we explore two coupled QDs with four electrons and extend the analysis to QD arrays. Using a mapping of the BLGQD chain to an effective bilinear-biquadratic (BLBQ) spin model, we demonstrate that BLGQD arrays can work as a quantum simulator for one-dimensional spin chains with emergent many-body topological phases.

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Advancing quantum information technologies and uncovering novel topological phases of matter are central goals in modern condensed matter physics and quantum engineering [1–4]. A primary focus is the development of robust qubits with extended coherence times, with various platforms, including superconducting [5–8], trapped ion [9–11], photonic [12–15], and semiconductor spin qubits [3,16–26]. Spin qubits and circuits, particularly spin chains, stand out as prototypes for investigating topologically strongly correlated quantum matter, particularly due to their capacity to host Haldane spin-1/2 quasiparticles at their edges [27–30].

The artificial synthetic spin-1 chains have been intensely studied, but their experimental realization is limited. Spin-1 chains can be engineered using various quantum dot architectures, such as gated triple quantum dots [31], linear arrays of semiconductor quantum dots embedded in nanowires [30,32–34], chains formed by triangular graphene quantum dots [35–38], or tunable hybrid platform of superconducting islands and quantum dots [39]. For instance, chains of InAsP quantum dots embedded in an InP nanowire have been proposed and predicted to host such synthetic spin-1 objects [33,40], realizing macroscopic quantum states in semiconductors. In these systems, four electrons per quantum dot can form a synthetic spin-1 state, and their low-energy behavior can be effectively described by a Hubbard-Kanamori Hamiltonian derived

from atomistic microscopic calculations [40]. This framework has successfully demonstrated that these arrays can emulate antiferromagnetic spin-1 chains, providing a foundation for engineering synthetic topologically nontrivial quantum matter. Gated bilayer graphene (BLG) quantum dots (QDs) [41,42] offer a particularly attractive platform for this purpose. Single and double BLGQDs have been experimentally realized, with evidence of spin-1 ground states in few-electron configurations [43,44]. BLG is unique among two-dimensional materials in that a perpendicular electric field can open a tunable band gap, enabling electrical control over confinement and access to both spin and valley degrees of freedom [43–45]. Recent theoretical and experimental studies of BLGQDs show that two confined electrons can robustly form a triplet ground state across a wide range of interaction strengths, often accompanied by spontaneous spin-valley polarization [43,44]. This robust triplet formation makes BLGQDs promising building blocks for electrically tunable synthetic spin-1 chains.

Here, we investigate single and coupled BLGQDs using an atomistic tight-binding model combined with exact diagonalization to treat Coulomb interactions and valley mixing on equal footing. For two coupled QDs, we show that the low-energy sector maps naturally onto a bilinear-biquadratic spin-1 model, establishing a direct link between microscopic BLGQD states and effective spin-1 chains. This mapping highlights BLGQDs as a practical and tunable platform for exploring one-dimensional quantum magnetism, topological phases, and electrically controlled quantum information architectures.

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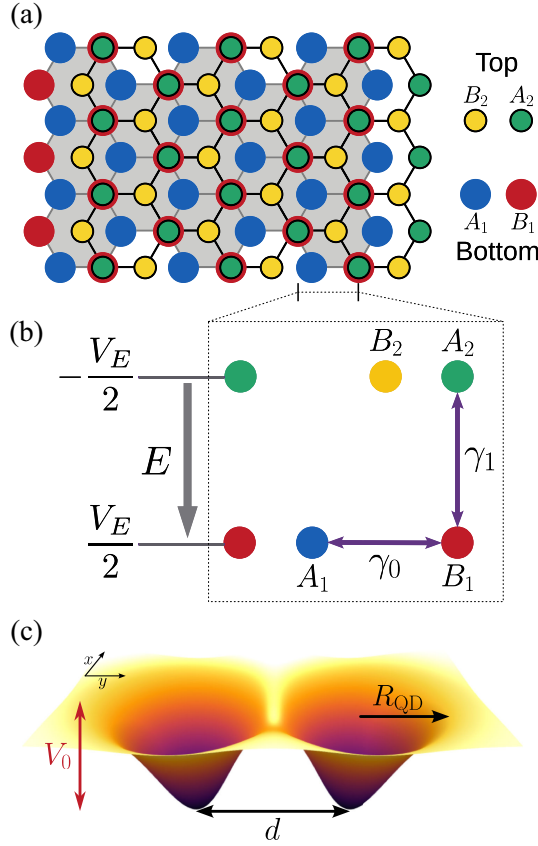


FIG. 1. (a) Top view of bilayer graphene, showing the four atoms within the unit cell: A_1 , B_1 , A_2 , B_2 , distinguished by color. (b) Side view (enlarged), highlighting the two primary hopping parameters: γ_0 for intralayer coupling and γ_1 for interlayer coupling. The applied perpendicular electric field is shown schematically; note that the diagram depicts the resulting potential energy profile rather than the electrostatic potential. (c) Schematic illustration of the confining potential for a double quantum dot. The parameters V_0 , R_{QD} , and d denote the potential depth, dot radius, and center-to-center distance between the dots, respectively.

We model Bernal-stacked bilayer graphene (BLG) [Fig. 1(a), whose bottom (top) layer contains sublattices A_1 and B_1 (A_2 and B_2)]. The electronic structure is described within a tight-binding model including the dominant in-plane and interlayer hopping parameters (γ_0 , γ_1) and a perpendicular electric field that opens a tunable band gap [Fig. 1(b)].

Gate-defined quantum dots (QDs) are created by adding smooth Gaussian confinement potentials [Fig. 1(c)], allowing us to realize single- or double-dot geometries. To obtain the confined single-particle states, we expand the full Hamiltonian $\hat{H}_{\text{QD}} = \hat{H}_{\text{bulk}} + \hat{V}_{\text{ext}}$ in a basis of bulk Bloch states near the $\pm K$ valleys and diagonalize it in momentum space. This approach captures the effect of the smooth confinement without resorting to large-scale real-space atomistic calculations [46].

Once the confined orbitals are obtained, we compute all relevant Coulomb matrix elements between them and perform

exact diagonalization of the many-body Hamiltonian to access the interacting few-electron spectrum. Numerically, we sample the Brillouin zone on a rhomboidal supercell of 901×901 k-points (equivalent to a real-space system of $\sim 3 \times 10^6$ atoms), ensuring convergence of both the single-particle and interacting energy levels. Full details of the tight-binding model, the momentum-space confinement treatment, and the numerical implementation are provided in Supplemental Material [47].

Many-body effects are incorporated by filling the lowest single-particle levels with N electrons and adding the Coulomb interaction,

$$H = \sum_s E_s c_s^\dagger c_s + \frac{\lambda}{2} \sum_{p,q,r,s} \langle pq|V|rs \rangle c_p^\dagger c_q^\dagger c_r c_s, \quad (1)$$

where E_s are the single-particle energies, $\langle pq|V|rs \rangle$ are the Coulomb matrix elements, and $\lambda \in [0, 1]$ tunes the interaction strength. We model electron-electron interactions using a scaled Coulomb potential with a dimensionless parameter λ that captures the effect of dielectric screening and gate geometry. The parameter λ effectively renormalizes the interaction strength, allowing us to explore different interaction regimes. In experimental BLGQD systems, λ can be tuned via the dielectric environment (e.g., hBN thickness and number of layers), the distance to metallic gates, and the quantum dot size. The spin structure is included explicitly, and all matrix elements are evaluated using the numerically obtained single-particle orbitals. Interdot electron-electron interactions are naturally included through the long-range Coulomb interaction, as we compute all matrix elements between all confined orbitals in the system.

The many-body Hamiltonian in the space of multi-electron configurations is solved via exact diagonalization to obtain the low-energy spectrum [44,46]. We perform the diagonalization in the full basis of confined orbitals, imposing only particle-number conservation as a constraint. At the single-particle level, the smooth confinement preserves valley symmetry (lacking Fourier components at $|K - K'|$), so Bloch states near $\pm K$ decouple and the numbers of K and K' states are conserved in the non-interacting problem. For the Coulomb interaction, we compute all matrix elements between all confined orbitals, including both intravalley and intervalley direct and exchange terms. Momentum-transfer selection rules make intervalley Coulomb scattering small but nonzero, allowing for valley mixing in the many-body states. Despite not explicitly imposing spin or valley conservation in the many-body calculation, the resulting eigenstates naturally organize according to total valley index, total spin S , and S_z , reflecting the underlying symmetries of the Hamiltonian.

For completeness, we first characterize the single quantum dot (QD) spectrum as a reference for the analysis of the double QD and BLG QD chain. For a representative

set of parameters, the single-particle levels form valley doublets, which become fourfold degenerate when spin is included, and display shell structures reminiscent of a two-dimensional harmonic oscillator with additional lattice-induced splittings. Including Coulomb interactions for two electrons, we find that the ground state remains a triplet over the entire interaction range considered, with a cross-over in valley polarization from polarized to unpolarized as the interaction weakens. The corresponding low-energy spectrum, along with its classification by total spin and valley polarization, is presented in Supplemental Material [47], which also includes full computational details and analytical considerations.

Motivated by the observation that the $N = 2$ ground state remains a triplet across a broad range of interaction strengths, we next examine the case of a double quantum dot (dQD) populated by four electrons. We aim to compare the resulting low-energy spectrum with that of an effective spin-1 Heisenberg model. To this end, we begin by analyzing the single-particle states of the double quantum dot, and then determine the interacting four-electron ground state along with its low-lying excitations.

Figure 2 shows the single-particle spectrum of the double quantum dot for an interdot separation $d = 40$ nm. Compared to the single QD case, each shell is now duplicated, with an energy splitting which reflects the presence of another coupled dot. This behavior can be understood by starting from the single-dot spectrum: as the dots are brought closer together, hybridization between the individual QD states increases, leading to the formation of bonding and antibonding combinations. Consequently, the splitting between these paired states grows as the interdot distance decreases and the depth of the potential increases [48–50].

Since higher QD states have a more extended wave function, the state hybridization is stronger for higher energy shells. We can see it on the probability density plot for each state (insets in Fig. 2). Low-energy states like the one in the s shell are not strongly modified compared to the single QD version. On the other hand, states composed of QD p and d shells are more modified, with the effect being bigger as the state energy increases.

Figure 2(b) shows the low-energy spectrum for four electrons confined in a double quantum dot as a function of interaction strength. In the weak-interaction regime, the ground state remains a valley-unpolarized singlet, with the first and second excited states being a triplet and a quintuplet, respectively. The main contribution to the singlet state comes from the configuration with the lowest shell completely full. This sequence closely matches the spectrum of a spin-1 Heisenberg antiferromagnetic chain with $L = 2$ sites, consistent with the mapping to an effective spin model.

As interaction strength increases, the system transitions to a valley-polarized quintuplet ground state ($S = 2$). This transition is driven by electron-electron repulsion and fine

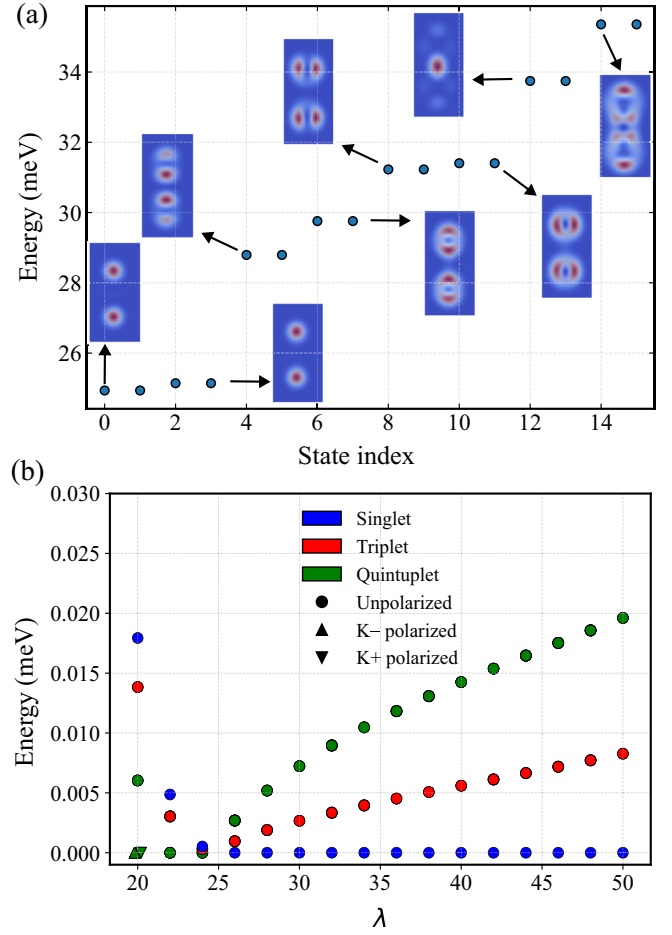


FIG. 2. (a) Low-energy levels of a double quantum dot. Each state shown is doubly degenerate by spin; only the valley-degenerate states are explicitly labeled. Insets show the corresponding electron densities for each shell, illustrating the increased coupling between interdot states with higher energy, as a consequence of the greater spatial extension of the wave functions. (b) Low-energy spectrum for four electrons in a double quantum dot as a function of interaction strength. For weak interactions, the ground state is a valley-unpolarized singlet, while for strong interactions, it becomes a valley-polarized quintuplet. There is an artificial horizontal shift for the valley-polarized quintuplets to show both valley cases. In the regime where the ground state is a singlet, the first and second excited states are a triplet and a quintuplet, respectively, closely resembling the spectrum of a spin-1 Heisenberg antiferromagnetic chain with $L = 2$ sites.

tuned by the exchange interaction: strong interactions make it energetically favorable to promote electrons to the second shell (at a kinetic energy cost), and since intravalley exchange is significantly larger than intervalley exchange, the electrons tend to concentrate in a single valley. In this case the next excited states are a quintuplet, followed by a triplet and then a singlet, mainly composed of electrons on the lowest shell. Remarkably, at intermediate interaction, when the ground state becomes a valley-unpolarized quintuplet, the first and second excited states are a triplet

and a singlet, respectively, mirroring the spectrum expected for two spin-1s coupled ferromagnetically.

The bilinear–biquadratic (BLBQ) spin-1 Hamiltonian

$$H = J \sum_i^{L-1} (\mathbf{S}_i \cdot \mathbf{S}_{i+1} + \beta (\mathbf{S}_i \cdot \mathbf{S}_{i+1})^2), \quad (2)$$

where $\mathbf{S}_i$ is the spin operator acting on site i , and J is the exchange coupling, β is a tunable parameter that controls the strength of the biquadratic interaction, and L is the number of sites. For even L , and $\beta < 1/3$, the low-energy spectrum consists of a singlet ground state, followed by a triplet and a quintuplet. In particular, for two spins, the energy gaps are given by [see Supplemental Material (SM) [47] for details], $\Delta_{TQ} = 2J$, $\Delta_{ST} = J(1 - 3\beta)$. The bilinear-biquadratic spin-1 model exhibits the Haldane phase for $-1 < \beta < 1$, a topologically ordered phase first predicted by Haldane for the $S = 1$ antiferromagnetic Heisenberg chain. This phase is characterized by a gapped bulk spectrum and fractionalized spin-1/2 edge states, despite the bulk being made of integer spins [27,29].

Using these expressions, we can map the dQD spectrum onto an effective BLBQ model and extract the corresponding J and β parameters. Figure 3(a) shows the low energy levels from the fitted BLBQ and Heisenberg models as a function of interaction strength. The blue dashed lines show the energies of the double quantum dot. The BLBQ Hamiltonian reproduces exactly the two low-energy gaps of two coupled quantum dots, containing two electrons each.

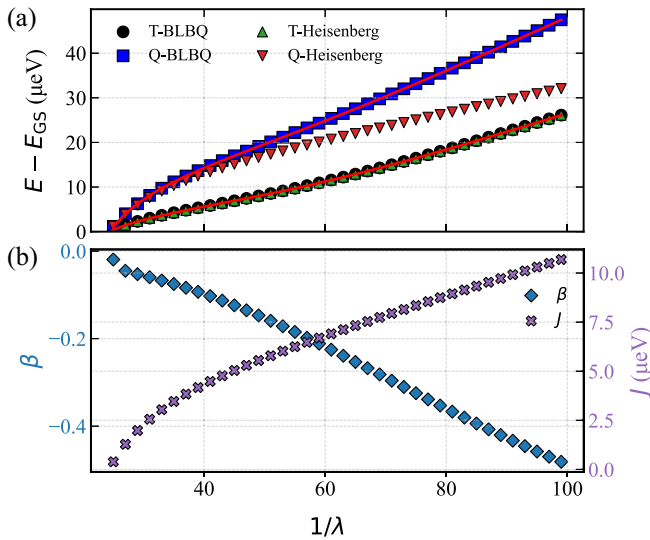


FIG. 3. (a) Triplet and quintuplet energies obtained from the fitted BLBQ and Heisenberg models as a function of interaction strength. The blue dashed lines show the energies of the double quantum dot. As discussed in the main text, the BLBQ Hamiltonian accurately reproduces the low-energy spectrum of two coupled quantum dots, each containing two electrons. (b) Fitted values of the BLBQ parameters β and J as functions of the electron-electron interaction strength.

Figure 3(b) shows the fitted value of J and β for different values of the ratio between kinetic and Coulomb interacting terms.

For stronger interactions, β is near zero, becoming more negative as the interaction strength decreases. This behavior is consistent with numerical [51] and perturbative [52] results in multiorbital Hubbard models, where the largest values of $|\beta|$ are obtained when the Hubbard interaction U is reduced. In the perturbative limit where an expansion in t/U is valid, the bilinear term J appears at lowest order while the biquadratic term (proportional to β) appears at next-to-leading order. The minimum value shown here for weak interaction is $\beta \approx -0.5$, which is still inside the Haldane phase region. The sign of β reflects the structure of the effective spin Hamiltonian emerging from multi-orbital superexchange [51,52].

To explore the behavior of the BLBQ Hamiltonian, we computed the low-energy spectrum of a spin-1 chain with $L = 100$ using matrix product state–based density matrix renormalization group (MPS-DMRG) approach [53–55]. Figure 4(a) shows the low-energy spectrum as a function of the biquadratic interaction parameter β in the range $-1/2 \leq \beta \leq 0$. In the entire Haldane phase ($-1 < \beta < 1$) and in the thermodynamic limit, the ground state is fourfold degenerate, consisting of a singlet and a triplet manifold separated from the lowest quintuplet by a finite topological gap. This degeneracy arises from two fractionalized spin-1/2 quasiparticles localized at the chain ends. In a finite chain, the singlet–triplet gap is nonzero but decreases exponentially with system size, reflecting the exponentially small coupling between the edge spins. In contrast, the topological gap to the quintuplet remains finite in the thermodynamic limit [27,29].

Figure 4(b) displays the spin-spin correlation function $g(n) = (-1)^n \langle S_1^z S_n^z \rangle$ for the first half of the chain. As β becomes more negative, the decay of $g(n)$ slows, indicating an increase in the correlation length ξ . This length scale is extracted by fitting the envelope of the data to an exponential form, $g(n) \propto \exp(-n/\xi)$. The inset shows the fitted values of ξ as a function of β . The correlation length ξ controls both the spatial extent of the edge quasiparticles and the range over which they can interact. When $L \gg \xi$, the system is effectively in the thermodynamic limit and the singlet–triplet splitting becomes negligible. However, for $\beta \approx -0.5$, ξ becomes comparable to $L = 100$, making finite-size effects visible in the spectrum and enhancing the singlet–triplet splitting. As $\beta \rightarrow -1$, the system approaches a quantum critical point characterized by a vanishing triplet–quintuplet gap and a diverging correlation length [56]. In this regime, DMRG convergence becomes increasingly challenging due to enhanced entanglement and critical fluctuations.

The mapping from BLGQDs to the BLBQ model can be understood within a broader theoretical framework. Similar to the two-site two-orbital Hubbard model [51,52], our graphene QD with two electrons exhibits a half-filled

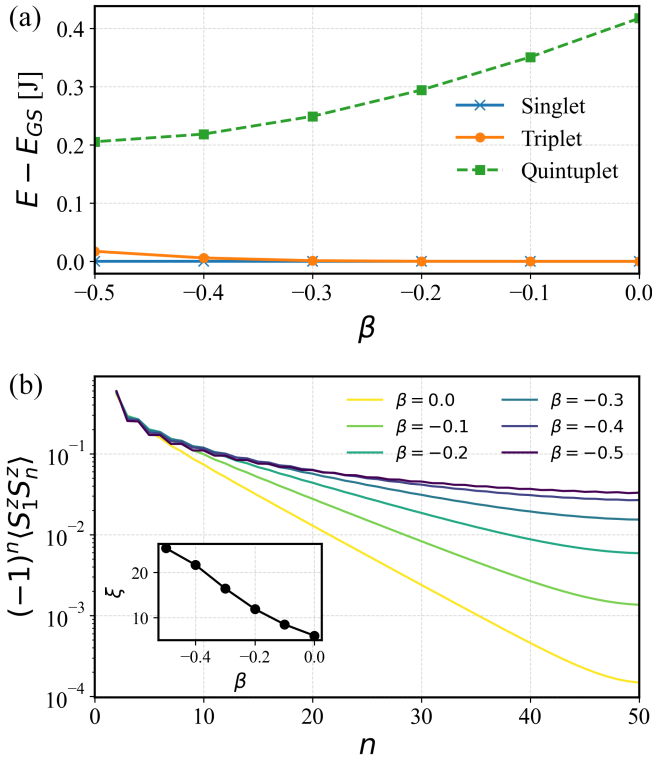


FIG. 4. (a) Low energies of the $L = 100$ BLBQ spin chain as a function of the β . (b) Spin-spin correlation function $g(n) = (-1)^n \langle S_1^z S_n^z \rangle$ plotted for the first half of the chain. The inset shows the correlation length ξ extracted from exponential fits to $g(n)$, illustrating its dependence on β . The correlation length ξ characterizes the spatial decay of spin correlations in the system.

degenerate s -shell at low energies, where the two orbital levels play the role of the two orbitals per site in the Hubbard model. Under the effective condition of weaker interactions, both systems map to the same BLBQ Hamiltonian, demonstrating the universality of this effective description for systems with degenerate orbital manifolds.

The correlation length ξ determines the minimal chain length required to observe topological properties. For the Haldane phase with β near 0, $\xi \sim 6$ sites, so chains of length $L \approx 10$ –20 sites should be sufficient to observe clear topological signatures such as the bulk gap and edge states. For example, topological effects have been observed in chains of 16 nanographene quantum dots [36]. Moreover, topological properties such as edge states and bulk gap are observable even in relatively short chains, making BLGQDs a promising platform for quantum simulation despite current experimental constraints.

The robustness of our results against model details has been verified by including the skew-hoppings γ_3 and γ_4 (see SM [47]). These terms introduce only quantitative corrections (trigonal warping and particle-hole asymmetry) but do not affect the qualitative physics: the spin structure, ground state nature, and valley ordering of the low-energy manifold remain unchanged [45,57]. This robustness stems from

the fact that the spin-1 character is determined by the fourfold degenerate lowest shell, which is insensitive to these corrections.

In summary, we have demonstrated that chains of electrostatically defined bilayer graphene quantum dots (BLGQDs) can become synthetic spin-1 chains, providing an atomistic platform for topological quantum matter. Using a realistic tight-binding description combined with exact diagonalization, we captured Coulomb interactions and valley mixing on equal footing for both single- and double-dot configurations. For two coupled QDs, the low-energy spectrum maps naturally onto a bilinear-biquadratic spin-1 model, establishing a direct link between microscopic electronic states in BLGQDs and effective quantum spin Hamiltonians. This correspondence opens a route to quantum simulators of Haldane topological quantum matter, one-dimensional quantum magnetism with electrically tunable parameters, offering new opportunities for quantum simulation, spintronics, and solid-state quantum information processing.

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Data availability—The data that support the findings of this article are not publicly available because of legal restrictions preventing unrestricted public distribution. The data are available from the authors upon reasonable request.

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