

Emergence of a Luttinger liquid phase in an array of chiral molecules

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We propose a robust platform for simulating chiral quantum magnetism using linear arrays of trapped asymmetric-top molecules, specifically 1,2-propanediol ($C_3H_8O_2$). By mapping the Stark-dressed rotational states onto an effective spin-1/2 subspace, we rigorously derive a generalized XXZ Heisenberg Hamiltonian governing the underlying many-body dynamics. Unlike standard solid-state models where the topological Dzyaloshinskii-Moriya interaction (DMI) is introduced phenomenologically, we demonstrate that DMI emerges *ab initio* from the molecular stereochemistry. Specifically, the interference between the transition dipole moments of heterochiral enantiomer pairs (left- and right-handed), which breaks inversion symmetry, generates a tunable DMI that stabilizes a chiral Luttinger liquid phase. Through a comprehensive phase-diagram analysis, we identify an optimal experimental regime characterized by intermolecular separations of $r \approx 1.5$ nm and intermediate electric-field strengths $d\epsilon/B \approx 2.5$. In this window, the system is protected from trivial field-polarized phases and exhibits a robust gapless spin-spiral texture. Our results establish 1,2-propanediol arrays as a versatile quantum simulator, providing a direct microscopic link between molecular chirality and topological many-body phases.

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I. INTRODUCTION

An object is said to be chiral if it cannot be superposed on its mirror image—like the right hand cannot be superposed on the left hand [1–3]. The chirality—or handedness—of molecules was first identified in 1848 by Louis Pasteur [4,5], who observed that the left- (L) and right-handed (R) isomers (termed enantiomers) of a given molecule interact differently with polarized light, rotating its plane of polarization either clockwise or counterclockwise. In 1927, Friedrich Hund analyzed molecular chirality quantum mechanically and, in the process, introduced the concept of tunneling through the potential barrier separating the L and R enantiomers [6,7].

Biological systems are homochiral, meaning that life uses only a single handedness, such as right-handed sugars and left-handed amino acids [8–11]. However, in the absence of a chiral bias, chemistry produces equal proportions of left- and right-handed enantiomers, known as racemic mixtures, which are achiral [12,13]. Chirality dictates function through a lock-and-key-like mechanism: A molecule must have the correct handedness in order to bind to a chiral receptor, as the interaction between two chiral molecules depends strongly on

their handedness and relative orientation. It has been proposed that homochirality may involve magnetic effects. Indeed, experimental studies suggest that magnetic surfaces could create a bias for the formation of one enantiomer over another, which could have played a role in the early stages of life on Earth [14–18].

Chirality-induced spin selectivity (CISS) describes the phenomenon where chiral molecules induce spin-dependent electron transport even in the absence of circularly polarized light or an external magnetic field [19–21]. So far, CISS has been studied in three experimental domains: (1) Electron transmission across chiral monolayers, such as DNA or peptides, which may exhibit spin polarization of up to 60% [22–24]. (2) Even greater spin selectivity (80%–90%) is produced by electron transport in chiral perovskite films and molecular junctions [25–27]. (3) Spin-polarized electron-mediated enantioselectivity in chemical processes, connecting CISS to spin-dependent catalysis and enantioseparation [28]. Spin-orbit coupling (SOC) is the primary mechanism proposed for CISS, but theoretical calculations based solely on SOC predict spin polarizations several orders of magnitude smaller than the experimentally determined values. The primary reason for this discrepancy is a lack of a unified theory of CISS that would integrate spin-orbit coupling, electron-phonon interactions, molecular structure, and environmental effects [29]. In this paper, we aim to develop an effective many-body model for an asymmetric-top molecule [1,2-propanediol (PDO)] rather than a simple, single-particle scattering model for a chiral molecular configuration.

Recently, several research groups have studied the many-body dynamics of linear, spherical, and symmetric-top

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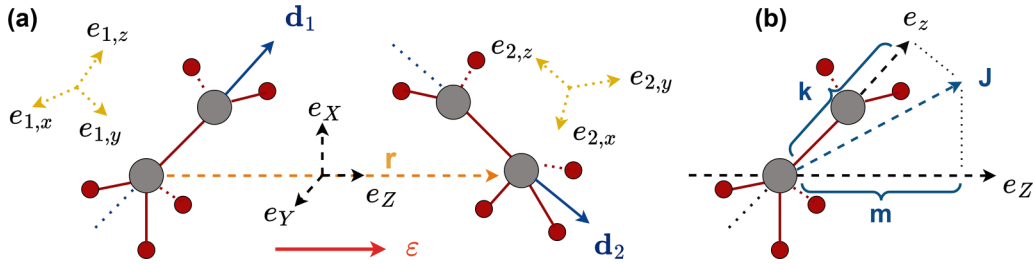


FIG. 1. (a) Schematic representation of two rotating prolate asymmetric-top molecules (1,2-propanediol) with permanent electric dipole moments $\mathbf{d}_1$ and $\mathbf{d}_2$, interacting with an external dc electric field $\boldsymbol{\varepsilon}$. The laboratory-fixed reference frame (X, Y, Z) is chosen such that the intermolecular separation vector satisfies $\mathbf{r} = r \mathbf{e}_Z$. Each molecule is associated with its own molecule-fixed frame (x_i, y_i, z_i) . Due to molecular chirality, the dipole moments $\mathbf{d}_i$ have opposite orientations with respect to their corresponding body-fixed frames for the two enantiomers. (b) Schematic representation of the total angular momentum $\mathbf{J}$ with respect to the laboratory-fixed axis $\mathbf{e}_Z$ and the molecule-fixed principal axis $\mathbf{e}_i$.

molecules by mapping their configurations onto the XXZ spin-1/2 model [30–32]. The realization of the XXZ model in a molecular system offers a key advantage: The ability to implement an effective spin degree of freedom without any fine-tuning of the field. Additionally, the study of ground-state phase diagrams is significantly easier. In this paper, we chose a chiral asymmetric-top molecule (1,2-propanediol), as its energy spectrum is particularly rich due to the absence of inversion symmetry [33]. For each fixed total angular momentum $\mathbf{J}$, the Hilbert space scales with the basis states j as $(2j + 1)^2$. The absence of a conserved molecular-fixed projection quantum number leads to strong mixing among the rotational states; therefore, the eigenstates cannot be labeled by a single value of the projection k on the figure axis of the molecule. In addition, unless the electric field is oriented along the laboratory Z axis, mixing among different m states further increases the complexity of the field-induced state. Together, these features result in highly coupled rotations and amide folds, making the theoretical description of 1,2-propanediol significantly more challenging than that of linear or symmetric-top molecules. Herein, we select the two lowest-energy states, $|j = 0, k = 0, m = 0\rangle$ and $|j = 1, k = -1, m = \pm 1\rangle$, which are identified as the pseudospin states $|\uparrow\rangle$ (dashed blue curve) and $|\downarrow\rangle$ (solid red curve) shown, respectively, in Fig. 2. We then demonstrate how an effective spin-1/2 XXZ model can be realized directly from these states without invoking any approximations.

The invariant XXZ model has been derived from achiral molecules, where the interactions are symmetric. In other spin models, the antisymmetric exchange term, $\vec{D} \cdot (\vec{S}_i \times \vec{S}_j)$, is introduced as a phenomenological fitting parameter; in our work, chiral coupling is derived *ab initio* from the molecular stereochemistry. We explicitly demonstrate how the biological handedness of enantiomers is encoded in dipole moments and aptly translated into a topological spin-spin interaction. This approach provides a detailed recipe for engineering Hamiltonian parameters through chemical synthesis and establishes a direct link between molecular chirality and the emergence of chiral spin textures. Our phase-diagram analysis reveals a broad sweet spot at intermediate electric fields ($d\varepsilon/B \approx 2.0$) and a lattice spacing of $r = 1.5$ nm. By mapping the complex rotational dynamics onto the universal phase diagram of the XXZ chain, we demonstrate that at the many-body level, the antisymmetric interactions qualitatively change the

low-energy physics of the spin chain. Instead of forming conventional Luttinger liquid phases of achiral XXZ models, the system can stabilize chiral quantum phases, such as the chiral Luttinger liquid (CLL).

The outline of this paper is as follows: In Sec. II, we calculate the eigenenergies of the asymmetric-top molecule ($\text{C}_3\text{H}_8\text{O}_2$) in an external electrostatic (dc) field as a function of the dimensionless parameter $d\varepsilon/B$, where d is the permanent electric dipole moment, ε is the electric-field strength, and B is the rotational constant. We then construct the matrix elements of the dipole-dipole interaction in the pseudospin basis $\{|\downarrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\uparrow\uparrow\rangle\}$ and finally realize the XXZ spin-1/2 model by projection onto an effective two-qubit subspace.

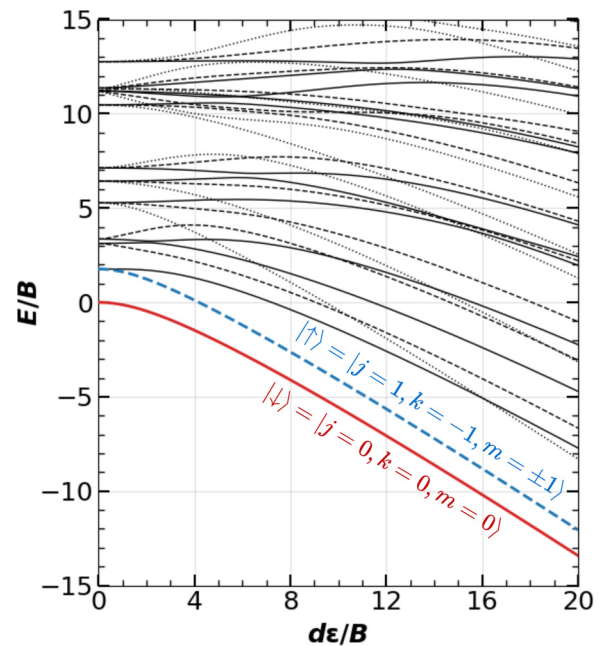


FIG. 2. Eigenenergies of the asymmetric-top molecule 1,2-propanediol ($\text{C}_3\text{H}_8\text{O}_2$) in an external dc electric field, plotted as a function of $d\varepsilon/B$, where d is the permanent dipole moment, ε is the electric-field strength, and B is the rotational constant. In the absence of the electric field, the states $|j = 0, k = 0, m = 0\rangle$ and $|j = 1, k = -1, m = \pm 1\rangle$ are identified as the pseudospin states $|\downarrow\rangle$ (solid red curve) and $|\uparrow\rangle$ (dotted blue curve), respectively.

In Sec. III, we present and analyze the coupling constants as functions of $d\epsilon/B$ for several intermolecular separations. In Sec. IV, we examine the resulting phase diagram of the array of asymmetric-top molecules. In Sec. V, we discuss the experimental implementation of a quantum simulator of a Luttinger liquid based on an array of asymmetric-top molecules. Finally, in Sec. VI we present our conclusions.

II. MODEL

A. Single-molecule Hamiltonian ($\hat{H}_{\text{rot}} + \hat{H}_{\text{dc}}$)

We consider two asymmetric-top molecules (1,2-propanediol) in their vibrational ground state, separated by a distance r as illustrated in Fig. 1. Each molecule is treated as a rigid rotor with a permanent dipole moment d_i ($i = 1, 2$), leading to dipole-dipole interactions, subject to a dc electric field ϵ applied along the laboratory Z axis at fixed intermolecular separation. The total Hamiltonian for the system reads

$$\hat{H}_{\text{total}} = \hat{H}_{\text{rot}} + \hat{H}_{\text{dc}} + \hat{H}_{\text{dd}}, \quad (1)$$

where $\hat{H}_{\text{rot}}$ describes the rotation of both molecules relative to each other, $\hat{H}_{\text{dc}}$ captures the interaction of the molecules with an external electric field, and $\hat{H}_{\text{dd}}$ gives the dipole-dipole interaction. We have ignored nuclear spins of the molecules. For a general asymmetric-top molecule (1,2-propanediol), the rotation of the molecules under rigid rotor approximation is given as [34,35]

$$\hat{H}_{\text{rot}} = A\hat{J}_a^2 + B\hat{J}_b^2 + C\hat{J}_c^2. \quad (2)$$

Here, $A = \frac{\hbar^2}{2I_a}$, $B = \frac{\hbar^2}{2I_b}$, and $C = \frac{\hbar^2}{2I_c}$ are the rotational constants associated with the principal moments of inertia I_a, I_b, I_c , respectively, with $I_a \neq I_b \neq I_c$. The operators $\hat{J}_a^2$, $\hat{J}_b^2$, and $\hat{J}_c^2$ denote the squared components of the angular momentum along the corresponding principal axes. For asymmetric-top molecules, where $I_a \neq I_b \neq I_c$, none of the angular momentum components $\hat{J}_a$, $\hat{J}_b$, and $\hat{J}_c$ commute with the rotational Hamiltonian:

$$[\hat{H}_{\text{rot}}, \hat{J}_a] \neq 0, \quad [\hat{H}_{\text{rot}}, \hat{J}_b] \neq 0, \quad [\hat{H}_{\text{rot}}, \hat{J}_c] \neq 0.$$

Consequently, no molecule-fixed projection of the total angular momentum $\hat{J}$ is conserved. Based on microwave spectroscopy measurements [36,37], the rotational constants for a conformer of 1,2-propanediol ($\text{C}_3\text{H}_8\text{O}_2$) are $A = 8572.05$ MHz, $B = 3640.11$ MHz, and $C = 2790.97$ MHz. The permanent electric dipole moment $\mathbf{d}$ possesses nonzero components along all three principal axes, with $d_a = 1.201$ D, $d_b = 1.916$ D, and $d_c = \pm 0.365$ D.

Since the rotational constants of 1,2-propanediol ($\text{C}_3\text{H}_8\text{O}_2$) satisfy $A > B > C$, the molecule is classified as a prolate asymmetric top. For a prolate asymmetric top, we define the molecule-fixed frame as $(x_i, y_i, z_i) = (b_i, c_i, a_i)$ for each molecule i , while the laboratory-fixed frame (X, Y, Z) is chosen such that the intermolecular separation vector satisfies $\mathbf{r} = r\mathbf{e}_Z$ [35].

The primary objective of this work is to investigate the interaction between two enantiomers of 1,2-propanediol ($\text{C}_3\text{H}_8\text{O}_2$). The two enantiomers are identical in all spectroscopic properties, including their rotational constants and

energy spectra; however, they differ in the orientation of one component of the permanent dipole moment in the molecule-fixed frame. Specifically, for the L enantiomer, the dipole-moment components are $d_a = 1.201$ D, $d_b = 1.916$ D, and $d_c = +0.365$ D, whereas for the R enantiomer the components d_a and d_b remain unchanged, while the c -axis component reverses sign, $d_c = -0.365$ D. Our goal is to encode this enantiomer-dependent dipole structure directly into the Hamiltonian in order to study its impact on molecular chirality and, indirectly, on the resulting phase diagrams.

For any rotating molecule—linear, symmetric-top, or asymmetric-top—the total angular momentum operator $\mathbf{J}$ satisfies $\mathbf{J}^2 = \hbar^2 j(j+1)$, where $j = 0, 1, 2, \dots$ is always a good quantum number. For 1,2-propanediol ($\text{C}_3\text{H}_8\text{O}_2$), each fixed value of j corresponds to $(2j+1)^2$ rotational basis states $|jkm\rangle$. The quantum number k represents the projection of $\mathbf{J}$ on the molecule-fixed z axis, which coincides with the a axis for prolate asymmetric tops. For a given j , k can take $2j+1$ values in the range $k = -j, -j+1, \dots, j-1, j$. Since k is not a conserved quantum number, the true eigenstates are labeled by $\tilde{k}$. Similarly, m denotes the projection of $\mathbf{J}$ on the laboratory Z axis and, for a given j , can take $2j+1$ values in the range $m = -j, -j+1, \dots, j-1, j$. The quantum number m remains conserved if the system possesses cylindrical symmetry about the z axis; i.e., if the applied external electric field ϵ is oriented along z . If the field is not oriented along the Z axis, different m states are mixed [38]. Overall, for a fixed value of j , all combinations of k and m satisfying $|k| \leq j$ and $|m| \leq j$ are allowed.

The interaction between the permanent dipole and the external dc electric field is given by

$$\hat{H}_{\text{dc}} = -\mathbf{d} \cdot \boldsymbol{\epsilon}, \quad (3)$$

where $\mathbf{d}$ is the dipole-moment operator and $\boldsymbol{\epsilon}$ is the external dc electric field. Both $\mathbf{d}$ and $\boldsymbol{\epsilon}$ can be expressed as rank-1 spherical tensors, leading to

$$\mathbf{d} \cdot \boldsymbol{\epsilon} = \sum_{q=-1}^1 (-1)^q d_q \epsilon_{-q}, \quad (4)$$

where d_q and ϵ_{-q} denote the spherical components in the laboratory frame. Consequently,

$$\hat{H}_{\text{dc}} = - \sum_{q=-1}^1 (-1)^q d_q \epsilon_{-q}. \quad (5)$$

Since the molecular dipole moment is naturally defined in the molecule-fixed frame through the components $d_r^{(\text{mol})}$, whereas d_q denotes the corresponding components in the laboratory frame, it is necessary to transform d_q into the molecule-fixed frame. This transformation is given by

$$d_q = \sum_{r=-1}^1 D_{q,r}^{1*}(\Omega) d_r^{(\text{mol})}, \quad (6)$$

where $d_r^{(\text{mol})}$ are fixed molecular constants and Ω specifies the molecular orientation. Substituting this expression into Eq. (5)

gives

$$\hat{H}_{\text{dc}} = - \sum_{q=-1}^1 \sum_{r=-1}^1 (-1)^q D_{q,r}^{1*}(\Omega) d_r^{(\text{mol})} \varepsilon_{-q}. \quad (7)$$

The corresponding matrix elements in the $|jkm\rangle$ basis become

$$\begin{aligned} \langle jkm | \hat{H}_{\text{dc}} | j'k'm' \rangle &= - \sum_{q=-1}^1 \sum_{r=-1}^1 (-1)^q \langle jkm | D_{q,r}^{1*}(\Omega) | j'k'm' \rangle \\ &\times d_r^{(\text{mol})} \varepsilon_{-q}. \end{aligned} \quad (8)$$

Since the external electric field has only a single component along the laboratory z axis ($\boldsymbol{\varepsilon} = \varepsilon \mathbf{e}_z$), its spherical components satisfy

$$\varepsilon_0 = \varepsilon_z, \quad \varepsilon_{\pm 1} = 0. \quad (9)$$

As a result, only the ($q = 0$) term contributes to the dc Stark Hamiltonian. The matrix elements, therefore, reduce to

$$\langle jkm | \hat{H}_{\text{dc}} | j'k'm' \rangle = -\varepsilon_z \sum_{r=-1}^1 \langle jkm | D_{0,r}^{1*}(\Omega) | j'k'm' \rangle d_r^{(\text{mol})}. \quad (10)$$

The detailed derivation of the matrix elements for $\hat{H}_{\text{rot}}$ and $\hat{H}_{\text{dc}}$ is provided in Appendix B. For a single molecule interacting with an external electric field, we compute the energy spectrum of the asymmetric-top molecule (1,2-propanediol) and plot it as a function of $d\varepsilon/B$ in Fig. 2. The results show that increasing the electric field leads to enhanced mixing of states with different j and k quantum numbers. For higher rotational quantum numbers $j \geq 2$, we clearly observe the onset of level crossings, reflecting the fact that asymmetric-top molecules exhibit mixing between states with different k . As a result, neither j nor k remains a good quantum number, and we therefore denote them by $\tilde{j}$ and $\tilde{k}$. In contrast, the quantum number m remains conserved, since the external electric field has only a single component along the laboratory z axis ($\boldsymbol{\varepsilon} = \varepsilon \mathbf{e}_z$), with spherical components $\varepsilon_0 = \varepsilon_z$ and $\varepsilon_{\pm 1} = 0$.

We select two rotational states—the ground state $|j = 0, k = 0, m = 0\rangle$ (red solid line) and the first excited state $|j = 1, k = -1, m = \pm 1\rangle$ (blue dotted line)—to define effective pseudospin states $|\downarrow\rangle$ and $|\uparrow\rangle$, respectively, as illustrated in Fig. 2. An external electric field is then used to couple these two states, forming an effective two-level system. Owing to field-induced mixing, the resulting pseudospin states are linear combinations of basis states characterized by different $\tilde{j}$ and $\tilde{k}$:

$$|\downarrow\rangle = \sum_{\tilde{j}, \tilde{k}} c_{\tilde{j}, \tilde{k}}^{(\downarrow)}(x = d\varepsilon/B) |\tilde{j}, \tilde{k}, m = 0\rangle, \quad (11)$$

$$|\uparrow\rangle = \sum_{\tilde{j}, \tilde{k}} c_{\tilde{j}, \tilde{k}}^{(\uparrow)}(x = d\varepsilon/B) |\tilde{j}, \tilde{k}, m = 1\rangle. \quad (12)$$

Figure 3 shows the expansion coefficients as functions of $d\varepsilon/B$. At $d\varepsilon/B = 0$, both $|\downarrow\rangle$ and $|\uparrow\rangle$ correspond to pure rotational states, consisting solely of the components $|j = 0, k = 0\rangle$ and $|j = 1, k = -1\rangle$, respectively. As $d\varepsilon/B$ increases, additional j and k components are progressively admixed, while the initially dominant contributions decrease accordingly. For $|\uparrow\rangle$, the dominant component $|j = 0, k = 0\rangle$

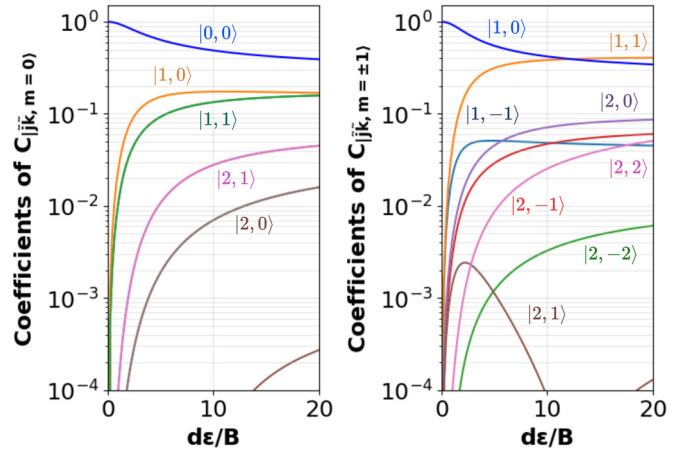


FIG. 3. Field dependence of the expansion coefficients $c_{j\tilde{k}, m}(x = d\varepsilon/B)$ for the pseudospin states $|\downarrow\rangle = |\tilde{j}, \tilde{k}, m = 0\rangle$ (left panel) and $|\uparrow\rangle = |\tilde{j}, \tilde{k}, m = \pm 1\rangle$ (right panel) [see Eq. (3)].

(shown in blue) decreases slowly and saturates along the component $|j = 1, k = 1\rangle$ for $d\varepsilon/B \gtrsim 5.0$. In contrast, for $|\downarrow\rangle$ the initial dominant component $|j = 1, k = -1\rangle$ (also shown in blue) decreases more gradually, but is eventually overtaken by the $|j = 1, k = 1\rangle$ component at sufficiently large $d\varepsilon/B$.

B. Dipole-dipole interaction

The dipole-dipole interaction is described by the Hamiltonian

$$\hat{H}_{\text{dd}} = - \frac{(\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2) - 3(\hat{\mathbf{d}}_1 \cdot \mathbf{e}_r)(\hat{\mathbf{d}}_2 \cdot \mathbf{e}_r)}{r^3}, \quad (13)$$

where $\mathbf{e}_r = \mathbf{e}_z$ in our chosen frame. For simplicity, in evaluating the matrix element of $\hat{H}_{\text{dd}}$ we set $1/(4\pi\varepsilon_0) = 1$, where ε_0 is the vacuum permittivity. We rewrite the dipole-dipole interaction in terms of spherical tensors [34,39], which allows $\hat{H}_{\text{dd}}$ to be written as [40]

$$\hat{H}_{\text{dd}} = - \frac{\sqrt{6}}{r^3} \sum_{q=-2}^2 (-1)^q C_q^{(2)}(\Omega_r) [\hat{\mathbf{d}}_1 \otimes \hat{\mathbf{d}}_2]_q^{(2)}. \quad (14)$$

Here, $\Omega_r = (\theta_r, \phi_r)$ denotes the polar and azimuthal angles specifying the orientation of the intermolecular separation vector $\mathbf{r}$ in the laboratory-fixed frame and $C_q^{(2)}(\Omega_r)$ denotes an unnormalized spherical harmonic. The molecular orientation with respect to the laboratory frame is described separately by the Euler angles $\Omega = (\alpha, \beta, \gamma)$. The term in the bracket,

$$[d_1 \otimes d_2]_q^{(2)} = \sum_{q'=-1}^1 \langle 1 q', 1 q - q' | 2 q \rangle d_{1,q'} d_{2,q-q'} \quad (15)$$

represent a rank-2 tensor product of the two dipole moments. Following the formulation detailed in Appendix A, the dipole operators are expressed in spherical tensor form as

$$d_0 = d_z, \quad d_{\pm 1} = \mp \frac{1}{\sqrt{2}}(d_x \pm id_y). \quad (16)$$

For molecules separated by a distance r with the electric field oriented along the laboratory z axis, the laboratory-frame orientation is $\Omega_r = (\theta_r, \phi_r) = (0, 0)$ and the $q = 0$ term is

the only nonvanishing contribution. Hence, the expression reduces to [31,41]

$$\hat{H}_{dd} = -\frac{1}{r^3}(2d_{1,0}d_{2,0} + d_{1,-1}d_{2,1} + d_{1,1}d_{2,-1}). \quad (17)$$

In this equation, the dipole components are expressed in the laboratory frame and must be transformed to the molecule-fixed frame. Accordingly,

$$d_{i,q} = \sum_{r_i=-1}^1 D_{q,r_i}^{1*}(\Omega_i) d_{i,r_i}^{(\text{mol})}, \quad (18)$$

where $q = -1, 0, 1$ and $r = -1, 0, 1$ label the laboratory-frame and molecule-fixed spherical components, respectively; $d_{i,r_i}^{(\text{mol})}$ are the known dipole components of the molecule i , and $D_{q,r_i}^{1*}(\Omega_i)$ is the Wigner D -matrix associated with the orientation Ω_i . Expressing the dipole-moment components in the molecule-fixed frame gives

$$\begin{aligned} \hat{H}_{dd} = & -\frac{1}{r^3} \sum_{r_1=-1}^1 \sum_{r_2=-1}^1 [2D_{0,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{0,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})} \\ & + D_{-1,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{1,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})} \\ & + D_{1,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{-1,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}]. \end{aligned} \quad (19)$$

The expression inside the brackets shows that the dipole-dipole interaction is projected onto the laboratory frame with the intermolecular axis oriented along the z axis. Physically, this implies the following contributions:

(1) The first term corresponds to both dipoles having a laboratory-frame projections $m_1 = 0$ and $m_2 = 0$; i.e., both dipoles are oriented along the laboratory Z axis.

(2) The second term describes a configuration in which dipole 1 has $m_1 = -1$, while dipole 2 has $m_2 = +1$.

(3) The third term corresponds to dipole 1 having $m_1 = +1$ and dipole 2 having $m_2 = -1$.

These are the only physically allowed combinations, as they conserve the total laboratory-frame projection quantum number ($m = m_1 + m_2$), when the intermolecular axis is oriented along the z axis. Consequently, the only nonzero matrix elements of $\hat{H}_{dd}$ are given by Eq. (C14).

We now construct the matrix elements of the dipole-dipole interaction in the dressed-state basis

$$\begin{aligned} |\downarrow\rangle &= \sum_{\tilde{j},\tilde{k}} c_{\tilde{j},\tilde{k}}^{(\downarrow)}(x = d\varepsilon/B) |\tilde{j},\tilde{k}, m=0\rangle, \\ |\uparrow\rangle &= \sum_{\tilde{j},\tilde{k}} c_{\tilde{j},\tilde{k}}^{(\uparrow)}(x = d\varepsilon/B) |\tilde{j},\tilde{k}, m=1\rangle. \end{aligned}$$

When the dipole-dipole Hamiltonian $\hat{H}_{dd}$ is projected onto the pseudospin basis $\{|\downarrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\uparrow\uparrow\rangle\}$, its structure naturally separates into two distinct physical contributions: a longitudinal (static) interaction, corresponding to diagonal matrix elements, and a transverse (resonant) exchange interaction, corresponding to off-diagonal matrix elements. The diagonal terms arise from the selection rules

$$\Delta m_1 = 0 \Rightarrow m_1 = m'_1, \quad \Delta m_2 = 0 \Rightarrow m_2 = m'_2,$$

and physically represent a classically interaction between the time-averaged static dipole moments induced by the external electric field. Although each molecule occupies a

rotational superposition, the strong dc field polarizes the molecules, resulting in a nonzero expectation value $\langle d_z \rangle$ along the quantization axis. Consequently, the dipole-dipole Hamiltonian reduces, by virtue of the selection rules discussed above, to purely diagonal contributions in the dressed pseudospin basis. These correspond to the matrix elements $\langle\downarrow\downarrow|\hat{H}_{dd}|\downarrow\downarrow\rangle$, $\langle\downarrow\uparrow|\hat{H}_{dd}|\downarrow\uparrow\rangle$, $\langle\uparrow\downarrow|\hat{H}_{dd}|\uparrow\downarrow\rangle$, and $\langle\uparrow\uparrow|\hat{H}_{dd}|\uparrow\uparrow\rangle$, which define the coefficients C_1 – C_4 , respectively:

$$\begin{aligned} C_1 &= - \sum_{\substack{\tilde{j}_1,\tilde{k}_1 \\ \tilde{j}'_1,\tilde{k}'_1}} \sum_{\substack{\tilde{j}_2,\tilde{k}_2 \\ \tilde{j}'_2,\tilde{k}'_2}} c_{\tilde{j}_1,\tilde{k}_1}^{(\downarrow)*} c_{\tilde{j}'_1,\tilde{k}'_1}^{(\downarrow)} c_{\tilde{j}_2,\tilde{k}_2}^{(\downarrow)*} c_{\tilde{j}'_2,\tilde{k}'_2}^{(\downarrow)} \\ &\quad \times [2 \langle\tilde{j}_1\tilde{k}_1 1|D_{0,\tilde{k}_1-\tilde{k}'_1}^1|\tilde{j}'_1\tilde{k}'_1 1\rangle \langle\tilde{j}_2\tilde{k}_2 1|D_{0,\tilde{k}_2-\tilde{k}'_2}^1|\tilde{j}'_2\tilde{k}'_2 1\rangle] \\ &\quad \times d_{1,\tilde{k}_1-\tilde{k}'_1} d_{2,\tilde{k}_2-\tilde{k}'_2}, \\ C_2 &= - \sum_{\substack{\tilde{j}_1,\tilde{k}_1 \\ \tilde{j}'_1,\tilde{k}'_1}} \sum_{\substack{\tilde{j}_2,\tilde{k}_2 \\ \tilde{j}'_2,\tilde{k}'_2}} c_{\tilde{j}_1,\tilde{k}_1}^{(\downarrow)*} c_{\tilde{j}'_1,\tilde{k}'_1}^{(\downarrow)} c_{\tilde{j}_2,\tilde{k}_2}^{(\uparrow)*} c_{\tilde{j}'_2,\tilde{k}'_2}^{(\uparrow)} \\ &\quad \times [2 \langle\tilde{j}_1\tilde{k}_1 1|D_{0,\tilde{k}_1-\tilde{k}'_1}^1|\tilde{j}'_1\tilde{k}'_1 1\rangle \langle\tilde{j}_2\tilde{k}_2 0|D_{0,\tilde{k}_2-\tilde{k}'_2}^1|\tilde{j}'_2\tilde{k}'_2 0\rangle] \\ &\quad \times d_{1,\tilde{k}_1-\tilde{k}'_1} d_{2,\tilde{k}_2-\tilde{k}'_2}, \\ C_3 &= - \sum_{\substack{\tilde{j}_1,\tilde{k}_1 \\ \tilde{j}'_1,\tilde{k}'_1}} \sum_{\substack{\tilde{j}_2,\tilde{k}_2 \\ \tilde{j}'_2,\tilde{k}'_2}} c_{\tilde{j}_1,\tilde{k}_1}^{(\uparrow)*} c_{\tilde{j}'_1,\tilde{k}'_1}^{(\uparrow)} c_{\tilde{j}_2,\tilde{k}_2}^{(\downarrow)*} c_{\tilde{j}'_2,\tilde{k}'_2}^{(\downarrow)} \\ &\quad \times [2 \langle\tilde{j}_1\tilde{k}_1 0|D_{0,\tilde{k}_1-\tilde{k}'_1}^1|\tilde{j}'_1\tilde{k}'_1 0\rangle \langle\tilde{j}_2\tilde{k}_2 1|D_{0,\tilde{k}_2-\tilde{k}'_2}^1|\tilde{j}'_2\tilde{k}'_2 1\rangle] \\ &\quad \times d_{1,\tilde{k}_1-\tilde{k}'_1} d_{2,\tilde{k}_2-\tilde{k}'_2}, \end{aligned}$$

and

$$\begin{aligned} C_4 &= - \sum_{\substack{\tilde{j}_1,\tilde{k}_1 \\ \tilde{j}'_1,\tilde{k}'_1}} \sum_{\substack{\tilde{j}_2,\tilde{k}_2 \\ \tilde{j}'_2,\tilde{k}'_2}} c_{\tilde{j}_1,\tilde{k}_1}^{(\uparrow)*} c_{\tilde{j}'_1,\tilde{k}'_1}^{(\uparrow)} c_{\tilde{j}_2,\tilde{k}_2}^{(\uparrow)*} c_{\tilde{j}'_2,\tilde{k}'_2}^{(\uparrow)} \\ &\quad \times [2 \langle\tilde{j}_1\tilde{k}_1 0|D_{0,\tilde{k}_1-\tilde{k}'_1}^1|\tilde{j}'_1\tilde{k}'_1 0\rangle \langle\tilde{j}_2\tilde{k}_2 0|D_{0,\tilde{k}_2-\tilde{k}'_2}^1|\tilde{j}'_2\tilde{k}'_2 0\rangle] \\ &\quad \times d_{1,\tilde{k}_1-\tilde{k}'_1} d_{2,\tilde{k}_2-\tilde{k}'_2}. \end{aligned}$$

The like-state interactions, C_1 and C_4 , correspond to the energy cost when both molecules occupy the same pseudospin manifold, either $|\downarrow\rangle$ or $|\uparrow\rangle$. In contrast, the unlike-state interactions, C_2 and C_3 , describe the interaction energy when the two molecules occupy different pseudospin states. These terms are particularly important in the context of the effective spin- $\frac{1}{2}$ description, as the longitudinal interactions give rise to the Ising anisotropy J_z and the effective longitudinal field h . Collectively, they play a central role in orienting the pseudospins along the z axis, favoring either ferromagnetic or antiferromagnetic order depending on the sign and relative magnitude of the interaction.

To quantify the longitudinal (static) interaction, we numerically evaluate the diagonal matrix elements C_1 – C_4 in the dressed-state basis as a function of the applied dc electric field. The results are shown in Fig. 4. At $\varepsilon = 0$, all coefficients vanish. Although chiral molecules possess indefinite parity due to the localization of enantiomers, the spatial isotropy of the field-free Hamiltonian ensures that the time-averaged dipole moment along any fixed laboratory axis satisfies $\langle \hat{d}_z \rangle = 0$, and thus no static dipole-dipole coupling is present.

As the electric field increases, the rotational states of opposite parity are mixed via the Stark effect, inducing a

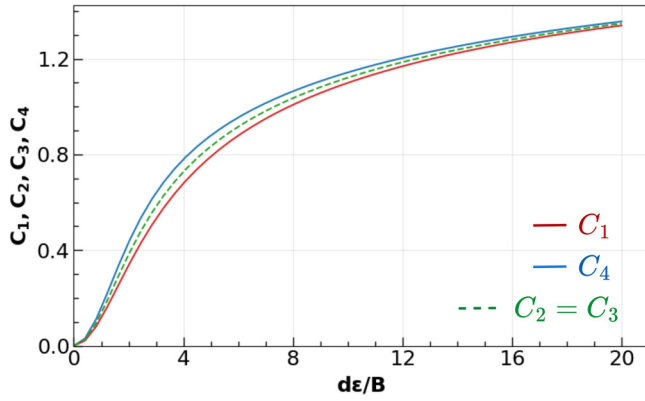


FIG. 4. Matrix elements of C_1 , C_2 , C_3 , and C_4 as functions of the dimensionless electric-field interaction parameter $d\epsilon/B$. The dotted green curves correspond to C_2 and C_3 , which are equal in magnitude for the (1,2-propanediol) molecule configuration. In contrast, C_1 (solid red) and C_4 (solid blue) are initially distinct but gradually converge at larger field strengths.

finite dipole moment oriented along the field direction. Since the diagonal interaction scales with the product of these induced moments, $C_{ij} \propto \langle \hat{d}_z \rangle_i \langle \hat{d}_z \rangle_j$, the coefficients increase monotonically with field strength. At larger fields ($d\epsilon/B \gtrsim 6$), the growth slows down and the coefficients begin to saturate, signaling the strong-field regime in which the molecules form highly oriented pendular states pinned along the field direction. In this limit, the dipole moment induced in the laboratory frame approaches the magnitude of the permanent molecular dipole, yielding a maximal static interaction strength.

While all four coefficients exhibit the same overall behavior, they separate into two distinct groups. The interactions between identical pseudospin states (C_1 for $|\downarrow\downarrow\rangle$ and C_4 for $|\uparrow\uparrow\rangle$, solid lines) are slightly stronger than the cross-state interactions (C_2 for $|\downarrow\uparrow\rangle$ and C_3 for $|\uparrow\downarrow\rangle$, dashed lines). This splitting arises from the different degrees of orientation of the dressed states $|\uparrow\rangle$ and $|\downarrow\rangle$, which lead to unequal induced dipole moments, $\langle \hat{d}_z \rangle_\uparrow \neq \langle \hat{d}_z \rangle_\downarrow$.

The off-diagonal coefficients C_{d_1} and C_{d_2} originate from the dipole-dipole selection rules

$$(m_1 - m'_1, m_2 - m'_2) = (-1, +1) \text{ or } (+1, -1),$$

which couple the pseudospin states $\langle \downarrow \uparrow | \hat{H}_{dd} | \uparrow \downarrow \rangle$ and $\langle \uparrow \downarrow | \hat{H}_{dd} | \downarrow \uparrow \rangle$, respectively:

$$\begin{aligned} C_{d_1} = & - \sum_{\tilde{j}_1, \tilde{k}_1} \sum_{\tilde{j}'_1, \tilde{k}'_1} c_{\tilde{j}_1, \tilde{k}_1}^{(\downarrow)*} c_{\tilde{j}'_1, \tilde{k}'_1}^{(\uparrow)} c_{\tilde{j}_2, \tilde{k}_2}^{(\uparrow)*} c_{\tilde{j}'_2, \tilde{k}'_2}^{(\downarrow)} \\ & \times \langle \tilde{j}_1 \tilde{k}_1 1 | D_{+1, \tilde{k}_1 - \tilde{k}'_1}^1 | \tilde{j}'_1 \tilde{k}'_1 0 \rangle \langle \tilde{j}_2 \tilde{k}_2 0 | D_{-1, \tilde{k}_2 - \tilde{k}'_2}^1 | \tilde{j}'_2 \tilde{k}'_2 1 \rangle \\ & \times d_{1, \tilde{k}_1 - \tilde{k}'_1} d_{2, \tilde{k}_2 - \tilde{k}'_2} \end{aligned}$$

and

$$\langle \uparrow \downarrow | \hat{H}_{dd} | \downarrow \uparrow \rangle$$

$$\begin{aligned} C_{d_2} = & - \sum_{\tilde{j}_1, \tilde{k}_1} \sum_{\tilde{j}'_1, \tilde{k}'_1} c_{\tilde{j}_1, \tilde{k}_1}^{(\uparrow)*} c_{\tilde{j}'_1, \tilde{k}'_1}^{(\downarrow)} c_{\tilde{j}_2, \tilde{k}_2}^{(\downarrow)*} c_{\tilde{j}'_2, \tilde{k}'_2}^{(\uparrow)} \\ & \times \langle \tilde{j}_1 \tilde{k}_1 0 | D_{-1, \tilde{k}_1 - \tilde{k}'_1}^1 | \tilde{j}'_1 \tilde{k}'_1 1 \rangle \langle \tilde{j}_2 \tilde{k}_2 1 | D_{+1, \tilde{k}_2 - \tilde{k}'_2}^1 | \tilde{j}'_2 \tilde{k}'_2 0 \rangle \\ & \times d_{1, \tilde{k}_1 - \tilde{k}'_1} d_{2, \tilde{k}_2 - \tilde{k}'_2}. \end{aligned}$$

These terms describe a quantum-coherent exchange, or flip-flop, process mediated by the transverse components of the dipole operator (d_+ , d_-). In this process, one molecule undergoes a transition from $|\uparrow\rangle \rightarrow |\downarrow\rangle$, while its neighbor is simultaneously excited from $|\downarrow\rangle \rightarrow |\uparrow\rangle$. This resonant energy transfer is driven by transition dipole moments that rotate in the plane perpendicular to the applied electric field.

Collecting all nonzero terms, the dipole-dipole Hamiltonian in the dressed basis $\{|\downarrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\uparrow\uparrow\rangle\}$ becomes

$$H_{dd} = -\frac{1}{r^3} \begin{pmatrix} C_1 & 0 & 0 & 0 \\ 0 & C_2 & C_{d_1} & 0 \\ 0 & C_{d_2} & C_3 & 0 \\ 0 & 0 & 0 & C_4 \end{pmatrix}. \quad (20)$$

Since we consider interactions between two nonidentical molecules—specifically, the L and R enantiomers of 1,2-propanediol ($C_3H_8O_2$)—the off-diagonal matrix elements C_{d_1} and C_{d_2} are related by Hermitian conjugation. These terms generally carry complex phases originating from the transverse dipole components $d_\pm$, whose molecule-fixed-frame expressions involve the combinations $\mp(d_b \pm id_c)/\sqrt{2}$. The presence of these complex phases constitutes the microscopic origin of the chirality-dependent interaction in the effective Hamiltonian given in Eq. (29). The coefficients C_{d_1} and C_{d_2} are directly proportional to the products $d_{1, \tilde{k}_1 - \tilde{k}'_1} d_{2, \tilde{k}_2 - \tilde{k}'_2}$, indicating that the interaction strength depends on which dipole-moment components couple the initial $\tilde{k}$ state to the final $\tilde{k}'$ state for each molecule. For the specific transition contributing to the C_{d_1} term, molecule 1 undergoes a transition $|\downarrow\rangle \rightarrow |\uparrow\rangle$, corresponding to a change in the laboratory-frame projection $m: 1 \rightarrow 0$. Conservation of angular momentum, therefore, requires the dipole operator to carry a laboratory-frame component $q = +1$, which maps in the molecule-fixed frame to the transverse dipole component $d_+^{(1)}$. Similarly, molecule 2 undergoes the transition $|\uparrow\rangle \rightarrow |\downarrow\rangle$, corresponding to $m: 0 \rightarrow 1$. In this case, angular momentum conservation requires a laboratory-frame component $q = -1$, which in the molecule-fixed frame corresponds to the transverse dipole component $d_-^{(2)}$. For these specific off-diagonal matrix elements, the generic indices $\tilde{k} - \tilde{k}'$ are fixed by the character of the transition, such that

$$d_{1, \tilde{k}_1 - \tilde{k}'_1} \rightarrow d_+^{(1)}, \quad d_{2, \tilde{k}_2 - \tilde{k}'_2} \rightarrow d_-^{(2)}.$$

As a result, the matrix element C_{d_1} is given by the product of these dipole components multiplied by a real Wigner coefficient W ,

$$C_{d_1} \propto W \times (d_+^{(1)} d_-^{(2)}).$$

Here, $d_{\pm} = \mp \frac{d_b \pm id_c}{\sqrt{2}}$ are the spherical-tensor components of the dipole operator in the molecule-fixed frame. The chirality of 1,2-propanediol ($C_3H_8O_2$) is encoded in the sign of the c -axis dipole component d_c . Consider a heterochiral pair in which molecule 1 is left-handed ($+d_c$) and molecule 2 is right-handed ($-d_c$). For the left-handed (L) enantiomer,

$$d_+^{(L)} = -(d_b + id_c), \quad d_-^{(L)} = (d_b - id_c),$$

while for the right-handed (R) enantiomer,

$$d_+^{(R)} = -(d_b - id_c), \quad d_-^{(R)} = (d_b + id_c).$$

The product of the transition dipoles for the L-R pair is therefore

$$d_+^{(1)} d_-^{(2)} = d_+^{(L)} d_-^{(R)} = -(d_b + id_c)(d_b + id_c), \quad (21)$$

which evaluates to

$$d_+^{(L)} d_-^{(R)} = -(d_b^2 - d_c^2) - 2id_b d_c. \quad (22)$$

The nonzero imaginary part constitutes the microscopic origin of the DMI, reflecting constructive interference between the handedness of the two molecules, leading to a chiral twisting of excitation transfer that breaks inversion symmetry. For the opposite ordering, where molecule 1 is right-handed and molecule 2 is left-handed, one finds

$$d_+^{(1)} d_-^{(2)} = d_+^{(R)} d_-^{(L)} = -(d_b - id_c)(d_b - id_c), \quad (23)$$

which yields

$$d_+^{(R)} d_-^{(L)} = -(d_b^2 - d_c^2) + 2id_b d_c. \quad (24)$$

Thus, exchanging the order of the enantiomers from an L-R pair to an R-L pair reverses the sign of the imaginary contribution, confirming that the interaction possesses a vector chirality determined by the spatial arrangement of the enantiomers. In contrast, for a homochiral pair (LL or RR), the transition dipoles carry conjugate phases that cancel exactly:

$$\begin{aligned} d_+^{(1)} d_-^{(2)} &= d_+^{(L)} d_-^{(L)} = -(d_b + id_c)(d_b - id_c), \\ d_+^{(L)} d_-^{(L)} &= (d_b + d_c)^2. \end{aligned} \quad (25)$$

This product is purely real. Consequently, the imaginary term vanishes ($D = 0$) and the interaction reduces to a pure symmetric exchange (J_{xy}). In order to validate the above analytic derivation of the origin of the DMI, we numerically evaluated the imaginary part of the off-diagonal coupling coefficient C_d as a function of the applied electric field. The results are shown in Fig. 5, providing a direct visualization of the symmetry breaking discussed in Eqs. (22) and (24). The solid blue curve corresponds to the right-left (RL) heterochiral pair and exhibits a positive DMI strength ($D > 0$), while the dashed red curve corresponds to the left-right (LR) configuration and shows an equal magnitude but opposite sign ($D < 0$). The mirror symmetry of the two curves is in perfect agreement with the analytical prediction that exchanging the spatial order

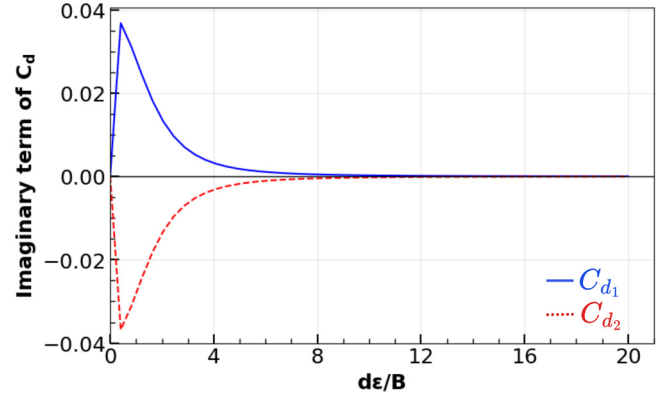


FIG. 5. Imaginary parts of the dipolar coefficients C_{d1} and C_{d2} as functions of the electric-field parameter $d\epsilon/B$. The two coefficients are equal in magnitude and opposite in sign ($C_{d1} = -C_{d2}$); this antisymmetry constitutes the microscopic origin of the effective Dzyaloshinskii-Moriya interaction, which causes neighboring spins to be slightly twisted relative to each other, rather than strictly parallel or antiparallel.

of the enantiomers reverses the sign of the interference term,

$$\Im[d_+^{(L)} d_-^{(R)}] = -\Im[d_+^{(R)} d_-^{(L)}],$$

confirming that the interaction possesses a vector chirality determined by the structural handedness of the molecules.

A further key feature apparent in Fig. 5 is that the DMI is not a static property, but is widely tunable by the external electric field. At $\epsilon = 0$, the interaction vanishes because the rotational eigenstates have well-defined parity, which forbids the simultaneous $\Delta m = \pm 1$ transitions required for the flip-flop process. At intermediate fields, $d\epsilon/B \approx 0.5$, the DMI reaches a pronounced maximum, corresponding to an optimal regime in which the Stark effect sufficiently mixes states of opposite parity to activate the transverse transition dipoles $d_{\pm 1}$ without locking the rotors so tightly that transverse fluctuations are suppressed. At larger fields, the DMI decreases as the molecules become strongly polarized along the z axis, approaching the Ising limit in which transverse flip-flop exchange processes are quenched in favor of static longitudinal interactions.

C. Realization of the XXZ spin-1/2 model from asymmetric-top molecules

As we are considering a pair of interacting 1,2-propanediol molecules ($C_3H_8O_2$), prepared in opposite enantiomer configurations, one left-handed and one right-handed. Restricting each molecule to an effective two-level dressed rotational manifold, the composite system is described in the pseudospin basis $\{|\downarrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\uparrow\uparrow\rangle\}$.

The total Hamiltonian consists of single-molecule rotational and Stark contributions together with the intermolecular dipole-dipole interaction,

$$\hat{H}_{\text{tot}} = \hat{H}_{\text{rot}}^{(L)} + \hat{H}_{\text{rot}}^{(R)} + \hat{H}_{\text{dc}}^{(L)} + \hat{H}_{\text{dc}}^{(R)} + \hat{H}_{\text{dd}}^{(LR)}. \quad (26)$$

In the pseudospin basis, the Hamiltonian takes the matrix form

$$\hat{H}_{\text{tot}} = \begin{pmatrix} 2E_{\downarrow} - \frac{C_1}{r^3} & 0 & 0 & 0 \\ 0 & E_{\downarrow} + E_{\uparrow} - \frac{C_2}{r^3} & \frac{C_{d_1}}{r^3} & 0 \\ 0 & \frac{C_{d_2}}{r^3} & E_{\uparrow} + E_{\downarrow} - \frac{C_3}{r^3} & 0 \\ 0 & 0 & 0 & 2E_{\uparrow} - \frac{C_4}{r^3} \end{pmatrix}. \quad (27)$$

Here, $E_{\uparrow}$ and $E_{\downarrow}$ denote the single-molecule dressed energies, while the coefficients C_1 – C_4 arise from diagonal dipole-dipole interactions. The off-diagonal coefficients C_{d_1} and C_{d_2} originate from resonant dipolar exchange processes between the two molecules. A crucial step in constructing the effective spin model is the unification of physical units. The single-molecule energies ($E_{\uparrow}$, $E_{\downarrow}$) are naturally expressed in frequency units set by the rotational constants, whereas the dipole-dipole interaction coefficients initially carry geometric units of electric dipole density (e.g., Debye²/nm³). To restore dimensional consistency, the interaction energy is converted to frequency units by normalizing with Planck's constant $\hbar$. We define a distance-dependent scaling factor

$$\Psi(r) = \frac{1}{\hbar} \left(\frac{|\mathbf{d}|^2}{4\pi\epsilon_0 r^3} \right), \quad (28)$$

which sets the maximal interaction strength between two dipoles of magnitude $|\mathbf{d}|$ separated by a distance r . The effective coupling constants are obtained as the product of this physical scale and the dimensionless geometric coefficients C_i .

Projecting onto the effective two-qubit subspace (see the details in Appendix D), the interacting molecular dipoles map onto an anisotropic spin- $\frac{1}{2}$ XXZ Heisenberg model,

$$\begin{aligned} \hat{H}_{\text{spin}} = & \sum_{j=1}^{N-1} [J_{xy} (\hat{\sigma}_j^x \hat{\sigma}_{j+1}^x + \hat{\sigma}_j^y \hat{\sigma}_{j+1}^y) - D (\hat{\sigma}_j^x \hat{\sigma}_{j+1}^y - \hat{\sigma}_j^y \hat{\sigma}_{j+1}^x) \\ & + J_z \hat{\sigma}_j^z \hat{\sigma}_{j+1}^z] + \hbar \sum_{j=1}^N \hat{\sigma}_j^z. \end{aligned} \quad (29)$$

The effective coupling constants are given by

$$\begin{aligned} J_{xy} &= -\frac{\Psi(r)}{2} \Re(C_{d_1}), \\ D &= \frac{\Psi(r)}{2} \Im(C_{d_1}), \\ J_z &= \frac{\Psi(r)}{4} [(C_2 + C_3) - (C_1 + C_4)], \\ \hbar &= \frac{2(E_{\uparrow} - E_{\downarrow}) + \Psi(r)(C_1 - C_4)}{4}. \end{aligned} \quad (30)$$

The transverse couplings J_{xy} and D originate from the off-diagonal dipole-dipole matrix elements and describe coherent resonant exchange of rotational excitation between the two molecules, corresponding to flip-flop processes $|\downarrow \uparrow\rangle \leftrightarrow |\uparrow \downarrow\rangle$. The imaginary component of this exchange gives rise to the chiral Dzyaloshinskii-Moriya interaction D . In contrast, the

longitudinal parameters J_z and $\hbar$ arise from diagonal dipole-dipole interactions and the single-molecule Stark splitting. These terms encode the static interaction landscape generated by field-induced permanent dipoles aligned along the laboratory axis.

To determine the ground-state phase diagram, we simplify the Hamiltonian by applying a gauge transformation that absorbs the Dzyaloshinskii-Moriya interaction into the transverse exchange coupling J_{xy} [42,43]. This is achieved by performing a site-dependent rotation about the z axis, defined by the unitary operator (see Appendix D 3 for details),

$$U = \prod_{i=1}^N e^{-i\phi_i \hat{\sigma}_i^z},$$

where the rotation angle ϕ_i increase along the chain as $\phi_i = j\theta$. If we choose the twist angle $\theta = \tan^{-1}(D/J_{xy})$, the antisymmetric DMI term and the symmetric exchange term will combine, and a renormalized planar interaction will form [44]

$$\tilde{J}_{xy} = \sqrt{J_{xy}^2 + D^2}.$$

The transformed Hamiltonian takes the form of a conventional XXZ spin model, characterized by transverse and longitudinal couplings $\tilde{J}_{xy}$ and J_z , respectively:

$$\hat{H}_{\text{spin}} = \sum_{j=1}^{N-1} [\tilde{J}_{xy} (\hat{\sigma}_j^x \hat{\sigma}_{j+1}^x + \hat{\sigma}_j^y \hat{\sigma}_{j+1}^y) + J_z \hat{\sigma}_j^z \hat{\sigma}_{j+1}^z] + \hbar \sum_{j=1}^N \hat{\sigma}_j^z.$$

Because the unitary transformation U leaves the energy spectrum invariant while rotating the eigenstates, the phase boundaries of the chiral Luttinger liquid in the laboratory frame are identical to those of the standard Luttinger liquid in the rotated frame. To characterize chirality, we examine spin-spin correlation functions, which quantify how a spin at site i couples to a spin at site j . In a standard LL, the transverse correlations decay algebraically as [45]

$$\langle \hat{\sigma}_i^- \hat{\sigma}_j^+ \rangle_{\text{LL}} \sim \frac{1}{|i-j|^{\frac{1}{2K}}},$$

where $K = 1$ for a Luttinger liquid. The physical spin operators in the laboratory frame are related to the effective spin operators in the rotated frame by a site-dependent twist,

$$\hat{\sigma}_{i,\text{lab}}^+ = e^{-i\theta j} \hat{\sigma}_{j,\text{eff}}^+.$$

As a result, when evaluating correlation functions in the laboratory frame, an additional phase factor emerges [46]. Using the relation between laboratory-frame and rotated-frame spin

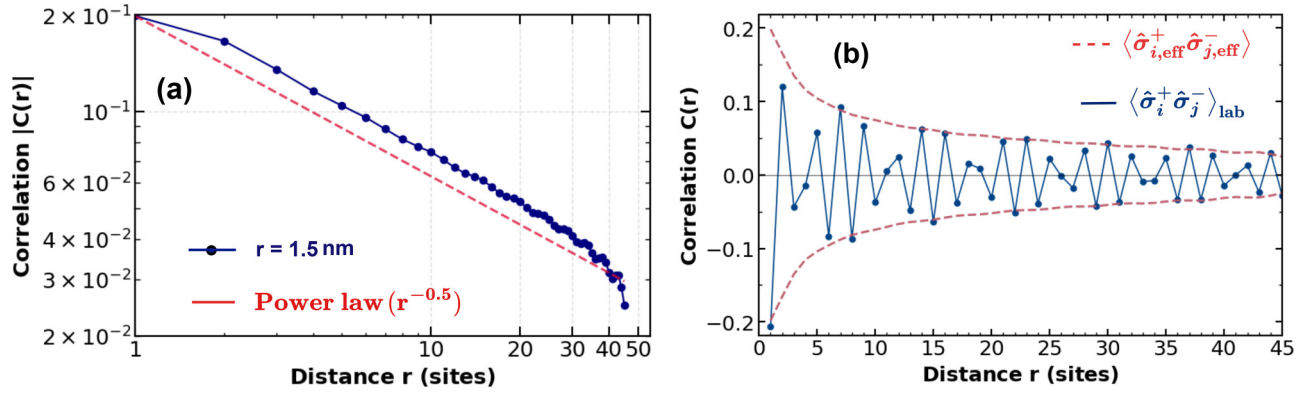


FIG. 6. (a) The red dotted line represents the reference power-law decay $r^{-0.5}$, while the solid blue line shows the standard Luttinger liquid behavior after the gauge transformation, in which J_{xy} and D combine into the effective coupling $\tilde{J}_{xy} = \sqrt{J_{xy}^2 + D^2}$. (b) The red dotted curve represents the standard Luttinger liquid correlation plotted on a linear scale and serves as an envelope for the correlations in the presence of the Dzyaloshinskii-Moriya interaction. The oscillations of the laboratory-frame correlations (solid blue curve) within this envelope arise from the phase factor $e^{i\theta(i-j)}$, providing clear evidence of the twisting induced by the DMI. The corresponding twist angle is given by $\theta = \tan^{-1}(D/J_{xy})$.

operators,

$$\hat{\sigma}_{j,\text{lab}}^+ = e^{-i\theta j} \hat{\sigma}_{j,\text{eff}}^+, \quad \hat{\sigma}_{j,\text{lab}}^- = e^{+i\theta j} \hat{\sigma}_{j,\text{eff}}^-,$$

the transverse spin-spin correlation function in the laboratory frame becomes

$$\langle \hat{\sigma}_i^+ \hat{\sigma}_j^- \rangle_{\text{lab}} = \langle e^{-i\theta i} \hat{\sigma}_{i,\text{eff}}^+ e^{+i\theta j} \hat{\sigma}_{j,\text{eff}}^- \rangle.$$

Factoring out the phase yields

$$\langle \hat{\sigma}_i^+ \hat{\sigma}_j^- \rangle_{\text{lab}} = e^{i\theta(j-i)} \langle \hat{\sigma}_{i,\text{eff}}^+ \hat{\sigma}_{j,\text{eff}}^- \rangle.$$

The correlator $\langle \hat{\sigma}_{i,\text{eff}}^+ \hat{\sigma}_{j,\text{eff}}^- \rangle$ is the standard Luttinger liquid correlation, which decays algebraically with distance. The additional phase factor $e^{i\theta(j-i)}$ induces an oscillatory spatial modulation, causing the laboratory-frame correlations to acquire a spiral structure—characteristic of a chiral Luttinger liquid.

Figure 6(a) shows the correlation function in the interacting limit for a system size of $N = 100$. The dotted black line represents the reference power-law decay $r^{-0.5}$ [45], while the solid blue line with filled circles illustrates the decay of correlations for a standard Luttinger liquid when $\tilde{J}_{xy} = \sqrt{J_{xy}^2 + D^2}$. The coupling constants are fixed at $\tilde{J}_{xy} \approx 6.4$ GHz, $J_z \approx -0.2454$, and $\hbar \approx 9.550$, and the objective is to study the response of the quantum state to this specific set of parameters. The coupling J_{xy} is responsible for generating quantum correlations that propagate along the spin chain, while the longitudinal field $\hbar$ competes with this process by favoring spin polarization, thereby tending to orient all spins either up or down. At an intermolecular separation of $r = 1.5$ nm, the ratio $\hbar/J_{xy} \approx 1.5$. Therefore, for this specific molecular geometry, the system remains well within the Luttinger liquid regime. This is clearly evidenced in the correlation function: For a long chain ($N = 100$), the correlations exhibit a power-law decay of the form

$$\langle S_i^+ S_j^- \rangle \sim \frac{1}{|i-j|^{0.5}}.$$

In Fig. 6(b), we plot the correlation function $\langle \hat{\sigma}_i^+ \hat{\sigma}_j^- \rangle_{\text{lab}}$, measured in the laboratory frame, which explicitly includes the

physical twist induced by the Dzyaloshinskii-Moriya interaction. The red curve represents the effective-frame correlation $\langle \hat{\sigma}_{i,\text{eff}}^+ \hat{\sigma}_{j,\text{eff}}^- \rangle$, previously shown in Fig. 6(a), plotted here on a linear scale. This curve acts as an envelope for the laboratory-frame correlations, since the oscillatory spiral structure (blue curve) is bounded by the condition $|e^{i\theta(i-j)}| = 1$ and therefore cannot exceed the effective-frame amplitude. The blue curve exhibits a clear wavelike pattern, confirming the presence of the phase factor $e^{i\theta(i-j)}$. The period of these oscillations is directly determined by the twist angle $\theta = \tan^{-1}(D/J_{xy})$.

III. RESULTS AND DISCUSSION

A. Effective coupling constants (J_{xy} , J_z , D , and $\hbar$)

Figures 7(a)–7(d) show the evolution of the effective coupling constants (J_{xy} , J_z , D , and $\hbar$) as functions of the dimensionless dc electric-field strength $d\varepsilon/B$ for the L and R enantiomers of 1,2-propanediol at different intermolecular separations. The symmetric exchange coupling J_{xy} varies non-monotonically with the electric-field strength $d\varepsilon/B$, as shown in Fig. 7(a). The nonmonotonic behavior of J_{xy} originates from Stark-induced mixing of the molecular rotational eigenstates, as k is not a good quantum number for asymmetric-top molecules and the electric field therefore mixes different k components.

In the weak-field regime, the coupling increases rapidly and attains a maximum near $d\varepsilon/B \approx 0.5$, indicating that the rotational states remain only weakly polarized. In this regime, the applied electric field breaks parity symmetry and induces strong mixing between states of opposite parity. As a result, the transition dipole matrix elements are significantly enhanced, giving rise to a pronounced coherent spin-exchange process described by $(\hat{\sigma}_1^+ \hat{\sigma}_2^- + \hat{\sigma}_1^- \hat{\sigma}_2^+)$. In contrast, in the strong-field regime ($d\varepsilon/B \gg 1$), the off-diagonal transition amplitudes that mediate spin-flip processes are progressively suppressed, leading to an asymptotic reduction of the exchange coupling J_{xy} toward zero. This behavior arises because the molecular states become strongly Stark localized and enter the pendular regime, where the wave functions acquire a

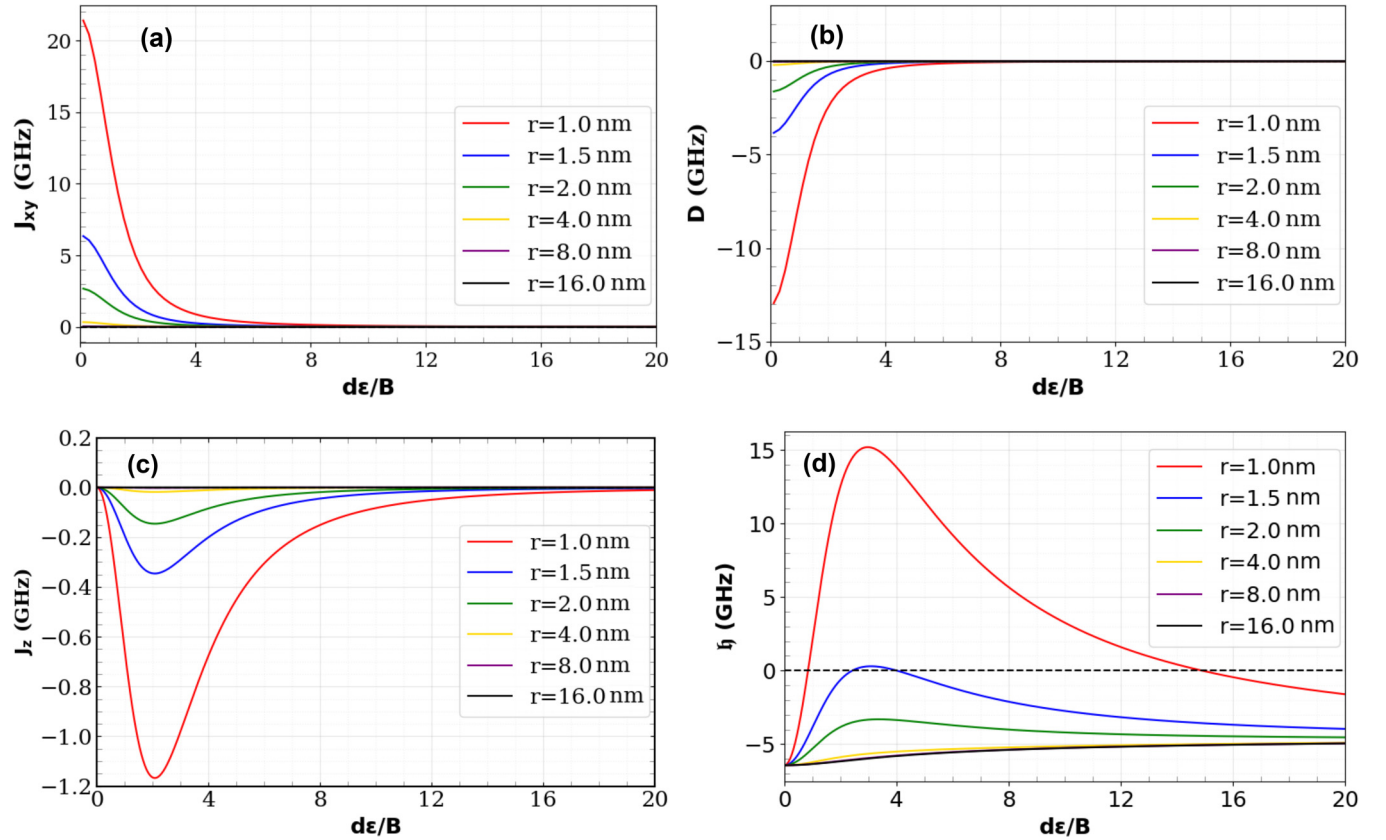


FIG. 7. Effective coupling parameters of an asymmetric-top molecular Hamiltonian mapped onto a spin-chain model, illustrating their dependence on the intermolecular separation r between L and R enantiomers. (a) Symmetric exchange coupling J_{xy} and (b) Dzyaloshinskii-Moriya interaction D , arising from the left- and right-handed enantiomers, decrease monotonically as the electric-field parameter $d\epsilon/B$ is increased. (c) The Ising interaction J_z remains negative throughout the entire parameter range, indicating a robust ferromagnetic bias. (d) The effective magnetic field h displays a strong nonmonotonic behavior, including a sign reversal at short intermolecular distances ($r = 1.0$ nm), followed by an asymptotic decay.

dominant magnetic quantum number (m) character, which remains a good quantum number.

Observing appreciable quantum correlations requires the enantiomers to be separated by an optimal intermolecular distance. As evident from Fig. 7(a), J_{xy} is the dominant interaction mechanism at very short separations, i.e., on the nanoscale. At the optimal field strength $d\epsilon/B \approx 0.5$, the exchange coupling reaches approximately 33 GHz for an intermolecular distance of $r = 1.0$ nm (red solid line) but is reduced to about 10 GHz when the separation increases to $r = 1.5$ nm (blue solid line). This strong sensitivity of the coupling to the intermolecular separation underscores the need for precise spatial control in experimental realizations, as subnanometer variations in distance can produce significant changes in the interaction strength. For larger separations, $r \geq 4.0$ nm (yellow, purple, and black solid lines), the exchange coupling becomes essentially negligible (< 1 GHz) for all values of the dc-field-induced detuning.

Figure 7(b) presents the Dzyaloshinskii-Moriya interaction parameter D for the real molecular system $C_3H_8O_2$ in a heterochiral RL molecular configuration. Notably, the magnitude of the resulting DMI is comparable to values typically encountered in solid-state systems containing heavy elements, where strong relativistic spin-orbit coupling is present. In contrast, herein the chiral interaction is generated purely by

molecular geometry because the intrinsic chirality of the 1,2-propanediol molecule locally breaks inversion symmetry. On the other hand, the applied dc electric field breaks inversion symmetry globally by mixing (hybridizing) rotational states and thereby creating states of indefinite parity, which renders the transition dipole moment $d_{\pm 1}$ nonzero. When a spin excitation propagates from a right-handed molecule ($|\downarrow\rangle$) to a left-handed molecule ($|\uparrow\rangle$), the transfer is no longer trivial; instead, the excitation undergoes a chiral twisting process and acquires a direction-dependent geometric (Berry) phase. This phase manifests itself microscopically as the Dzyaloshinskii-Moriya interaction, favoring orthogonal spin orientations over collinear alignment and stabilizing chiral spin textures. For the RL configuration, the acquired phase is $-\pi/2$, and the resulting DMI is antisymmetric under site exchange, ($D_{ij} = -D_{ji}$). Reversing the ordering to an LR configuration, therefore, flips the sign of D while leaving its magnitude unchanged. Physically, this sign reversal corresponds to inverting the handedness of the effective synthetic spin-orbit coupling. This realization of DMI thus renders the molecular spin-orbit coupling highly tunable, in contrast to its atomic counterpart, which is intrinsically fixed by the material composition. Such tunability constitutes the principal advantage of mapping the DMI term onto the underlying molecular geometry. We note that the handedness of the molecular frame is defined by the

scalar triple product $\mathbf{d}_a \cdot (\mathbf{d}_b \times \mathbf{d}_c)$, whose sign changes under inversion between L and R enantiomers [36,47]. This sign reversal constitutes the microscopic origin of the corresponding sign change of the DMI parameter D . Figure 7(b) clearly demonstrates that the strength of the chiral interaction can be continuously tuned from zero up to ~ 15 GHz by varying the dc electric-field strength $d\epsilon/B$. This tunability enables controlled access to phase transitions between nonchiral and chiral spin-spiral phases.

The Ising coupling J_z for the heterochiral RL molecular pair is shown in Fig. 7(c). Over the examined range of the applied electric-field parameter $d\epsilon/B$, J_z remains consistently negative, indicating a robust ferromagnetic bias ($J_z < 0$). Physically, this implies that the dipole-dipole interaction energy is minimized when neighboring molecules occupy identical rotational states, either $|\uparrow\uparrow\rangle$ or $|\downarrow\downarrow\rangle$. Since the Ising coupling arises from the difference between like-spin and unlike-spin interactions ($C_{\uparrow\uparrow} + C_{\downarrow\downarrow}$ versus $C_{\uparrow\downarrow} + C_{\downarrow\uparrow}$), a weak applied field only slightly polarizes the molecules. Consequently, the induced dipole moments $\langle d_z \rangle$ associated with the two spin states remain small and nearly identical, rendering the energy difference between oriented and antioriented spins negligible and driving J_z toward zero, a trend that also appears in the strong-field limit. At intermediate values of $d\epsilon/B$, however, the Stark effect mixes the rotational states differently for $|\uparrow\rangle$ and $|\downarrow\rangle$, producing a maximal contrast in the attainable laboratory orientation. This enhances the energy difference between oriented and antioriented dipole configurations, thereby stabilizing a ferromagnetic regime. Accessing a quantum phase transition requires the transverse couplings (J_{xy} and D) to become comparable to J_z , which can be achieved by operating either at $d\epsilon/B < 2.5$ or $d\epsilon/B > 2.5$.

Figure 7(d) presents the effective transverse magnetic field $\hbar$, which sets the single-site energy cost for creating a spin excitation. The magnitude and sign of $\hbar$ result from a competition between two distinct physical contributions: (1) The first is the internal Stark splitting, given by the energy difference between the dressed rotational states $E_{\uparrow} - E_{\downarrow}$, which is strictly positive and increases monotonically with the applied electric field. (2) The second is an external dipolar bias, arising from the difference in electrostatic environments experienced by the two states and proportional to the geometric factor $\Omega(r) [C_1 - C_4]$. At large intermolecular separations ($r \gtrsim 2.0$ nm), the dipolar bias is strongly suppressed by its $1/r^3$ scaling and becomes negligible. In this regime, $\hbar$ is dominated by the Stark splitting and remains large and positive, favoring a paramagnetic configuration in which spins are effectively frozen into the ground state.

For the smallest separation $r = 1.0$ nm (red solid line), Fig. 7(d) reveals a nontrivial reentrant behavior characterized by two distinct resonance points. The first crossing occurs at a low field ($d\epsilon/B \approx 0.8$), where the rising Stark splitting initially compensates the static dipolar bias. Upon further increasing the field, the system enters a regime of strong dipolar saturation, and a second zero crossing appears near $d\epsilon/B \approx 15$, beyond where $\hbar$ becomes negative again. This double crossing indicates that at extremely short distances, the geometric dipolar bias remains sufficiently strong to overwhelm the Stark splitting even at high fields. While physically

rich, this behavior creates a narrow and potentially unstable window for quantum simulation, as small fluctuations in the electric field may drive the system out of the critical regime.

In contrast, the intermediate separation $r = 1.5$ nm (blue solid line) exhibits a single, well-defined resonance at $d\epsilon/B \approx 2.5$. In this case, the dipolar interaction is strong enough to achieve field cancellation, yet sufficiently moderate to avoid the reentrant zero-field crossing ($\hbar \rightarrow 0$) observed at high fields ($d\epsilon/B \approx 15$) for shorter distances. For fields above the resonance, $\hbar$ remains small and positive, providing a stable and controllable platform for exploring the phase diagram. This identifies $r = 1.5$ nm as an optimal distance for experimental realization, balancing large interaction strengths with a monotonic and predictable tuning parameter.

IV. PHASE DIAGRAM

Figure 8(a) presents the ground-state phase diagram of the nearest-neighbor spin-1/2 XXZ model [48–50]. The horizontal axis is scaled by the anisotropy parameter $J_z/\tilde{J}_{xy}$, where $\tilde{J}_{xy} = \sqrt{J_{xy}^2 + D^2}$ denotes the effective transverse exchange coupling after the local gauge transformation and the vertical axis is scaled by the field strength $\hbar/\tilde{J}_{xy}$. The phase diagram exhibits two gapped phases: (1) an antiferromagnetic phase for $J_z/\tilde{J}_{xy} > 1$ and (2) a ferromagnetic phase for $J_z/\tilde{J}_{xy} < -1$. These two phases are separated by a gapless Luttinger liquid phase [51], which occurs for $-1 < J_z/\tilde{J}_{xy} < 1$. According to Eq. (30), for asymmetric-top molecules the ratio $J_z/\tilde{J}_{xy}$ depends solely on the dimensionless field strength $d\epsilon/B$. Accordingly, Fig. 8(b) illustrates the evolution of $J_z/\tilde{J}_{xy}$ as $d\epsilon/B$ is increased from 0 to 20. The results show that the anisotropy remains consistently negative but is strictly confined to the interval $-0.3 \lesssim J_z/\tilde{J}_{xy} \lesssim 0$. The ratio attains a minimum value of approximately -0.27 at an intermediate field strength $d\epsilon/B \approx 8$, coinciding with a pronounced dip in J_z , where the molecular polarizability is maximal. Because $-0.27 > -1$, the molecular system never enters the pure Ising ferromagnetic phase and instead remains in the easy-plane regime in which quantum fluctuations are dominant. To fully characterize the phase behavior of the asymmetric molecular configuration, it is therefore necessary to determine the accessible range of $\hbar/\tilde{J}_{xy}$. Figure 8(c) shows the dependence of $\hbar/\tilde{J}_{xy}$ on $d\epsilon/B$ for several intermolecular separations r . At low electric fields ($d\epsilon/B \approx 0.5$ – 1.0), pronounced dips appear in the curves. These features do not indicate any physical inconsistency of the system; rather, they are a consequence of the logarithmic scale used, since $\hbar/\tilde{J}_{xy} \rightarrow 0$ leads to a divergence in the logarithm, producing sharp features in Fig. 8(c). The use of a logarithmic scale on the vertical axis is required by the strong sensitivity of the dipolar interaction to the intermolecular distance; a linear scale would compress the critical transition boundary ($\hbar/\tilde{J}_{xy} = 1$) onto the horizontal axis, thereby obscuring the Luttinger liquid window. We identify these dips as optimal operating points, where quantum fluctuations are maximized due to the competition between two mechanisms.

Specifically, at low electric fields, the exchange interaction $\tilde{J}_{xy}$ is enhanced relative to the Stark splitting energy $\hbar$. Within this narrow regime, the kinetic energy associated with flip-flop

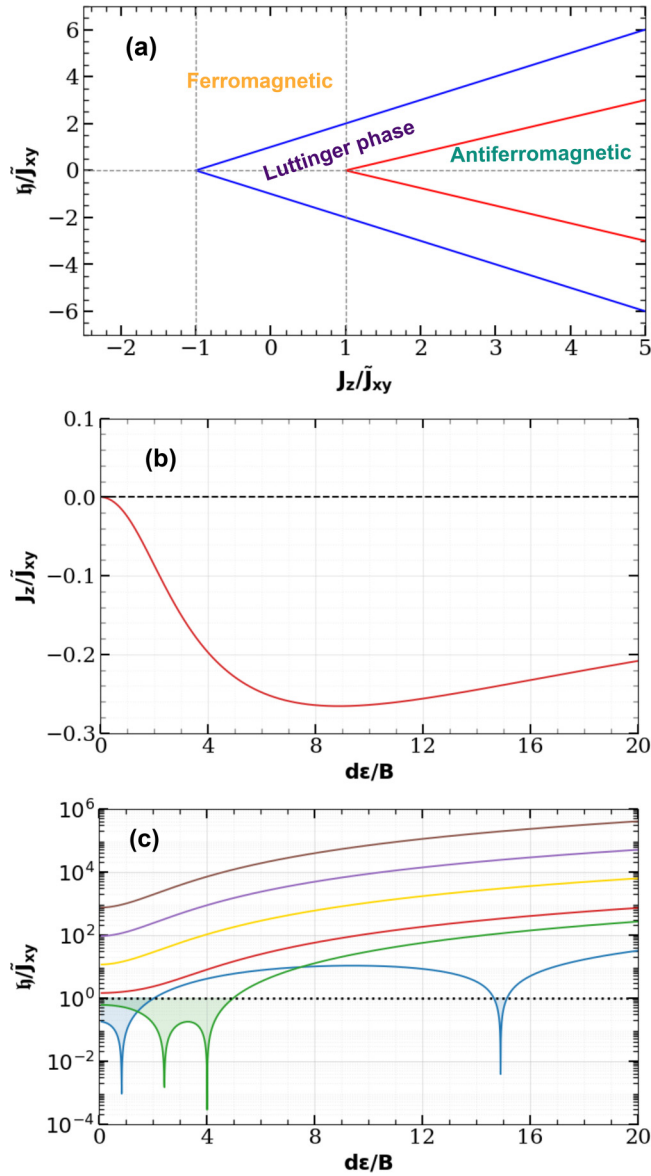


FIG. 8. (a) Universal ground-state phase diagram of the XXZ spin chain as a function of the normalized Ising coupling $J_z/\tilde{J}_{xy}$ and field $\hbar/\tilde{J}_{xy}$, where the effective transverse coupling is defined as $\tilde{J}_{xy} = \sqrt{J_{xy}^2 + D^2}$. (b) The anisotropy ratio $J_z/\tilde{J}_{xy}$ for Stark-dressed asymmetric-top molecules indicates a robust ferromagnetic Ising bias ($J_z < 0$) that persists across the entire field range. (c) Ratio $\hbar/\tilde{J}_{xy}$ as a function of the dimensionless electric-field parameter $d\epsilon/B$ for several intermolecular separations r . The dotted horizontal line marks the quantum critical value $\hbar/\tilde{J}_{xy} \approx 1$; regions below (above) this line correspond to the Luttinger liquid (ferromagnetic) phase. The shaded region highlights an experimentally accessible window where quantum correlations are stabilized by the interplay between Stark dressing and dipole-dipole interactions.

processes dominates over the static energy cost of spin flips ($\tilde{J}_{xy} \gg \hbar$), driving the system well below unity and into the shaded green and blue regions. These shaded regions correspond to parameter regimes that predominantly support the formation of a gapless Luttinger liquid phase. As the electric field is increased ($d\epsilon/B > 5$), the Stark splitting $\hbar$ continues

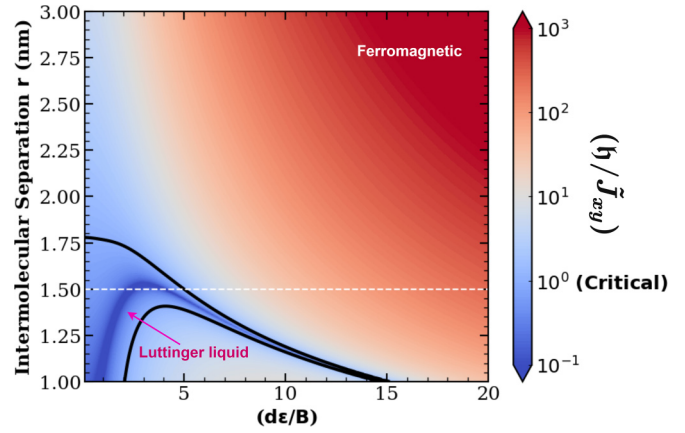


FIG. 9. Ground-state phase diagram of the XXZ spin chain realized in a linear array of asymmetric-top molecules, shown as a function of the field parameter $d\epsilon/B$ and the intermolecular separation r . The color scale represents the ratio $\hbar/\tilde{J}_{xy}$, while the solid black lines denote the quantum critical boundary $\hbar \approx \tilde{J}_{xy}$, which separates the Luttinger liquid phase from the ferromagnetic phase.

to grow while $\tilde{J}_{xy}$ is progressively suppressed, thereby driving the system into a trivial field-polarized phase. At the shortest intermolecular separation ($r = 1.0$ nm), the effective field approaches zero ($\hbar \rightarrow 0$) at two distinct points: first at very low fields ($d\epsilon/B \approx 0.5$ – 1.0) and again near a specific higher field ($d\epsilon/B \approx 15$). The latter point is unstable and highly sensitive to geometric details, making it a challenging regime for controlled simulations. The green curve in Fig. 8(c), corresponding to $r = 1.5$ nm, identifies the Goldilocks regime for realizing the Luttinger liquid phase. This separation is optimal because, unlike $r = 1.0$ nm, which exhibits sharp dives toward zero and unstable resonances, or $r = 2.0$ nm (red curve), which only marginally approaches the phase boundary, the $r = 1.5$ nm geometry provides a broad and stable operational window. The shaded green region further indicates that the system remains in the Luttinger liquid phase over a wide range of electric fields ($d\epsilon/B \approx 0.5 \rightarrow 4.5$), providing experimentalists with substantial flexibility to tune the system without risking the onset of uncontrolled higher-order multipole interactions.

The contour plot in Fig. 9 shows the ground-state phase diagram of a one-dimensional (1D) 1,2-propanediol chain with nearest-neighbor interactions, plotted as a function of the dimensionless electric-field strength $d\epsilon/B$ and the intermolecular separation. The color scale represents the ratio $\hbar/\tilde{J}_{xy}$, which quantifies the competition between electric-field-induced polarization and dipole-mediated quantum exchange processes. The white dashed line corresponds to a realistic experimental scenario in which the L and R enantiomers are fixed at an intermolecular separation of $r = 1.5$ nm. Tracing this line from left to right, the system initially resides in the trivial (red) phase, then enters the quantum (blue) phase near $d\epsilon/B \approx 2.0$, and remains in this phase over a broad and stable parameter window before exiting at higher fields. This behavior further confirms that $r = 1.5$ nm represents an optimal lattice spacing, providing a robust operational window for realizing the chiral Luttinger liquid phase. The solid black lines mark the critical quantum phase boundary defined

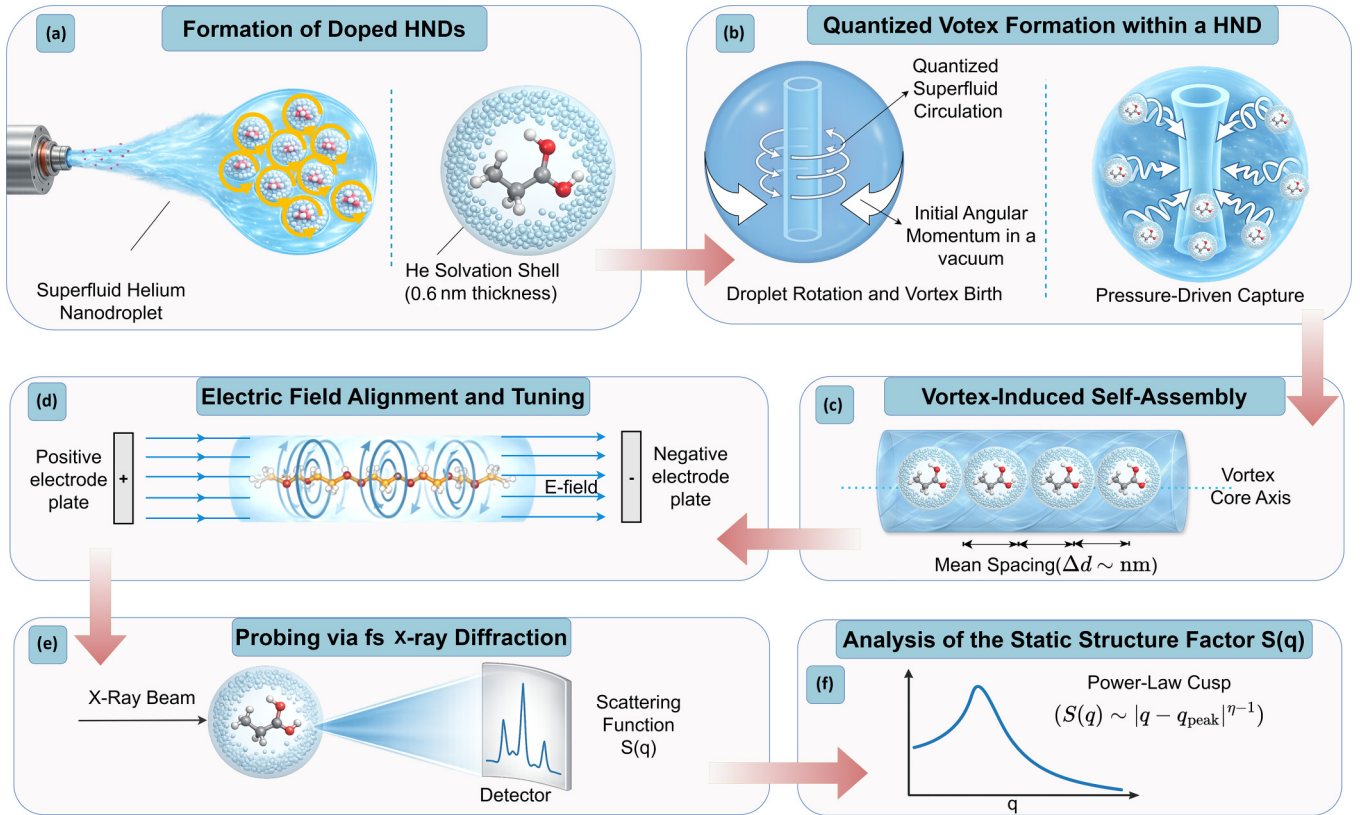


FIG. 10. Schematic of the experimental protocol for realizing and detecting a CLL in superfluid helium nanodroplets. (a) Formation of superfluid HND doped with 1,2-propanediol molecules via coexpansion of ^4He gas seeded with 1,2-propanediol with superfluid ^4He into vacuum through a cryogenic nozzle. The panel depicts the cooling and solvation of the dopant molecules (left) as well as a dopant molecule within a helium solvation shell that forms due to van der Waals attraction between the molecule and helium (right). (b) Quantized vortex formation within an HND. As the droplet expands into vacuum through the nozzle, it cools and acquires angular momentum. Below the superfluid transition, a quantized vortex core is formed to conserve angular momentum. The resulting pressure gradient (Bernoulli force) drives the solvated molecules radially inward, trapping them in the vortex core. (c) Vortex-induced self-assembly. The quantized vortex core within a given superfluid droplet guides the solvated molecules doped therein into a 1D linear filament. The mean spacing is defined by the effective diameter of the solvation shell. Based on density functional theory (DFT) calculations [52,53], the shell thickness is $\Delta R_{\text{shell}} \approx 6 \text{ \AA}$. Thus, the spacing is estimated as $\Delta r \approx 2 \times (R_{\text{mol}} + \Delta R_{\text{shell}}) \approx 2 \times (2.5 \text{ \AA} + 6.0 \text{ \AA}) \approx 1.7 \text{ nm}$. (d) Electric-field alignment and tuning of the interaction strength. An external dc electric field $\mathbf{E}$, applied via parallel plate electrodes, aligns the molecular dipoles along the vortex axis (head-to-tail), stabilizes the chain, and allows for tuning the interaction strength. (e) Structural probing via femtosecond x-ray diffraction to measure the scattering function. (f) Analysis of the static structure factor $S(q)$. The CLL phase is distinguished from a crystalline solid by the absence of a sharp Bragg peak and the emergence of a power-law singularity, $S(q) \sim |q - q_{\text{peak}}|^{\eta-1}$, characteristic of the algebraic decay of correlations in 1D quantum fluids [45].

by $\hbar/\tilde{J}_{xy} \approx 1$, separating the gapless chiral Luttinger liquid phase from the field-polarized ferromagnetic phase. The blue region enclosed by these boundaries does not form a flat band; instead, it develops distinct lobelike structures that extend upward at specific electric-field values ($d\epsilon/B \approx 2.5$). These lobes correspond to resonance points where $\hbar \approx 0$, as analyzed in Fig. 8(c). Within these lobes, the external Stark splitting is nearly perfectly canceled by the geometric dipolar interactions. This cancellation stabilizes the quantum phase at larger intermolecular separations, allowing it to persist up to $r \approx 1.8 \text{ nm}$, which would otherwise be inaccessible.

V. PROPOSED EXPERIMENTAL IMPLEMENTATION

Conventional experimental platforms

Figure 10 summarizes this protocol formation of doped helium nanodroplets (a), quantized vortex formation (b),

vortex-induced molecular self-assembly (c), electric-field alignment (d), structural probing via femtosecond x-ray diffraction (e), and analysis of the static structure factor (f). Studying the CLL phase requires an intermolecular spacing of $r \approx 1.7 \text{ nm}$, which lies in a regime that poses severe challenges for conventional experimental platforms. Although advanced optical trapping techniques, such as structured light, allow precise control over neutral particles [54], a major limitation remains the attainable trap size. Conventional optical lattices are restricted by the diffraction limit of light, leading to minimum lattice site spacings of about 300–500 nm [55,56]. Such separations are too large to accomplish sufficiently strong dipolar interactions and, as a result, preclude the formation of a Luttinger liquid phase.

Turning to nanophotonics, where evanescent electromagnetic fields are exploited to generate traps that are much smaller than the wavelength of light, such as optical

nanofiber traps [57] and metallic plasmonic nanostructures [58], enabled trapping and manipulating single atoms with nanometer-scale precision. In particular, plasmonic nano-optical tweezers [59] and nanoplasmonic lattices [60] have been successful in this regard. However, close proximity of trapped atoms or molecules to surfaces introduces deleterious decoherence channels, including Johnson-Nyquist noise and Casimir-Polder interactions [61], which are known to destabilize fragile coherent quantum phases. A robust realization of the CLL phase thus requires a confinement platform that provides subnanometer positional control while remaining free from surface-induced noise. Such combination of requirements is naturally met by superfluid helium nanodroplets.

Superfluid helium nanodroplets (HNDs), comprised of ^4He atoms provide a cold environment (0.38 K), which is largely perturbation-free to the extent that molecules embedded in it can retain their rotational degrees of freedom [52,62,63]. Imparting angular momentum to the HNDs results in quantized vortex lines that act as effective one-dimensional confinement channels. While doping the droplets via pick up of molecules during passage through a vapor cell has a limited efficiency (leading to loading of just a few molecules per droplet), a coexpansion with a helium beam seeded with the atoms or molecules of interest (such as 1,2-propanediol in our case or Xe in Ref. [63]) upstream from the nozzle greatly increases the doping efficiency. For Xe, the number of Xe atoms, N_{Xe} , loaded into a droplet comprised of $N_{\text{He}} \approx 10^6$ helium atoms was shown to be as high as $N_{\text{Xe}} \approx 10^{-3}N_{\text{He}}$ [63].

Meanwhile, coherent diffractive imaging experiments showed that heavy dopants preferentially localize along vortex cores [64], producing characteristic Bragg features in the diffraction signal. These experiments thus provide direct structural evidence of vortex-confined matter—and a concrete roadmap for achieving the nanometer-scale, quasi-one-dimensional regime, which is key to creating and studying the chiral Luttinger phase. In particular, x-ray diffraction could be used to probe the unique periodicity of the collective density wave of a chiral Luttinger liquid phase and thereby obtain its definitive signature. The diffraction intensity is directly proportional to the static structure factor $S(q)$, defined as the Fourier transform of the density-density correlation function:

$$S(q) = \frac{1}{N} \sum_{j,l} e^{-iq(r_j - r_l)} \langle \hat{n}_j \hat{n}_l \rangle, \quad (31)$$

where $\hat{n}_j$ denotes the local density (or effective spin projection) at site j . This quantity measures the strength of spatial correlations at a given wave vector q , revealing the dominant ordering pattern of the system. Detecting diffraction features at a wave vector $q = 2\pi/(1.7 \text{ nm})$, corresponding to intermolecular spacing $r = 1.7 \text{ nm}$, would therefore provide direct structural evidence for the formation of the CLL phase. Crucially, the Luttinger liquid state could be distinguished from trivial ordered or fluid phases by analyzing lineshape of the diffraction peak. Unlike the sharp, delta-function-like Bragg peaks of a crystalline solid or the broad, smooth features characteristic of a conventional fluid, a Luttinger liquid exhibits power-law singularities at $2k_F$ [45,65,66]. As a result, the diffraction peaks are expected to display a characteristic

cusplike profile governed by an algebraic decay exponent, thereby providing a rigorous signature of the underlying quantum critical state.

More generally, a chiral Luttinger liquid is characterized by a chiral excitation spectrum, a well-defined collective plasmon velocity, and power-law correlation dynamics. In solid-state electronic systems, these properties are typically inferred from transport observables, such as quantized conductance or Hall response. For isolated quantum fluids such as HNDs, however, direct contact-based measurements are not possible. In this case, the appropriate experimental analog is time-resolved pump-probe spectroscopy. In this experimental framework, a femtosecond pump pulse generates a localized excitation within the vortex-confined molecular filament, and a time-delayed probe pulse subsequently interrogates its propagation and relaxation. If the underlying phase is chiral, the excitation dynamics should display strict directionality, indicative of broken time-reversal symmetry, and follow the non-Fermi-liquid power-law decay associated with Luttinger physics. Although conventional transport measurements are not feasible in this platform, the excitation spectrum and relaxation dynamics offer a rigorous and experimentally meaningful alternative to transport-based evidence.

A key challenge in neutral droplet experiments is the interaction time, limited to millisecond-scale due to high beam velocities (200–400 m/s). To enable the extended observation times necessary for pump-probe spectroscopy, we propose utilizing electrostatic trapping of charged nanodroplets, a technique recently demonstrated to store HNDs for up to 1 min [67]. A critical concern is whether the presence of a net charge on the droplet surface would generate electrostatic noise disruptive of the delicate dipole-dipole interactions that drive the CLL phase. The intermolecular dipole-dipole interaction at $r = 1.7 \text{ nm}$ is approximately $V_{\text{dd}} \approx 1.0 \text{ K}$. In contrast, the Stark perturbation from a surface charge on a macroscopic droplet ($R \approx 500 \text{ nm}$) is estimated at $V_{\text{charge}} \approx 0.003 \text{ K}$. Since the signal-to-noise ratio $V_{\text{dd}}/V_{\text{charge}} \sim 300$, the phase can thus be expected to be robust against electrostatic perturbations. For more details, see Appendix E. Furthermore, although achieving strong dc electric fields in superfluid droplets can be challenging due to breakdown limits, fields on the order of 10 kV/cm (corresponding to $d\varepsilon/B \approx 2$) are achievable and sufficient to hybridize the rotational states required [68].

VI. CONCLUSION

In this paper, we demonstrate that a heteroenantiomer (R-L) pair can be mapped onto an effective Heisenberg XXZ spin model, exhibiting an emergent DMI in the presence of an external electric field and dipole-dipole coupling. The effective spin-1/2 description is realized by projecting onto the two lowest pendular states of the molecules, each formed as a coherent superposition of rotational basis states, and labeled by different values of $\tilde{j}$ and $\tilde{k}$. An external electric field is then used to couple these two states, forming an effective two-level system that provides a platform for studying the Heisenberg model based on chiral molecules. In order to characterize the model comprehensively, we investigate a wide parameter regime involving the rotational constants, the strength and

orientation of the external electric field, and the strength of the dipole-dipole coupling between the molecules. The external electric field is essential for controlling and engineering the effective spin interactions: It is required for hybridizing the rotational states as well as generating nonzero transition dipole moments C_{d_1} and C_{d_2} that make the requisite one-photon transition $|\downarrow\rangle \leftrightarrow |\uparrow\rangle$ fully allowed.

In previous studies, linear, spherical, and symmetric-top molecules were employed, with the pendular states $|00\rangle$ and $|10\rangle$ chosen to represent an effective two-level system. However, because these systems possess inversion symmetry, they did not allow for the study of heteroenantiomer (R-L) interactions. In our model, the Dzyaloshinskii-Moriya interaction arises naturally from the interference between the transition dipole moments of heterochiral (R-L) enantiomers. This establishes a clear connection between molecular chirality and the resulting spin-spin interactions, thus providing a distinct advantage for controlling and engineering effective spin couplings. Through a rigorous analysis of the quantum phase diagrams shown in Figs. 8 and 9, we identify an optimal experimental regime characterized by an intermolecular separation of $r \approx 1.5$ nm and intermediate electric-field strengths $d\varepsilon/B$, where the chiral Luttinger liquid phase emerges and remains protected against trivial phases. A further advantage of the model presented herein is that, in contrast to solid-state platforms where spin-orbit coupling is fixed by material properties, the chiral Dzyaloshinskii-Moriya interaction can be continuously tuned using an external field. In their sum, these properties establish arrays of 1,2-propanediol molecules as a promising platform for quantum simulation, as this work bridges the gap between single-molecule rotational spectroscopy and many-body condensed-matter physics.

Moreover, utilizing an array of asymmetric-top molecules has the following virtues:

Topological switching. By replacing L enantiomers with R enantiomers in the molecular array, the sign of D is instantaneously reversed. As a consequence, the chirality of the resulting Luttinger liquid is inverted, switching the spiral ordering from clockwise to counterclockwise (or vice versa).

Engineered domain walls. If a heterogeneous molecular chain is constructed, for example, $\cdots \text{L-L-L-R-R-R} \cdots$, the sign of the DMI parameter D necessarily changes at the interface between the two enantiomeric regions. This enforced sign change generates a topological domain wall, which can host exotic fractionalized excitations or localized zero modes. In this way, quantum defects can be engineered deterministically through chemical substitution alone.

As a future extension, the present model can be engineered to simulate the Su-Schrieffer-Heeger (SSH) model [69] by introducing lattice dimerization, thereby giving rise to topologically protected zero-energy modes localized at the chain boundaries [70]. This suggests that molecular arrays may serve as decoherence-free quantum memories [71], in which quantum information is stored in topologically protected edge states and remains immune to thermal noise in the interior of the chain. The present study thus contributes to the exploration of complex topological phases, which may yield insights into the microscopic mechanisms responsible for CISS in biological and chemical systems

[23,72]. Electron transfer in biological systems, such as photosynthesis, occurs over long distances with surprisingly low scattering. While highly speculative, if such systems were to exhibit a topological phase akin to the SSH model [73], one might expect transport pathways involving edgewise channels that are comparatively resilient to biological environmental noise.

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M.A.A. conceptualized the study, performed theoretical derivations for the Stark-dressed rotational states and dipole-dipole interactions, conducted numerical simulations for the phase-diagram analysis, and drafted the manuscript. B.F. contributed to the theoretical framework of molecular chirality and the quantum mechanical analysis of asymmetric-top molecules in external fields. S.K. conceptualized the study, supervised the research project, provided guidance on the many-body dynamics and mapping to the XXZ Heisenberg Hamiltonian, and finalized the manuscript for submission.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: DERIVATION OF THE INTERACTING HAMILTONIAN

In quantum mechanics and rotational spectroscopy, spherical tensors are preferred over Cartesian tensors because they can transform naturally between the laboratory and molecule frames. In spherical representation, spatial directions are labeled by angular momentum quantum numbers rather than Cartesian components (x, y, z) . Consequently, to describe two interacting asymmetric-top molecules such as 1,2-propanediol, it is natural to work in the basis $|jkm\rangle$, which follows directly from the spherical-tensor formalism and allows a symmetric transformation of vector operators between frames.

A spherical tensor of rank l represents an object with angular momentum l . Since an angular momentum l has $2l + 1$ possible projections along a quantization axis, labeled by $m = -l, -l + 1, \dots, l$, a spherical tensor of rank l consists of $2l + 1$ components $T_m^{(l)}$.

Consider a two-dimensional vector $\mathbf{v}$, such as the dipole moment of a molecule, rigidly attached to the molecular frame. The same vector may be described from two complementary perspectives: (1) in the molecule-fixed frame, where the vector remains fixed relative to the molecular body and has components $\mathbf{v} = (v_x, v_y)$; and (2) in the laboratory frame, where the observer is fixed and the molecule rotates, yielding rotated components $\mathbf{v} = (v_X, v_Y)$. If the molecule is rotated

by an angle θ , the transformation between these components follows directly from elementary trigonometry:

$$v_X = v_x \cos \theta - v_y \sin \theta, \quad (\text{A1})$$

$$v_Y = v_x \sin \theta + v_y \cos \theta. \quad (\text{A2})$$

We can write this as a rotation matrix:

$$\begin{pmatrix} v_X \\ v_Y \end{pmatrix} = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} v_x \\ v_y \end{pmatrix}. \quad (\text{A3})$$

In three dimensions, transformations of spherical components are described using Wigner D -matrices rather than ordinary rotation matrices. Thus, the molecule-to-laboratory frame transformation can be expressed as

$$T_q^{(l)} = \sum_{r=-l}^l D_{q,r}^{l*}(\Omega) T_r^{(l)}. \quad (\text{A4})$$

Here, $T_r^{(l)}$ denotes the tensor components in the molecule-fixed frame, $T_q^{(l)}$ denotes the corresponding components in the laboratory frame, and $D_{q,r}^{l*}(\Omega)$ is the Wigner D -matrix element that specifies how the r th molecular-frame component contributes to the q th laboratory-frame component. The inverse transformation, which maps laboratory-frame measurements back to the molecule's intrinsic properties, is given by

$$T_r^{(l)} = \sum_{q=-l}^l D_{q,r}^{l*}(\Omega) T_q^{(l)}. \quad (\text{A5})$$

For a vector (rank-1 tensor, $l = 1$), we have three spherical components: $T_{-1}^{(1)}$, $T_0^{(1)}$, and $T_1^{(1)}$. Suppose we know the vector in the molecule frame: $(T_{-1}^{(1)}, T_0^{(1)}, T_1^{(1)})$. To find it in the laboratory frame, we perform the summation over $r = -1, 0, 1$:

$$T_{-1}^{(1)} = D_{-1,-1}^{1*}(\Omega) T_{-1}^{(1)} + D_{-1,0}^{1*}(\Omega) T_0^{(1)} + D_{-1,1}^{1*}(\Omega) T_1^{(1)}, \quad (\text{A6a})$$

$$T_0^{(1)} = D_{0,-1}^{1*}(\Omega) T_{-1}^{(1)} + D_{0,0}^{1*}(\Omega) T_0^{(1)} + D_{0,1}^{1*}(\Omega) T_1^{(1)}, \quad (\text{A6b})$$

$$T_1^{(1)} = D_{1,-1}^{1*}(\Omega) T_{-1}^{(1)} + D_{1,0}^{1*}(\Omega) T_0^{(1)} + D_{1,1}^{1*}(\Omega) T_1^{(1)}. \quad (\text{A6c})$$

In this paper, we employ this transformation to study the dipole-dipole interaction:

$$\hat{H}_{\text{dd}} = \frac{\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2 - 3(\hat{\mathbf{d}}_1 \cdot \mathbf{e}_r)(\hat{\mathbf{d}}_2 \cdot \mathbf{e}_r)}{r^3}, \quad (\text{A7})$$

where $\hat{\mathbf{d}}_1$ and $\hat{\mathbf{d}}_2$ represent molecular dipole moments, which are rank-1 tensor operators ($l = 1$). Their spherical components in the molecule-fixed frame are denoted by $d_{1,r}^{(1)}$ and $d_{2,r}^{(1)}$, with $r = -1, 0, 1$. For dipole 1, the laboratory-frame spherical components are related to the molecule-fixed components by

$$d_{1,q}^{(\text{lab})} = \sum_{r_1=-1}^1 D_{q,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})}, \quad (\text{A8})$$

where Ω_1 specifies the orientation of molecule 1. Similarly, for dipole 2:

$$d_{2,q}^{(\text{lab})} = \sum_{r_2=-1}^1 D_{q,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}, \quad (\text{A9})$$

with Ω_2 denoting the orientation of molecule 2.

We are primarily interested in the dot product of the two dipole-moment operators, which in spherical components is given by

$$\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2 = \sum_{s=-1}^1 (-1)^s d_{1,s}^{(\text{lab})} d_{2,-s}^{(\text{lab})}. \quad (\text{A10})$$

Substituting the laboratory-frame transformations yields

$$\begin{aligned} \hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2 &= \sum_{s=-1}^1 (-1)^s \left[\sum_{r_1=-1}^1 D_{s,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} \right] \\ &\times \left[\sum_{r_2=-1}^1 D_{-s,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})} \right]. \end{aligned} \quad (\text{A11})$$

Similarly, the term $(\hat{\mathbf{d}}_1 \cdot \mathbf{e}_r)(\hat{\mathbf{d}}_2 \cdot \mathbf{e}_r)$ requires a transformation of the dipole components along the intermolecular axis. Since $\mathbf{e}_r$ is chosen along the laboratory Z axis, its spherical components satisfy $e_{r,0} = 1$ and $e_{r,\pm 1} = 0$, leading to

$$\hat{\mathbf{d}}_1 \cdot \mathbf{e}_r = d_{1,0}^{(\text{lab})} = \sum_{r_1=-1}^1 D_{0,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})}, \quad (\text{A12})$$

with an analogous expression for dipole 2. Consequently, the dipole-dipole Hamiltonian $\hat{H}_{\text{dd}}$ can be written as a double sum over r_1 and r_2 involving products of Wigner D -matrices.

To evaluate the matrix elements of the interaction, $\langle j_1 k_1 m_1, j_2 k_2 m_2 | \hat{H}_{\text{dd}} | j'_1 k'_1 m'_1, j'_2 k'_2 m'_2 \rangle$, one must compute matrix elements of the form

$$\langle j_1 k_1 m_1 | D_{q,r_1}^{1*}(\Omega_1) | j'_1 k'_1 m'_1 \rangle \quad \text{and} \quad \langle j_2 k_2 m_2 | D_{q,r_2}^{1*}(\Omega_2) | j'_2 k'_2 m'_2 \rangle. \quad (\text{A13})$$

These are given by the Wigner-Eckart theorem using Wigner 3- j symbols:

$$\begin{aligned} &\langle j_1 k_1 m_1 | D_{q,r_1}^{1*}(\Omega_1) | j'_1 k'_1 m'_1 \rangle \\ &= (-1)^{m_1+k_1} \sqrt{(2j_1+1)(2j'_1+1)} \\ &\times \begin{pmatrix} j_1 & 1 & j'_1 \\ -m_1 & q & m'_1 \end{pmatrix} \begin{pmatrix} j_1 & 1 & j'_1 \\ -k_1 & r_1 & k'_1 \end{pmatrix} \end{aligned} \quad (\text{A14})$$

and

$$\begin{aligned} &\langle j_2 k_2 m_2 | D_{q,r_2}^{1*}(\Omega_2) | j'_2 k'_2 m'_2 \rangle \\ &= (-1)^{m_2+k_2} \sqrt{(2j_2+1)(2j'_2+1)} \\ &\times \begin{pmatrix} j_2 & 1 & j'_2 \\ -m_2 & q & m'_2 \end{pmatrix} \begin{pmatrix} j_2 & 1 & j'_2 \\ -k_2 & r_2 & k'_2 \end{pmatrix}. \end{aligned} \quad (\text{A15})$$

The only nonzero matrix elements are those satisfying the selection rules $m_1 = q + m'_1$, $m_2 = q + m'_2$, $k_1 = r_1 + k'_1$, and $k_2 = r_2 + k'_2$.

APPENDIX B: $\hat{H} = \hat{H}_{\text{rot}} + \hat{H}_{\text{dc}}$

We have derived a comprehensive formulation, based on Wigner D -matrices, for transforming operators between the laboratory and molecule-fixed frames. This formalism applies directly to the rotational and Stark terms of the Hamiltonian, $\hat{H}_{\text{rot}}$ and $\hat{H}_{\text{dc}}$.

The dc Stark interaction is given by

$$\hat{H}_{\text{dc}} = -\mathbf{d} \cdot \boldsymbol{\varepsilon}, \quad (\text{B1})$$

where $\mathbf{d}$ is the dipole-moment operator and $\boldsymbol{\varepsilon}$ is the external dc electric field. Both $\mathbf{d}$ and $\boldsymbol{\varepsilon}$ may be expressed as rank-1 spherical tensors, leading to

$$\mathbf{d} \cdot \boldsymbol{\varepsilon} = \sum_{q=-1}^1 (-1)^q d_q \varepsilon_{-q}, \quad (\text{B2})$$

where d_q and ε_{-q} denote the spherical components in the laboratory frame. Consequently,

$$\hat{H}_{\text{dc}} = - \sum_{q=-1}^1 (-1)^q d_q \varepsilon_{-q}. \quad (\text{B3})$$

Since the molecular dipole moment is naturally defined in the molecule-fixed frame through the components $d_r^{(\text{mol})}$, whereas d_q denotes the corresponding components in the laboratory frame, it is necessary to transform d_q into the molecule-fixed

frame. This transformation is given by

$$d_q = \sum_{r=-1}^1 D_{q,r}^{1*}(\Omega) d_r^{(\text{mol})}, \quad (\text{B4})$$

where $d_r^{(\text{mol})}$ are fixed molecular constants and Ω specifies the molecular orientation.

Substituting Eq. (B3) into Eq. (B4) yields

$$\hat{H}_{\text{dc}} = - \sum_{q=-1}^1 (-1)^q \left[\sum_{r=-1}^1 D_{q,r}^{1*}(\Omega) d_r^{(\text{mol})} \right] \varepsilon_{-q}, \quad (\text{B5})$$

which may be written equivalently as

$$\hat{H}_{\text{dc}} = - \sum_{q=-1}^1 \sum_{r=-1}^1 (-1)^q D_{q,r}^{1*}(\Omega) d_r^{(\text{mol})} \varepsilon_{-q}. \quad (\text{B6})$$

The corresponding matrix elements in the $|jkm\rangle$ basis are therefore

$$\begin{aligned} \langle jkm | \hat{H}_{\text{dc}} | j'k'm' \rangle \\ = - \sum_{q=-1}^1 \sum_{r=-1}^1 (-1)^q \langle jkm | D_{q,r}^{1*}(\Omega) | j'k'm' \rangle d_r^{(\text{mol})} \varepsilon_{-q}. \end{aligned} \quad (\text{B7})$$

Using the standard expression for the matrix elements of the Wigner D -matrix, this becomes

$$\langle jkm | \hat{H}_{\text{dc}} | j'k'm' \rangle = - \sum_{q=-1}^1 \sum_{r=-1}^1 (-1)^q d_r^{(\text{mol})} \varepsilon_{-q} (-1)^{m+k} \sqrt{(2j+1)(2j'+1)} \begin{pmatrix} j & 1 & j' \\ -m & q & m' \end{pmatrix} \begin{pmatrix} j & 1 & j' \\ -k & r & k' \end{pmatrix}. \quad (\text{B8})$$

Since the external electric field has only a single component along the laboratory z axis, $\boldsymbol{\varepsilon} = \varepsilon_z \mathbf{e}_z$, its spherical components satisfy

$$\varepsilon_0 = \varepsilon_z, \quad \varepsilon_{\pm 1} = 0. \quad (\text{B9})$$

As a result, only the $q = 0$ term contributes to the dc Stark Hamiltonian. The matrix elements therefore reduce to

$$\langle jkm | \hat{H}_{\text{dc}} | j'k'm' \rangle = - \sum_{r=-1}^1 (-1)^0 d_r^{(\text{mol})} \varepsilon_0 (-1)^{m+k} \sqrt{(2j+1)(2j'+1)} \begin{pmatrix} j & 1 & j' \\ -m & 0 & m' \end{pmatrix} \begin{pmatrix} j & 1 & j' \\ -k & r & k' \end{pmatrix}. \quad (\text{B10})$$

Using $\varepsilon_0 = \varepsilon_z$, this expression simplifies to

$$\langle jkm | \hat{H}_{\text{dc}} | j'k'm' \rangle = -\varepsilon_z \sum_{r=-1}^1 d_r^{(\text{mol})} (-1)^{m+k} \sqrt{(2j+1)(2j'+1)} \begin{pmatrix} j & 1 & j' \\ -m & 0 & m' \end{pmatrix} \begin{pmatrix} j & 1 & j' \\ -k & r & k' \end{pmatrix}. \quad (\text{B11})$$

From this expression, several selection rules follow immediately. First, the condition $m = m'$ reflects conservation of the laboratory-frame projection quantum number. Second, the relation $k' = k - r$ with $r = -1, 0, 1$ implies that the molecule-fixed projection can change only according to $\Delta k = 0, \pm 1$. Finally, the Wigner 3- j symbol

$$\begin{pmatrix} j & 1 & j' \\ -m & 0 & m' \end{pmatrix} \quad (\text{B12})$$

is nonzero only when $|j - j'| \leq 1$. Combined with the condition $m = m'$, this yields the selection rule $\Delta j = 0, \pm 1$.

When we consider the full system of two interacting molecules, the dc interaction part becomes

$$\hat{H}_{\text{dc}} = \hat{H}_{\text{dc},1} + \hat{H}_{\text{dc},2} = - \sum_{i=1}^2 \hat{\mathbf{d}}_i \cdot \boldsymbol{\varepsilon}. \quad (\text{B13})$$

As $\hat{H}_{\text{dc},1}$ acts only on molecule 1, the wave function of molecule 2 remains unchanged, so

$$\begin{aligned} \langle j_1 k_1 m_1, j_2 k_2 m_2 | \hat{H}_{\text{dc},1} | j'_1 k'_1 m'_1, j'_2 k'_2 m'_2 \rangle \\ = \langle j_1 k_1 m_1 | \hat{H}_{\text{dc},1} | j'_1 k'_1 m'_1 \rangle \delta_{j_2, j'_2} \delta_{k_2, k'_2} \delta_{m_2, m'_2}. \end{aligned} \quad (\text{B14})$$

Similarly, for $\hat{H}_{dc,2}$,

$$\begin{aligned} & \langle j_1 k_1 m_1, j_2 k_2 m_2 | \hat{H}_{dc,2} | j'_1 k'_1 m'_1, j'_2 k'_2 m'_2 \rangle \\ &= \langle j_2 k_2 m_2 | \hat{H}_{dc,2} | j'_2 k'_2 m'_2 \rangle \delta_{j_1, j'_1} \delta_{k_1, k'_1} \delta_{m_1, m'_1}. \end{aligned} \quad (B15)$$

Combining Eqs. (B14) and (B15),

$$\begin{aligned} & \langle j_1 k_1 m_1, j_2 k_2 m_2 | \hat{H}_{dc} | j'_1 k'_1 m'_1, j'_2 k'_2 m'_2 \rangle \\ &= \langle j_1 k_1 m_1 | \hat{H}_{dc,1} | j'_1 k'_1 m'_1 \rangle \delta_{j_2, j'_2} \delta_{k_2, k'_2} \delta_{m_2, m'_2} \\ &+ \langle j_2 k_2 m_2 | \hat{H}_{dc,2} | j'_2 k'_2 m'_2 \rangle \delta_{j_1, j'_1} \delta_{k_1, k'_1} \delta_{m_1, m'_1}. \end{aligned} \quad (B16)$$

We can apply the same logic to the rotational part, $\hat{H}_{rot}$:

$$\hat{H}_{rot} = \hat{H}_{rot,1} + \hat{H}_{rot,2}, \quad (B17)$$

where $\hat{H}_{rot,i} = A_i \hat{J}_{i,a_i}^2 + B_i \hat{J}_{i,b_i}^2 + C_i \hat{J}_{i,c_i}^2$. The nonzero matrix elements will be

$$\begin{aligned} & \langle j_1 k_1 m_1, j_2 k_2 m_2 | \hat{H}_{rot} | j'_1 k'_1 m'_1, j'_2 k'_2 m'_2 \rangle \\ &= \langle j_1 k_1 m_1 | \hat{H}_{rot,1} | j'_1 k'_1 m'_1 \rangle \delta_{j_2, j'_2} \delta_{k_2, k'_2} \delta_{m_2, m'_2} \\ &+ \langle j_2 k_2 m_2 | \hat{H}_{rot,2} | j'_2 k'_2 m'_2 \rangle \delta_{j_1, j'_1} \delta_{k_1, k'_1} \delta_{m_1, m'_1}. \end{aligned} \quad (B18)$$

APPENDIX C: DERIVATION OF THE DIPOLE-DIPOLE INTERACTION TERM

The dipole-dipole interaction is described by the Hamiltonian

$$\hat{H}_{dd} = - \frac{(\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2) - 3(\hat{\mathbf{d}}_1 \cdot \mathbf{e}_r)(\hat{\mathbf{d}}_2 \cdot \mathbf{e}_r)}{r^3}, \quad (C1)$$

where $\mathbf{e}_r = \mathbf{e}_Z$ in our chosen frame. For simplicity in evaluating the matrix element of $\hat{H}_{dd}$, we set $1/(4\pi\epsilon_0) = 1$, where ϵ_0 is the vacuum permittivity. We rewrite the dipole-dipole interaction in terms of spherical tensors, which allows $\hat{H}_{dd}$ to be written as

$$\hat{H}_{dd} = - \frac{\sqrt{6}}{r^3} \sum_{p=-2}^2 (-1)^p C_{-p}^{(2)}(\Omega_r) [\hat{\mathbf{d}}_1 \otimes \hat{\mathbf{d}}_2]_p^{(2)}. \quad (C2)$$

For molecules separated by a distance r with the electric field aligned along the laboratory Z axis, the laboratory-frame orientation is $\Omega_r = (\theta_r, \phi_r) = (0, 0)$, and only the $p = 0$ term contribution remains. Using the property of spherical harmonics $C_m^{(l)}(0, 0) = \delta_{m,0}$, we find that $C_{-p}^{(2)}(0, 0) = 1$ only for $p = 0$, and zero otherwise. Thus, the sum collapses to a single term:

$$\begin{aligned} \hat{H}_{dd} &= - \frac{\sqrt{6}}{r^3} (-1)^0 C_0^{(2)}(0, 0) [\hat{\mathbf{d}}_1 \otimes \hat{\mathbf{d}}_2]_0^{(2)} \\ &= - \frac{\sqrt{6}}{r^3} \sum_{p'=-1}^1 \langle 1 p', 1 p - p' | 2 p \rangle d_{1,p'} d_{2,p-p'}. \end{aligned} \quad (C3)$$

According to Eq. (C3), three distinct contributions arise:

(1) Term with $p' = 0$:

$$\langle 1 0, 1 0 | 2 0 \rangle = \sqrt{\frac{2}{3}} \Rightarrow \sqrt{\frac{2}{3}} d_{1,0} d_{2,0}. \quad (C4)$$

(2) Term with $p' = +1$:

$$\langle 1 1, 1 - 1 | 2 0 \rangle = \sqrt{\frac{1}{6}} \Rightarrow \sqrt{\frac{1}{6}} d_{1,1} d_{2,-1}. \quad (C5)$$

(3) Term with $p' = -1$:

$$\langle 1 - 1, 1 1 | 2 0 \rangle = \sqrt{\frac{1}{6}} \Rightarrow \sqrt{\frac{1}{6}} d_{1,-1} d_{2,1}. \quad (C6)$$

Inserting these values into Eq. (C3) gives

$$\hat{H}_{dd} = - \frac{\sqrt{6}}{r^3} \left(\sqrt{\frac{2}{3}} d_{1,0} d_{2,0} + \sqrt{\frac{1}{6}} d_{1,1} d_{2,-1} + \sqrt{\frac{1}{6}} d_{1,-1} d_{2,1} \right). \quad (C7)$$

Hence, the expression reduces to

$$\hat{H}_{dd} = - \frac{1}{r^3} (2 d_{1,0} d_{2,0} + d_{1,-1} d_{2,1} + d_{1,1} d_{2,-1}). \quad (C8)$$

In Eq. (C8), the dipole components are expressed in the laboratory frame and must be transformed to the molecule-fixed frame. Accordingly,

$$d_{i,q} = \sum_{r_i=-1}^1 D_{q,r_i}^{1*}(\Omega_i) d_{i,r_i}^{(\text{mol})}, \quad (C9)$$

where $q = -1, 0, 1$ and $r = -1, 0, 1$ label the laboratory-frame and molecule-fixed spherical components, respectively; $d_{i,r_i}^{(\text{mol})}$ are the known dipole components of molecule i , and $D_{q,r_i}^{1*}(\Omega_i)$ is the Wigner D -matrix associated with the orientation Ω_i .

The first term in the equation is given by $2 d_{1,0} d_{2,0}$:

$$\begin{aligned} d_{1,0} &= \sum_{r_1=-1}^1 D_{0,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})}, \\ d_{2,0} &= \sum_{r_2=-1}^1 D_{0,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}, \\ \Rightarrow 2 d_{1,0} d_{2,0} &= 2 \sum_{r_1=-1}^1 \sum_{r_2=-1}^1 D_{0,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{0,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}. \end{aligned} \quad (C10)$$

The second term $d_{1,-1} d_{2,1}$:

$$d_{1,-1} d_{2,1} = \sum_{r_1=-1}^1 \sum_{r_2=-1}^1 D_{-1,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{1,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}. \quad (C11)$$

Similarly, the third term $d_{1,1} d_{2,-1}$:

$$d_{1,1} d_{2,-1} = \sum_{r_1=-1}^1 \sum_{r_2=-1}^1 D_{1,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{-1,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}. \quad (C12)$$

Collecting all terms, one obtains

$$\begin{aligned} \hat{H}_{dd} &= - \frac{1}{r^3} \sum_{r_1=-1}^1 \sum_{r_2=-1}^1 [2 D_{0,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{0,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})} \\ &+ D_{-1,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{1,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})} \\ &+ D_{1,r_1}^{1*}(\Omega_1) d_{1,r_1}^{(\text{mol})} D_{-1,r_2}^{1*}(\Omega_2) d_{2,r_2}^{(\text{mol})}]. \end{aligned} \quad (C13)$$

By substituting Eqs. (A14) and (A15) into Eq. (C13), we obtain a general expression for the matrix elements

of the dipole-dipole interaction,

$$\begin{aligned}
 \langle j_1 k_1 m_1, j_2 k_2 m_2 | \hat{H}_{dd} | j'_1 k'_1 m'_1, j'_2 k'_2 m'_2 \rangle = & -\frac{1}{r^3} [2 \langle j_1 k_1 m_1 | D_{0, k_1-k'_1}^{1*} | j'_1 k'_1 m'_1 \rangle \langle j_2 k_2 m_2 | D_{0, k_2-k'_2}^{1*} | j'_2 k'_2 m'_2 \rangle \delta_{0, m_1-m'_1} \delta_{0, m_2-m'_2}] \\
 & -\frac{1}{r^3} [\langle j_1 k_1 m_1 | D_{-1, k_1-k'_1}^{1*} | j'_1 k'_1 m'_1 \rangle \langle j_2 k_2 m_2 | D_{1, k_2-k'_2}^{1*} | j'_2 k'_2 m'_2 \rangle \delta_{-1, m_1-m'_1} \delta_{1, m_2-m'_2}] \\
 & -\frac{1}{r^3} [\langle j_1 k_1 m_1 | D_{1, k_1-k'_1}^{1*} | j'_1 k'_1 m'_1 \rangle \langle j_2 k_2 m_2 | D_{-1, k_2-k'_2}^{1*} | j'_2 k'_2 m'_2 \rangle \delta_{1, m_1-m'_1} \delta_{-1, m_2-m'_2}] \\
 & \times d_{1, k_1-k'_1} d_{2, k_2-k'_2}.
 \end{aligned} \tag{C14}$$

APPENDIX D: PROJECTION ONTO THE EFFECTIVE SPIN-1/2 MODEL

In this Appendix, we derive the matrix elements of the dipole-dipole Hamiltonian in the dressed pseudospin basis, map the total Hamiltonian onto an effective spin-1/2 XXZ model, and detail the gauge transformation used to absorb the Dzyaloshinskii-Moriya interaction into a renormalized transverse coupling.

1. Matrix elements of $\hat{H}_{dd}$

We now construct the matrix elements of the dipole-dipole interaction in the dressed-state basis

$$|\uparrow\rangle = \sum_{J,K} c_{J,K}^{(\uparrow)}(x) |J, K, M=0\rangle, \tag{D1}$$

$$|\downarrow\rangle = \sum_{J,K} c_{J,K}^{(\downarrow)}(x) |J, K, M=1\rangle. \tag{D2}$$

Basis: $\{|\downarrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\uparrow\uparrow\rangle\}$, using the selection rules $\Delta m_1 = m_1 - m'_1$, $\Delta m_2 = m_2 - m'_2$. There are three rules that give nonzero elements according to Eq. (C14):

(1) $\Delta m_1 = 0$, $\Delta m_2 = 0$, implying $m_1 = m'_1$ and $m_2 = m'_2$.

(2) $m_1 - m'_1 = -1$, $m_2 - m'_2 = +1$.

(3) $m_1 - m'_1 = +1$, $m_2 - m'_2 = -1$.

(1) $\langle\downarrow\downarrow|H_{dd}|\downarrow\downarrow\rangle$

$$m_1 = 1, m'_1 = 1, m_2 = 1, m'_2 = 1,$$

$$\Delta m_1 = 0, \Delta m_2 = 0.$$

From Eq. (C14), we define

$$\begin{aligned}
 C_1 = & - \sum_{\substack{J_1, K_1 \\ J'_1, K'_1}} \sum_{\substack{J_2, K_2 \\ J'_2, K'_2}} c_{J_1, K_1}^{(\downarrow)*} c_{J'_1, K'_1}^{(\downarrow)} c_{J_2, K_2}^{(\downarrow)*} c_{J'_2, K'_2}^{(\downarrow)} \\
 & \times [2 \langle j_1 k_1 1 | D_{0, k_1-k'_1}^1 | j'_1 k'_1 1 \rangle \langle j_2 k_2 1 | D_{0, k_2-k'_2}^1 | j'_2 k'_2 1 \rangle] \\
 & \times d_{1, k_1-k'_1} d_{2, k_2-k'_2}.
 \end{aligned}$$

(2) $\langle\downarrow\downarrow|H_{dd}|\downarrow\uparrow\rangle$

$$m_1 = 1, m'_1 = 1, m_2 = 1, m'_2 = 0,$$

$$\Delta m_1 = 0, \Delta m_2 = 1 - 0 = 1.$$

Zero.

(3) $\langle\downarrow\downarrow|H_{dd}|\uparrow\downarrow\rangle$

$$m_1 = 1, m'_1 = 0, m_2 = 1, m'_2 = 1,$$

$$\Delta m_1 = 1, \Delta m_2 = 0.$$

Zero.

(4) $\langle\downarrow\downarrow|H_{dd}|\uparrow\uparrow\rangle$

$$m_1 = 1, m'_1 = 0, m_2 = 1, m'_2 = 0,$$

$$\Delta m_1 = 1, \Delta m_2 = 1.$$

Zero.

(5) $\langle\downarrow\uparrow|H_{dd}|\downarrow\downarrow\rangle$

$$m_1 = 1, m'_1 = 1, m_2 = 0, m'_2 = 1,$$

$$\Delta m_1 = 0, \Delta m_2 = -1.$$

Zero.

(6) $\langle\downarrow\uparrow|H_{dd}|\downarrow\uparrow\rangle$

$$m_1 = 1, m'_1 = 1, m_2 = 0, m'_2 = 0,$$

$$\Delta m_1 = 0, \Delta m_2 = 0.$$

Nonzero, defining from Eq. (C14):

$$\begin{aligned}
 C_2 = & - \sum_{\substack{J_1, K_1 \\ J'_1, K'_1}} \sum_{\substack{J_2, K_2 \\ J'_2, K'_2}} c_{J_1, K_1}^{(\downarrow)*} c_{J'_1, K'_1}^{(\downarrow)} c_{J_2, K_2}^{(\uparrow)*} c_{J'_2, K'_2}^{(\uparrow)} \\
 & \times [2 \langle j_1 k_1 1 | D_{0, k_1-k'_1}^1 | j'_1 k'_1 1 \rangle \langle j_2 k_2 0 | D_{0, k_2-k'_2}^1 | j'_2 k'_2 0 \rangle] \\
 & \times d_{1, k_1-k'_1} d_{2, k_2-k'_2}.
 \end{aligned}$$

(7) $\langle\downarrow\uparrow|H_{dd}|\uparrow\downarrow\rangle$

$$m_1 = 1, m'_1 = 0, m_2 = 0, m'_2 = 1,$$

$$\Delta m_1 = 1, \Delta m_2 = -1.$$

From Eq. (C14), nonzero (exchange-type term) is given as

$$\begin{aligned}
 C_{d_1} = & - \sum_{\substack{J_1, K_1 \\ J'_1, K'_1}} \sum_{\substack{J_2, K_2 \\ J'_2, K'_2}} c_{J_1, K_1}^{(\downarrow)*} c_{J'_1, K'_1}^{(\uparrow)} c_{J_2, K_2}^{(\uparrow)*} c_{J'_2, K'_2}^{(\downarrow)} \\
 & \times \langle j_1 k_1 1 | D_{+1, k_1-k'_1}^1 | j'_1 k'_1 0 \rangle \langle j_2 k_2 0 | D_{-1, k_2-k'_2}^1 | j'_2 k'_2 1 \rangle \\
 & \times d_{1, k_1-k'_1} d_{2, k_2-k'_2}.
 \end{aligned}$$

(8) $\langle\uparrow\downarrow|H_{dd}|\uparrow\uparrow\rangle$

$$m_1 = 0, m'_1 = 1, m_2 = 1, m'_2 = 1,$$

$$\Delta m_1 = -1, \Delta m_2 = 0.$$

Zero.

(9) $\langle\uparrow\downarrow|H_{dd}|\downarrow\downarrow\rangle$

$$m_1 = 0, m'_1 = 1, m_2 = 1, m'_2 = 1,$$

$$\Delta m_1 = -1, \Delta m_2 = 0.$$

Zero.

$$(10) \langle \uparrow \downarrow | H_{dd} | \downarrow \uparrow \rangle$$

$$m_1 = 0, m'_1 = 1, m_2 = 1, m'_2 = 0,$$

$$\Delta m_1 = -1, \Delta m_2 = 1.$$

Nonzero, giving the Hermitian-conjugate exchange term C_{d_2} from Eq. (C14):

$$C_{d_2} = - \sum_{J_1, K_1} \sum_{J'_1, K'_1} c_{J_1, K_1}^{(\uparrow)*} c_{J'_1, K'_1}^{(\downarrow)} c_{J_2, K_2}^{(\downarrow)*} c_{J'_2, K'_2}^{(\uparrow)}$$

$$\times \langle j_1 k_1 0 | D_{-1, k_1 - k'_1}^1 | j'_1 k'_1 1 \rangle \langle j_2 k_2 1 | D_{+1, k_2 - k'_2}^1 | j'_2 k'_2 0 \rangle$$

$$\times d_{1, k_1 - k'_1} d_{2, k_2 - k'_2}.$$

$$(11) \langle \uparrow \downarrow | H_{dd} | \uparrow \downarrow \rangle$$

$$m_1 = 0, m'_1 = 0, m_2 = 1, m'_2 = 1,$$

$$\Delta m_1 = 0, \Delta m_2 = 0.$$

We define

$$C_3 = - \sum_{J_1, K_1} \sum_{J'_1, K'_1} c_{J_1, K_1}^{(\uparrow)*} c_{J'_1, K'_1}^{(\uparrow)} c_{J_2, K_2}^{(\downarrow)*} c_{J'_2, K'_2}^{(\downarrow)}$$

$$\times [2 \langle j_1 k_1 0 | D_{0, k_1 - k'_1}^1 | j'_1 k'_1 0 \rangle \langle j_2 k_2 1 | D_{0, k_2 - k'_2}^1 | j'_2 k'_2 1 \rangle]$$

$$\times d_{1, k_1 - k'_1} d_{2, k_2 - k'_2}.$$

$$(12) \langle \uparrow \uparrow | H_{dd} | \uparrow \downarrow \rangle$$

$$m_1 = 0, m'_1 = 0, m_2 = 1, m'_2 = 0,$$

$$\Delta m_1 = 0, \Delta m_2 = 1.$$

Zero.

$$(13) \langle \uparrow \uparrow | H_{dd} | \downarrow \downarrow \rangle$$

$$m_1 = 0, m'_1 = 1, m_2 = 1, m'_2 = 1,$$

$$\Delta m_1 = -1, \Delta m_2 = 0.$$

Zero.

$$(14) \langle \uparrow \uparrow | H_{dd} | \downarrow \uparrow \rangle$$

$$m_1 = 0, m'_1 = 1, m_2 = 0, m'_2 = 0,$$

$$\Delta m_1 = -1, \Delta m_2 = 0.$$

Zero.

$$(15) \langle \uparrow \uparrow | H_{dd} | \uparrow \downarrow \rangle$$

$$m_1 = 0, m'_1 = 0, m_2 = 0, m'_2 = 1,$$

$$\Delta m_1 = 0, \Delta m_2 = -1.$$

Zero.

$$(16) \langle \uparrow \uparrow | H_{dd} | \uparrow \uparrow \rangle$$

$$m_1 = 0, m'_1 = 0, m_2 = 0, m'_2 = 0,$$

$$\Delta m_1 = 0, \Delta m_2 = 0.$$

From Eq. (C14), we define

$$C_4 = - \sum_{J_1, K_1} \sum_{J'_1, K'_1} c_{J_1, K_1}^{(\uparrow)*} c_{J'_1, K'_1}^{(\uparrow)} c_{J_2, K_2}^{(\uparrow)*} c_{J'_2, K'_2}^{(\uparrow)}$$

$$\times [2 \langle j_1 k_1 0 | D_{0, k_1 - k'_1}^1 | j'_1 k'_1 0 \rangle \langle j_2 k_2 0 | D_{0, k_2 - k'_2}^1 | j'_2 k'_2 0 \rangle]$$

$$\times d_{1, k_1 - k'_1} d_{2, k_2 - k'_2}.$$

Collecting all nonzero contributions, the dipole-dipole Hamiltonian in the dressed basis $\{|\downarrow \downarrow\rangle, |\downarrow \uparrow\rangle, |\uparrow \downarrow\rangle, |\uparrow \uparrow\rangle\}$ takes the form

$$H_{dd} = -\frac{1}{r^3} \begin{pmatrix} C_1 & 0 & 0 & 0 \\ 0 & C_2 & C_{d_1} & 0 \\ 0 & C_{d_2} & C_3 & 0 \\ 0 & 0 & 0 & C_4 \end{pmatrix}.$$

Here, the off-diagonal terms C_{d_1} and C_{d_2} are Hermitian conjugates:

$$C_{d_2} = C_{d_1}^*.$$

Because the molecules under consideration are chiral, the dipole components $d_{\pm 1}$ carry complex phase $\mp \frac{d_b \pm i d_c}{\sqrt{2}}$, and the resulting matrix elements are therefore complex. We decompose these matrix elements into their real and imaginary parts as

$$C_{d_1} = J_{xy} + i\mathcal{D},$$

$$C_{d_2} = J_{xy} - i\mathcal{D}.$$

To map this system to a spin-1/2 model, we define Pauli operators acting on the dressed states:

(1) $|\uparrow\rangle$ (state $M = 0$) corresponds to spin “up” (+z).

(2) $|\downarrow\rangle$ (state $M = 1$) corresponds to spin “down” (−z).

The ladder operators are defined as

$$\hat{\sigma}^+ = |\uparrow\rangle\langle\downarrow| \quad (\text{flips } \downarrow \rightarrow \uparrow),$$

$$\hat{\sigma}^- = |\downarrow\rangle\langle\uparrow| \quad (\text{flips } \uparrow \rightarrow \downarrow),$$

with the Pauli matrices:

$$\hat{\sigma}^x = \hat{\sigma}^+ + \hat{\sigma}^-, \quad (D3)$$

$$\hat{\sigma}^y = -i(\hat{\sigma}^+ - \hat{\sigma}^-) \Rightarrow \hat{\sigma}^{\pm} = \frac{1}{2}(\hat{\sigma}^x \pm i\hat{\sigma}^y), \quad (D4)$$

$$\hat{\sigma}^z = |\uparrow\rangle\langle\downarrow| - |\downarrow\rangle\langle\uparrow|. \quad (D5)$$

The off-diagonal matrix elements connect $|\downarrow \uparrow\rangle$ and $|\uparrow \downarrow\rangle$. We analyze the transition corresponding to C_{d_1} .

(1) Initial state (ket): $|\downarrow \uparrow\rangle$ (Mol 1 is $\uparrow$, Mol 2 is $\downarrow$).

(2) Final state (bra): $\langle \uparrow \uparrow |$ (Mol 1 is $\downarrow$, Mol 2 is $\uparrow$).

(3) Action: Mol 1 flips $\uparrow \rightarrow \downarrow$ ($\hat{\sigma}_1^-$) and Mol 2 flips $\downarrow \rightarrow \uparrow$ ($\hat{\sigma}_2^+$).

This term is represented by the operator

$$H_{\text{off}} = -C_{d_1} \hat{\sigma}_1^- \hat{\sigma}_2^+ - C_{d_2} \hat{\sigma}_1^+ \hat{\sigma}_2^-. \quad (D6)$$

Substitute the Pauli-matrix forms

$$\hat{\sigma}_1^- \hat{\sigma}_2^+ = \left[\frac{1}{2}(\hat{\sigma}_1^x - i\hat{\sigma}_1^y) \right] \left[\frac{1}{2}(\hat{\sigma}_2^x + i\hat{\sigma}_2^y) \right]$$

$$= \frac{1}{4} [\hat{\sigma}_1^x \hat{\sigma}_2^x + \hat{\sigma}_1^y \hat{\sigma}_2^y + i(\hat{\sigma}_1^x \hat{\sigma}_2^y - \hat{\sigma}_1^y \hat{\sigma}_2^x)],$$

and similarly for the conjugate term

$$\begin{aligned}\hat{\sigma}_1^+ \hat{\sigma}_2^- &= \left[\frac{1}{2} (\hat{\sigma}_1^x + i \hat{\sigma}_1^y) \right] \left[\frac{1}{2} (\hat{\sigma}_2^x - i \hat{\sigma}_2^y) \right] \\ &= \frac{1}{4} [\hat{\sigma}_1^x \hat{\sigma}_2^x + \hat{\sigma}_1^y \hat{\sigma}_2^y - i (\hat{\sigma}_1^x \hat{\sigma}_2^y - \hat{\sigma}_1^y \hat{\sigma}_2^x)].\end{aligned}$$

Substitute $C_{d1} = J_{xy} + i\mathcal{D}$ and $C_{d2} = J_{xy} - i\mathcal{D}$ into Eq. (D6):

$$\begin{aligned}H_{\text{off}} &= -(J_{xy} + i\mathcal{D}) \frac{1}{4} [A + iB] - (J_{xy} - i\mathcal{D}) \frac{1}{4} [A - iB] \\ &= -\frac{J_{xy} + i\mathcal{D}}{4} [A + iB] - \frac{J_{xy} - i\mathcal{D}}{4} [A - iB],\end{aligned}$$

where, for convenience,

$$\begin{aligned}A &= \hat{\sigma}_1^x \hat{\sigma}_2^x + \hat{\sigma}_1^y \hat{\sigma}_2^y, \\ B &= \hat{\sigma}_1^x \hat{\sigma}_2^y - \hat{\sigma}_1^y \hat{\sigma}_2^x.\end{aligned}$$

Collecting the real contributions (J_{xy}), we obtain

$$\begin{aligned}-\frac{J_{xy}}{4} [(A + iB) + (A - iB)] &= -\frac{J_{xy}}{4} (2A) = -\frac{J_{xy}}{2} A \\ &= -\frac{J_{xy}}{2} (\hat{\sigma}_1^x \hat{\sigma}_2^x + \hat{\sigma}_1^y \hat{\sigma}_2^y),\end{aligned}$$

which is the XY exchange interaction.

The imaginary terms contribution ($\mathcal{D}$) is given by

$$\begin{aligned}-\frac{i\mathcal{D}}{4} [(A + iB) - (A - iB)] &= -\frac{i\mathcal{D}}{4} (2iB) = \frac{\mathcal{D}}{2} B \\ &= \frac{\mathcal{D}}{2} (\hat{\sigma}_1^x \hat{\sigma}_2^y - \hat{\sigma}_1^y \hat{\sigma}_2^x),\end{aligned}$$

which is the DMI.

2. Mapping of Hamiltonian

The total Hamiltonian for the two-molecule system is

$$\hat{H}_{\text{total}} = (\hat{H}_{\text{rot}}^{(1)} + \hat{H}_{\text{dc}}^{(1)}) + (\hat{H}_{\text{rot}}^{(2)} + \hat{H}_{\text{dc}}^{(2)}) + \hat{H}_{\text{dd}}. \quad (\text{D7})$$

Since the single-molecule Hamiltonian ($\hat{H}_{\text{rot}} + \hat{H}_{\text{dc}}$) has already been solved, its eigenvalues are (1) spin up: $|\uparrow\rangle$ (state $M = 0$) has energy $E_{\uparrow}$ and (2) spin down: $|\downarrow\rangle$ (state $M = 1$) has energy $E_{\downarrow}$.

The full diagonal energies of the product states are obtained by adding the single-molecule energies to the diagonal dipole terms (C_1, C_2, C_3, C_4):

(1) State $|\downarrow\downarrow\rangle$:

$$E_{\text{tot}}(\downarrow\downarrow) = E_{\downarrow} + E_{\downarrow} - C_1 = 2E_{\downarrow} - C_1. \quad (\text{D8})$$

(2) State $|\downarrow\uparrow\rangle$:

$$E_{\text{tot}}(\downarrow\uparrow) = E_{\downarrow} + E_{\uparrow} - C_2. \quad (\text{D9})$$

(3) State $|\uparrow\downarrow\rangle$:

$$E_{\text{tot}}(\uparrow\downarrow) = E_{\uparrow} + E_{\downarrow} - C_3. \quad (\text{D10})$$

(4) State $|\uparrow\uparrow\rangle$:

$$E_{\text{tot}}(\uparrow\uparrow) = E_{\uparrow} + E_{\uparrow} - C_4 = 2E_{\uparrow} - C_4, \quad (\text{D11})$$

$$\hat{H}_{\text{diag}} = J_z \hat{\sigma}_1^z \hat{\sigma}_2^z + \mathfrak{h}_1 \hat{\sigma}_1^z + \mathfrak{h}_2 \hat{\sigma}_2^z + E_0. \quad (\text{D12})$$

(a) Eq. (1) ($|\downarrow\downarrow\rangle$, spins $-1, -1$):

$$J_z(-1)(-1) + \mathfrak{h}_1(-1) + \mathfrak{h}_2(-1) + E_0 = 2E_{\downarrow} - C_1, \quad (\text{D13})$$

$$J_z - \mathfrak{h}_1 - \mathfrak{h}_2 + E_0 = 2E_{\downarrow} - C_1. \quad (\text{D14})$$

(b) Eq. (2) ($|\downarrow\uparrow\rangle$, spins $-1, +1$):

$$J_z(-1)(+1) + \mathfrak{h}_1(-1) + \mathfrak{h}_2(+1) + E_0 = E_{\downarrow} + E_{\uparrow} - C_2 \quad (\text{D15})$$

$$-J_z - \mathfrak{h}_1 + \mathfrak{h}_2 + E_0 = E_{\downarrow} + E_{\uparrow} - C_2. \quad (\text{D16})$$

(c) Eq. (3) ($|\uparrow\downarrow\rangle$, spins $+1, -1$):

$$-J_z(+1)(-1) + \mathfrak{h}_1(+1) + \mathfrak{h}_2(-1) + E_0 = E_{\uparrow} + E_{\downarrow} - C_3 \quad (\text{D17})$$

$$-J_z + \mathfrak{h}_1 - \mathfrak{h}_2 + E_0 = E_{\uparrow} + E_{\downarrow} - C_3. \quad (\text{D18})$$

(d) Eq. (4) ($|\uparrow\uparrow\rangle$, spins $+1, +1$):

$$J_z(+1)(+1) + \mathfrak{h}_1(+1) + \mathfrak{h}_2(+1) + E_0 = 2E_{\uparrow} - C_4, \quad (\text{D19})$$

$$J_z + \mathfrak{h}_1 + \mathfrak{h}_2 + E_0 = 2E_{\uparrow} - C_4. \quad (\text{D20})$$

(i) Solving for J_z .

We isolate J_z by computing

$$[\text{Eq. (D13)} + \text{Eq. (D19)}] - [\text{Eq. (D15)} + \text{Eq. (D17)}].$$

Left-hand side:

$$\begin{aligned}&[(J_z - 2\mathfrak{h} + E_0) + (J_z + 2\mathfrak{h} + E_0)] \\ &\quad - [(-J_z + E_0) + (-J_z + E_0)] \\ &= [2J_z + 2E_0] - [-2J_z + 2E_0] = 4J_z.\end{aligned}$$

Right-hand side:

$$\begin{aligned}&[(2E_{\downarrow} - C_1) + (2E_{\uparrow} - C_4)] \\ &\quad - [(E_{\downarrow} + E_{\uparrow} - C_2) + (E_{\uparrow} + E_{\downarrow} - C_3)] \\ &= (2E_{\downarrow} + 2E_{\uparrow} - C_1 - C_4) - (2E_{\downarrow} + 2E_{\uparrow} - C_2 - C_3) \\ &= -C_1 - C_4 + C_2 + C_3.\end{aligned}$$

Thus,

$$J_z = \frac{1}{4}(C_2 + C_3 - C_1 - C_4). \quad (\text{D21})$$

(ii) Solving for fields.

Compute Eqs. (D19)–(D13):

$$(J_z + \mathfrak{h}_1 + \mathfrak{h}_2 + E_0) - (J_z - \mathfrak{h}_1 - \mathfrak{h}_2 + E_0) = 2(\mathfrak{h}_1 + \mathfrak{h}_2), \quad (\text{D22})$$

$$2(\mathfrak{h}_1 + \mathfrak{h}_2) = 2(E_{\uparrow} - E_{\downarrow}) + (C_1 - C_4), \quad (\text{D23})$$

$$\mathfrak{h}_1 + \mathfrak{h}_2 = (E_{\uparrow} - E_{\downarrow}) + \frac{1}{2}(C_1 - C_4). \quad (\text{D24})$$

Compute Eqs. (D17)–(D15):

$$(-J_z + \mathfrak{h}_1 - \mathfrak{h}_2 + E_0) - (-J_z - \mathfrak{h}_1 + \mathfrak{h}_2 + E_0) = 2\mathfrak{h}_1 - 2\mathfrak{h}_2, \quad (\text{D25})$$

$$2(\mathfrak{h}_1 - \mathfrak{h}_2) = (E_{\uparrow} + E_{\downarrow} - C_3) - (E_{\downarrow} + E_{\uparrow} - C_2), \quad (\text{D26})$$

$$2(\mathfrak{h}_1 - \mathfrak{h}_2) = C_2 - C_3, \quad (\text{D27})$$

$$\mathfrak{h}_1 - \mathfrak{h}_2 = \frac{1}{2}(C_2 - C_3). \quad (\text{D28})$$

By adding and subtracting the results of Eqs. (D24) and (D28), we obtain

$$\hbar_1 = \frac{1}{2}(\hbar_1 + \hbar_2) + \frac{1}{2}(\hbar_1 - \hbar_2) \quad (\text{D29})$$

$$= \frac{1}{2}[(E_\uparrow - E_\downarrow) + \frac{1}{2}(C_1 - C_4)] + \frac{1}{2}[\frac{1}{2}(C_2 - C_3)] \quad (\text{D30})$$

$$= \frac{1}{2}[(E_\uparrow - E_\downarrow) + \frac{1}{2}(C_1 - C_4)] + \frac{1}{4}[(C_2 - C_3)], \quad (\text{D31})$$

and similarly

$$\hbar_2 = \frac{1}{2}(\hbar_1 + \hbar_2) - \frac{1}{2}(\hbar_1 - \hbar_2) \quad (\text{D32})$$

$$= \frac{1}{2}[(E_\uparrow - E_\downarrow) + \frac{1}{2}(C_1 - C_4)] - \frac{1}{2}[\frac{1}{2}(C_2 - C_3)] \quad (\text{D33})$$

$$= \frac{1}{2}[(E_\uparrow - E_\downarrow) + \frac{1}{2}(C_1 - C_4)] - \frac{1}{4}[(C_2 - C_3)]. \quad (\text{D34})$$

As from Fig. 4, it is confirmed that $C_2 = C_3$, and then $\hbar_1 = \hbar_2 = \hbar = \frac{1}{2}(E_\uparrow - E_\downarrow) + \frac{1}{4r^3}(C_1 - C_4)$. Collecting all terms gives the exact spin-1/2 Hamiltonian of the system

$$\begin{aligned} \hat{H}_{\text{spin}} = & \underbrace{J_{xy}(\hat{\sigma}_1^x \hat{\sigma}_2^x + \hat{\sigma}_1^y \hat{\sigma}_2^y)}_{\text{symmetric exchange}} - \underbrace{D(\hat{\sigma}_1^x \hat{\sigma}_2^y - \hat{\sigma}_1^y \hat{\sigma}_2^x)}_{\text{chiral DMI}} \\ & + \underbrace{J_z \hat{\sigma}_1^z \hat{\sigma}_2^z}_{\text{Ising interaction}} + \hbar(\hat{\sigma}_1^z + \hat{\sigma}_2^z), \end{aligned} \quad (\text{D35})$$

where

$$J_{xy} = -\frac{1}{2r^3} \Re(C_{d_1}), \quad D = \frac{1}{2r^3} \Im(C_{d_1}),$$

$$J_z = \frac{1}{4r^3}(C_2 + C_3 - C_1 - C_4), \quad (\text{D36})$$

$$\hbar = \frac{1}{2}(E_\uparrow - E_\downarrow) + \frac{1}{4r^3}(C_1 - C_4).$$

3. Guage transformation

From Eq. (D6), the real and imaginary parts of the off-diagonal Hamiltonian H_{off} described as

$$H_{\text{off}} = J_{xy}[2(\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- + \hat{\sigma}_i^- \hat{\sigma}_{i+1}^+)] - D[2i(\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- - \hat{\sigma}_i^- \hat{\sigma}_{i+1}^+)]. \quad (\text{D37})$$

Collecting terms, this expression can be rewritten as

$$H_{\text{off}} = 2(J_{xy} - iD)\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- + 2(J_{xy} + iD)\hat{\sigma}_i^- \hat{\sigma}_{i+1}^+. \quad (\text{D38})$$

It is convenient to express the complex coefficients $(J_{xy} \pm iD)$ in polar form as $Re^{\pm i\theta}$. Defining the effective transverse coupling $\tilde{J}_{xy} = \sqrt{J_{xy}^2 + D^2}$ and the phase $\theta = \tan^{-1}(D/J_{xy})$, we may write

$$J_{xy} \mp iD = \tilde{J}_{xy} e^{\mp i\theta}. \quad (\text{D39})$$

Substituting Eq. (D39) into Eq. (D37), the Hamiltonian yields the compact form

$$H_{\text{off}} = 2\tilde{J}_{xy}(e^{-i\theta}\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- + e^{+i\theta}\hat{\sigma}_i^- \hat{\sigma}_{i+1}^+). \quad (\text{D40})$$

We perform a local unitary transformation about the z axis

$$U = \prod_{i=1}^n e^{-i\frac{\phi_i}{2}\sigma_i^z}. \quad (\text{D41})$$

This transforms the ladder operators as

$$U^\dagger \hat{\sigma}_i^+ U = e^{i\phi_i} \hat{\sigma}_i^+, \quad (\text{D42})$$

$$U^\dagger \hat{\sigma}_i^- U = e^{-i\phi_i} \hat{\sigma}_i^-. \quad (\text{D43})$$

The term $\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^-$ becomes

$$\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- = (e^{i\phi_i} \hat{\sigma}_i^+)(e^{-i\phi_{i+1}} \hat{\sigma}_{i+1}^-) = e^{-i(\phi_{i+1} - \phi_i)} \hat{\sigma}_i^+ \hat{\sigma}_{i+1}^-. \quad (\text{D44})$$

To eliminate the complex phase appearing in $(J_{xy} - iD) = \tilde{J}_{xy}e^{-i\theta}$ and $(J_{xy} + iD) = \tilde{J}_{xy}e^{+i\theta}$, we require the rotation phases to satisfy

$$e^{-i(\phi_{i+1} - \phi_i)} = e^{i\theta} \implies \phi_{i+1} - \phi_i = -\theta. \quad (\text{D45})$$

This condition is satisfied by choosing the rotation angle to vary linearly along the chain,

$$\phi_i = -i\theta, \quad \theta = \tan^{-1}\left(\frac{D}{J_{xy}}\right). \quad (\text{D46})$$

Substituting values of $\phi_i = -i\theta$ into Eq. (D40), the transformed off-diagonal Hamiltonian becomes

$$\tilde{H}_{\text{off}} = 2\tilde{J}_{xy}(e^{-i\theta}e^{i(\phi_{i+1} - \phi_i)}\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- + e^{+i\theta}e^{-i(\phi_{i+1} - \phi_i)}\hat{\sigma}_i^- \hat{\sigma}_{i+1}^+), \quad (\text{D47})$$

and using $\phi_{i+1} - \phi_i = -\theta$, we obtain

$$e^{-i\theta}e^{i(\phi_{i+1} - \phi_i)} = e^{-i\theta}e^{i\theta} = 1, \quad (\text{D48})$$

which yields the simplified form

$$\tilde{H}_{\text{off}} = 2\tilde{J}_{xy}(\hat{\sigma}_i^+ \hat{\sigma}_{i+1}^- + \hat{\sigma}_i^- \hat{\sigma}_{i+1}^+). \quad (\text{D49})$$

Converting back to Pauli matrices,

$$\hat{H}_{\text{spin}} = \sum_{j=1}^{N-1} [\tilde{J}_{xy}(\hat{\sigma}_j^x \hat{\sigma}_{j+1}^x + \hat{\sigma}_j^y \hat{\sigma}_{j+1}^y) + J_z \hat{\sigma}_j^z \hat{\sigma}_{j+1}^z] + \hbar \sum_{j=1}^N \hat{\sigma}_j^z. \quad (\text{D50})$$

APPENDIX E: ROBUSTNESS OF THE CHIRAL LUTTINGER LIQUID PHASE AGAINST DROPLET SURFACE CHARGE

We assume a macroscopic droplet doped with PDO forming a vortex filament.

(1) Dipole moment of PDO (μ): ≈ 2.5 Debye $\approx 8.34 \times 10^{-30}$ C m.

(2) Intermolecular spacing (r): 1.7 nm (critical density for CLL phase).

(3) Droplet radius (R_{drop}): 500 nm (assuming that $N \approx 10^8$ He atoms).

(4) Droplet charge (q): $+1e \approx 1.602 \times 10^{-19}$ C (localized on the surface).

The stabilization energy of the CLL phase is governed by the interaction between nearest neighbors in the filament. Assuming a head-to-tail alignment along the vortex core:

$$V_{\text{dd}} = \frac{1}{4\pi\epsilon_0} \frac{\mu^2}{r^3}. \quad (\text{E1})$$

Substituting the values:

$$V_{\text{dd}} \approx \frac{(8.34 \times 10^{-30})^2}{4\pi(8.85 \times 10^{-12})(1.7 \times 10^{-9})^3},$$

$$V_{\text{dd}} \approx 1.35 \times 10^{-23} \text{ J}.$$

Converting to temperature units (kelvin) by dividing by the Boltzmann constant (k_B):

$$V_{dd} \approx 0.98 \text{ K} \approx 1.0 \text{ K}. \quad (\text{E2})$$

In a charged helium droplet, the charge localizes on the surface due to the electrostriction of the helium. The vortex filament is located at the center of the droplet. The electric field E generated by the surface charge at the center is

$$E_{\text{surf}} = \frac{1}{4\pi\epsilon_0} \frac{q}{R_{\text{drop}}^2}. \quad (\text{E3})$$

The interaction energy with a molecular dipole is $V_{\text{charge}} = -\vec{\mu} \cdot \vec{E}_{\text{surf}}$. The maximum perturbation magnitude is

$$|V_{\text{charge}}| \approx \mu \times \left(\frac{1}{4\pi\epsilon_0} \frac{q}{R_{\text{drop}}^2} \right),$$

$$|V_{\text{charge}}| \approx (8.34 \times 10^{-30})$$

$$\times \frac{1.602 \times 10^{-19}}{4\pi(8.85 \times 10^{-12})(500 \times 10^{-9})^2},$$

$$|V_{\text{charge}}| \approx 4.8 \times 10^{-26} \text{ J}.$$

Converting to kelvin:

$$V_{\text{charge}} \approx 0.0035 \text{ K}. \quad (\text{E4})$$

Comparing the two energy scales:

$$\frac{V_{dd}(\text{signal})}{V_{\text{charge}}(\text{noise})} = \frac{1.0 \text{ K}}{0.0035 \text{ K}} \approx 285. \quad (\text{E5})$$

The intermolecular interaction is nearly 300 times stronger than the electrostatic perturbation. Furthermore, because the source of the electric field ($R = 500 \text{ nm}$) is distant compared to the molecular spacing ($r = 1.7 \text{ nm}$), the field appears as a uniform background that may define a quantization axis without disrupting the internal Luttinger liquid dynamics.

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- Correction:* An erroneous reference was included in the published version and has been removed.