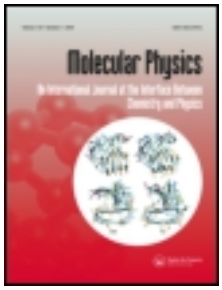

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INVITED ARTICLE

Kitaev honeycomb and other exotic spin models with polar molecules

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We show that ultracold polar molecules pinned in an optical lattice can be used to access a variety of exotic spin models, including the Kitaev honeycomb model. Treating each molecule as a rigid rotor, we use DC electric and microwave fields to define superpositions of rotational levels as effective spin degrees of freedom, while dipole–dipole interactions give rise to interactions between the spins. In particular, we show that, with sufficient microwave control, the interaction between two spins can be written as a sum of five independently controllable Hamiltonian terms proportional to the five rank-2 spherical harmonics $Y_{2,q}(\theta, \phi)$, where (θ, ϕ) are the spherical coordinates of the vector connecting the two molecules. To demonstrate the potential of this approach beyond the simplest examples studied in [S.R. Manmana *et al.*, Phys. Rev. B. **87**, 081106 (2013)], we focus on the realisation of the Kitaev honeycomb model, which can support exotic non-Abelian anyonic excitations. We also discuss the possibility of generating spin Hamiltonians with arbitrary spin S, including those exhibiting SU(N=2S+1) symmetry.

Keywords: polar molecules; spin models; quantum magnetism; dipole–dipole interactions; Kitaev honeycomb model

1. Introduction

Recent experimental progress in the control of ultracold polar molecules [1–6] has stimulated the study of many-body systems featuring strong dipole–dipole interactions [7–10]. A particularly promising research area [11–25] of implementing lattice Hamiltonians from dipole–dipole-interacting molecules has emerged following the pioneering works of [26,27]. This research area largely owes its promise to the great degree of controllability that external DC, microwave, and optical fields can provide over rotational levels and dipole–dipole interactions [11–15,18,27–33]. In [34], we demonstrated that this controllability can yield a variety of spin Hamiltonians, in which spin states are encoded in superpositions of rotational states of the molecules. While dipole–dipole interactions are often thought of as being proportional to $1 - 3\cos^2\theta$, where $\mathbf{R} = (R, \theta, \phi)$ are the spherical coordinates of the vector connecting the two dipoles, we showed that not only the overall amplitude and sign of the spin Hamiltonian but also its individual terms can depend on $\hat{\mathbf{R}}$. We harnessed this dependence to propose the realisation of spin models known to harbour topological phases of matter [34].

In contrast to [34], which focused on the simplest examples, in the present paper we discuss a range of models that are more interesting but harder to implement experimentally. Specifically, as in [34], we emphasise that the interaction Hamiltonian between two molecules can, in principle, be a sum of five independently controllable terms proportional to the five rank-2 spherical harmonics $Y_{2,q}(\theta, \phi)$.

We then study a specific class of Hamiltonians with an arbitrary spin S. This class of Hamiltonians can be used to realise [34] the general SU(2) symmetric $S = 1$ model, i.e. the bilinear–biquadratic spin model, which has a rich phase diagram even in one dimension [11,35,36]. It can also give rise to SU(N=2S+1) spin models, which have recently attracted a great deal of theoretical and experimental attention due to their potential to access exotic strongly correlated phases in alkaline-earth atoms [37–40]. The strong dipole–dipole interactions of the polar-molecule implementation of SU(N) models can be larger than the SU(N)-symmetric superexchange interactions in cold-atom implementations [37].

Then we turn to the primary example of this paper: the implementation of the Kitaev honeycomb model. In contrast to the proposal of [29], our implementation of the Kitaev honeycomb model relies on direct – rather than perturbative – dipole–dipole interaction and hence gives rise to stronger interactions, which are easier to access experimentally. Other proposals for implementing the Kitaev honeycomb model use solid-state Mott insulators [41], trapped ions [42], superconducting quantum circuits [43], weak superexchange interaction between cold atoms in optical lattices [44], and an experimentally challenging system of coupled cavity arrays [45]. Related models involving

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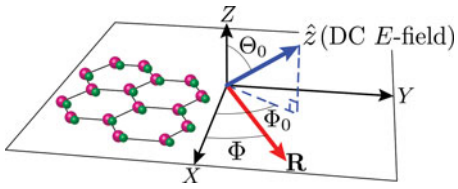

Figure 1. (colour online). The lattice of molecules is in the XY plane. The direction of the DC electric field is $\hat{z}$. The xyz coordinate system is obtained from the XYZ coordinate system by rotating the former around $\hat{Z}$ by Φ_0 and then rotating it around $\hat{y}$ by Θ_0. A typical vector $\mathbf{R}$ with spherical coordinates (R, θ, ϕ) in the xyz coordinate system has polar coordinates (R, Φ) in the XY plane.

many-body spin interactions can be implemented with polar molecules by employing time-dependent microwave fields [25]. We end the manuscript with a discussion of how we use microwave fields to create the dressed states that enable our implementations, followed by an outlook.

2. Setup

Consider a deep optical lattice in the XY plane shown in Figure 1 and loaded with one polar molecule per site. Each molecule is described by a rigid rotor with rotational angular momentum operator $\mathbf{N}$, rotational constant B, and dipole moment operator $\mathbf{d}$. In the presence of an electric field E along $\hat{z}$, each molecule is described by the Hamiltonian

$$H_0 = B\mathbf{N}^2 - Ed^z. \tag{1}$$

In general, the molecules can be allowed to hop to give rise to Hubbard-type [26,46] or t-J-type [47–49] models with highly tunable anisotropic long-range spin–spin interactions. In this paper, however, we will assume that the lattice is so deep that tunnelling is negligible and molecules are pinned in the motional ground state on each site. The field-free (i.e. $E = 0$) eigenstates of H_0 are the simultaneous eigenstates of $\mathbf{N}^2$ and N_z with eigenvalues $N(N+1)$ and M, respectively. Let us denote with $|N, M\rangle$ the eigenstates of H_0, which the field-free eigenstates connect to as E is turned on. For each molecule, we define [29] $d^{\pm} = \mp(d^x \pm id^y)/\sqrt{2}$, which changes the M of the molecule by ± 1, while $d^0 = d^z$ couples rotational states with the same M. There are no selection rules on N for $E \neq 0$.

Consider two molecules i and j separated by $\mathbf{R}_{ij}$, which has polar coordinates (R_{ij}, Φ_{ij}) in the XY plane and spherical coordinates $(R_{ij}, \theta_{ij}, \phi_{ij})$ in the xyz coordinate system. The dipole–dipole interaction between these two molecules is [50]

$$H_{ij} = \frac{\mathbf{d_i}\cdot\mathbf{d_j} - 3(\mathbf{d_i}\cdot\hat{\mathbf{R}}_{ij})(\mathbf{d_j}\cdot\hat{\mathbf{R}}_{ij})}{R_{ij}^3} \tag{2}$$

$$= -\frac{\sqrt{6}}{R_{ij}^3}\sum_{q=-2}^{2}(-1)^q C^2_{-q}(\theta_{ij},\phi_{ij})T^2_q(\mathbf{d_i},\mathbf{d_j}), \tag{3}$$

where $C^k_q(\theta,\phi) = \sqrt{\frac{4\pi}{2k+1}}Y_{kq}(\theta,\phi)$, Y_{kp} are spherical harmonics, $T^2_{\pm2} = d^{\pm}_i d^{\pm}_j$, $T^2_{\pm1} = (d^0_i d^{\pm}_j + d^{\pm}_i d^0_j)/\sqrt{2}$, and $T^2_0 = (d^-_i d^+_j + 2d^0_i d^0_j + d^+_i d^-_j)/\sqrt{6}$, so that T^2_q changes the total M of the two molecules by q. $T^2_{\pm1}$ contributes only at those values of the DC electric field that have $\sigma^{\pm}$ ($M \to M \pm 1$) and π (M-conserving) transitions of matching frequency.

In each molecule, we couple states $|N, M\rangle$ with several microwave fields and choose $2S+1$ dressed states (linear combinations of states $|N, M\rangle$) to define an effective spin S system. We will focus in this paper on homogeneous driving, which is easier to achieve as it can be done with microwave – as opposed to optical – fields. Extensions to inhomogeneous driving allow for an even richer class of Hamiltonians [51,52]. Assuming dipole–dipole interactions are too weak to take the molecules out of the $2S+1$ chosen dressed states, we can project dipole–dipole interactions onto these states to obtain a spin-S interaction Hamiltonian of the form $H = \frac{1}{2}\sum_{i\neq j} H_{ij}$, where

$$R^3_{ij}H_{ij} = \mathbf{v}(\theta_{ij},\phi_{ij})\cdot\mathbf{H}. \tag{4}$$

Here

$$\mathbf{v}(\theta,\phi) = \left(-2C^2_0, -\sqrt{6}\mathrm{Re}[C^2_2], \sqrt{6}\mathrm{Im}[C^2_2], \mathrm{Re}[C^2_1], \mathrm{Im}[C^2_1]\right)$$

is a real five-component vector describing the five different angular dependences (the prefactors are chosen for later convenience). Each of the five components of $\mathbf{H}$ is a Hamiltonian acting on the Hilbert space of the two spin-S particles i and j and comes with its own angular dependence. Specific examples for the components of $\mathbf{H}$ are given below, for example Equation (7) for the spin-1/2 case. Due to Hermiticity and symmetry under the exchange of the two particles, each component of $\mathbf{H}$ has $[(2S+1)^4 + (2S+1)^2]/2$ independent real coefficients. With an appropriate choice of rotational states, DC electric field strength, and a sufficient number of microwave fields, one might envision achieving full control over all $5[(2S+1)^4 + (2S+1)^2]/2$ coefficients, which, together with Θ_0, Φ_0, and a choice of lattice, determine the system. Requiring in addition that the total number of particles in any given internal state is conserved gives only $(2S+1)^2$ independent coefficients in each angular dependence.

Equation (4) allows one to access a great variety of exotic spin Hamiltonians. Since we will discuss $S = 1/2$ examples below in Equation (7), here we only point out that,

in [34], we showed how Equation (4) can be used to realise the most general SU(2)-symmetric $S = 1$ Hamiltonian (the so-called bilinear–biquadratic Hamiltonian) restricted to the C_0^2 angular dependence. In [34], we also briefly mentioned the possibility of realising the Kitaev honeycomb model. In the present paper, we discuss the details behind the realisation of the Kitaev honeycomb model and provide additional insights into how to generate spin Hamiltonians with an arbitrary S.

3. Spin Hamiltonians with arbitrary S

In this section, we show how to obtain a variety of spin Hamiltonians with an arbitrary S. For simplicity, in each molecule, we choose Q distinct $|N, M\rangle$ states and label them as $|a\rangle$, where $a = 1, \ldots, Q$. We break this set of Q states into $2S + 1$ disjoint sets labelled by $p = -S, \ldots, S$. We couple the states within each set with microwave fields to form dressed states in the rotating frame. We will show in detail below that $\sim n$ microwave fields are needed to couple n states and to create any desired linear combination out of them. We then choose one dressed state from each set to create the single-spin basis $|p\rangle = \sum_{a(p)} \sqrt{x_a}|a\rangle$ with rotating frame energies E_p. Here $\sum_{a(p)}$ means that a is summed over the states belonging to the set p. The coefficients x_a are assumed to be non-negative real numbers: allowing for superpositions with complex coefficients does not allow any additional tunability in this example [1]. The states $|p\rangle$ and $|q\rangle$ will refer to dressed states, while states $|a\rangle$ and $|b\rangle$ will refer to bare states. (One could also choose fewer sets and choose more than one dressed state from the same set.) For simplicity, we further assume that dipole–dipole interactions are so weak and the states are chosen in such a way that the two-molecule state $|pq\rangle$ is connected via H_{ij} only to itself and to $|qp\rangle$, while all the other processes are off-resonant and are negligible. Similarly, now in terms of the bare non-microwave-dressed states, we assume that the states are chosen in such a way that the two-molecule state $|ab\rangle$ is connected via H_{ij} only to itself and to $|ba\rangle$, while all the other processes are off-resonant and are negligible. This implies, in particular, that only C_0^2 contributes (an extension that includes the other four components of $\mathbf{v}$ is straightforward). The assumptions in the previous three sentences are generically satisfied if, for example, all the states $|a\rangle$ involved have $M \geq 0$ and no accidental degeneracies occur. We then have

$$H_{ij} = \frac{1 - 3\cos^2\theta_{ij}}{R_{ij}^3}\left[\sum_p B_p |pp\rangle\langle pp| + \sum_{p,q} A_p A_q |pq\rangle\langle pq| + \sum_{p<q} \frac{J_{p,q}}{2}(|qp\rangle\langle pq| + \text{h.c.})\right], \tag{5}$$

where $A_p = \sum_{a(p)} x_a \mu_a$, $B_p = \sum_{a(p) < b(p)} x_a x_b d_{a,b}$, $J_{p,q} = \sum_{a(p),\, b(q)} x_a x_b d_{a,b}$, where

$$d_{a,b} = \begin{cases} 2\mu_{a,b}^2 & \text{if } M(a) = M(b) \\ -\mu_{a,b}^2 & \text{if } |M(a) - M(b)| = 1. \\ 0 & \text{otherwise} \end{cases} \tag{6}$$

Here $\mu_{a,b} = \langle a|d^{M(a)-M(b)}|b\rangle$ and $\mu_a = \langle a|d^0|a\rangle$. A_p can be regarded as an effective dipole moment of state $|p\rangle$, while B_p encompasses the contribution to the interaction from the transition dipole moments between the states $a(p)$. On the other hand, $J_{p,q}$ describes the flip–flop transition involving states $|p\rangle$ and $|q\rangle$ and is driven by transition dipole moments coupling $a(p)$ to $b(q)$. The factor of 2 and the minus sign in Equation (6) arise from the fact that two identical dipoles rotating in phase in the x-y plane experience, on average, an interaction equal to negative one-half of that felt by these dipoles had they been pointing along $\hat{\mathbf{z}}$ [30].

The implementation of SU(N=2S+1)-symmetric models is an attractive case to consider given that such models have great potential for generating exotic phases including chiral spin liquids [53,54]. The expression in the square brackets in Equation (5) possess SU(N=2S+1) symmetry if and only if it is proportional to $\sum_{p,q} |pq\rangle\langle qp|$ up to an additive constant. Necessary and sufficient conditions for this are that $A_p A_q$, $J_{p,q}$, and $B_p + A_p^2$ do not depend on p and q for all $p < q$ and that $B_p + A_p^2 = A_p A_q + J_{p,q}/2$ for all $p < q$. As an example, for $N = 3$, it is sufficient to satisfy $A_1 = A_2 = 0$ and $B_1 = B_2 = B_3 + A_3^2 = \frac{1}{2}J_{1,2} = \frac{1}{2}J_{1,3} = \frac{1}{2}J_{2,3}$. In [34], we showed how to realise this and, in fact, an arbitrary SU(2) symmetric $S = 1$ interaction. Finding specific level configurations for higher N is postponed until future work.

4. The general $S = 1/2$ Hamiltonian

The most general $S = 1/2$ Hamiltonian implementable with dipole–dipole interactions is

$$\begin{aligned} R_{ij}^3 H_{ij} = {} & \mathbf{v}(\theta_{ij}, \phi_{ij}) \cdot [\mathbf{V} n_i n_j + \mathbf{W}_x (n_i S_j^x + S_i^x n_j) \\ & + \mathbf{W}_y (n_i S_j^y + S_i^y n_j) + \mathbf{W}_z (n_i S_j^z + S_i^z n_j) \\ & + \mathbf{J}_{xx} S_i^x S_j^x + \mathbf{J}_{yy} S_i^y S_j^y + \mathbf{J}_{zz} S_i^z S_j^z \\ & + \mathbf{J}_{xy} (S_i^x S_j^y + S_i^y S_j^x) + \mathbf{J}_{xz} (S_i^x S_j^z + S_i^z S_j^x) \\ & + \mathbf{J}_{yz} (S_i^y S_j^z + S_i^z S_j^y)], \end{aligned} \tag{7}$$

where we have 10 real 5-component vectors in the square brackets. By analogy with [34,47,48], we expect that these vectors might be independently controllable, an issue whose details we leave for future investigation. For this, we need $\sim$50 microwave fields. In Equation (7), n_i is the occupation

of site i. In the present manuscript, $n_i = 1$ for all sites. However, this more general form of the Hamiltonian is necessary when hopping is allowed, such as in [26,46–49].

In general, the interaction in Equation (7) is also accompanied by on-site Hamiltonian terms. In particular, if the on-site energy difference between $|\uparrow\rangle$ and $|\downarrow\rangle$ (the two dressed basis states making up the spin-1/2) is much larger than the strength of H_{ij}, only those interaction terms that conserve the total S^z play a role [47,48]. In order to realise models such as the quantum compass model [55], the Kitaev honeycomb model [56], and the Kitaev quantum double models [57] implemented with two-body interactions [58], we need to break S^z conservation and realise terms, such as $S_i^x S_j^x$. To do this, we simply tune $|\uparrow\rangle$ and $|\downarrow\rangle$ to be degenerate.

For $n_i = 1$, the Hamiltonian in Equation (7) is the special case of Equation (4): its restriction to $S = 1/2$. Despite the restriction to $S = 1/2$, the Hamiltonian in Equation (7) is still extremely powerful, as illustrated by the following examples. First, the commonly used density–density interaction with the $1 - 3\cos^2\theta_{ij}$ angular dependence (see e.g. [7,9]) comes from the first component of $\mathbf{V}$, while density–density interactions with more exotic angular dependences can be obtained by making use of all five components of $\mathbf{V}$. Second, the first components of $\mathbf{J}_{xy}$, $\mathbf{J}_{zz}$, $\mathbf{W}_z$, and $\mathbf{V}$ give rise to the spin–spin, density–spin, and density–density interactions of the generalised t-J model (referred to as the t-J-V-W model) of [47–49]. Third, the first three components of $\mathbf{J}_{xy}$ and $\mathbf{J}_{zz}$ give rise to an XXZ model featuring a direction-dependent spin-anisotropy [34]. In addition to providing access to symmetry protected topological phases in spin ladders [34], such an XXZ model can also be used to obtain a variety of exotic antiferromagnets including a square-lattice Heisenberg model, in which the ratio between coupling strengths on $\hat{X}$ and $\hat{Y}$ bonds is tunable [59]. Fourth, the $\mathbf{J}_{xx}$ and $\mathbf{J}_{zz}$ terms can be used to realise the quantum compass model [55]. In the present paper, we demonstrate the power of Equation (7) on another important example: the Kitaev honeycomb model [56].

5. Kitaev honeycomb

In Figure 1 and Figure 2(a), we show the Kitaev honeycomb lattice with Hamiltonian [56]

$$H = J_{xx} \sum_{x-\text{link}} S_i^x S_j^x + J_{yy} \sum_{y-\text{link}} S_i^y S_j^y + J_{zz} \sum_{z-\text{link}} S_i^z S_j^z. \tag{8}$$

We are interested in phase B, which contains the point $J_{xx} = J_{yy} = J_{zz} < 0$. This phase is gapless, but, in the presence

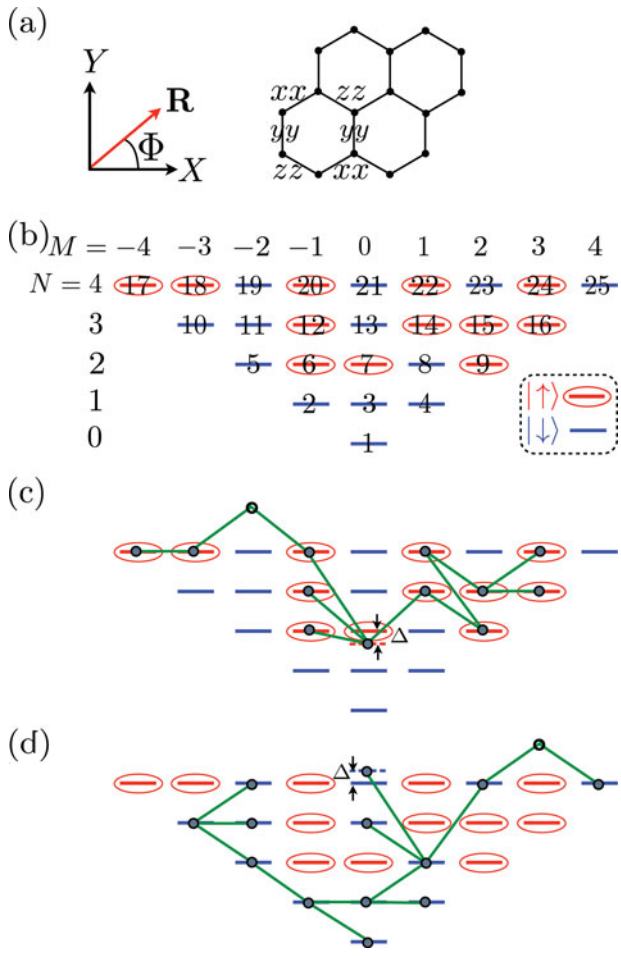

Figure 2. (colour online). Implementation of the Kitaev honeycomb model. (a) The honeycomb model: $S_i^x S_j^x$, $S_i^y S_j^y$, and $S_i^z S_j^z$ interactions along $\Phi_{ij} = \pi/6, \pi/2$, and $5\pi/6$, respectively. (b) The rotational levels used. The diagram is schematic: the real system is anharmonic and levels $|N, M\rangle$ with the same N are non-degenerate (unless the levels have the same $|M|$). The dressed states are $|\uparrow\rangle$ (linear combination of states indicated by ovals) and $|\downarrow\rangle$ (the rest). (c) Microwave fields that can be used to create the dressed state $|\uparrow\rangle$. Notice that level $|2, 0\rangle$ is used off-resonantly with a detuning Δ. (d) Microwave fields that can be used to create the dressed state $|\downarrow\rangle$. Notice that level $|4, 0\rangle$ is used off-resonantly with a detuning Δ.

of a magnetic field,

$$H_B = \sum_j (B_x S_j^x + B_y S_j^y + B_z S_j^z), \tag{9}$$

with all three components B_x, B_y, and B_z being non-zero, acquires a gap and supports non-Abelian anyonic excitations. Non-Abelian anyons have quantum computing applications and can be used, for example, for topologically protected quantum state transfer [60].

We use Equation (7) for a system of $n = 1$ molecules per site on a honeycomb optical lattice – which can be implemented with three laser beams [61–63] – to implement

the Kitaev honeycomb model. Specifically, we would like to obtain $S_i^x S_j^x$ interactions along $\Phi_{ij} = \pi/6$, $S_i^y S_j^y$ interactions along $\Phi_{ij} = \pi/2$, and $S_i^z S_j^z$ interactions along $\Phi_{ij} = 5\pi/6$ [see Figure 2(a)]. It is worth noting that interactions along Φ_{ij} and $\Phi_{ij} + \pi$ are the same.

Let us first point out some general facts that hold independently of the direction and magnitude of the applied DC electric field and of the dressed states $|\uparrow\rangle$ and $|\downarrow\rangle$ that we choose. The terms in Equation (7) involving $\mathbf{V}$ play no role since they are proportional to identity. Ignoring boundary effects, the terms in Equation (7) involving $\mathbf{W}_x$, $\mathbf{W}_y$, and $\mathbf{W}_z$ give rise to a uniform magnetic field, precisely as required by Equation (9); we will compute this field below.

Having taken care of the $\mathbf{V}$ and $\mathbf{W}_\alpha$ terms, we omit them from the Hamiltonian. We would like the remaining terms in the Hamiltonian ($\mathbf{J}_{xx}, \mathbf{J}_{yy}, \mathbf{J}_{zz}, \mathbf{J}_{xy}, \mathbf{J}_{xz}, \mathbf{J}_{yz}$) to satisfy

$$H_{ij}\left(\left\{\frac{\pi}{6}, \frac{\pi}{2}, \frac{5\pi}{6}\right\}\right) = -\frac{J}{R_{ij}^3}\left\{S_i^x S_j^x, S_i^y S_j^y, S_i^z S_j^z\right\} \quad (10)$$

with $J > 0$ for the three indicated values of Φ_{ij}. Since, for any Φ_{ij}, $\mathbf{v}(\Phi_{ij})$ can be written as a linear combination of $\mathbf{v}(\pi/6)$, $\mathbf{v}(\pi/2)$, and $\mathbf{v}(5\pi/6)$, Equation (10) completely determines the Hamiltonian at all Φ_{ij}:

$$\begin{aligned} H_{ij}(\Phi_{ij}) &= \sum_{n=1}^{3} \frac{1 - 2\cos\left(2\Phi_{ij} - 2(n+1)\pi/3\right)}{3} \\ &\quad \times H_{ij}\left(\frac{(2n-1)\pi}{6}\right) \quad (11) \\ &= -\frac{J}{3R_{ij}^3}\Big\{\left[1 - 2\cos\left(2\Phi_{ij} - 4\pi/3\right)\right] S_i^x S_j^x \\ &\qquad + \left[1 - 2\cos\left(2\Phi_{ij}\right)\right] S_i^y S_j^y \\ &\qquad + \left[1 - 2\cos\left(2\Phi_{ij} - 2\pi/3\right)\right] S_i^z S_j^z\Big\}. \end{aligned}$$

We will show how to realise this momentarily. First, observe that, for example, along $\Phi_{ij} = \pi/3$, which is halfway between $S_i^x S_j^x$ and $S_i^y S_j^y$,

$$H_{ij}(\pi/3) = -\frac{J}{3R_{ij}^3}(2S_i^x S_j^x + 2S_i^y S_j^y - S_i^z S_j^z). \quad (12)$$

Because of the $1/R^3$ dependence, the strength of such next-nearest-neighbour interactions (along $\Phi_{ij} = 0, \pi/3, 2\pi/3$) is reduced relative to the nearest-neighbour interactions by $1/3^{3/2} \approx 1/5$, which will be the largest correction introduced by long-range interactions. While in some cases long-range corrections are weak enough to ensure the survival of the desired phases [34], it is an open question whether this holds for the present example.

It now remains to find the DC electric field strength and direction, as well as the dressed states, that yield Equation (10). We assume that the DC electric field is along the Z axis (i.e. $\Theta_0 = 0$) and take $\Phi_0 = 0$. In that case, the last two components of $\mathbf{v}$ vanish, which means that $C_{\pm 1}^2$ terms do not contribute ($v_4 = v_5 = 0$). This is fine because those terms are the hardest to generate experimentally as they are non-vanishing only at specific values of the DC electric field, and it will turn out they are not required to generate the Kitaev Hamiltonian. At $\Theta_0 = \Phi_0 = 0$, the uniform magnetic field coming from $\mathbf{W}_\alpha$ terms is easy to calculate. Indeed, v_2 and v_3 do not contribute to the magnetic field since the honeycomb lattice is invariant under $2\pi/3$ rotations and since $\sum_{n=0}^{2}\{v_2(\Phi_1 + n2\pi/3), v_3(\Phi_1 + n2\pi/3)\} = \{0, 0\}$ for any Φ_1. Finally, summing over all bonds connected to a given site, the term $\mathbf{W}\cdot(\mathbf{S}_i + \mathbf{S}_j)$ with angular dependence v_1 gives rise to a uniform magnetic field of strength $6.58\mathbf{W}$.

Since $v_4 = v_5 = 0$, we drop the last two components of $\mathbf{v}$ and $\mathbf{H}$ to obtain

$$\mathbf{v}(\Phi) = (1, -\frac{3}{2}\cos(2\Phi), \frac{3}{2}\sin(2\Phi)) \quad (13)$$

and

$$\begin{aligned} \mathbf{H} = \{&d_i^0 d_j^0 + \frac{1}{2}(d_i^- d_j^+ + d_i^+ d_j^-), \\ &d_i^+ d_j^+ + d_i^- d_j^-, i(d_i^+ d_j^+ - d_i^- d_j^-)\}. \end{aligned} \quad (14)$$

The linear independence of $\mathbf{v}(\pi/6)$, $\mathbf{v}(\pi/2)$, and $\mathbf{v}(5\pi/6)$ allows, in principle, for the possibility of obtaining Equation (10).

We will use the 25 states shown and numbered in Figure 2(b). We write the two dressed states as $|\sigma\rangle = \sum_{a(\sigma)} y_a |a\rangle$, where y_a are complex, $\sum_{a(\sigma)} |y_a|^2 = 1$, $\sigma = \uparrow, \downarrow$, and $\sum_{a(\sigma)}$ means that one sums a over the 12 (13) states belonging to $|\uparrow\rangle$ ($|\downarrow\rangle$) in Figure 2(b). We now keep only resonant terms and project $\mathbf{H}$ in Equation (14) onto dressed states $|\uparrow\rangle$ and $|\downarrow\rangle$.

We work at a DC electric field $E = 10B/d$, where d is the permanent dipole moment of the molecule. The following transitions contribute to H_1: $|N, M\rangle|N', M'\rangle \to |N, M\rangle|N', M'\rangle$, $|N, M\rangle|N', M'\rangle \to |N', M'\rangle|N, M\rangle$ (for $|M - M'| \leq 1$, as dictated by electric-dipole selection rules), and $|N, M\rangle|N', -M\rangle \to |N', M\rangle|N, -M\rangle$. The transition $|N, -M-1\rangle|N', M\rangle \to |N', -M\rangle|N, M+1\rangle$ is the only one contributing to $d_i^+ d_j^+$ in H_2 and H_3, while $d_i^- d_j^-$ is the Hermitian conjugate of $d_i^+ d_j^+$. Projecting these terms on $|\uparrow\rangle$ and $|\downarrow\rangle$, we obtain an expression for $\mathbf{H}$ in terms of y_a. This allows us to get an expression for $\mathbf{W}$ and for the left-hand side of Equation (10). Since a magnetic field that is too large will eventually take us out of the Kitaev B phase, we first verify that we can obtain Equation (10) with a minimal magnetic field $\mathbf{W}$. One can then easily verify that the set of y_a given in footnote [2]

gives $J = 0.0284d^2$ and $6.58|\mathbf{W}| = 7 \times 10^{-7}d^2$ (which is negligibly small).

By adjusting the coefficients y_a, one can find points with larger values of $|\mathbf{W}|$. For example, the coefficients y_a given in footnote [3] achieve $J = 0.0264d^2$ and $6.58\mathbf{W} = \{0.00072, 0.00084, -0.03937\}$.

6. Microwave dressing

So far, we have not discussed in detail what exact microwave couplings are required to create the dressed states $|\uparrow\rangle$ and $|\downarrow\rangle$ needed to implement the Kitaev honeycomb model. In this section, we provide such a discussion. The methods we present here are generally applicable to the creation of generic spin models from dipolar interactions.

Although generating large numbers of precisely tuned microwaves is experimentally feasible [64], the effort in obtaining and verifying the spectrum of microwaves increases with the number of frequencies. It is thus experimentally desirable to use as few microwave fields as possible. In order to create a dressed state out of n bare states, one needs at least $n-1$ microwaves (assuming the same microwave cannot be used to couple more than one transition). Therefore, let us first address the question of whether $n-1$ microwaves suffice to prepare a dressed state at any prescribed rotating frame energy featuring any prescribed superposition of the bare states.

Suppose we would like to prepare a dressed state $|D\rangle = \sum_{a=1}^{n} y_a |a\rangle$ out of the n bare states $|a\rangle$ ($a = 1, \ldots, n$) with prescribed coefficients y_a and a prescribed rotating-frame energy E relative to the bare state $|m\rangle$. Suppose state $|a\rangle$ is coupled to state $|a+1\rangle$ using a microwave with Rabi frequency Ω_a for $a = 1, \ldots, n-1$. The rotating-frame Hamiltonian is then

$$\begin{pmatrix} \Delta_1 & \Omega_1 & 0 & \cdots & 0 & 0 \\ \Omega_1^* & \Delta_2 & \Omega_2 & \cdots & 0 & 0 \\ 0 & \Omega_2^* & \Delta_3 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & \Delta_{n-1} & \Omega_{n-1} \\ 0 & 0 & 0 & \cdots & \Omega_{n-1}^* & \Delta_n \end{pmatrix} \begin{pmatrix} y_1 \\ y_2 \\ y_3 \\ \vdots \\ y_{n-1} \\ y_n \end{pmatrix} = E \begin{pmatrix} y_1 \\ y_2 \\ y_3 \\ \vdots \\ y_{n-1} \\ y_n \end{pmatrix}.$$

Here $\Delta_m = 0$ (recall that $|m\rangle$ is the reference sate), while $\Delta_{a+1} - \Delta_a$ is the detuning of the a'th microwave field from the $|a\rangle \to |a+1\rangle$ transition. Writing

$$\Omega_a = \tilde{\Omega}_a \exp\{i \arg(y_a/y_{a+1})\}, \tag{15}$$

where $\tilde{\Omega}_a$ are real, we obtain n real-number equations

$$\begin{aligned} \tilde{\Omega}_1 \left|\frac{y_2}{y_1}\right| &= E - \Delta_1, \\ \tilde{\Omega}_1 \left|\frac{y_1}{y_2}\right| + \tilde{\Omega}_2 \left|\frac{y_3}{y_2}\right| &= E - \Delta_2, \\ &\vdots \\ \tilde{\Omega}_{a-1} \left|\frac{y_{a-1}}{y_a}\right| + \tilde{\Omega}_a \left|\frac{y_{a+1}}{y_a}\right| &= E - \Delta_a, \\ &\vdots \\ \tilde{\Omega}_{n-2} \left|\frac{y_{n-2}}{y_{n-1}}\right| + \tilde{\Omega}_{n-1} \left|\frac{y_n}{y_{n-1}}\right| &= E - \Delta_{n-1}, \\ \tilde{\Omega}_{n-1} \left|\frac{y_{n-1}}{y_n}\right| &= E - \Delta_n. \end{aligned} \tag{16}$$

One can satisfy the equations using $n-1$ Rabi frequencies $\tilde{\Omega}_a$ and any one non-zero detuning Δ_c. Allowing other detunings to be non-zero, one obtains freedom in how to choose the magnitudes of $\tilde{\Omega}_a$. This can be useful, for example, in cases when the magnitudes of $\tilde{\Omega}_a$ need to be larger than a certain value (e.g. larger than the strength of hyperfine interactions, as in [34,48] – see our Outlook).

The approach we have just described works nicely in the cases where the only off-diagonal bare-states processes that contribute to the final spin Hamiltonian have the form $|a\rangle|b\rangle \to |b\rangle|a\rangle$. This holds for all the configurations studied in [47,48]. It also holds [4] for the level configuration used to implement the bilinear–biquadratic model in [34].

On the other hand, consider a model where $\{|\uparrow\rangle, |\downarrow\rangle\} = \{|0,0\rangle, y_{-1}|1,-1\rangle + y_1|1,1\rangle\}$ (as in [34]) and where we want the process $|1,-1\rangle|0,0\rangle \to |0,0\rangle|1,1\rangle$ to contribute. We can couple states $|1,-1\rangle$ and $|1,1\rangle$ with two microwave fields via state $|2,0\rangle$. It is crucial that these two microwave fields have the same frequency; otherwise, in the rotating frame, the process $|1,-1\rangle|0,0\rangle \to |0,0\rangle|1,1\rangle$ would pick up an oscillatory time dependence and will average out to zero. This means that, in the cases where off-diagonal bare-state processes other than $|a\rangle|b\rangle \to |b\rangle|a\rangle$ contribute, there are restrictions on what detunings can be chosen as non-zero.

In the case of the configuration for the Kitaev honeycomb model shown in Figure 2(b), it is easy to check that the microwave couplings shown in Figure 2(c) and 2(d) for $|\uparrow\rangle$ and $|\downarrow\rangle$, respectively, work. In particular, in Figure 2(c) all microwaves are assumed to be resonant except for the microwaves coupled to state $|2,0\rangle$, which are all detuned by the same amount Δ. This detuning provides the extra tuning parameter one has to add to the Rabi frequencies in order to fully control the coefficients y_j defining state $|\uparrow\rangle$. The fact that $|2,0\rangle$ has $M = 0$ ensures that it is non-degenerate. Hence, any contributing bare-state process containing $|2,0\rangle$ in the initial two-molecule state will also contain it in the final two-molecule state, meaning that a detuning of this state keeps the process resonant. Similar arguments apply to Figure 2(d), where all microwaves are assumed to be resonant except for the microwave coupling to state $|4,0\rangle$. To ensure that $|\uparrow\rangle$ and $|\downarrow\rangle$ are degenerate, we use $|2,2\rangle$ and $|2,-2\rangle$ as the two reference states and set the two dressed-state energies to be equal. It is also worth pointing out that an additional $N = 5$ state is introduced in both Figure 2(c)

and 2(d) to act as an intermediate state for coupling two states with $\Delta M = 2$. We also note that one has to be careful using π transitions to couple states with $M \neq 0$ since the same microwave will act on both M and $-M$.

A solution alternative to the introduction of a detuning is an introduction of additional resonant microwave couplings, resulting in configurations such as three states coupled with three microwave fields. At the same time, it is worth pointing out that, in some cases, one may wish to introduce additional detunings on purpose with the goal of eliminating certain interaction processes (written in terms of bare non-microwave-dressed states as $|a, b\rangle \rightarrow |a', b'\rangle$), which can be regarded as an additional control knob.

7. Outlook

The most straightforward implementation of our proposal would involve polar molecules with no nuclear spin. This is possible, for example, in the case of a SrO molecule, which has a large permanent dipole moment of 8.9 Debye [29]. In the case where hyperfine structure is present, such as in alkali dimers, we assume that the Rabi frequencies Ω_a are larger than the strength of hyperfine interactions $H_{\rm hf}$. In situations where this poses a significant restriction, the opposite limit, in which $\Omega_a \ll H_{\rm hf}$, can also be considered.

As in [48], we assume that the Rabi frequencies Ω_a are also larger than the amplitude of the optical lattice potential. In this case, one can first compute the microwave-dressed states and only then include the effect of the optical lattice by finding what potential each dressed state experiences. Several approaches to ensuring that each dressed state sees the same potential are outlined in [11,29,48,65,66]. However, even in the cases where dressed states see different potentials, the resulting Hamiltonian is modified only slightly as some interaction coefficients are somewhat reduced [48,67].

While we have focused on pinned molecules, allowing the molecules to hop in the lattice gives rise to *t–J*-type [47–49] or Hubbard-type [26] models with highly tunable long-range spin–spin interactions featuring a direction-dependent spin anisotropy. As a particularly simple example, one can obtain a single-component system with a density–density interaction whose angular dependence is given by a linear combination of all five C_q^2 [see the $\mathbf{V}$ term in Equation (7)]. This may allow one to study, for example, exotic quantum Hall physics [68].

The approaches we presented can be used to tune dipole–dipole interactions to zero [12,30]. In that case, one has to consider corrections due to dipole–dipole interactions to second order (which give van-der-Waals-type $1/R^6$ dependence) and interactions between higher multipole moments [69]. These interactions can also be viewed as extra control knobs.

We have focused in the present manuscript on polar molecules. However, the ideas that we presented should be extendable in a straightforward manner to other systems interacting via dipole–dipole interactions such as magnetic atoms [70,71], Rydberg atoms [72,73], and magnetic solid-state defects [74,75].

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Notes

1. Allowing for complex coefficients does not allow any additional tunability in this example because we assume that, in terms of the bare non-microwave-dressed states, the two-molecule state $|ab\rangle$ is connected via H_{ij} only to itself and to $|ba\rangle$.
2. $\{y_1, \ldots, y_{25}\} = \{-0.0016 - 0.0230i, -0.1077 + 0.1635i, 0.2272 + 0.0323i, 0.0168 - 0.0024i, -0.0277 - 0.1580i, 0.2258 + 0.1997i, 0.0503 + 0.0694i, -0.4994 + 0.1819i, 0.3618 - 0.0554i, 0.3558 + 0.1635i, 0.0530 - 0.1025i, 0.1942 - 0.0515i, -0.1135 + 0.0599i, 0.2284 + 0.2537i, -0.3417 - 0.1720i, -0.3946 + 0.0264i, -0.0739 + 0.0092i, -0.3268 - 0.0782i, -0.0200 + 0.0088i, -0.1105 - 0.0720i, 0.5198 + 0.0203i, -0.0598 - 0.0452i, -0.1838 + 0.1808i, -0.3090 - 0.2669i, -0.1163 + 0.2564i\}$.
3. $\{y_1, \ldots, y_{25}\} = \{0.0041 - 0.0220i, -0.1773 + 0.1736i, 0.2159 + 0.0282i, 0.0212 - 0.0025i, -0.0394 - 0.1639i, 0.2020 + 0.1610i, 0.0082 + 0.1014i, -0.5091 + 0.1405i, 0.2452 - 0.0578i, 0.4216 + 0.1754i, 0.0255 - 0.0924i, 0.1975 - 0.0497i, -0.1510 + 0.0547i, 0.2955 + 0.2858i, -0.4215 - 0.1541i, -0.4519 + 0.0592i, -0.0477 + 0.0083i, -0.3147 - 0.0823i, -0.0449 + 0.0088i, -0.0298 - 0.1000i, 0.4665 + 0.0034i, -0.0981 - 0.0361i, 0.0425 + 0.1662i, -0.1913 - 0.2707i, -0.1028 + 0.2820i\}$.
4. The process $|1, 0\rangle |1, 0\rangle \rightarrow |0, 0\rangle |2, 0\rangle$ is not of the form $|a\rangle |b\rangle \rightarrow |b\rangle |a\rangle$ but the presented microwave construction still applies since we use the three bare states involved ($|1, 0\rangle$, $|1, 1\rangle$, and $|1, 2\rangle$) as the reference states for the zero of the dressed-state energies.

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