Far from equilibrium quantum magnetism with ultracold polar molecules

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Recent theory has indicated how to emulate tunable models of quantum magnetism with ultracold polar molecules. Here we show that present molecule optical lattice experiments can accomplish three crucial goals for quantum emulation, despite currently being well below unit filling and not quantum degenerate. The first is to verify and benchmark the models proposed to describe these systems. The second is to prepare correlated and possibly useful states in well-understood regimes. The third is to explore many-body physics inaccessible to existing theoretical techniques. Our proposal relies on a non-equilibrium protocol that can be viewed either as Ramsey spectroscopy or an interaction quench. It uses only routine experimental tools available in any ultracold molecule experiment.

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Excitement about the recent achievement of near-degenerate ultracold polar molecules [1] in optical lattices [6] stems from their strong dipolar interactions and rich internal structure, including rotational, vibrational, and hyperfine states. These features may be applied to tests of fundamental constants [7], quantum information [8], ultracold chemistry [9], and quantum emulation of condensed matter models [10, 11]. In this paper our focus is on molecules as emulators of quantum magnetism [12–24], specifically as proposed in Refs. [25, 26]. Models of quantum magnetism have some of the simplest many-body Hamiltonians, yet describe numerous materials [27–29] and display condensed matter phases ranging from fundamental to exotic: antiferromagnets, valence bond solids, symmetry protected topological phases, and spin liquids. Emulating quantum magnetism with molecules is appealing because, like cold atoms, the systems are clean and the microscopics well understood. Advantages over cold atom emulations of quantum magnetism [30] include orders of magnitude larger energy scales and more tunable Hamiltonians [25, 26]. These prior studies have focused on spin ground states of unit filling insulators. In contrast, we propose a simple dynamic procedure applicable to present experiments, which are ultracold, but non-degenerate and low density. We show that interesting many body quantum magnetism can be studied immediately.

Specifically we show how experiments may use this dynamics to achieve major goals for emulating quantum magnetism, and we outline these goals to motivate our calculations. First, although interesting models of quantum magnetism are predicted to describe ultracold molecules under appropriate circumstances, this has yet to be experimentally demonstrated. The proposed dynamic protocol allows such a demonstration as well as benchmarking of the emulator’s accuracy. Second, one wishes to prepare interesting correlated — and possibly useful — states. This protocol can generate such states in well-understood regimes. Finally, one wants to explore behavior in these models in regimes inaccessible to present theoretical tools. This is the generic case for the proposed dynamics. We emphasize that all of these goals are achievable under existing experimental conditions [6], despite present experiments being non-quantum degenerate and at low density. Furthermore, they require only routinely used measurement and preparation tools [31].

Background.—Ref. [23] show how molecule rotational states can serve as effective spins, and that dipolar interactions provide an effective spin-spin interaction. In the simplest case, one populates two rotational levels in a dc electric field $E$ [67] and works in a deep lattice to allow no tunneling. In this limit, a spin-1/2 dipolar quantum XXZ model describes the molecules [68]:

$$H = \frac{1}{2} \sum_{i \neq j} V_{dd}(i,j) \left[ J z S_i^z S_j^z + \frac{J}{2} (S_i^+ S_j^- + H.c.) \right].$$

The sum runs over all molecules, $S_i^\mp$ and $S_j^\mp$ are the spin-1/2 operators satisfying $[S_i^+, S_j^-] = \pm S_i^z$, and $V_{dd}(i,j) = (1 - 3 \cos^2 \Theta_{ij}) / |r_i - r_j|^3$ with $r_i$ the $i$’th molecule’s position in lattice units and $\Theta_{ij}$ the angle between $E$ and $r_i - r_j$. For simplicity and concreteness we assume a dimension $d = 2$ system with $E$ perpendicular to it, so
\( V_{dd}(i,j) = 1/|\mathbf{r}_i - \mathbf{r}_j|^3 \), but our ideas apply in arbitrary geometries. One may tune \( J_{L}/J_z \) by changing \( E \) and the choice of rotational state. We denote by \(|0\rangle\), \(|1\rangle\), and \(|2\rangle\) the three lowest energy rotational eigenstates in the applied \( E \)-field with zero angular momentum along the quantization axis. Choosing \(|0\rangle\) and \(|1\rangle\) to make the spin-1/2, one can tune \( \infty > J_{L}/J_z > 0.35 \) using (readily achievable) \( E \)-fields from 0 to 16 kV/cm. Choosing \(|0\rangle\) and \(|2\rangle\) for the spin-1/2, one can tune \( 0 < J_{L}/J_z < 0.1 \) for similar \( E \)-fields. A characteristic scale for these couplings is 400 Hz in KRB and 40 kHz in LiCs [32], compared to \( \lesssim 100\) Hz in cold atoms using superexchange [33]. KRB molecules recently have been loaded in a deep three-dimensional lattice with 25 second lifetimes [6], allowing dynamics lasting thousands of \( J_{L}^{-1} \) and \( J_z^{-1} \).

One important aspect of the ongoing experiments is that the filling \( f \) is much less than one molecule per site. The JILA experiments estimate \( f \sim 0.1 \). As a result, although the molecules’ positions are static throughout one shot, they fluctuate shot-to-shot. Thus, rather than forming a regular lattice, the spins’ locations have significant disorder. Our calculations show that the dynamic protocol’s utility persists with disorder.

We use a simple disorder model that likely describes current experiments. We assume that each site is occupied with a probability \( p \) that is independent of other sites [69]. If the molecules are fermions (e.g., KRB [6]) then for current temperatures, which occupy only the lowest band, no sites can be doubly occupied and \( p = f \). This also applies to bosons with a strong on-site density-density interaction (e.g., RbCs [51]). The trap causes \( f \) to vary spatially. Although we show results only for the homogeneous system, we have taken the trap into account and found that our conclusions remain valid [35].

Remarkably, close relatives of such seemingly unusual models exist in the literature, for example the Blume-Emery-Griffiths model [30]. These mainly focus on the classical equilibrium limit, \( J_z = 0 \). They were introduced to understand materials [31, 32], \(^3\)He-\(^4\)He mixtures [30], Griffiths phases [39], glassy dynamics [40, 41], and the interplay of the underlying lattice’s statistical mechanics (site-dilution percolation) with that of the magnetism living on that lattice [42, 44]. Rather than studying unique disorder-induced behavior, we focus on showing that \( f = 1 \) behavior survives disorder.

Only through “disorder” does temperature enter, because the deep lattice freezes out the motion. In particular, one must distinguish motional temperature from spin temperature. The former may be large but is entirely captured by the disorder, while the latter is ill-defined since we consider non-equilibrium spin states. However, experimental microwave manipulation can produce essentially zero entropy spin states. While one could worry that disorder washes out the behavior, we will show that strong correlations, entanglement, and interesting many body physics survive large amounts of disorder.

**Dynamic protocol.**—Our dynamic procedure may be alternatively viewed as Ramsey spectroscopy or an interaction quench (Fig. 1). Ref. [45] studied closely related Rabi spectroscopy. In Ramsey spectroscopy, a well established tool in atomic physics, one begins with all molecules in the rotational ground state and applies two strong, resonant microwave pulses separated by time \( t \). The first pulse initializes the spin states along \( \theta \), specifically to \( \cos(\theta/2)e^{i\varphi/2}(|\uparrow\rangle + \sin(\theta/2)e^{-i\varphi/2}(|\downarrow\rangle) \), for an angle \( \theta \) set by the pulse area, with high fidelity (\( > 99\% \)). We take \( \varphi = 0 \) with no loss of generality. The second pulse rotates a desired spin component, chosen by the pulse area and phase, to the \( z \) axis. In this way one can measure any desired collective spin component \( \langle \hat{n} \cdot \mathbf{S} \rangle \), where \( \hat{n} \) is a unit vector and \( S^\alpha = \sum_i S_i^\alpha \) with \( \alpha \in \{x, y, z\} \). One can also obtain higher moment correlations, e.g. \( \langle (\hat{n} \cdot \mathbf{S})^2 \rangle \), from the measurement record. Between these pulses the spins evolve for a time \( t \) under the Hamiltonian in Eq. (1). We note that molecule experiments have recently began using this protocol [31] and Ref. [45] applied it to long-range Ising models in recent Penning trap experiments with \( \sim 300 \) ions.

If one imagines adding a transverse field term \( \hbar \mathbf{S} \cdot \hat{n}_\theta \) to the Hamiltonian in Eq. (1), with \( \hat{n}_\theta \) a unit vector pointing \( \theta \) from the \( -z \) axis (see Fig. 1), the Ramsey protocol corresponds to a quench from \( h = \infty \) to \( h = 0 \). One may therefore be able to explore, for example, Kibble-Zurek physics (e.g., entropy production, topological defects) [46, 47].

**Theoretical methods.**—We calculate dynamics in four limits: (1) short times, \( \{J_L, J_z\}t \ll 1 \), (2) Ising, \( J_L = 0 \), (3) near-Heisenberg [SU(2)], \( |J_z - J_L| \ll J_z \), and (4) one dimension for arbitrary \( J_L/J_z \). The first three limits’ results are analytic and valid in any dimension, while the last is from numerically exact adaptive time-dependent density matrix renormalization group (adaptive t-DMRG) [49, 51]. Details of the calculations will be presented elsewhere [55]. In all cases \( S^Z \) is conserved, with \( \langle S_i^Z(t) \rangle = -(f/2) \cos \theta \).

**Short time limit, \( \{J_L, J_z\}t \ll 1 \).** For short times, \( \langle \mathcal{O}(t) \rangle = \langle \mathcal{O} \rangle - \text{i} t \langle [\mathcal{O}, H] \rangle - \frac{t^2}{2} \langle [\mathcal{O}, H], H \rangle + O(t^3) \) for an operator \( \mathcal{O} \). We calculate the commutators and time dependence of \( \langle S_i^\alpha(t) \rangle \) to leading non-zero order, and \( \langle S^\alpha(t)S^\gamma(t) \rangle \) to linear order. We find

\[
\langle S_i^\alpha(t) \rangle = \frac{f}{2} \sin \theta \left\{ 1 - \frac{f t^2}{8} \left[ \Xi_2 + f \Upsilon \cos^2 \theta \right] \right\} + O(\tau^4),
\]

\[
\langle S_i^\alpha(t)S^\gamma(t) \rangle = -\left( f^2 \tau \Xi_1/8 \right) \sin(2\theta) + O(\tau^3),
\]

where \( \tau = (J_z - J_L)t \), \( \Xi_m = \sum_{i \neq j} V_{dd}^{m\alpha}(i, i + j) \), and \( \Upsilon = \Xi_1 - \Xi_2 \). Note that for these homogeneous systems, these observables are independent of \( i \). Similarly, defining
The expectation $\langle S^y_i(t) \rangle$ takes the imaginary (rather than real) part of the square-bracketed expression in Eq. (4).

$$\langle S^y_i(t) \rangle = f \frac{\sin(\theta)}{2} \text{Re} \left[ \prod_{j \neq i} g(\rho_j) e^{\frac{1}{2} i \tau J_z V_{dd}(i,j)} \right]$$

where the sum runs over $\rho = 0$ (unoccupied site) and $\rho = \pm 1$ ($S^z = \pm 1/2$), and

$$g(\rho) = \begin{cases} 1 - f & \text{if } \rho = 0 \\ f \sin^2(\theta/2) & \text{if } \rho = 1 \\ f \cos^2(\theta/2) & \text{if } \rho = -1 \end{cases}$$

The expectation $\langle S^y_i(t) \rangle$ is the corresponding imaginary part. Unlike the other approximations, this is valid only for finite $N$.

**One dimension.**—We use adaptive t-DMRG [29, 32] to calculate dynamics of one dimensional chains. We treat 20 site chains and find finite size effects to be fairly small. We discretize time in steps of $t = 0.05 J^{-1}_z$, and find a discarded weight of $\lesssim 10^{-9}$ for times $\lesssim 10 J^{-1}_z$, adaptively keeping $m = 50-500$ reduced density matrix states. Altogether, we expect errors dominated by the disorder average, which is taken over 100 random configurations.

**Results: global perspective.**—Fig. 2 overviews dynamics, from the calculations above, as a function of $J_z / J_x$, $f$, and $\theta$. Experimentally, these are controlled by electric field [23, 24], temperature/density, and first Ramsey
timescales are roughly independent of dynamics’ magnitude at times ∼ {f} for f = 1, but the damping oscillations. Approaching J⊥ = Jz, the dynamics slows down, since at J⊥ = Jz the initial state is an eigenstate of the Hamiltonian. As J⊥/Jz increases further, the dynamics is damped with characteristic timescale (J⊥ − Jz)^-1.

For f ≪ 1, the behavior crosses over to that of independent clusters, eventually with only two particles. The largest frequency is roughly half that for f = 1, since there is a single neighbor instead of two. Thus, the dynamics remains roughly as fast as for f = 1, but the dynamics’ magnitude at times ∼ {Jz^-1, J⊥^-1} is smaller since there are fewer molecules and only a fraction of them are close enough to interact. At any f the overall timescales are roughly independent of θ but the damping vanishes as θ → 0.

Achieving goals of emulating quantum magnetism.— Fig. 3 shows the characteristic dependence of the XXZ model’s short time dynamics on θ, f, and {Jz, J⊥}. This can be used to experimentally verify the emulation of the XXZ model and benchmark its accuracy.

To achieve the second goal of generating interesting well-understood states, both the near-Heisenberg and Ising limits are useful. For f ≈ 1 and θ = π/2 near the Heisenberg point, the state at t = π/(2χ) is |GHZ⟩ = 1/√2(|+→ − −→⟩ + e^{iφ} |−→ + −→⟩) for some φ [47, 60]. This is a cat state, specifically the GHZ or NOON state, which suffices for universal measurement based quantum computing [42]. The presence of long range interactions perturbs the cluster state, and an interesting question is how this affects its utility. Decoherence can also limit the creation of entangled states.

A generic implementation of the proposed dynamics in d > 1 achieves the third goal, emulating quantum magnetism in theoretically intractable regimes. Away from the short time, Ising, and Heisenberg limits, no solution is known in d > 1. As Fig. 4 shows, in d = 1 strong correlations develop, suggesting the difficulty of d > 1 calculations. The long time f = 1 correlations are even larger at large distance than in the ground state. Interestingly, the dynamics shows a light-cone-like spreading to an apparent steady state.

Experimental outlook.— Though our discussion focused on molecules, we point out that the dynamics studied here can have direct application in other physical systems, including condensed matter [41], trapped ions [45, 63], and optical lattice clocks [64, 65].

We close by noting technical details for molecule experiments. Rotational states’ polarizabilities differ [25, 66], so the optical trap induces a spatially varying field \( \sum_i h_i S^z_i \). Also, Eq. (1) ignores density-density \( n_in_j \) and density-spin \( n_iS^z_j \) interactions [25, 26]. For f < 1, the latter gives a spatially varying magnetic field that depends on molecules’ random positions. Spin-echo pulses common in Ramsey experiments remove both effects.

Summary.— We have shown that Ramsey spectroscopy enables ongoing ultracold polar molecule experiments to accomplish three goals for emulating quantum magnetism: (1) benchmarking the emulation’s accuracy (using short time dynamics), (2) generating strongly correlated and entangled states in well-understood limits (Ising, near-Heisenberg, one dimension), and (3) exploring strongly correlated dynamics in regimes inaccessible to theory (generic case in dimensions d > 1).

Finally, we mention that in addition to the XXZ Hamiltonian explored in this paper, our dynamic protocol should be useful for verifying emulation of more complicated spin models that may be realized with ultracold molecules, as in Refs. [25, 26] and beyond.

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[67] The electric field ensures that the two rotational levels are sufficiently off-resonant from other levels that they form an isolated spin-1/2 [26].
[68] We neglect hyperfine coupling, as Refs. [25, 26] justify.
[69] In addition to simplicity, this disorder distribution results from suddenly quenching tunneling $t$ to zero for lattice fermions initially at a temperature $T \gg t$.
[70] These results also extend straightforwardly to arbitrary spatially varying initial angles and longitudinal magnetic fields, as we will present elsewhere [55].