

Quantum Computation and Quantum Simulation with Ultracold Molecules

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Ultracold molecules confined in optical lattices or tweezer traps can be used to process quantum information and simulate the behaviour of many-body quantum systems. Molecules offer several advantages for these applications. They have a large set of stable states with strong transitions between them and long coherence times. They can be prepared in a chosen state with high fidelity, and the state populations can be measured efficiently. They have controllable long-range dipole-dipole interactions that can be used to entangle pairs of molecules and generate interesting many-body states. We review the advances that have been made and the challenges still to overcome, and describe the new ideas that will unlock the full potential of the field.

I. INTRODUCTION

Producing gases of polar molecules cooled to within a millionth of a degree of absolute zero is now routine in many laboratories, either by associating pairs of alkali-metal atoms [1–13] or by direct laser cooling [14–18]. At such ultracold temperatures both the motional and internal states of the molecules can be exquisitely controlled. Moreover, standard techniques from the field of ultracold atoms, such as optical lattices [19] and optical tweezers [20], can be used to confine molecules in ordered arrays. These developments unlock many new opportunities in quantum science that exploit the rich properties of molecules.

Molecules, unlike atoms, may possess an intrinsic electric dipole moment allowing the generation of controlled long-range dipole-dipole interactions (DDI) through the application of electric and/or microwave fields. Typical interaction strengths lie between those found in highly magnetic atoms [21, 22] and Rydberg atoms [23, 24], providing a different regime for study. Additionally, vibration and rotation of the molecule lead to a rich internal structure of long-lived states. This combination of properties leads to new experimental possibilities and a broad range of applications, spanning ultracold chemistry [25], few-body physics [26], precision measurement [27], quantum simulation [28] and quantum computation [29].

In this review we focus on the use of ultracold molecules for quantum simulation and quantum computation. We outline the new opportunities in these fields stemming from the properties of molecules. We give a snapshot of the current experimental state of the art and describe the challenges specific to molecules. We present

a vision for the immediate next steps in the field and describe several nascent ideas which could unlock the full potential of ultracold molecules.

The interested reader is directed to earlier reviews [30–35] that chart the remarkable progress in the field.

II. THEORETICAL CONSIDERATIONS

We briefly review the basic theory of molecular structure and interactions that motivates the growing interest in ultracold molecules. We explicitly considers a $^1\Sigma$ diatomic molecule in its vibrational ground state, but the long-ranged coupling of rotational states holds more generally [37]. Such molecules are described by a rigid-rotor Hamiltonian with eigenstates $|N, M_N\rangle$ labeled by the rotational angular momentum N and its projection M_N along the quantisation axis. The rotational state energies form a ladder with anharmonic spacing given by $E_N = B_v N(N + 1)$, where B_v is the rotational constant (see Fig. 1(a)). Transitions between neighboring rotational states typically lie in the microwave domain. The rotational levels are $(2N + 1)$ -fold degenerate, but in practise, there is hyperfine structure and the degeneracy is lifted by an applied magnetic field. We therefore can consider $|N, M_N\rangle$ states as isolated, and a pair of states may encode a spin-1/2 or qubit.

The symmetry of the $|N, M_N\rangle$ states leads to vanishing electric dipole moments in the laboratory frame. However, controllable DDI can be engineered in several ways. An electric field can be used to create static dipole moments (see Fig. 1(b) & (c)). Microwave fields can be used to create rotational-state superpositions with oscillating dipole moments. Finally a pair of molecules can be prepared in neighboring rotational states connected by an allowed electric-dipole transition. The DDI between molecules i and j with dipole moments $\vec{\mu}_k$ separated by

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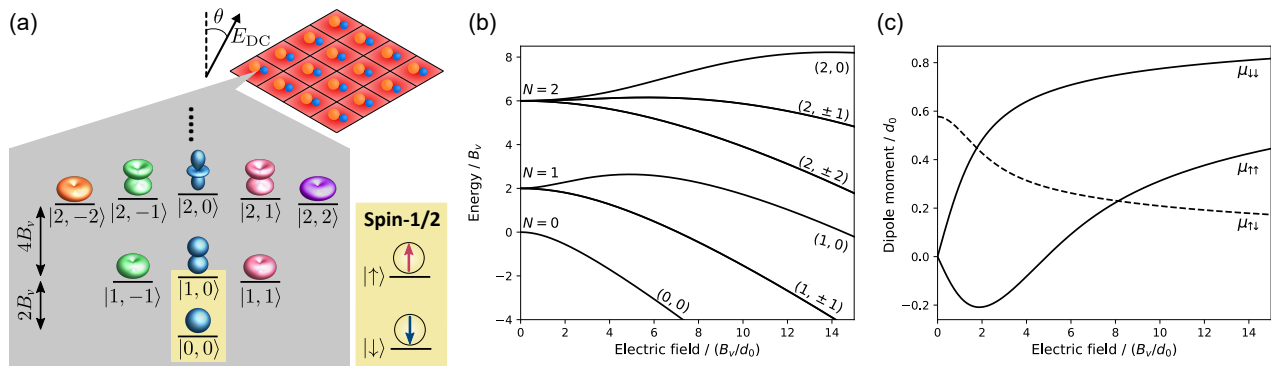


FIG. 1. (a) Pseudo-spins (or qubits) may be encoded in the rotational states of ultracold polar molecules confined in optical lattices (or tweezer arrays) for applications in quantum simulation and computation. Typical rotational constants, B_v , lead to transitions between rotational states in the microwave domain. Spin-exchange and Ising interactions between molecules may be controlled through the applied field, E_{DC} . (b) Energy and (c) dipole moments of a rigid rotor in an electric field. Energies are labelled by (N, M_N) at high field. Dipole moments are shown for $|\downarrow\rangle = |0, 0\rangle$ and $|\uparrow\rangle = |1, 0\rangle$. All quantities are scaled to the rotational constant B_v and molecule-frame dipole moment d_0 . (c) The tunability of the spin-exchange ($\propto \mu_{\uparrow\downarrow}^2$) and Ising interaction ($\propto (\mu_{\uparrow\uparrow} - \mu_{\downarrow\downarrow})^2$). (a) Adapted from [36] with permission from the authors.

$\vec{r}_{ij} = \vec{r}_i - \vec{r}_j = r_{ij}\vec{e}_{ij}$ is

$$V_{ij}^{\text{DDI}} = \frac{1}{4\pi\epsilon_0} \frac{\vec{\mu}_i \cdot \vec{\mu}_j - (\vec{\mu}_i \cdot \vec{e}_{ij})(\vec{\mu}_j \cdot \vec{e}_{ij})}{r_{ij}^3}. \quad (1)$$

In Sections III&IV below, we present how the interaction in Eqn. (1) can be mapped onto a subset of states to encode a spin or qubit and perform quantum simulations and quantum information processing. We will explain how the choice of states and applied fields dictates the nature of the interactions, and how some non-interacting state combinations can be useful for storing quantum information. Throughout it is useful to keep in mind that typical electric dipole moments of diatomic molecules are ~ 1 Debye resulting in interaction strengths ~ 1 kHz for molecules spaced by 532 nm in an optical lattice.

III. MOLECULES IN LATTICES FOR QUANTUM SIMULATION

A. Model Hamiltonians and many-body phases

Quantum simulations with ultracold molecules divide into two classes, one where molecules are frozen in place by a lattice or microtraps to eliminate losses, and one where the molecules can move and losses are tolerated or mitigated. In the former, frozen motion necessitates using rotational states to have interesting many-body physics and generally leads to interacting spin models as studied in quantum magnetism. In the latter, interesting physics can occur already with the rotational ground state.

When molecules are frozen in place, projecting Eqn. (1) onto the rotational states populated experimentally gives a spin model. With two rotational states, a

unit-filling array is described by the dipolar XXZ model,

$$H_{\text{XXZ}} = \frac{1}{2} \sum_{i \neq j} \left[\frac{J_{\perp}}{2} (S_i^+ S_j^- + S_i^- S_j^+) + J_z S_i^z S_j^z \right] \quad (2)$$

where the $\{S_j^a\}$ are the site- j spin operators. Refs. [38, 39] theoretically showed how experiments could engineer Eqn. (2), building on a body of research starting in Ref. [40] and reviewed in Ref. [32]. J_{\perp} and J_z depend on the states chosen and the electric field, illustrated in Fig. 1. As an example, internal states $|\downarrow\rangle = |N=0, M_N=0\rangle$ and $|\uparrow\rangle = |N=1, M_N=0\rangle$ yield

$$J_{\perp} = \frac{1 - 3 \cos^2 \theta_{ij}}{4\pi\epsilon_0 r_{ij}^3} \times 2\mu_{\uparrow\downarrow}^2, \quad (3)$$

$$J_z = \frac{1 - 3 \cos^2 \theta_{ij}}{4\pi\epsilon_0 r_{ij}^3} \times (\mu_{\uparrow\uparrow} - \mu_{\downarrow\downarrow})^2, \quad (4)$$

where $\mu_{\sigma\tau} = \langle \sigma | \hat{\mu}^{(z)} | \tau \rangle$ and $\hat{\mu}^{(z)}$ is the z -component of $\hat{\mu}_i$. The J_{\perp} term describes spin-exchange; the J_z term describes Ising interactions, which vanish absent an electric field. Both prefactors depend on the angle θ_{ij} between the dipole moments and the intermolecular vector, highlighting the anisotropy of the DDI: side-by-side dipoles repel, while head-to-tail dipoles attract. The form of the interactions is the same for other state choices, but magnitudes change [32, 39]. More generally, below unit filling, Eqn. (1) gives two additional interactions: spin-density $[W(S_i^z n_j + n_i S_j^z)]$ and density-density $[V n_i n_j]$, where n_i is the site- i number operator. Allowing tunneling results in a highly-tunable t - J - V - W model [38, 39].

The model Eqn. (2) is a long-ranged interacting variant of the widely-studied XXZ model. It generalizes Ising ($J_{\perp} = 0$), XY ($J_z = 0$), and Heisenberg ($J_{\perp} = J_z$) models that are studied intensely in statistical mechanics, condensed matter, and many-body physics [41–43].

Their ground states display a broad range of symmetry breaking phases, topological phases, including, spin liquids [44], and quantum phase transitions depending on J_{\perp}/J_z and the lattice geometry. Driven out of equilibrium, integrable dynamics, integrability breaking, and unconventional superdiffusive scaling in one dimensional systems have been predicted [45–48].

Several theoretical proposals give blueprints for spin-1/2 and higher-spin models beyond the “naturally-occurring” XXZ model. The most common techniques are microwave dressing the rotational states [38] and Floquet engineering with periodic microwave pulse sequences [49–51], as recently demonstrated experimentally [52]. Both allow flexible control of the two-body interaction – sums of $S_i^{\alpha}S_j^{\beta}$ and significant spatial anisotropy can be created. These can engineer spin models with interacting symmetry-protected topological ground states [53], spin-orbit coupling [54] (related physics has been seen in Rydberg atoms [55]), and the famous Kitaev honeycomb model that harbors exotic non-Abelian topological order [56, 57] and can robustly encode and process quantum information. Raman lasers can induce spatially-varying interactions and exotic fractional Chern [58] and Hopf insulators [59]. Recent work discussed in Sec. VB has explored the use of more rotational states to realise synthetic dimensions [60–62]. We note that earlier influential work suggested an alternative implementation of spin models with ultracold molecules [63, 64]. Although extremely flexible, this approach requires ${}^2\Sigma_{1/2}$ molecules and has yet to be attempted.

The molecules need not be pinned to a lattice site, or confined in a lattice at all, and this opens up quantum simulations of itinerant particles. However, this allows fast inelastic losses to occur, even in molecules that are ostensibly non-reactive (see also the Perspective Ref. [65] in this issue), which must be suppressed or circumvented. In a lattice, the continuous quantum Zeno effect can suppress losses: if molecules start on different sites, they are forbidden from hopping onto the same site by continuous strong “measurements” associated with the on-site reactive losses, and long lifetimes are experimentally observed [66, 67]. In weaker lattices or bulk gases, dc electric-field shielding [68–70] or microwave shielding of reactions [71–77] can suppress losses by adding a repulsive barrier to the short-ranged interaction potential.

If reactive losses can be successfully suppressed, itinerant models offer a wealth of important physics. The t - J - V - W model mentioned above can occur in either bosonic or fermionic variants. The fermionic version is an extension of the usual t - J model, which in certain limits approximates the Hubbard model [78]. The Hubbard model is central to condensed-matter physics, in part as a minimal model capturing the interplay of tunneling and interactions and in part for its relation to high-temperature superconducting cuprates [79–81]. The t - J_{\perp} model arising without an electric field was predicted in Ref. [82] to have several ground-state phases even in one dimension,

including spin-density waves, charge-density waves, spin-gapped superconductors, and phase separated regimes, and correlations whose long-distance power-law decay relates non-trivially to the dipolar interaction power law, and Ref. [83] predicted d -wave superfluids on a checkerboard. The two-dimensional case is numerically challenging and an excellent quantum simulation target, with important open questions, e.g. how holes bind.

Itinerant physics requires neither a lattice nor the use of multiple rotational states; molecules in the bulk in their rotational ground state are expected to show interesting phases of matter, including p -wave topological superfluids in two-dimensional fermionic molecules [84–86], superfluid liquid crystals [87], supersolids in self-bound droplets [88], and Wigner crystals when interactions are dressed [89–91]. With a lattice, but using only rotational ground states, extended Hubbard Hamiltonians can be realized, which possess a variety of ordered patterns with and without superfluidity [28, 92].

While itinerant quantum simulations potentially display a wide variety of phenomena, earlier theory neglected both reactive losses and any shielding to suppress them. The detailed structure of the interaction potential will differ when shielding is present, requiring modification of previous theory. While this may change details, many phenomena are likely to survive.

B. Experimental and theoretical challenges

Many quantum simulations discussed above require molecules in coherent rotational-state superpositions in lattices with fillings approaching one molecule per site, and in the lowest energy band for experiments where tunneling plays a role. This is challenging for several reasons.

Molecules exhibit an anisotropic polarisability where, e.g. for a diatomic molecule, the polarisability along the bond axis generally differs from that orthogonal to the bond. This leads to light shifts that depend on N , M_N , and the angle between the laser polarisation and the quantisation axis [93, 94]. Further complications from hyperfine states result in a rich, trap intensity-dependent level structure [36, 95]. The resulting differential light shifts severely limit the bare single-molecule coherence times and can mask the (typically $\sim 0.1 - 10$ kHz) DDI.

Notably, achieving high filling of molecules in a lattice remains challenging. An early proposal [96] suggested dual-species atomic Mott insulators [97, 98] could produce one atom of each species per site, with atom pairs subsequently converted to molecules. In practise this approach is difficult: it is sensitive to the species-dependence of the interactions, spatial overlaps, and the relative atomic polarisabilities. A modified protocol using superfluid ${}^{87}\text{Rb}$ and Mott insulating ${}^{133}\text{Cs}$ with tuned interactions achieved a filling exceeding 30% for Feshbach molecules [99]. Similar fillings have been reported for ground state ${}^{40}\text{K}{}^{87}\text{Rb}$ by combining a Mott insulator of ${}^{87}\text{Rb}$ with a band insulator of ${}^{40}\text{K}$ [100].

Although these allow access to significant many-body physics, higher filling is desirable. The recent creation of quantum degenerate molecular gases (Section VA) provides a promising path to low-entropy molecules in lattices.

A major additional challenge for quantum simulation with molecules is suppressing losses to explore the many-body phases in itinerant molecules, though shielding techniques may alleviate this. Other frontiers include implementing single-site addressing and entropy engineering, realising more complex Hamiltonians, and extending to new ultracold molecules, including those produced by direct laser cooling.

Finally, calculating the properties of molecular systems is often extremely difficult, whether one- and two-molecule properties, such as susceptibilities and scattering cross sections, or many-body behaviors. This causes issues for quantum simulation experiments: difficulty preparing low-temperature equilibrium states, measuring their temperature, or diagnosing their validity. However, progress is being made even where properties cannot be calculated, for example fluctuation-dissipation relations for thermometry [101–103], Lieb-Robinson bounds to guarantee stability of local observables under small imperfections [104], and quantum control for state preparation [105]. Although these techniques need to be adapted to molecules, their general principles apply. These challenges are also a blessing: the difficulty predicting the behaviour provides an opportunity for experiments to act as powerful quantum simulators that give insights into physics that cannot be predicted classically.

C. Measurements of spin dynamics

The field of quantum simulation of spin models with ultracold molecules has just begun, with three experiments beginning to show its power. The spin-exchange interactions (J_{\perp} term in Eqn. (2)) were first observed using $^{40}\text{K}^{87}\text{Rb}$ molecules in a three-dimensional optical lattice [66] following a technique proposed in Ref. [107]. The molecules were initialised in $|\downarrow\rangle = |0, 0\rangle$ and interrogated with a Ramsey sequence on a ~ 2.2 GHz microwave transition to $|\uparrow\rangle = |1, -1\rangle$. Experiments observed that the Ramsey contrast oscillated as a function of the time between $\pi/2$ pulses with a frequency matching the expected nearest-neighbor interaction strength, $J_{\perp}/2 \simeq h \times 52$ Hz, and decayed with a dependence on density characteristic of DDI [66]; Ref. [108] used $|\uparrow\rangle = |1, 0\rangle$ and observed the expected change of DDI strength. Ref. [66] also allowed the molecules to tunnel along one direction and observed the continuous quantum Zeno effect induced by fast on-site loss rates. These experiments showed that even lattice fillings of 10-25% and bulk measurements can observe DDI effects, laying the foundations for the study of many-body dynamics in disordered dipolar systems.

Recent work used a quantum gas microscope to mea-

sure site-resolved spin dynamics of $^{23}\text{Na}^{87}\text{Rb}$ in a two-dimensional lattice, again with two rotational states as an effective spin-1/2 [52]. This builds on the observation of the Hanbury Brown-Twiss effect in molecules, the first application of a molecular quantum gas microscope [109]. Spin correlations were measured (Fig. 2(c) and (d)), isolating first, second, and third-nearest neighbors which, combined with varying the quantisation axis, directly evidenced the DDI's anisotropy. They also engineered the XXZ Hamiltonian starting from the $J_z = 0$ by using a Floquet technique [110, 111] where a microwave pulse sequence creates the desired time-averaged Hamiltonian. Quantum gas microscopy of ultracold polar molecules is a pivotal result with great promise for the future.

Li et al. have studied dynamics involving both spin and motion [106]. They combine DC electric and AC microwave fields to control $^{40}\text{K}^{87}\text{Rb}$ in isolated 2D planes (see Fig. 2(d)). Their DC electric field gives $J_z \neq 0$ in Eqn. (2), and the spin dynamics depend on $\chi = J_z - J_{\perp}$. The DDI slightly shifts the rotational transition frequency (~ 100 Hz), which is measured using a Ramsey sequence with XY8 dynamical decoupling to achieve sufficiently long coherence times. Their measurements (Fig. 2(f)) demonstrate control of the interactions using both the strength and orientation of the electric field. The field orientation affects the spin-exchange and Ising interactions similarly (Eqn. (4)), whereas its magnitude controls their relative strength (see Fig. 1(c)). At short times the molecules move little so dynamics are primarily of the spins, and switching the internal state of the molecule can coherently reverse the spin dynamics. At longer times, collisions irreversibly decohere and thermalise the molecules [68]. Stronger DDI was observed to lead to faster collisions and, hence, faster coherence loss. The authors point to future studies of the Heisenberg model ($\chi = 0$) by tuning the electric field to ~ 7 kV/cm, as shown in Fig. 2(f). Collectively the results highlight the tunability of spin Hamiltonians implemented using ultracold polar molecules.

Importantly, much of the recent progress stems from improved control and understanding of molecular quantum states in applied fields, as discussed in a Perspective in this issue [65]. Methods to eliminate or suppress differential light shifts of rotational transitions in optical potentials, thereby extending coherence times, have been crucial. Often the intrinsic single-molecule coherence time is only ~ 1 ms [36, 106]. Methods to match the polarisabilities of different rotational states have included tuning the trap polarisation [93, 94, 112–114], intensity [36] and ellipticity [115]. Recently magic-wavelength trapping [116, 117] has emerged as an effective solution [118–121]. Indeed, the experiments using $^{23}\text{Na}^{87}\text{Rb}$ molecules discussed above used a near-magic wavelength lattice where the polarisabilities of the two relevant rotational states differed by less than 1%, leading to bare single-molecule coherence times of 56(2) ms. In very recent work, second-scale rotational coherences have been reported for $^{87}\text{Rb}^{133}\text{Cs}$ molecules, achieved by tuning the

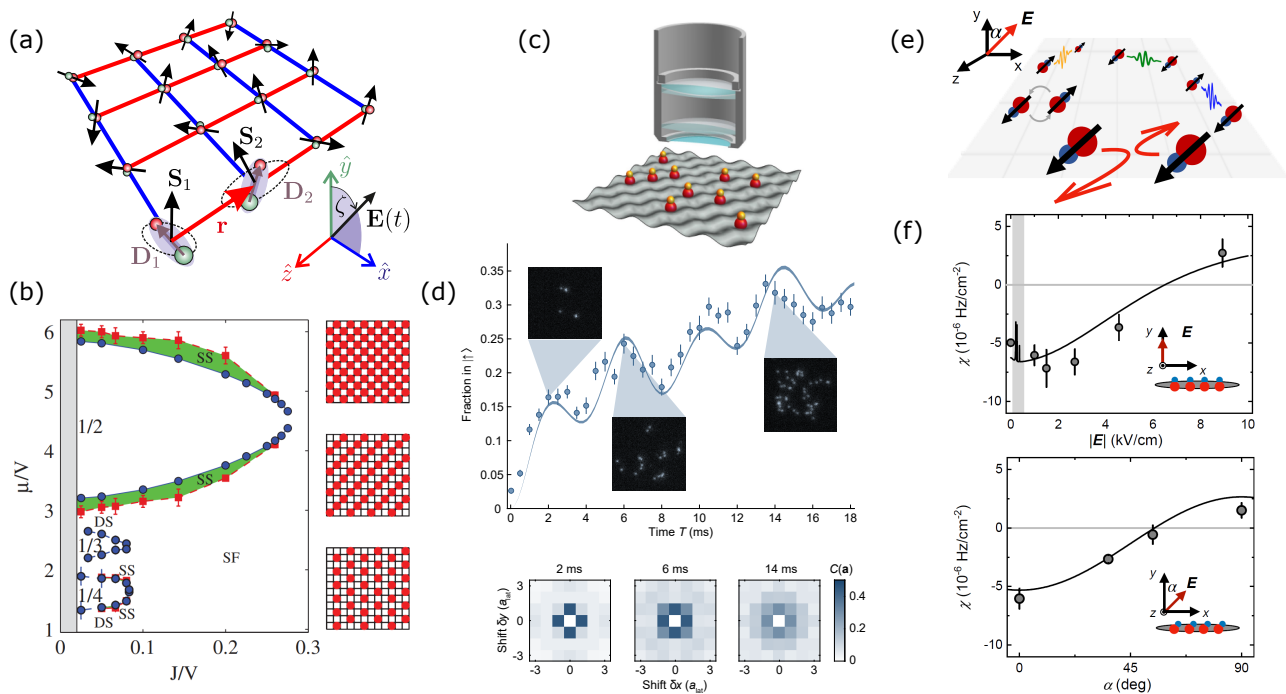


FIG. 2. Quantum simulation with ultracold polar molecules. (a) Molecules pinned to the sites of an optical lattice can be used to engineer spin models. (b) Novel quantum phases become accessible when the molecules are able to tunnel in the lattice. The zero-temperature phase diagram as a function of tunnelling rate J and chemical potential μ (both scaled to the DDI strength, V) exhibits a devil's staircase (DS) with checkerboard, stripe and star Mott lobes (shown right), as well as superfluid (SF) and supersolid (SS) phases. Other situations show topological phases. (c) Schematic of a molecular quantum gas microscope used to image individual $^{23}\text{Na}^{87}\text{Rb}$ molecules in a 2D lattice with single-site resolution. (d) Observing correlations between molecules interacting in an XY spin model. The insets show typical fluorescence images and, below, their associated correlation matrices. (e) Exploration of itinerant quantum magnetism in a thermal 2D gas of $^{40}\text{K}^{87}\text{Rb}$ molecules. (f) Controlling the spin-spin interaction (χ) with the strength (top) and direction (bottom) of the applied electric field. (a) Adapted from [63], (b) adapted from [92], (c)-(d) adapted from [52] and (e)-(f) adapted from [106], with permissions from the authors.

trap wavelength near weakly-allowed transitions to the $b^3\Pi-A^1\Sigma^+$ potential [122]. At the same time, more elaborate microwave pulse sequences, such as XY8 dynamical decoupling, have been used [106, 123, 124] to reduce single-particle dephasing; Ref. [106] improved the coherence time by a factor of ~ 70 from 0.24(1) to 17(1) ms. With these techniques, rotational coherences have become a usable resource and further quantum simulation experiments are eagerly anticipated.

IV. MOLECULES IN TWEEZERS FOR QUANTUM COMPUTATION

Molecules provide many stable hyperfine and rotational states that make attractive qubits, and long-range interactions suitable for entangling operations. Fast single qubit gates can be driven using microwave fields or two-photon optical processes. State-selective detection can be achieved by driving an optical cycling transition in the molecule or, following coherent dissociation, in one of the constituent atoms.

A. Tweezer arrays

An array of optical tweezer traps, each containing a single molecule, is an ideal starting point for quantum computation with molecules. In 2019, Anderegg et al. [125] trapped laser-cooled CaF molecules in an array of five tweezers. The molecules were cooled from a MOT into a dipole trap using Λ -enhanced gray molasses cooling to increase the density, then loaded into tweezers and detected with high fidelity by fluorescence imaging. Light-assisted collisions result in a collisional blockade, ensuring zero or one molecule per tweezer. These methods have now been extended to larger arrays and tighter confinement, enabling entanglement of pairs as described below.

In 2021, Cairncross et al. [12] created single ground-state NaCs molecules in optical tweezers. First, Na and Cs were loaded into separate tweezers from a dual species MOT. Fluorescence imaging was used to select experiments containing an atom pair. These atoms were then cooled to the motional ground state by polarization gradient cooling followed by Raman sideband cooling, and subsequently prepared in single quantum states. Then,

the traps were merged adiabatically and the pair converted into a weakly-bound molecule by sweeping a magnetic field through a Feshbach resonance. Finally, the weakly-bound molecule was transferred to a single hyperfine component of the rovibrational ground state by a two-photon Raman transition. The efficiency of producing the ground state molecule from a pair of atoms was $31 \pm 4\%$, the probability of occupying the motional ground state was $65 \pm 5\%$, and the lifetime of the ground-state molecule in the tweezer was 3.4 ± 1.6 s. This pioneering work was soon extended to small 1D arrays [126]. Similar results have now been reported for RbCs molecules [127].

B. Qubits and coherence times

High-fidelity gate operations require a coherence time much longer than the gate time. For molecules, long hyperfine and rotational coherence times require a qubit that has low sensitivity to fluctuations in magnetic field and tweezer trap intensity. The qubit transition should also be well separated from other transitions so that single-qubit gates can be driven quickly without coupling to any other states. This can be challenging for molecules with large nuclear spins and small rotational constants, where the density of states is high.

In their ground-states, alkali molecules have no electronic angular momentum so the hyperfine structure originates exclusively from the nuclear spins and is highly insensitive to external fields. Nuclear spin superpositions can be prepared by driving a two-photon transition. Using NaK molecules in an optical dipole trap, Park et al. [128] demonstrated a hyperfine coherence time close to one second. To go further, Gregory et al. [129] identified a pair of nuclear spin states in the ground state of RbCs that have identical magnetic moments at a specific magnetic field. At this field the magnetically-insensitive qubit has a long coherence time, limited by a tiny differential ac Stark shift coupled with the non-uniform intensity of the trapping light. For light linearly polarized at angle θ to the magnetic field, this tensor shift is proportional to the Legendre polynomial $P_2(\cos\theta)$. Setting $\cos\theta = 1/\sqrt{3}$ thus eliminates the differential Stark shift. In this case, no decoherence of the hyperfine qubit was discernible and a coherence time exceeding 5.6 s was deduced.

Rotational coherence times have been studied for CaF in tweezer traps [113]. Here, a pair of states in neighboring rotational manifolds forms a qubit that, to first order, is magnetically insensitive around zero field. Although there is a differential light shift, its gradient with intensity, at some chosen intensity, can be tuned to zero through the choice of polarization angle. Combined with active cancellation of magnetic field noise, the experiment measured a rotational coherence time of 93 ± 7 ms for molecules cooled to $5 \mu\text{K}$. A spin echo extended the coherence time to 470 ± 40 ms, probably limited by residual sensitivity to the intensity variations across the trap.

C. Entanglement

DDI can be used to entangle molecules and implement two-qubit gates [29, 130–133]. The first paper on this topic [29] proposed a 1D array of molecules polarised by an electric field, E_{DC} , with qubits $|\downarrow\rangle = |\tilde{N} = 0, M_N = 0\rangle$ and $|\uparrow\rangle = |1, 0\rangle$, whose dipole moments depend on E_{DC} (Fig. 1). A small electric field gradient along the array shifts the qubit frequency to provide single-site addressability. The two-qubit gate uses the Ising interaction from Eqn. (2), proportional to $(\mu_{\uparrow\uparrow} - \mu_{\downarrow\downarrow})^2$, to distinguish the frequencies of the molecule-pair transitions $|\downarrow\rangle|\downarrow\rangle \leftrightarrow |\downarrow\rangle|\uparrow\rangle$ and $|\uparrow\rangle|\downarrow\rangle \leftrightarrow |\uparrow\rangle|\uparrow\rangle$. A microwave or two-photon pulse that resolves this frequency difference implements a two-qubit gate.

Alternatively, molecules can be entangled without applying E_{DC} by using the spin-exchange interaction of Eqn. (2), proportional to $\mu_{\uparrow\downarrow}^2$. A molecule pair prepared in $|\downarrow\rangle|\uparrow\rangle$ will evolve into $|\uparrow\rangle|\downarrow\rangle$. Reference [132] shows one way of using this spin-exchange for two-qubit gates. Here, the qubit states $|0\rangle$ and $|1\rangle$ are hyperfine levels of the ground rotational state, and a rotationally excited state $|e\rangle$ is used for the interaction. A pair of molecules from an array is moved to an interaction zone, a microwave π -pulse transfers $|1\rangle \rightarrow |e\rangle$, the pair are brought close together and then separated again so that the time-integrated interaction swaps $|1\rangle|e\rangle$ and $|e\rangle|1\rangle$, and then a second π -pulse transfers $|e\rangle \rightarrow |1\rangle$ to complete the gate. All other molecules have a different tweezer intensity to the selected pair, so are light-shifted out of resonance with the microwave field.

A similar method uses stationary molecules in stationary states. In the $|\downarrow\rangle, |\uparrow\rangle$ representation, the eigenstates of the DDI are $|\downarrow\rangle|\downarrow\rangle, |\uparrow\rangle|\uparrow\rangle$ and $|\Psi^\pm\rangle = \frac{1}{\sqrt{2}}(|\downarrow\rangle|\uparrow\rangle \pm |\uparrow\rangle|\downarrow\rangle)$. The entangled states $|\Psi^\pm\rangle$ are separated by the interaction energy, $2J_\perp$. A 2π -pulse resonant with $|0\rangle|0\rangle \leftrightarrow |\Psi^+\rangle$ implements a two-qubit gate. In practice, it is better to use qubit states with no electric dipole coupling and an auxiliary state for the two-qubit gate. A simple implementation is described in Ref. [134]. A more sophisticated one uses an optimized microwave field pulse to produce a gate that is robust to errors in the trapping and control fields [133], and gate fidelities exceeding 99.9% are predicted to be feasible.

Two remarkable papers [123, 124] have demonstrated deterministic entanglement of molecule pairs using dipolar spin-exchange interactions. Both used rotational qubits of CaF trapped in tweezer arrays (Fig. 3(a)). After bringing molecule pairs initialized in $|\uparrow\uparrow\rangle$ close, the experiments applied a Ramsey sequence – a pair of $\pi/2$ pulses separated by an evolution period t – and a dynamical decoupling sequence to extend coherence times, and then measured the population in $|\uparrow\uparrow\rangle$, $P_{\uparrow\uparrow}$. Spin-exchange causes $P_{\uparrow\uparrow}$ to oscillate with an angular frequency of $J_\perp/(2\hbar)$. Figure 3(c), from Ref. [123], shows these oscillations. The measured value of J_\perp as a function of molecule separation r (Fig. 3(d)) is found to fol-

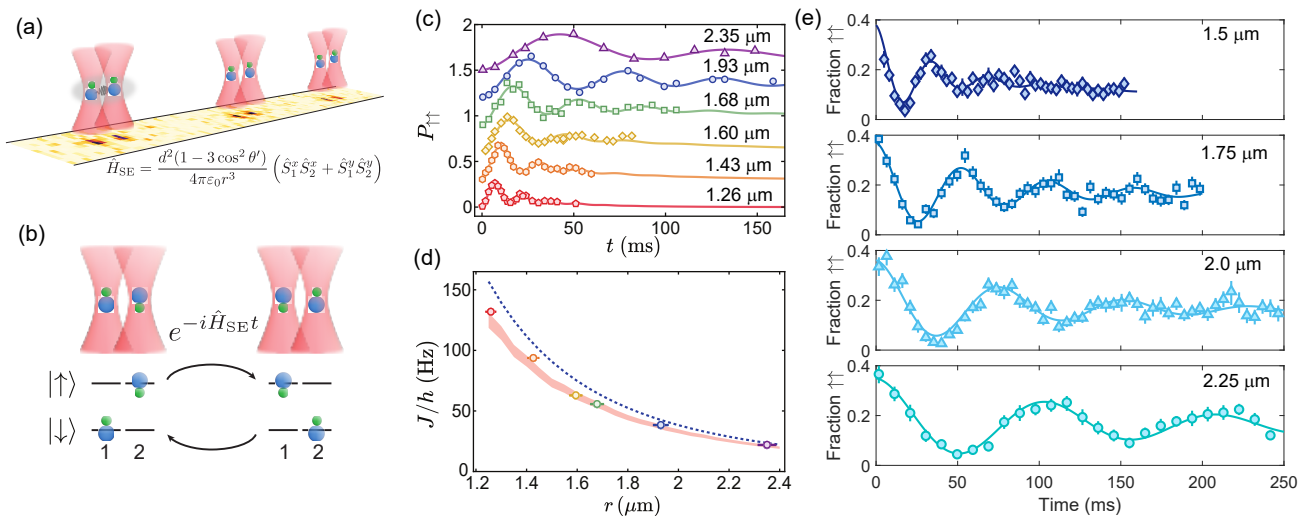


FIG. 3. Entangling gate between CaF molecules using dipolar spin-exchange interactions. (a) Single molecules in a tweezer array with dipolar interactions H_{SE} . (b) Exchange of rotational excitation induced by H_{SE} . (c) Spin-exchange oscillations at various tweezer separations, from [123]. (d) Points: measured interaction strength versus separation; Dashed line: zero-temperature theory; Red band: theory at the finite temperature of the molecules. (e) Spin-exchange oscillations at various tweezer separations, from [124]. (a)-(d) adapted from [123], (e) adapted from [124], with permissions from the authors.

low the theoretical prediction once the temperature of the molecules is accounted for. Figure 3(e) shows another set of spin-exchange oscillations, from Ref. [124]. By choosing $J_{\perp}t/\hbar = \pi$, the initial state $|\uparrow\uparrow\rangle$ is converted to the maximally entangled Bell state $\frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle + i|\downarrow\downarrow\rangle)$. The Bell state fidelity, an important figure of merit for quantum computing, was found to be ≈ 0.8 in both experiments, after correcting for state preparation and measurement errors. The fidelity is limited by motional dephasing of the spin-exchange oscillations, since the interaction energy $\propto \langle r_{ij}^{-3} \rangle$ differs for each motional state, and the experiments were done using molecules in thermal states with a broad motional-state distribution. Sideband cooling to the ground state [135–137] will eliminate motional dephasing and also eliminate single-particle decoherence arising from non-uniform differential light shifts.

D. Scaling up

Tweezer arrays can be scaled up to many hundreds of molecules, but this brings challenges. It will be necessary to control the tweezer separation with high precision and reproducibility throughout the array, maintain single-site addressability, avoid cross-talk and next-nearest neighbor interactions, and ensure the uniformity of the trap intensity and the addressing fields. In this context, it will be important to design protocols that are robust to inhomogeneous intensity and other non-uniformities.

V. OUTLOOK AND NEW DIRECTIONS

A. Controlling collisions and quantum degeneracy

Understanding and controlling ultracold collisions is critical for studying molecular gases and cooling them to quantum degeneracy. To date, experiments in optical traps have shown molecular collisions lead to loss for both reactive and non-reactive species [2, 4, 9, 138–142]. Although not fully understood [143], these losses are attributed to the transient formation of a collision complex [144, 145] which subsequently absorbs photons from the optical trap leading to the observed loss [146–148]. To circumvent this problem, several shielding methods [65] have been developed that exploit the dipolar interactions between molecules to engineer a repulsive barrier that prevents the molecules from reaching short range where the loss occurs. These methods include confining the molecules in 2D with their dipoles aligned perpendicular to the plane of confinement [149], using a dc electric field tuned to a level crossing within the Stark-shifted rotational levels of the molecule pair [150] and dressing with strong microwave fields [151–153].

The dipolar interactions responsible for shielding collisions from loss can also increase the rate of elastic collisions. This is ideal for evaporative cooling, as spectacularly demonstrated with KRb [68, 70] and NaK [75] to obtain the first quantum degenerate Fermi gases of molecules. Microwave shielding has recently been demonstrated for bosonic NaCs [154] and NaRb [155] molecules, resulting in the very recent first molecular Bose-Einstein condensates [156]. This opens new avenues, including dipolar droplets and supersolids [157], and provides a

new route to the high-filling molecules in optical lattices for quantum simulation. Already it is evident that techniques to shield molecules from loss and to control their interactions will become an intrinsic feature of future experiments.

B. Beyond qubits: qudits and synthetic dimensions

Molecules offer internal states across a vast range of energy scales: hyperfine, rotational, vibrational, and electronic. As Fig. 4(a) illustrates for RbCs, hundreds of long-lived states are accessible with microwave fields in just a handful of rotational levels. Finding ways to harness this wealth of states is an emerging research area.

For quantum computing, Ref. [162] proposed encoding qudits of dimension $d = 4$ in the hyperfine states of a single rotational level of either RbCs or CaF molecules, and presented a protocol to implement the Deutsch algorithm. Ref. [163] considered quantum error-correcting codes encoding quantum information in rotational-state superpositions, demonstrating theoretically robustness to relevant noise sources.

An exciting direction in quantum simulation is to harness the internal structure to create *synthetic dimensions*, in which motion in internal states is engineered to mimic motion in real spatial dimensions [60, 62]. For example, sites of a lattice can be mapped onto the rotational states, with coherent “tunneling” between sites controlled using microwave fields [61]. The single-particle Hamiltonian is highly tunable: lattice geometry, tunneling rates (including gauge fields), and on-site potentials can all be programmed (see Fig. 4(b) for examples). Moreover, with interactions, even the simplest one-dimensional synthetic dimension configuration is predicted to give rise to several phases of matter, and quantum and thermal phase transitions [164–166].

C. Increasing the interaction strength

Although typical dipolar interaction strengths between molecules of $\sim 0.1 - 10$ kHz suffice for high-fidelity computation and simulation, access to stronger interactions is desirable. One way to achieve this is to use pairs of molecules in opposite spin states together with the state-dependent optical tweezers, in order to control the spacing of the molecules on a sub-wavelength scale. Ref. [134] examined this, concluding that 2-3 orders of magnitude increase in interaction strength is feasible.

An alternative approach is to use Rydberg atoms to mediate interactions between molecules [159, 160, 167–169]. Rydberg atoms have transitions throughout the microwave domain, which an electric or magnetic field can bring into resonance with a rotational transition in the molecule. The resulting resonant atom-molecule DDI can be much stronger than the interaction between

molecules because of the large (typically \sim kD) transition dipole moment of the Rydberg atom. If the atom is placed between a pair of molecules, as illustrated in Fig. 4(c) & (d), it can mediate a strong dipolar interaction between the pair. Two recent studies [159, 160] have examined different protocols that exploit this mediated interaction to entangle molecules. In both cases, a gate duration of $\sim 1 \mu$ s and fidelity of 99.9% look achievable. Very recently, the first observation of Rydberg blockade due to the charge-dipole interaction between an atom and a polar molecule has been reported [127]. The authors used species-specific optical tweezers to position a single RbCs molecule around 300 nm from a single Rb atom, sufficiently close to cause blockade of the transition to the Rb(52s) Rydberg state. Extensions of these experiments to perform non-destructive read-out of the molecule [168, 169] and to engineer entanglement [159, 160, 167, 169] are eagerly anticipated.

D. Different species

To date, research into ultracold molecules has mostly focused on diatomic molecules. However, molecular diversity is enormous and attention is rightly turning to other species, including organic molecules and polyatomic species [170]. Remarkably, the highly diagonal Franck-Condon factors needed for laser cooling can be found in a wide range of complex molecules. The common feature of these molecules is a ligand attached to an atom or diatomic species that acts as the optical cycling center. Already, magneto-optical trapping, sub-Doppler cooling and optical trapping of CaOH has been demonstrated [18, 171] and many other species are under investigation (for a recent review see [172]). In parallel, efforts are underway to extend the range of molecular species formed by associating ultracold atoms. Interspecies Feshbach resonances have been observed in several mixtures of alkali atoms with alkaline-earth-like atoms [173–175], although molecule formation has thus far proved elusive. In contrast, recent progress on ^{161}Dy - ^{40}K [176] and ^6Li - ^{53}Cr [177] mixtures looks promising. Evidence for the association of triatomic molecules in ultracold mixtures of ^{40}K atoms and ground-state $^{23}\text{Na}^{40}\text{K}$ molecules has been reported [178]. Recently, “field-linked resonances” have been observed in collisions between microwave-dressed $^{23}\text{Na}^{40}\text{K}$ molecules [76], opening up a route to coherent “electroassociation” of tetramer molecules [179, 180].

Cooling new molecules to ultracold temperatures will allow new opportunities stemming from their different structure and interactions with external fields. For example, polyatomic molecules exhibit parity doublets where quantum states of opposite parity lie close in energy. This near degeneracy means that the states can be mixed with a small electric field, typically $\lesssim 100$ V/cm, leading to linear Stark shifts and making it exceptionally easy to produce laboratory frame dipole moments. These properties have stimulated a number of proposals advocating

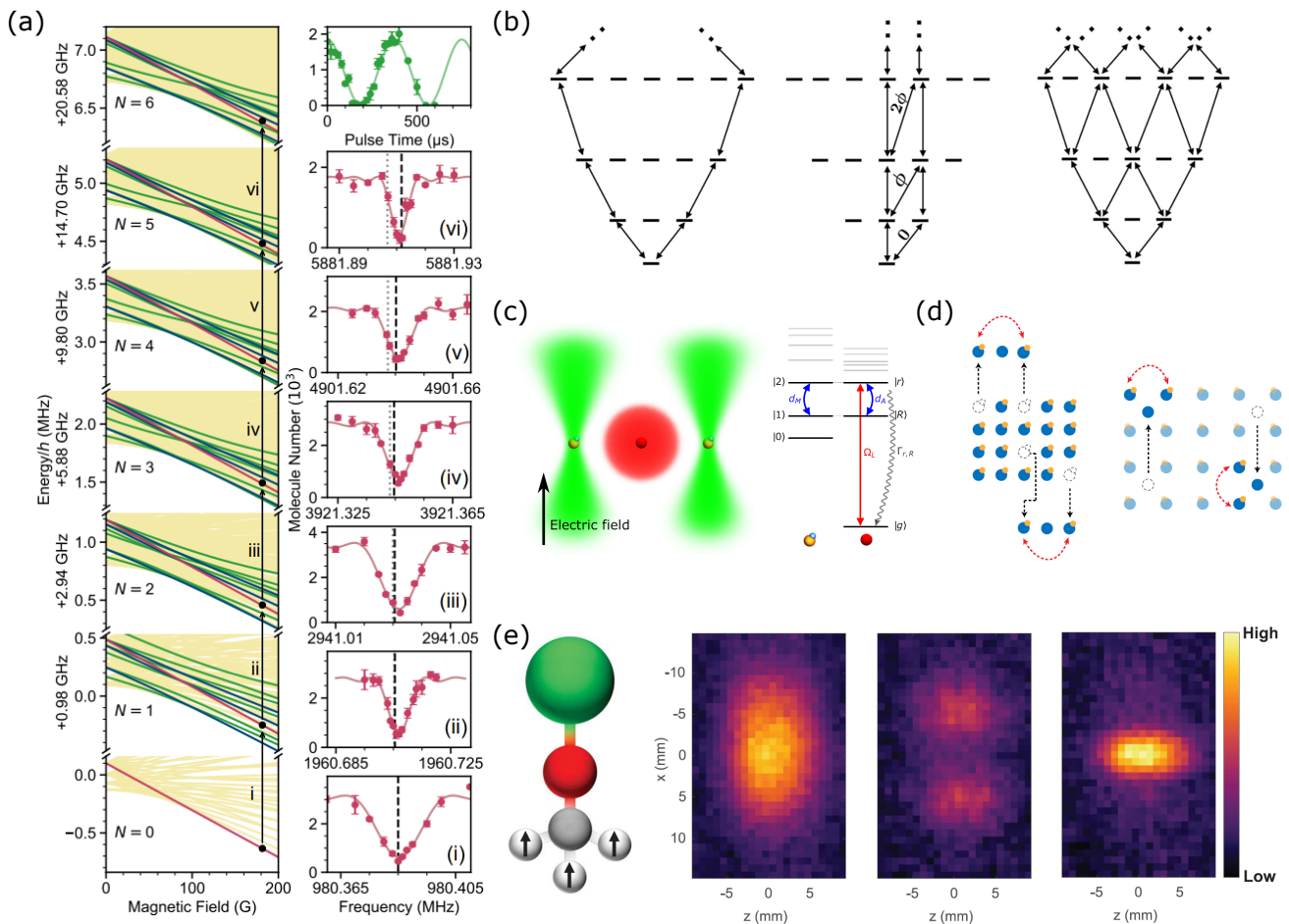


FIG. 4. Future research directions. (a) Ultracold molecules offer a rich internal structure of long-lived rotational and hyperfine states easily accessible with microwave fields, illustrated for $^{87}\text{Rb}^{133}\text{Cs}$ up to $N = 6$ prepared in the hyperfine ground state of $N = 0$ at ~ 181 G. The microwave spectra (right) are labelled to match the transitions (left). The top panel shows Rabi oscillations on the $N = 5 \rightarrow 6$ transition. The nuclear spins of ^{87}Rb ($i = \frac{3}{2}$) and ^{133}Cs ($i = \frac{7}{2}$) result in $32 \times (2N + 1)$ hyperfine states per total angular momentum, N . (b) Rotational states can encode synthetic dimensions with the “tunneling” rates controlled by microwave fields (arrows). Examples include (left to right) a 1D chain with periodic boundaries, a two-leg ladder with a gauge field and a square lattice with open boundaries. (c) & (d) Interfacing molecules with Rydberg atoms may increase the effective interaction strength, allowing fast Rydberg-mediated gates between molecules. Moving molecules and/or atoms using optical tweezers allows a scalable quantum computing architecture. (e) Cooling new molecules will open new opportunities, such as the laser cooling of CaOCH_3 symmetric-top molecules: the unperturbed beam (left), Sisyphus heating (centre) and Sisyphus cooling (right). (a) Adapted from [158], (b) adapted from [61], (c) adapted from [159], (d) adapted from [160] and (e) adapted from [161], with permissions from the authors.

the use of symmetric-top molecules for quantum information processing [181–183], and laser cooling of CaOCH_3 has already been reported (Fig. 4(e)) [161].

For quantum simulation, Ref. [184] highlighted the correspondence between the linear Stark shifts of a symmetric-top molecule and a magnetic dipole in a magnetic field. They proposed that these molecules in lattices could be used to simulate spin models with arbitrary integer spin and in later work that they may be used to study other quantum spin models, such as the XYZ model [185].

VI. CONCLUSION

The field of ultracold molecules has seen rapid progress over the last decade. Through the combined effort of many research groups, the understanding and control of molecular systems has now reached a point where scientific applications beyond AMO physics are emerging. We have presented notable highlights in the realms of quantum simulation and quantum computation. However, this is undoubtedly just the beginning and we can expect many exciting developments in the near future. Looking further ahead, as the number of molecular species far exceeds the number of atomic elements, perhaps we should

expect a day to come when molecules dominate the field of ultracold physics.

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