

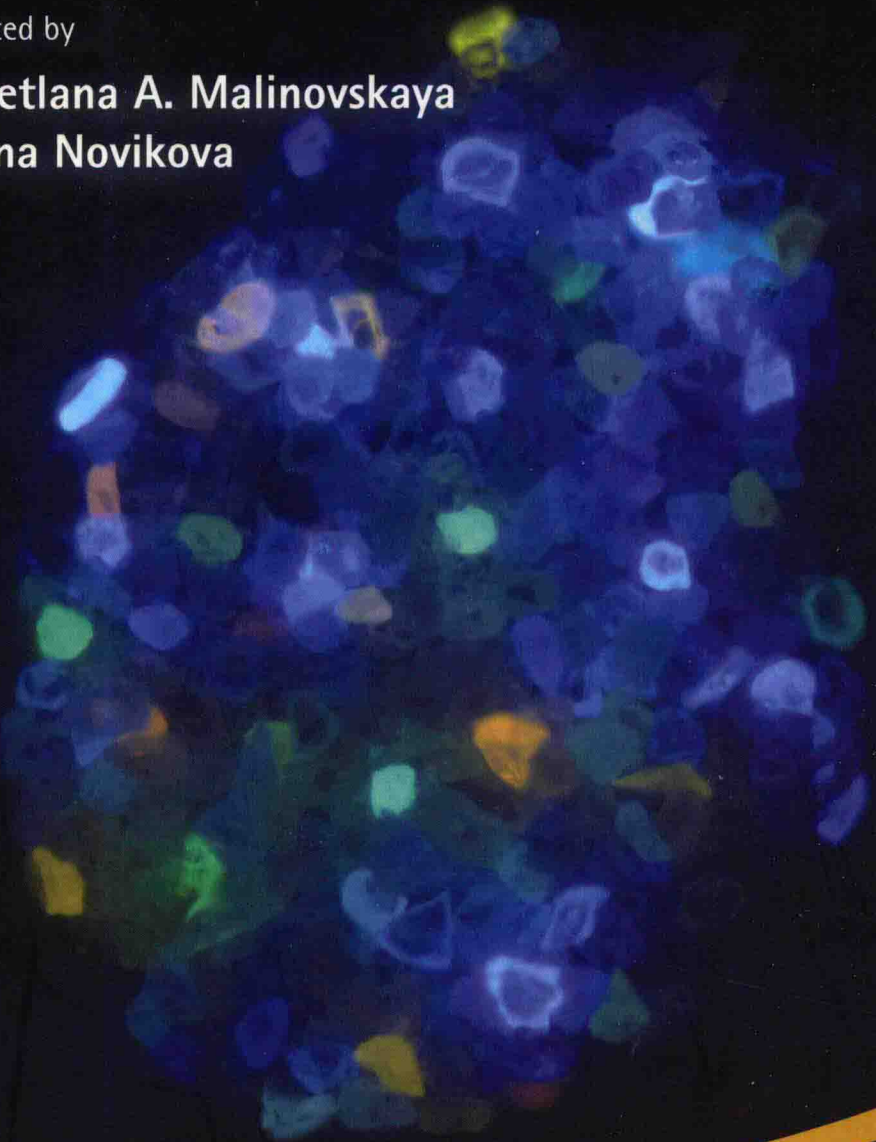
# FROM ATOMIC TO MESOSCALE

*The Role of Quantum Coherence in Systems of Various Complexities*

Edited by

**Svetlana A. Malinovskaya**

**Irina Novikova**



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# FROM ATOMIC TO MESOSCALE

*The Role of Quantum Coherence in Systems of Various Complexities*

## Foreword

The latest advancements and future directions of atomic, molecular and optical (AMO) physics and their vital role in modern sciences and technologies form the content of this volume. The chapters are devoted to studies of a wide range of quantum systems, with an emphasis on understanding of quantum coherence and other quantum phenomena originated from light-matter interactions. The book intends to survey the current research landscape and to highlight major scientific trends in AMO physics as well as those interfacing with interdisciplinary sciences. The volume may be particularly useful for young researchers working on establishing their scientific interests and goals.

The content of the book was inspired by presentations at the workshop from Atomic to Mesoscale, which was held at the Institute for Theoretical Atomic, Molecular and Optical Physics (ITAMP) in March, 2014. All talks are available on the ITAMP website.

The book is organized in six sections; most of them are a synergy of theory and experiment. Section on “Collective phenomena and long-range interactions in ultracold atoms and molecules” describes the realization of quantum magnetism with ultracold molecules in an optical lattice and reviews the experimental and theoretical progress in this field. This section provides an overview of quantum optics in high-density, cold atomic matter in which disorder-driven electromagnetic interactions can develop strongly correlated character. This section also discusses spin dynamics that occur within spinor Bose-Einstein gases, composed of bosonic atoms. It highlights the advantages of spin-sensitive imaging for understanding and utilizing such dynamics.

The section on “Atom-like coherent solid state systems” introduces the nitrogen vacancy (NV) center, being a quantum defect in diamonds and belonging to cutting edge scientific and technological objects. The imaging of biological cells, cellular temperature gradient measurement and nanoscale electric field sensing are discussed as applications of NV-diamond quantum sensors. This section also presents the studies on self-assembled quantum dots interacting with light to investigate coherence properties and confirm entanglement between a spontaneously emitted photon and an electron spin qubit confined to the quantum dot. The combination of the

solid-state nature of quantum dots and their close approximation to atomic systems makes them an attractive platform for quantum information-based technologies.

The section on “Coherent nanophotonics and plasmonics” emphasizes the central role of quantum photonics in the development of quantum computation and communication technologies owing to the high transmission capacity and outstanding low-noise properties of photonic information channels. Theoretical and experimental studies of single-photon sources are presented focusing on the NV center in a diamond placed near a hyperbolic metamaterial. This section also presents the studies of the optical properties of hybrid systems in the linear regime accompanied by strong coupling between the molecules and the surface plasmon polaritons.

The section on “Fundamental Physics” reports on a boson-sampling computational model as the key to a non-universal quantum computer and discusses the feasibility of building a boson-sampling device using existing technology. Moreover, this section introduces a new approach to quantum amplification by superradiant emission of radiation (QASER).

The section on “Ultrafast dynamics in strong laser fields” describes an approach to create attosecond magnetic fields by making use of few cycle circularly polarized attosecond UV pulses produced by HHG. Those pulses generate electronic currents in molecular media on an attosecond time scale, which are the sources of attosecond magnetic pulses. Another chapter of this section reviews theoretical methods to study and predict the response of fullerenes with interior atoms to UV and soft x-ray radiation. Various effects of coherence are discussed here as they are manifested in plasmon resonances, many-body correlations and Auger-intercoulombic hybrid multicenter decays of inner-shell holes.

The section “Ultracold chemistry” presents theoretical investigations of ultracold chemical reactions at a single partial wave level such as barrierless chemical reactions. Studies of highly efficient energy transfer processes are introduced involving rotational and vibrational energy exchange in ultracold molecular collisions.

We hope this overview will be useful and inspirational to many scientists. We would like to acknowledge ITAMP and Dr. Hossein Sadeghpour for support in carrying out the workshop, which became so important in the preparation of the volume. We are grateful to Dr. Christian Buth for technical help at the initial stage of the project. We would also like to thank Dr. Duncan Steel for insightful discussions regarding the volume proposal.

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I

Collective Phenomena and Long-Range  
Interactions in Ultracold Atoms  
and Molecules



## Chapter 1

### Quantum Magnetism with Ultracold Molecules

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This article gives an introduction to the realization of effective quantum magnetism with ultracold molecules in an optical lattice, reviews experimental and theoretical progress, and highlights future opportunities opened up by ongoing experiments. Ultracold molecules offer capabilities that are otherwise difficult or impossible to achieve in other effective spin systems, such as long-ranged spin-spin interactions with controllable spatial and spin anisotropy and favorable energy scales. Realizing quantum magnetism with ultracold molecules provides access to rich many-body behaviors, including many exotic phases of matter and interesting excitations and dynamics. Far-from-equilibrium dynamics plays a key role in our exposition, just as it did in recent ultracold molecule experiments realizing effective quantum magnetism. In particular, we show that dynamical probes allow the observation of correlated many-body spin physics, even in polar molecule gases that are not quantum degenerate. After describing how quantum magnetism arises in ultracold molecules and discussing recent observations of quantum magnetism with polar molecules, we survey prospects for the future, ranging from immediate goals to long-term visions.

#### 1. Introduction

The realization of a Bose-Einstein condensate (BEC) in an ultracold, dilute gas of alkali atoms<sup>1-3</sup> was a landmark achievement in several respects. For one, the production of an atomic BEC required significant technical advances in cooling and trapping atoms with electromagnetic radiation, as well as evaporative cooling. In addition, atomic BECs provided the most direct evidence for a many-body phenomenon predicted more than 80 years prior. The fact that nearly all aspects of the atomic system are amenable to experimental control not only enabled the realization of this new state of matter, but also facilitated the study of its properties out of equilibrium, such as the dynamics of vortices<sup>4</sup> and solitons.<sup>5</sup> As we shall see, ultracold molecules parallel this, providing entirely new phases of matter and non-equilibrium behaviors that are otherwise unrealized in ultracold matter.

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Following the realization of a BEC the field of many-body physics with ultracold atoms has grown steadily,<sup>6</sup> including degenerate fermionic gases<sup>7</sup> as well as many non-alkali species.<sup>8–11</sup> A burgeoning subfield of research involves optical lattices standing wave arrangements of laser light that form a periodic potential for atoms or molecules.<sup>12</sup> Such a periodic potential mimics the crystal potential felt by electrons in a solid, enabling the atoms to “simulate” the behavior of interacting electrons in a crystal lattice. The power of the atom-electron analogy comes from the fact that characteristics of the atomic system, such as lattice geometry, degree of disorder, and strength of interactions, are all highly tunable. This enables the atoms to behave as a *quantum simulator*, a quantum system that behaves analogously to another system (which may be much harder to microscopically control or measure),<sup>13</sup> has already led to many spectacular observations, such as the transition from a weakly-interacting gas to a Mott insulator for both bosonic<sup>14</sup> and fermionic<sup>15,16</sup> atoms.

In spite of the successes of ultracold atom experiments, some phenomena remain difficult to manifest and probe in cold atoms. One such phenomenon is quantum magnetic interactions between effective spins. Quantum magnetism, which studies the many-body physics of coupled spins, is of key importance in condensed matter physics (see Sec. 2). In atomic realizations of quantum magnetism, usually the “spin” is formed from some discrete set of internal states, for example hyperfine sublevels. The reason why realizing effective magnetic interactions between such spins is difficult, as Sec. 2.2 describes in more detail, is that the dominant interactions between neutral atoms are short ranged; this requires any effective non-local spin-spin interactions between internal states to be mediated by motion. Hence, magnetic correlations become visible only when the motional temperature is less than the effective spin-spin coupling energy. To date, such temperatures are extraordinarily difficult to reach.

Ultracold ground state molecules are a newly realized platform for quantum magnetism in which the magnetism arises in a qualitatively distinct way from atoms, and this underlies the molecules’ many favorable qualities. As opposed to atoms, polar molecules have strong, long-range electric dipole-dipole interactions<sup>a</sup>. The basic idea is to encode effective degrees of freedom in long-lived, low-lying, and easily accessible internal degrees of freedom such as rotational and vibrational modes. Dipole-dipole interactions can mediate coupling between these effective spins even when molecules are pinned in a lattice (i.e. when their tunneling is completely suppressed). One consequence is that polar molecules can be used to study far-from-equilibrium quantum magnetism even in non-degenerate quantum gases. Such far-from-equilibrium dynamics has been observed in ultracold polar molecule experiments and will play a central role in our exposition. Additionally, taking a broad view of molecular diversity and experimental constraints such as temperature and lattice-scale probe resolution, we provide an overview of the “quantum

<sup>a</sup>Homonuclear molecules are not polar, and so are not amenable for simulating quantum magnetism in the fashion discussed in this work.

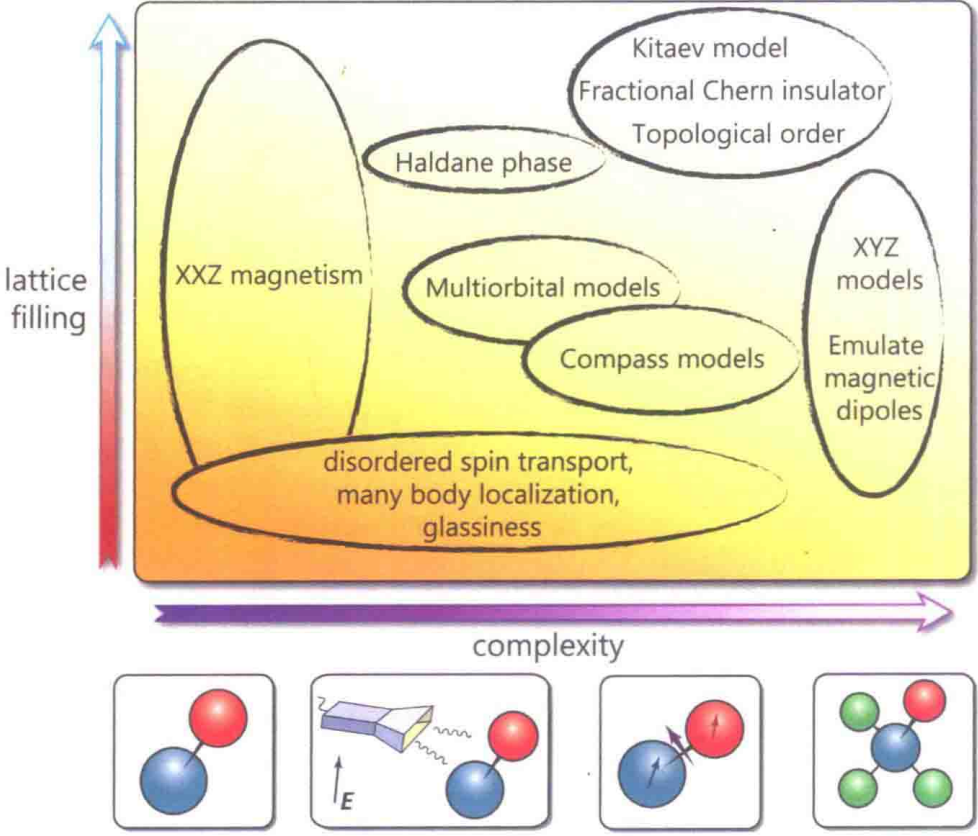


Fig. 1. Regimes of quantum magnetism in ultracold ground state polar molecules in optical lattices, encompassing ongoing experiments (lower left) and directions being pursued (upper right). The vertical “lattice filling” axis represents the fraction of occupied lattice sites, that is the molecule density relative to the lattice spacing. Higher fillings generally correspond to lower motional entropies and temperatures. Current experiments reach filling fractions in the range of 10%-20%. Interesting physics exists in this regime, but exciting prospects also occur as one increases the filling fraction towards unity. The horizontal “complexity” axis represents complexity in two senses: the molecules’ degrees of freedom and the experimental requirements to harness them. Diverse, rich, and novel phenomena at the forefront of modern quantum many-body physics occur in all of the indicated regimes.

simulation landscape” of quantum magnetic phenomena achievable with ultracold molecules, and summarize it in Fig. 1. This figure shows the new regimes of quantum magnetism that become available both as the motional entropy and temperature decrease (vertical axis) and as the complexity of the internal molecular structure increases (horizontal axis). This figure will be discussed in more detail in Sec. 5.

Our paper is organized as follows. Sec. 2 derives effective spin Hamiltonians describing the internal state dynamics of polar molecules in optical lattices (Sec. 2.1) and neutral atoms in optical lattices for comparison (Sec. 2.2), focusing on the simplest scenarios. Sec. 3 describes recent experiments in which effective quantum magnetism has been experimentally probed via far-from-equilibrium dynamics, as well



as new theoretical tools that were developed to verify and understand the experimental observations. In Sec. 4 we explore molecules with complex internal structure, review methods of producing ultracold molecules, and go over basic molecular structure. In Sec. 5, we identify future directions for quantum magnetism with ultracold molecules, considering both advances in experimental technology and the structural complexity of molecules on the experimental horizon. Finally, in Sec. 6, we conclude.

## 2. Quantum Magnetism with Ultracold Molecules and Atoms

Exploring quantum magnetism with ultracold matter is a particularly fruitful direction of research, in part because quantum magnetic phenomena lie at the core of condensed matter physics. Moreover, despite the apparent simplicity of many models of quantum magnetism, these models are in general extraordinarily hard to solve with classical resources. This makes them excellent candidates for “quantum simulation” with ultracold systems. Although quantum magnetism is a vast field that is well beyond the capacity of this review to cover, we mention here some of the broad ideas that motivate its study. More complete reviews and introductions can be found, e.g., in Refs. 17–19.

One reason for the intense study of quantum magnetism is its relevance to materials and experimental phenomena – for example, antiferromagnets, multiferroic materials, spin glasses, and spin nematics – and the frequent proximity of quantum magnetism to unconventional superconductivity. Another motivation is the numerous *exotic* phenomena that have been theoretically predicted, including topologically ordered phases and (algebraic) spin liquids. These harbor physics which cannot be described within the “Landau paradigm” of symmetry breaking, as is also the case with the fractional quantum Hall effect. Observing a broader range of such phenomena, which lie outside of conventional classification, would clearly deepen our knowledge of quantum many-body physics. Finally, an understanding of quantum magnetism can have a great impact in advancing current technology including better and faster hard drives, computers, and spintronic devices.

Quantum magnetism in the solid state usually refers to interactions between electron spins localized in a crystal lattice. As the Coulomb interaction, which provides the microscopic interaction between electrons, is spin-independent, interactions between spins should be interpreted as *effective* interactions which arise from Coulomb interactions in conjunction with Fermi statistics, i.e. the required antisymmetry of electrons under exchange. In Sec. 2.2, we show how such effective magnetic interactions arise from particles with short-range interactions when they can tunnel in a lattice. This spin interaction mechanism, known as superexchange, is the most common mechanism by which effective magnetic phenomena arise in cold *atomic* gases loaded in optical lattices. Before we discuss the superexchange mechanism, however, Sec. 2.1 describes the simplest example of effective quantum magnetism mediated by dipole-dipole interactions in polar molecules, the main focus



of this review. As we shall see, the resulting models for dipole-mediated quantum magnetism and the superexchange mechanism are very similar, even though the physical mechanism is very different. Finally, Sec. 2.3 discusses control and experimental consequences of the terms appearing in the effective spin models.

### 2.1. *Effective Magnetism with Polar Molecules*

Let us now consider how effective quantum magnetism arises for polar molecules.<sup>20–23</sup> For clarity, we discuss the simplest manifestation in this section before discussing how more complex magnetic interactions may be engineered in Secs. 4.2 and 5. Our starting point is shown schematically in the top left panel of Fig. 2. Here, molecules are pinned in a deep optical lattice with exactly one molecule per lattice site. By “pinned” we mean that molecules do not move between lattice sites on the timescales of an experiment. We now wish to encode an effective spin-1/2 in the internal degrees of freedom of the molecule. Considering  $^1\Sigma$  molecules, in which there are no unpaired spins or orbital angular momentum<sup>b</sup>, and neglecting hyperfine structure, the lowest-lying degrees of freedom to encode spin in are the rotational degrees of freedom<sup>c</sup>. The rotational states are described by a rigid rotor Hamiltonian<sup>24</sup> and can be labeled by  $|NM_N\rangle$ , where  $N$  is the rotational angular momentum quantum number and  $-N \leq M_N \leq N$  is the projection of the rotational angular momentum along a space-fixed quantization axis. In the absence of external fields, the rotational energy spectrum is  $E_{NM_N} = B_N N(N+1)$ , where  $B_N$  is called the rotational constant and is inversely proportional to the moment of inertia of the molecule. Typical rotational constants are a few GHz, which is much larger than the dipolar interaction energies of molecules at typical ( $\sim 500$  nm) optical lattice spacings, and also much larger than ultracold temperatures. These facts, together with the anharmonic spectrum and very long ( $> 10$  s) lifetimes of rotational excitations, imply that the number of molecules in each of the excited rotational states is conserved over the timescale of an experiment<sup>d</sup>.

The  $(2N+1)$ -fold degeneracy of rotational excited states makes isolating a pair of rotational levels in which to encode a spin-1/2 challenging, and so we would like to split this degeneracy. One way to do so is to introduce a DC electric field  $\mathbf{E}_{\text{DC}}$ ; the resulting splitting is illustrated in Fig. 2. The key feature that we need in order to understand the emergence of quantum magnetism is that states such as  $|0, 0\rangle$  and  $|1, 0\rangle$  in Fig. 2, are now well-isolated from all other states, and so we can use them to encode a spin-1/2. The details of the coupling and notation will be explained below.

<sup>b</sup>A review of molecular structure and terminology is given in Sec. 4.2.

<sup>c</sup>In fact, these non-rotational degrees of freedom can sometimes also be neglected in molecules with more complex molecular structures under appropriate circumstances.

<sup>d</sup>Provided, of course, that rotational excitations are not generated by external means, e.g. by a microwave field.

Now that we have isolated an effective spin-1/2, we investigate the effect of dipole-dipole interactions within this subspace of states. The dipole-dipole interaction between molecules  $i$  and  $j$  is

$$\hat{H}_{\text{DDI}} = \frac{\hat{\mathbf{d}}_i \cdot \hat{\mathbf{d}}_j - 3 (\hat{\mathbf{d}}_i \cdot \mathbf{e}_r) (\hat{\mathbf{d}}_j \cdot \mathbf{e}_r)}{r^3}, \quad (1)$$

where  $\mathbf{e}_r$  is a unit vector connecting molecules  $i$  and  $j$ ,  $r$  is the distance between these molecules, and  $\hat{\mathbf{d}}_i$  is the dipole operator of molecule  $i$ . Within the subspace of states  $\{|\downarrow\rangle, |\uparrow\rangle\} \equiv \{|0,0\rangle, |1,0\rangle\}$  forming our spin-1/2, and in the limit of the interaction being much smaller than the rotational splitting, the interaction Eq. (1) is simple; for example

$$\hat{H}_{\text{DDI}} |\uparrow\downarrow\rangle = a |\uparrow\downarrow\rangle + b |\downarrow\uparrow\rangle \quad (2)$$

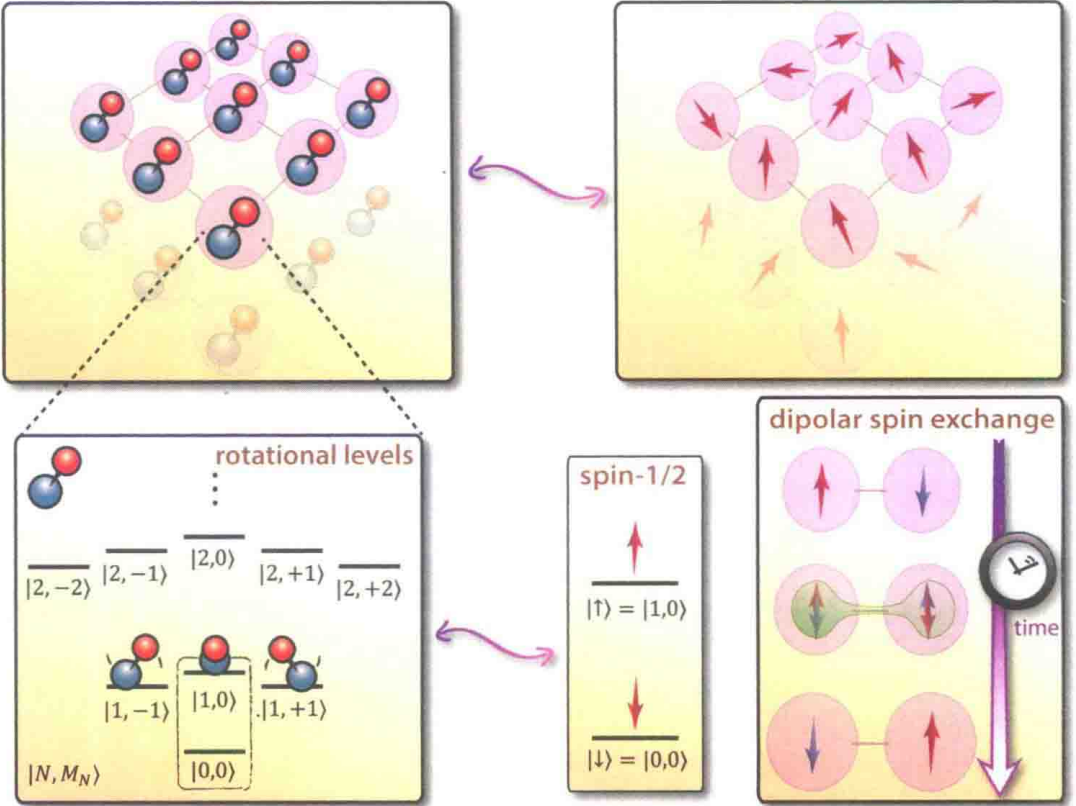


Fig. 2. Quantum magnetism of ultracold molecules in a lattice. Molecules in a deep lattice (top left) realize lattice spin models (top right), when the lattice is deep enough to *suppress* tunneling. The spin degree of freedom is encoded in rotational states of the molecule (bottom left). Two types of interactions occur naturally: a “spin exchange” interaction that exchanges pairs of spin states (illustrated, bottom right) and, in the presence of a dc electric field, an “Ising” interaction that splits the energies of aligned and anti-aligned pairs of spins [see Eq. (17)]. Both processes are capable of correlating and entangling spins.

with  $a = \langle \uparrow\downarrow | \hat{H}_{\text{DDI}} | \uparrow\downarrow \rangle$  and  $b = \langle \downarrow\uparrow | \hat{H}_{\text{DDI}} | \uparrow\downarrow \rangle$ . Processes that change the total magnetization, such as  $|\uparrow\downarrow\rangle \rightarrow |\uparrow\uparrow\rangle$ , are energetically far off-resonant and therefore negligible. This implies that the spin-spin interaction of the molecules is

$$\hat{H}_{ij} = \left[ \frac{J_{\perp}(\hat{\mathbf{d}}_i, \hat{\mathbf{d}}_j, \mathbf{r}_i - \mathbf{r}_j)}{2} (\hat{S}_i^+ \hat{S}_j^- + \text{h.c.}) + J_z(\hat{\mathbf{d}}_i, \hat{\mathbf{d}}_j, \mathbf{r}_i - \mathbf{r}_j) \hat{S}_i^z \hat{S}_j^z \right] \quad (3)$$

with  $S_i^{\pm, z}$  the usual spin-1/2 operators acting on molecule  $i$ . We will derive below the forms of  $J_{\perp}(\hat{\mathbf{d}}_i, \hat{\mathbf{d}}_j, \mathbf{r}_i - \mathbf{r}_j)$  and  $J_z(\hat{\mathbf{d}}_i, \hat{\mathbf{d}}_j, \mathbf{r}_i - \mathbf{r}_j)$  and their dependence on electric field and choice of rotational states, as well as determine additional single spin terms that are omitted in Eq. (3) [see Eq. (11) for the final result].

However, before giving a more complete derivation of Eq. (3) and a determination of the coefficients in it, we first will describe how an electric field may be used to achieve the level splitting required to energetically isolate the spin-1/2 degree of freedom. This also provides useful background for how the interactions in Eq. (3) may be manipulated with electric fields.

The coupling Hamiltonian of the molecule's dipole operator,  $\hat{\mathbf{d}}$ , with the external field is  $-\hat{\mathbf{d}} \cdot \mathbf{E}_{\text{DC}}$ . We take  $\mathbf{E}_{\text{DC}}$  to set the space-fixed  $z$  axis,  $\mathbf{E}_{\text{DC}} = E_{\text{DC}} \mathbf{e}_z$  and thus the coupling Hamiltonian has matrix elements

$$\langle N' M_{N'} | -\hat{d}_z E_{\text{DC}} | N M_N \rangle = -E_{\text{DC}} \delta_{M_{N'}, M_N} \langle N' M_{N'} | \hat{d}_0 | N M_N \rangle \quad (4)$$

where

$$\langle N' M_{N'} | \hat{d}_p | N M_N \rangle = d (-1)^{M_{N'}} \sqrt{(2N' + 1)(2N + 1)} \begin{pmatrix} N' & 1 & N \\ -M_{N'} & p & M_N \end{pmatrix} \begin{pmatrix} N' & 1 & N \\ 0 & 0 & 0 \end{pmatrix} \quad (5)$$

are the matrix elements of the dipole operator in the basis of rotational states. In Eq. (5),  $(\dots)$  is a  $3j$ -symbol and  $\hat{d}_{\pm 1} = \mp(\hat{d}_x \pm i\hat{d}_y)/\sqrt{2}$  and  $\hat{d}_0 = \hat{d}_z$  are spherical components of the dipole operator. Only the  $p = 0$  component is used above, but the  $p = \pm 1$  components will be useful to us later. The electric field mixes rotational states while preserving their projection  $M_N$  on the field axis. Because the electric field does not cause level crossings, we can still label the eigenstates of rotation in the presence of a DC field with  $|N M_N\rangle$ , where  $N$  is now interpreted as a label corresponding to the number of rotational quanta if the field were to be ramped adiabatically to zero. The energies of these states in weak fields, calculated to lowest order in the small parameter  $\beta_{\text{DC}} = dE_{\text{DC}}/B_N$ , are

$$E_{N M_N}/B_N = N(N + 1) + \frac{\beta_{\text{DC}}^2}{2} \frac{N(N + 1) - 3M_N^2}{(2N - 1)(2N + 3)N(N + 1)}. \quad (6)$$

As shown in Fig. 2, all states with the same value of  $|M_N|$  remain degenerate, and are separated from all other states with the same value of  $N$  by an energy  $\sim \beta_{\text{DC}}^2 B_N$ .

Now turning to the full derivation of how the dipole-dipole interaction, Eq. (1), projects onto the spin-1/2 degree of freedom we note that the dipolar interaction is the contraction of two rank-two tensors, one acting on the internal state of the