A molecular crystal with tunable disorder: Controlled localization of rotational and spin excitons

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Quantum Simulation

Design *simple controllable* systems with some of the same properties as complex quantum systems, such as solid-state crystals.
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This talk

I. Ultracold molecules on an optical lattice as a crystal with tunable exciton–impurity interactions

II. Ultracold molecules on an optical lattice as a crystal with tunable magnetic properties
Frenkel exciton

\[ \phi_n = |00\rangle_1 |00\rangle_2 \ldots |10\rangle_n |00\rangle_{n+1} \ldots |00\rangle_N \]

\[ \psi = \sum_n C_n \phi_n \]
Frenkel exciton

\[ \phi_n = |00\rangle_1 |00\rangle_2 \ldots |10\rangle_n |00\rangle_{n+1} \ldots |00\rangle_N \]

\[ \psi_k = \sum_n \frac{e^{ik \cdot r_n}}{\sqrt{N}} \phi_n \]
Dispersion Curves

\[ E(k) \text{ (in units of } 10^{-6} B) \]

\[ m_* < 0 \]

\[ |1,-1\rangle \rightarrow |1,0\rangle \rightarrow |1,1\rangle \rightarrow |0,0\rangle \]

\[ |1,-1\rangle \rightarrow |1,0\rangle \rightarrow |1,1\rangle \rightarrow |\alpha\rangle, |\beta\rangle \]

\[ |0,0\rangle \rightarrow |\gamma\rangle \]
Negative effective mass => negative refraction of EM field
$E(k)$ (kHz)

$E_{\perp x}$

$E_{|| x}$

$\alpha$, $\beta$

$\gamma$
Impurities

Pure Exciton Hamiltonian:

\[
H = \left( \sum_{n} E_0 B_n^\dagger B_n + \sum_{n} J_{mn} B_m^\dagger B_n \right)
\]
Impurities

One impurity:

\[
H = \sum_{n \neq 0} E_0 B_n^\dagger B_n + E_{imp} B_{n=0}^\dagger B_{n=0} + \sum_n J_{mn} B_m^\dagger B_n
\]

Scatterer with the strength = difference in transition energies:

\[
H = \left( \sum_n E_0 B_n^\dagger B_n + \sum_n J_{mn} B_m^\dagger B_n \right) + (E_{imp} - E_0) B_{n=0}^\dagger B_{n=0}
\]

Breaks translational symmetry \(\Rightarrow\) Mixes states with different \(k\)

\[
H = \sum_k E(k) B^\dagger(k) B(k) + \frac{V_0}{N} \sum_{k,q} B^\dagger(k) B(q)
\]
Tunable impurities

\[ \Delta E_{eg} \text{ (x10^4 MHz)} \]

\[ \sigma_{2D} \text{ (Å)} \]

\[ E \text{ (kV/cm)} \]

\[ E \text{ (mV/cm)} \]

- **CsF**
- **LiCs**
- **LiRb**

\[ k = 10^{-8} \text{ Å}^{-1} \]
\[ k = 10^{-6} \text{ Å}^{-1} \]
\[ k = 10^{-5} \text{ Å}^{-1} \]
Exciton – impurity Hamiltonian matrix

\[ \langle \hat{H}_0 \rangle_{q,k} = E(k) \delta_{k,q}, \]

\[ \langle \hat{W} \rangle_{q,k} = \frac{2\Delta J(a)}{N_{\text{mol}}} (\cos q \cdot a + \cos k \cdot a) \sum_{i_n=1}^{N_i} e^{i(q-k) \cdot i_n} \]

Off-diagonal disorder

Diagonal disorder

\[ \langle \hat{V} \rangle_{q,k} = \frac{V_0}{N_{\text{mol}}} \sum_{i_n=1}^{N_i} e^{i(q-k) \cdot i_n} \]
No diagonal disorder

Strong diagonal disorder

Diagonal disorder \sim off-diagonal disorder
\[ |\Psi(x)|^2 \left( \frac{1}{N_{\text{mol}}} \right) \]

- **No diagonal disorder**
- **Diagonal disorder \sim off-diagonal disorder**
- **Large diagonal disorder**
\[ |\Psi(x)|^2 \] 

\[ f(t) \]

\[ t (\mu s) \]

\[ |C(k)|^2 \]

\[ x (\text{a}) \]

\[ ka \]
Applications

• Time-domain quantum simulation of localization of quantum particles:
  timescale of Anderson localization
dynamics of exciton localization as a function of effective mass, exciton bandwidth, and exciton-impurity interaction strength
effect of disorder correlations on localization and delocalization

• Negative refraction of MW fields

• Controlled preparation of many-body entangled states of molecules

• Effects of dimensionality and finite size on energy transfer in crystals
Energy diagram of a $^2\Sigma$ diatomic molecule

How do electric fields affect spin relaxation?

- Induce couplings between the rotational levels ($N = 1$)
- Increase the energy gap between the rotational levels

Enhancement of spin relaxation

- **First-order Stark effect**

$^2\Sigma$ molecules

Energy diagram of a $^2\Sigma$ diatomic molecule
Energy diagram of a $^2\Sigma$ diatomic molecule

$^2\Sigma$ molecules

$\Delta E$

Spin-orbit interaction

Interaction potential

$J$
$^2 \Sigma$ molecules

Energy diagram of a $^2 \Sigma$ diatomic molecule
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Frenkel exciton

\[ \phi_n = |00\rangle_1 |00\rangle_2 \ldots |10\rangle_n |00\rangle_{n+1} \ldots |00\rangle_N \]

\[ \psi_k = \sum_n e^{ik \cdot r_n} \frac{1}{\sqrt{N}} \phi_n \]

\[ \Psi = \frac{1}{\sqrt{N_{\text{mol}}}} \sum_i C_i \Phi_i^S \]

\[ \Phi_i^S = |M_S = 1/2\rangle_{r_i} \prod_{j \neq i} |M_S = -1/2\rangle_{r_j}. \]
Frenkel exciton

\[ \phi_n = |00\rangle_1 |00\rangle_2 \ldots |10\rangle_n |00\rangle_{n+1} \ldots |00\rangle_N \]

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\[ \Phi^S_i = |M_S = 1/2\rangle_{r_i} \prod_{j \neq i} |M_S = -1/2\rangle_{r_j} \]

\[ \alpha |\uparrow\rangle |\downarrow\rangle + \beta |\downarrow\rangle |\uparrow\rangle \]
Applications

Crystal with tunable impurities:

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  timescale of Anderson localization
  dynamics of exciton localization as a function of effective mass, exciton bandwidth, and exciton-impurity interaction strength
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Optical lattice of magnetic molecules:

• Crystal with tunable magnetic properties, tunable spin waves

• Preparation of many-body entangled states of spin up-down pairs

• ???