Collective excitations of ultracold molecules trapped on an optical lattice

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Ultracold molecules on optical lattices = molecular crystals with unusual properties:

Intermolecular interactions are very weak, much weaker than the energy of rotational splitting in molecules.

Molecules are held in the crystal by optical field forces, not intermolecular interactions.
Is rotational excitation of a molecule a single-particle or collective excitation?
What can we do with molecules on a lattice that cannot be done with conventional crystals?
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

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

\[ k a \]

\[ m_* < 0 \]

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

\[ |0, 0\rangle \]

\[ |\alpha\rangle, |\beta\rangle \]

\[ |\gamma\rangle \]
What are rotational excitons good for?
Rotational excitons are controllable...
Excitons are sensitive to impurities ...

Exciton – impurity interactions can be controlled!
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:

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

Scatterer with the strength = difference in transition energies:

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

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

\[ H = \sum_k E(k) B^\dagger (k) B(k) + \frac{V_0}{N} \sum_{k,q} B^\dagger (k) B(q) \]
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 ~ off-diagonal disorder
Tunable exciton – phonon interactions = Tunable Holstein Hamiltonian
\[ \hat{H} = \hat{H}_{ex} + \hat{H}_{ph} + \hat{H}_I \]

\[ \hat{H}_{ex} = \sum_i (\epsilon_{eg} + D_{ij}) \hat{B}_i^\dagger \hat{B}_i + \sum_{i,j \neq i} J_{i,j} \hat{B}_i^\dagger \hat{B}_j \]

\[ \hat{H}_{ph} = \hbar \omega_0 \sum_{\mathbf{q},\lambda} (\hat{a}_{\mathbf{q},\lambda}^\dagger \hat{a}_{\mathbf{q},\lambda} + \frac{1}{2}) \]

\[ \hat{H}_I = \frac{1}{2} \sum_{i,j \neq i} \left( \hat{a}_i^\dagger + \hat{a}_i - \hat{a}_j^\dagger - \hat{a}_j \right) \times \]

\{ g_{D_{ij}} \left[ \hat{B}_i^\dagger \hat{B}_i + \hat{B}_j^\dagger \hat{B}_j \right] + g_{J_{ij}} \left[ \hat{B}_i^\dagger \hat{B}_j + \hat{B}_j^\dagger \hat{B}_i \right] \} \]
One dimensional array of LiCs molecules

Electric field (kV/cm)

Energy (kHz)

$\Theta = 90$

$\Theta = 0$

$g_J$

$g_D$
Quantum particles with tunable quantum statistics...
Kinematic Interaction

\[ H = \sum_M \sum_n E_M P_{n,M}^\dagger P_{n,M} + \sum_{M,L} J_{mn}^{M,L} P_{m,M}^\dagger P_{n,L} = \sum_{\rho=1}^{\sigma} \sum_k E_\rho(k) P_\rho^\dagger(k) P_\rho(k) \]

Operators \( P_{n,F}, P_{n,F}^\dagger \) are neither bosonic, nor fermionic

One-particle state: \( H \left| \Psi_{\rho_1}(k_1) \right\rangle = H \left[ P_{\rho_1}^\dagger(k_1) \left| \text{vac} \right\rangle \right] = E_{\rho_1}(k_1) \left| \Psi_{\rho_1}(k_1) \right\rangle \)

Two-particle state: \( \left| \Psi_{\rho_1,\rho_2}(k_1, k_2) \right\rangle = P_{\rho_1}^\dagger(k_1) P_{\rho_2}^\dagger(k_2) \left| \text{vac} \right\rangle \)

\[ H \left| \Psi_{\rho_1,\rho_2}(k_1, k_2) \right\rangle = \left( E_{\rho_1}(k_1) + E_{\rho_2}(k_2) \right) \left| \Psi_{\rho_1,\rho_2}(k_1, k_2) \right\rangle \quad \text{Bose/Fermi} \]

\[ H \left| \Psi_{\rho_1,\rho_2}(k_1, k_2) \right\rangle = \left( E_{\rho_1}(k_1) + E_{\rho_2}(k_2) \right) \left| \Psi_{\rho_1,\rho_2}(k_1, k_2) \right\rangle + \{ \ldots \text{K.I.} \ldots \} \quad \text{other} \]
What does this mean?

The same molecule cannot be excited twice!

The two excitations are coupled!

\( k_1 \) and \( k_2 \) are not conserved, however the total wavevector \( K = k_1 + k_2 \) should be conserved
Two-Particle Schroedinger Equation

\[ H = \sum_{k, \rho} E_{\rho}(k) P_{\rho}^{\dagger}(k) P_{\rho}(k) \]

\[ \ket{\psi_{2-B}} = \sum_{k_1, k_2, \rho_1, \rho_2} C_{\rho_1, \rho_2}(k_1, k_2) P_{\rho_1}^{\dagger}(k_1) P_{\rho_2}^{\dagger}(k_2) \ket{\text{vac}} \]

\[ (E_{\rho_1}(k_1) + E_{\rho_2}(k_2) - E) C_{\rho_1, \rho_2}(k_1, k_2) = \frac{E_{\rho_1}(k_1) + E_{\rho_2}(k_2)}{N} \sum_q C_{\rho_1, \rho_2}(k_1 - q, k_2 + q) \]

In terms of \( K \) and \( k \):

\[ K = k_1 + k_2; \]
\[ k = (k_1 - k_2)/2. \]

\[ (E_{\rho_1}(K/2 - k) + E_{\rho_2}(K/2 + k) - E) C_{\rho_1, \rho_2}^{K}(k) = \frac{E_{\rho_1}(K/2 + k) + E_{\rho_2}(K/2 - k)}{N} \left( \sum_q C_{\rho_1, \rho_2}^{K}(q) \right) \]
Bound state solutions?

\[ 1 = \frac{1}{N} \sum_k \frac{E_{\rho_1}(K/2 + k) + E_{\rho_2}(K/2 - k)}{(E_{\rho_1}(K/2 + k) + E_{\rho_2}(K/2 - k) - E)} \]

Notation: \( E_{\rho_1}(K/2 + k) + E_{\rho_2}(K/2 - k) \rightarrow E_0 + J_0(k) \)

\[ \frac{1}{N} \sum_{k=1}^{N} \frac{E_0 + J_0(k)}{(E_0 + J_0(k) - E)} = \frac{1}{N} \sum_{k=1}^{N} \frac{(E_0 + J_0(k) - E) + E}{(E_0 + J_0(k) - E)} = 1 + \frac{E}{N} \sum_{k=1}^{N} \frac{1}{(E_0 + J_0(k) - E)} = 1 \]

\[ \sum_k \frac{1}{E_0 + J_0(k) - E} = 0; \quad \frac{1}{N} \sum_k \frac{J_0(k)}{E_0 + J_0(k) - E} = 1 \]

No solutions ...
However,

Schroedinger equation has a simple solution

\[
\begin{cases}
  E_{\rho_1} (K/2 - k) + E_{\rho_2} (K/2 + k) = \text{const} = E_* \\
  \sum_k C^K_{\rho_1, \rho_2} (k) = 0
\end{cases}
\]

Under this condition \( \{... \text{K.I.} ...\} = 0 \)

if the dispersion curve shape is such that for a specific value of the total wave vector \( K = K_* \) (and specific branches \( \rho_1 \) and \( \rho_2 \)) the sum \( E_{\rho_1} (K/2 - k) + E_{\rho_2} (K/2 + k) \) does not depend on the relative wave vector \( k \). This solution is \((N-1)\)-time degenerate \((n\)-degeneracy\) and has the following wave function:

\[
\left| \psi_{K_*, n>0}^{\rho_1, \rho_2} \right> = \frac{1}{\sqrt{N}} \sum_k B^K_{\rho_1, \rho_2} (n) e^{i k n a} P^\dagger_{\rho_1} (K_* / 2 + k) P^\dagger_{\rho_2} (K_* / 2 - k) \left| \text{vac} \right>
\]

\( n \) is the (fixed) distance between excitations in site representation!
Can happen for branches with high symmetry:

\[ E_1(k) = E_1 + \frac{J}{2} \cos(ak); \quad E_2(k) = E_2 + \frac{J}{2} \cos(ak) \]

\[ E_{2-B} = E(K/2 + k) + E(K/2 - k) \]
Can be realized with Frenkel rotational excitons in an optical lattice with oblique electric field:

\[ E_\alpha (k, \theta) \approx E_1 + 2 |\alpha_-|^2 j(k) (\cos^2 \theta - 2/3), \]
\[ E_\beta (k) \approx E_1 + 2/3 |\alpha_-|^2 j(k), \]
\[ E_\gamma (k, \theta) \approx E_0 - 2 |\alpha_-^* \beta_+ + \beta_-^* \alpha_+| j(k) (\cos^2 \theta - 1/3) \]

\[ \cos \theta = \frac{1}{\sqrt{3}} : \quad J_\alpha = -J_\beta, \text{ and } J_\gamma = 0. \]
K (1/cm)

\( E(a) \)
\( E(a, r_e) \)

\( \Gamma \) (1/cm)

\( t \) (s)

negative binding energy

positive binding energy
What can we do with molecules on a lattice that cannot be done with conventional crystals?

1. Study rotational excitons:
   Rotational excitons are controllable
   * Electric field can be used to control exciton effective mass, exciton – impurity interactions and exciton – exciton interactions

2. Study quantum statistics of excitons
   * The role of kinematic interactions remains an open question

3. Study energy transfer in molecular ensembles
   * Could be used for quantum simulation of energy transfer in photosynthetic complexes and polaron physics
References


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Alisdair Wallis and RK, JCP, in press (2011); arXiv:1109.1008