Formulating the Few-Body Problem for Ultracold Nonreactive Molecules in an Optical Lattice

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KITP FBS-16 Theory Talk

Collaborators

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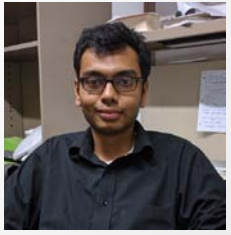

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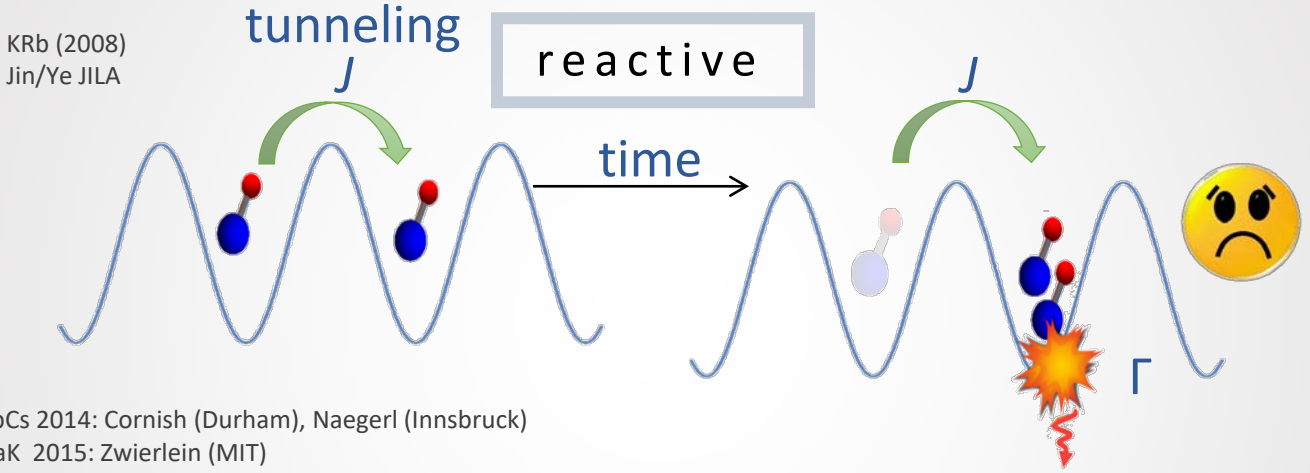
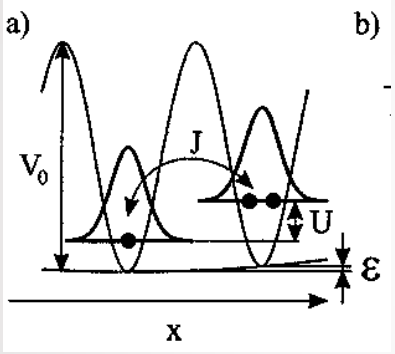
Types of molecules



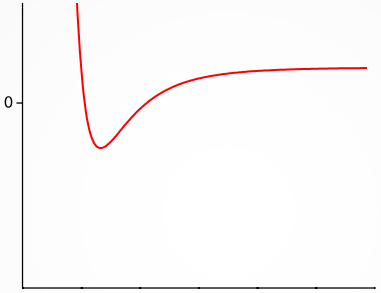
Conventional wisdom from atoms

$$H = -J \sum_{\langle i,j \rangle} b_i^\dagger b_j + \sum_i \epsilon_i \hat{n}_i + \frac{1}{2} U \sum_i \hat{n}_i(\hat{n}_i - 1)$$



Jaksch, et al PRL (1998)

Feshbach resonance



Chin, et al RMP (2010)

Isolated resonance

$$\approx \quad V(\mathbf{R}) = \frac{2\pi\hbar^2}{\mu} a(B)\,\delta(\mathbf{R})\frac{\partial}{\partial R}R,$$

$$U = 4\pi a_s \hbar^2 \int d^3x\,|w(\mathbf{x})|^4/m$$

Simple assumption is that molecules are no different (unless electric field)

Big question: Ignoring rovibrational excitations?

Ultracold Nonreactive Molecules in an Optical Lattice: Connecting Chemistry to Many-Body Physics

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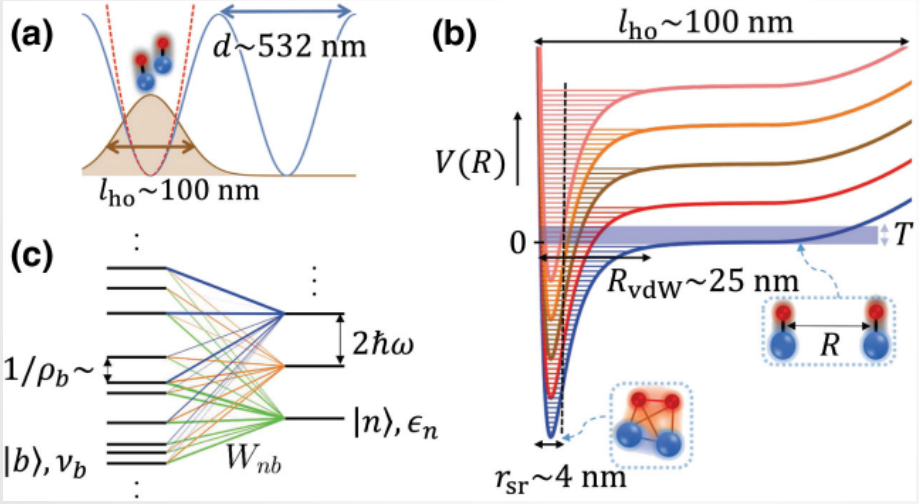
Basic 2-molecule model:

$$H_{\mathrm{rel}} = \sum_n \epsilon_n |n\rangle\langle n| + \sum_b \nu_b |b\rangle\langle b| + \sum_{bn} (W_{nb}|n\rangle\langle b| + \mathrm{H.c.})$$



- $|n\rangle$: long-range oscillator state
- $|b\rangle$: bimolecular complex
- W_{nb}: coupling

Basic bimolecular model:

$$H_{\rm rel} = \underbrace{\sum_n \epsilon_n |n\rangle\langle n|}_{\hat{H}_{\rm lr}} + \underbrace{\sum_b \nu_b |b\rangle\langle b|}_{\hat{H}_{\rm b}} + \underbrace{\sum_{bn} (W_{nb}|n\rangle\langle b| + {\rm H.c.})}_{\hat{H}_{\rm cp}}$$

$\hat{H}_{\rm b}$ describes the short-range physics where the motion is classically chaotic and QM RMT applies.

$\nu_{\rm b}$ should be determined as eigenvalues of a random matrix:

$$P(\hat{H}_{\rm b}) = \mathcal{N}_H e^{-{\rm Tr}\,\hat{H}_{\rm b}^2/2\sigma^2} \qquad \sigma = \frac{\sqrt{2N_b}}{\pi\rho_b}$$

N_b = total number of eigenvalues of H_b

ρ_b = density of states

$$W_{nb} = w_b M_n / I_{\rm ho}^{3/2}$$

$$M_n = \frac{\Gamma(n+3/2)}{\Gamma(n+1)}$$

Basic bimolecular model:

$$H_{\mathrm{rel}} = \underbrace{\sum_n \epsilon_n |n\rangle\langle n|}_{\hat{H}_{\mathrm{lr}}} + \underbrace{\sum_b \nu_b |b\rangle\langle b|}_{\hat{H}_{\mathrm{b}}} + \underbrace{\sum_{bn} (W_{nb}|n\rangle\langle b| + \mathrm{H.c.})}_{\hat{H}_{\mathrm{cp}}}$$

W_{nb} describes the coupling between the long-range oscillator states and the short ranged bimolecular complex states.

Separation of length scales means the coupling factors:

$$W_{nb} = w_b M_n / l_{\mathrm{ho}}^{3/2} \qquad M_n = \sqrt{\frac{\Gamma(n + 3/2)}{\Gamma(n + 1)}}$$

w_{b} is also governed by RMT physics:

$$P_w(w_b) = N_w e^{-w_b^2/(2\sigma_w^2)}$$

But... What σ_w should we use?

Determining σ_w: TST + QDT

Fermi's golden rule yields

$$\gamma_b = 2\pi \frac{w_b^2}{l_{\rm ho}^3} \sum_n M_n^2 \delta(\nu_b - \epsilon_n).$$

Average dissociation rate

$$\gamma_b = \frac{\pi \mu^{3/2} \sqrt{\nu_b} \sigma_w^2}{\sqrt{2}}$$

$$\boxed{\sigma_w = \left(\frac{2}{\mu^3 \nu_b}\right)^{1/4} \sqrt{\frac{\gamma_b}{\pi}}.}$$

"unimolecular dissociation"

$l_{\rm ho} \sim$

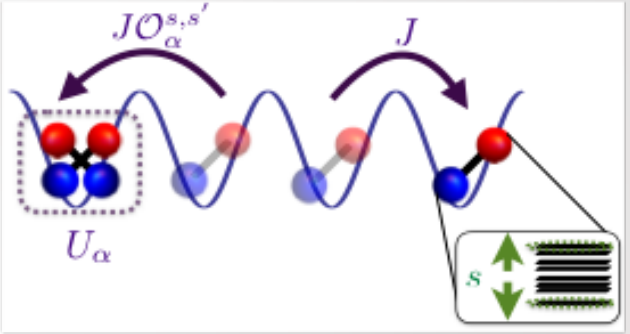
$$\gamma_{\rm TST} = \frac{2}{\pi \rho_b}.$$

$$\mathcal{A}(E) = \frac{\Gamma(3/4)^2}{\pi} R_{\rm vdW} \sqrt{2\mu E}\,.$$

$$\gamma_b = \gamma_{\rm TST} \mathcal{A}(\nu_b).$$

The lattice model:

$$\hat{H} = -J \sum_{\langle i,j\rangle,s} \left[\hat{c}^{\dagger}_{i,s}\hat{c}_{j,s} + \text{H.c.}\right] + \sum_{i}\left(\sum_{\alpha} U_{\alpha}\hat{n}_{i,\alpha} + \frac{3\omega}{2}\hat{n}_{i}\right),$$



- s runs over the internal molecule states in the open channel
- J tunneling
- U_{α} interaction energy of a pair of NRMs in state α. $\widehat{H}_{\text{rel}}|\alpha\rangle = E_{\alpha}|\alpha\rangle$

$$\hat{c}^{\dagger}_{i,s}|0\rangle_i = |s\rangle_i\,,$$
$$\hat{c}^{\dagger}_{i,s}|s'\rangle_i = P_{s,s'}\sqrt{1+\delta_{s,s'}}\sum_{\alpha}\mathcal{O}_{\alpha}^{s,s'}|\alpha\rangle_i\,,$$
$$\hat{c}^{\dagger}_{i,s}|\alpha\rangle_i = 0\,.$$

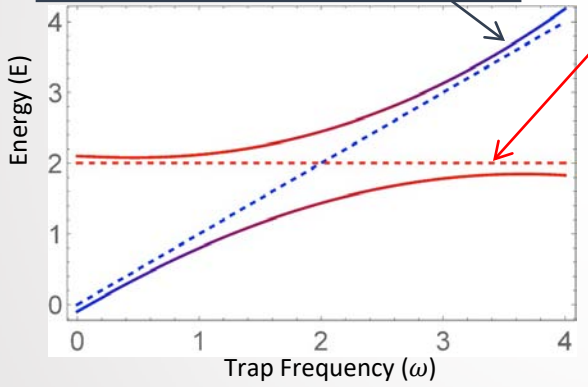
$$\mathcal{O}_{\alpha}^{s,s'} \equiv \langle\alpha|s,s'\rangle$$

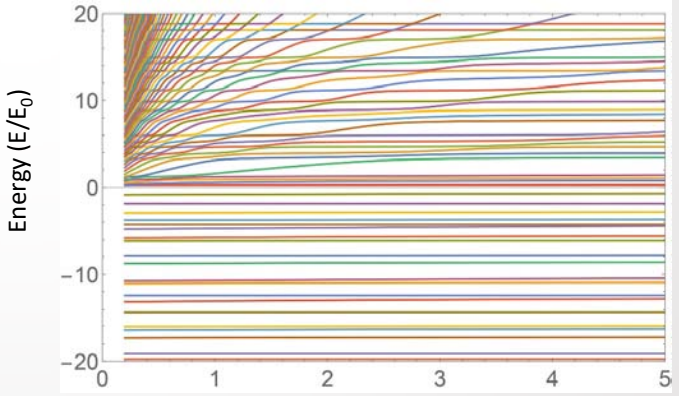
Eigenenergies of two molecules in a trap

$r_{sr} \ll l_{ho} \longrightarrow$ bound states independent of ω

harmonic oscillator state

bound State





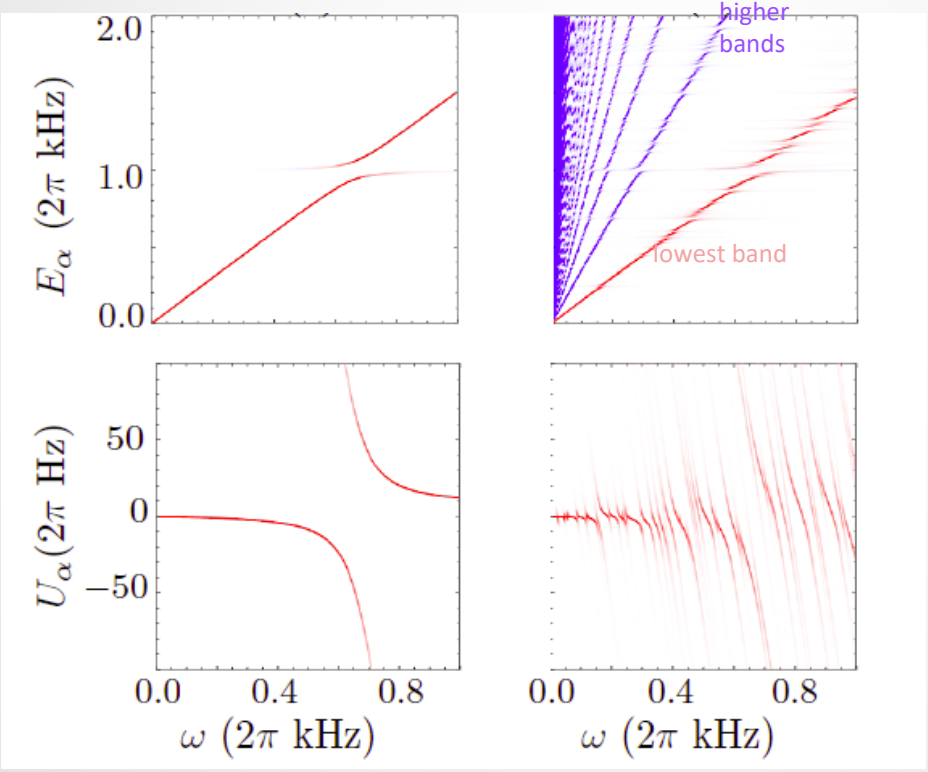
on-site interaction energy

$$U = E - 4n\hbar\omega - 3\hbar\omega$$

Interaction energy and amplitude for hopping

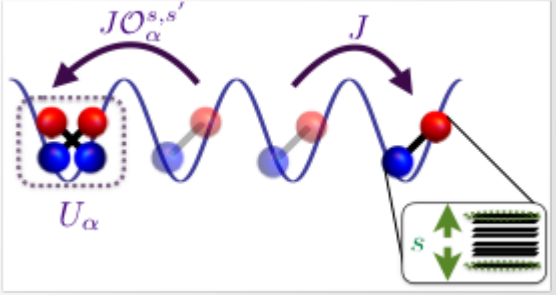
isolated state

realistic parameters (RbCs)



Opacity of line is

$$\mathcal{O}_\alpha^{s,s'} \equiv \langle \alpha | s, s' \rangle$$



$$U_\alpha = E_\alpha - 3\omega/2$$

Current model operates under a suite of approximations : RMT+TST+QDT

- Random Matrix Theory (RMT): describes short range classically chaotic regime
- Transition State Theory (TST): standard tool in chemistry that describes rates of chemical reactions. Is it good here? Who knows?
- Quantum Defect Theory (QDT): Accounts for the propagation of the solutions in the van der Waals tail. Very likely the most reliable of the three approximations.

It sure would be nice if we could test these approximations with a full calculation..

The hyperspherical formulation of this problem...

$$H_{\rm rel} = \sum_n \epsilon_n |n\rangle\langle n| + \sum_b \nu_b |b\rangle\langle b| + \sum_{bn} (W_{nb}|n\rangle\langle b| + \text{H.c.})$$

$$\hat{H}_{\rm rel} = \frac{-1}{2\mu_{\rm 4B} R^{d-1}} \frac{\partial}{\partial R}\left(R^{d-1}\frac{\partial}{\partial R}\right) + \frac{1}{2}\mu_{\rm 4B}\omega^2 R^2 + \underbrace{\frac{\hat{\Lambda}^2(\Omega)}{2\mu_{\rm 4B}R^2} + \hat{V}(R,\Omega)}.$$

$$\hat{H}_{\rm ad}(R;\Omega)\Phi_\alpha(R;\Omega) = \mathcal{U}_\alpha(R)\Phi_\alpha(R;\Omega)$$

In each channel, choose a basis of channel eigenstates:

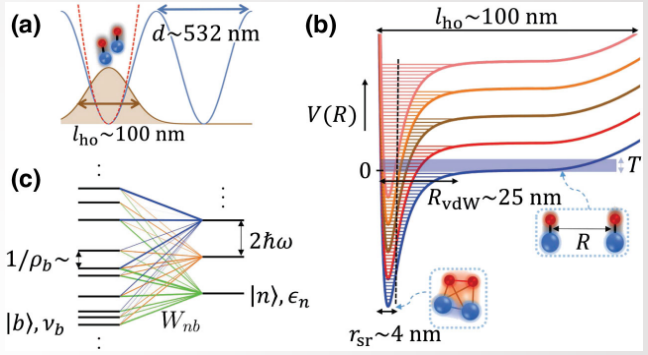
$$|\psi_\alpha^j \alpha\rangle = \begin{cases} |\psi_B^a B\rangle & \alpha \in B \\ |\psi_O^n O\rangle & \alpha \in O \end{cases},$$

$$\mathcal{X}_{\alpha\beta}^{ij} = -\frac{1}{2\mu_{\rm 4B}}\int_0^{R_m} \Big[P_{\alpha\beta}(R)\Big(\psi_\alpha^i \frac{d\psi_\beta^j}{dR} - \psi_\beta^j \frac{d\psi_\alpha^i}{dR}\Big) - (1-\delta_{\alpha\beta})\,\psi_\alpha^i \psi_\beta^j \tilde{Q}_{\alpha\beta}(R)\Big] R^2 dR\,.$$

$$P_{\alpha\beta}(R) = \left\langle \Phi_\alpha \middle| \frac{\partial \Phi_\beta}{\partial R} \right\rangle \qquad \tilde{Q}_{\alpha\beta}(R) = \left\langle \frac{\partial \Phi_\alpha}{\partial R} \middle| \frac{\partial \Phi_\beta}{\partial R} \right\rangle$$

$$\begin{aligned}\hat{H}_{\rm rel} = &\sum_n |\psi_O^n O\rangle \epsilon_n \langle \psi_O^n O| + \sum_{Ba} |\psi_B^a B\rangle \lambda_B^a \langle \psi_B^a B| \\ &+ \sum_n \sum_{B,a} (|\psi_O^n O\rangle \mathcal{X}_{OB}^{na} \langle \psi_B^a B| + \text{H.c.}) \\ &+ \sum_{B,B',a,a'} |\psi_B^a B\rangle \mathcal{X}_{BB'}^{aa'} \langle \psi_{B'}^{a'} B'|\,.\end{aligned}$$

$$\hat{H}_{\rm rel} = \sum_n |\psi_O^n O\rangle \epsilon_n \langle \psi_O^n O| + \sum_{Ba} |\psi_B^a B\rangle \lambda_B^a \langle \psi_B^a B|$$
$$+ \sum_n \sum_{B,a} \left(|\psi_O^n O\rangle \mathcal{X}_{OB}^{na} \langle \psi_B^a B| + \text{H.c.} \right)$$
$$+ \sum_{B,B',a,a'} |\psi_B^a B\rangle \mathcal{X}_{BB'}^{aa} \langle \psi_{B'}^{a'} B'| \,.$$



$$\hat{H}_{\rm rel} = \left(\begin{array}{c|cc|c} OO & OB_1 & OB_2 & \cdots \\ \hline B_1 O & B_1 B_2 & B_1 B_3 & \cdots \\ B_2 O & B_2 B_1 & B_2 B_2 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{array} \right) \rightarrow \left(\begin{array}{c|cc|c} \epsilon_n & \cdots & W_{nb} & \cdots \\ \hline \vdots & \ddots & 0 & 0 \\ W_{bn} & 0 & \nu_b & 0 \\ \vdots & 0 & 0 & \ddots \end{array} \right) .$$

So, in terms of solutions to the four-atom Schrodinger equation:

$$\nu_b = \sum_{B,B',a,a'} \langle b|\psi^a_B B\rangle\langle\psi^{a'}_{B'} B'|b\rangle \left(\lambda^a_B \delta_{aB,a'B'} + \mathcal{X}^{aa'}_{BB'}\right),$$

$$W_{nb} = \frac{w_b}{l_{\rm ho}^{3/2}} \sqrt{\frac{\Gamma(n+3/2)}{\Gamma(n+1)}}$$

$$w_b = \frac{-\sqrt{2}}{\sqrt{\pi}\mu_{\rm 4B}} \sum_{B,a} \langle\psi^a_B B|b\rangle \int_0^{R_m} \mathcal{I}^a_{OB}(R) R^2\, dR\,.$$

$$\mathcal{I}^a_{OB}(R) = P_{OB}(R)\left(\frac{d}{dR} - \tilde{Q}_{OB}(R)\right)\psi^a_B(R)$$

$$P_{\alpha\beta}(R) = \left\langle \Phi_\alpha \left| \frac{\partial \Phi_\beta}{\partial R} \right. \right\rangle$$

$$\tilde{Q}_{\alpha\beta}(R) = \left\langle \frac{\partial \Phi_\alpha}{\partial R} \left| \frac{\partial \Phi_\beta}{\partial R} \right. \right\rangle$$

Final Comments...

- Full calculation of four-body problem with realistic atom-atom potentials is daunting.
- Reduced dimensionality?
  - Freeze one molecule? Use a toy model? See Chris Ticknor's talk up next!
  - Try formulating a 1D model? Can we even expect RMT physics in a 1D model?
- Could learn a lot from such a 4-body calculation:
  - What is a realistic value for the density of states?
  - Are the widths of the distributions of the RMT statistics correctly estimated?
  - Is the dissociation rate from TST reliable?

THANK YOU!

Thanks to these collaborators

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