Entanglement Manipulation in Cold Molecular Gases using Strong Laser Pulses

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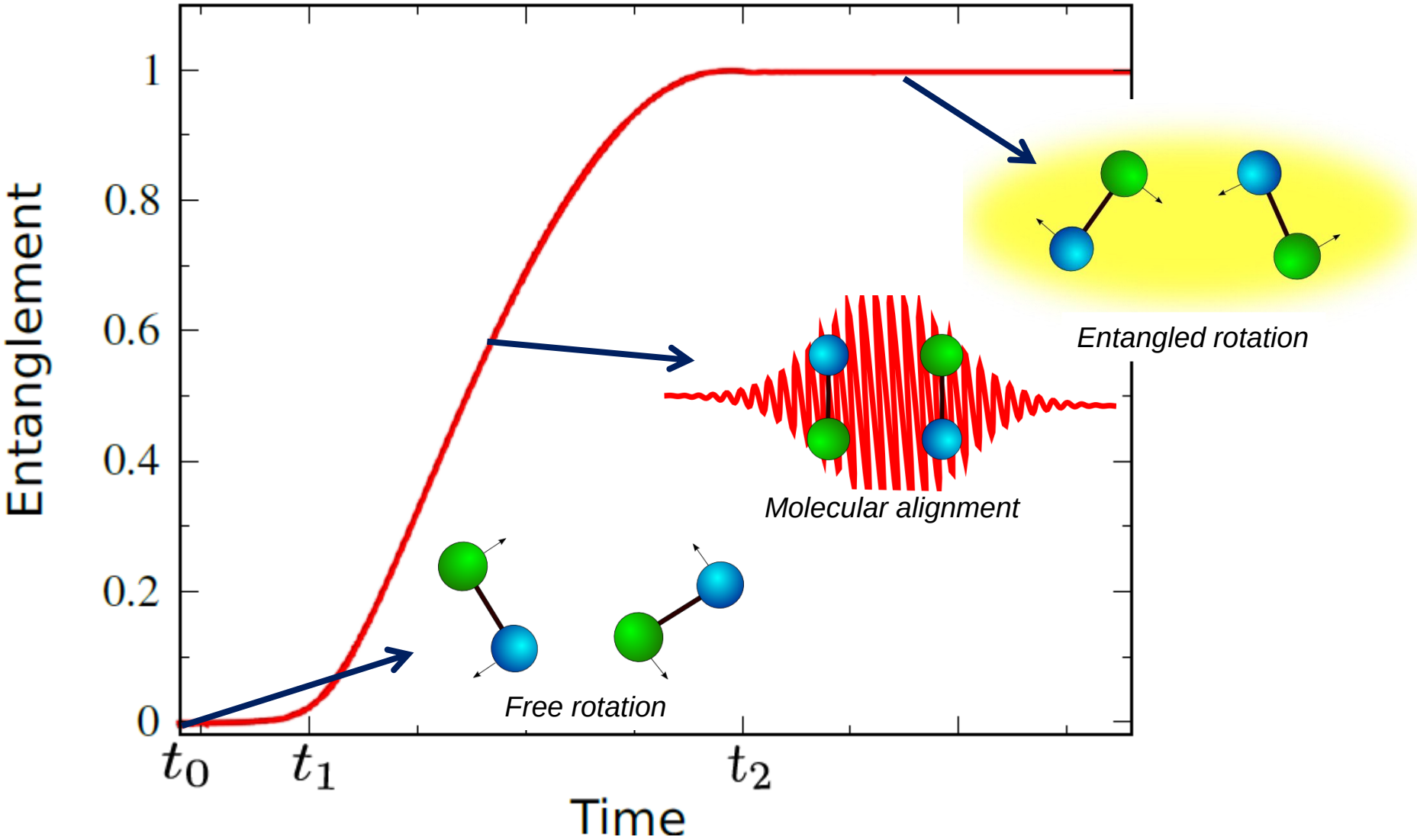
Brief review

- How to create entanglement in optical lattices?
  - Tuning interparticle distance to control scattering dynamics:
    - s-wave interaction induces a phase shift for conditional logic

      Nature 425, 937, **2003**; *Nature* 448, 452, **2007**
  - Excitation of particles to strongly interacting states:
    - Long-range interaction induces a conditional phase shift

      Nat. Phys. 5, 110, **2009**; PRL 104, 010502, **2010**; PRL 104, 010503, **2010**
  - Other theoretical proposals:
    - NMR-type "always-on" dipole-dipole interaction PRL 88, 067901, **2002**
    - Topologically protected states in spin chains *Nat. Phys.* 2, 341, **2006**

The idea of this talk



arXiv 1302.6444

Molecules in off-resonant laser fields

- Optical field far-detuned from any vibronic resonance
  - Low intensity fields give optical trapping potentials

- Eliminating excited electronic state from the system dynamics gives the effective light-matter interaction*

$$\hat{H}_{\mathrm{AC}} = -\frac{|E_0|^2}{4}\left\{\alpha_\perp + (\alpha_\parallel - \alpha_\perp)\cos^2\theta\right\}$$

- For a wide range of intensities, laser field only couples to rotational degrees of freedom
  - Field strength below ionization threshold ($E_0 < 10^9$ V/cm)

*Linear molecule & linear field polarization, see *Rev. Mod. Phys* .75, 543, 2003

Molecular alignment

- Optical field creates a double-well potential in angular space
- Rotational motion is constrained to potential minima.
- The effective Hamiltonian in angular space is given by

$$H = -B_e \frac{d^2}{d\theta^2} + V_{\text{eff}}(\theta)$$

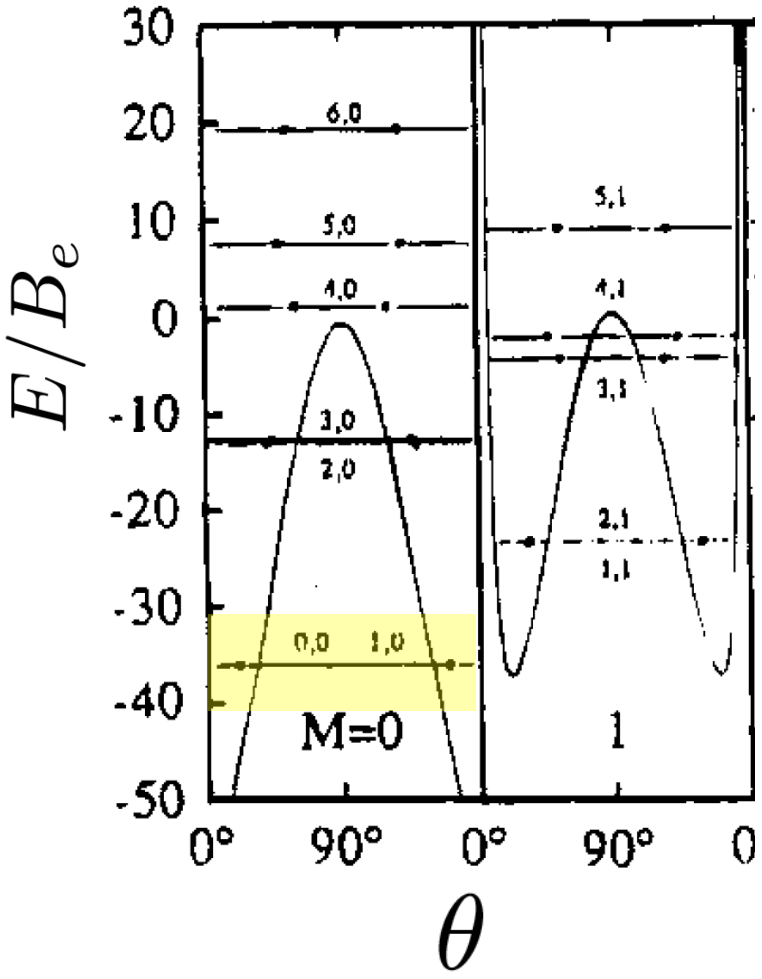
Anisotropic double-well angular potential

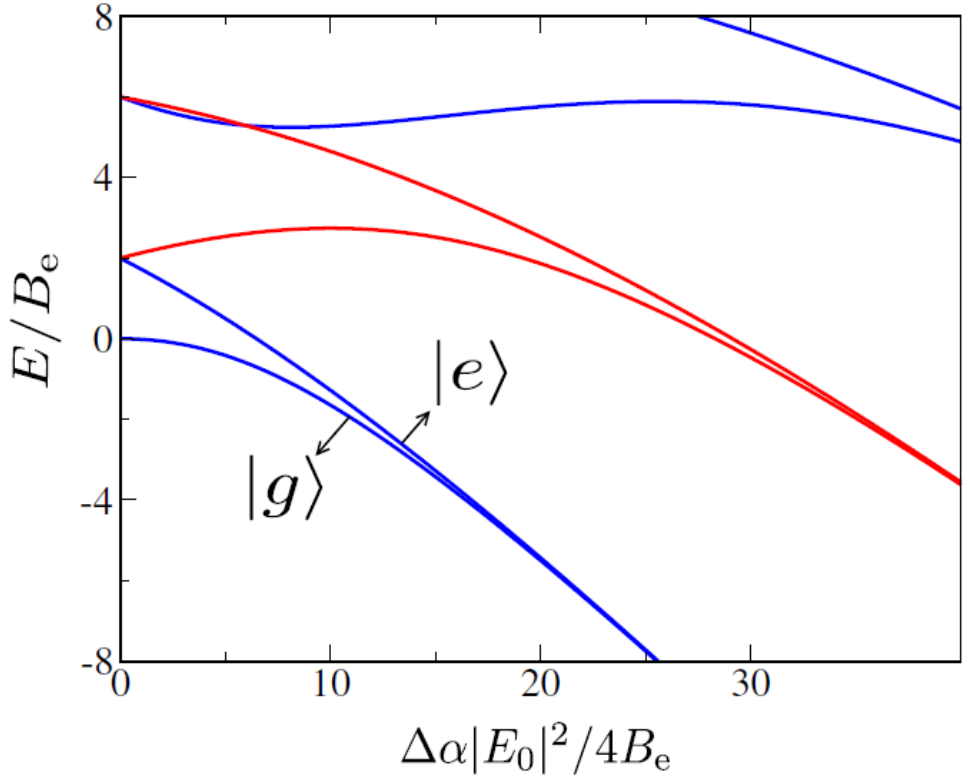
$$V_{\text{eff}}(\theta) = B_e \left(\frac{|M|^2 - 1/4}{\sin^2 \theta} - \frac{1}{4} \right) + \bar{H}_{\text{AC}}$$

Friedrich & Herschbach, *J. Phys. Chem.* 99, 15686, 1995

Two-level approximation

High-field angular potentials





The lowest two rotational states with M=0 form a qubit basis

J. Phys. Chem. 99, 15686, 1995

Field-dressed dipolar interaction

- Dipole-dipole interaction between molecules given by

$$\hat{V}_{\mathrm{dd}} = \gamma(d^2/R_{ij}^3)(1 - 3\cos^2\Theta)\left\{|e_i e_j\rangle\langle g_i g_j| + |e_i g_j\rangle\langle g_i e_j| + \mathrm{H.c.}\right\}$$

- Universal parameter $0 \leq \gamma \leq 1$ depends on field strength

$$\gamma = |\langle e|\hat{d}_Z|g\rangle|^2/d^2$$

- If the field breaks parity:
  - γ is suppressed with increasing field strength
  - Additional channels become allowed

- Far-detuned optical fields preserve parity (Raman process)

Double exchange interaction

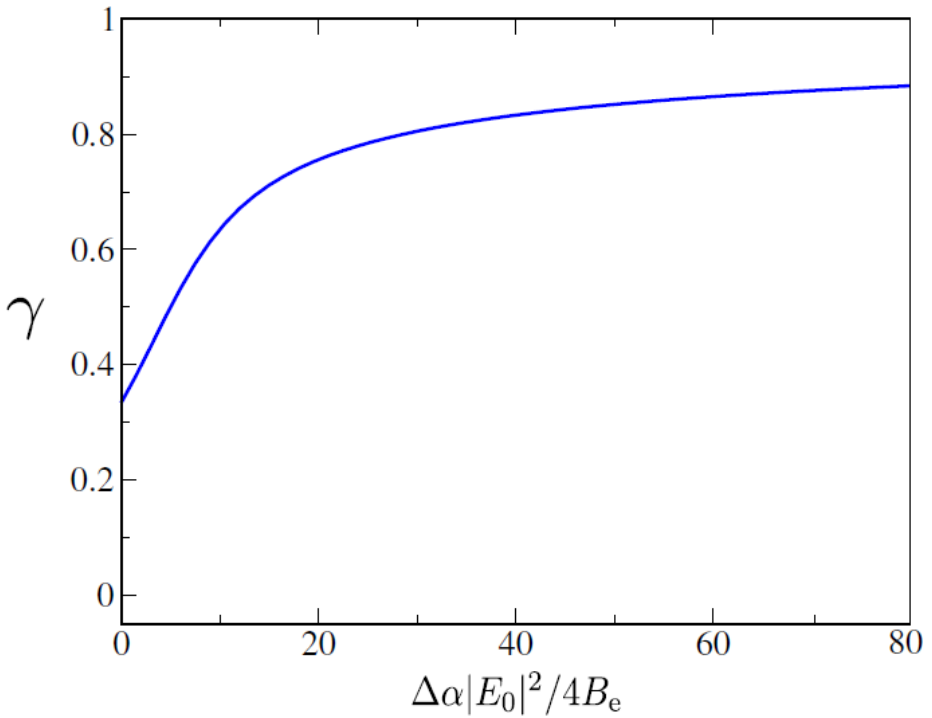
- Strong laser field suppresses the qubit energy gap
- Double excitations become energetically allowed

$$J_{ij} = \langle e_i e_j | \hat{V}_{\mathrm{dd}} | g_i g_j \rangle$$
$$= \langle e_i g_j | \hat{V}_{\mathrm{dd}} | g_i e_j \rangle$$

Parity forbidden channels

$$V_{ij}^{gg} = \langle g_i g_j | \hat{V}_{\mathrm{dd}} | g_i g_j \rangle = 0$$

$$V_{ij}^{ee} = \langle e_i e_j | \hat{V}_{\mathrm{dd}} | e_i e_j \rangle = 0$$



Two-body dynamics in strong fields

- Consider dipolar coupling driven by strong laser pulse
  - Intermolecular distance is fixed
  - Initial state is separable
  - Pulse is **long** compared with molecular rotation period

- Two-body evolution determined by simple Hamiltonian
  - Time-dependence induced by the pulse

$$\mathcal{H} = \begin{pmatrix} 0 & 0 & 0 & J_{12}(t) \\ 0 & \epsilon_{eg}(t) & J_{12}(t) & 0 \\ 0 & J_{12}(t) & \epsilon_{eg}(t) & 0 \\ J_{12}(t) & 0 & 0 & 2\epsilon_{eg}(t) \end{pmatrix}$$

- Relevant timescales for optical traps
  - Laser pulse duration much larger than rotational period (~1 ps)
  - Pulse duration not larger than dipolar interaction time (~10-100 μs)
  - Decoherence time is the longest timescale (~1 s)

- Solve two-body evolution for Gaussian pulse
  - Ignore dissipation and solve Hamiltonian dynamics numerically
  - Initial condition is the separable ground state $|g_1 g_2\rangle$
  - Single-excitation manifold is uncoupled from the dynamics
  - System undergoes loop in parameter space without geometrical phase

Alignment-mediated entanglement

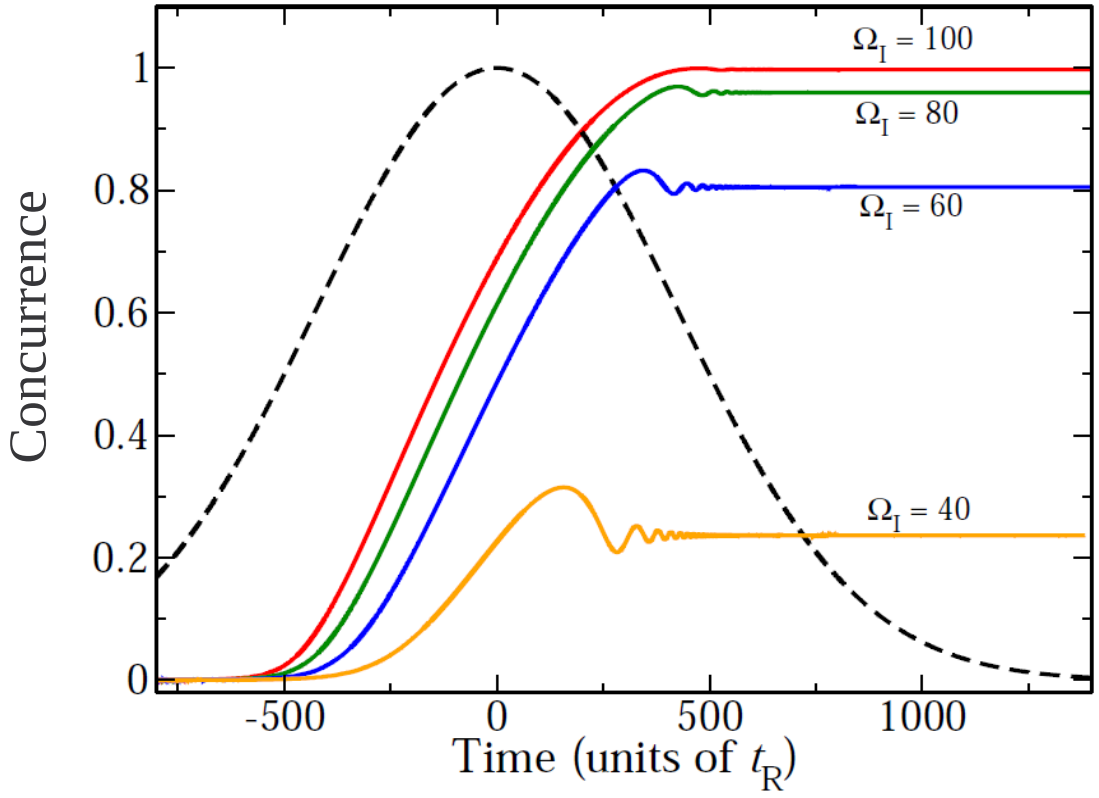
- Molecules become entangled during alignment period
  - Entanglement persists after the pulse is over
  - Concurrence depends on laser parameters

Light-matter interaction strength

$$\Omega_{\mathrm{I}} = \frac{\Delta\alpha |E_0|^2}{4B_e}$$

Rotational timescale

$$t_{\mathrm{R}} = \hbar / B_{\mathrm{e}}$$



Laser manipulation of entanglement

- Relevant system parameters
  - Intermolecular distance R
  - Pulse peak intensity Ω_0
  - Pulse duration τ_{p}

Dipole-dipole interaction time

$$t_{\mathrm{dd}} = \hbar R^3 / d^2$$

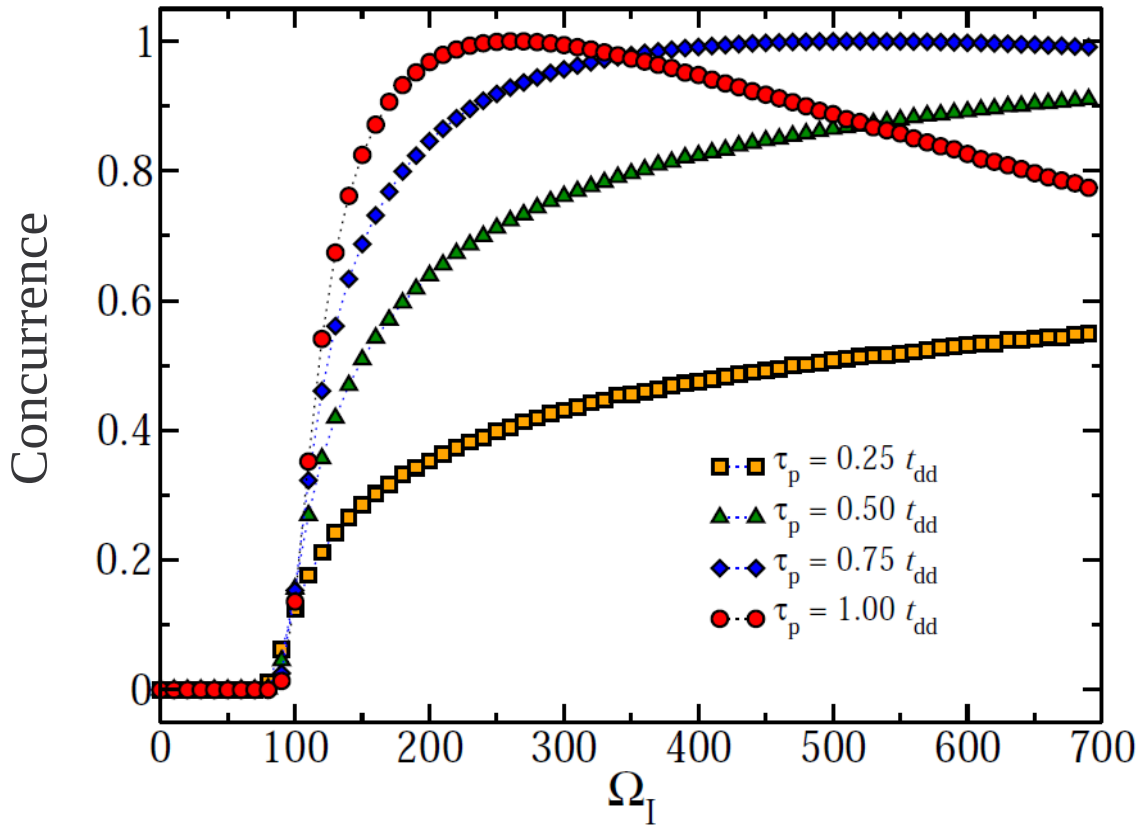
Plot for fixed distance

$$R = 100\, R_0$$

Dipole radius

$$R_0 = (d^2 / B_e)^{1/3}$$

Asymptotic concurrence vs peak intensity



Alkali-metal dimers in optical lattices

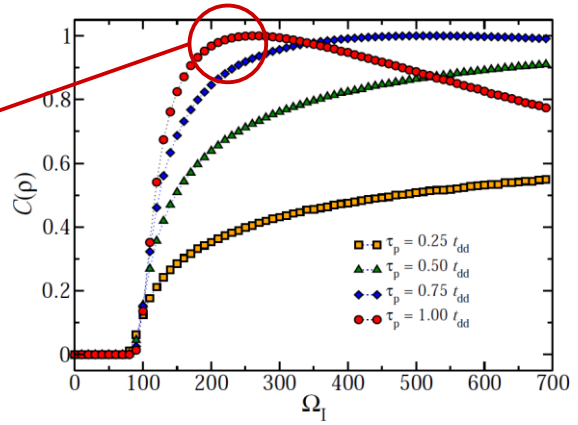
- System parameters for selected species

	d	$\Delta\alpha_V$	B_e	I_0	R_0	t_R
	(D)	(a_0^3)	(cm^{-1})	$(10^8$ W/cm$^2)$	(nm)	(ps)
RbCs	1.238	441	0.0290	0.4	6.4	1.15
KRb	0.615	360	0.0386	0.7	3.7	0.86
LiCs	5.529	327	0.1940	3.8	9.3	0.17
LiRb	4.168	280	0.2220	5.0	7.3	0.15

Example:
LiRb molecules in a 1460 nm wavelength lattice

Maximally entangled molecular pairs with

- Peak Intensity = 1.35 × 10^{11} W/cm^2
- FWHM = 150 ns



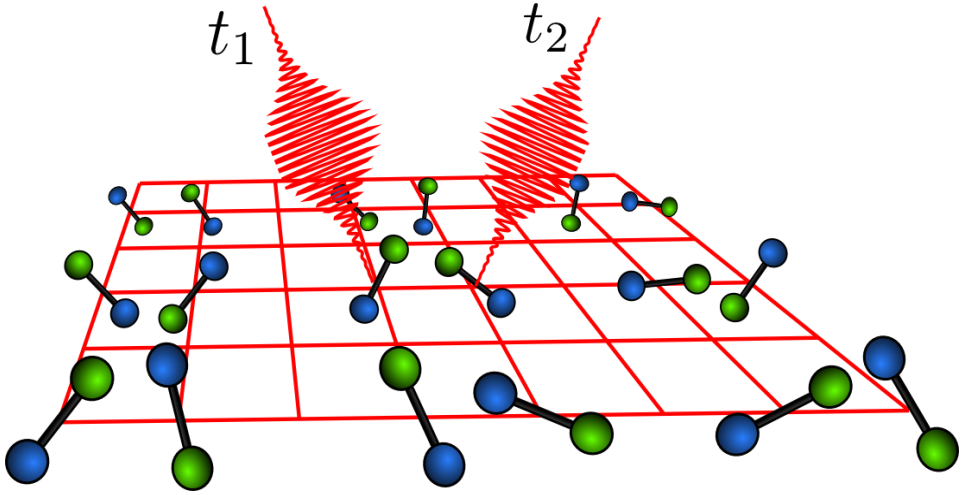
Entanglement detection in optical traps

- We propose two ways to detect rotational entanglement
  - Laser-induced fluorescence measurements with site resolution
  - Microwave absorption of the array
  - Molecules are initially prepared in entangled state

- Can we establish violations of Bell's inequality?

  - Measure molecular orientation at different times

  - Simple case: *double-well lattice*



Molecular orientation

- There is a direct analogy between rotational evolution and orientation of Stern-Gerlach apparatus

  - Spin orientation is replaced by the orientation of internuclear axis. For linear molecules the operator is $\hat{O} = \cos\theta$
  - In the two-level subspace this reduces to $\hat{O} = a_X \hat{\sigma}_X$, with $a_X = 1/\sqrt{3}$
  - In the Heisenberg picture:

$$\hat{O}(\tau) = \mathrm{e}^{i\sigma_Z \tau} \hat{O} \mathrm{e}^{-i\sigma_Z \tau} = \vec{a}(\tau) \cdot \vec{\sigma}$$

Dimensionless time $\tau = 2B_\mathrm{e} t/\hbar$

(Pseudo) Stern-Gerlach direction

$$\vec{a}(\tau) = a_X(\cos\tau, -\sin\tau, 0)$$

  - Rotational time evolution is equivalent to a rotation of the Stern-Gerlach apparatus in the XY plane.

Bell inequality for orientation correlations

- By measuring molecular orientation at different sites it is possible to establish violations of the CHSH-type inequality

$$|E(t_a, t_b) + E(t_a, t'_b) + E(t'_a, t_b) - E(t'_a, t'_b)| \leq 2/3$$

Orientation correlation function

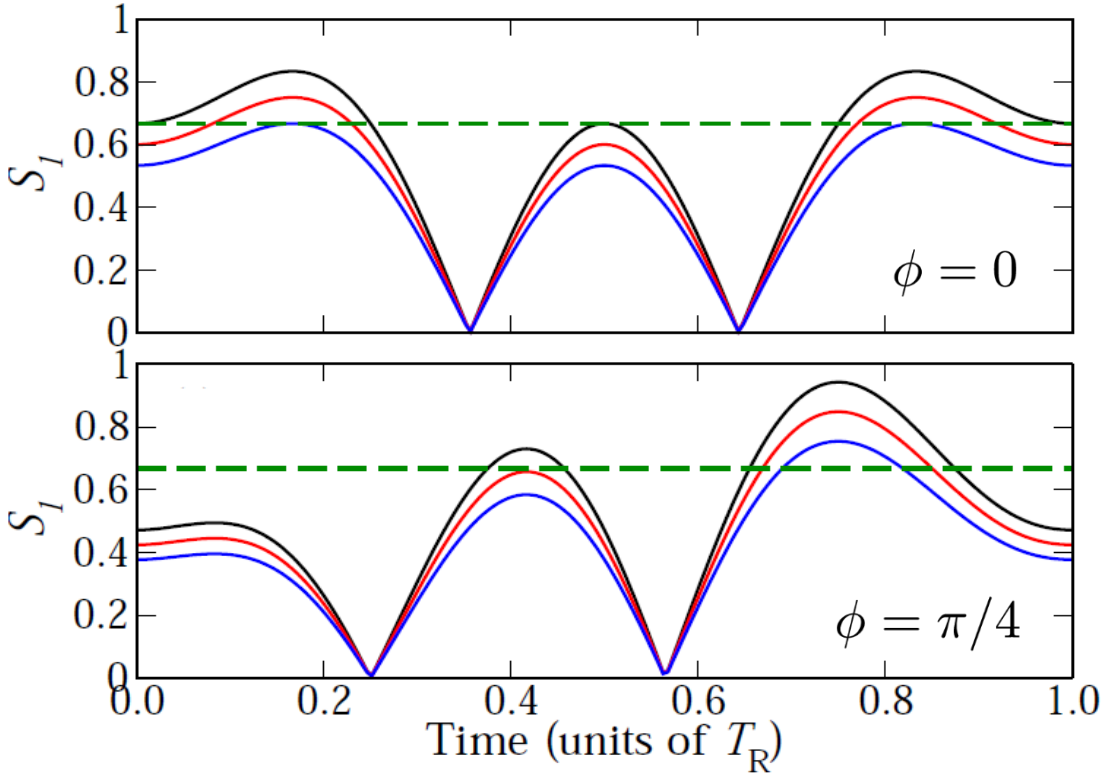
$$E(t, t') = \langle \hat{O}(t) \otimes \hat{O}(t') \rangle$$
$$= \langle \vec{a} \cdot \vec{\sigma} \otimes \vec{a}' \cdot \vec{\sigma} \rangle$$

Initial entangled state

$$|\Phi_0\rangle = a_0 |g_1 g_2\rangle + b_0 |e_1 e_2\rangle$$

Relative phase

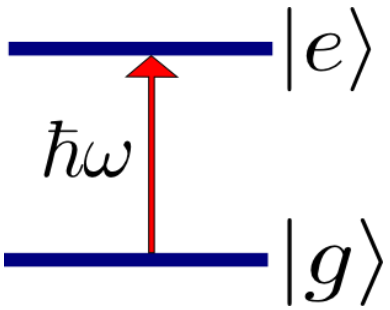
$$\phi = \arg\{b_0\}$$



Microwave entanglement detection

- Single site resolution is not straightforward to achieve
  - Although experiments with atoms are promising
    Nature 471, 319, **2011**; PRL 104, 010502, **2010**; PRL 104, 010503, **2010**

- Microwave field addresses the ensemble globally
  - For thermal ensembles, the linear absorption at frequency $\omega_{eg} = 2B_e/\hbar$ has Lorentzian lineshape.



$$\mathcal{A}(\omega) = \mathcal{N}\left(\frac{d^2}{3\hbar}\right) P_0(T) \frac{\gamma_e}{(\omega-\omega_{eg})^2+\gamma_e^2}$$

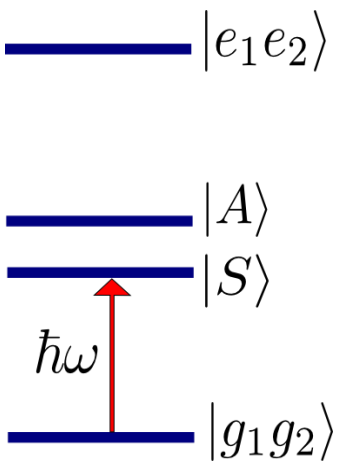
Ensemble of non-interacting molecules

- How does this change for an ensemble of entangled dimers?
  - Consider identical pairs in the state $|\Phi\rangle = a|g_1 g_2\rangle + b|e_1 e_2\rangle$
  - Frequency near resonance with lowest dipole-allowed transition
  - The absorption lineshape has a dynamical contribution proportional to the dimer concurrence

$$\mathcal{A}(\omega) = \mathcal{N}\left(\frac{d^2}{3\hbar}\right)\left[|a|^2 \frac{\gamma_S}{(\omega-\omega_S)^2+\gamma_S^2} + |ab| \frac{\mathcal{F}_\omega(t)}{(\omega-\omega_S)^2+\gamma_S^2}\right]$$

Dynamical lineshape factor

$$\mathcal{F}_\omega(t) = \mathrm{e}^{-\gamma t}\left[(\omega_S - \omega)\sin(\phi t) + \gamma_S \cos(\phi t)\right]$$



  - Detecting oscillations in the absorption peak indicates presence of entanglement

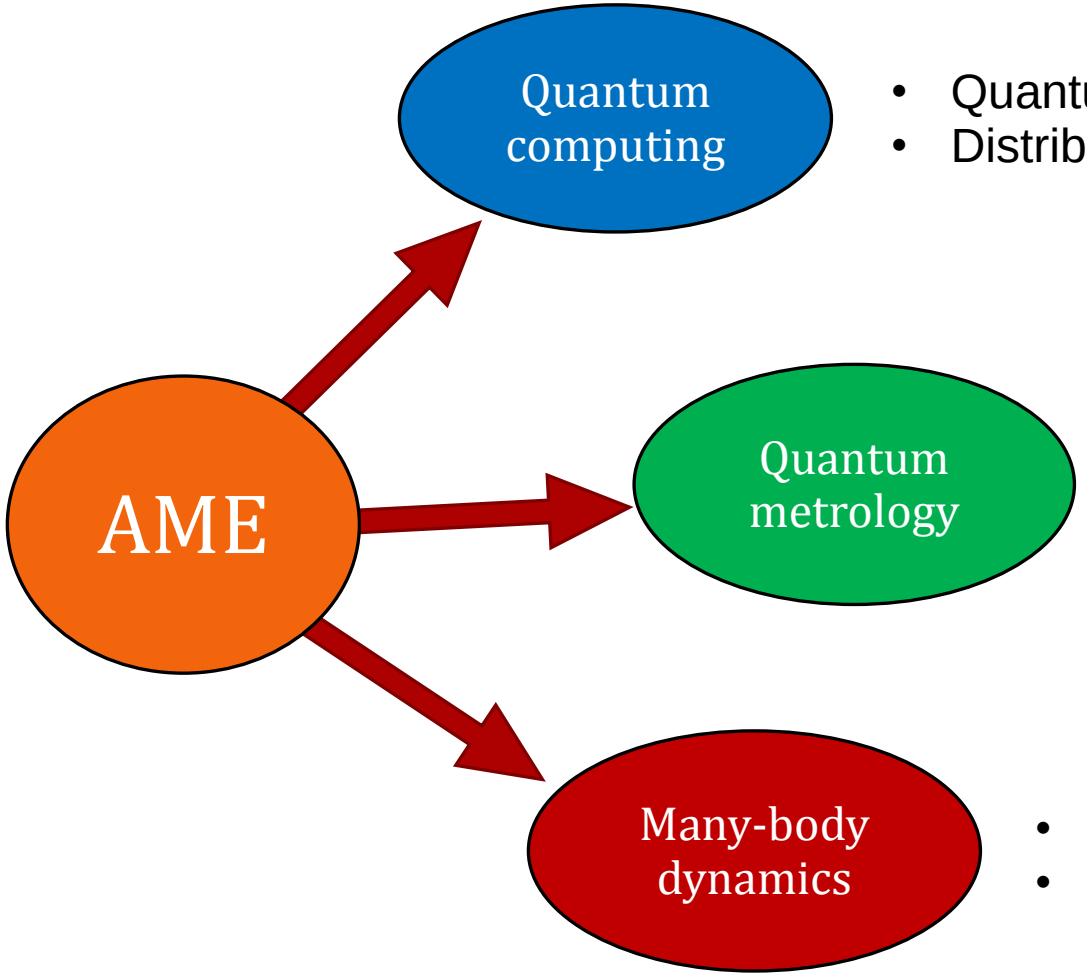
Summary

- Strong laser pulses mediate long-range entanglement between polar molecules.
- Degree of binary entanglement can be manipulated by tuning pulse parameters.
- Violation of Bell's inequality in optical traps can be established by measuring site-resolved fluorescence.
- Signatures of entanglement are present in the coherent beating of the *linear* microwave spectra.

Solved the two-body problem,

what can be done with larger molecular arrays?

Applications for larger systems



- Quantum circuit model
- Distributed quantum computing

- Single photon detection
- Nonlinear quantum optics

- Non-equilibrium phase transitions
- Entanglement dynamics

Acknowledgements

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