Center for Quantum Research and Technology

Pump-Probe Spectroscopy Of Weakly-Bound Molecules

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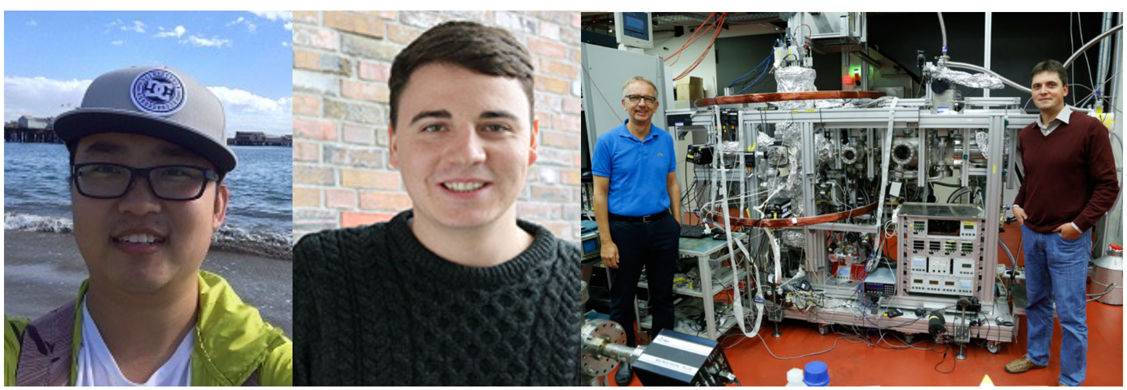


Supported by the NSF.

Two Exciting Fields

cold atoms:
universal physics

fast intense
lasers

(ultra)cold atoms:
universal physics

fast intense
lasers

(Selected) Works in Related Directions

PHYSICAL REVIEW LETTERS **124**, 253201 (2020)

Ultrafast Creation of Overlapping Rydberg Electrons in an Atomic BEC and Mott-Insulator Lattice

M. Mizoguchi,[1,2] Y. Zhang,[1,3] M. Kunimi,[1] A. Tanaka,[1] S. Takeda,[1,2,†] N. Takei,[1,2,‡] V. Bharti,[1] K. Koyasu,[1,2] T. Kishimoto,[4] D. Jaksch,[5,6] A. Glaetzle,[5,6] M. Kiffner,[5,6] G. Masella,[7] G. Pupillo,[7] M. Weidemüller,[8,9] and K. Ohmori[1,2,*]

ARTICLE

DOI: 10.1038/s41467-018-04556-3 OPEN

Quantum simulation of ultrafast (
trapped ultracold atoms

Ruwan Senaratne[1], Shankari V. Rajagopal[1], Toshihiko Shimasaki[1], Peter E. Dotti[1], Kurt M. Fujiwara[1], Kevin Singh[1]
Zachary A. Geiger[1] & David M. Weld[1]

(ultra)cold atoms: universal physics

fast intense lasers

Found Phys (2014) 44:813–818
DOI 10.1007/s10701-014-9773-5

Optically Engineered Quantum States in Ultrafast and Ultracold Systems

Kenji Ohmori

RAPID COMMUNICATIONS

PHYSICAL REVIEW A **95**, 011403(R) (2017)

Ultracold-atom quantum simulator for attosecond science

Simon Sala, Johann Förster, and Alejandro Saenz
AG Moderne Optik, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße 15, 12489 Berlin, Germany

(Received 23 November 2016; published 25 January 2017)

PRL **103**, 260401 (2009) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2009

Pump-Probe Spectroscopy of Two-Body Correlations in Ultracold Gases

Christiane P. Koch[1,*] and Ronnie Kosloff[2]

Pump-Probe Experiments: Field Induced Alignment

Long history of electric-field induced alignment of molecules: Unique rotational dynamics for molecules such as I_2, N_2,...

From: "Colloquium: Aligning molecules with strong laser pulses", RMP 75, 543 (2003) by Stapelfeldt and Seideman (>1000 citations):

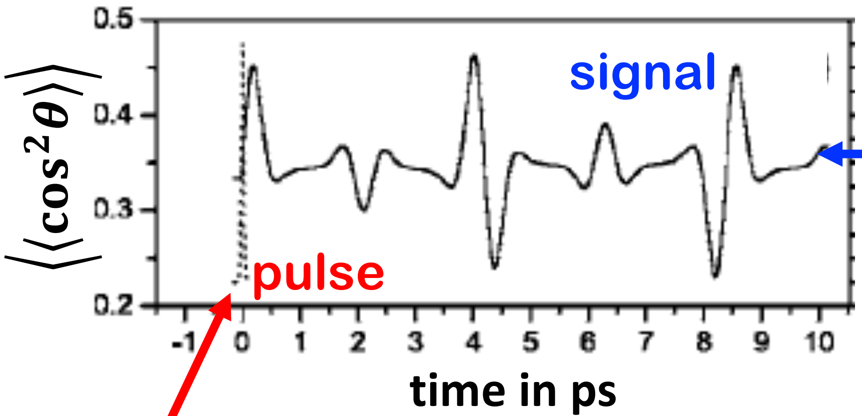
"We review the theoretical and experimental status of intense laser alignment—a field at the interface between intense laser physics and chemical dynamics with potential applications ranging from high harmonic generation and nanoscale processing to stereodynamics and control of chemical reactions."

Work on weakly-bound molecules adds "physical dynamics" to the list!

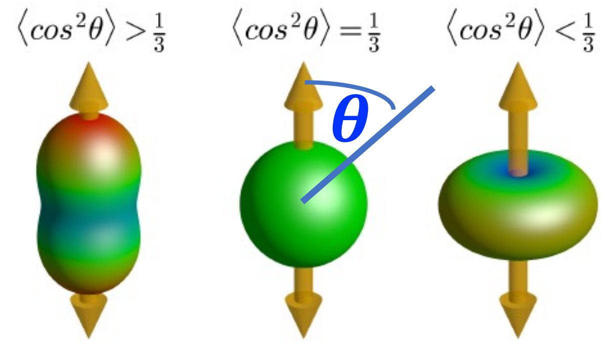
Alignment Signal $\langle\langle \cos^2\theta \rangle\rangle$ for N_2

Torres et al., PRA 72, 023420 (2005):

Integration over θ (R frozen; thermal distribution).
R: internuclear distance.
θ: angle between $\vec{R}$ and polarization vector.



Alignment signal
(1/3 ≡ spherically symmetric).



Pulse length 50 fs.

Intensity $2.5 \times 10^{13} \frac{\mathrm{W}}{\mathrm{cm}^2}$.

Impulse regime.

"Rotational revivals" require particular phase relation:

$E_J = B_0 J(J+1) - D_0 J^2 (J+1)^2$.

Typically:

$$E_{electronic} \gg E_{vibr.} \gg E_{rot.}$$

Can work in regime where one excites rotational wave packet.

In Contrast: Weakly-Bound Van der Waals Molecules

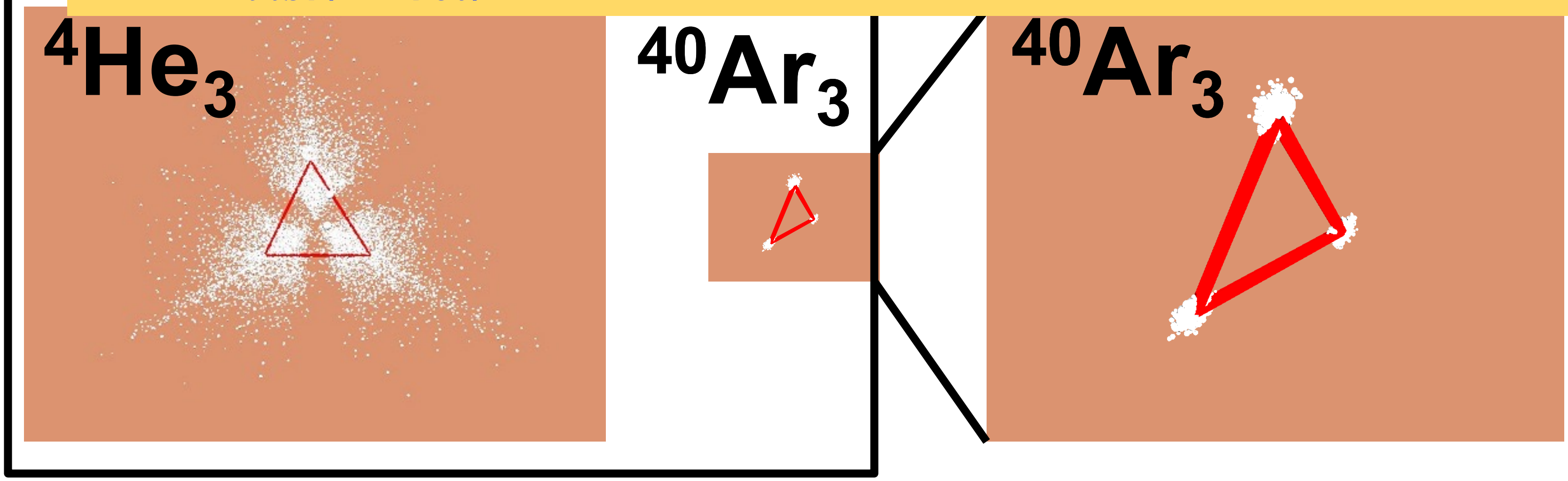
- ^{4}H
  at
  de
- Di

Typically: $E_{electronic} \gg E_{vibr.} \gg E_{rot.}$
Can work in regime where one excites rotational wave packet.

However, for light molecules, there is no clear ordering:

$$E_{vibr.} \sim E_{rot.}$$

For example:
He_2: only one bound state ($E_{vibr.}$ and $E_{rot.}$ "undefined").
He_3: two $J = 0$ bound states and no $J > 0$ bound states.
Ne_2: $E_{vibr.} \sim E_{rot.}$.

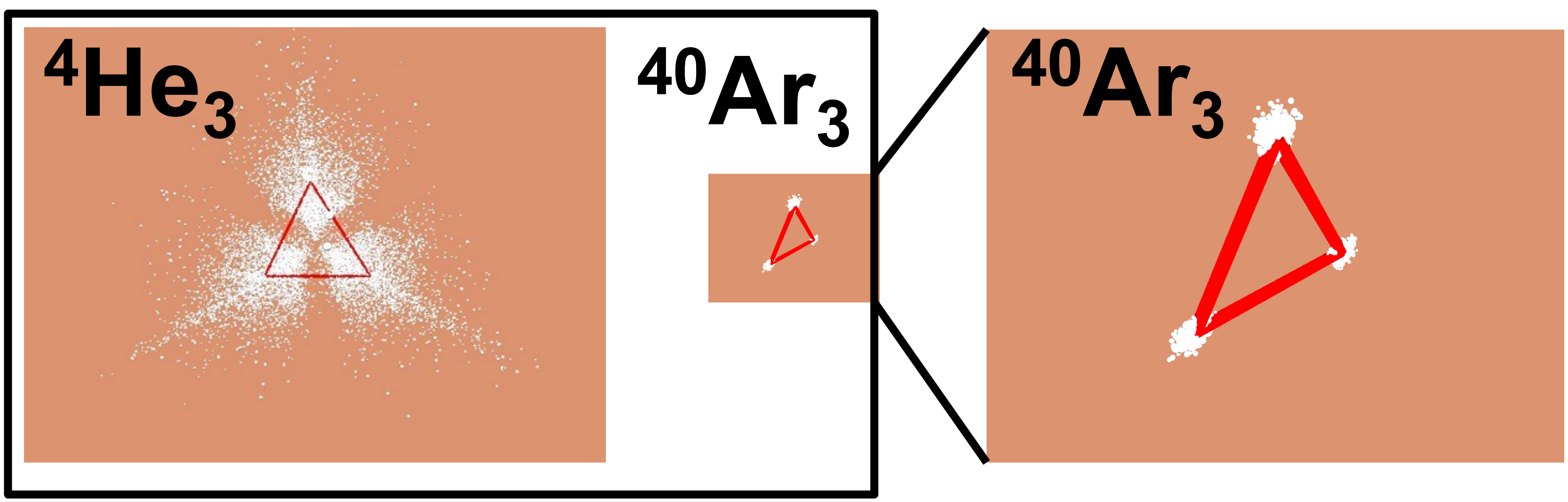


In Contrast: Weakly-Bound Van der Waals Molecules

- ^{4}He, ^{10}Ne, ^{20}Ar: composite bosons (energy scales are such that these atoms can be considered as point particles; consider only nuclear degrees of freedom).

- ^{4}He$_N$ binding energy: $E_{dimer} = -1.3$mK. $E_{trimer} = -131.8$mK and -2.65mK.
- ^{10}Ne-^{10}Ne binding energy: $E_{dimer} = -20.1$ K.
- ^{20}Ar-^{20}Ar binding energy: $E_{dimer} = -101$ K.

1 K = 8.6 x 10^{-5} eV



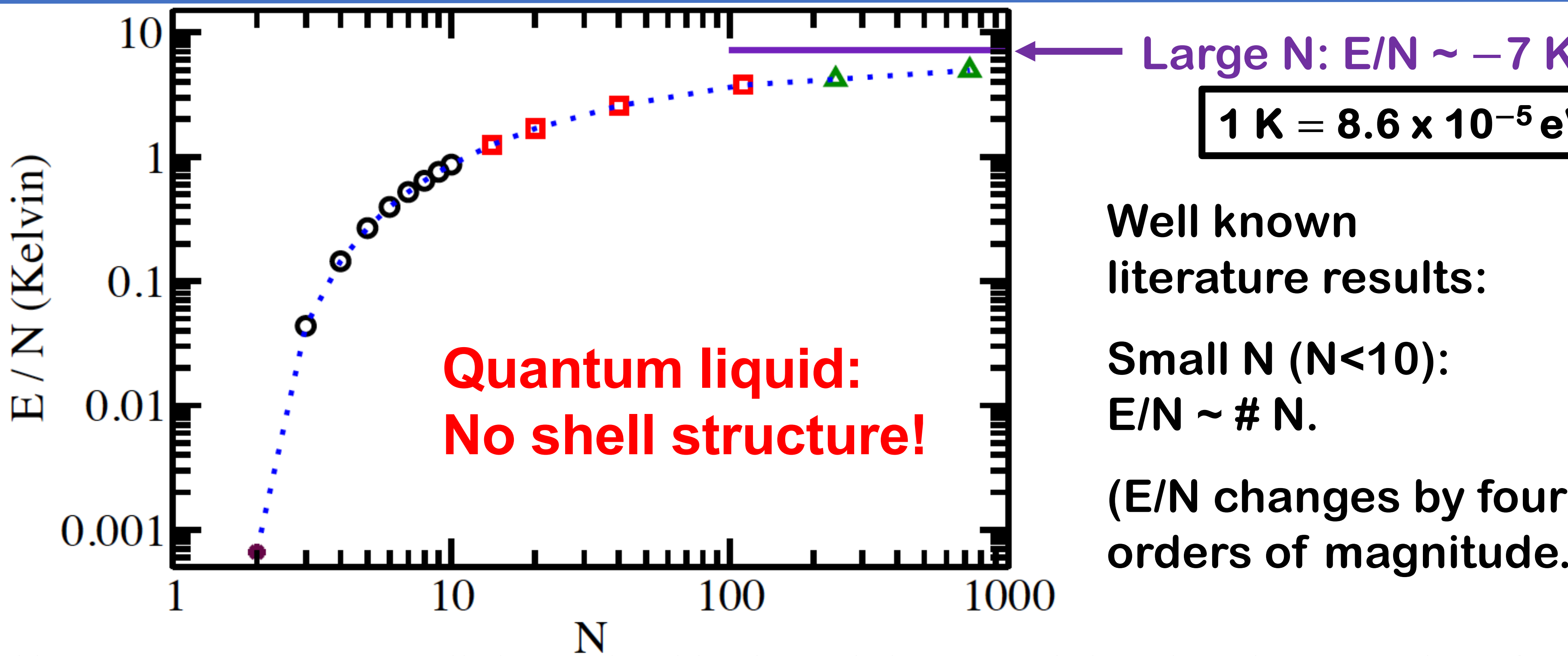
Why Weakly-Bound Van der Waals Molecules?

- Universality:
  - Nuclear physics: Deuteron (dimer), triton (trimer), and alpha-particle (trimer).
  - Helium dimer, trimer, and tetramer.
  - Atomic clusters as "model systems".

- Helium trimer state is Efimov state.

- Few- to many-body transition in highly diffuse quantum systems:
  - Ground state properties.
  - Dynamics probes excitation spectrum.

Bosonic Helium (^4He_N) Droplets = Quantum Liquid



1 K = 8.6 x 10^{-5} eV

Well known literature results:

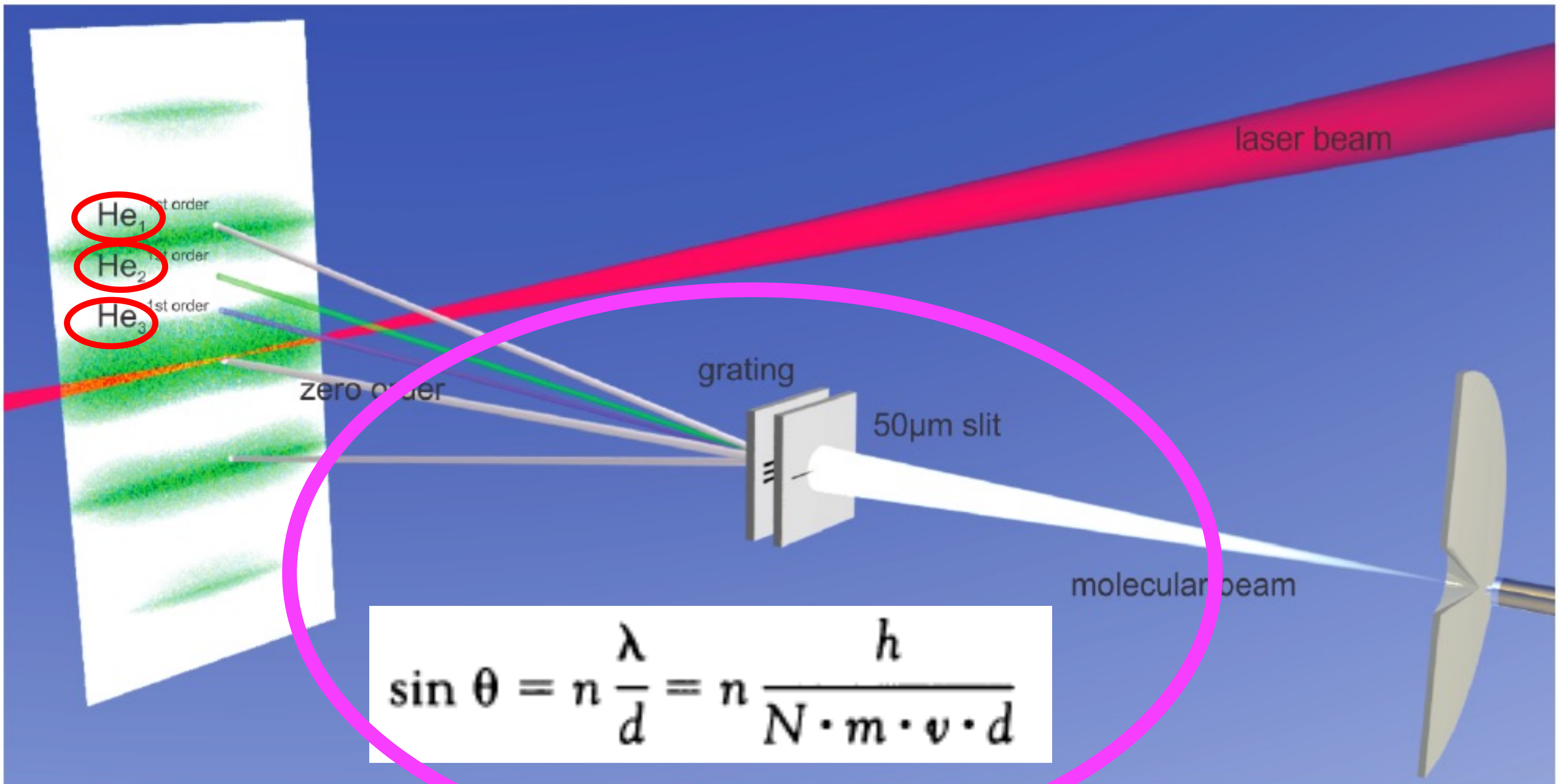
Small N (N<10): E/N ~ # N.

(E/N changes by four orders of magnitude.)

N>20 energies are well described by liquid drop model with volume and surface terms (no Coulomb, asymmetry, or pairing terms).

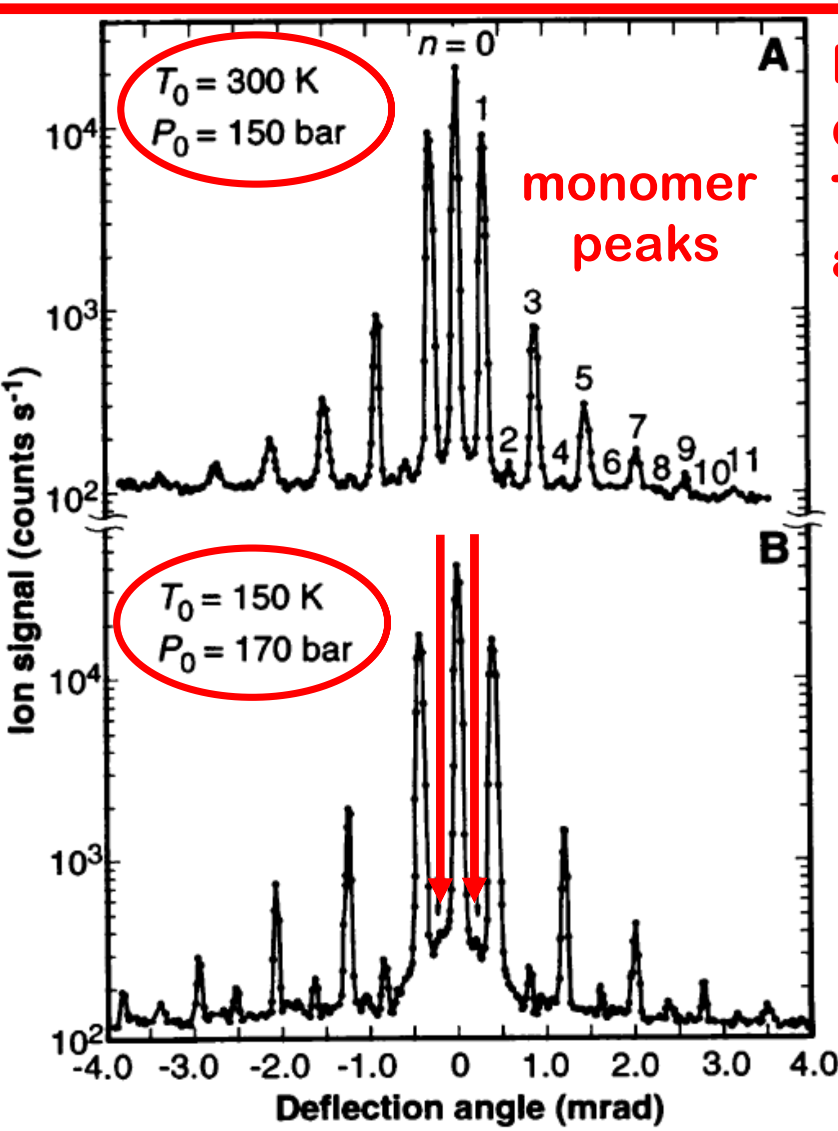
Rich interplay between many-body nuclear physics and quantum droplet community [e.g., Pandharipande et al., PRL 50, 1676 (1983); Stringari et al., JCP 87, 5021 (1987); Sindzingre et al., PRL 63, 1601 (1989)].

Creating Isolated Van der Waals Molecules



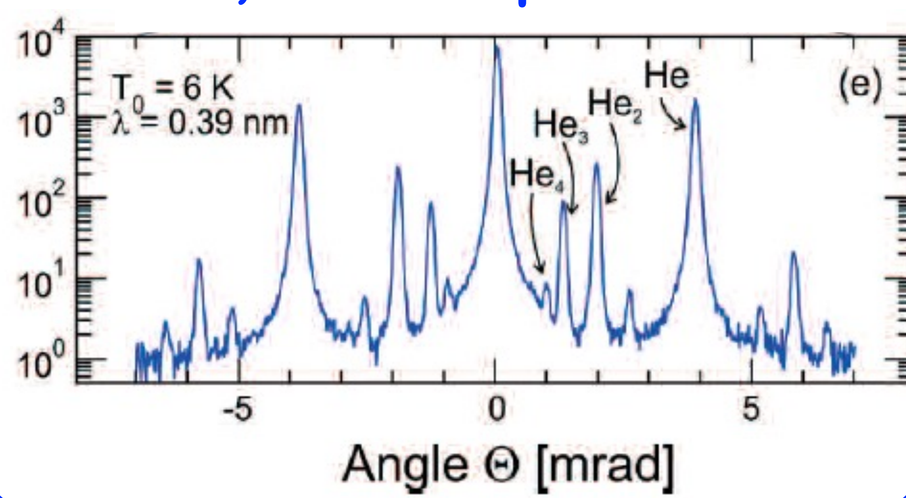
matter wave diffraction

Observation Of Bosonic Helium Dimer: $^4He_2$

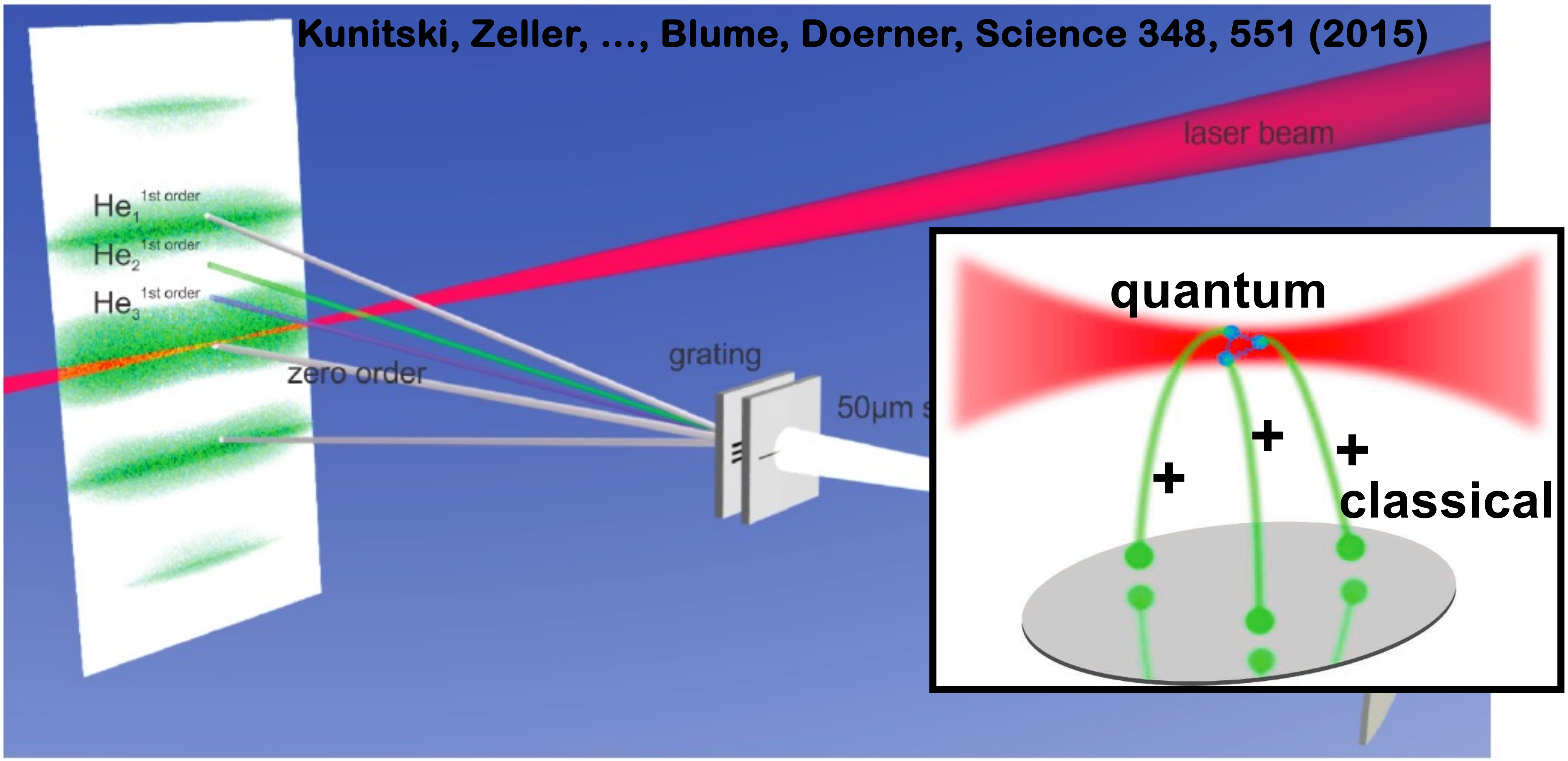


Fragile helium dimer forms in beam and can be isolated. Schoellkopf and Toennies, Science 266, 1345 (1994); see also Luo et al., JCP 98, 3564 (1993).

Nozzle temperature and pressure can be adjusted. Kornilov, Toennies, 10.1051/epn:2007003

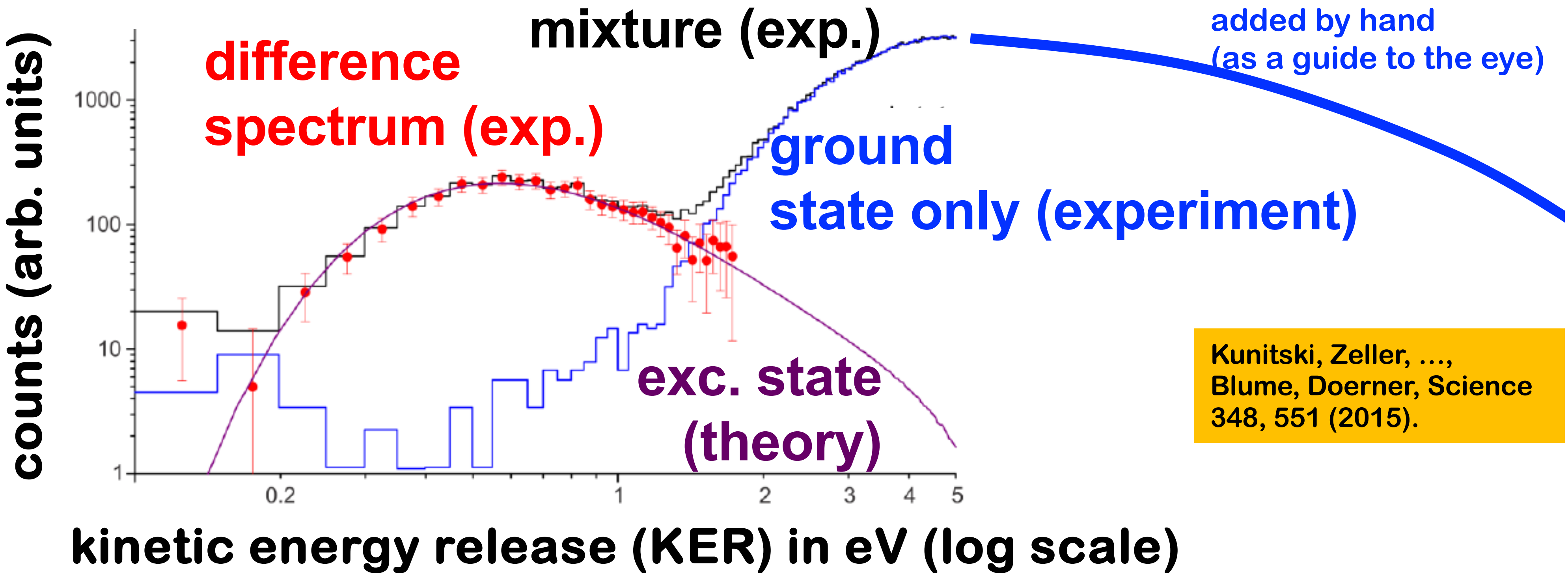


Imaging Helium Trimer (COLTRIMS, "Probe Only")



$^4He_3$ signal contains ground state trimer *and* excited state trimer.
Laser beam ionizes trimer: Coulomb explosion of $^4He_3$ (3 ions).

Kinetic Energy Release Measurement: Observing $(^4He_3)^*$

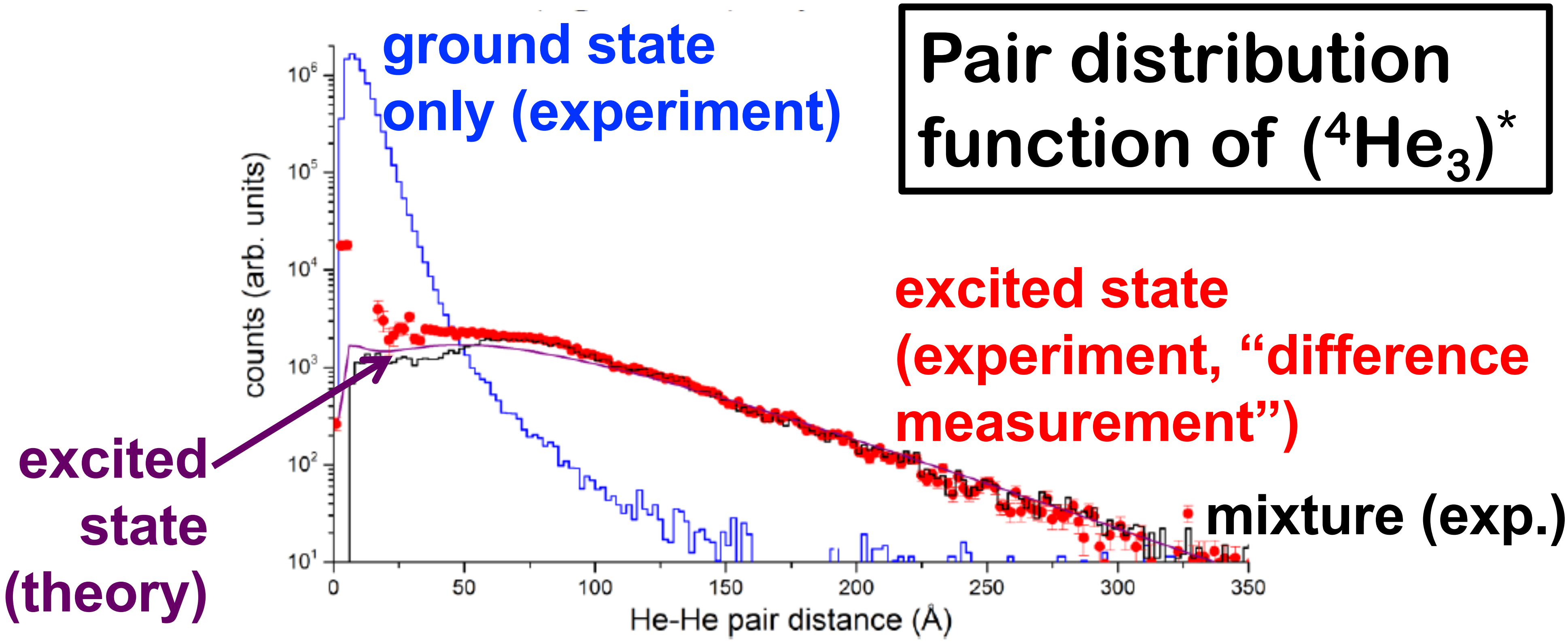


Kunitski, Zeller, …, Blume, Doerner, Science 348, 551 (2015).

The ionization is instantaneous and the He-ions are distributed according to the quantum mechanical eigen states of the ground and excited helium trimers.

Large r_{12}, r_{23} and r_{31} correspond to small $KER = 1/r_{12} + 1/r_{23} + 1/r_{31}$.

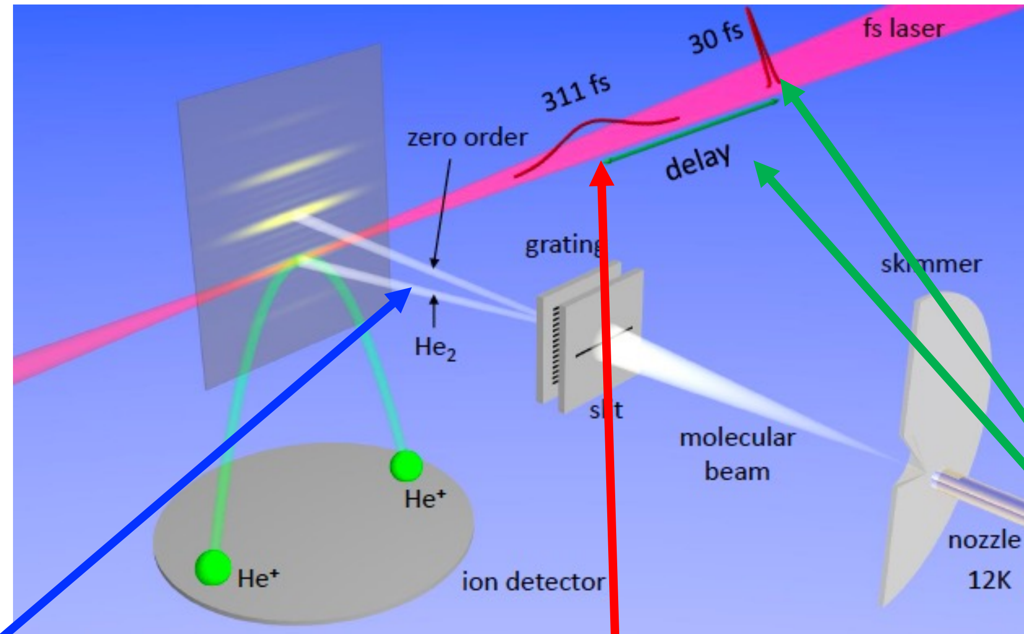
Reconstructing Real Space Properties



The excited state is eight times larger than the ground state.
Assuming an “atom-dimer geometry”, the tail can be fit to extract the binding energy of the excited helium trimer.
Fit to experimental data yields 2.6(2)mK.
Theory 2.65mK [Hiyama et al., PRA 85, 062505 (2012)].

Kunitski, Zeller, …, Blume, Doerner, Science 348, 551 (2015).

Basic Concept of Pump-Probe Spectroscopy



Prepare initial state (e.g., state that is dominated by s-wave scattering length).

Interrogate the initial state: fast and intense pump laser that takes the system out of equilibrium.

Wait for a variable time (delay) and apply even shorter and more intense probe laser that allows us to look at time-evolved system.

Two Identical Atoms (Diatomic Molecule) in E-Field

$H = H_0^{(1)} + H_0^{(2)} + \Delta H$. Unperturbed atomic Hamiltonian $H_0^{(1)}, H_0^{(2)}$.

Perturbation $\Delta H = -\vec{d}^{(1)} \cdot \vec{\mathcal{E}} - \vec{d}^{(2)} \cdot \vec{\mathcal{E}} + \frac{\vec{d}^{(1)} \cdot \vec{d}^{(2)} - 3(\vec{d}^{(1)} \cdot \hat{R})(\vec{d}^{(2)} \cdot \hat{R})}{R^3}$.

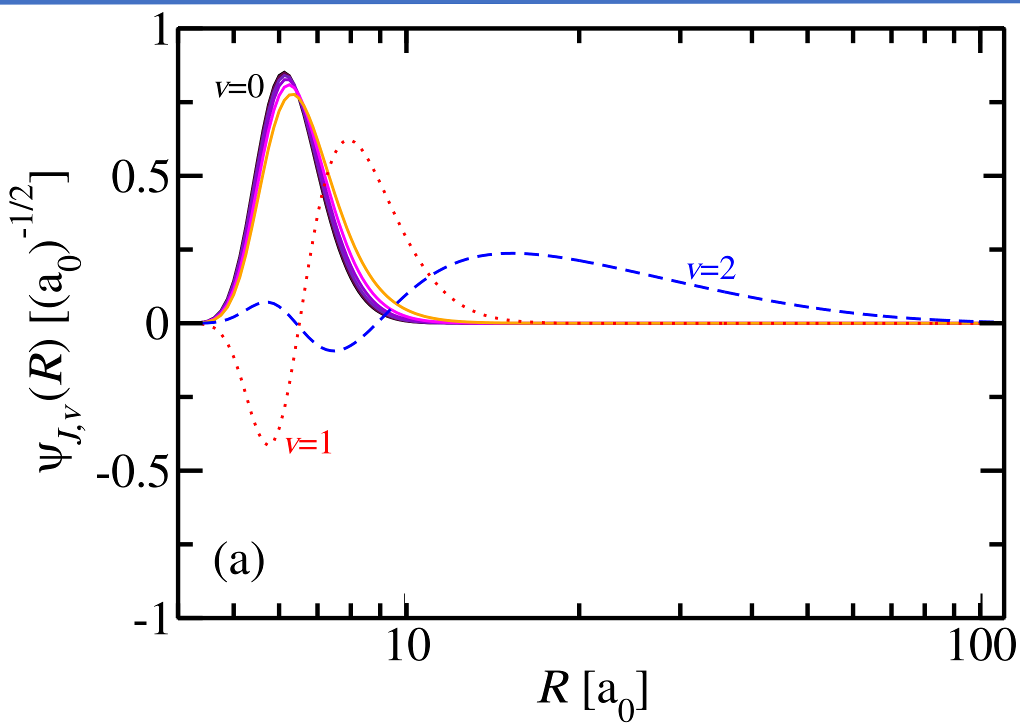
2nd-order PT: $\Delta E^{(2)} = -\frac{1}{2}\alpha\mathcal{E}^2 - \frac{1}{2}\alpha\mathcal{E}^2 - \frac{C_6}{R^6}$ ("trapping terms" + E-field independent long-range dipole-dipole interaction).

3rd-order PT: $\Delta E^{(3)} = \alpha^2\mathcal{E}^2 \frac{1-3\cos^2\theta}{R^3}$ (E-field induced interaction between two classical dipoles $\vec{D}^{(1)}$ and $\vec{D}^{(2)}$, where $\vec{D}^{(j)} = \alpha\vec{\mathcal{E}}$).

4th-order PT: $\Delta E^{(4)} = -4\sqrt{\pi}\alpha^3\mathcal{E}^2 \frac{Y_{00}(\theta) + \frac{1}{\sqrt{5}} Y_{20}(\theta)}{R^6} + \cdots$ (equal to 2nd-order PT shift of atom interacting with classical dipole).

Buckingham and Watts, Mol. Phys. 26, 7 (1973); Nielsen et al., PRL 82, 2844 (1999).

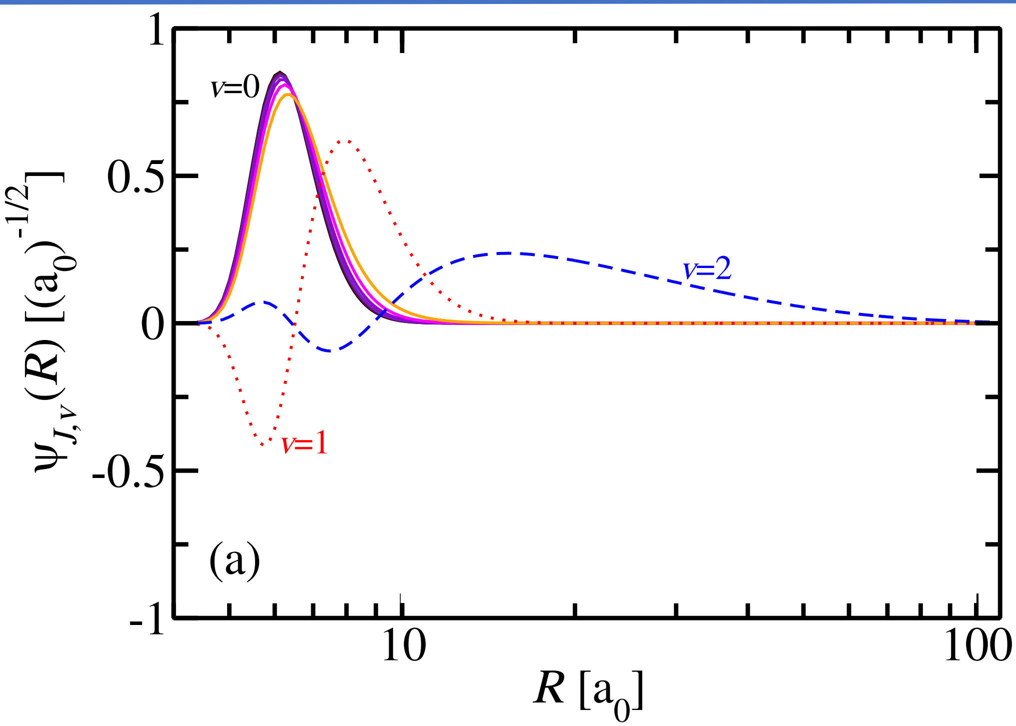
First Example: Neon Dimer



J	v	$E_{J,v}$ [K]	$E_{J,v}$ [K] [21]	$E_{J,\text{rigid}}$ [K]	$E_{J,\text{barrier}}$ [K]
0	0	−24.0939	−24.0941	−24.0939	0.0000
0	1	−4.2493	−4.2494		
0	2	−0.0188	−0.0187		
2	0	−22.7504	−22.7506	−22.7922	0.0999
2	1	−3.3838	−3.3838		
4	0	−19.6336	−19.6338	−19.7549	0.5977
4	1	−1.4399	−1.4398		
6	0	−14.7900	−14.7901	−14.9820	1.7885
6	res.		1.2791		3.9473
8	0	−8.3030	−8.3031	−8.4734	3.9473
10	0	−0.3186	−0.3187	−0.2293	7.3322
12	res.		8.8362	9.7505	12.1977
14		unbound		21.4658	18.8179

a) Rotational energy ~ vibrational energy: hybridization.

First Example: Neon Dimer



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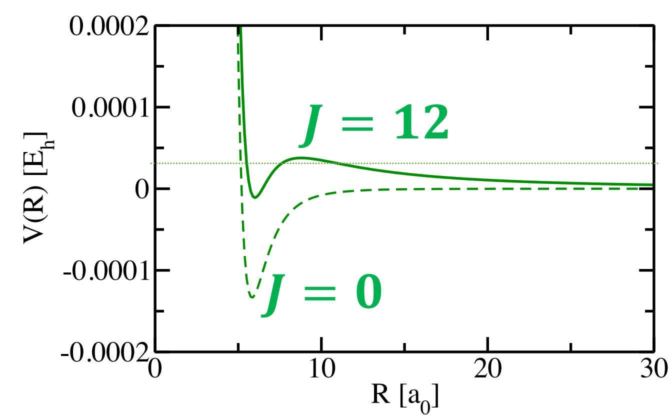
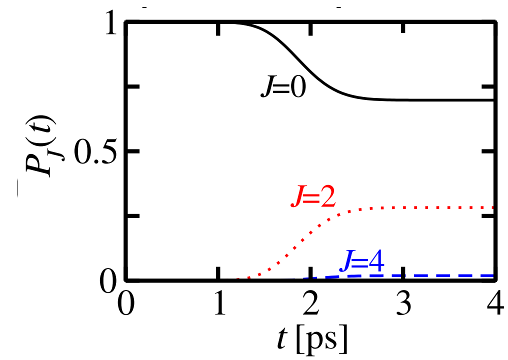
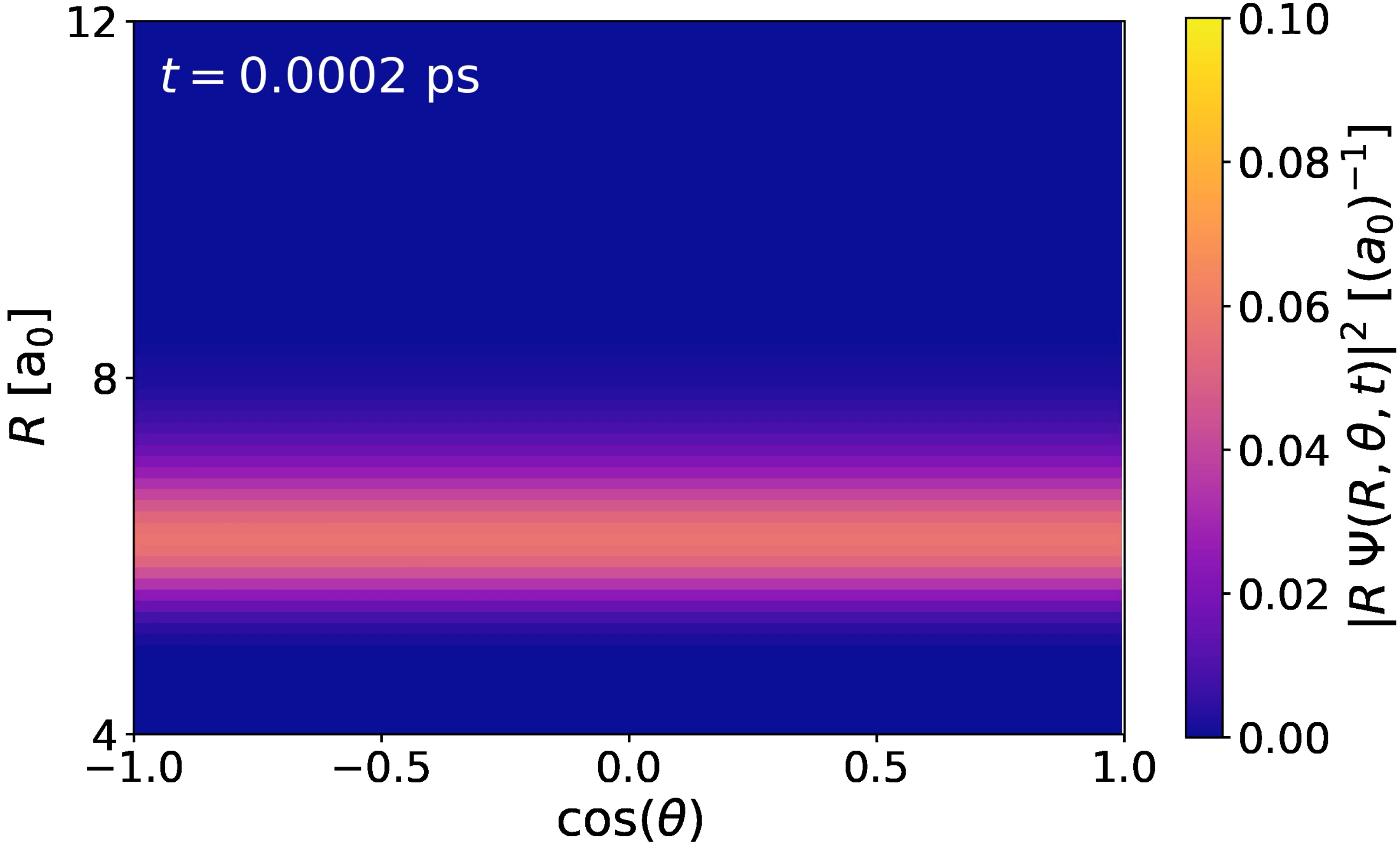


a) Rotational energy ~ vibrational ener

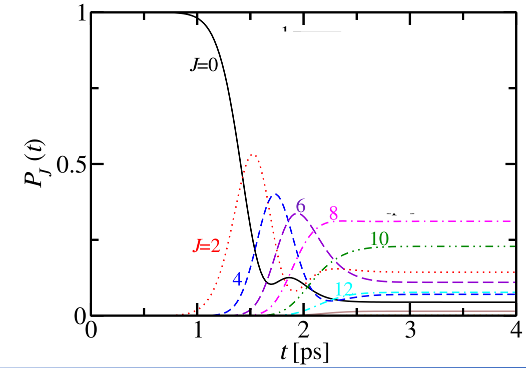
b) Resonance states: tunneling.

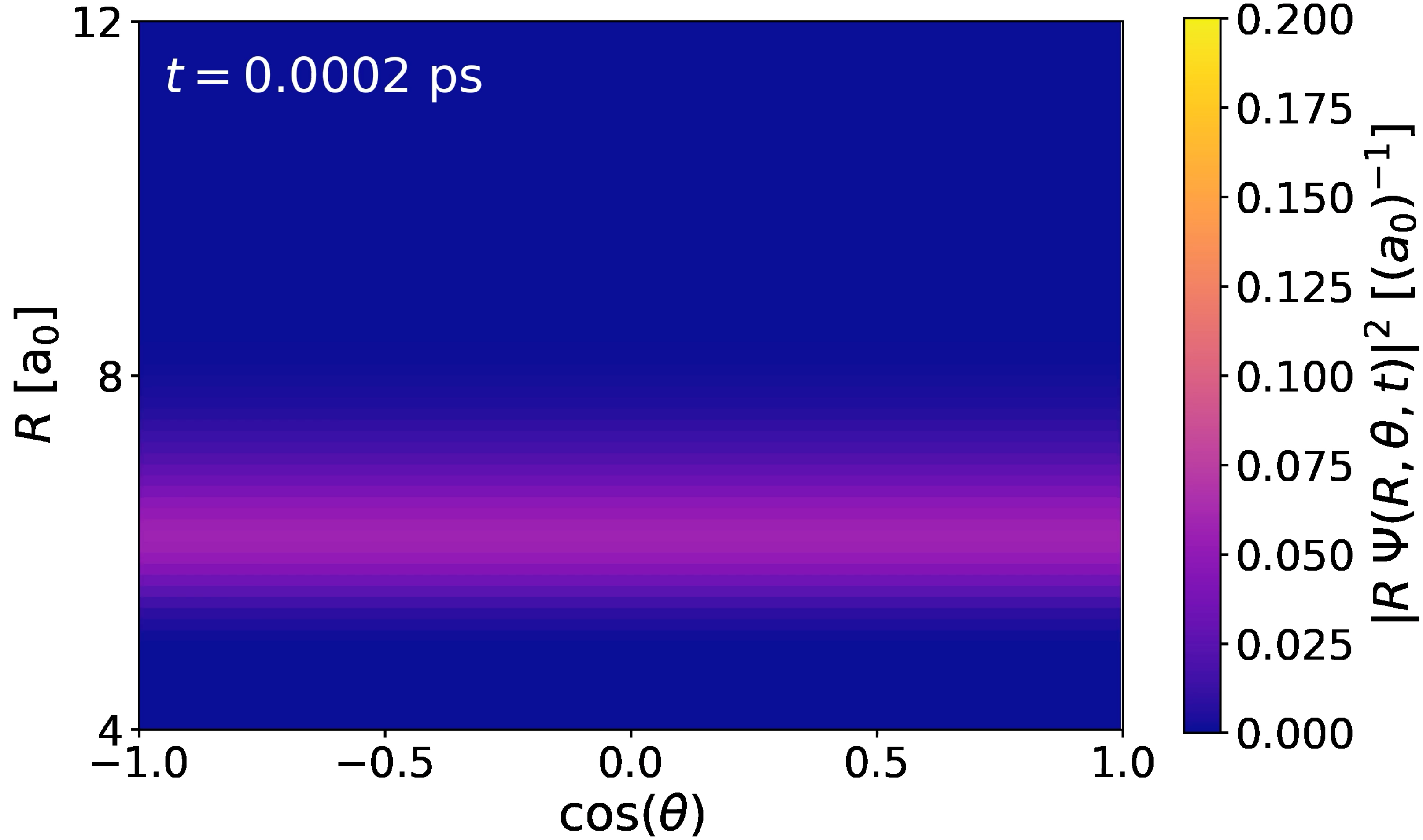
Density Evolution: Low Kick Strength



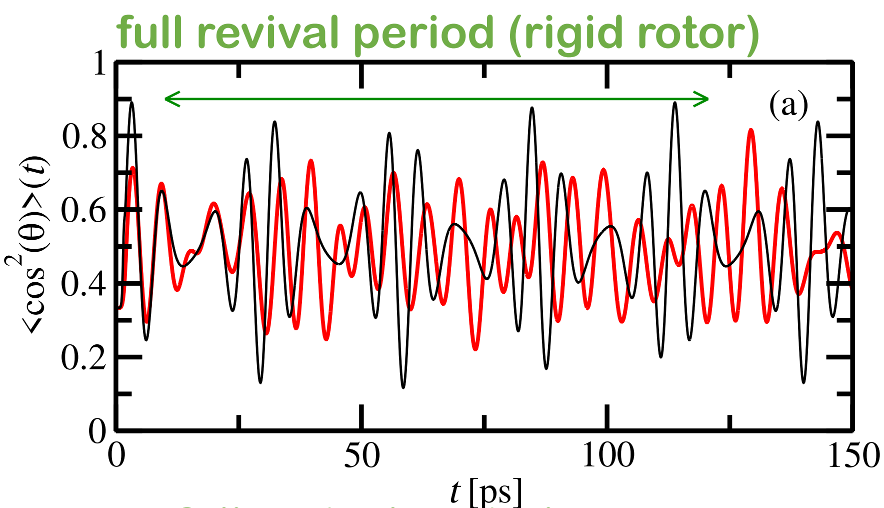


Density Evolution: High Kick Strength



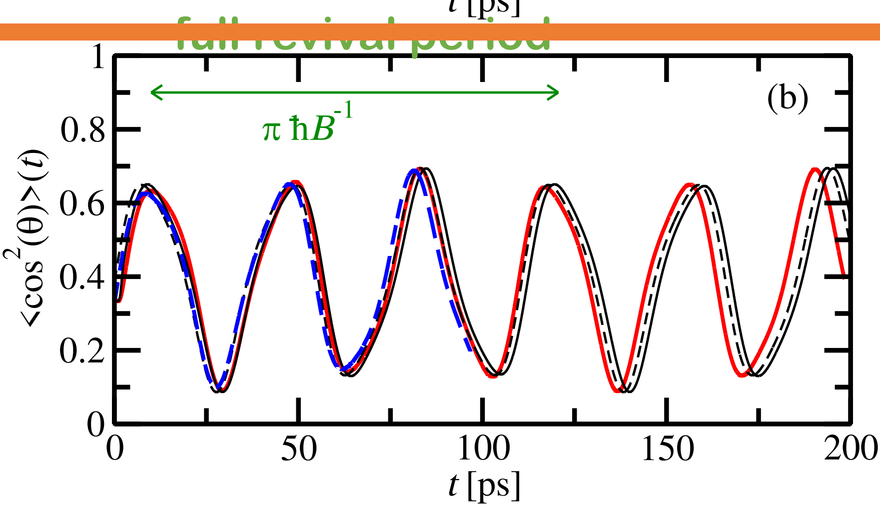


First Example: Neon Dimer Alignment Signal



"High" kick strength of ~10.

Red: Full calculation.
Black: Rigid rotor model (neglects vibrational motion).
Hybridization (vibrational degree of freedom cannot be neglected).

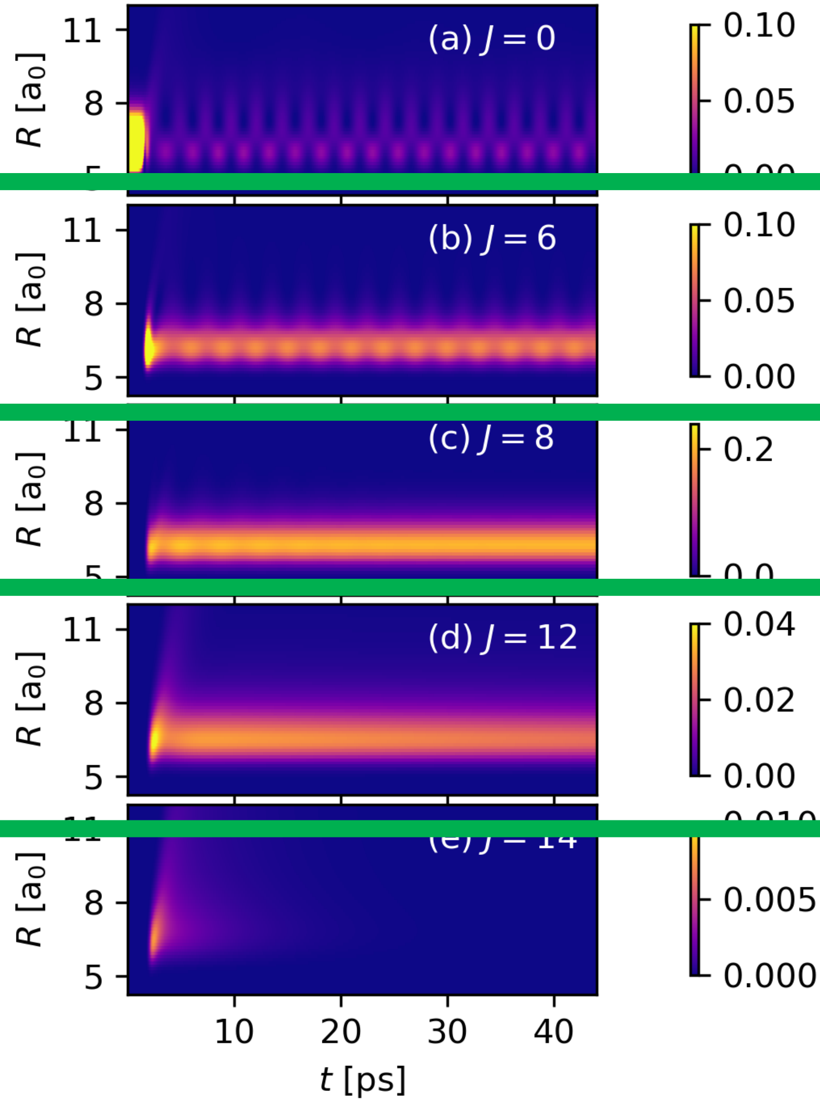


"Low" kick strength of ~2.

Red: Full calculation (long pulse and low intensity).
Blue: Full calculation (short pulse and high intensity).
Black: Rigid rotor model model (neglects vibrational motion).

First Example: Fingerprint of Resonance State

Density in *J*-channel



$J = 0$: Supports three bound states.

$J = 6$: Supports ground state and resonance state.
Oscillations are due to vibrationally excited resonance state.

$J = 8$: Supports one bound state.

$J = 12$: Supports only resonance state.
Finite population is due to resonance state (tunneling).

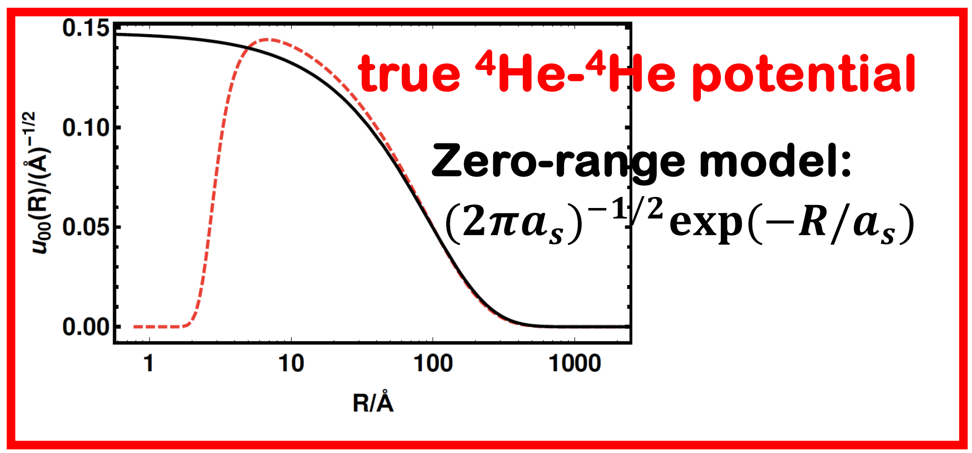
$J = 14$: Does not support bound state.

Second Example: Helium Dimer

- ^{4}He-^{4}He bound state energy $E_{dimer} = -1.625$mK.
- No J > 0 bound states.
- ^{4}He-^{3}He does not support bound state.
- Two-body s-wave scattering length $a_s = 170.86a_0$.
- Two-body effective range $r_{eff} = 15.2a_0$
  (alternatively, two-body van der Waals length $r_{vdW} = 5.1a_0$).

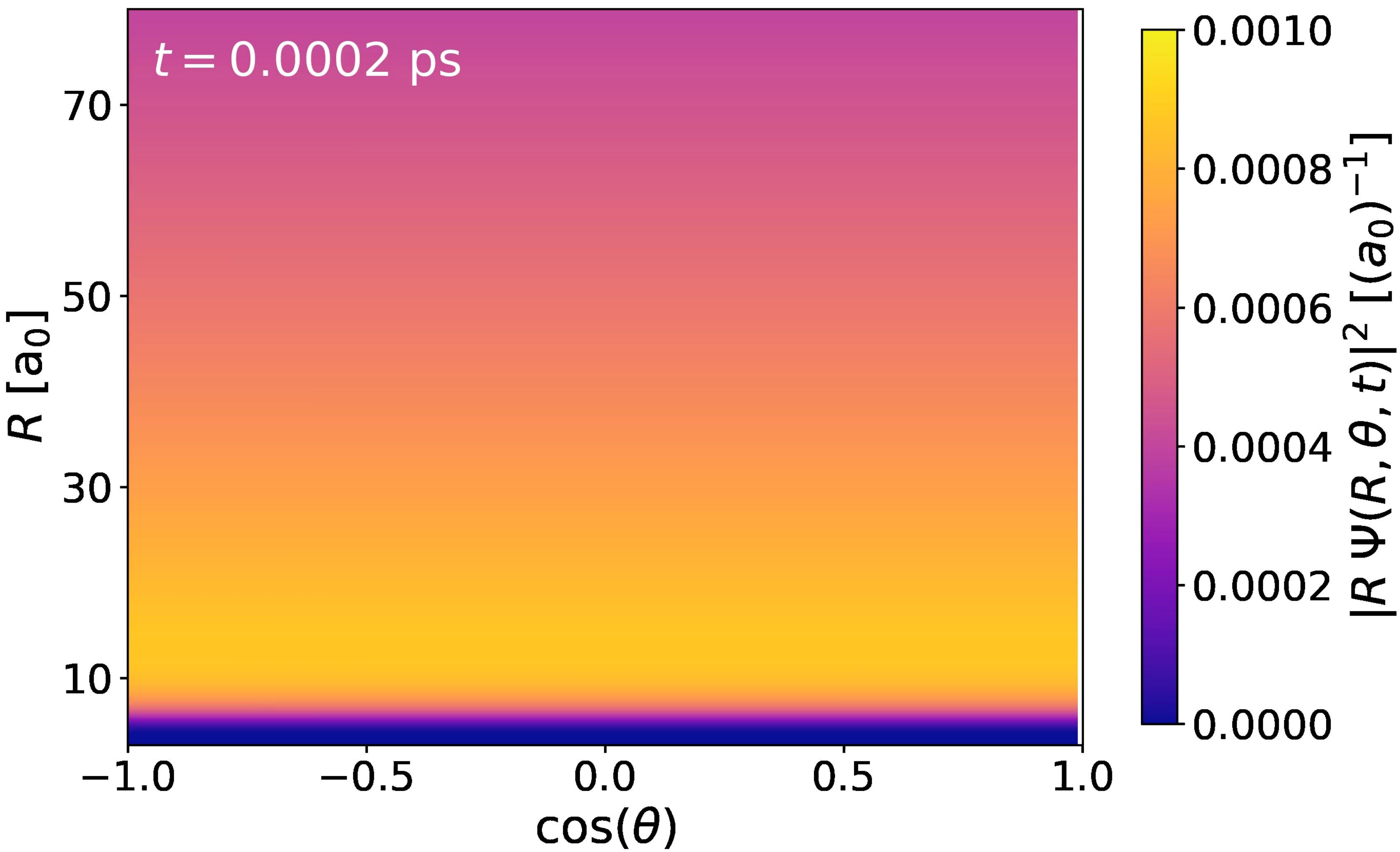
Large positive a_s:

- Reminiscent of Feshbach molecules observed in the ultracold.
- Here: universal dimer is the true ground state.



Born-Oppenheimer potential curves tractable by *ab initio* methods (quantum chemistry + asymptotics).

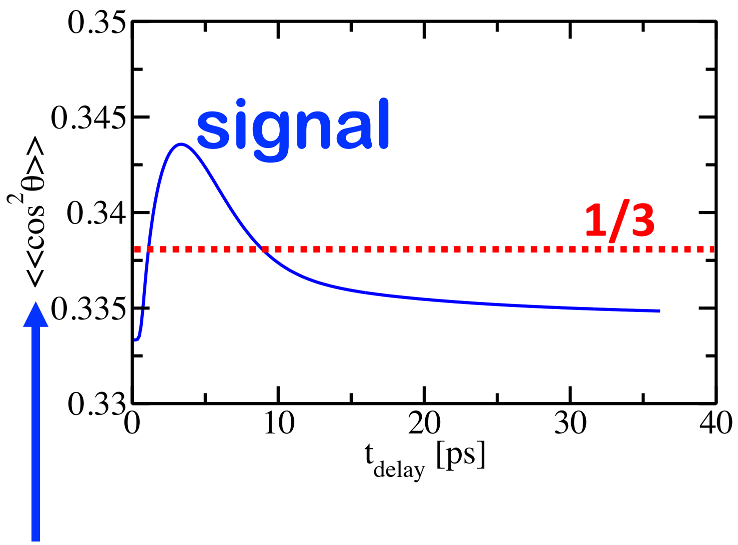
Helium Dimer: Density Evolution



Pump-Probe Spectroscopy of ^{4}He-^{4}He: No Rotational Revivals

Variety of theory predictions:
Friedrich et al., Collect. Czech. Chem. Commun. 63, 1089 (1998); Nielsen et al., PRL 82, 2844 (1999); Bruch, JCP 112, 9773 (2000).

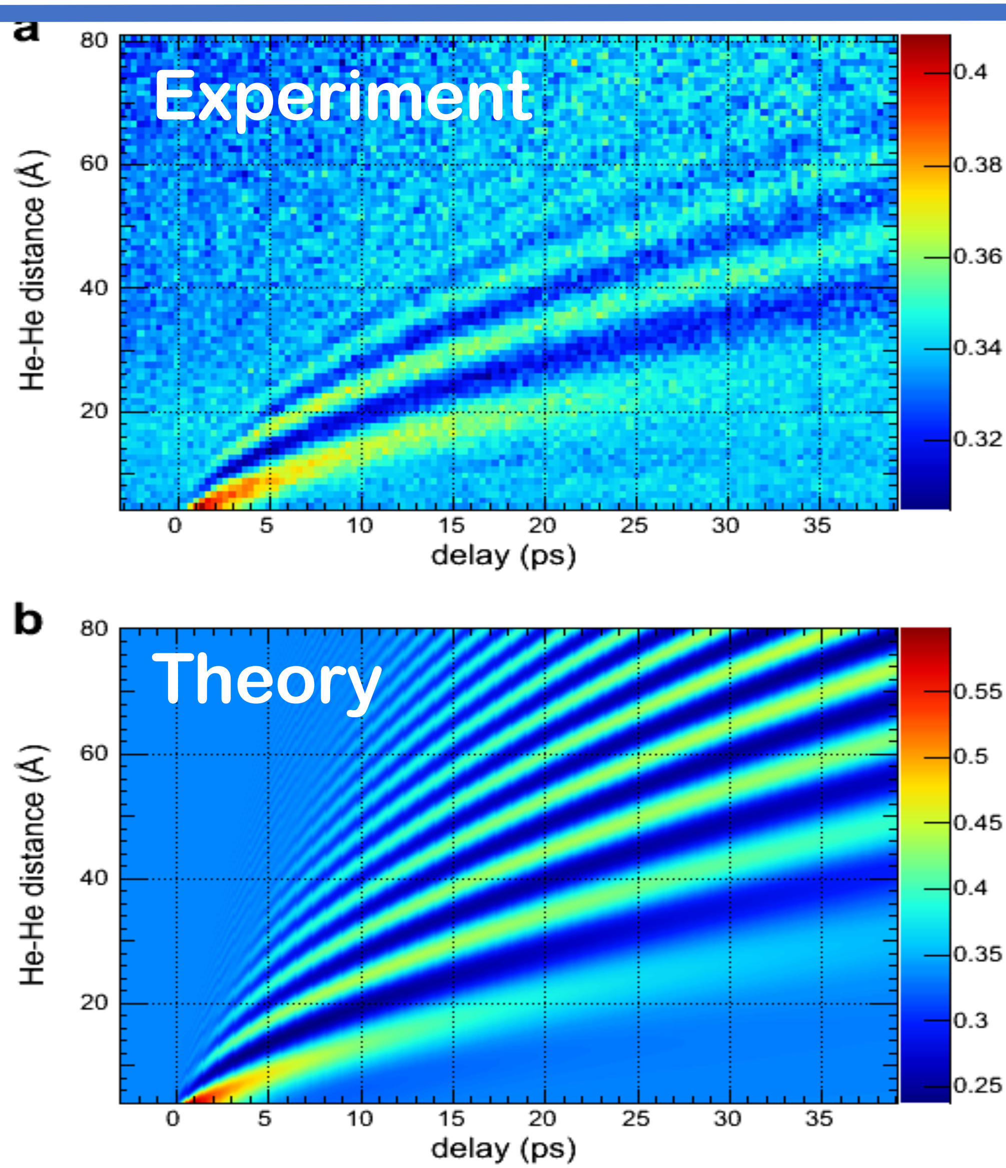
After averaging over *R*: Tiny signal.



Averaged over *R* and *θ*: Maximum change of 3%.

Disappointingly low response…

Distance-Resolved Alignment Signal $\langle \cos^2\theta \rangle(R,t)$



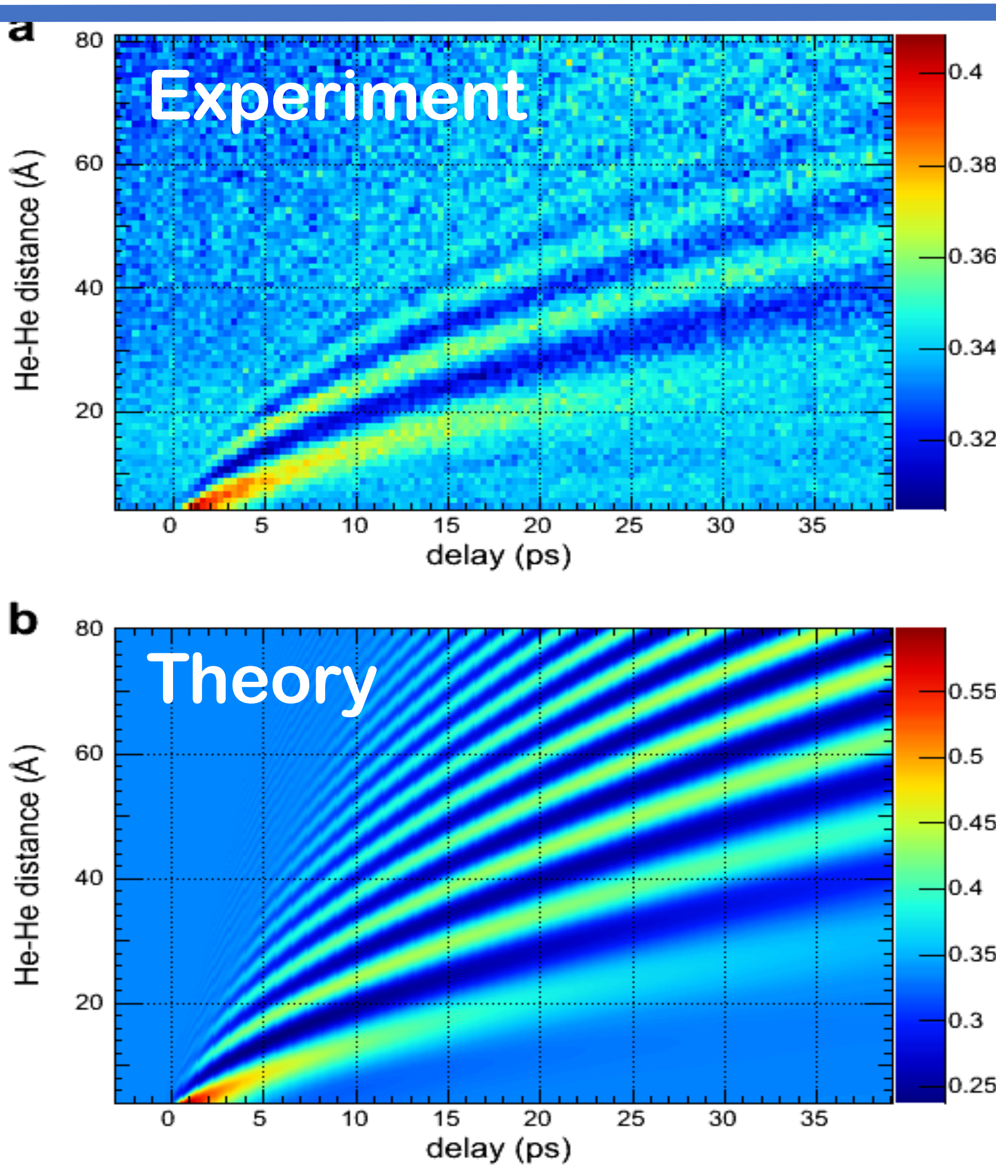
No integration over R !!!

$$\langle \cos^2\theta \rangle = \frac{\int_0^\pi \Psi^*(R,\theta,t)\cos^2\theta\Psi(R,\theta,t)\sin\theta d\theta}{\int_0^\pi |\Psi(R,\theta,t)|^2 \sin\theta d\theta}$$

Experimental data by Maksim Kunitski, Reinhard Doerner et al. (Frankfurt University)

Agreement is qualitative but not quantitative.

Distance-Resolved Alignment Signal $\langle \cos^2\theta \rangle (R,t)$



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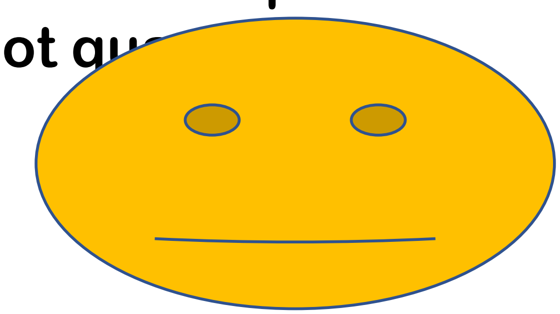
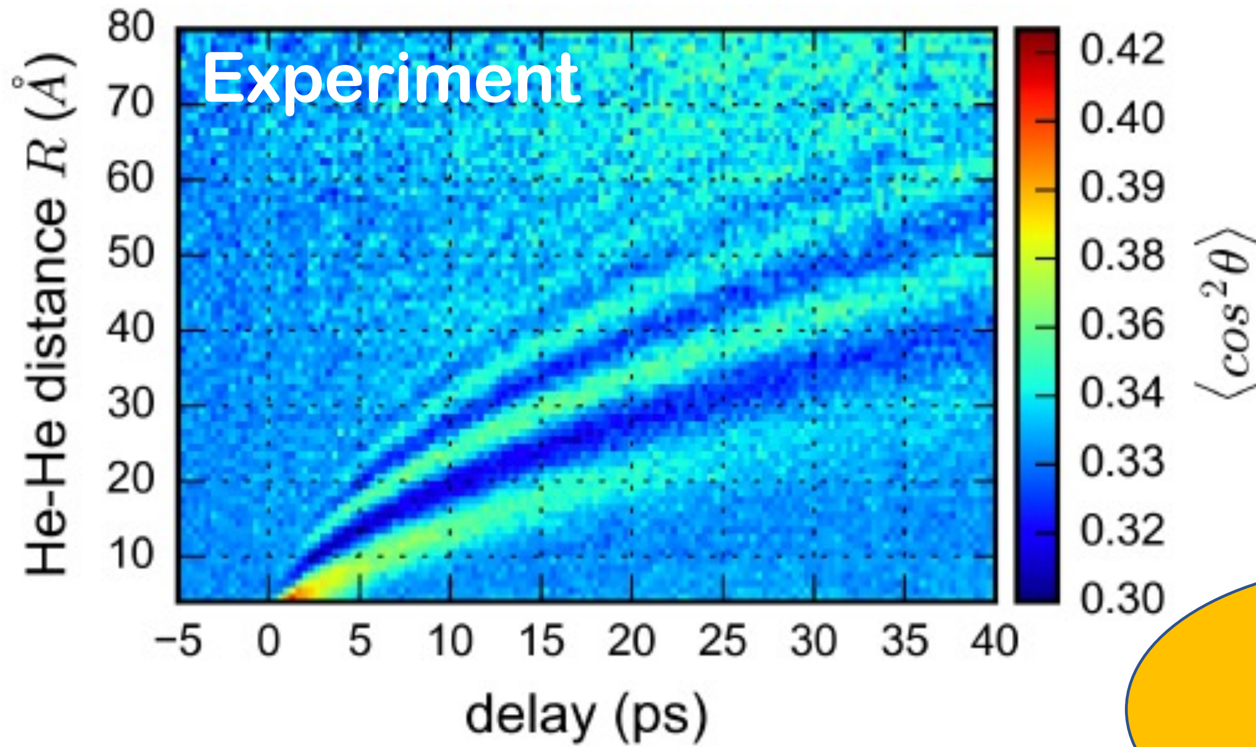
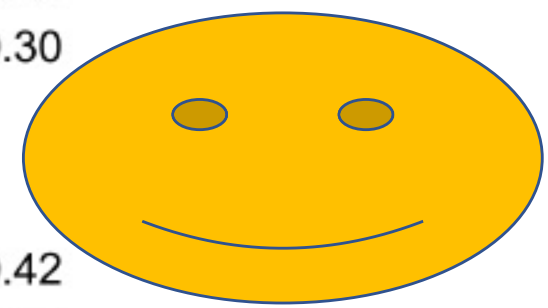
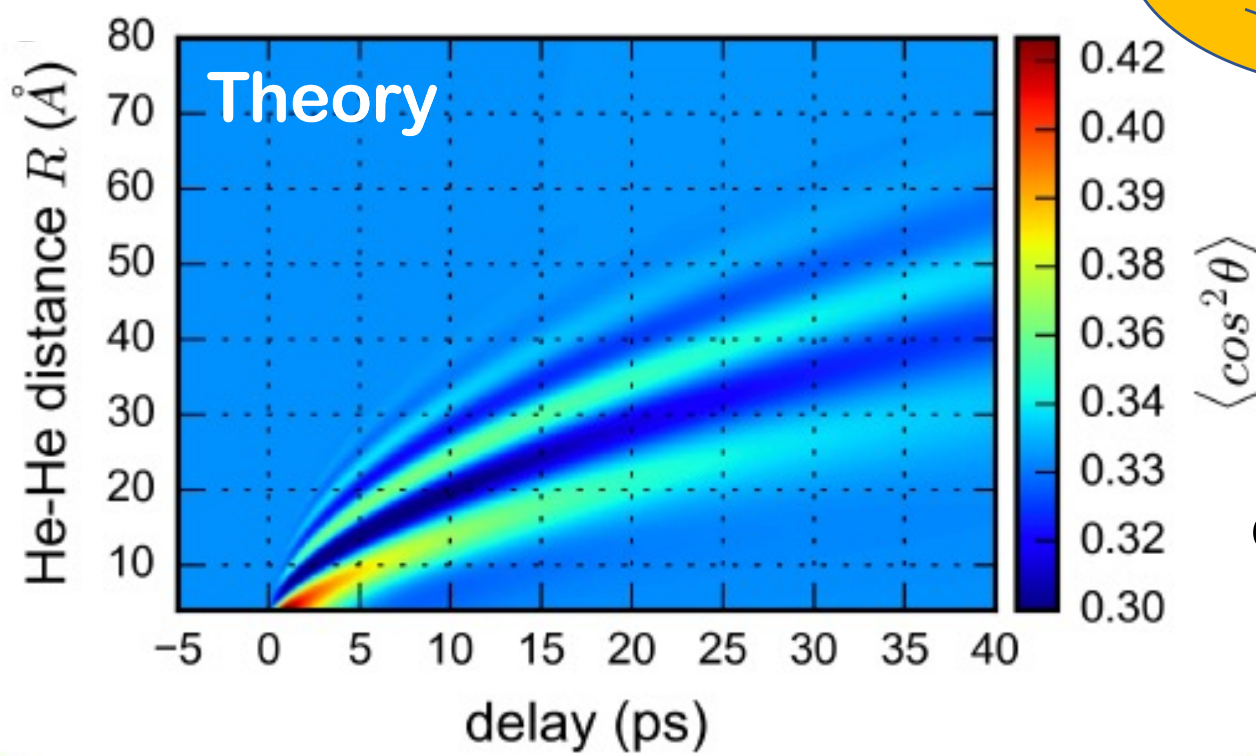






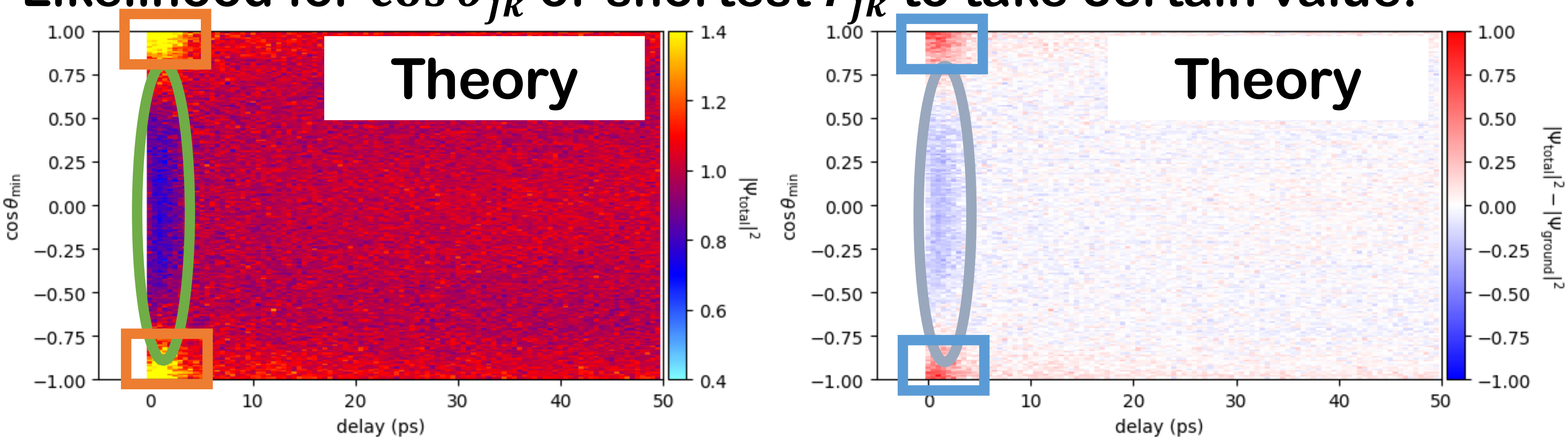
Parameter-free theory (using measured pulse length, intensity, spatial imaging resolution)



Kunitski, Guan,…,Blume, Doerner, Nature Physics (2021).

Third Example: Helium Trimer (A First Glimpse)

Likelihood for $\cos\theta_{jk}$ of shortest $\vec{r}_{jk}$ to take certain value:



Provides us with a tool to explore how the presence of third particle “perturbs” the dynamics of the other two particles in spatially- and time-resolved manner:

Access to time scale required to redistribute energy!

Summary

- Unambiguous detection and characterization of excited helium Efimov trimer.

- Entirely new regime: Pump-probe spectroscopy (pump = laser kick) of weakly-bound molecules.
- Examples:
  - Neon dimer (theory).
  - Helium dimer (theory and experiment).
  - Helium trimer (theory and experiment comparison in progress).

- Observed rich interplay of rotational and vibrational degrees of freedom.
- Completely different from rotational revivals observed for heavy molecules.


Any questions, comments, discussion?