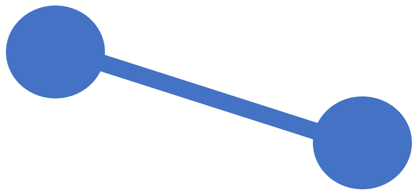


Dynamics Of The Helium Dimer

Doerte Blume
Center for Quantum Research and Technology (CQRT)
Department of Physics and Astronomy
The University of Oklahoma, Norman.

Supported by the NSF.

This work was done in collaboration with Qingze Guan and Reinhard Doerner's group at Frankfurt U. (lead postdoc Maksim Kunitski).

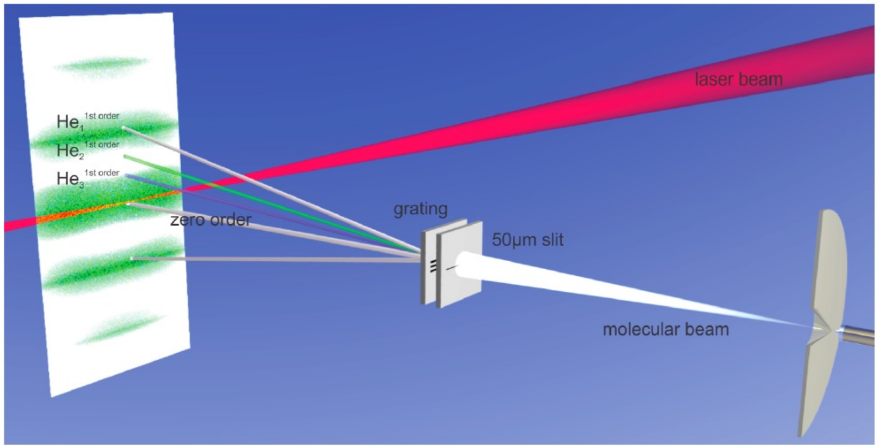
Helium Droplets: Cold But Not Ultracold Samples

Cold ^{4}He atoms (sub-Kelvin temperatures):

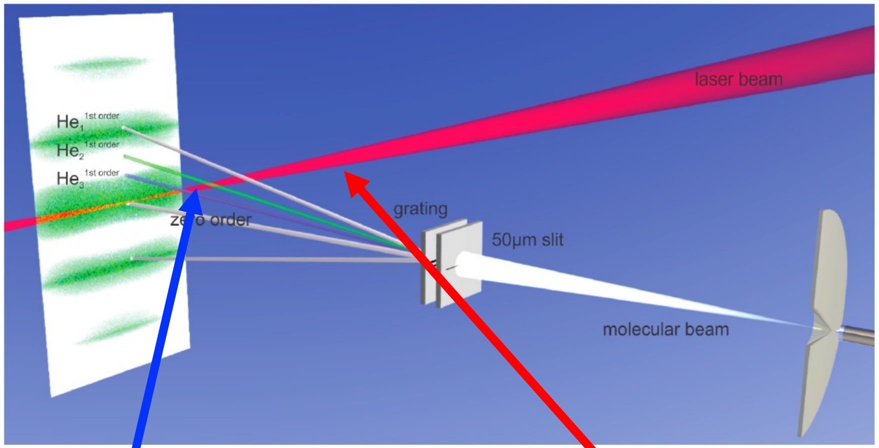
Two-body (real-time dynamics) → today's topic…

Three-body (three-body Efimov state;
no real-time dynamics) → see Pascal Naidon's talk…

Size-selected nozzle beam expansion experiments and theory



Basic Concept

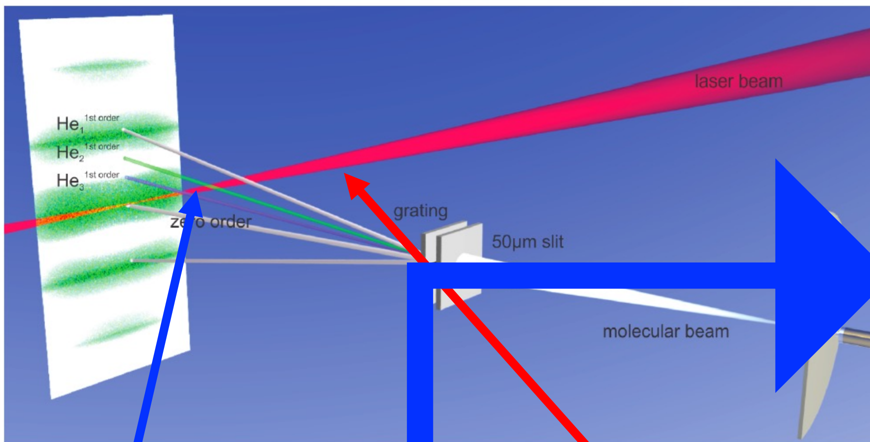


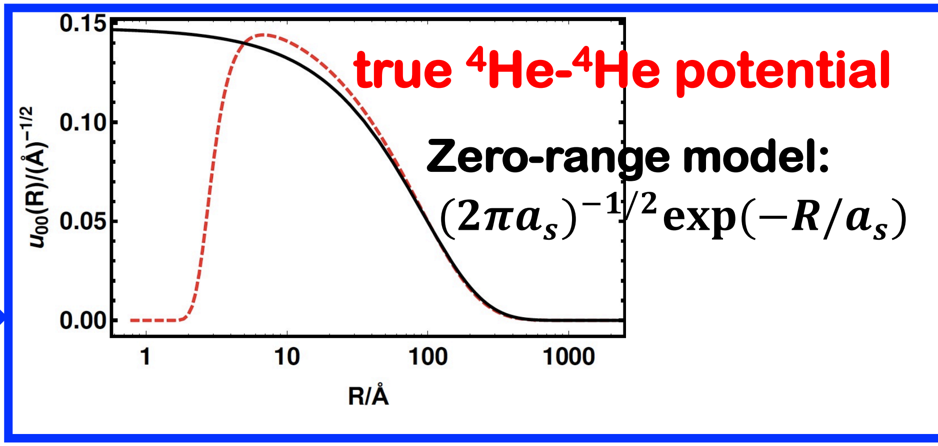
Prepare universal initial state (i.e., state that is dominated by s-wave scattering length).

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.

Basic Concept





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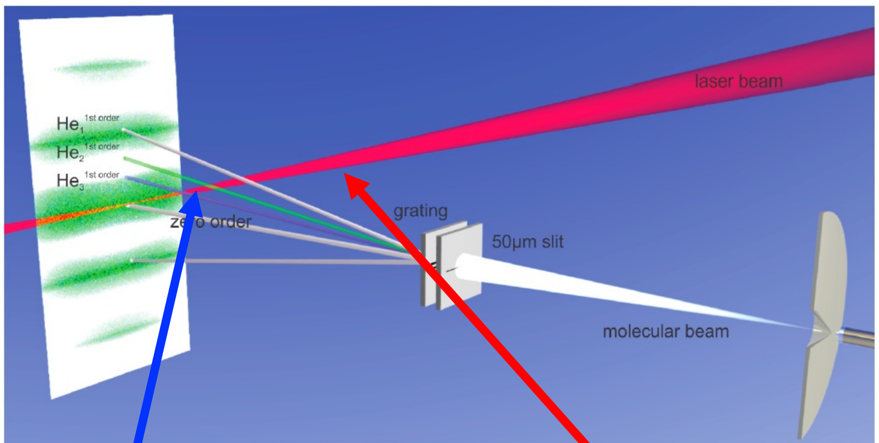
Basic Concept



Sort of like…

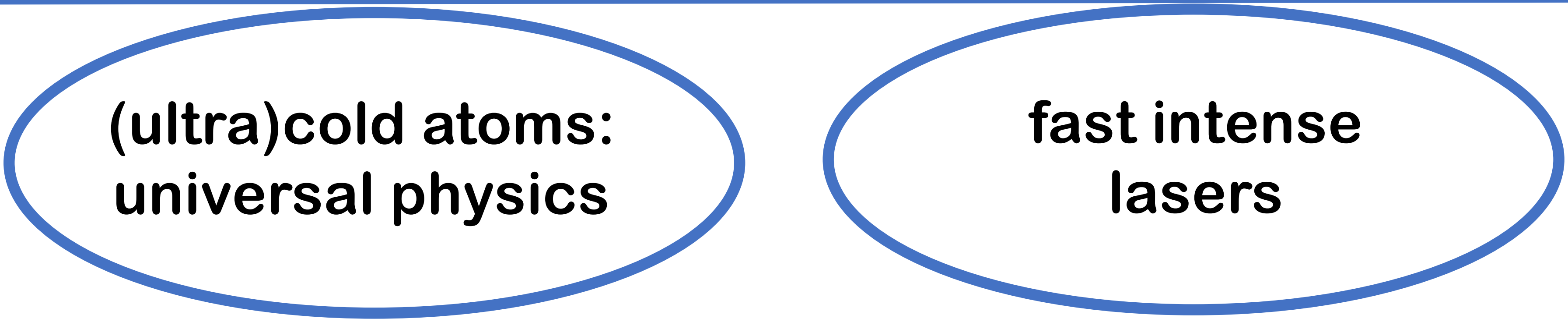
From vectorstock.com

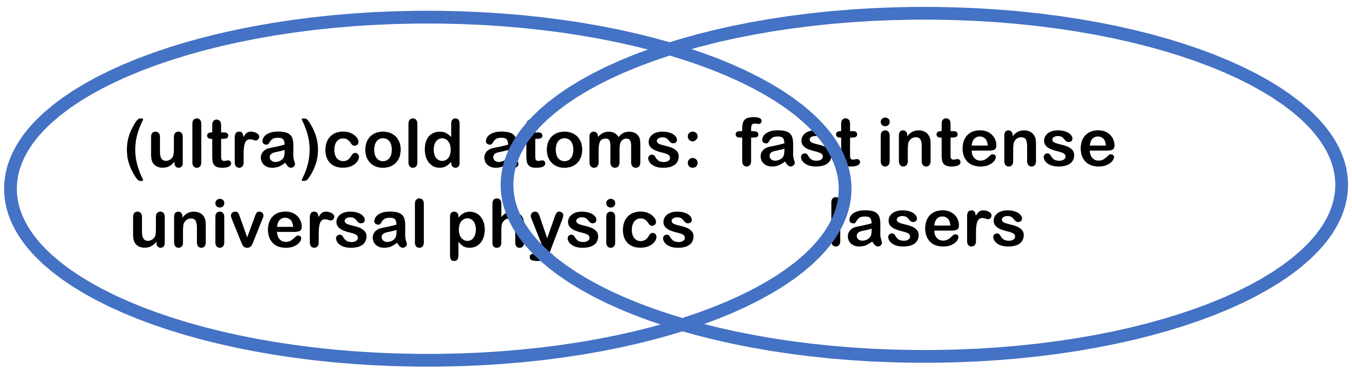
Prepare universal initial state (i.e., state that is dominated by s-wave scattering length).

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 Exciting Fields





One may hope: Two good things combined should be better than two good things separated...

But you may object: Aren't we just gonna blow everything up?

Yes, we will… and it's fun and useful…

Selected Works in This Direction

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], … E. Dotti[1], Kurt M. Fujiwara[1], Kevin … [1]
Zachary A. Geiger[1] & David M. Weld[1]

(ultra)cold atoms: universal physics

fast intense lasers

RAPID COMMUNICATIONS

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

Optically Engineered Quantum States in Ultrafast and Ultracold Systems

Kenji Ohmori

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]

Helium Dimer

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

- **Using modern Born-Oppenheimer potential:**
  - **^{4}He-^{4}He bound state energy $E_{dimer} = -1.625$ mK.**
  - **Two-body s-wave scattering length $a_s = 170.86\ \mathrm{a_0}$.**

- **Question: Is the ^{4}He dimer universal?**

Helium Dimer

$1\ \text{K} = 8.6 \times 10^{-5}\ \text{eV}$

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- **Question: Is the ^{4}He dimer universal?**

- **Strategy:**
  - **Convert to consistent set of units.**
  - **Dimer energy according to zero-range theory (one-parameter theory).**
  - **Correction? Dimer energy according to effective-range theory (two-parameter theory → need second parameter).**

Helium Dimer

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

- Using modern Born-Oppenheimer potential:
  - ^{4}He-^{4}He bound state energy $E_{dimer} = -1.625$ mK.
  - Two-body s-wave scattering length $a_s = 170.86\ \mathrm{a_0}$.

- **Question: Is the ^{4}He dimer universal?**

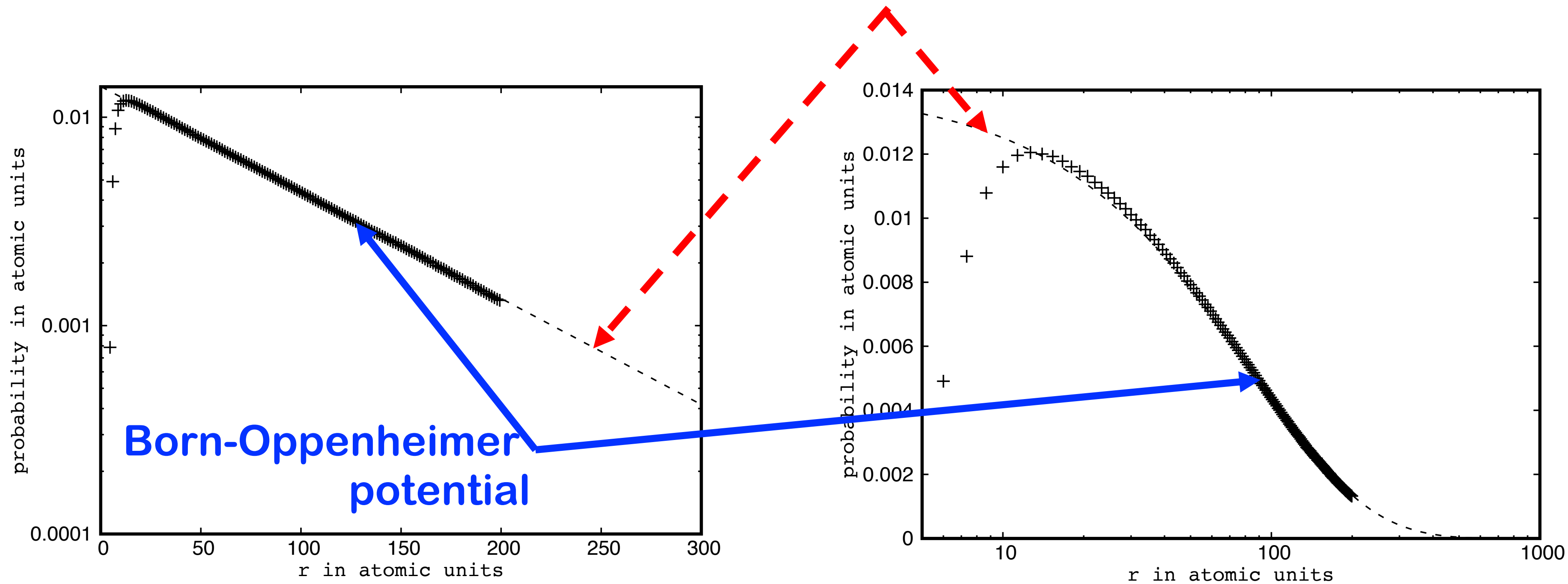
  - Convert E_{dimer} to atomic units: $E_{dimer} = -5.147 \cdot 10^{-9}$ a.u.

  - Zero-range theory: $E_{dimer} = -\frac{\hbar^2}{m\, a_s^2} = -4.69 \cdot 10^{-9}$ a.u. (≈91%)
  - Include effective range correction ($r_{eff} = 15.2\ \mathrm{a_0}$):

$$E_{dimer} = -\frac{\hbar^2}{m\, r_{eff}^2}\left(1 - \sqrt{1 - \frac{2r_{eff}}{a_s}}\right)^2 = -5.17 \cdot 10^{-9}\ \mathrm{a.u.}\ (\approx 100\%)$$

Visualizing The Difference

- **Using modern Born-Oppenheimer potential:**
  - **^{4}He-^{4}He bound state energy $E_{dimer} = -1.625$ mK.**
  - **Two-body s-wave scattering length $a_s = 170.86\ \mathrm{a_0}$.**
  - **Zero-range theory: probability $= \frac{2}{a_s} \exp\left(-2\frac{r}{a_s}\right)$.**



Visualizing The Difference

- **Using modern Born-Oppenheimer potential:**
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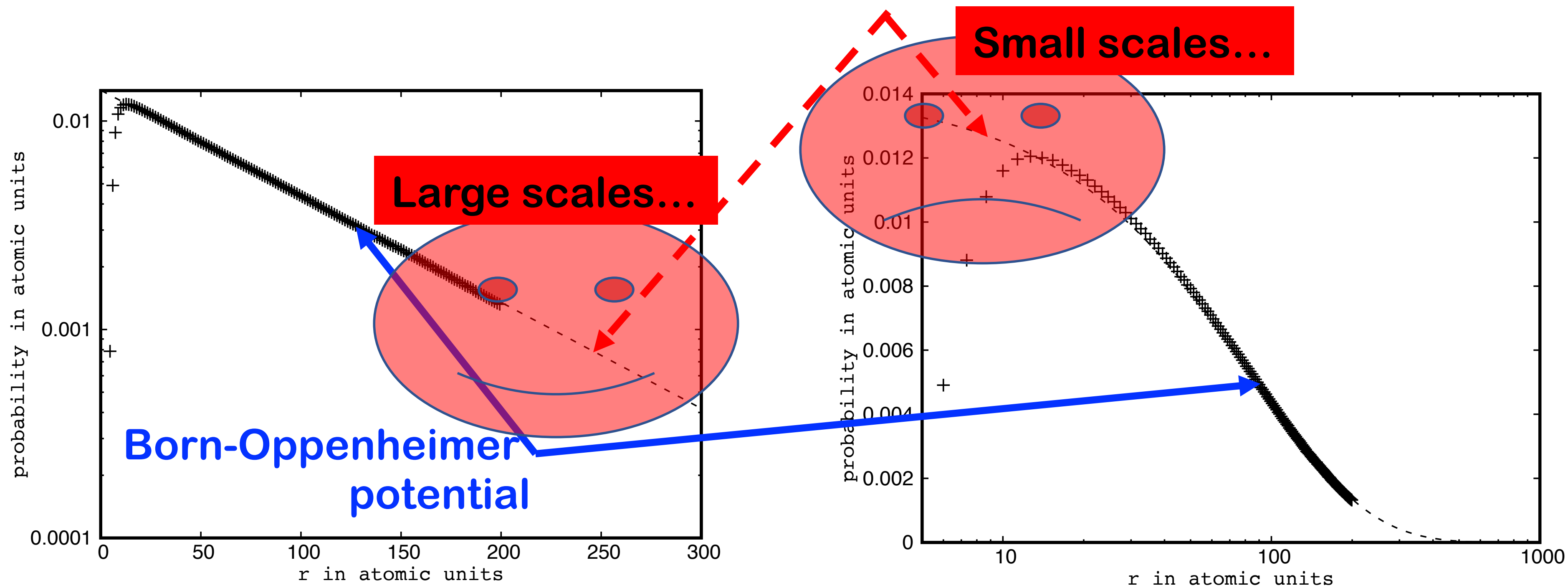
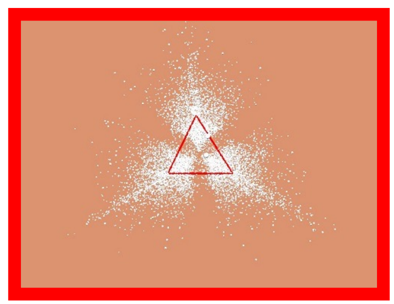
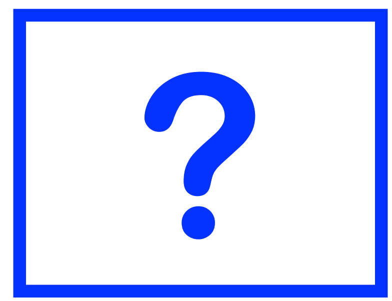


More Background on the Helium System

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

- Dimer:
  - ^4He-^4He bound state energy $E_{dimer} = -1.625$mK.
  - No J > 0 bound states.
  - 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 $r_{vdW} = 5.1a_0$).
- Trimer:
  - Two J = 0 bound states with $E_{trimer} = -131.8$mK and -2.65mK.
  - No J > 0 bound states.

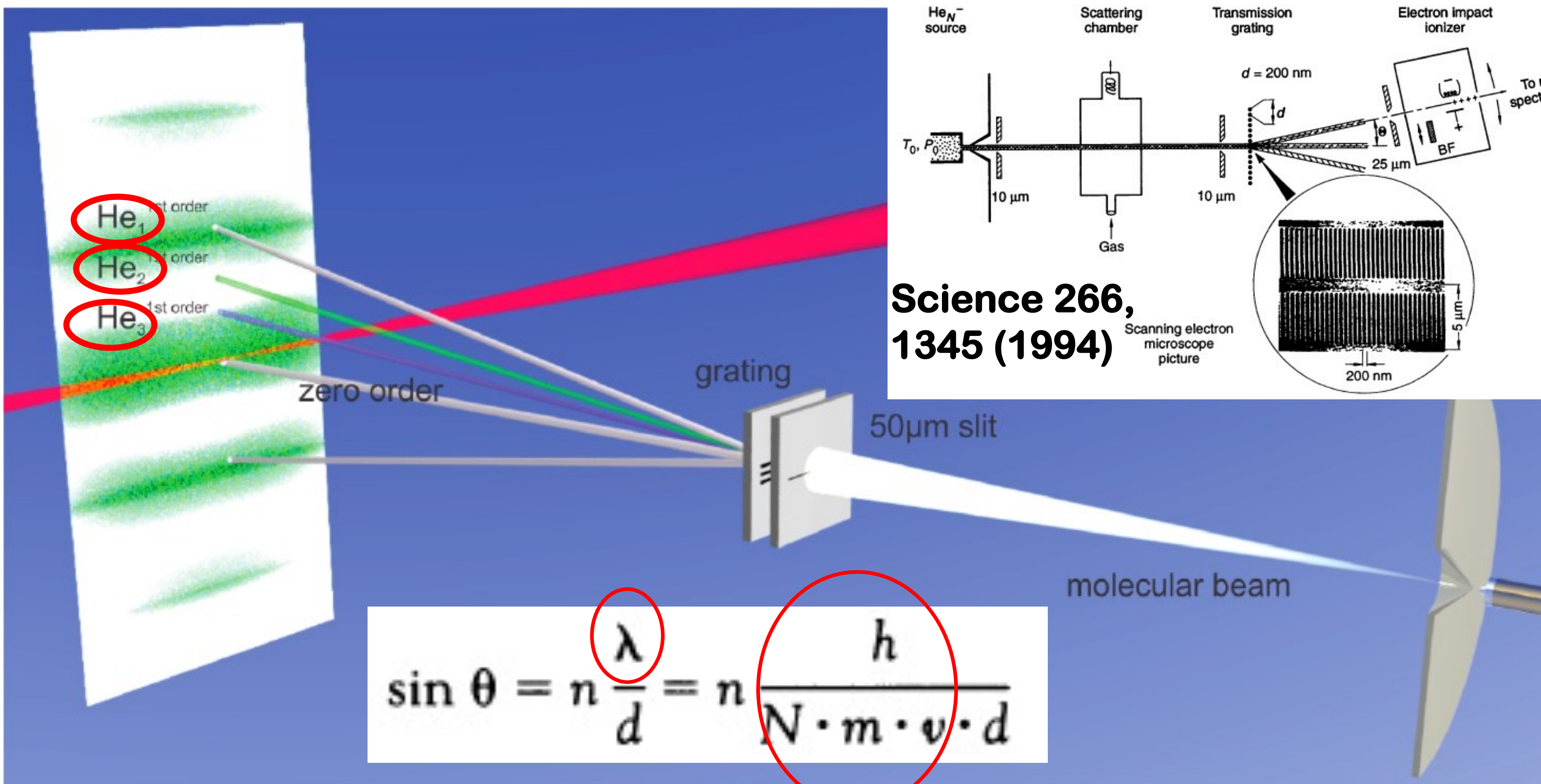




Discussed in Pascal Naidon's talk.

- Binding energy of liquid helium is E/N = −7K.

How to Prepare Helium Clusters?



Science 266, 1345 (1994)

$$\sin\theta = n\frac{\lambda}{d} = n\frac{h}{N\cdot m\cdot v\cdot d}$$

Grating serves as mass selector (N times atom mass m).
For fixed order n, larger N yields smaller angle θ.

What Is The Order Of Magnitude Of The Deflection Angle θ?

What Is The Order Of Magnitude Of The Deflection Angle θ?

Recall:

$$\sin\theta = n\frac{\lambda}{d} = n\frac{h}{N\cdot m\cdot v\cdot d}$$

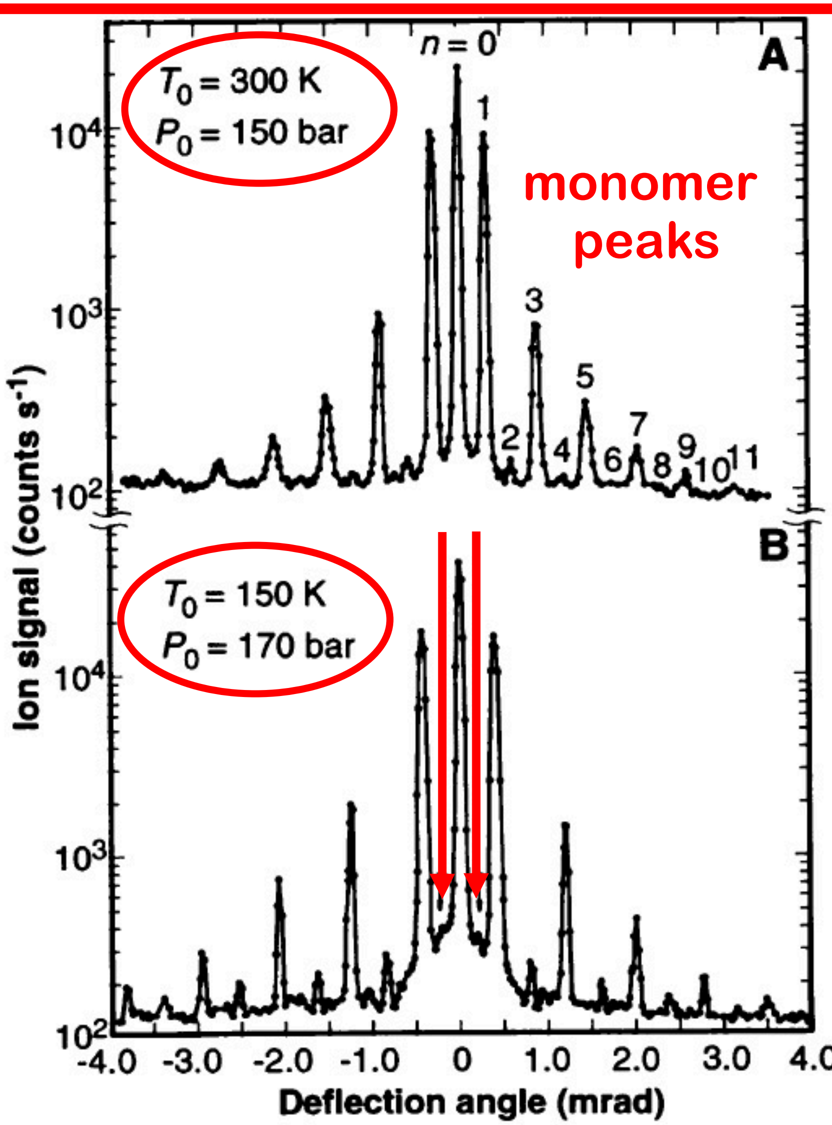
Need to know thermal de Broglie wave length.

Use diffraction order $n = 1$.
From previous slide: $d = 200\ \mathrm{nm} = 2,000\ \text{Å}$.

For $\lambda = 1\ \text{Å}$ (can be adjusted somewhat by changing nozzle temperature), we find $\sin\theta = \frac{\lambda}{d} \approx \frac{1\ \text{Å}}{2{,}000\ \text{Å}} = 0.5\cdot 10^{-3}$.

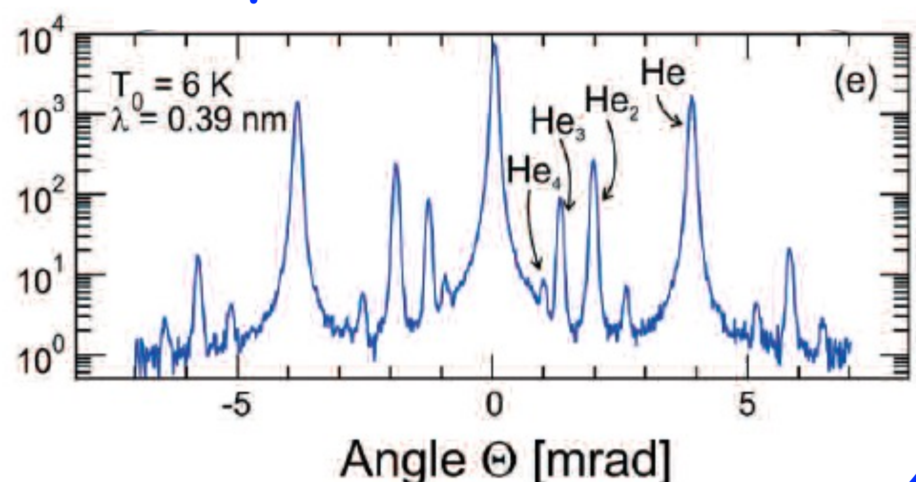
Thus, $\sin\theta \approx \theta \approx 0.5\ \mathrm{mrad}$ (this is small!).

Observation of Helium Dimer: $^{4}He_2$

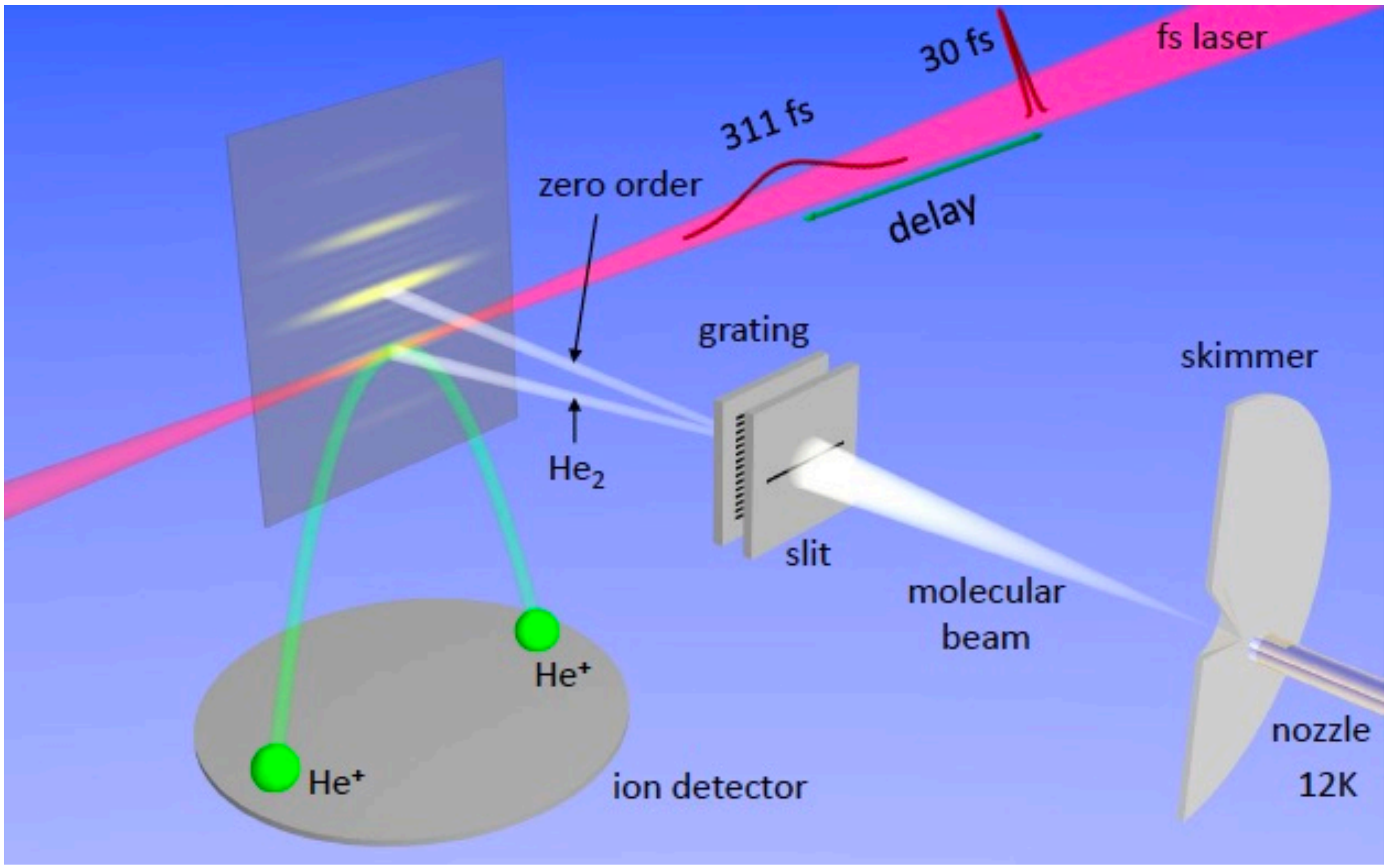


Fragile helium dimer forms in beam and can be isolated. Schoellkopf and Toennies, Science 266, 1345 (1994)

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



Pump-Probe Spectroscopy of Isolated Helium Dimers



Pump pulse: pulse length of 311 fs and intensity of 1.3×10^{14} W/cm^2.
Probe pulse rips off two electrons (Coulomb explosion).
What do we expect to happen as a function of the delay time???

What Do The Numbers Mean?

Pump pulse: pulse length of 311 fs and intensity of 1.3×10^{14} W/cm^2.
Probe pulse rips off two electrons (Coulomb explosion).
What do we expect to happen as a function of the delay time???

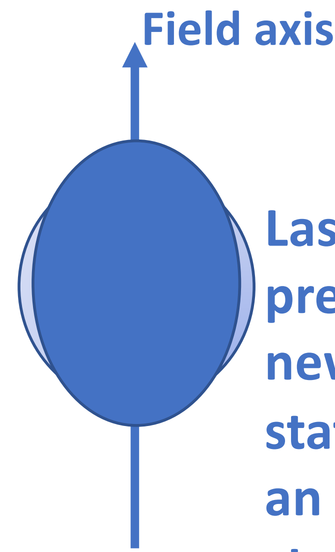
Binding energy of 1mK corresponds to $50\ \mathrm{ns} = 5 \cdot 10^7\ \mathrm{fs}$. The 311 fs pump laser is extremely short compared to the natural time scale of the helium dimer: laser pulse acts as a "kick."

Solar: $\frac{10^3 \mathrm{W}}{\mathrm{m}^2}$.

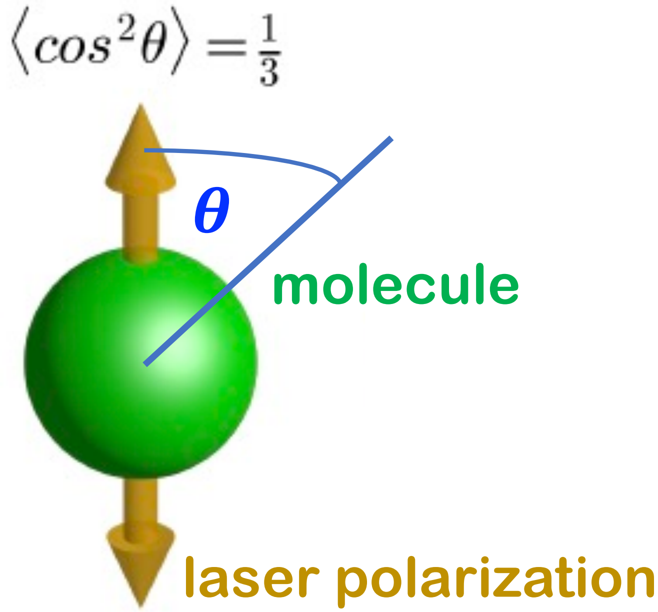
Laser pointer: $\frac{10^6 \mathrm{W}}{\mathrm{m}^2}$.

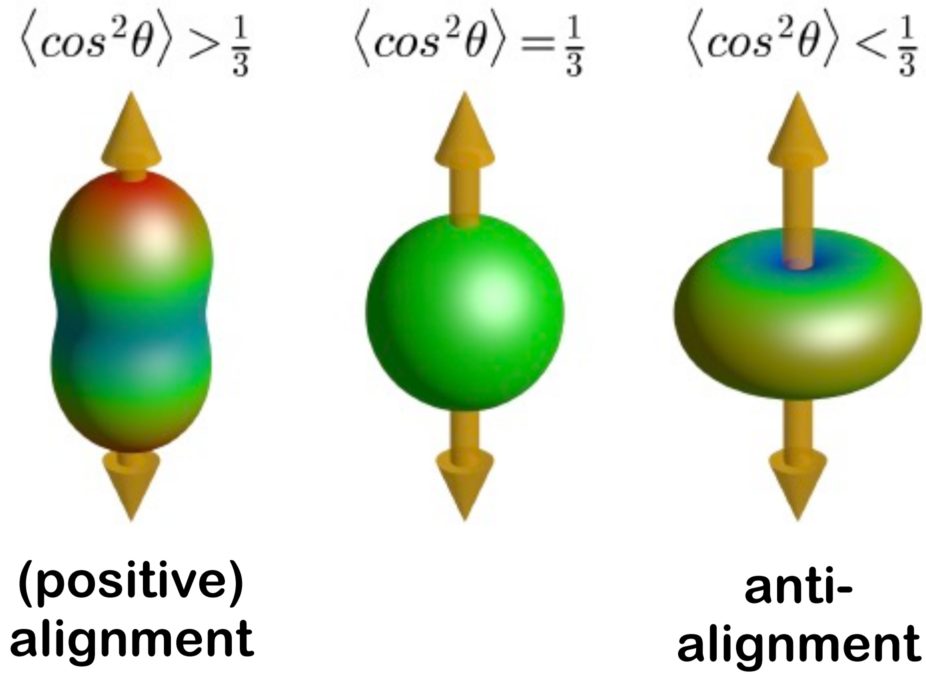
Pump pulse: $1.3 \cdot \frac{10^{13} W}{\mathrm{cm}^2} = 1.3 \cdot \frac{10^{17} \mathrm{W}}{\mathrm{m}^2}$.

Roughly, we need to worry about electronic degrees of freedom at intensities $> \frac{10^{15} \mathrm{W}}{\mathrm{cm}^2}$ (probe pulse).



What Does The Pulse Do To Helium Dimer?





Without the laser, the molecule is spherically symmetric (no alignment): The helium dimer has vanishing relative orbital angular momentum.

Will show: Helium dimer can be aligned. However, since the $J > 0$ partial wave components are not bound, they will "run away" (dissociative wave packet). Heavier non-universal dimers behave very differently.

Pump-Probe Experiments: Field Induced Alignment

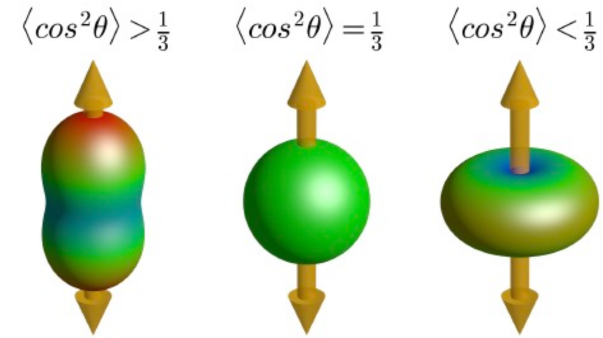
Long history of electric-field induced alignment of molecules: Unique rotational dynamics for molecules such as I_2, N_2,…

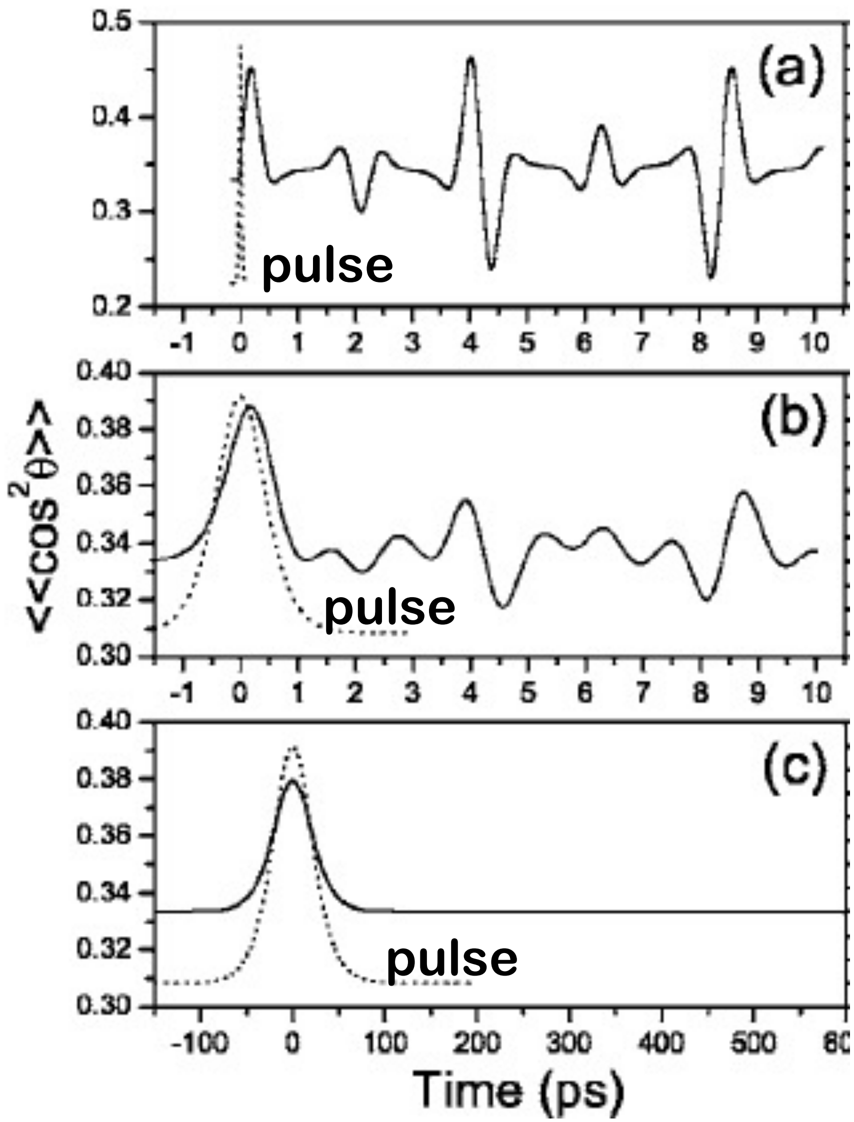
E.g., "Colloquium: Aligning molecules with strong laser pulses", RMP 75, 543 (2003) by Stapelfeldt and Seideman, >1000 citations:

"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 helium dimer adds "physical dynamics" to the list!

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





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

Pulse length 50 fs.

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

<u>**Impulse regime.**</u>

Alignment signal of 1/3 = spherically symmetric. "Rotational revivals" require particular phase relation:
$E_J = B_0 J(J+1) - D_0 J^2 (J+1)^2$.

Pulse length 50 ps.

Intensity $2.5 \times 10^{12} \frac{W}{cm^2}$.

<u>**Adiabatic regime.**</u>

“Kicking” the $^4He_2$: Pump-Probe Experiments

Entirely new regime:

Recall: $^4He_2$ dimer supports exactly one (extremely weakly-bound) state. State is largely universal.
What happens when one applies short (~310fs), intense (~10^{14}W/cm^2) kick?

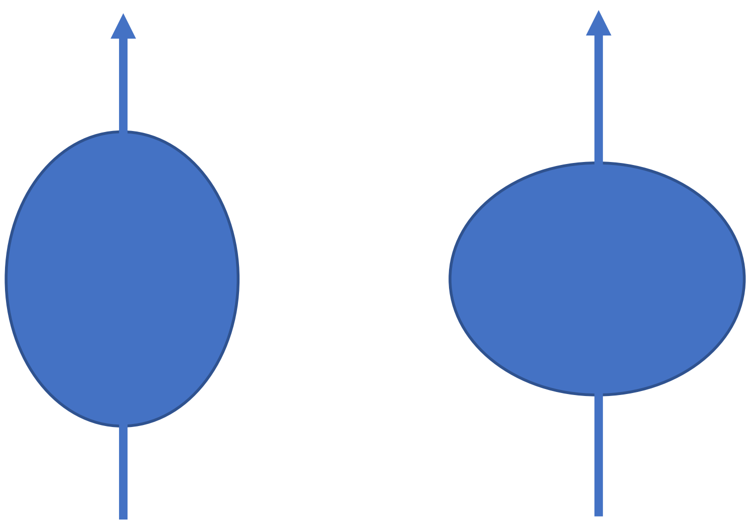
Separation of time scales (binding energy of 1mK corresponds to 50ns):
Kick is non-adiabatic (quench); in fact, we can simulate it by a delta-function pulse.

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).

Theoretical Treatment: Laser-Molecule Interaction

$$V_{\rm lm}(r,\theta,t) = \frac{|\epsilon(t)|^2\alpha_0^2}{4\pi\mathcal{E}_0}\left[-\frac{\alpha_0}{2\,(4\pi\mathcal{E}_0)\,r^6} + \alpha_0\frac{1-3\cos^2\theta}{(4\pi\mathcal{E}_0)\,r^6} + \frac{1-3\cos^2\theta}{r^3}\right]$$

dipole-dipole interaction



Question: Which of these "configurations" is energetically favored?

Theoretical Treatment: Laser-Molecule Interaction

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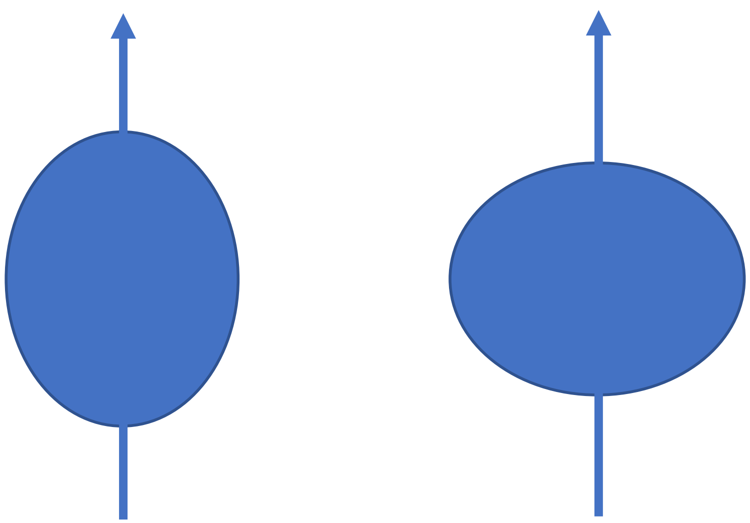

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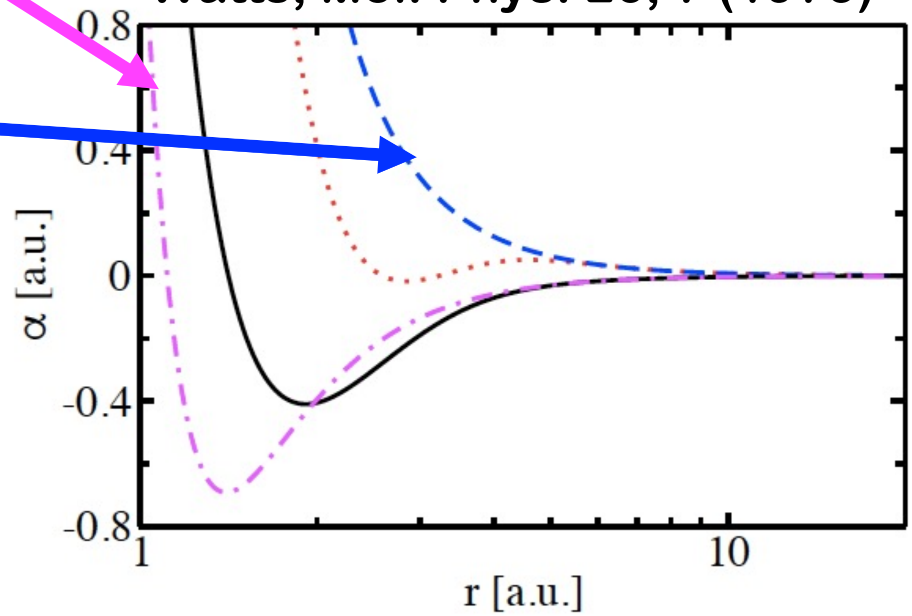
$$V_{\rm lm}(r,\theta,t) = -\frac{1}{2}|\epsilon(t)|^2\,\left[\alpha_{\|}(r)\cos^2\theta+\alpha_{\perp}(r)\sin^2\theta\right]$$

V_{lm} couples different partial waves.

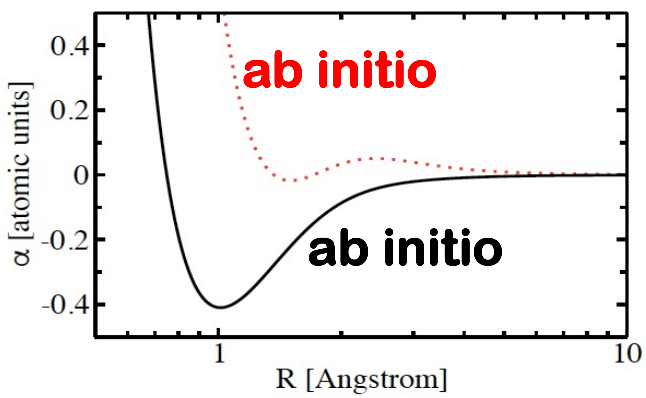
$$\alpha_{\perp}(r) = 2a_0 - \frac{2\alpha_0^2}{4\pi\mathcal{E}_0\,r^3}+\frac{2\alpha_0^3}{(4\pi\mathcal{E}_0)^2\,r^6}$$

$$\alpha_{\|}(r) = 2a_0 + \frac{4\alpha_0^2}{4\pi\mathcal{E}_0\,r^3}+\frac{8\alpha_0^3}{(4\pi\mathcal{E}_0)^2\,r^6}$$

Analytical: Buckingham and Watts, Mol. Phys. 26, 7 (1973)

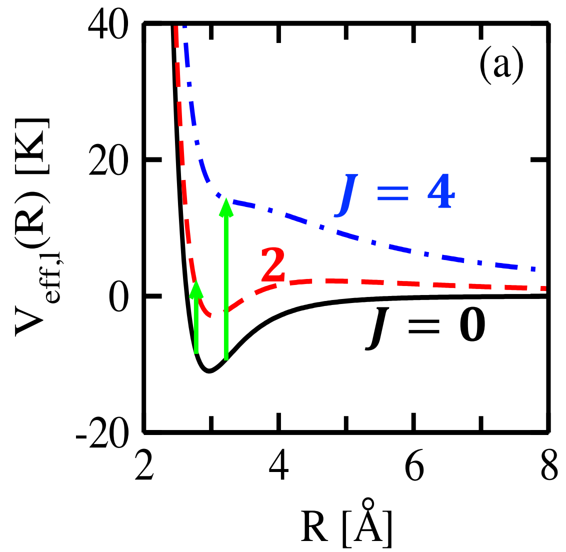


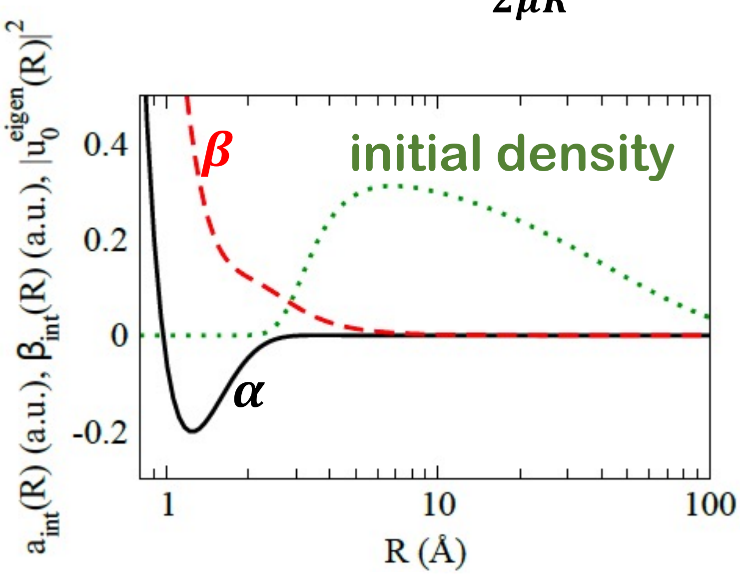
Ab initio: Cencek et al., JCP 135, 014301 (2011)



Theoretical Treatment

Dimer potential $V_{eff,J}(R) = V_{He-He}(R) + \frac{\hbar^2 J(J+1)}{2\mu R^2}$





Laser-molecule interaction:

$$V_{lm} = -\frac{1}{2}\varepsilon^2(t)\left[\alpha(R)\,Y_{00}(\hat{R}) + \beta(R)\,Y_{20}(\hat{R})\right]$$

Gaussian profile

Solve time-dependent Schroedinger equation using spherical coordinates:

$$\Psi(R,\theta,t) = \sum_{J=0,2,\ldots} \frac{u_J(R,t)}{R} Y_{J0}(\hat{R})$$

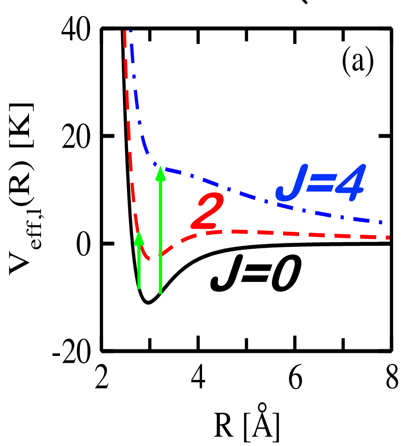
Pulse couples different partial waves.
When pulse is off, the channels are decoupled.

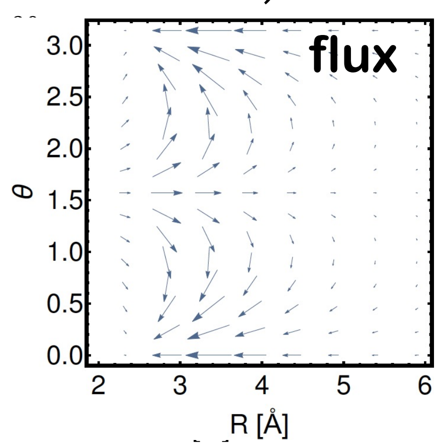
^{4}He-^{4}He In Time-Dependent Electric Field

In what follows, the initial state will be the $J = 0$ eigenstate of the zero-field Hamiltonian of ^{4}He-^{4}He system.

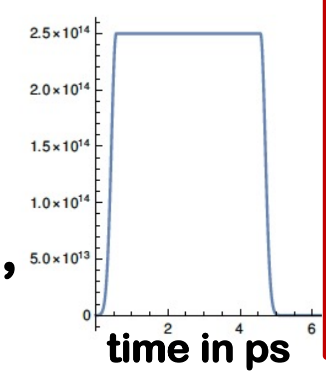
Scenario 1 (non-adiabatic laser kick):

$$\varepsilon(t) = \varepsilon_0 \exp\left(-2\ln 2 \left(\frac{t-t_{ref}}{\tau}\right)^2\right); \tau \approx 300\text{fs}.$$





Scenario 2 ("slow"): Gaussian turn-on, hold for several ps, Gaussian turn-off.



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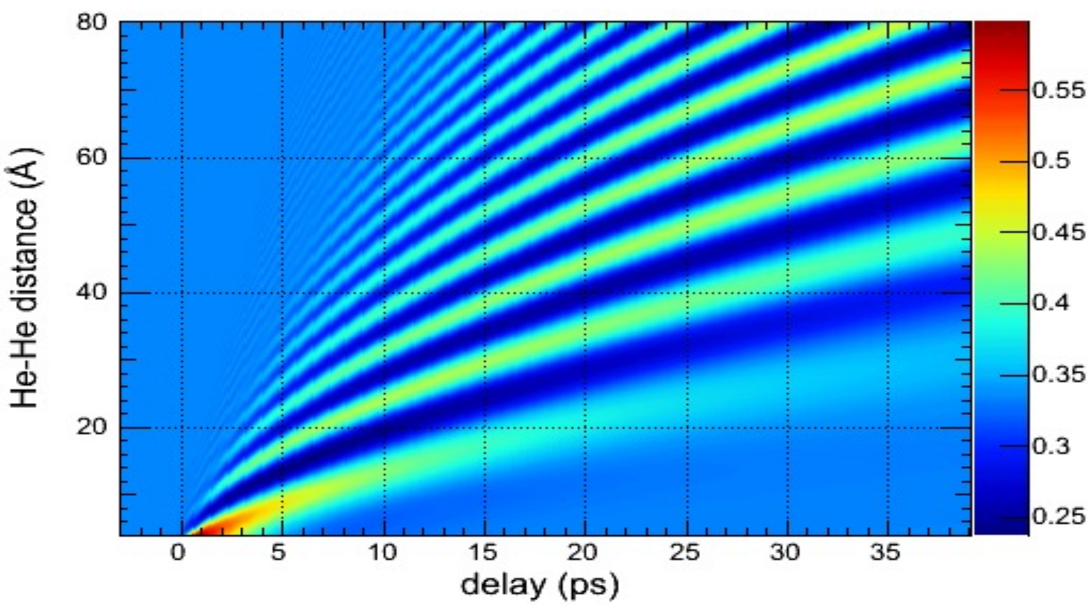
$$\Psi(R,\theta,t) = \sum_{J=0,2,\ldots} \frac{u_J(R,t)}{R} Y_{J0}(\hat{R})$$

Laser couples different partial waves.

When laser is off, the channels are decoupled.

Scenario 1: Theory Result

$$C_2(R,t) = \frac{\int_0^\pi \Psi^*(R,\theta,t)\cos^2\theta\Psi(R,\theta,t)\sin\theta d\theta}{\int_0^\pi |\Psi(R,\theta,t)|^2 \sin\theta d\theta}$$



Interference between J=0 and J=2 partial waves. J=2 portion "travels" on structureless background.

Solve time-dependent Schroedinger equation using spherical coordinates:

$$\Psi(R,\theta,t) = \sum_{J=0,2,\ldots} \frac{u_J(R,t)}{R} Y_{J0}(\hat{R})$$

Pulse couples different partial waves.

When pulse is off, the channels are decoupled.

Origin Of The Interference Pattern?

Expand: $\Psi(R,\theta,t) = \sum_{J=0,2,4,\cdots} R^{-1} u_J(R,t) Y_{J0}(\theta)$

$u_J(R,t) = \exp(\imath\gamma_J(R,t))|u_J(R,t)|$ & $\tan(\gamma_J(R,t)) = \frac{\mathrm{Im}(u_J(R,t))}{\mathrm{Re}(u_J(R,t))}$

Plug in: $C_2(R,t) = \frac{\int_0^\pi \Psi^*(R,\theta,t)\cos^2\theta\Psi(R,\theta,t)\sin\theta d\theta}{\int_0^\pi |\Psi(R,\theta,t)|^2 \sin\theta d\theta}$

$$C_2(R,t) = \frac{1}{3} + \frac{4}{3\sqrt{5}}\mathrm{Re}\left(\frac{u_2(R,t)}{u_0(R,t)}\right) + \cdots$$

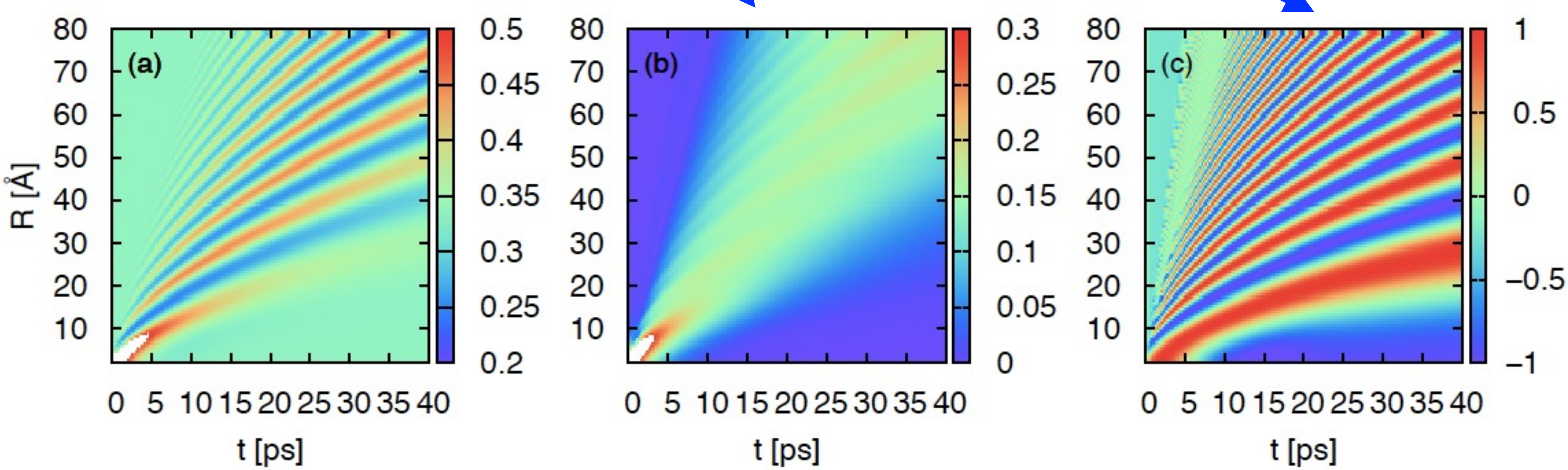
$$C_2(R,t) = \frac{1}{3} + \frac{4}{3\sqrt{5}}\left|\frac{u_2(R,t)}{u_0(R,t)}\right| \cos(\gamma_2(R,t) - \gamma_0(R,t)) + \cdots$$

Interference Pattern Due To $J = 0$ and $J = 2$ Phases

$$C_2(R,t) = \frac{1}{3} + \frac{4}{3\sqrt{5}}\left|\frac{u_2(R,t)}{u_0(R,t)}\right|\cos(\gamma_2(R,t) - \gamma_0(R,t)) + \cdots$$

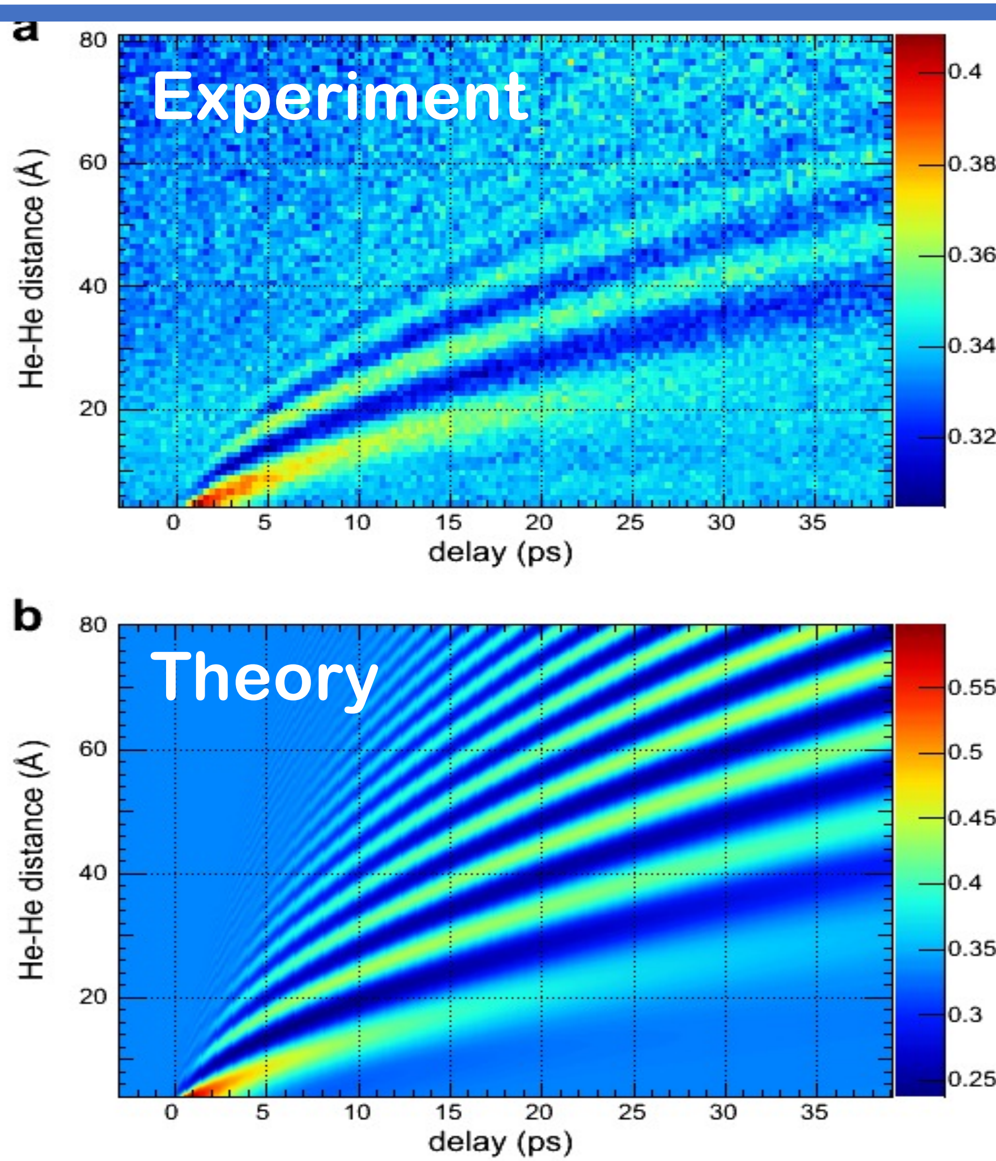
Only plotting $J = 0$ and 2

$\gamma_0 \approx const$



Alignment signal $cos^2\theta$ can be interpreted as measuring $\gamma_2(R,t)$.

Comparison With Experiment

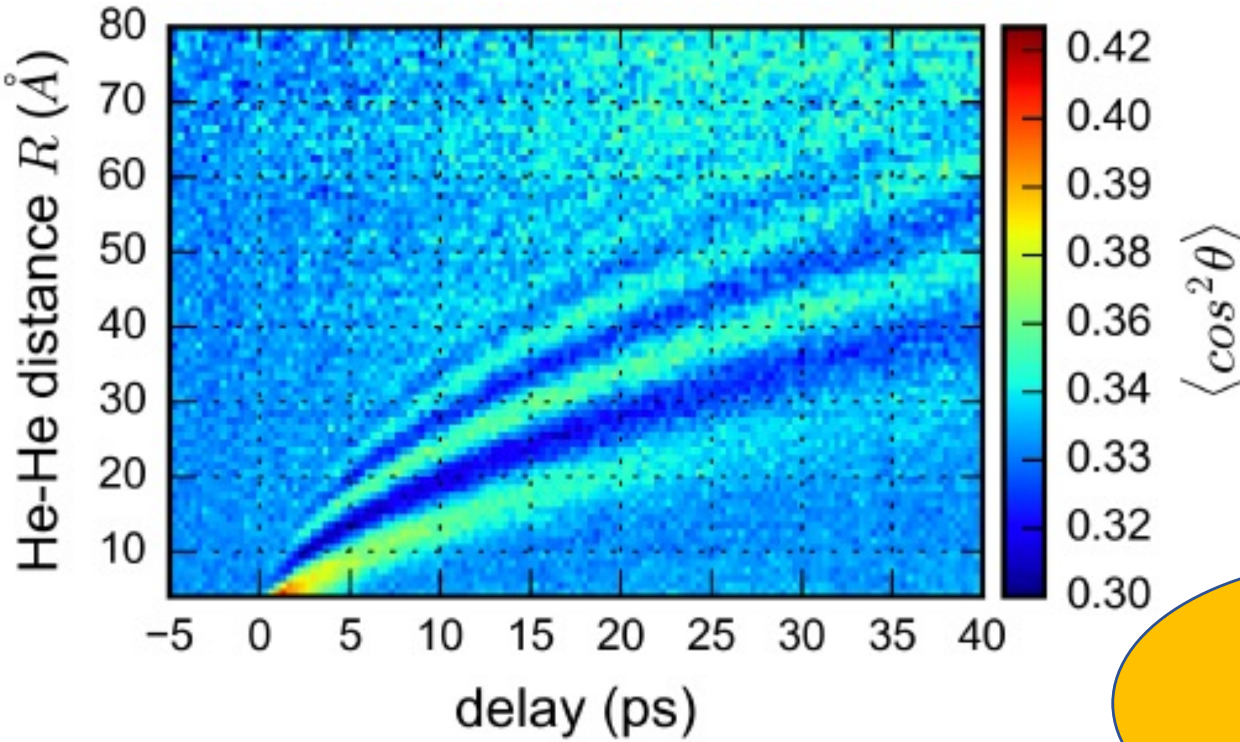


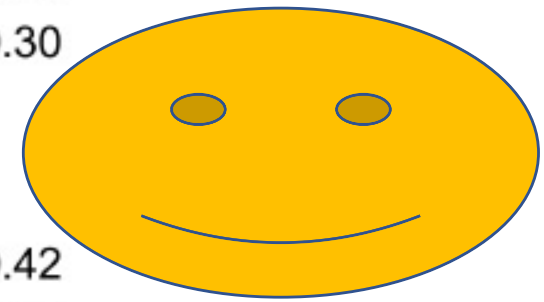
Experimental data by Maksim Kunitski, Reinhard Doerner et al. (Frankfurt University)

Agreement is qualitative but not quantitative.

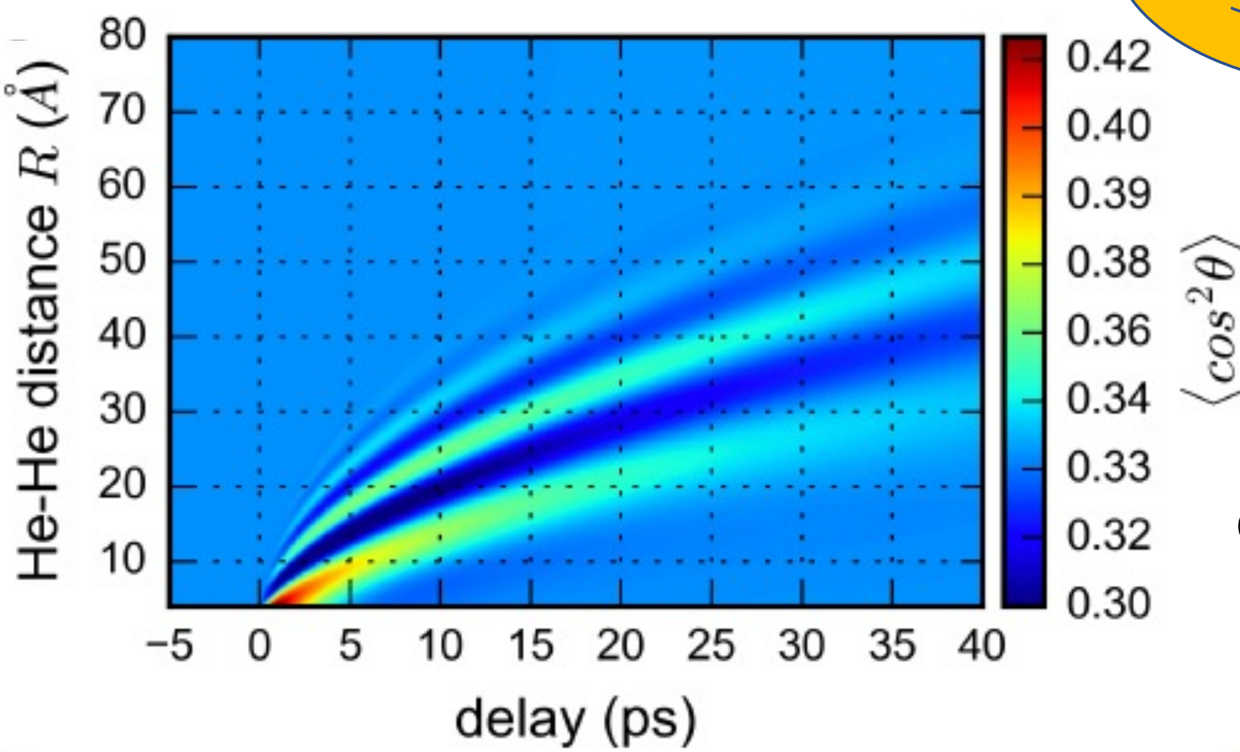
Need to account for finite experimental resolution.

Experiment





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



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

Kicking the ^{4}He Dimer

For the first time: Intense laser used to probe dynamics at single-atom level using universal, scattering length dominated initial state.

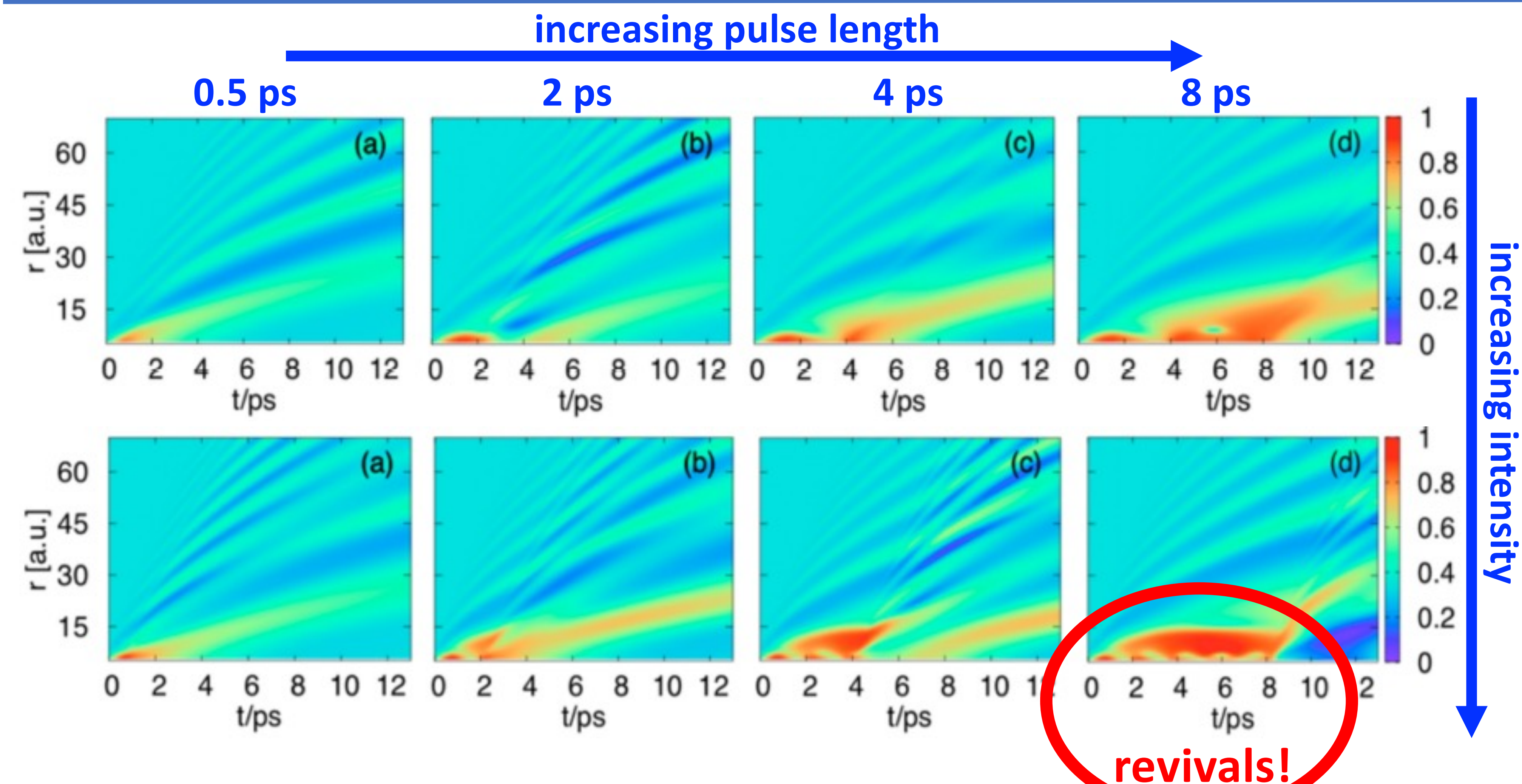
"Rotationless" ^{4}He dimer can be aligned! Note, it's the continuum portion of the wave packet…

Pattern due to interference between J=0 and J=2 channels: Measurement of spatially and time dependent relative phase between these two partial wave channels. State tomography!

Many outstanding challenges:

Resonances as in ultracold atoms? Need longer pulses…
Time-dependent modulation of interaction strength?
Dynamics of (Efimov) trimers? Need to populate it first…
Larger clusters.

Scenario 2: Longer Pulses



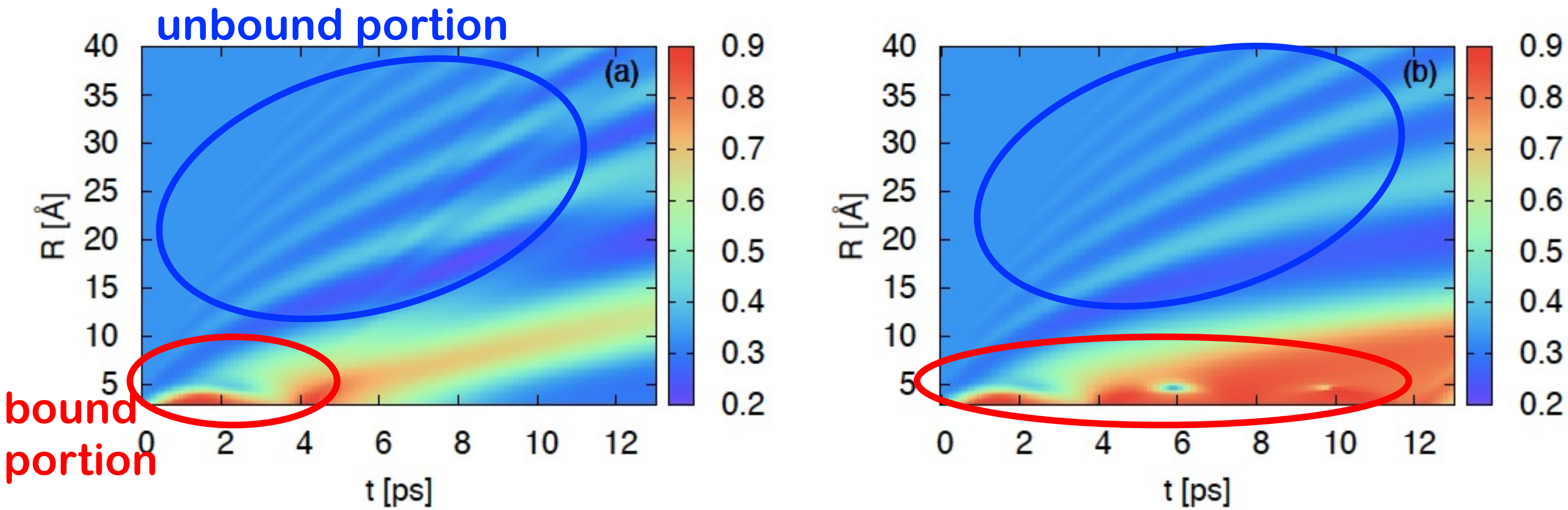
Guan and Blume, PRA 99, 033416 (2019). Awaiting experimental realization…

Signature Of Field-Induced $^4He_2$ Bound States?

$$C_2(R,t) = \frac{\int_0^\pi \Psi^*(R,\theta,t)\cos^2\theta\Psi(R,\theta,t)\sin\theta d\theta}{\int_0^\pi |\Psi(R,\theta,t)|^2 \sin\theta d\theta}$$

max. intensity
$2.5 \cdot 10^{14}$W/cm²

4ps hold time **12ps hold time**



Fingerprint of revivals in time-dependent response of system: Dimer oscillates between deeply-bound state and weakly-bound state.

Helium Dimer In Static External Electric Field

Plugging $\Psi(R,\theta) = \sum_{J=0,2,\ldots} \frac{u_J(R)}{R} Y_{J0}(\hat{R})$ into $H\Psi(R,\theta) = E\Psi(R,\theta)$ yields

$$\left(-\frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial R^2} + \frac{\hbar^2 J(J+1)}{2\mu R^2} + V_{He-He}(R)\right)u_J(R) + \sum_{K=0,2,\ldots}^{\infty} W_{JK}(R)u_K(R) = Eu_J(R),$$

where the coupling matrix elements read

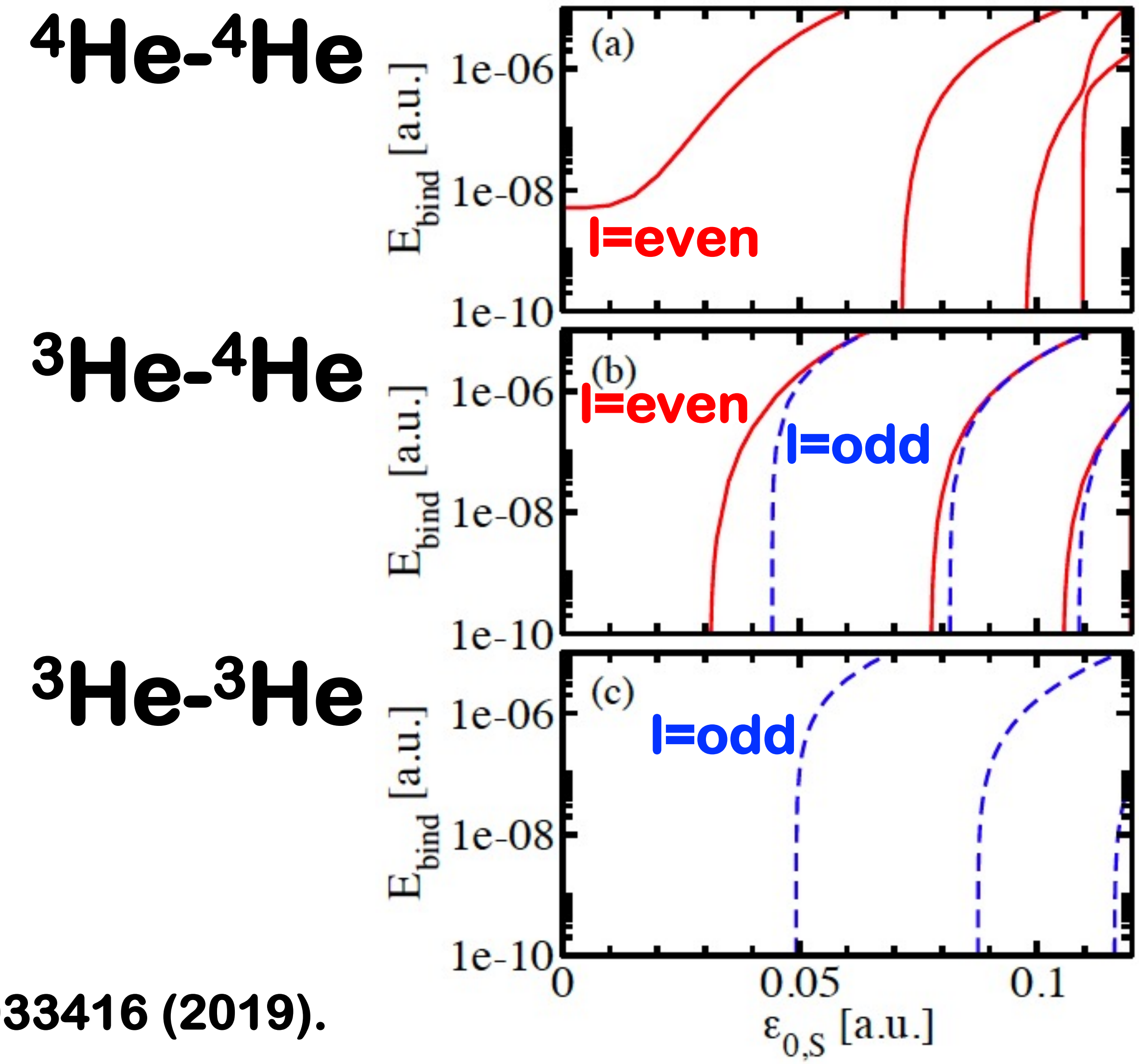
$$W_{JK}(R) = \int_0^{2\pi}\int_0^{\pi} u_J^*(R)V_{lm}(R,\theta)u_K(R)\sin\theta\, d\theta\, d\varphi.$$

Bound states depend on R and θ!

Scattering states are characterized by scattering length matrix that contains a_{00} (come in in 0 and go out in 0), a_{02} (come in in 2 and go out in 0), a_{20}, a_{22},…!

Tunability of Dimer: Pure and Mixed Isotopes

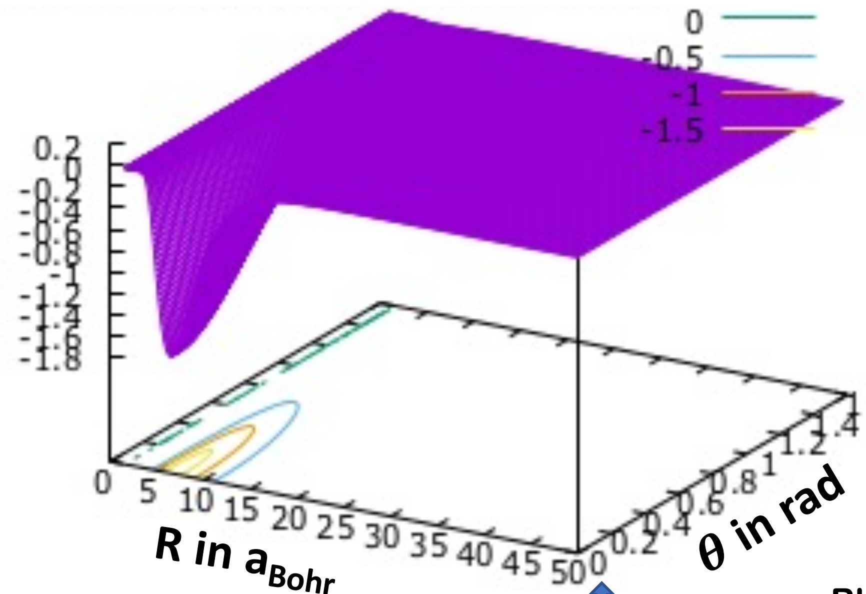
Static electric field (infinitely long pulse): We are no longer looking at dynamics but instead solving the time-independent Schrodinger equation in the presence of static external field.



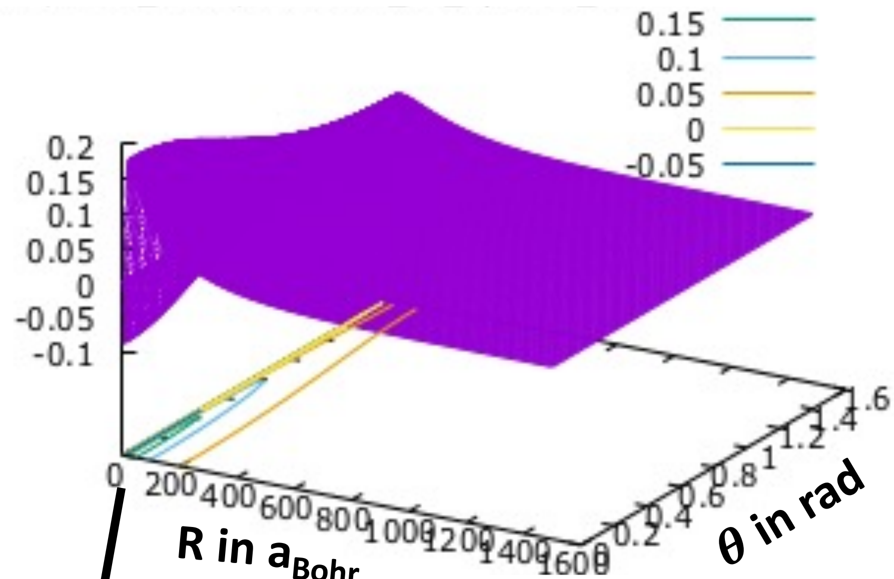
Guan and Blume, PRA 99, 033416 (2019).

Static External Electric Field: Results for $^4He_2$

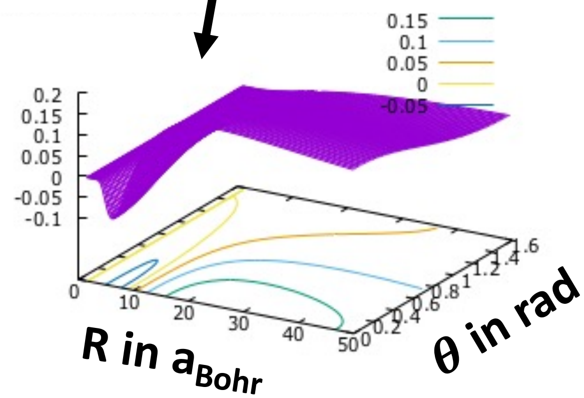
Ground state wave function



Excited state wave function



Blow-up of excited state wave function



Wave functions near the first field-induced resonance: Newly supported bound state is large.

Anisotropy is stronger at small R than at large R.

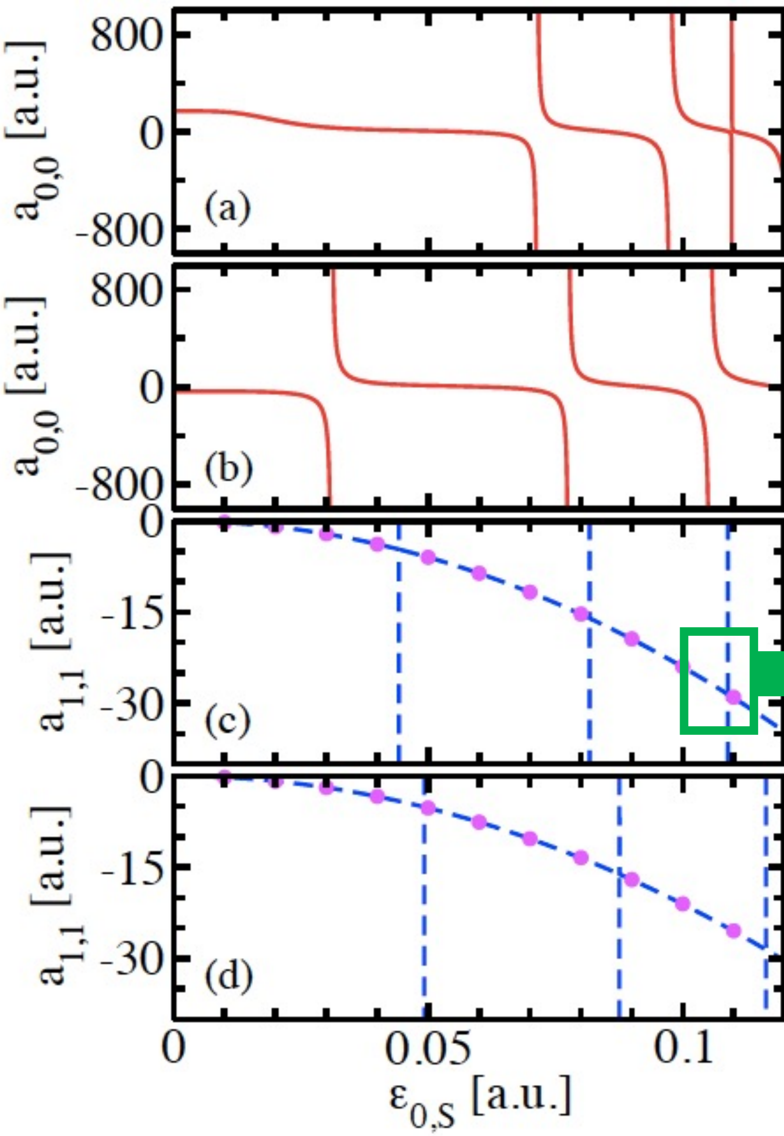
Static External Electric Field: Scattering Lengths For He-He

^{4}He-^{4}He even *J*

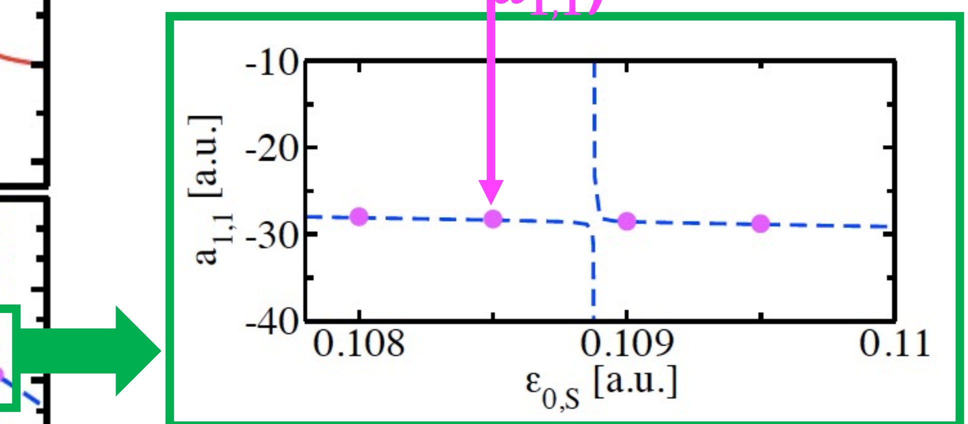
^{3}He-^{4}He even *J*

^{3}He-^{4}He odd *J*

^{3}He-^{3}He odd *J*

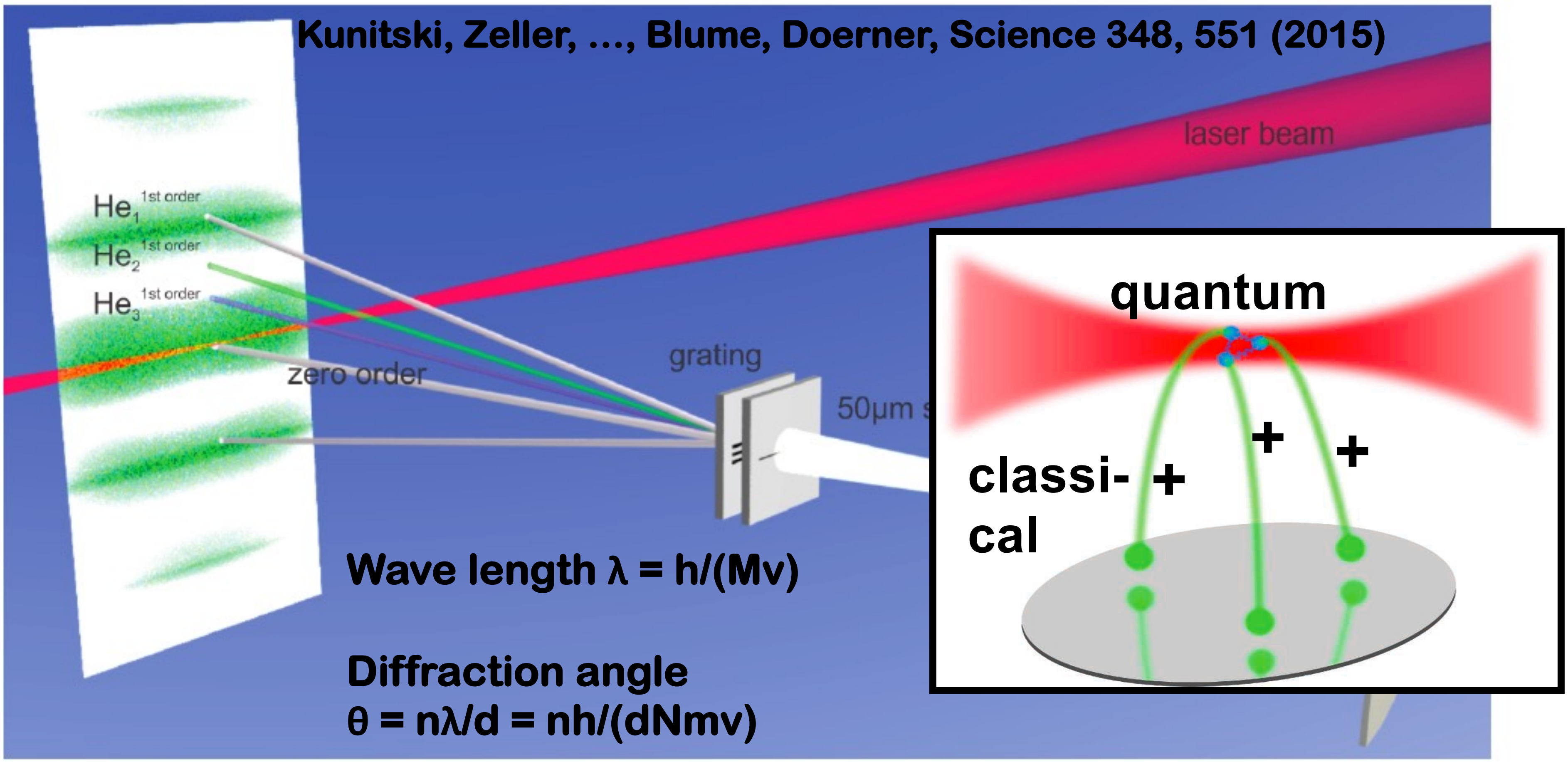


Born approximation (expected to work, away from resonances, for $a_{1,1}$)



Tunability of ^{3}He-^{4}He and ^{3}He-^{3}He discussed in Nielsen et al., PRL 82, 2844 (1999). Qualitative agreement with ^{3}He-^{4}He results.

Finally: Helium Trimer Excited Efimov State?

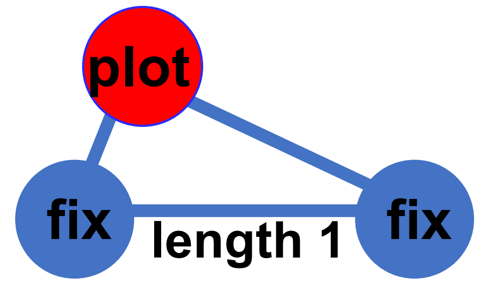


Grating serves as mass selector (N times atom mass m):

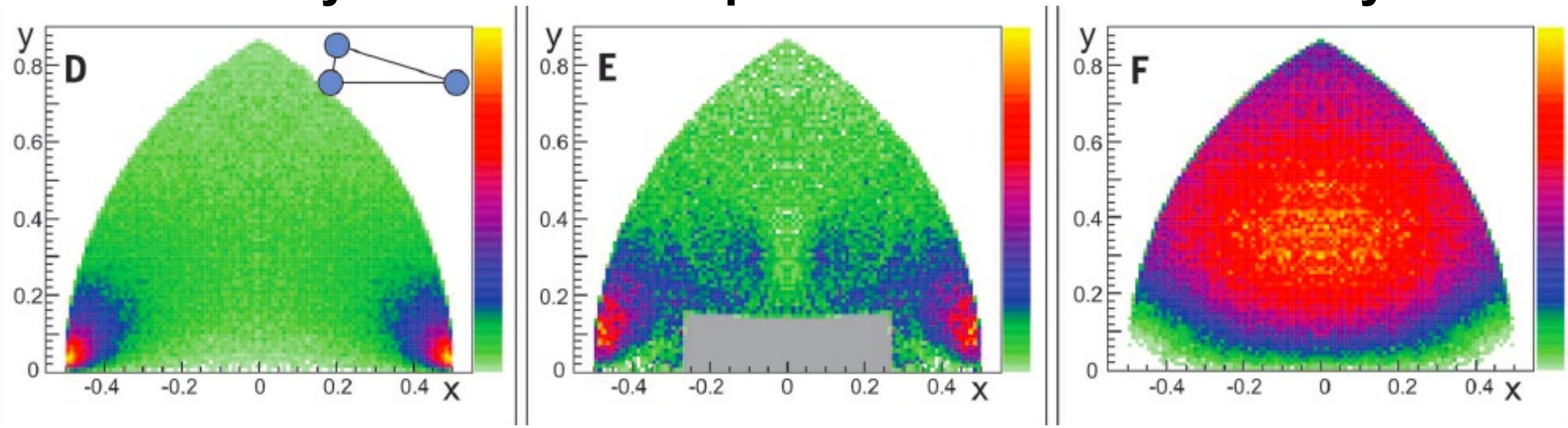
He_3 signal contains ground state trimer *and* excited state trimer.

Laser beam ionizes trimer: Coulomb explosion of $^4He_3$ (3 ions).

Normalized Structural Properties of $^4He_3$



excited state: theory | excited state: experiment | ground state: theory

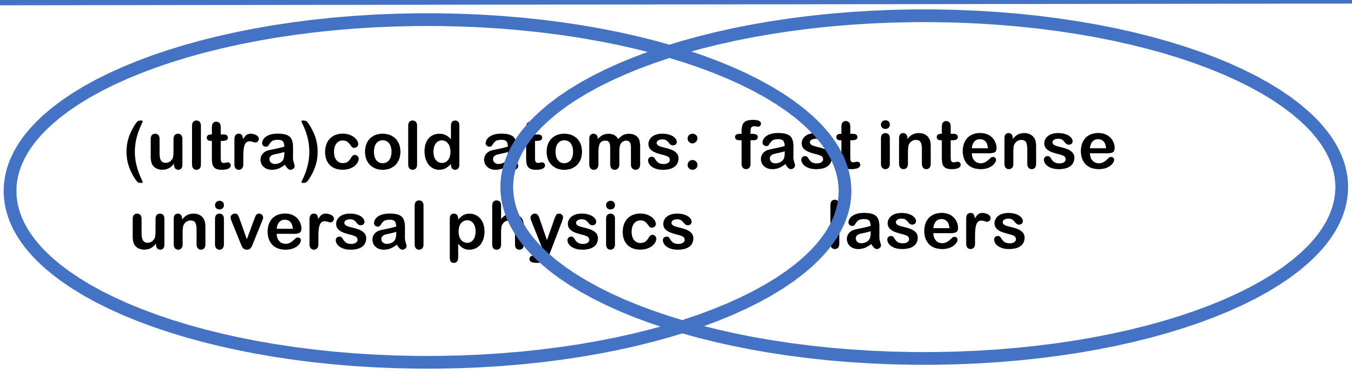


Divide all three interparticle distances by largest r_{ij} and plot k^{th} atom (positive y): Corresponds to placing atoms i and j at $(-1/2,0)$ and $(1/2,0)$.

Ground state and excited states have distinct characteristics!!!

Message: Reconstruction of quantum mechanical trimer density.

New Opportunities



Many thanks to Qingze Guan, Maksim Kunitski, Reinhard Doerner, and Doerner's group at Frankfurt University

One may hope: Two good things combined should be better than two good things separated...

But you may object: Aren't we just gonna blow everything up?

Yes, we will... and it's fun and useful...

Hopefully, I was able to convince you:

A different type of non-equilibrium dynamics (no rotational revivals,...).

Interesting future prospects.

Thank You!