Center for Quantum Research and Technology

Dynamics of Weakly-Bound Molecules

Doerte Blume

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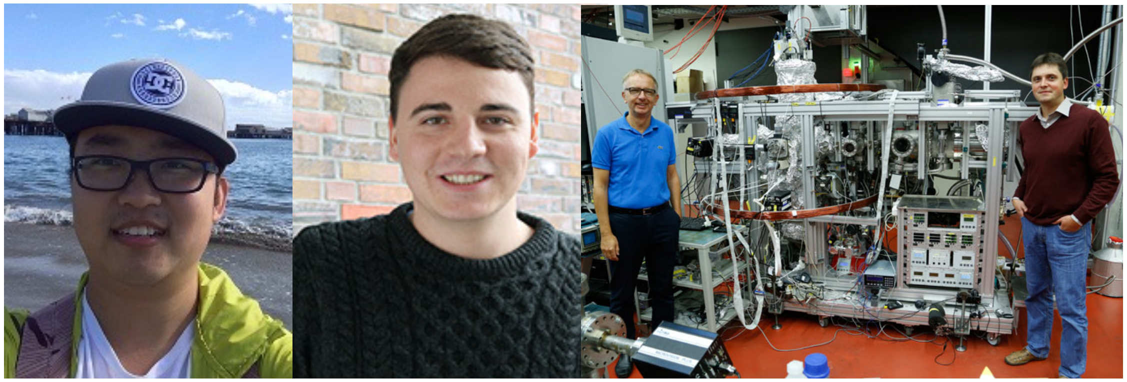

Supported by the NSF.

This Talk

- Interaction between laser and atom/molecule:
  - Critical in many fields of physics.
- Helium dimer dynamics (theory and experiment):
  - Limit of infinitely long pulse.
  - Inducing rotational and vibrational dynamics with a femto-second laser.
- Dynamics of heavier rare gas dimers:
  - Rich interplay of bound and unbound eigen states.

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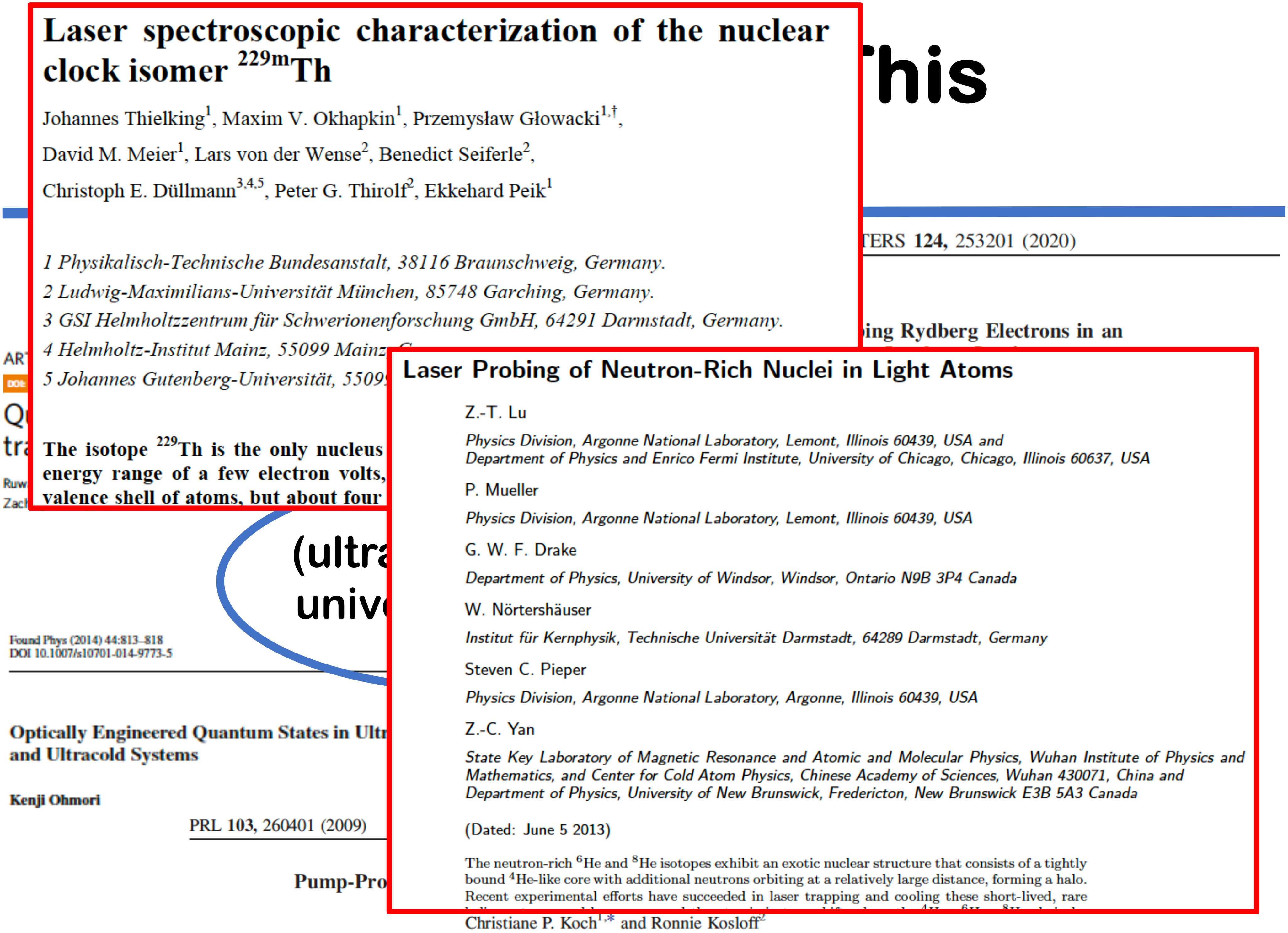

Laser spectroscopic characterization of the nuclear clock isomer ^{229m}Th

Johannes Thielking[1], Maxim V. Okhapkin[1], Przemysław Głowacki[1,†],
David M. Meier[1], Lars von der Wense[2], Benedict Seiferle[2],
Christoph E. Düllmann[3,4,5], Peter G. Thirolf[2], Ekkehard Peik[1]

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4 Helmholtz-Institut Mainz, 55099 Mainz
5 Johannes Gutenberg-Universität, 5509

The isotope ^{229}Th is the only nucleus energy range of a few electron volts, valence shell of atoms, but about four

TERS **124,** 253201 (2020)

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

Optically Engineered Quantum States in Ultr and Ultracold Systems

Kenji Ohmori

PRL **103,** 260401 (2009)

Pump-Pro

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

Laser Probing of Neutron-Rich Nuclei in Light Atoms

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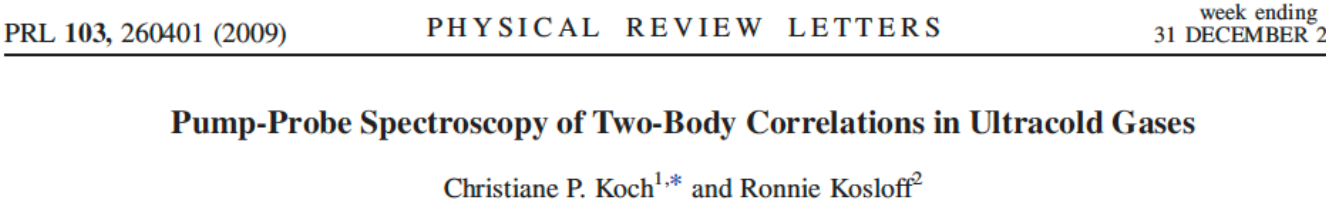
State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, and Center for Cold Atom Physics, Chinese Academy of Sciences, Wuhan 430071, China and
Department of Physics, University of New Brunswick, Fredericton, New Brunswick E3B 5A3 Canada

(Dated: June 5 2013)

The neutron-rich ^{6}He and ^{8}He isotopes exhibit an exotic nuclear structure that consists of a tightly bound ^{4}He-like core with additional neutrons orbiting at a relatively large distance, forming a halo. Recent experimental efforts have succeeded in laser trapping and cooling these short-lived, rare

Atomic Physics

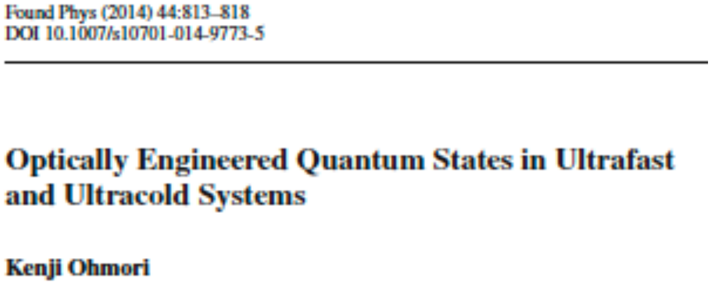
- Cooling of atoms and molecules.
- Trapping of atoms and molecules.
    - My group, e.g., is working on polarizabilities of neutral atoms, including Rydberg atoms (needed for determining magic wave lengths).
- Manipulating atom-atom interactions.



PRL **103,** 260401 (2009) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2

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

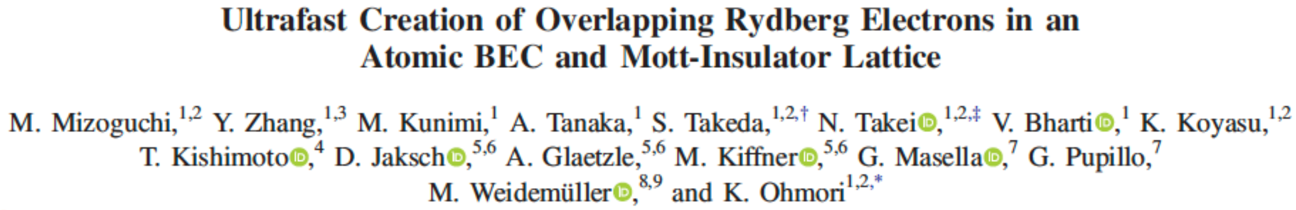
Christiane P. Koch[1,*] and Ronnie Kosloff[2]



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

Optically Engineered Quantum States in Ultrafast and Ultracold Systems

Kenji Ohmori



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,*]

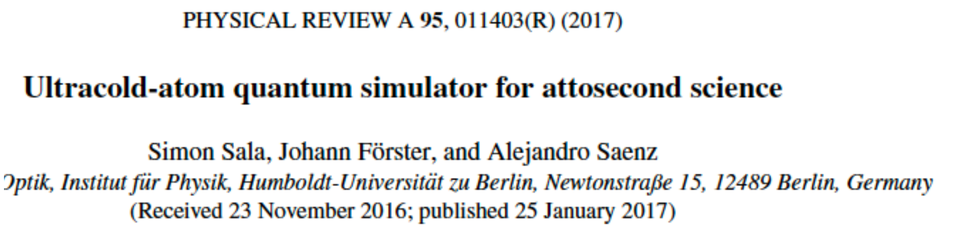
ARTICLE

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

Quantum simulation of ultrafast dynamics using 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]

RAPID COMMUNIC



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

Ultracold-atom quantum simulator for attosecond science

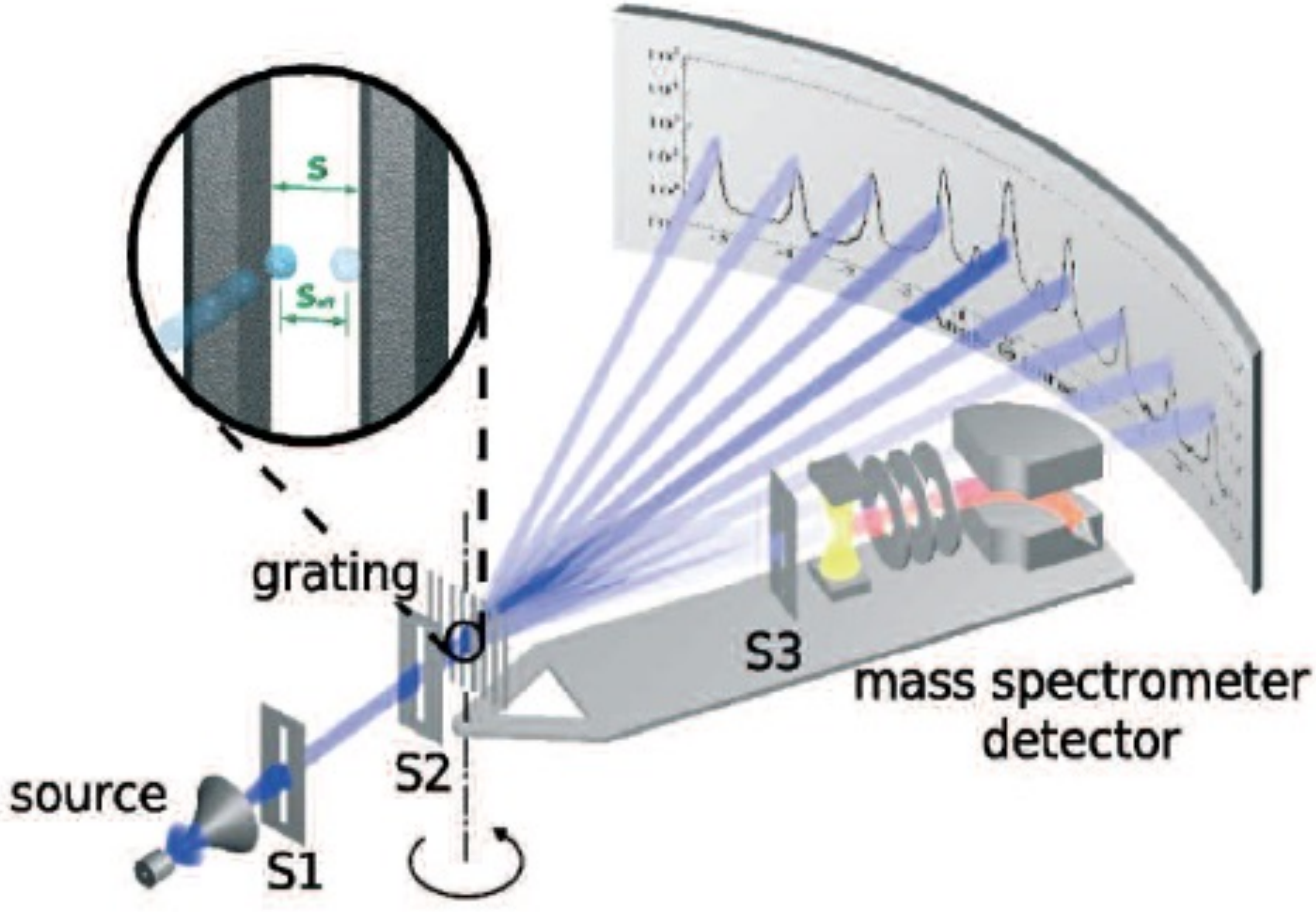
Simon Sala, Johann Förster, and Alejandro Saenz

Optik, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße 15, 12489 Berlin, Germany

(Received 23 November 2016; published 25 January 2017)

Rare Gas Molecules Prepared in Matter Wave Diffraction Experiment

Kornilov, Toennies, 10.1051/epn:2007003



De Broglie wave length λ:

$\lambda = h/(Mv)$

Diffraction angle θ:

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

v: velocity

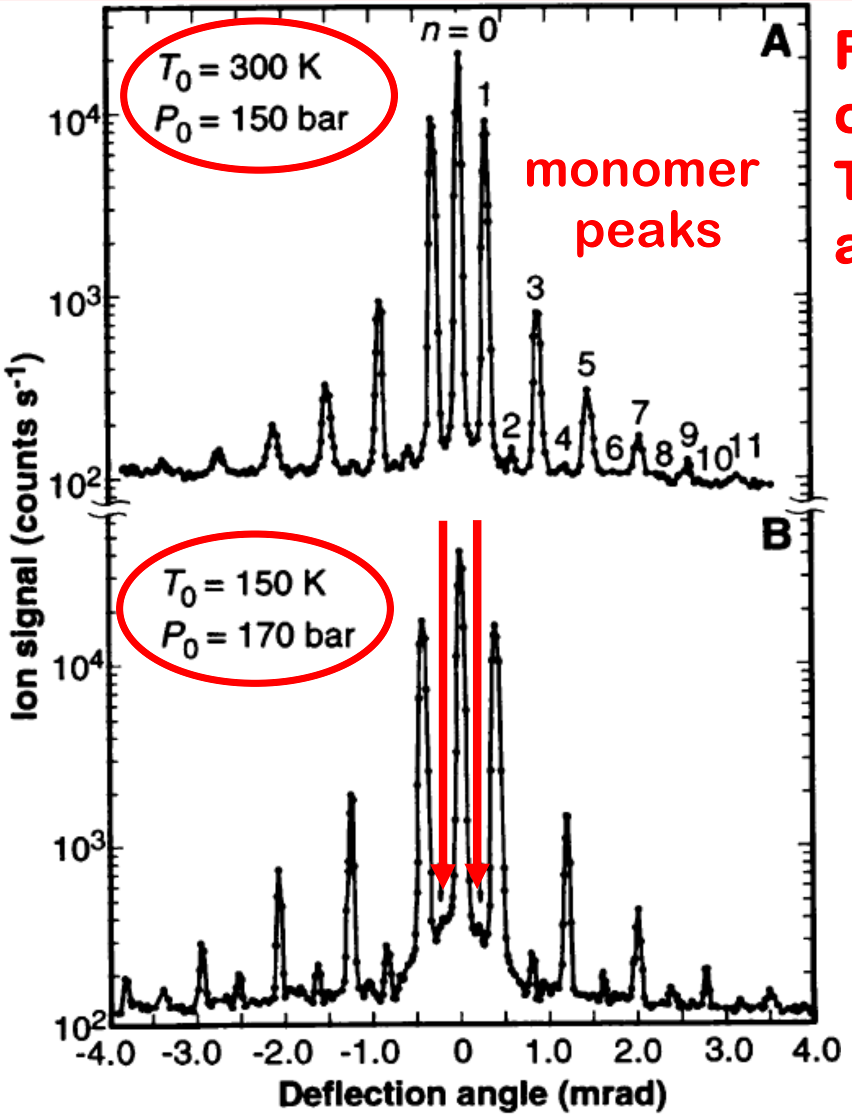
n: diffraction order

m: mass of helium atom

N: number of helium atoms

Nm: mass of molecule or cluster

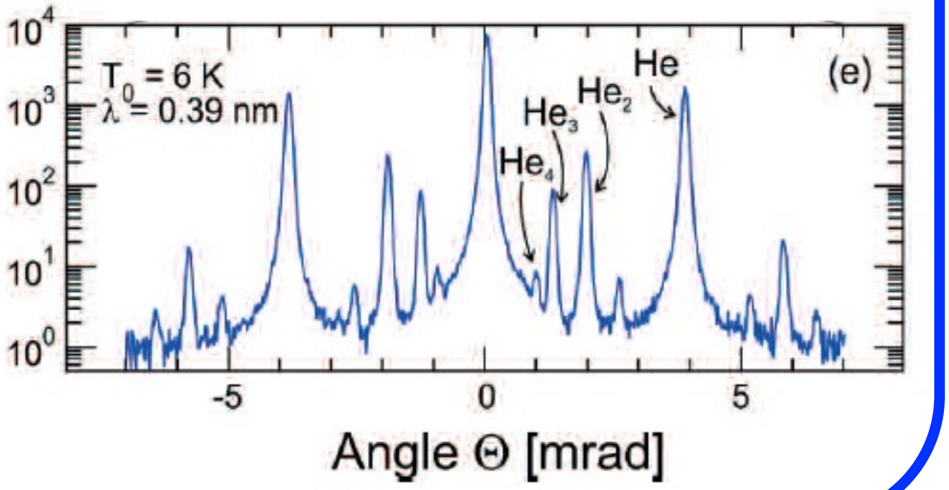
Observation of Bosonic Helium Dimer: $^4He_2$



monomer peaks

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



Helium Dimer

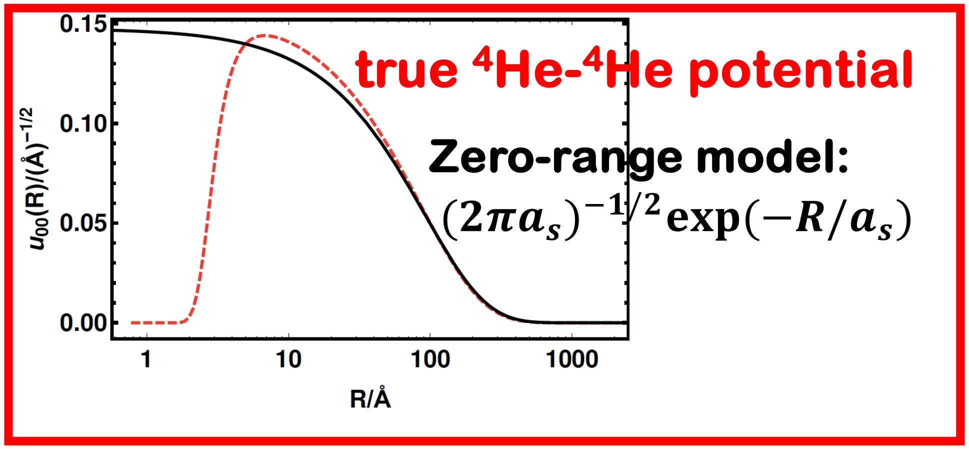
$1\text{ K} = 8.6 \times 10^{-5}\text{ eV}$

- ^{4}He-^{4}He bound state energy $E_{\text{dimer}} = -1.625$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 $r_{\text{eff}} = 15.2a_0$

  (alternatively, two-body van der Waals length $r_{\text{vdW}} = 5.1a_0$).

Large positive a_s:

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



Dynamics discussed in this talk.

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

Bound States of Other Selected Rare Gas Dimers

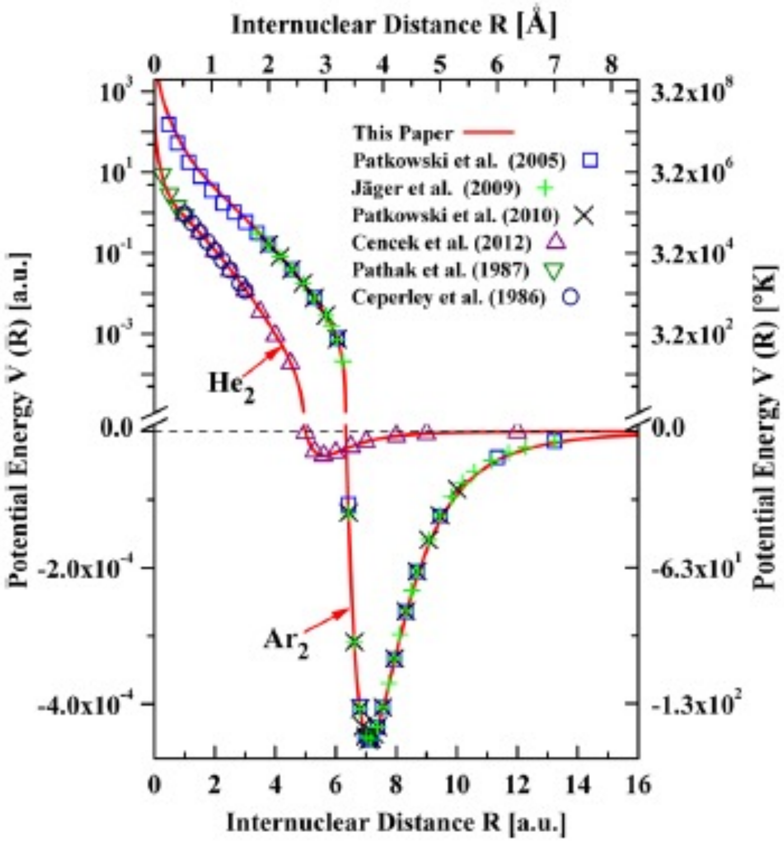
"Conformal analytical potential for all the rare gas dimers over the full range of internuclear distances"

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

Bound states labeled by rotational quantum number J and vibrational quantum number v.

Even J bound states:

^{4}He-^{4}He: 1 bound state ($J = 0$).
Number of bound states increases for ^{4}He-^{20}Ne, ^{20}Ne-^{20}Ne, ^{40}Ar-^{40}Ar.



Sheng, Toennies, Tang, PRL 125, 253402 (2020).

Parametrization of Laser-Molecule Interaction

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

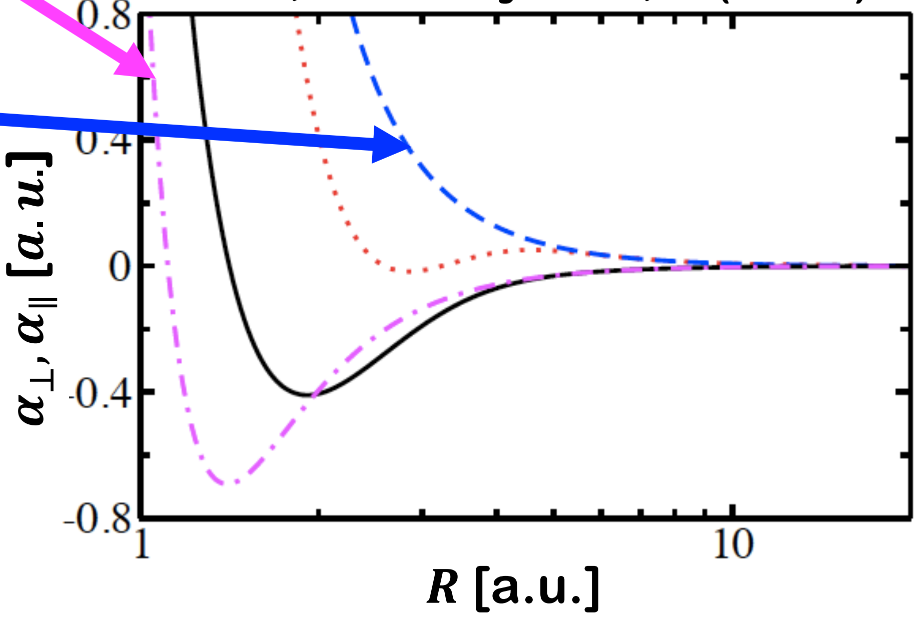
$$V_{\rm lm}(r,\theta,t) = -\frac{1}{2}|\epsilon(t)|^2\,\left[\alpha_{\parallel}(r)\cos^2\theta+\alpha_{\perp}(r)\sin^2\theta\right]$$

$V_{\rm lm}$ **couples different partial waves.**

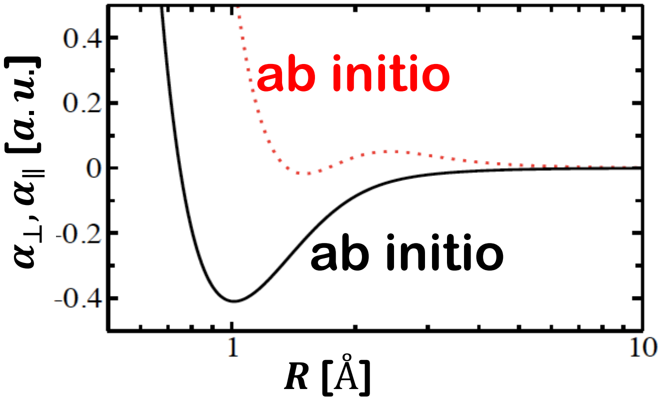
$$\alpha_{\perp}(r) = 2a_0 - \frac{2\alpha_0^2}{4\pi\mathcal{E}_0\,r^3}+\frac{2\alpha_0^3}{(4\pi\mathcal{E}_0)^2\,r^6}$$

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

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



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

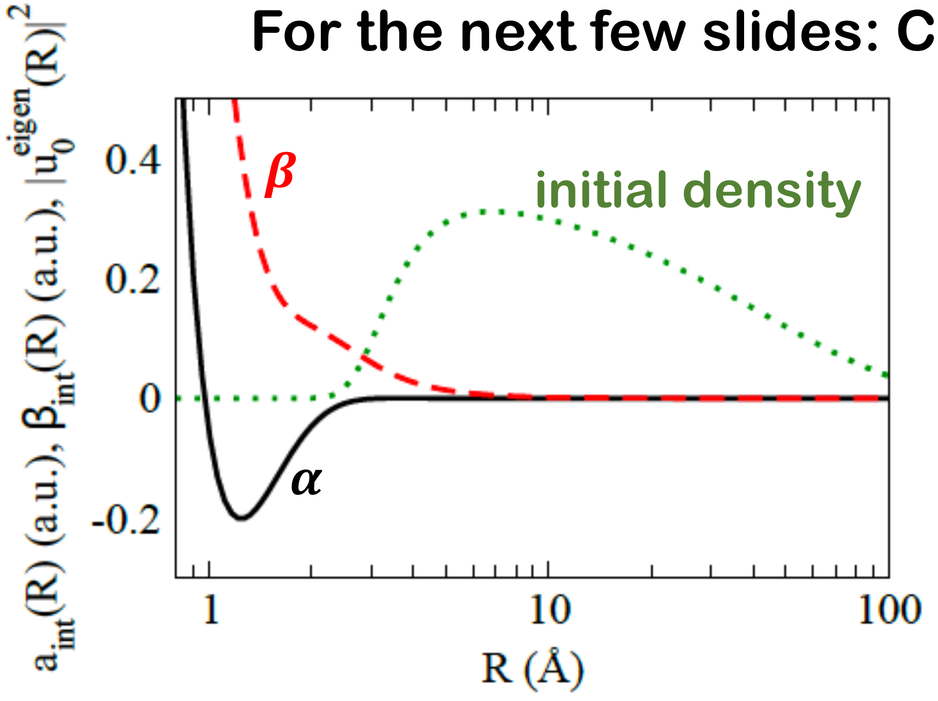


Parametrization of Laser-Molecule Interaction

Laser-molecule interaction:

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

Throughout this talk: Linearly polarized laser.
For the next few slides: Continuous electric field.



Solve coupled channel problem:

Diverging $a_{0,0}$ corresponds to Emergence of new bound state.

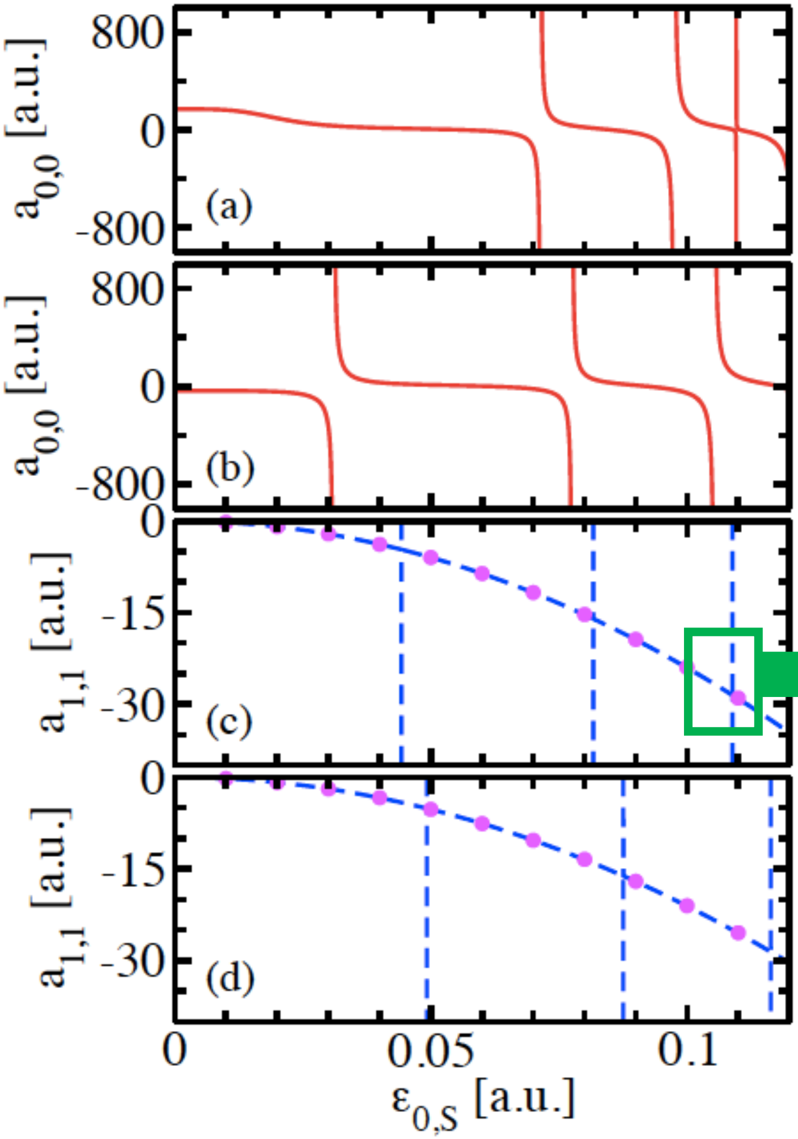
Static External Electric Field: Scattering Lengths For He-He

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

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

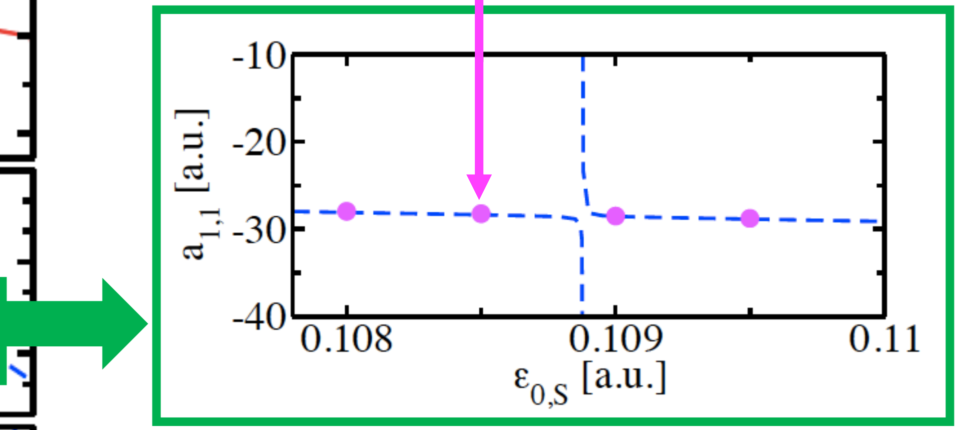
^{3}He-^{4}He odd *J*

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



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

Born approximation works away from resonances



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

Tunability of Helium Dimer: Pure and Mixed Isotopes

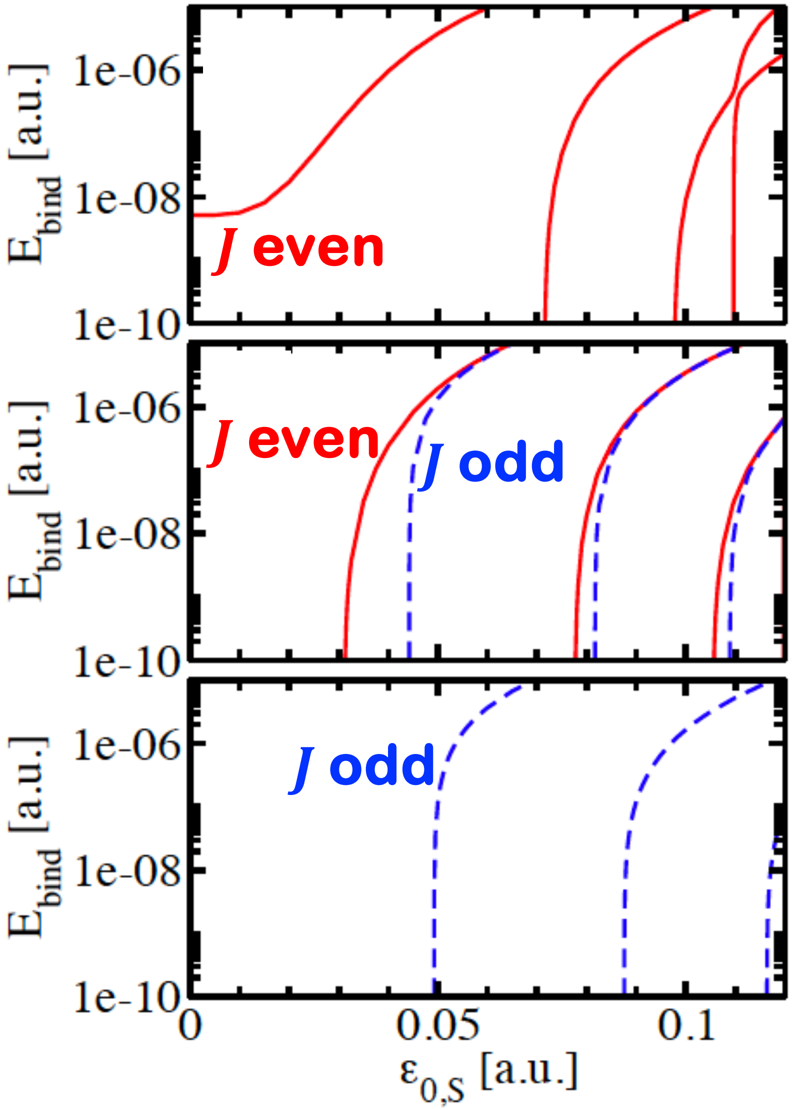
Static electric field (infinitely long pulse).

New bound states appear when $a_{0,0}$ diverges.

^{4}He-^{4}He

^{3}He-^{4}He

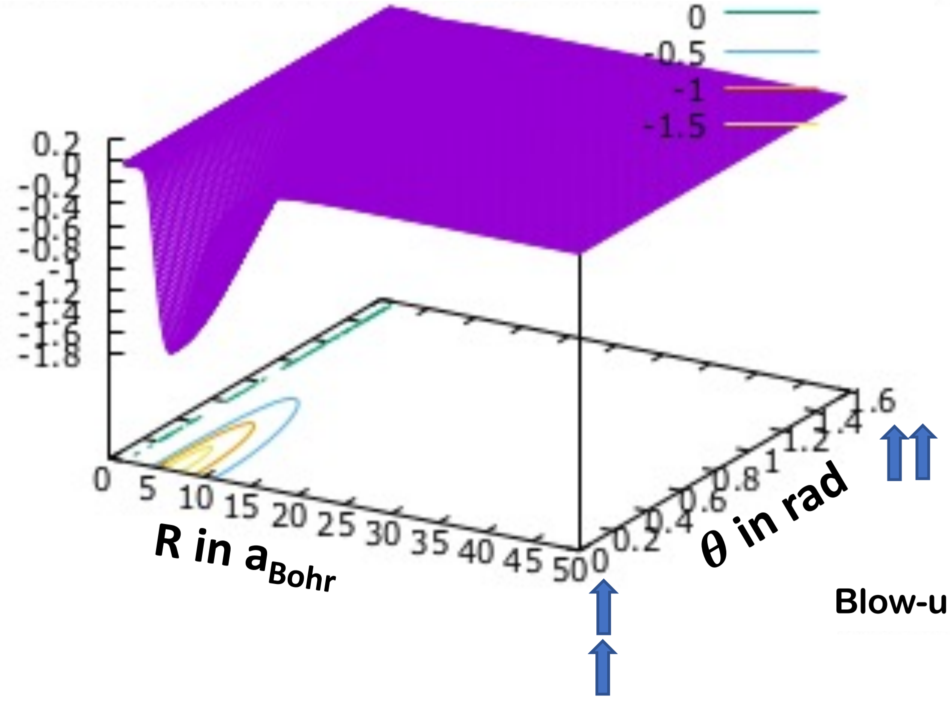
^{3}He-^{3}He



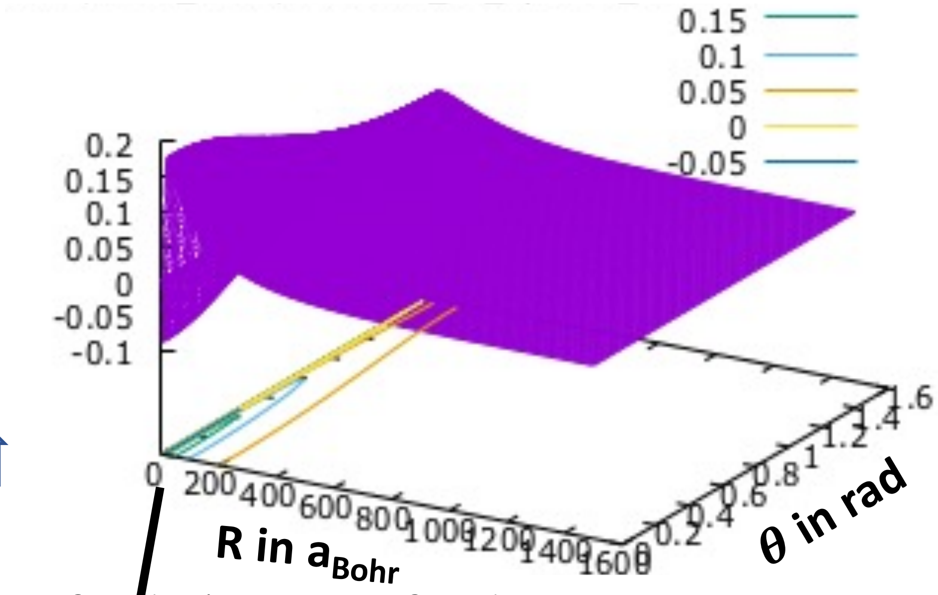
Guan and Blume, PRA 99, 033416 (2019).

Static External Electric Field: Results for $^4\mathrm{He}_2$

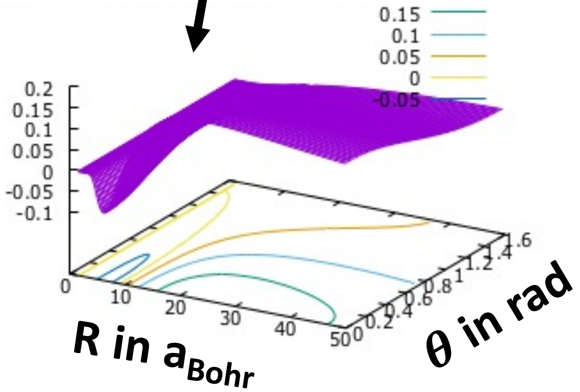
Ground state wave function



Excited state wave function



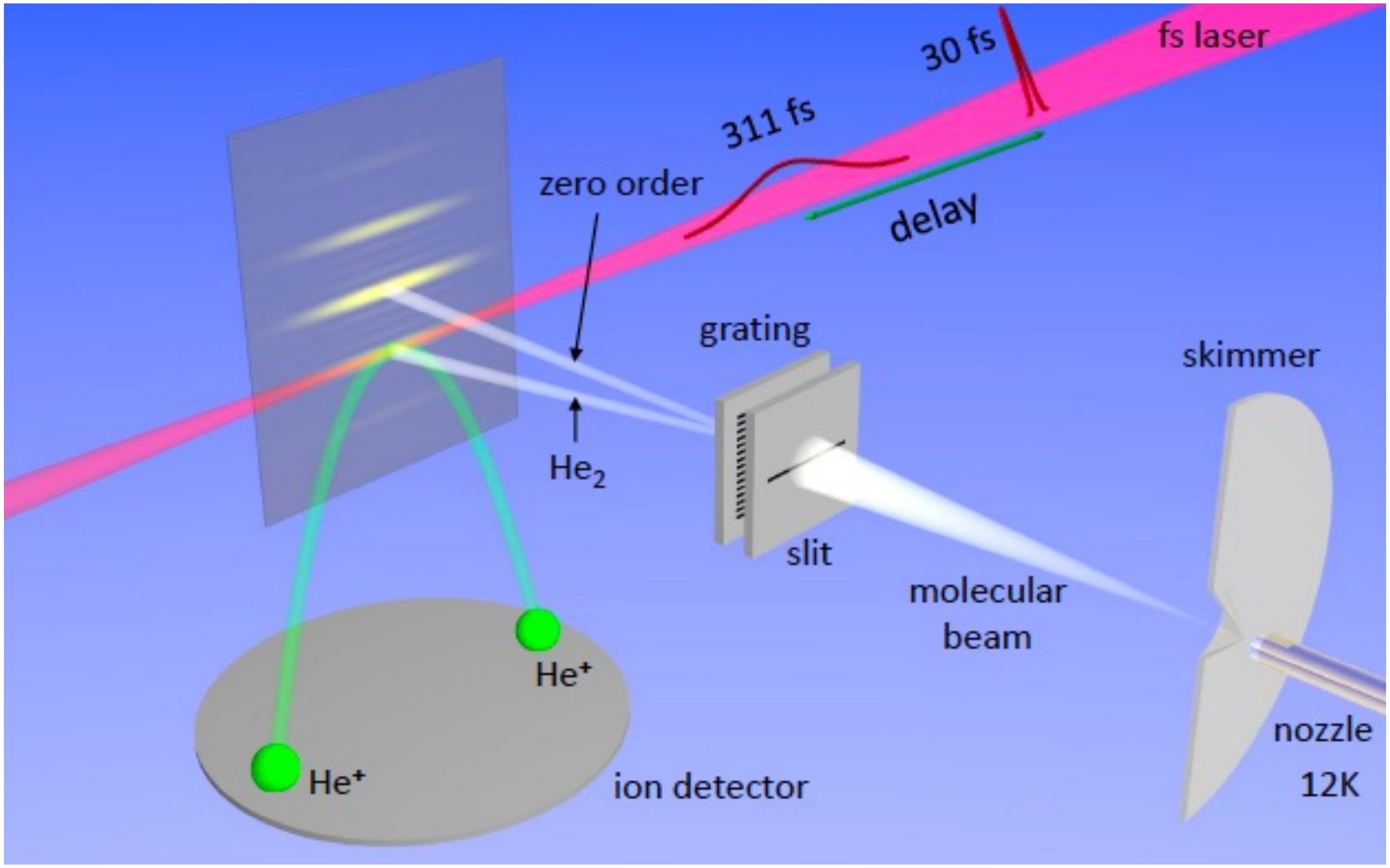
Blow-up of excited state wave function



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

Anisotropy is stronger at small R than at large R .

Pump-Probe Spectroscopy of Isolated Helium Dimers

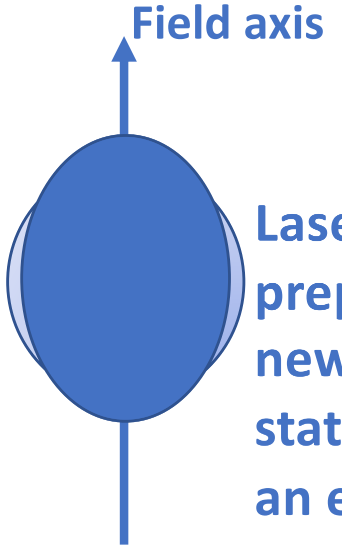


Pump pulse: pulse length of 311 fs and intensity of 1.3 × 10^{14} W/cm^2.
Probe pulse rips off two electrons (Coulomb explosion).
What happens as a function of the delay time???

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 happens as a function of the delay time???

Binding energy of 1 mK 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 “rotational kick.”



Laser pulse prepares new initial state (not an eigen state).

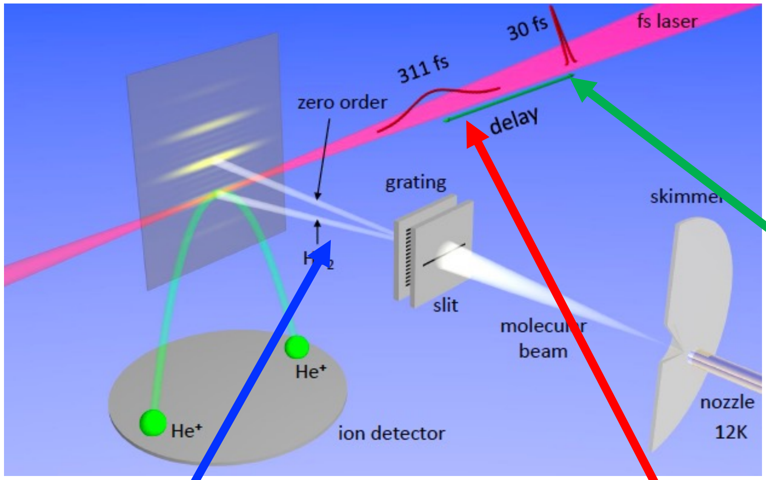
Solar: $\frac{10^3\mathrm{W}}{\mathrm{m}^2}$.

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).

Basic Concept



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

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.

Diatomic Molecule In Time-Dependent Electric Field

Solve time-dependent Schroedinger equation using spherical coordinates:

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

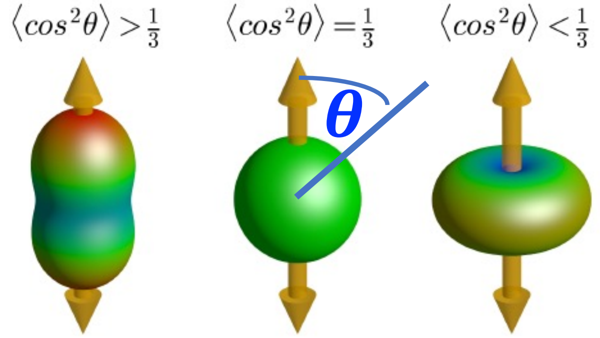
Laser couples different partial waves.
When laser is off, the channels are decoupled.

Initial state:
$J = 0$ eigenstate of the zero-field Hamiltonian of diatomic system.

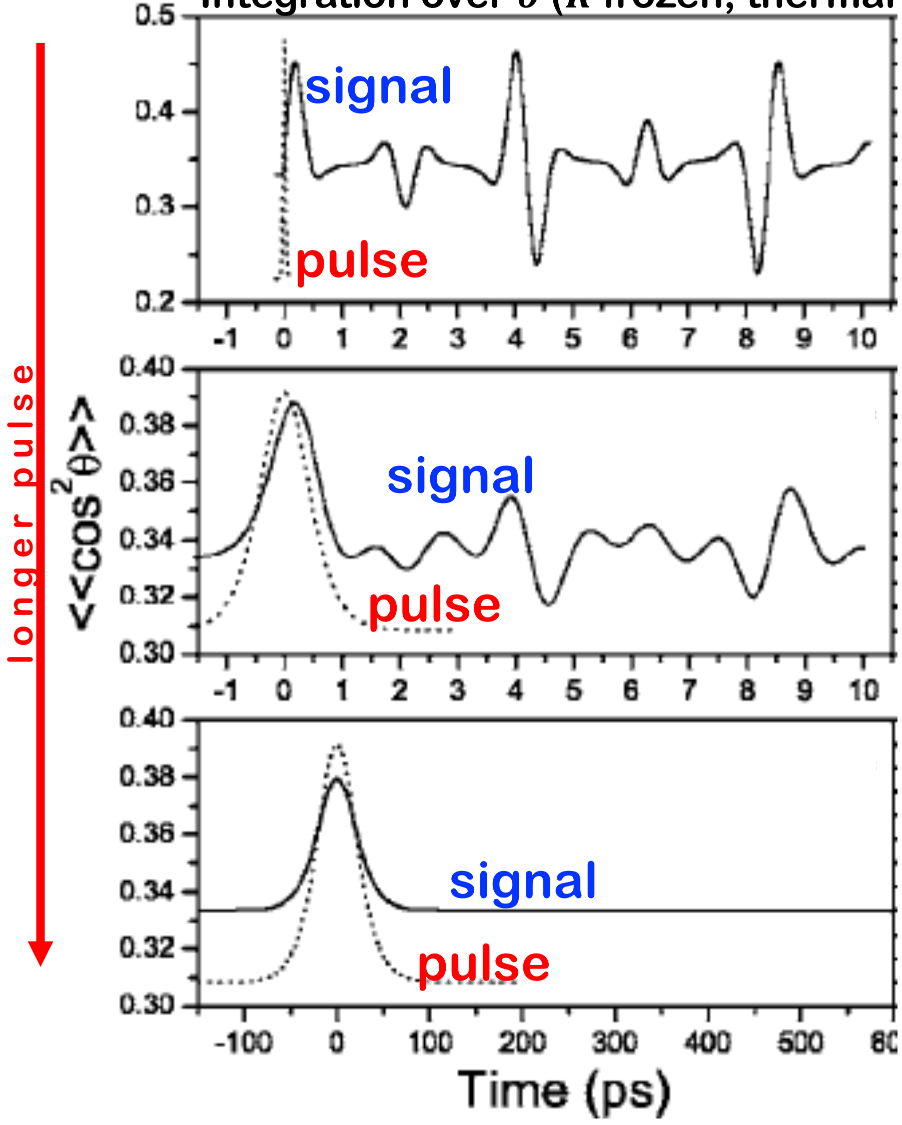
Non-adiabatic Gaussian pump pulse (“rotational kick”):

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

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



Integration over θ (R frozen; thermal distribution).



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

Pulse length 50 fs.

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

Impulse regime.

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

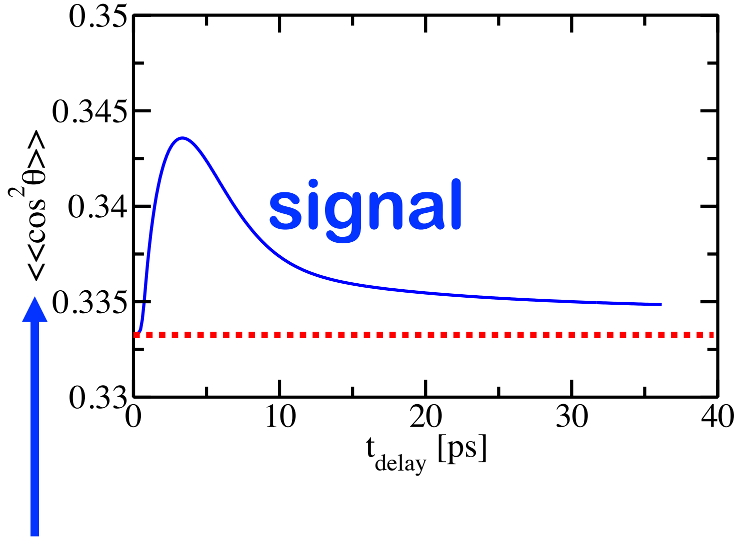
Pulse length 50 ps.

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

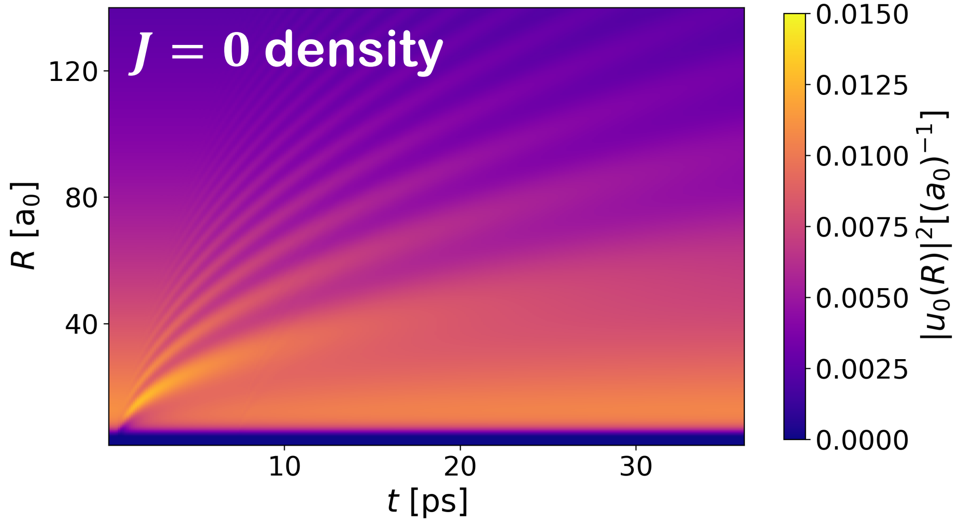
Adiabatic regime.

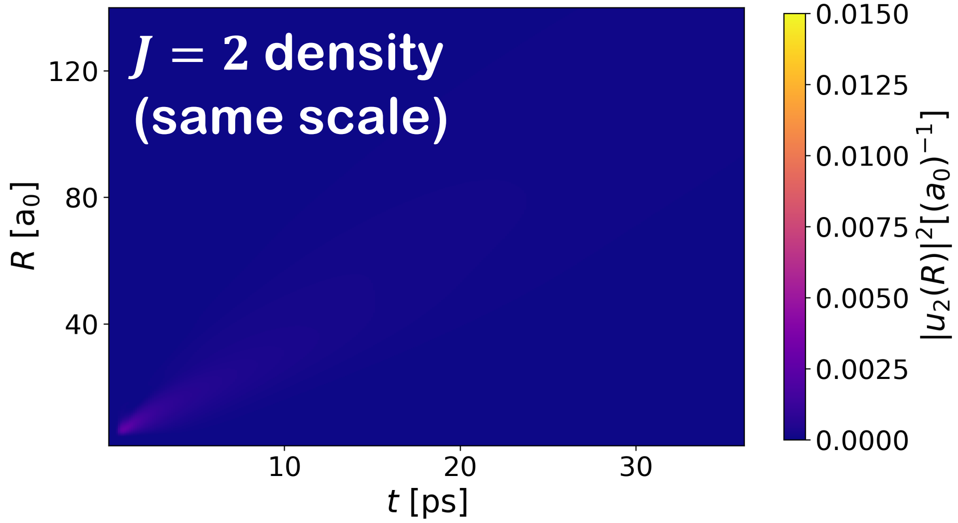
Pump-Probe Spectroscopy of ^{4}He-^{4}He: Rotational Revivals?

Tiny signal, and possibly not overly exciting (?):

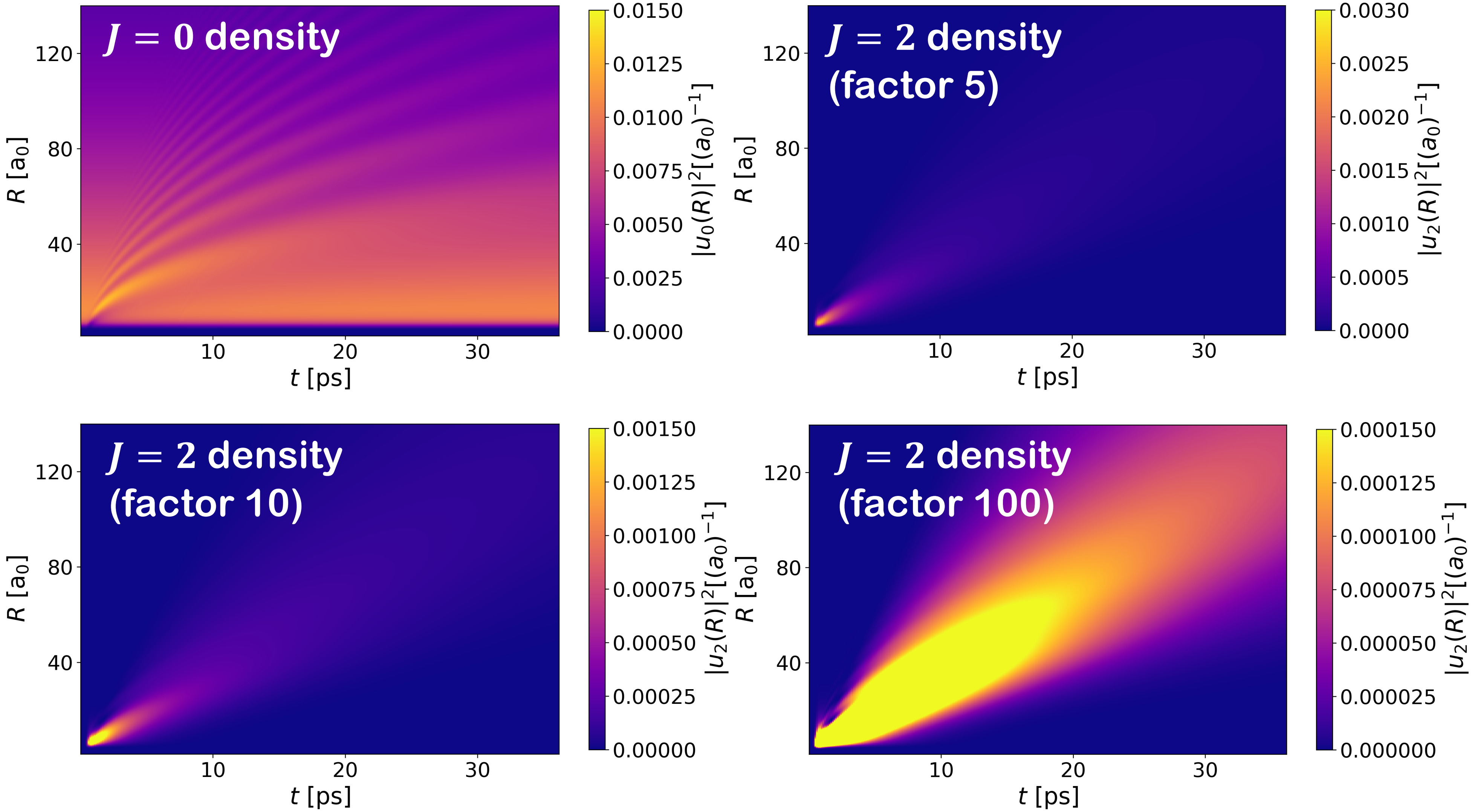


Averaged over R and θ:
Maximum change of 3%.

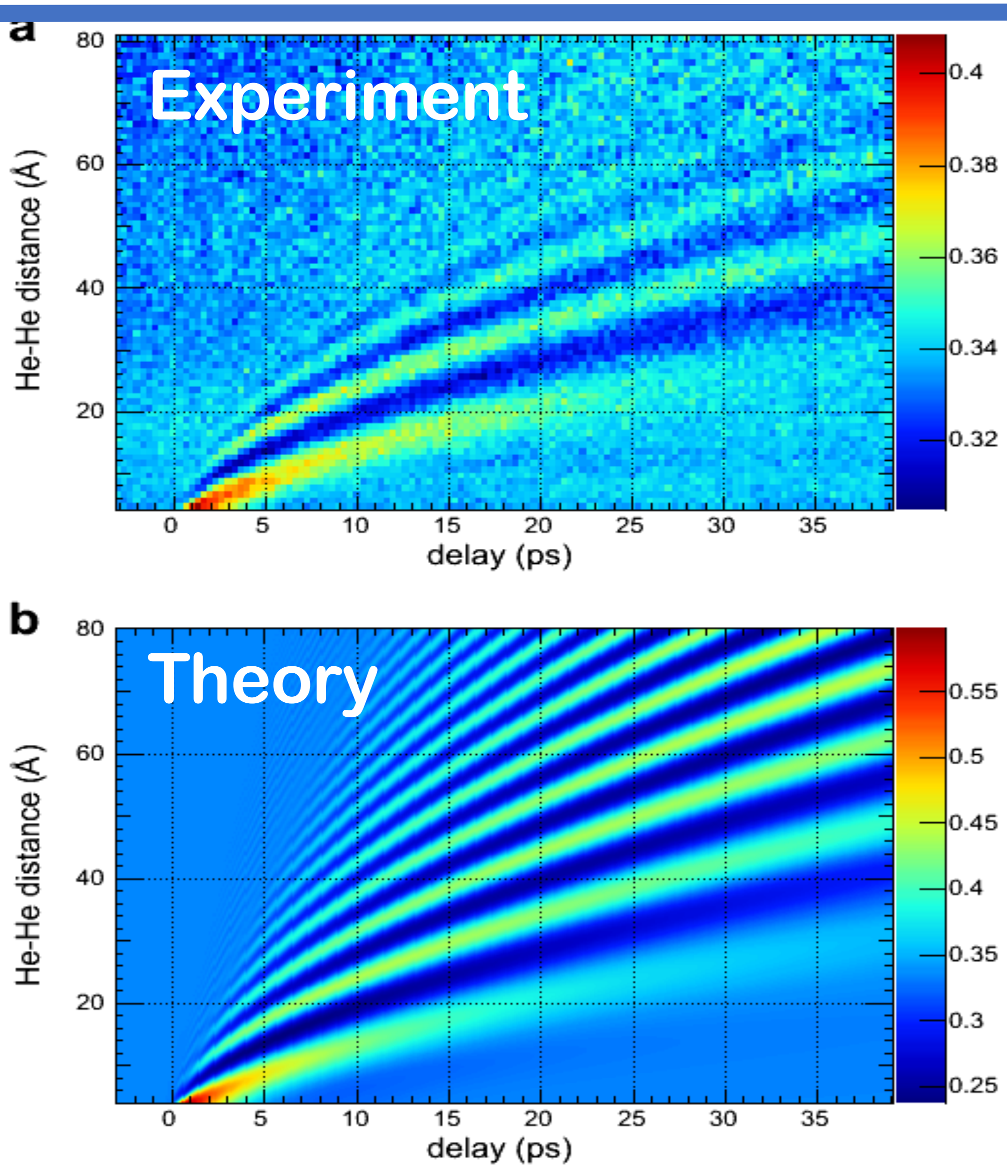




Wave Packet Components of ^{4}He-^{4}He



Alignment Signal $\langle cos^2\theta \rangle$: Experiment and Theory



No integration over R !!!

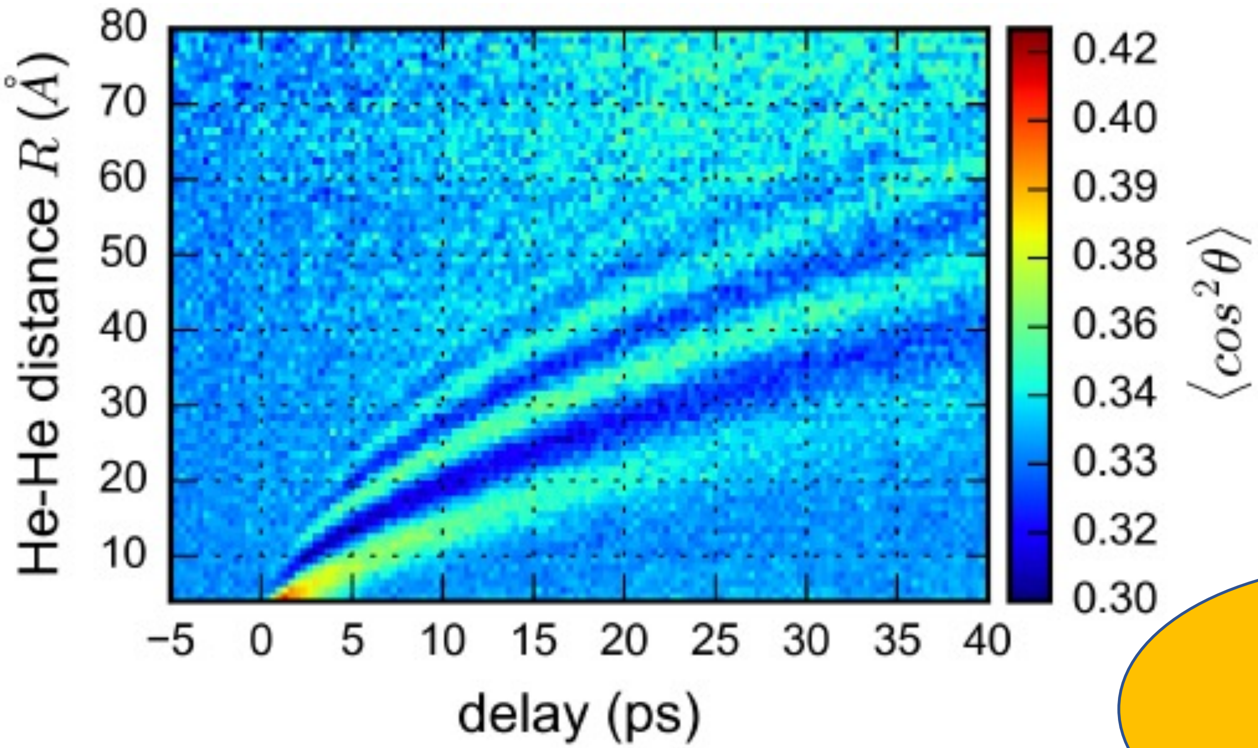
$$\langle cos^2\theta \rangle = \frac{\int_0^\pi \Psi^*(R,\theta,t)\cos^2\theta\Psi(R,\theta,t)\sin\theta d\theta}{\int_0^\pi |\Psi(R,\theta,t)|^2 \sin\theta d\theta}$$

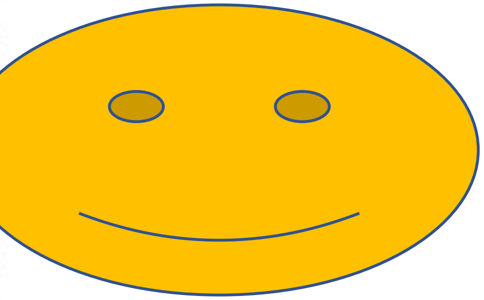
Experimental data by Maksim Kunitski, Reinhard Doerner et al. (Frankfurt University)

Agreement is qualitative but not quantitative.

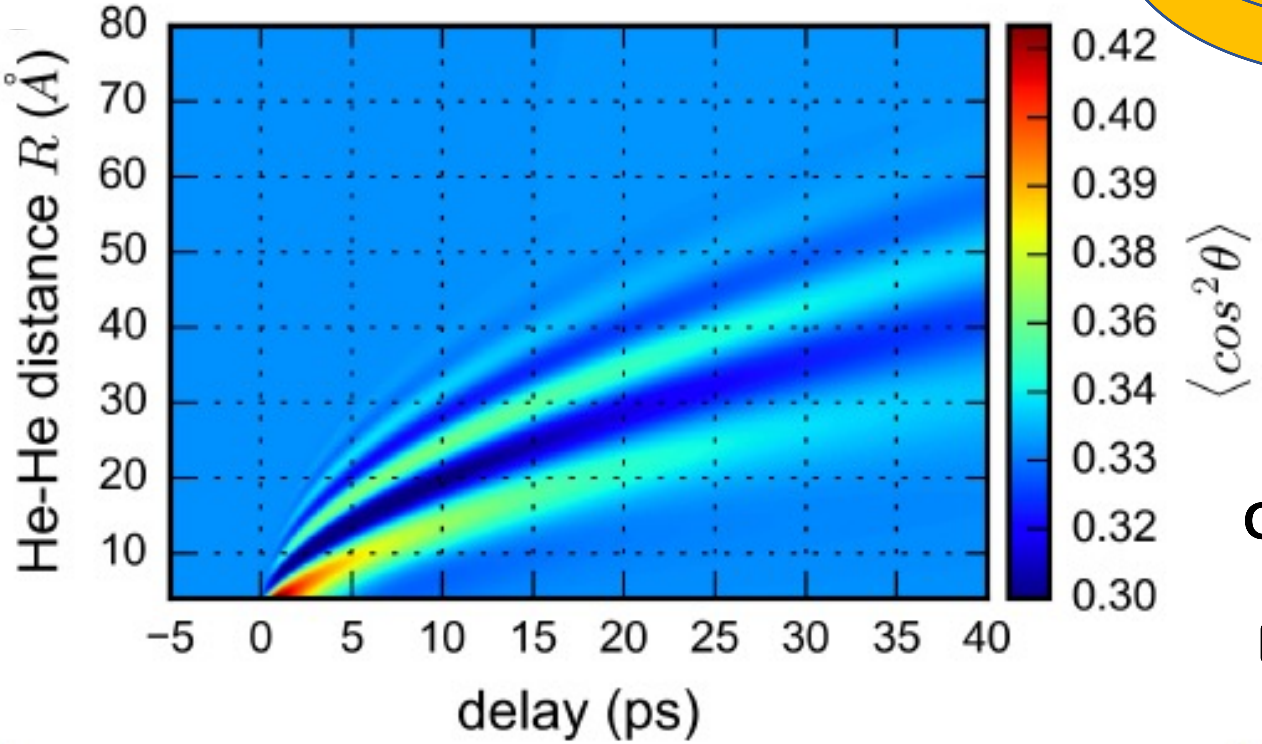
Need to account for finite experimental resolution.

Experiment





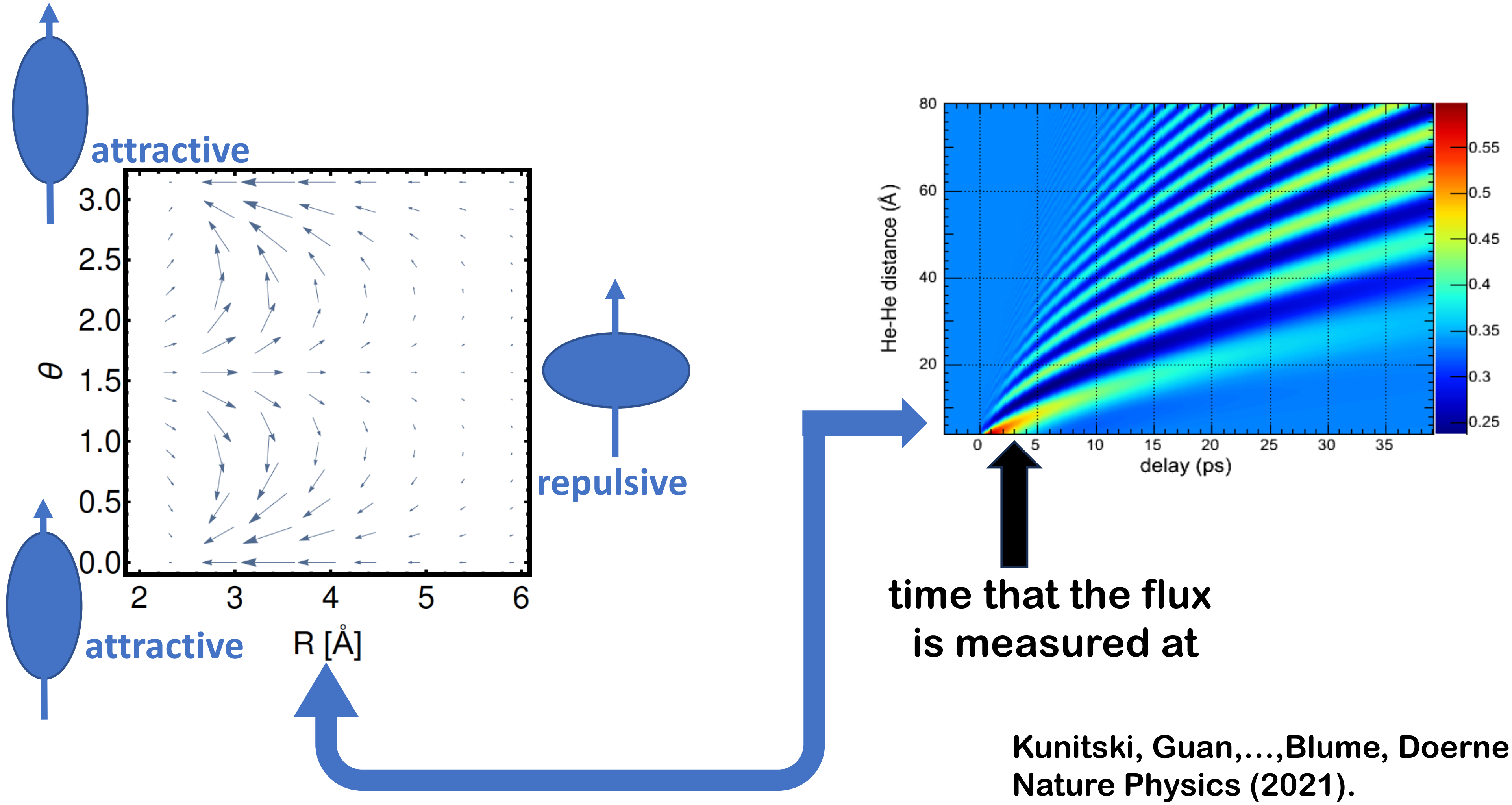
Parameter-free theory (using measured pulse length, intensity, spatial imaging resolution)



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

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

Flux after the pump pulse has decayed to zero:



time that the flux is measured at

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

Origin Of The Interference Pattern?

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

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

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

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

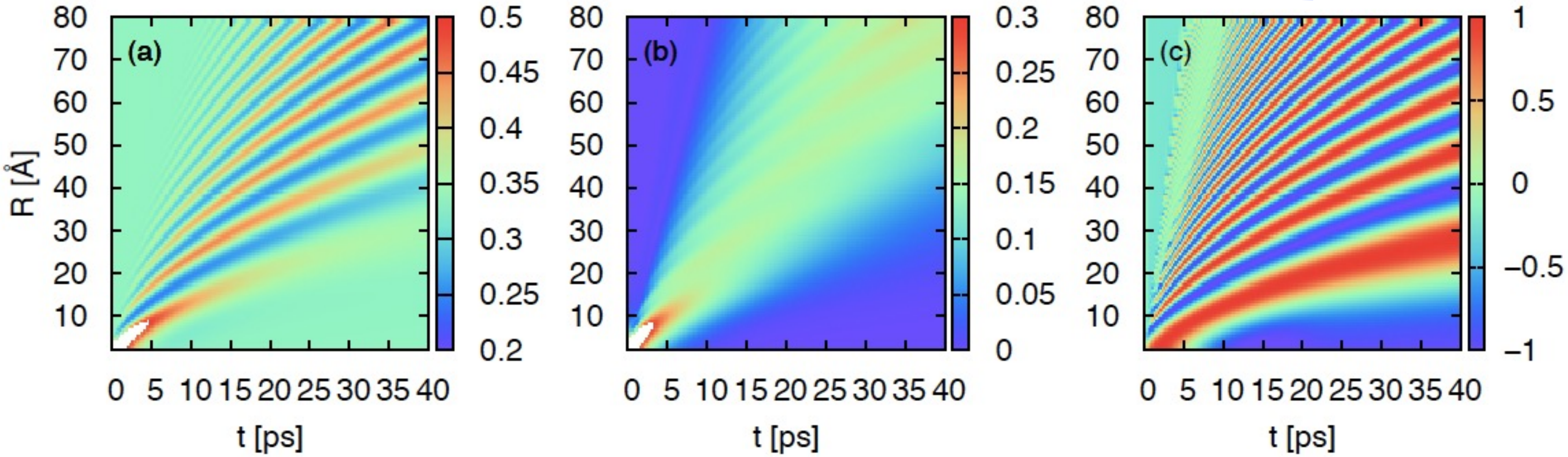
$$\boxed{C_2(R,t) = \frac{1}{3} + \frac{4}{3\sqrt{5}}\left|\frac{u_2(R,t)}{u_0(R,t)}\right| \cos(\gamma_2(R,t) - \gamma_0(R,t)) + \cdots}$$

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

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

Only plotting $J = 0$ and 2

$\gamma_0 \approx const$

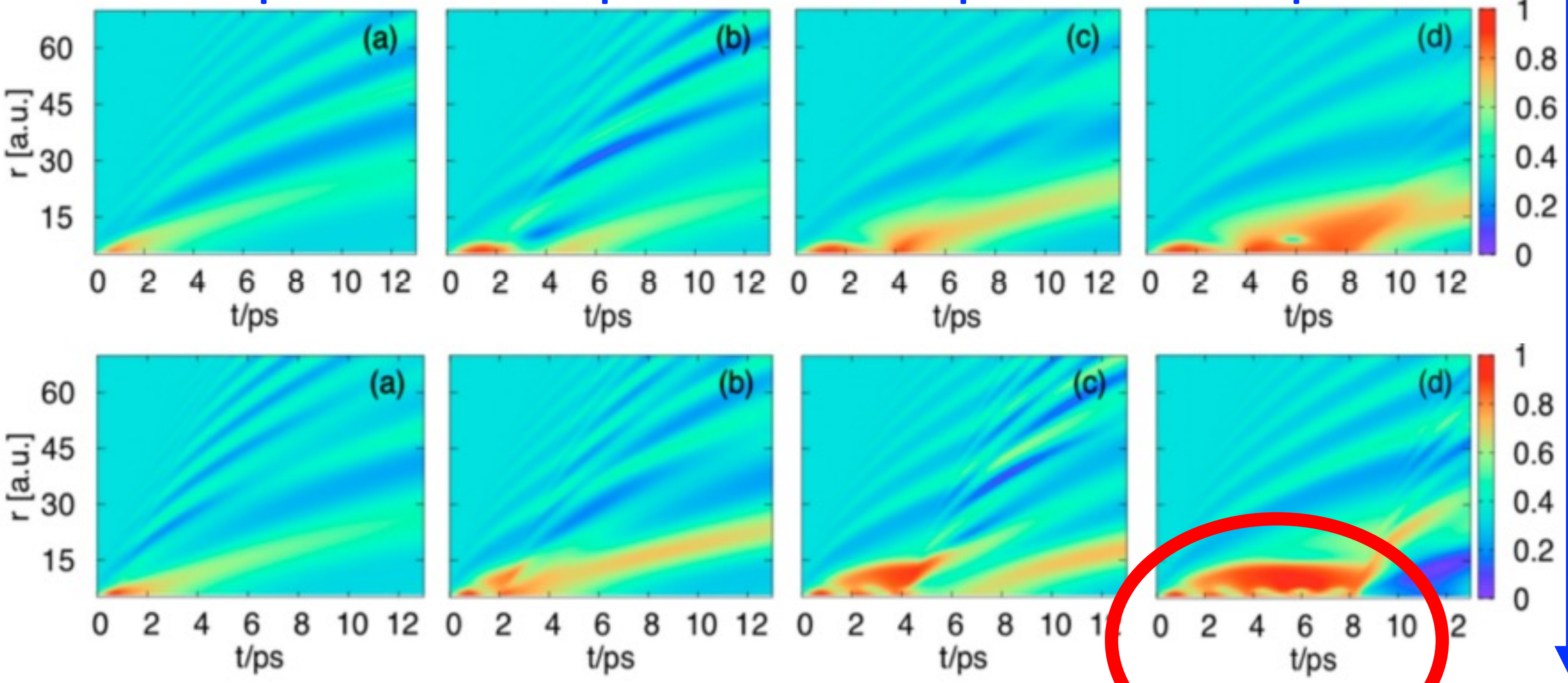


Alignment signal $cos^2\theta$ can be interpreted as measuring $\gamma_2(R,t)$.

^{4}He-^{4}He: Longer Pulses

increasing pulse length

0.5 ps 2 ps 4 ps 8 ps

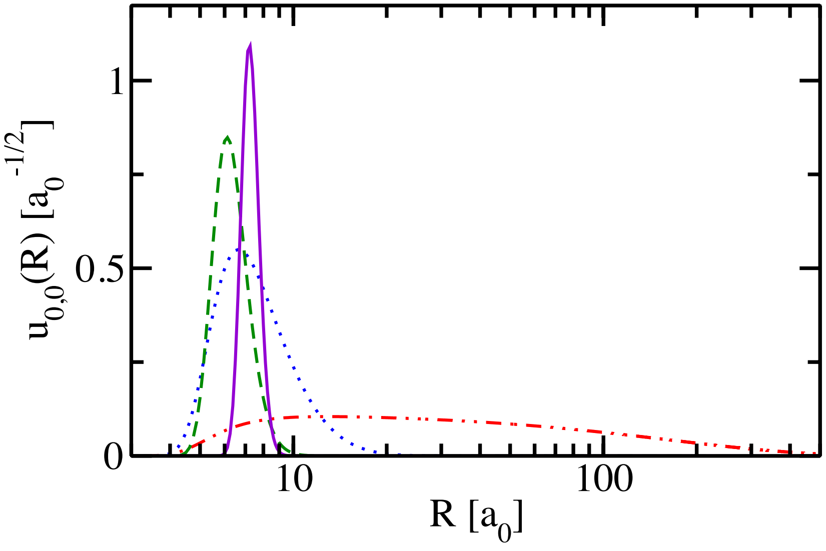


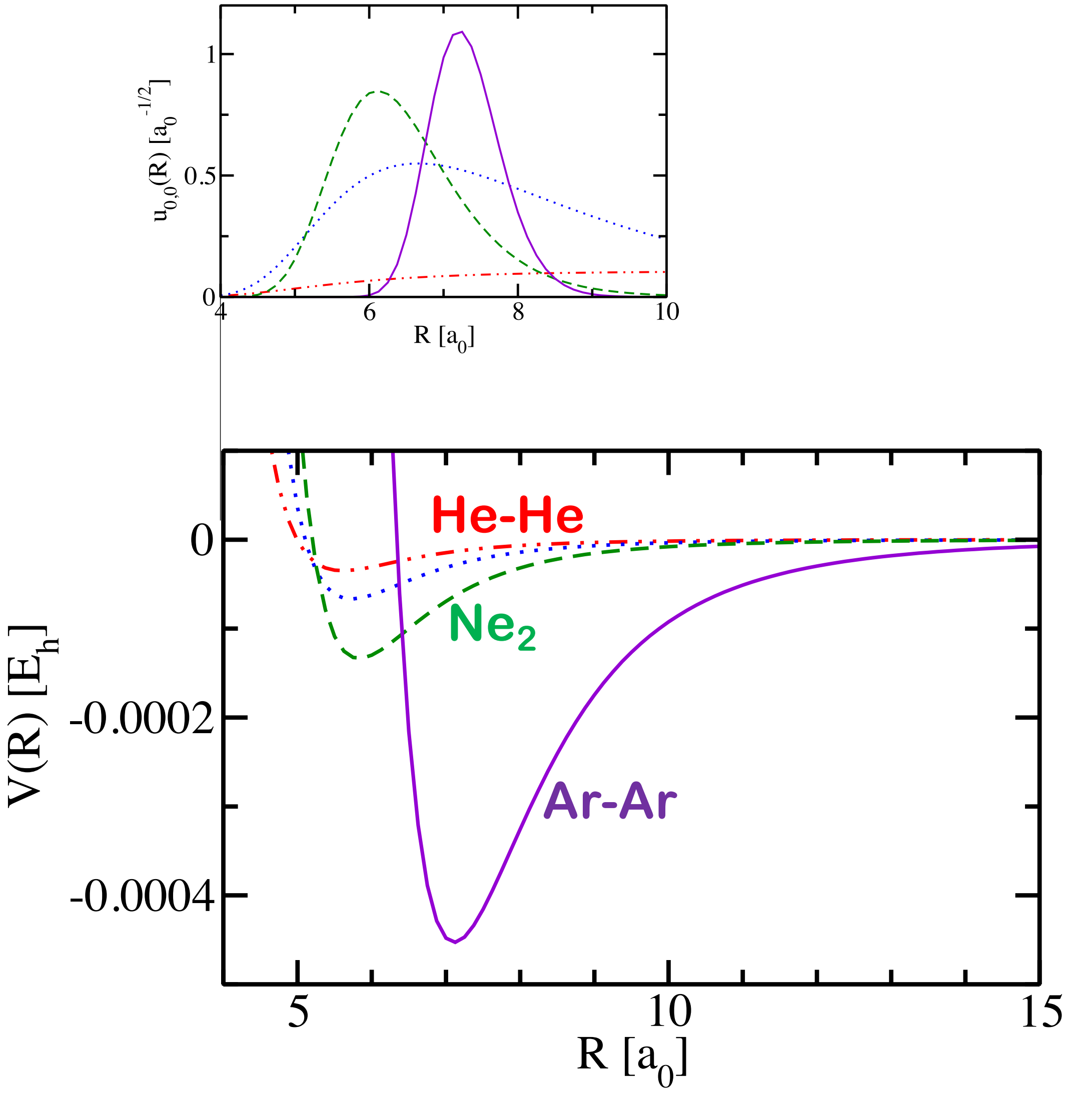
increasing intensity

revivals!

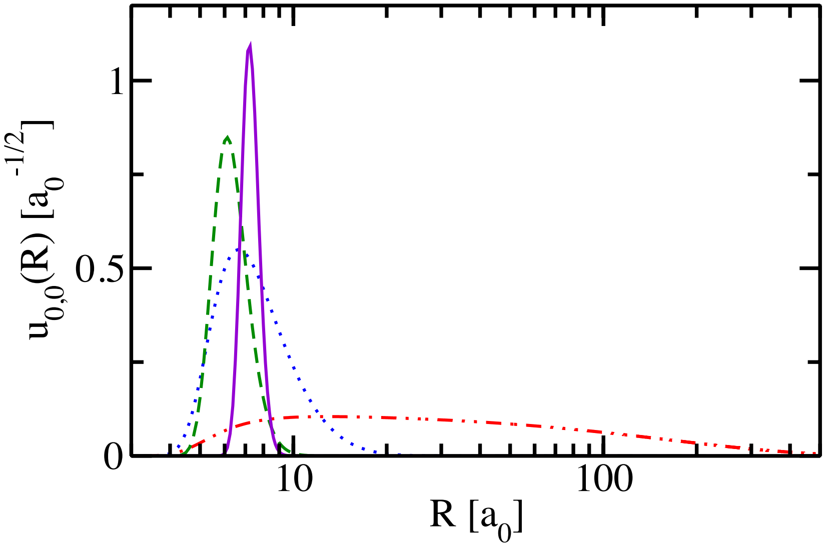
Guan and Blume, PRA 99, 033416 (2019). Awaiting experimental realization…

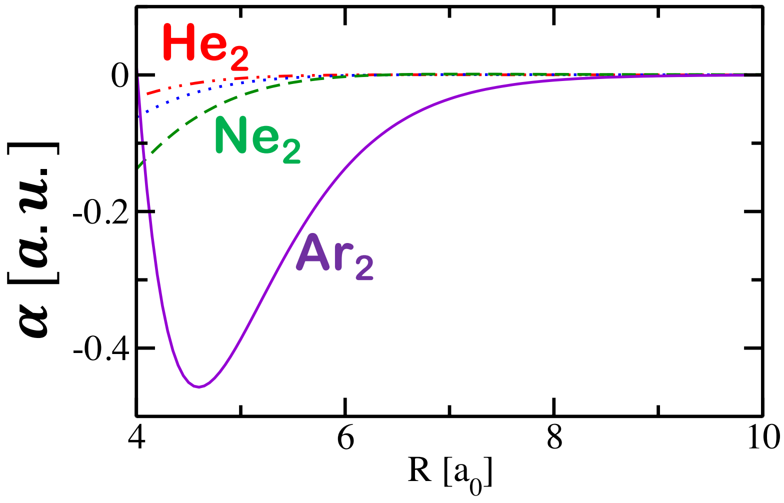
Other Rare Gas Dimers: Initial State





Other Rare Gas Dimers: Polarizabilities

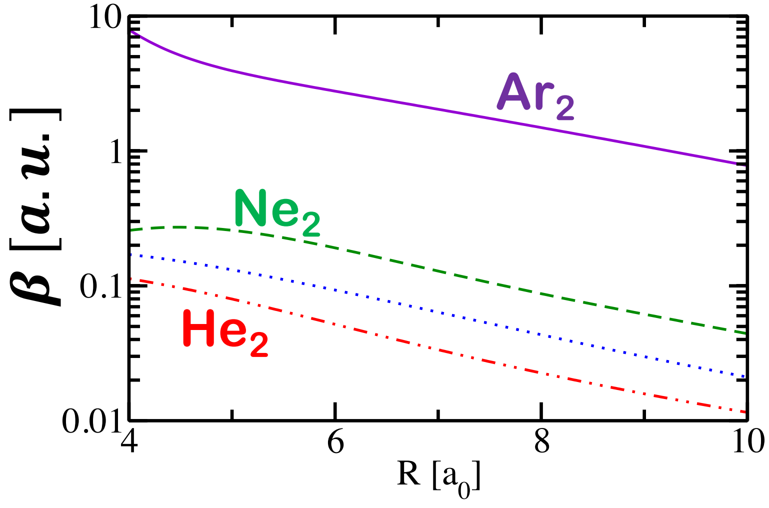




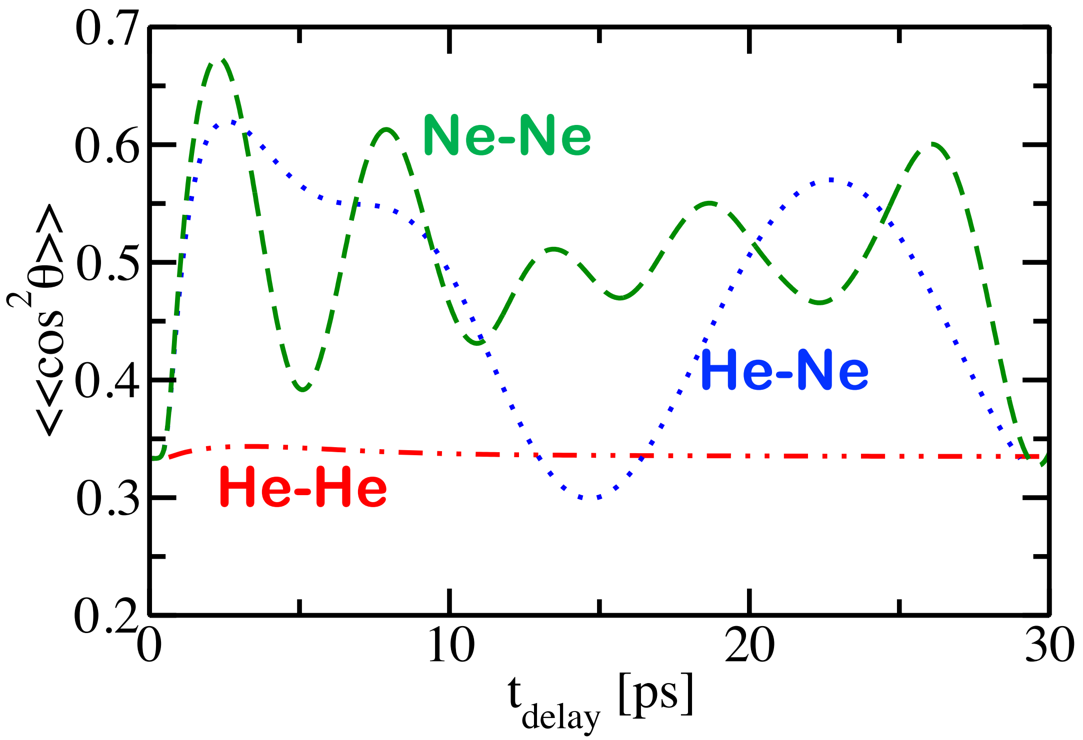
Laser-molecule interaction:

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

$|\alpha|$ and $|\beta|$ increase as the molecule becomes heavier and more compact.



Comparison of $\left\langle\langle cos^2\theta\rangle\right\rangle$ for He-He, He-Ne, and Ne-Ne

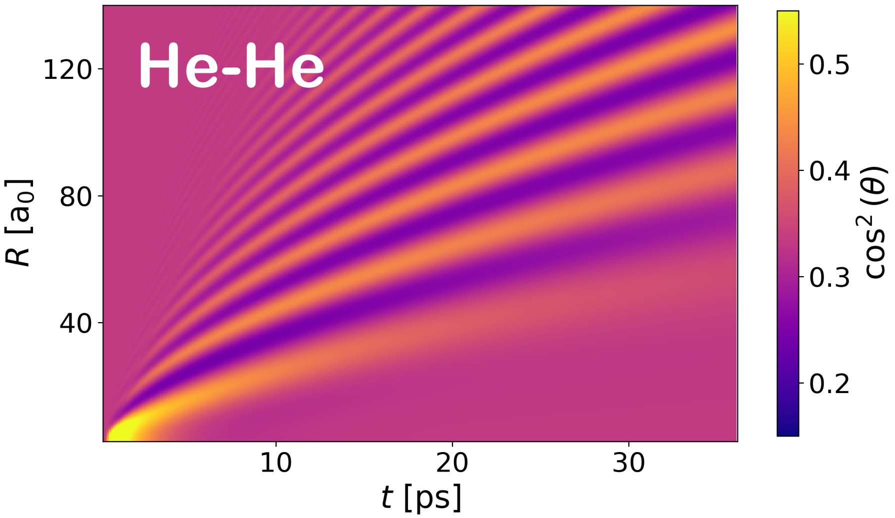


^{20}Ne-^{20}Ne:

Vibrational energies ($J = 0$):
Energy difference = 19.798 K
$\Rightarrow$ 2.44 ps

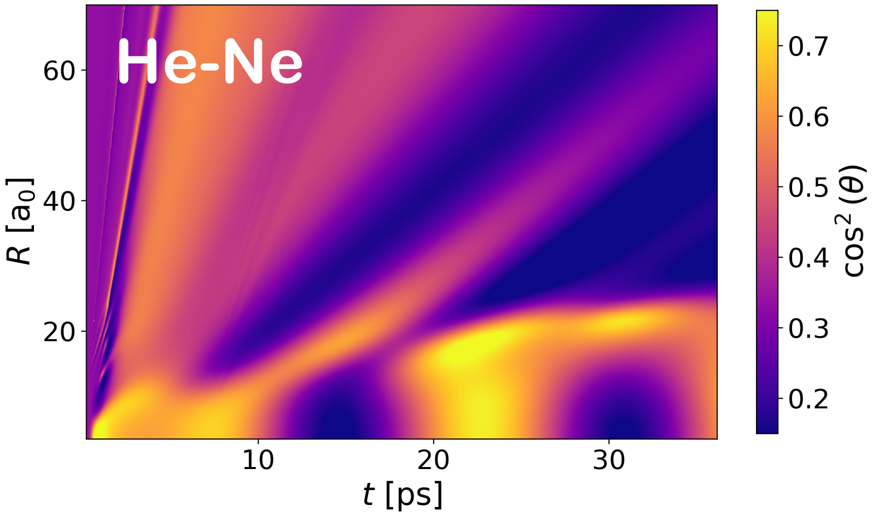
Rotational energies ($J = 0$ and $J = 2$):
Energy difference = 1.349 K
$\Rightarrow$ 35.87 ps

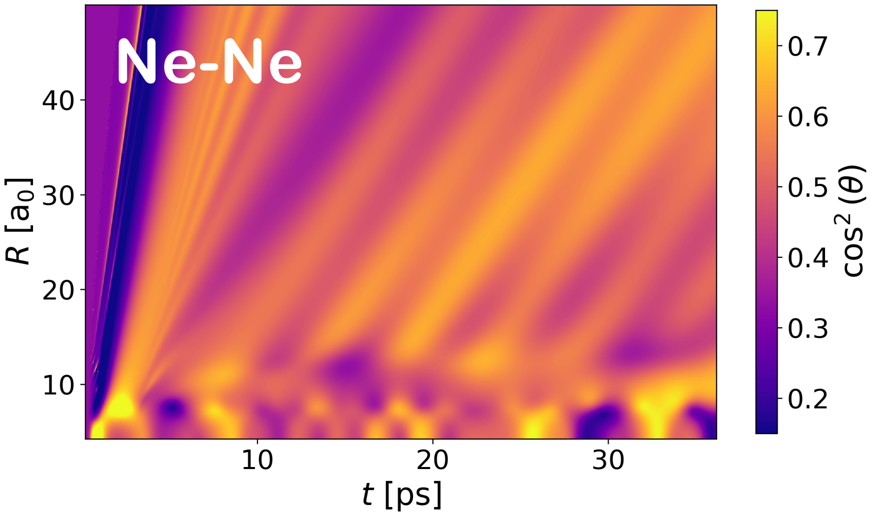
Comparison of $\langle cos^2\theta \rangle$ for He-He, He-Ne, and Ne-Ne



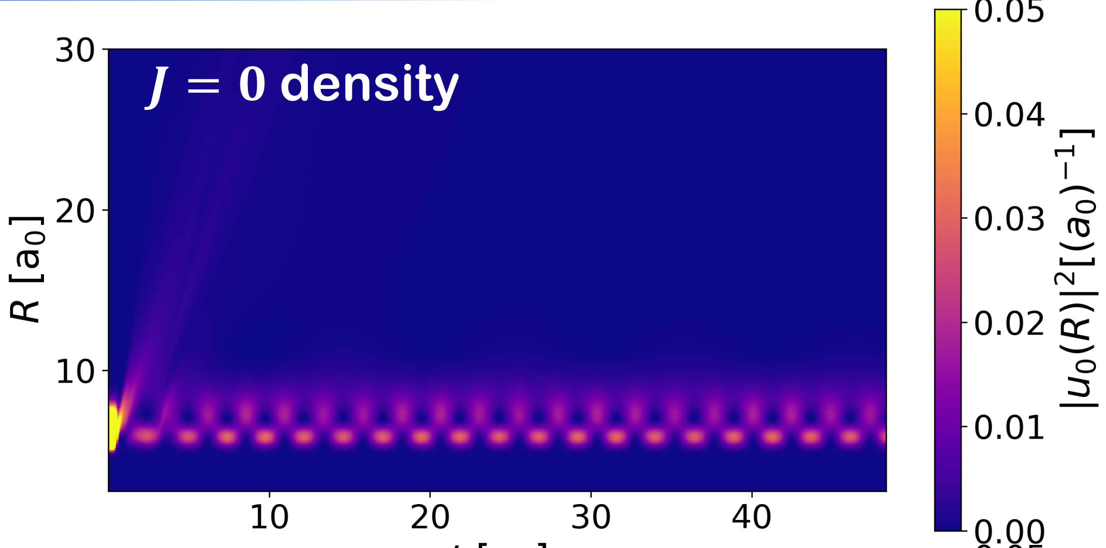
Helium-helium systems is clearly unique:

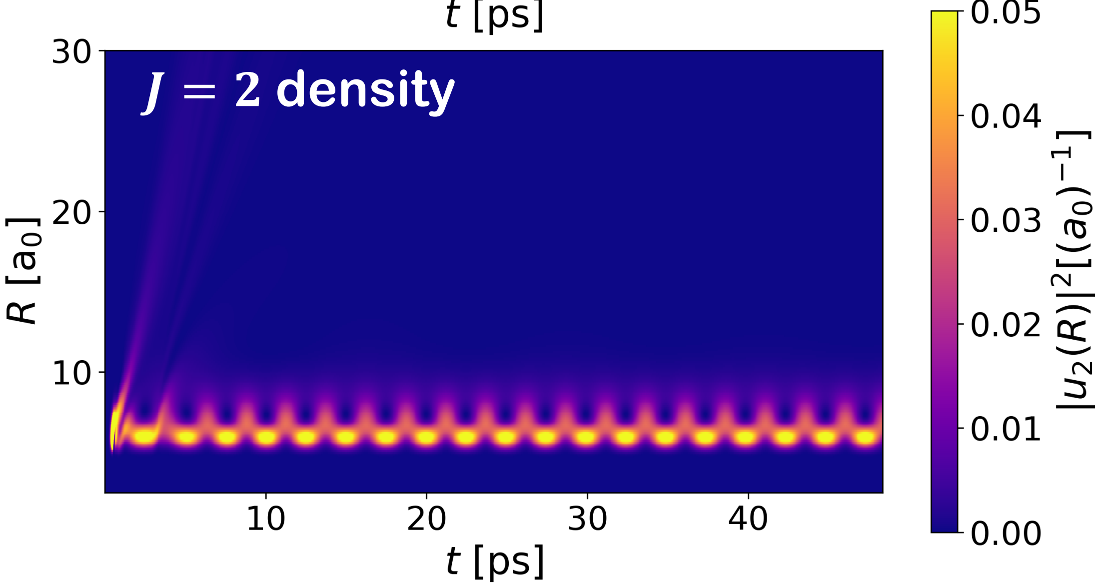
- **Only one bound state.**
- **Broad s-wave initial state.**

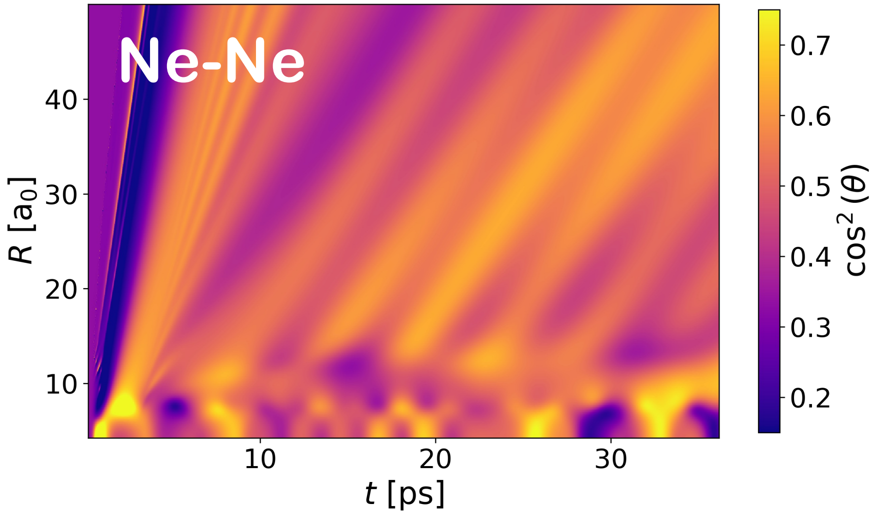




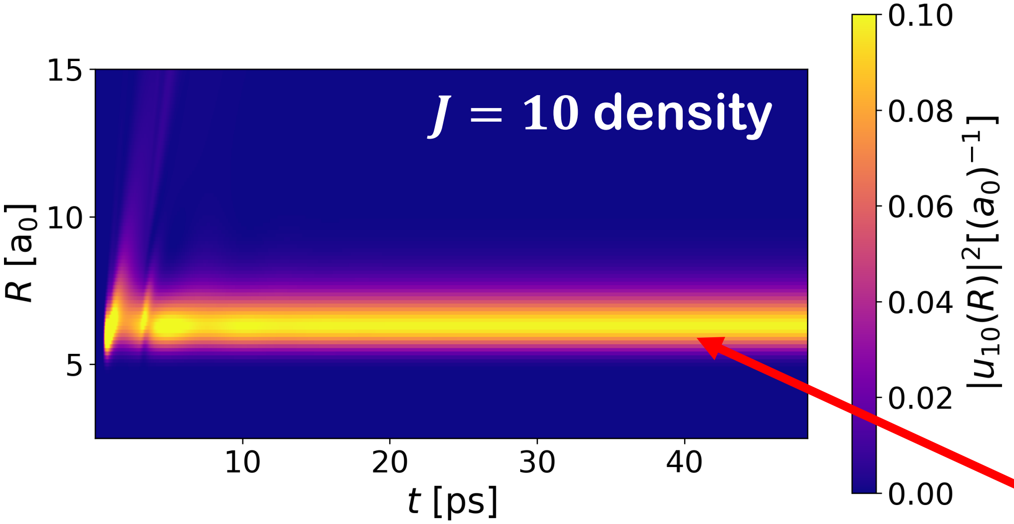
Ne-Ne: Clear Signature of Vibrational Dynamics

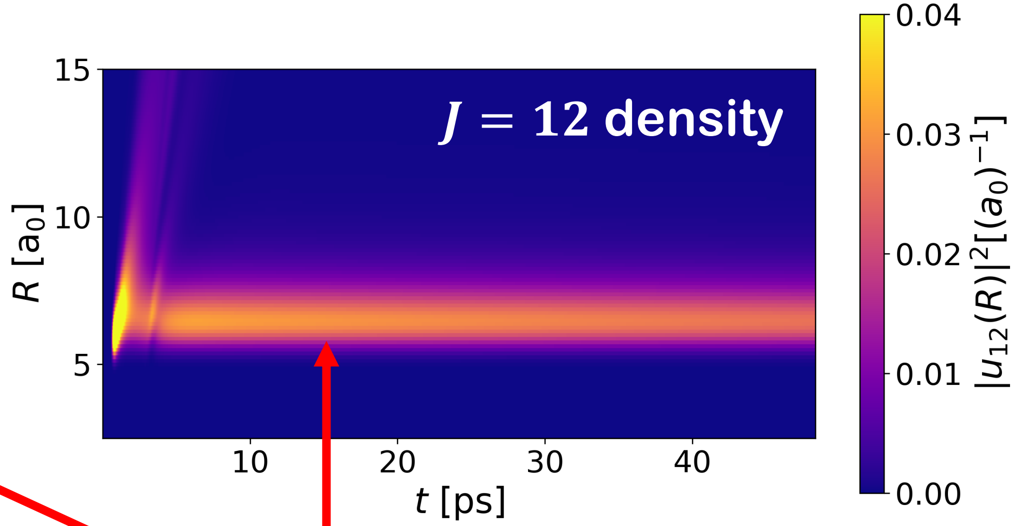


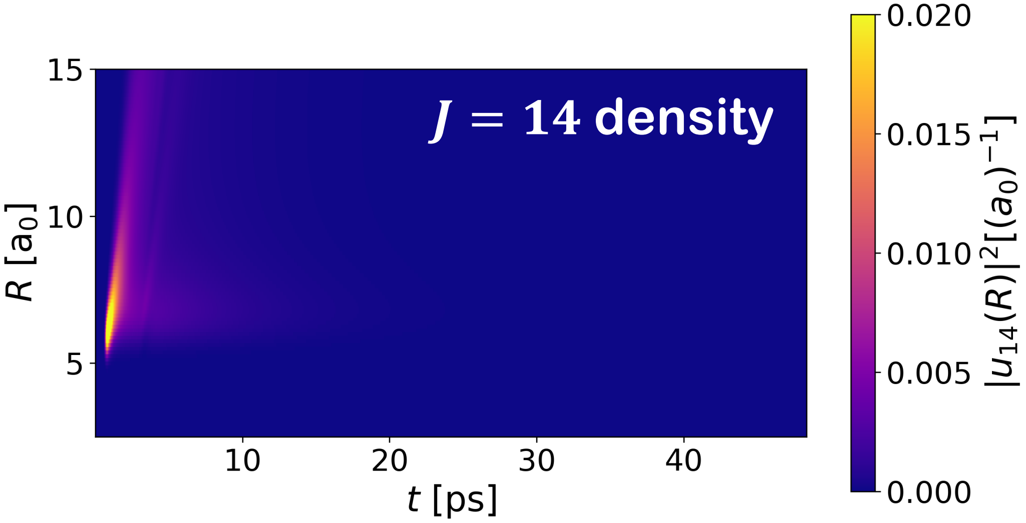




Ne-Ne: Clear Signature of Quasi-Bound State



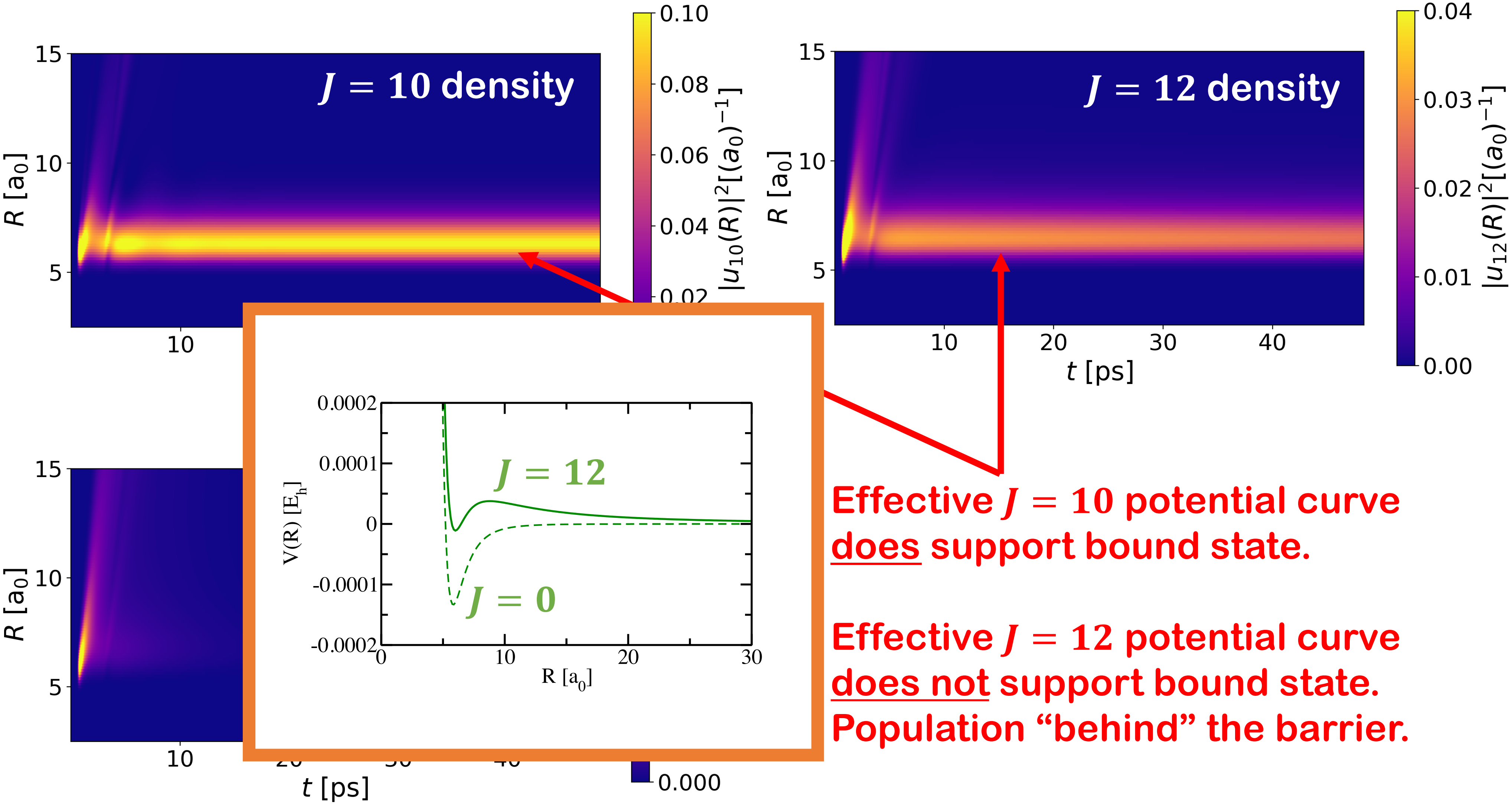




Effective J = 10 potential curve <u>does</u> support bound state.

Effective J = 12 potential curve <u>does not</u> support bound state. Population "behind" the barrier.

Ne-Ne: Clear Signature of Quasi-Bound State



Effective J = 10 potential curve <u>does</u> support bound state.

Effective J = 12 potential curve <u>does not</u> support bound state. Population “behind” the barrier.

Pump-Probe Spectroscopy: 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 and other “light” rare-gas dimers adds “physical dynamics” to the list!

Summary

- Pump-probe spectroscopy (pump laser = rotational kick) of weakly-bound molecules: Entirely new regime (completely different from rotational revivals observed for heavy molecules).
- Observed rich interplay of rotational and vibrational degrees of freedom.

- Helium dimer:
  - Excellent agreement between theory and experiment.
  - Non-zero R-dependent dissociative alignment.
  - Tiny population of $J = 2$ partial wave component.
- Neon dimer:
  - Low-lying J-component display vibrational dynamics.
  - Appreciable population of many J-components (for same pump laser intensity and pulse length as for helium dimer).
  - Pump-pulse occupies quasi-bound $J = 12$ partial wave component.

Dynamics of Weakly-Bound Molecules

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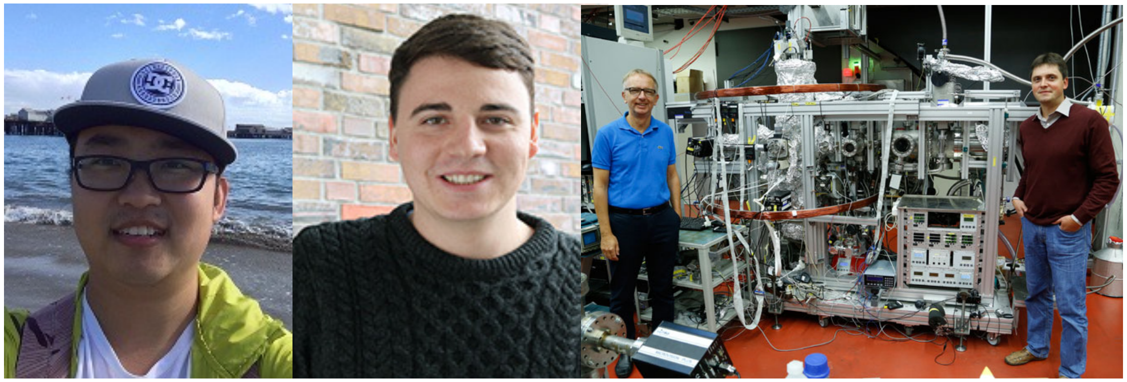
Jan Kruse, Maksim Kunitski, Reinhard Doerner + group (Frankfurt)



Supported by the NSF.