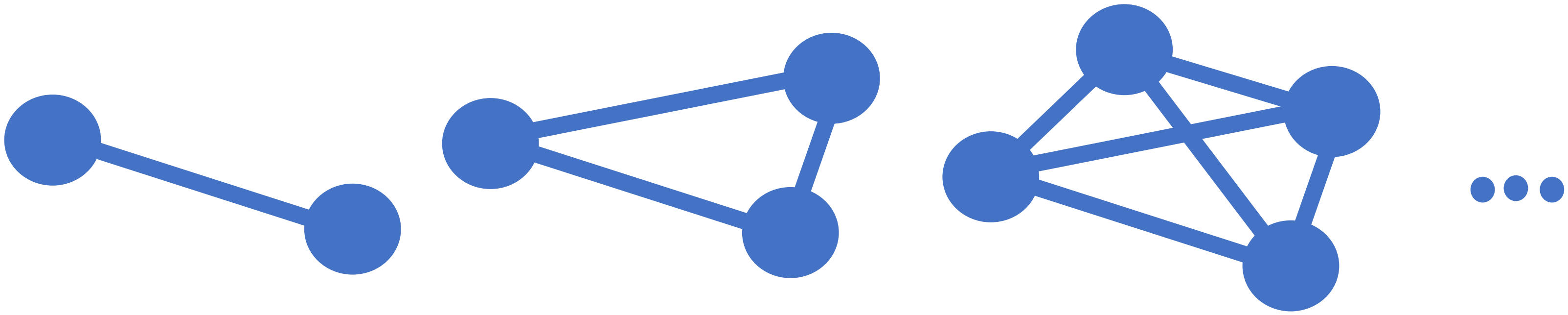


Loosely bound van der Waals clusters

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

Collaborators:
Qingze Guan, AJ Yates, Maksim Kunitski,
Reinhard Doerner and group.

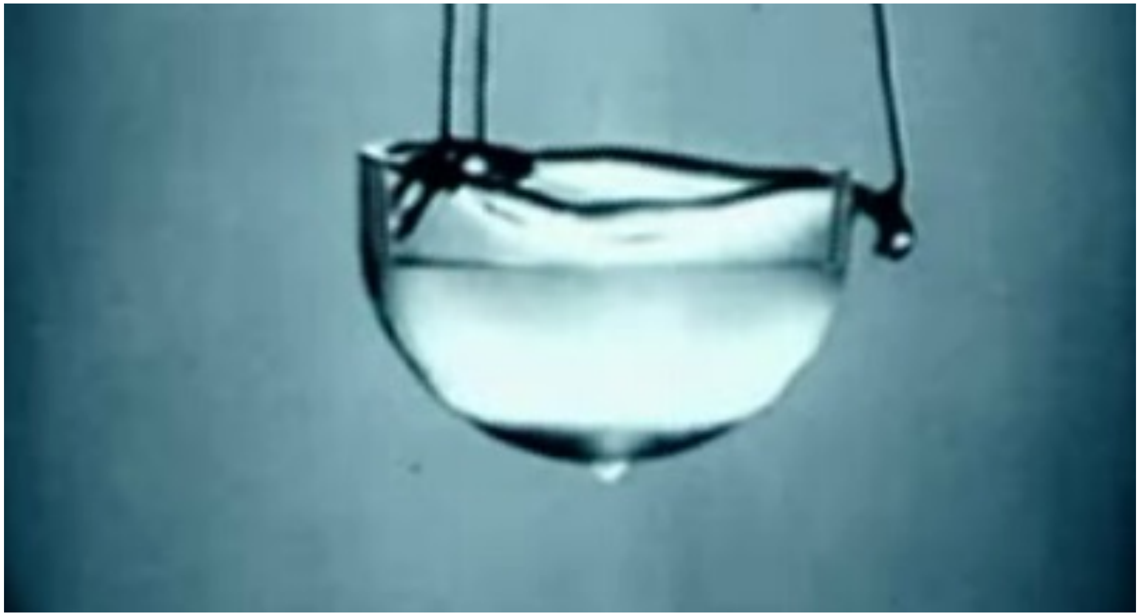
Supported by the NSF.

Superfluid Bulk Helium-4

- ^{4}He is the only substance that remains liquid under normal pressure at zero temperature (superfluid with condensate fraction of around 8%).
- Normal to superfluid transition at 2.17K.

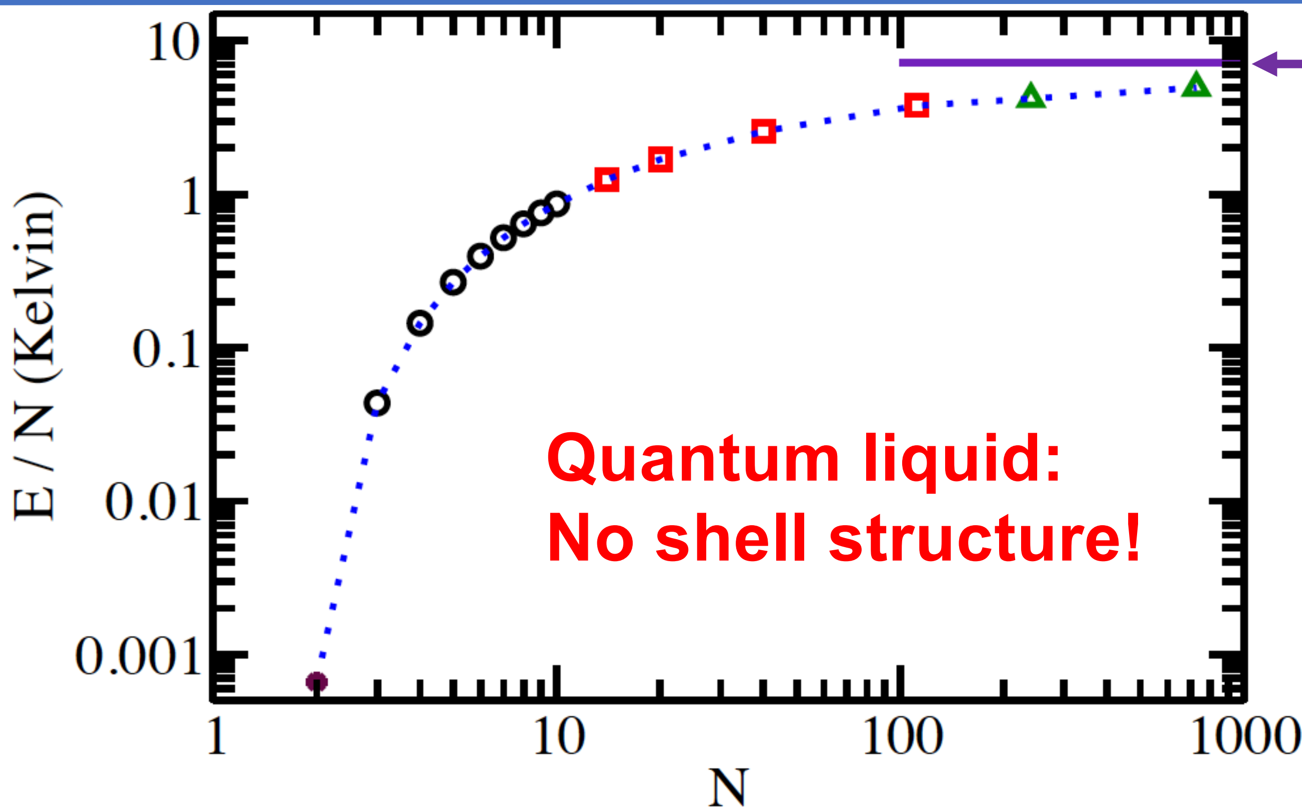
Helium named after
the sun (greek “helios”).
Discovered in 1868.

Bulk liquid helium-4:
Binding energy per
particle $E/N = -7$K
(1 K = 8.6 x 10^{-5} eV).



From Wikipedia

Helium Droplets = Quantum Liquid



Large N: E/N ~ −7K

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).

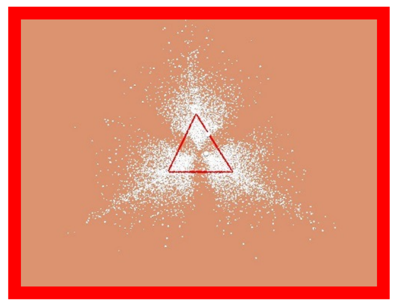
Rich interplay between many-body nuclear physics and quantum droplet community [e.g., Pandharipande et al., PRL 50, 1676 (1983); Stringari et al., JCP 87, 5021 (1987); Sindzingre et al., PRL 63, 1601 (1989)].

Helium Dimer and Trimers

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

- Dimer:
  - ^{4}He-^{4}He bound state energy $E_{dimer} = -1.625\mathrm{mK}$. 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 $r_{vdW} = 5.1a_0$).

- Trimer:
  - Two $J = 0$ bound states with $E_{trimer} = -131.8\mathrm{mK}$ and $-2.65\mathrm{mK}$.
  - No $J > 0$ bound states.



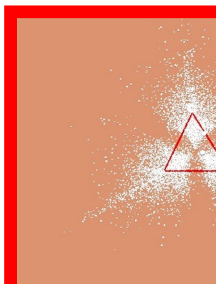
$(^4He_3)^*$

Discussed later in this talk.

Helium Dimer and Trimers

1 K = 8.6×10^{-5} eV

- 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 $r_{vdW} = 5.1a_0$).
- Trimer:
  - Two J = 0 bound states with $E_{trimer} = -13$ -2.65mK.
  - No J > 0 bound states.



Close to hard wall at small internuclear distances is a challenge for some numerical approaches: "Less soft" than "typical" nuclear potentials.

k

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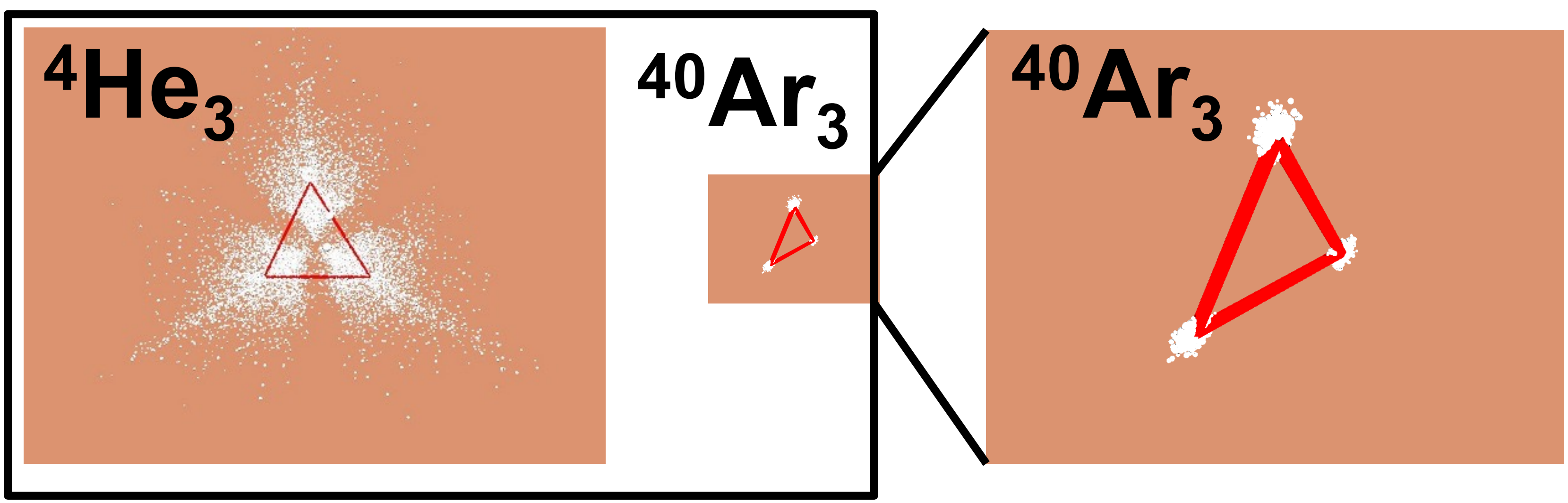
in this talk.

- Nuclear physics: Deuteron and triton.

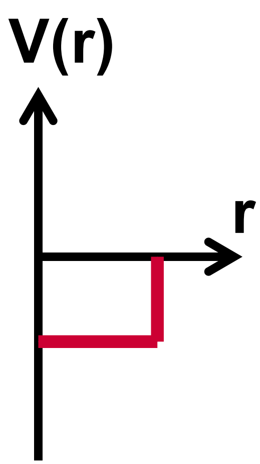
Comparison With Other Neutral Rare Gas Clusters

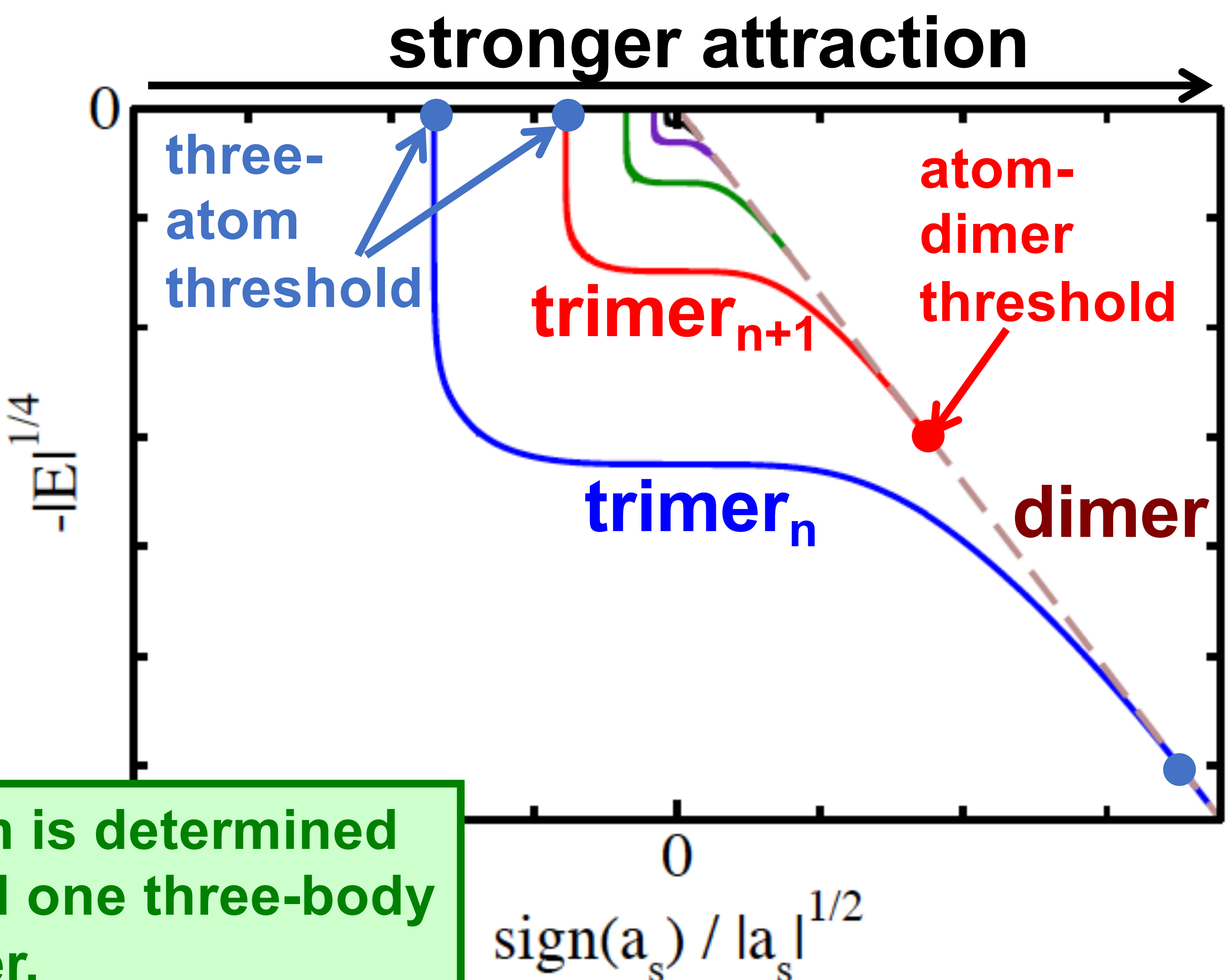
- ^{4}He, ^{10}Ne, ^{20}Ar: composite bosons (energy scales are such that these atoms can be considered as point particles; consider only nuclear degrees of freedom).

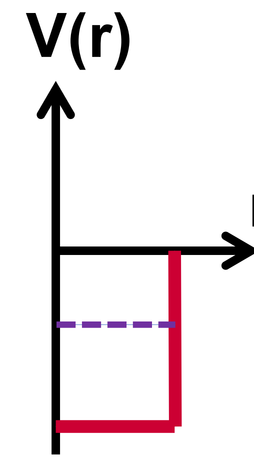
- Dimer (potential minimum at $5-10a_0$):
  - ^{4}He-^{4}He binding energy: $E_{dimer} = -1.3$mK.
  - ^{10}Ne-^{10}Ne binding energy: $E_{dimer} = -20.1$K.
  - ^{20}Ar-^{20}Ar binding energy: $E_{dimer} = -101$K.



Finite s-wave Scattering Length: Universally Linked States







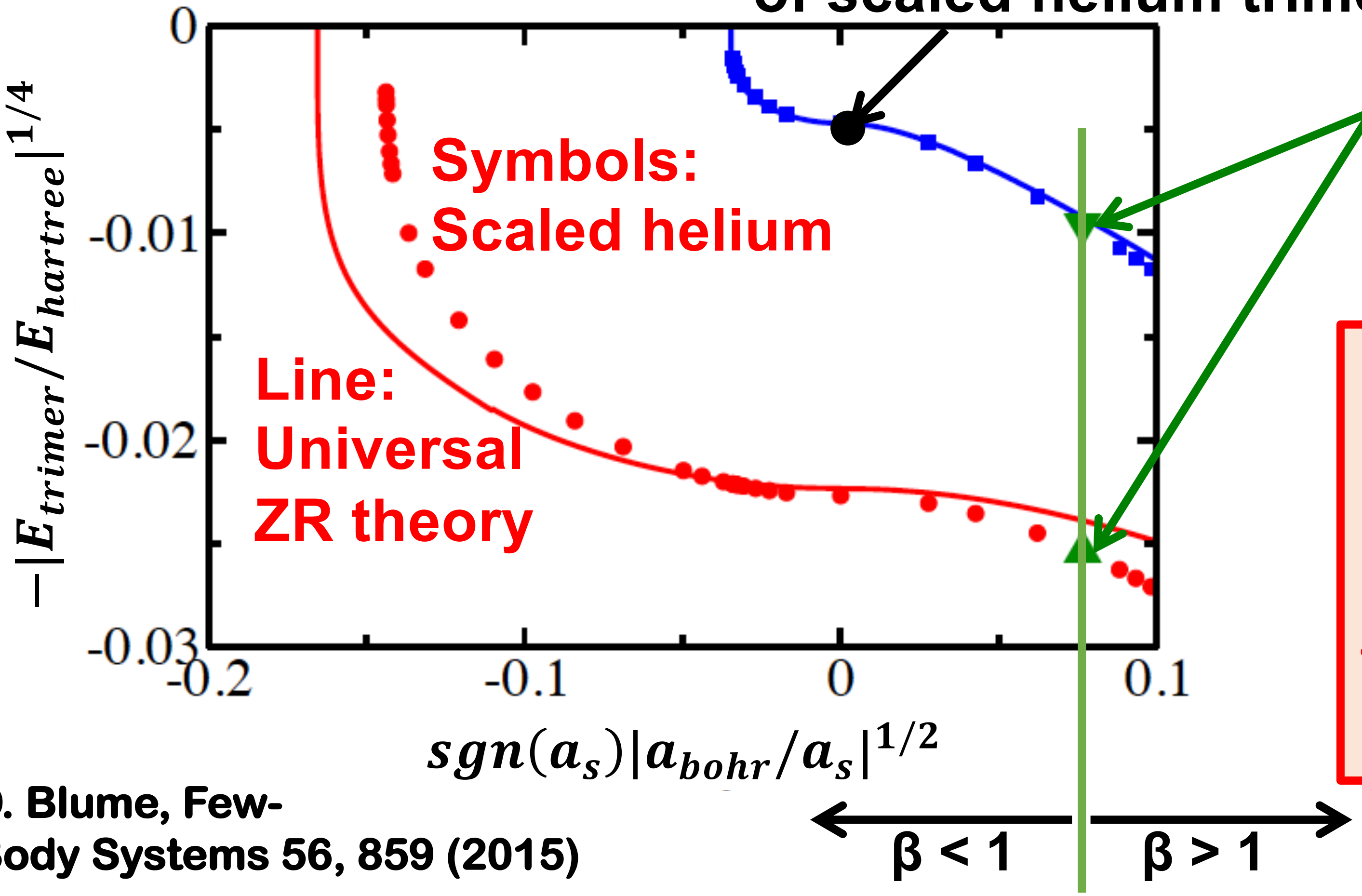
Spectrum is determined by a_s and one three-body parameter.

Review by Braaten and Hammer, Physics Reports 428, 259 (2006).

Helium Trimer Excited State is an Efimov State

$\beta V_{He\text{-}He}(r_{12}) + \beta V_{He\text{-}He}(r_{23}) + \beta V_{He\text{-}He}(r_{31})$.

Three-body parameter is chosen such that ZR energy agrees with energy of scaled helium trimer excited state.

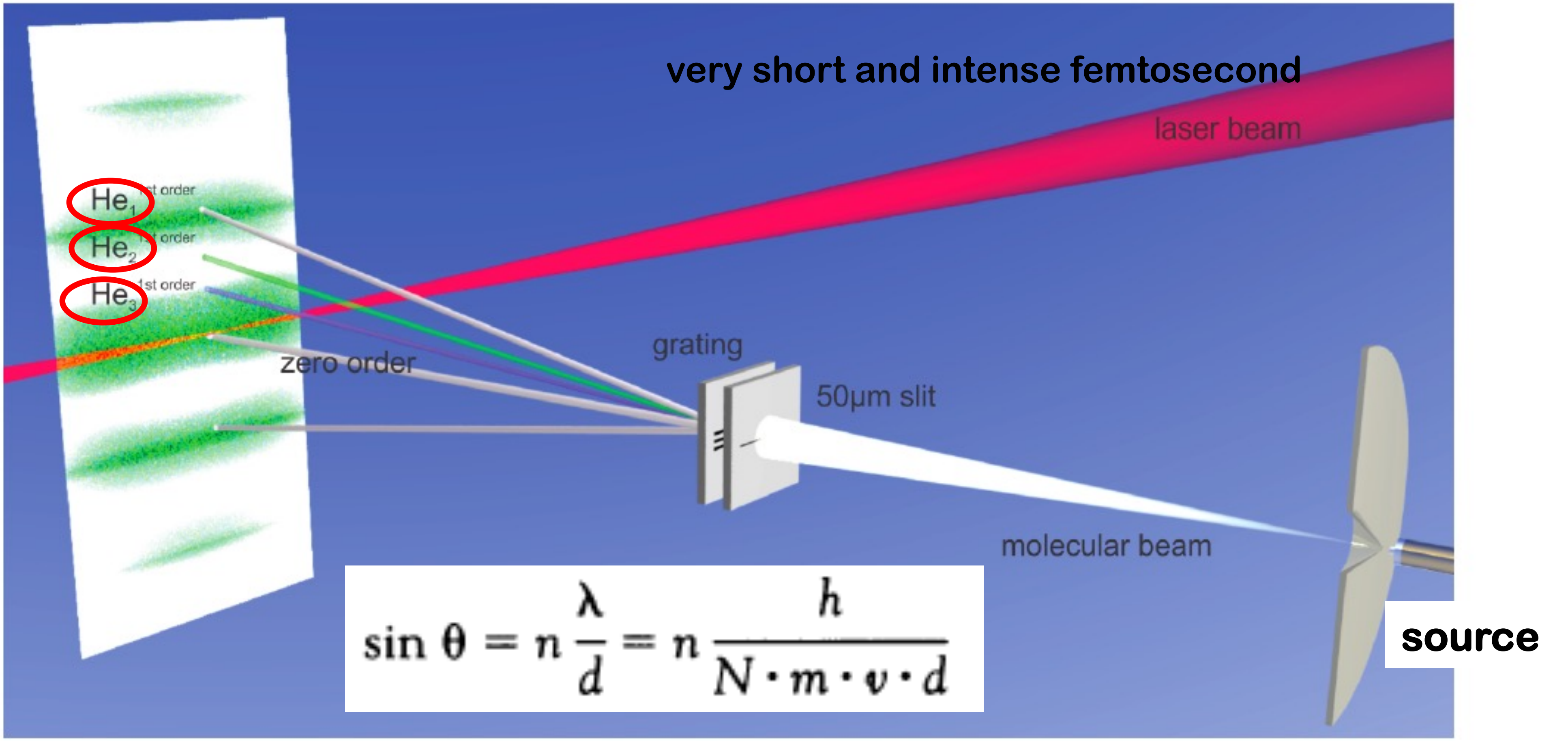


True helium (ground and excited states)

For the excited state, symbols agree with line! Molecular system follows predicted Efimov behavior.

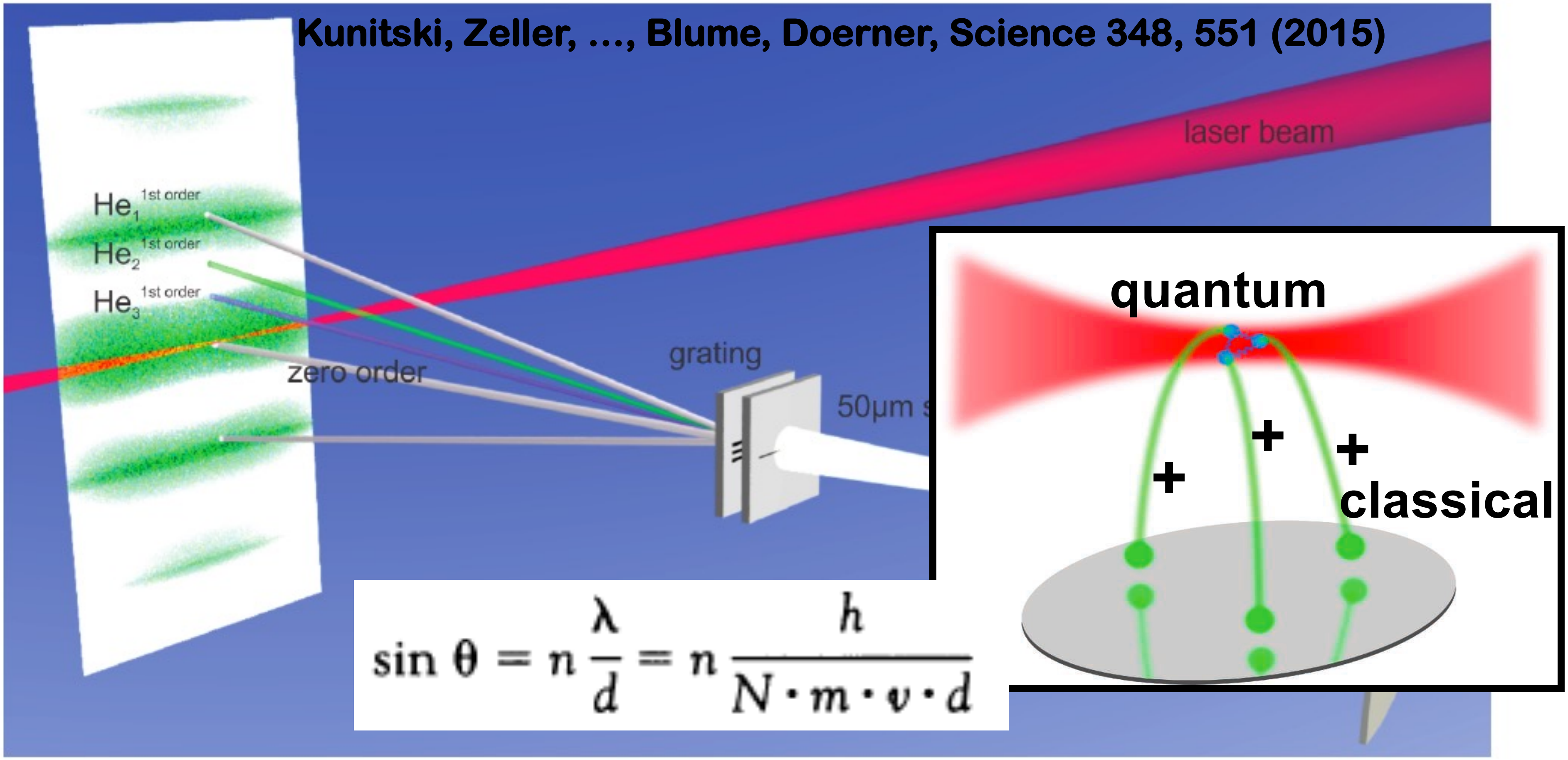
D. Blume, Few-Body Systems 56, 859 (2015)

Probing Helium Trimer Excited Efimov State



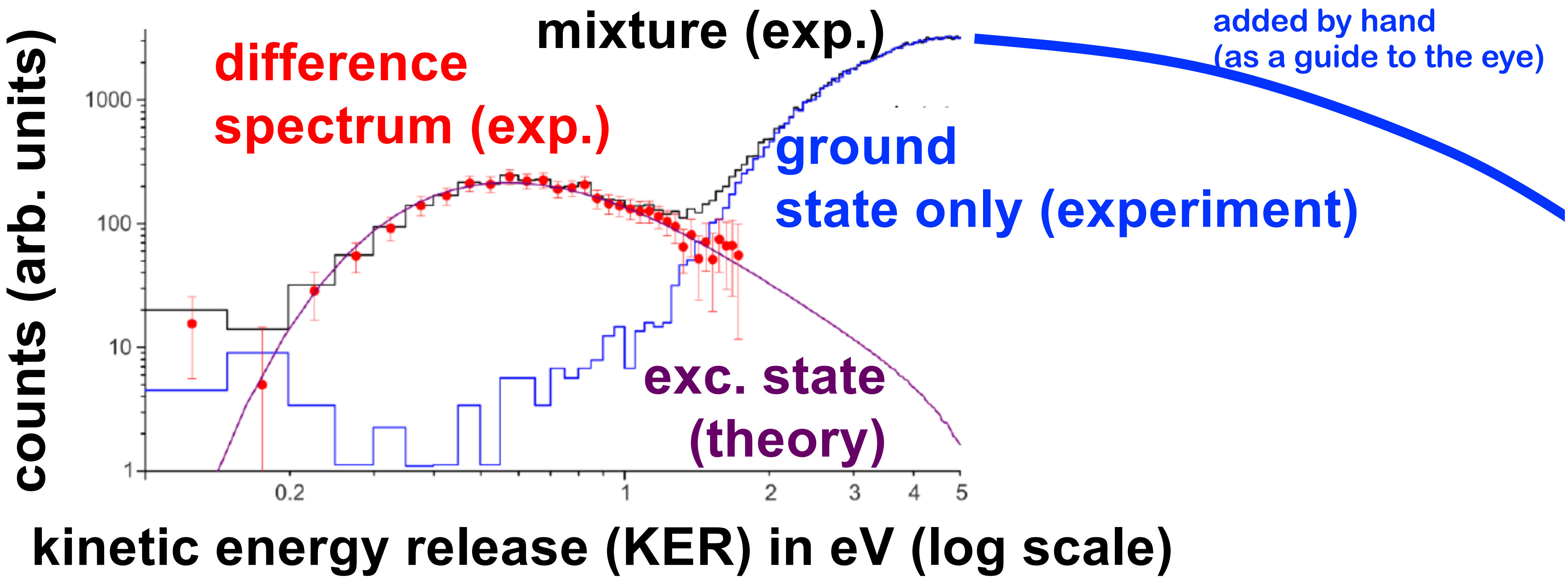
$$\sin\theta = n\frac{\lambda}{d} = n\frac{h}{N\cdot m\cdot v\cdot d}$$

Probing Helium Trimer Excited Efimov State



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

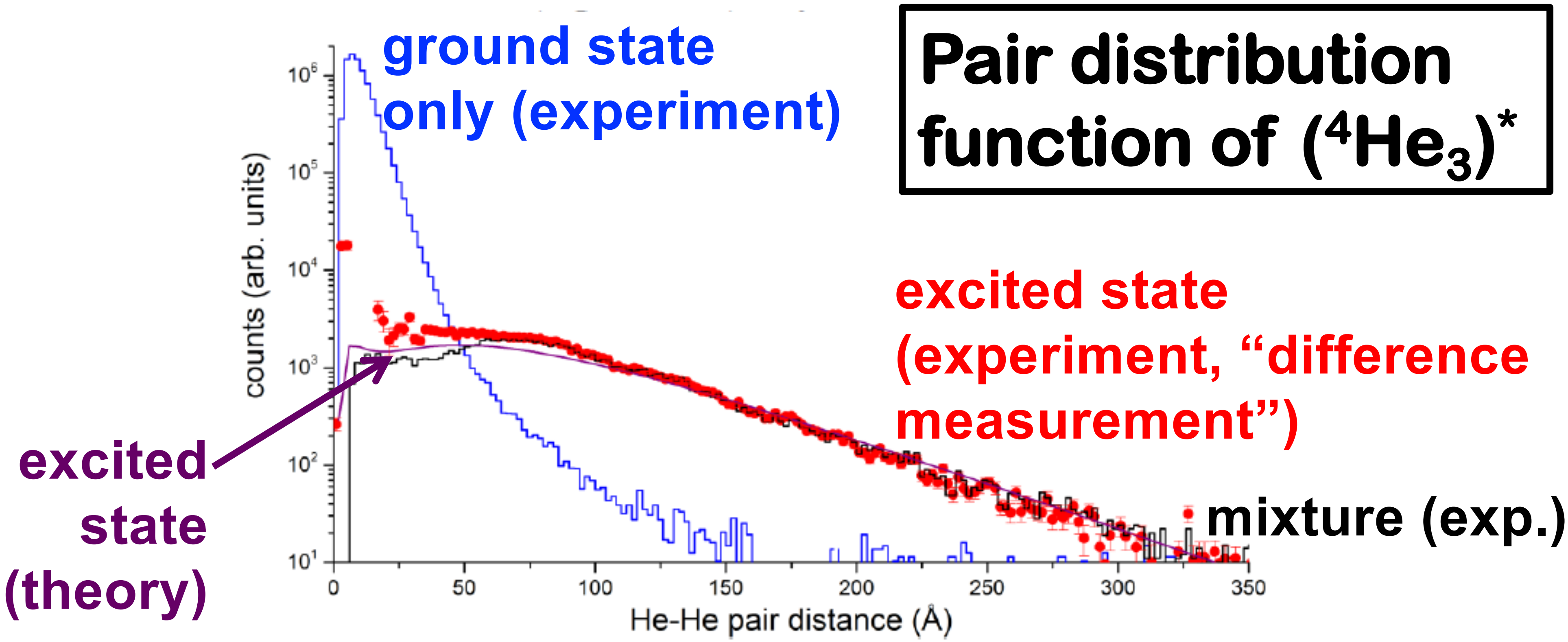
Kinetic Energy Release Measurement: Observing $(^4He_3)^*$



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

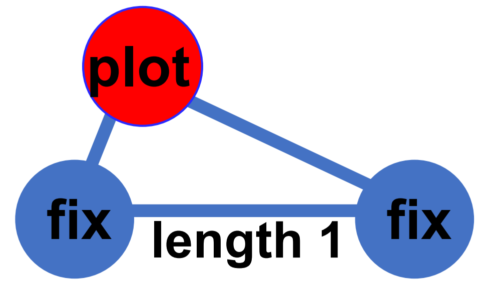
Large r_{12}, r_{23} and r_{31} correspond to small $KER=1/r_{12}+1/r_{23}+1/r_{31}$.

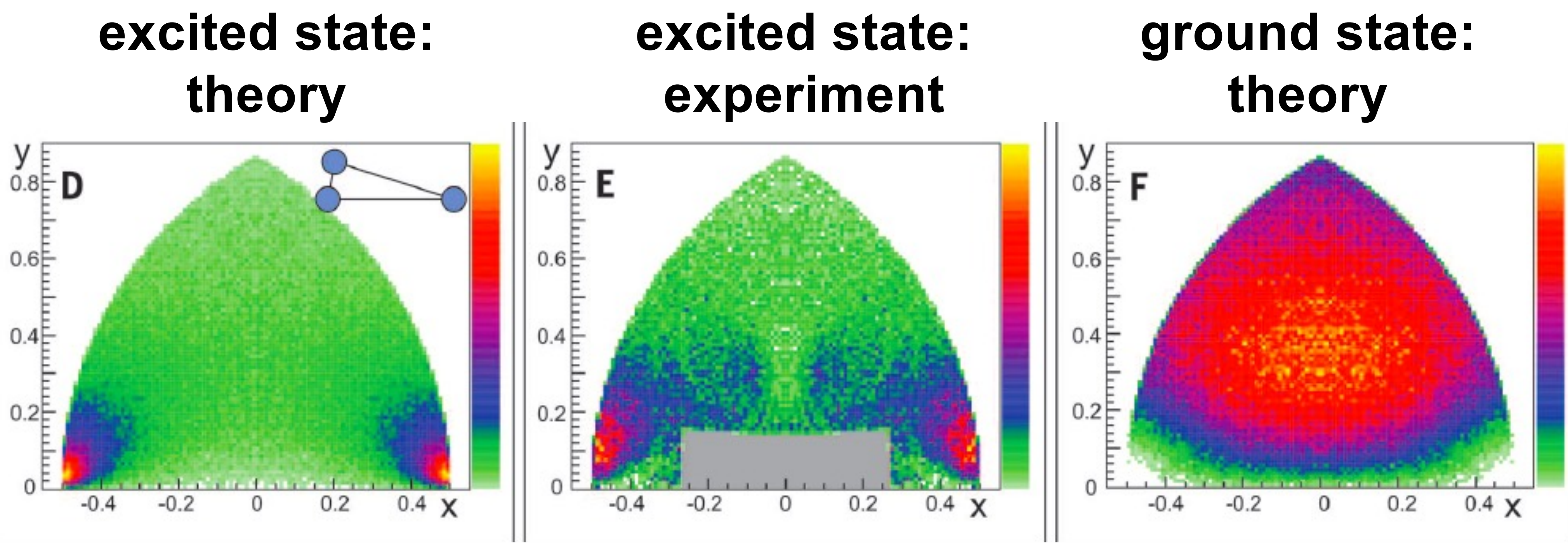
Reconstructing Real Space Properties



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)].

Normalized Structural Properties of $^4He_3$





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.

Structural Properties: Short- and Long-Range Characteristics

Atomic systems:

- Van der Waals universality.
- Highly repulsive short-range potential.

Which long-range behaviors “collapse”?
Which short-range behaviors “collapse”?

Strategy:
Consider different two-body potentials (realistic and effective models), both at the physical point and at unitarity.

Different (Helium-Helium) Interaction Potentials

Strategy: Consider different two-body potentials [realistic **(Model I)** and effective models **(Model II)**], both at the physical point and at unitarity.

Model I: Realistic potential with hard inner wall.

$$V_{tot} = \sum_{j=1}^{N-1} \sum_{k>j}^{N} V_{realistic}(r_{jk})$$

Unitarity realized by applying overall scaling factor.

HFD-HE2.
CPKMJS.
TTY.
LM2M2.

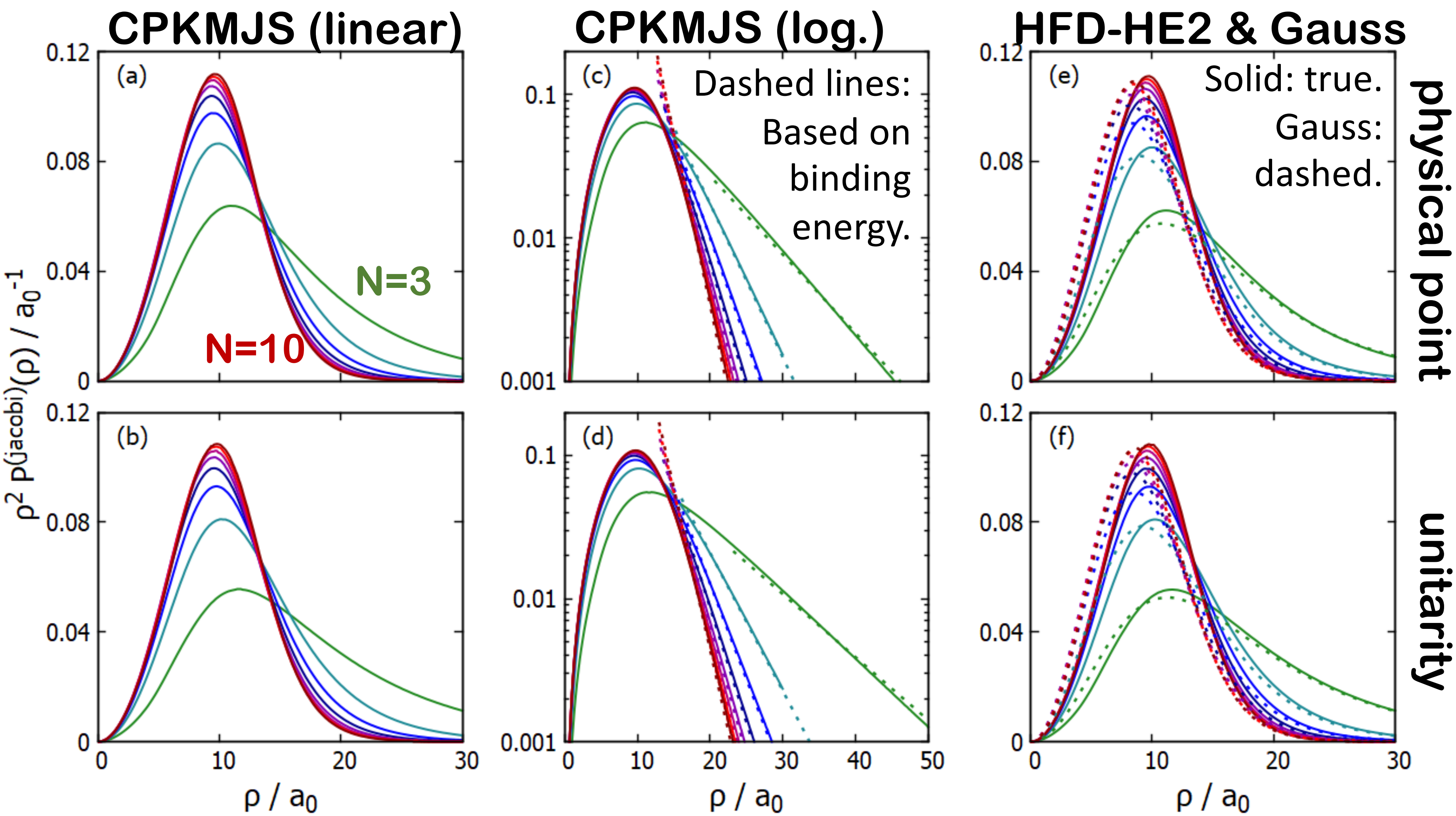
Model II: Effective low-energy potential model.

$$V_{tot} = \sum_{j=1}^{N-1} \sum_{k>j}^{N} V_{2,gauss}(r_{jk}) + \sum_{j=1}^{N-2} \sum_{k>j}^{N-1} \sum_{l>k}^{N} V_{3,gauss}(R_{jkl})$$

matched to HFD-HE2 and scaled-HFD-HE2.

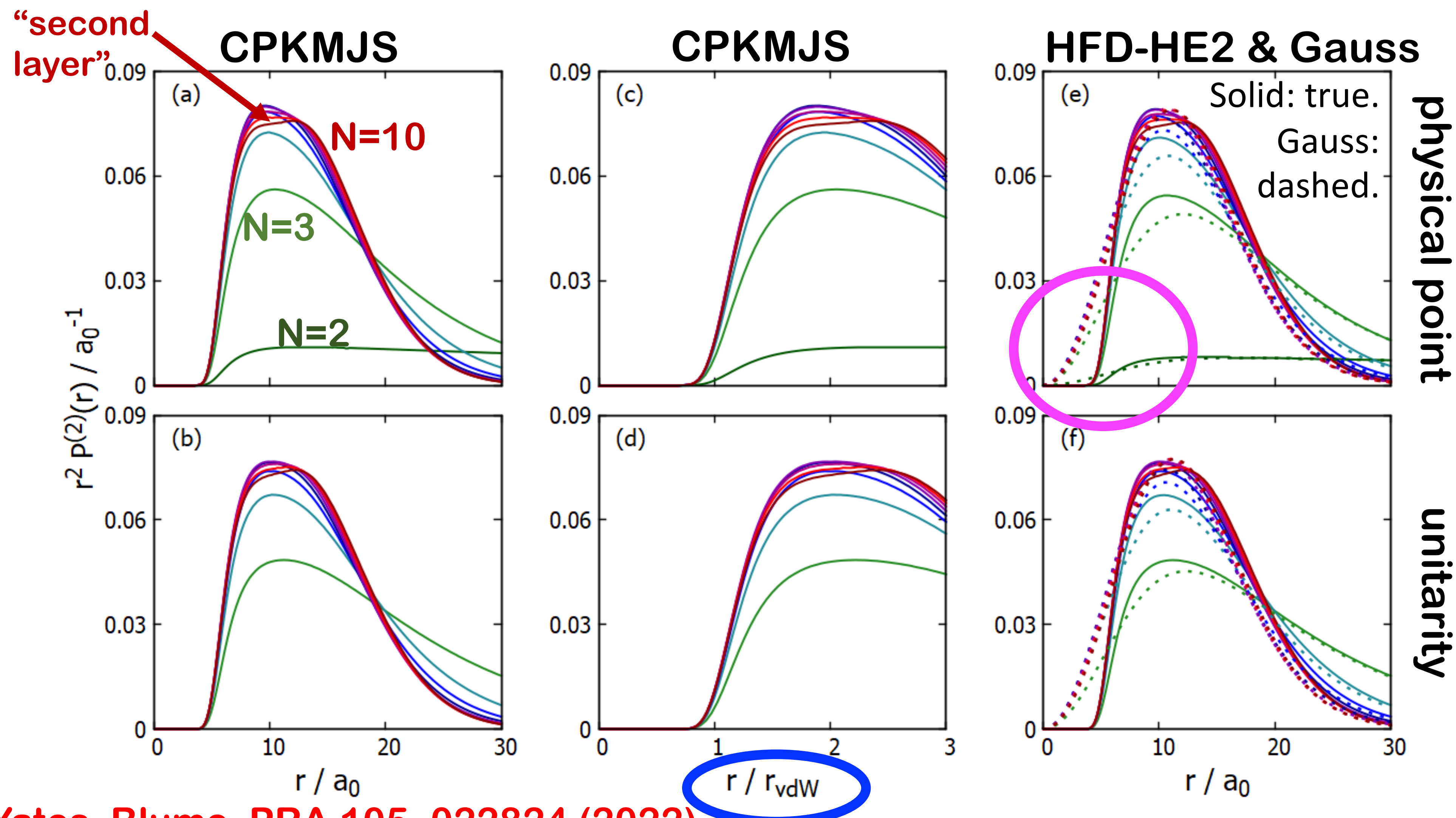
See Kievsky et al., PRA 96, 040501 (2017); PRA 102, 063320 (2020):
2-body range and depth → s-wave scattering length and effective range.
3-body range and depth → three- and four-body energy.

^4He_N: N^{th} Atom Relative to Center-of-Mass of N-1 Atoms



Results obtained using forward walking: Reynolds et al., J. Stat. Phys. 43, 1017 (1986).

Pair Distribution Function $P^{(2)}(r)$ for $N = 2 - 10$



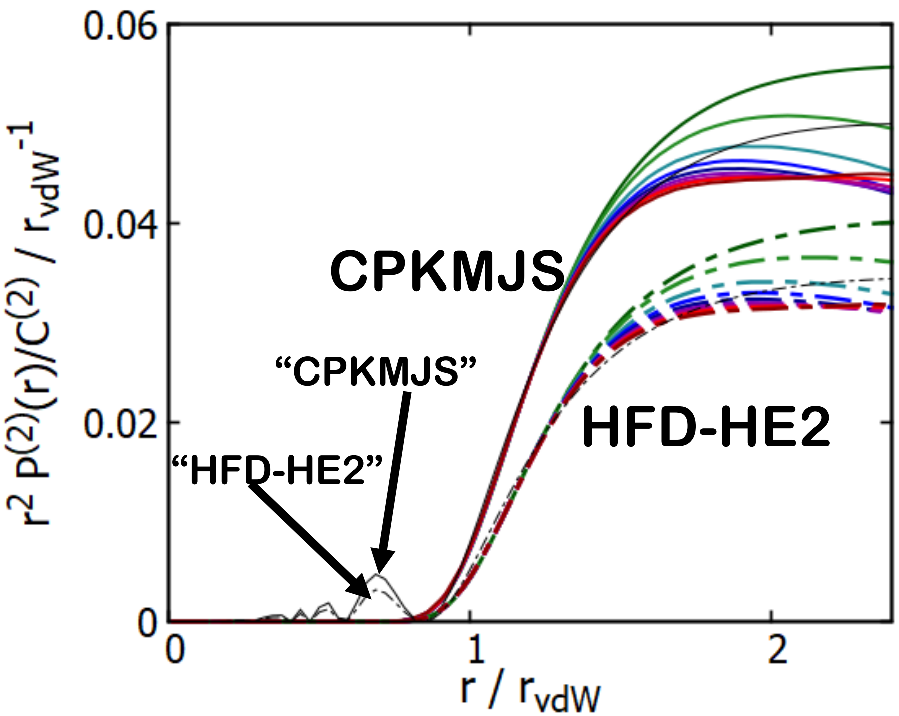
Yates, Blume, PRA 105, 022824 (2022)

$\mathrm{P}^{(2)}(\mathrm{r})$, Two-Body Contact $C_N^{(2)}$, van der Waals Universality

$$\hat{P}_N^{(2)}(r) = \frac{2}{N(N-1)} \sum_{j=1}^{N-1} \sum_{k>j}^{N} \frac{\delta(r_{j,k}-r)}{r^2}$$

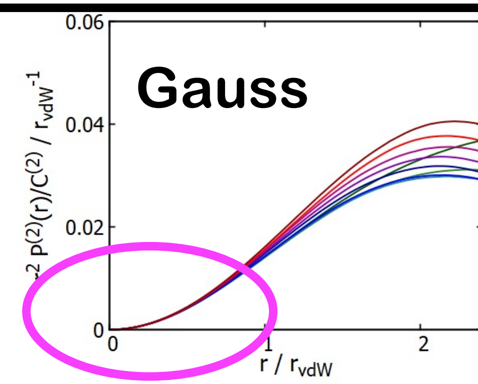
$$\int_0^\infty P_N^{(2)}(r) r^2 dr = 1$$

$$P_N^{(2)}(r) \underset{\text{small } r}{\rightarrow} C_N^{(2)} P_2^{(2)}(r)$$



$\frac{a_s}{r_{vdW}} = 46.95$ for HFD-HE2.

$\frac{a_s}{r_{vdW}} = 33.63$ for CPKMJS.



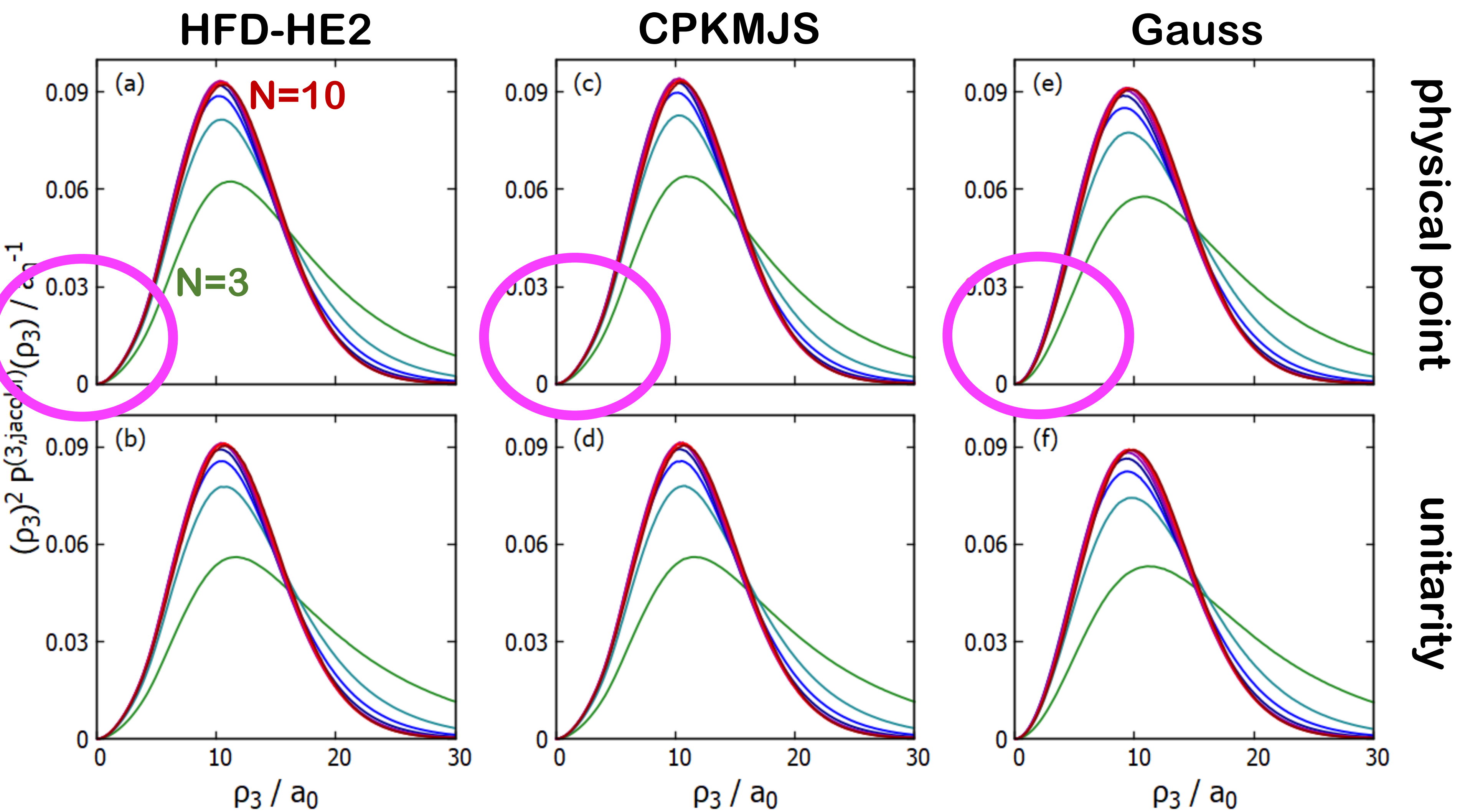
See Naidon et al., PRL 112, 105301 (2014); PRA 90, 022106 (2014) for vdW universality of trimer at unitarity.

$$\varphi_{\mathrm{vdW}}(r) = B\left[\Gamma(5/4)x^{1/2}J_{1/4}(2x^{-2}) - \frac{r_{\mathrm{vdW}}}{a_s}\Gamma(3/4)x^{1/2}J_{-1/4}(2x^{-2})\right]$$

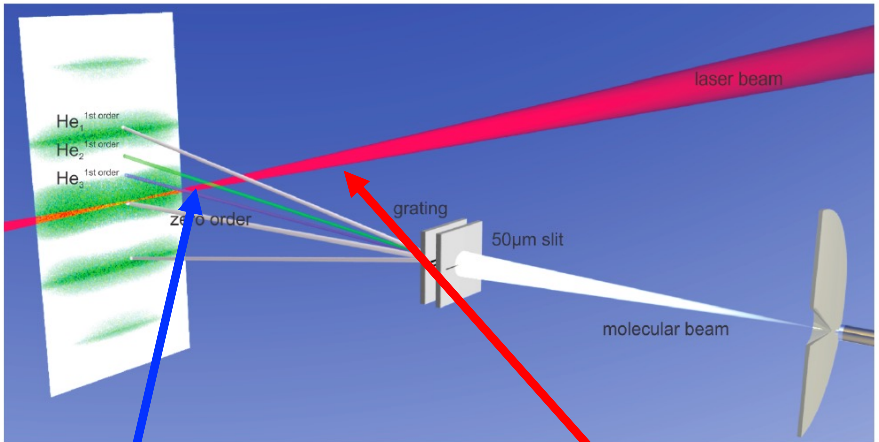
Flambaum et al., PRA 59, 1998 (1999).
Gao, PRA 58, 4222 (1998).

See Kim et al., Annu. Rev. Nucl. Sci. 24, 96 (1974): "asymptotic normalization constant"

Three-Body (Sub-Cluster) Correlations for $N = 3 - 10$



Next: Dynamics of Dimer

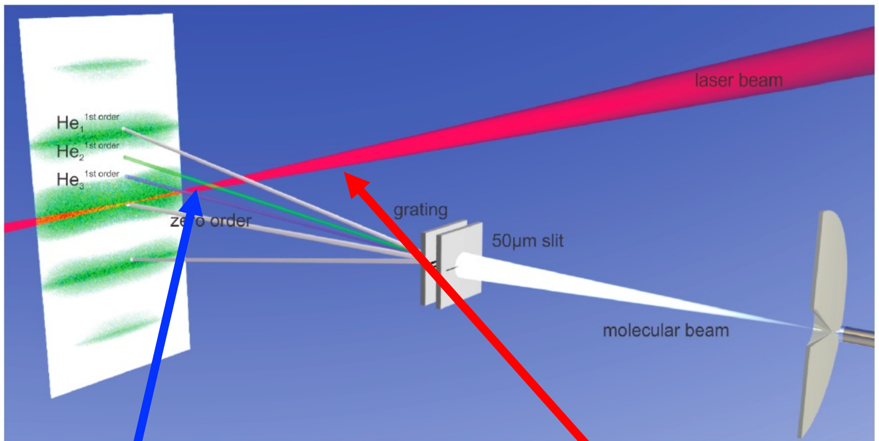


Prepare universal initial state (i.e., state that is dominated by s-wave scattering length).

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.

Next: Dynamics of Dimer



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.

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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, G
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 Ult 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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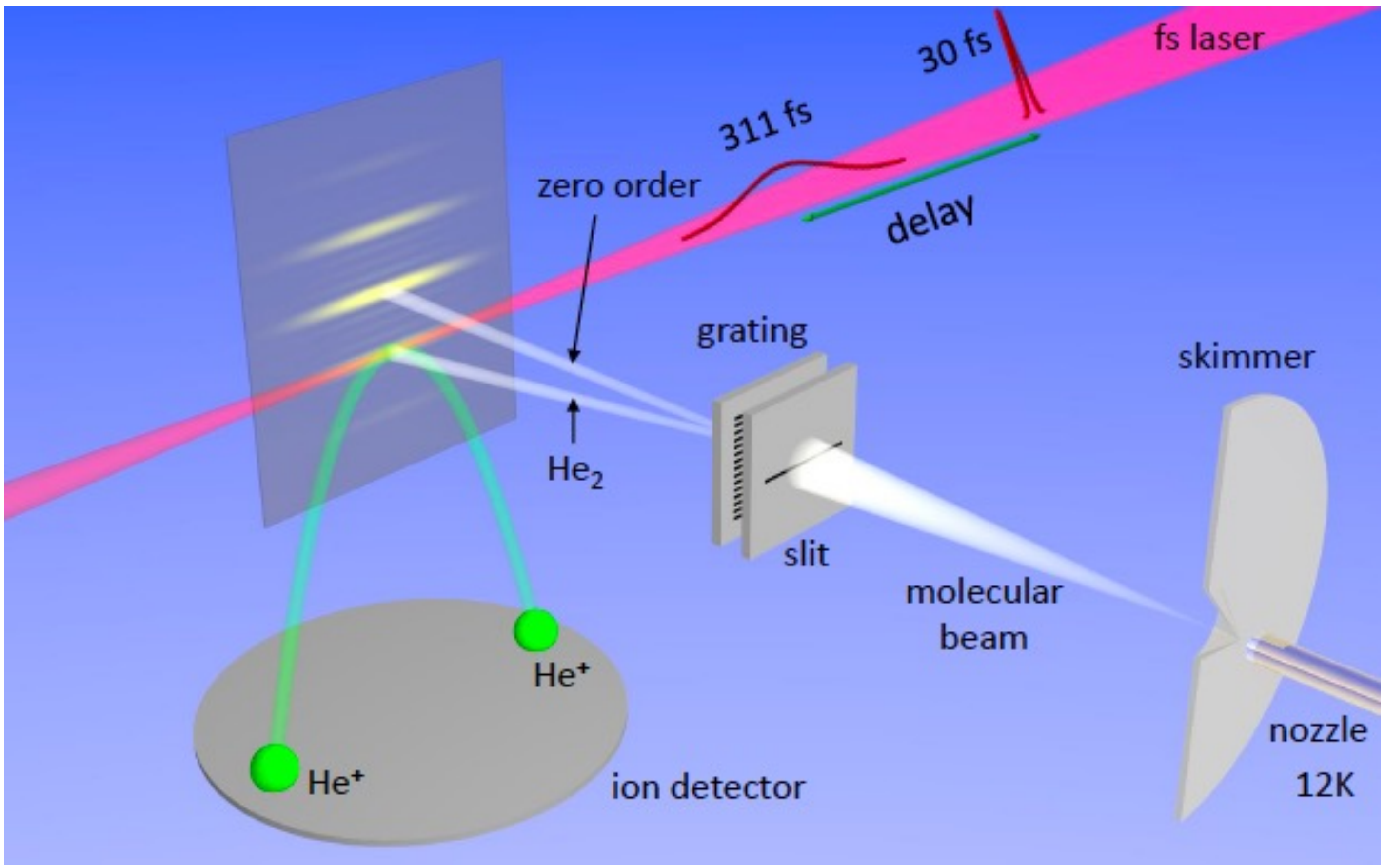
(Dated: June 5 2013)

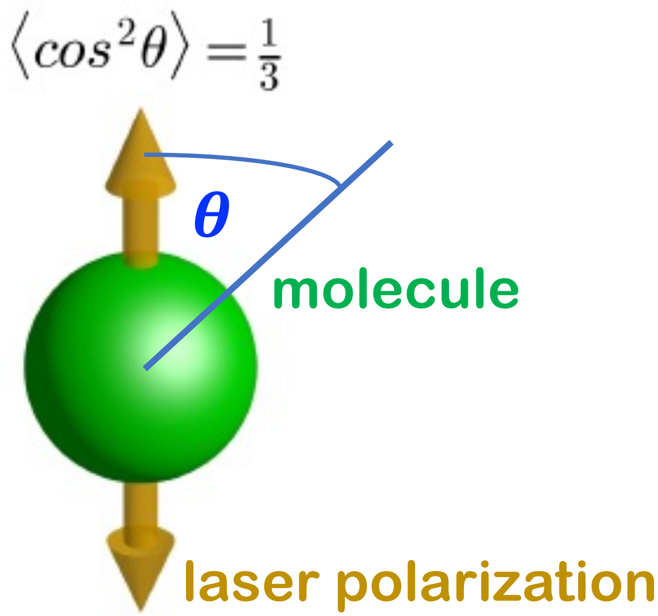
The neutron-rich ^{6}He and ^{8}He isotopes exhibit an exotic nuclear structure that consists of a tightly bound ^{4}He-like core with additional neutrons orbiting at a relatively large distance, forming a halo. Recent experimental efforts have succeeded in laser trapping and cooling these short-lived,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

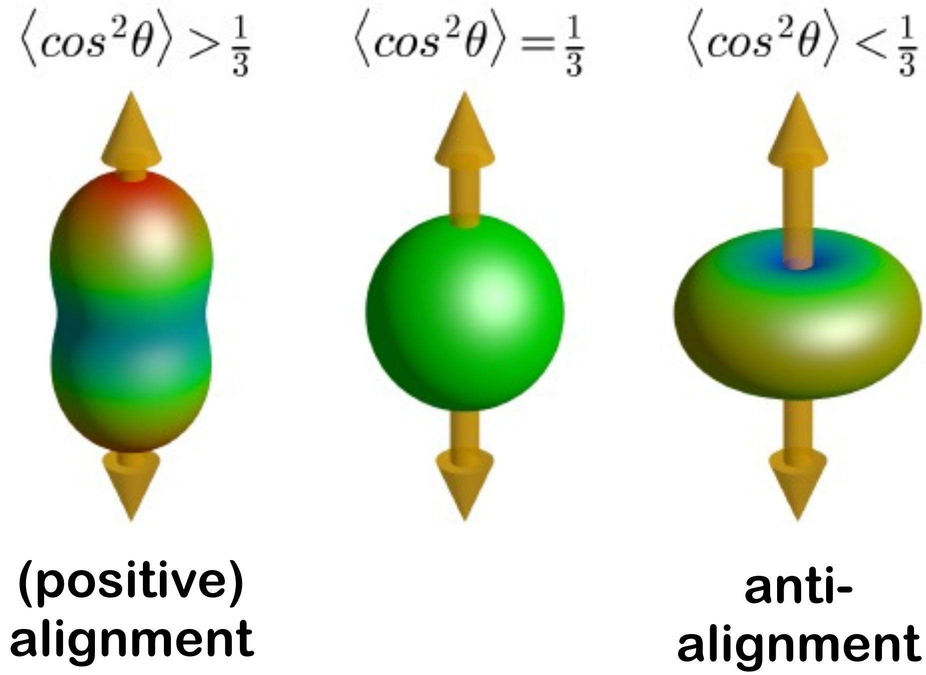
Pump-Probe Spectroscopy of Isolated Helium Dimers



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 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.

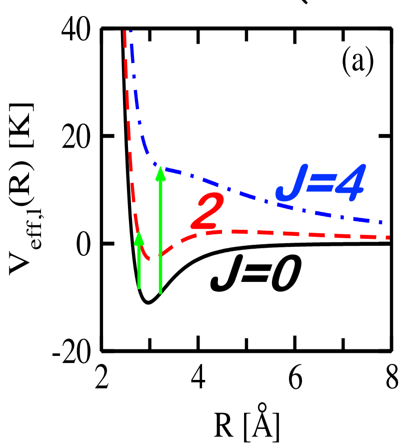
Will show: Helium dimer can be aligned. However, since the $J > 0$ partial wave components are not bound, they will “run away” (dissociative wave packet). Heavier non-universal dimers behave very differently.

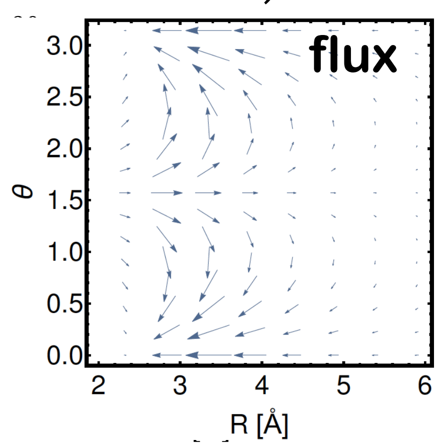
^{4}He-^{4}He In Time-Dependent Electric Field

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

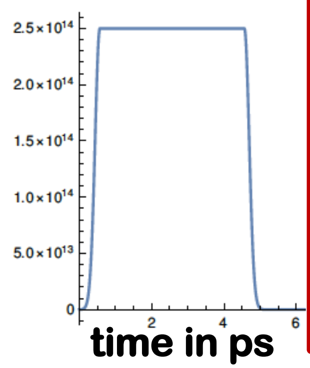
Scenario 1 (non-adiabatic laser kick):

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





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



Solve time-dependent Schroedinger equation using spherical coordinates:

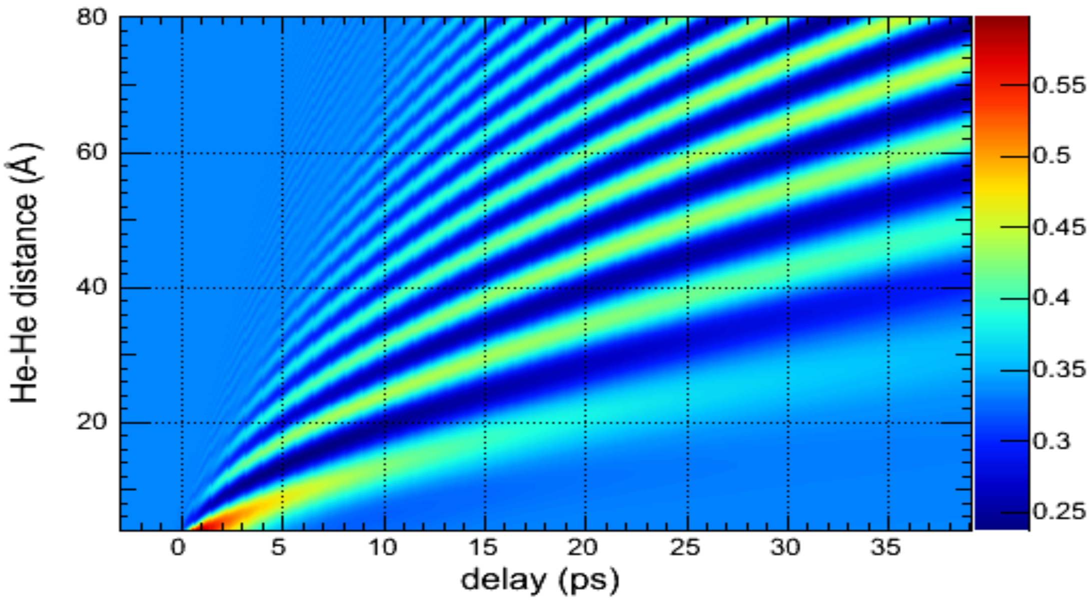
$$\Psi(R,\theta,t) = \sum_{J=0,2,\ldots} \frac{u_J(R,t)}{R} Y_{J0}(\hat{R})$$

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.

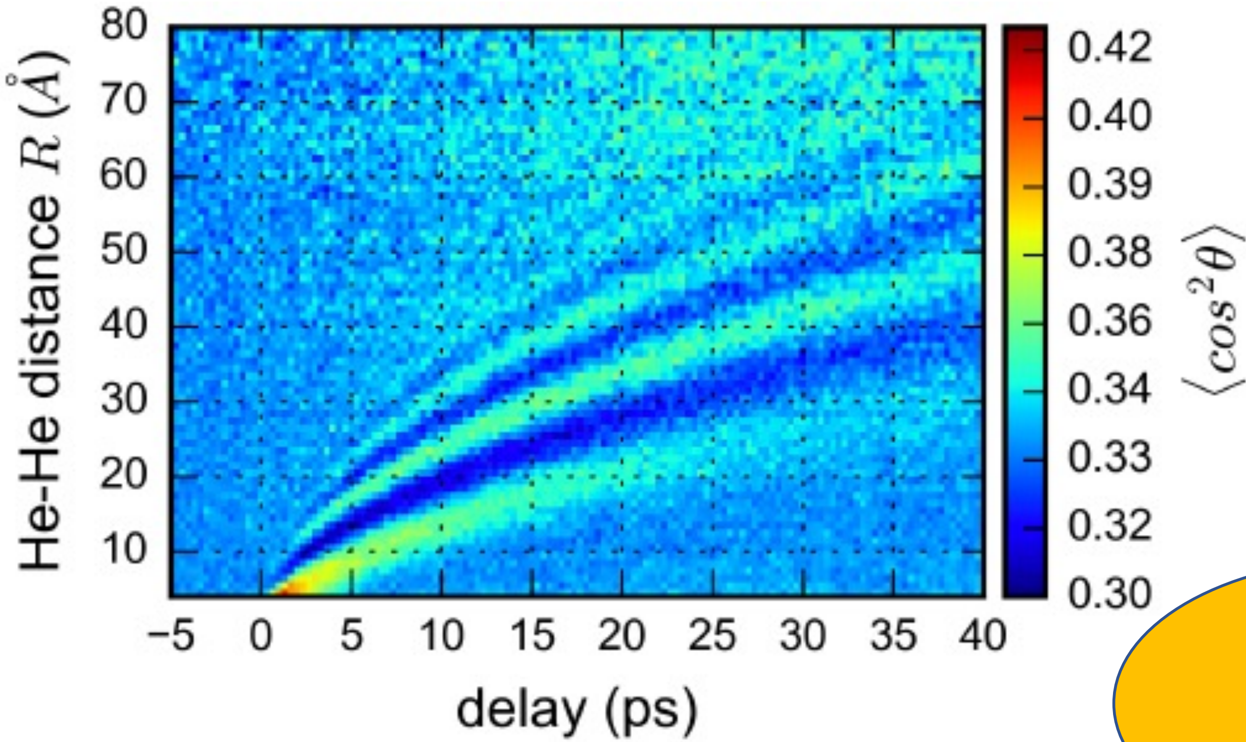
Solve time-dependent Schroedinger equation using spherical coordinates:

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

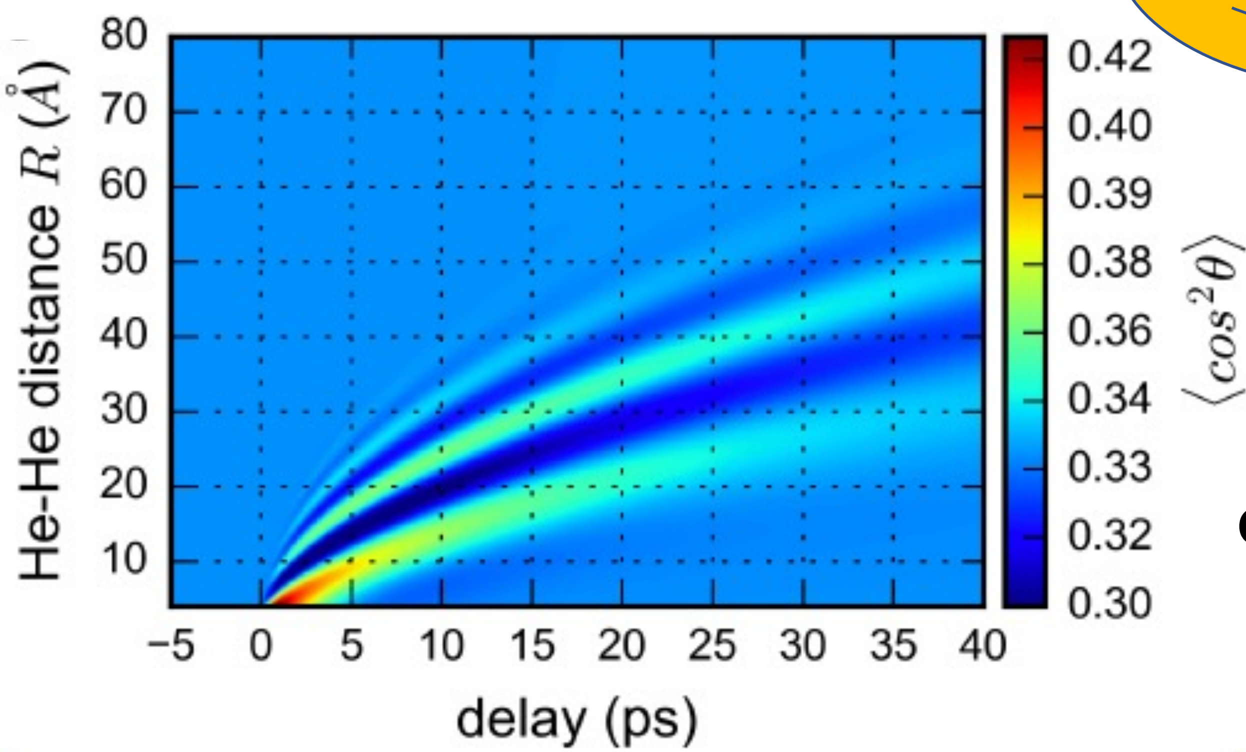
Pulse couples different partial waves.

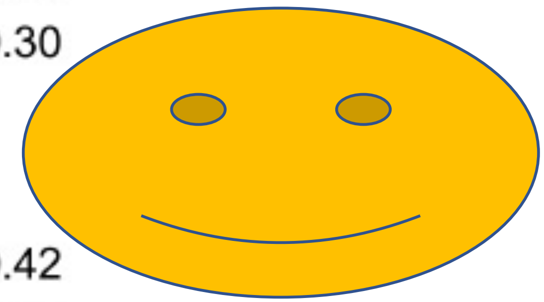
When pulse is off, the channels are decoupled.

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:

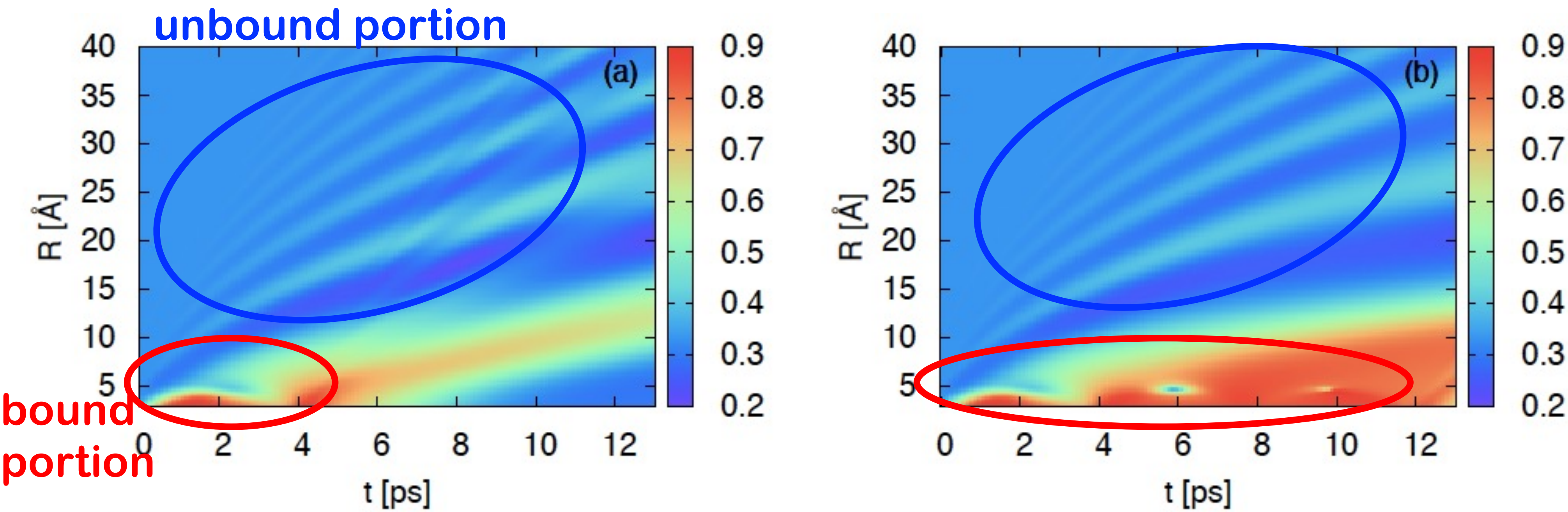
Resonances as in ultracold atoms? Need longer pulses…
Time-dependent modulation of interaction strength?
Dynamics of (Efimov) trimers? Need to populate it first…
Larger clusters.

Scenario 2: Longer Pulses

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

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 electric field-induced deeply-bound state and weakly-bound state.

Summary

- ^4He_N droplets can be realized experimentally in size-selective manner.
- Access to structural properties (accessing real-space structures beyond N=3 is non-trivial due to reconstruction algorithm).
- $^4He_3$: Ground state has Efimov characteristics and excited state is Efimov trimer (s-wave scattering length and three-body parameter).
- Some aspects of ^4He_N clusters well described by effective low-energy model.
- $^4He_2$: Alignment of dissociate wave packet portion.

Thank You!