Spectroscopy of (Helium)$_N$-Molecule Clusters: Tracing the Onset of Superfluidity

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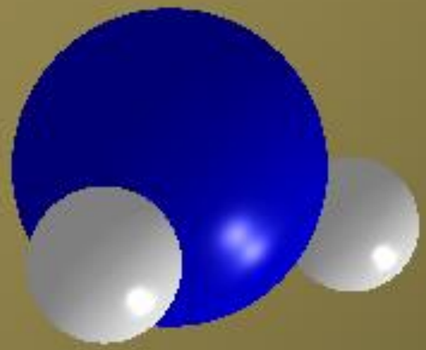
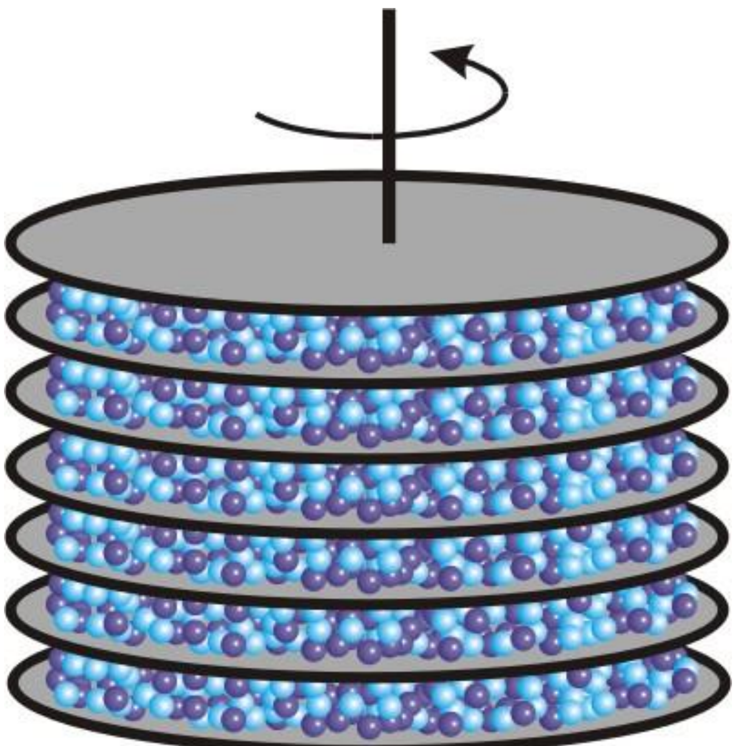
Collaborations:
Bob McKellar, Jiang Tang, NRC (mid-IR)
PN Roy, Nick Blinov, UofA (theory)

From the Molecular Regime to the Bulk Phase



A Case in Point: Superfluidity

- ^{4}He becomes superfluid below the λ-point (2.17 K)
- Frictionless flow, irrotationality, quantized vorticity, fountain effect …

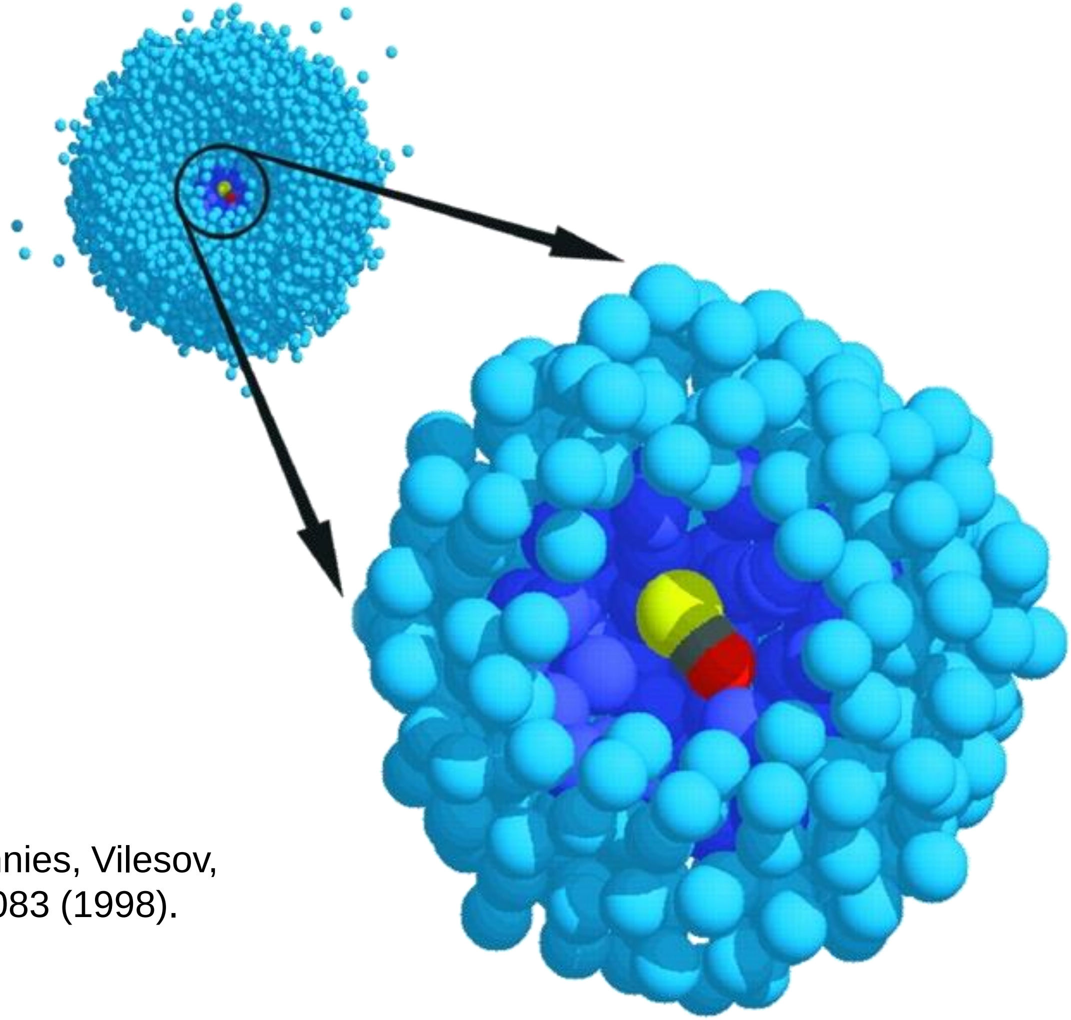


Andronikashvili experiment

‘Drag’ from normal fluid component causes increase of moment-of-inertia of disk stack.

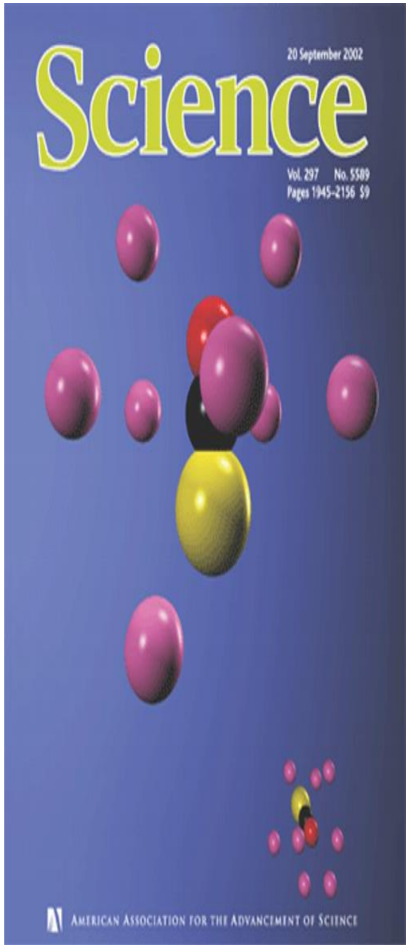
Confirmation of two fluid model.

The Microscopic Andronikashvili Experiment



Grebenev, Toennies, Vilesov,
Science **279**, 2083 (1998).

The Small Cluster Approach



Rotational (microwave) spectra of He_N-molecule clusters.

Clusters are produced in pulsed molecular expansion.

Instrument: FT Microwave Spectrometer, 4 - 26 GHz (~0.1 - 1 cm^{-1}).

He_N – Molecule Clusters

1. He_N-OCS (N=1-8)

Multidimensional Assignment Procedure

a) infrared predictions

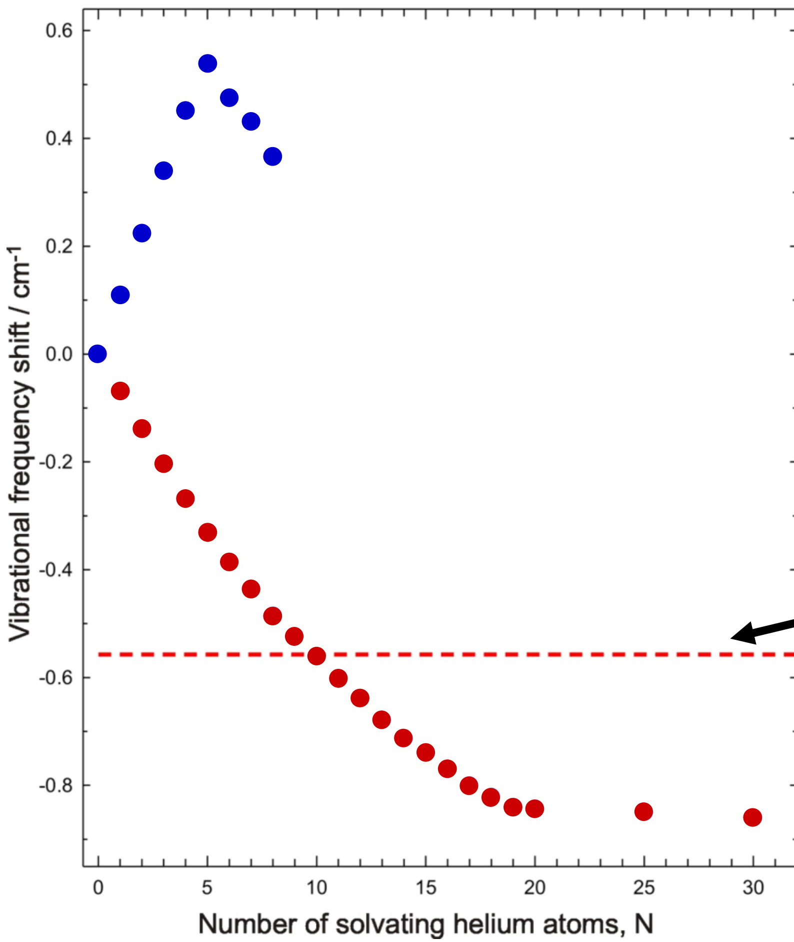
b) sample conditions (pressure, temperature)

c) double resonance experiments

d) consistency of isotopic data

e) spectral fits

Vibrational Frequency Shifts of He_N-OCS Clusters



experimental values,
Tang, Xu, McKellar, Jäger,
Science **297**, 5030 (2002).

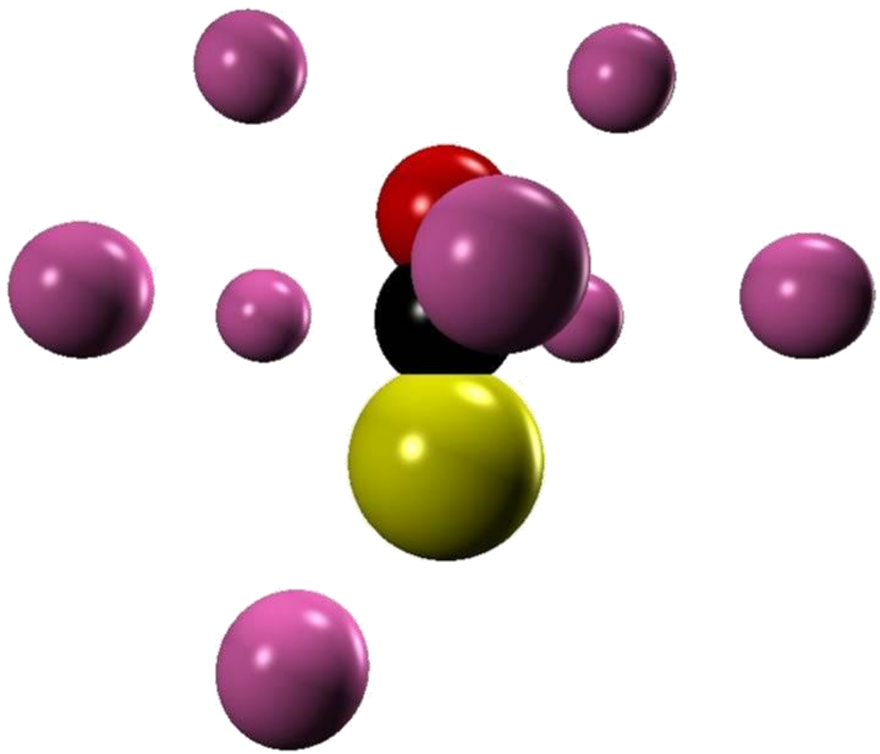
Helium droplet value

values from Whaley and co-workers,
JCP **115**, 10225 (2001).

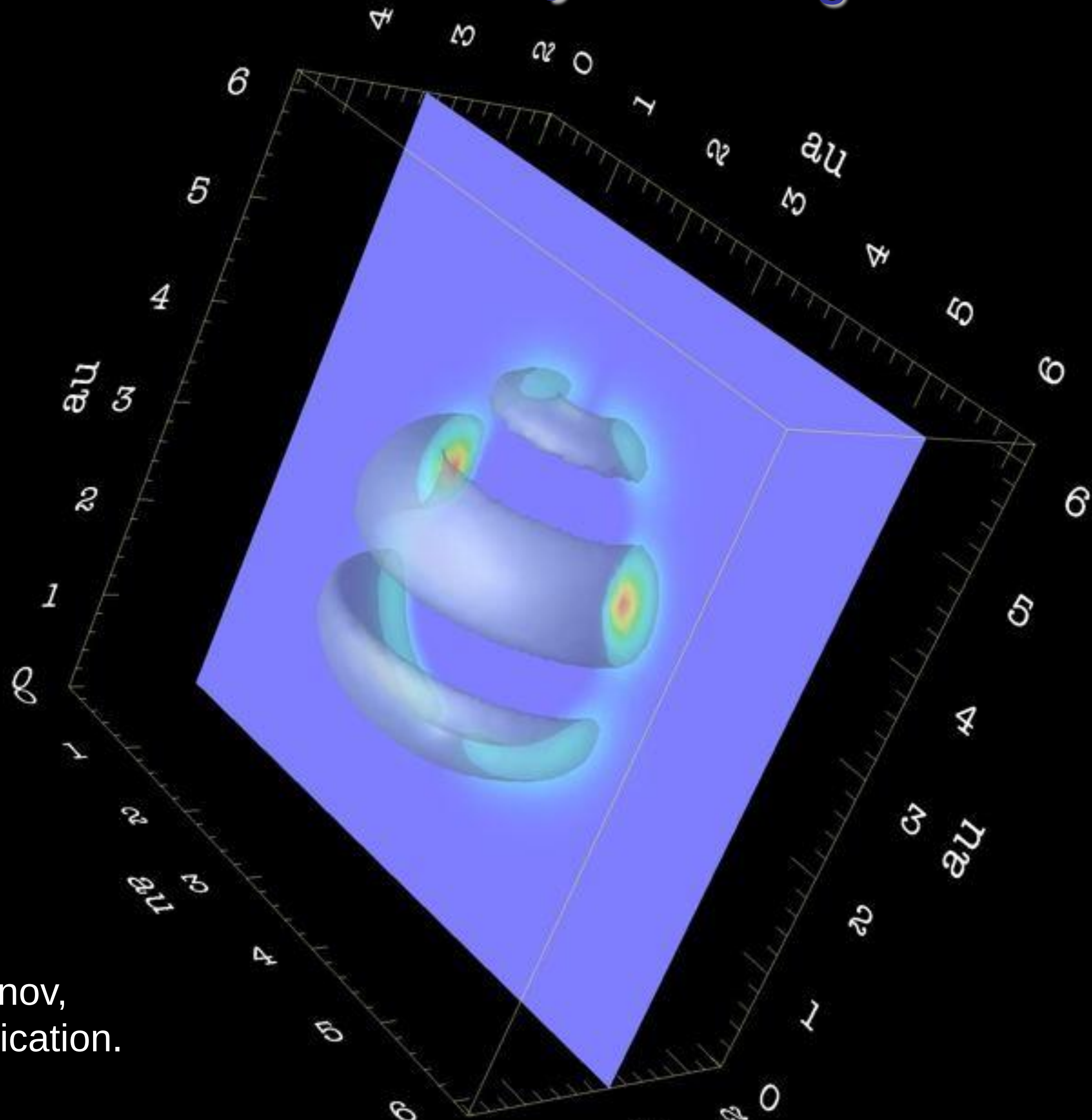
Spectroscopic Constants of He_N-OCS Clusters

Molecule	B		D
Free OCS		6081.59	1.31×10^{-5}
He-OCS	13208.57 5504.18 3661.42	4582.80	0.950
He_2-OCS	5803.39 4546.34 3019.28	3782.81	---
He_3-OCS		3104.57	5.11
He_4-OCS		2591.95	0.881
He_5-OCS		2225.15	0.234
He_6-OCS		1910.49	2.60
He_7-OCS		1682.98	1.29
He_8-OCS		1447.73	2.00
OCS in ^{4}He droplet (N~3,000)		2194.5(90)	11.4(3)

Proposed Structure of He_8-OCS



Helium density in He_8-OCS

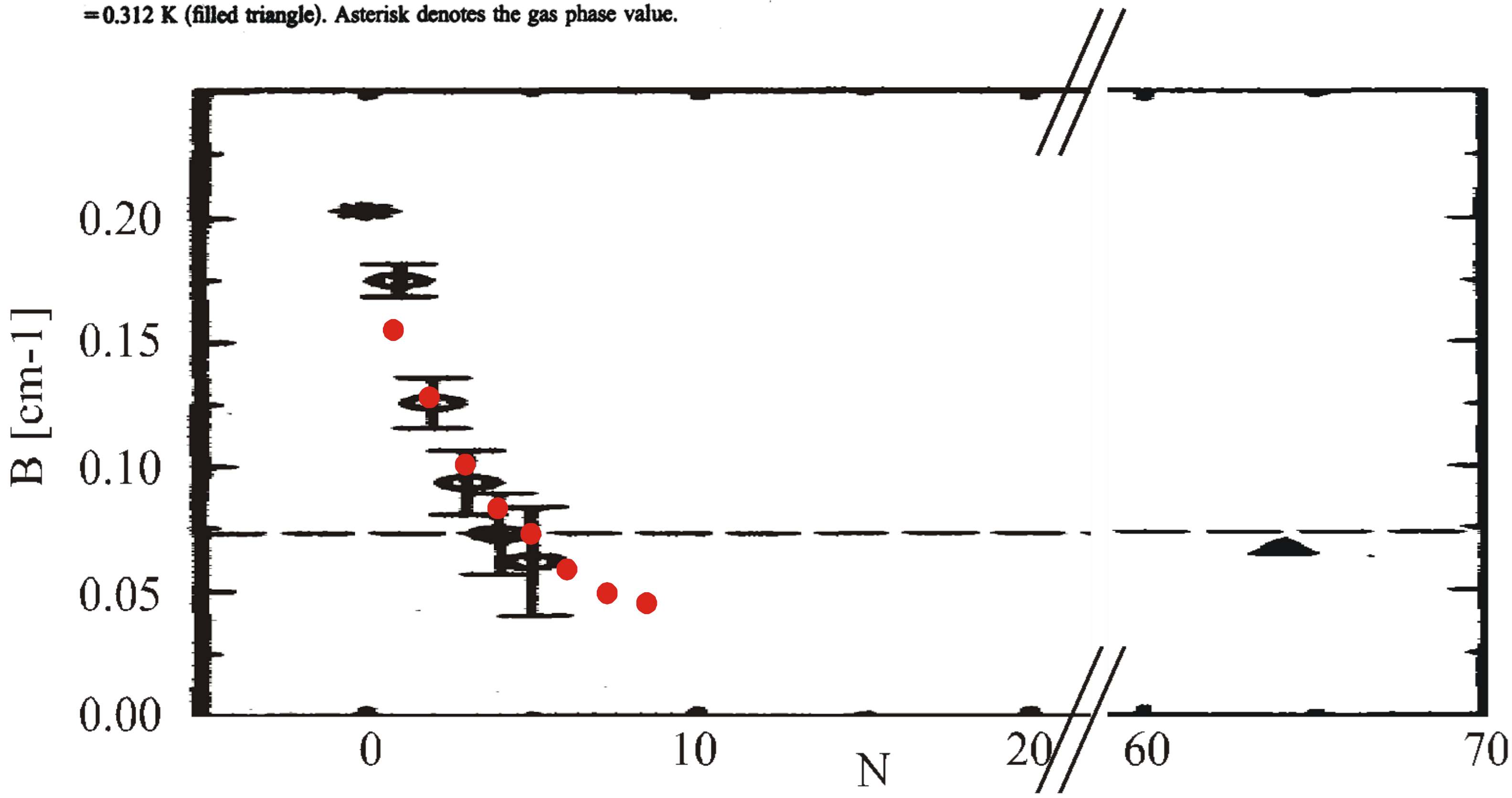


P. N. Roy, N. Blinov,
private communication.

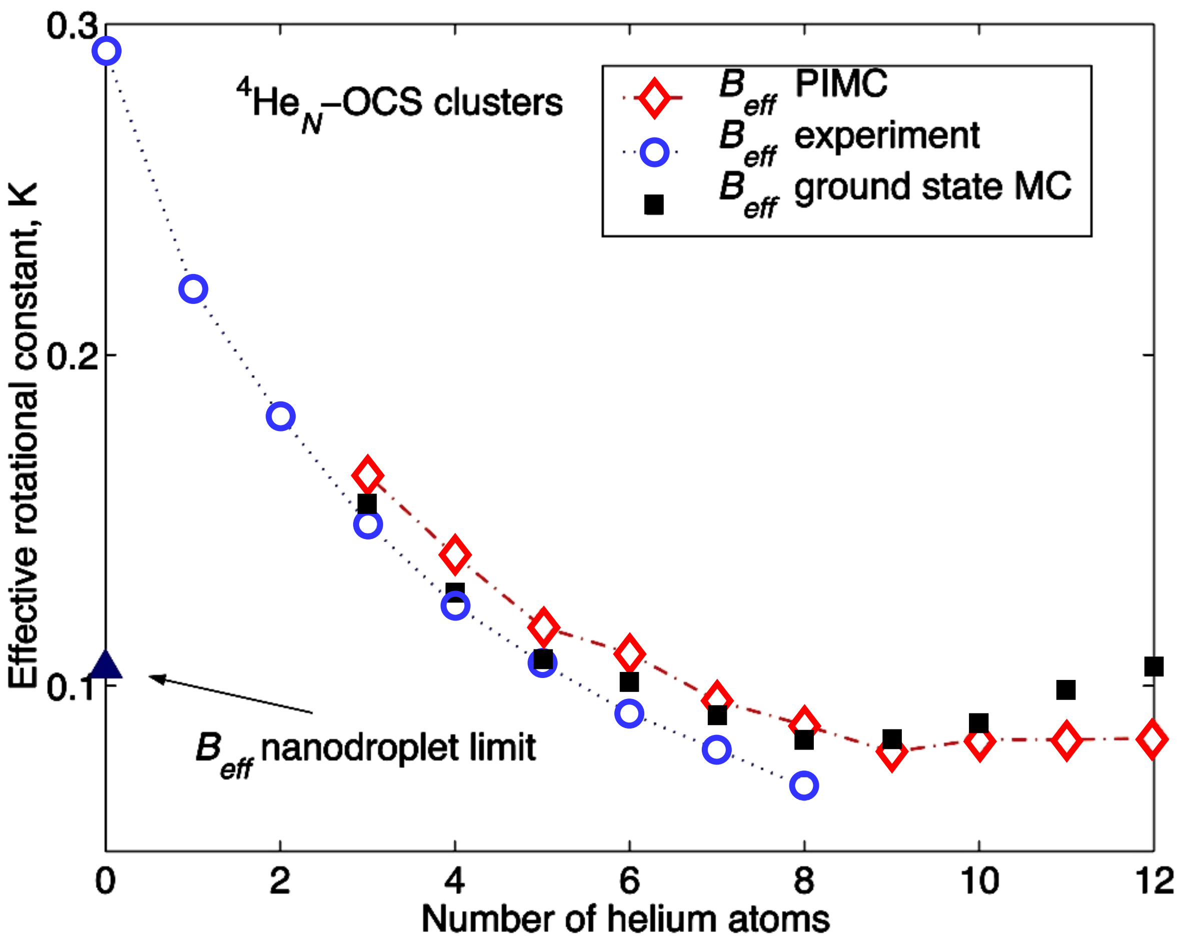
Rotational Constant vs.Number of He Atoms

Whaley and coworkers, J. Chem. Phys. 113, 6469 (2000).

FIG. 16. Rotational constant B for OCS in ^4He_N as a function of N, derived from quasi-adiabatic DMC calculations (Ref. 113) (circles) and from the two-fluid continuum model based on PIMC densities at temperature $T = 0.312$ K (filled triangle). Asterisk denotes the gas phase value.



… and very recent Calculations.



N. Blinov, X. Song, P. N. Roy, *JCP* **120**, 5916 (2004).
S. Moroni *et al.*, *Phys. Rev. Lett.* **90**, 143401 (2003).

Helium Density Profiles in He_N-OCS

N=5

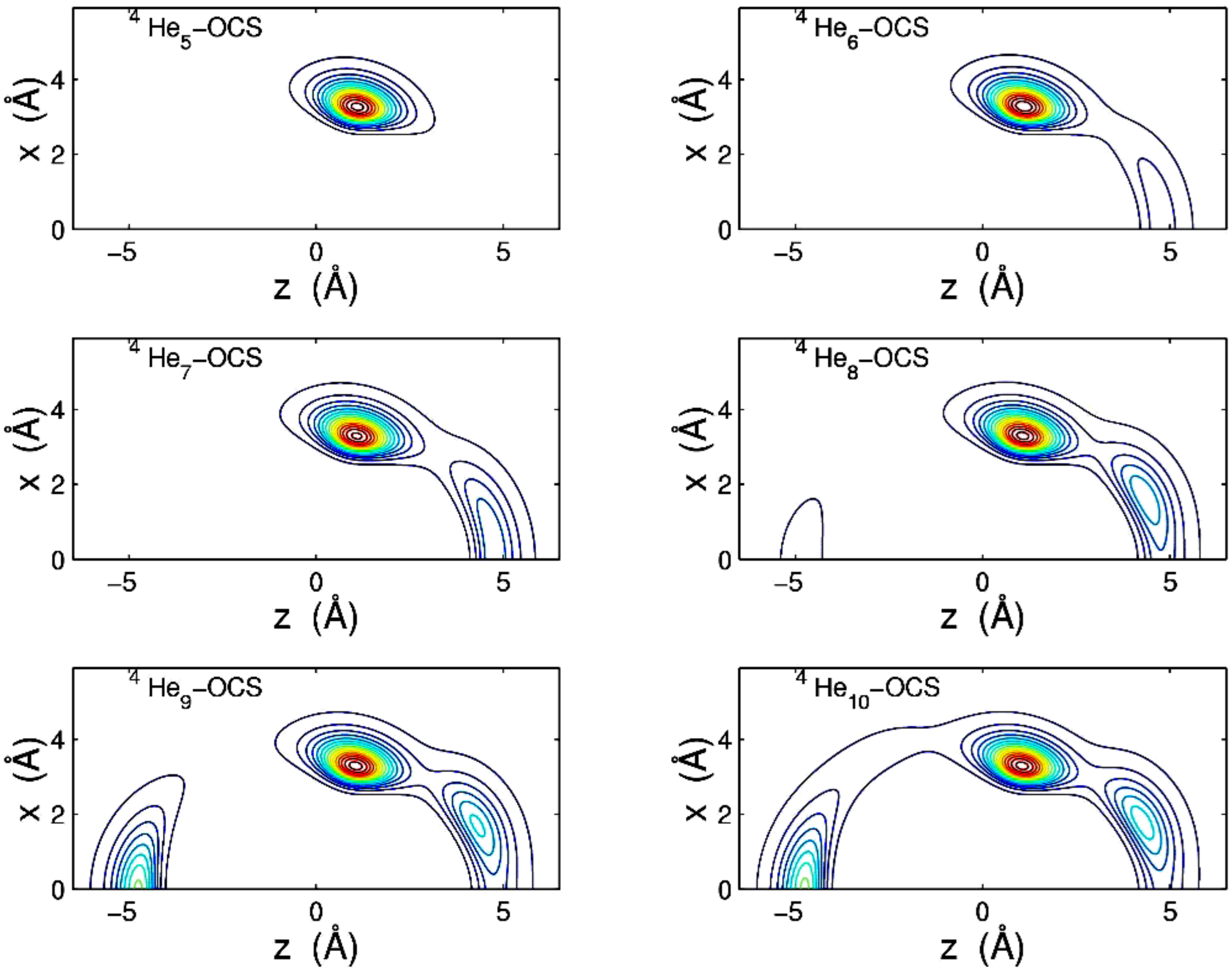
N=6

N=7

N=8

N=9

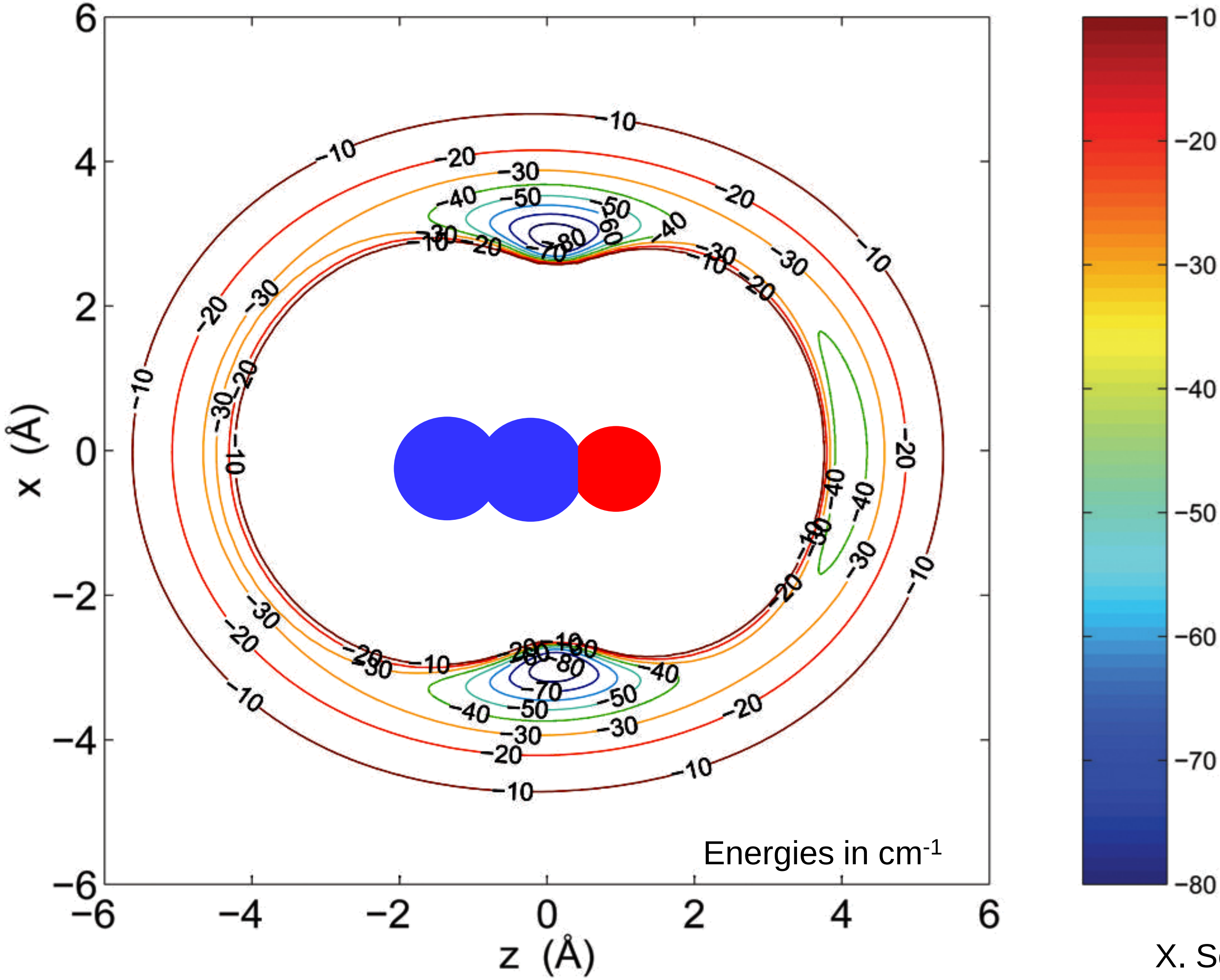
N=10



N. Blinov, X. Song, P. N. Roy, *JCP* **120**, 5916 (2004).

2. He_N-N_2O (N=1-19)

Potential Energy Surface of He-N_2O



level of theory:
CCSD(T)

basis set:
aug-cc-pVTZ

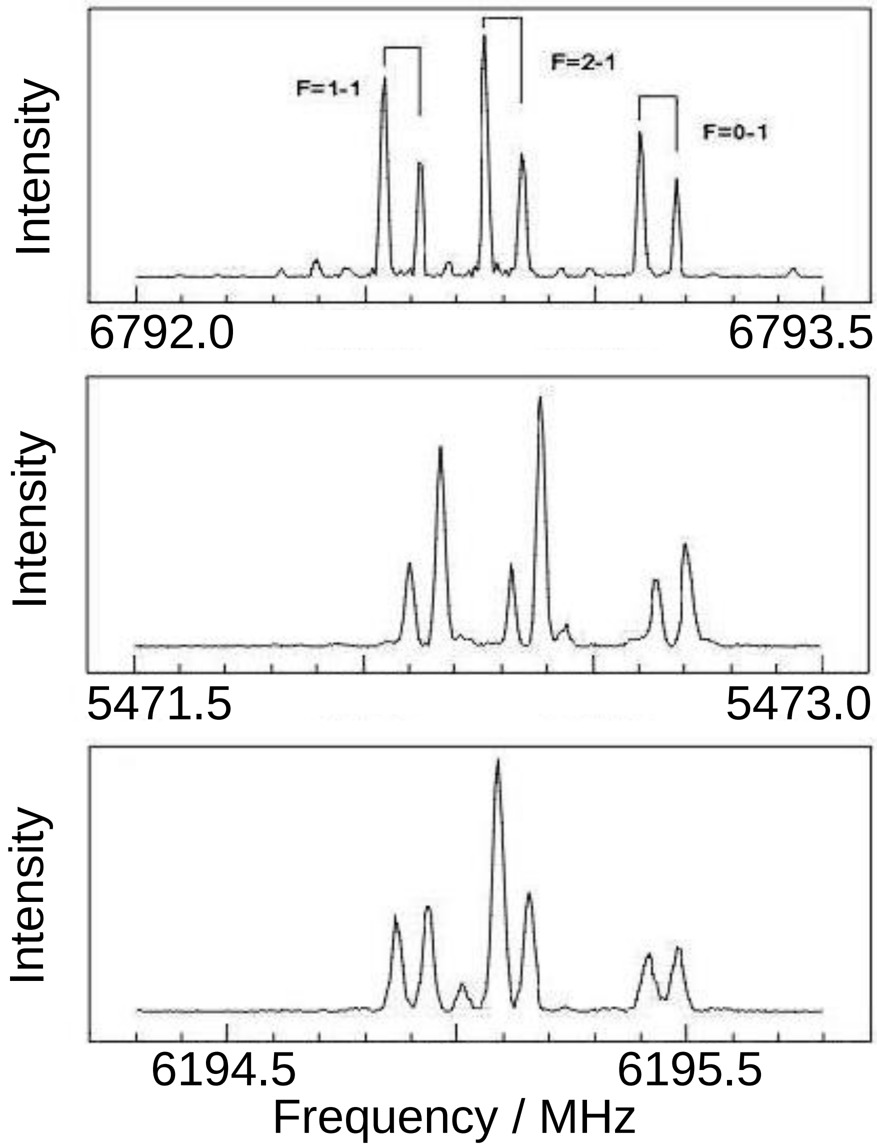
bond functions:
3s, 3p, 2d, 1f, 1g

X. Song, P. N. Roy, Y. Xu, and W. Jäger, submitted.

Bound State Calculations for $He\text{-}N_2O$

Transition	Experiment	Bound	Difference
1_{01} - 0_{00}	18560.5 MHz	18435.1 MHz	0.68 %
1_{11} - 0_{00}	19743.3 MHz	19704.6 MHz	0.20 %
1_{10} - 1_{11}	6295.0 MHz	6222.1 MHz	1.16 %
1_{10} - 1_{01}	7477.5 MHz	7491.5 MHz	-0.19 %
2_{20} - 2_{21}		5035.0 MHz	
2_{11} - 2_{12}		18465.7 MHz	
2_{02} - 1_{11}		30342.8 MHz	
2_{11} - 1_{10}		30657.1 MHz	
2_{02} - 1_{01}		31612.3 MHz	
2_{11} - 1_{10}		42900.7 MHz	

J=1-0 Rotational Transition



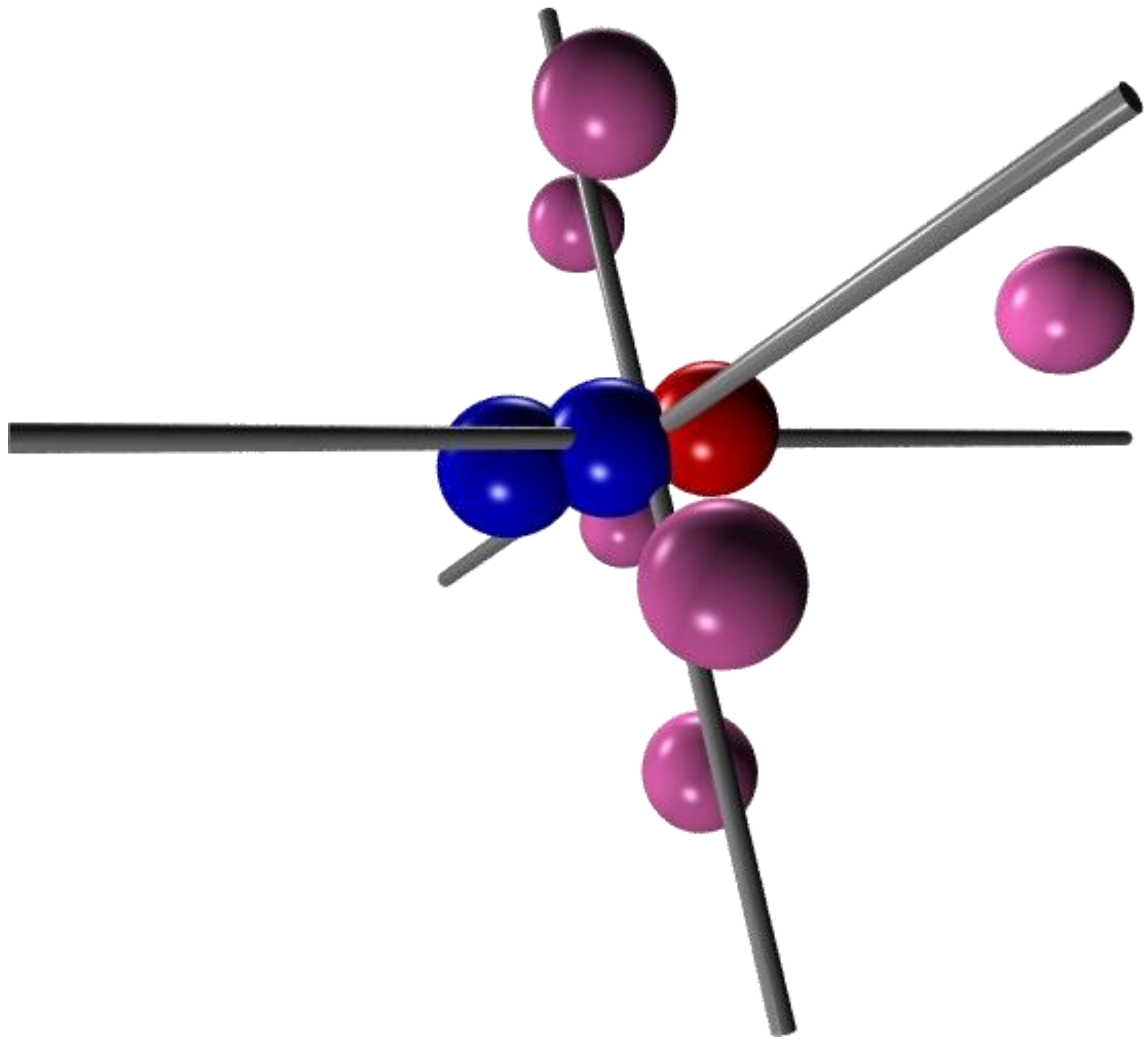
He_5-$^{14}N^{15}NO$

He_7-$^{14}N^{15}NO$

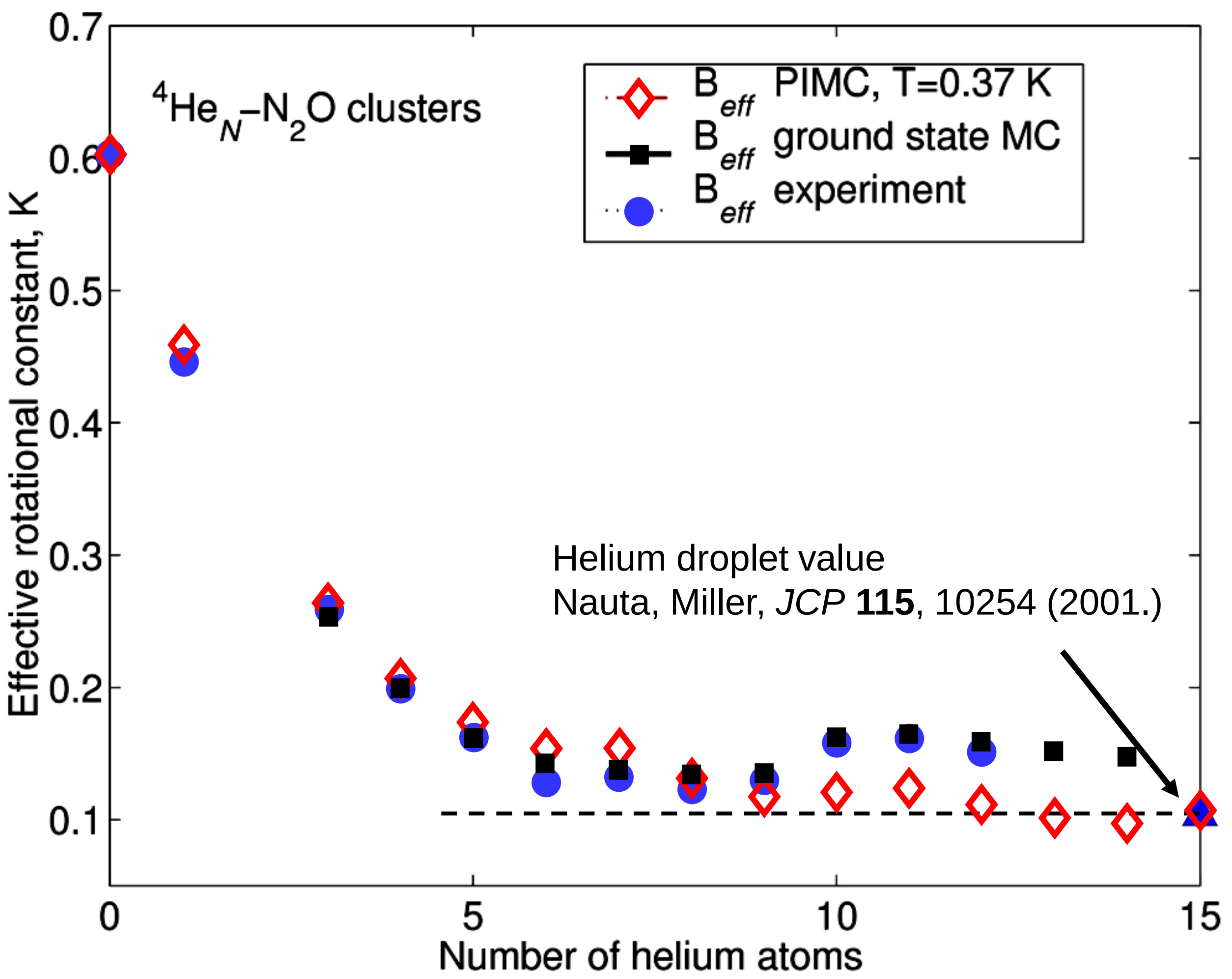
He_{12}-$^{14}N^{15}NO$

Xu, Jäger, Tang, McKellar,
Phys. Rev. Lett. **91,** 163401 (2003).

He_6-N_2O in its Principal Inertial Axes System



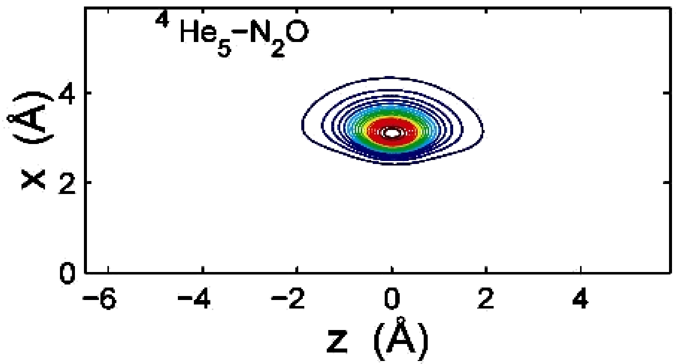
Rotational Constant vs.Number of He Atoms

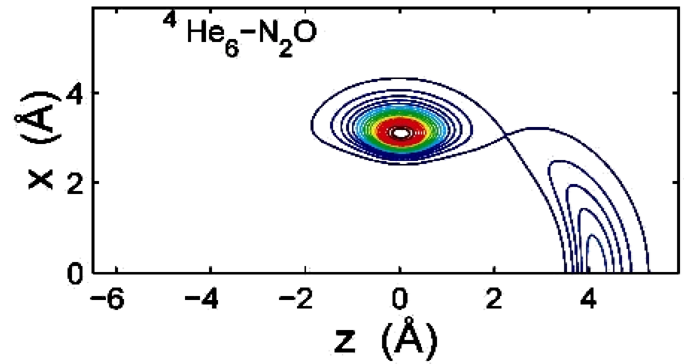


S. Moroni, N. Blinov, P. N. Roy, JCP, accepted.

Helium Density Distributions in He_N-N_2O

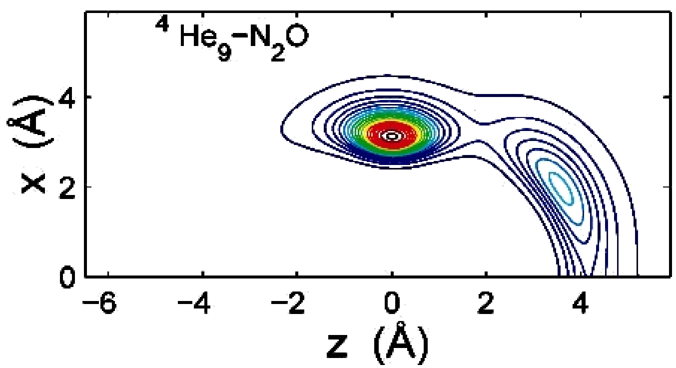
N=5

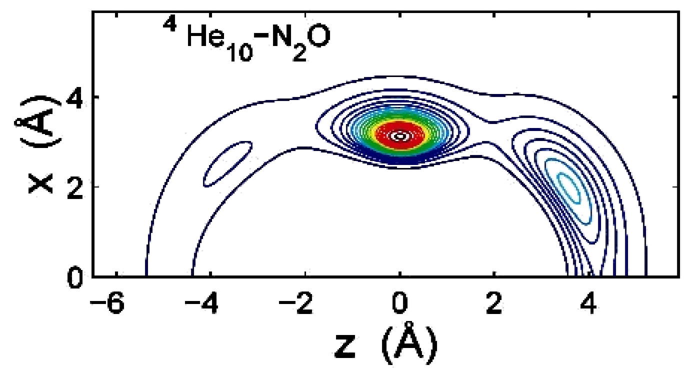




N=6

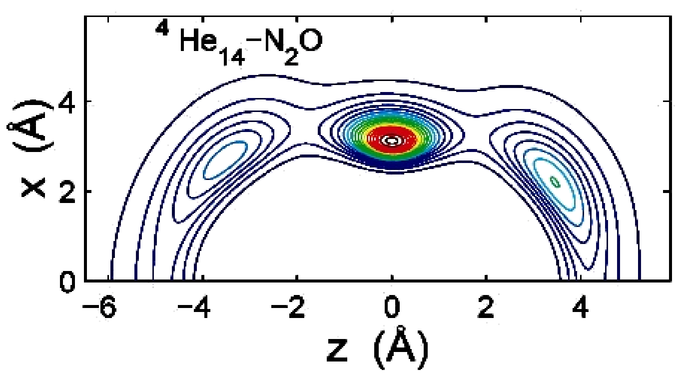
N=9

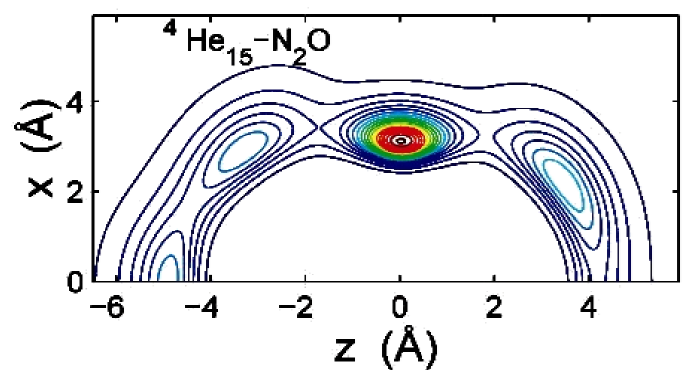




N=10

N=14





N=15

Our Plan for the Future

- Push to even larger cluster sizes (N~60?).
- Use non-linear dopant molecules.
- $(H_2)_N$-molecule systems (already in progress).

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