Molecular Approach to Superfluid Nano Clusters

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第5回クラスター階層領域研究会

Self-introduction

Susumu Kuma
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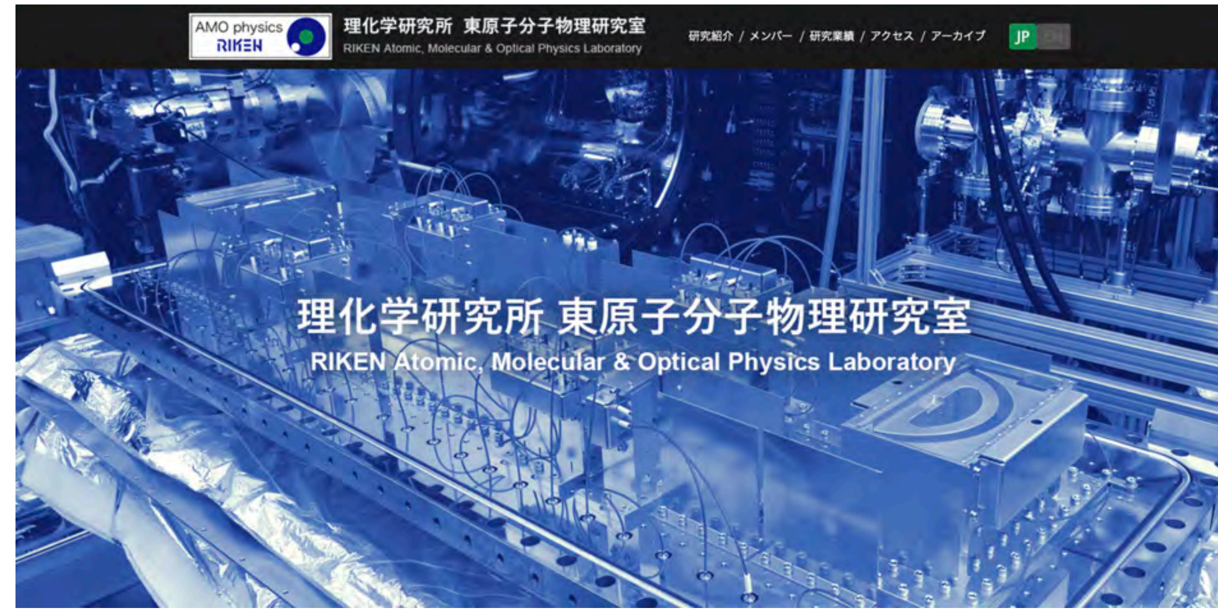
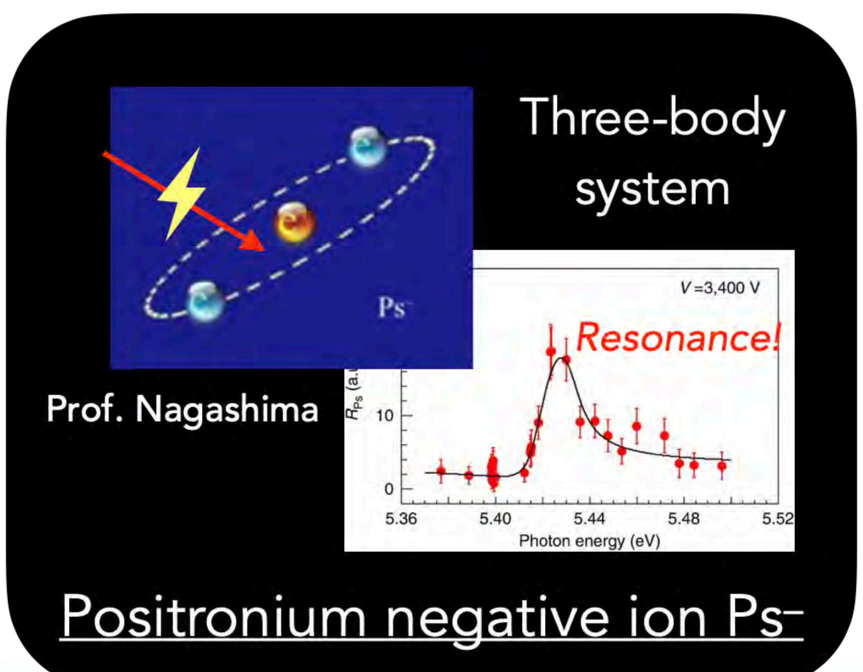
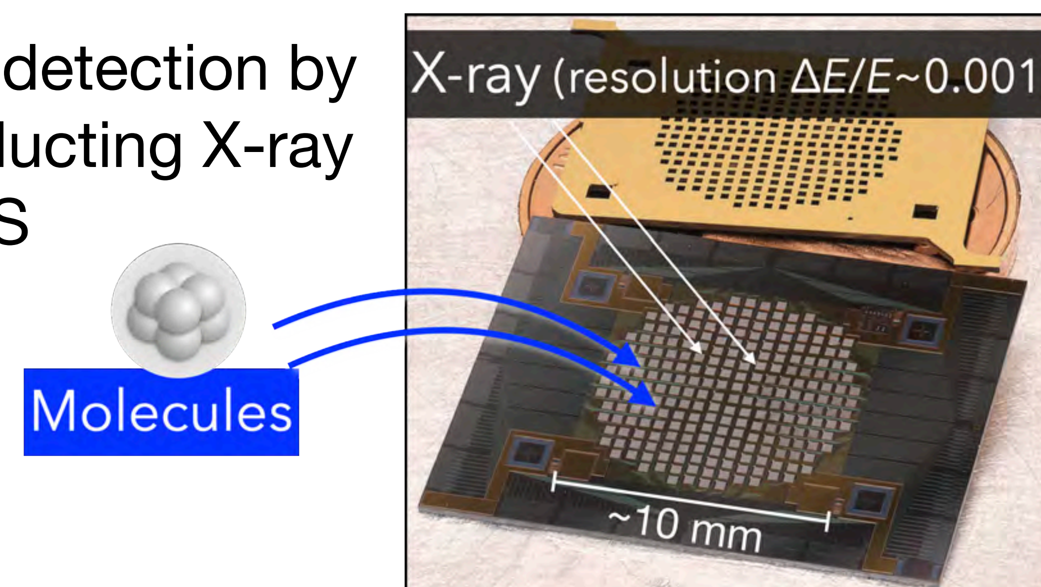


My research activities: experiments using ~~ultra~~cold atoms and molecules

And



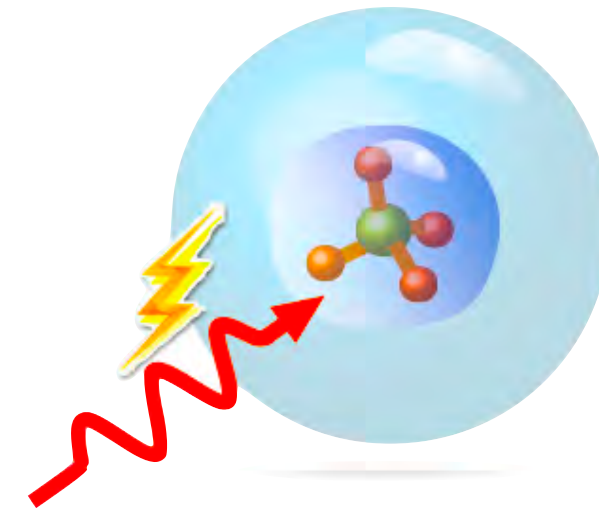
Molecular detection by superconducting X-ray sensor TES

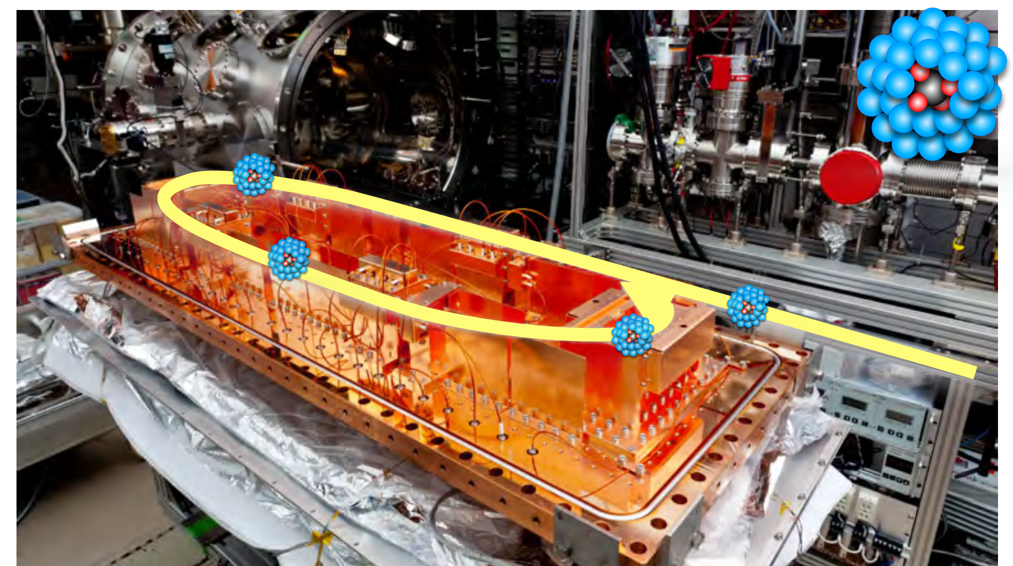


Contents

of this talk

1. Helium droplets as superfluid nano clusters
2. Nano-scale superfluidity probed by molecules
    1. Superfluid response
    2. Superfluid hydrogen!?
3. On-going projects
    1. Slow dynamics
    2. *Micro*- to *macroscopic*

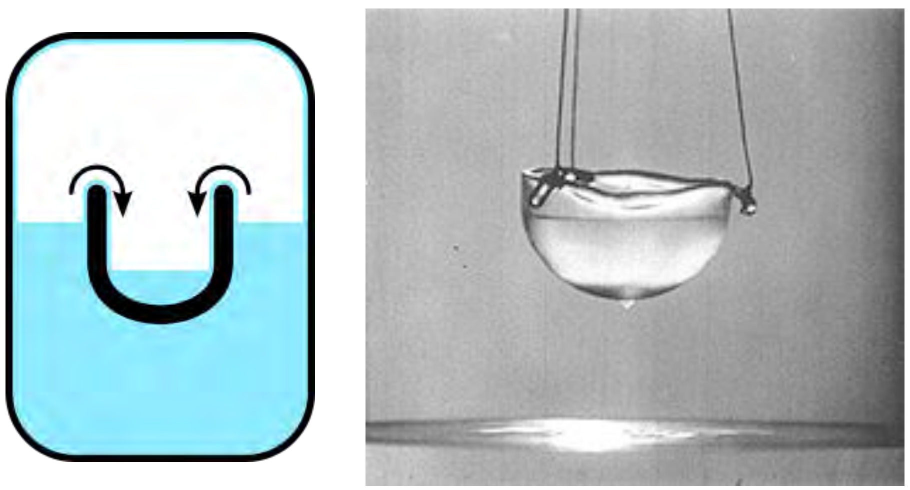




Superfluidity of liquid helium

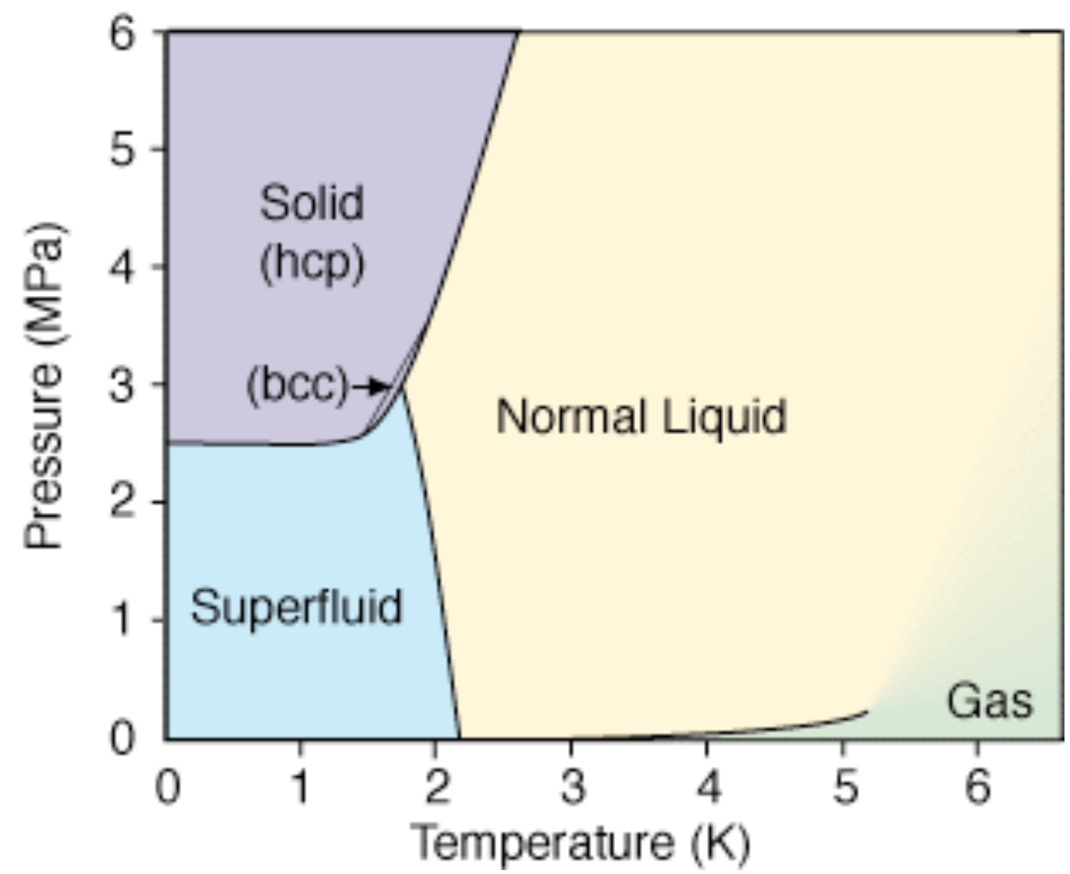
What is superfluid?

Flow without viscosity



etc.

^{4}He phase diagram



Bosonic nature

(Delocalization of atoms)

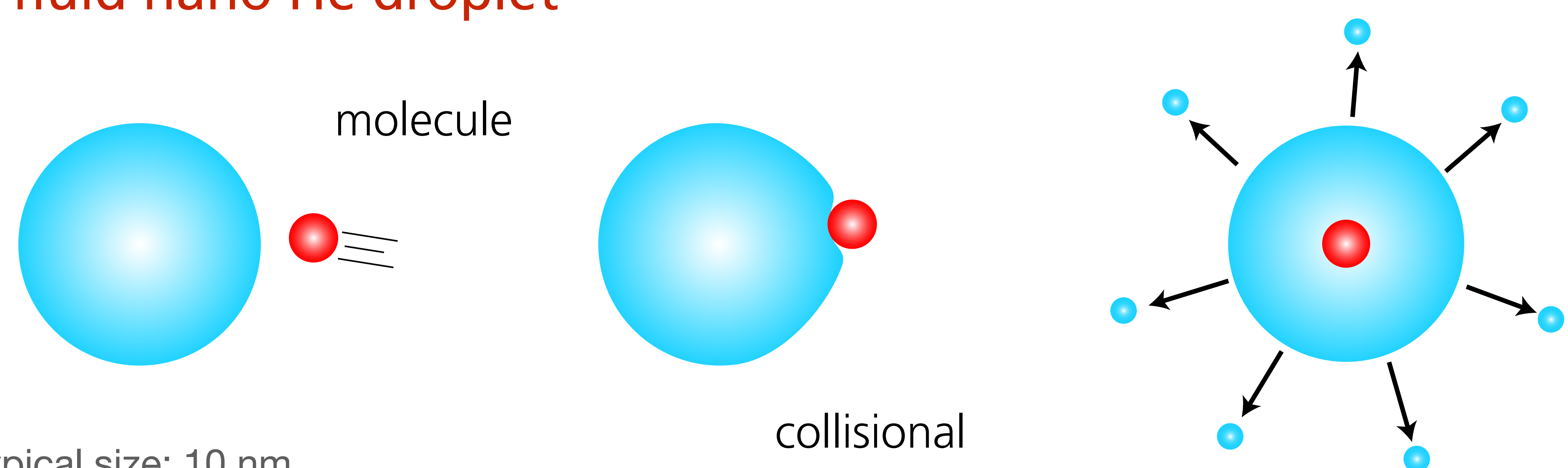
Firstly realized in liquid ^{4}He

Differences from classical liquids

1. Remains liquid at T=0 (at p=0)
2. Becomes superfluid at T = 2.17K

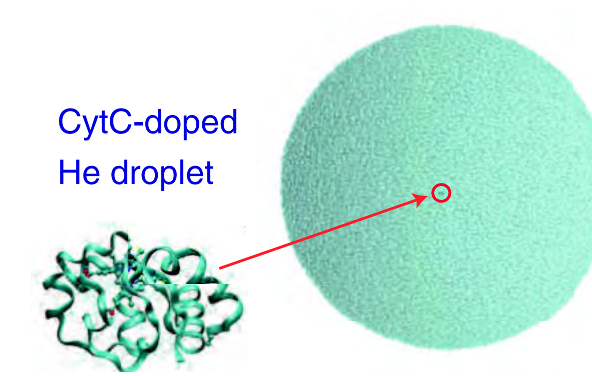
Capturing molecules in He droplets

Superfluid nano He droplet



Typical size: 10 nm

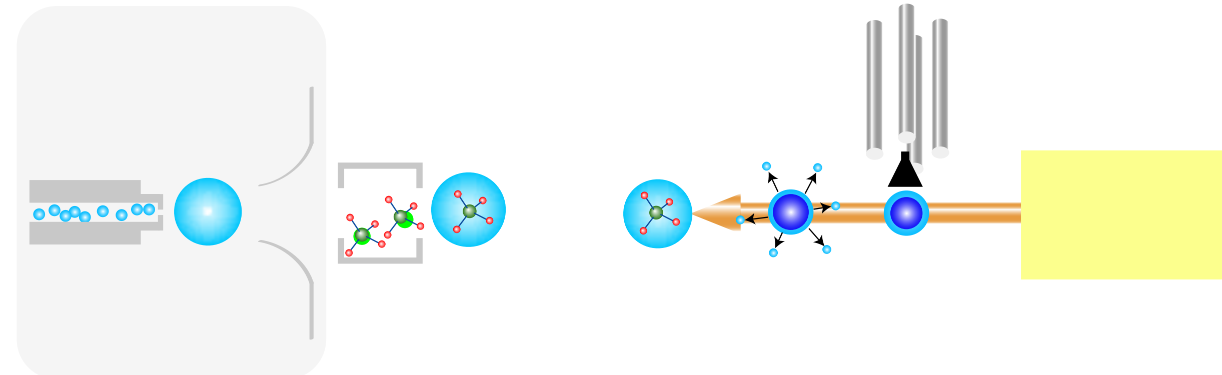
Molecular isolation is easy!



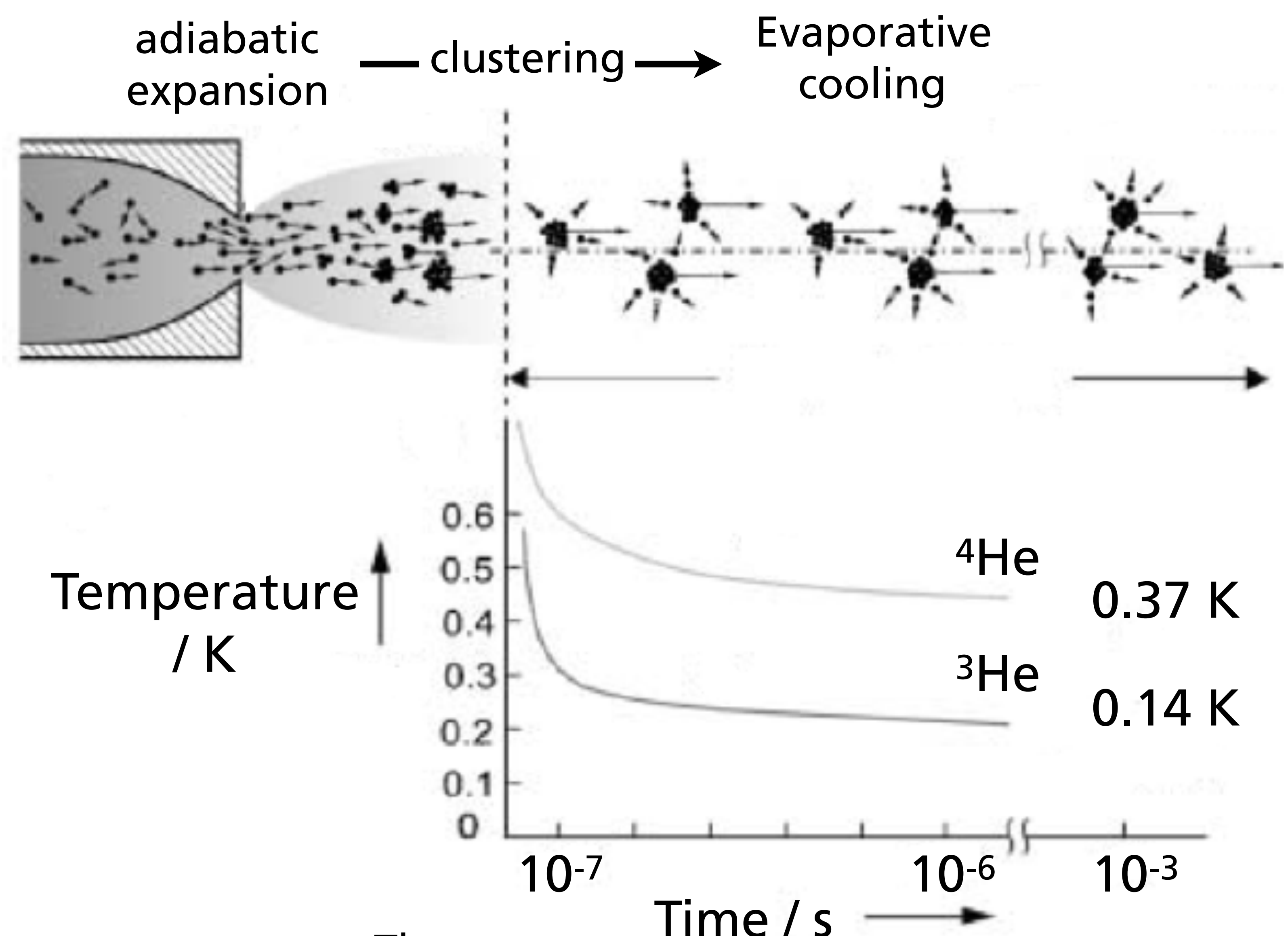
12.3 kDa

Atoms to protein molecules

Droplet production

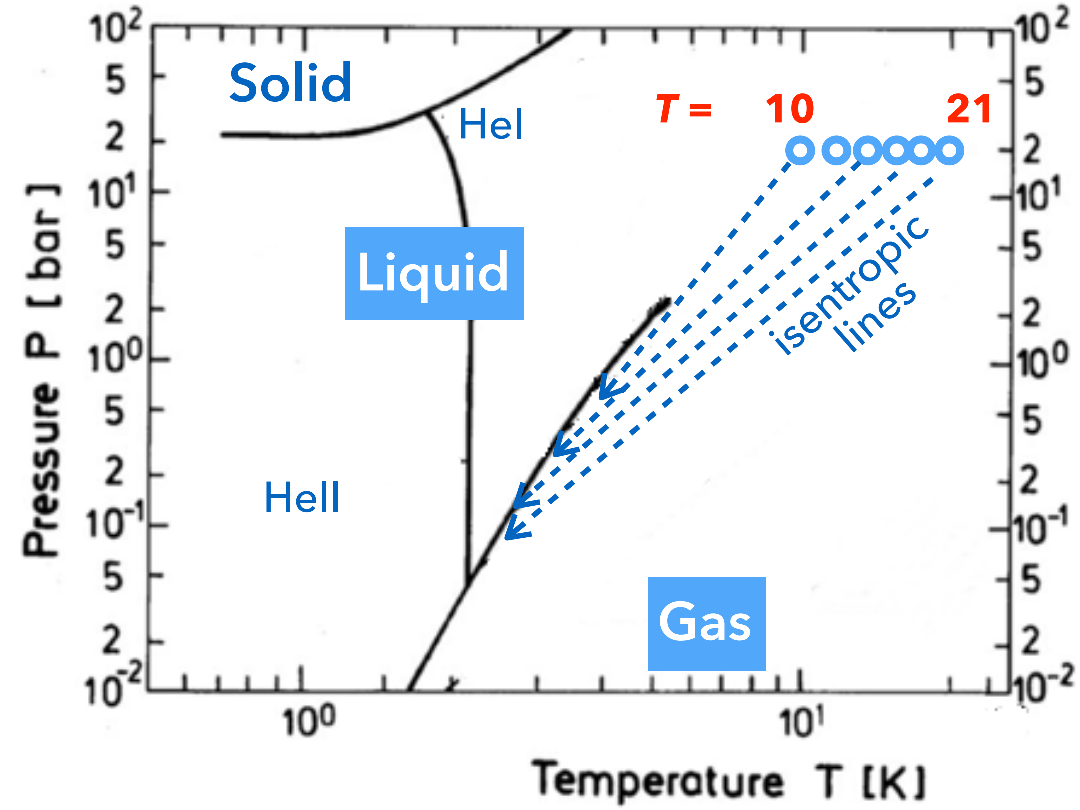


"Molecular beam" =
Nozzle expansion of cold, high-pressure gas



Theory
Stringari *et al. Z. Phys. D* **15**, 257 (1990)

Phase diagram

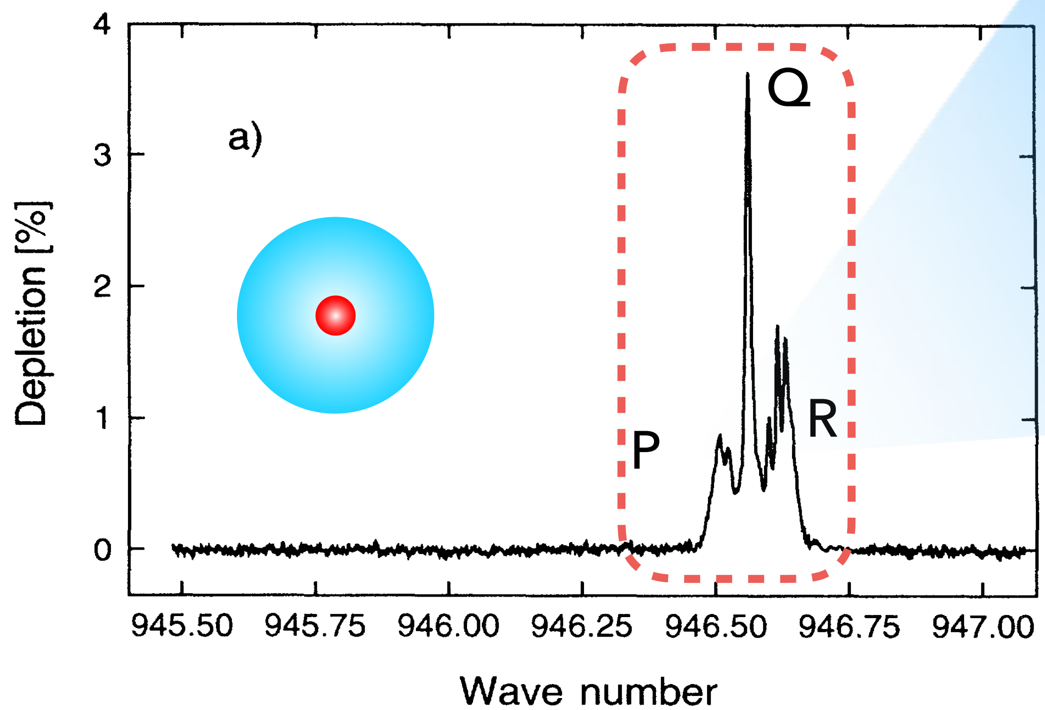


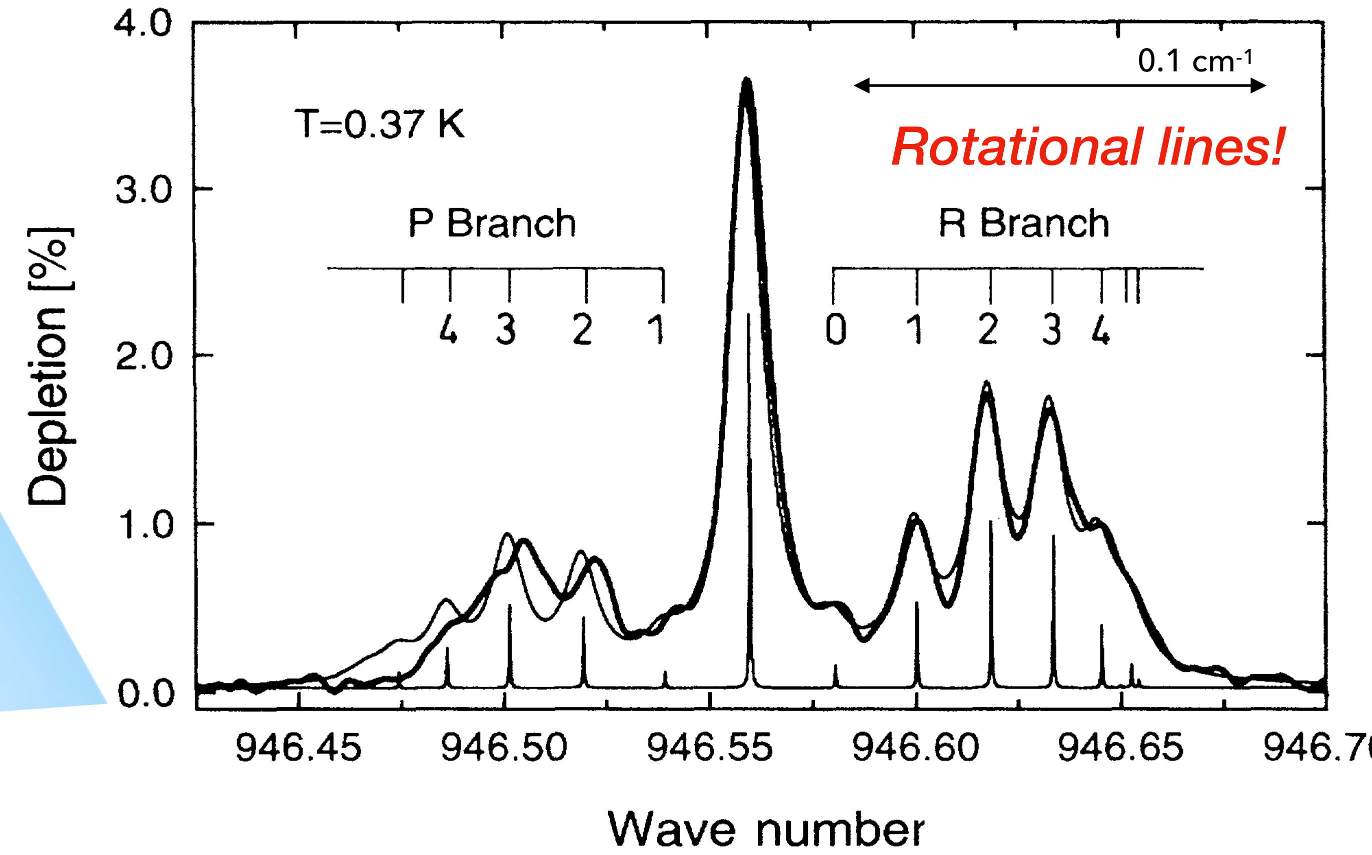
0.37 K within ~10 µs
=> *Superfluidity*

Molecular rotation in He droplets

SF_6 rotational spectroscopy

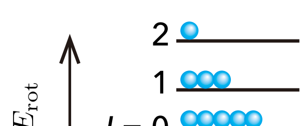
Hartmann et al, Phys. Rev. Lett. 75, 1566 (1995)





<u>Two important issues were found.</u>

1. Droplet *temperature* can be determined from rotational population of the dopant molecules.



$g\exp(-\frac{E_{\rm rot}}{kT})$ ➡ **$T = 0.37$ K**

$E = BJ(J+1) + DJ^2(J+1)^2$

"Thermometer"

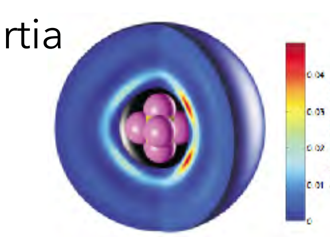
2. Rotational constants decreases by about 1/3.

Increase in the "effective" moment of inertia

rotational constant $B = \hbar^2/2I$

➡ **6-8 He atoms attached**

"Helium response"



Theory (PIMC): Whaley et al. (2000)

Superfluidity of helium droplets

Compare ^{3}He and ^{4} He droplets

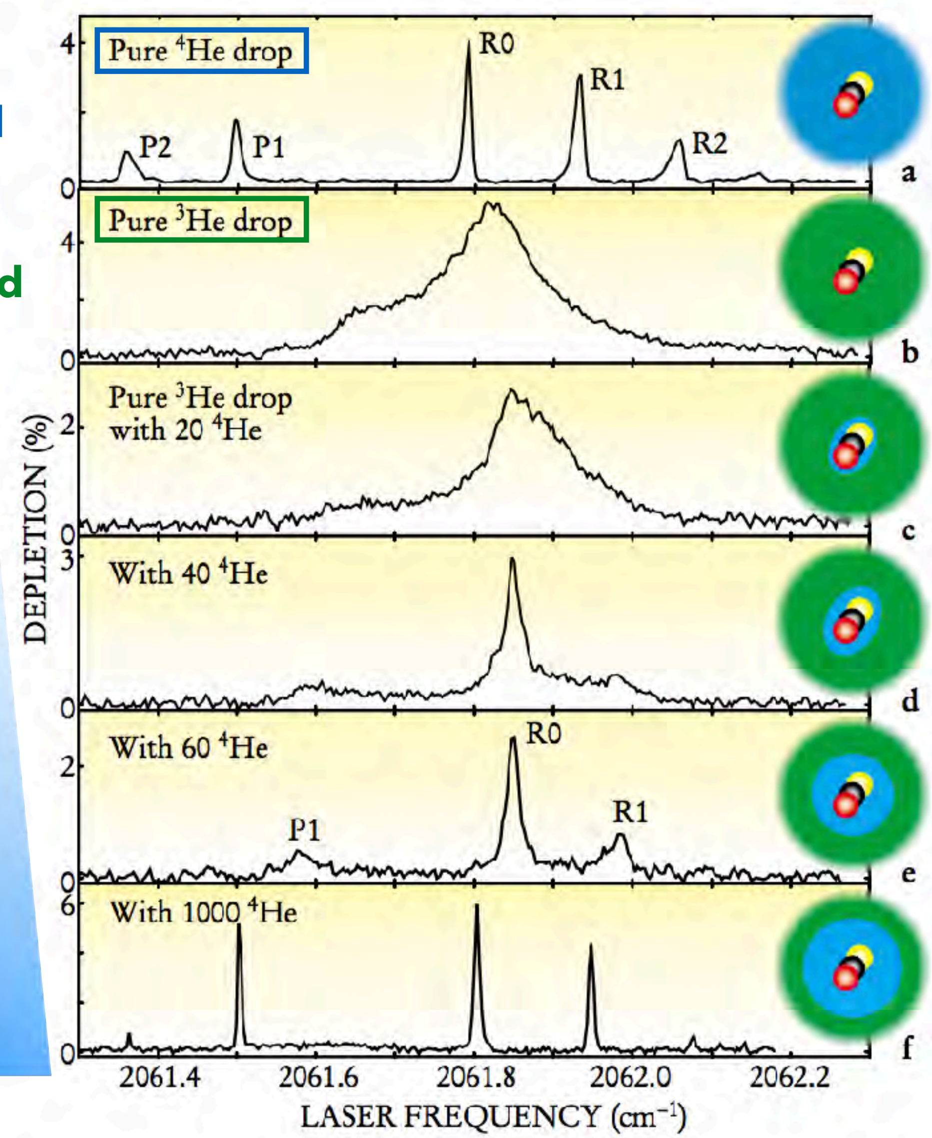
Tennis and Vilesov, Science 279, 2083 (1998).

OCS in ^{4}He/^{3}He droplets

superfluid

normal fluid

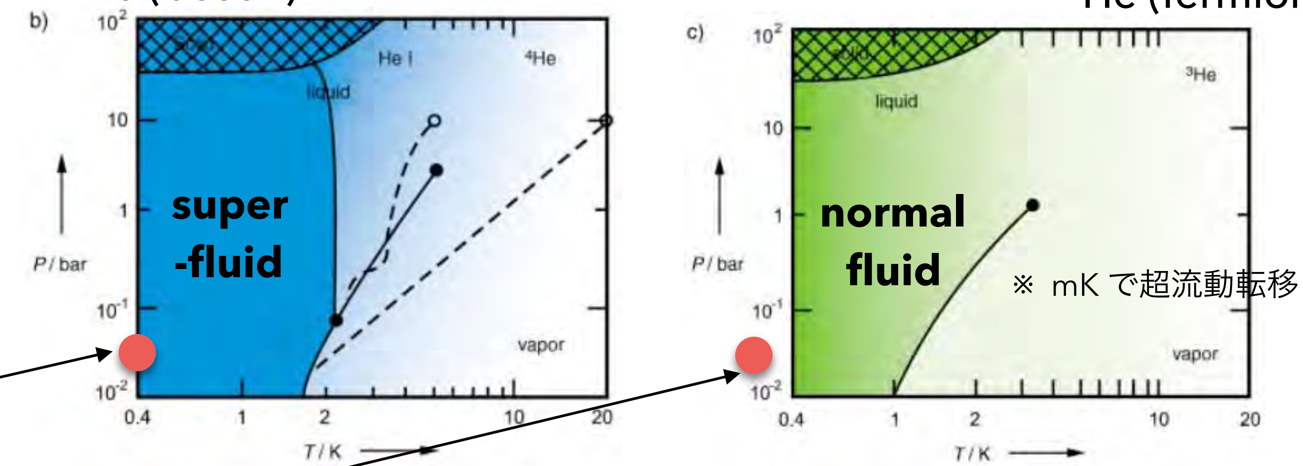
superfluidity



Phase diagrams

^{4}He (boson)

^{3}He (fermion)



Angew. Chem. Int. Ed. 2004, 43, 2622 – 2648

1. _Sharp_ rotational structure originates from the **superfluidity** of helium droplets.

2. _N = 60_ ^{4}He atoms are required to observe the **superfluid** character.

Size effect bridging over the neighboring hierarchies

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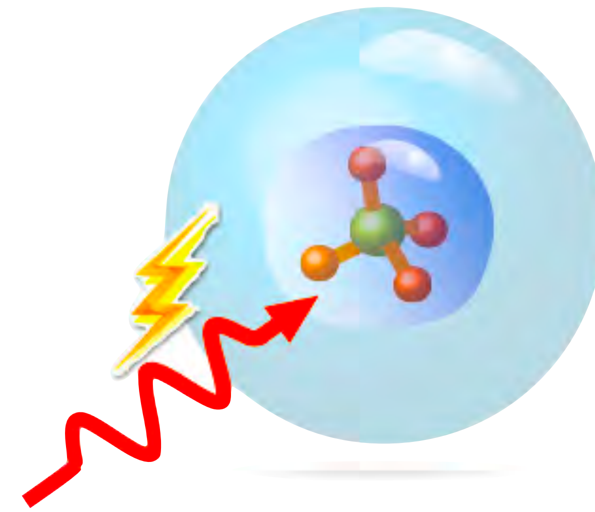
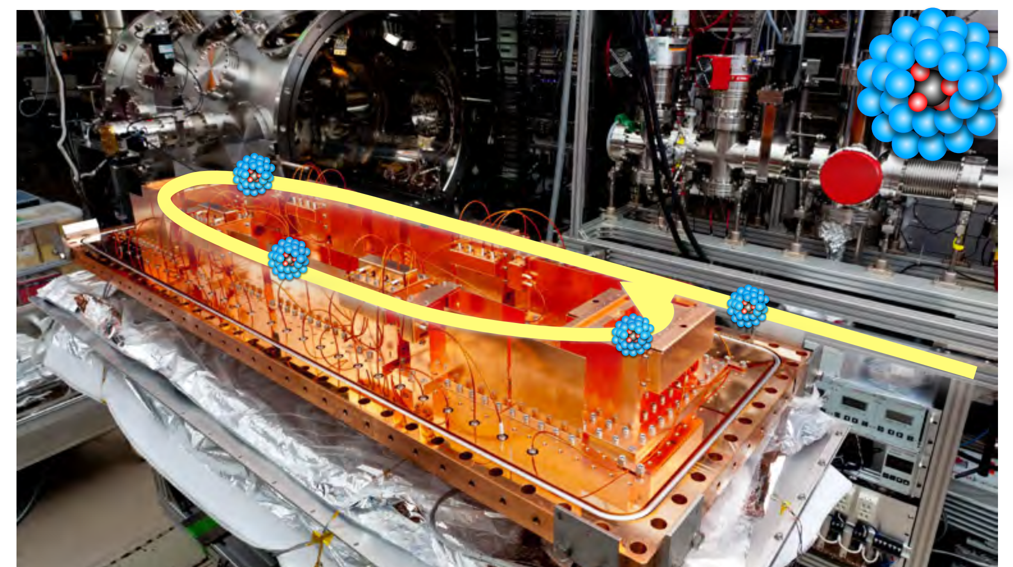
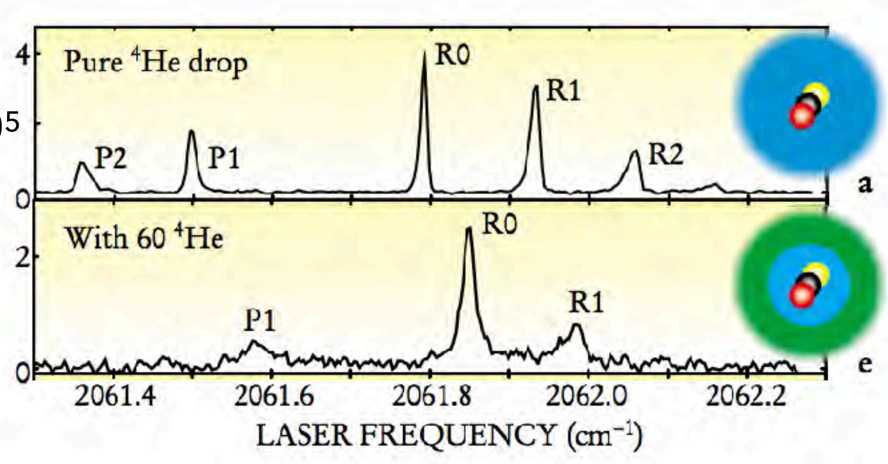




Relation between micro-&macroscopic

Microscopic superfluidity

N = 10^5



Response to molecular rotation

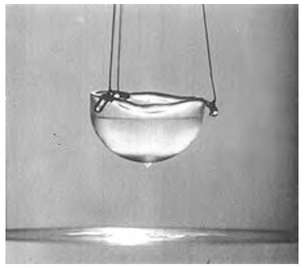
⇓

Microscopic probe of superfluidity

?

Macroscopic superfluidity

Flow without viscosity



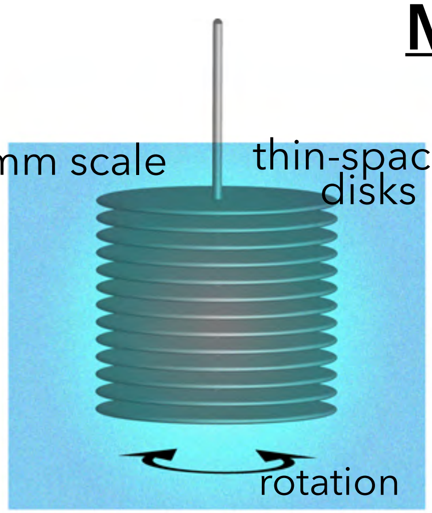
&

fountain effect

second sound

etc…

Macroscopic effects



in superfluid He (bulk)

Andronikashvili's experiment

"two-fluid model"

Quests for the next stage of understanding superfluidity in helium droplets

New probe of microscopic superfluidity

Other superfluids

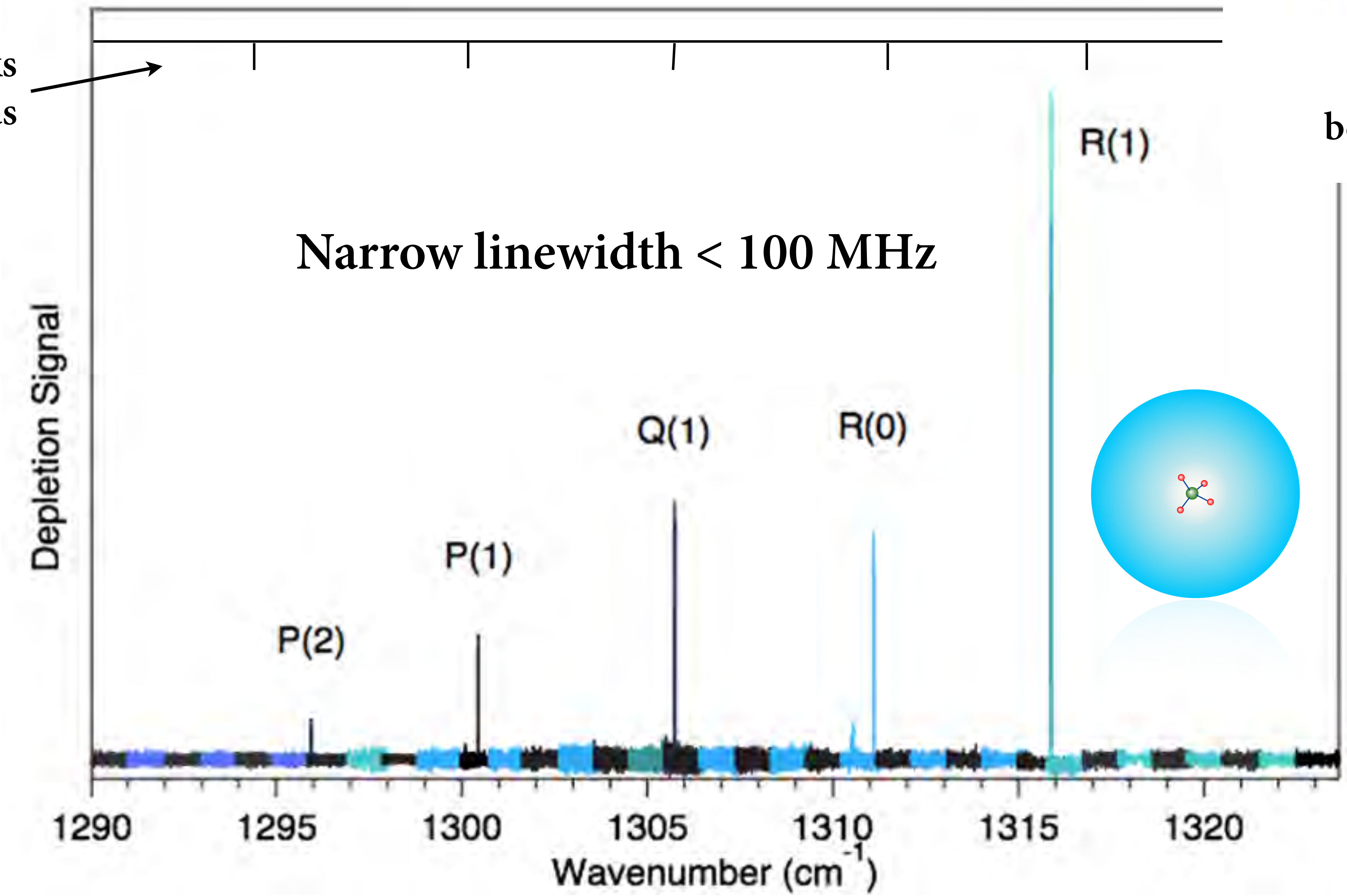
Superfluidity in large droplets

Aakash, Kuma et al., Phys. Rev. A 84, 020502 (2011)

Rotational probe of molecules

ν_4 of CH_4 in He droplets with N_{He} = 2000

peaks in gas



Narrow linewidth < 100 MHz

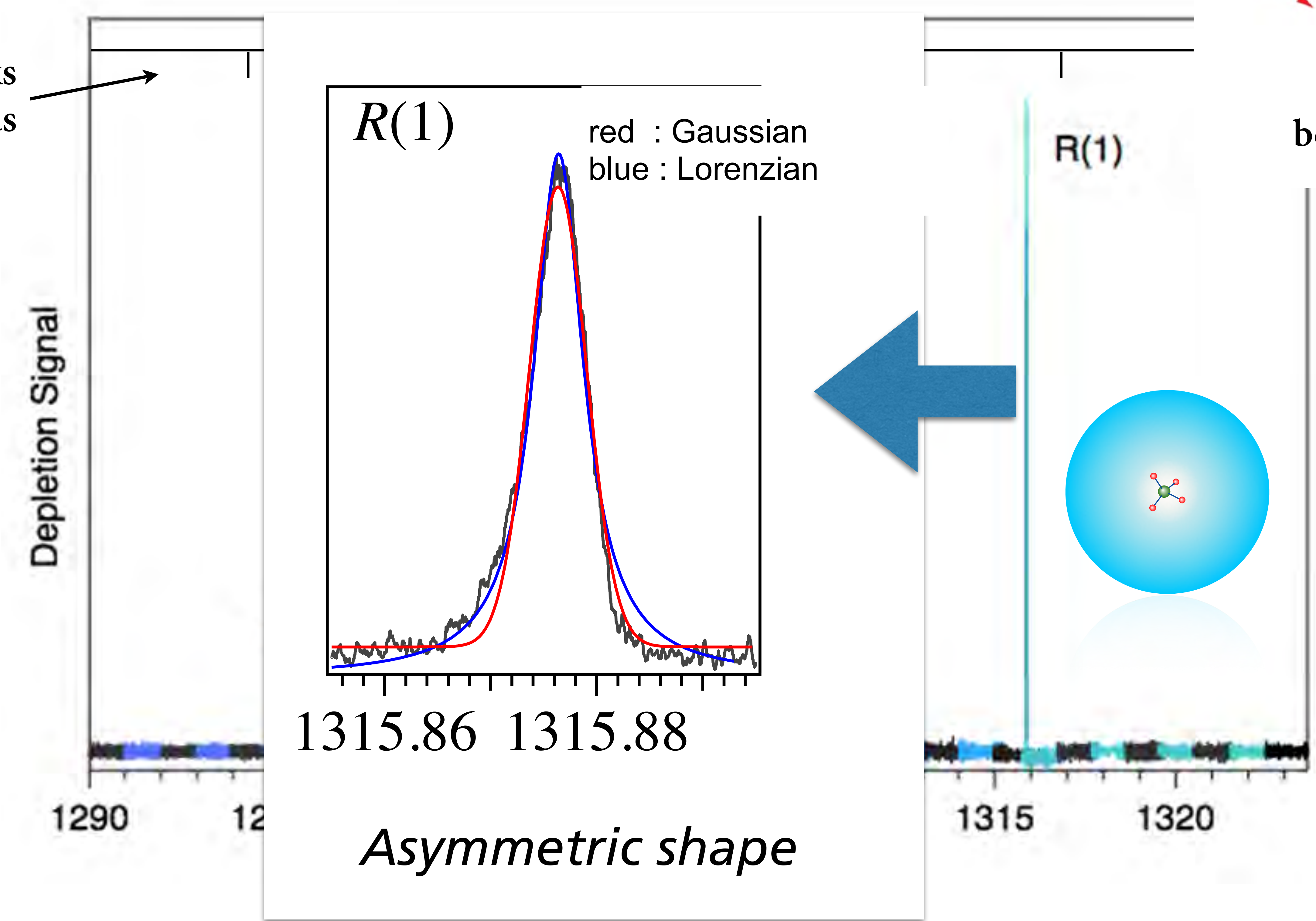
Aakash, Kuma et al., Phys. Rev. A 84, 020502 (2011)

Results

ν_4 of CH_4 in He droplets with N_{He} = 2000

ν_4
bend

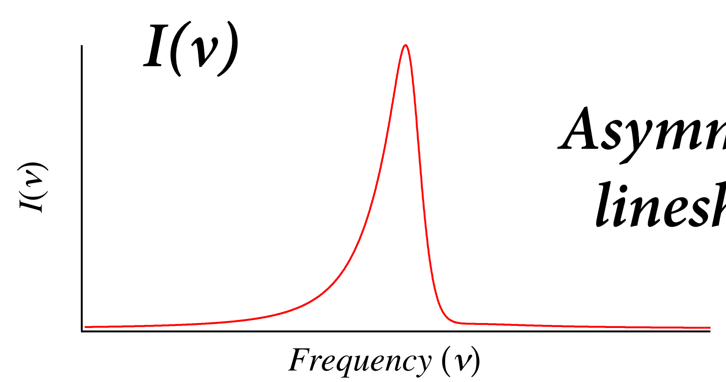
peaks
in gas



Lineshape analysis for dynamics

Lineshape

$$I(\nu) \propto \mathrm{Re}\left[\int_0^{\infty} c(t) e^{-2\pi i \nu t} dt\right]$$

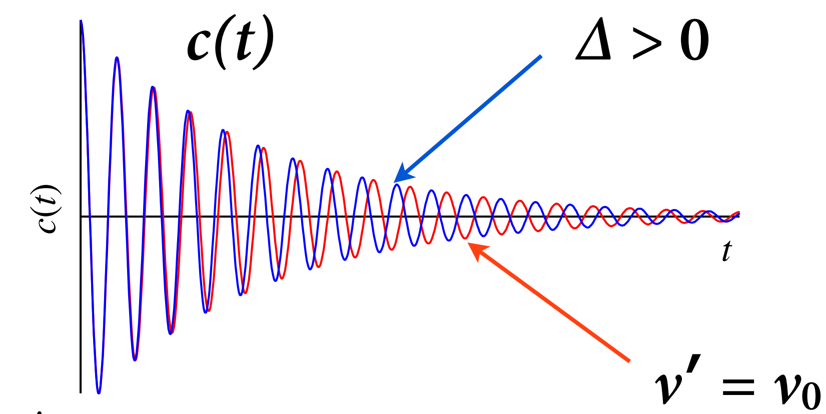


Asymmetric lineshape

Fourier transform

Correlation function of the dipole moment

$$\begin{aligned} c(t) &= \langle \hat{\mu}(0)\hat{\mu}(t)\rangle \\ &= \exp(2\pi i \nu' t)\exp(-2\pi\gamma t) \end{aligned}$$



$$\nu'(t) = \nu_c + \Delta[1 - \exp(-2\pi\beta t)]$$

Dynamically skewed spectral lines arise for chirped damped oscillators.

Apkarian et al.
[JCP 133, 054506(2010)]

Aakash, Kuma et al., Phys. Rev. A 84, 020502 (2011)

Results of skewed line analysis

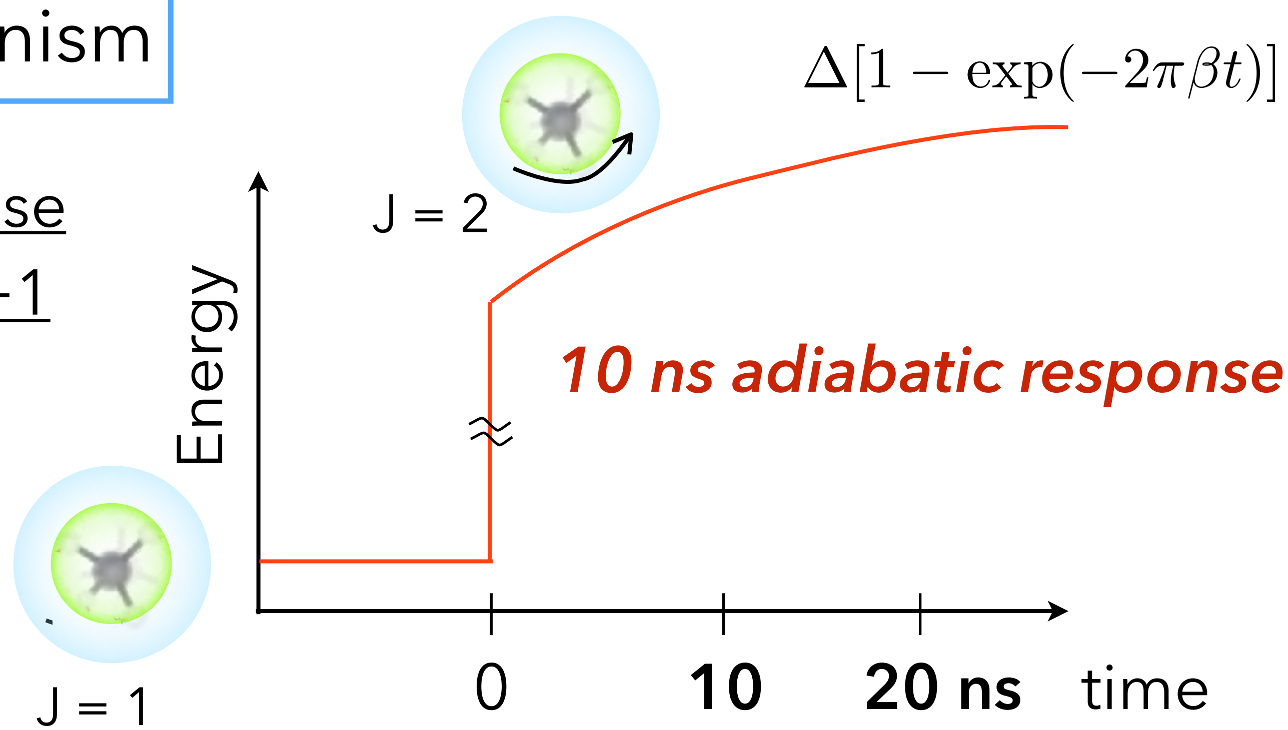
Skewed parameters $\nu'(t) = \nu_c + \Delta[1 - \exp(-2\pi\beta t)]$

	Parameters				
	ν_c (cm^{-1}) [a]	γ (MHz)	Δ (MHz)	β (MHz)	
$P(2)$	1295.9475(25)	160.5	−68.8	47.5	$\Delta < 0$
$P(1)$	1300.4409(11)	36.1	0	–	
$Q(1)$	1305.7325(32)	32.0	0	–	
$R(0)$	1311.0918(5)	40.0	0	–	
$R(1)$	1315.8753(19)	76.3	59.0	35.9	$\Delta > 0$

Mechanism

R(1) case

ΔJ = +1



Increase in the rotational constant after the transition (rotor speed-up)

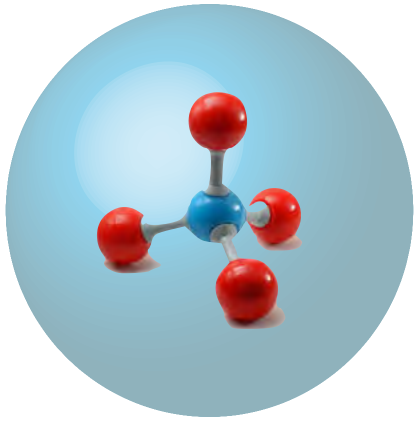
Decrease in the moment of inertia

Detachment of He atoms

Normal- to superfluid transition

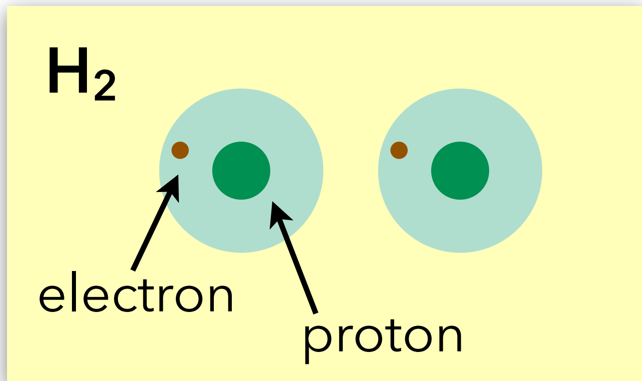
Summary of the CH_4 skewed lineshape

- CH_4, light rotor: sensitive probe of superfluid response of helium droplets.
- Skewed lineshape: the dynamics in the surrounding helium environment.
- Finite time (nanosecond) adiabatic response of helium in the normal- to superfluid transition



Superfluidity of p-H_2?

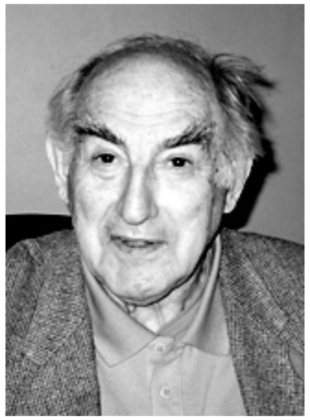
Idea



Composite boson

= Candidate for molecular superfluidity

Ginzburg & Sobayanin (1972)



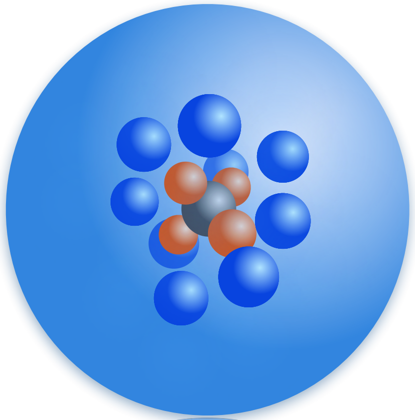
Superfluid transition temperature T_S

$$T_S = \frac{3.31\hbar^2}{k_B}\frac{n^{2/3}}{g^{2/3}m}$$

n: density
m: mass
g: degeneracy

Species	Ts (pred.)
^{4}He	3.1 K (2.2 K, obs.)
H_2, para	6 K
H_2, ortho	1.5 K

← **Next candidate**



Supercooled H_2 Clusters in He droplets @ 0.4 K

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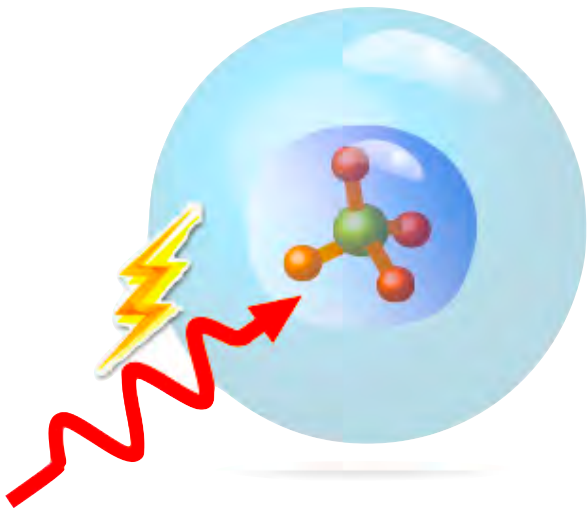
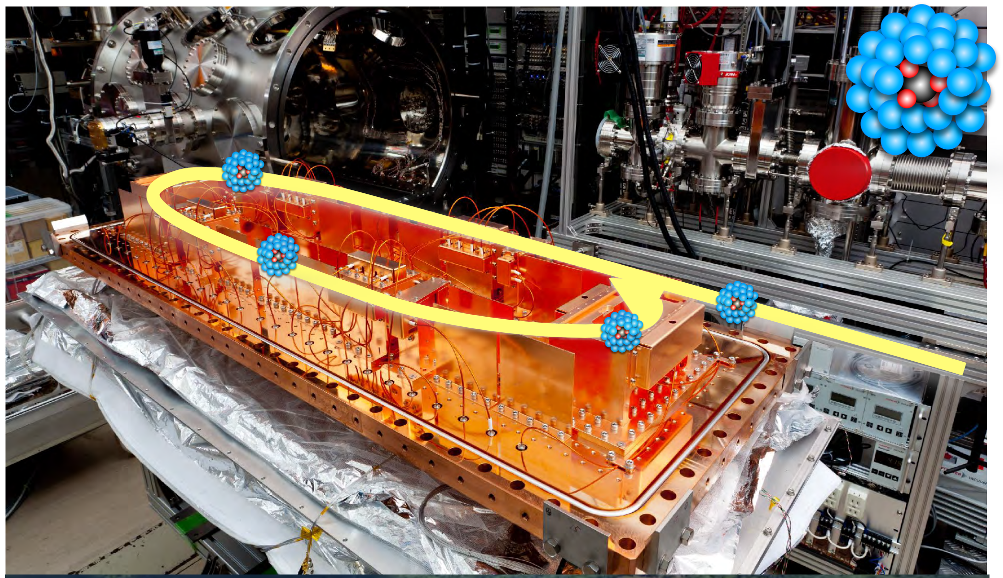
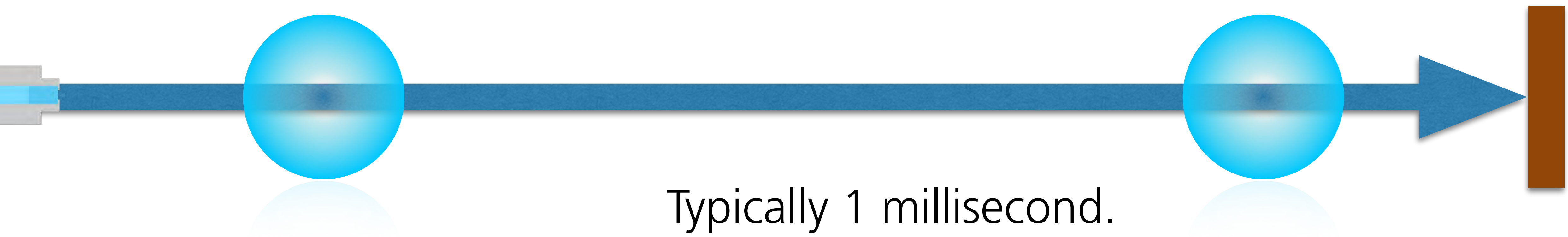


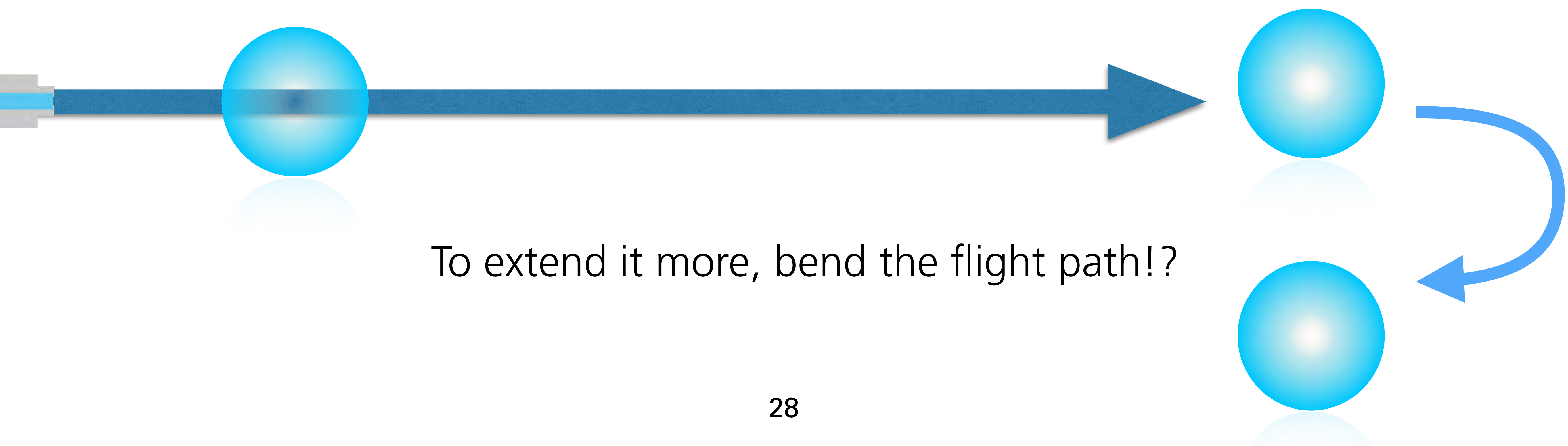


“Lifetime” of He droplets

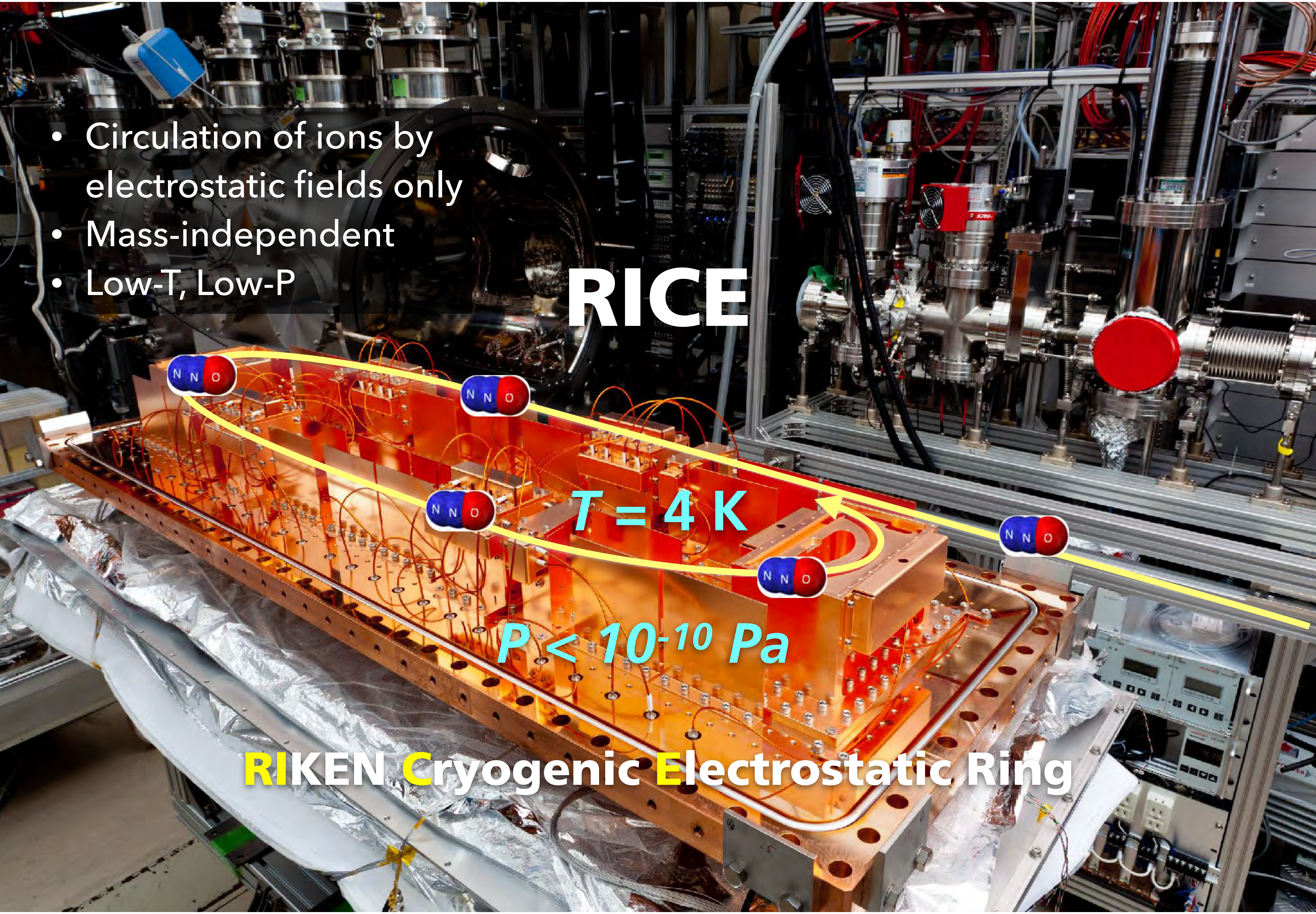


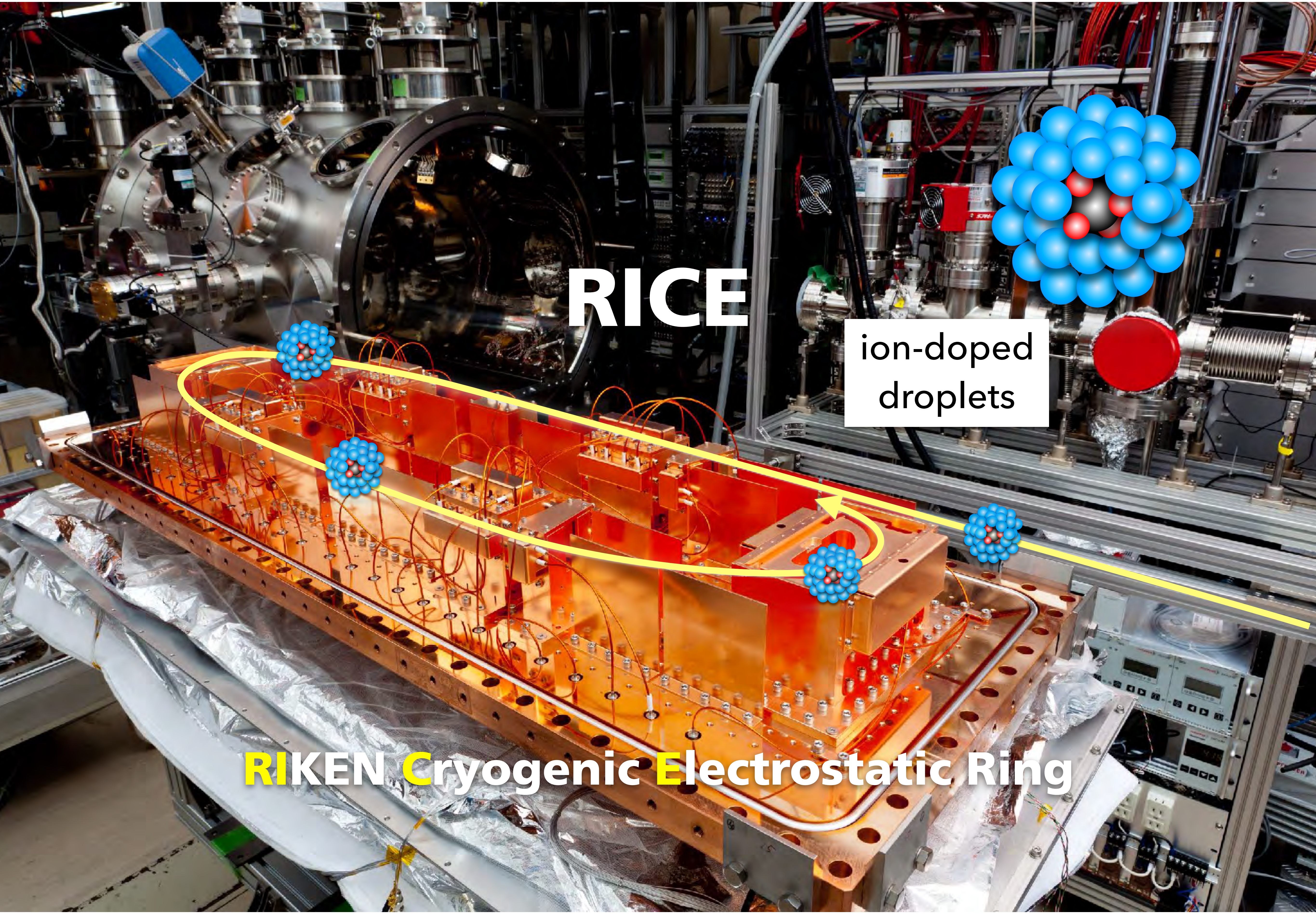
Typically 1 millisecond.

wall

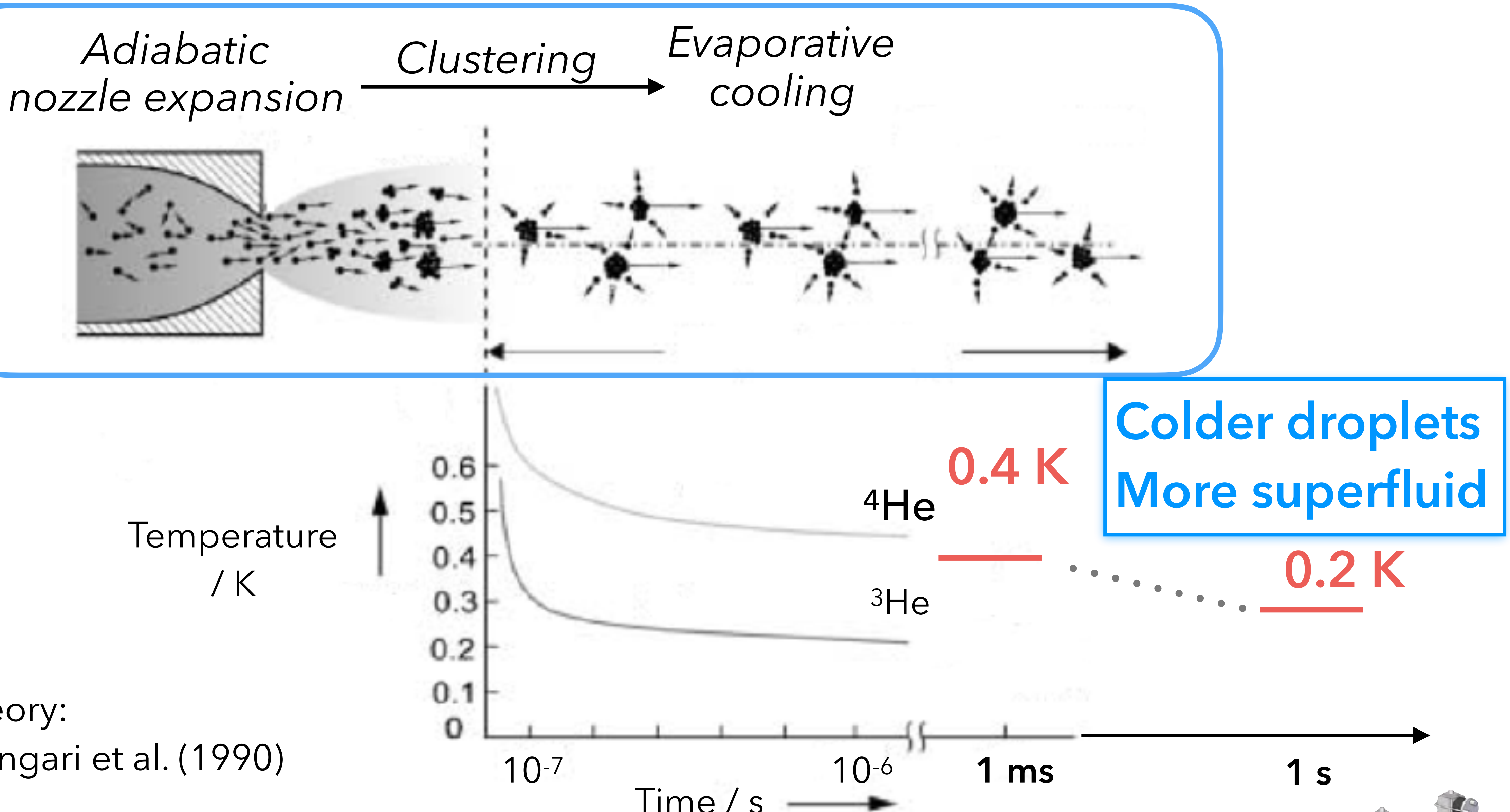


To extend it more, bend the flight path!?


• Circulation of ions by electrostatic fields only
• Mass-independent
• Low-T, Low-P
RICE
N N O
T = 4 K
P < 10-10 Pa
RIKEN Cryogenic Electrostatic Ring

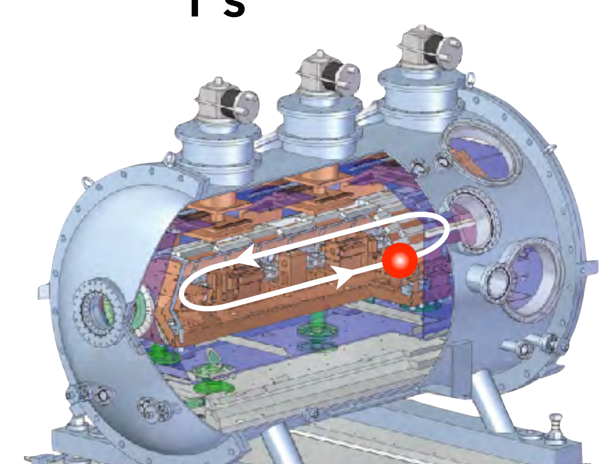

RICE
ion-doped
droplets
RIKEN Cryogenic Electrostatic Ring

Colder droplets expected in the ring

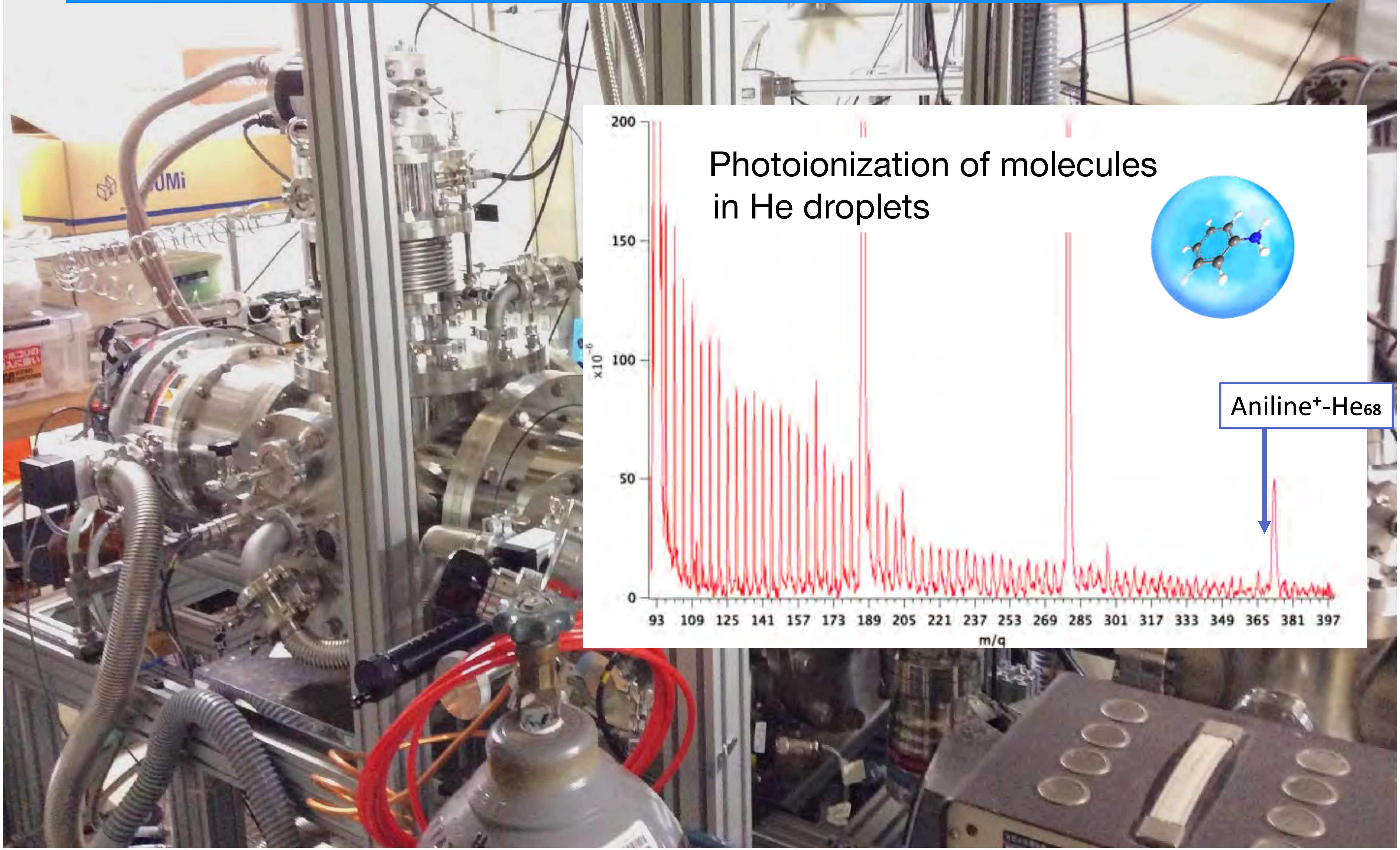


Theory:
Stringari et al. (1990)

0.4 K:
determined by thermal balance
of evaporation vs internal energy in 1 ms



He Droplet Machine @ RIKEN



Direct observation in He droplets

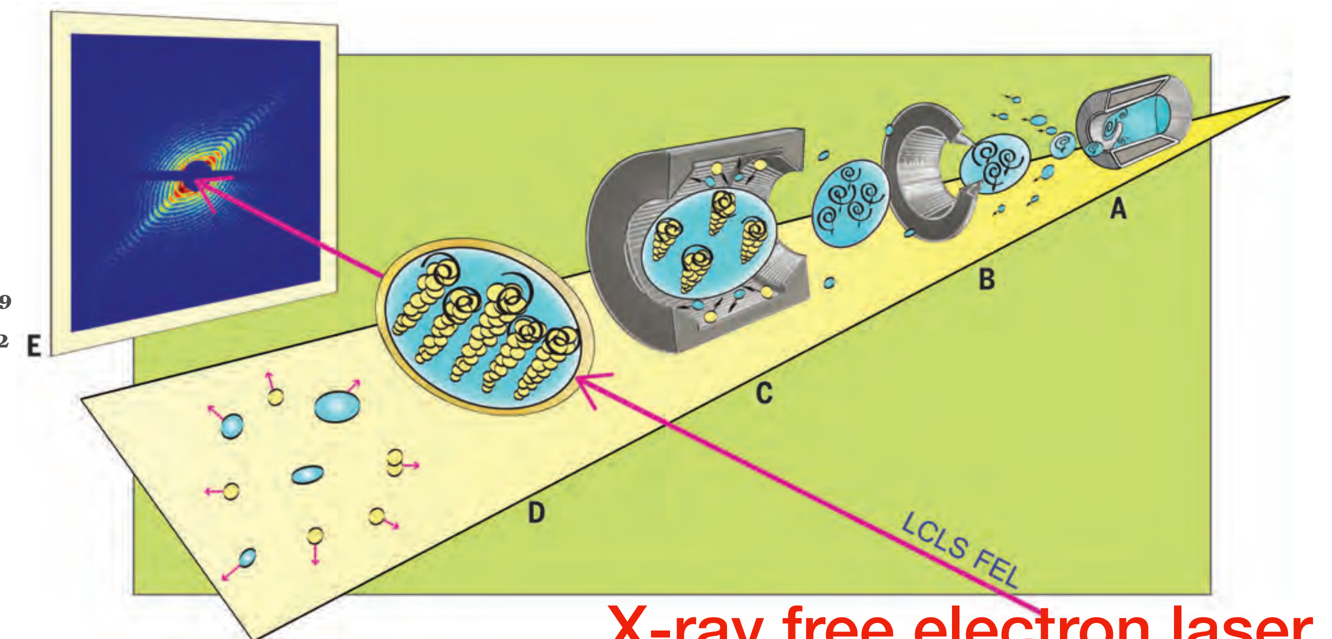
HELIUM SUPERFLUIDITY

Shapes and vorticities of superfluid helium nanodroplets

Luis F. Gomez,[1*] Ken R. Ferguson,[2] James P. Cryan,[3] Camila Bacellar,[3,4] Rico Mayro P. Tanyag,[1] Curtis Jones,[1] Sebastian Schorb,[2] Denis Anielski,[5,6] Ali Belkacem,[3] Charles Bernando,[7] Rebecca Boll,[5,6,8] John Bozek,[2] Sebastian Carron,[2] Gang Chen,[9†] Tjark Delmas,[10] Lars Englert,[11] Sascha W. Epp,[5,6] Benjamin Erk,[5,6,8] Lutz Foucar,[6,12] Robert Hartmann,[13] Alexander Hexemer,[9] Martin Huth,[13] Justin Kwok,[14] Stephen R. Leone,[3,4,15] Jonathan H. S. Ma,[3,16] Filipe R. N. C. Maia,[17‡] Erik Malmerberg,[18,19] Stefano Marchesini,[9,20] Daniel M. Neumark,[3,4] Billy Poon,[18] James Prell,[4] Daniel Rolles,[6,8,12] Benedikt Rudek,[5,6§] Artem Rudenko,[5,6,21] Martin Seifrid,[1] Katrin R. Siefermann,[3‖] Felix P. Sturm,[3] Michele Swiggers,[2] Joachim Ullrich,[5,6§] Fabian Weise,[3¶] Petrus Zwart,[18] Christoph Bostedt,[2,22#] Oliver Gessner,[3#] Andrey F. Vilesov[1,7#]

Gomez et al.

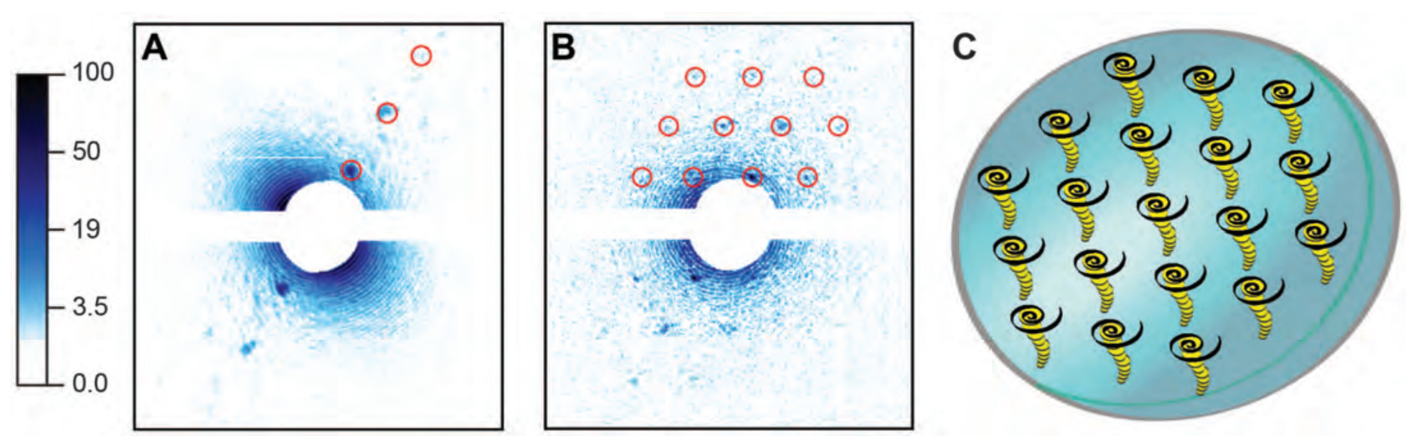
Shapes and vorticities of superfluid helium nanodroplets
Luis F. Gomez *et al.*
Science **345**, 906 (2014);
DOI: 10.1126/science.1252395



X-ray free electron laser

Fig. 1. Experimental setup. (**A**) Rotating droplets are produced by expanding He fluid into a vacuum through a 5-μm nozzle at a temperature of $T_0 \approx 5$ K and backing pressure $P_0 = 20$ bar. (**B**) Quantum vortices form upon evaporative cooling of rotating droplets to below T_λ. (**C**) Droplets are doped with Xe atoms in a ce filled with Xe gas. (**D** and **E**) X-ray diffraction images of single droplets are recorded using single FEL light pulses.

Size: 1000 nm



vortex lattice

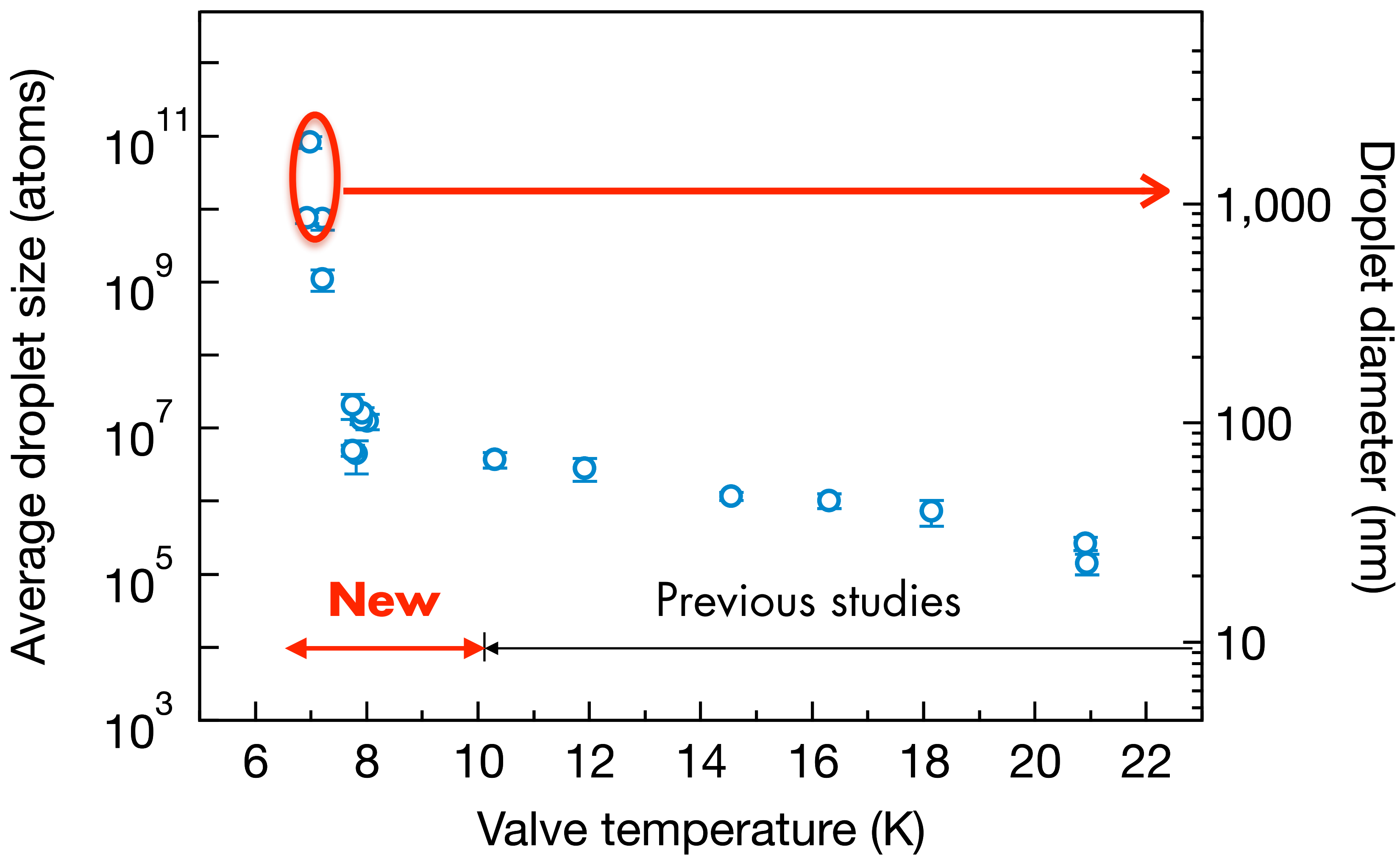
Fig. 3. He droplets doped with Xe atoms. (**A** and **B**) X-ray diffraction images of doped droplets, displayed in a logarithmic intensity scale. (**C**) Droplet and embedded Xe clusters. Images in (A) and (B) correspond to tilted and parallel alignments of the vortex axes with respect to the incident x-ray beam, respectively.

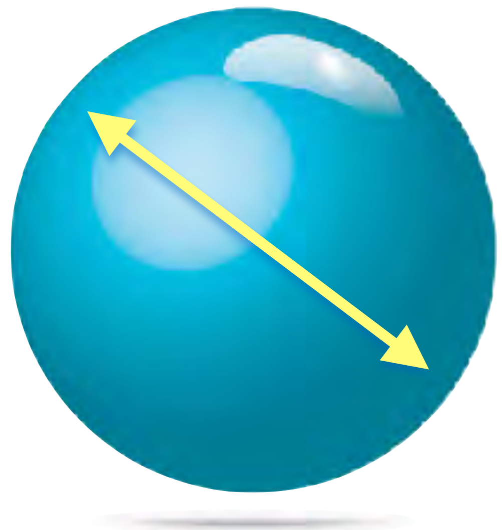
Large Helium Droplets

Kuma & Azuma, Cryogenics **88**, 78 (2017)

Size measurement by beam depletion method under collision with gas

Valve condition:
Pressure: 2 MPa
Duration: 200 μs
Repetition: 1–10 Hz





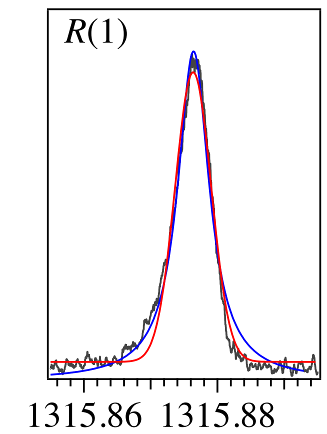
2-μm in diameter

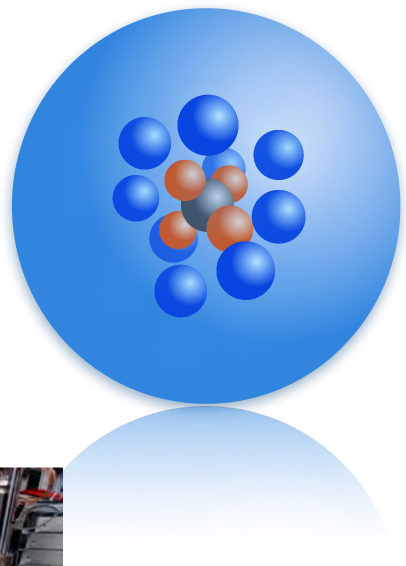
Micron-size helium droplets in a pulsed beam

SUMMARY

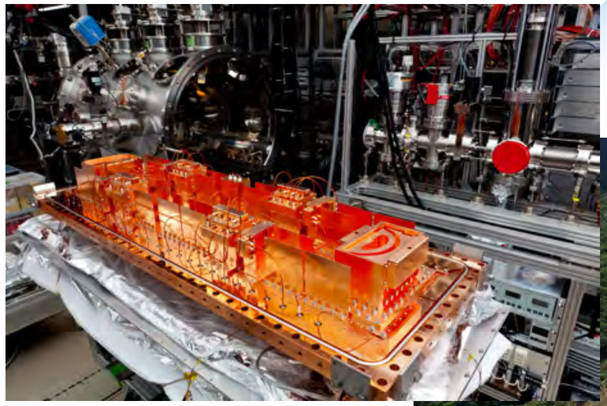
He droplets are unique nano-size superfluid systems.

Microscopic probe of superfluidity
- Rotational response
- New superfluid of molecular hydrogen





On-going efforts
- Slow dynamics with storage ring
- Direct probe of quantized vortices



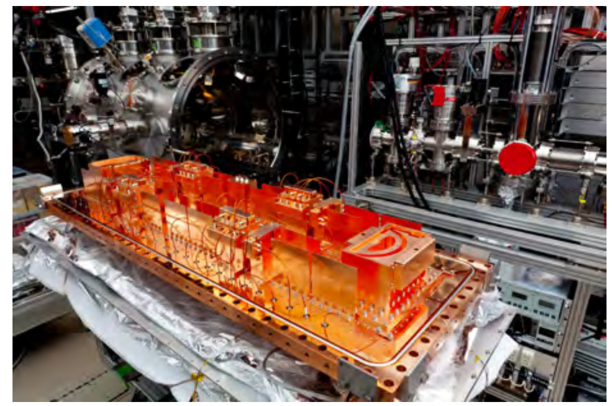
Collaborators

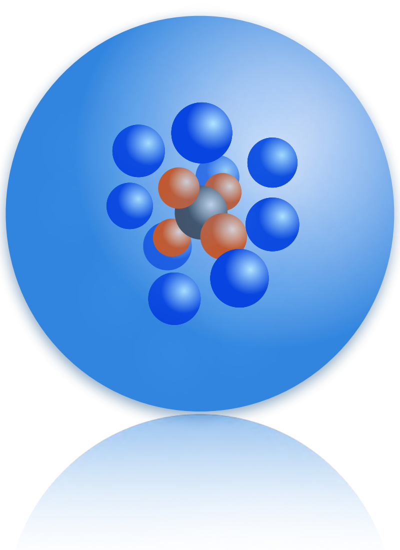
He droplet + Storage ring

Arisa Iguchi (TMU)

Hajime Tanuma (TMU)

Toshiyuki Azuma (RIKEN)





H_2 superfluidity

Hatsuki Otani (UBC)

Takamasa Momose (UBC)

Shinichi Miura (Kanazawa-U)

Quantized Vortices with X-FEL

James R. Harries (QST)

Hiroshi Iwayama (IMS)