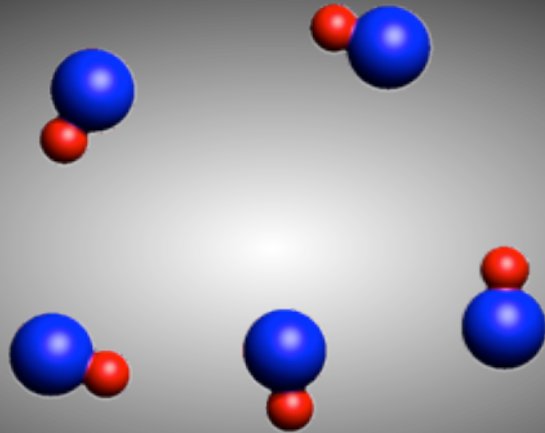


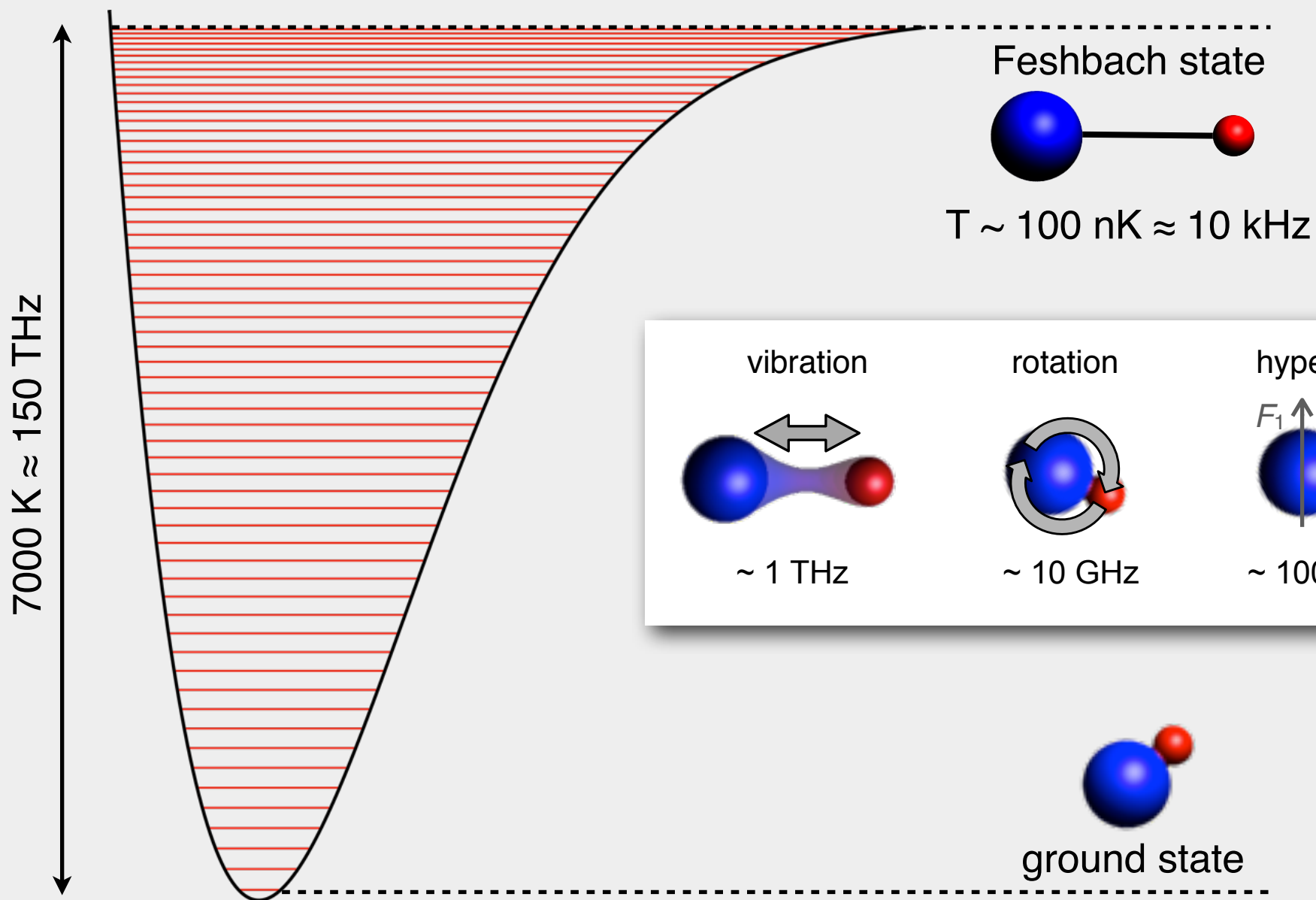
# Towards Chemically Stable Ground State Molecules of NaK



**Sebastian Will**  
Jee Woo Park  
Jennifer Schloss  
**Martin Zwierlein**



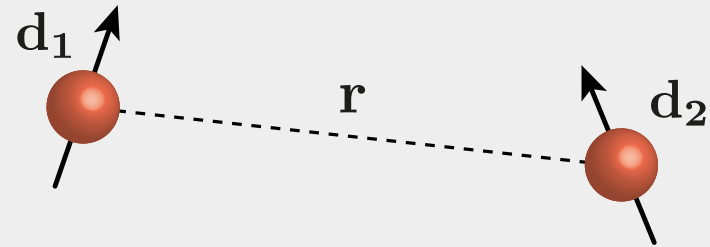
# Feshbach molecules - a far way from home...



# Why dipolar interactions?

## Dipolar interactions:

- are long-range
- are anisotropic
- can be attractive or repulsive



$$V_{dd} = \frac{1}{4\pi\epsilon_0} \frac{\mathbf{d}_1 \cdot \mathbf{d}_2 - 3(\mathbf{d}_1 \cdot \hat{\mathbf{r}})(\mathbf{d}_2 \cdot \hat{\mathbf{r}})}{r^3}$$

## Ultracold dipolar many-body systems:

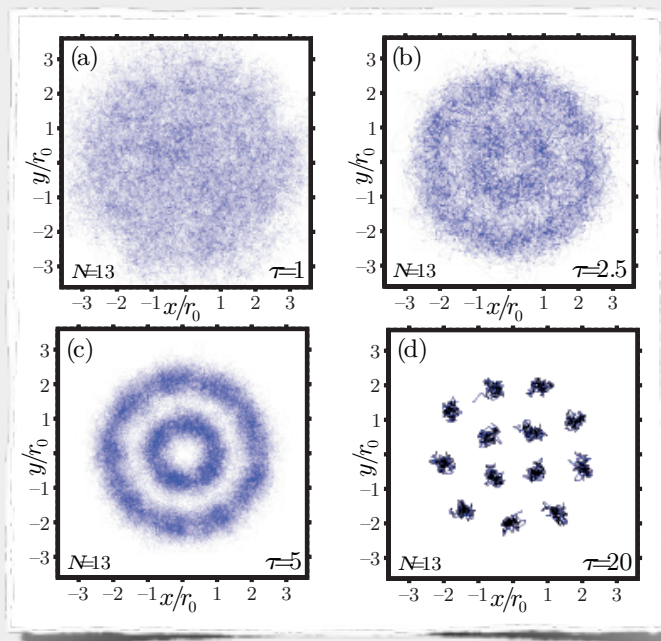
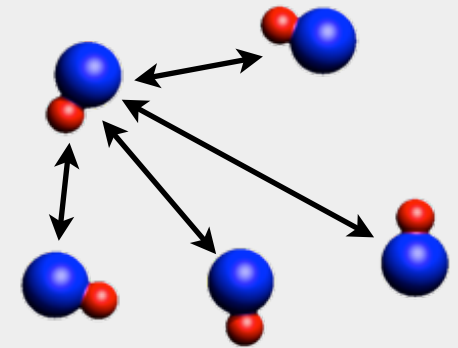
- interesting Hamiltonians
- dimensionality: 0D, 1D, 2D, 3D

see e.g. Carr *et al.*, NJP, 11 055049 (2009),  
Baranov *et al.*, Chem. Rev. 112, 5012 (2012)

# Why dipolar fermionic molecules?

Alkali **fermionic molecules** in the rovibrational **ground state**:

- clean implementation of a **dipolar quantum gas**
- direction and **strength of interactions** are **tunable**
- **dipolar interactions can dominate**



Pupillo *et al.*, PRL **104**, 223002 (2010)

Intriguing experimental possibilities:

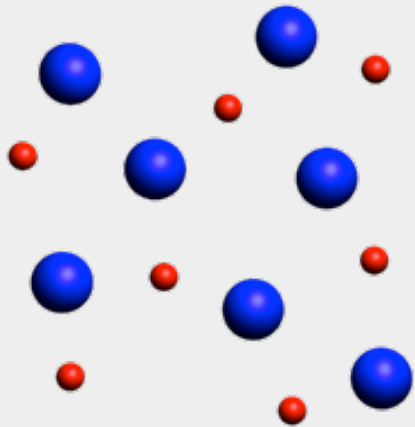
- **exotic superfluids**
- **non-trivial ordering (1D, 2D)**
- **quantum magnetism**
- **quantum information applications**
- **quantum chemistry**

$$p_x + ip_y$$

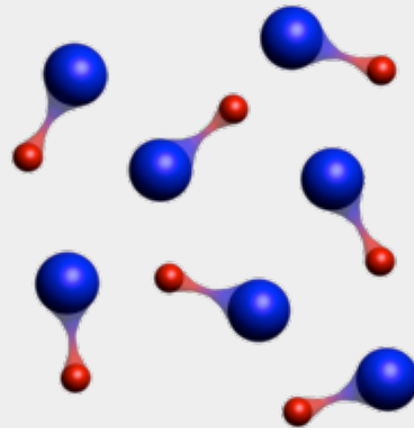
Cooper and Shlyapnikov, PRL **103**, 155302 (2009)

# Pioneering work at JILA and Innsbruck

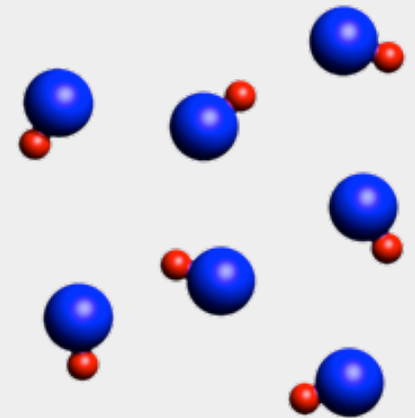
Degenerate Bose-Fermi mixture of  $^{40}\text{K}$  and  $^{87}\text{Rb}$



Near-degenerate gas of KRb Feshbach molecules



Near-degenerate gas of ground state molecules



## Feshbach association:

Binding energy: 100 kHz  
Internuclear distance: 300  $a_0$   
Dipole moment:  $5 \times 10^{-11}$  D

## Raman transfer (STIRAP)

→ 100 THz  $\sim$  0.5 eV  
→ 8  $a_0$   
→ up to 0.56 D

K.-K. Ni *et al.*, Science 322, 231 (2008)  
J. G. Danzl *et al.*, Science 321, 1062 (2008)

# Chemical stability in the ground state

fermionic bialkali molecule	chemically stable ground state
LiNa	✗
LiK	✗
LiRb	✗
LiCs	✗
NaK	✓
KRb	✗
KCs	✓

**NaK** and **KCs** are the only stable combinations!

Also **stable against trimer formation** in molecule-molecule collisions

TABLE II. Energy changes  $\Delta E_2$  for the reactions  $2XY \rightarrow X_2 + Y_2$  (in  $\text{cm}^{-1}$ ). The quantities in parentheses are uncertainties in the final digit(s).

	Na	K	Rb	Cs
Li	-328(2)	-533.9(3)	-618(200)	-415.38(2)
Na		74.3(3)	45.5(5)	236.75(20)
K			-8.7(9)	37.81(13)
Rb				29.1(1.5)

see Zuchowski and Hutson, PRA, **81**, 060703 (2010)

# A molecule with a long history...

Starting in 1924...

*The Absorption Spectra of Mixed Metallic Vapours.*

By S. BARRATT, B.A. (Oxon.), Dept. of Chemistry, University of Leeds.

Received Jan. 1, 1924.)

## The Band Spectrum of NaK

F. W. LOOMIS AND M. J. ARVIN, *University of Illinois*

(Received June 4, 1934)

The spectrum of the NaK molecule has been studied in absorption and in magnetic rotation throughout the visible and photographic infrared regions. By means of magnetic rotation the upper vibrational levels of the orange system have been followed nearly to dissociation, permitting infrared, ha

infrared, ha  
theoretically  
presumably  
magnetic rot  
simple relati

## High resolution laser spectroscopy of the $B^1\Pi-X^1\Sigma^+$ transition of $^{23}\text{Na}^{39}\text{K}$ , and the perturbation between the $B^1\Pi$ and $c^3\Sigma^+$ states

Hajime Katô, Mina Sakano, Naoki Yoshie, Masaaki Baba,<sup>a)</sup> and Kiyoshi Ishikawa  
*Department of Chemistry, Faculty of Science, Kobe University, Nada-ku, Kobe 657, Japan*

(Received 20 February 1990; accepted 2 May 1990)

Free laser polarization spectrum of the  $B^1\Pi - X^1\Sigma^+$  transition of  $^{23}\text{Na}^{39}\text{K}$  was  
the molecular constants of the  $B^1\Pi$  state of  $v = 0 \sim 16$  were determined. The  
between the  $B^1\Pi(v = 8)$  and the  $c^3\Sigma^+(v = 22)$  levels at small  $J$  were studied in

## Ground state potentials of the NaK molecule

A. Gerdes<sup>a</sup>, M. Hobein, H. Knöckel, and E. Tiemann

Institut für Quantenoptik, Welfengarten 1, 30167 Hannover, Germany

Received 9 May 2008/ Received in final form 25 June 2008  
Published online 16 July 2008 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 200

**Abstract.** We present a simultaneous analysis of the  $X^1\Sigma^+$  and the  $a^3\Sigma^+$  electronic ground  
the NaK molecule. Excitation of the  $[B^1\Pi, c^3\Sigma^+]$ -system made it possible to record fluores  
rovibrational levels of both ground states simultaneously with Fourier transform spectroscopy  
first time highly resolved energy curves, the  
for  $^{23}\text{Na}$ - $^{39}\text{K}$  were determined. Cold collision pr  
energy curves, the  
Cold collision pr  
potentials and co

## An improved potential energy curve for the ground state of NaK

I Russier-Antoine, A J Ross, M Aubert-Frécon, F Martin and P Crozet  
Laboratoire de Spectrométrie Ionique et Moléculaire (UMR 5579), Bâtiment 205, Université  
Lyon I et CNRS, Campus La Doua, 69622 Villeurbanne Cédex, France

Received 13 January 2000, in final form 17 April 2000

## The $c^3\Sigma^+$ , $b^3\Pi$ , and $a^3\Sigma^+$ states of NaK revisited

R. Ferber  
*Department of Physics, University of Latvia, Riga LV-1586, Latvia*  
E. A. Pazyuk, A. V. Stolyarov, and A. Zaitsevskii  
*Department of Chemistry, Moscow State University, Moscow, 119899, Russia*

*Warsaw University, ul. Hoza 69, 00-681 Warsaw, Poland*

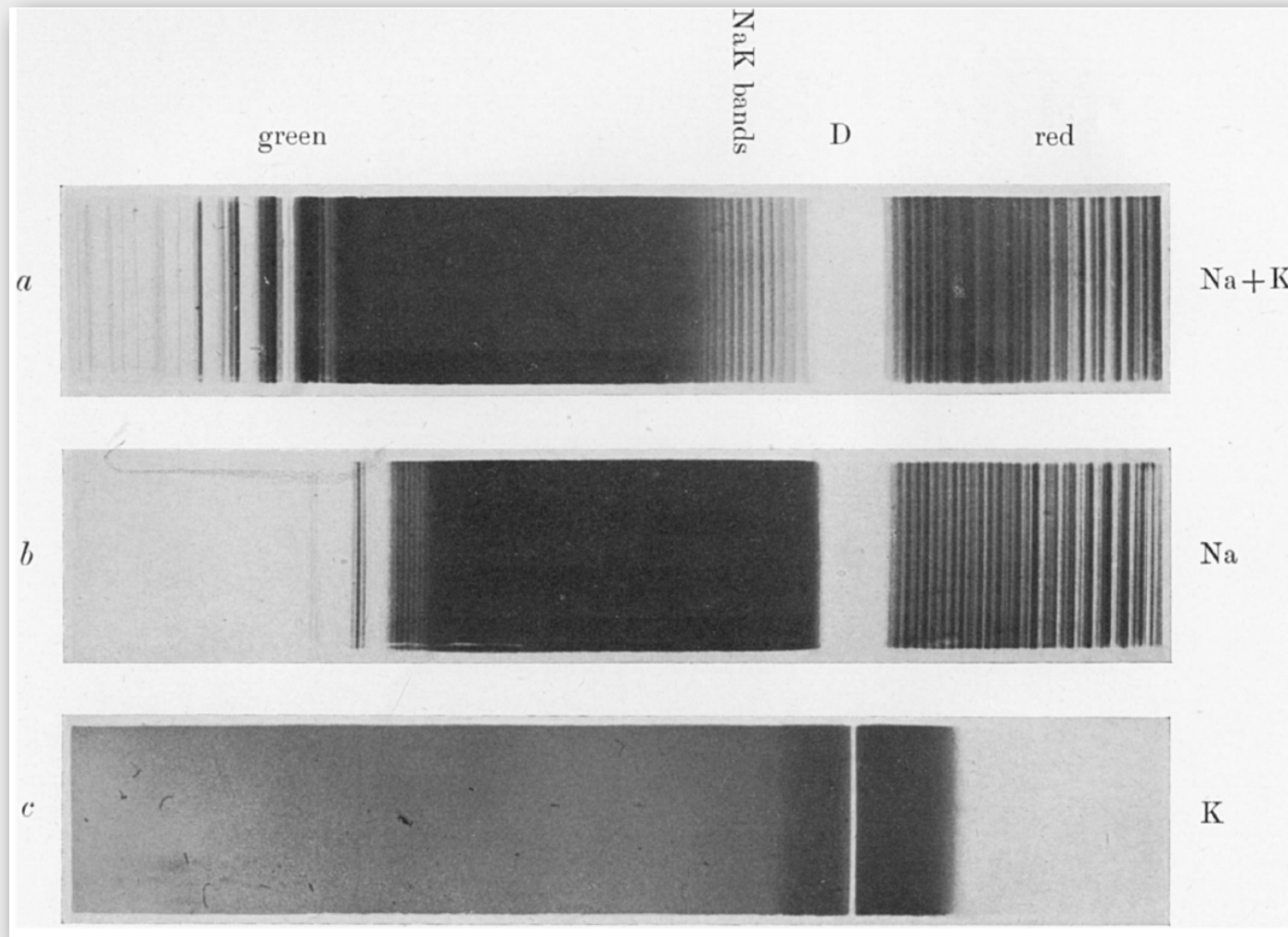
William C. Stwalley  
*Department of Chemistry, and Institute of Material Science,  
Connecticut 06269-3046*

Accepted 4 January 2000)

...and many more!

# Features of NaK

- **textbook diatomic molecule** - first spectra recorded in 1924



*“The Absorption Spectra of Mixed Metallic Vapours”,  
S. Barratt, Proc. Roy. Soc. London, Series A, 105, 221 (1924)*

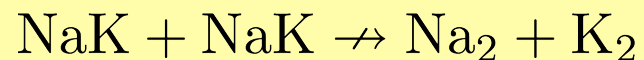


# Features of NaK

- **textbook diatomic molecule** - first spectra recorded in 1924

Barratt (1924), Walter & Barratt (1928), Ritschl & Villars (1928), Loomis & Arven (1934), Sinha (1948), Cowley (1969), Breford & Engelke (1978), Demtröder (1979), McCormack & McCafferey (1979), Ross (1985), Kato (1989),...

- **chemically stable** in its ground state



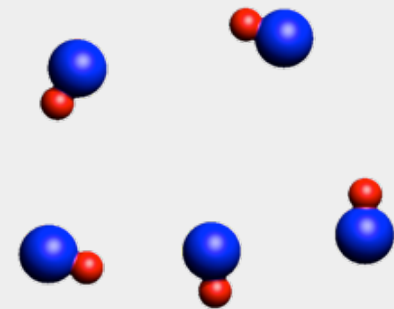
$$2 \times 5274 \text{ cm}^{-1} = 10548 \text{ cm}^{-1} > 10474 \text{ cm}^{-1} = 6022 \text{ cm}^{-1} + 4451 \text{ cm}^{-1}$$

- large induced electric dipole moment of **2.72 Debye**

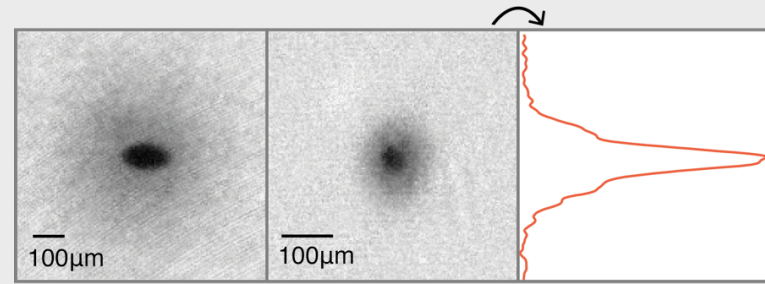
Dipolar interaction energy up to 30% of  $E_{\text{Fermi}}$

Worldwide efforts:

MIT, Hannover, Munich, Trento, Chin. Acad. of Science, HKUST

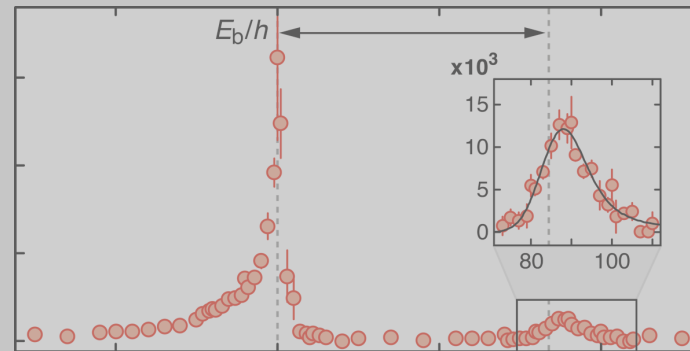


# Quantum degenerate $^{23}\text{Na}^{40}\text{K}$ Bose-Fermi mixture and its Feshbach resonances



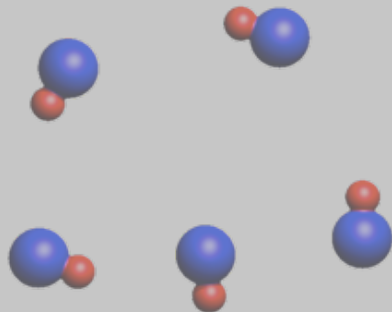
see Park *et al.*, Phys. Rev. A 85, 051602 (R) (2012)

## Ultracold fermionic Feshbach molecules of $^{23}\text{Na}^{40}\text{K}$



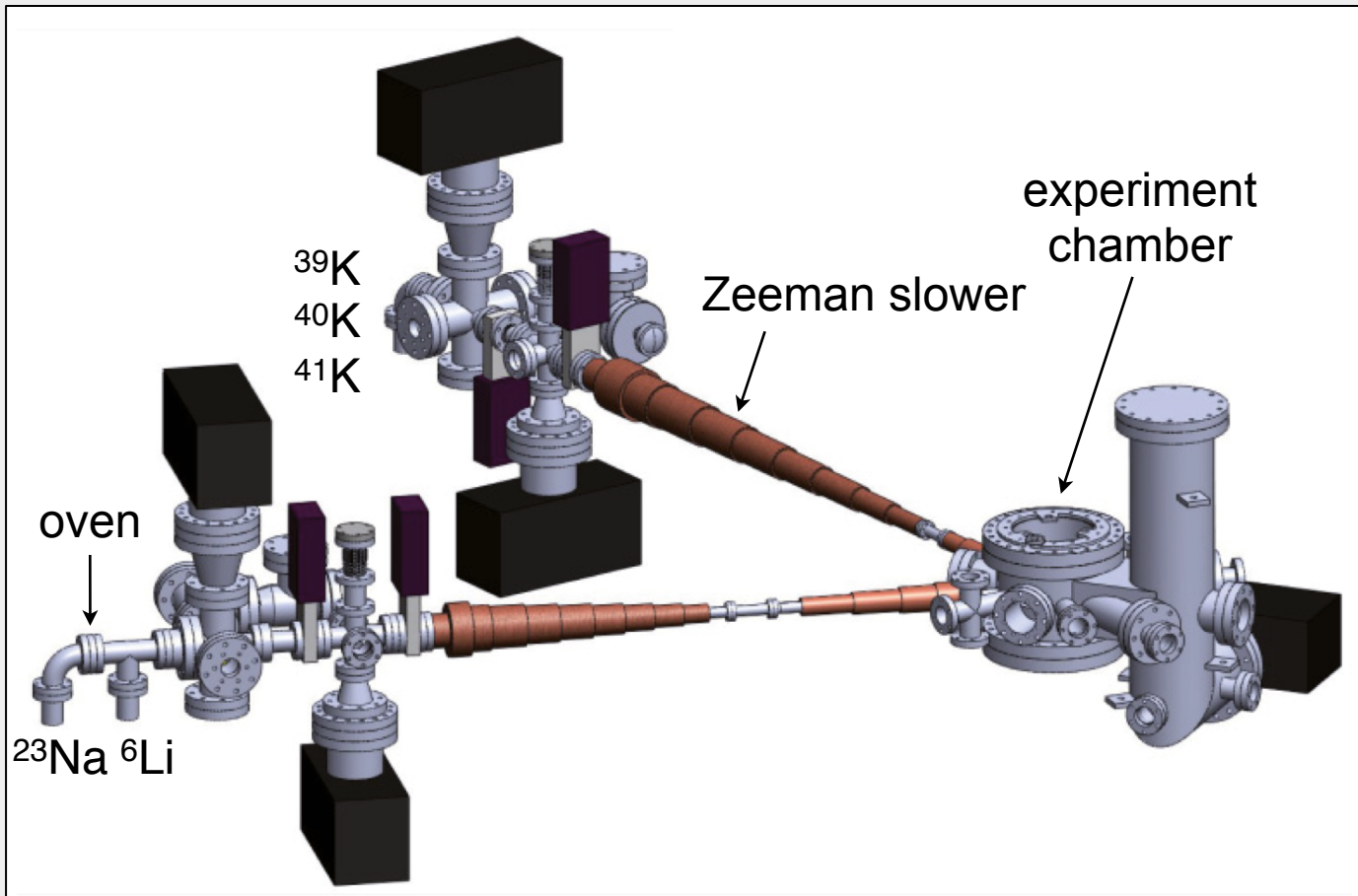
see Wu *et al.*, PRL 109, 085301 (2012)

## Towards fermionic ground state molecules of $^{23}\text{Na}^{40}\text{K}$



# Experimental setup

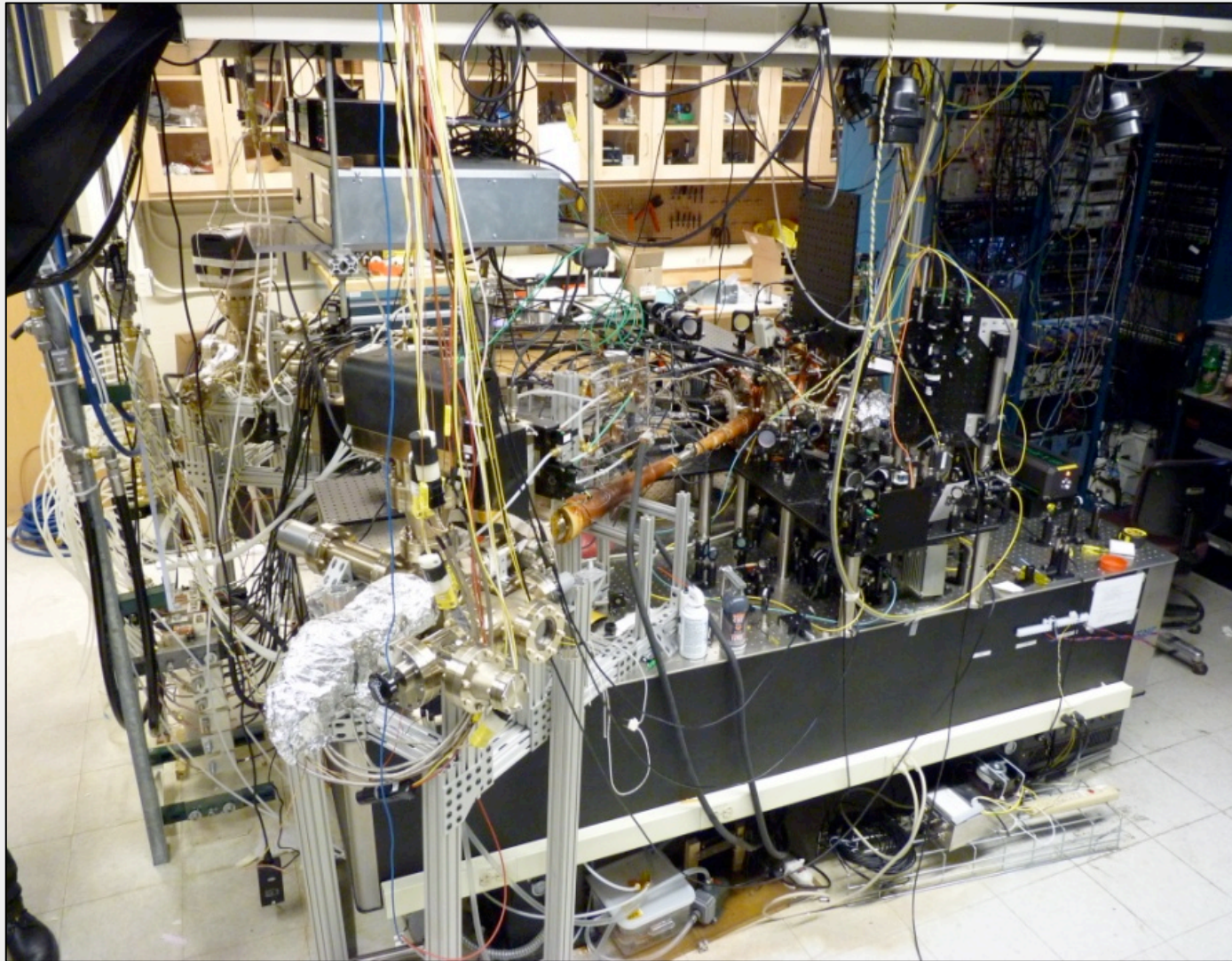
see Wu *et al.*, Phys. Rev. A **84**, 011601 (R) (2011)



Studies with  
**Bose**  
**Fermi**  
**Bose-Bose**  
**Bose-Fermi**  
**Fermi-Fermi**  
systems  
possible!

## *Experimental setup - for real*

---



**Simultaneous  
loading of all  
species possible!**

**$^{23}\text{Na}$  MOT:  $10^9$  atoms**

**$^{40}\text{K}$  MOT:  $10^7$  atoms**

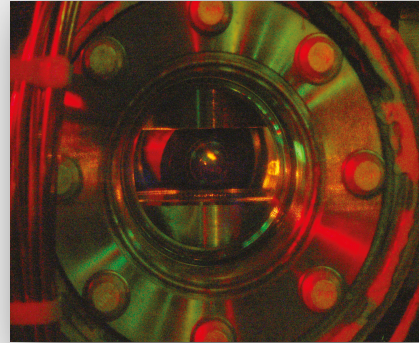
only 0.01%  
abundance

# Triple degeneracy

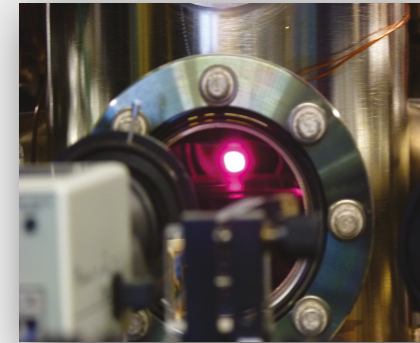
see Wu *et al.*, Phys. Rev. A **84**, 011601 (R) (2011)



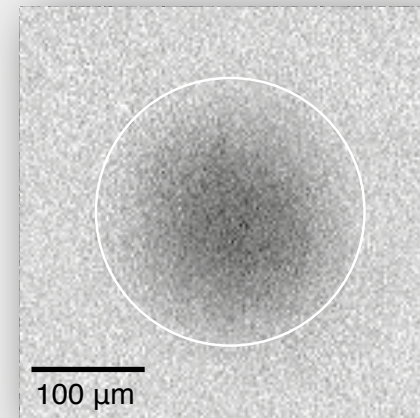
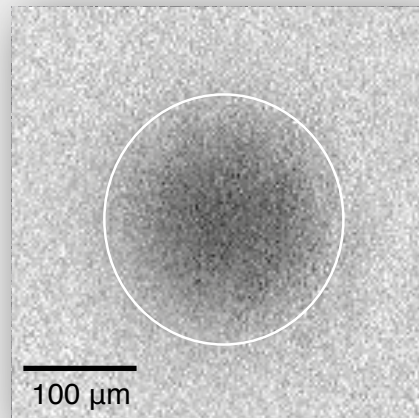
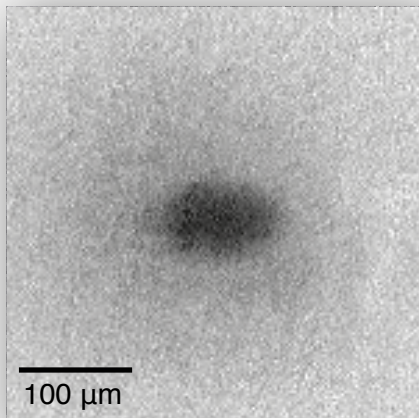
$^{41}\text{K}$



$^{40}\text{K}$

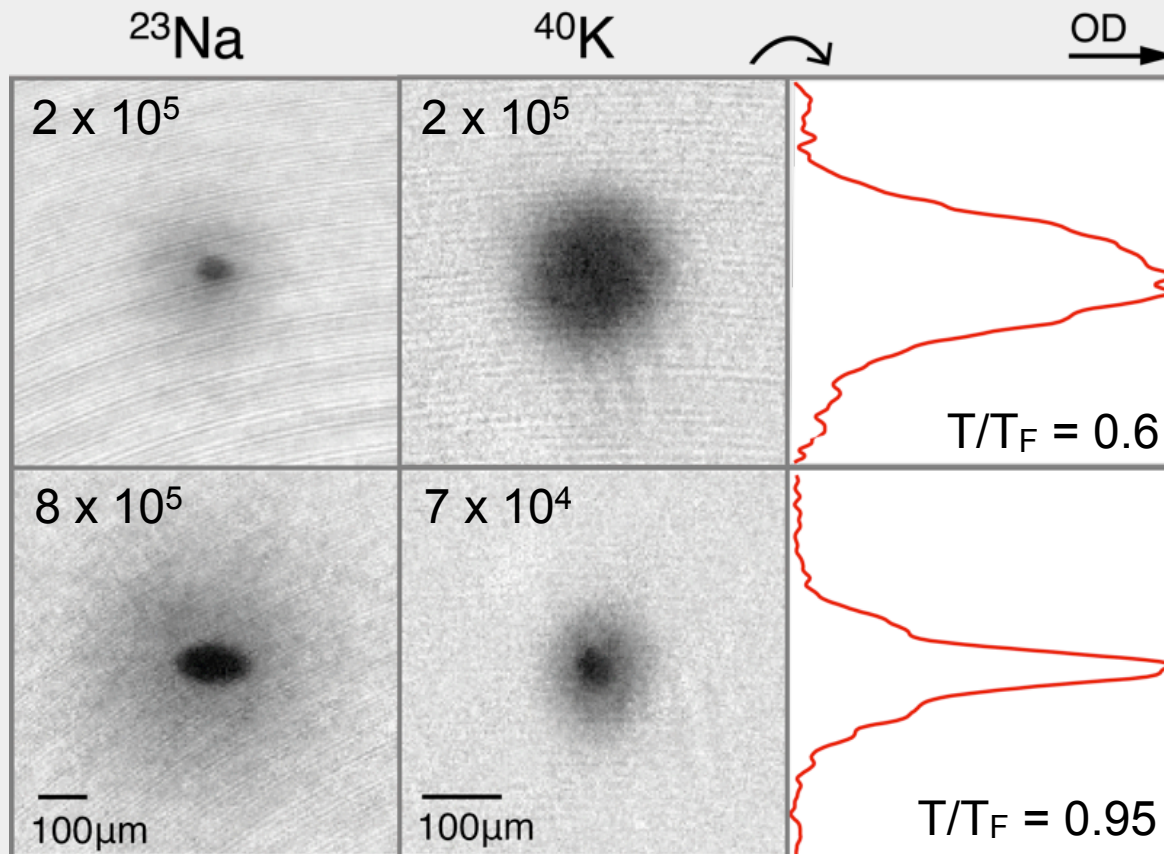


$^6\text{Li}$



# Sodium-Potassium: A new Bose-Fermi mixture

see Park et al., Phys. Rev. A **85**, 051602 (R) (2012)



**Efficient sympathetic cooling of  $^{40}\text{K}$  using  $^{23}\text{Na}$**

**Sharp increase of central density in  $^{40}\text{K}$**

**Strongly attractive interactions between Na and K!**

Mean-field energy  $k_B \times 6 \mu\text{K}$   
(for  $a_{bg} = -690 a_0$ )

**Only 4 sec RF evaporation in magnetic trap plus 4 sec in optical trap!**

# Sympathetic cooling of NaK

see Park et al., Phys. Rev. A **85**, 051602 (R) (2012)

## Cooling efficiency

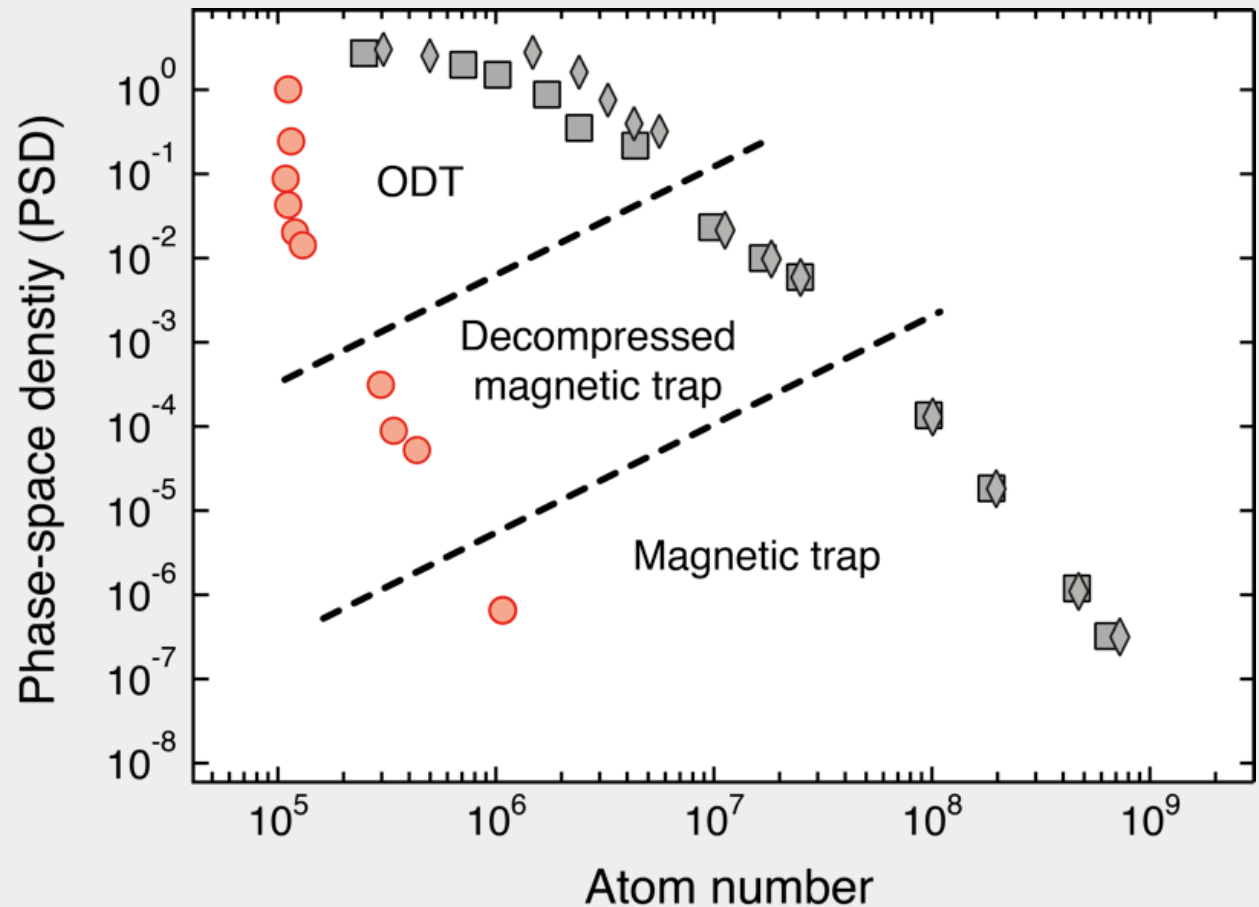
$$\Gamma = -\frac{d \ln(\text{PSD})}{d \ln(N)}$$

Magnetic trap:  $\Gamma_{\text{Na}} = 2.7$

$\Gamma_{\text{K}} = 4.6$

Optical trap:  $\Gamma_{\text{Na}} = 1$

$\Gamma_{\text{K}} = 15.3$

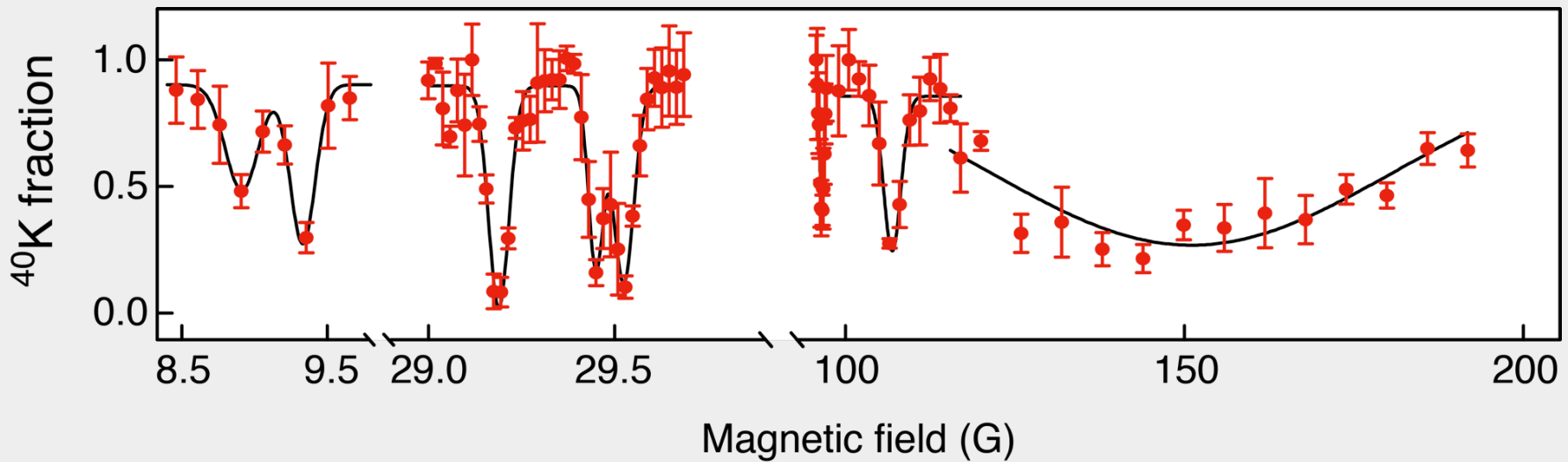


**$^{40}\text{K}$  can be efficiently cooled by  $^{23}\text{Na}$ !**

# Observation of Feshbach resonances

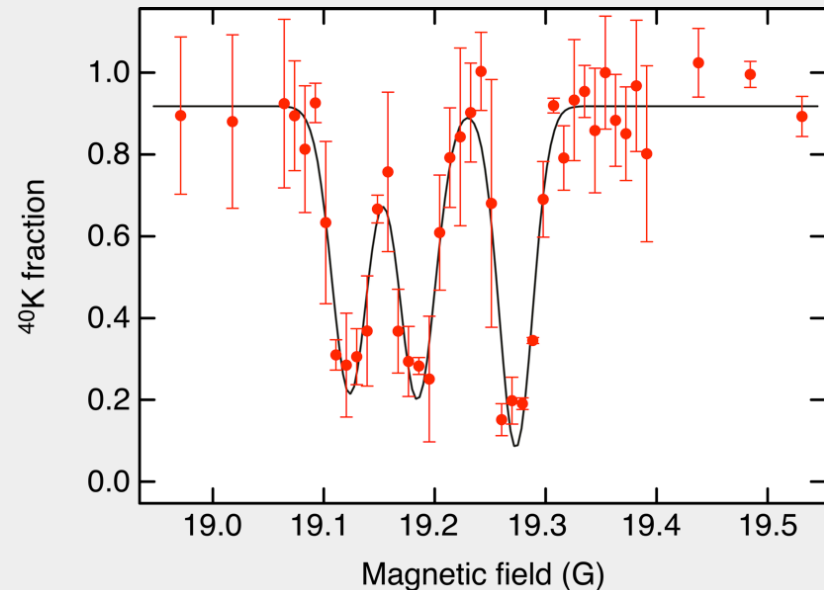
see Park *et al.*, Phys. Rev. A **85**, 051602 (R) (2012)

Feshbach loss spectroscopy for Na  $|1,+1\rangle$  and K  $|9/2,-5/2\rangle$



*p*-wave resonances:

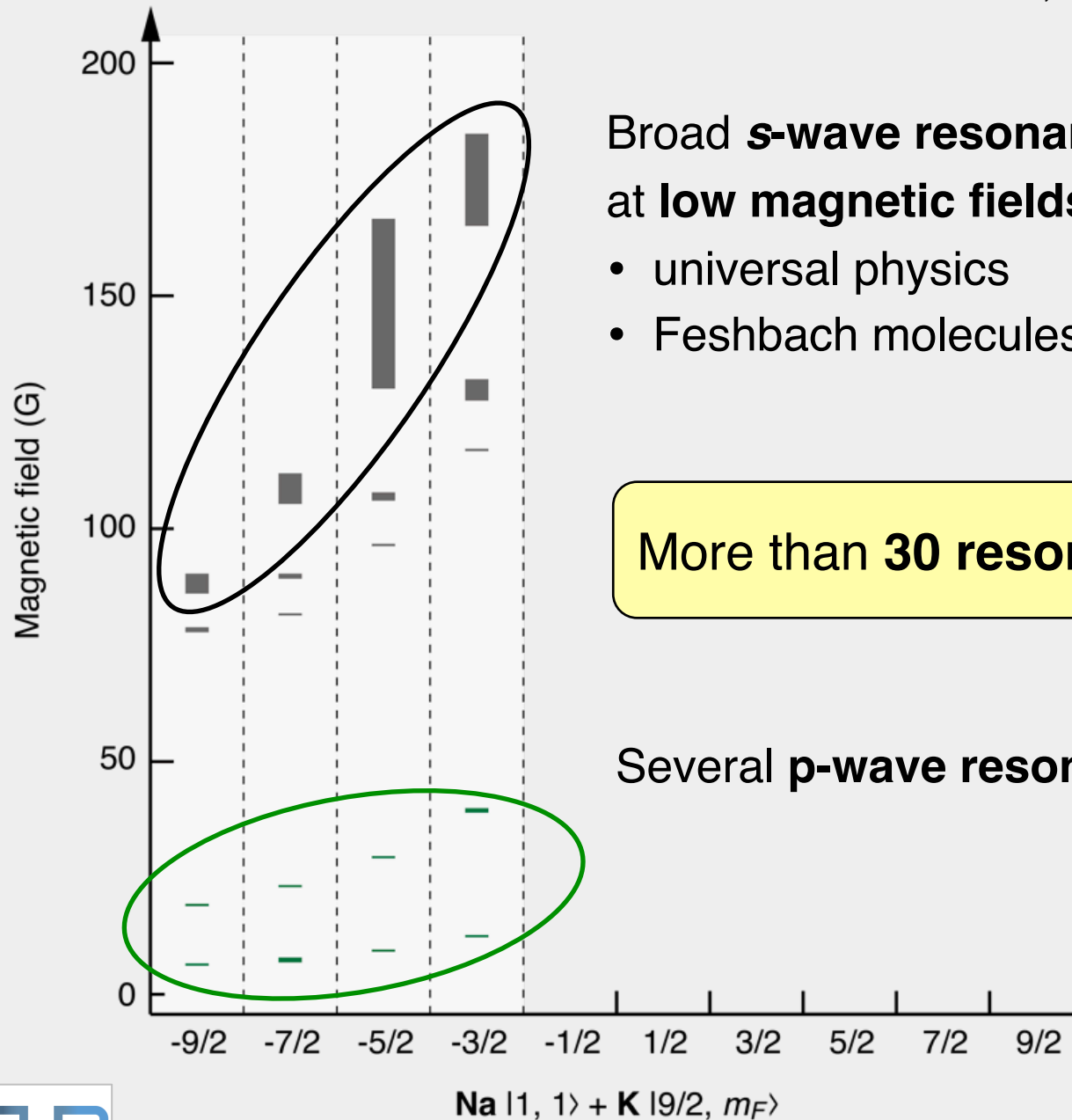
Several *p*-wave multiplets  
at low fields resolved!





# Experimentally observed resonances

see Park *et al.*, Phys. Rev. A **85**, 051602 (R) (2012)



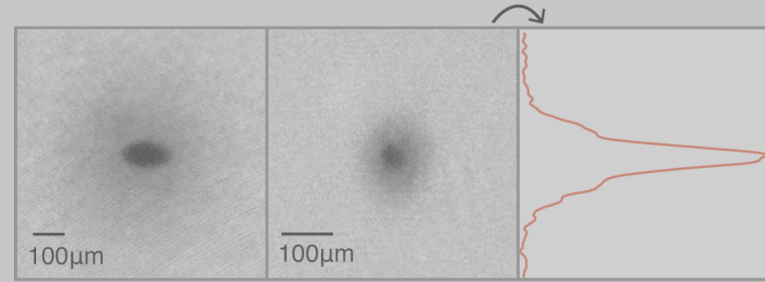
Broad **s-wave resonances** in a Bose-Fermi mixture at **low magnetic fields**:

- universal physics
- Feshbach molecules

More than **30 resonances below 200 G!**

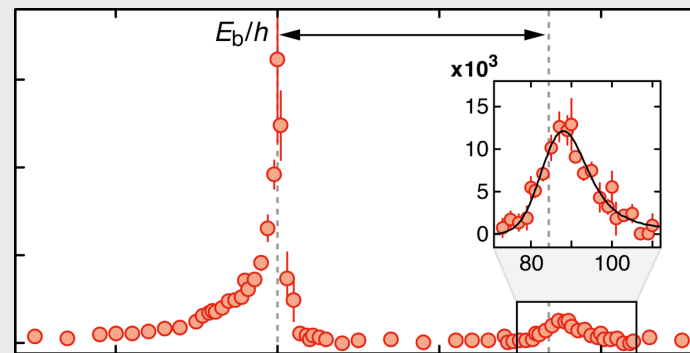
Several **p-wave resonances** at low field

# Quantum degenerate $^{23}\text{Na}^{40}\text{K}$ Bose-Fermi mixture and its Feshbach resonances



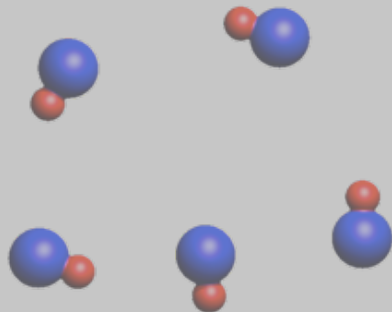
see Park *et al.*, Phys. Rev. A 85, 051602 (R) (2012)

## Ultracold fermionic Feshbach molecules of $^{23}\text{Na}^{40}\text{K}$



see Wu *et al.*, PRL 109, 085301 (2012)

## Towards fermionic ground state molecules of $^{23}\text{Na}^{40}\text{K}$

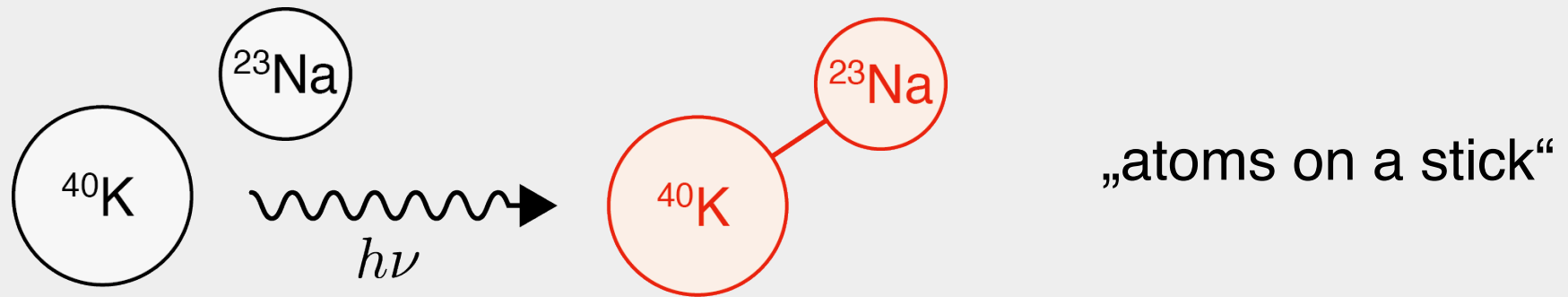


# RF spectroscopy on molecules

---

## Why RF spectroscopy?

- Yields a lot of **information about the system** ( $\leftrightarrow$  Feshbach ramp)
- For wide Feshbach resonances: **Molecular wavefunction can have large extent and good overlap with two unbound atoms.**



### Typical atom numbers:

$^{23}\text{Na}$ : 150.000 atoms

$^{40}\text{K}$ : 150.000 atoms

### T for optimized phase space overlap:

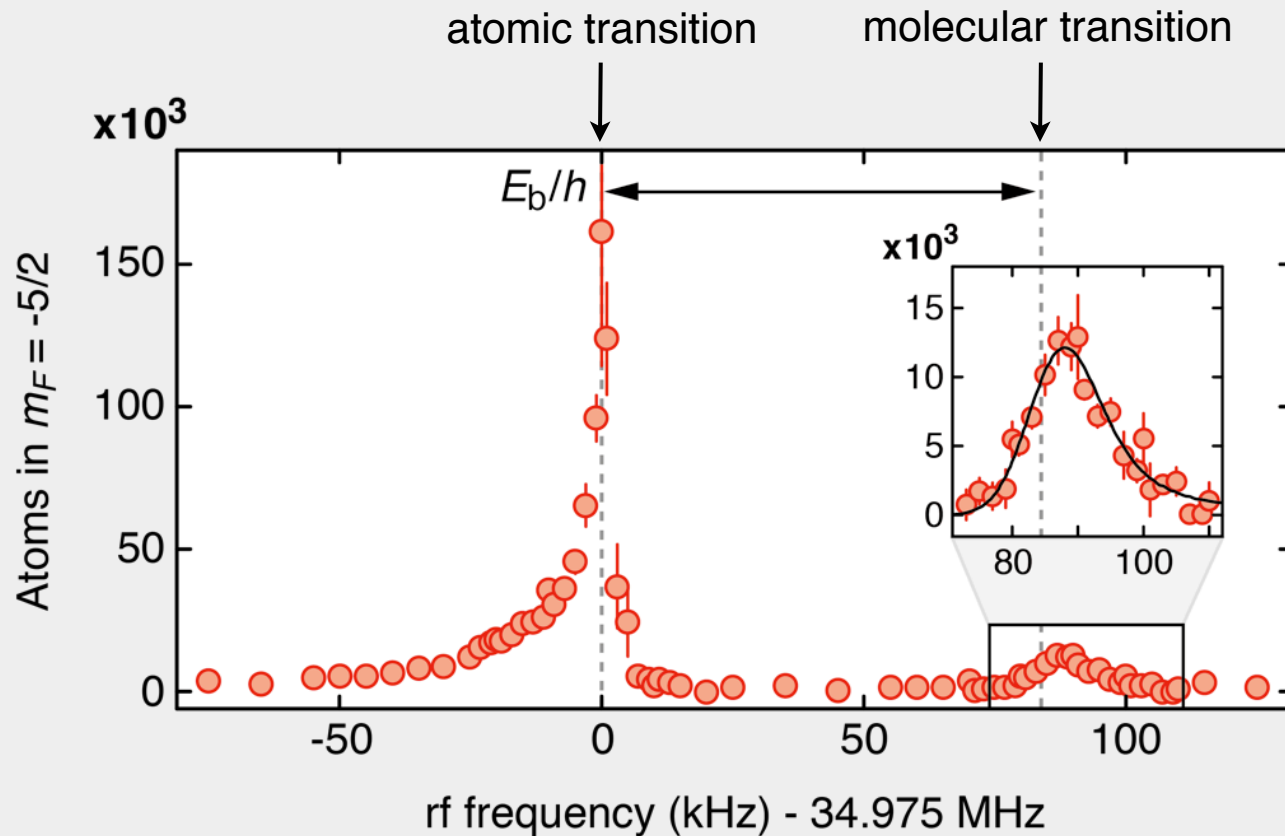
$^{23}\text{Na}$ : around  $T_C$

$^{40}\text{K}$ : corresponding to  $T/T_F = 0.4$

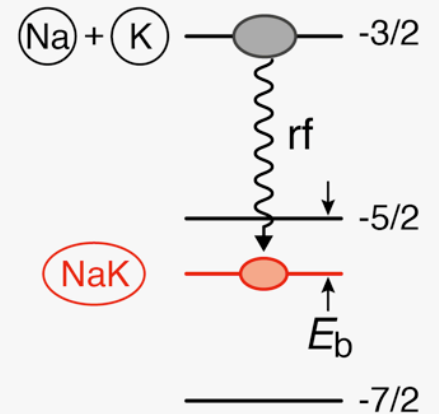
### Blackman pulse

# Association spectrum

$B = 129.4 \text{ G}$

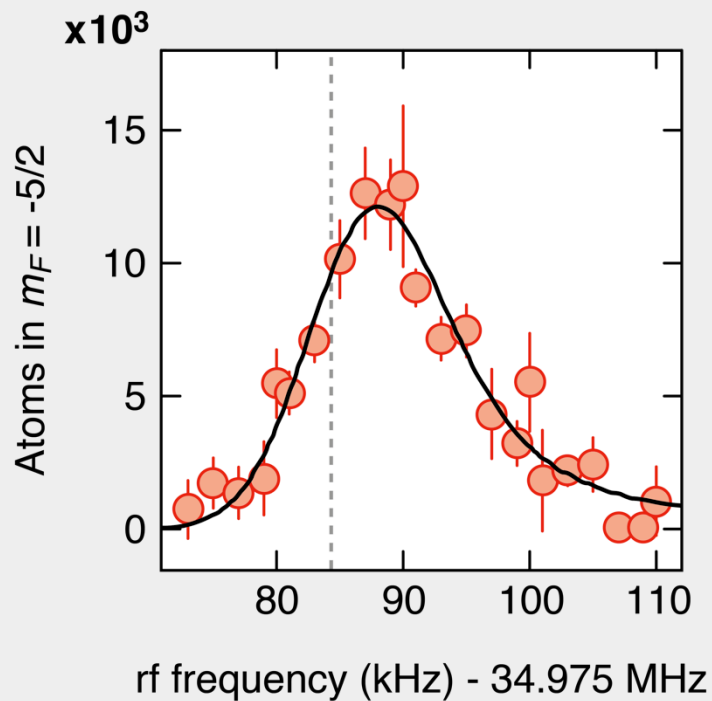


$\text{Na } |1, 1\rangle + \text{K } |9/2, m_F\rangle$



- atomic **mean-field tail** due to repulsion
- about **15%** molecule **conversion efficiency**

# Lineshape of the molecule peak



$$E_b = h \times 84(6) \text{ kHz}$$

- convoluted with rf Fourier width
- width compatible with temperature

**Probability density** for Na-K pairs  
with relative kinetic energy  $\epsilon$  :

$$p(\epsilon) = \rho(\epsilon) \lambda_M^3 \exp\left(-\frac{\mu}{M} \frac{\epsilon}{k_B T}\right)$$

$$\Gamma_{\text{mol}}(\nu) \propto \mathcal{F}(h\nu - E_b) p(h\nu - E_b)$$

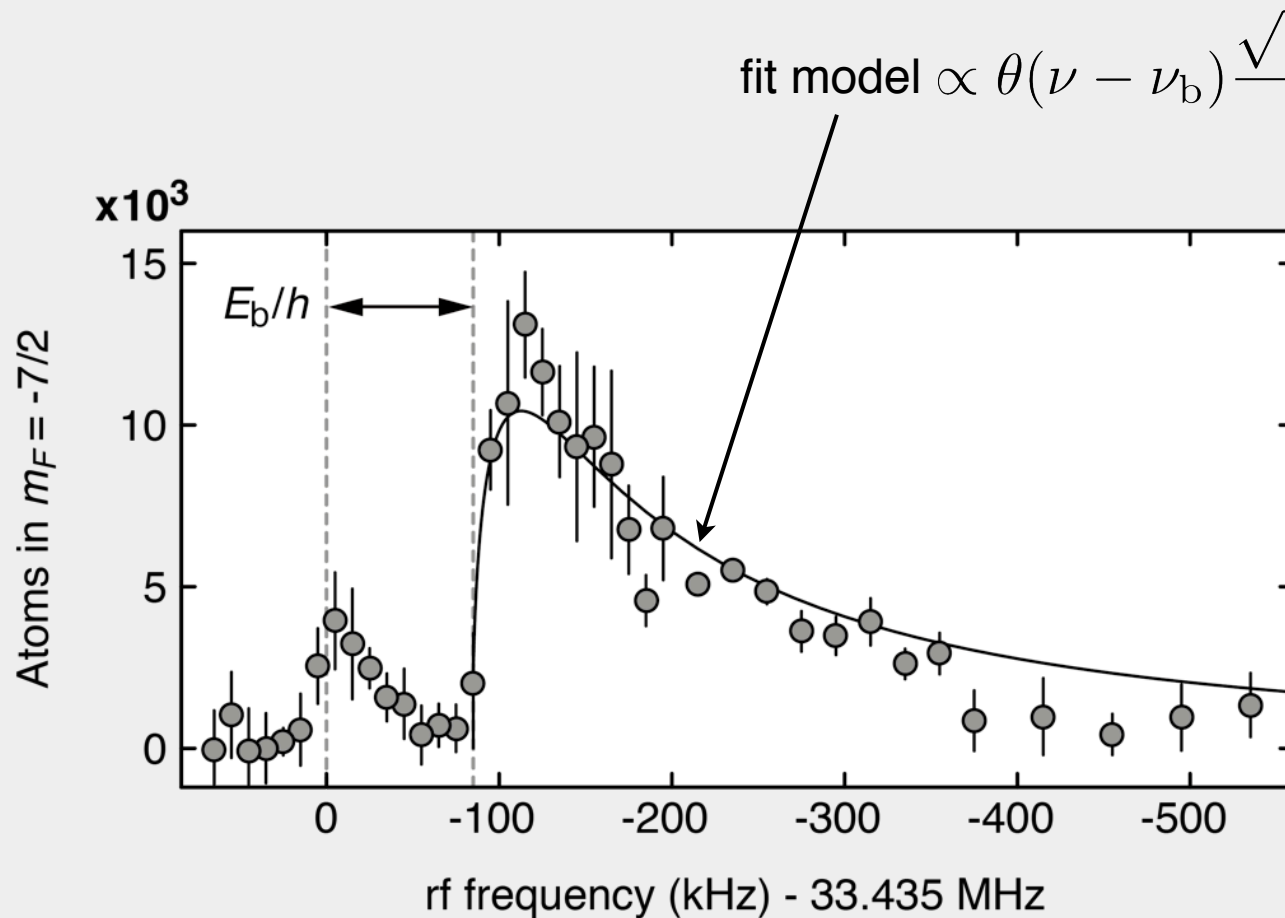
$$\mathcal{F}(\epsilon) \propto (1 + \epsilon/E_b)^{-2}$$

**Franck-Condon factor** between unbound  
Na-K pair and bound Feshbach molecule

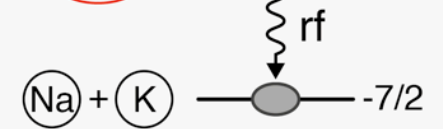
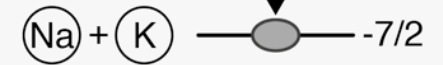
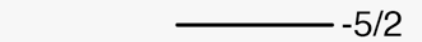
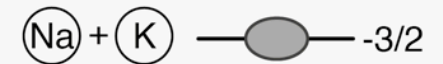
Related work: Chin and Julienne, PRA 71, 012713 (2005), Klempt *et al.*, PRA 78, 061602 (2008)

# Dissociation spectrum

$B = 129.4 \text{ G}$



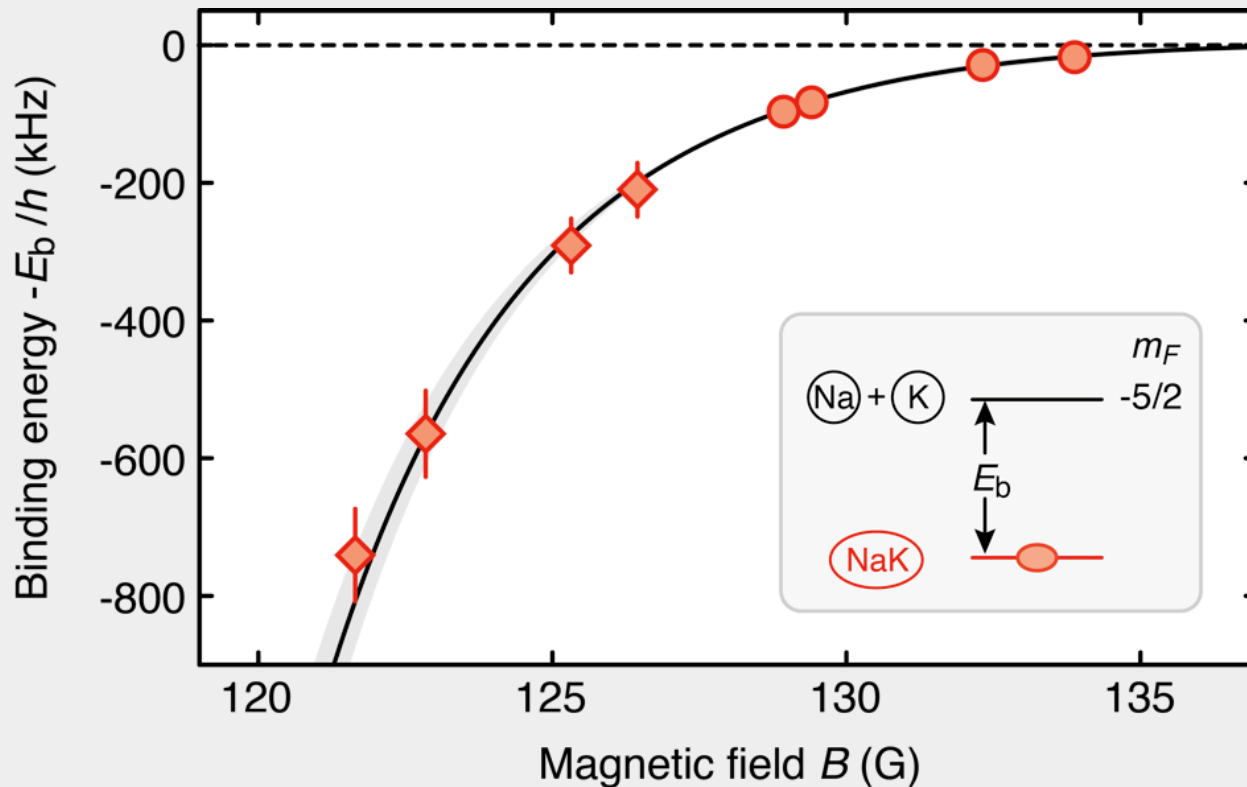
**Na**  $|1, 1\rangle + \text{K}$   $|9/2, m_F\rangle$



$E_b = h \times 85(8) \text{ kHz}$

In lifetime measurements: Dissociation ensures **exclusive detection of molecules!**

# Binding energy versus magnetic field



$$E_b \approx \frac{\hbar^2}{2\mu(a - \bar{a})^2}$$

$$a(B) \approx a_{\text{bg}} \left( 1 + \frac{\Delta B}{B - B_0} \right)$$

$$a_{\text{bg}} = -690(130) a_0$$

$$\Delta B = 29(2) \text{ G}$$

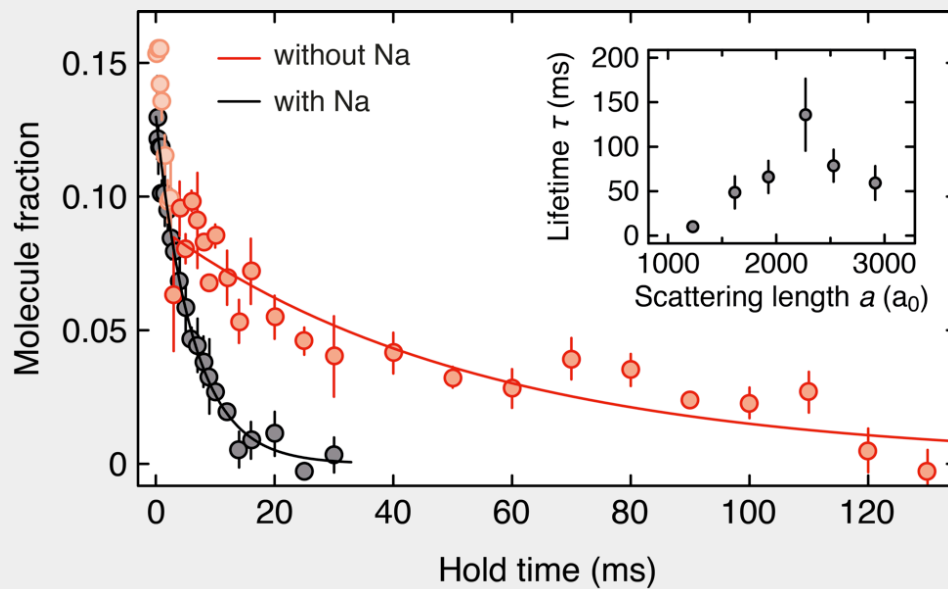
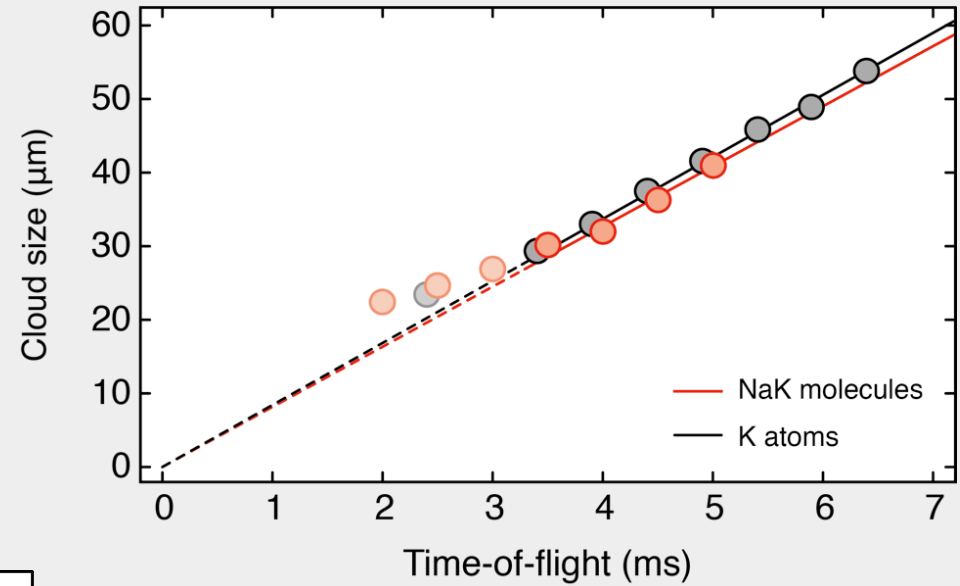
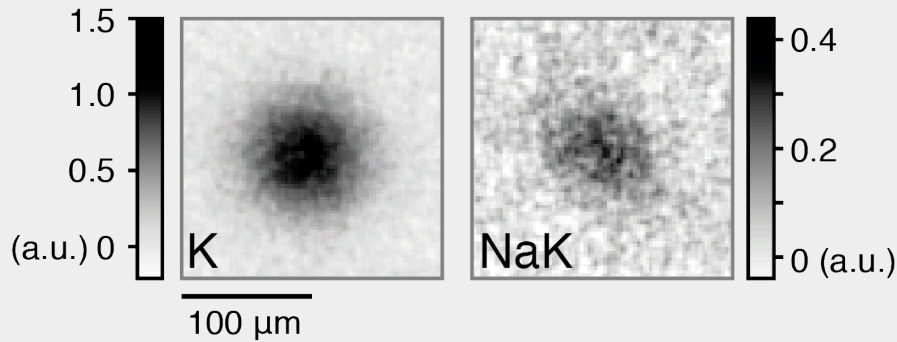
$$B_0 = 139.7(2.1) \text{ G}$$

Molecules are open-channel dominated over a large field range

“two atoms on a stick”

→ Significant singlet admixture

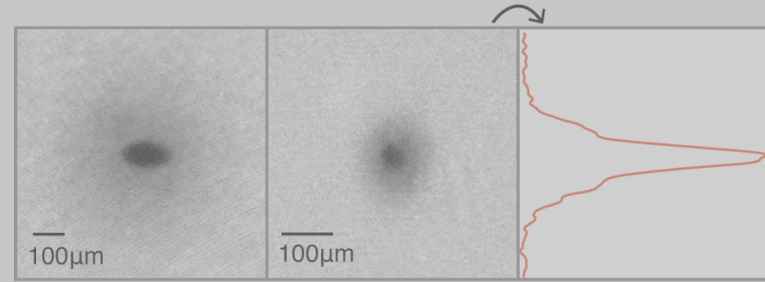
# Number, Temperature and Lifetime



- up to 20% conversion efficiency of  $^{40}\text{K}$  yields  $2 \times 10^4$  molecules
- average kinetic energy **500 nK**
- longest lifetime of  **$\sim 140$  ms** observed

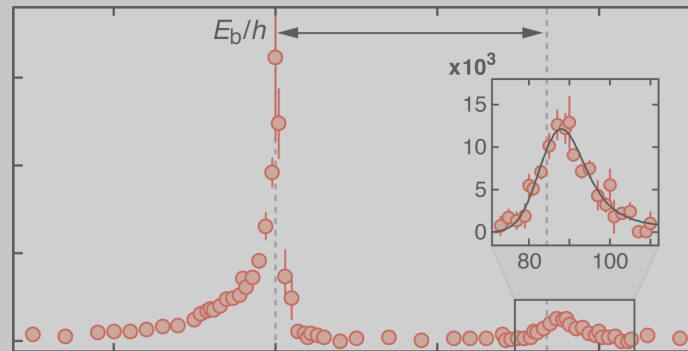


# Quantum degenerate $^{23}\text{Na}^{40}\text{K}$ Bose-Fermi mixture and its Feshbach resonances



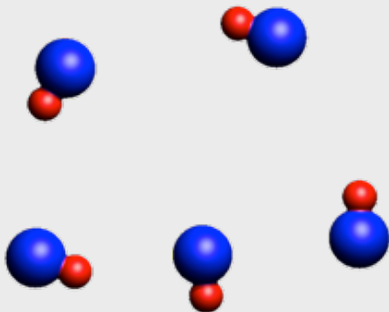
see Park *et al.*, Phys. Rev. A 85, 051602 (R) (2012)

# Ultracold fermionic Feshbach molecules of $^{23}\text{Na}^{40}\text{K}$

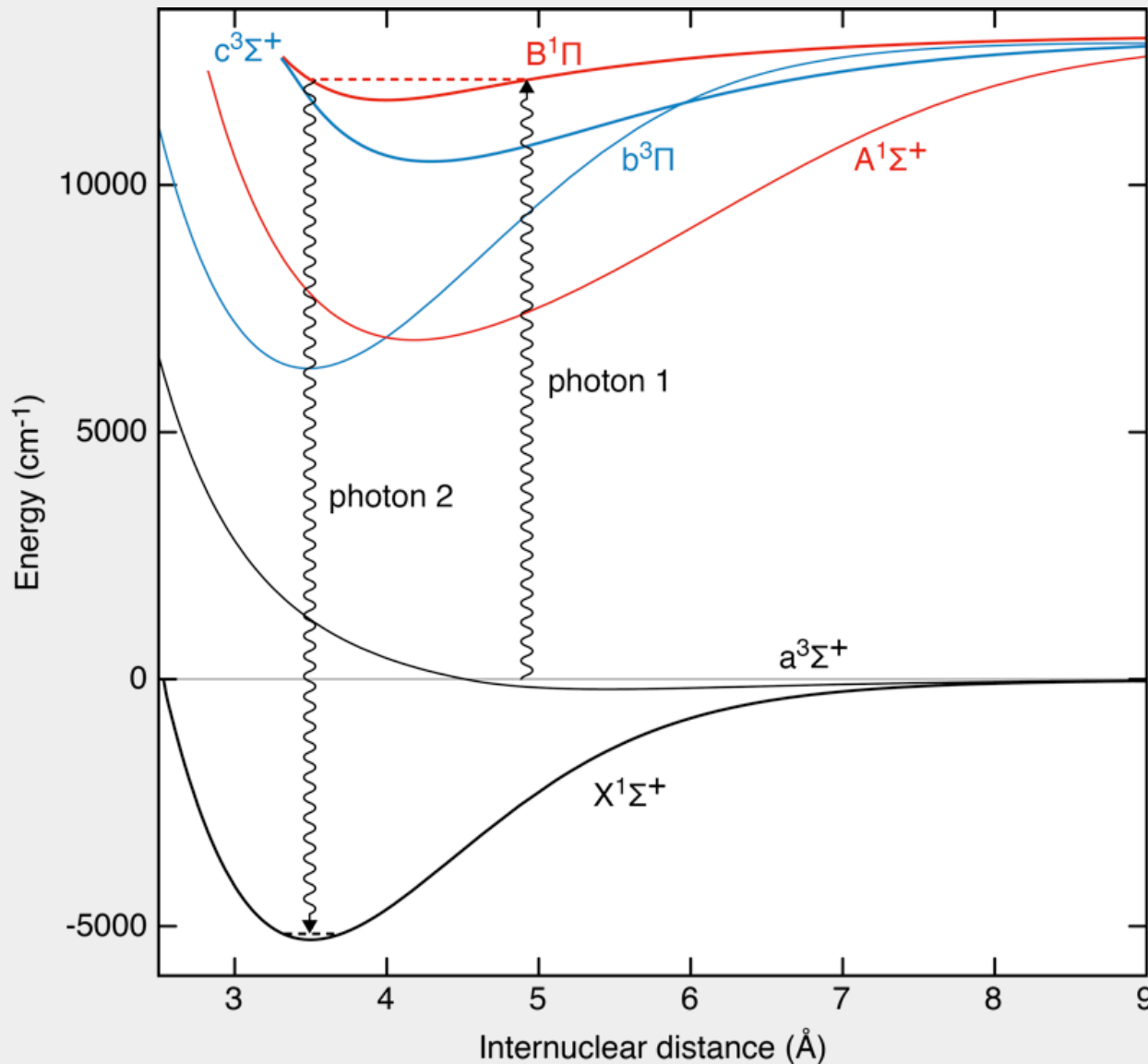


see Wu *et al.*, PRL 109, 085301 (2012)

# Towards fermionic ground state molecules of $^{23}\text{Na}^{40}\text{K}$



# Towards ground state molecules of NaK



- Similar strategy as for KRb

**STIRAP: Coherent, adiabatic transfer of loosely bound Feshbach molecules to the  $X^1\Sigma^+$  rovibrational ground state**

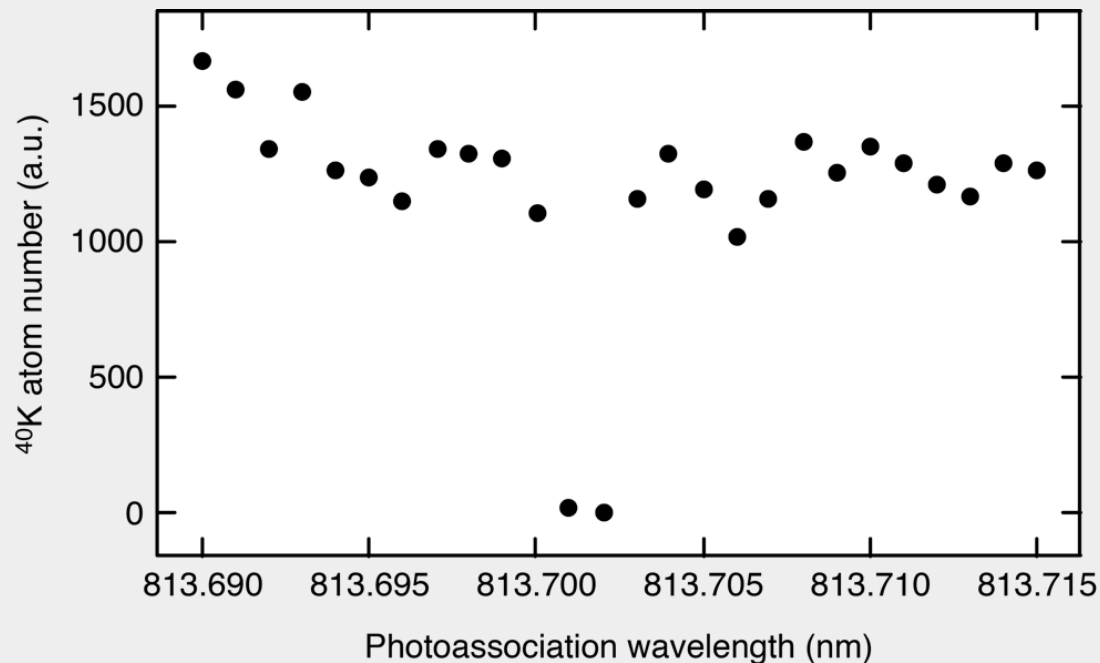
- Spin-orbit coupling mixes  $B(1)^1\Pi \leftrightarrow c(2)^3\Sigma^+$
- Must know which intermediate excited state to use → need spectroscopic information
- $^{23}\text{Na}^{39}\text{K}$  is a well studied bialkali molecule!

see e.g. Gerdes, Hobein, Knöckel, Tiemann, Eur. Phys. J. D 49, 67-73 (2008)  
for vast data on NaK, see references therein

# Photoassociation

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- based on **mass-scaled prediction**, do photoassociation spectroscopy
- for simplicity, Photoassociation performed on **atomic mixture**
- tunable Ti:Sapph laser capable of doing **20 GHz scans**

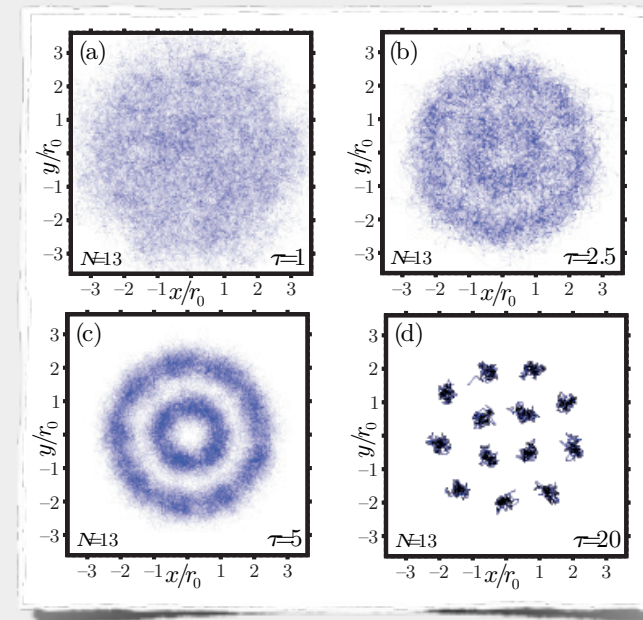
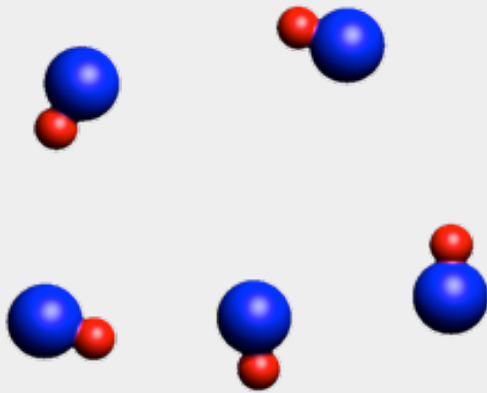


## Signature of an NaK transition:

- Simultaneous loss of Na and K
- No loss of K without presence of Na (and vice versa)

# What's next?

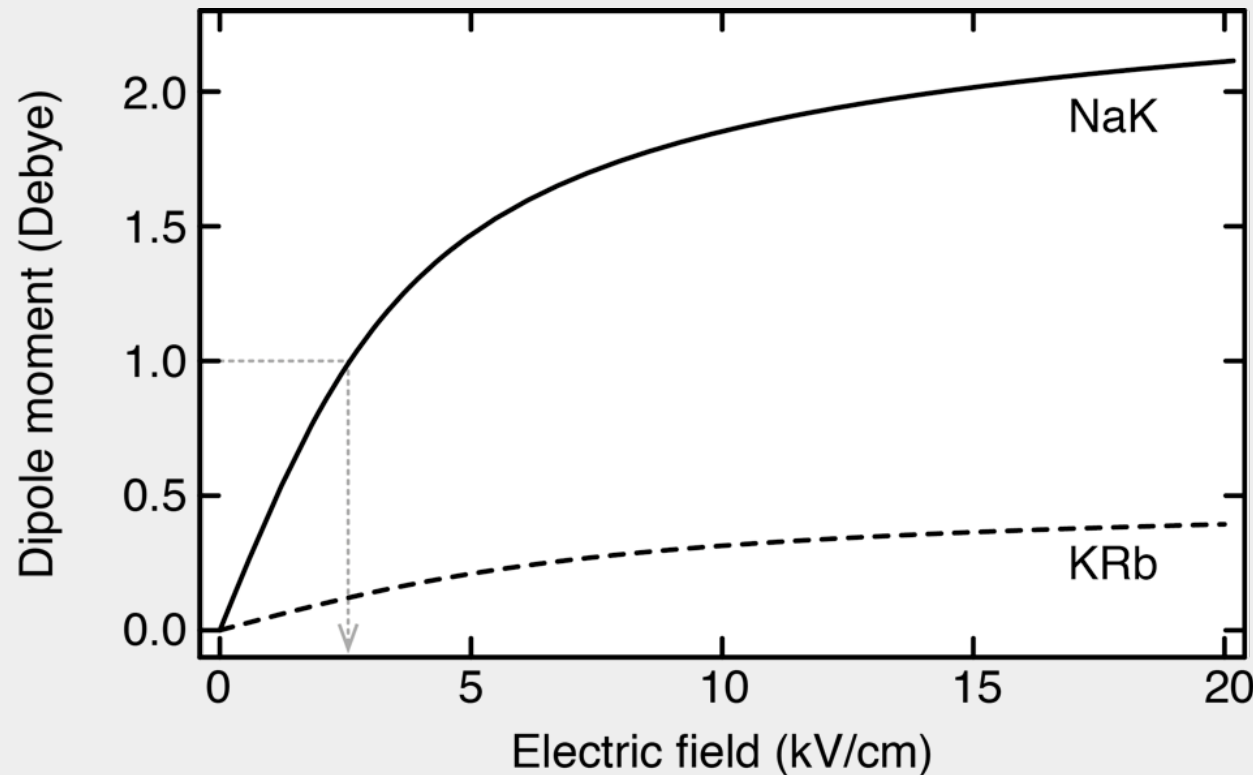
- **Photoassociation measurements and understand lines**
- Phase-coherent **Raman laser system** (lock to comb or cavity)
- **Dark-resonance/Autler-Townes** spectroscopy
- Getting ground state molecules via **STIRAP**
- **Lifetime measurement**
- **Installing electrodes**
- ...



Pupillo *et al.*, PRL **104**, 223002 (2010)

## Aligning the dipoles...

Induced dipole moment of NaK in  $v=0$ :



**Dipole moment:**

NaK:  $d = 2.72$  Debye

RbK:  $d = 0.574$  Debye

**Rotational constant:**

NaK:  $B = 2.71$  GHz

RbK:  $B = 1.11$  GHz

**Dipole moment larger than 1 Debye already at 3 kV/cm!**

# Dipole length and its consequences for NaK

Characteristic range of dipole-dipole interactions:

$$a_d = \frac{d^2 m}{4\pi\epsilon_0 \hbar^2}$$

“dipole length”

$$a_d \approx 1\mu\text{m} \text{ for 1 Debye}$$

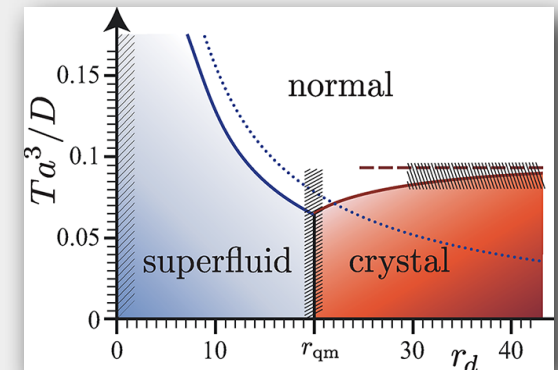
$$a_d \approx 7\mu\text{m} \text{ for 2.7 Debye}$$

Ratio of interaction energy and kinetic energy at mean interparticle spacing:

$$r_d = \frac{a_d}{a_{sp}}$$

“dipole interaction parameter”

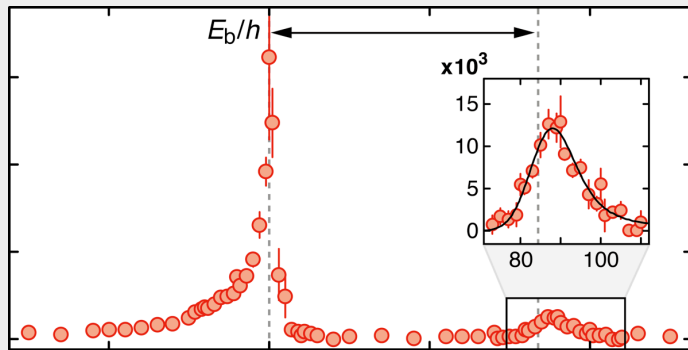
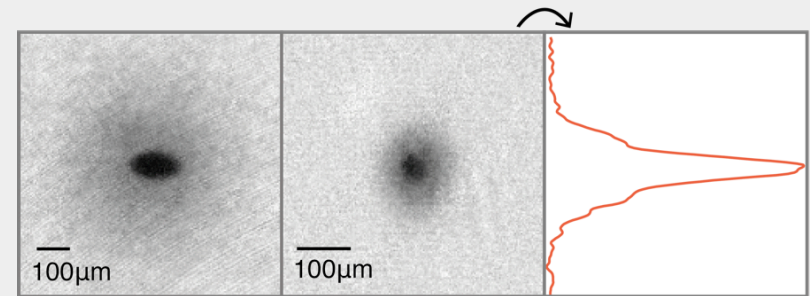
For  $n_{\text{NaK}} \approx 10^{11} \dots 10^{13} \text{ cm}^{-3} \rightarrow r_d \approx 0.4 \dots 15$



$r_d \gg 1$ : For example, **competition** between **superfluidity** and **crystallization!**

# Conclusions

- More than **30 Feshbach resonances** at low magnetic fields

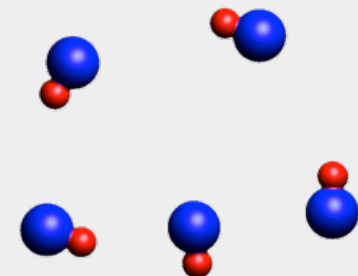


**NaK Feshbach molecules** have nice properties:

- **long lifetime** close to FBR
- **open-channel dominated** over wide range

Unique opportunity to create **fermionic NaK ground state molecules**

- **chemically stable**
- **large induced dipole moment** (2.72 Debye)
- **strongly dipolar Fermi gases** possible



# Thank you!

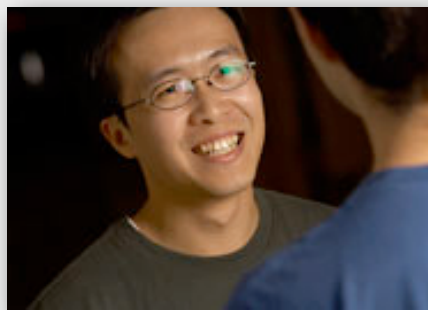
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SW

## Thanks to:

