# **Ion-atom physics**

## **Carlo Sias**

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Gressoney, 13 September 2018

## Roots..

## Understanding physics enables new technology

#### Thermodynamics





#### Optics, spectroscopy





# ..and future

## Were are we with quantum mechanics?



# ..and future

## Were are we with quantum mechanics?



# ..and future

## Were are we with quantum mechanics?



# Can we think of a composite system formed by two **coupled** quantum systems?



## Same fields, different approaches



**Coherent** matter



**Single** particles

## Same fields, different approaches



**Coherent** matter



Single particles



Many-body physics



Blatt, Roos Nat. Phys. 2012

# Same fields, different approaches



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Blatt, Roos Nat. Phys. 2012



Greiner group

# Single particle manipulation



Blatt group

# Same fields, different approaches



**Coherent** matter





Single particles



Many-body physics



Blatt, Roos Nat. Phys. 2012



Greiner group

# Single particle manipulation



Blatt group

# Hybrid quantum systems

# Quantum degenerate atoms Image: Construct of the second secon

#### A new approach to

- Ultracold collisions & quantum chemistry
- Quantum information processing and decoherence
- Quantum many-body physics with impurities
- Metrology

Review: C. Sias, M. Koehl, arXiv 1401.3188

# **Ion-neutral interactions**

An old problem in physics..

Ion-neutral collisions (1905)

#### UNE FORMULE FONDAMENTALE DE THÉORIE CINÉTIQUE;

PAR M. P. LANGEVIN.

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#### Scattering of Ions by Polarization Forces

ERICH VOGT\* AND GREGORY H. WANNIER Bell Telephone Laboratories, Murray Hill, New Jersey (Received May 19, 1954) Scattering from neutralion potential

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First atom-ion experiment (He<sup>+</sup>-Cs)

#### Exchange-Collision Technique for the rf Spectroscopy of Stored Ions\*†

F. G. MAJOR<sup>‡</sup> AND H. G. DEHMELT Department of Physics, University of Washington, Seattle, Washington 98105 (Received 6 September 1967)

# Buffer gas sympathetic cooling

### First application: sympathetic cooling of charged particles

Increase of confinment lifetime of Hg<sup>+</sup> by buffer gas cooling (Ne)

The Three-Dimensional Quadrupole Ion Trap

P. H. DAWSON and N. R. WHETTEN

Received December 16, 1968

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Received December 16, 1968

Appl. Phys. 25, 249-251 (1981)

Trapped Ion Density Distribution in the Presence of He-Buffer Gas

H. Schaaf, U. Schmeling, and G. Werth



# Cooling of Ba<sup>+</sup> by the presence of He buffer gas

Computed energy and spatial statistical properties of stored ions cooled by a buffer gas

F. Vedel, J. André, M. Vedel, and G. Brincourt

Brownian Motion of a Parametric Oscillator: A Model for Ion Confinement in Radio Frequency Traps

#### + theoretical analysis

# Ion-neutral quantum mixtures

Can we exploit our knowledge in trapping and cooling particles to study collisions at lower temperature?

## Ion-neutral quantum mixtures

# Can we exploit our knowledge in trapping and cooling particles to study collisions at lower temperature?

#### Yes!

PRL 102, 223201 (2009)

#### PHYSICAL REVIEW LETTERS

#### **Observation of Cold Collisions between Trapped Ions and Trapped Atoms**

Andrew T. Grier, Marko Cetina, Fedja Oručević, and Vladan Vuletić



FIG. 3 (color online). (a) (1,0,1) camera image of the ion crystal (blue or dark gray) and cross-section showing highly non-Gaussian shape of crystal (red or gray). (b) (0,1,1) camera image of the ion crystal. (c) Typical low-overlap setting between MOT (colored or shaded contours) and  $1/e^2$  contour of ions (white). (d) Same as (c) but for a higher overlap setting.

# Ion-neutral quantum mixtures

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PHYSICAL REVIEW LETTERS

Yes!

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#### A fast growing community

Yb-Yb<sup>+</sup> Rb-Yb<sup>+</sup>

Li-Yb<sup>+</sup>

Rb-Ba⁺ Yb-Yb⁺ Rb-Rb<sup>+</sup> Rb-Ca<sup>+</sup> Ca-Yb<sup>+</sup> Rb-Sr<sup>+</sup> Na-Na<sup>+</sup>

Rb-N<sub>2</sub>+ Li-Ca+

- Ca-BaCl<sup>+</sup>
- Rb-K<sup>+</sup>

. . . .



FIG. 3 (color online). (a) (1,0,1) camera image of the ion crystal (blue or dark gray) and cross-section showing highly non-Gaussian shape of crystal (red or gray). (b) (0,1,1) camera image of the ion crystal. (c) Typical low-overlap setting between MOT (colored or shaded contours) and  $1/e^2$  contour of ions (white). (d) Same as (c) but for a higher overlap setting.

# Outline

A brief intro of atom-ion collisions

Hybrid atom-ion experiments – state-of-the-art

Hybrid atom-ion experiments: perspectives

Atom-ion systems for improving ion clocks

# **Atom-ion interactions**



#### Longer ranged than atom-atom interactions!

<u>**R**\* ≥ 100nm</u>

For atom-atom interactions typically R\*≈1nm

# **Atom-ion interactions**



# **Atom-ion interactions**



 $\rightarrow$  independent from the ion's element!

# **Atom-ion collisions**

First theory by Langevin (classical)

$$V(r) = -\frac{C_4}{2 r^4}$$
  $C_4 = \frac{\alpha e^2}{(4\pi \epsilon_0)^2}$ 

We can define a critical impact parameter

$$b_c = \left(\frac{2 C_4}{E_c}\right)^{1/4}$$

Large deflection b < bc

Small deflection b > bc



C. Zipkes, Ph.d. Thesis, University of Cambridge

Colliding particles – Schroedinger eq.  $\left(-\frac{\hbar^2}{2\mu}\nabla^2 + V(\boldsymbol{r})\right)\Psi(\boldsymbol{r}) = \frac{\hbar^2k^2}{2\mu}\Psi(\boldsymbol{r})$ 

Solution (at large r):

In: plane wave Out: spherical wave

$$\Psi(\boldsymbol{r}) \sim e^{ikz} + f(\theta) \frac{e^{ikr}}{r}$$

Colliding particles – Schroedinger eq.

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Hypothesis: V(r) is central  $\rightarrow$  cylindrical simmetry Ok in atom-atom, atom-ion NOT in dipole-dipole!

 $\rightarrow$  Noether theorem, a quantity is conserved: angular momentum

# Expansion in partial waves

$$\Psi(\mathbf{r}) \sim e^{ikz} + f(\theta) \frac{e^{ikr}}{r}$$

Expansion in partial waves  

$$\Psi(r) \sim e^{ikz} + f(\theta) \frac{e^{ikr}}{r}$$
(from expansion of spherical  
Bessel function)  

$$e^{ikz} \sim \frac{1}{2ikr} \sum_{l=0}^{\infty} (2l+1) \left( e^{ikr} - (-1)^l e^{-ikr} \right) P_l(\cos \theta)$$

Expansion in partial waves  

$$\Psi(\mathbf{r}) \sim e^{ikz} + f(\theta) \frac{e^{ikr}}{r}$$

$$f(\theta) = \sum_{l=0}^{\infty} (2l+1) f_l P_l(\cos \theta)$$

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Expansion in partial waves  

$$\Psi(r) \sim e^{ikz} + f(\theta) = \sum_{l=0}^{\infty} (2l+1) f_l P_l(\cos \theta)$$
Legendre polynomials  

$$e^{ikz} \sim \frac{1}{2ikr} \sum_{l=0}^{\infty} (2l+1) (e^{ikr} - (-1)^l e^{-ikr}) P_l(\cos \theta)$$
Let's re-write these terms  $f_l = \frac{1}{k} e^{i\delta_l} \sin \delta_l$  and solve the S.E.  
(crunch crunch...)  

$$R_l(r) \sim \frac{1}{2ikr} \frac{1}{\sqrt{4\pi(2l+1)}} e^{2i\delta_l} e^{ikr} - (-1)^l e^{-ikr})$$
Scattered wave

#### Cross section:

$$\sigma = \frac{4\pi}{k^2} \sum_{l} (2l+1)(\sin\delta_l)^2$$

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**Summarizing**: a plane wave – written in terms of sum of spherical waves – is changed after the collision since each spherical wave of the expansion gets a **different** phase shift

#### Cross section:

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**Summarizing**: a plane wave – written in terms of sum of spherical waves – is changed after the collision since each spherical wave of the expansion gets a **different** phase shift

<u>Important</u>: energy conservation! The only effect of collisions are phase shifts that modify the «envelope» of the wave but not its amplitude!

We solve the Schroedinger equation! (radial part)



Inguscio, Fallani «Atomic Physics», Oxford University Press

How do we calculate the phase shifts?

We solve the Schroedinger equation! (radial part)



Inguscio, Fallani «Atomic Physics», Oxford University Press

# **Atom-ion collisions**

## Large deflection b < bc

# Collisions from the hard wall





# **Atom-ion collisions**

## Large deflection b < bc

# Collisions from the hard wall





Small deflection b > bc

# Collisions from the centrifugal barrier


#### Large deflection b < bc

# Collisions from the hard wall



#### Small deflection b > bc

# Collisions from the centrifugal barrier











**Cross section** 

$$\sigma_{tot} = \pi \left( 1 + \frac{\pi}{16} \right) \left( \frac{\mu C_4^2}{\hbar^2} \right)^2 E_c^{-1/3}$$

# Langevin cross section





R. Côté, A. Dalgarno, PRA 2000

$$\sigma_{tot} = \pi \left( 1 + \frac{\pi}{16} \right) \left( \frac{\mu C_4^2}{\hbar^2} \right)^2 E_c^{-1/3}$$

Langevin cross section

$$\sigma_L = \pi \sqrt{2C_4} E_c^{-1/2}$$

Scattering rate:

$$\gamma = n \sigma_L v$$

$$\gamma_{tot} = n \, \sigma_{tot} v \propto E_c^{1/6}$$

 $\gamma_L = n \sigma_L v = cost.$ 



R. Côté, A. Dalgarno, PRA 2000

Cross section 
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10<sup>9</sup>



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#### Paul trap + Atom trap

(Magneto-optical trap, magnetic trap, optical trap..)

#### Paul trap + Atom trap

Overlap Calculation Camera 1 Camera 2 Yb Ions Timm Ca Atoms CEM Cooling Lasers MOT Beams Ca and Yb + clouds

Hudson group, UCLA

(Magneto-optical trap, magnetic trap, optical trap..)

#### Paul trap + Atom trap



Hudson group, UCLA

(Magneto-optical trap, magnetic trap, optical trap..)



Ozeri group, Weizmann

#### Paul trap + Atom trap



Hudson group, UCLA

PROs: high repetition rate
large atom number
CONs: relatively high temperature,
poor control of internal state

(Magneto-optical trap, magnetic trap, optical trap..)



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#### Paul trap + Atom trap



Hudson group, UCLA

PROs: high repetition rate
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CONs: relatively high temperature,
poor control of internal state

(Magneto-optical trap, magnetic trap, optical trap..)



Ozeri group, Weizmann

PROs: lower temperature, coherence CONs: slower repetition rate, high collisional rate

### A hybrid apparatus





### **Elastic** collisions: sympathetic cooling

Bose-Einstein condensate

 $T \approx 100 \text{ nK}$ 

<u>Goal: continuous cooling</u> of an ion-based quantum <u>hardware</u>

lon trap (level spacing ≈ 5 mK)

## Elastic collisions: sympathetic cooling

#### Bose-Einstein condensate

 $T \approx 100 \text{ nK}$ 

lon trap (level spacing ≈ 5 mK) Goal: continuous cooling of an ion-based quantum hardware



C. Zipkes, et al. Nature 2010; C. Zipkes et al. PRL 2010; S. Schmid et al. PRL 2010

## **Elastic** collisions: sympathetic cooling



C. Zipkes, et al. Nature 2010; C. Zipkes et al. PRL 2010; S. Schmid et al. PRL 2010

#### Possible effects of inelastic collisions: (example: Rb-Yb<sup>+</sup> mixture)

- $Yb^+ + Rb \rightarrow Yb + Rb^+$  Charge Exch
- $Yb^+ + Rb \rightarrow (YbRb)^+$
- $(Yb^+)^* + Rb \rightarrow Yb^+ Rb$
- Charge Exchange
  - Molecular association
- Collisional quenching

L. Ratschbacher et al. Nat. Phys. 2011, F.H.J. Hall et al. PRL (2011), L. Ratschbacher et al. PRL 2012

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- $(Yb^+)^* + Rb \rightarrow Yb^+ Rb$
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<u>Problems</u>: 1. New ion invisible to spectroscopy
 2. Non-radiative decays cause trap losses
 3. Heating mechanism

L. Ratschbacher et al. Nat. Phys. 2011, F.H.J. Hall et al. PRL (2011), L. Ratschbacher et al. PRL 2012

**Detection of inelastic processes** 

# Creation of a dark ion







Not enough to ascertain what happened

#### **Detection of inelastic processes**



#### Not enough to ascertain what happened

We can perform mass spectrometry of a dark ion by modulating e.g. the cooling laser of an ancillary ion



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Reaction rate independent from collisional energy: Langevin!



## **Controlling chemical reactions**

everything depends on the ion-neutral pair

Charge exchange rate	<b>Rb-Yb<sup>+</sup></b> (Cambridge) $\sim 10^{-5} \gamma_L$	<b>Rb-Sr</b> <sup>+</sup> (Weizmann) $\sim 2 \times 10^{-5} \gamma_L$	<b>Rb-Ca</b> <sup>+</sup> (Basel) $< 10^{-3} \gamma_L$
Creation of molecules			

## **Controlling chemical reactions**

# **everything** depends on the ion-neutral pair ... and on the state

#### Table 1 | Measured proportionality constant $\epsilon$ and branching ratios.

	<sup>2</sup> S <sub>1/2</sub>	<sup>2</sup> D <sub>3/2</sub>	<sup>2</sup> F <sub>7/2</sub>	<sup>2</sup> P <sub>1/2</sub>
$\epsilon$	$10^{-5\pm0.3}$	$1.0 \pm 0.2$	$0.018 \pm 0.004$	$0.1 \pm 0.2$
Charged particle lost	65%	87%	84%	
Rb <sup>+</sup> identified	35%	12%	15%	
Dark Yb <sup>+</sup> identified		< 1%		Rb-Yb <sup>+</sup>
Hot ion (unidentified)			1%	(Cambridge)
Number of events	283	754	225	(Cambridge)

$$\gamma_{react} = \gamma_L \epsilon$$
 State-dependent reactivity

L. Ratschbacher et al. Nature Phys. 2012; T. Sikorsky et al. Nature Comm. 2018

How COLD can we get?



#### How COLD can we get? а 5 At these temperature atom-ion Temperature (K) collisions are **CLASSICAL** 3 2 energy $\Delta E_{p}$ ∆E<sub>p</sub> /k<sub>B</sub>≈ 1-10uK internuclear separation 0 20 30 50 60 40 0 10 Interaction time (ms)

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# → Problem 1: coherence does not survive

L. Ratschbacher et al. PRL 2013





L. Ratschbacher et al. PRL 2013

#### Z. Idziaszek et al. PRA 2009

#### **Origin of problems: micromotion**

$$r_{ion,x} = A_x \sin(\omega_x t + \varphi_x) \left[ 1 + \frac{q}{2} \sin(\Omega_T t) \right]$$

$$r_{ion,y} = A_y \sin(\omega_y t + \varphi_y) \left[ 1 - \frac{q}{2} \sin(\Omega_T t) \right]$$

$$\rightarrow Secular motion$$



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Collisions in the presence of micromotion can couple energy from the radiofrequency field to the colliding particles
#### .. a problematic story

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Secular motion  
Micromotion

Collisions in the presence of micromotion can couple energy from the radiofrequency field to the colliding particles

#### **Problem = Opportunity!**

We can use micromotion to tune the collisional energy

### Tuning the collisional energy



By applying an offset electric field  $\vec{E}_0$ the mean kinetic energy changes:

$$\Delta E \approx \frac{1}{2} m \Omega^2 a_{mm}^2 \propto E_0^2$$

Where  $a_{mm}$  is the micromotion amplitude



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### **Studying ion-neutral collisions**

## **Langevin collisions** (no energy dependence of the scattering rate constant)



C. Zipkes, S. Palzer, L. Ratschbacher, C.S., M. Koehl PRL 105, 133201 (2010)

### **Studying ion-neutral collisions**

## **Langevin collisions** (no energy dependence of the scattering rate constant)



**«Soft» collisions** (energy dependent scattering rate constant)



C. Zipkes, S. Palzer, L. Ratschbacher, C.S., M. Koehl PRL 105, 133201 (2010)

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# The energy distribution of an ion in a buffer gas is *not* a thermal distribution



#### It's like playing Texas hold'em only with all-ins!<sup>©Roee Ozeri</sup>

## The energy distribution of an ion in a buffer gas is *not* a thermal distribution



C. Zipkes et al., New J. Phys. 2011

It's a power law distribution.. This is complex physics!

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#### **Open problems**

#### Still many issues:

- Can we enter a full quantum regime (few partial waves) in atom-ion interactions?
- Can we reach a coherent evolution of the ionneutral mixture?
- Can we control (in a more deterministic way) inelastic processes?

#### Atoms are an exceptional tool to measure micromotion!

Method	$\Delta \varepsilon  (V/m)$
Photon-correlation spectroscopy	0.9
Micromotional sideband spectroscopy	7 / 1 / 0.4
Ion-cavity emmission spectroscopy	1.8
Parametric excitation of secular motion	6/0.4
Neutral atom loss	0.02
Monitor displacement	$\leq 11.8$
Trajectory analysis	0.09

Gloger et al. PRA 92, 043421 (2015)

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Haerter et al. APL 102, 221115 (2013)

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Gloger et al. PRA 92, 043421 (2015)

#### Is that sufficient?



Haerter et al. APL 102, 221115 (2013)



What mass ratio to reach s-wave scattering? → Yb+ - Li Cetina et al. PRL 109, 253201 (2012) → Yb<sup>+</sup> - Li Krych et al. PRA **91**, 023430 (2015) → Ba<sup>+</sup> - Li

#### Static ion trapping

Paul trap  $\rightarrow$  dipolar trap

### Optical trapping of an ion

Ch. Schneider, M. Enderlein, T. Huber and T. Schaetz\*

First demonstrated in 2010

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#### **PROS:**

Intrinsically without micromotion

No limitations in mass ratio

### Static ion trapping

#### Paul trap $\rightarrow$ dipolar trap



### **Optical trapping of an ion**

Ch. Schneider, M. Enderlein, T. Huber and T. Schaetz\*

First demonstrated in 2010

#### CONS:

me because of

**PROS**:

SHORT lifetime because of difficulties in laser cooling

BUT atoms can provide cooling!

Intrinsically without micromotion No limitations in mass ratio



S-wave regime - collisions described by a single phase shift: scattering length



S-wave regime - collisions described by a single phase shift: scattering length

Observe Feshbach resonances to control ion-neutral interactions



Z. Idziaszek et al. PRA 2009



S-wave regime - collisions described by a single phase shift: scattering length

Observe Feshbach resonances to control ion-neutral interactions



 Use atom-ion pair with "tall" (>10uK) p-wave barrier



Z. Idziaszek et al. PRA 2009



S-wave regime - collisions described by a single phase shift: scattering length

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#### Some advice:

- Use atom-ion pair with "tall" (>10uK) p-wave barrier
- Use atom-ion pair with a sufficiently large mass ratio
   OR optical trap for the ions



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#### Control the formation of molecules and achieve sympathetic cooling

#### 0.05 а С -v = 0<sup>1</sup>П -v = 160 0.04 ••• 300 K sim 0.04 0.03 0.02 0.02 Energy (10<sup>3</sup> cm<sup>-1</sup>) 50 Data Ba+(6s) + Cl(3p 40 30 20 0.01 🔊 42,000 44,000 46,000 48,000 50,000 40,000 Internuclear radius $(a_0)$ Energy (cm<sup>-1</sup>) b

W.G. Rellergert et al. Nature 2013

Photodissociation thermometry of a BaCl+ ions immersed in a Ca MOT

Also Heidelberg, Basel, Bangalore, ...

#### **Inelastic collisions**

#### More pieces of advice:

- Use atom-ion pairs for which you know you get a molecule
- Alternatively: begin from molecules!

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# Langevin: phase shift during clock operation



#### **Effects:** Decoherence (Langevin, soft collisions) Quench (Langevin collisions)



# Langevin: phase shift during clock operation



Model (Rosenband et al. Science 2008):

- assume pi/2 phase shift at each Langevin collision
- 2. Integrate over optical Bloch equation
- $\rightarrow$  Average frequency shift:

 $\Delta v_{\text{coll}}$ =0.15  $\gamma_{\text{L}}$ 

### **Effects:** Decoherence (Langevin, soft collisions) Quench (Langevin collisions)



# Langevin: phase shift during clock operation



estimated shift  $\Delta v_{coll} = 0.15 \gamma_{L}$ uncertainty Al<sup>+</sup>: 0.5 x 10<sup>-18</sup> Sr<sup>+</sup>: 2 x 10<sup>-18</sup>

Model (Rosenband et al. Science 2008):

- assume pi/2 phase shift at each Langevin collision
- 2. Integrate over optical Bloch equation
- $\rightarrow$  Average frequency shift:

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Soft collisions: differential phase



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Complicated structure!



Soft collisions: differential phase



Li-Ca<sup>+</sup> (Mukaiyama, Dulieu)

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Complicated structure!



#### Soft collisions: differential phase



Li-Ca<sup>+</sup> (Mukaiyama, Dulieu) Model (Rosenband et al. Science 2008): Consider the induced dipole on the atom as a fluctuating E-field estimated shift uncertainty  $Al^+ < 1 \times 10^{-19}$ 

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Precise detection of Langevin and soft collisions rates



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Precise detection of Langevin and soft collisions rates





L. Ratschbacher, et al. PRL 110, 160402 (2012) Similar on Sr<sup>+</sup>-Rb measurements at Weizmann "knobs" that we can use:

- Density of the neutrals
- Collisional energy
- Optical transitions
- Internal states of the ions

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Are collisionally-created molecular ions a common phenomena? NO: Yb<sup>+</sup>-Rb, Ba<sup>+</sup>-Rb, Sr<sup>+</sup>-Rb, Yb<sup>+</sup>-Li, ... YES: Ca<sup>+</sup>-Rb, many hydrates (like YbH<sup>+</sup>)

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Questions: How big can Coulomb crystals be before being too much affected by these inelastic processes? Can we photodissociate these molecules and recycle the ion?

## Conclusions

- Neutral-ion interaction's main term is a long-range R-4 potential
- Collisions can be of two types: Langevin and soft collisions
- Langevin collisions' scattering rate is independent from the collisional energy, while soft collisions' scattering rate is not
- Hybrid atom-ion experiments have dramatically advanced our understanding of neutral-ion collisions
- In hybrid experiments we can control the density, collisional energy, the internal state of the colliding particles, the occurrence of chemical reactions
- Applications to ion clocks: searching for frequency shifts and for procedures to "recycle" an ion undergoing a chemical reaction

## The group and funds



#### **Our experimental setup:** a novel atom-ion mixture

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Reactive collisions only "on demand", i.e. only by placing the ions or the atoms in a different internal state

#### **Our experimental setup:** a novel atom-ion mixture



By using a spin-polarized Fermi gas, 3-body collisions (in s-wave) are suppressed by Pauli principle  $\rightarrow$  less heating

#### **Our experimental setup:** a novel atom-ion mixture



(Landé factor of opposite sign)