Lecture 1





Towards quantum impurity physics with atoms and ions Rene Gerritsma



Lecture 1

- Introduction
- Atom-ion interactions
- Ion trapping
- Reaching the quantum regime

Motivation

Why study atom-ion mixtures?





- → A crystal of ions overlapped with p cloud of ultracold fermions: an artificial solid??
- → Mediated interactions?



	Nat. solid	⁶ Li / ¹⁷⁴ Yb+
Lattice spacing d (nm)	0.3-0.6	10 ³ - 10 ⁴
Length scale R* (nm)	0.026	71
d/R*	10-20	14-140
mi / mf	10 ⁴	29

U. Bissbort et al., PRL 111, 080501 (2013).

Quantum chemistry with trapped ions and atoms

- Prepare ions and atoms in particular quantum state
- Detect chemical reactions at the single particle level
- Measure the output states, released or absorbed energy, branching ratios, etc...



Phys. Rev. Lett. 107, 243202 (2011). Phys. Rev. Lett. 107, 243201 (2011). Nat. Phys. 8, 649 (2012). Phys. Rev. A 96, 030703(R) (2017). Nature Communications. 9, 920 (2018) Phys. Rev. Lett. 109, 123201 (2012) And many more....

Atoms and ions used

Alkali atoms and earth alkaline or alkali (i.e. same as parent atom) ions

		2																
Ι	II												III	IV	V	VI	VII	VIII
1	2		3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
${\rm H}^{1}$														(<i>r</i>	$p \mathrm{shel}$	lls)	~	${\rm He}^2$
Ľi	${\operatorname{Be}}^4$												B ⁵ B	$\overset{6}{\mathrm{C}}$	7 N	8 0	$^{9}{ m F}$	10 Ne
Na	${\rm Mg}^{12}$				tra	ansitic	on elen	nents	$(nd \sinh$	ells)			Al	¹⁴ Si	$\overset{15}{\mathrm{P}}$	$\overset{16}{\mathrm{S}}$	Cl	${\rm Ar}^{18}$
${\rm K}^{19}$	$\overset{20}{\mathrm{Ca}}$		${\operatorname{Sc}}^{21}$	${\mathop{\rm Ti}\limits^{22}}$	23 V	Cr^{24}	${\stackrel{25}{ m Mn}}$	${\mathop{\rm Fe}}^{26}$	$\begin{array}{c} 27\\ \mathrm{Co} \end{array}$	28 Ni	29 Cu	$\frac{30}{\text{Zn}}$	Ga 31	Ge^{32}	$\stackrel{33}{\mathrm{As}}$	³⁴ Se	${\operatorname{Br}^{35}}$	$\frac{36}{\mathrm{Kr}}$
Řb	${ m Sr}^{ m 38}$		39 Y	${ m }^{40}_{ m Zr}$	⁴¹ Nb	$\stackrel{42}{\mathrm{Mo}}$	$\frac{43}{\mathrm{Tc}}$	${\operatorname{Ru}}^{44}$	$\frac{45}{Rh}$	Pd^{46}	$\mathop{\mathrm{Ag}}\limits^{47}$	$\overset{48}{\mathrm{Cd}}$	In	${\mathop{\rm Sn}\limits^{50}}$	$\frac{51}{\text{Sb}}$	$\stackrel{52}{\mathrm{Te}}$	53 I	$\overset{54}{ ext{Xe}}$
$\overset{55}{\mathrm{Cs}}$	$\mathbf{\overset{56}{Ba}}$	57-70 *	${ m Lu}^{71}$	${\rm Hf}^{72}$	73 Ta	\mathbf{W}^{74}	$\stackrel{75}{\mathrm{Re}}$	$\overset{76}{\mathrm{Os}}$	77 Ir	$^{78}_{\mathrm{Pt}}$	${\mathop{\rm Au}\limits^{79}}$	${}^{80}_{\rm Hg}$	81 Tl	$^{82}_{ m Pb}$	83 Bi	84 Po	At^{85}	$\frac{86}{Rn}$
${ m Fr}^{87}$	$\frac{88}{\text{Ra}}$	89-102 **	${ m Lr}^{103}$	${ m Rf}^{104}$	105 Db	${}^{106}_{\mathrm{Sg}}$	Bh	${}^{108}_{\mathrm{Hs}}$	${ m Mt}^{109}$	${ m Ds}^{110}$	$\stackrel{111}{\mathrm{Rg}}$							
	<u> </u>					r	are-ea	rth ele	ement	s $(nf$	shells)							
				57	58	50	60	61	62	63	64	65	66	67	68	60	70	

*lanthanides	(4f)

**actinides (5f)

L^{57}	$\overset{58}{\mathrm{Ce}}$	\Pr^{59}	$\overset{60}{\mathrm{Nd}}$	${\mathop{\rm Pm}}^{61}$	${ m Sm}^{62}$	63 Eu	${\operatorname{Gd}}^{64}$	$\frac{65}{\text{Tb}}$	$\overset{66}{\mathrm{Dy}}$	67 Ho	${\mathop{\rm Er}\limits^{68}}$	${}^{69}_{Tm}$	${ m Yb}^{70}$
$\stackrel{89}{\mathrm{Ac}}$	$^{90}_{\mathrm{Th}}$	91 Pa	92 U	$\stackrel{93}{\mathrm{Np}}$	${\mathop{\rm Pu}\limits^{94}}$	${ m Am}^{95}$	${ m }^{96}_{ m Cm}$	$^{97}_{ m Bk}$	$^{98}_{\mathrm{Cf}}$	$\overset{99}{\mathrm{Es}}$	${ m Fm}^{100}$	Md ¹⁰¹	102 No

X. 8

Some of the groups:

Amsterdam: Yb⁺/Li, Freiburg: Ba⁺/Li, LENS: Ba⁺/Li, Ulm: Ba⁺ and Rb⁺/Rb, Weizmann: Sr⁺/Rb, Stuttgart: Rb⁺/Rb, Arhus Ba⁺/Li (under construction), ...

Yb⁺

\downarrow Same as earth alkaline ions

 $\nvdash \downarrow \downarrow$ Configuration interactions severely limit theoretical predictions



Already pretty complicated, but still a favorite in ion trapping

Atom/ion potentials

PRA 96, 030703(R) (2017)



Note: no spin-orbit!



Atom-ion interactions

- Interaction potential
- The s-wave limit
- The Langevin rate

Atom-ion interactions

- An ion polarizes a nearby atom
- Leads to attraction at large distances



Typical units

- An ion polarizes a nearby atom
- Leads to attraction at large distances

$$V_{a-i}(r) = -\frac{C_4}{2 r^4}$$

Equate potential energy with kinetic energy to obtain typical length and energy scales:

$$R^* = \frac{\sqrt{\mu C_4}}{\hbar} \quad \leftarrow \text{Typical scattering} \qquad E^* = \frac{\hbar^2}{2\mu (R^*)^2} \leftarrow \text{s-wave energy}$$

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Atom	Ion	Mass ratio $\frac{m_i}{m_a}$	E^*/h (kHz)	$E^*/k_B \ (\mu \mathrm{K})$	R^* (nm)	References
⁶ Li	$^{174}\mathrm{Yb}^{+}$	28.92	178.583	8.57	69.77	[106]
	$^{171}\mathrm{Yb}^+$	28.42	178.793	8.58	69.75	
	$^{138}Ba^{+}$	22.93	181.716	8.72	69.46	
87 Rb	$^{174}\mathrm{Yb}^{+}$	2.00	0.924	0.044	307.23	[112, 113]
	$^{171}\mathrm{Yb}^+$	1.97	0.935	0.045	306.33	
	$^{138}\mathrm{Ba^{+}}$	1.59	1.092	0.052	294.67	
	$^{88}\mathrm{Sr}^+$	1.01	1.624	0.078	266.80	

Langevin collisions

- Atoms that come close to the ion undergo close-range 'Langevin' collisions
- Write down Hamiltonian for the relative motion:

$$H = \frac{\vec{p}^2}{2\mu} - \frac{C_4}{2r^4}$$

Reduced mass $\mu = \frac{m_{atom}m_{ion}}{m_{atom}+m_{ion}}$

Transform to spherical coordinates:



Langevin collisions

- Angular momentum and collision energy determine whether particles can get close enough to each other to react
- Closeby collisions are called 'Langevin collisions'



Impact parameter b

When the particles are far away from each other, their motion is not affected yet by the force:

$$\vec{L} = \vec{r} \times \vec{p} \rightarrow |L| = \mu v b$$

• The particle can only make it over the barrier if the collision energy is larger than $E_{barrier}$:



$$E_{barrier} = \frac{L^4}{8C_4\mu^2}$$
$$E_{col} = \frac{1}{2}\mu v^2$$

The Langevin rate

There is a critical impact parameter below which all collisions are short range 'Langevin collisions'



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

From: Contemporary Physics, 55:1, 33-4

The Langevin rate

There is a critical impact parameter below which all collisions are short range 'Langevin collisions'



From: Contemporary Physics, 55:1, 33-4

→ Famous result from physical chemistry: reaction rates between neutral and charged particles is independent of energy

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

Langevin collision rate: $\gamma = n v \sigma$ Cross section $\sigma = \pi b_c^2$ Atomic density n

→ Langevin rate independent of Velocity (collision energy)

$$\gamma = 2\pi n \sqrt{C_4/\mu}$$

Then why look at cold chemistry?

- Quantum effects change the picture: The angular momentum is quanitized!
- ▶ When the collision energy gets very low, only L = 0 collisions are allowed → the s-wave limit
- Langevin theory breaks down! \rightarrow What happens in the ultracold regime?





lon trapping

- ► The Paul trap
- Equations of motion
- Micromotion and the secular approximation
- The quantum case and the breakdown of the secular approximation

Some history



Wolfgang Paul

Hans Dehmelt

- Ion traps were developed by Paul and Dehmelt in the 50s
- They were awarded the Nobel price in 1989
- Two types of ion traps: Paul (or radio frequency) traps Penning traps





Ion trapping: Paul traps

Ions trapped by a combination of static and oscillating (rf) electric fields

$$\begin{aligned} \boldsymbol{\mathcal{E}}(t) &= \boldsymbol{\mathcal{E}}_{s} + \boldsymbol{\mathcal{E}}_{rf}(t) \\ \boldsymbol{\mathcal{E}}(t) &\approx \left(\mathbf{G}_{s} + \mathbf{G}_{rf}(t)\right) \cdot \mathbf{r} \end{aligned}$$

$$G_{s,i,j} = \frac{d\mathcal{E}_{s,i}}{dr_j}$$
$$G_{\mathrm{rf},i,j}(t) = \frac{d\mathcal{E}_{\mathrm{rf},i}(t)}{dr_j}$$
$$i, j \in \{x, y, z\}$$



Ion trapping: Paul traps

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 $i,j \in \{x,y,z\}$



Earnshaw: cannot hold stationary charged particle with static fields alone

Solving the equations of motion

Ions trapped by a combination of static and oscillating (rf) electric fields

$$G_{\mathrm{s},i,j} = -(m_{\mathrm{i}}\Omega_{\mathrm{rf}}^{2}/e)a_{ij}$$

$$G_{\mathrm{rf},i,j} = -(m\Omega_{\mathrm{rf}}^{2}/(2e))q_{ij}$$
Stability parameters
$$G_{\mathrm{rf},i,j} = -(m_{\mathrm{rf}}^{2}/(2e))q_{ij}$$

eqs. Of motion:

$$m_{\rm i}\ddot{\mathbf{r}}(\mathbf{t}) = e(\mathbf{G}_{\rm s} + \mathbf{G}_{\rm rf}(t)) \cdot \mathbf{r}$$

Solving the equations of motion

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$$G_{\mathrm{rf},i,j} = -(m_{\mathrm{rf}}^{2}/(2e))q_{$$

Assume gradient tensors can be simultaneously diagonalized \rightarrow 3 uncoupled eqs. Of motion:

$$m_i \ddot{x} = (a_x - 2q_x \cos 2\xi)x$$

With $\xi=\Omega_{rf}t/2$

And similar for the y and z directions



Summary

- An ion in a Paul trap behaves like a harmonic oscillator with small amplitude, high frequency micromotion on top
- Just choose stability parameters such that $a \ll q^2 \ll 1$
- Build a good ion trap (there is also something called excess micromotion.....)
- And forget about micromotion ③

"The career of a young theoretical physicist consists of treating the harmonic oscillator in ever-increasing levels of abstraction."

Sidney Coleman (according to google)...

Summary

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Stick to 1D

$$H_{\rm ion}(t) = \frac{p_i^2}{2m_i} + \frac{1}{8}m_i\Omega^2 z_i^2[a + 2q\cos(\Omega t)]$$

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Cook and Shankland: Write wavefunction as:

$$\Psi(z_i,t) = \exp\left(-\frac{i}{4\hbar}m_i q \,\Omega z_i^2 \sin(\Omega t)\right) w(z_i,t)$$

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The wavefunction $w(z_i,t)$ evolves according to the Hamiltonian:

$$H_{\rm eff}(t) = \frac{p_i^2}{2m_i} + \frac{1}{2}m_i\omega_i^2 z_i^2 + H_{mm}(t)$$

 $H_{mm}(t) = -m_i g^2 \omega_i^2 z_i^2 \cos(2\Omega t) - g \omega_i \{z_i, p_i\} \sin(\Omega t)$ $g = [2(1 + 2a/q^2)]^{-1/2}$

Stick to 1D

$$H_{\rm ion}(t) = \frac{p_i^2}{2m_i} + \frac{1}{8}m_i\Omega^2 z_i^2[a + 2q\cos(\Omega t)]$$

Cook and Shankland: Write wavefunction as:

$$\Psi(z_i,t) = \exp\left(-\frac{i}{4\hbar}m_i q \Omega z_i^2 \sin(\Omega t)\right) w(z_i,t)$$

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Another look....

-

This extra micromotion term looks troubling though....

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However....

This extra micromotion term looks troubling though....

Another look....

Parametric excitations in trapped ions: well known

- ← In practice, the multipole expansion never ends with quadrupole!
- → Solution: low q (or beta)
- \rightarrow This is a ringtrap btw

Alheit R, Hennig C, Morgenstern R, Vedel F and Werth G 1995 Appl. Phys. B 61 277-83

What happens when we add atoms?

• Unfortunately, it is hard to imagine anything more anharmonic than $\frac{1}{r^4}$

$$H_{\rm eff}(t) = \frac{p_i^2}{2m_i} + \frac{1}{2}m_i\omega_i^2 z_i^2 + H_{mm}(t) - \frac{C_4}{2|r_i - r_a|^4}$$

$$H_{mm}(t) = -m_i g^2 \omega_i^2 z_i^2 \cos(2\Omega t) - g \omega_i \{z_i, p_i\} \sin(\Omega t)$$

 \rightarrow Interaction with an atom will cause parametric excitation all over the place....





Ultracold atom-ion mixtures?

• Micromotion of ion in a Paul trap limits attainable temperatures

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DeVoe, PRL 102, 063001 (2009). Zipkes et al., NJP 13, 053020 (2011). M. Cetina et al., PRL 109, 253201 (2012). Chen et al., PRL 112, 143009 (2014). Rouse & Willitsch, PRL 118, 143401 (2017). Höltkemeier et al., PRL 116, 233003 (2016). Fürst et al., J. Phys. B 51, 195001 (2018).

So what to do?

Live with it The quantum regime is in reach for excellent Paul traps and some atom-ion combinations	Optical traps Optical traps eliminate micromotion -induced heating
Nature Physics 16, 413–416 (2020).	Phys. Rev. Lett. 124, 053402 (2020).
No trap No trap, no micromotion	<text></text>
Phys. Rev. Lett. 126, 033401 (2021).	Phys. Rev. Lett. 118, 263201 (2017).

⁶ Li ³ 171 70 Yb⁺

Why live with it?

- The s-wave limit can still be reached in a Paul trap
- The Paul trap offers key benefits:
 - >100000 K deep \rightarrow very long storage time
 - Can use lasers to accurately measure motion and state of ions
- Optical traps have their own disadvantages (for example photoninduced chemistry)
- Using no trap makes it hard to sustain the ultracold regime (sensitive to offset fields)



The role of the masses

- All polarizabilities of alkalis are pretty similar (within factor ~2)
- Mass ratio between ion and atom can be varied more

$$W_0 = 2 \left(\frac{m_a}{m_i + m_a} \right)^{5/3} \left(\frac{m_i^2 \omega^4 C_4}{q^2} \right)^{1/3}$$

← Estimated minimum heating in one low energy collision, based on classical physics

Phys. Rev. Lett. 109, 253201 (2012)

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The role of the masses

$$W \sim \left(\frac{m_a}{m_i + m_a}\right)^{5/3}$$



Micromotion heating effect

S-wave limit

 $E_{col} \sim \frac{\mu}{m_i} T_{ion} + \frac{\mu}{m_a} T_{atom}$

Collision energy

- Sticking to much used ions and atoms that can be easily laser cooled:
- Largest practical mass ratio → Yb⁺/⁶Li
- Also good choice: Ba⁺/⁶Li

H. A. Fürst, N. V. Ewald, T. Secker, J. Joger, T. Feldker and R. Gerritsma, J. Phys. B 51, 195001 (2018).

What to expect?

Equilibrium energy of ion \downarrow

Li

Ion	Atom	a_x	q	$\Omega_{ m rf}/(2\pi)$	$W_0^{3D}/k_{ m B}$	$E_{\infty}^{ m ion}/k_{ m B}$	Refs.
				(MHz)	(μK)	(μK)	
¹⁷¹ Yb ⁺	⁶ Li	-0.00073	0.22	2	0.31	28.0	Hirzler et al. (2020a)
$^{138}Ba^{+}$	⁶ Li	-0.00011	0.24	1.433	0.30	35.5	Weckesser et al. (2021a)
$^{40}\mathrm{Ca^{+}}$	⁶ Li	-0.0016	0.16	4.8	3.29	87.1	Saito et al. (2017)
$^{138}\mathrm{Ba^{+}}$	⁸⁷ Rb	-0.00093	0.13	5.24	56.3	1.47×10^3	Schmid et al. (2012)
$^{138}Ba^{+}$	⁸⁷ Rb	-0.00032	0.40	1.4	20.7	667	Schmidt et al. (2020b)
$^{88}\mathrm{Sr}^+$	⁸⁷ Rb	-0.052	0.58	4.22	104	313×10^3	Katz et al. (2022)
$^{87}\mathrm{Rb}^+$	⁸⁷ Rb	-0.0015	0.21	5.24	87.5	552×10^3	Schmid et al. (2012)

Molecular dynamics simulations for various combinations and traps in a dilute atomic bath of 2 μK with realistic excess micromotion

R.S. Lous and R. Gerritsma, Advances In Atomic, Molecular, and Optical Physics 71, 65-133 (2022).

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Collision energy below s-wave

R.S. Lous and R. Gerritsma, Advances In Atomic, Molecular, and Optical Physics 71, 65-133 (2022).

So let's do it!



lon trap

⁶ Li ³ 171 70 Yb

Doppler-cooled trapped ¹⁷¹Yb⁺





+

Transport up using piezo-controlled mirrors

Li

6 3

Buffer gas cooling



⁶ Li ³ 171 70 Yb⁺

Release atoms



Rabi frequency depends on motional quantum number n $\Omega \approx \Omega_0 (1 - \eta^2 n)$

Spectroscopy on ion

Leibfried et al., PRL 77, 4281{4285 (1996). Meir, Z. et al. PRL 117, 243401 (2016).



70 +

Wait for decay of $D_{5/2}$ state



70 +

State detection





Interactions in quantum regime

Type of motion	$E_{\rm kin}/{ m k_B}(\mu{ m K})$	$E_{\rm col}/k_{\rm B}(\mu{\rm K})$
Radial secular ion	$2 \times 21(9)$	1.4(0.6)
Intrinsic micromotion	$2 \times 21(9)$	1.4(0.6)
Axial secular ion	65(18)	2.2(0.4)
Excess micromotion	44(13)	1.5(0.4)
Total ion energy	193(42)	6.6(1.4)
Atom temperature	$3/2 \times 2.3(0.4)$	3.3(0.6)
Total collision energy		9.9(2.0)

Measurement of all types of motion

$$E_{col} = 1.15(23) \times E_{s} \longrightarrow Cross$$

Crossover to quantum regime

Li

Coldest results

T. Feldker et al., *Nature Physics, 16, 413–416 (2020)*.

Summary

- Plenty of interesting physics to explore with atoms and ions!
- We introduced the atom-ion interaction potential
- We introduced ion trapping
- We explored micromotion-induced heating and what to do about it
- We now have two systems, Yb⁺/⁶Li and Ba⁺/⁶Li that have reached the crossover into the quantum regime
- Tomorrow: Some quantum chemistry and controlling interactions between atoms and ions