Towards ultracold RbSr ground-state molecules
Quantum many-body physics

Strong quantum correlations render description difficult.

Especially difficult:

Fermionic systems

Frustrated systems

Dynamics

\[ \langle \hat{O} \rangle \]
Quantum simulation (and beyond)

Difficult to study quantum system

Simplified model

Well-controlled ultracold system

Control everything

Potential

Interactions

Internal state
New ultracold systems

**Alkaline-earth(-like) elements**
Yb, Sr, Ca

**Magnetic atoms**
Cr, Dy, Er

**Rydberg atoms**

**Ground-state molecules**
closed-shell: KRb, NaK, RbCs,...
open-shell: RbYb, RbSr, YbCs

**Benefits**

- More internal structure
- More interesting interactions
  - long-range
  - anisotropic

- More interesting interactions
Molecules

Richer structure than atoms
rotational and vibrational states

Dipole-dipole interactions
due to electric dipole moment
Example: quantum magnetism

Magnetism requires spin dependent interactions.

Provided by super-exchange in ultracold ground-state atoms

\[ t \quad U \quad t \]

spin dependent interaction

Requires extremely low motional temperature

In dipolar molecules simply provided by spin-state dependent dipole interaction

Theory:
Micheli et al., nature physics 2, 341 (2006)
Gorshkov et al., PR A, 84, 033619 (2011)

Requires only low spin temperature
Examples: dipolar gases in 2D

**Bosonic molecules**

Monte-Carlo density distributions in dependence of interaction strength

- (a) $N=13$, $\tau=1$
- (b) $N=13$, $\tau=2.5$
- (c) $N=13$, $\tau=5$
- (d) $N=13$, $\tau=20$

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

**Fermionic molecules**

Interlayer Cooper pairing

Pikovski, A. et al., PRL 105, 215302 (2010)

Reviews

Two paths to ultracold molecules

Path 1
- Hot atoms associate to form cold molecules

Path 2
- Cold atoms associate to form hot molecules
Status

Four systems, all based on alkali atoms:

- KRb  0.5 Debye dipole moment  Science 322, 231 (2008)  Ye, Jin group
- Cs$_2$  0 Debye  Faraday Disc., 284 (2009)  Nägerl group
- NaK  2.7 Debye  PRL 114, 205302 (2015)  Zwierlein group

Other alkali-alkali molecule experiments being set up by many groups.

First experiments studying magnetism:

*Observation of dipolar spin-exchange interactions with lattice-confined polar molecules*


*Many-body dynamics of dipolar molecules in an optical lattice*

PRL 113, 195302 (2014)

Challenge

Cooling to quantum degeneracy
Evaporative cooling
Challenge for evaporative cooling

Chemistry

\[ \text{KRb} + \text{KRb} \rightarrow \text{K}_2 + \text{Rb}_2 + \text{energy} \]
Counter measures

Non-reactive species
e.g. RbCs, NaK,...

Dangerous „sticky collisions“?
Bohn group, PR A 87, 012709 (2013)

Repulsive molecules in 1D lattice

Zoller group, PRL 98, 060404 (2007)
Bohn group, PR A 81, 060701(R) (2010)

Repulsive van der Waals interactions

Bohn group, PR A 73, 022707 (2006)
Gorshkov et al. PRL 101, 073201 (2008)

—> might allow evaporative cooling to quantum gas
RbSr ground-state molecules

So far
alkali + alkali atom

Our goal
alkali + alkaline earth atom

- only paired electrons
- unpaired electron

Our group          RbSr
Görlitz group      RbYb
Gupta, Takahashi groups LiYb
Cornish group      CsYb
Repulsive van der Waals interactions

instantaneous dipole – induced dipole interaction

\[ V_{\text{vdW}}(r) = \sum_n \frac{|\langle m | V_{\text{dd}}(r) | n \rangle|^2}{E_m - E_n} \equiv \frac{C_6}{r^6} \]

Can be repulsive in excited state: \( E_m - E_n > 0 \)

→ evaporative cooling of OH at mK temperatures, Ye group, Nature 492, 396 (2012)
Repulsive van der Waals interactions

RbSr energy, neglecting hyperfine structure

E = 150 V/cm

magnetic field

383 G

N=0

N=1

repulsive vdW interaction!
Repulsive van der Waals interactions

preliminary result by John Bohn, JILA, neglecting hyperfine interaction

$RbSr$ interaction potential

$E = 150 \text{ V/cm} \quad B = 383 \text{ G}$

Evaporative cooling possible?

$\frac{C_6}{r^6} > 0$

expected temperature of molecular gas
Rb-Sr mixtures

\[ ^{87}\text{Rb} \quad N = 1.3 \times 10^5 \]
\[ ^{84}\text{Sr} \quad N = 2.3 \times 10^5 \]
Molecule association

**Standard technique**
- magneto association
- Sr is non-magnetic $\rightarrow$ magneto association not trivial
  
  Hutson group, PRL **105**, 153201 (2010)

**Alternative technique**
- STIRAP association
  
  $\rightarrow$ let's demonstrate it by creating Sr$_2$
Scheme

BEC

ramp on optical lattice

Mott insulator

associate molecules
Molecule association by STIRAP

\begin{align*}
|e> & \quad \Omega_2 \\
\Gamma & \quad \Omega_1 \\
|m> & \quad |a> \\
{^1S_0} + {^3P_1} & \quad {^1S_0} + {^1S_0}
\end{align*}

external trap (lattice well)
Molecule association by STIRAP

\[ |e> \quad \Gamma \quad 1S_0 + 3P_1 \]

\[ |m> \quad \Omega_2 \quad 1S_0 + 1S_0 \]

external trap (lattice well)
Molecule association by STIRAP

\[ |e> \quad {^1S_0 + ^3P_1} \quad |a> \quad {^1S_0 + ^1S_0} \]

\[ \Omega_1 \quad \Gamma \quad \text{external trap (lattice well)} \]
Molecule association by STIRAP

\[ |\Psi\rangle = \Omega_1 |m\rangle + \Omega_2 |a\rangle \]

\[ ^1S_0 + ^3P_1 \]

\[ ^1S_0 + ^1S_0 \]

external trap
(lattice well)
Molecule association by STIRAP

$|e\rangle$  $|a\rangle$  $|m\rangle$

$\Omega_2$  $\Omega_1$  $\Gamma$

$^1S_0 + ^3P_1$

$^1S_0 + ^1S_0$

external trap (lattice well)
STIRAP transition

Singlet states

\[ ^1S_0 \]

\[ ^1P_1 \]

Triplet states

\[ ^3P_J \]

461 nm
30 MHz

671 nm
mHz

689 nm
7.4 kHz

698 nm
mHz
STIRAP transition

singlet states  triplet states

$^{1}\text{P}_1$

$^{3}\text{P}_J$

$^1\text{S}_0$

689 nm
7.4 kHz
See also $^{88}\text{Sr}_2$ work by Tanya Zelevinsky, PRL 109, 115303 (2012).
Result

30% conversion efficiency
60µs lifetime

What are the limitations?
Can we improve performance?

See also $^{88}$Sr$_2$ work by Tanya Zelevinsky, PRL 109, 115303 (2012).
Short lifetime of molecules

Sr$_2$ molecular potentials

A. Stein et al., Eur. Phys. J. D 64, 227 (2011)

532 nm (Verdi) used in Innsbruck

1064 nm

Energy [cm$^{-1}$] vs. $R$ [Å]
Lifetime in 1064 nm lattice

110(25) s lifetime

STIRAP efficiency: 55%
Lightshift on binding energy

\[ |e> \]

\[ L_1 \]

\[ 228 \text{ MHz} \]

\[ 75 \text{ MHz} \]

\[ 1S_0 + 3P_1 \]

\[ 1S_0 + 1S_0 \]

\[ |m> \]

\[ |a> \]

\[ \Omega_1 = 2\pi \times 45 \text{ kHz} \]

\[ -18.3 \frac{\text{kHz}}{\text{W/cm}^2} \]

Graph: Binding energy - 644.825 MHz (kHz)
Compensating binding energy shift

\[ |e\rangle \quad 228 \text{ MHz} \quad 75 \text{ MHz} \quad |a\rangle \quad \text{compensation beam} \]

\[ ^1S_0 + ^3P_1 \]

\[ ^1S_0 + ^1S_0 \]

\[ \text{compensation beam intensity / } L_1 \text{ intensity (\%)} \]

\[ \text{Binding energy shift (kHz)} \]
STIRAP with compensation

~50% single pass efficiency

80% single pass efficiency

Sr atom number ($10^4$)

Time (ms)

Intensity (abr)

Time (ms)
STIRAP with compensation

80% single pass STIRAP efficiency

efficiency increase by compensation beam
Why initially lower efficiency?

- ~50% single pass efficiency
- 80% single pass efficiency
STIRAP parameter dependence

atom number after STIRAP round-trip

![Graphs showing the dependence of Sr atom number on δ and Δ](image)

- δ (kHz) vs. Sr atom number ($10^4$) with error bars
- Δ (MHz) vs. Sr atom number ($10^4$) with error bars

Energy level diagram:

$|m\rangle \rightarrow |a\rangle \rightarrow |e\rangle$ with control parameters $\delta$ and $\Delta$.
What limits STIRAP efficiency?

1) off-resonant scattering of photons on atomic line

2) dark state lifetime of 3.5 ms
Next goal

$^{87}\text{Rb}$ $^{84}\text{Sr}$

$\text{Sr}_2$ molecules

$\text{RbSr}$ molecules
Rb-Sr mixture

$^{87}$Rb $N = 1.3 \times 10^5$

$^{84}$Sr $N = 2.3 \times 10^5$
RbSr molecular potentials

Zuchowski et al., PR A 90, 012507 (2014)
RbSr molecular potentials

Zuchowski et al., PR A 90, 012507 (2014)
Photoassociation spectroscopy

88Sr-87Rb  84Sr-87Rb
Photoassociation spectroscopy

Observations

- Transition linewidth increases from 20kHz to 400kHz with increasing binding energy
- $^{84}\text{Sr}^{87}\text{Rb}$ Rabi frequencies $\sim 10x$ lower than $\text{Sr}_2$ or $^{88}\text{Sr}^{87}\text{Rb}$ Rabi frequencies at given intensity

Observation

Rb hyperfine splitting decreases by 3 MHz for 2 GHz binding energy of F=2 state
To fit data, ground-state molecular potential needs to be 10% lower than predicted.

Scattering lengths obtained from fit (preliminary)

<table>
<thead>
<tr>
<th></th>
<th>(^{84}\text{Sr})</th>
<th>(^{86}\text{Sr})</th>
<th>(^{87}\text{Sr})</th>
<th>(^{88}\text{Sr})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{85}\text{Rb})</td>
<td>300 (a_0)</td>
<td>90 (a_0)</td>
<td>50 (a_0)</td>
<td>-10 (a_0)</td>
</tr>
<tr>
<td>(^{87}\text{Rb})</td>
<td>90 (a_0)</td>
<td>-15 (a_0)</td>
<td>-400 (a_0)</td>
<td>275 (a_0)</td>
</tr>
<tr>
<td>(\text{Sr intraspecies})</td>
<td>122 (a_0)</td>
<td>800 (a_0)</td>
<td>96 (a_0)</td>
<td>-2 (a_0)</td>
</tr>
</tbody>
</table>

Simplest mixtures to work with

- \(^{84}\text{Sr}-^{87}\text{Rb}\): Bose - Bose
- \(^{87}\text{Sr}-^{87}\text{Rb}\): Fermi - Bose
1-color PA: thermal cloud vs. BEC

Rb atom number (10^4)

Detuning (MHz)

thermal clouds
Spatial overlap

Phase-separation if \( g_{12} > \sqrt{g_{11} g_{22}} \); \( g = \frac{4\pi\hbar^2}{m} a \)

low interspecies scattering length

\[ \text{density} \]
\[ \text{position} \]

large interspecies scattering length

\[ \text{density} \]
\[ \text{position} \]

interspecies scattering length

\( ^{87}\text{Rb} - ^{84}\text{Sr}: 90 \ a_0 \)

intraspecies scattering lengths

\( ^{87}\text{Rb}: 105 \ a_0 \), \( ^{84}\text{Sr}: 124 \ a_0 \)

\rightarrow \text{no phase separation expected}
Spatial overlap

We use 1064 nm dipole trap: 3x deeper for Rb than for Sr

- gravitational sagging
- weaker trap for Sr
Bi-chromatic dipole trap

Balance trap by 532nm dipole trap
(attractive for Sr, repulsive for Rb)
PA with trap compensation

- Thermal clouds
- BECs, different potentials
- BECs, similar potentials
$^{87}\text{Rb}-^{84}\text{Sr}$ Mott insulator
PA of double Mott insulator

- 20,000 sites with atoms of each species
STIRAP attempts with $^{87}$Rb-$^{84}$Sr

All attempts failed because of weak free-bound transition

Why is this transition so weak?
Free-bound transition strength

Predictions from ab-initio model fitted to data

<table>
<thead>
<tr>
<th></th>
<th>Excited state binding energy (MHz)</th>
<th>Franck-Condon factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{84}\text{Sr} - ^{87}\text{Rb}$</td>
<td>174</td>
<td>$7 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{88}\text{Sr} - ^{87}\text{Rb}$</td>
<td>273</td>
<td>$4 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{87}\text{Sr} - ^{87}\text{Rb}$</td>
<td>115</td>
<td>$3 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

Try with $^{87}\text{Sr} - ^{87}\text{Rb}$!

40 times higher Rabi frequency!
Towards the RbSr ground state

\[ 2S_{1/2} + 3P_{1} \]

\[ 2S_{1/2} + \, ^1S_{0} \]

\(~500 \text{ \textit{a}_0 \text{ radius}~}\)

\(~50 \text{ \textit{a}_0 \text{ radius}~}\)

\(30 \text{ \textit{THz} binding energy} \)

\(~9 \text{ \textit{a}_0 \text{ radius}~}\)

\(~\text{few 100 MHz binding energy}~\)
Spectroscopy of "hot" RbSr
Spectroscopy of "hot" RbSr


absorption [arb. units]

simulation

experiment

E [cm⁻¹]
Current projects

- RbSr molecules
- Li-Sr quantum gas microscope
- Perpetual atom laser
- K quantum gases
The team

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