Ultracold Metastable Helium Atoms

Collisions, trapping effects, long-range bound states

Ian Whittingham

School of Mathematical and Physical Sciences,

James Cook University, Townsville

Collaborators: Tim Beams (JCU), Gillian Peach (UCL)

Overview

- Ultracold collisions in traps
- Ultracold He(2s $^3S_1$)+He(2s $^3S_1$)
  - Potentials, collisions
  - Isotropic and anisotropic harmonic traps
- Ultracold He(2s $^3S_1$)+He(np ^3P_j)
  - Bound states
  - Multichannel and perturbative calculations
- Current and future calculations
  - Anharmonicity corrections for $S+S$
  - Photoassociation to long-range $S+P$ bound states
- Conclusions/Outlook

Ultracold Collisions in Traps

Understanding collisions is crucial to design and operation of traps.

- High *elastic* collision rate required for efficient thermalisation.
- Low *inelastic* rates required to minimize trap loss.
- For He* *Penning ionisation*

$$\mathrm{He}^* + \mathrm{He}^* \rightarrow \mathrm{He} + \mathrm{He}^+ + e^-$$

causes rapid trap loss unless *spin polarised* $s = 1, m_s = +1$ He* is used so that the total spin of the colliding atoms is $S = 2$.

Effects of Trapping Environment

- Ultracold collisions usually studied under weak trapping conditions where confining field is either
  - Ignored or
  - Assumed to have parabolic or harmonic spatial variation of sufficiently low frequency (typically 10^2 Hz) to be treated as constant.
- Tight trapping conditions (optical lattices, microtraps) with trapping frequencies $10^5 - 10^6$ Hz are expected to modify the properties of colliding atoms.

Ultracold He(2s $^3S_1$)+He(2s $^3S_1$)

- Binary collisions represented by the potentials $^{1,3,5}\Sigma^+_{u/g}$.
- Spin polarised systems: $^5\Sigma^+_g$ (14 bound states) with BE_{14} (MHz) and scattering length a_5 (nm).
  - Stärck and Meyer (1994): $BE_{14} = 135.9, a_5 = 8.298$
  - Dickinson *et al* (2004): $8.044 \leq a_5 \leq 12.17$
  - Przybytek and Jeziorski (2005): $BE_{14} = 87.4 \pm 6.7, a_5 = 7.64 \pm 0.20$
  - Experiment (Moal *et al* 2005): $BE_{14} = 91.35 \pm 0.06, a_5 = 7.512 \pm 0.005$
  - Beams and Peach (2006) $(1+\alpha)V^{\mathrm{SM}}_{\mathrm{Disp}}$ with $\alpha = 1.1773 \times 10^{-3}$ gives $BE_{14} = 91.35, a_5 = 7.512$.
- At low incident kinetic energies of ultracold collisions, $l = 0$ (s-wave) scattering dominates.

Collisions Between Two Atoms

- Hamiltonian

$$\hat{H} = \hat{T}_A + \hat{T}_B + \hat{H}_{\text{el}} + \hat{H}_{\text{ext}}.$$

  - $\hat{T}_{A,B}$ represent the kinetic energy of the nuclei.
  - $\hat{H}_{\text{el}}$ is the electronic Hamiltonian (kinetic energy of the electrons plus electrostatic interactions).
  - $\hat{H}_{\text{ext}}$ describes interaction with external field.
- Harmonic approximation near trapping minimum:
  - Decouples CM and relative motions.
  - CM obeys dynamics of a pure harmonic oscillator.
  - Relative motion states $|\psi\rangle$ satisfy

$$[\hat{T}_r + V^{\text{el}}(r) + V^{\text{trap}}(\mathbf{r})]|\psi\rangle = E^{\text{rel}}|\psi\rangle.$$

Isotropic Harmonic Trap

- Trapping potential for the relative motion is also harmonic

$$V^{\text{trap}}(\mathbf{r}) = \frac{1}{2}\mu \sum_{i=1}^{3} \omega_i^2 (r^i)^2.$$

- For isotropic traps, all trapping frequencies are equal $\omega_i = \omega$. Relative motion states have the form

$$\psi_{nlm}(\mathbf{r}) = \frac{1}{r} F_{nl}(r) Y_{lm}(\theta, \phi),$$

where the radial functions $F_{nl}(r)$ satisfy

$$\left[-\frac{\hbar^2}{2\mu}\frac{d^2}{dr^2} + \frac{l(l+1)\hbar^2}{2\mu r^2} + V^{\text{el}}(r) + \frac{1}{2}\mu\omega^2 r^2\right] F_{nl}(r) = E_{nl} F_{nl}(r).$$

Quantum Defect Method I

- V^{el} modifies trap energies $E_0 = \hbar\omega(2n_r + l + \frac{3}{2})$ to $E = \hbar\omega(2n_r^* + l + \frac{3}{2})$ where $n_r^* = n_r - \delta$.
- Quantum Defect Theory based upon observation that present problem (characterised by long asymptotic trap region beyond effective range of V^{el}) shows similar features to the well studied modified Coulomb problem (long-range attractive Coulomb potential supplemented by a short-range interaction).

Quantum Defect Method II

- Transform harmonic oscillator equation in variables $\rho = r/\xi$, $\xi = \sqrt{\hbar/\mu\omega}$, $\kappa = 2E/\hbar\omega$:

$$\left[\frac{d^2}{d\rho^2} - \frac{l(l+1)}{\rho^2} + \kappa - \rho^2\right] F(\rho) = 0$$

  into

$$\left[\frac{d^2}{dy^2} - \frac{\lambda(\lambda+1)}{y^2} + \frac{2}{y} + \epsilon\right] Y(y) = 0$$

  where $y = \kappa\rho^2$, $Y(y) = \sqrt{\rho}F(\rho)$ and $\epsilon = -1/n^{*\,2}$ with $n^* = \kappa/4 = \nu - \delta$.

- This is the modified Coulomb problem (Seaton 1958,1983) for a bound state with $(n, l) \rightarrow (\nu, \lambda)$ where $\nu = n_r + \lambda + 1$ and $\lambda = l/2 - 1/4$.

Self-Consistent Solution

- $V^{\rm el}(r)$ rapidly approaches zero ($\sim r^{-6}$ for $\rm He^*$) outside about $50a_0$ where the trap solution is

$$F^{\rm as}(r) = Ae^{-\frac{1}{2}r^2} r^{l+1} W_{lE}(r^2). \qquad (V^{\rm el}(r) = 0)$$

- Large intermediate region typically $10^2 - 10^4 a_0$ where

$$F^{\rm int}(r) = Br\left[j_l(kr) - \tan\delta_l(E) n_l(kr)\right]. \quad (V^{\rm trap}(r) = 0)$$

- Matching these solutions yields energy eigenvalues in terms of scattering phase shifts

$$\tan\delta_l(E) = -\left(\frac{E}{4}\right)^{l+\frac{1}{2}} \tan\left(\frac{\pi}{4}[E - 2l + 1]\right) \frac{\Gamma(\frac{1}{4}[E-2l+1])}{\Gamma(\frac{1}{4}[E+2l+3])}.$$

- Note: Result *does not* depend on the use of a $\delta-$ function pseudopotential for $V^{\rm el}$.

Discrete Variable Representation

- Solve radial equation in the form $\hat{H}\psi(x) = E\psi(x)$ using a DVR.
- Introduce set of orthonormal basis functions $\{\phi_k\}$ and set of discrete grid points $\{x_\alpha\}$ and choose expansion coefficients to give matrix eigenvalue equation

$$\hat{H}\psi(x_\beta) = \sum_\alpha H_{\beta\alpha}\psi(x_\alpha) = E\psi(x_\beta),$$

where the matrix elements of $\hat{H}$ are

$$H_{\beta\alpha} = \sum_k \phi_k^*(x_\beta)\hat{H}\phi_k(x_\alpha).$$

- For bound states, a Fourier sine basis is used.

Scaling Transformation

- Current problem involves two disparate length scales
  1. the short-range molecular interaction ($\sim 50a_0$), and
  2. the long-range trapping potential ($\sim 10^3 - 10^6 a_0$).
- To sample the small ρ region sufficiently without using an excessive number of grid points in the trapping region, introduce nonlinear coordinate transformation $\rho = U(t)$. This transforms $[-\hat{D}_\rho^2 + V(\rho)]\psi(\rho) = E\psi(\rho)$ into

$$\left[-f^2(t)\hat{D}_t^2 f^2(t) + V[U(t)] + f^3(t)f''(t)\right]\phi(t) = E\phi(t),$$

  where $f(t) \equiv [U'(t)]^{-\frac{1}{2}}$ and $\phi(t) \equiv \psi[U(t)]/f(t)$.
- The choice $\rho = \zeta t^p$ with $\zeta = 20, p = 10$ gives $\approx 17\%$ of scaled mesh points inside 20.

Spin-Dipole Interaction

Non-spherically symmetric interaction

$$\hat{H}_{\mathrm{sd}} = -\frac{\beta}{\hbar^2 r^3}\left[3\frac{(\hat{\mathbf{s}}_A\cdot\mathbf{r})(\hat{\mathbf{s}}_B\cdot\mathbf{r})}{r^2} - \hat{\mathbf{s}}_A\cdot\hat{\mathbf{s}}_B\right] = V_{\mathrm{p}}(r)\mathbf{T}^2\cdot\mathbf{C}^2.$$

- Interaction causes transitions from spin-polarised $S = 2$ state to the $S = 0$ state from which there is a high probability of ionization.
- SD interaction is relatively weak so use perturbation theory.
- Rates sensitive to form used for Penning decay width.
- For frequencies above 100 kHz the lifetimes are less than typical trapping times.

Anisotropic Traps

General harmonic trap can be expanded into an isotropic $V^{(0)}(r)$ and anisotropic part $V^{(2)}(r,\theta,\phi)$

$$V^{\mathrm{trap}}(\mathbf{r}) = \frac{1}{2}\mu \sum_{i=1}^{3} \omega_i^2 (r^i)^2 = \frac{1}{2}\mu\bar{\omega}^2 r^2 \left[1 + \alpha(Y_{22} + Y_{2,-2}) + \beta Y_{20}\right].$$

Writing the relative Hamiltonian as $\hat{H} = \hat{H}_0 + V^{(2)}$ and expanding the state $|\Psi\rangle = \sum_\alpha c_\alpha |\psi_\alpha\rangle$ in terms of the eigenstates of $\hat{H}_0$ we obtain the matrix eigenvalue equation

$$\sum_\alpha \left[E_\alpha \delta_{\alpha'\alpha} + \langle\psi_{\alpha'}|V^{(2)}|\psi_\alpha\rangle\right] c_\alpha = E c_{\alpha'}.$$

The matrix elements are given by $D_{\alpha'\alpha}\int_0^\infty F^*_{\alpha'}(r) r^2 F_\alpha(r) dr$.

Cylindrical Trap ($\alpha = 0$)

- Asymmetry parameter β ranges from 2 (pancake) to -1 (cigar).
- Effects of collisions increases significantly with $|\beta|$ and is most marked for $l = 0$.
- Self-consistent solution for asymptotic spherically symmetric V^{el} matched to axially symmetric V^{trap} is currently under investigation.

He(2s ^3S) + He(np ^3P_j)

- *Photoassociation spectroscopy* of bound states can provide
  - Valuable knowledge about the He* system.
  - Accurate determination of the $S+S$ scattering length a_5.
- *Optical Feshbach Resonance* can modify a_5 through laser coupling to a $S+P$ bound state.

Some Recent Studies

- Herschbach *et al* (2000) observed bound states (NOT long-range) that dissociate to the 2s $^3S_1$+2p $^3P_2$ limit.
- Léonard *et al* (2003) reported theoretical and experimental studies of some purely long-range bound states (binding energies ≤ 1.43 GHz) below the 2s $^3S_1$+2p $^3P_0$ limit.
- Kim *et al* (2004) and van Rijnbach (2004) observed detailed structure (40 peaks) associated with bound states ≤ 13.57 GHz below the 2s $^3S_1$+2p $^3P_2$ limit.
- Moal *et al* (2005) reported high accuracy determination of a_5 from two-photon photoassociation determination of the least bound $(v = 14)$ state of $S + S$.

Collisions of Atoms with Fine-Structure

- Hamiltonian

$$\hat{H} = \hat{T}_r + \hat{H}_{\mathrm{el}} + \hat{H}_{\mathrm{fs}}.$$

where

$$\hat{T}_r \equiv -\frac{\hbar^2}{2\mu}\nabla_r^2 = -\frac{\hbar^2}{2\mu r}\frac{\partial^2}{\partial r^2}r + \frac{\hat{l}^2}{2\mu r^2} \equiv \hat{T}^{\mathrm{rad}} + \hat{H}^{\mathrm{rot}}$$

- Expand system eigenstate in a channel basis :

$$|\Psi(r,q)\rangle = \sum_b \frac{1}{r} G_b(r) |\Phi_b(r,q)\rangle$$

to give (q denotes electronic and angular coords)

$$\sum_b \left[T_{ab}^{\mathrm{rad}} + H_{ab}^{\mathrm{rot}} + H_{ab}^{\mathrm{el}} + H_{ab}^{\mathrm{fs}} \right] G_b(r) = E G_a(r).$$

Long-Range Bound States

- Multichannel calculations (Venturi *et al* 2003; Leduc *et al* 2003):
  - 12 Born-Oppenheimer potentials ($^{2S+1}\Sigma_\sigma^+, {}^{2S+1}\Pi_\sigma; S = 1, 3, 5; \sigma = u/g$) with retarded long-range dispersion contribution

$$f_{3\Lambda}(r/\lambda)C_{3\Lambda}/r^3 + C_{6\Lambda}/r^6$$

  - *Ad hoc* short-range potentials of two forms.
- Penning ionisation at $r \le 5a_0$ ignorable as long-range bound states occur at $r \ge 100a_0$.
- Four sets of purely long-range states found:
  - 1_g (3 levels) and 0_u^+ (6 levels) below 2s $^3S_1$+2p $^3P_0$.
  - 0_u^- (1 level) and 2_u (4 levels) below 2s $^3S_1$+2p $^3P_1$.

Perturbative Calculations

- Multichannel calculations necessary to include couplings that substantially alter nature and lifetimes of bound states, however insight can be obtained from a decoupled perturbative approach.
- *Movre-Pichler with rotation*: Diagonalise at each r

$$V_{ab}^{\mathrm{MPR}} = H_{ab}^{\mathrm{el}} + H_{ab}^{\mathrm{fs}} + H_{ab}^{\mathrm{rot}}(\Omega)$$

  where $H(\Omega)$ is that part of $\hat{l}^2$ that leaves Ω unchanged.
- Neglect radial couplings in kinetic energy terms

$$T_{ab}^{\mathrm{rad}} = -\frac{\hbar^2}{2\mu}\left[\delta_{a,b}\frac{d^2G_b}{dr^2} + 2\langle\Phi_a|\frac{\partial}{\partial r}|\Phi_b\rangle\frac{dG_b}{dr} + \langle\Phi_a|\frac{\partial^2}{\partial r^2}|\Phi_b\rangle G_b\right]$$

Single Channel Results

- Bound states determined from

$$\left[-\frac{\hbar^2}{2\mu}\left(\frac{d^2}{dr^2}+\langle\Phi_a|\frac{d^2}{dr^2}|\Phi_a\rangle\right)+V_a^{\mathrm{MPR}}(r)\right]G_a(r)=EG_a(r).$$

- Results for He(2s $^3S_1$)+ He(2p $^3P_{0,1}$) reproduce multichannel results to better than 0.5 MHz for all but the $J=3\ 0_u^+$ levels where differences are ≤ 3 MHz.
- For He(2s $^3S_1$)+ He(3p $^3P_{0,1}$) we find only one bound state, situated in the 0_u^+ potential with $J=1$. Unfortunately this state extends in to $\approx 80a_0$ and is sensitive to the *ad hoc* short-range potentials added. Estimates of its binding energy range from 25 to 40 MHz.

Bound States Below $j = 2$ Limit

- Dickinson *et al* (2005):
  - Calculated *ab initio* short-range ${}^5\Sigma^+_{g/u}$ and ${}^5\Pi_{g/u}$ potentials plus retarded dispersion.
  - Single channel determination of quintet levels gave assignments (Ω_u, J, v) of $\approx 75\%$ of the 40 observed PA peaks
- Léonard *et al* (2005):
  - Compared PA peaks ≤ 10 GHz below asymptote observed in MOT and magnetic traps.
  - Single channel calculations of *ungerade* levels revealed evidence of weak non-diagonal rotational couplings from quintet to singlet symmetry for $1_u(J = 1, 3)$ and $2_u(J = 3)$. $2_u(J = 2)$ is purely quintet.

Current/Future Calculations I

1. He(2s $^3S_1$)+He(2s $^3S_1$)
   Anharmonic quartic corrections for isotropic trap:

$$V^{\text{trap}} = \frac{\alpha}{2}(M^2R^4) + 4\mu^2r^2R^2 + 8\mu^2(\mathbf{R}\cdot\mathbf{r})^2$$

2. He(2s $^3S_1$)+He(2p ^3P_j)
   - Multichannel calculation of the numerous bound states below the $j = 2$ asymptote when short-range $^{1,3}\Sigma_{g/u}$ and $^{1,3}\Pi_{g/u}$ potentials become available.
   - Trapping effects on long-range bound states.
3. He(2s $^3S_1$)+He(3p ^3P_j)
   Further study of $(0_u^+, J = 1)$ bound state below $j = 0$ asymptote. Requires numerous potentials into $r \approx 20a_0$.

Current/Future Calculations II

Photoassociation to long-range bound states of He(2s $^3S_1$)+He(2p ^3P_j), especially the $(0_u^+, v=0)$ state just below the $j=0$ asymptote.

- Investigate dependence upon laser intensity for small detunings comparable to laser coupling.
- Use fully dressed multichannel calculation.
- Limitations on existing calculations:
  - Napolitano (1998): Multichannel, dressed $s+d$ waves, high detunings $-\hbar\Delta \gg \hbar\Omega_{\mathrm{Rabi}}$.
  - Bohn and Julienne (1999); perturbative radiative coupling.
  - Simoni *et al* (2002): radiative coupling vanishes asymptotically.

Conclusions/Outlook

- Several interesting and important calculations worth undertaking for 2s ^3S+2s ^3S and 2s ^3S+np ^3P He* systems.
- Progress is critically dependent upon availability of
  - Short-range singlet and triplet potentials for 2s ^3S+2p ^3P and numerous potentials for 2s ^3S+3p ^3P.
  - Postgraduate and postdoctoral students!