Effects of electric static and laser fields on cold collisions of polar molecules

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Outline

- Introduction
  What’s interesting about cold molecules?
  Methods to produce cold molecules

- Electric-field-control of molecular collisions
  How electric fields can help cool molecules
  Collisions of molecules in a microwave cavity

- Threshold laws for collisions in 2D

- Outlook: Collision physics and ultra-cold molecules
Applications of cold molecules

- Tunable interactions
- Quantum computation
- High-precision spectroscopy
- Controlled chemistry

\[ V_{\text{dip}} = \frac{d_1 \cdot d_2 - 3(R \cdot d_1)(R \cdot d_2)}{R^3} \]

The dipole-dipole interaction

Cold molecules

- Tunable interactions

\[ V_{\text{dip}} = \frac{d_1 \cdot d_2 - 3(R \cdot d_1)(R \cdot d_2)}{R^3} \]

- Orientation and alignment

- Controlled chemistry
How to create ultracold molecules?
How to create ultracold molecules?

- Photoassociation of ultracold atoms
  (Stwalley, DeMille, Bigelow, Heinzen, Massignon-Sieuws, ...)

- Feshbach resonance sweep
  (Jin, Ketterle, Wieman, Cornell, Heinzen, ...)

- Stark deceleration of molecular beams
  (Meijer, Barker, Peters, Friedrich, ...)

- Skimming
  (Abraham, Shafer-Ray, ...)

- Free expansion
  (Gupta, Friedrich, Hershbach, ...)

- Buffer gas loading
  (Doyle, Peters, ...)

- Optical dipole force slowing
  (DeMille, ...)

- Mechanical slowing
  (Gupta, Hershbach, ...)

- Sympathetic cooling by collisions with ultracold atoms
  (Meijer, ...)

- Billiard-ball-like collisions to stop molecules
  (Chandler, ...)

Notes:
Experimental methods for cooling molecules

- Coherent control of dynamics: \( \text{Li}_2, \text{RbCs} \)
- Testing fundamental symmetries of nature: \( \text{YbF, PbO} \)
- Quantum computation: \( \text{CaF, CaCl, MgCl} \)

**Cryogenic cell:**

- To dilution refrigerator
- Superfluid helium
- Mirror
- Trapped molecules
- Magnetic field
- Cu wire
- Ablation Laser
- Probe laser
Magnetic trap

Cryogenic cell:

- Magnetic field
- Trapped molecules
- Ablation Laser
- Probe laser
- Cell Window
- Superfluid Helium
- G-10
- Mirror
- Heatlink
- To dilution refrigerator

Energy vs. Magnetic field diagram:
- Energy increases with increasing Magnetic field.
- Spin relaxation indicated by downward arrow.

Graphical representation of the magnetic trap showing the distribution of particles in the magnetic field and the cryogenic cell components.
$V_{AB}(R, r_A, r_B) = (4\pi)^{3/2} \sum_{\lambda_A\lambda_B \lambda} V_{\lambda_A\lambda_B\lambda}(R, r_A, r_B) \times \sum_{m_{\lambda_A} m_{\lambda_B} m_\lambda} \begin{pmatrix} \lambda_A & \lambda_B & \lambda \\ m_{\lambda_A} & m_{\lambda_B} & m_\lambda \end{pmatrix} Y_{\lambda_A m_{\lambda_A}}(\hat{r}_A) Y_{\lambda_B m_{\lambda_B}}(\hat{r}_B) Y_{\lambda m_\lambda}(\hat{R})$
Trap loss...
How do electric fields affect spin relaxation?

- Induce couplings between the rotational levels ($\Delta N = 1$)
- Increase the energy gap between the rotational levels

Theory of collisions in external fields

\[ H_{\text{mol}} = -\frac{1}{2\mu_m} \frac{d^2}{dr^2} + \frac{N^2(\vec{r})}{2\mu_m r^2} + V(r) + \gamma S \cdot N - E \cdot d + 2\mu_B B \cdot S \]

\[ -Ed \cos \chi = -Ed \frac{4\pi}{3} \sum_q Y_1^*(\hat{r})Y_1(\hat{E}). \]

\[ H = -\frac{1}{2\mu R} \frac{d^2}{dR^2} R + \frac{\ell^2(\hat{R})}{2\mu R^2} + V(R, r, \theta) + H_{\text{mol}} \]

Coupled equations

\[ \left[ \frac{d^2}{dR^2} + 2\mu E_{\text{tot}} - \frac{\ell(\ell + 1)}{R^2} \right] F_{NN'NS'S\ell M_\ell}(R) \]

\[ = 2\mu \sum_{N',M',S',M',\ell',M'} \langle NM_N S M_S \ell M_\ell | V(R, r, \theta) + H_{\text{mol}} | N'M'_{N'} S'M'S' \ell' M'_\ell \rangle F_{N'M'_{N'} S'M'S' \ell' M'_\ell}(R) \]
Controlling Electronic Spin Relaxation of Cold Molecules with Electric Fields

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Spin relaxation is suppressed
Enhancement of spin relaxation

- First-order Stark effect

Enhancement of spin relaxation (a 3D view)
Spin-changing reactions

\[ \text{Na}^2S \text{ + CaH}^2\Sigma \rightarrow \text{NaH} \text{ + Ca} \]
Trap loss...
Microwave traps for polar molecules

Fig. 2. Energies of dressed states vs. applied microwave electric field strength. State labels are zero-field basis states $|\psi_{Jmn}\rangle$.

Energy levels of a diatomic molecule in a microwave field

Energy (in units of rotational constant) vs. $\mu \varepsilon_0$ (in units of rotational constant)
Energy levels of a diatomic molecules in a microwave field

Energy (in units of rotational constant)

$\mu \varepsilon_0$ (in units of rotational constant)
Energy levels of a diatomic molecule in a microwave field

\[ \text{Energy (in units of rotational constant)} \]

\[ N=0, \ n=Q \]

\[ N=0, \ n=Q-1 \]
Collisions of molecules in a microwave cavity

Molecular Hamiltonian: $H_{\text{mol}} = BN^2$

Field Hamiltonian: $H_f = \hbar \omega (aa^\dagger - \bar{n})$

Molecule - Field Hamiltonian: $H_{\text{mol},f} = -\mu \sqrt{\frac{\hbar \omega}{2\epsilon_0 V}} \cos \theta \left( a + a^\dagger \right)$

Basis set: $|NM_N\rangle|n\rangle$

The matrix elements of the molecule - field Hamiltonian:

$$\langle n|NM_N|H_{\text{mol},f}|N'M_N'\rangle|n'\rangle \sim \langle NM_N|\cos \theta|N'M_N'\rangle \times \left( \delta_{n,n'+1} + \delta_{n,n'-1} \right)$$

$$\langle NM_N|\cos \theta|N'M_N'\rangle \sim \delta_{M_N,M_N'} \left( \delta_{N,N'+1} + \delta_{N,N'-1} \right)$$
Energy levels of a diatomic molecules in a microwave field

\[ a(N=0, n=Q) + b(N=1,n=Q-1) + c(N=1,n=Q+1) \]

\[ a(N=0, n=Q-1) + b(N=1, n=Q) + c(N=1,n=Q-2) \]
Superposition states of molecules in a microwave field

\[ \Psi_{\text{“ground”}} = a|N = 0, n = \bar{N}\rangle + b|N = 1, n = \bar{N} - 1\rangle + c|N = 1, n = \bar{N} + 1\rangle + d|N = 0, n = \bar{N} - 2\rangle + e|N = 0, n = \bar{N} + 2\rangle \]

\( \bar{N} \) is the average number of photons

Off-resonant light: \( w = 0.01B \).

<table>
<thead>
<tr>
<th>( \mu\epsilon_0 = 0.1B )</th>
<th>( \mu\epsilon_0 = 0.3B )</th>
<th>( \mu\epsilon_0 = 0.5B )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( a = 0.9994 )</td>
<td>( a = 0.9637 )</td>
<td>( a = 0.7497 )</td>
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<tr>
<td>( b = 0.0143 )</td>
<td>( a = 0.0397 )</td>
<td>( a = 0.0178 )</td>
</tr>
<tr>
<td>( c = 0.0145 )</td>
<td>( c = 0.0471 )</td>
<td>( c = 0.0441 )</td>
</tr>
<tr>
<td>( d = 0.0207 )</td>
<td>( a = 0.182 )</td>
<td>( a = 0.445 )</td>
</tr>
<tr>
<td>( e = 0.0209 )</td>
<td>( a = 0.184 )</td>
<td>( a = 0.448 )</td>
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Collisionally induced transitions between field-dressed states

Cross section (Å²)

Collision energy: 0.3 cm⁻¹

με₀ (in units of rotational constant)
Change of a shape resonance in the presence of microwave radiation

Cross section for elastic scattering (Å$^2$)

Collision energy (cm$^{-1}$)

ω = 1.9 B; $\mu \varepsilon_0 =$ 0.5 B

no field
Threshold laws for collisions in 2D
Threshold laws for collisions in 2D

In 3D, we have Wigner’s threshold laws for elastic scattering:

\[ \text{collision cross section} \sim v^{2l+2l'} \]

In 2D, there is no \( l \). The Hamiltonian is

\[ H = -\frac{1}{2\mu \rho} \frac{d}{d\rho} \rho \frac{d}{d\rho} + \frac{l_z^2}{2\mu \rho^2} + H_{\text{as}} + V(\rho), \]

The role of \( l \) is played by \( m \), the projection quantum number.

How are the Wigner’s threshold laws modified, if we confine the system in 2D?
Let’s look at low-energy scattering:

In 3D, the Schrödinger’s equation is

\[
\left[ -\frac{1}{2\mu R^2} \frac{d}{dR} R^2 \frac{d}{dR} + \frac{l(l+1)}{2\mu R^2} - 2\mu V(R) \right] \psi(k, R) = -k^2 \psi(k, R)
\]

Consider first the solution to this equation with \( V = 0 \) and \( k = 0 \):

\[
\left[ -\frac{1}{2\mu R^2} \frac{d}{dR} R^2 \frac{d}{dR} + \frac{l(l+1)}{2\mu R^2} \right] \psi(k, R) = 0
\]

Let’s look for the solution in the form \( \psi(R, k = 0) = \text{const} R^s \)

The derivative:

\[
\frac{1}{2\mu R^2} \frac{d}{dR} R^2 \frac{d}{dR} R^s = s(s + 1) R^s
\]

Hence, \( s(s + 1) = l(l + 1) \) or \( s = l \) and \( s = -(l + 1) \).
A general solution at $k = 0$ is therefore

$$\psi(k = 0, R) = A_1 R^l + A_2 R^{-(l+1)}$$

Now, for $k \neq 0$, we have a Bessel equation and the general solution

$$\psi(k, R) = A j_l(kR) + B \eta_l(kR)$$

which can be re-written at small $k$ as

$$\psi(k, R) = (kR)^l + \tan \delta_l(kR)^{-(l+1)}$$

For smooth and continuous matching to $k = 0$, we must require

$$\tan \delta_l \sim k^{2l+1}$$

which gives after some manipulation:

$$\text{elastic scattering cross section} \sim k^{4l}$$
Repeating this derivation for 2D, we get

\[ \text{cross section} \sim \frac{1}{k \ln^2 k}, \quad \text{when } m = 0 \]

Using the formalism of Wigner, it is also possible to get the off-diagonal cross sections:

\[ \text{cross section for } m = 0 \rightarrow m' \text{ transitions} \sim k^{2|m|-1} \frac{1}{\ln^2 k} \]

and

\[ \text{cross section for } m > 0 \rightarrow m' > 0 \text{ transitions} \sim k^{2|m|+2|m'|-1} \]
Threshold collision laws

<table>
<thead>
<tr>
<th>Transition</th>
<th>3D</th>
<th>2D</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s$-wave elastic</td>
<td>$\sigma = \text{const}$</td>
<td>$\sigma \sim \frac{1}{k \ln^2 k}$</td>
</tr>
<tr>
<td>$s$-wave to non-$s$-wave</td>
<td>$\sigma \sim k^{2l'}$</td>
<td>$\sigma \sim k^2</td>
</tr>
<tr>
<td>non-$s$-wave to non-$s$-wave</td>
<td>$\sigma \sim k^{2l+2l'}$</td>
<td>$\sigma \sim k^2</td>
</tr>
</tbody>
</table>

Why is this interesting?
Consider ultracold collisions of molecules in 2D.

Angular momentum transfer of molecules - such as spin relaxation - must be accompanied by changes of $m$, if the magnetic field axis is directed perpendicularly to the plane of confinement.

If the magnetic field axis is tilted, collisions do not have to conserve the total angular momentum projection.

Inelastic angular momentum transfer - such as spin relaxation - will then be much more efficient if the axis of the external field is not perpendicular to the plane of confinement.
Suppressed collisional spin relaxation

Enhanced collisional spin relaxation
Conclusions

- Electric fields may suppress collisional loss from a magnetic trap
- Evaporative cooling in a microwave trap might be difficult (?)
- Microwave fields modify interactions of cold molecules
- Elastic and Inelastic Two-Body Collisions are modified in 2D
Outlook: Collision Physics and Ultracold Molecules

Experiments with cold molecules may

- confirm or disprove Wigner’s threshold laws
  → more insight into long-range interactions

- elucidate rates for chemical reactions at ultracold Ts
  → ultracold chemistry → lots of applications

- demonstrate the possibility of controlling chemical reactions
  → controlled chemistry → lots of applications

- make coherent control of bimolecular reactions possible
  → controlled chemistry

- provide new testground for statistical theories of molecules
  → new reaction rate theories
References

R. V. Krems, PRL 93, 013201 (2004).

Reviews