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

Roman Krems
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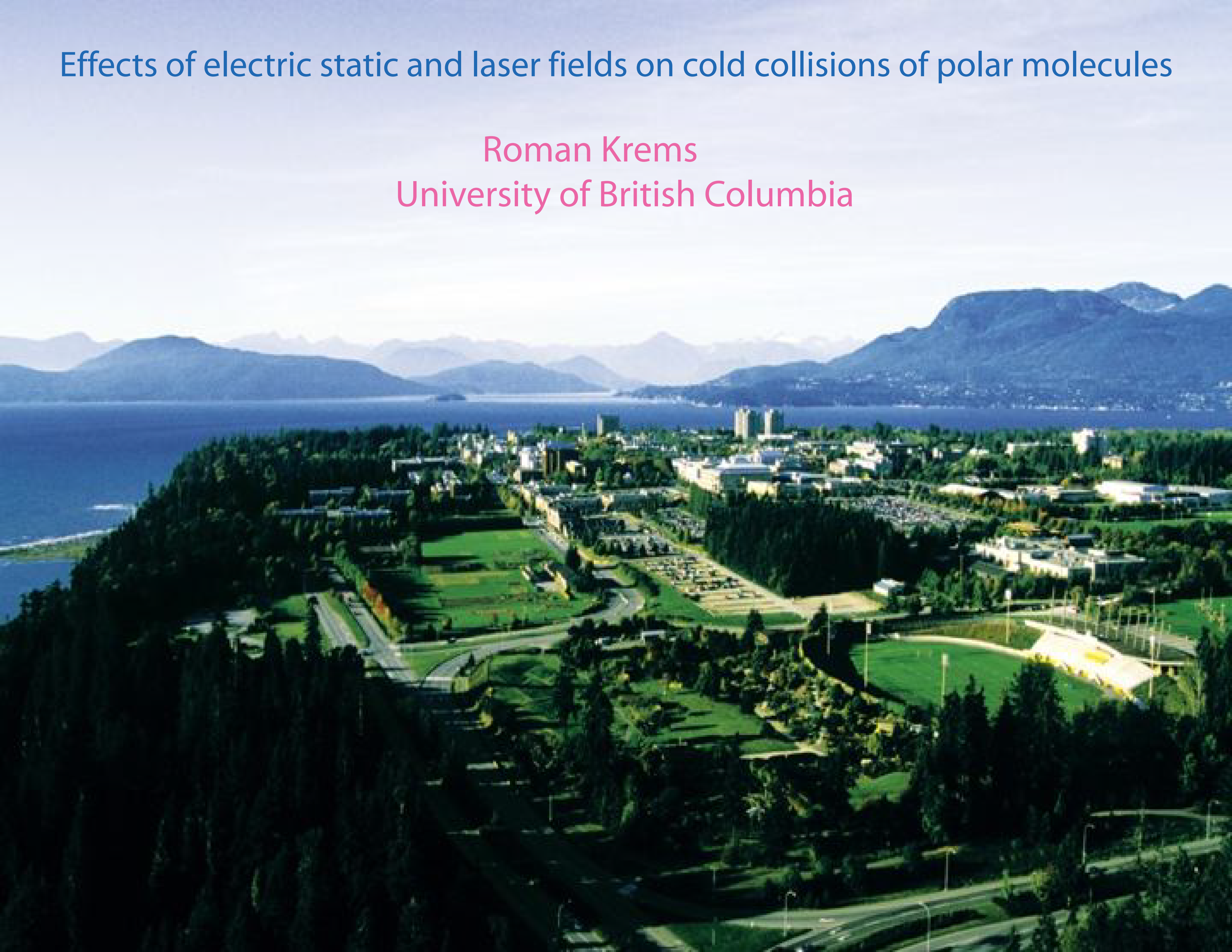

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Outline

- 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

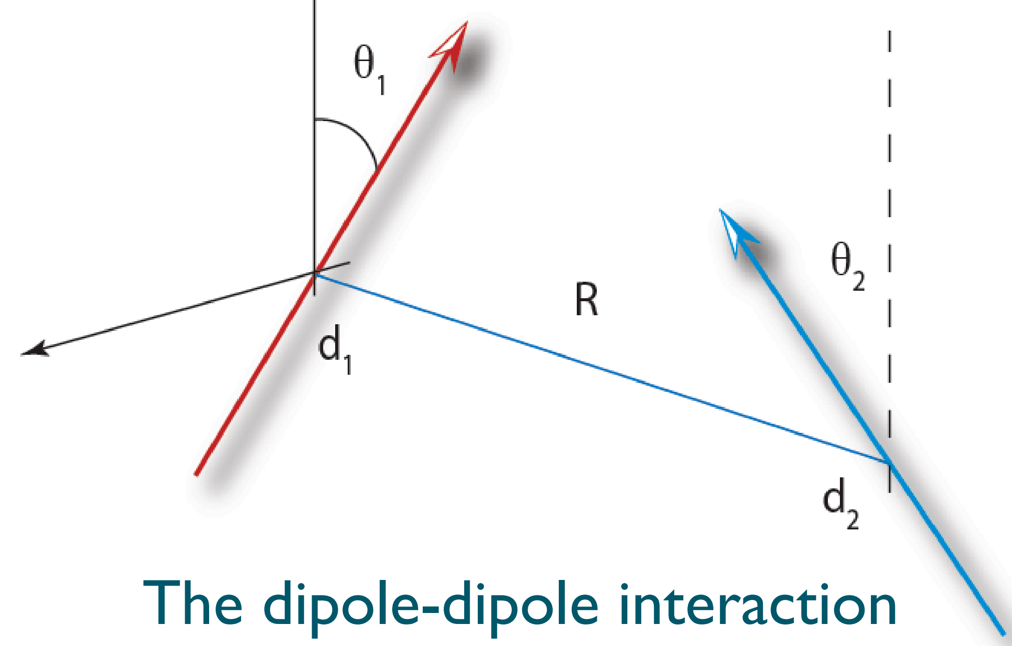
- Threshold laws for collisions in 2D

- Outlook: Collision physics and ultra-cold molecules

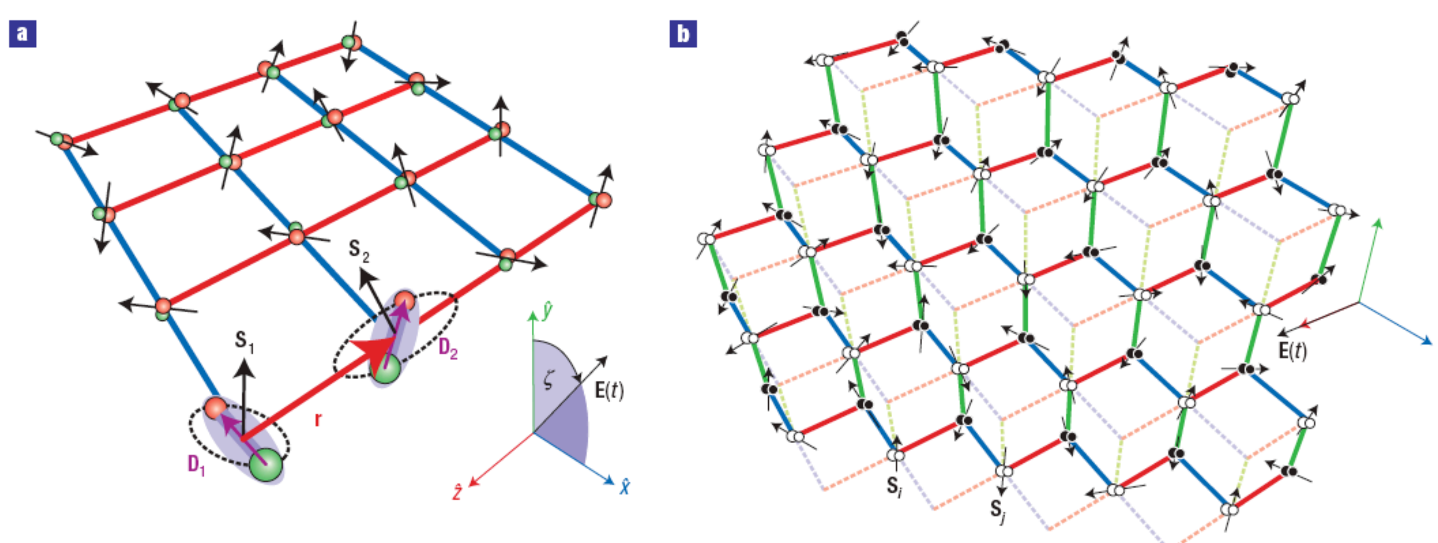
Applications of cold molecules

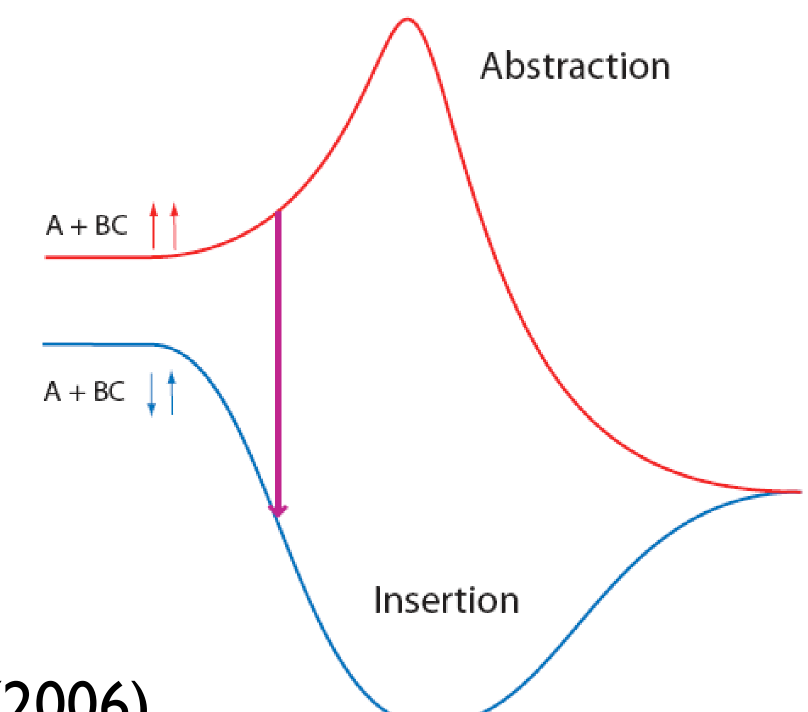
- Tunable interactions
- Quantum computation
- High-precision spectroscopy
- Controlled chemistry

$$V_{\rm dip} = \frac{\mathbf{d}_1 \cdot \mathbf{d}_2 - 3(\mathbf{R}\cdot\mathbf{d}_1)(\mathbf{R}\cdot\mathbf{d}_2)}{R^3}$$



The dipole-dipole interaction



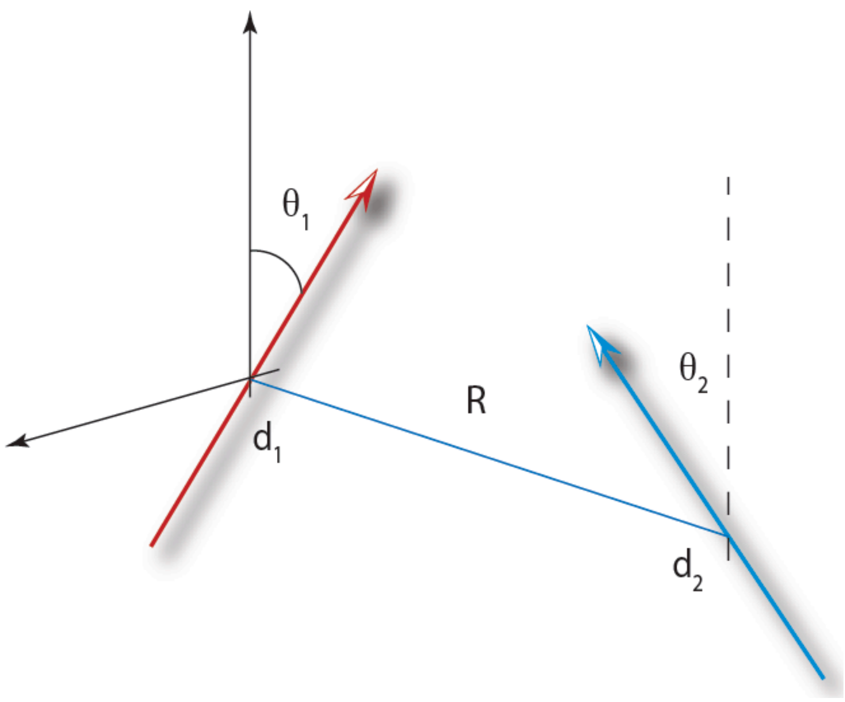


A. Micheli, G. K. Brennen, and P. Zoller, Nature Phys. **2**, 341 (2006)

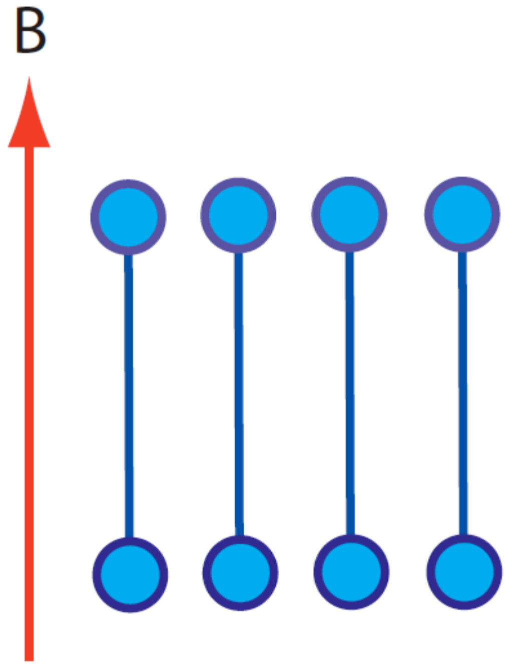
Cold molecules

- Tunable interactions

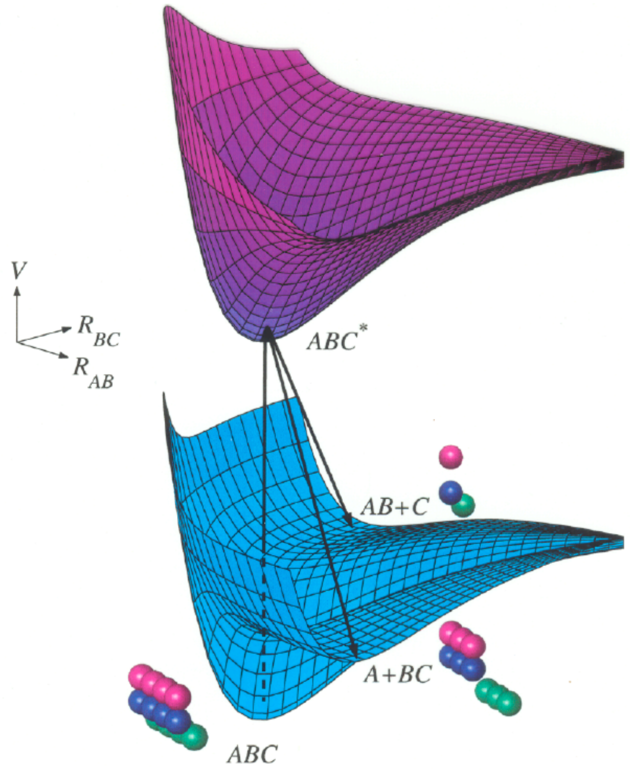
$$V_{\mathrm{dip}} = \frac{\mathbf{d}_1 \cdot \mathbf{d}_2 - 3(\mathbf{R} \cdot \mathbf{d}_1)(\mathbf{R} \cdot \mathbf{d}_2)}{R^3}$$



- Orientation and alignment

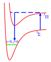


- Controlled chemistry

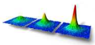


How to create ultracold molecules?

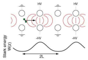
How to create ultracold molecules?



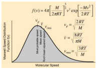
Photoassociation of ultracold atoms
(Stwalley, DeMille, Bigelow, Heinzen, Masnou-Seeuws, ...)



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

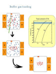


Stark deceleration of molecular beams
(Meijer, Barker, Peters, Friedrich, ...)

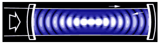


Skimming
(Abraham, Shafer-Ray, ...)

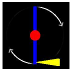
Free expansion
(Gupta, Friedrich, Hershbach, ...)



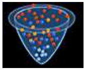
Buffer gas loading
(Doyle, Peters, ...)



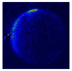
Optical dipole force slowing
(DeMille, ...)



Mechanical slowing
(Gupta, Hershbach, ...)

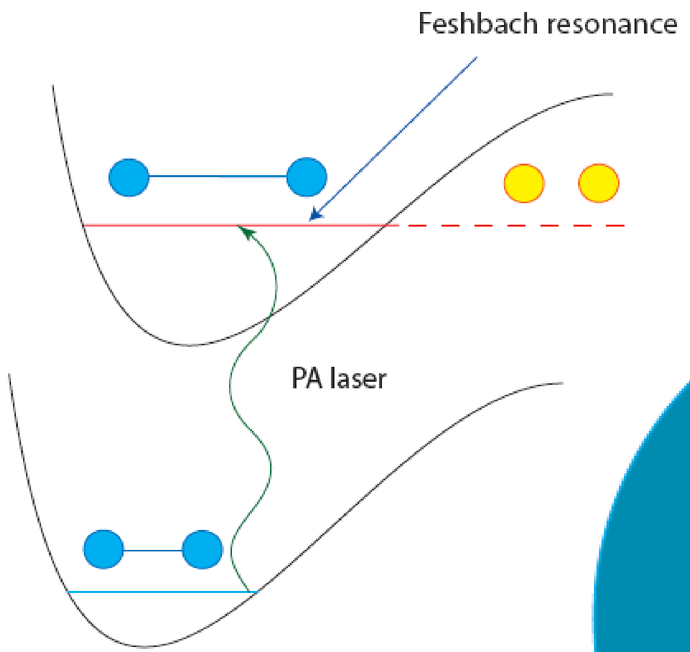


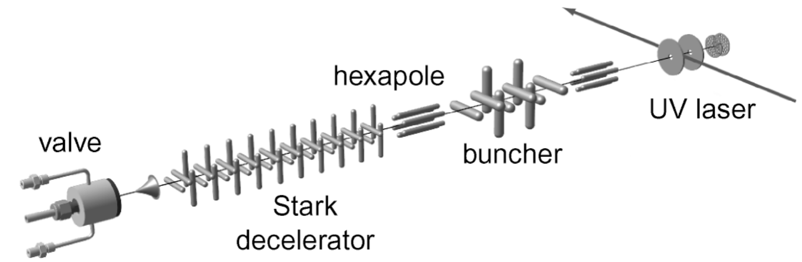
Sympathetic cooling by collisions with ultracold atoms
(Meijer, ...)



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

Experimental methods for cooling molecules





Coherent control of dynamics

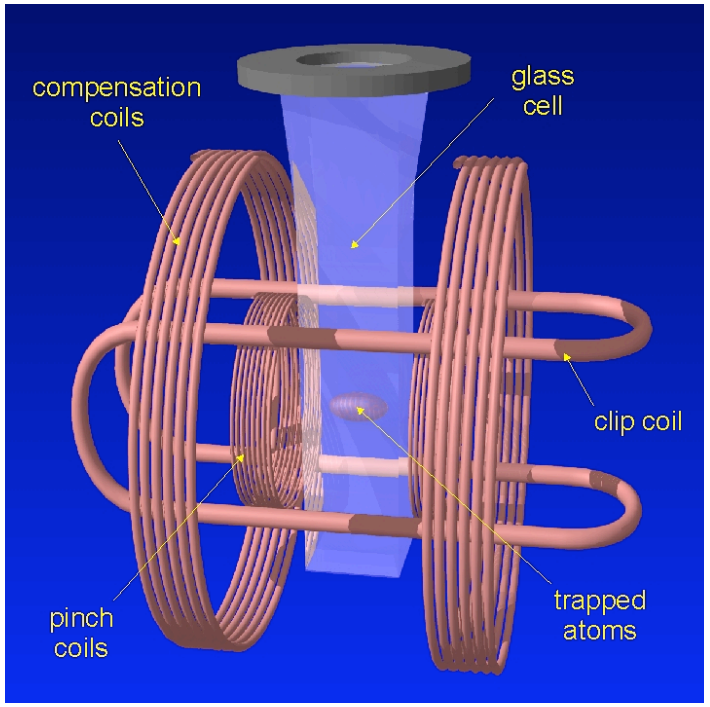
Li_2, RbCs

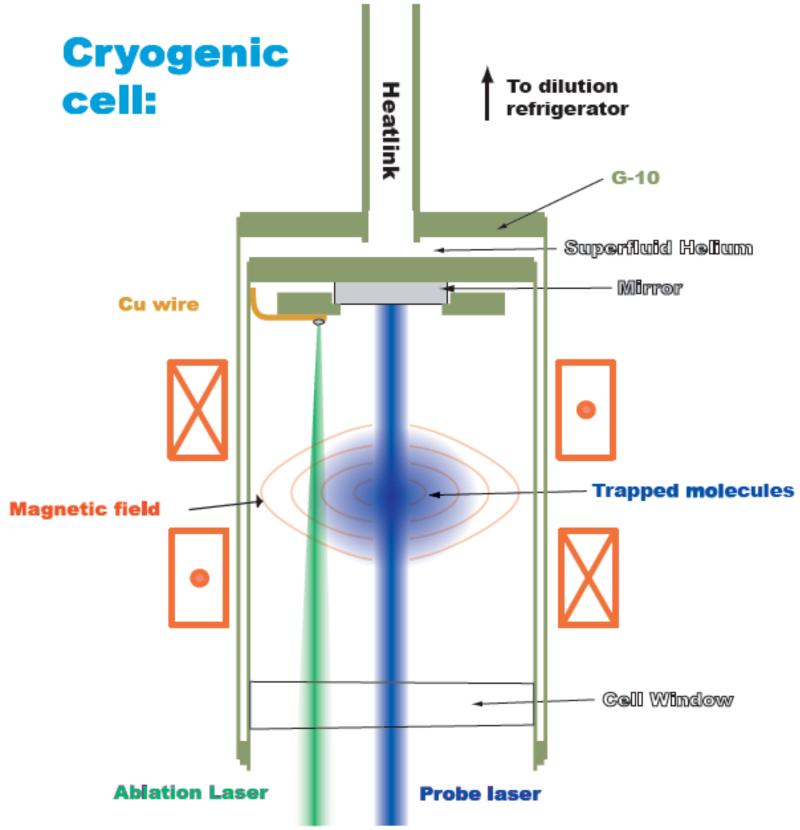
Testing fundamental symmetries of nature

YbF, PbO

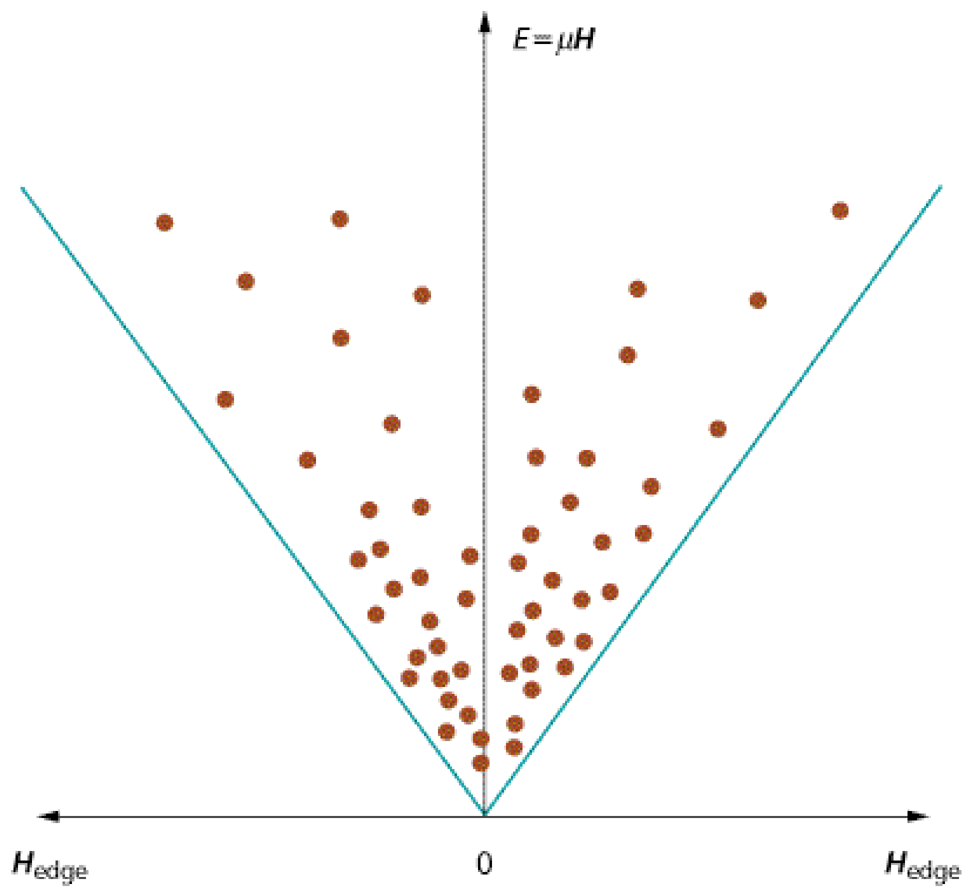
Quantum computation

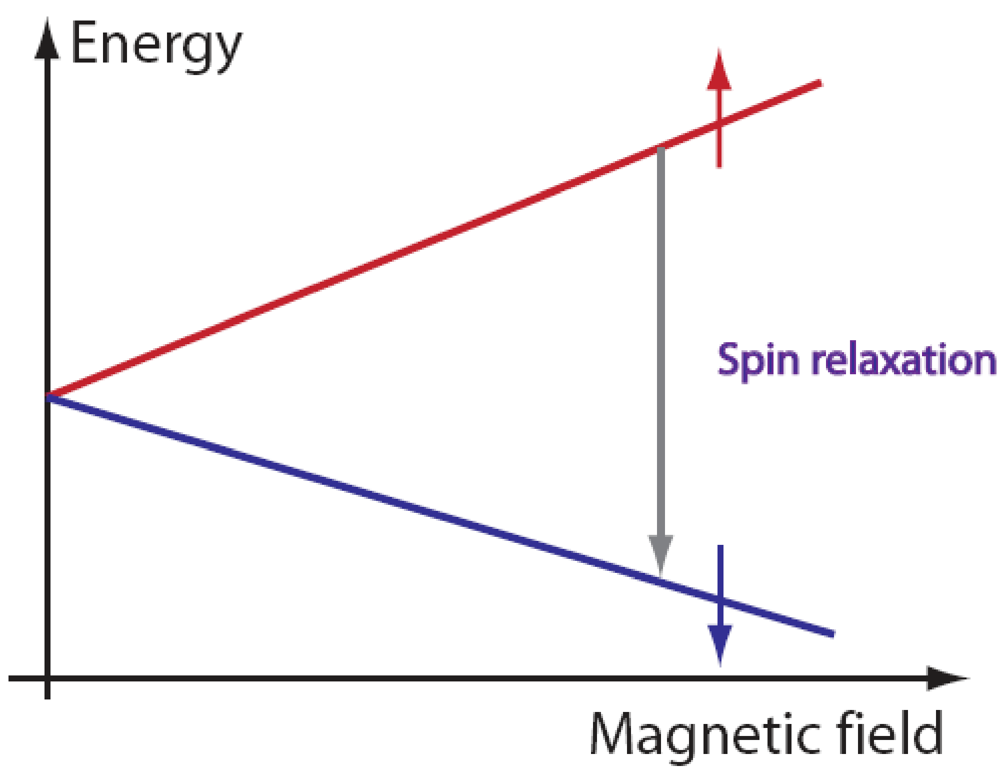
CaF, CaCl, MgCl

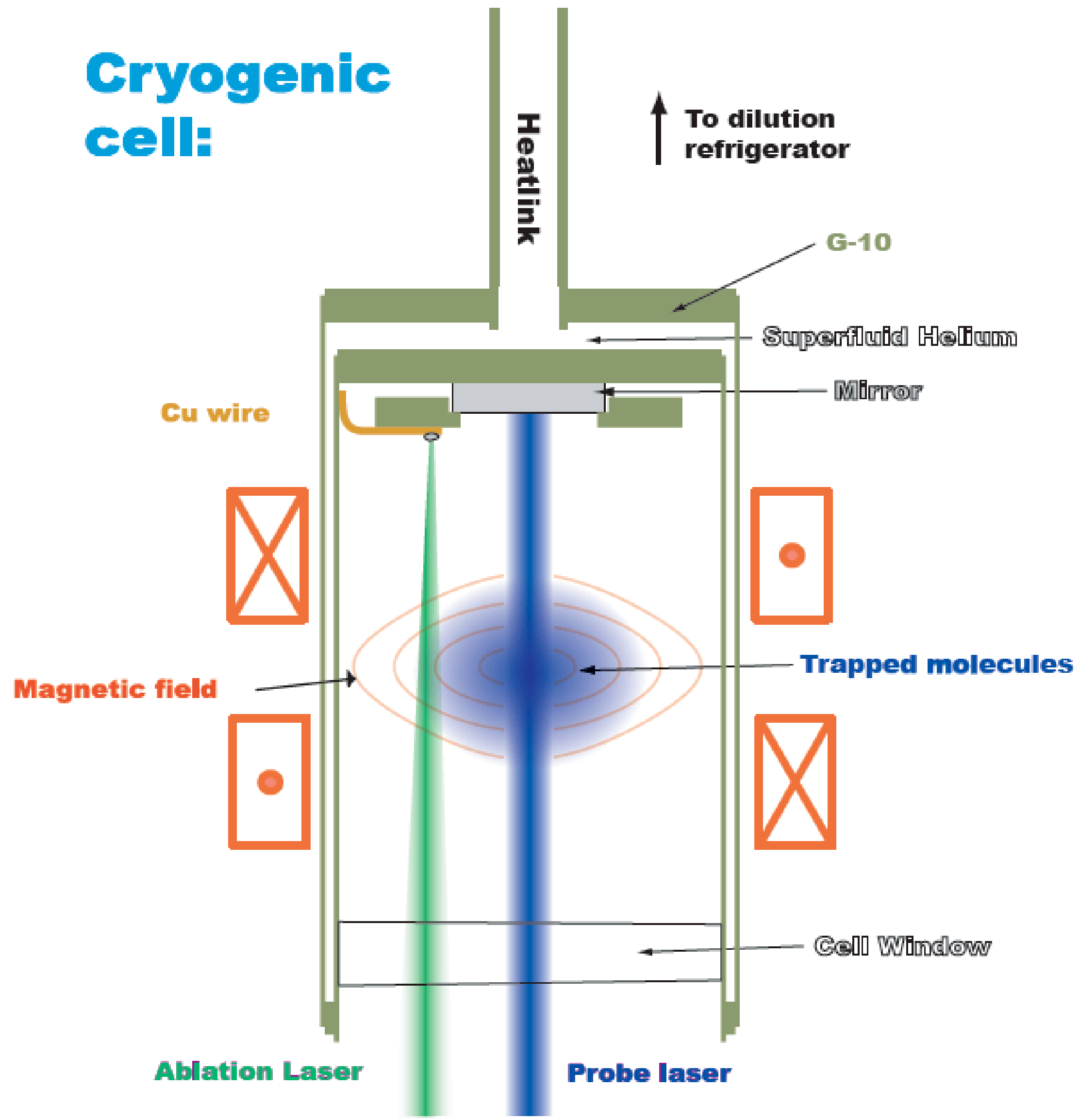


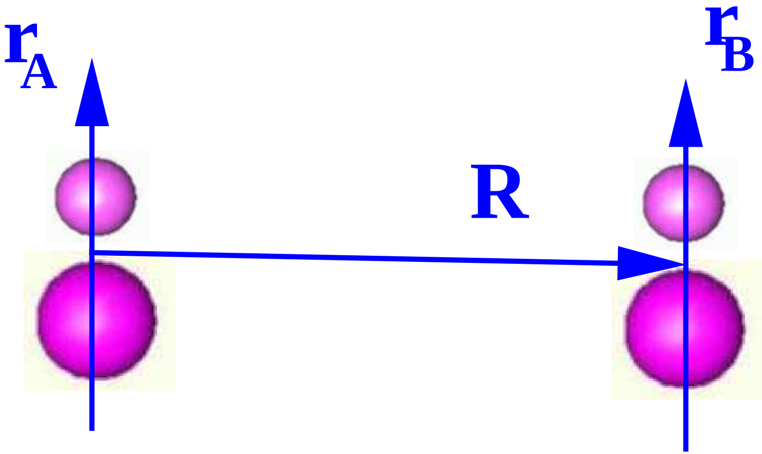


Magnetic trap



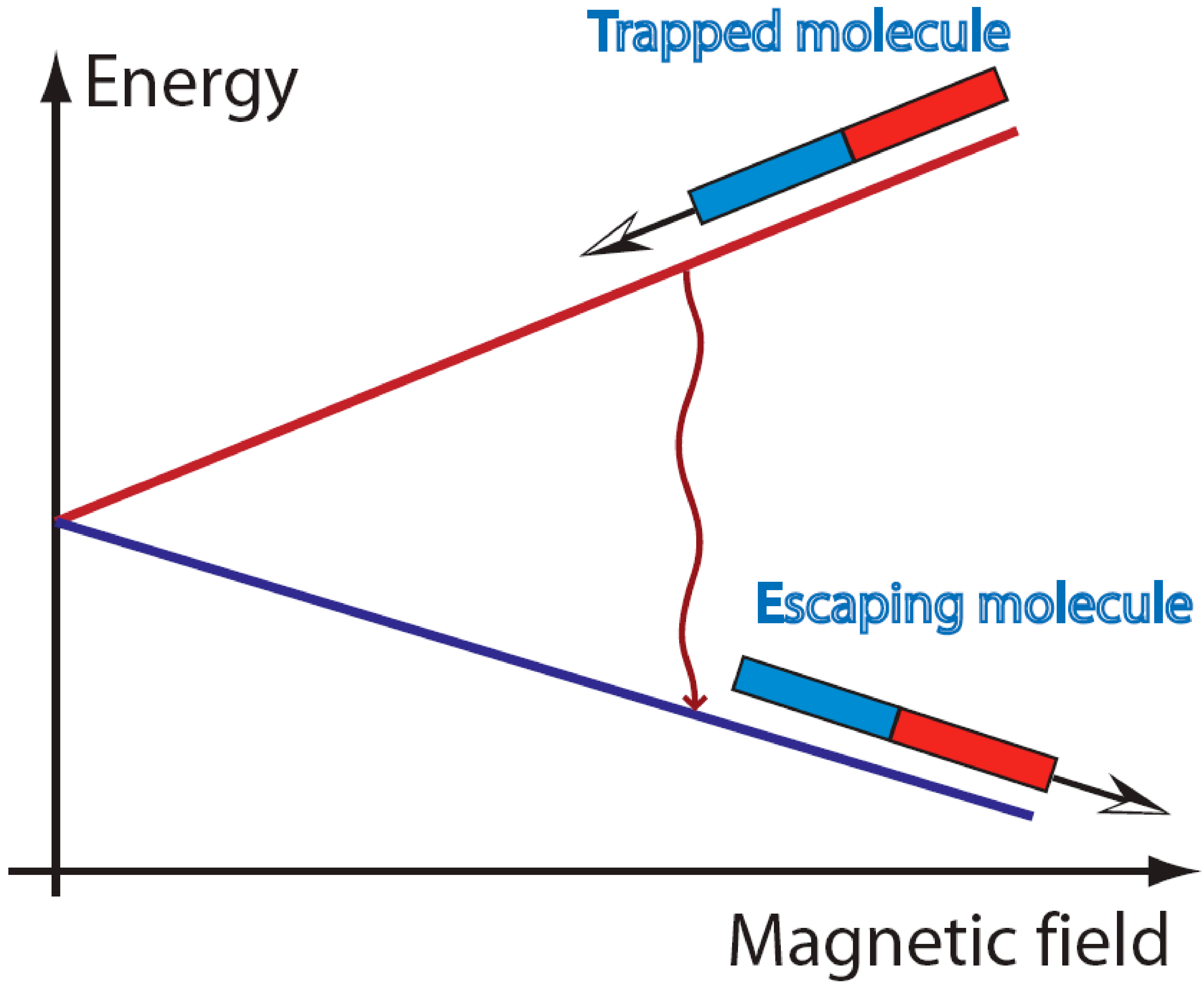






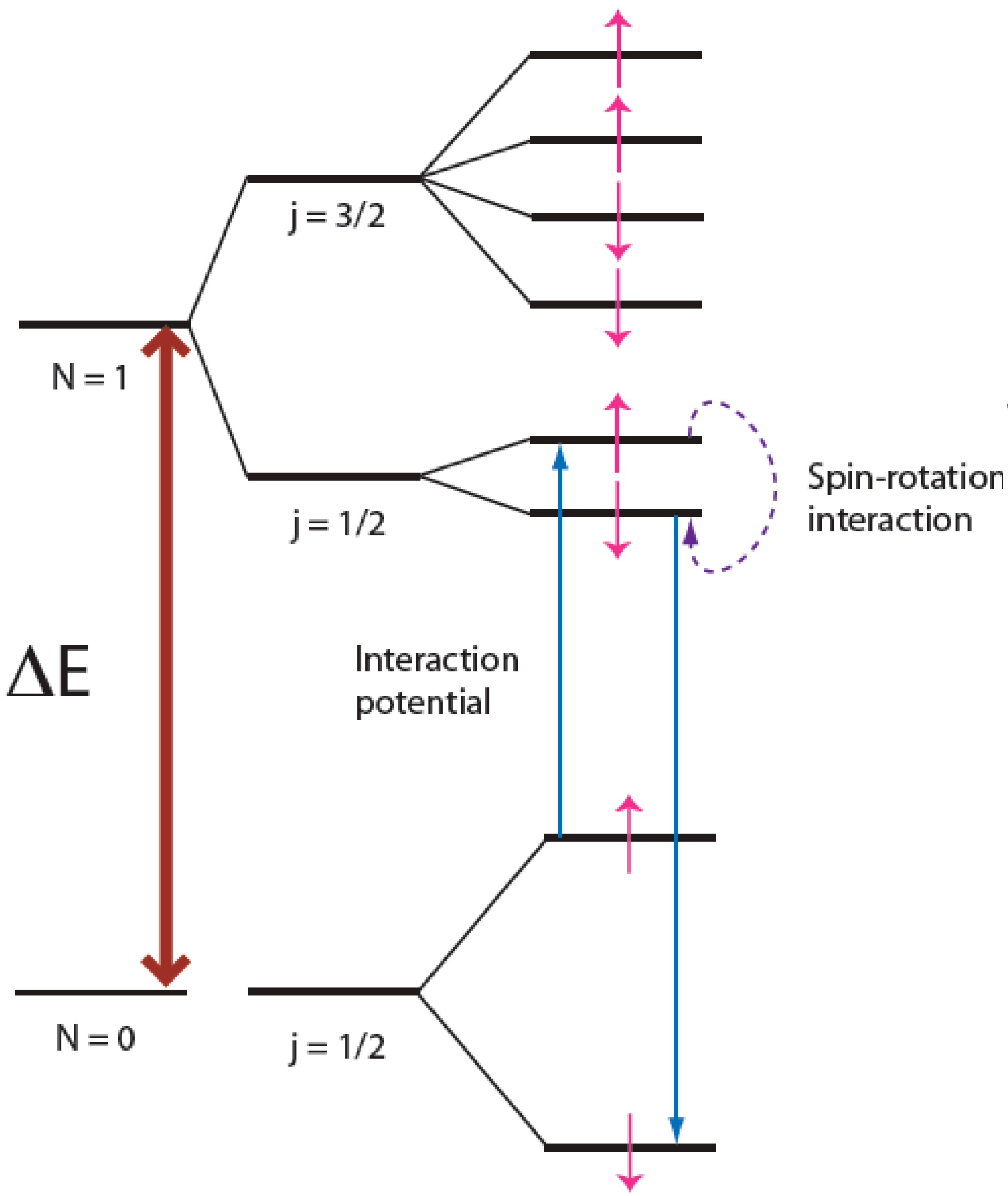
$$V_{\mathrm{AB}}(\mathbf{R},\mathbf{r}_{\mathrm{A}},\mathbf{r}_{\mathrm{B}}) = (4\pi)^{3/2} \sum_{\lambda_{\mathrm{A}}\lambda_{\mathrm{B}}\lambda} V_{\lambda_{\mathrm{A}}\lambda_{\mathrm{B}}\lambda}(R, r_{\mathrm{A}}, r_{\mathrm{B}})$$
$$\times \sum_{m_{\lambda_{\mathrm{A}}} m_{\lambda_{\mathrm{B}}} m_{\lambda}} \begin{pmatrix} \lambda_{\mathrm{A}} & \lambda_{\mathrm{B}} & \lambda \\ m_{\lambda_{\mathrm{A}}} & m_{\lambda_{\mathrm{B}}} & m_{\lambda} \end{pmatrix} Y_{\lambda_{\mathrm{A}} m_{\lambda_{\mathrm{A}}}}(\hat{r}_{\mathrm{A}}) Y_{\lambda_{\mathrm{B}} m_{\lambda_{\mathrm{B}}}}(\hat{r}_{\mathrm{B}}) Y_{\lambda m_{\lambda}}(\hat{R})$$

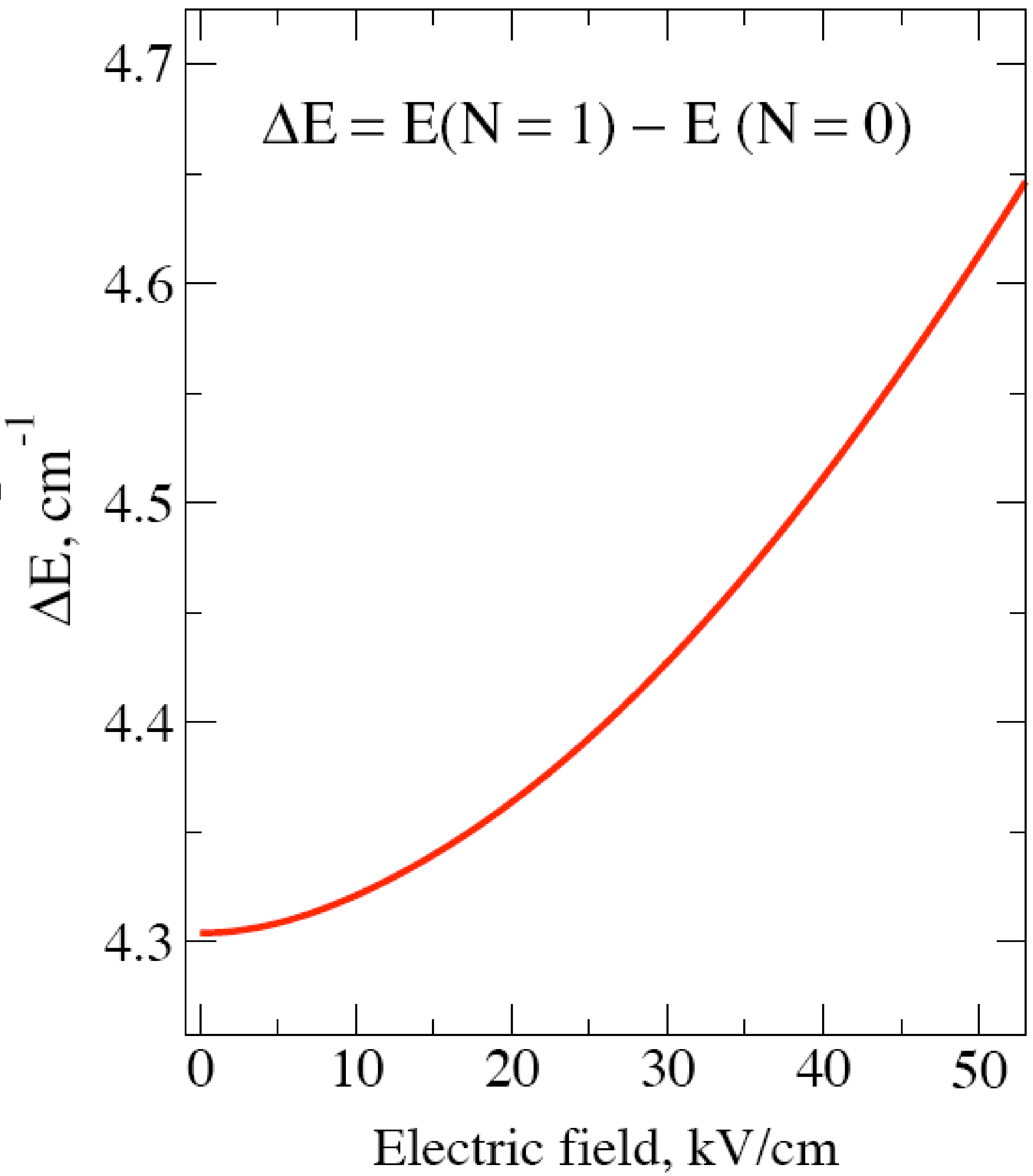
Trap loss...


Trapped molecule
Energy
Escaping molecule
Magnetic field

How do electric fields affect spin relaxation?

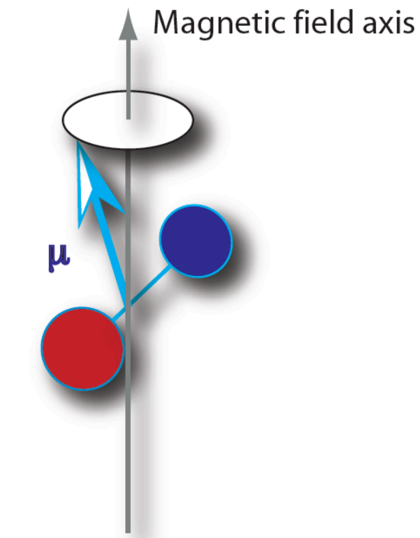
- Induce couplings between the rotational levels ($\Delta N = 1$)
- Increase the energy gap between the rotational levels



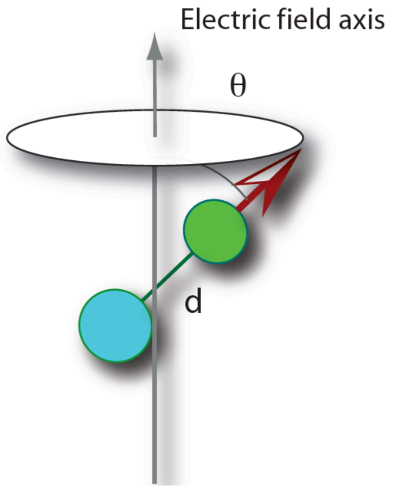


R.V. Krems, A.Dalgarno, N.Balakrishnan, and G.C. Groenenboom, PRA **67**, 060703(R) (2003)

Theory of collisions in external fields

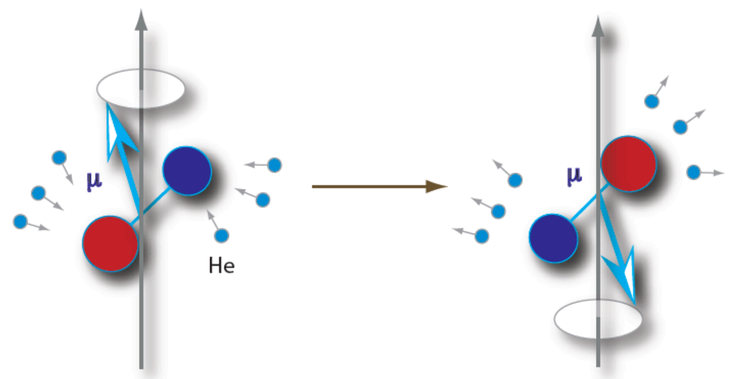


$$H_{\mathrm{mol}} = -\frac{1}{2\mu_m}\frac{d^2}{dr^2} + \frac{\boldsymbol{N}^2(\hat{r})}{2\mu_m r^2} + V(r) + \gamma \boldsymbol{S}\cdot\boldsymbol{N} - \boldsymbol{E}\cdot\boldsymbol{d} + 2\mu_B \boldsymbol{B}\cdot\boldsymbol{S}$$



$$-Ed\cos\chi = -Ed\frac{4\pi}{3}\sum_q Y^*_{1q}(\hat{r})Y_{1q}(\hat{E}).$$

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



Coupled equations

$$\left[\frac{d^2}{dR^2} + 2\mu E_{\mathrm{tot}} - \frac{\ell(\ell+1)}{R^2}\right] F_{NM_NSM_S\ell M_\ell}(R)$$

$$= 2\mu \sum_{N',M_N',S',M_S',\ell',M_\ell'} \langle NM_NSM_S\ell M_\ell | V(R,r,\theta) + H_{\mathrm{mol}} | N'M_N'S'M_S'\ell' M_\ell' \rangle F_{N'M_N'S'M_S'\ell' M_\ell'}(R)$$

PRL **97,** 083201 (2006) PHYSICAL REVIEW LETTERS week ending 25 AUGUST 2006

Controlling Electronic Spin Relaxation of Cold Molecules with Electric Fields

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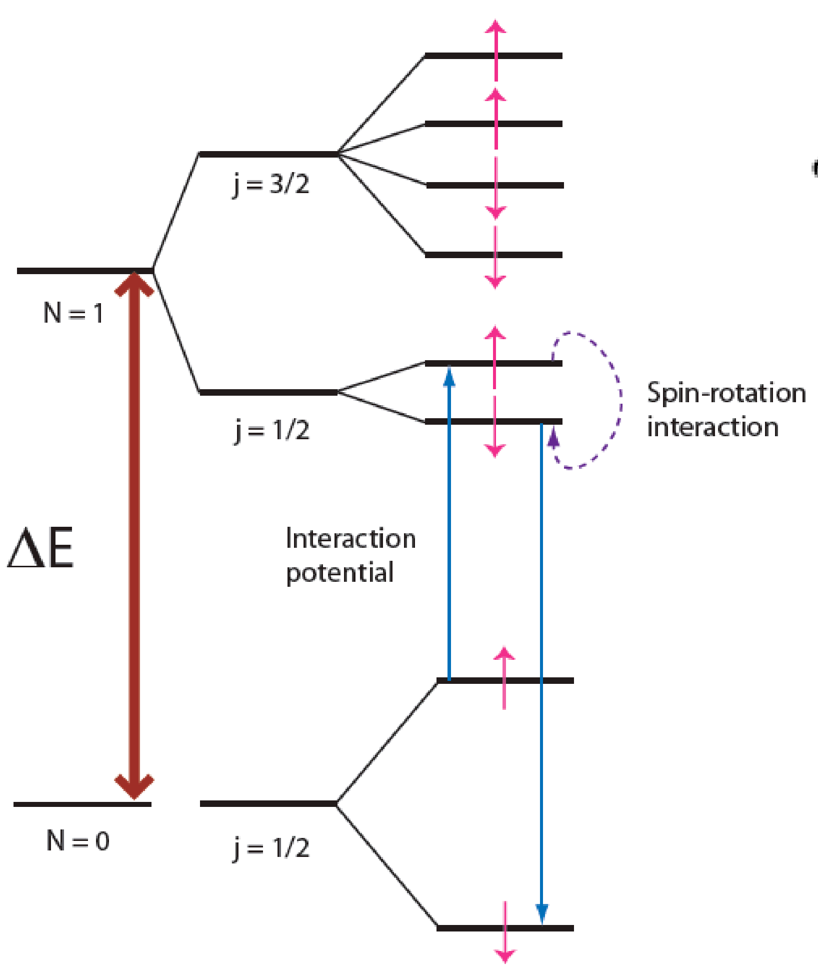
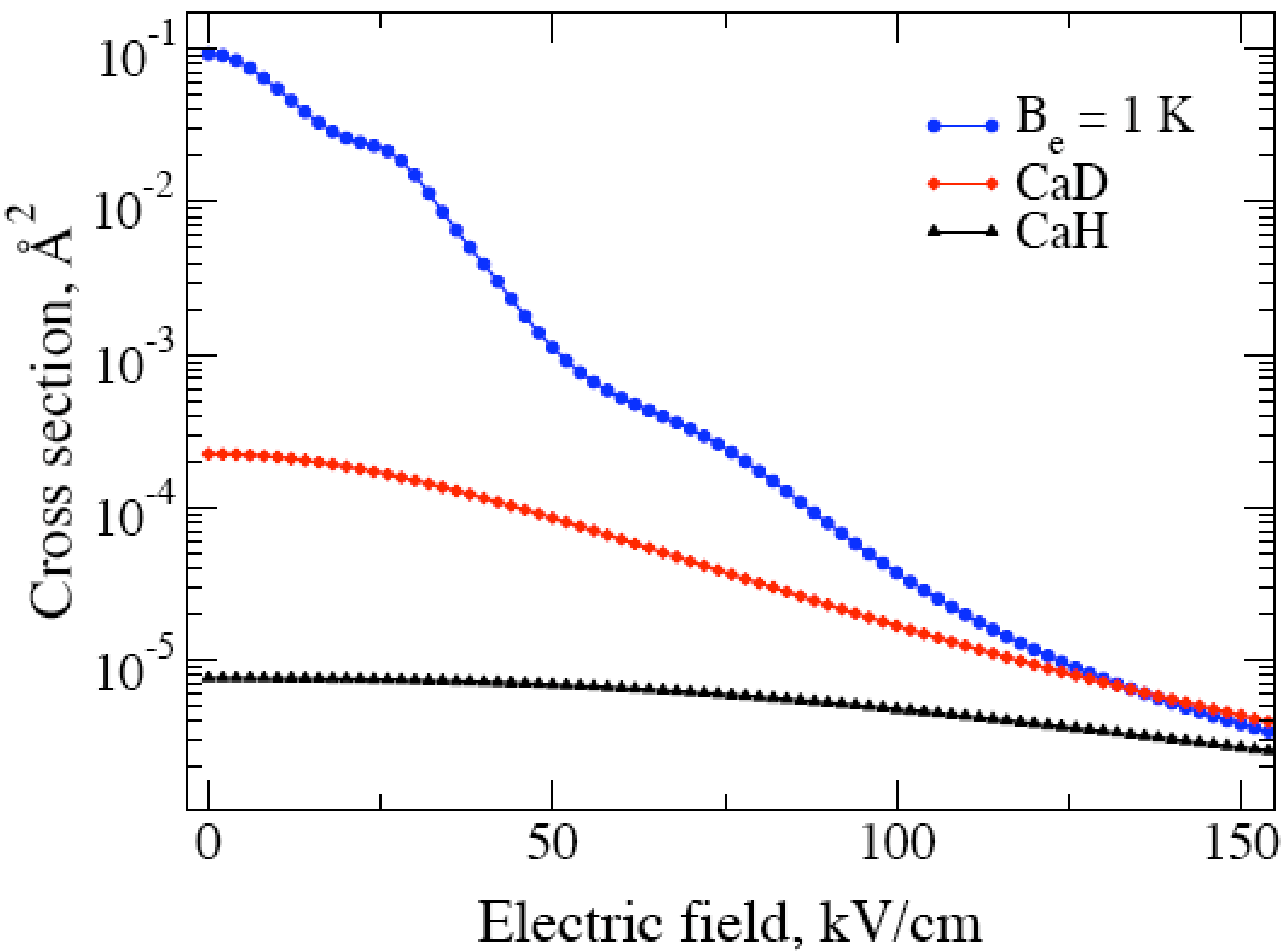
(Received 3 June 2006; published 22 August 2006)

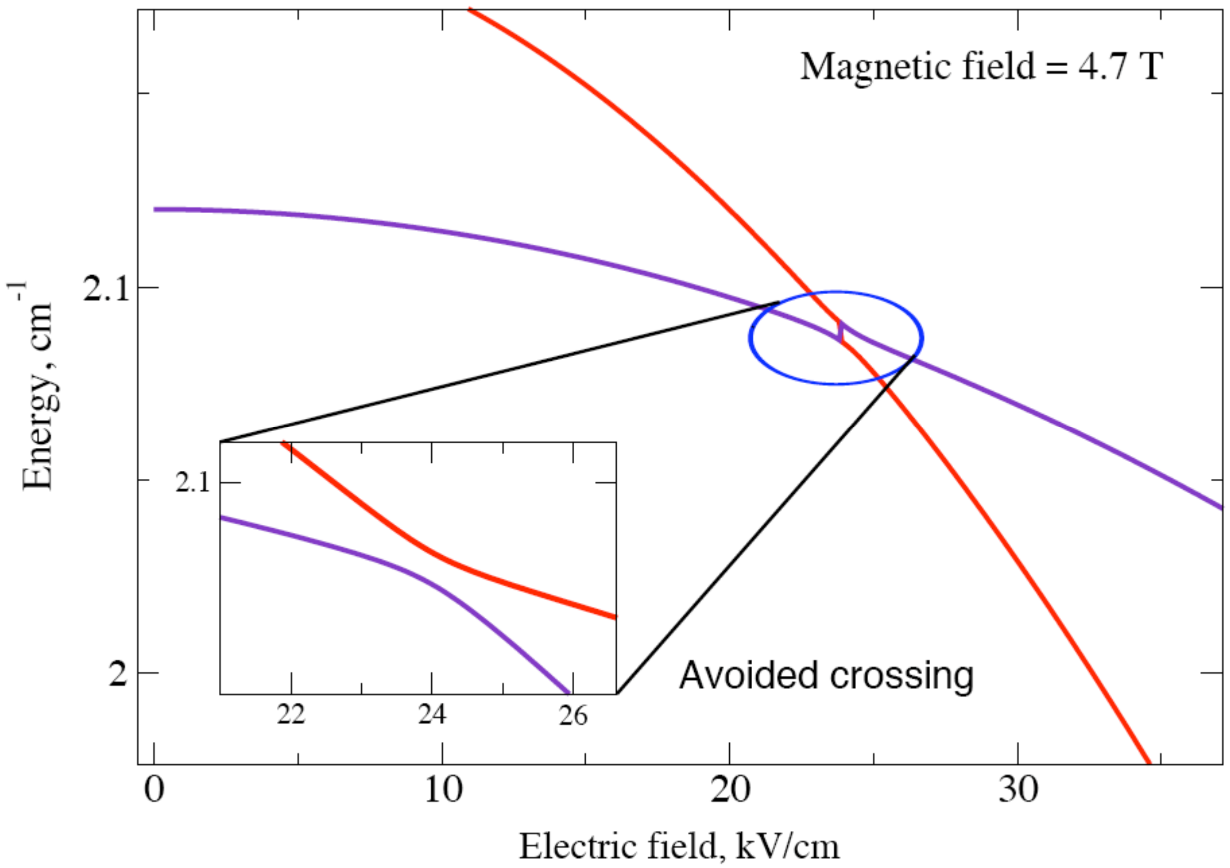
Spin relaxation is suppressed

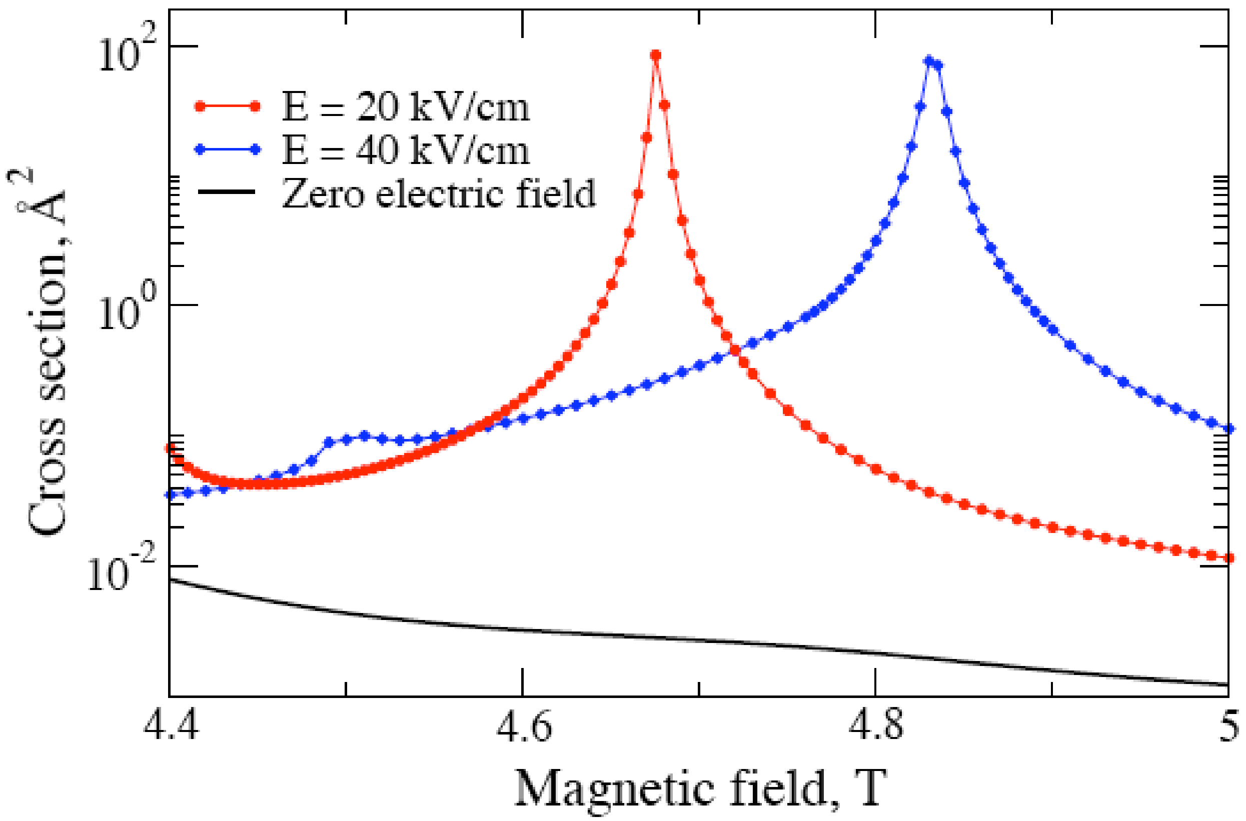


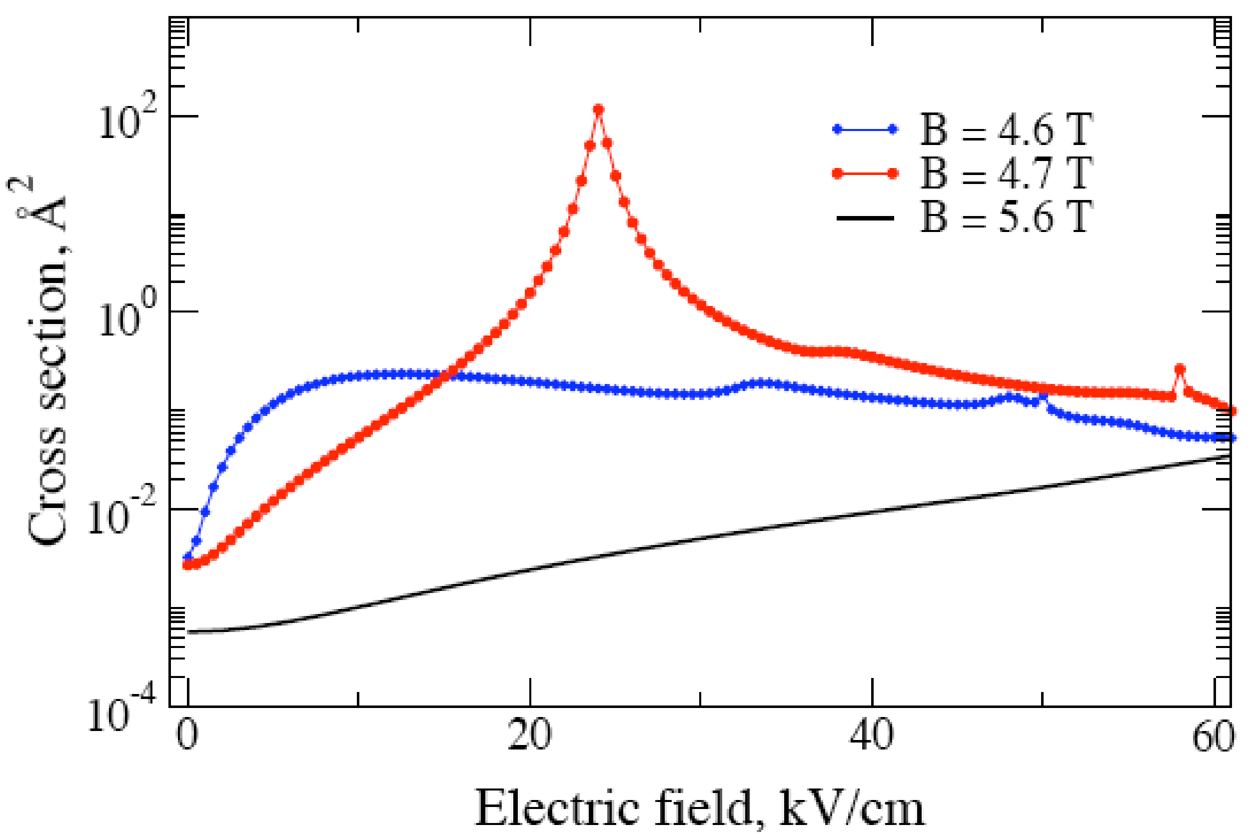


Enhancement of spin relaxation

- First-order Stark effect

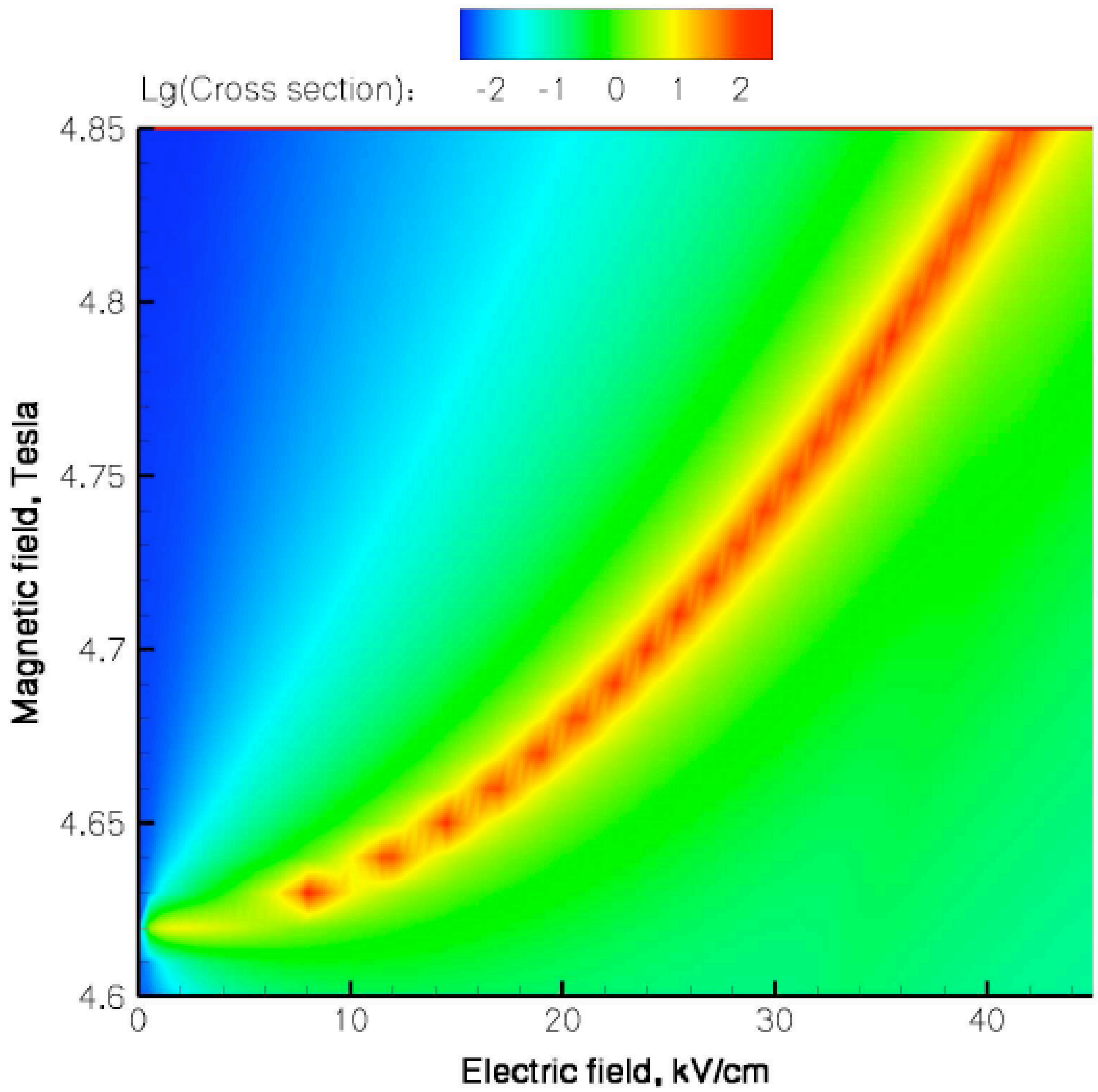




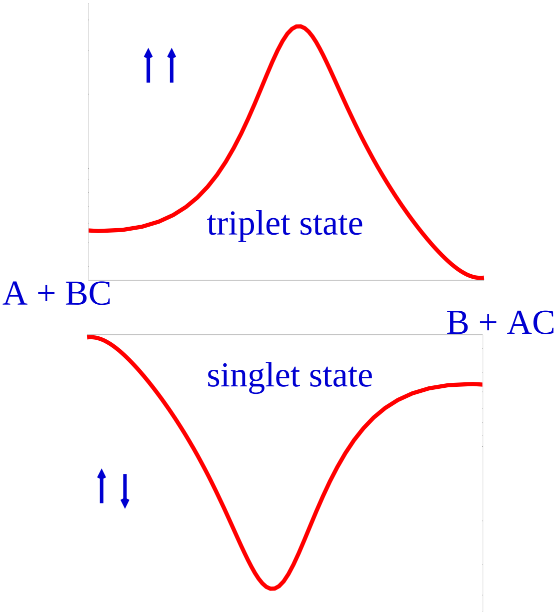


T.V. Tscherbul and R.V. Krems, PRL **97**, 083201 (2006)

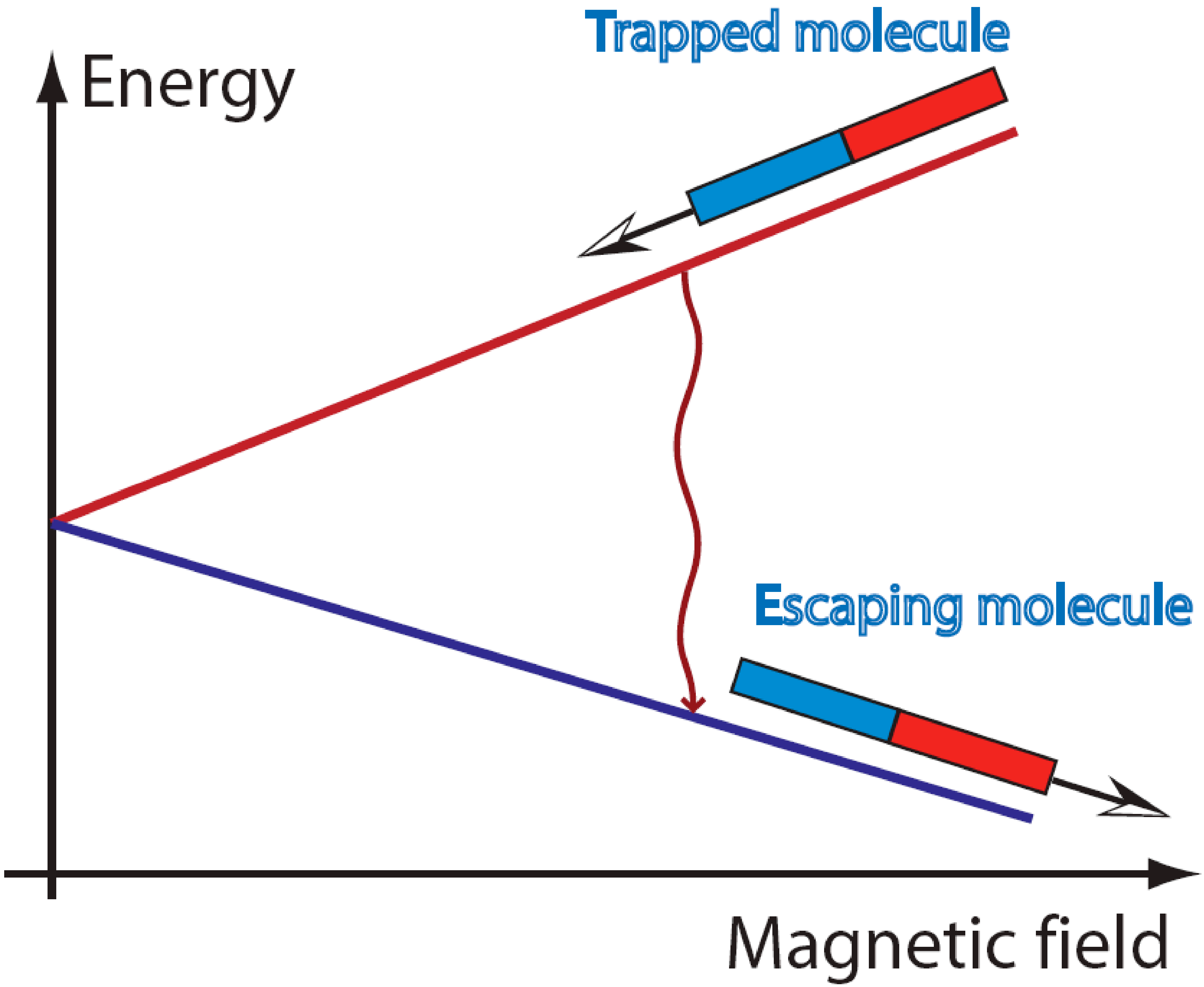
Enhancement of spin relaxation (a 3D view)



$$\mathrm{Na}(^2S) + \mathrm{CaH}(^2\Sigma) \to \mathrm{NaH} + \mathrm{Ca}$$



Trap loss...


Trapped molecule
Energy
Escaping molecule
Magnetic field

Microwave traps for polar molecules

D. DeMille et al.: Microwave traps for cold polar molecules 379

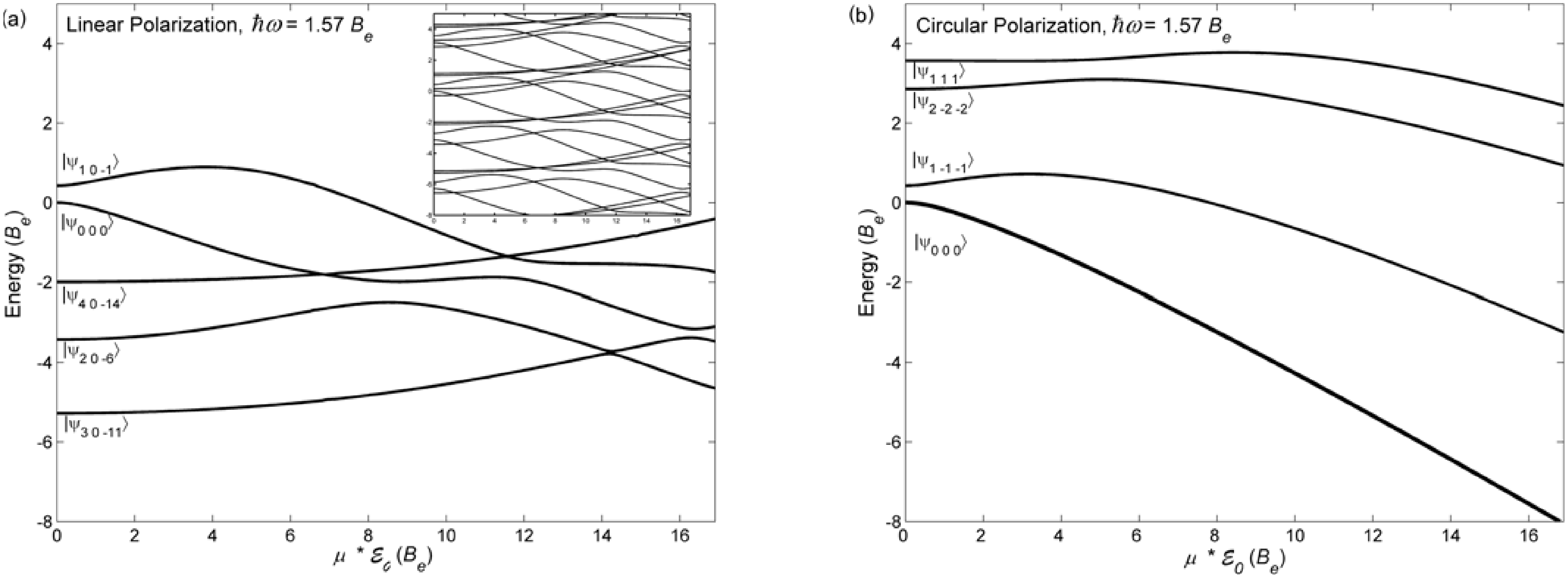
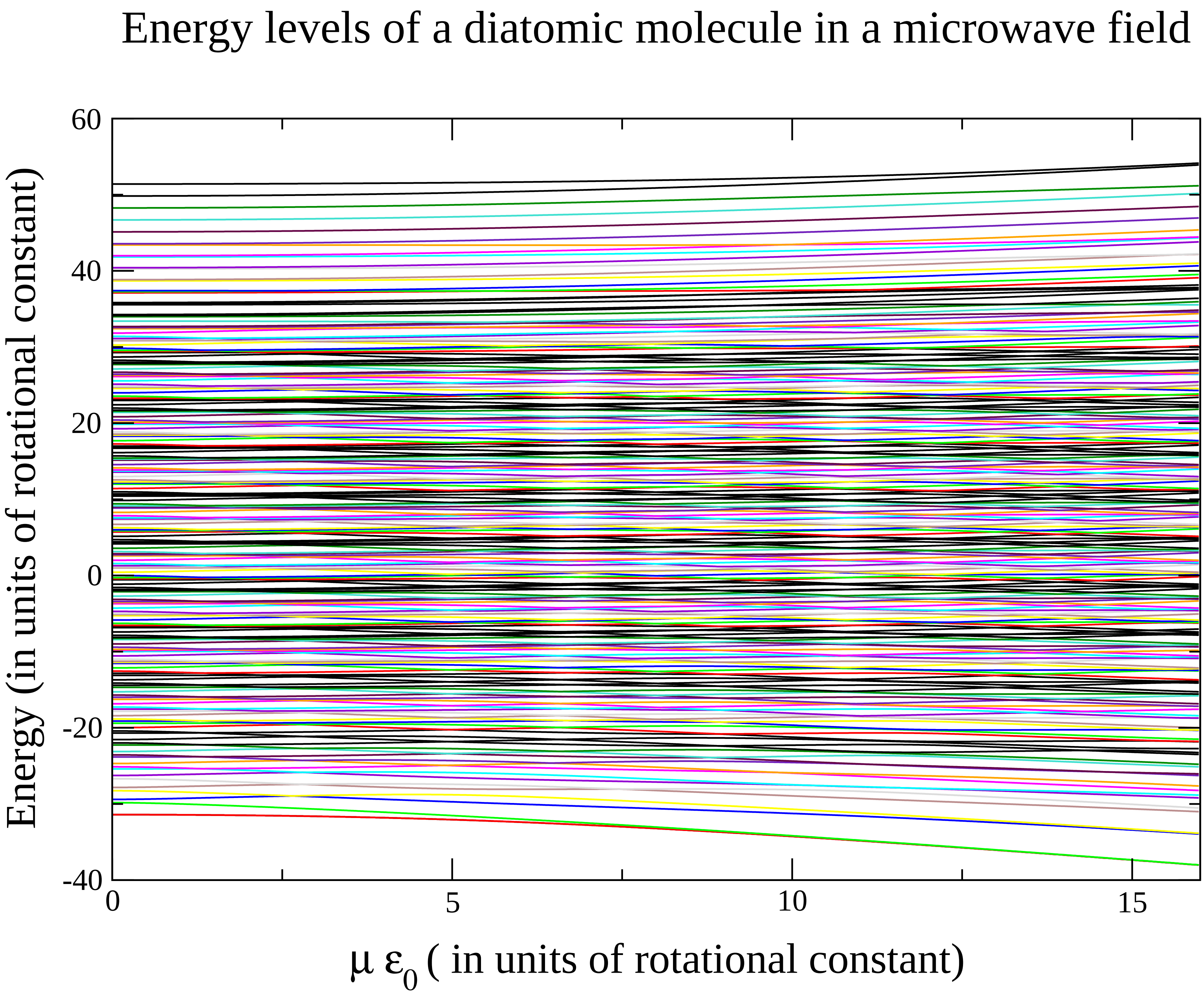


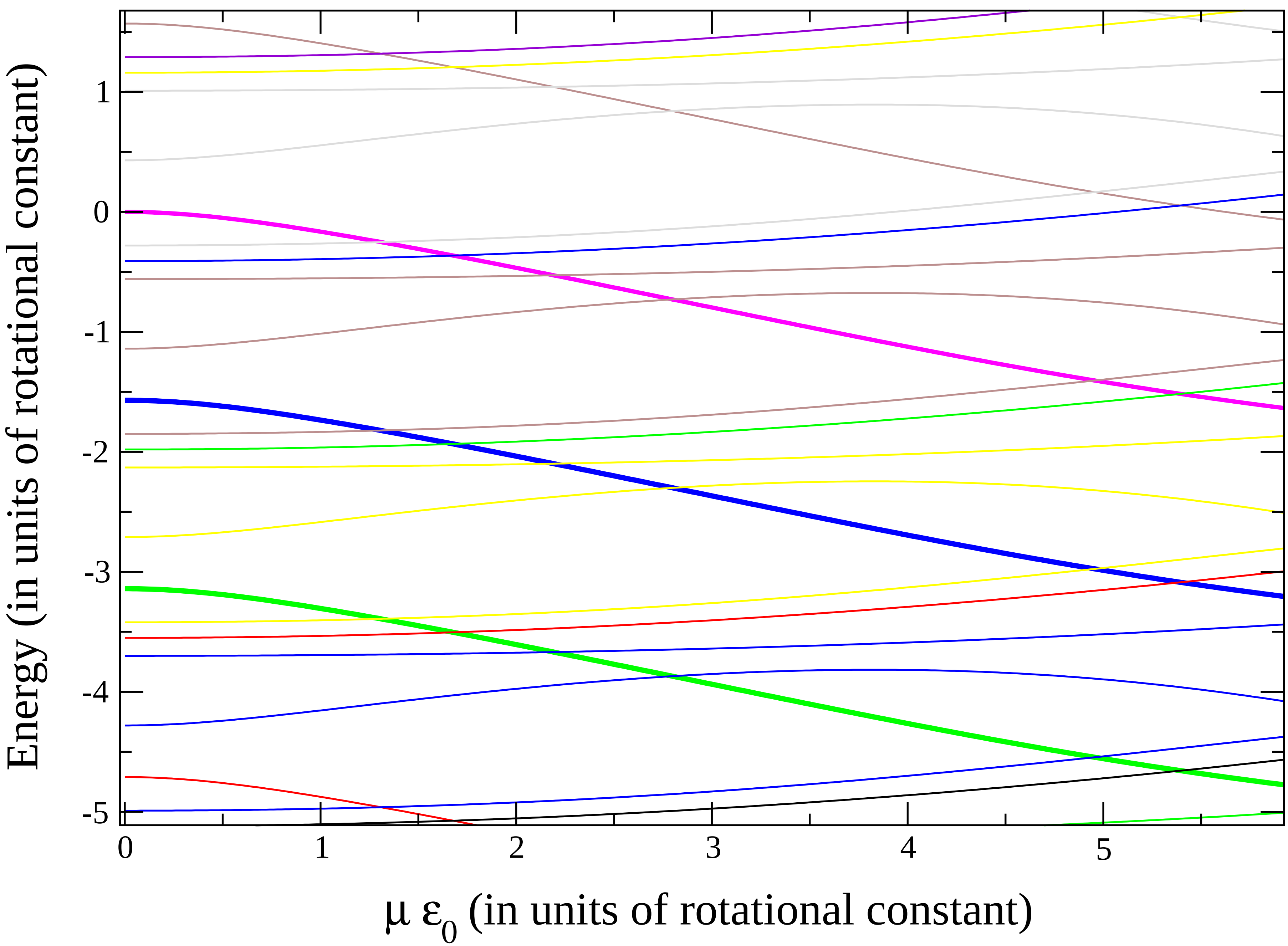
Fig. 2. Energies of dressed states vs. applied microwave electric field strength. State labels are zero-field basis states $|\psi_{Jmn}\rangle$,

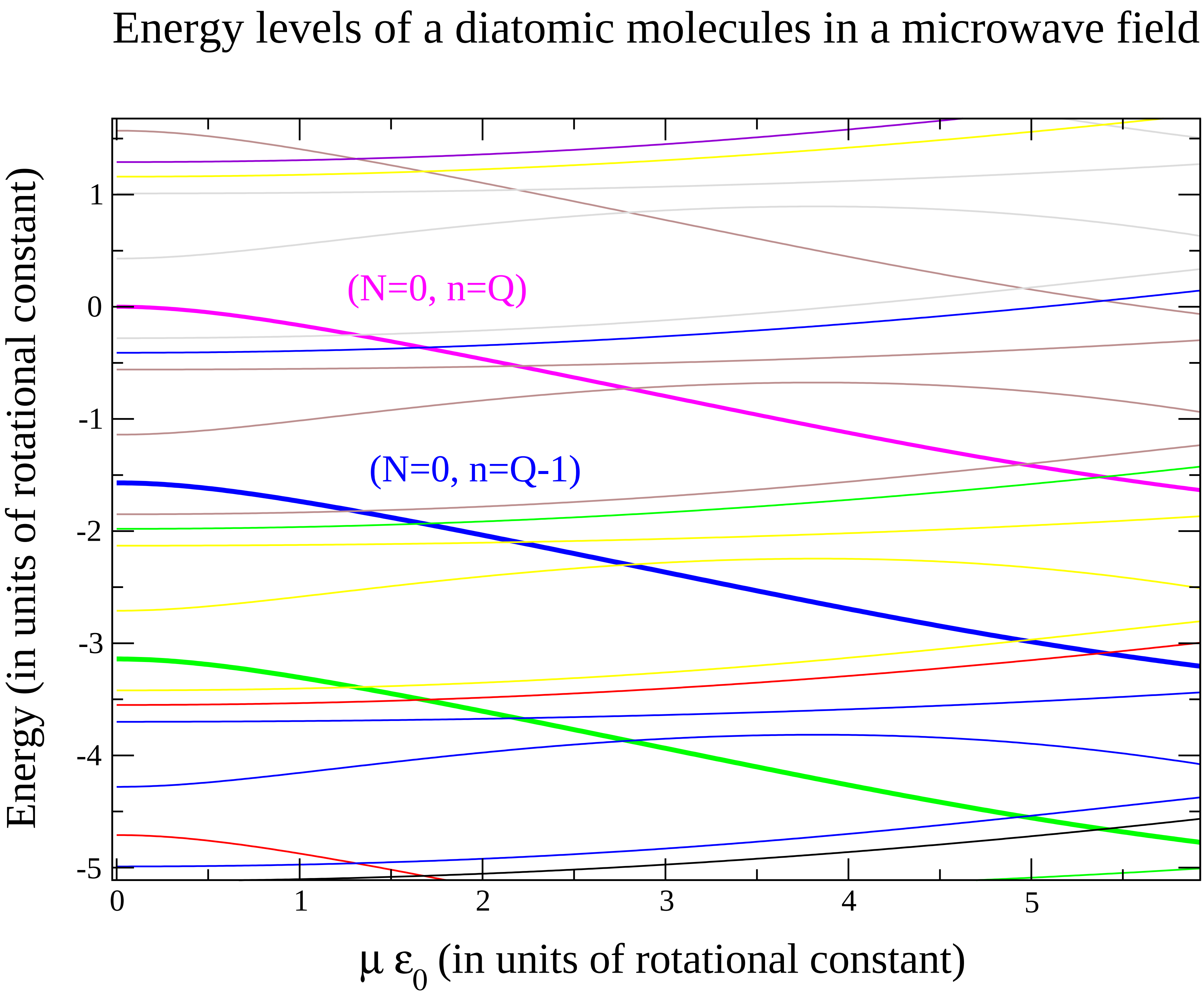
DeMille, Glenn, and Petricka, *Eur. J. Phys. D* **31**, 375 (2004)



Energy levels of a diatomic molecule in a microwave field
60
40
20
0
-20
-40
0
5
10
15
Energy (in units of rotational constant)
μ ε0 (in units of rotational constant)

Energy levels of a diatomic molecules in a microwave field




Energy levels of a diatomic molecules in a microwave field
(N=0, n=Q)
(N=0, n=Q-1)
Energy (in units of rotational constant)
μ ε0 (in units of rotational constant)
1
0
-1
-2
-3
-4
-5
0
1
2
3
4
5

Collisions of molecules in a microwave cavity

Molecular Hamiltonian: $H_{\text{mol}} = B\boldsymbol{N}^2$

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

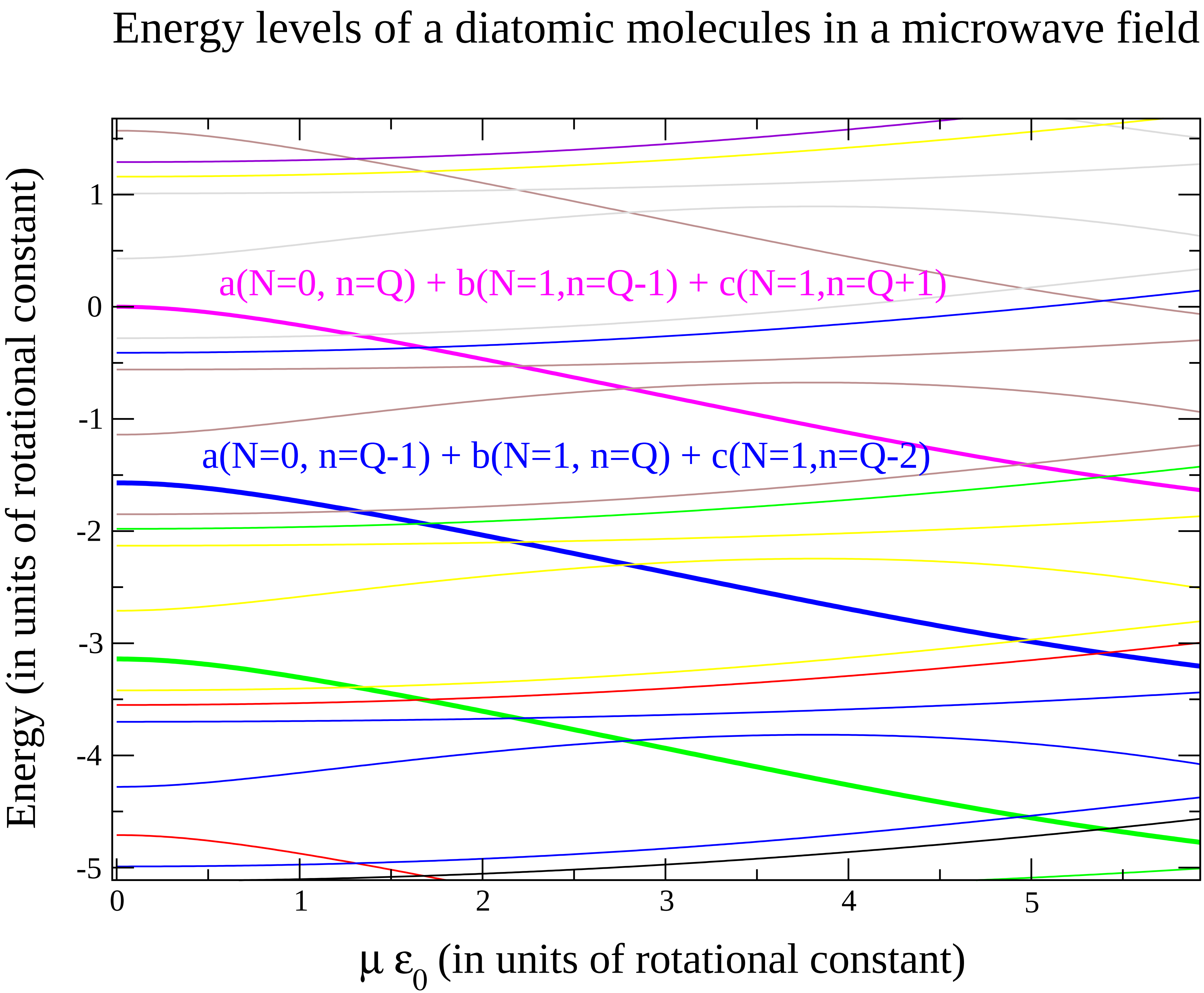
Molecule - Field Hamiltonian: $H_{\text{mol,f}} = -\mu\sqrt{\frac{\hbar\omega}{2\epsilon_0 V}}\cos\theta\left(a + a^\dagger\right)$

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

The matrix elements of of the molecule - field Hamiltonian:

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

$$\langle NM_N|\cos\theta|N'M'_N\rangle \sim \delta_{M_N,M'_N}\left(\delta_{N,N'+1} + \delta_{N,N'-1}\right)$$



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)
Energy (in units of rotational constant)
μ ε0 (in units of rotational constant)
1
0
-1
-2
-3
-4
-5
0
1
2
3
4
5

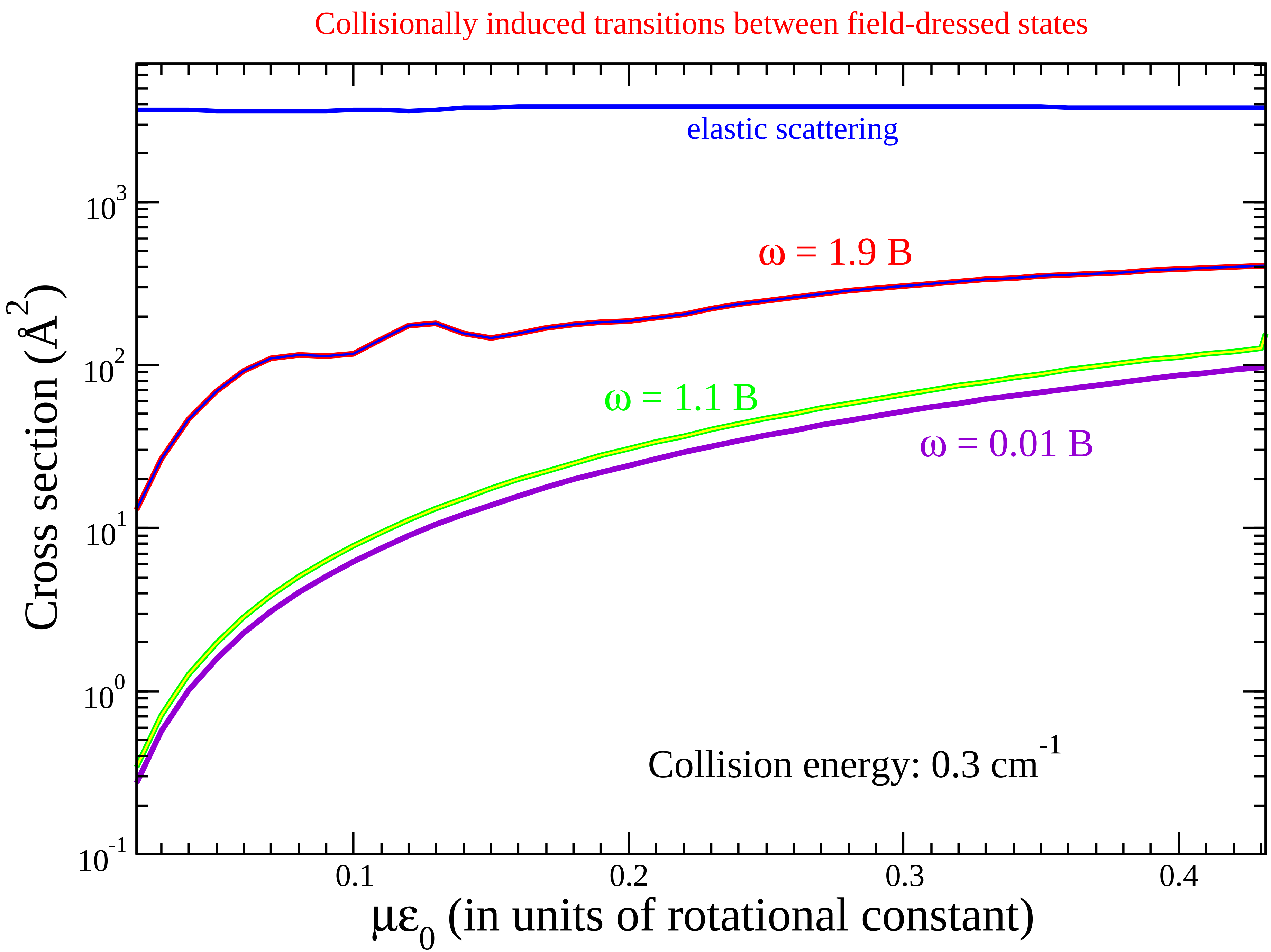
Superposition states of molecules in a microwave field

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

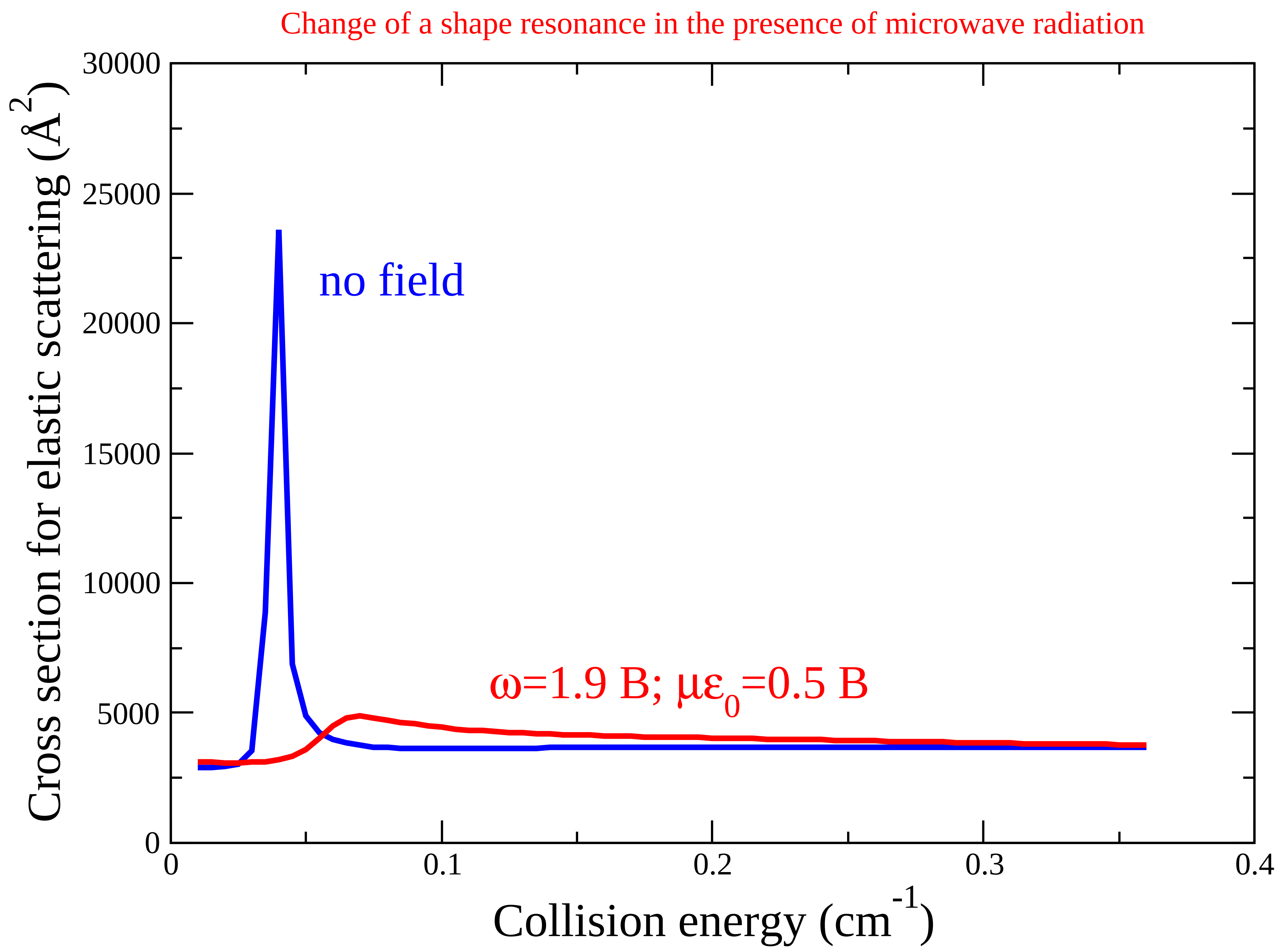
$\bar{N}$ is the average number of photons

Off-resonant light: $w = 0.01B$.

$\mu\epsilon_0 = 0.1B$	$\mu\epsilon_0 = 0.3B$	$\mu\epsilon_0 = 0.5B$
$a = 0.9994$	$a = 0.9637$	$a = 0.7497$
$b = 0.0143$	$a = 0.0397$	$a = 0.0178$
$c = 0.0145$	$c = 0.0471$	$c = 0.0441$
$d = 0.0207$	$a = 0.182$	$a = 0.445$
$e = 0.0209$	$a = 0.184$	$a = 0.448$



Collisionally induced transitions between field-dressed states
elastic scattering
ω = 1.9 B
ω = 1.1 B
ω = 0.01 B
Collision energy: 0.3 cm-1
Cross section (Å2)
με0 (in units of rotational constant)
10-1
100
101
102
103
0.1
0.2
0.3
0.4


Change of a shape resonance in the presence of microwave radiation
30000
25000
20000
15000
10000
5000
0
0
0.1
0.2
0.3
0.4
no field
ω=1.9 B; με$_0$=0.5 B
Cross section for elastic scattering (Å^2)
Collision energy (cm^{-1})

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

$$H = -\frac{1}{2\mu\rho}\frac{d}{d\rho}\rho\frac{d}{d\rho} + \frac{\boldsymbol{l}_z^2}{2\mu\rho^2} + H_{\mathrm{as}} + V(\rho),$$

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 $\psi(R,k=0)=\mathrm{const}R^s$
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:

elastic scattering cross section $\sim k^{4l}$

Repeating this derivation for 2D, we get

$$\text{cross secion} \sim \frac{1}{k \ln^2 k}, \quad \text{when } \; m = 0$$

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

$$\text{cross secion for } \; m = 0 \to m' \; \text{ transitions } \; \sim k^{2|m|-1} \frac{1}{\ln^2 k}$$

and

$$\text{cross secion for } \; m > 0 \to m' > 0 \; \text{ transitions } \; \sim k^{2|m|+2|m'|-1}$$

Threshold collision laws

Transition	3D	2D
s-wave elastic	$\sigma = \text{const}$	$\sigma \sim \frac{1}{k \ln^2 k}$
s-wave to non-s-wave	$\sigma \sim k^{2l'}$	$\sigma \sim k^{2\lvert m\rvert-1}\frac{1}{\ln^2 k}$
non-s-wave to non-s-wave	$\sigma \sim k^{2l+2l'}$	$\sigma \sim k^{2\lvert m\rvert+2\lvert m'\rvert-1}$

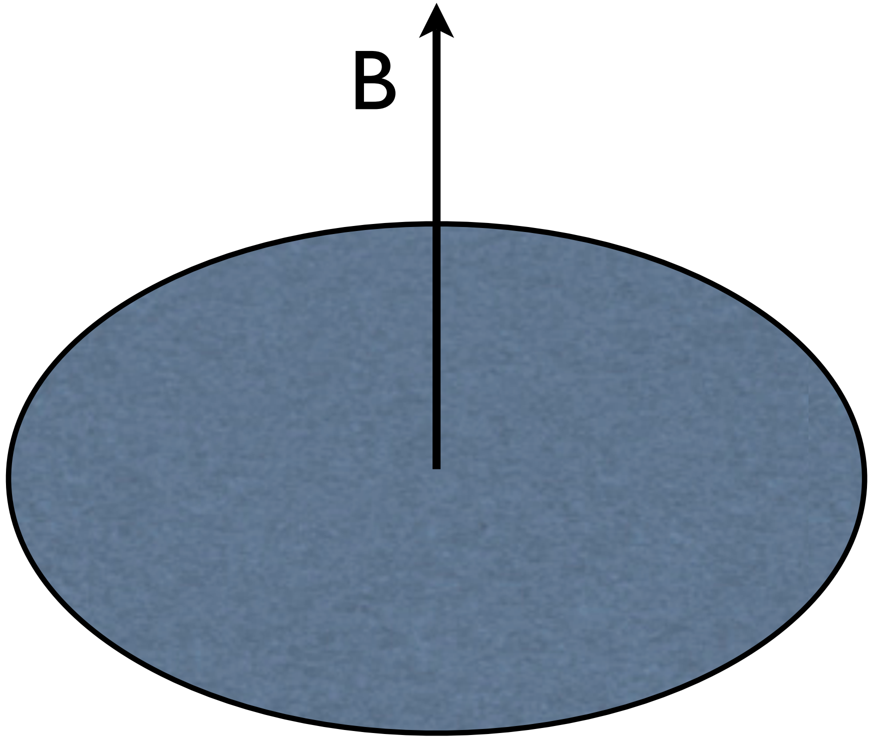
Why is this interesting?

Consider ultracold collisions of molecules in 2D.

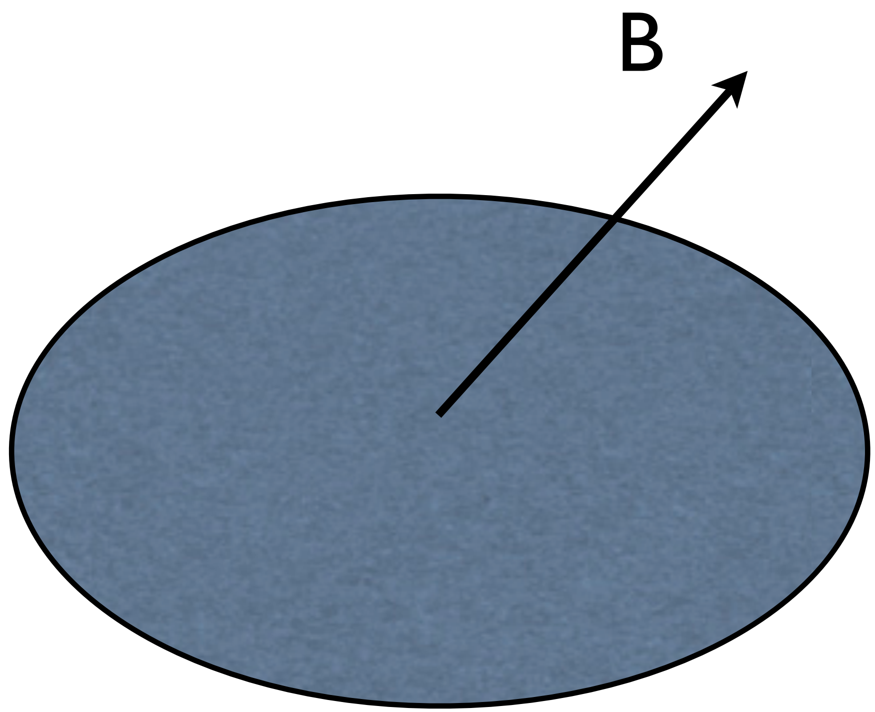
Angular momentum transfer of molecules - such as spin relaxation - must be accompanied by changes of m, if the magnetic field axis is directed perpendicularly to the plane of confinement

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
  $\longrightarrow$ more insight into long-range interactions
- elucidate rates for chemical reactions at ultracold Ts
  $\longrightarrow$ ultracold chemistry $\longrightarrow$ lots of applications
- demonstrate the possibility of controlling chemical reactions
  $\longrightarrow$ controlled chemistry $\longrightarrow$ lots of applications
- make coherent control of bimolecular reactions possible
  $\longrightarrow$ controlled chemistry
- provide new testground for statistical theories of molecules
  $\longrightarrow$ new reaction rate theories

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