Interactions and Collisions involving Cold Molecules

Jeremy M. Hutson
University of Durham, UK

KITP, Santa Barbara
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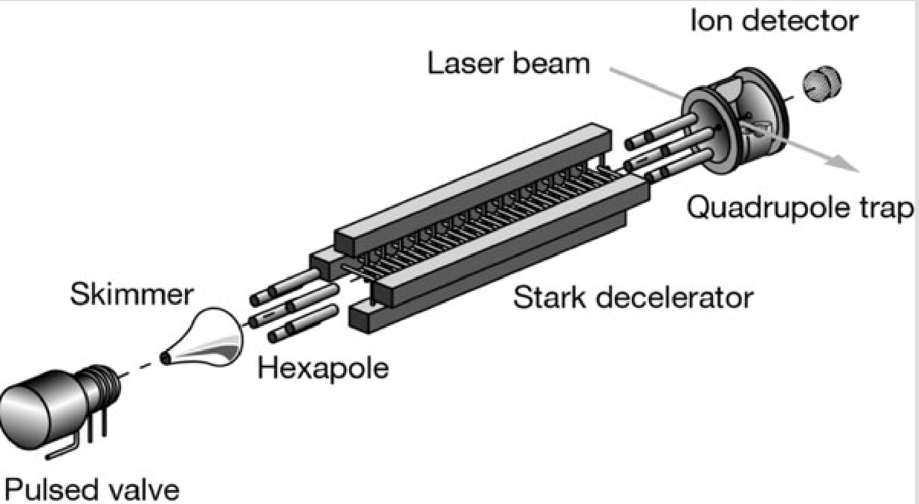
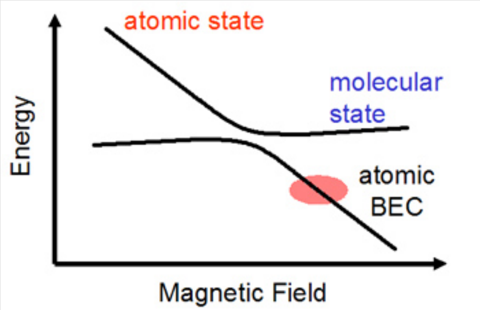
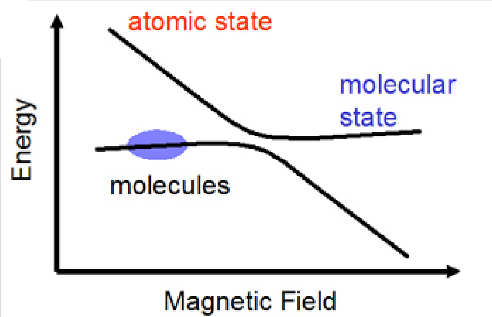
Cold molecules offer new possibilities:

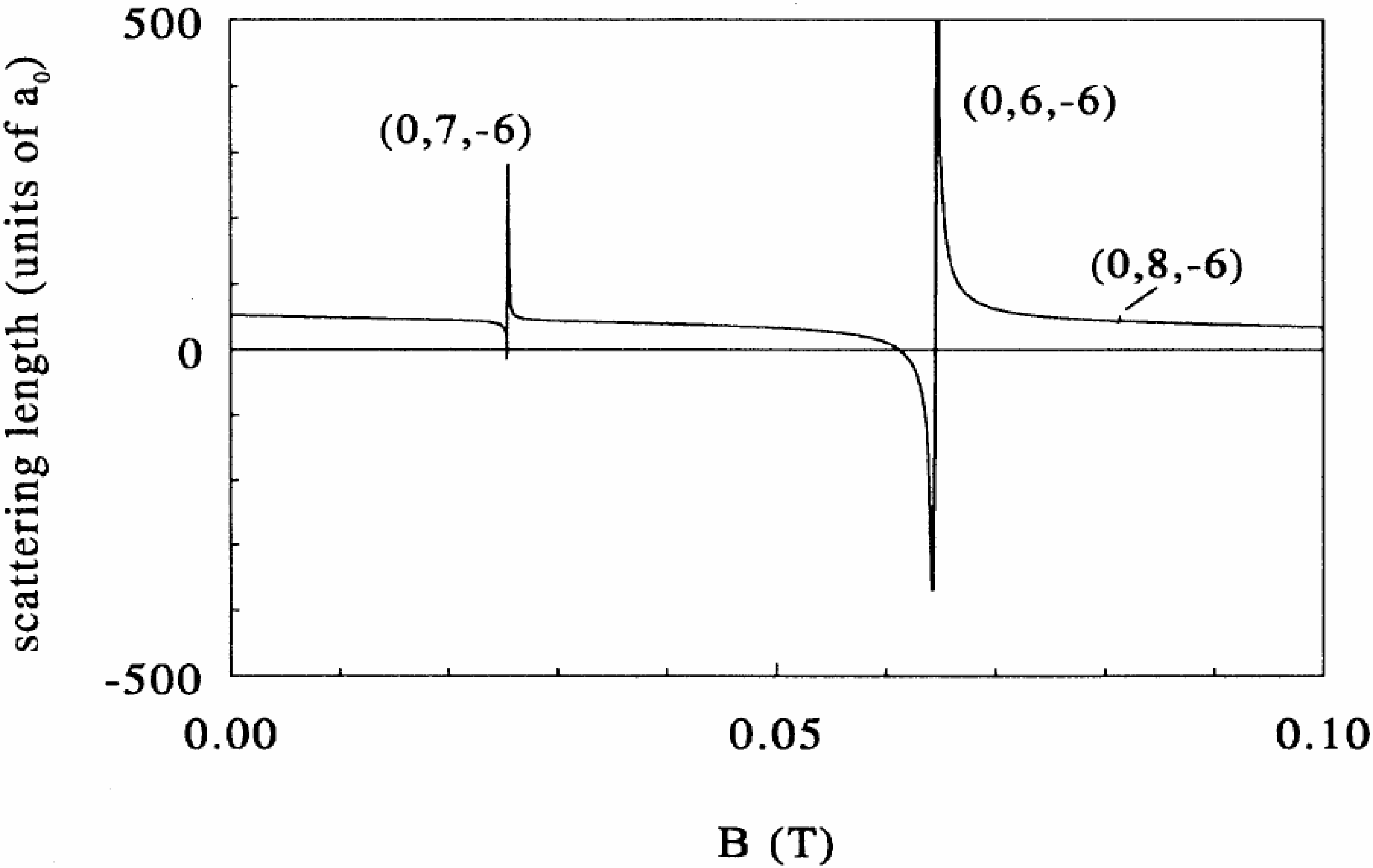
1. Dipole moments
2. Long-range molecule-molecule forces
3. Richer energy level structure (more handles)
4. Two general classes:
   i. Molecules cooled from room temperature
   ii. Dimers formed from ultracold atoms



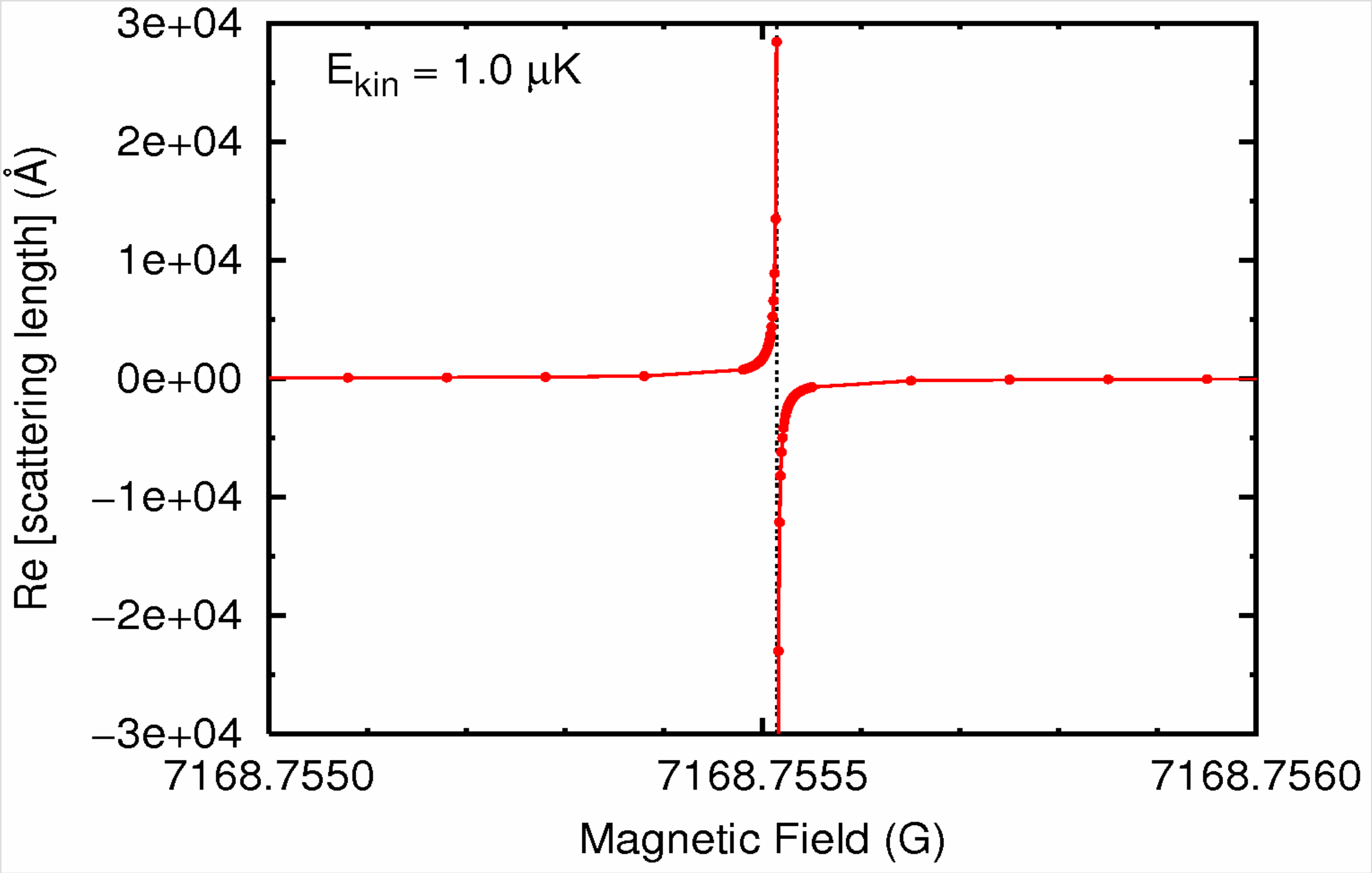




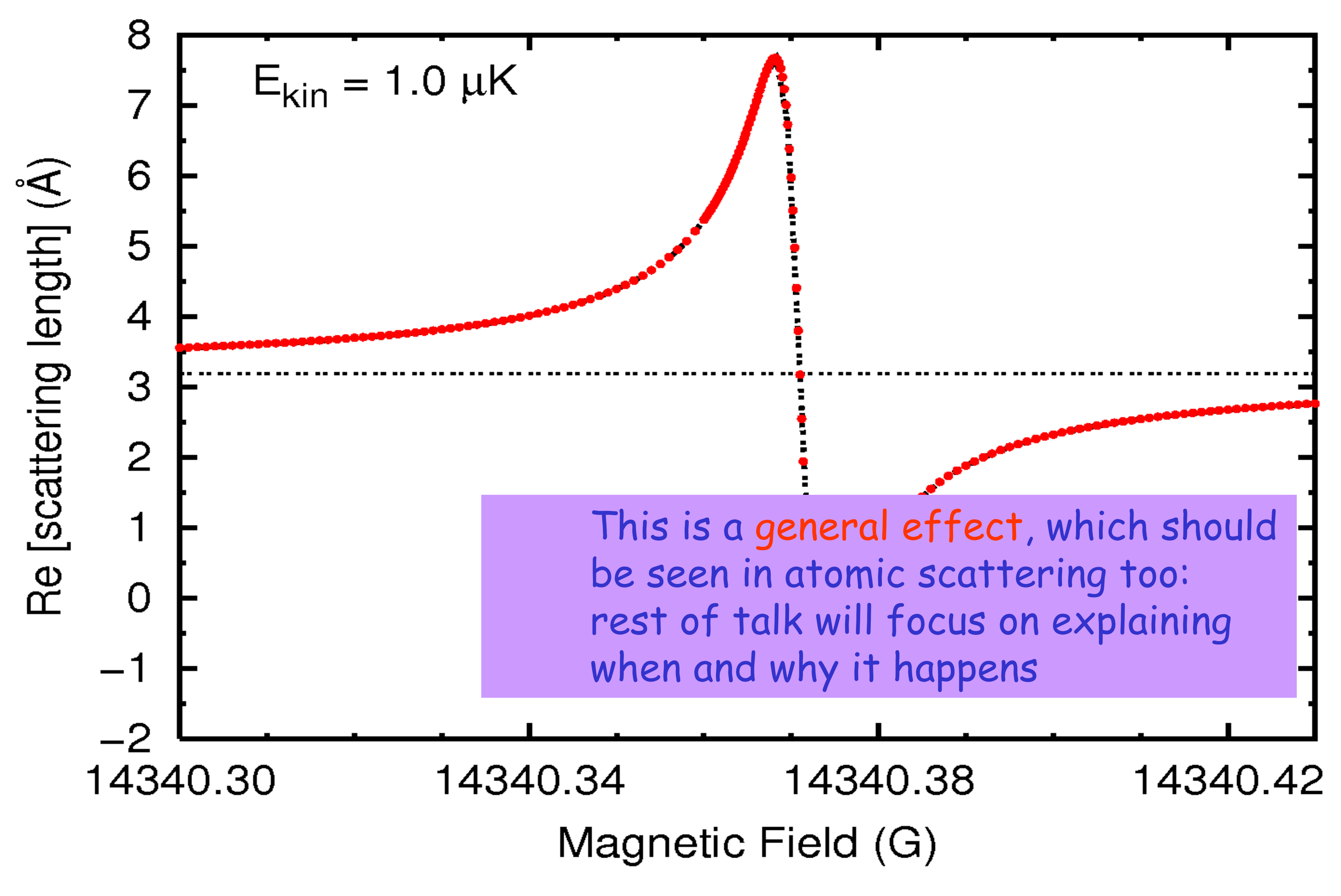
Point of contact between collisions and condensed matter is scattering length: passes through pole at resonance



For molecular scattering, *some* resonances show poles



For molecular scattering, *some* resonances show poles: others show only small oscillation



This is a general effect, which should be seen in atomic scattering too: rest of talk will focus on explaining when and why it happens

Origin of scattering length in collision theory

- Quantum scattering amplitudes defined by the S matrix:

$$\psi = \psi_0^- + \sum_i S_{0i}\, \psi_i^+$$

- The S matrix is unitary (complex symmetric).
  For condensed matter we are most interested in the elastic scattering submatrix (usually a single element) S_{00},

$$S_{00} = \exp[2i\delta(k)]$$

  If scattering is purely elastic, $\delta(k)$ is real.

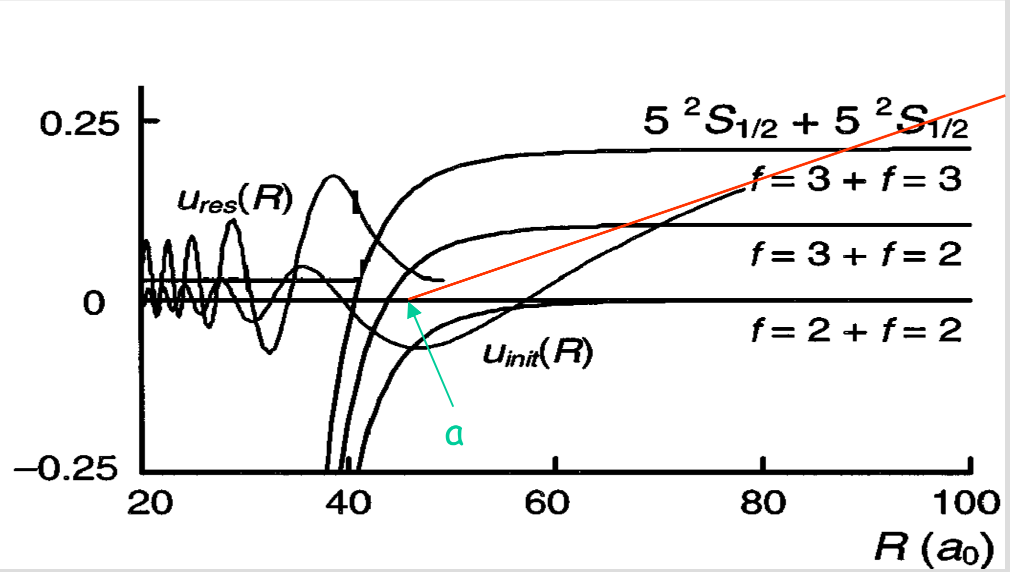
- Scattering length a(k) related to phase shift by

$$a(k) = \frac{-\tan\delta(k)}{k} = \frac{1}{ik}\left(\frac{1 - S_{00}}{1 + S_{00}}\right)$$

  In limit of low kinetic energy, a(k) is independent of E_{kin}.

Low-energy collisions characterized by scattering length

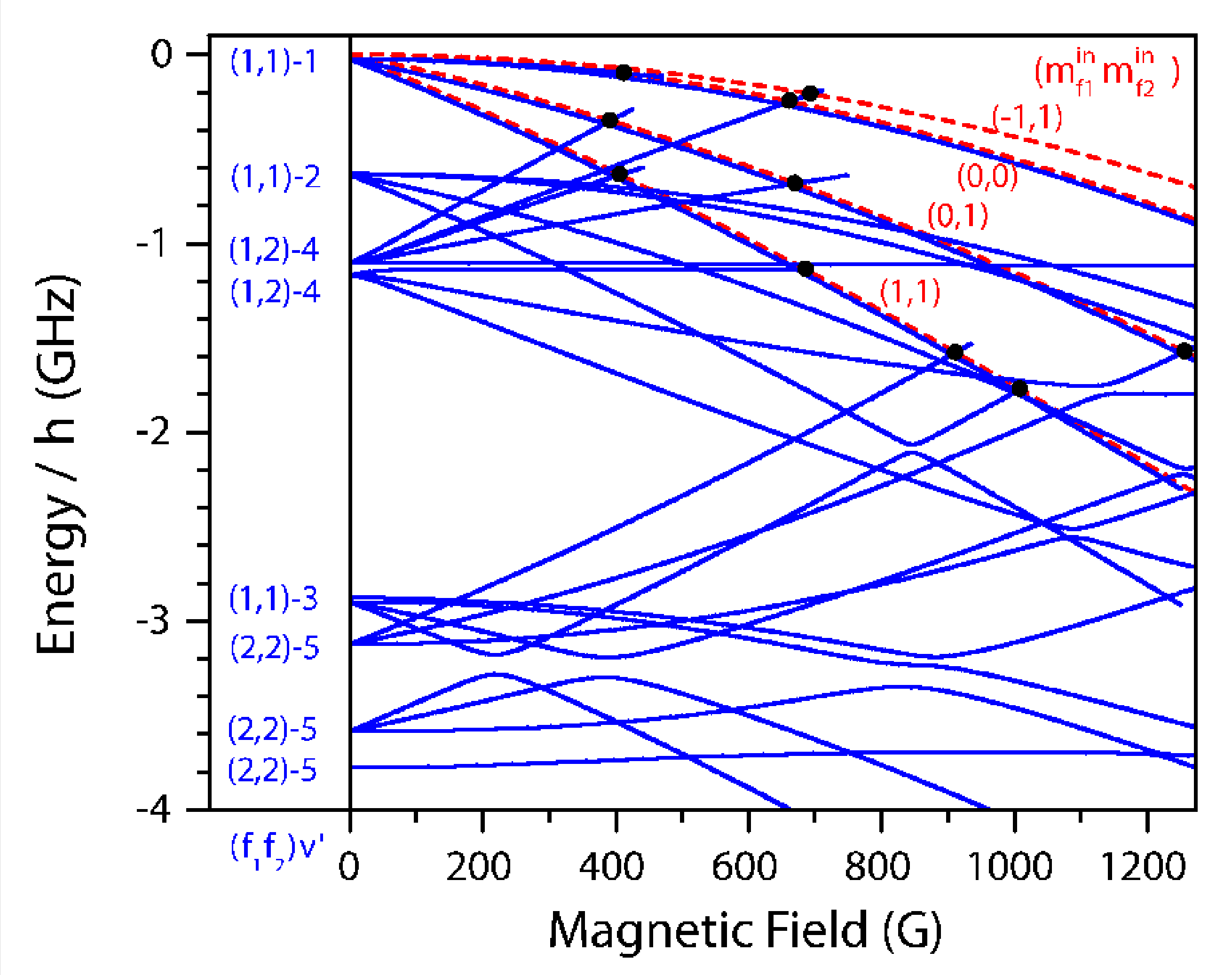
- If $E_{kin}=0$, $V(R) = E$ at long range
- $d^2\psi/dR^2 = 0$, so wavefunction is straight line at long range,
  $\psi = 1\text{-}r/a$
- Scattering length a is distance where extrapolated line crosses zero



- Elastic cross section
  $\sigma_{el} = 4\pi a^2/(1+k^2a^2) \approx 4\pi a^2$
- The scattering length is a measure of the overall strength of the interaction:
  - A positive scattering length corresponds to a repulsive interaction
  - A negative scattering length corresponds to an attractive interaction

Remember how Feshbach resonances work for atoms:

Atomic dimers have many bound states near threshold.
Bound states (blue) and thresholds (red) have different Zeeman effects: zero-energy Feshbach resonances occur where they cross

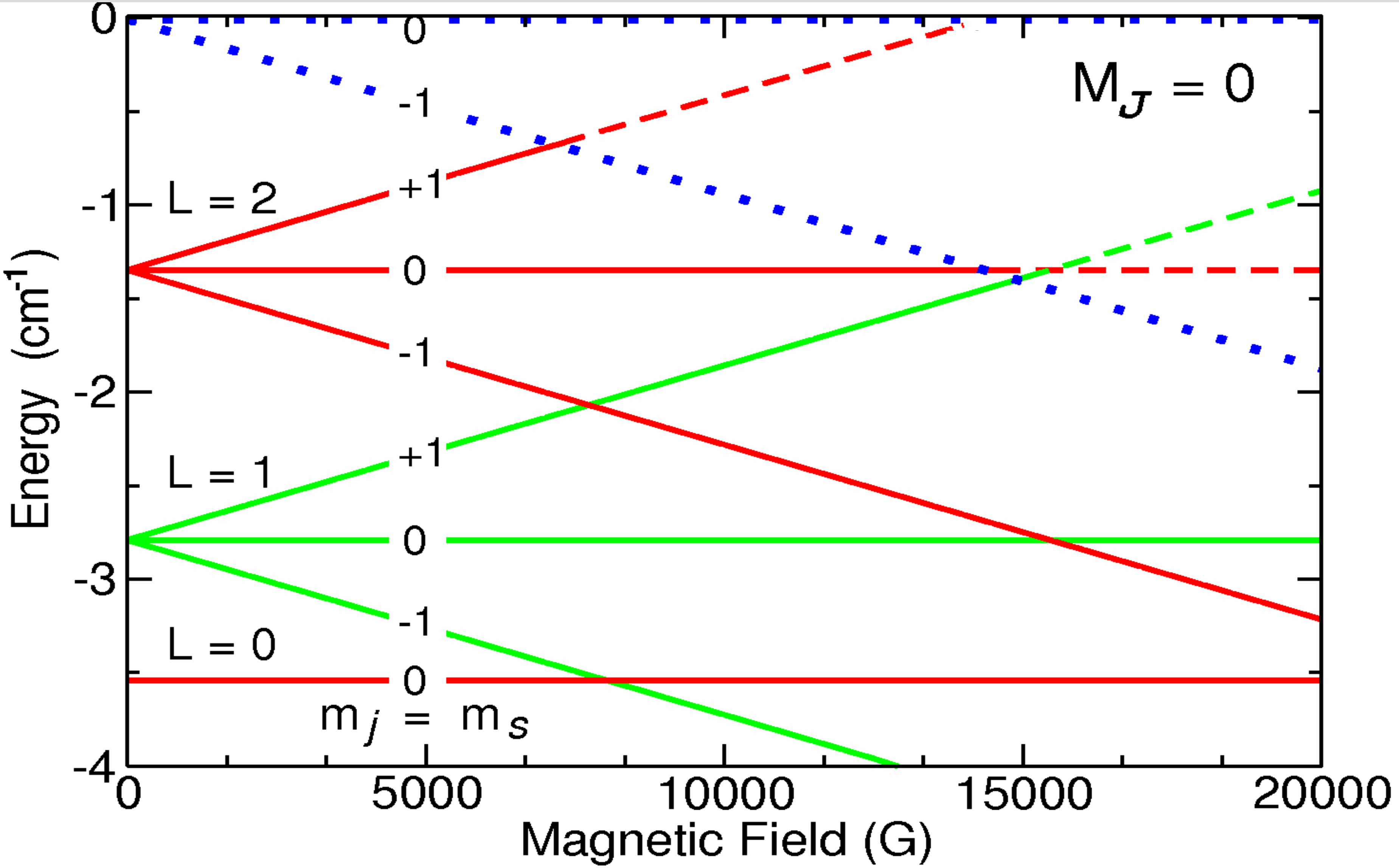


$^{87}Rb_2$: Marte et al., 2002

We need to be able to carry out *molecular* bound state and scattering calculations in magnetic fields

1. Carry out bound-state calculations as function of magnetic field:
   we have modified our BOUND package to handle atom + molecule bound states in magnetic fields

Prototype: bound states crossing threshold in ^3He + NH



Bound states: red and green; thresholds: blue dots

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2. Find fields at which bound states cross thresholds: these are the positions of zero-energy Feshbach resonances

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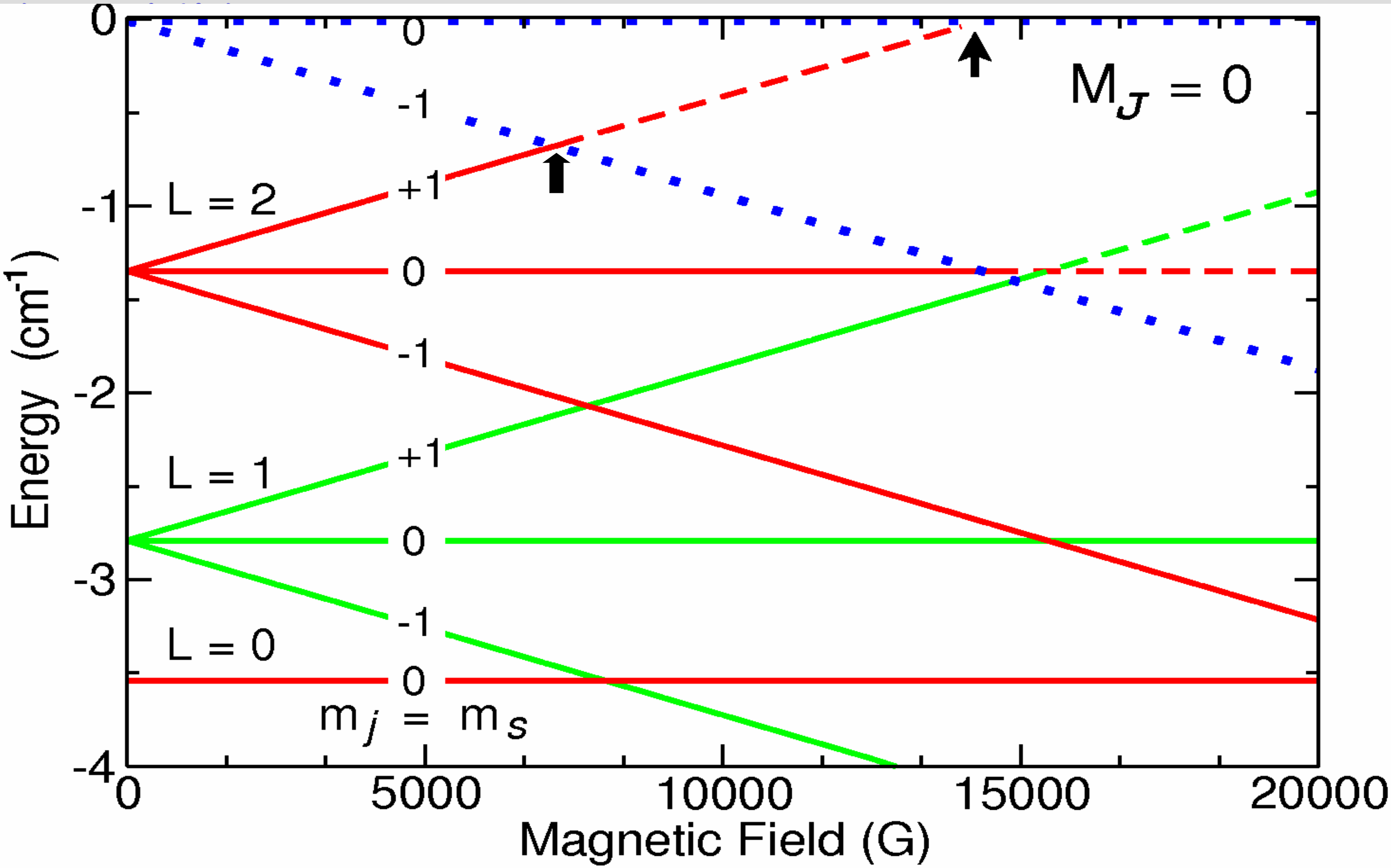

Bound states: red and green; thresholds: blue dots

Resonances expected at 7169 G and 14340 G

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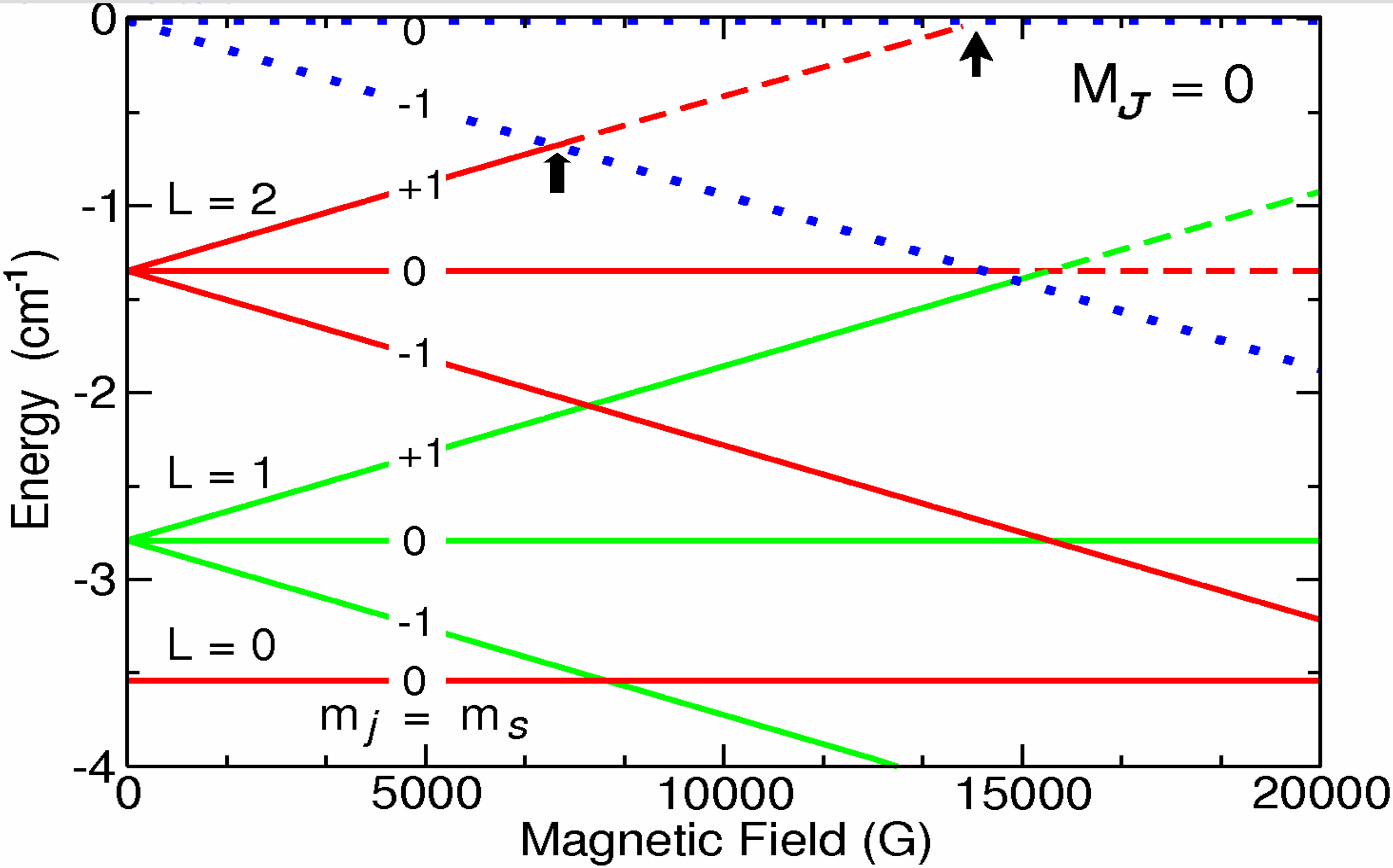
2. Find fields at which bound states cross thresholds: these are the positions of zero-energy Feshbach resonances

3. Carry out scattering calculations as function of magnetic field across resonances:
   we have modified our MOLSCAT package to handle atom-molecule collisions in magnetic fields

Prototype: bound states crossing threshold in



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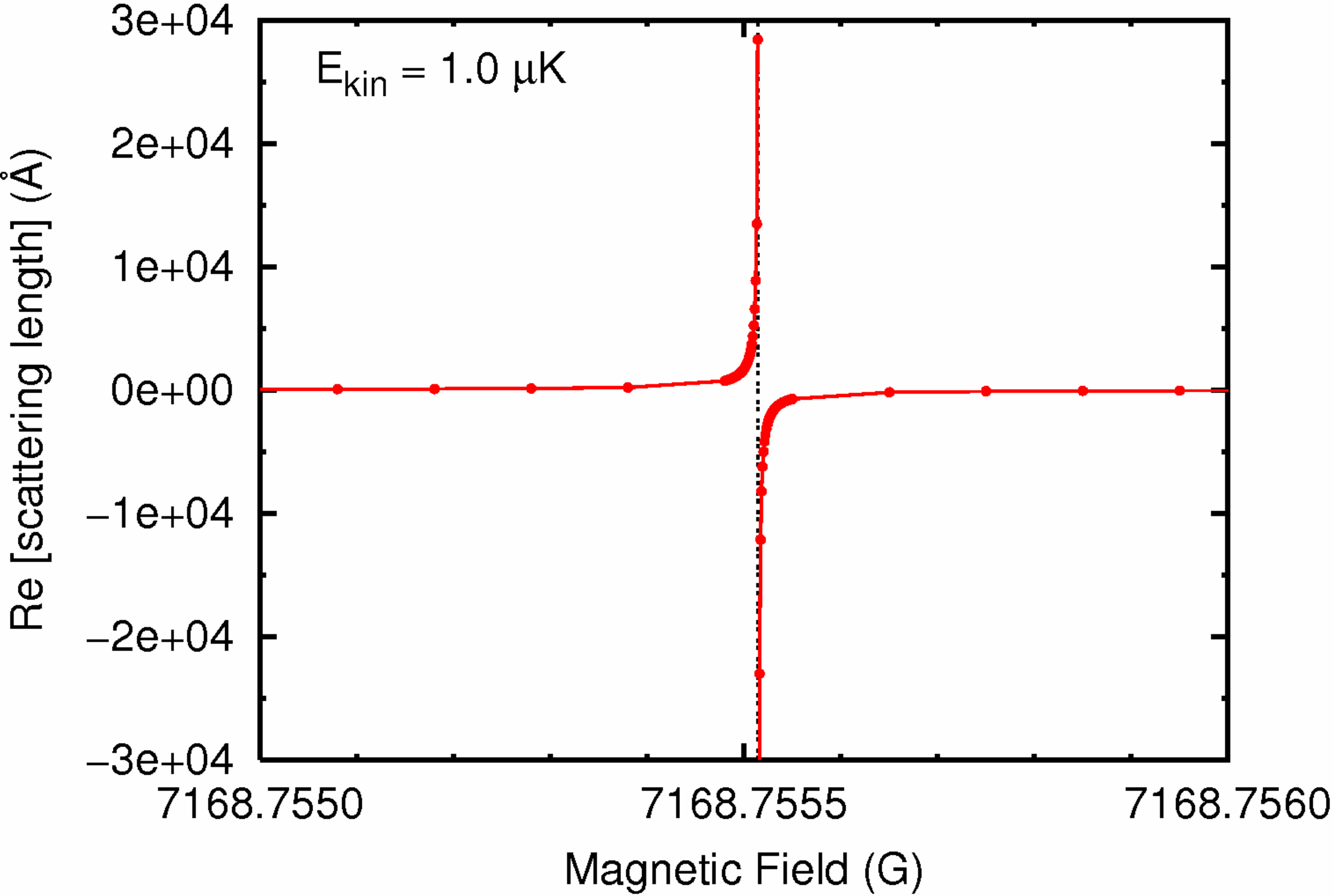
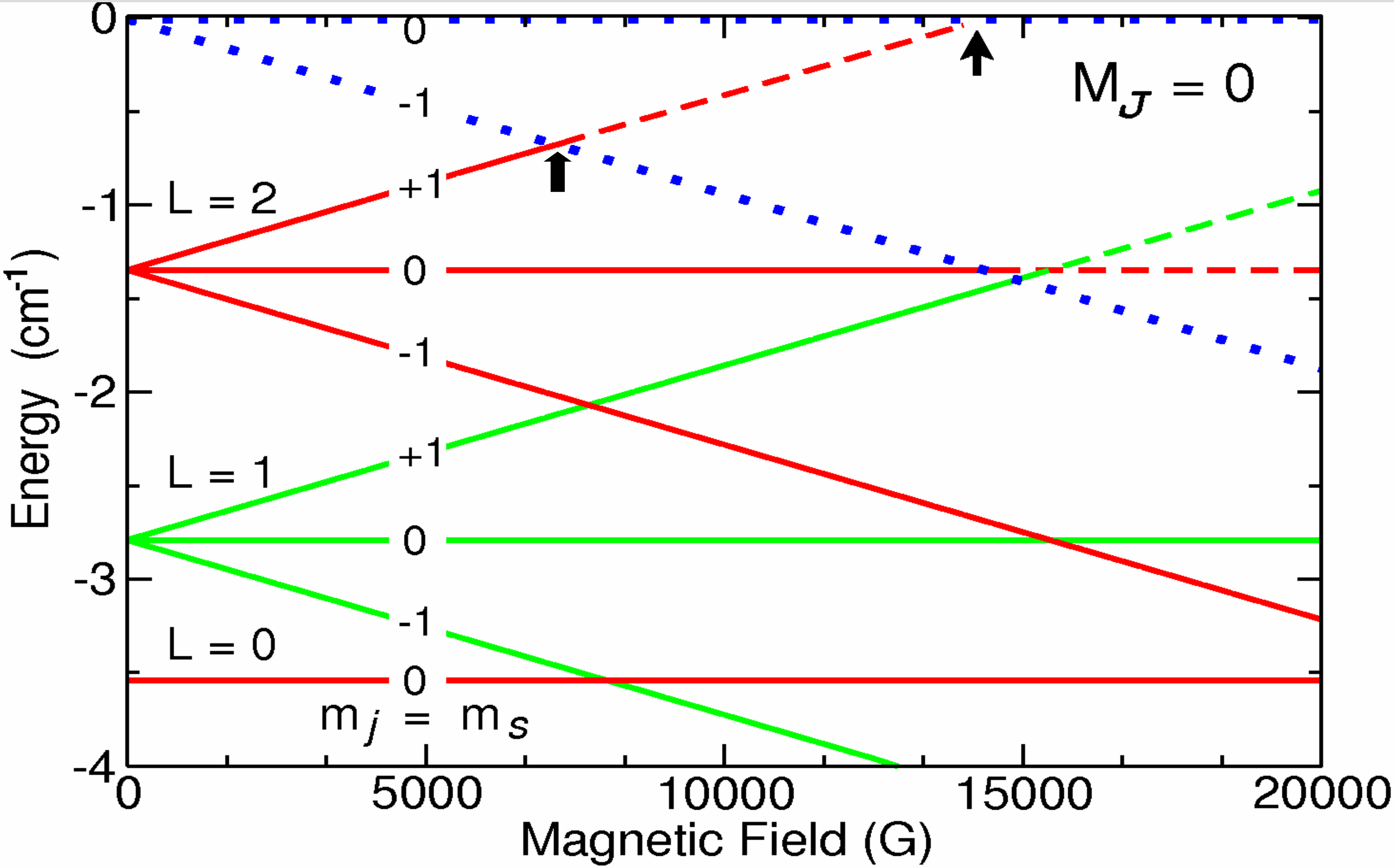
Resonances expected at 7169 G and 14340 G

He+NH scattering resonance 1: pole in scattering length



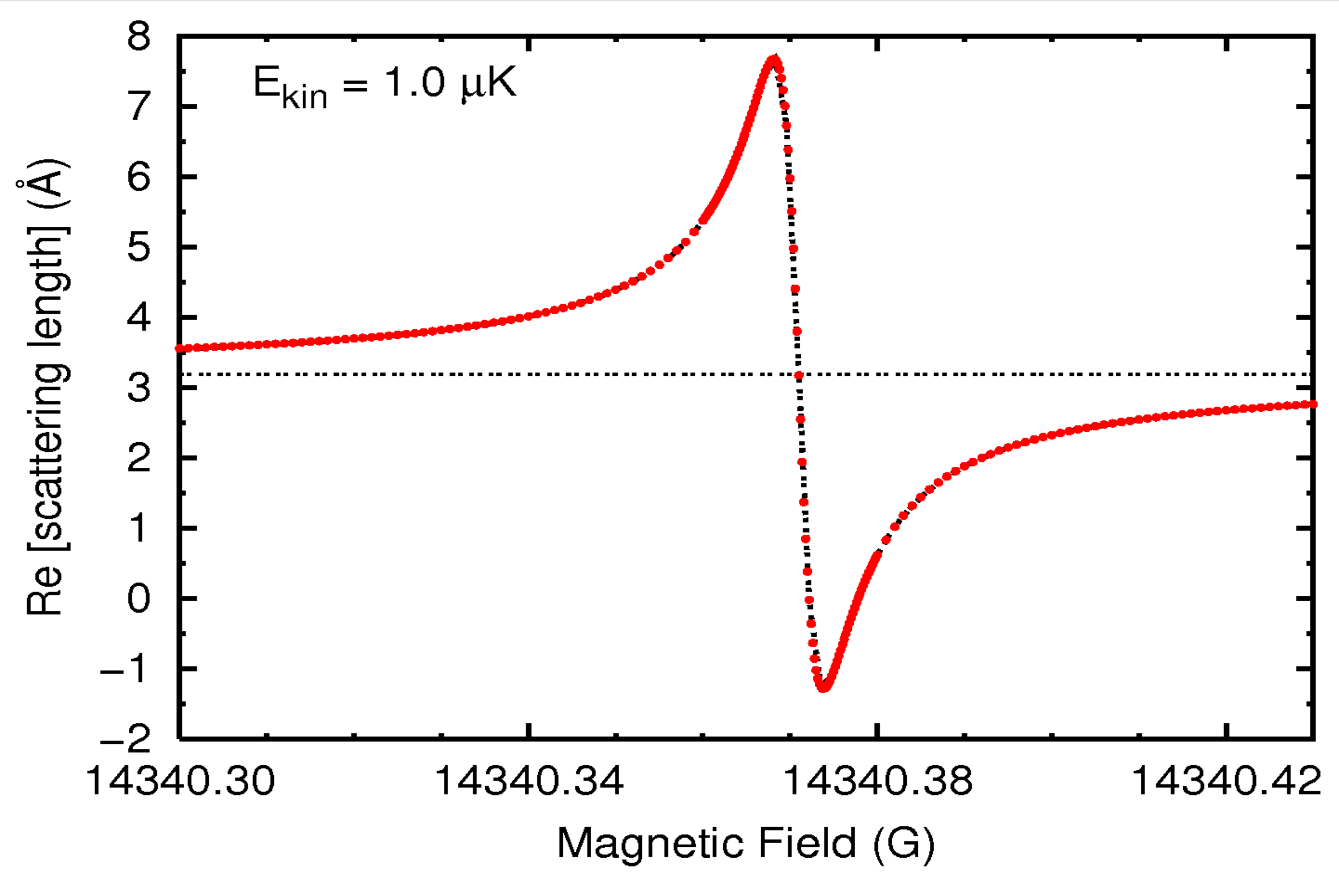
^3He + NH: bound states crossing threshold



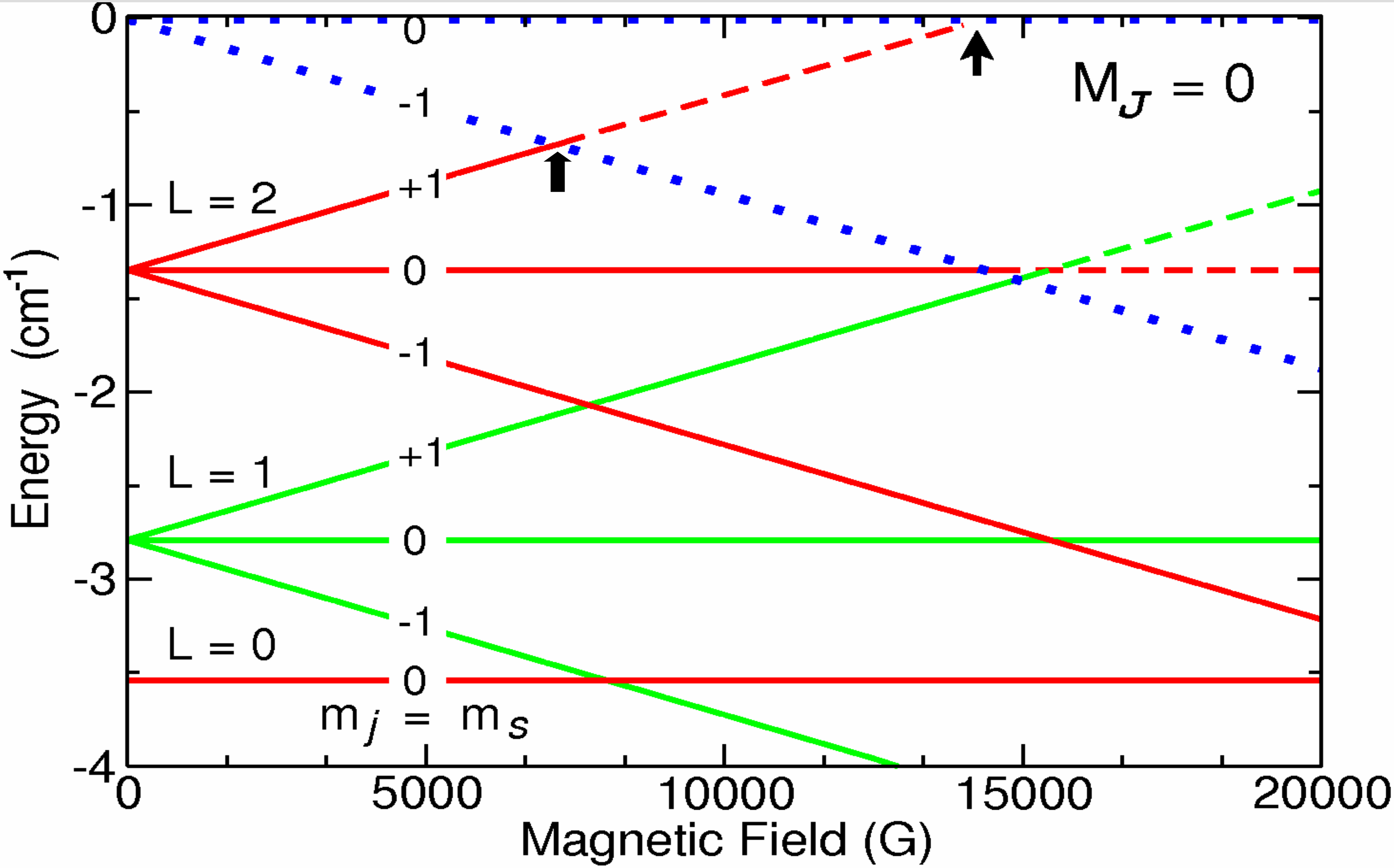
Bound states: red and green; thresholds: blue dots

Resonances expected at 7169 G and 14340 G

He+NH scattering resonance 2:
scattering length shows small wobble, *not* pole



Why the difference in behaviour?



Inelastic scattering is allowed for resonance at 14340 G

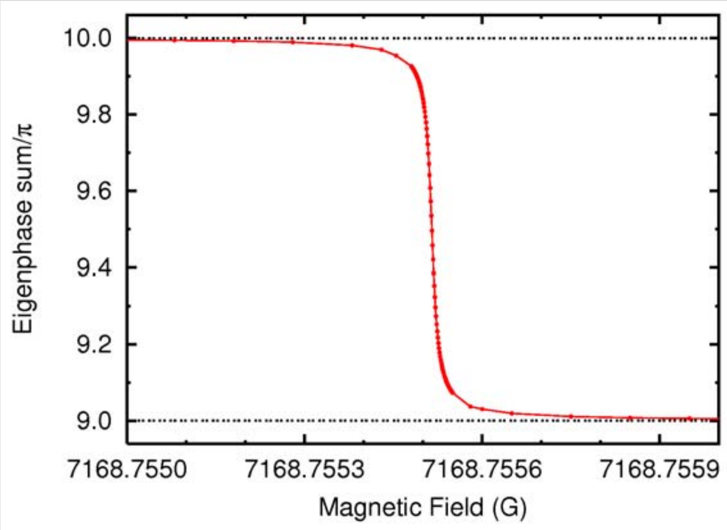
Characterising resonant behaviour

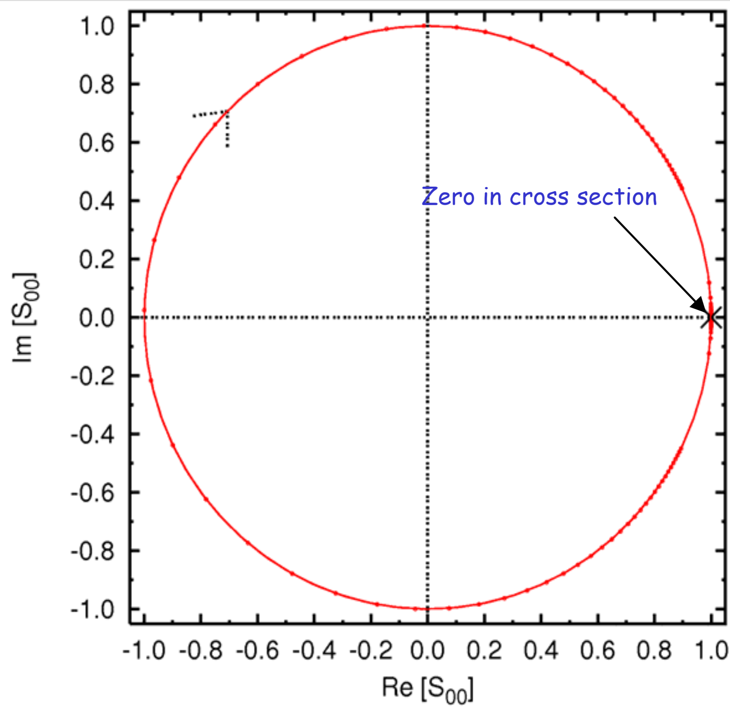
- Scattering phase shift $\delta(k)$,

$$S_{00} = \exp[2i\delta(k)]$$

Across an elastic scattering resonance, the phase shift changes sharply by π

- The S matrix element describes a circle in the complex plane as the energy (or field) is tuned across resonance

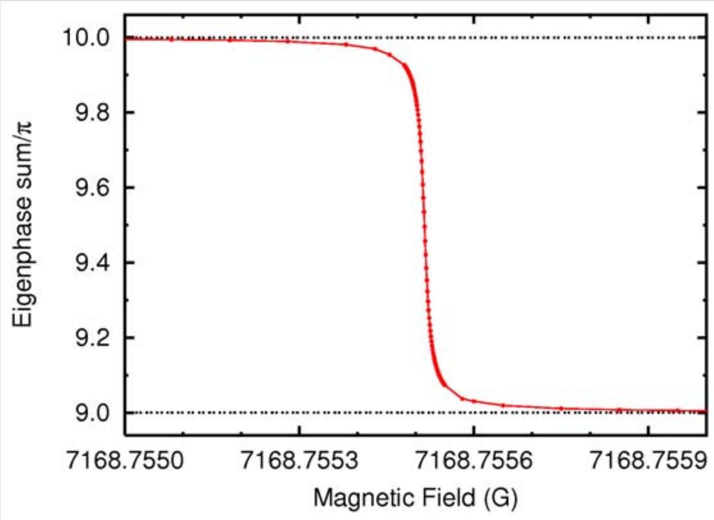




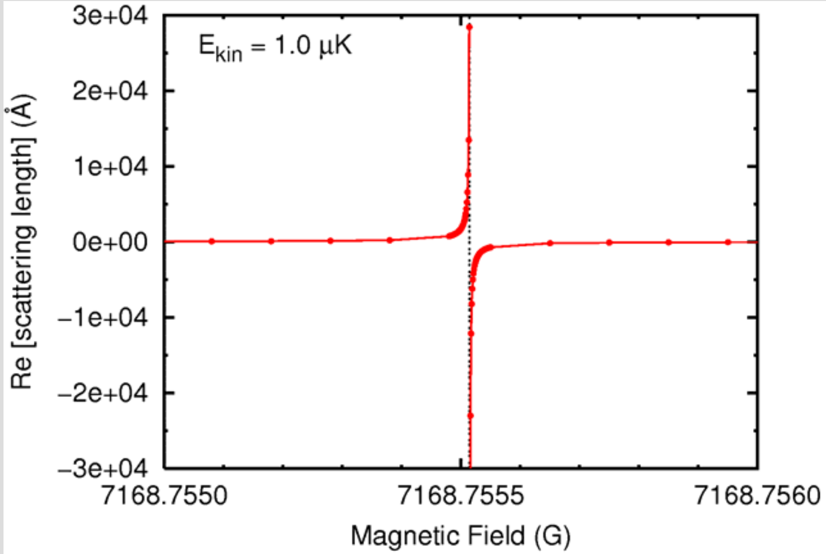
Characterising resonant behaviour

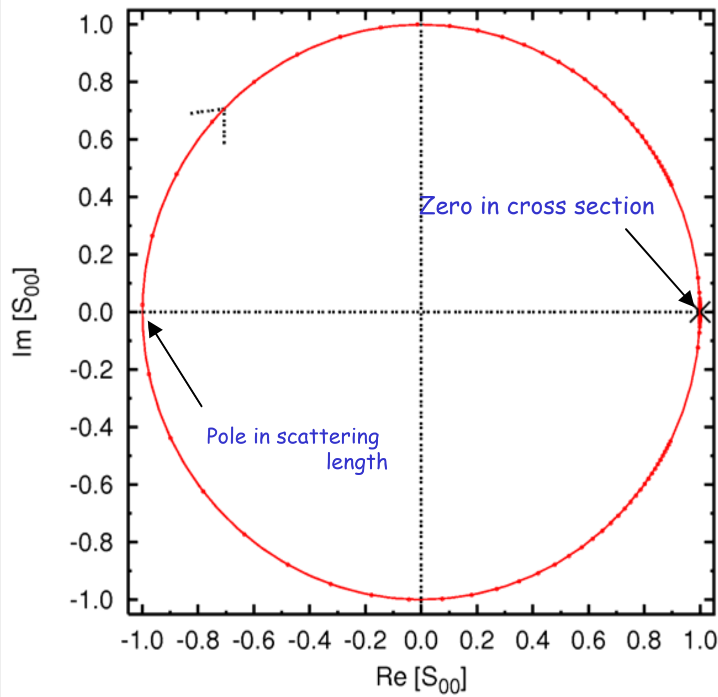
- Scattering length a(k) related to phase shift by

$$a(k) = \frac{-\tan\delta(k)}{k} = \frac{1}{ik}\left(\frac{1-S_{00}}{1+S_{00}}\right)$$

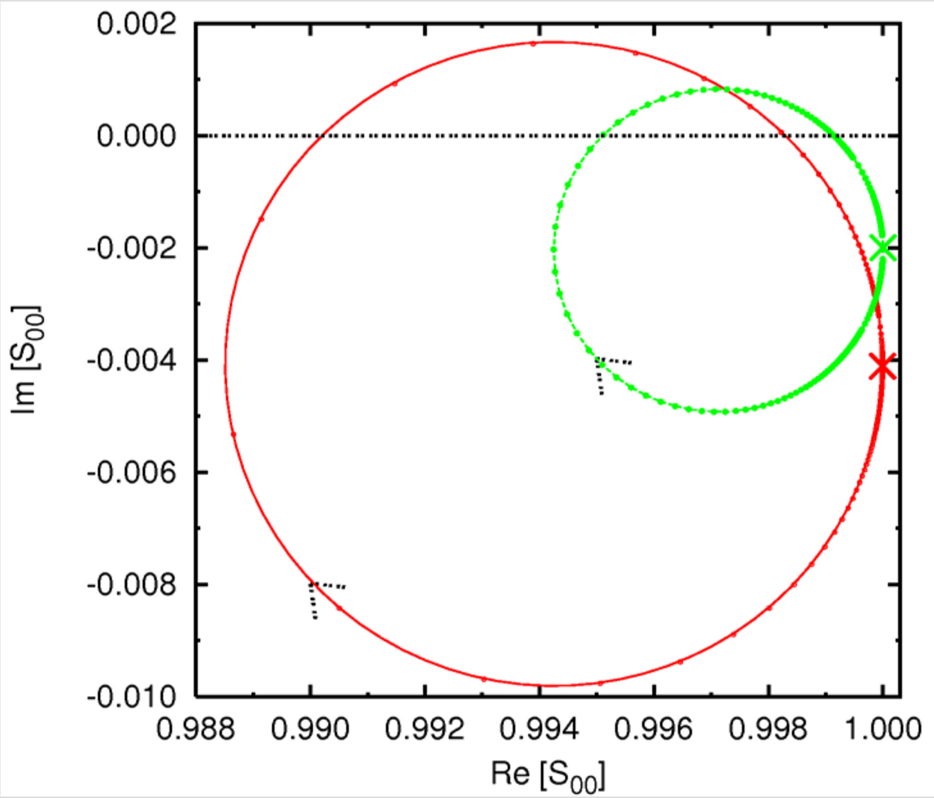


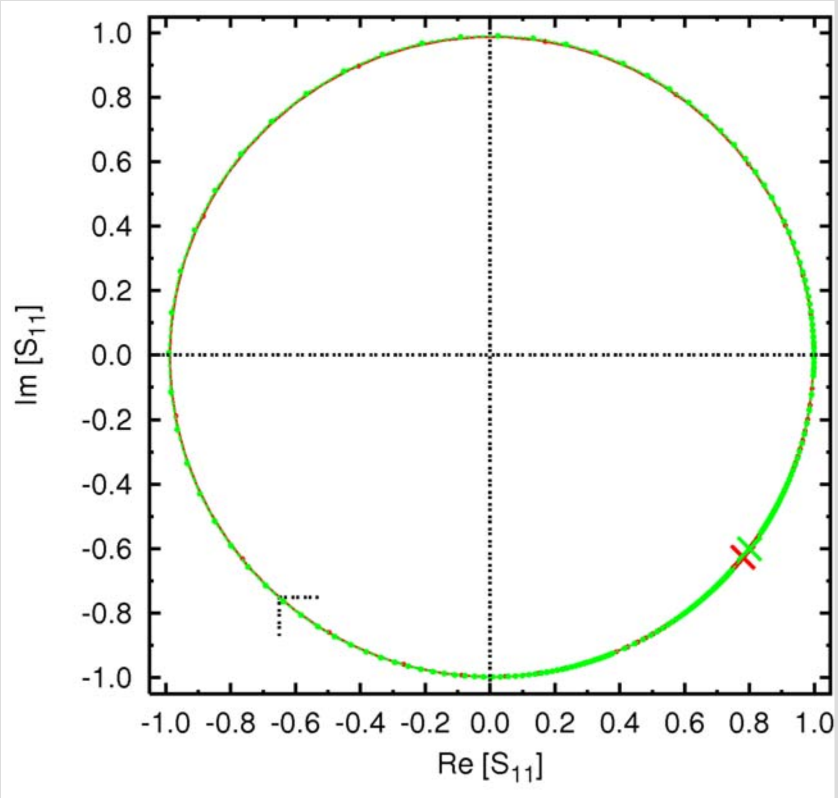
- When S_{00} = -1, a(k) = ∞: corresponds to $\delta(k)/\pi = n + \frac{1}{2}$.





He + NH inelastic scattering resonance: S-matrix element describes *small* circle in complex plane





Elastic S-matrix element in incoming (low-energy) channel
Radius depends on energy

Diagonal S-matrix element in inelastic channel
Radius independent of energy

E_{kin} = 1 μK (green); 4 μK (red)

Threshold behaviour

- The circle in the elastic S-matrix element S_{00} gets smaller with decreasing energy: proportional to k (i.e. $E_{kin}^{1/2}$) [wavenumber k related to E_{kin} by $E_{kin} = \hbar^2 k^2/2\mu$]

- Phase shift $\delta(k)$ defined as before by

$$S_{00} = \exp[2i\delta(k)]$$

but now complex because $|S_{00}| < 1$.

- Complex scattering length a(k) related to phase shift by

$$a(k) = \alpha(k) - i\beta(k) = \frac{-\tan\delta(k)}{k} = \frac{1}{ik}\left(\frac{1 - S_{00}}{1 + S_{00}}\right)$$

- If the circle in S_{00} is small, δ(k) and a(k) show a small oscillation instead of a pole

How general is the suppression of poles?

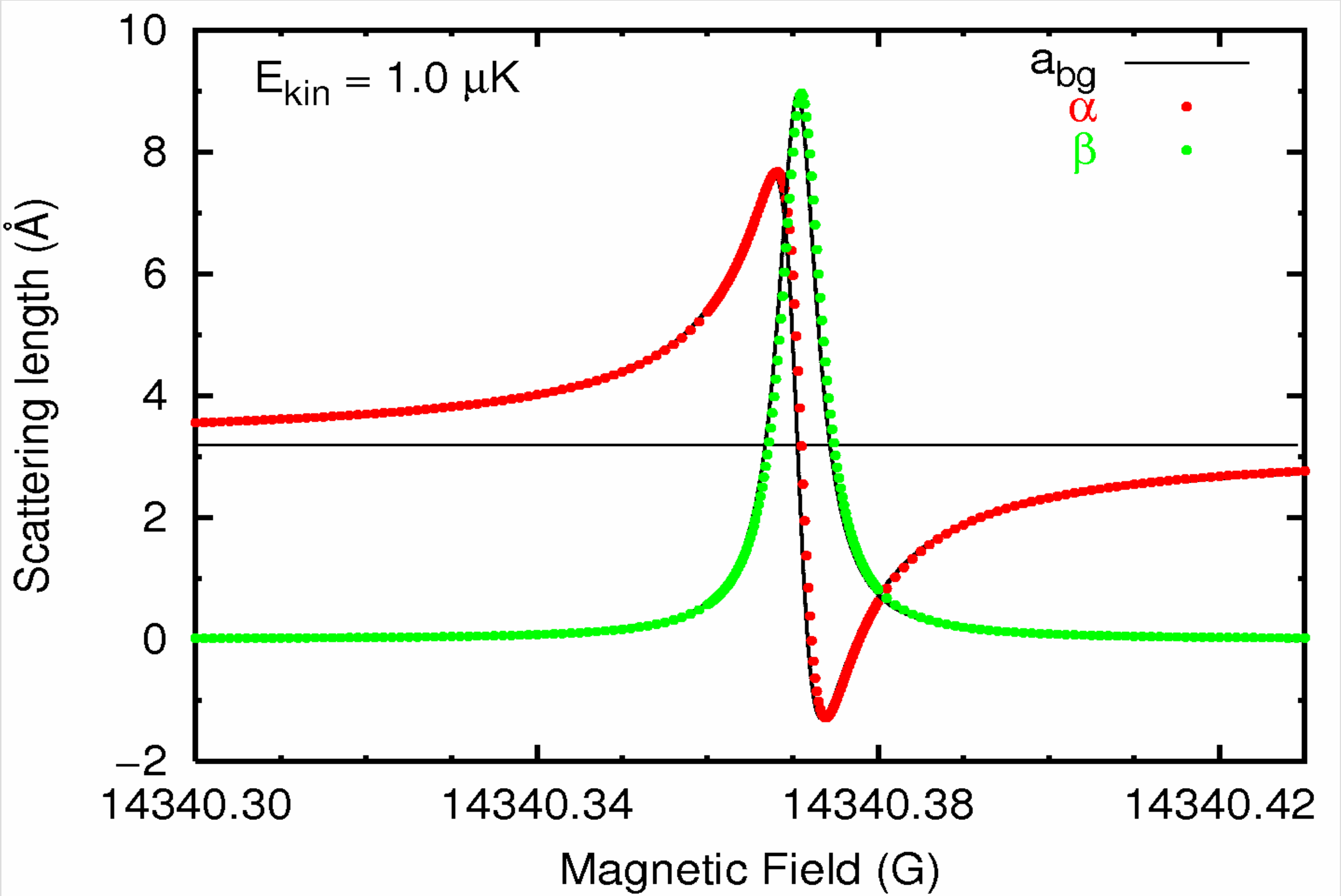
- Radius of circle in S_{00} depends on partial widths Γ_i:
  Radius is $\Gamma_0/\Gamma^{tot} \approx \Gamma_0/\Gamma^{inel}$
  [Γ_i is a measure of the coupling between the resonant state and open channel i]
- Partial width for elastic S-matrix element is proportional to k, $\Gamma_0 = 2k\gamma_0$
- Partial widths for inelastic S-matrix elements are independent of k, Γ^{inel}
- Complex scattering length is described by

$$a(B) = a_{\mathrm{bg}} + \frac{a_{\mathrm{res}}}{2(B - B_{\mathrm{res}})/\Gamma^{\mathrm{inel}} + \mathrm{i}}$$

with $a_{res} = 2\gamma_0/\Gamma^{inel}$

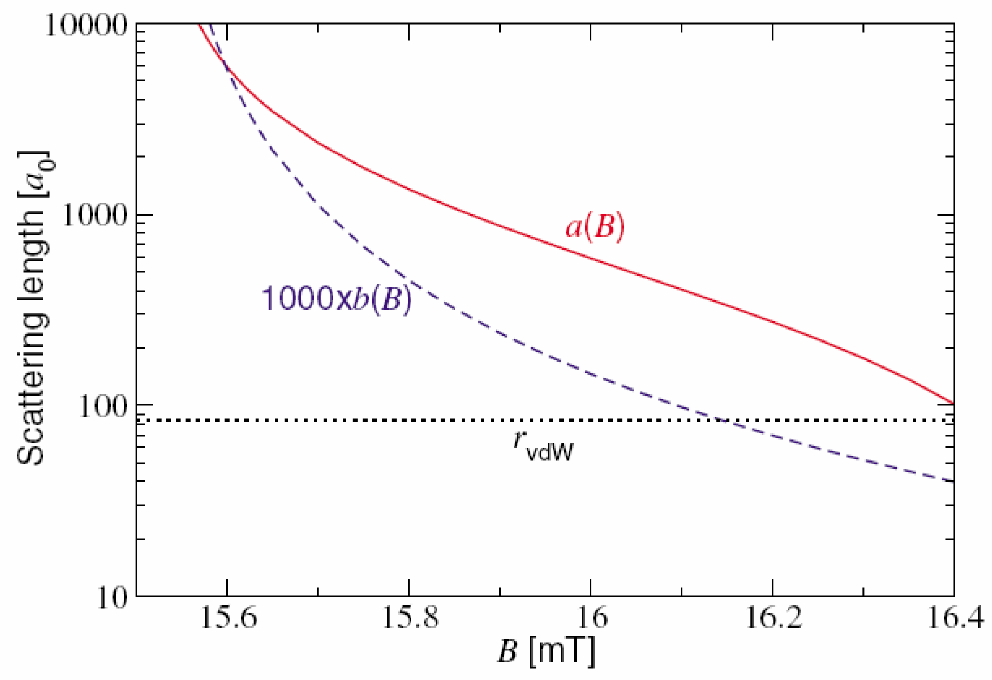
- Size of oscillation depends on *ratio* of coupling of resonant state to elastic and inelastic channels

He+NH inelastic scattering resonance:
Re(a) shows small oscillation, -Im(a) shows peak



How general is the suppression of poles?

- Size of oscillation characterized by a_{res}: depends on *ratio* of coupling of resonant state to elastic and inelastic channels
- Consider example cases:
  - Atom-atom scattering with spin exchange forbidden
    - Coupling to elastic channel is via central potential terms
    - Coupling to inelastic channels is via weak dipolar (spin-spin) coupling
    - Resonance amplitude is strong ($a_{res} > 10^4\ a_0$): pole-like behaviour in a



Coupled channel calculations: Köhler et. al., PRL 94, 020402 (2005)

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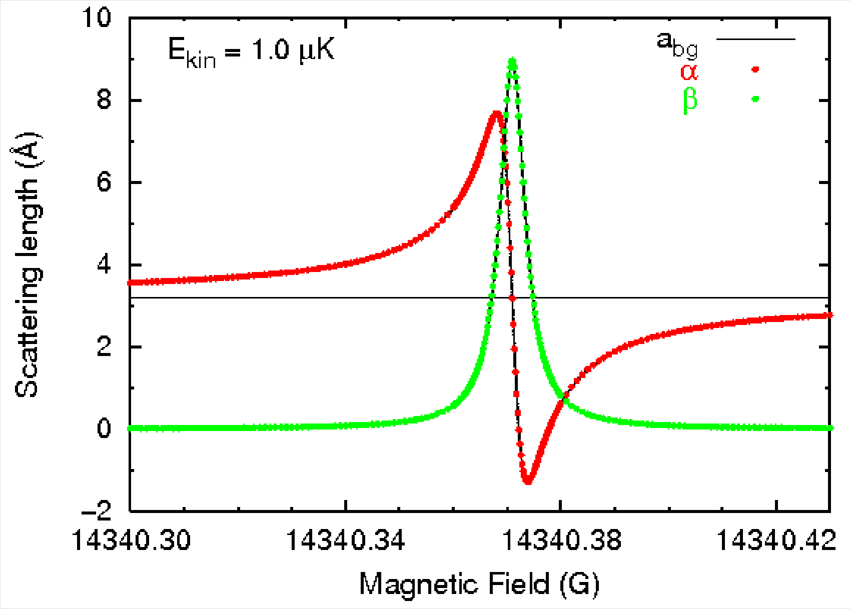
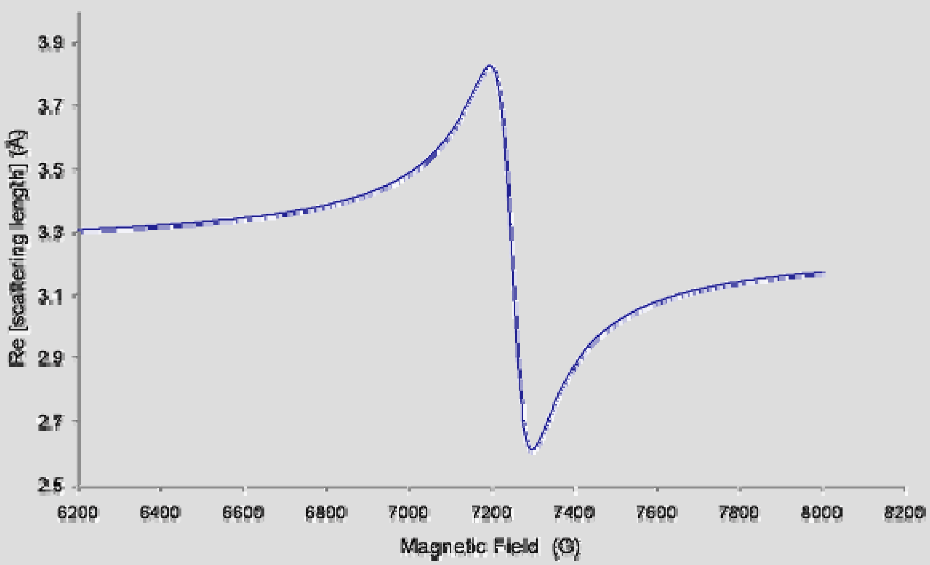
  - He – NH (N=0 states)
    - Coupling to elastic and inelastic channels is indirect coupling involving both potential energy terms and NH spin-spin coupling
    - Strengths of elastic and inelastic couplings are comparable, so resonance is weak
    - $a_{res} \approx 9$ Å: only small oscillation
    - Resonance width 0.006 G

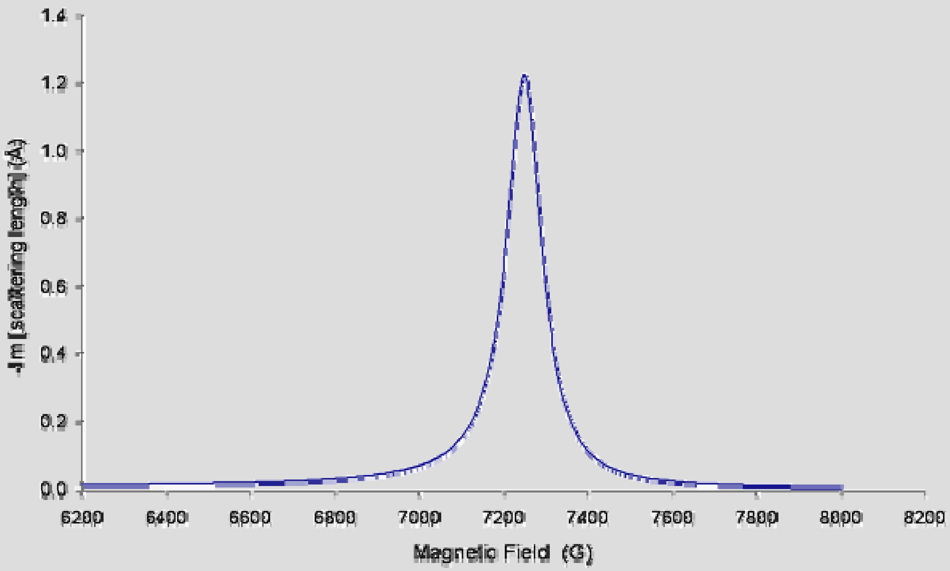


What about other cases?

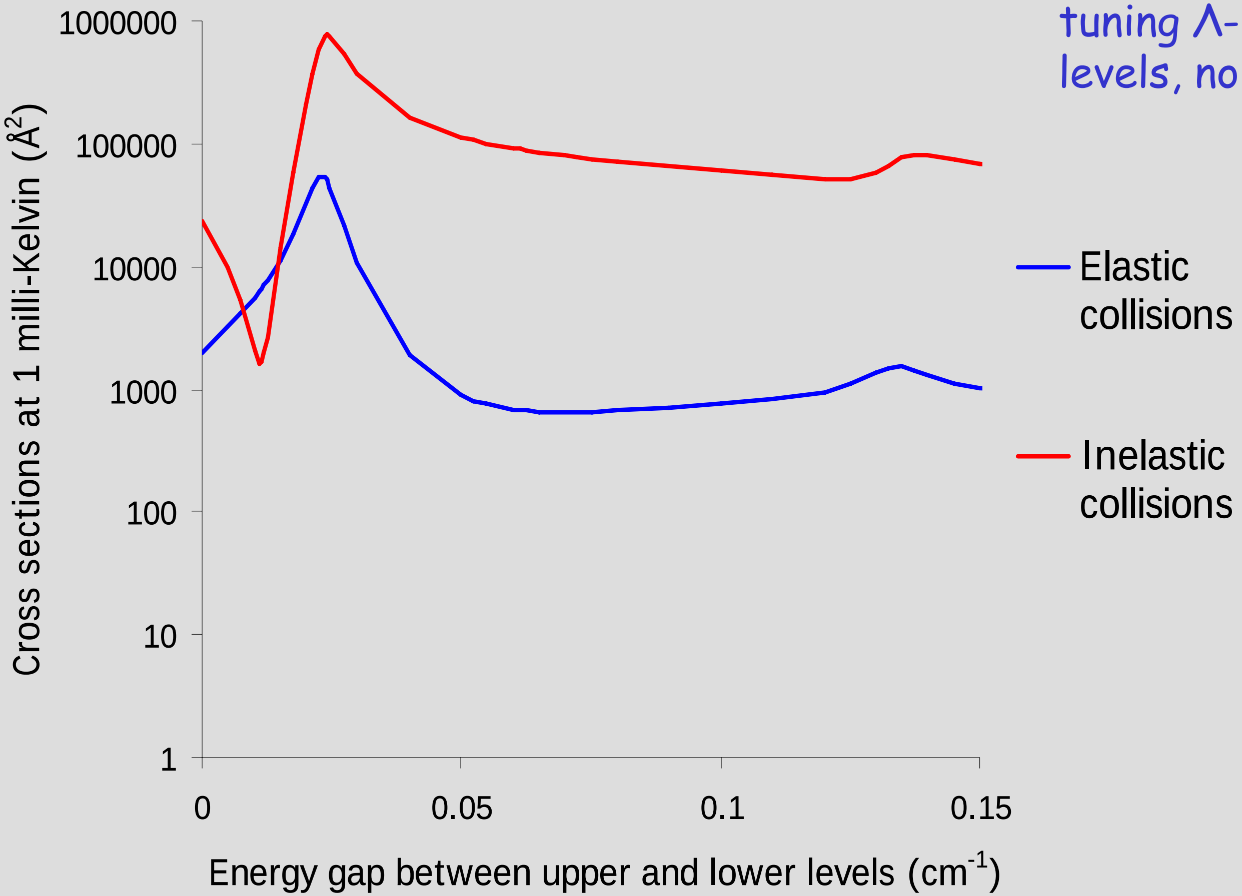
- Spin relaxation in non-zero rotational states of $^3\Sigma$ molecule:
  - Coupling to elastic and inelastic channels is via direct spin-spin coupling
  - Strengths of elastic and inelastic couplings are comparable, so resonance is suppressed: no pole-like behaviour in a

- He + NH (N=1): even weaker than for N=0
  - Amplitude of oscillation/peak a_{res} is only 1.2 Å
  - Resonance width ≈ 100 G





If the background scattering is inelastic, the inelastic cross sections can go down as well as up



Crude model of Rb + OH: tuning Λ-doubling to shift levels, no spin-orbit coupling

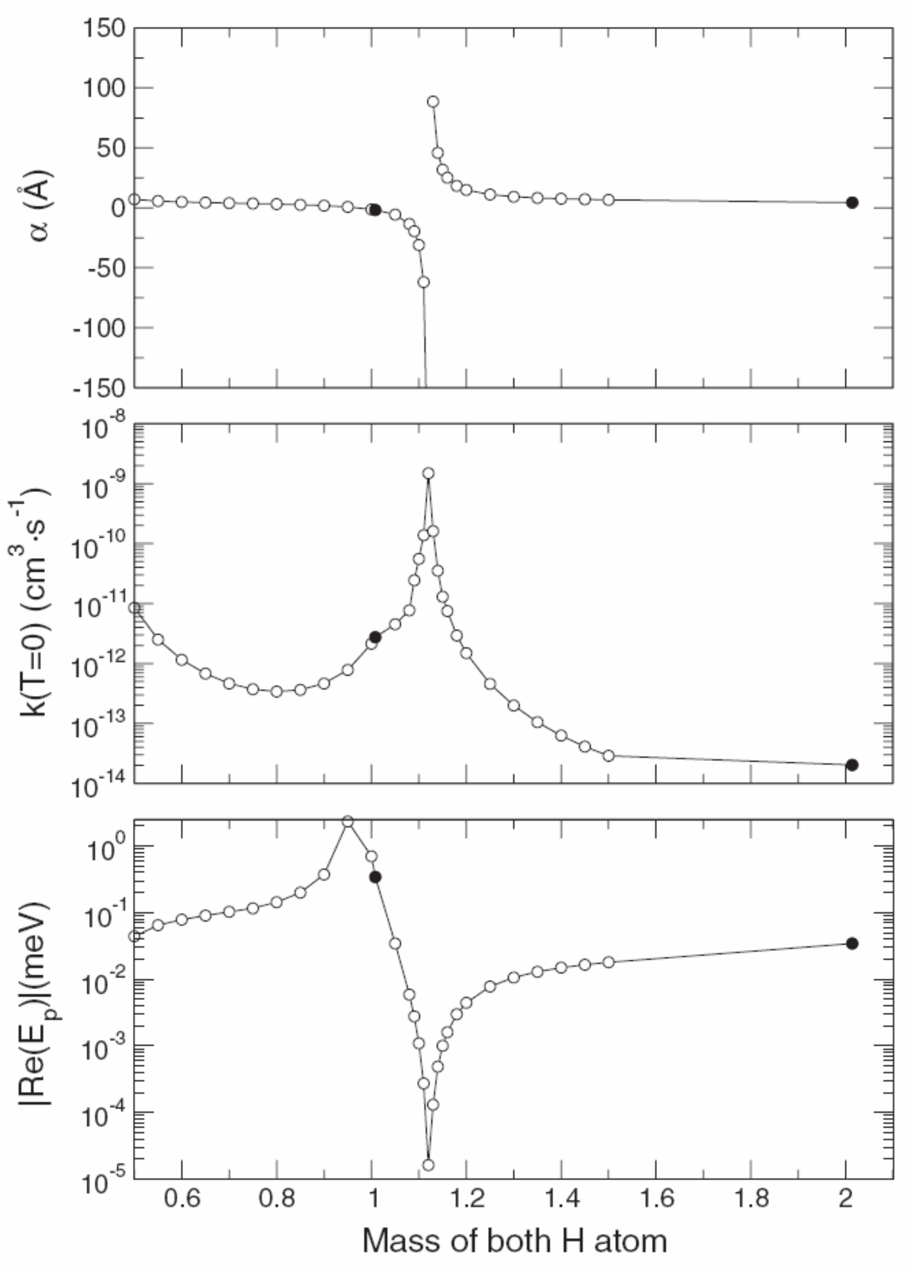
Cross section at fixed energy fluctuates as resonance crosses it: can achieve major changes in ratio of elastic to inelastic cross sections

What about other cases?

- Rotationally and vibrationally inelastic molecular collisions
  - Coupling to elastic and inelastic channels is via anisotropic potential terms
  - Strengths of elastic and inelastic couplings are comparable, so resonance is suppressed: no pole-like behaviour in a

- Atom-atom scattering with spin exchange allowed,
  - Coupling to elastic and inelastic channels is via central potential terms
  - Strengths of elastic and inelastic couplings are comparable, so resonance will be suppressed: no pole-like behaviour in a

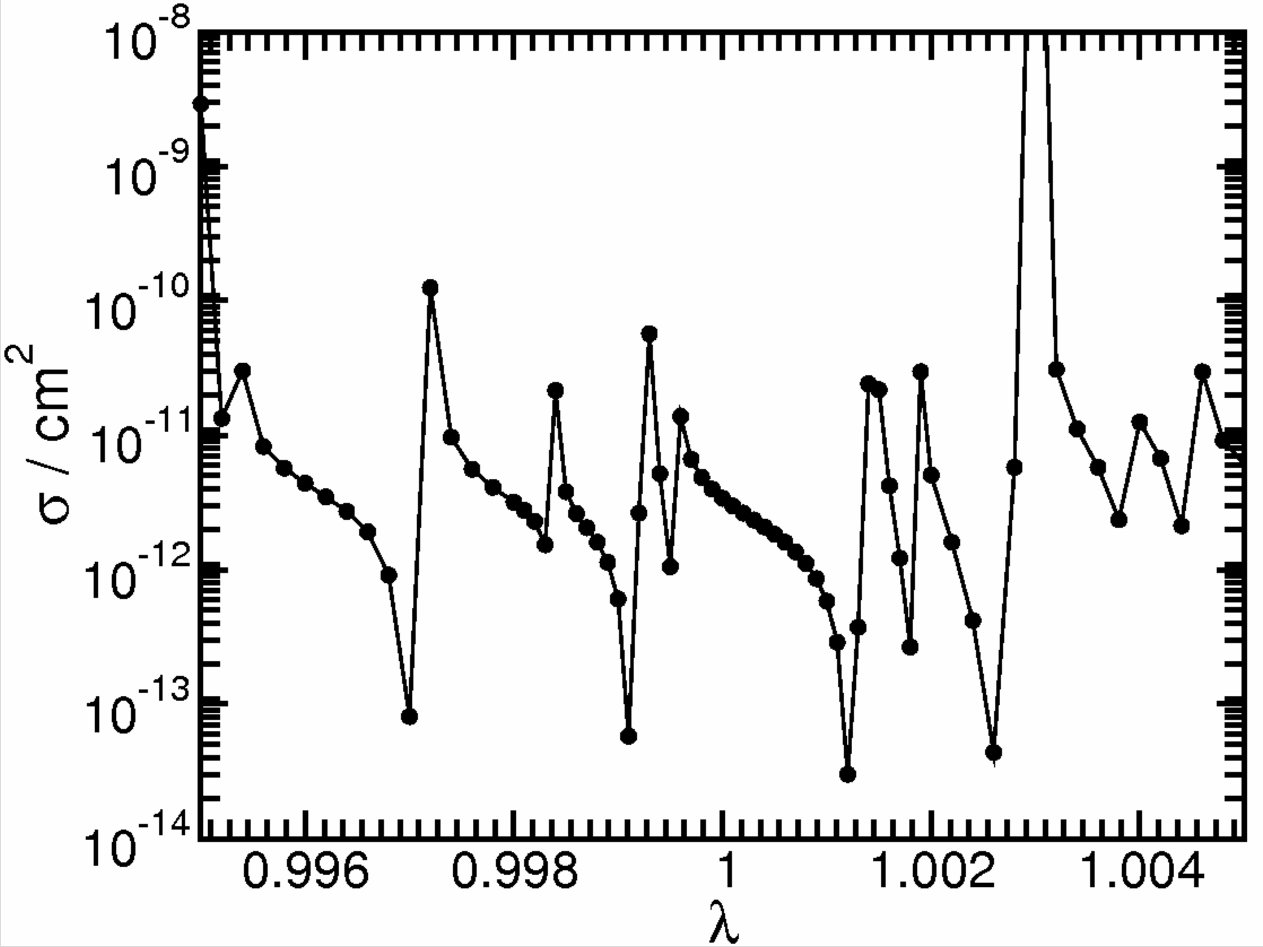
F + H_2 reactive scattering (with a barrier)



- For entrance channel resonances, the coupling to the elastic channel is strong but the coupling to exoergic (reactive) channels (through the barrier) is weak
- Resonance amplitude is strong: pole-like behaviour in a
- Real part of scattering length shows pole-like signature (peaks at least ± 100 Å)
- Reactive rate shows substantial peak.

Coupled channel calculations by Bodo et al., JPB 37, 3641 (2004)

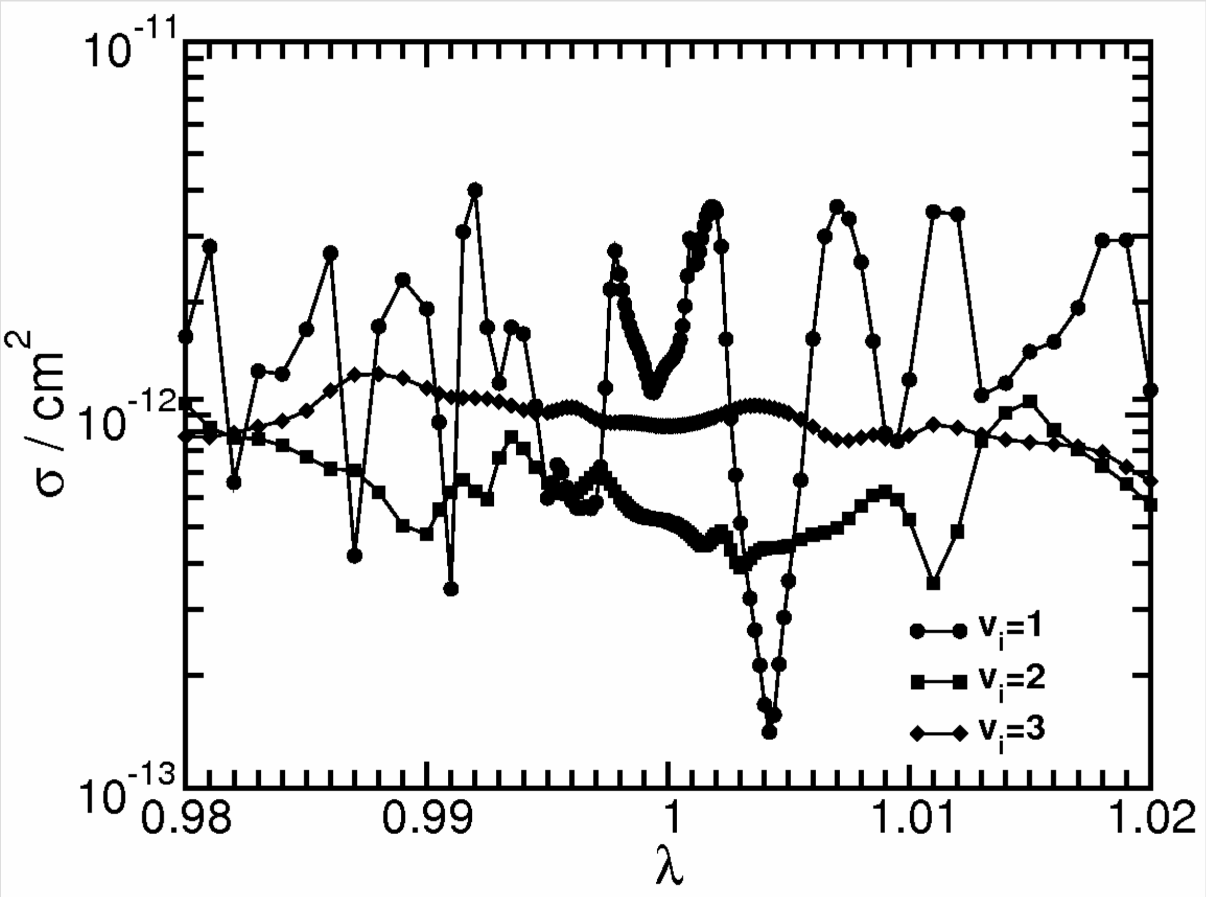
Reactive scattering calculations on Li + Li_2 (barrierless)



Elastic cross sections as function of potential scaling factor λ

σ_{el} for v=0 shows large peaks due to poles in scattering length

Reactive scattering calculations on Li + Li_2 (barrierless)



Elastic cross sections for vibrationally excited Li_2 as function of potential scaling factor λ

- Coupling to inelastic channels is reduced by large kinetic energy release
- Weakish resonances still observed for v=1, but almost completely suppressed for v=3

Feshbach resonance conclusions

- Inelastic scattering can strongly suppress resonant peaks in scattering lengths and cross sections
- For systems where resonances are very weakly coupled to inelastic channels, there is still (nearly) a pole in the scattering length:
  - Atom-atom systems with spin exchange forbidden e.g. for $^{85}Rb + ^{85}Rb$ in high $|m_f|$ states, inelastic scattering comes only from very weak dipolar spin-spin interactions
  - Reactive scattering in $F+H_2$: coupling to exoergic (reactive) channels is suppressed by high barrier
- For most molecular systems (and some atomic systems), the peaks will be strongly suppressed.
- The suppression has important consequences for control of quantum gases
- When resonances are strongly coupled to inelastic channels, cross sections are not so sensitive to details of the potential

Who did what, and where can I find it?

- RbOH surfaces:
  - Pavel Soldán (Post-doc, 2000-2005: now faculty in Prague)
  - Daniel Potter (MSc student)

- Scattering:
  - RbOH: John Bohn and Manuel Lara (JILA)
    - PRL, 97, 183201 (2006).
    - PRA 75, 012704 (2007).
  - He + NH: Maykel Leonardo González-Martínez (Cuba)
    - Formal theory, arXiv:physics/0610210
    - He + NH application, PRA 75, 022702 (2007).

- Recent reviews on alkali metal dimers:
  - Molecule formation in ultracold atomic gases, Int. Rev. Phys. Chem. 25, 497 (2006)
  - Molecular collisions in ultracold atomic gases, Int. Rev. Phys. Chem. 26, 1 (2007); arXiv:physics/0610219