Semiclassical Analysis of Quantum Mechanical Calculations of Rotationally Inelastic Collisions of He and Ar with NaK

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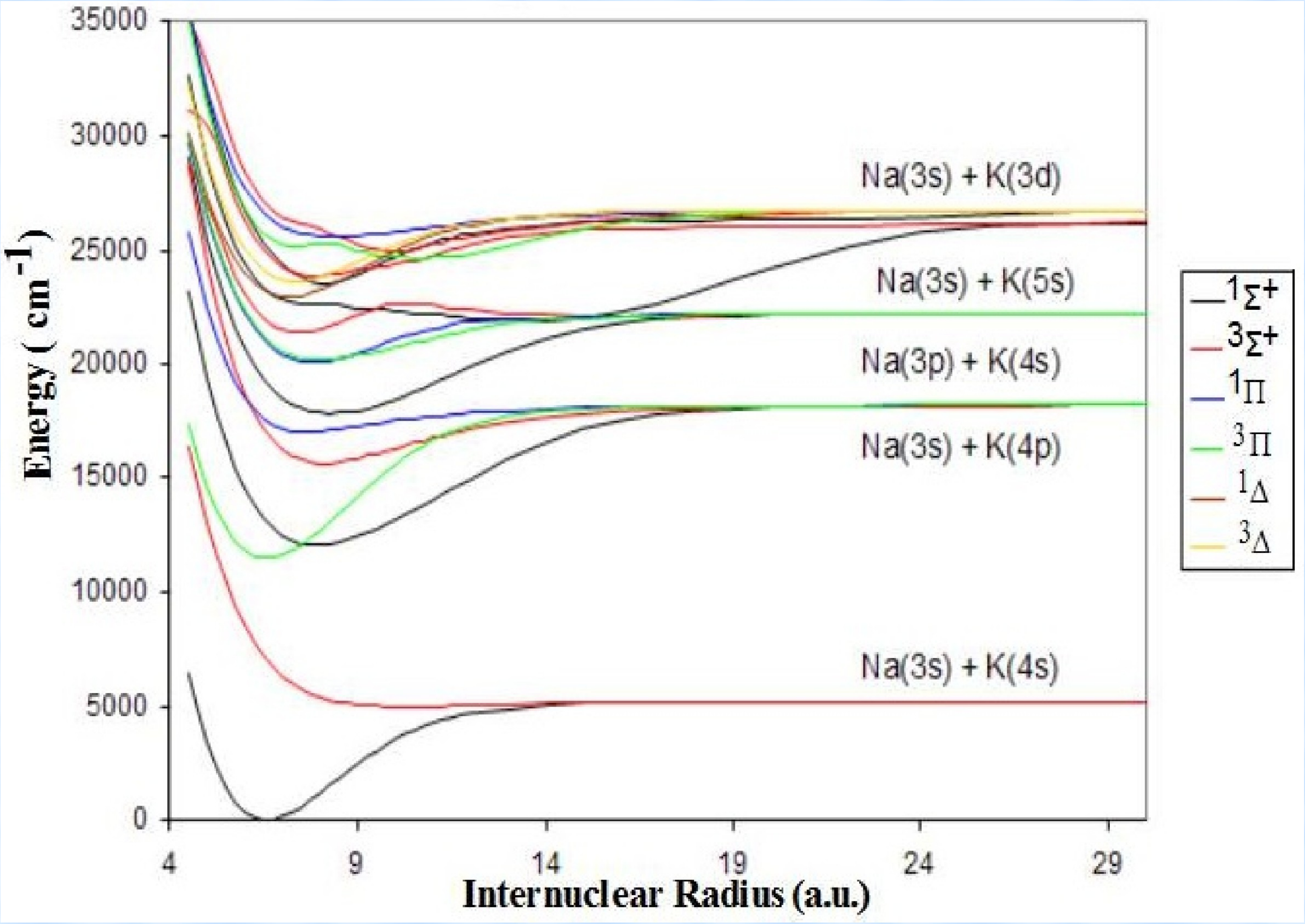
Background

- Experiment – Prof. Huennekens' group
  - Rotationally inelastic collisions at thermal energies

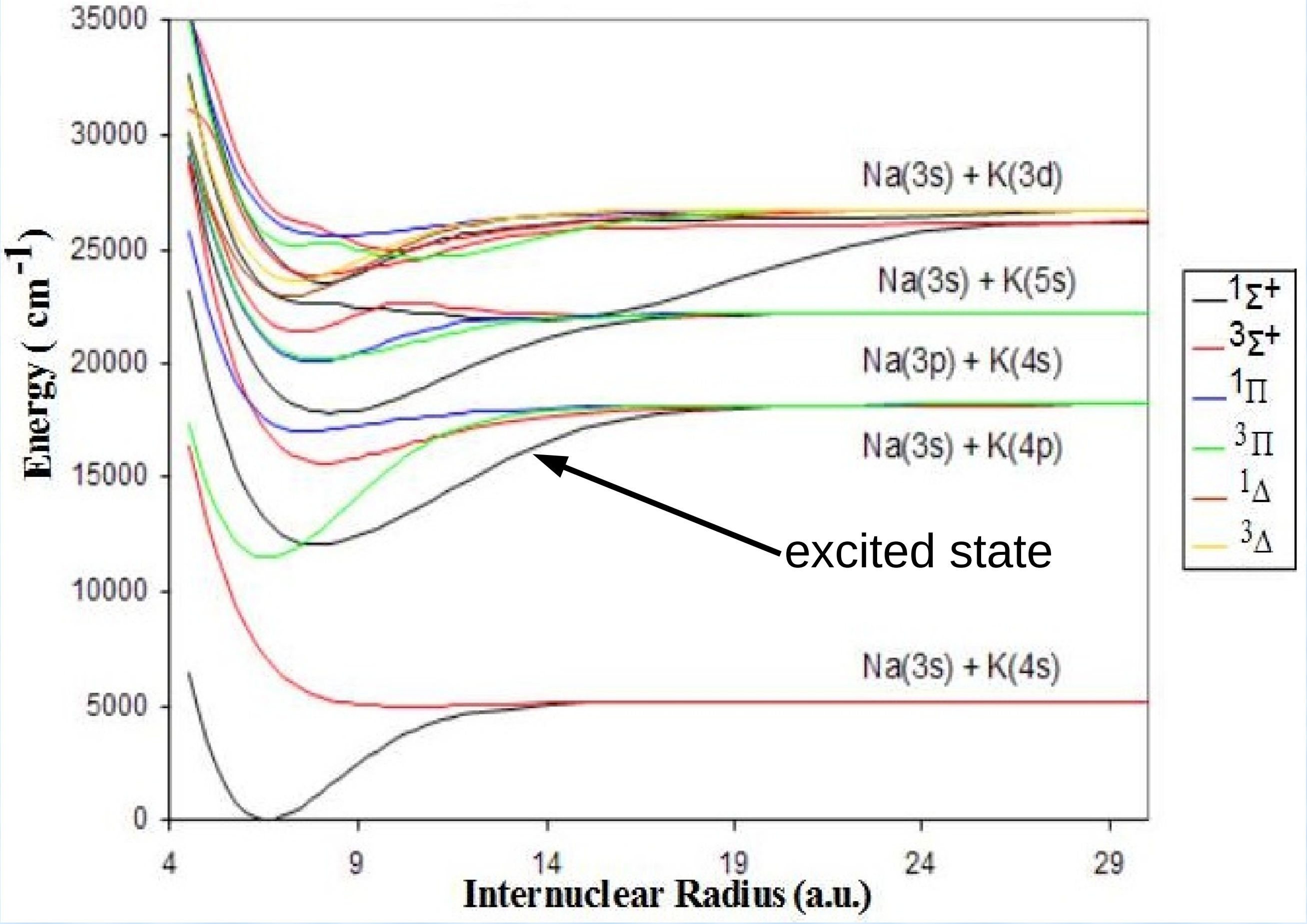
$$He + NaK(v,j,m) \rightarrow He + NaK(v,j',m')$$
$$Ar + NaK(v,j,m) \rightarrow Ar + NaK(v,j',m')$$

- Theory – Prof. Hickman's group

NaK potential curves



NaK potential curves

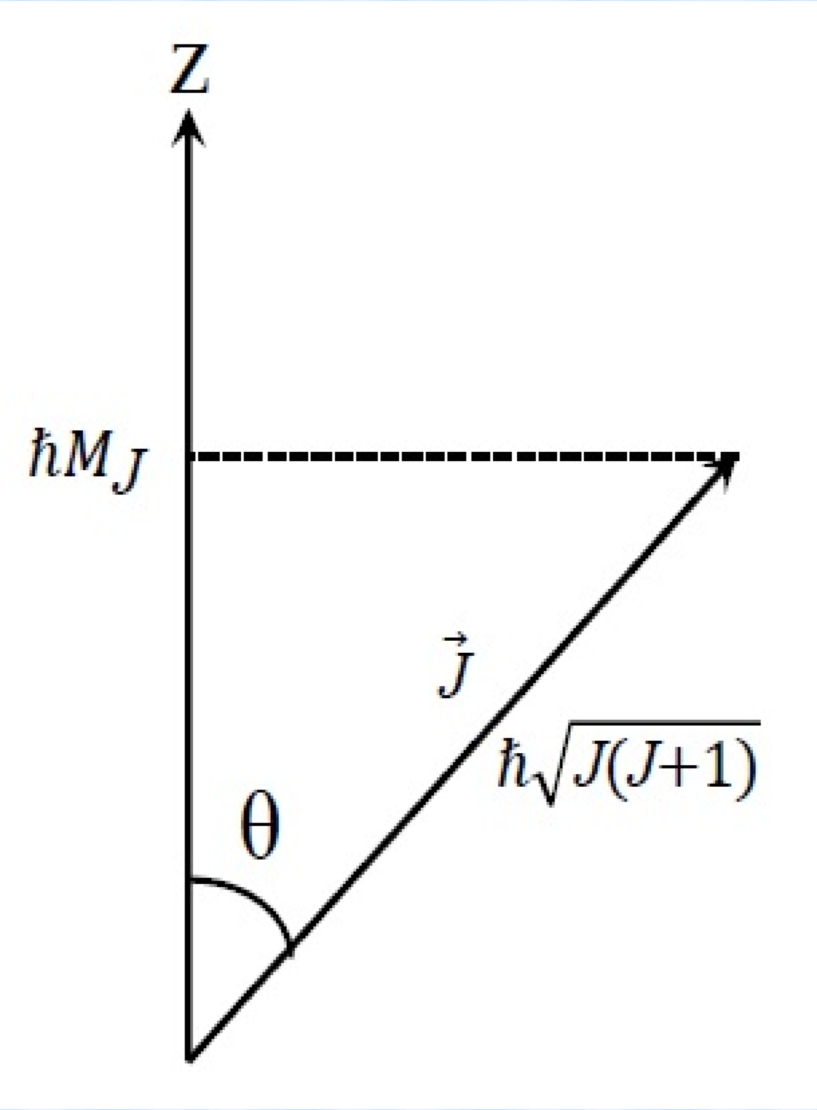


Experimental Measurements

- Cross sections for $j \rightarrow j'$ transitions
- Fraction of orientation preserved in collisions
  - Typically orientation is random, <m>=0
  - Laser polarization can create nonrandom <m>

$$O^j = \frac{\langle m \rangle}{\sqrt{j(j+1)}}$$

Vector Model

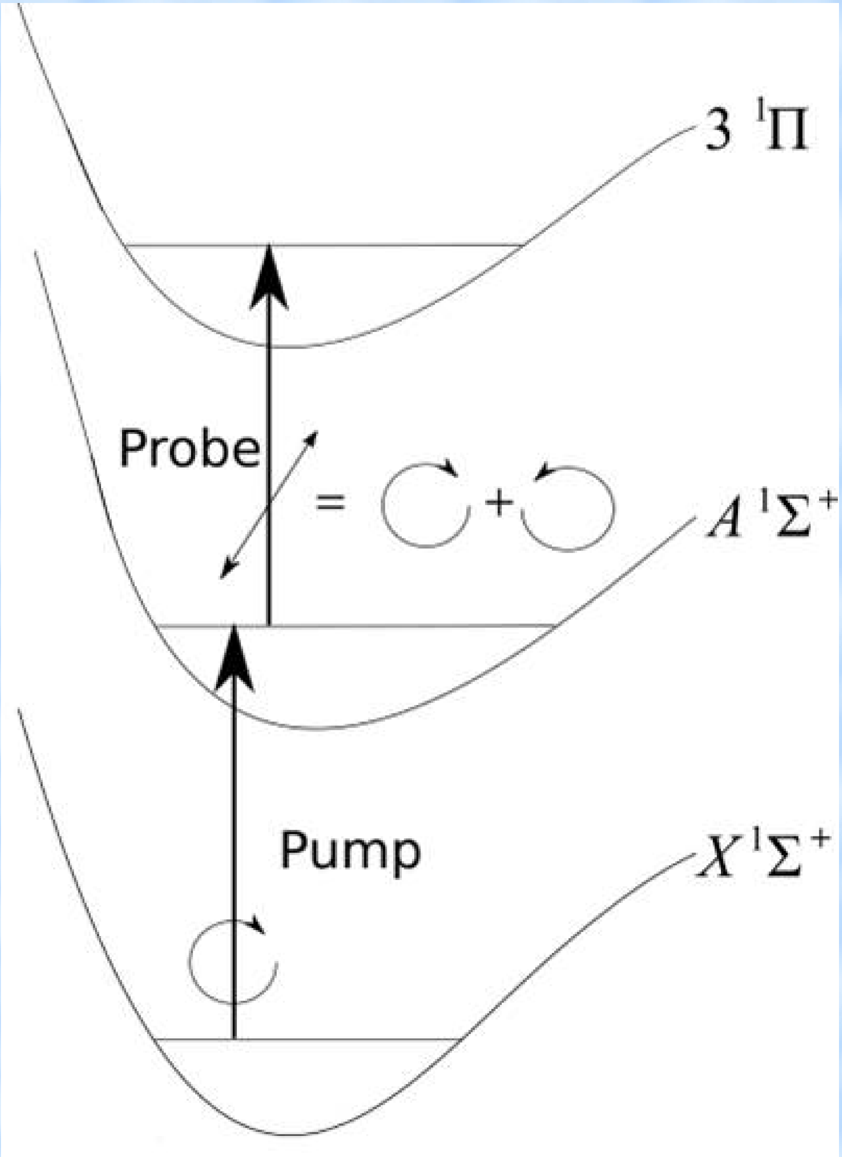


$$\cos\theta = \frac{m}{\sqrt{j(j+1)}}$$

$$O^j = \frac{\langle m \rangle}{\sqrt{j(j+1)}}$$

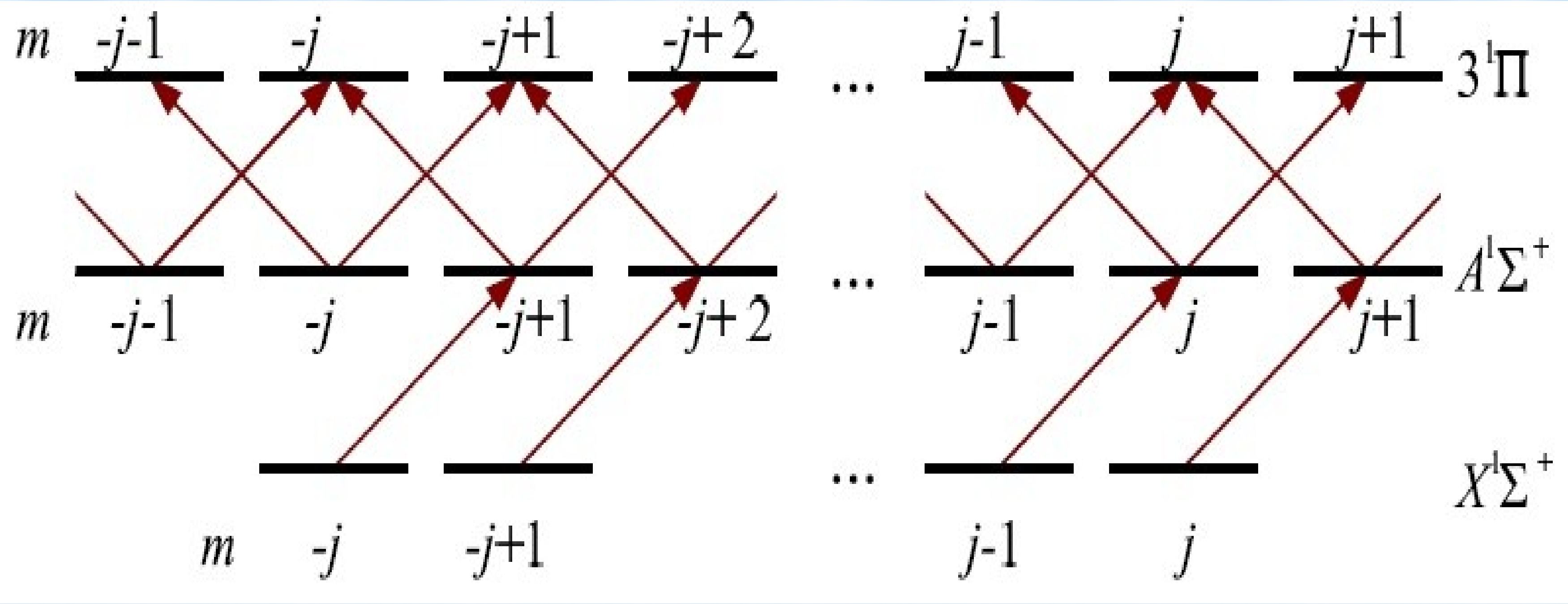
Creating nonzero orientation

- Use polarized lasers
- Pump creates an orientation in the A state
- Probe is used to measure polarization after collisions



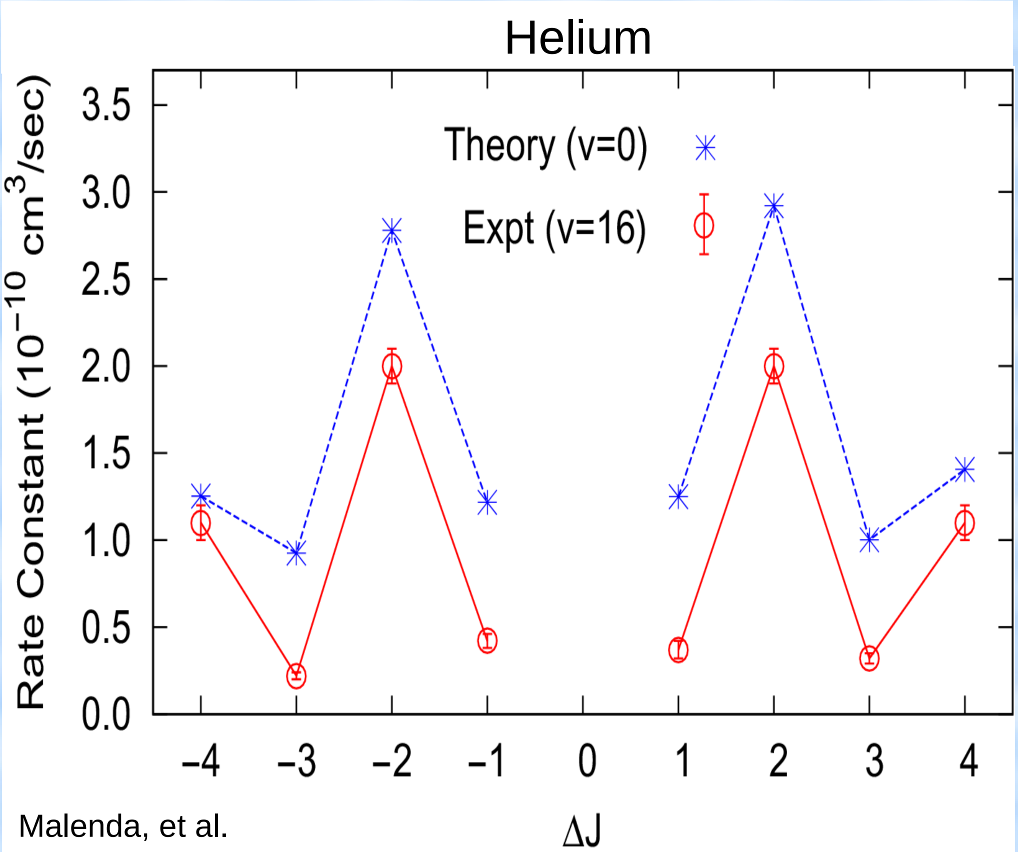
Creating nonzero orientation

- Selection rule $\Delta m = +1$ leads to preferential population of large m



Comparison for $\sigma(j \rightarrow j')$

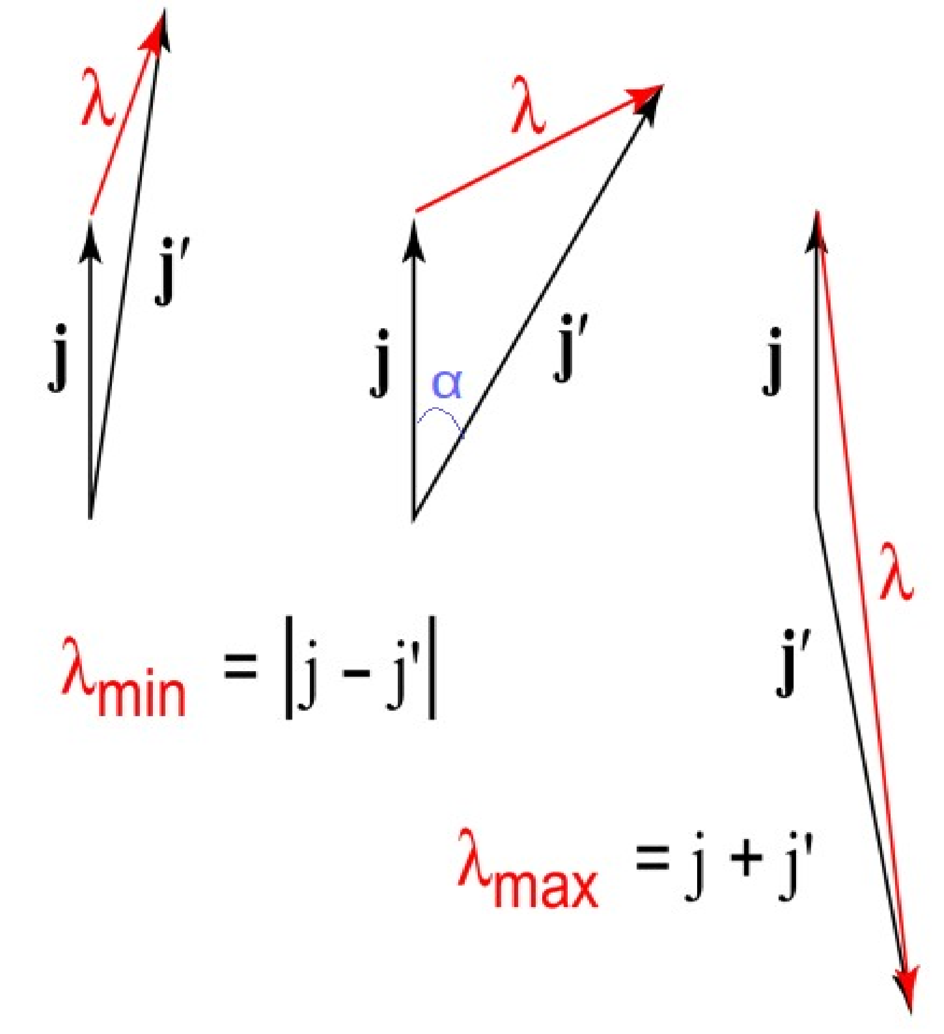
- Propensity for Δj to be even
  - Strict selection rule for homonuclear molecules
  - Detailed calculations necessary for each system



Theory – quantum mechanical

$$\sigma(j \rightarrow j') = \frac{\pi}{(2j+1)k_j^2} \sum_{\lambda=|j-j'|}^{j+j'} (2\lambda+1) B_\lambda(j, j')$$

- λ is related to transfer of angular momentum from projectile (He or Ar) to target NaK



Theory – semiclassical

- λ is related to the tipping angle α between j and j'

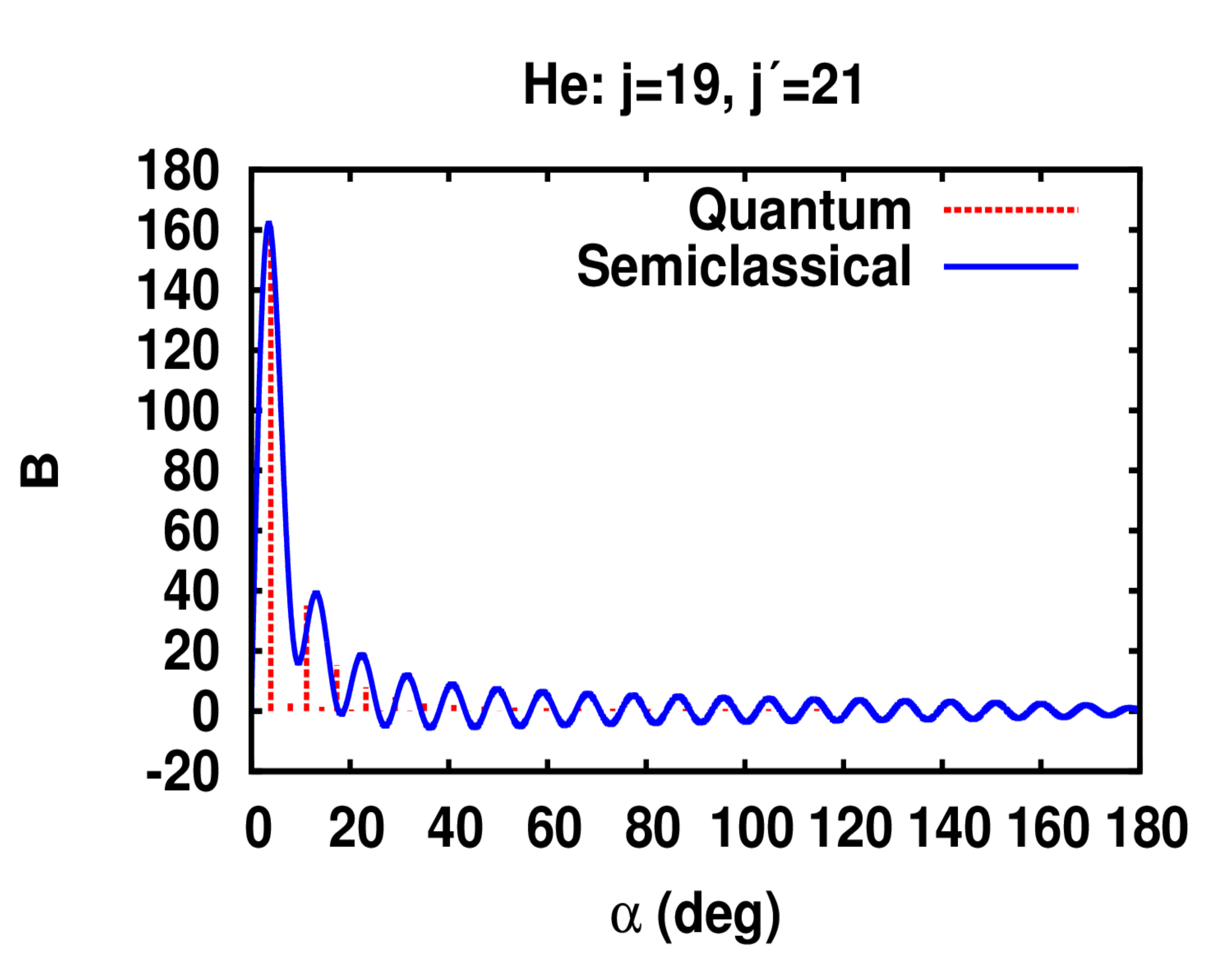
$$\lambda(\lambda+1)=j(j+1)+j'(j'+1)-2\sqrt{j(j+1)j'(j'+1)}\cos\alpha$$

- Approximate cross sections

$$\sigma(j\rightarrow j')=\frac{\pi(j'+1/2)}{k_j^2}\int_0^\pi B(j,j',\cos\alpha)\sin\alpha\,d\alpha$$

- B(j,j',cosα) gives the distribution of tipping angles

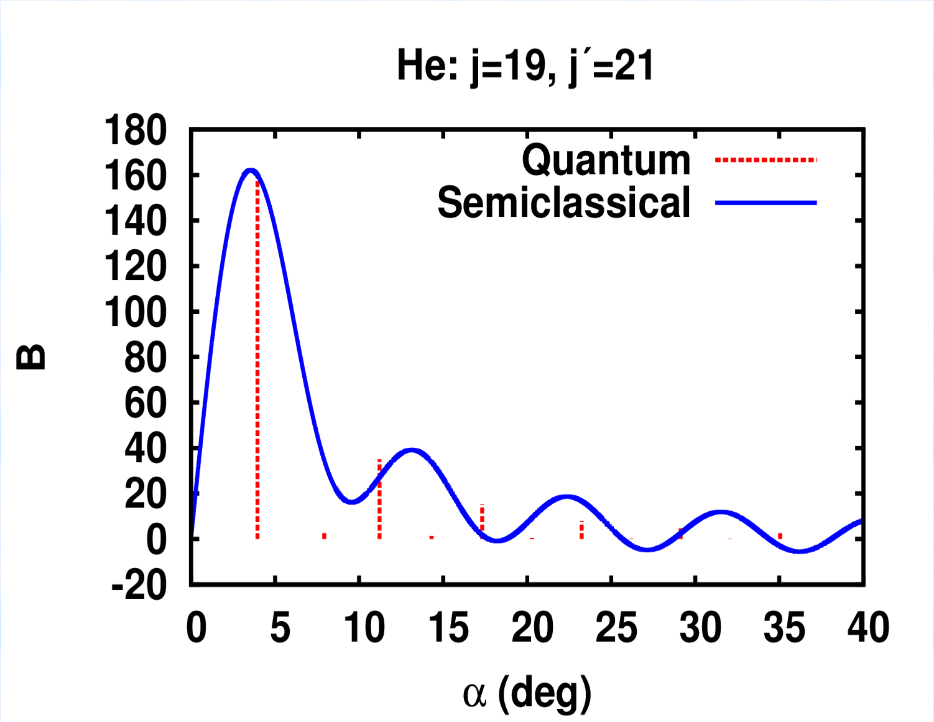
He + NaK; Δj=2



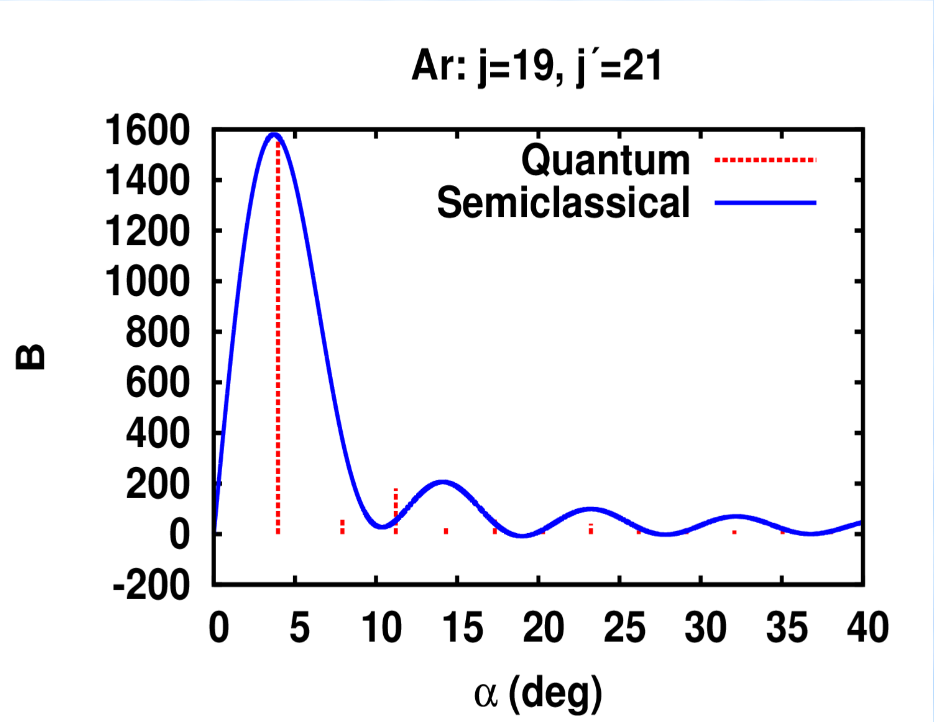
Distribution of tipping angles α

Comparison Δj=2

He + NaK



Ar + NaK



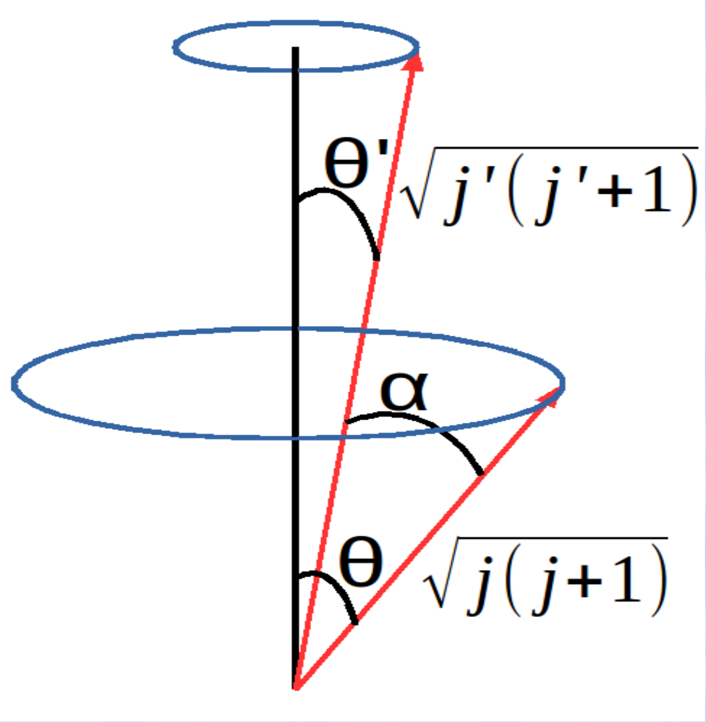
Distribution of tipping angles α

Trends in $B(j,j,\cos\alpha)$

- Helium
  - Larger average tipping angle for odd Δj than even Δj
  - α tends to be small for the more probable transitions
- Argon
  - Larger $\langle\alpha\rangle$ as Δj increases
  - No even/odd Δj propensities
- Comparison
  - Argon B values are larger
  - As $\bar{j}$ (mean value of j) increases, $\langle\alpha\rangle$ decreases

Relation between Δθ, Δm, B

- σ(jm → j'm')
- Δθ is related to Δm
  - If Δθ is small, and j changes, m must also change



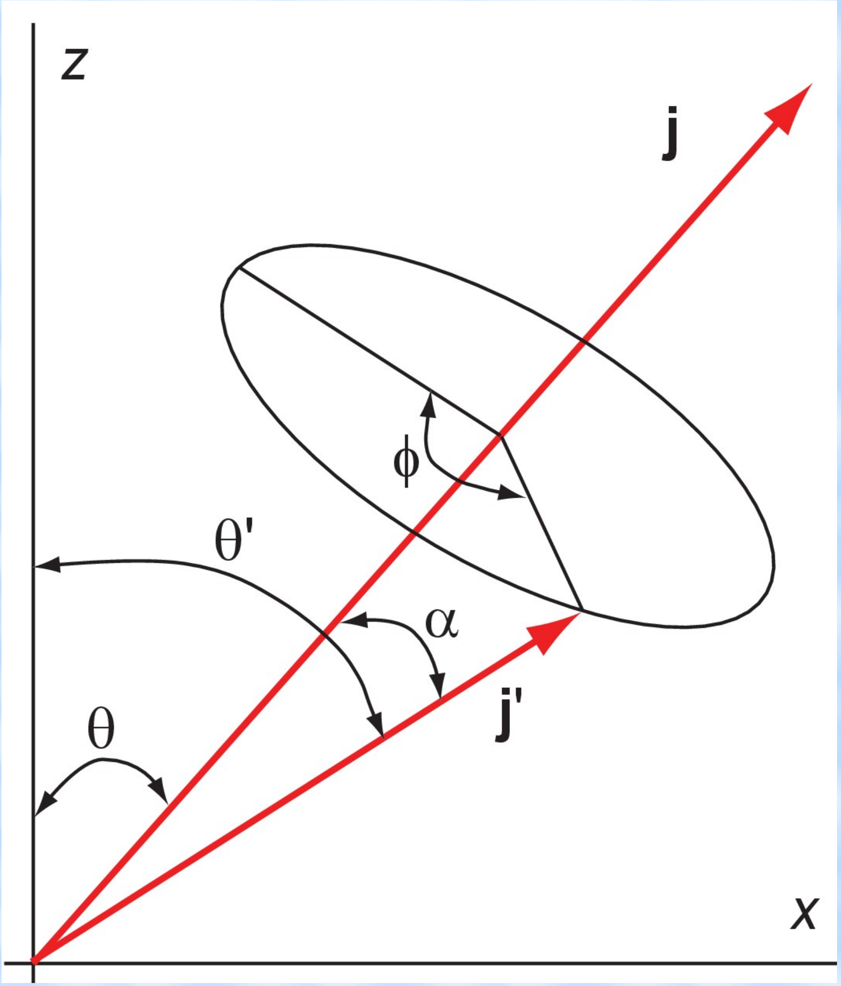
$$\cos\theta = \frac{m}{\sqrt{j(j+1)}}$$

$$\Delta\theta = \theta' - \theta \neq \alpha$$

$$\Delta\theta = \cos^{-1}\left(\frac{m'}{\sqrt{j'(j'+1)}}\right) - \cos^{-1}\left(\frac{m}{\sqrt{j(j+1)}}\right)$$

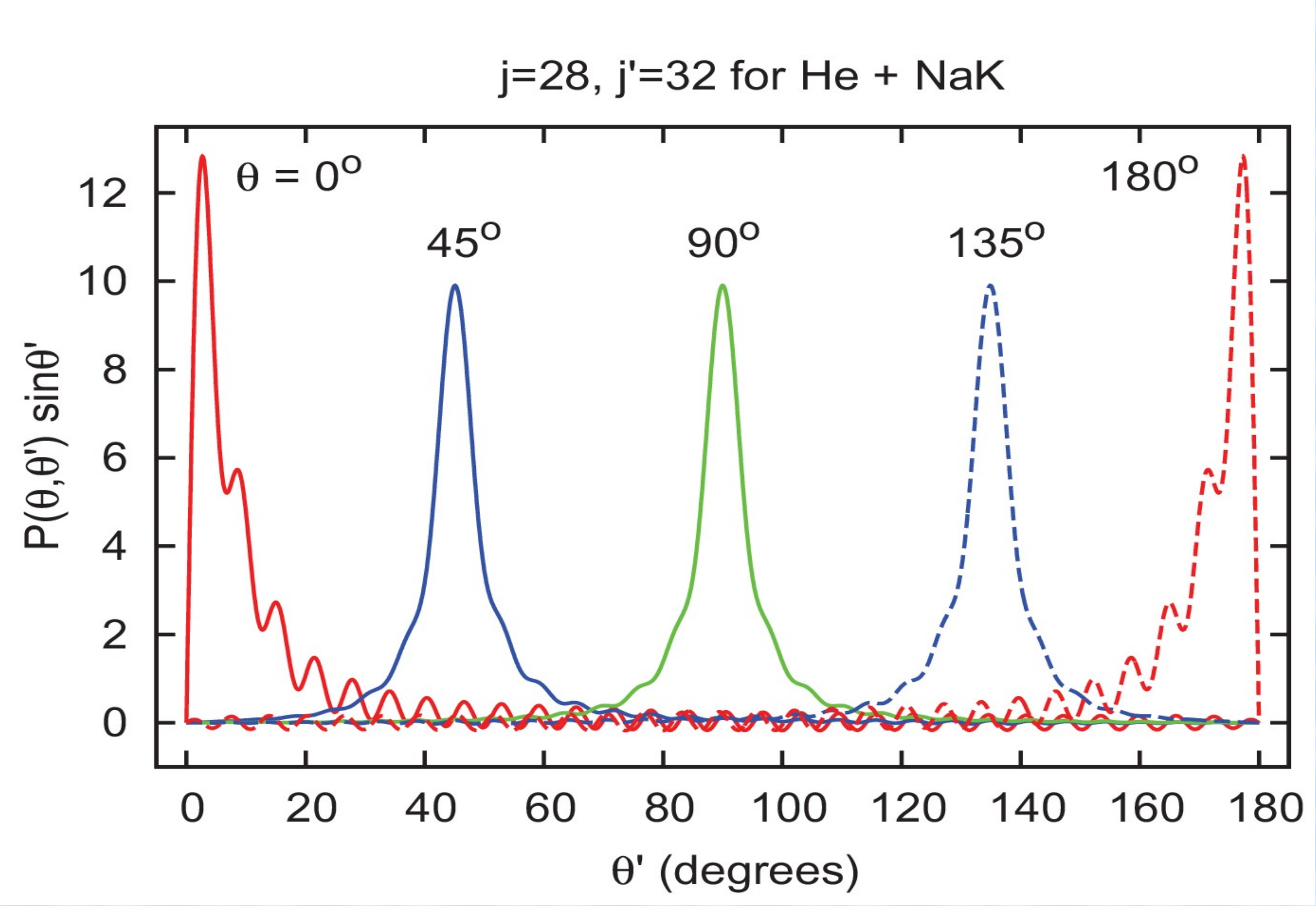
Methods for Δθ analysis

- Quantum mechanical model
  - Compute Δθ from j, j', m, m'
  - Create histogram angle bins
  - Convolve with Gaussian
- Compute semiclassical model



$$P(\theta,\theta')\sin\theta' = \frac{\sin\theta'}{\pi}\int_{-1}^{1}\frac{B(j,j';\cos\theta\cos\theta' + y\sin\theta\sin\theta')}{\sqrt{1-y^2}}\,dy$$

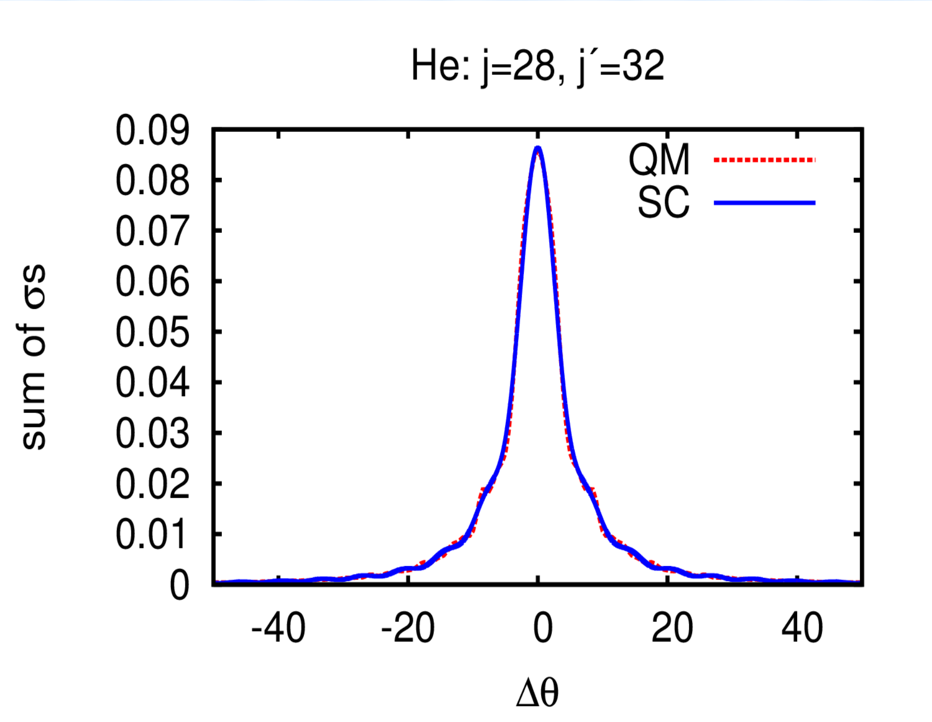
Semiclassical $\theta \rightarrow \theta'$

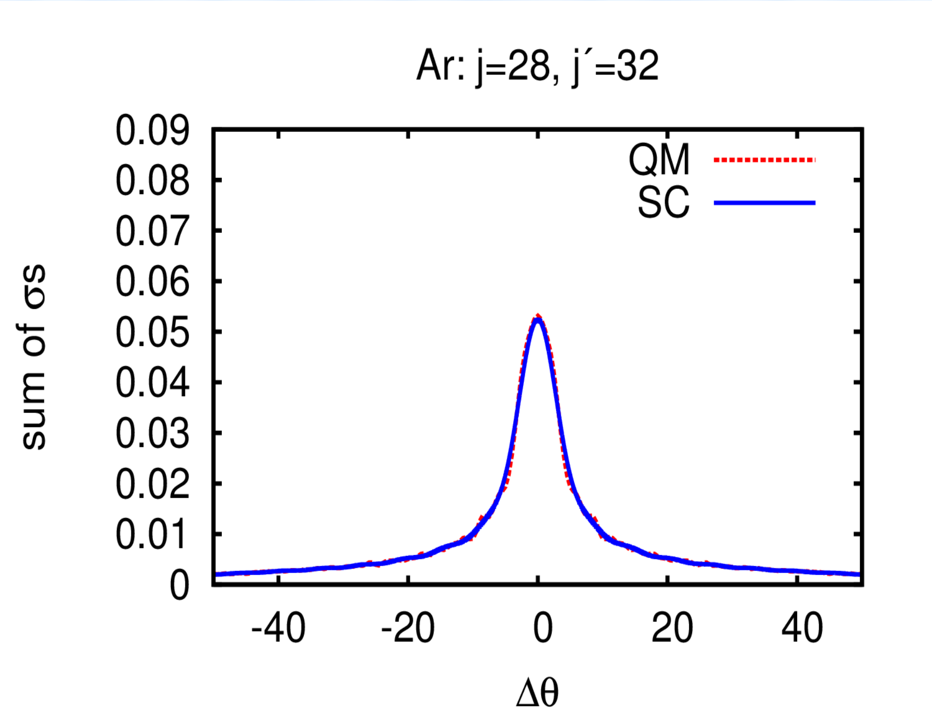


$\bar{j}=30$, $\Delta j=4$

He + NaK

Ar + NaK



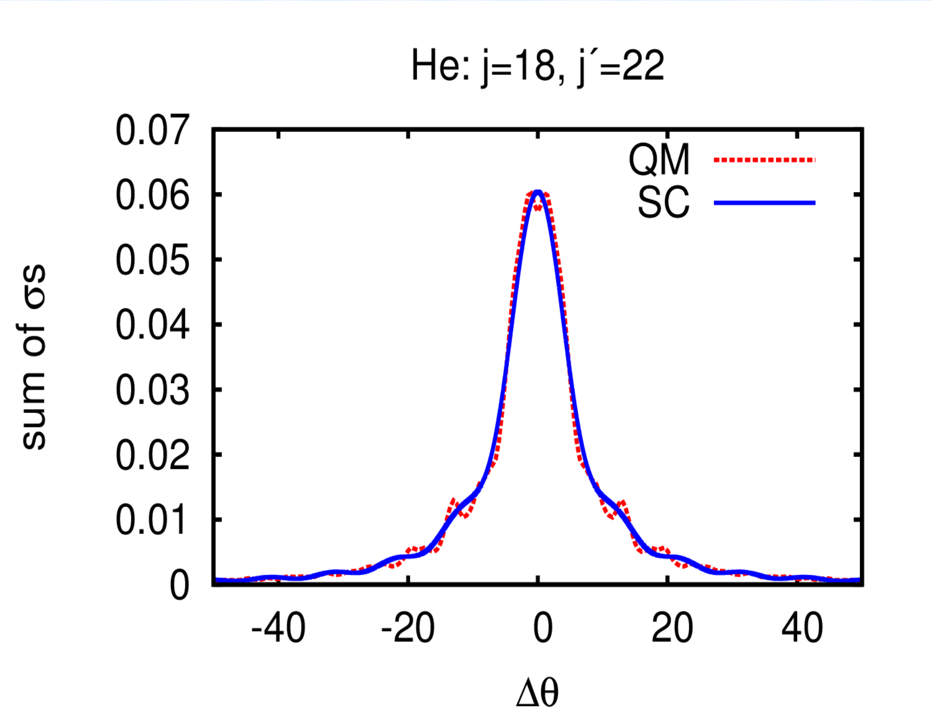


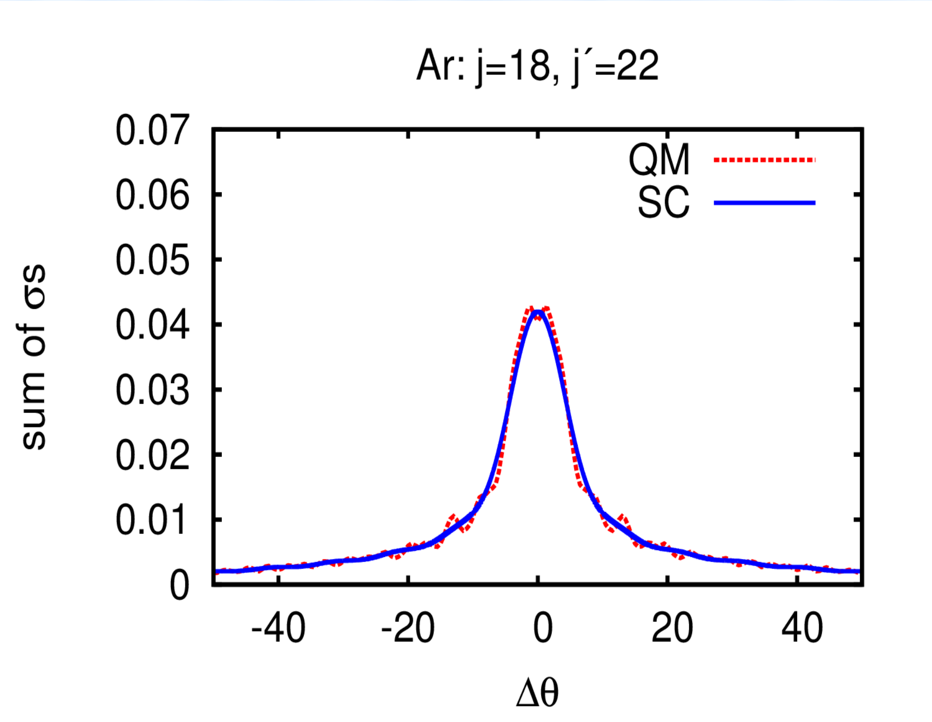
Distribution of $\Delta\theta$ averaged over all collisions

$\bar{j}$=20, Δj=4

He + NaK

Ar + NaK

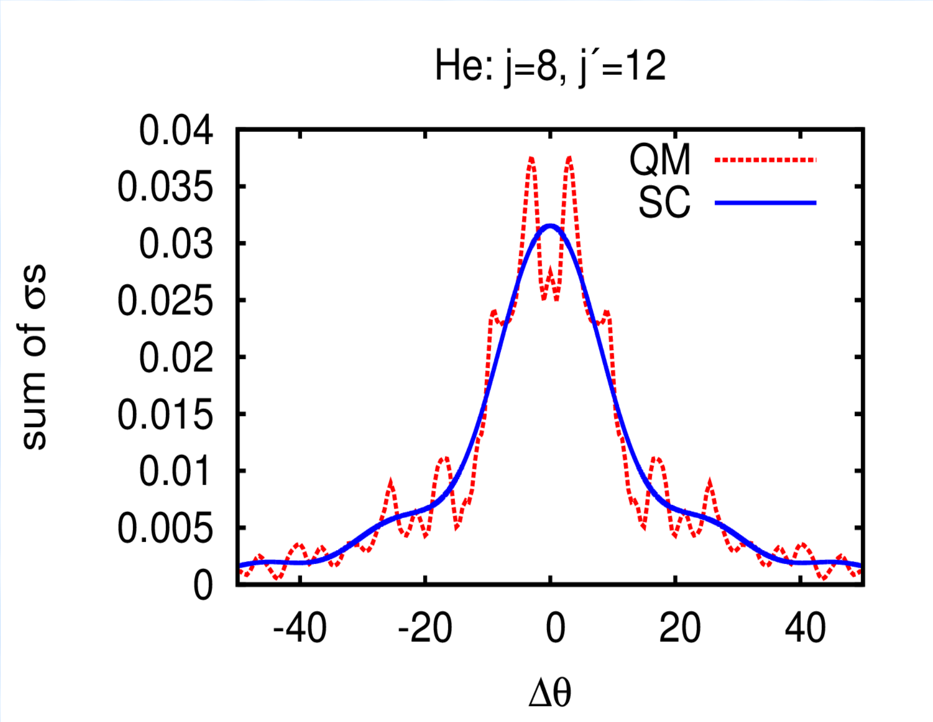




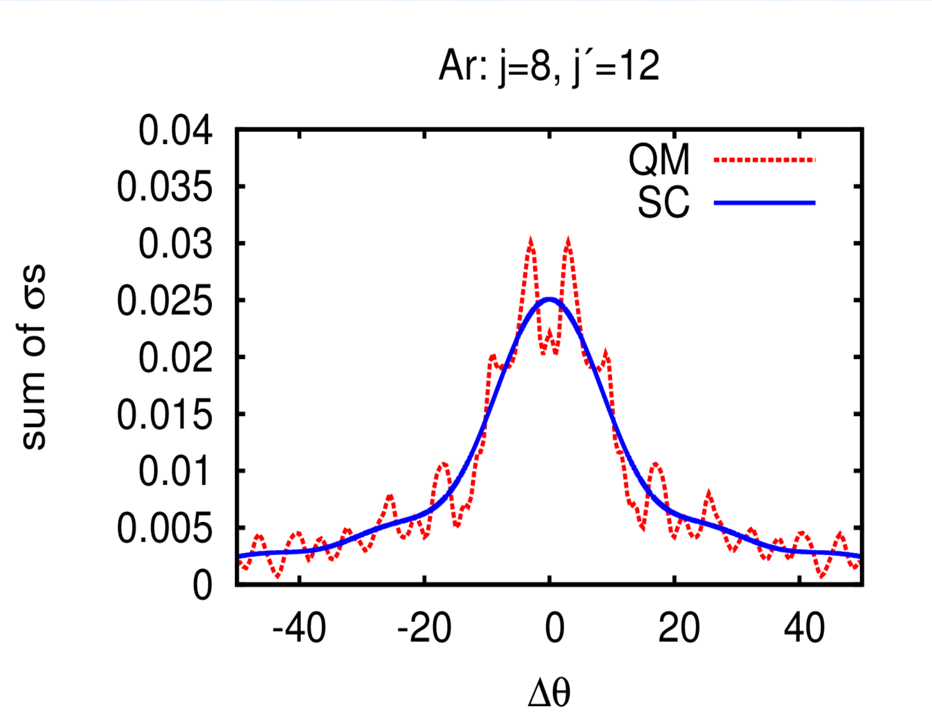
Distribution of Δθ averaged over all collisions

$\bar{j}$=10, Δj=4

He + NaK

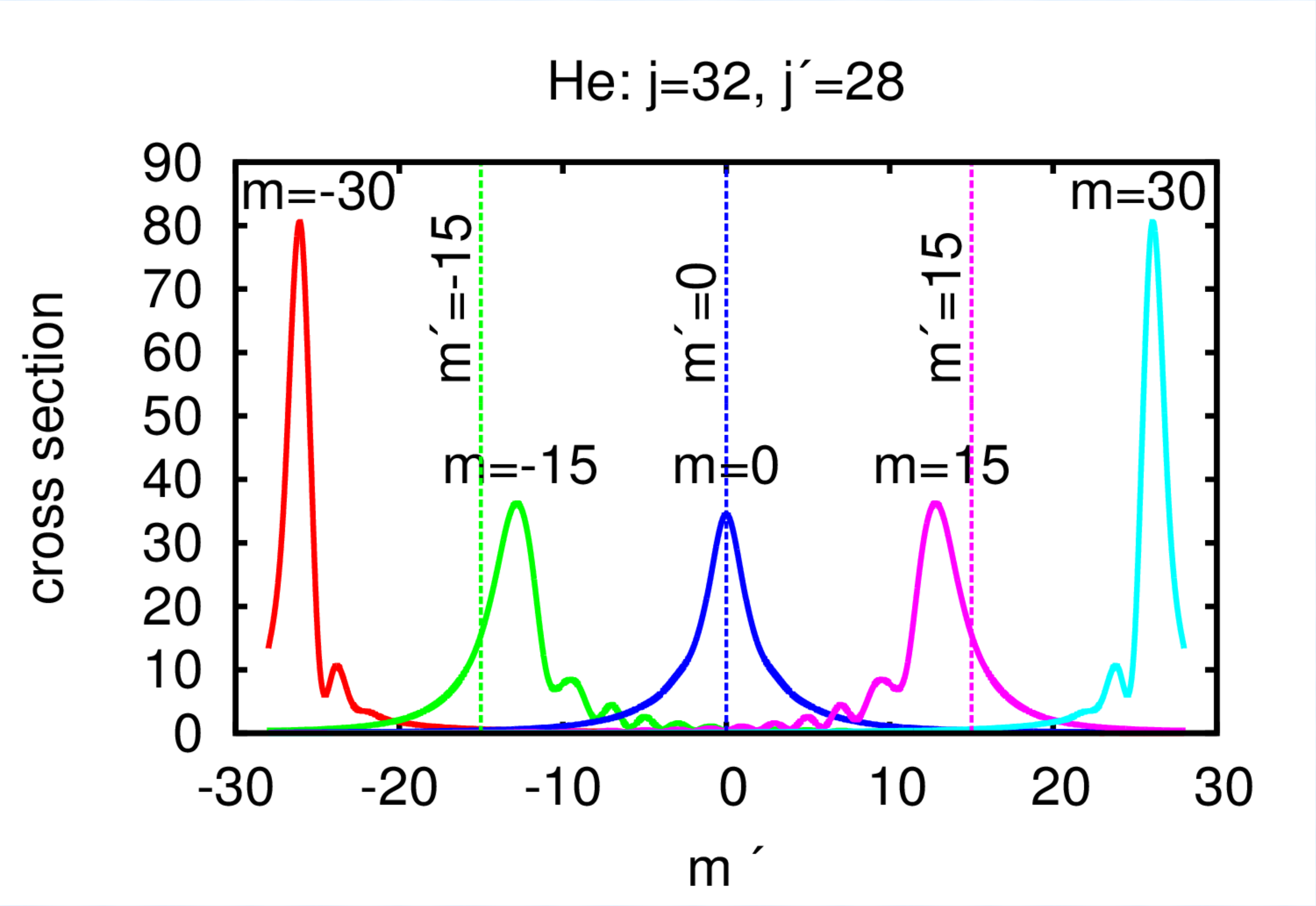


Ar + NaK



Distribution of Δθ averaged over all collisions

Quantum m → m'



He: j=32, j´=28
cross section
m´
m=-30
m=30
m´=-15
m´=0
m´=15
m=-15
m=0
m=15
90
80
70
60
50
40
30
20
10
0
-30
-20
-10
0
10
20
30

Conclusions

- $\Delta\theta$ tends to be small
- m tends to change
- Physical interpretations
  - Vector model
  - Semiclassical model

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- Citation
  - Malenda, R. F., T. J. Price, J. Stevens, S. L. Uppalapati, A. Fragale, P. M. Weiser, A. Kuczala, D. Talbi, and A. P. Hickman. "Theoretical Calculations of Rotationally Inelastic Collisions of He with NaK($A^1\Sigma^+$): Transfer of Population, Orientation, and Alignment." J. Chem. Phys. **142**, 224301 (2015).