Scattering of identical molecules: Imaging CO+CO rotational energy transfer

28-11-2018

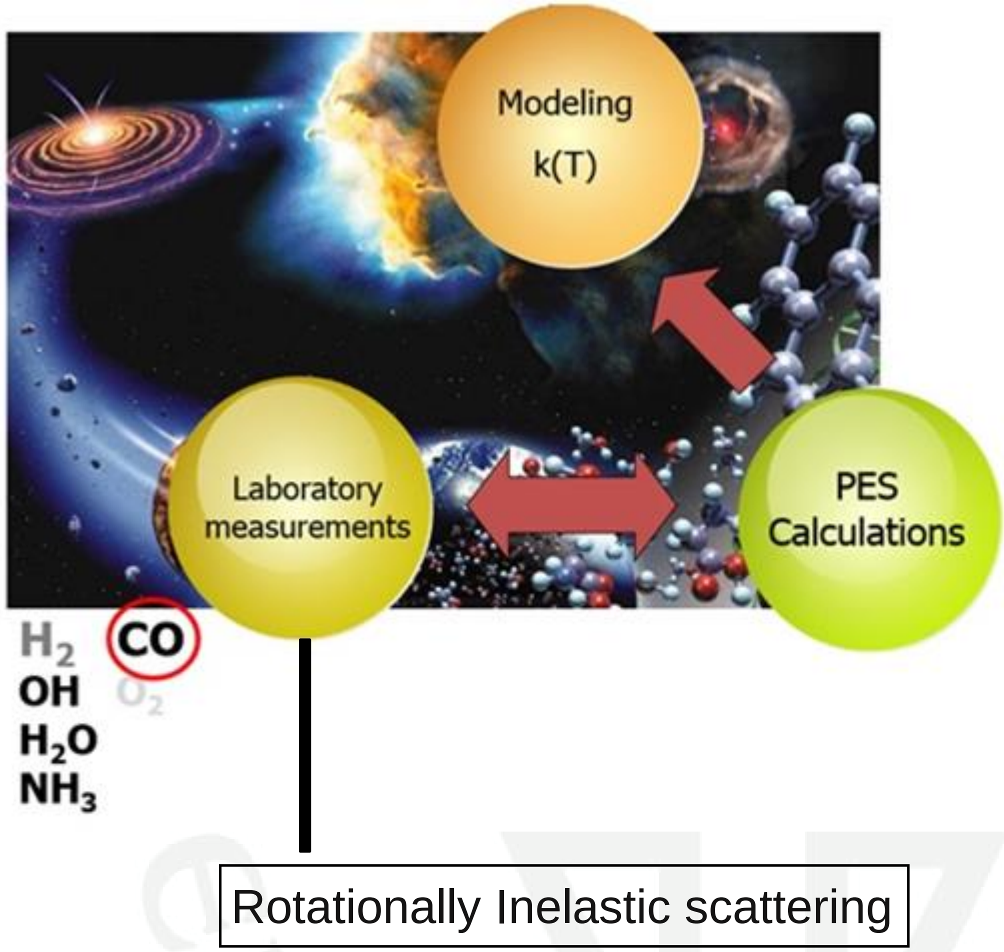
Molecular Laser Physics Department,
Radboud University

Zhongfa Sun

Why inelastic scattering involving CO?

CO is very important molecule

- Second most abundant molecule in the universe
- Tracer for all molecules in interstellar medium



CO scattering is important

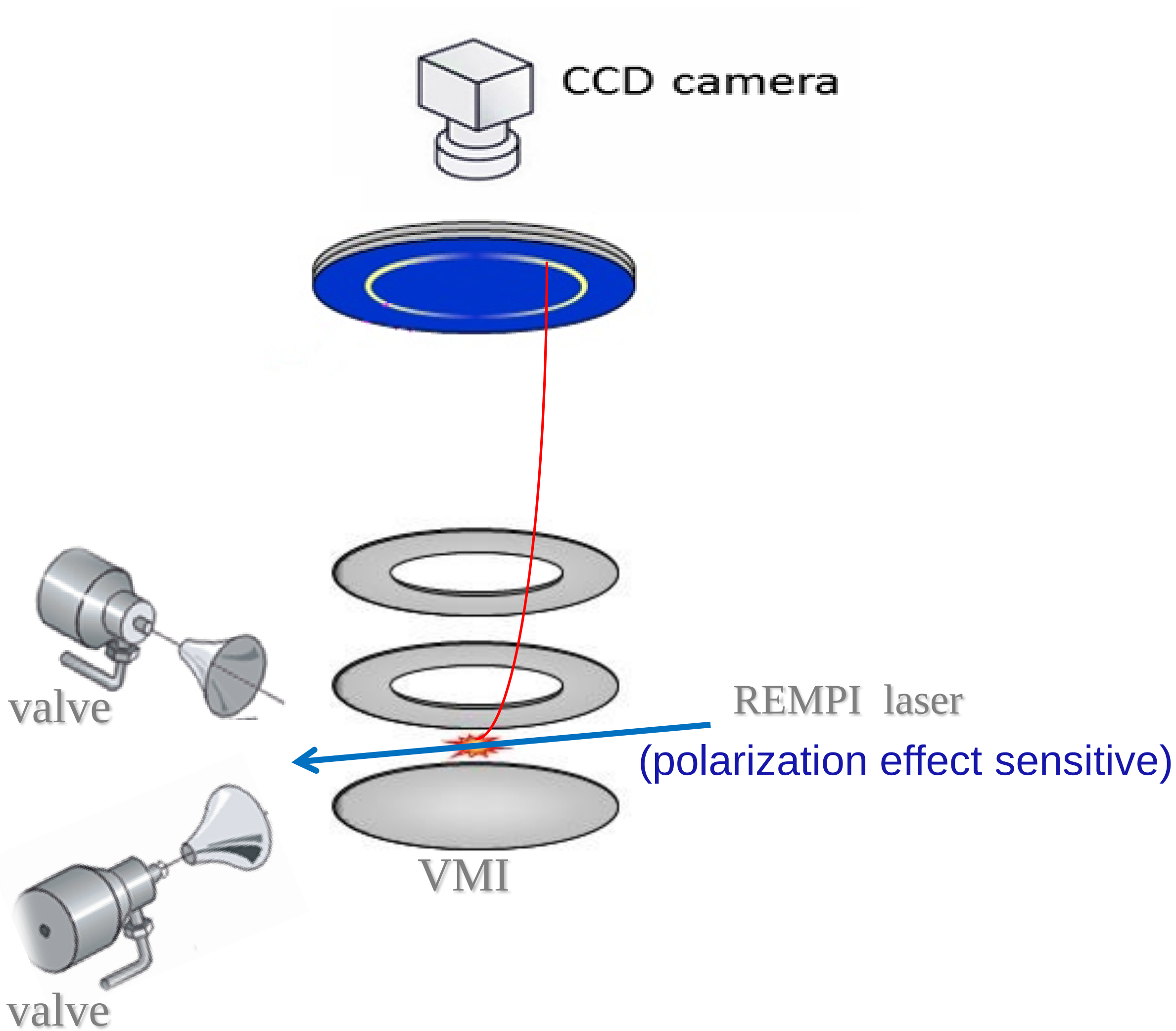
- Test PES
- To interpret telescope data

From **CO+Rg atom** (well understood) **to** **CO+molecule** (virtually uncharted territory)

(CO+CO)

Exp challenges & Solutions

General experimental methods to study "Rotationally Inelastic Scattering"

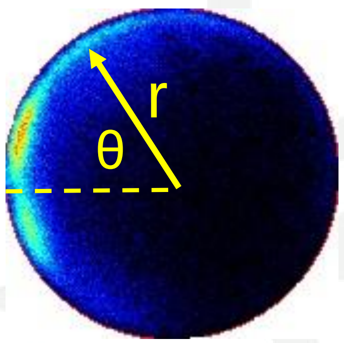


Crossed-molecular beam setup

REMPI : state selectivity

VMI : directly measures **Newton Spheres**

Ion Image



Radius I(r) (r – v - KE) → Rotational enegy transfer channel information

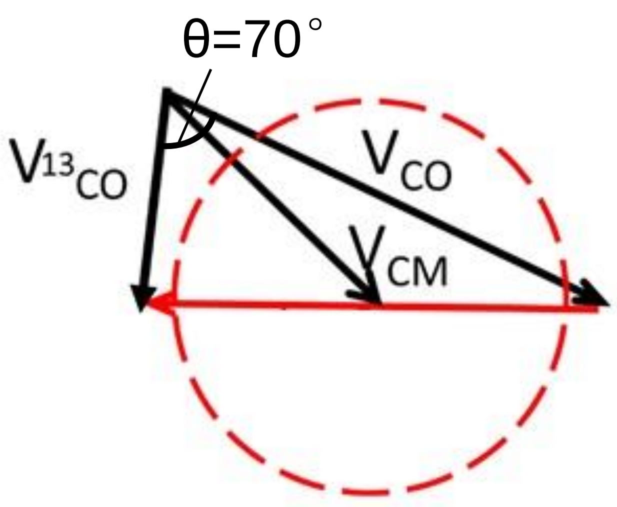
Angular distributions I(θ) (PD) DCSs → stringent probe for PESs

Bonus: Collision Induced Polarization info

Exp challenges & Solutions

However imaging scattering of identical molecules is extremely challenging!

Challenges for CO+CO experiment:		Possible solutions:
• Preparation of high initial state purity	⟵	Supersonic expansion with carrier gas
• Image overlap problem among accompanied processes(kill signal)	⟵	Adequate kinematic expt conditions to avoid overlap
• Indistinguishability of identical molecules	⟵	Isotope substitution i.e. ^{13}CO detected
• Pair correlation is difficult to resolve in imaging	⟵	Detecting moderate and high-J scattered products



Final experimental condition settings

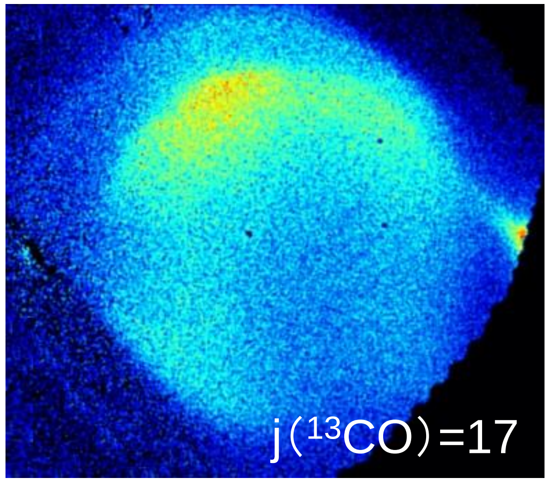
Primary beam: 5% ^{13}CO/Ar ($v_1 \approx 650$m/s)

Secondary beam: 10% CO/He ($v_2 \approx 1650$m/s)

θ_{col}: 70° (minimal for current setup)

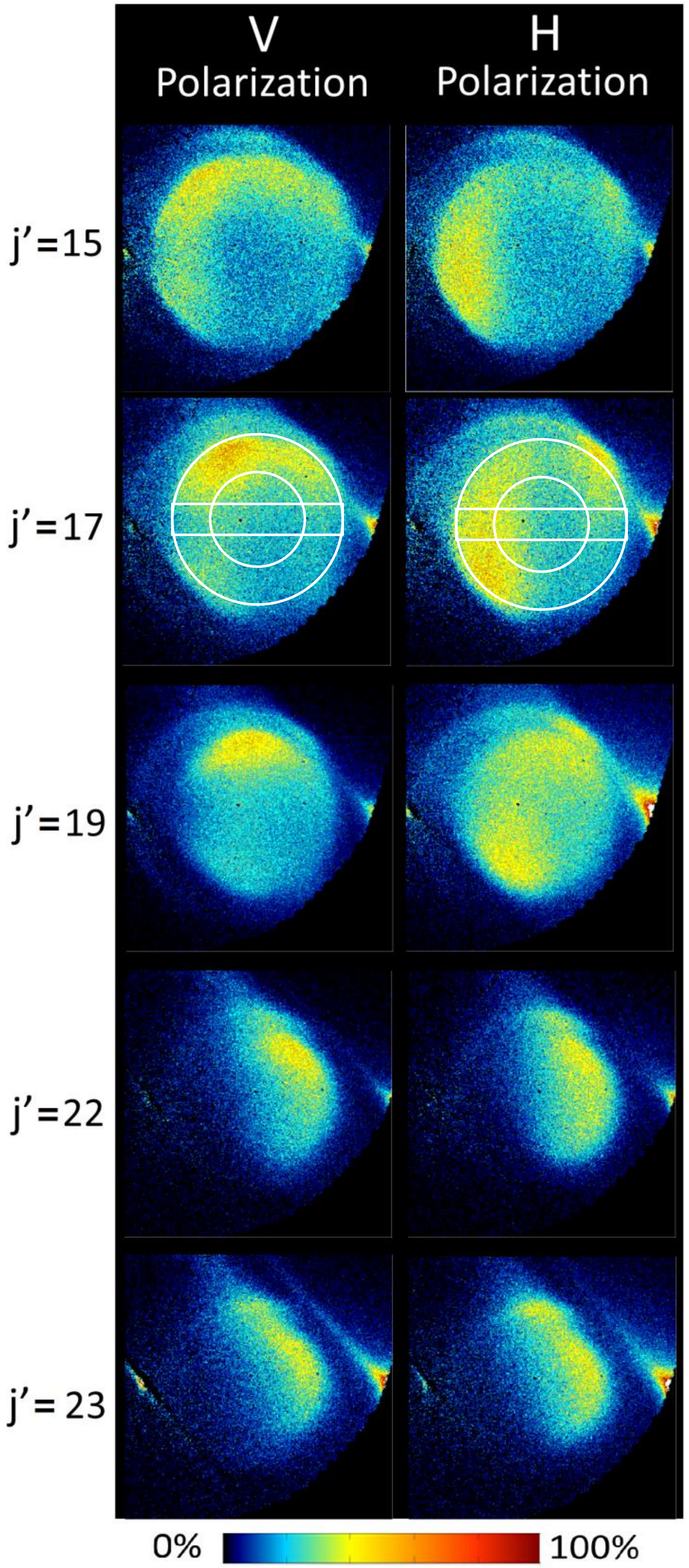
Detection: 1+1' VUV REMPI with H / V polarization

→ Polarization-dependent images



Pure ^{13}CO+CO were obtained !

Results of ^{13}CO+CO collisions



extraction

DCSs

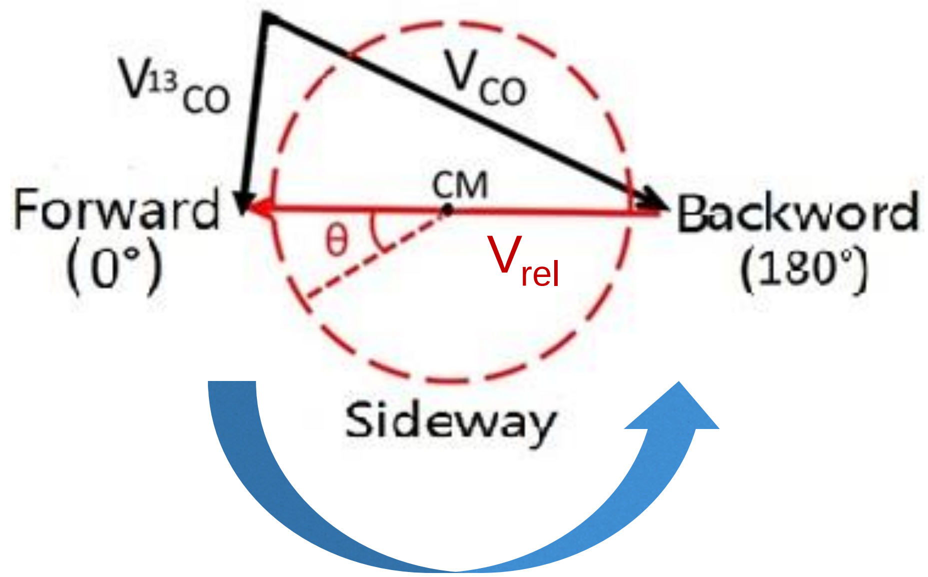
&

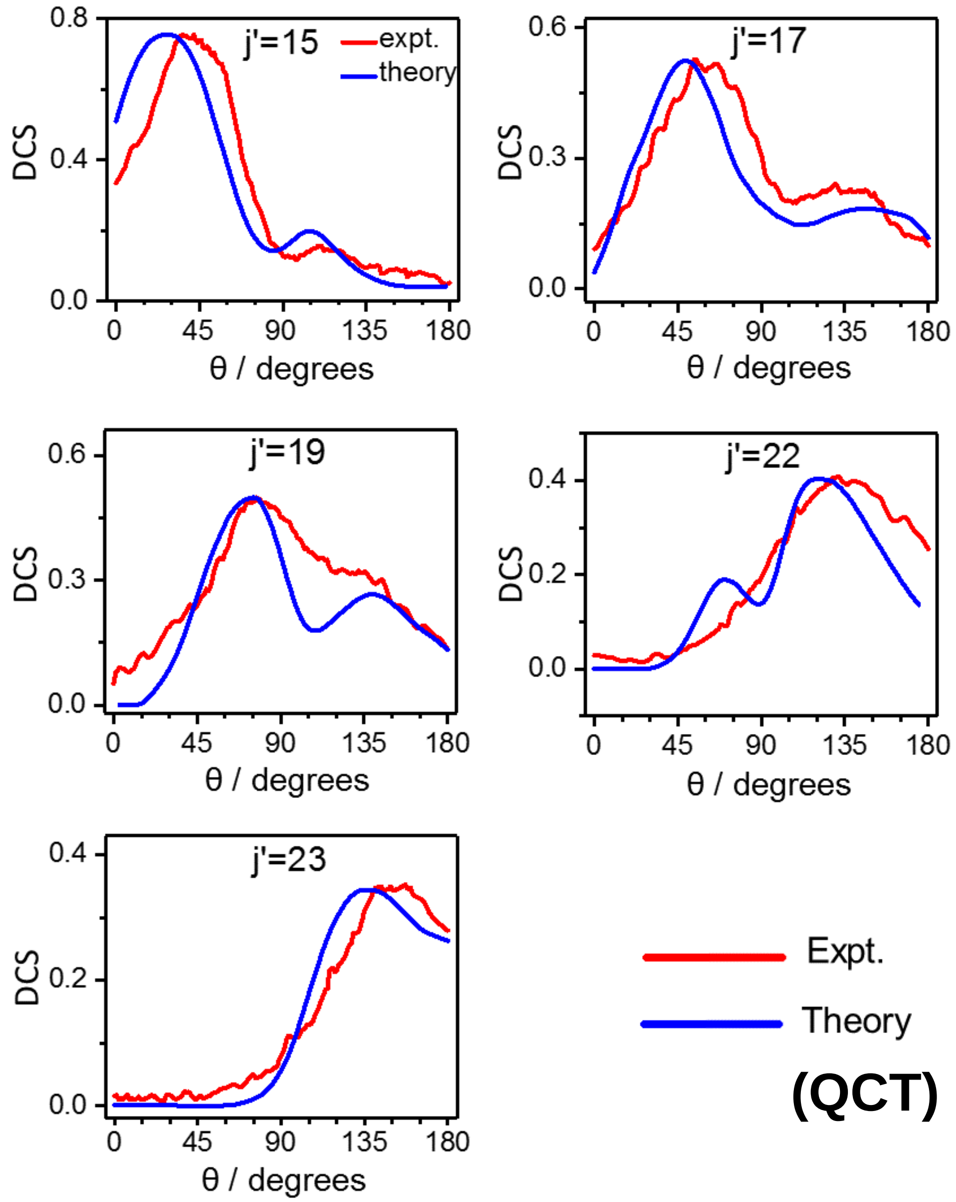
Alignment moments

(describe j' spatial polarization in collision frame)

Alignment moments	Directional meaning	**limits**
$A_0^{\{2\}}$	alignment of j' about k (k-j' correlation)	[-1,2]
$A_{1+}^{\{2\}}$	alignment of j w.r.t X±Z	[-1,1]
$A_{2+}^{\{2\}}$	alignment of j along X or Y axis	[-1,1]

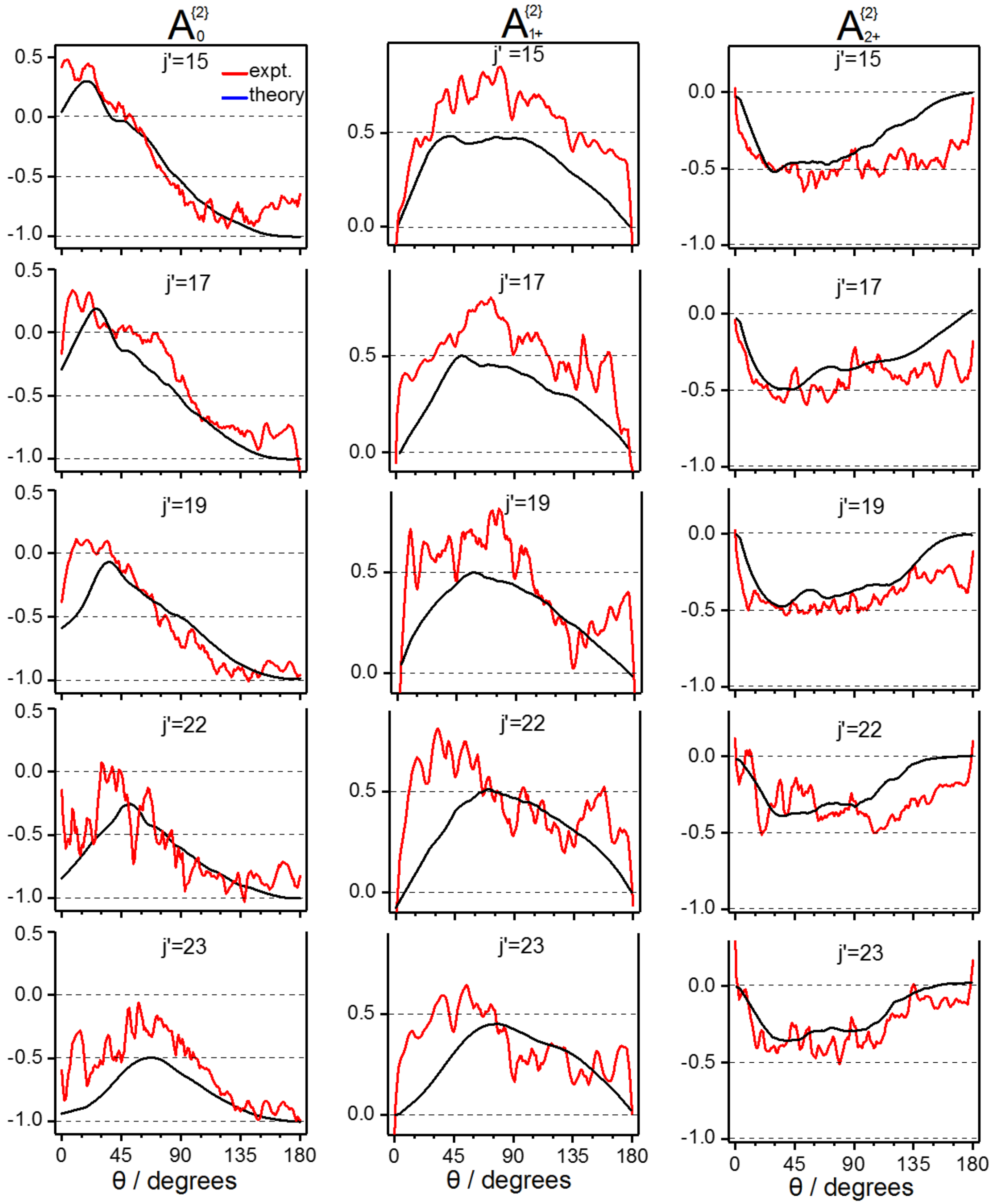
Results of ^{13}CO+CO collisions





Experimental DCSs in good agreement with theory

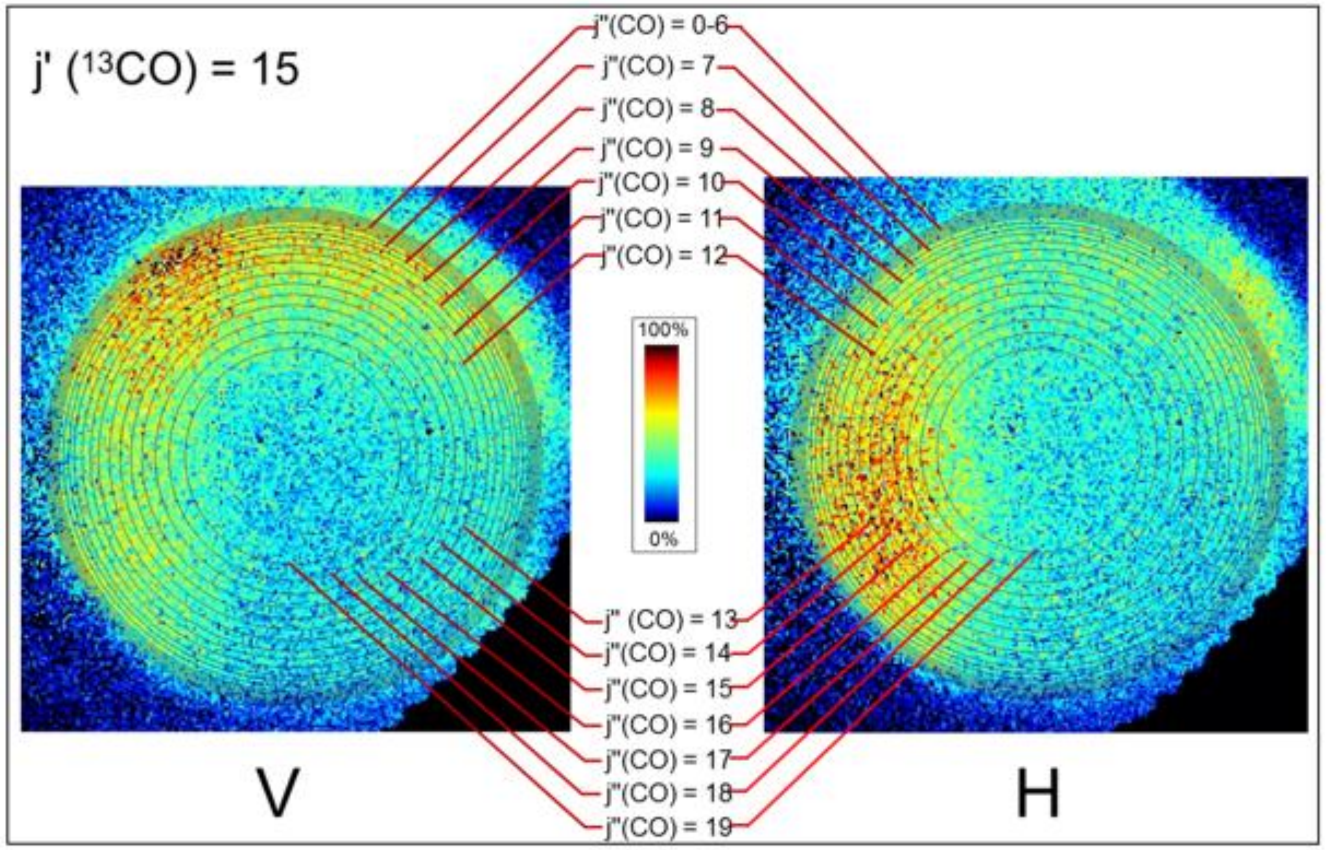
Results of ^{13}CO+CO collisions

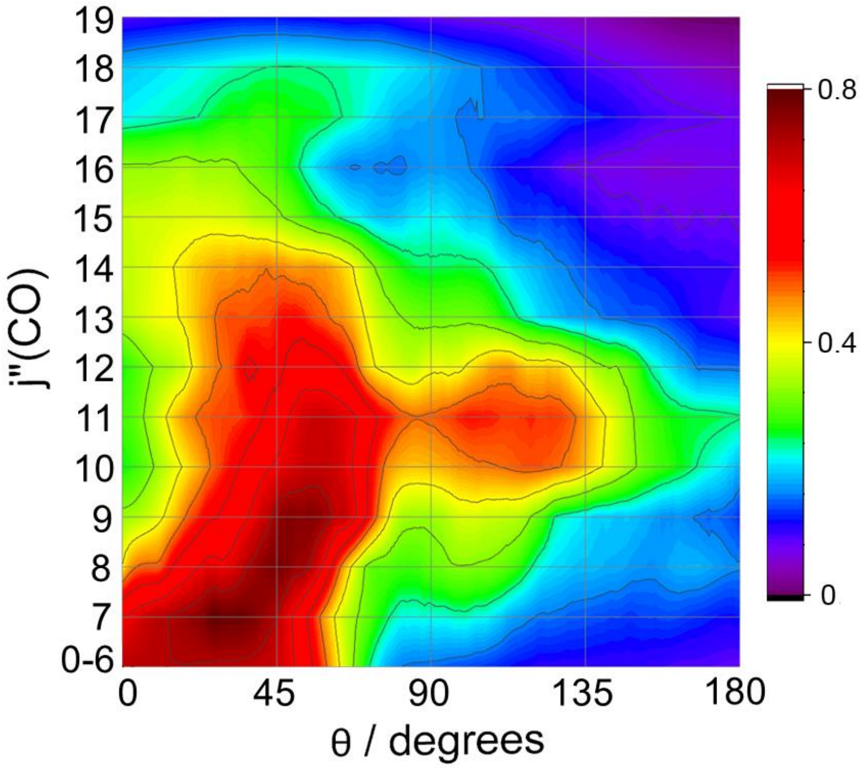


Experimental Alignment moments agree reasonably with theory; the reliability of PESs employed again confirmed.

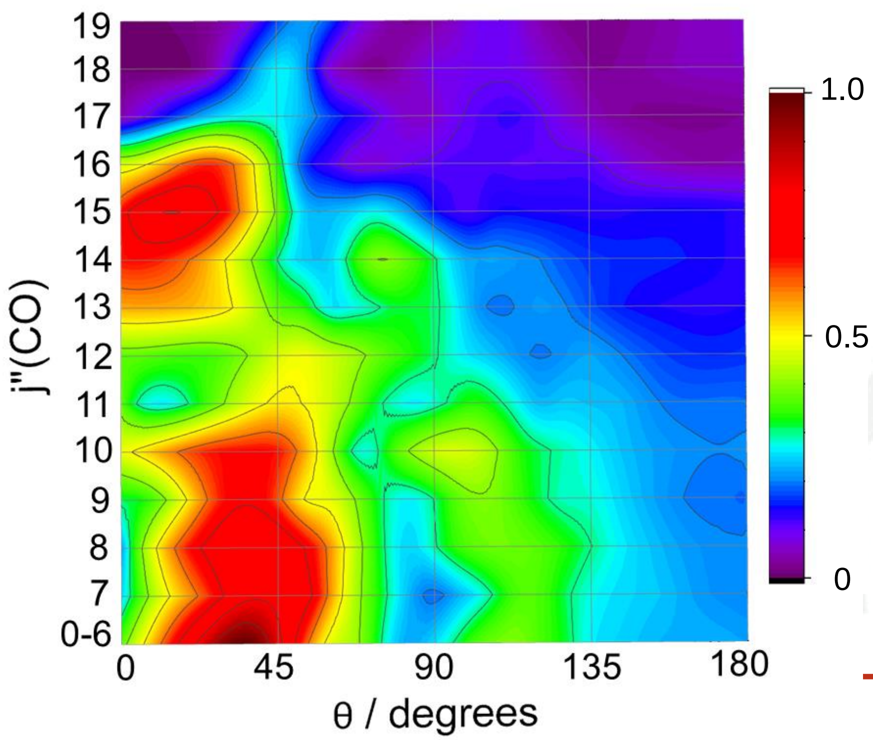
Results of $^{13}CO+CO$ collisions

Pair correlated DCSs comparison





Expt



Theory

Conclusion and outlook

For the first time, state-to-state PDDCSs for CO collisions with CO were measured experimentally, and compared with theory;

Good agreement between experiment and theory predictions implies that the features of PESs are quantitatively correct within the E_{coll} range sampled;

New insights into energy partitioning, propensity rules, and collision induced alignment effects are revealed;

Offering a general and reliable approach to study bimolecular inelastic scattering, can be applied to other systems.

Acknowledgements

Theory

- Marc van Hemert (Universiteit Leiden)
- Ad van der Avoird

Experiment

- Dave Parker
- George McBane
- Arthur Siuts
- Roy Scheidsbach
- Agniva Banerjee
- Evaggelia Sakkoula

- Bas van de Meerakker & SCM group

Technical support:

- Chris Berkhout
- Andre van Roij
- Edwin Sweers
- Niek Janssen
- Paul Assman

Administration

- Magda Speijers

Thank you for your attention!

Radboud Universiteit Nijmegen

Netherlands Organisation for Scientific Research

1[st] Beam
CO/carrier gas I

+

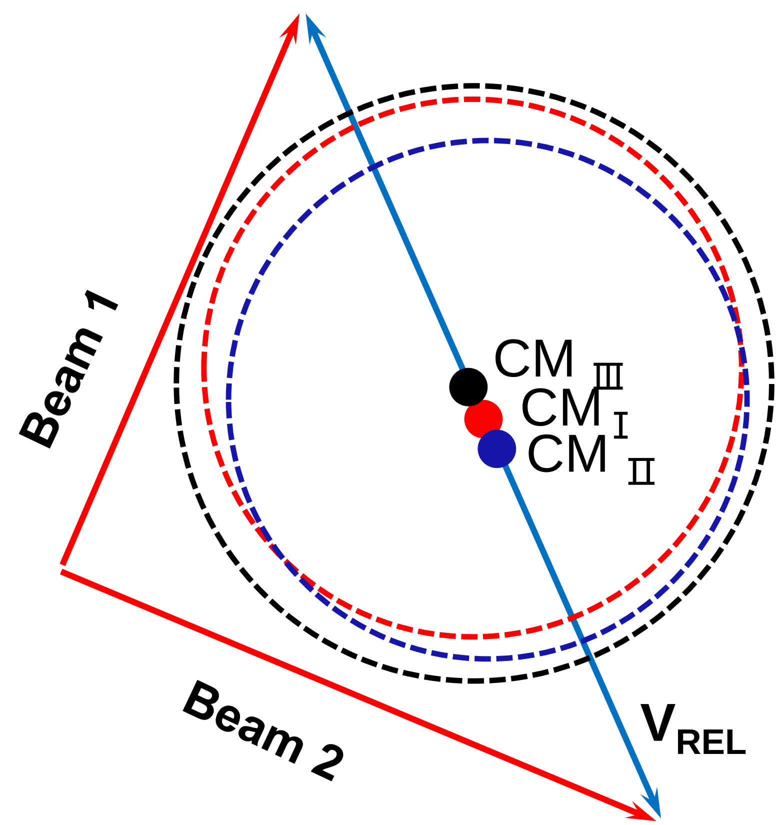
2[nd] Beam
CO/carrier gas II

=

CO+CO — system 1

CO + carrier gas atom I — system 2

CO + carrier gas atom II — system 3

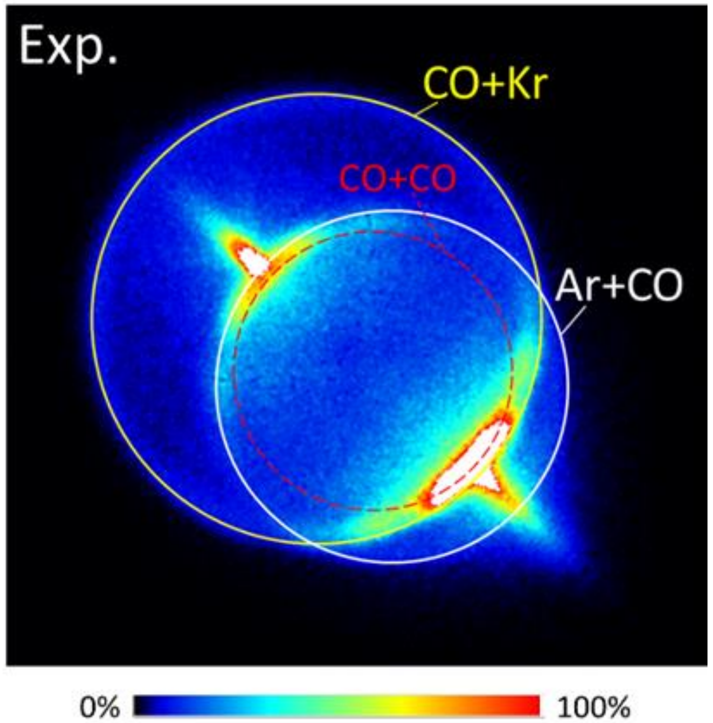


Overlap problem can kill the desired CO+CO signal !!

Adequate experimental conditions seeking

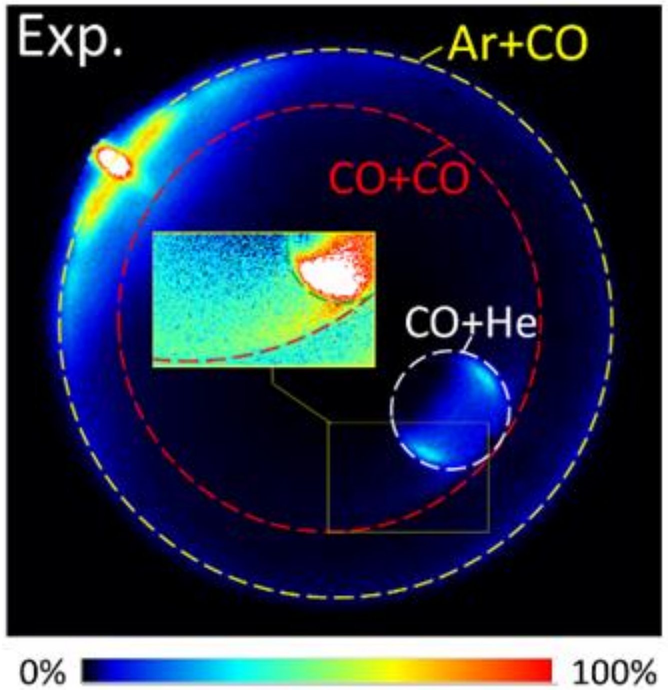
1st beam: 5%CO/Ar ($V_1 \approx$ 650m/s)

2nd beam:10%CO/Kr ($V_2 \approx$ 420m/s)



1st beam: 5% CO/Ar ($V_1 \approx$ 650m/s)

2nd beam: 10%CO/He ($V_2 \approx$1650m/s)



✕ √

Large relative velocity, mass difference

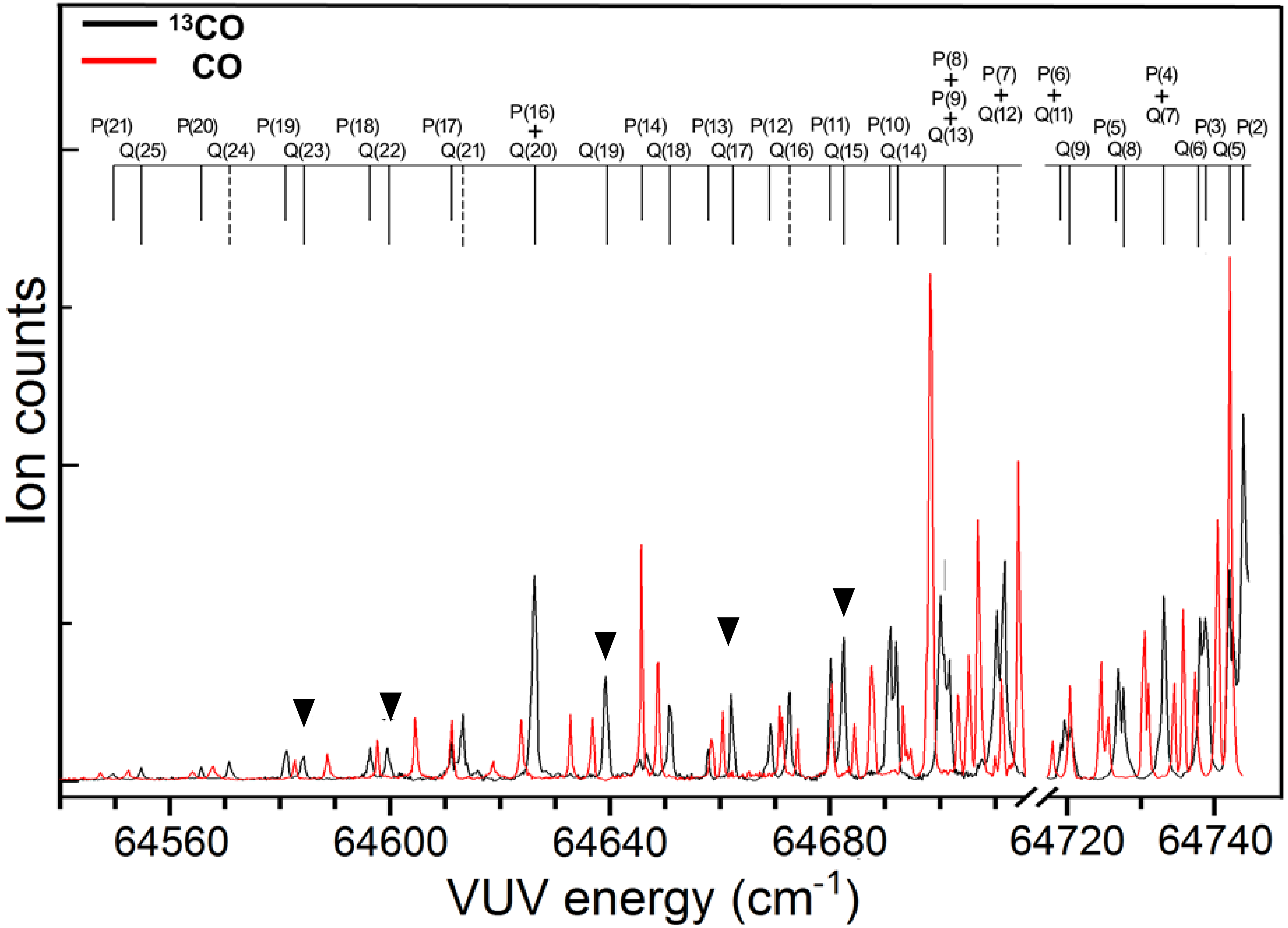
↓

overlap problem solved

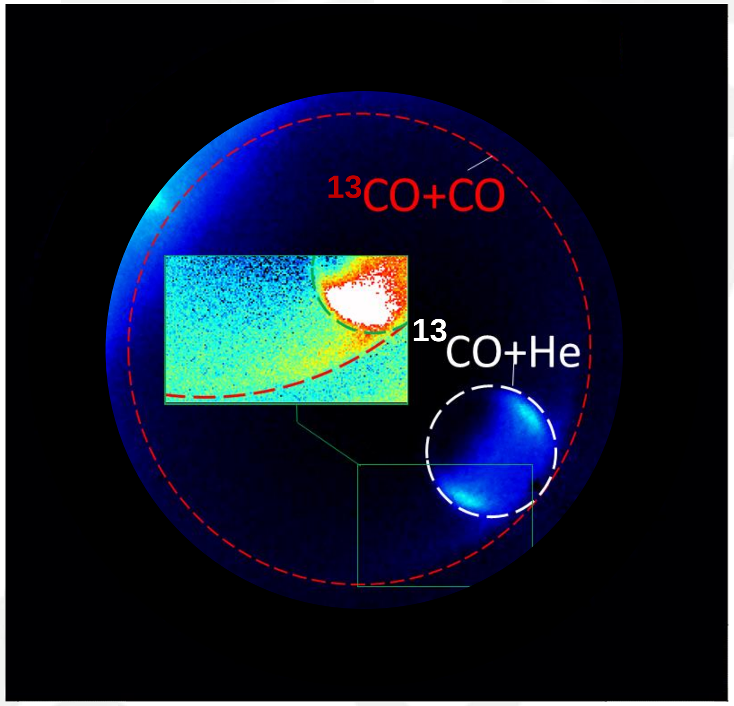
adequate!

Isotope substitution foundation

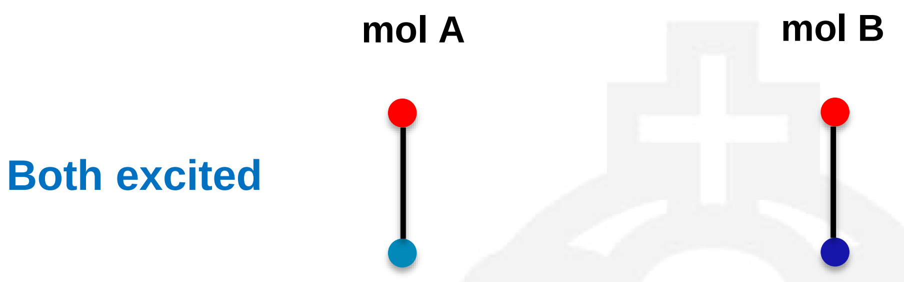
^{13}CO REMPI vs. CO REMPI



Isotope substitution (^{13}CO) for elimination



For bimolecular collisions:



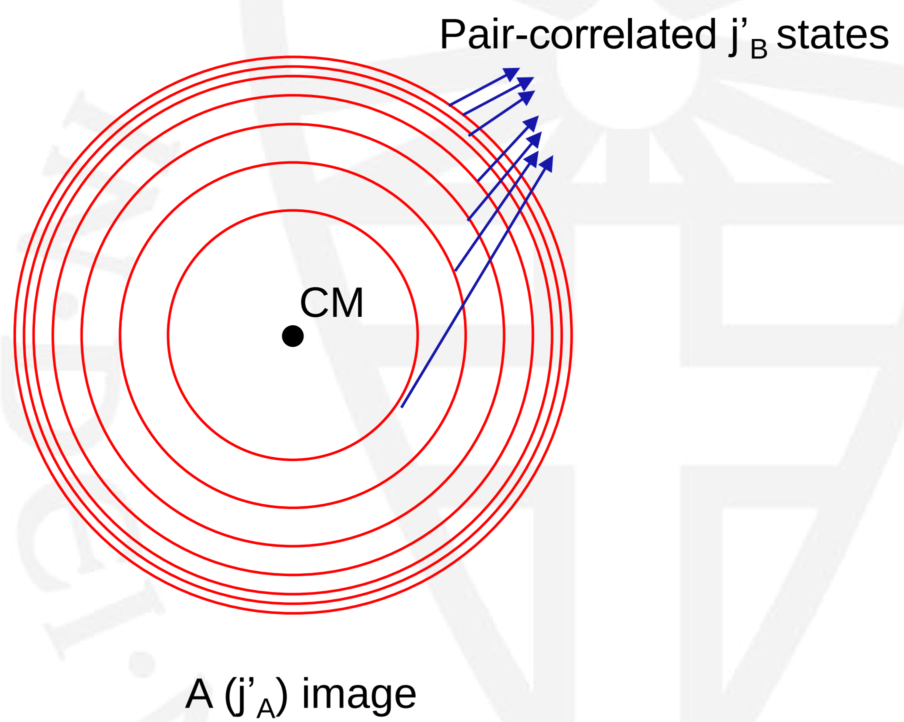
$$r = \frac{1}{f} * \frac{m_B}{m_A + m_B} \sqrt{\frac{2\left(E_{coll} - \Delta E_{rot}(A) - \Delta E_{rot}(B)\right)}{\mu}}$$

where

f : pixel-to-velocity factor

$\Delta E_{rot}(A)$: specified by REMPI

$\Delta E_{rot}(B)$ = $E_{rot}(B)' - E_{rot}(B) = B_B j'_B(j'_B+1) - B_B j_B(j_B+1)$



Detecting moderate and high-j' scattered products