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Spin-spin and spin-rotational fine structure of the metastable $a\ ^3\Sigma_u^+$ states of molecular helium

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Spin-spin and spin-rotational fine structure of the metastable $a\ ^3\Sigma_u^+$ states of molecular helium and He_2^+

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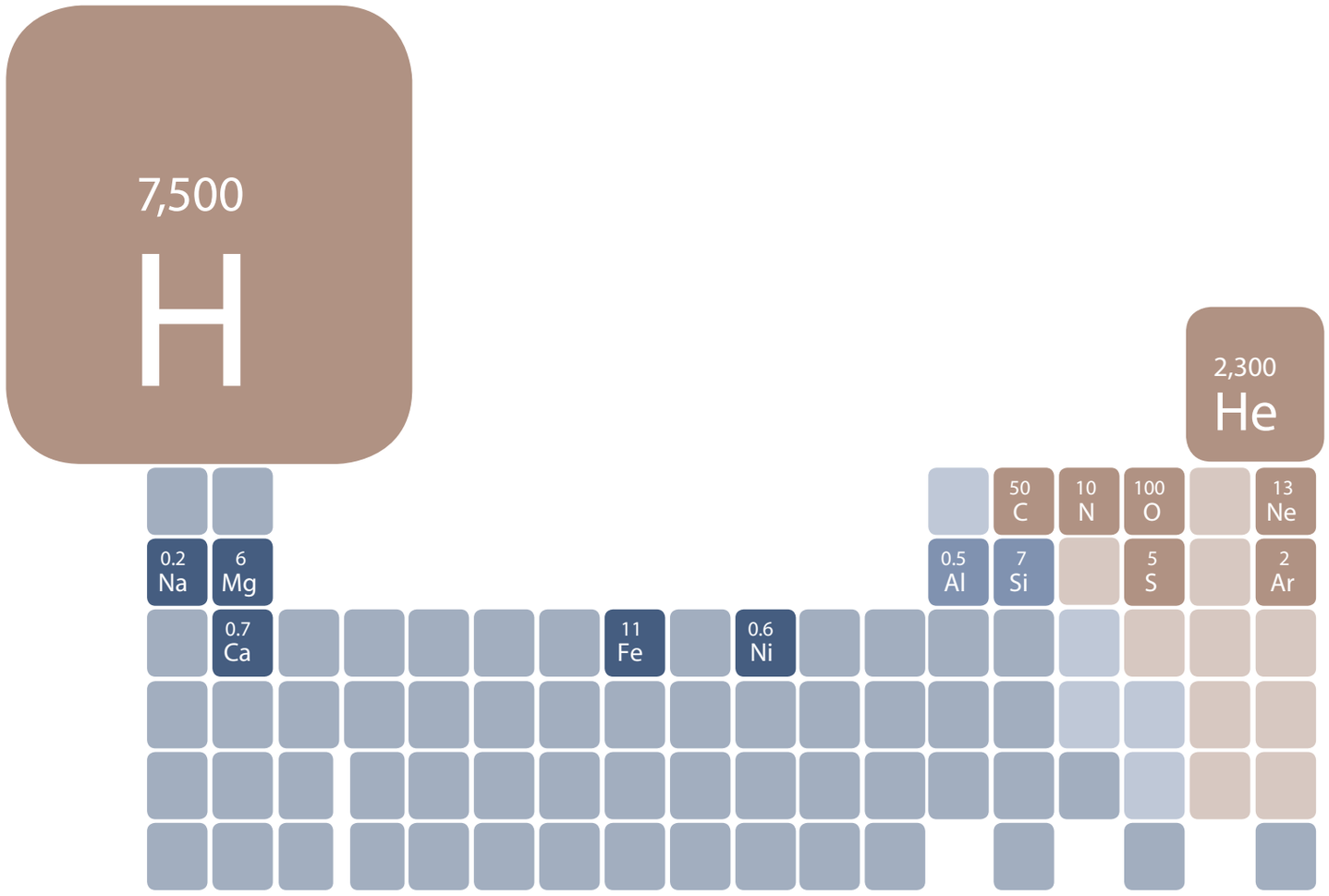
The periodic table for astronomy

- Helium was the first atom to become neutral after Big Bang.
- He_2^+ was among the first molecules to be formed [1].

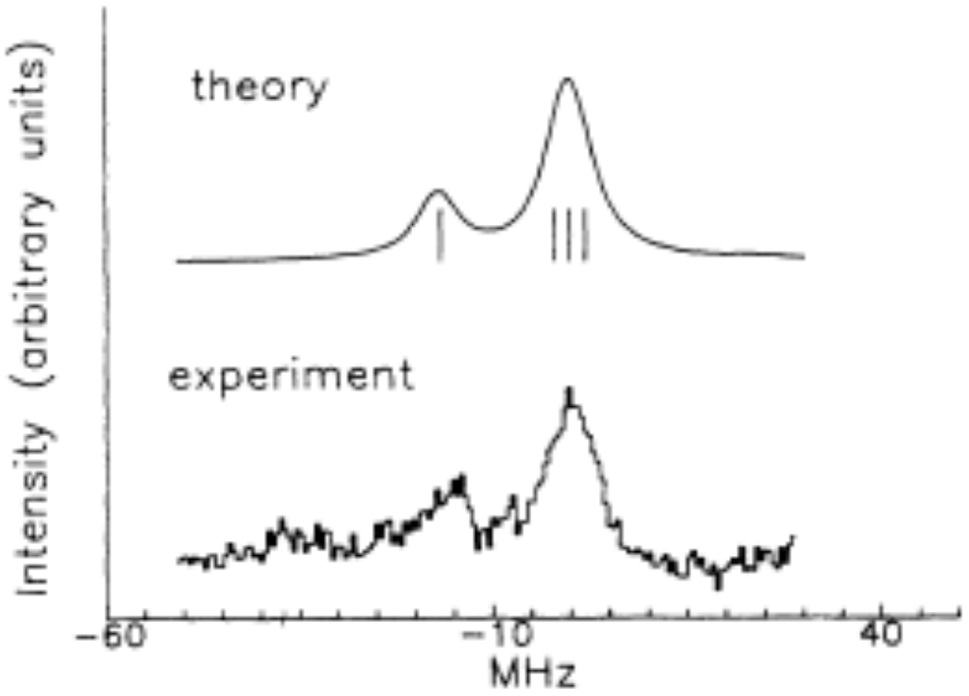


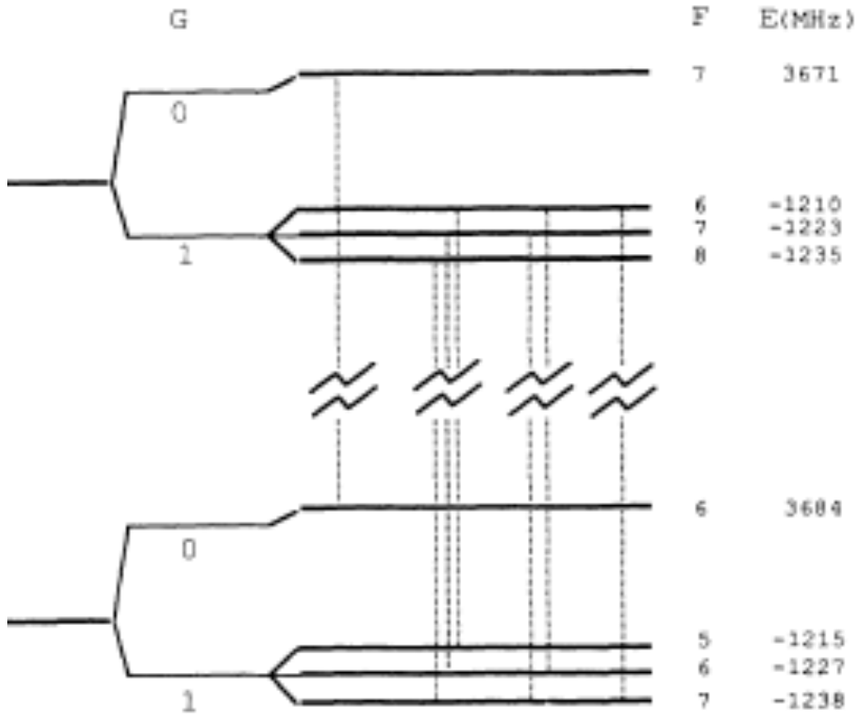
Location	$^3He/^4He$
Earth atmosphere	$\sim10^{-7}$
Moon	$\sim10^{-5}$
Sun	$\sim10^{-4}$
Local ISC	$\sim10^{-4}$
Stars	$\sim10^{-4}$ - 5

[1] Dalgarno, *J. Phys. Conf. Ser.* **4**, 10 (2005)

Previous studies of He_2^+

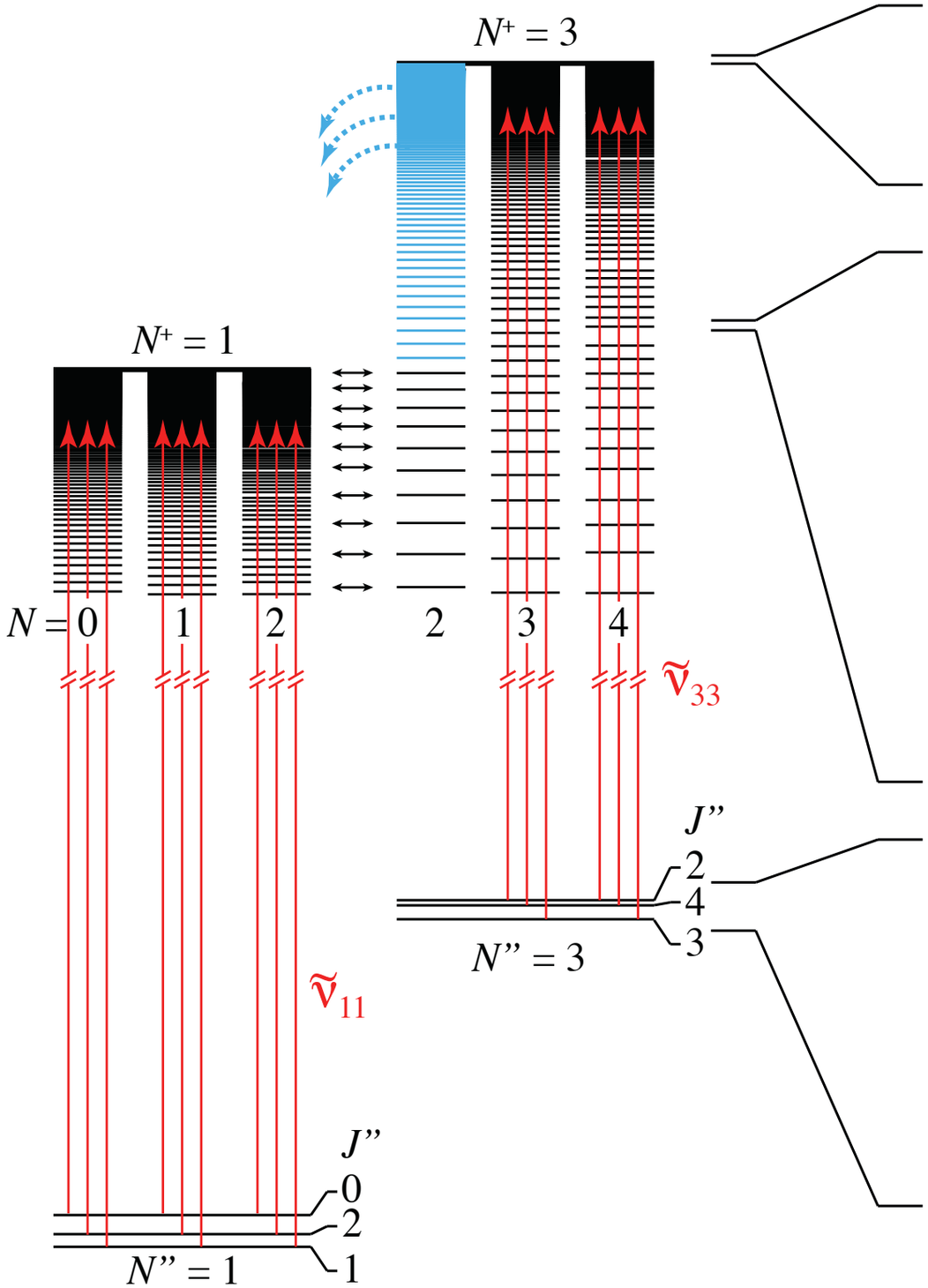
- He_2^+ has no electric dipole moment or easily accessible electronic states.
- Only a few transitions have been observed experimentally:
  - 7 rovibronic $X^+ \rightarrow A^+$ ($v^+ = 22, 23 \rightarrow 0,1$) in $^4He_2^+$ (uncertainty ~ 2 MHz) [1]
  - 9 robivrational ($v^+ = 0$) transitions in $^3He^4He^+$ (uncertainty ~ 18 MHz) [2]

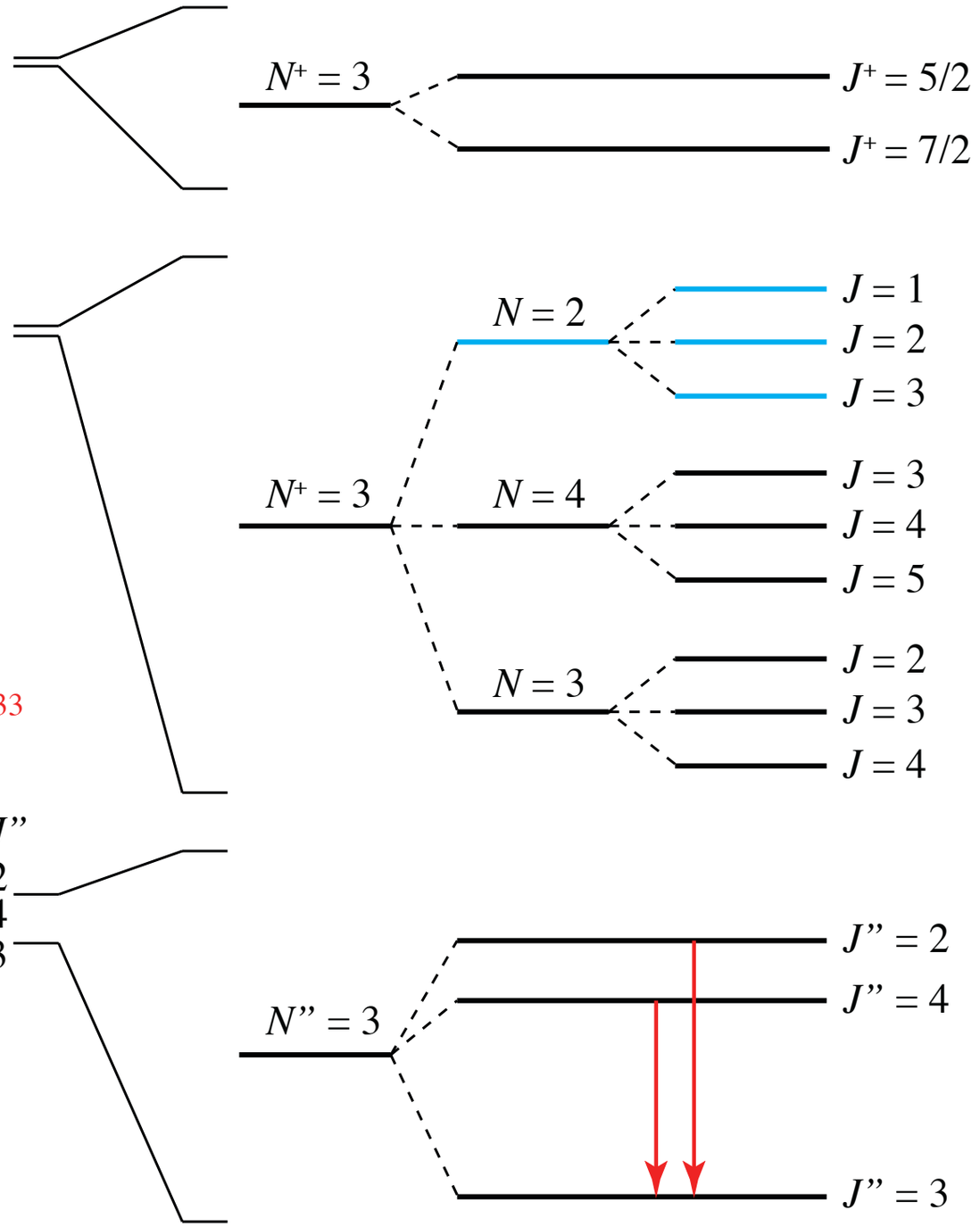




[1] Carrington *et al.*, *J. Chem. Phys.* **102**, 5979 (1995).
[2] Yu and Wing, *Phys. Rev. Lett.* **59**, 2055 (1987).

Rydberg states of He_2



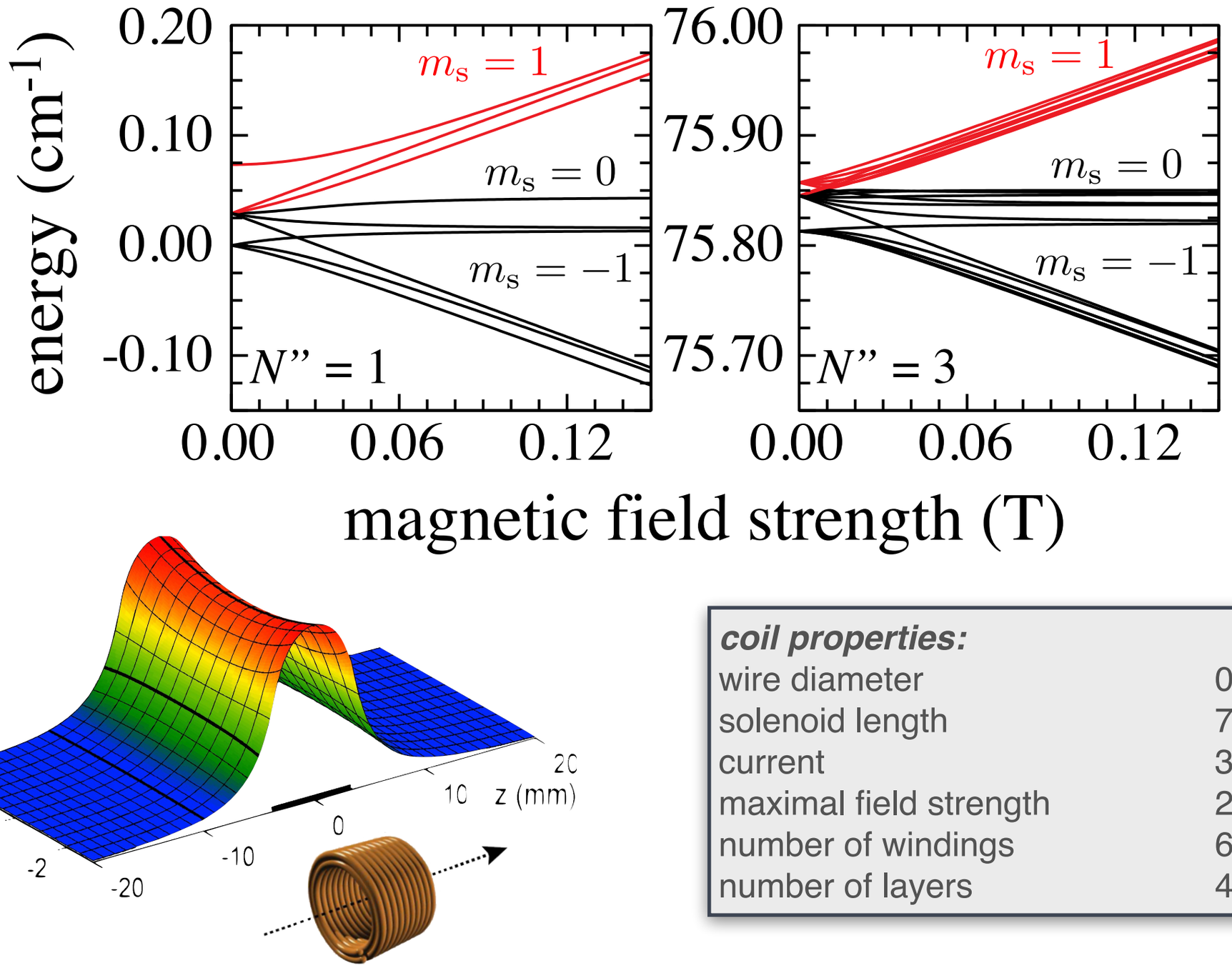


$X^+\,^2\Sigma_u^+$

np Rydberg states

$a\,^3\Sigma_u^+$

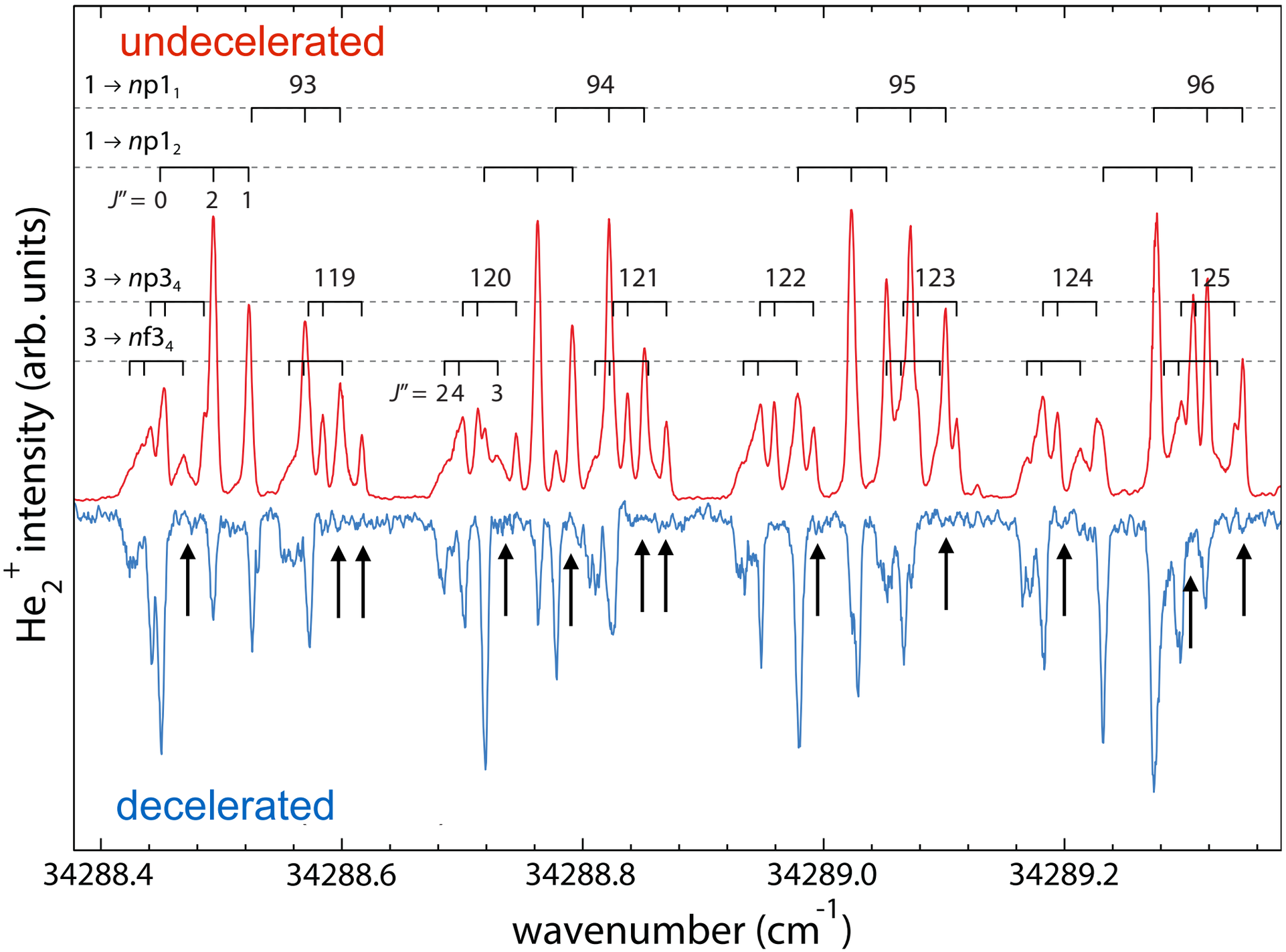
He_2^* in an inhomogeneous magnetic field



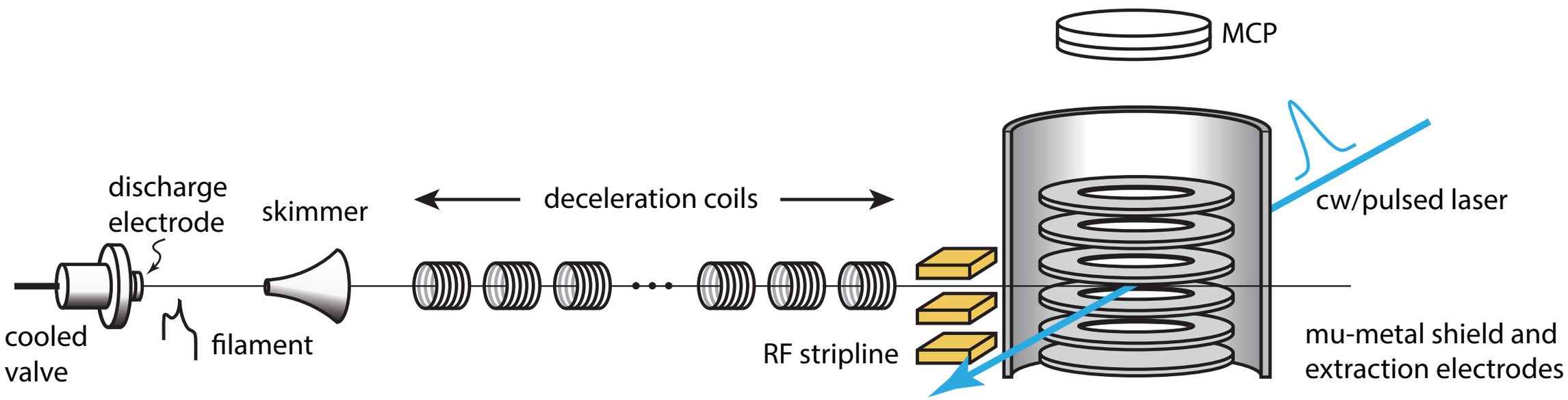
coil properties:	
wire diameter	0.4 mm
solenoid length	7.2 mm
current	300 A
maximal field strength	2.2 T
number of windings	62
number of layers	4

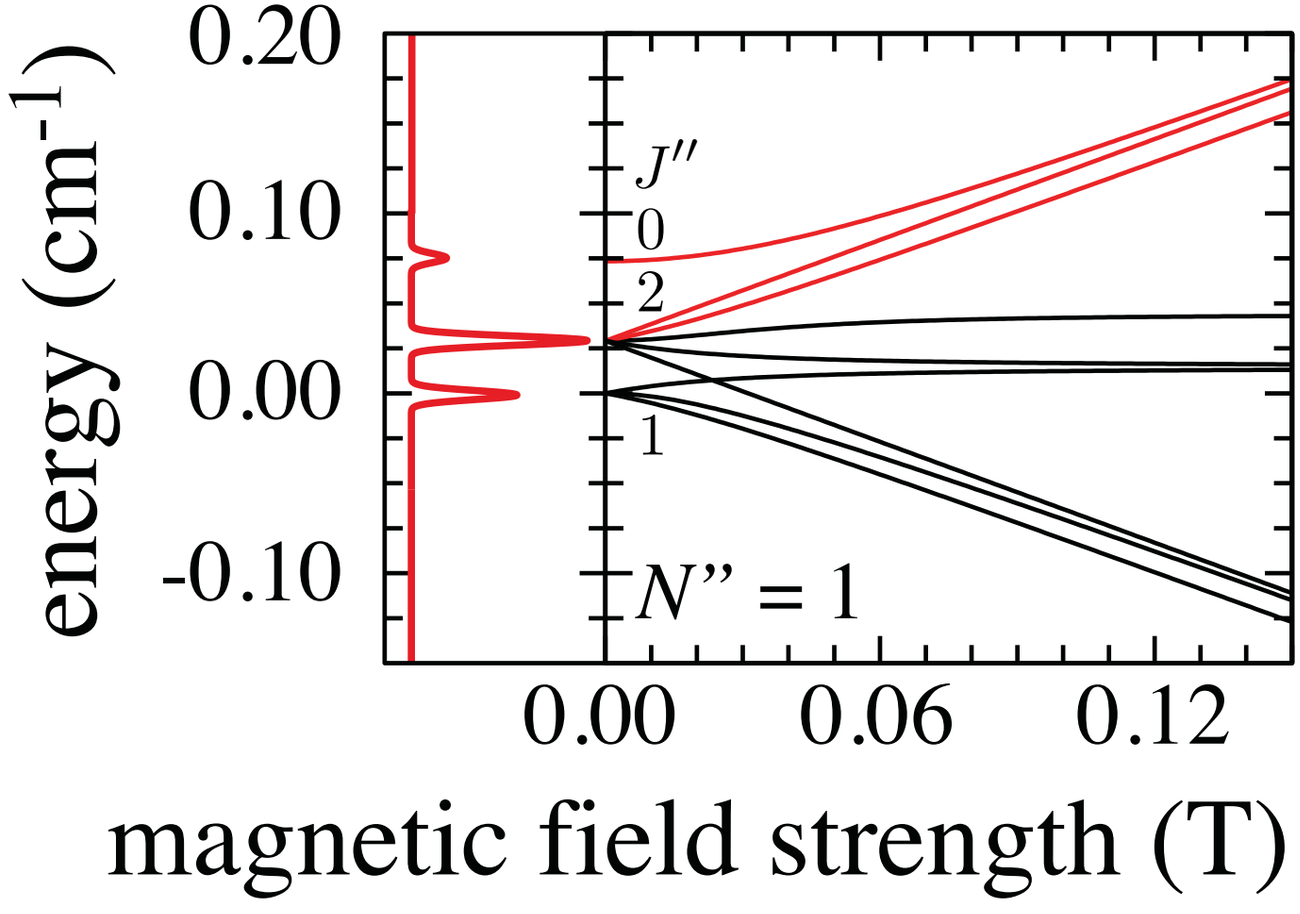
Vanhaecke *et al.*, *Phys. Rev. A* **75**, 031402(R) (2007).
Hogan *et al.*, *Phys. Chem. Chem. Phys.* **13**, 18705 (2011).

Spin-polarized molecular beams by Zeeman deceleration



A Zeeman-decelerated molecular-beam magnetic-resonance method





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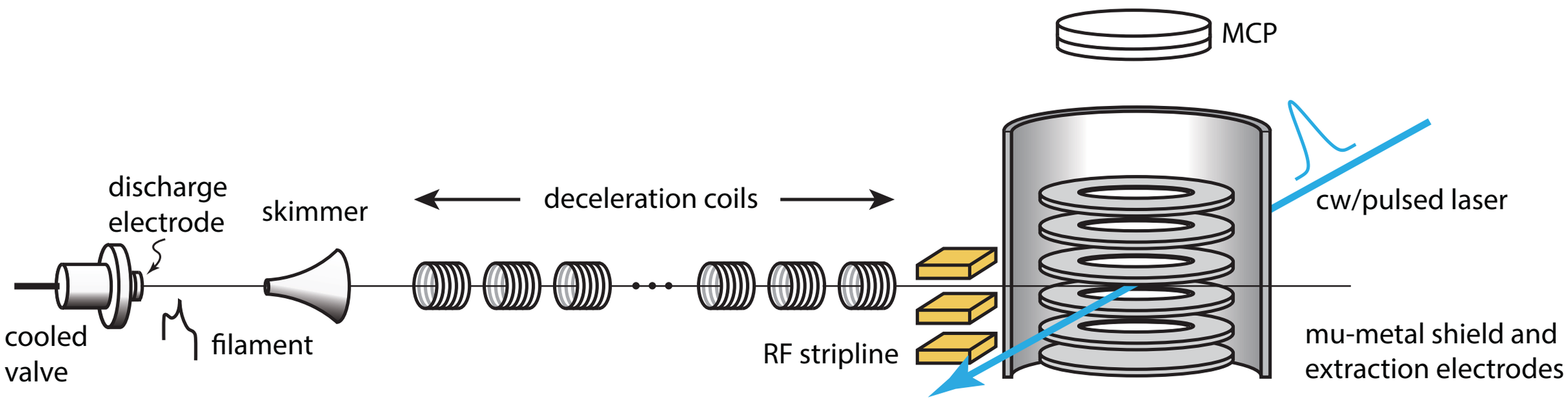

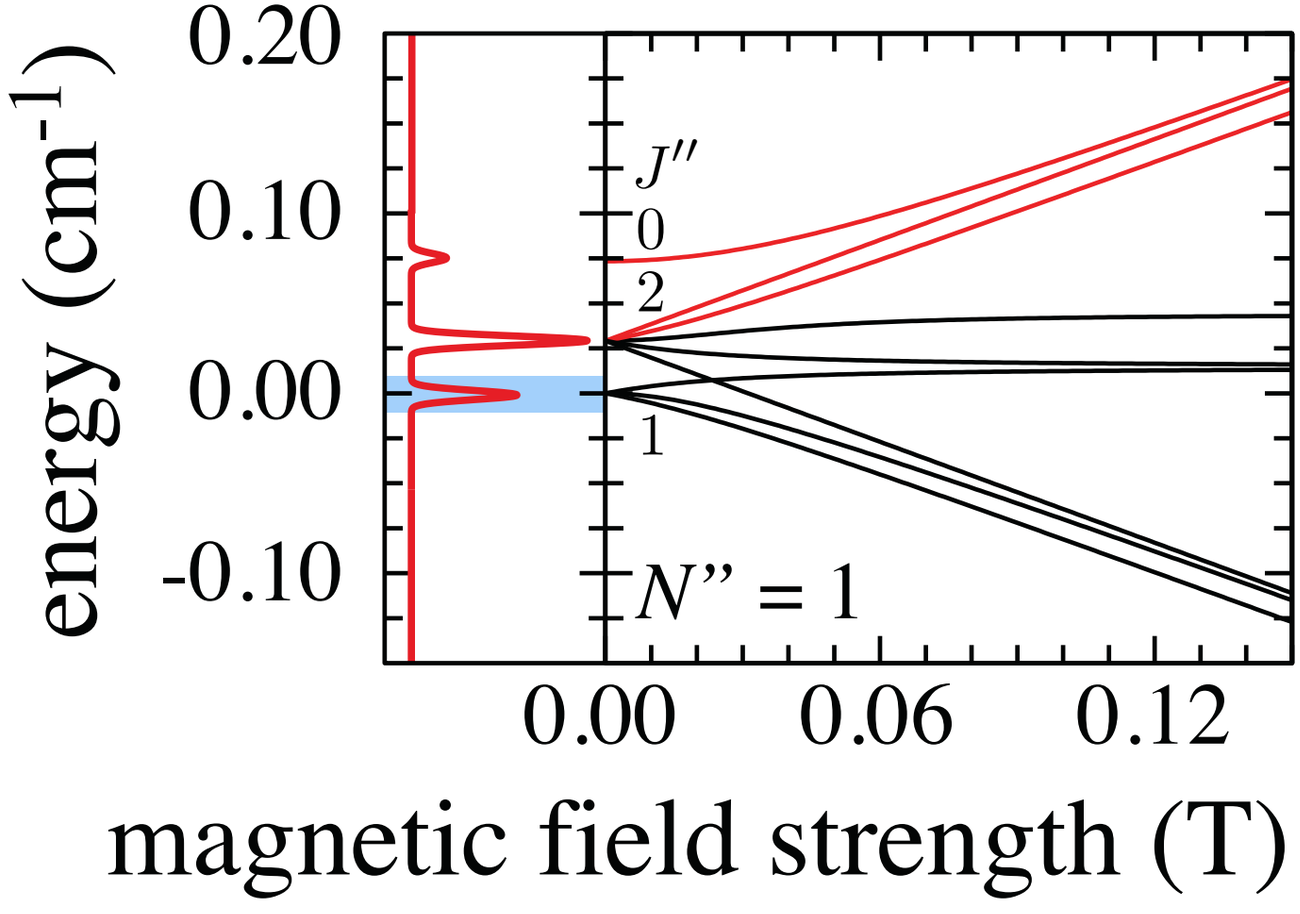

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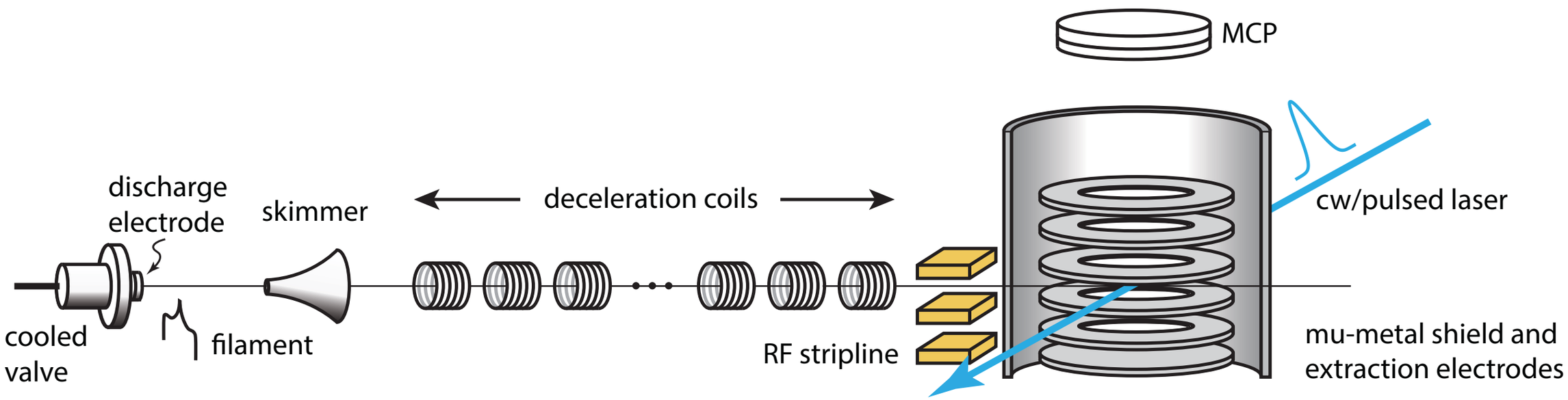

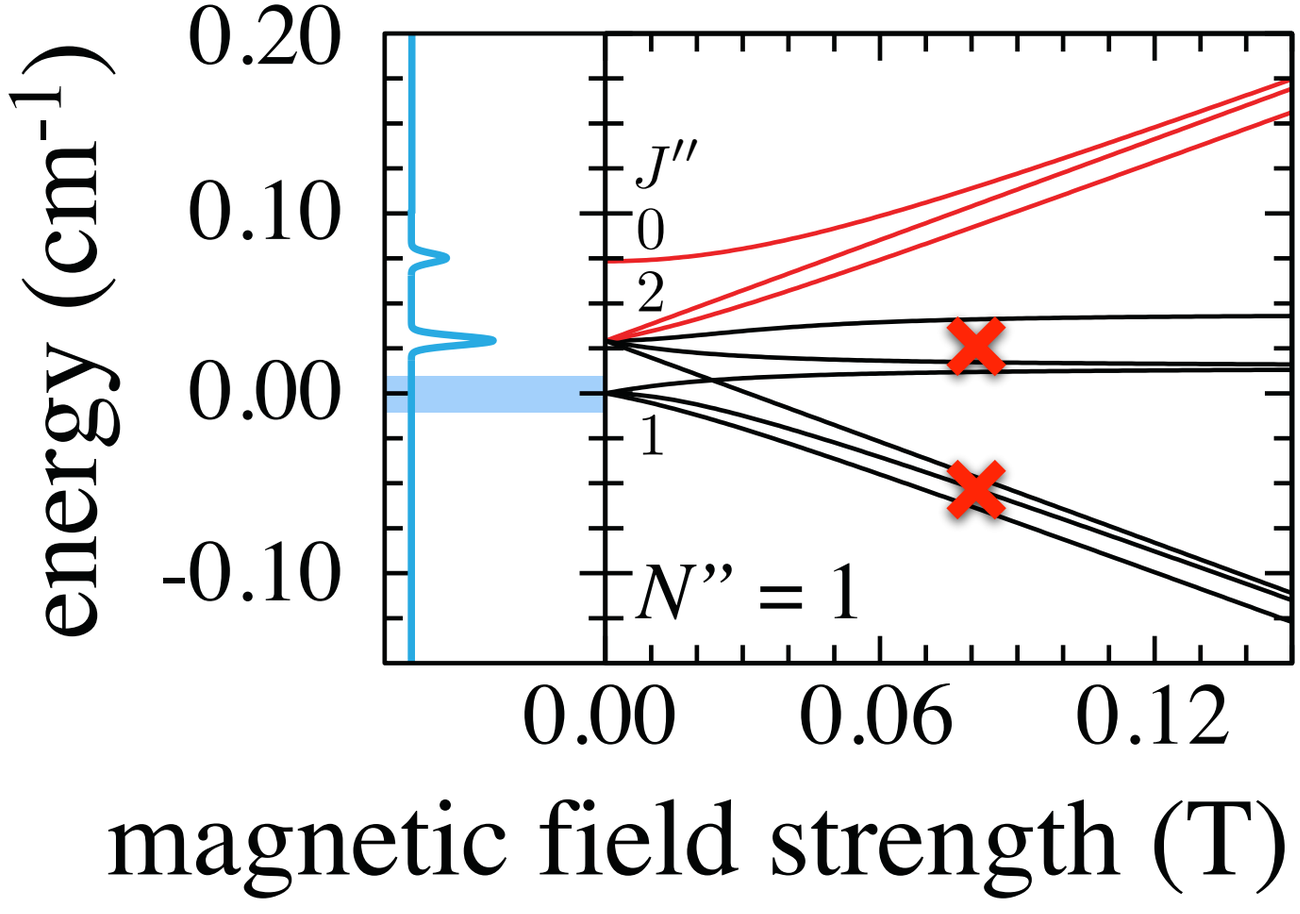

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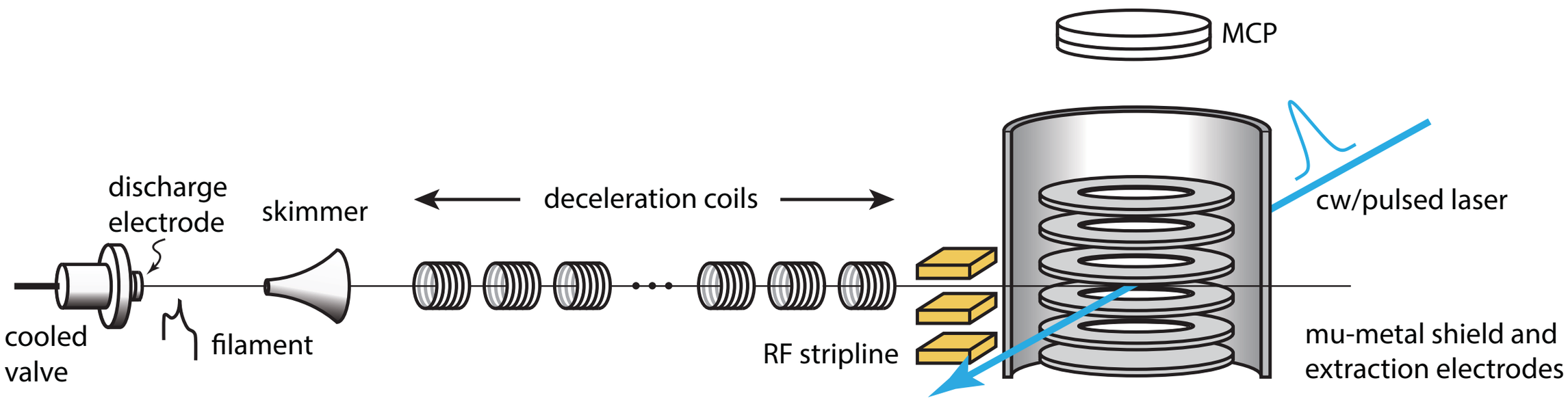

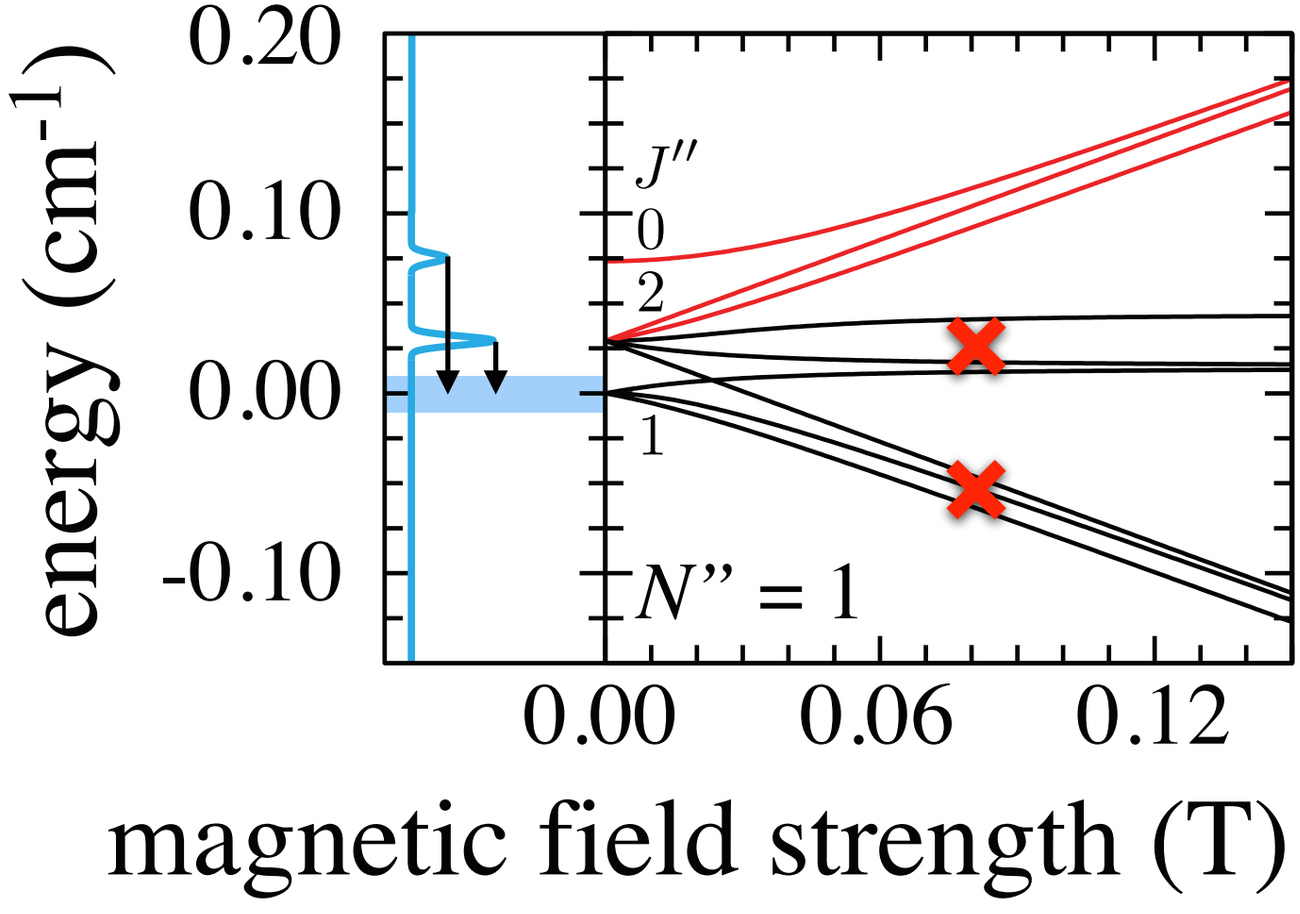

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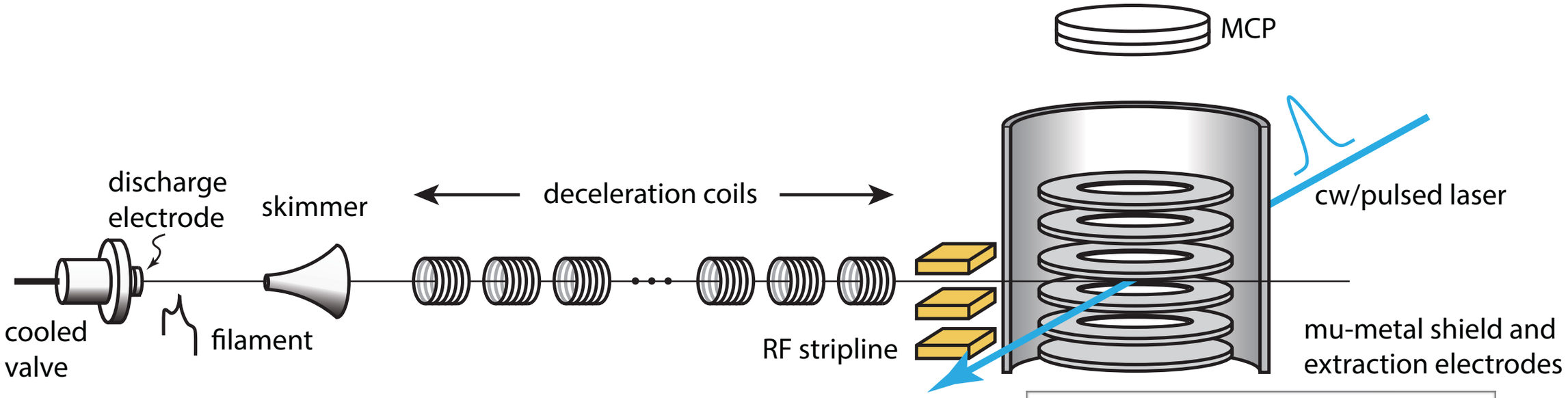
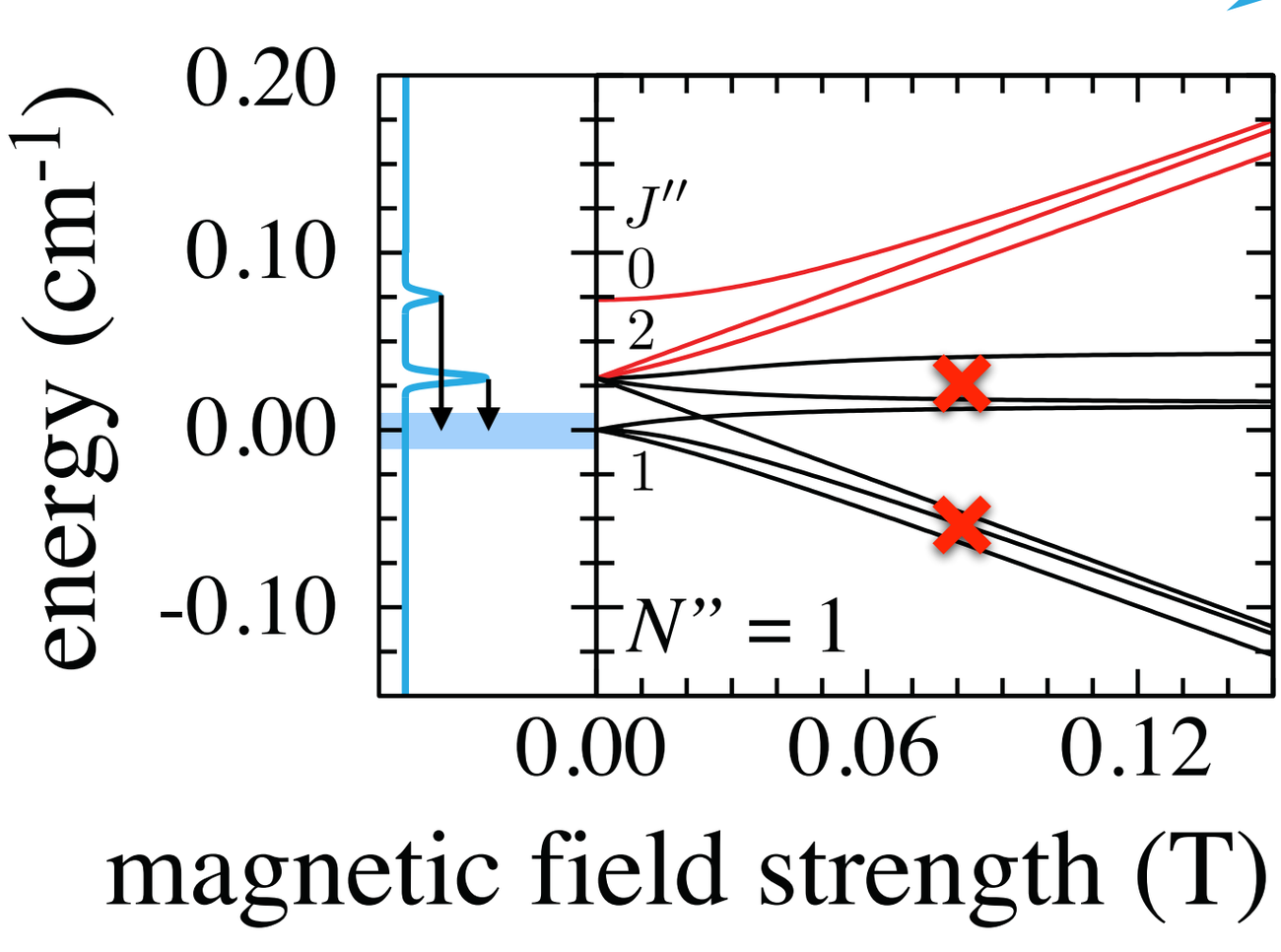
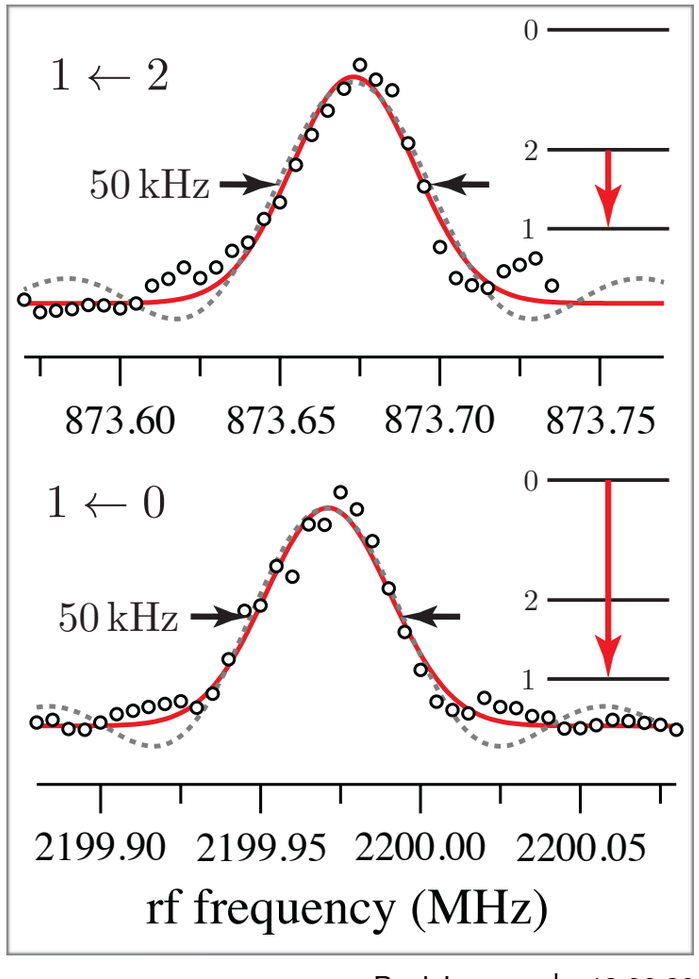
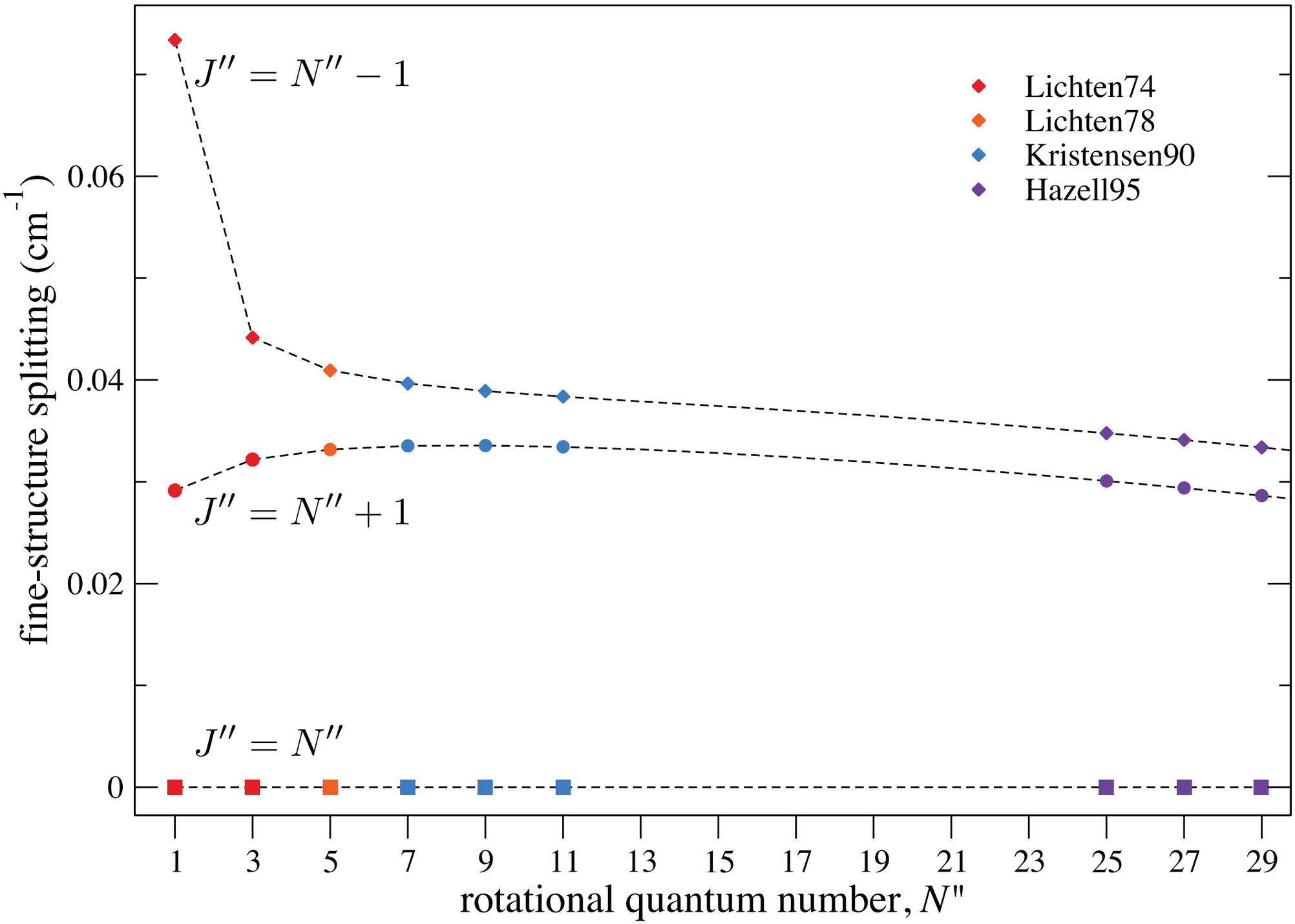






Remeasurement of the fine structure of the metastable *a* $^3\Sigma_u^+$ state of He_2^*



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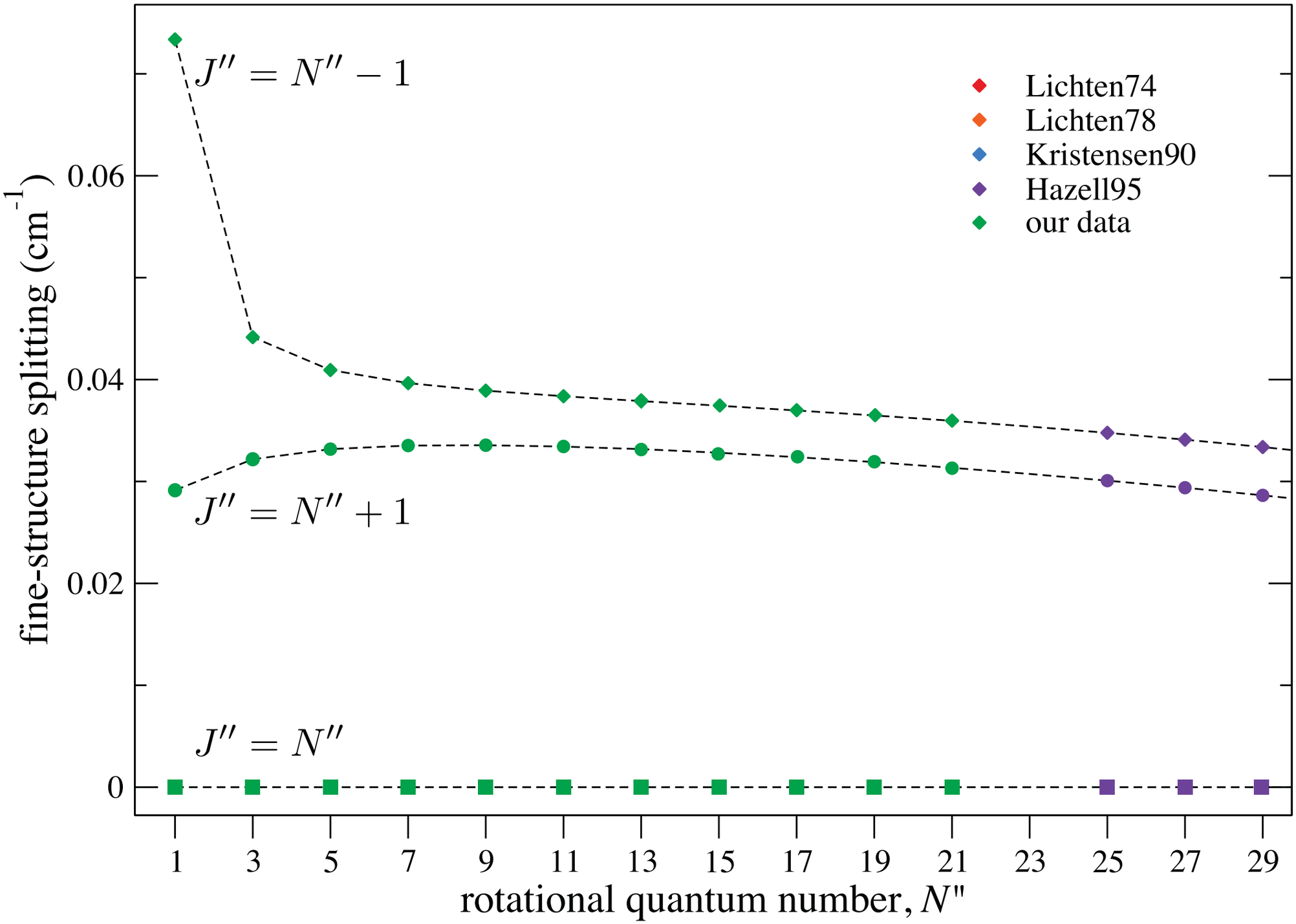
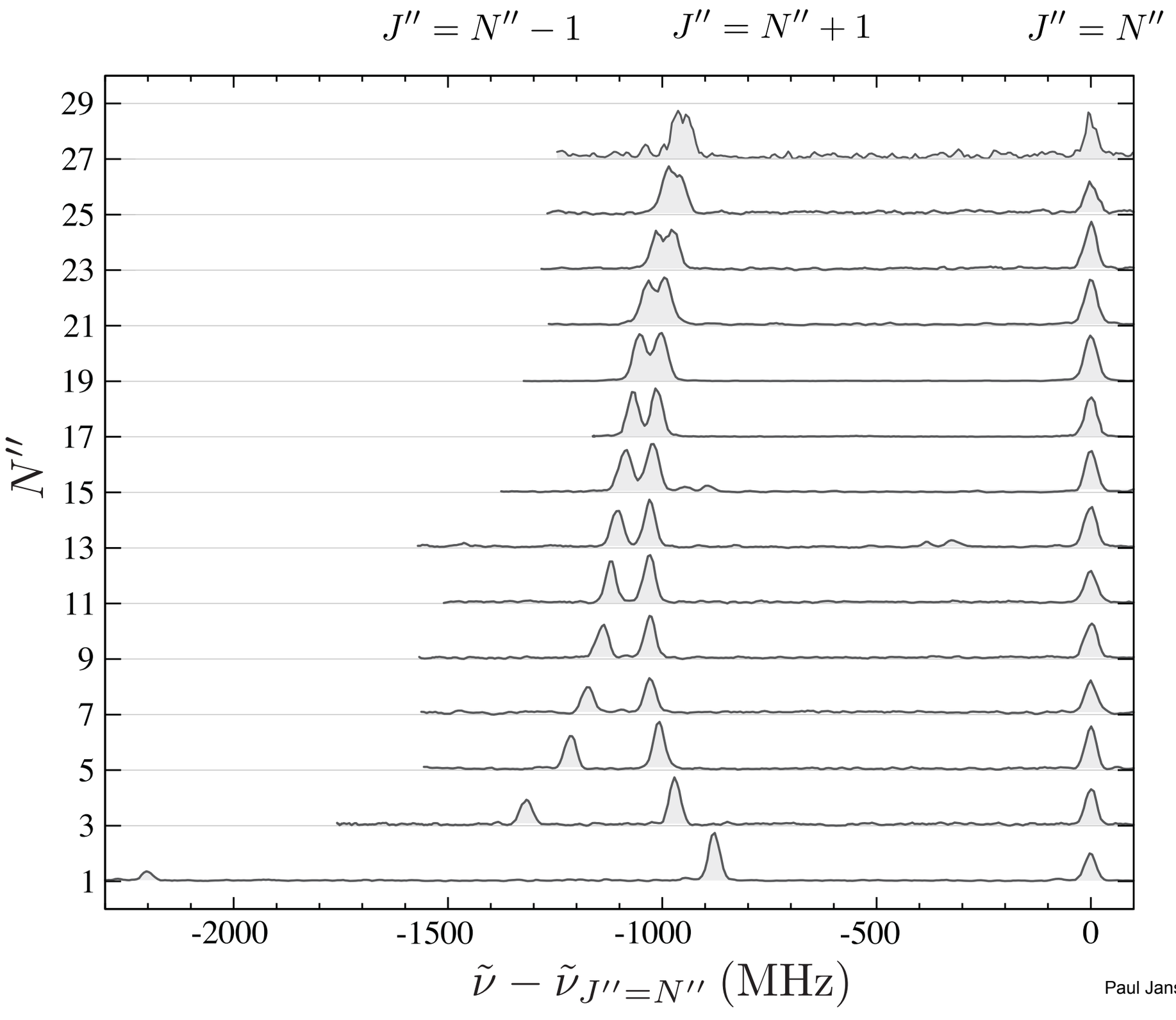
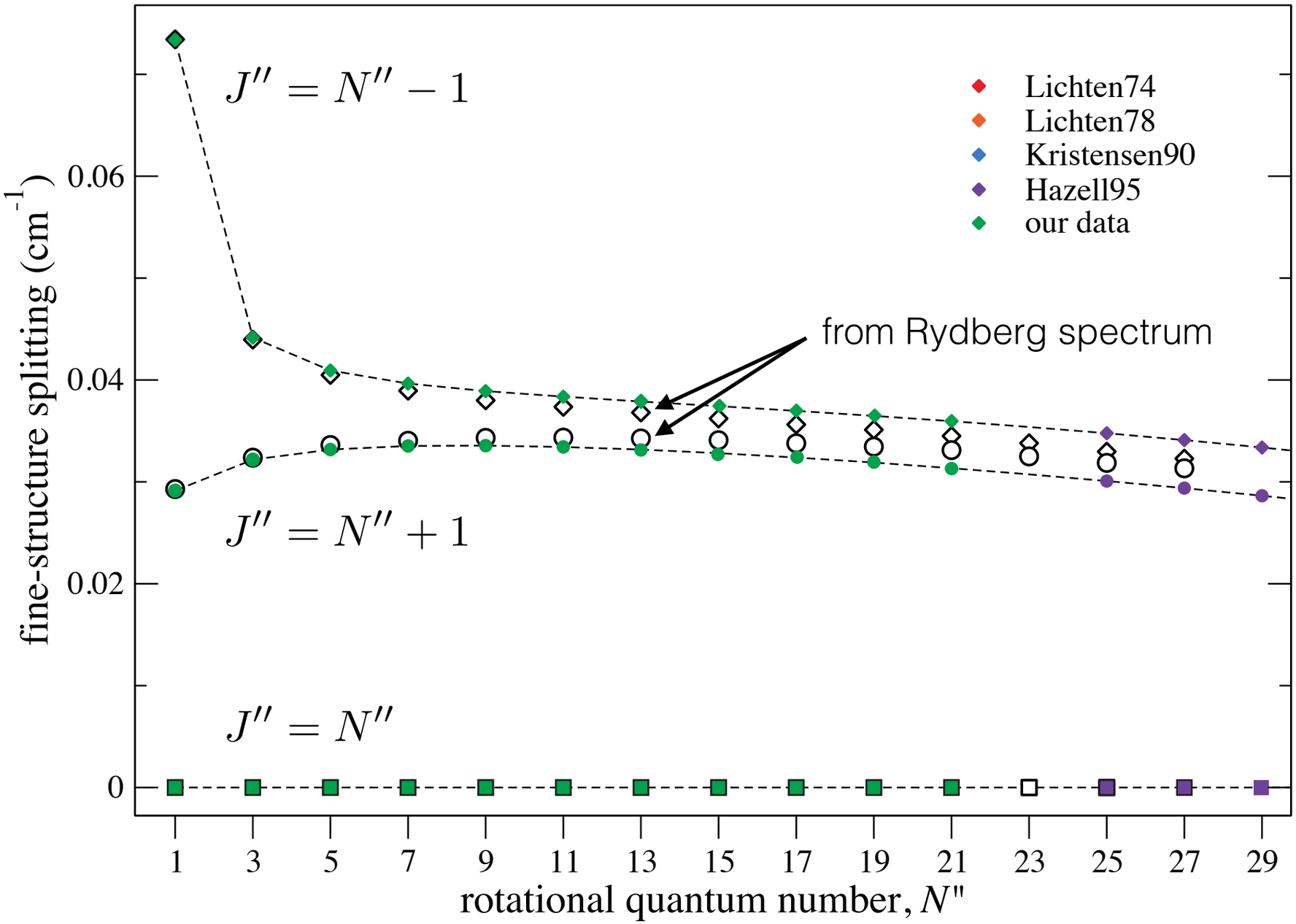
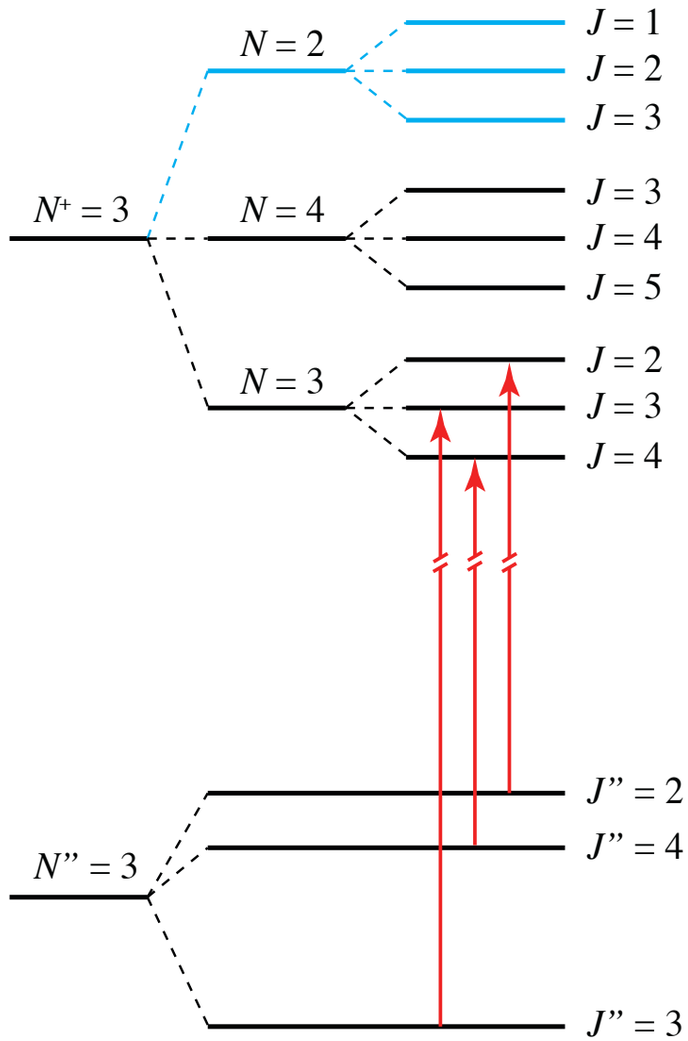


High-resolution Rydberg spectrum of He_2

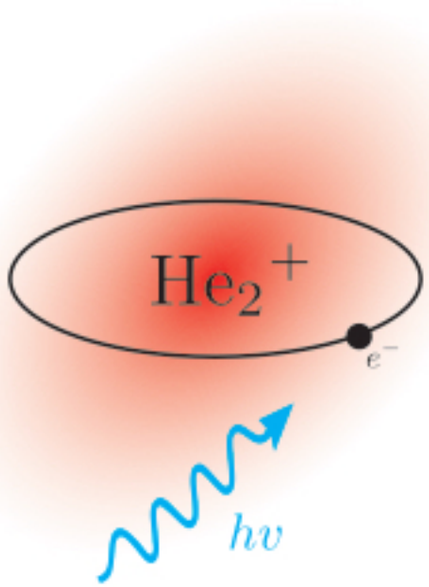


Determination of the fine structure of the Rydberg states of He_2



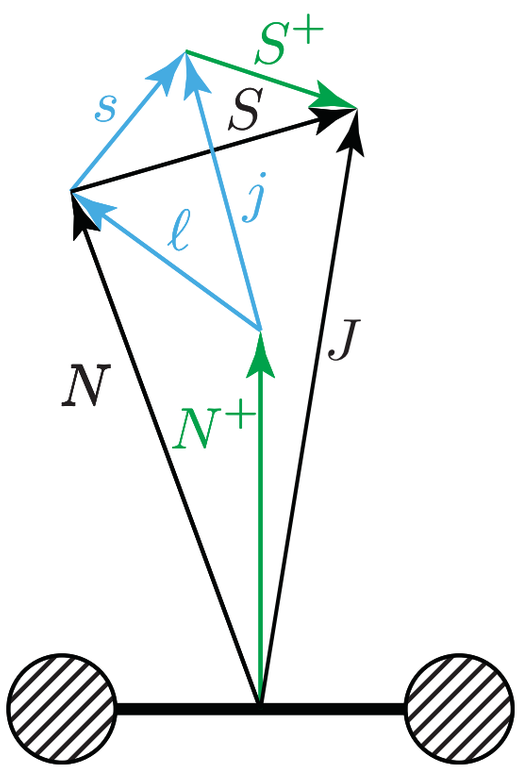


Rotational selection rules (heuristic)



$2s\,(N'', S'', J'')$

$np\,(N^+, \ell, S, J)$



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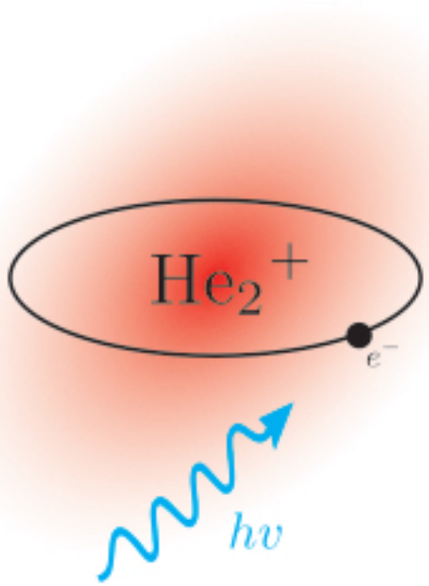
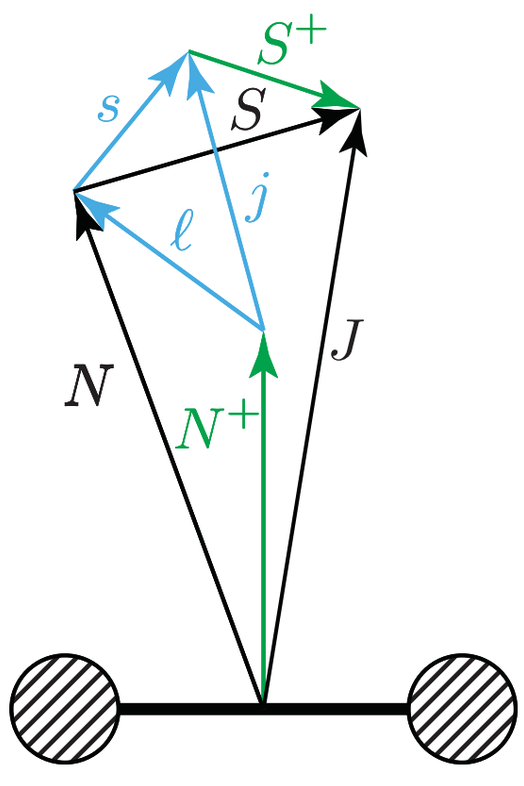


$2s\,(N'', S'', J'')$ $\qquad$ $np\,(N^+, \ell, S, J)$

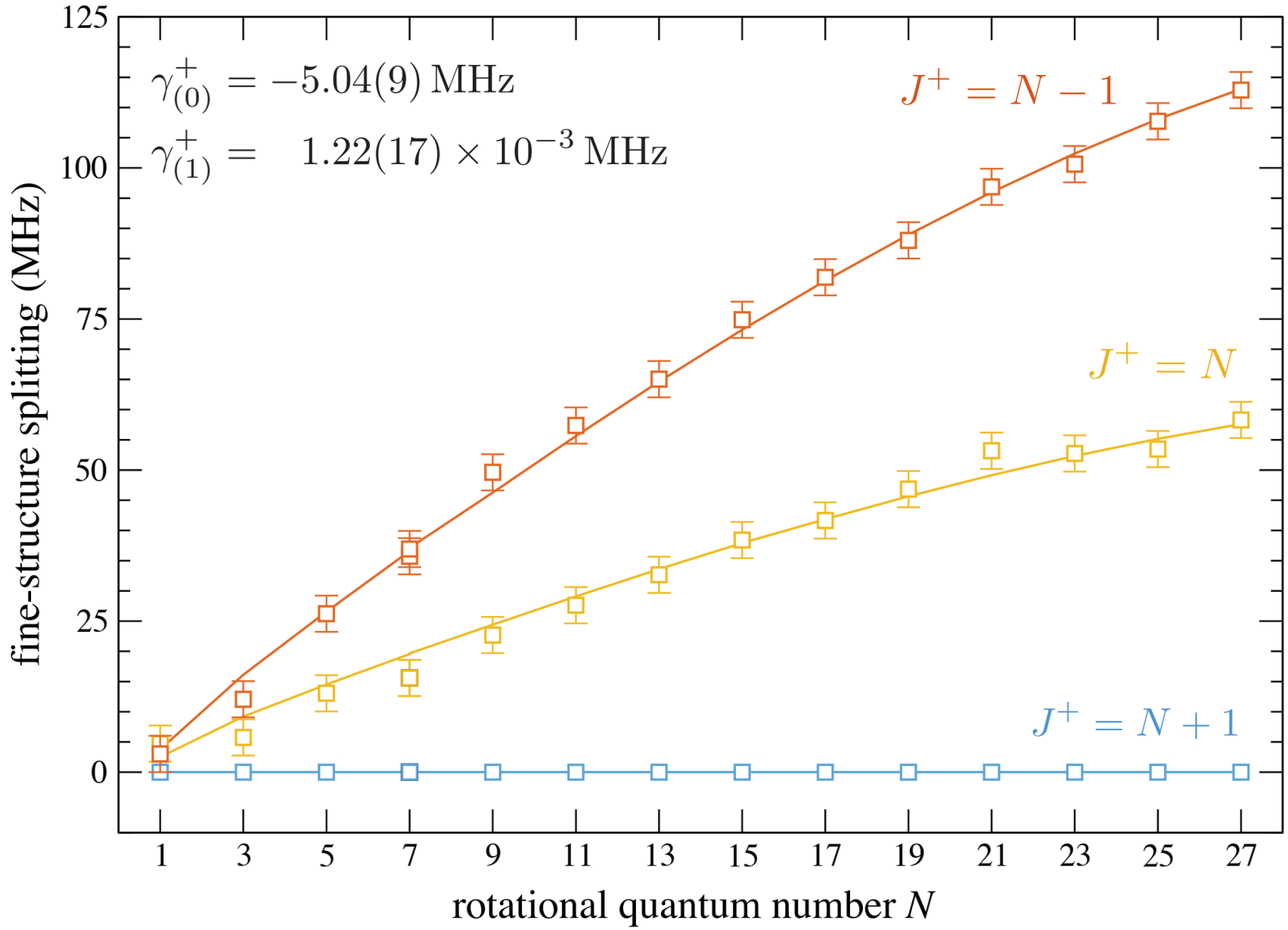
$$S = S'' \qquad \left.\begin{aligned} \vec{J}'' &= \vec{N}'' + \vec{S} \\ \vec{J} &= \underbrace{\vec{N}^+ + \vec{\ell}}_{\vec{N}} + \vec{S} \end{aligned}\right\} \Delta J = \Delta N$$



Determining the fine structure of *n*p Rydberg states of He_2

- MQDT was used to fit spin-rotation constants to experimental data

$$\gamma^+(N^+) = \gamma^+_{(0)} + \gamma^+_{(1)} N^+(N^+ + 1)$$



- Scaling the *ab initio* value for $^3He^4He^+$ [1] and ignoring core electrons as well as mixing of nearby states gives

$$\gamma^+_{(0)} = -3\,\mathrm{MHz},$$
$$\gamma^+_{(1)} = 1.2 \times 10^{-3}\,\mathrm{MHz}$$

[1] Yu *et al.*, *Phys. Rev. Lett.* **62**, 253 (1989).

Conclusions & Outlook

- We have observed the fine-structure of metastable He_2 and of the ion-core of $^4He_2$ Rydberg states and determined effective spin-rotational and distortion constants of $^4He_2^+$.

- These results indicate a need to improve *ab initio* calculations in 3-electron molecules.

- These magnetic-dipole allowed transitions might be observable under astrophysical conditions.

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