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 $^3\Sigma_u^+$ states of molecular helium and $\text{He}_2^+$

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The periodic table for astronomy

- Helium was the first atom to become neutral after Big Bang.
- $\text{He}_2^+$ was among the first molecules to be formed [1].

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$^+$ ($\nu^+ = 22, 23 \rightarrow 0,1$) in $^4$He$_2^+$ (uncertainty $\sim 2$ MHz) [1]
  - 9 rovibrational ($\nu^+ = 0$) transitions in $^3$He$^4$He$^+$ (uncertainty $\sim 18$ MHz) [2]

Rydberg states of He$_2$

\[ X + 2 \Sigma^+_u \]

\( np \) Rydberg states

\[ a \ 3 \Sigma^+_u \]
He$_2^*$ in an inhomogeneous magnetic field

Spin-polarized molecular beams by Zeeman deceleration

![Graph showing spin-polarized molecular beams](image)

- He$_2^+$ intensity (arb. units)
- Wavenumber (cm$^{-1}$)

- Undecelerated (1000 m/s)
- Decelerated (120 m/s)

- Transitions:
  - $1 \rightarrow np_{1_1}$
  - $1 \rightarrow np_{1_2}$
  - $3 \rightarrow np_{3_4}$
  - $3 \rightarrow nf_{3_4}$

- Wavenumbers:
  - $93, 94, 95, 96$
  - $119, 120, 121, 122, 123, 124, 125$

- Quantum states:
  - $J'' = 0, 1, 2$
  - $J'' = 24, 3$
A Zeeman-decelerated molecular-beam magnetic-resonance method

- skimmer
- deceleration coils
- MCP
- mu-metal shield and extraction electrodes
- cooled valve
- filament
- discharge electrode
- RF stripline
- cw/pulsed laser

Energy (cm⁻¹) vs. Magnetic Field Strength (T)

- $N'' = 1$
- $J'' = 0$
- $J'' = 2$
- $J'' = 1$

Energy (cm⁻¹):
- -0.10
- 0.00
- 0.10
- 0.20

Magnetic Field Strength (T):
- 0.00
- 0.06
- 0.12
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Energy (cm\(^{-1}\)) vs. Magnetic Field Strength (T)

- \(N'' = 1\)
- \(J''\) values: 0, 1, 2

- Graph showing the relationship between energy (cm\(^{-1}\)) and magnetic field strength (T) for different values of \(J''\).
A Zeeman-decelerated molecular-beam magnetic-resonance method

- Skimmer deceleration coils
- MCP
- Mu-metal shield and extraction electrodes
- Cooled valve
- Discharge electrode
- Filament
- RF stripline
- CW/pulsed laser

**Graph:**
- Energy (cm$^{-1}$) vs. magnetic field strength (T)
- $J''$ levels:
  - $J'' = 0$
  - $J'' = 1$
  - $J'' = 2$
- $N'' = 1$

**Data Points:**
- $J'' = 0$: $E = 0.00$
- $J'' = 1$: $E = -0.10$
- $J'' = 2$: $E = -0.20$
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deceleration coils
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Energy (cm⁻¹) vs. Magnetic Field Strength (T)

- N'' = 1
- J'' = 0, 1, 2

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A Zeeman-decelerated molecular-beam magnetic-resonance method

- Discharge electrode
- Skimmer
- Deceleration coils
- RF stripline
- Cool valve
- Filament
- MCP
- CW/pulsed laser
- Mu-metal shield and extraction electrodes

Energy (cm⁻¹)

<table>
<thead>
<tr>
<th>Energy (cm⁻¹)</th>
<th>0.20</th>
<th>0.10</th>
<th>0.00</th>
<th>-0.10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnetic field strength (T)</td>
<td>0.00</td>
<td>0.06</td>
<td>0.12</td>
<td></td>
</tr>
</tbody>
</table>

- Magnetic field strength (T)
- RF frequency (MHz)
- 1 ← 2
- 50 kHz

- Energy levels: N" = 1
  - 1 ← 2
  - 1 ← 0
  - 50 kHz

- Energy transitions:
  - 1 ← 2
  - 1 ← 0
  - 50 kHz
Remeasurement of the fine structure of the metastable $a \ ^3\Sigma_u^+ \ \text{state of He}_2^*$

$J'' = N'' - 1$

$J'' = N'' + 1$

$J'' = N''$

fine-structure splitting (cm$^{-1}$)

rotational quantum number, $N''$

Lichten74  
Lichten78  
Kristensen90  
Hazell95
Remeasurement of the fine structure of the metastable $a \ ^3\Sigma_u^+$ state of $\text{He}_2^*$

$$J'' = N'' - 1$$

$$J'' = N'' + 1$$

$$J'' = N''$$

fine-structure splitting (cm$^{-1}$)

rotational quantum number, $N''$

- Lichten74
- Lichten78
- Kristensen90
- Hazell95
- our data
High-resolution Rydberg spectrum of He$_2$

\[ J'' = N'' - 1 \quad J'' = N'' + 1 \quad J'' = N'' \]

\[ \tilde{\nu} - \tilde{\nu}_{J''=N''} \text{ (MHz)} \]
Determination of the fine structure of the Rydberg states of $\text{He}_2$

\[ J'' = N'' - 1 \]

\[ J'' = N'' + 1 \]

\[ J'' = N'' \]

rotational quantum number, $N''$

fine-structure splitting (cm$^{-1}$)

Lichten74
Lichten78
Kristensen90
Hazell95
our data

from Rydberg spectrum
Rotational selection rules (heuristic)

\[ 2s\ (N'', \ S'', \ J'') \qquad n\ell \ (N^+, \ \ell, \ S, \ J) \]
Rotational selection rules (heuristic)

\[ S = S'' \]
\[ J = N + \ell + S \]

\[ \Delta J = \Delta N \]
Determining the fine structure of np 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 $^3\text{He}^4\text{He}^+$ [1] and ignoring core electrons as well as mixing of nearby states gives

\[ \gamma_{(0)}^+ = -3 \text{ MHz}, \]
\[ \gamma_{(1)}^+ = 1.2 \times 10^{-3} \text{ MHz} \]

\[ J^+ = N - 1 \]
\[ J^+ = N \]
\[ J^+ = N + 1 \]

Conclusions & Outlook

- We have observed the fine-structure of metastable He$_2$ and of the ion-core of $^4$He$_2$ Rydberg states and determined effective spin-rotational and distortion constants of $^4$He$_2^+$.

- 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.