Precision spectroscopy of molecular hydrogen and the search for new physics

Wim Ubachs
Vrije Universiteit Amsterdam

International Symposium on Molecular Spectroscopy
Urbana-Champaign
22 June 2017
The Standard Model of Physics

What do we know?

- Quarks: up, charm, top, down, strange, bottom
- Leptons: electron, muon, tau neutrino

What do we not know?

- Dark Matter
- Dark Energy
- How does Gravity fit to SM?
- Why is Gravity so weak?
- Are there only 3+1 dimensions?
- Are there only 4 forces?

Fundamental constants drift?

- Variation on cosmological scale
- Dependencies on fields as indicators?
New urgent question: proton-radius puzzle

**Hydrogen (H)**

- Energy levels: 1S, 2S
- Proton size: 0.8768(69) fm

**Muonic hydrogen (µH)**

- Energy levels: 1S, 2S, 2P
- Proton size: 0.84184(67) fm
- Transitions: 2S → 2P, 1S → 2S, 6 μm (2 keV), 0.6 nm

---

C.G. Parthey et al., PRL *107*, 203001 (2011)

Varying Constants of Nature?

Coupling constants are free parameters in Standard Model
But cannot be varied at will

\[
S = \int \left( L_{\text{mat}} + \frac{j_\mu}{c} A^\mu - \frac{\varepsilon_0}{4} F_{\mu\nu} F^{\mu\nu} e^{-2\phi} - \frac{\hbar c}{2l^2} \partial_\mu \partial^\mu \phi \right) d\Omega
\]

Bekenstein – Barrow – Flambaum : consistent models

\[ \phi \rightarrow \Delta \alpha, \Delta \mu \]

1) Variation on cosmological time scales
   “Connection to Dark Energy scenarios”

2) Coupling to environment
   -> “chameleons”
   Dependence on local density
   Dependence on gravity
Empirical search for a change in $\mu$

Compare $H_2$ in different epochs

Lab today
90-112 nm

QSO 12 Gyr ago
~300-500 nm

Cosmological redshift

$$\frac{\lambda_i^z}{\lambda_i^0} \equiv 1 + z_i$$

$$T = T_0 \left[ 1 - \frac{1}{(1 + z_{abs})^{3/2}} \right]$$

Practical: atmospheric transmission only for $z > 2$
H$_2$ laboratory wavelengths
The Amsterdam “XUV-laser”

XUV-laser excitation (90-115 nm)

For HD


### TABLE I: Comprehensive list of measured transition wavelengths of the Lyman (L) and Werner (W) lines using the ultranarrowband XUV laser source in Amsterdam. Values in nm.

<table>
<thead>
<tr>
<th>Line</th>
<th>$\lambda_0$ (nm)</th>
<th>Line</th>
<th>$\lambda_0$ (nm)</th>
<th>Line</th>
<th>$\lambda_0$ (nm)</th>
<th>Line</th>
<th>$\lambda_0$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>L0</td>
<td>110.006 251</td>
<td>L8</td>
<td>100.838 615</td>
<td>L13</td>
<td>95.894 665</td>
<td>W1</td>
<td>99.138 046</td>
</tr>
<tr>
<td>L0</td>
<td>110.812 733</td>
<td>L8</td>
<td>100.182 387</td>
<td>L13</td>
<td>96.215 297</td>
<td>W1</td>
<td>98.679 800</td>
</tr>
<tr>
<td>L0</td>
<td>110.863 326</td>
<td>L8</td>
<td>100.245 210</td>
<td>L13</td>
<td>94.751 403</td>
<td>W1</td>
<td>98.797 445</td>
</tr>
<tr>
<td>L1</td>
<td>109.405 198</td>
<td>L8</td>
<td>100.398 545</td>
<td>L14</td>
<td>94.616 931</td>
<td>W1</td>
<td>98.972 929</td>
</tr>
<tr>
<td>L1</td>
<td>109.643 894</td>
<td>L8</td>
<td>100.641 416</td>
<td>L14</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L1</td>
<td>109.978 718</td>
<td>L9</td>
<td>99.280 968</td>
<td>L14</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L1</td>
<td>109.219 523</td>
<td>L9</td>
<td>99.137 891</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L1</td>
<td>109.273 423</td>
<td>L9</td>
<td>99.201 637</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L1</td>
<td>109.424 460</td>
<td>L9</td>
<td>99.355 061</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L1</td>
<td>109.672 534</td>
<td>L9</td>
<td>99.597 278</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L2</td>
<td>107.892 547</td>
<td>L10</td>
<td>98.283 533</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L2</td>
<td>107.713 874</td>
<td>L10</td>
<td>98.486 398</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L2</td>
<td>107.769 894</td>
<td>L10</td>
<td>98.776 882</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L2</td>
<td>107.922 542</td>
<td>L10</td>
<td>98.143 871</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L2</td>
<td>108.171 124</td>
<td>L10</td>
<td>98.207 427</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L2</td>
<td>108.514 554</td>
<td>L10</td>
<td>98.359 107</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L3</td>
<td>106.460 539</td>
<td>L10</td>
<td>98.596 279</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L3</td>
<td>106.690 068</td>
<td>L11</td>
<td>97.334 458</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L3</td>
<td>106.288 214</td>
<td>L11</td>
<td>97.534 576</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L3</td>
<td>106.346 014</td>
<td>L11</td>
<td>97.821 804</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L3</td>
<td>106.499 481</td>
<td>L11</td>
<td>97.978 623</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L3</td>
<td>106.747 855</td>
<td>L11</td>
<td>97.263 275</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L4</td>
<td>105.103 253</td>
<td>L12</td>
<td>97.415 791</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L4</td>
<td>104.936 744</td>
<td>L12</td>
<td>97.655 283</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L4</td>
<td>104.995 976</td>
<td>L12</td>
<td>97.980 512</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L4</td>
<td>105.149 857</td>
<td>L12</td>
<td>98.389 096</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L4</td>
<td>105.397 610</td>
<td>L12</td>
<td>96.431 064</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L5</td>
<td>103.815 713</td>
<td>L12</td>
<td>96.627 550</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L5</td>
<td>103.654 581</td>
<td>L12</td>
<td>96.908 984</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L5</td>
<td>103.714 092</td>
<td>L12</td>
<td>96.297 800</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L5</td>
<td>103.869 027</td>
<td>L12</td>
<td>96.360 800</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L5</td>
<td>104.115 892</td>
<td>L12</td>
<td>96.504 574</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L6</td>
<td>102.593 517</td>
<td>L12</td>
<td>96.767 695</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L6</td>
<td>102.437 395</td>
<td>L12</td>
<td>97.083 820</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
<tr>
<td>L6</td>
<td>102.498 790</td>
<td>L12</td>
<td>97.488 449</td>
<td>L15</td>
<td>94.698 040</td>
<td>W1</td>
<td>98.563 371</td>
</tr>
</tbody>
</table>

>160 lines measured at $\sim 5 \times 10^{-8}$

Some lines at $< 1 \times 10^{-8}$
"Sensitivity"

\[ K_i = \frac{d \ln \lambda_i}{d \ln \mu} \]

\[ \frac{\lambda_i^z}{\lambda_i^0} \equiv 1 + z_i = \left(1 + z_{abs}\right) \left(1 + K_i \frac{\Delta \mu}{\mu}\right) \]
Various systems observed

Relative velocity [km/s] around \( z_0 \)

(a) J2123 +0050, \( z_0 = 2.0594 \)

\( \lambda_{lab} = 1108.1273 \) [Å]

L0R0, \( \lambda_{lab} = 1108.6332 \) [Å]

(b) Q2348−011, \( z_0 = 2.4266 \)

(c) B0642−5038, \( z_0 = 2.6586 \)

(d) J1443 +2724, \( z_0 = 4.224 \)

Normalized flux

Observed wavelength [Å]
Analysis method: “comprehensive fitting”

Produce molecular fingerprint

\[ \lambda_i \text{ – set of accurate wavelengths} \]
\[ f_i \text{ – set of line oscillator strengths (from ab initio theory)} \]
\[ \Gamma_i \text{ – set of damping coefficients (from ab initio theory)} \]

Astrophysical conditions

\[ b \text{ – Doppler width parameter} \]
\[ z \text{ – red shift} \]
\[ N_J \text{ – column densities} \]

Fit equation onto spectrum

“Treat” HI and metal lines

Multiple velocity components (?)

\[ \frac{\lambda_i^z}{\lambda^0_i} \equiv 1 + z_i = (1 + z_{abs}) \left(1 + K_i \frac{\Delta \mu}{\mu}\right) \]

\[ K_i \text{ – set of sensitivity coefficients} \]

\[ \frac{\Delta \mu}{\mu} \]
H$_2$ high redshift absorption systems

10 H$_2$ proper absorption systems towards quasars analyzed:

<table>
<thead>
<tr>
<th>Quasar</th>
<th>$z_{\text{abs}}$</th>
<th>$z_{\text{em}}$</th>
<th>RA (J2000)</th>
<th>Decl. (J2000)</th>
<th>$N$(H$_2$)</th>
<th>$N$(HD)</th>
<th>$N$(CO)</th>
<th>N(Hr)</th>
<th>$R_{\text{mag}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>HE0027−1836</td>
<td>2.42</td>
<td>2.55</td>
<td>00:30:23.62</td>
<td>−18:19:56.0</td>
<td>17.3</td>
<td></td>
<td></td>
<td>21.7</td>
<td>17.37</td>
</tr>
<tr>
<td>Q0347−383</td>
<td>3.02</td>
<td>3.21</td>
<td>03:49:43.64</td>
<td>−38:10:30.6</td>
<td>14.5</td>
<td></td>
<td></td>
<td>20.6</td>
<td>17.48</td>
</tr>
<tr>
<td>Q0405−443</td>
<td>2.59</td>
<td>3.00</td>
<td>04:07:18.08</td>
<td>−44:10:13.9</td>
<td>18.2</td>
<td></td>
<td></td>
<td>20.9</td>
<td>17.34</td>
</tr>
<tr>
<td>Q0528−250</td>
<td>2.81</td>
<td>2.81</td>
<td>05:30:07.95</td>
<td>−25:03:29.7</td>
<td>18.2</td>
<td></td>
<td>13.3</td>
<td>21.1</td>
<td>17.37</td>
</tr>
<tr>
<td>B0642−5038</td>
<td>2.66</td>
<td>3.09</td>
<td>06:43:26.99</td>
<td>−50:41:12.7</td>
<td>18.4</td>
<td></td>
<td></td>
<td>21.0</td>
<td>18.06</td>
</tr>
<tr>
<td>Q1232+082</td>
<td>2.34</td>
<td>2.57</td>
<td>12:34:37.58</td>
<td>+07:58:43.6</td>
<td>19.7</td>
<td></td>
<td>15.5</td>
<td>20.9</td>
<td>18.40</td>
</tr>
<tr>
<td>J1237+064</td>
<td>2.69</td>
<td>2.78</td>
<td>12:37:14.60</td>
<td>+06:47:59.5</td>
<td>19.2</td>
<td></td>
<td>14.5</td>
<td>14.2</td>
<td>20.0</td>
</tr>
<tr>
<td>J2123−0050</td>
<td>2.06</td>
<td>2.26</td>
<td>21:23:29.46</td>
<td>−00:50:52.9</td>
<td>17.6</td>
<td></td>
<td>13.8</td>
<td>19.2</td>
<td>15.83</td>
</tr>
<tr>
<td>Q2348−011</td>
<td>2.42</td>
<td>3.02</td>
<td>23:50:57.87</td>
<td>−00:52:09.9</td>
<td>18.4</td>
<td></td>
<td></td>
<td>20.5</td>
<td>18.31</td>
</tr>
</tbody>
</table>

+ 23 additional H$_2$ absorption systems towards quasars known
+ some 20 tentative detections [Balashev et al (2014)]
Status/Review

\[ |\Delta \mu / \mu| = (3.1 \pm 1.6) \times 10^{-6} \]

W. Ubachs, J. Bagdonaite, E.J. Salumbides, M.T. Murphy, L. Kaper

Search for a drifting proton-electron mass ratio from $\text{H}_2$

Rev. Mod. Phys. 88, 021003 (2016)
Perspectives

Wavelength distortions ThAr relate to beam pointing.

Solutions for ‘ESPRESSO’
- Frequency comb calibration
- Fiber feeding
- Fibers designed for $\lambda > 3700$ Å

Problematic for $H_2$ studies, only $z > 3$

**Take Home**

$|\Delta \mu / \mu| < 5 \times 10^{-6}$ at $3\sigma$ level for redshifts $z = 2-4$

**EELT**

More light collection (39 m dish)
Improved signal to noise ratio
“Chameleon” Scenario

Dependence of $\mu$ on gravitational field?

$$\phi_{WD} = \frac{GM}{Rc^2} \sim 10^4 \times \phi_{Earth}$$

Spectrum of GD-133 and GD29-38 Photosphere of White Dwarf stars

In the Galaxy!
Contributions of many lines in the B-X Lyman system

High temperatures
High \( v \) populated
Franck-Condon factors
Dependence of $\Delta \mu/\mu$ on gravitational field

Invoke partition function:

$$P_{vJ}(T) = \frac{1}{\sum_{v=0}^{v_{\text{max}}} \sum_{J=0}^{J_{\text{max}(v)}} g_J(J)(2J+1) \exp \left( -\frac{E_{vJ}}{kT} \right)}$$

Invoke intensities (1500 lines):

$$I_i = N_{\text{col}} \int_{v'v''} f_{v'v''J'J''} P_{v'J'}(T)$$

Lines

$$\frac{\lambda_i^z}{\lambda_i^0} \equiv 1 + z_i = \left(1 + z_{\text{abs}}\right) \left(1 + K_i \frac{\Delta \mu}{\mu}\right)$$

Fit $T$ and $\Delta \mu/\mu$

**GD133:** $\Delta \mu/\mu = (-2.7 +/- 4.7) \times 10^{-5}$

**GD29-38:** $\Delta \mu/\mu = (-5.9 +/- 3.8) \times 10^{-5}$

Molecules as a metrology test system

Search for BSM-physics from laboratory spectroscopy experiment

\[ \Delta E = E_{\text{exp}} - E_{\text{theory}} \]

\[ \delta E = \sqrt{\delta E_{\text{exp}}^2 + \delta E_{\text{theory}}^2} \]

\[ \Delta E < \delta E \quad \text{Validate theory (QED)} \]

\[ \Delta E > \delta E \quad \text{New Physics:} \]

Theory is needed – only for “calculable” systems

\[ \text{H}_2 \] – Krzysztof Pachucki & team

\[ \text{H}_2^+ \] - Jean-Philippe Karr, Laurent Hilico, Vladimir Korobov

Discover new physics \[ \langle \Delta V_{\text{new}} \rangle > \delta E \]

Constrain new physics \[ \langle \Delta V_{\text{new}} \rangle < \delta E \]
Historical Inspiration

Willis E Lamb

Measurement of the tiny $2S_{1/2} - 2P_{1/2}$ splitting

Breakdown of the Dirac theory of the electron
The advent of Quantum Electro Dynamics
Measurement of IP in H$_2$
3 step approach (Zürich-Amsterdam collaboration)

H$_2^+$ : $X^2\Sigma_g^+$, $v^+=0$, $N^+=0,1$

$\tau \sim 150$ ns

EF$^1\Sigma_g^+$, $v=0$, $N=0,1$

$\Delta \nu$
100 kHz

$\Delta \nu$
10 MHz

$\Delta \nu$
5 MHz

$X^1\Sigma_g^+$, $v=0$, $N=0,1$

$E_i$ (ortho) = 124 357.237 97 (36) cm$^{-1}$

$E_i$ (para) = 124 417.491 13 (37) cm$^{-1}$
Benchmark: Dissociation energy $H_2$

$$D_0(H_2) = E_{IP}(H_2) + D_0(H_2^+) - E_{IP}(H)$$

$$D_0(H_2^+) = 21379.350232(50) \text{ cm}^{-1}$$

$$E_{IP}(H) = 109678.7717426(10) \text{ cm}^{-1}$$

$$E_{IP}(H_2) \rightarrow D_0(H_2)$$
## Comparison Theory/Experiment

(Theory: Pachucki, Komasa, et al.: 2010 values)

### \( D_0(H_2) \):
- **Experiment [1]**: \( 36118.0696(4) \text{ cm}^{-1} \)
- **Theory [2]:**
  - Born–Oppenheimer: \( 36112.5927(1) \text{ cm}^{-1} \)
  - Adiabatic: \( + 5.7711(1) \text{ cm}^{-1} \)
  - Nonadiabatic: \( + 0.4339(2) \text{ cm}^{-1} \)
  - Total \( \alpha^0 \): \( 36118.7978(2) \text{ cm}^{-1} \)
  - \( \alpha^2 \) all relativistic: \( - 0.5319(5) \text{ cm}^{-1} \)
  - \( \alpha^3 \) all QED: \( - 0.1948(3) \text{ cm}^{-1} \)
  - \( \alpha^4 \) one-loop term: \( - 0.0016(8) \text{ cm}^{-1} \)
  - Total theory: \( 36118.0695(10) \text{ cm}^{-1} \)

### \( D_0(D_2) \):
- **Total theory [2]**: \( 36748.3633(9) \text{ cm}^{-1} \)
- **Experiment [4]**: \( 36748.3629(7) \text{ cm}^{-1} \)
Fundamental vibration in $\text{H}_2$

Collision-free measurement

Features
- Narrowband UV sources
- Absolute frequency calibration
- 2-photon Doppler-free REMPI

Sagnac alignment
- Delayed ionisation
- ac-Stark extrapolation

$E = 4161.16632(18) \text{ cm}^{-1}$

$\delta E_{\text{exp}} \sim 1.8 \times 10^{-4} \text{ cm}^{-1}$

Precision study of $\text{H}_2 \, X^1\Sigma_g^+ \, v=12, \, 11$

Production of $\text{H}_2$, $v$
Photolysis of $\text{H}_2\text{S}$
Steadman & Baer (1989)

Now:
Three independent lasers

$V=12$: Niu et al.
JCP Comm 142 (2015) 081102

$V=11$: New results
Trivikram et al.

<table>
<thead>
<tr>
<th>$J''$</th>
<th>$E_{\text{exp}}$</th>
<th>$E_{\text{the}}$</th>
<th>$\Delta E_{\text{exp-the}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>34 302.1823 (35)</td>
<td>34 302.1741 (47)</td>
<td>0.008 (6)</td>
</tr>
<tr>
<td>1</td>
<td>34 343.8531 (35)</td>
<td>34 343.8483 (46)</td>
<td>0.005 (6)</td>
</tr>
<tr>
<td>2</td>
<td>34 426.2216 (35)</td>
<td>34 426.2179 (46)</td>
<td>0.004 (6)</td>
</tr>
<tr>
<td>3</td>
<td>34 547.3362 (35)</td>
<td>34 547.3332 (45)</td>
<td>0.003 (6)</td>
</tr>
</tbody>
</table>
Progress in theory

PHYSICAL REVIEW A 82, 032509 (2010)

Born-Oppenheimer potential for H₂

Krzysztof Pachucki

In the whole range of internuclear distance, about $10^{-15}$ precision is achieved; as an example, at the equilibrium distance $r = 1.4011$ a.u., the Born-Oppenheimer potential amounts to $-1.1744759314002167(3)$.

THE JOURNAL OF CHEMICAL PHYSICS 141, 224103 (2014)

Accurate adiabatic correction in the hydrogen molecule

Krzysztof Pachucki¹,a) and Jacek Komasa²,b)

¹Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland
²Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland

(Received 8 October 2014; accepted 18 November 2014; published online 8 December 2014)

For the ground state of H₂ the estimated precision is $3 \times 10^{-7}$ cm⁻¹, which is almost three orders of magnitude higher than that of the best previous result.


Leading order nonadiabatic corrections to rovibrational levels of H₂, D₂, and T₂

Krzysztof Pachucki¹,a) and Jacek Komasa²,b)

¹Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland
²Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland

(Received 22 May 2015; accepted 8 July 2015; published online 21 July 2015)

An estimated accuracy of the leading nonadiabatic correction to the rovibrational energy levels is of the order of $10^{-7}$ cm⁻¹.
Progress in theory

Complete $\alpha^6 m$ corrections to the ground state of H$_2$

Mariusz Puchalski and Jacek Komasa
Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland

Paweł Czachorowski and Krzysztof Pachucki
Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland
(Dated: July 29, 2016)


Relativistic corrections for the ground electronic state of molecular hydrogen

Mariusz Puchalski and Jacek Komasa
Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland

Krzysztof Pachucki
Faculty of Physics, University of Warsaw, Pasteura 5, 02-093 Warsaw, Poland
(Received 13 April 2017; published 25 May 2017)

More importantly, the updated theoretical energies became discrepant with the known experimental values and we conclude that the yet unknown relativistic recoil corrections might be larger than previously anticipated.

Important estimate: 10 kHz on rovibrational energies ~ proton radius at 1%
Strategies to improve the experimental side

Three step approach to IP has been probed via nanosecond excitation
Accuracy: 11 MHz

GK level splitting to IP is known to 1.2 MHz
Sprecher, Beyer, Merkt

Improvements ongoing (2017) GK and EF to IP
Max Beyer, Frederic Merkt
Measurement of the GK-X transition - ongoing

Home-built long-pulse narrowband Ti:Sa laser @ 716 nm

Adjustable pulse duration (match $\tau=30$ ns)
Two-photon Doppler-free
Study of auto-ionization
Improved molecular beam source
Chirp measurement
AC Stark studies
Seed-laser locked to Freq. comb

KBBF crystal for doubling to 179 nm
VUV mirror for retroreflect

2+1$'$ REMPI
@ 179 nm
HD$^+$ ions in a trap; measurement of (8,0)

Look for loss of HD$^+$

Signal detection by REMPD
Signal = fractional loss of ions

During secular excitation, $T_{\text{Be}^+}$ rises to $T_{\text{max}}$

$A_i \propto T \propto N_{\text{HD}^+}$ for $T_{\text{max}} < 400 \text{ mK}$

BUT in practice $T_{\text{max}} \approx 4 \text{ K} \ldots$

$\text{Be}^+$ fluorescence modeled (non-linear)
**HD$^+$ spectrum**

Theory

![Diagram showing transitions in HD$^+$ spectrum]

Experiment

![Experiment data plot]

**Experiment:** 383,407,177.38(41) MHz

**Theory**: 383,407,177.150(15) MHz

Tests of QED in molecules

\[ \Delta E < \delta E \]

<table>
<thead>
<tr>
<th>Species</th>
<th>Splitting</th>
<th>( \delta E_{\text{exp}} )</th>
<th>Ref.</th>
<th>( \delta E_{\text{calc}} )</th>
<th>( \delta E )</th>
<th>( \Delta E )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{H}_2 )</td>
<td>( v = 0,J = 6 - 12 )</td>
<td>150&lt;sup&gt;c&lt;/sup&gt;</td>
<td>[56]</td>
<td>12</td>
<td>150&lt;sup&gt;c&lt;/sup&gt;</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>( v = 0,J = 13 - 16 )</td>
<td>300&lt;sup&gt;c&lt;/sup&gt;</td>
<td>[56]</td>
<td>27</td>
<td>300&lt;sup&gt;c&lt;/sup&gt;</td>
<td>90</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 1 )</td>
<td>4.5&lt;sup&gt;a&lt;/sup&gt;</td>
<td>[54]</td>
<td>2.7</td>
<td>5</td>
<td>7</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 2 )</td>
<td>30</td>
<td>[57]</td>
<td>50</td>
<td>60</td>
<td>12</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 3 )</td>
<td>1.3</td>
<td>[58]</td>
<td>75</td>
<td>75</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 12 )</td>
<td>105</td>
<td>[59]</td>
<td>140</td>
<td>170</td>
<td>150&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>( D_0 )</td>
<td>12</td>
<td>[44]</td>
<td>30</td>
<td>3</td>
<td>36</td>
</tr>
<tr>
<td>( \text{HD} )</td>
<td>( v = 0 \rightarrow 1 )</td>
<td>7&lt;sup&gt;a&lt;/sup&gt;</td>
<td>[54]</td>
<td>2.4</td>
<td>8</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>( D_0 )</td>
<td>11</td>
<td>[49]</td>
<td>30</td>
<td>27</td>
<td>32</td>
</tr>
<tr>
<td>( \text{D}_2 )</td>
<td>( v = 0 \rightarrow 1 )</td>
<td>4.5&lt;sup&gt;a&lt;/sup&gt;</td>
<td>[54]</td>
<td>2.1</td>
<td>5</td>
<td>-0.6</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 2 )</td>
<td>30</td>
<td>[60]</td>
<td>12</td>
<td>30</td>
<td>-12</td>
</tr>
<tr>
<td></td>
<td>( D_0 )</td>
<td>21</td>
<td>[48]</td>
<td>27</td>
<td>30</td>
<td>12</td>
</tr>
<tr>
<td>( \text{H}_2^+ )</td>
<td>( v = 0,J = 0 \rightarrow 2 )</td>
<td>2.3</td>
<td>[61]</td>
<td>0.003</td>
<td>2.3</td>
<td>-1.0</td>
</tr>
<tr>
<td>( \text{HD}^+ )</td>
<td>( v = 0 \rightarrow 1 )</td>
<td>0.064</td>
<td>[62]</td>
<td>0.002</td>
<td>0.064</td>
<td>-0.156</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 4 )</td>
<td>0.50</td>
<td>[63]</td>
<td>0.008</td>
<td>0.50</td>
<td>-0.35</td>
</tr>
<tr>
<td></td>
<td>( v = 0 \rightarrow 8 )</td>
<td>0.41</td>
<td>[64]</td>
<td>0.015</td>
<td>0.41</td>
<td>0.22</td>
</tr>
</tbody>
</table>
Fifth-force searches

Assume: Extra *hadron-hadron* interaction

Parametrize (quantum field theory) as:

**Yukawa potential**

\[
V_5(r) = N_1 N_2 \left\{ \alpha_5 \frac{\exp\left(-\frac{r}{\lambda}\right)}{r} \right\} \hbar c
\]

**Strength:** \( \alpha_5 \)

**Range:** \( \lambda = \hbar / m_5 c \)

**Mass of force carrying particle:** \( m_5 \)

**Hadron numbers:** \( N_1, N_2 \)

See for analysis:

Salumbides, Koelmeij, Komasa, Pachucki, Eikema, Ubachs,

Calculate the expectation value of the energy operator

Level shifts:
\[ \langle \Psi_1 | V_5 | \Psi_1 \rangle; \langle \Psi_0 | V_5 | \Psi_0 \rangle \]

Transition shift:
\[ \langle \Psi_1 | V_5 | \Psi_1 \rangle - \langle \Psi_0 | V_5 | \Psi_0 \rangle \]

Differential effect larger for very high \( v \)'s (\( D_0 \) limit)

\[
\langle \Delta V_{5,\hat{\lambda}} \rangle = \alpha_5 N_1 N_2 \hbar c \left\{ \left| \Psi'_{J'}(r) \frac{\exp(-r/\lambda)}{r} \Psi_{J'}(r) \right| - \left| \Psi''_{J''}(r) \frac{\exp(-r/\lambda)}{r} \Psi''_{J''}(r) \right| \right\}
\]

Parameters \( \alpha_5 \) and \( \lambda \)
Impose constraints on 5\textsuperscript{th} force from spectroscopy HD\textsuperscript{+}/H\textsubscript{2}

\[ \langle \Delta V_5 \rangle < \delta E \quad \text{hence} \quad \alpha_5 < \frac{\delta E}{N_1 N_2 \hbar c \langle \Delta \Psi \rangle (\lambda)} \]

\begin{figure}
\includegraphics[width=\textwidth]{figure}
\end{figure}
Physics of extra spatial dimensions

Immanuel Kant

Number of dimensions consequence of Newton's Universal law of gravitation

\[ \int_F \cdot dA = kQ_{encl} \]

3-dim: \[ A_V \propto r^2 \Rightarrow F \propto \frac{1}{r^2} \]

N-dim: \[ A_V \propto r^{N-1} \Rightarrow F \propto \frac{1}{r^{N-1}} \]
Physics of extra spatial dimensions

Immanuel Kant
Number of dimensions consequence
of Newton's Universal law of gravitation

\[ \int_{V} \frac{\vec{F}}{m} \cdot d\vec{A}_n = -\hat{\Lambda}_n G_n M \]

\[ n\text{-dim: } A_n = \hat{\Lambda}_n r^{n-1} = \left[ \frac{n \pi^{n/2}}{\Gamma\left(\frac{n}{2} + 1\right)} \right] r^{n-1} \Rightarrow F \propto \frac{1}{r^{n-1}} \]

Gravitational attraction depends on dimensionality

W. Ubachs, J. Mol. Spectr. 320 (2016) 1-12
“Compactification”

Theory of consistent EM + Gravity in 5 dimensions (Kaluza)

Extra dimensions are not observed in the macroscopic world
They may be compactified: rolled up (Klein 1926)

String theory: “M-Theory” (Witten) is consistent in 11 dimensions
ADD-theory and Large Extra Dimensions

Arkani–Hamed, Dimopoulos, Dvali

Hierarchy Problem:
Why is gravity so much weaker?
For protons

Why is the Planck mass so much bigger than SM masses?

\[
\frac{V_G}{V_{em}} = 8 \times 10^{-37}
\]

\[
\frac{M_{P1}}{M_Z} \sim 10^{17}
\]

Solution:
Gravity "escapes" into higher dimensions

3-brane (SM) and "Bulk" (gravity)
Large Extra Dimensions (compactified $n$ extra)

$V_{ADD}(r) = -G_{(3+n)} \frac{m_1 m_2}{R_{comp}^n} \frac{1}{r}$ for $r > R_n$

Gravity outside Klein radius

$V_{Newton}(r) = -G_3 m_1 m_2 \frac{1}{r}$

Gravity inside Klein radius

derive $G_{(3+n)} = \left( R_{comp} \right)^n G_3$

$V_{ADD}(r) = -G_3 \frac{m_1 m_2}{r} \left( \frac{R_{comp}}{r} \right)^n$ for $r < R_n$

Enhancement factor for gravity in $n$ extra dimensions
A Cavendish torsion balance at 1 Å distance

Two protons act as Cavendish gravitating balls
ADD in Molecules

Expectation value for the ADD-compactification in a molecule:

\[
\langle V_{ADD}(r) \rangle = \alpha_G N_1 N_2 \left[ \alpha_G N_1 N_2 \right] \left[ \int_0^{R_n} \Psi^*(r) \frac{1}{r^{n+1}} \Psi(r) r^2 dr + \int_{R_n}^{\infty} \Psi^*(r) \frac{1}{r} \Psi(r) r^2 dr \right]
\]

Difference between two quantum states:

\[
\langle \Delta V_{ADD}(r) \rangle = \alpha_G N_1 N_2 \left[ \left\langle \frac{1}{r^{n+1}} \right\rangle_{\Psi_1} - \left\langle \frac{1}{r^{n+1}} \right\rangle_{\Psi_0} \right]
\]

Test for:

\[
\langle \Delta V_{ADD} \rangle_{\text{transition}} < \delta E
\]
Constraints from H$_2$ D$_0$

\[
\left( R_{\text{comp}} \right)^n < \frac{\delta E}{\alpha_g \hbar c N_1 N_2 \Delta \langle r^{-(n+1)} \rangle}
\]

Effect of extra dimensions on the H$_2$ D$_0$ constraint

<table>
<thead>
<tr>
<th>$n$</th>
<th>$R_n$ (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>$2.2 \times 10^4$</td>
</tr>
<tr>
<td>3</td>
<td>$1.9 \times 10^{-1}$</td>
</tr>
<tr>
<td>4</td>
<td>$8.5 \times 10^{-4}$</td>
</tr>
<tr>
<td>5</td>
<td>$3.2 \times 10^{-5}$</td>
</tr>
<tr>
<td>6</td>
<td>$3.7 \times 10^{-6}$</td>
</tr>
</tbody>
</table>

M-theory (10 dim): Compactification on $\mu$m scale !!

$R_c < 0.6 \mu$m
OUTLOOK:
A future molecular test system for physics

Lifetimes $10^6$ seconds (!)

Quadrupole transitions $\sim 10^{14}$ Hz

Possible precision 20-digit

Natural linewidth: $\sim 10^{16}$ cm$^{-1}$

There is room at the bottom guys
OUTLOOK

HD⁺: Doppler-free spectroscopy in Lamb-Dicke regime
