Title: Resonant dipole-dipole interactions between Rydberg atoms and polar molecules at temperatures below 1 K

Speakers: Stephen Hogan

Collection: Cold Atom Molecule Interactions (CATMIN)

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Abstract: Junwen Zou and Stephen Hogan

Resonant dipole-dipole interactions between Rydberg helium atoms and cold ground-state ammonia molecules allow Förster resonance energy transfer between the electronic degrees of freedom in the atom, and the nuclear degrees of freedom associated with the inversion of the molecule [1,2]. In this talk I will describe recent experiments in which we have exploited the Stark effect in the triplet Rydberg states in helium, with values of the principal quantum number n between 38 and 40, to tune these interactions through resonance using electric fields below 10 V/cm. Resonance widths as narrow as 70 MHz have been observed in this work. These are indicative of mean centre-of-mass collision speeds on the order of 10 m/s, and collisions that occur at temperatures significantly below 1 K. Studies of Förster resonances in this collision system are of interest in the search for dipole-bound states [3] of Rydberg atoms or molecules and polar ground-state molecules, in the exploitation of long-range dipole-dipole interactions to regulate access to ion-molecule chemistry that can occur if the polar molecule penetrates inside the Rydberg electron charge distribution [4], and for coherent control and non-destructive detection [5,6].

[1] V. Zhelyazkova and S. D. Hogan, Phys. Rev. A 95, 042710 (2017)
[2] K. Gawlas and S. D. Hogan, J. Phys. Chem. Lett. 11, 83 (2020)
[3] S. M. Farooqi, D. Tong, S. Krishnan, J. Stanojevic, Y. P. Zhang, J. R. Ensher, A. S. Estrin, C. Boisseau, R. Côté, E. E. Eyler and P. L. Gould, Phys. Rev. Lett. 91, 183002 (2003).
[4] V. Zhelyazkova, F. B. V. Martins, J. A. Agner, H. Schmutz and F. Merkt, Phys. Rev. Lett. 125, 263401 (2020)
[5] E. Kuznetsova, S. T. Rittenhouse, H. R. Sadeghpour and S. F. Yelin, Phys. Chem. Chem. Phys. 13, 17115 (2011)
[6] M. Zeppenfeld, Euro. Phys. Lett. 118, 13002 (2017)

Resonant dipole-dipole interactions between Rydberg atoms and polar molecules at temperatures below 1 K

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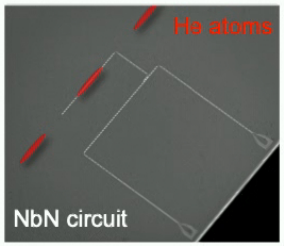
14.7.2022

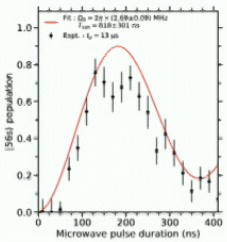
erc European Research Council
UKRI Science and Technology Facilities Council

Interacting/hybrid systems

- Hybrid quantum interfaces
  - Rydberg He atoms & superconducting circuits
  - Coherent coupling to resonator field (19.5 GHz)

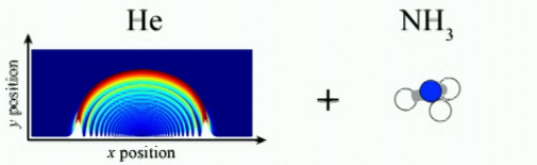
Morgan and Hogan, *Phys. Rev. Lett.* **124**, 193604 (2020)





- Gas-phase collisions ($T < 1$ K)
  - Rydberg He atoms & ground-state NH_3
  - Tuneable interactions
    controlled using weak electric fields

Gawlas and Hogan, *J. Phys. Chem. Lett.* **11**, 83 (2020)



Outline

- Resonant energy transfer between Rydberg He atom and NH_3
- Intrabeam collision apparatus
- Electric-field controlled resonant energy transfer
  - resonance widths
  - collision speeds

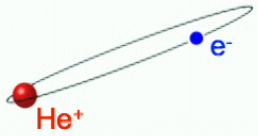
Resonant energy transfer

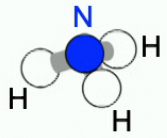
- Resonant dipole-dipole interaction

$$V_{\mathrm{dd}} = \frac{1}{4\pi\varepsilon_0}\left[\frac{\mu_A\,\mu_B}{R^3} - 3\,\frac{(\vec{\mu}_A\cdot\vec{R})\,(\vec{\mu}_B\cdot\vec{R})}{R^5}\right]$$

Perrin, *Ann. Chim. Phys.* **17**, 283 (1932)
Förster, *Ann. Phys.* **2**, 55 (1948)

$$\mathrm{He}(1s\mathit{n}s\ {}^3S_1) + \mathrm{NH_3} \rightarrow \mathrm{He}(1s\mathit{n}p\ {}^3P) + \mathrm{NH_3'}$$

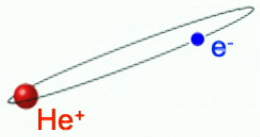


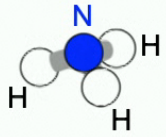


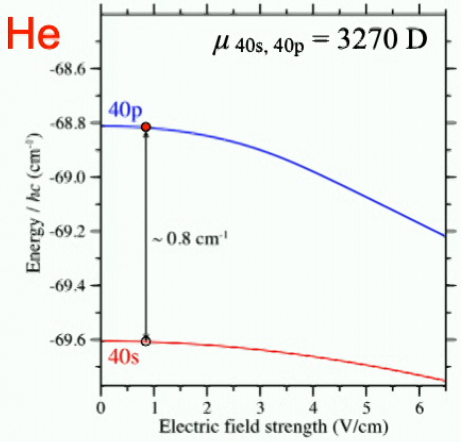
Zhelyazkova and Hogan, *Phys. Rev. A* **95**, 042710 (2017)
Zhelyazkova and Hogan, *J. Chem. Phys.* **147**, 244302 (2017)
Ling, Frey, Smith and Dunning, *J. Chem. Phys.* **98**, 2486 (1993)

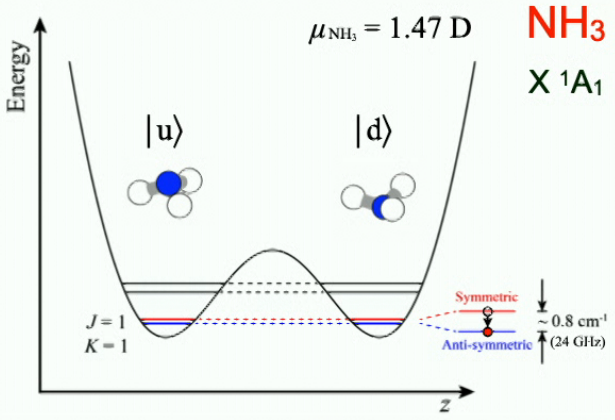
Resonant energy transfer

$$He(1s40s\ ^3S_1) + NH_3 \rightarrow He(1s40p\ ^3P) + NH_3'$$



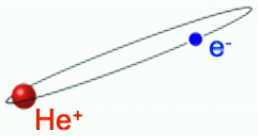


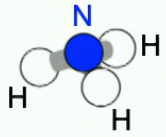




Collisions of NH_3 with Rydberg He

$$He(1s40s\ ^3S_1) + NH_3 \rightarrow He(1s40p\ ^3P) + NH_3'$$

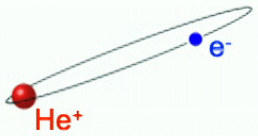


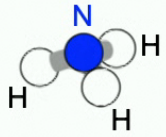


- Model hybrid system in which to study resonant energy transfer
  - energy transfer between nuclear and electronic degrees of freedom
  - effects particle orientation (stereochemistry)
  - exploit long-range dipole interactions to regulate access to short-range chemistry, e.g., ion-molecule reactions inside Rydberg electron orbit

Collisions of NH_3 with Rydberg He

$$He(1s40s\,^3S_1) + NH_3 \rightarrow He(1s40p\,^3P) + NH_3'$$



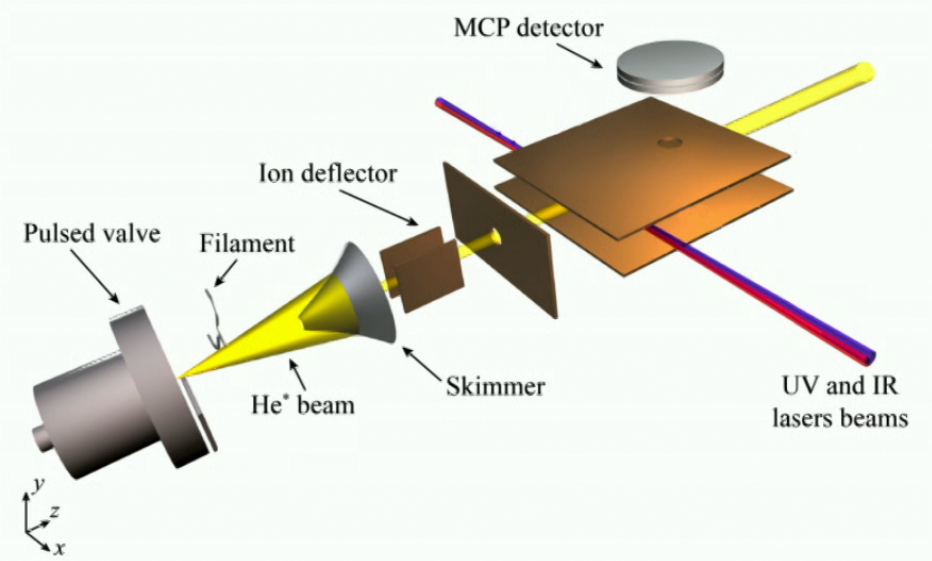


- Connection to experiments with cold/ultracold molecules
  - resonant dipole-dipole interactions between polar molecules and Rydberg atoms for *cooling*, *coherent control* and *non-destructive detection*

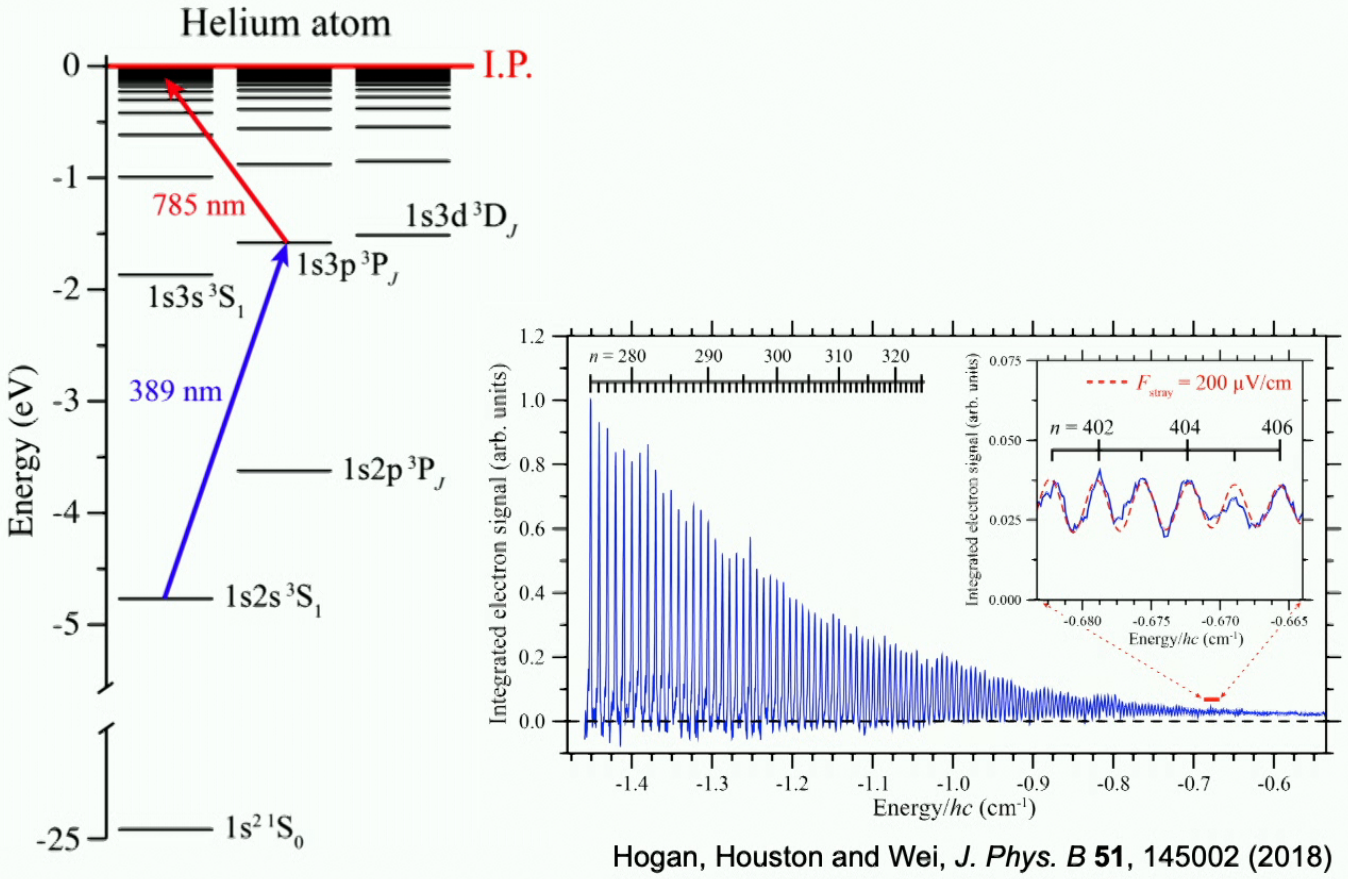
Huber and Buchler, *Phys. Rev. Lett.* **108**, 193006 (2012)
Zhao, Glaetzle, Pupillo and Zoller, *Phys. Rev. Lett.* **108**, 193007 (2012)
Kuznetsova, Rittenhouse, Sadeghpour and Yelin, *Phys. Rev. A* **94**, 032325 (2016)
M. Zeppenfeld, *EuroPhys. Lett.* **118**, 13002 (2017)

Experiment

- Pulsed supersonic beam of NH_3 seeded in He (ratio 1:99 by pressure)
- Two-photon laser excitation from 1s2s $^3S_1$ level to 1s*n*s $^3S_1$ Rydberg levels
- Rydberg-state selective detection by pulsed electric field ionisation

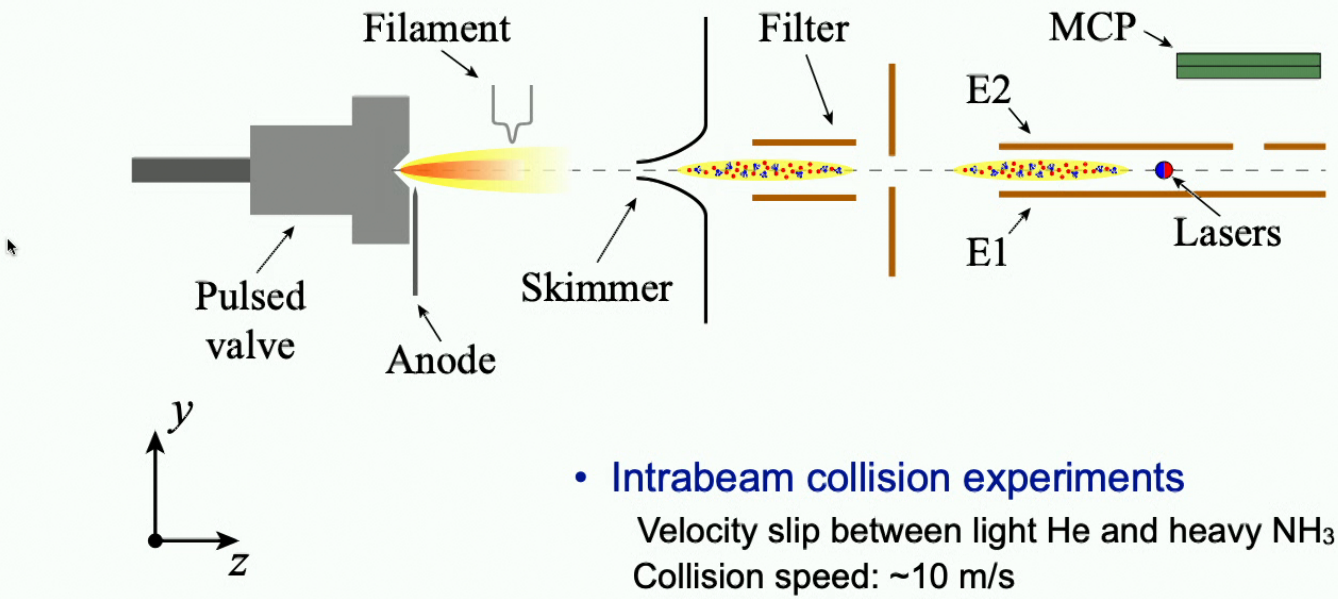


Rydberg state photoexcitation



Hogan, Houston and Wei, *J. Phys. B* **51**, 145002 (2018)

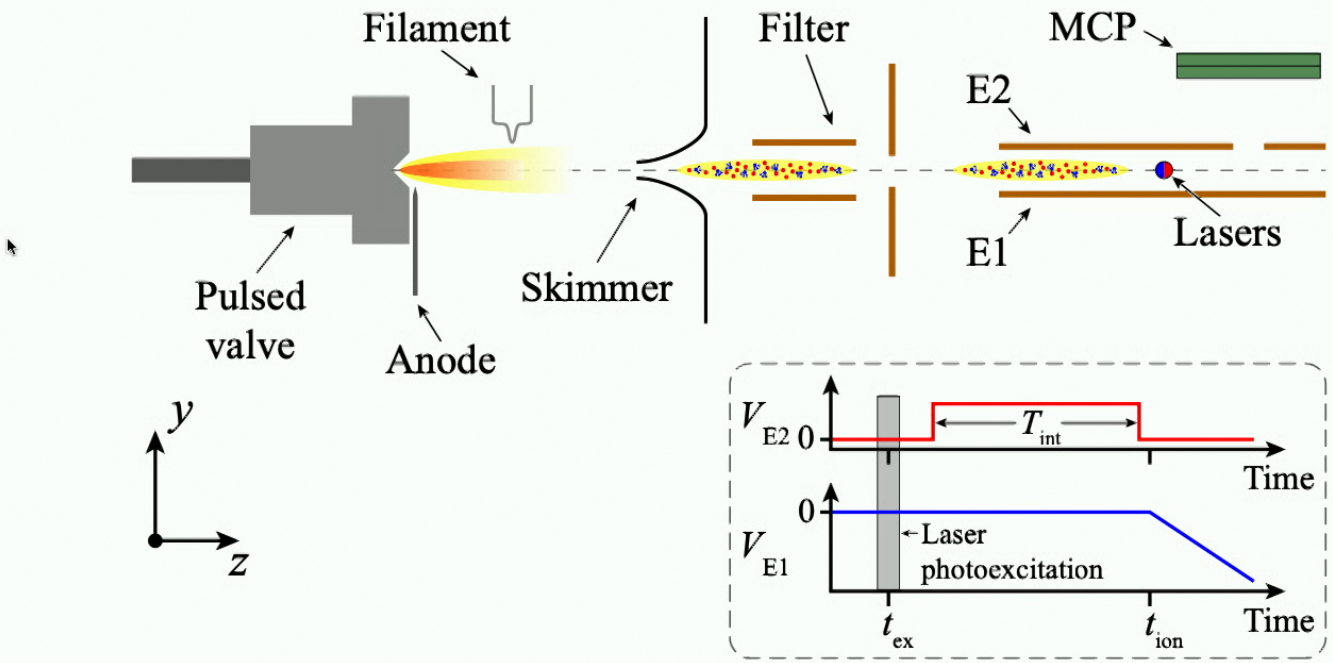
Experimental procedure



- Intrabeam collision experiments
  - Velocity slip between light He and heavy NH_3
  - Collision speed: ~10 m/s
  - Translational temperature: E_{kin}/k_B = 10 — 100 mK

Amarasinghe and Suits, *J. Phys. Chem. Lett.* **8**, 5153 (2017)
Gawlas and Hogan, *J. Phys. Chem. Lett.* **11**, 83 (2020)

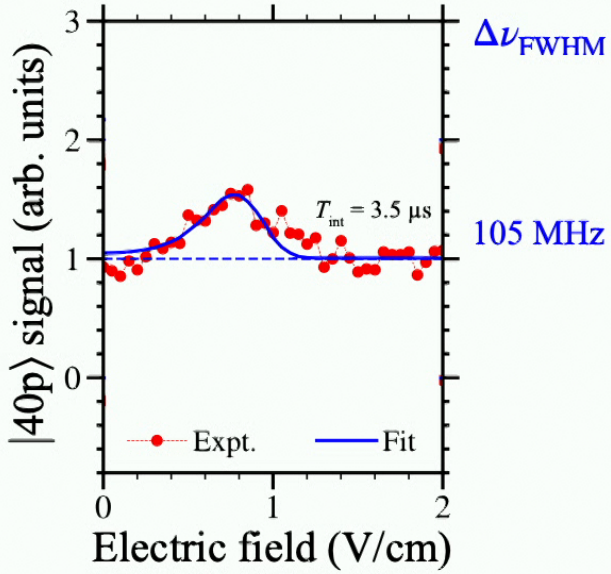
Experimental procedure



Gawlas and Hogan, *J. Phys. Chem. Lett.* **11**, 83 (2020)

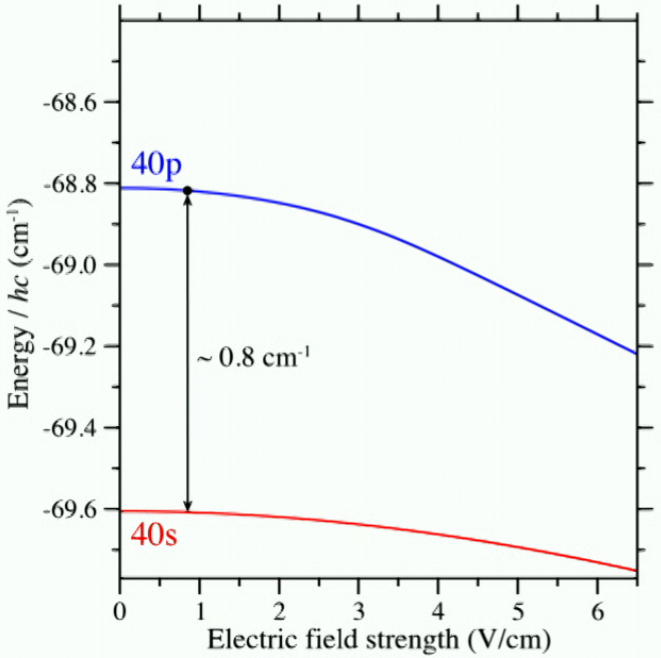
Weak electric fields

- Electric-field controlled resonant energy transfer



$\Delta\nu_{FWHM}$

105 MHz



Weak electric fields

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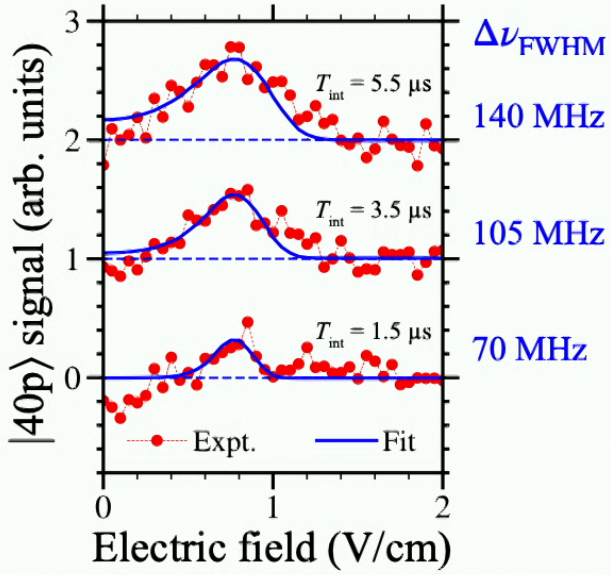


Since $\mu_{NH_3} = 3270$ D in field of 0.8 V/cm

$$\frac{\Delta E_{FWHM}}{h} \simeq \frac{v}{b} = \sqrt{\frac{4\pi\epsilon_0 v^3 h}{\mu_{NH_3}\mu_{He}}}$$

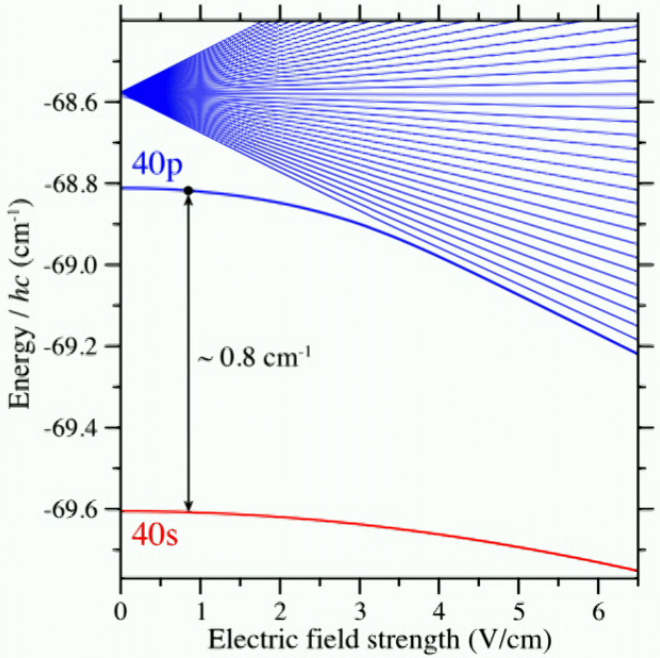
If $\Delta\nu_{FWHM} = 100$ MHz collision speed: ~20 m/s

Hence $E_{kin}/k_B \simeq 80$ mK

Gallagher, *Phys. Rep.* **210**, 319 (1992)

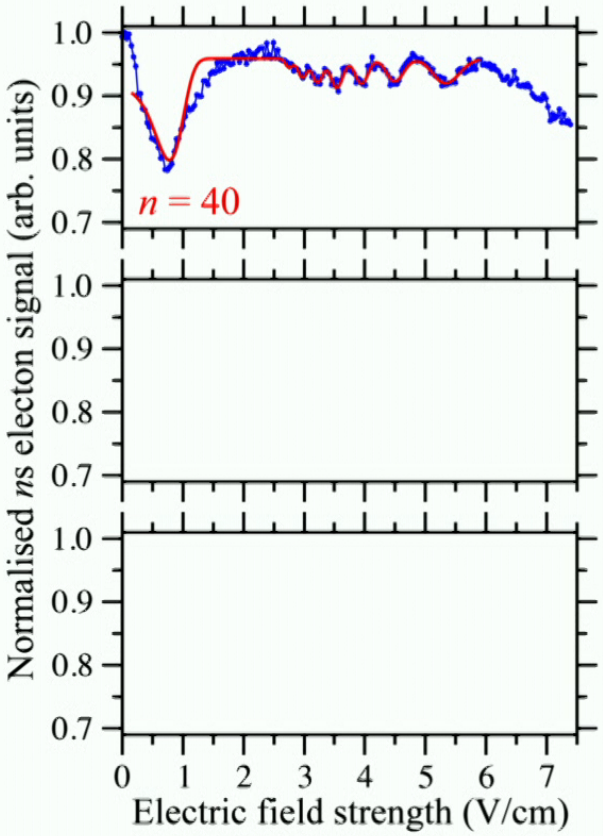
Larger electric fields

- Rydberg-Stark states



Larger electric fields

- Rydberg-Stark states
  — depletion of 40s population



Larger electric fields

- Rydberg-Stark states
  — depletion of 40s population

- Parameter range

  Interaction time $T_{\text{int}} = 11.5\ \mu\text{s}$

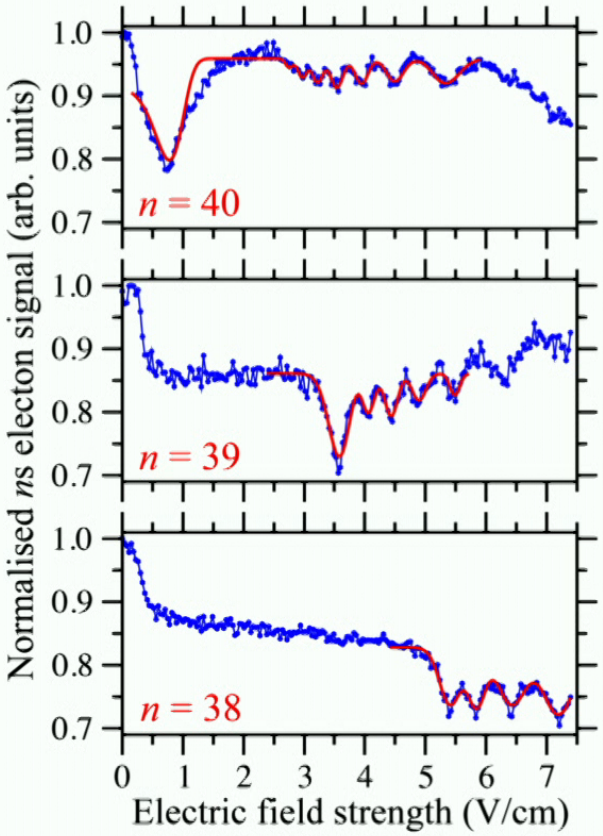
  Energy transfer rate ~100 kHz

  For energy transfer cross-section

$$\sigma = \pi b^2 = \frac{\mu_{\text{He}}\mu_{\text{NH}_3}}{4\epsilon_0 v h}$$

  Assuming pseudo-first-order kinetics

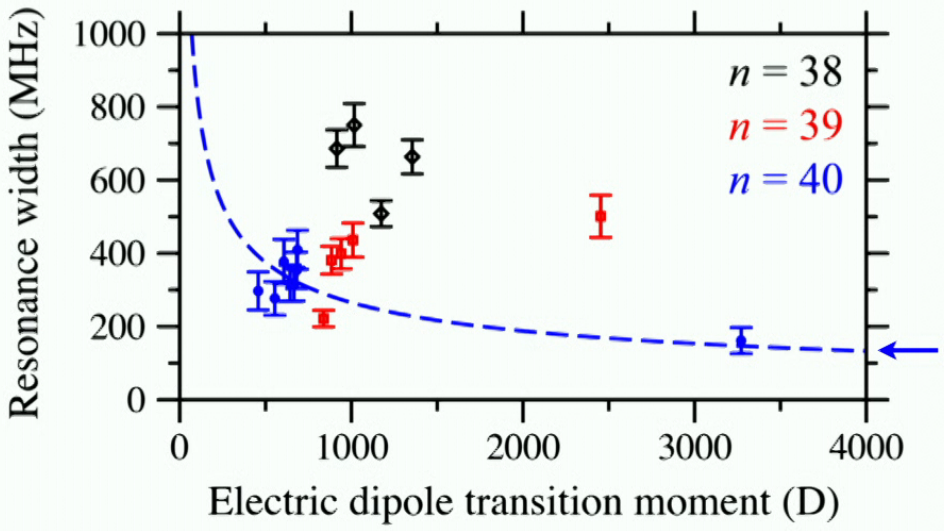
  NH_3 number density ~8×10^{10} cm^{-3}



Resonance widths

- All states/transitions studied

  Interaction time $T_{\mathrm{int}} = 11.5\ \mu\mathrm{s}$



Impact parameter model

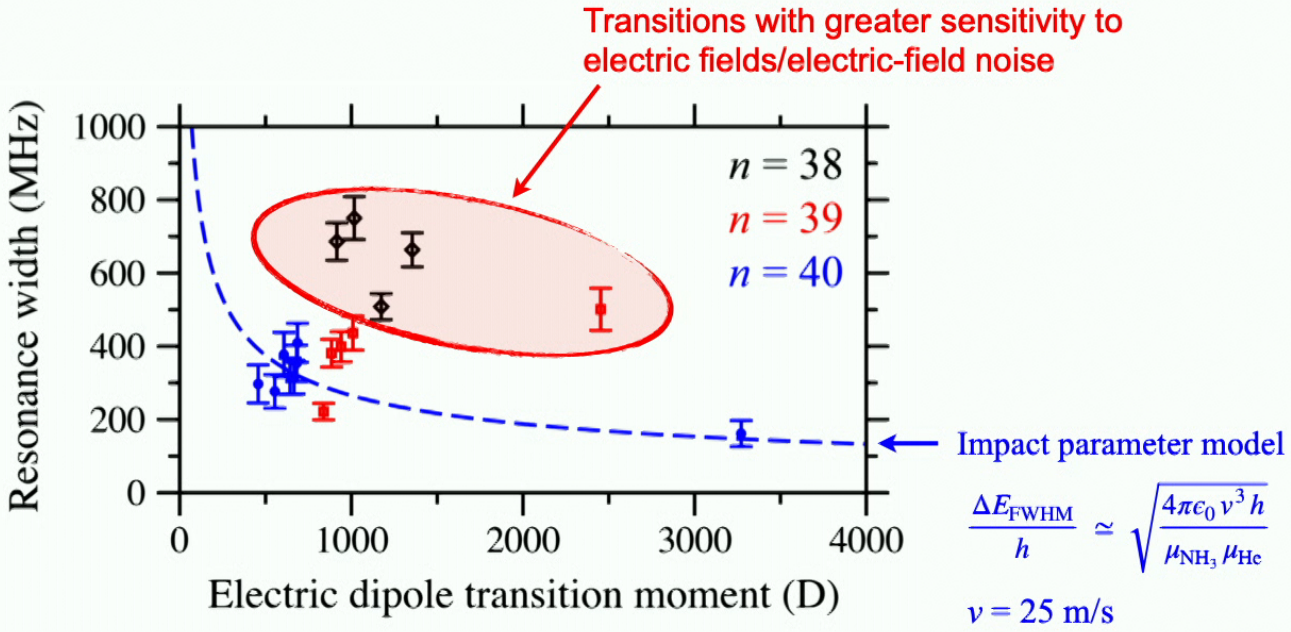
$$\frac{\Delta E_{\mathrm{FWHM}}}{h} \simeq \sqrt{\frac{4\pi\epsilon_0 v^3 h}{\mu_{\mathrm{NH_3}}\mu_{\mathrm{He}}}}$$

$v = 25$ m/s

Resonance widths

- All states/transitions studied

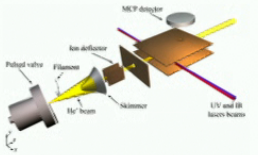
  Interaction time $T_{\mathrm{int}} = 11.5\ \mu\mathrm{s}$



Summary

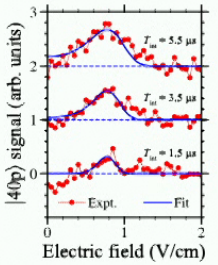
- Cold collisions of NH_3 with Rydberg He atoms

  Electric-field controlled resonant energy transfer



  $E_{kin}/k_B = 10 - 100$ mK

  NH_3 number densities 10^{10} — 10^{11} cm^{-3}



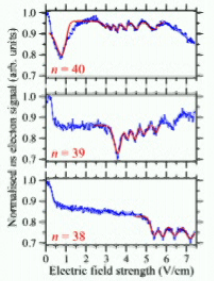
  Resonance widths affected by
  - electric dipole transition moments in Rydberg states
  - time-varying electric fields/electric field noise

- Future work

  Quantum-state-selected and oriented molecules: NH_3, ND_3, …

  Dipole-bound states of He^* and NH_3

  Effects of dipole-quadrupole interactions



  Patsch, Zeppenfeld, and Koch, *arXiv:2205.04327* (2022)