Title: Polyatomic ultralong range Rydberg molecules

Speakers: Rosario Gonzalez-Ferez

Collection: Cold Atom Molecule Interactions (CATMIN)

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Abstract: In cold and ultracold mixtures of atoms and molecules, Rydberg interactions with surrounding atoms or molecules may, under certain conditions, lead to the formation of special long-range Rydberg molecules [1,2,3]. These exotic molecules provide an excellent toolkit for manipulation and control of interatomic and atom-molecule interactions, with applications in ultracold chemistry, quantum information processing and many-body quantum physics.

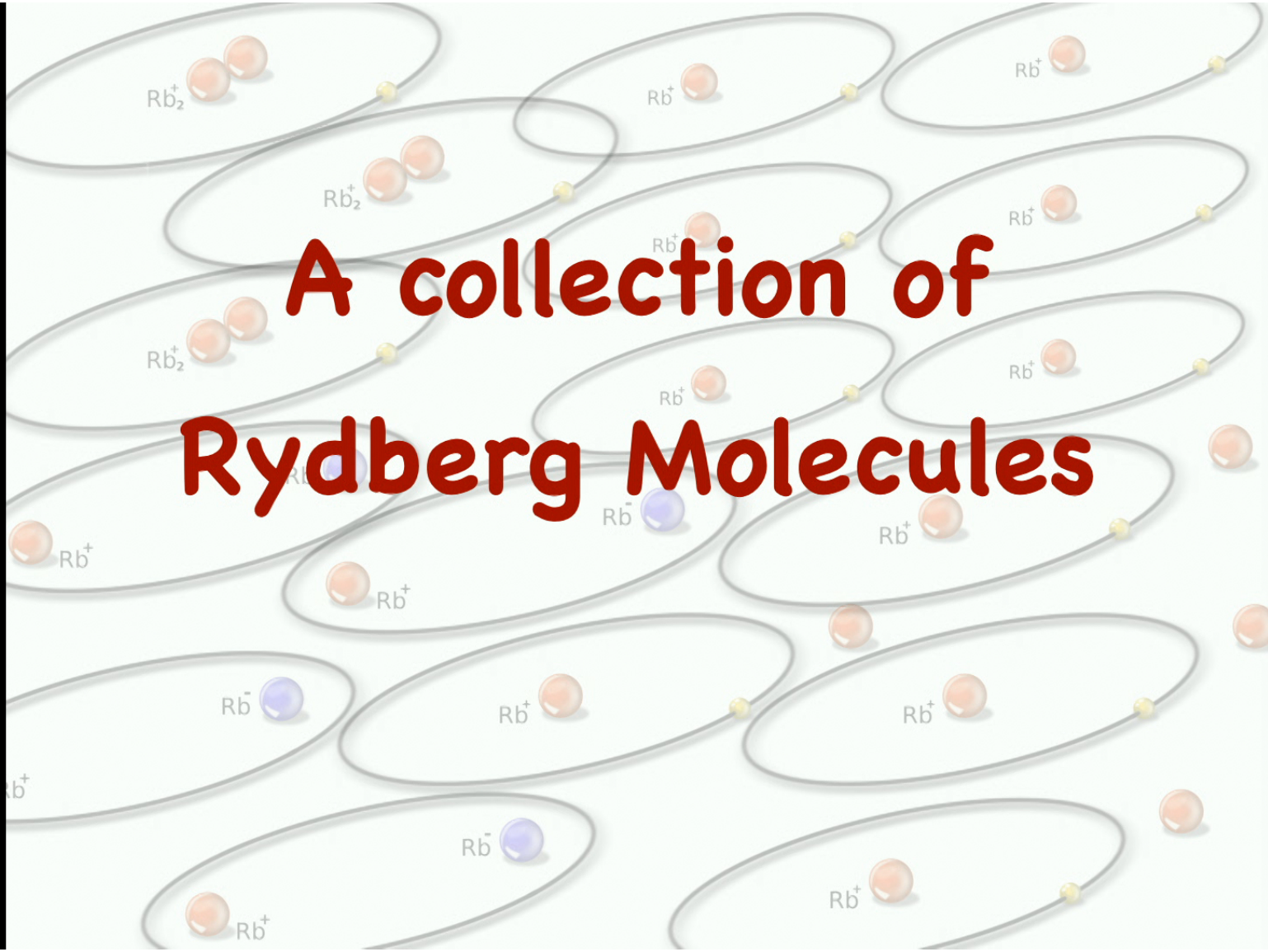
In this talk, we will discuss ultralong-range polyatomic Rydberg molecules formed when a heteronuclear diatomic molecule is bound to a Rydberg atom [3,4]. The binding mechanism appears due to anisotropic scattering of the Rydberg electron from the permanent electric dipole moment of the polar molecule. We propose an experimentally realizable scheme to produce these triatomic ultralong-range Rydberg molecules in ultracold RbCs traps, which might use the excitation of cesium or rubidium [5]. By exploiting the Rydberg electron-molecule anisotropic dipole interaction, we induce a near resonant coupling of the non-zero quantum defect Rydberg levels with the RbCs molecule in an excited rotational level. This coupling enhances the binding of the triatomic ultralong-range Rydberg molecule and produces favorable Franck-Condon factors.

References

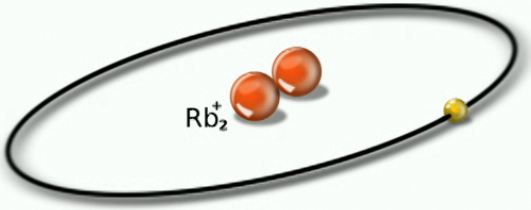
[1] C. H. Greene, A. S. Dickinson, and H. R. Sadeghpour, Phys. Rev. Lett. 85, 2458 (2000).
[2] S. T. Rittenhouse and H. R. Sadeghpour, Phys. Rev. Lett. 104, 243002 (2010).
[3] V. Bendkowsky, B. Butscher, J. Nipper, J. P. Shaffer, R. Löw, and T. Pfau, Nature 458, 1005 (2009).
[4] R. González-Férez, H. R. Sadeghpour, and P. Schmelcher, New J. Phys. 17, 013021 (2015).
[5] R. González-Férez, S.T. Rittenhouse, P. Schmelcher and H.R. Sadeghpour, J. Phys. B 53, 074002 (2020)."

Polyatomic ultralong range Rydberg molecule

Rosario González-Férez
Universidad de Granada

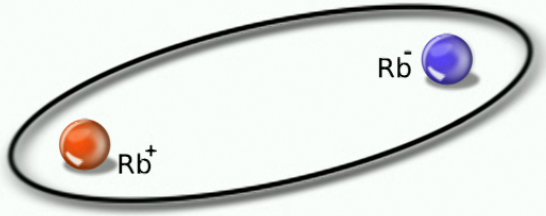

A collection of
Rydberg Molecules

Rydberg states of molecule



A highly excited electron interacting with the ionic core.

Heavy Rydberg states of molecules



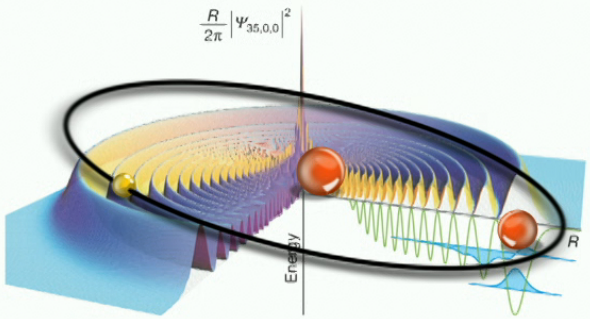
very high vibrational structure approaches a Rydberg series

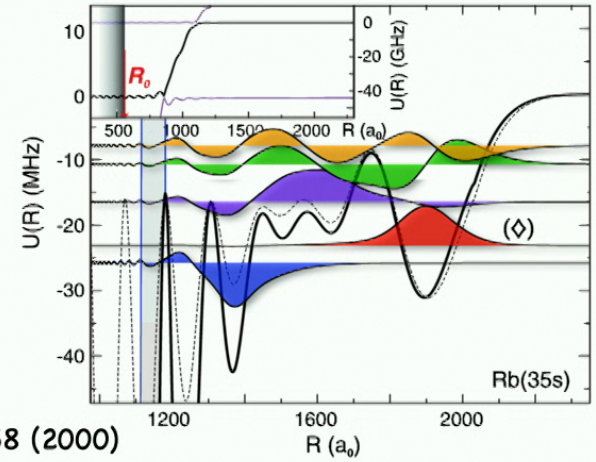
Macrodimers

Rydberg atom + Rydberg atom

Ultralong-range Rydberg molecule

Rydberg atom + ground-state atom

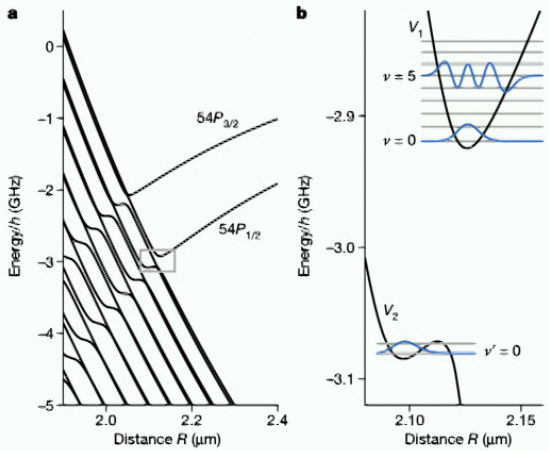




V. Bendkowsky et al, Nature **458**, 1005 (2009)

C.H. Greene, A.S. Dickinson & H.R. Sadeghpour, PRL **85**, 2458 (2000)

Long-range ion-Rydberg atom molecule

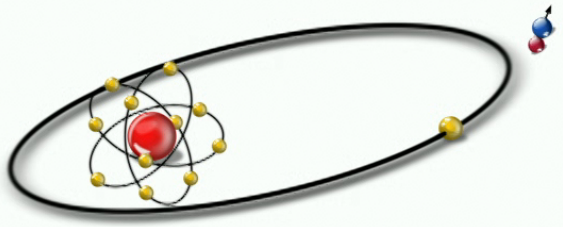


A. Duspayev et al, Phys. Rev. Research 3, 023114 (2021)

M Deiß, et al, Atoms **9**, 34 (2021)

N. Zuber et al, Nature **605**, 453 (2022)

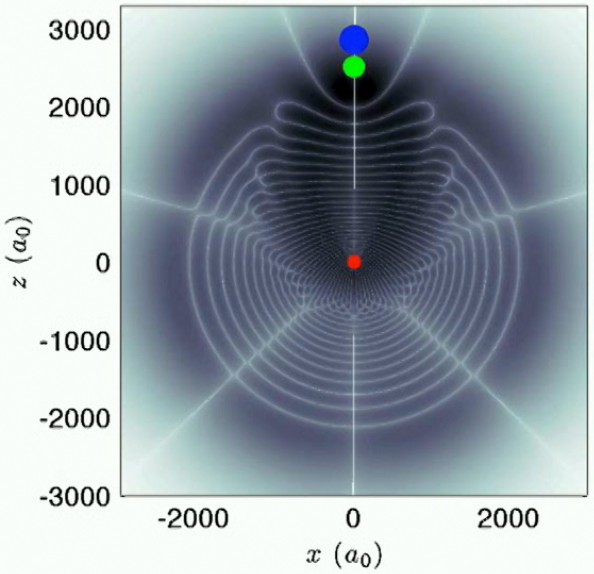
Rydberg molecules: Rydberg atom + polar molecule

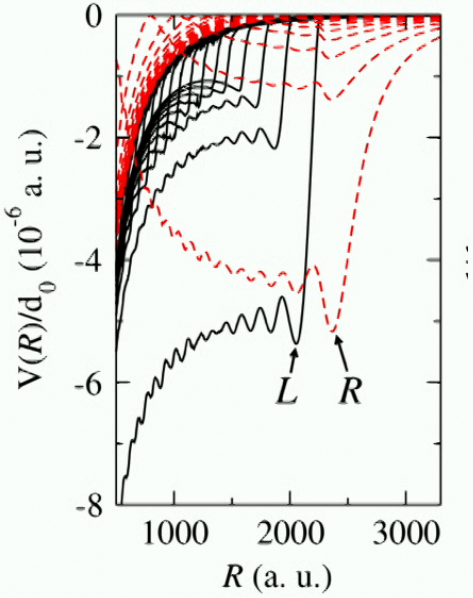


Binding mechanism: anisotropic scattering of the Rydberg electron with the dipole moment of the dimer

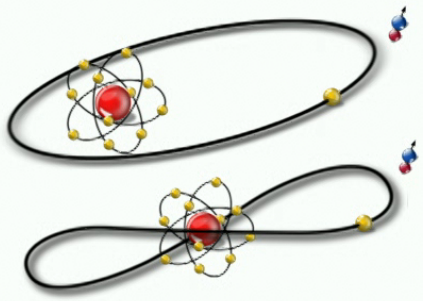
S.T. Rittenhouse & H.R. Sadeghpour, PRL (2010)

Electronic structure Rb-Λ-Doublet dimer





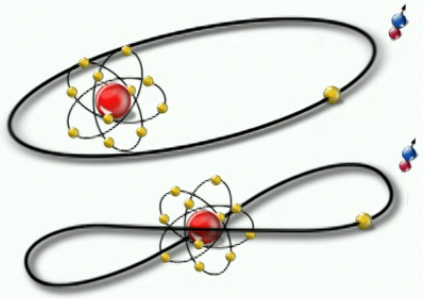
Rydberg atom + heteronuclear molecule



Core & Rydberg electron electric fields:

$$\mathbf{F}_{ryd}(\mathbf{R},\mathbf{r}) \quad = \quad e\frac{\mathbf{R}}{R^3}+\frac{\mathbf{r}-\mathbf{R}}{|\mathbf{r}-\mathbf{R}|^3}$$

Rydberg atom + heteronuclear molecule



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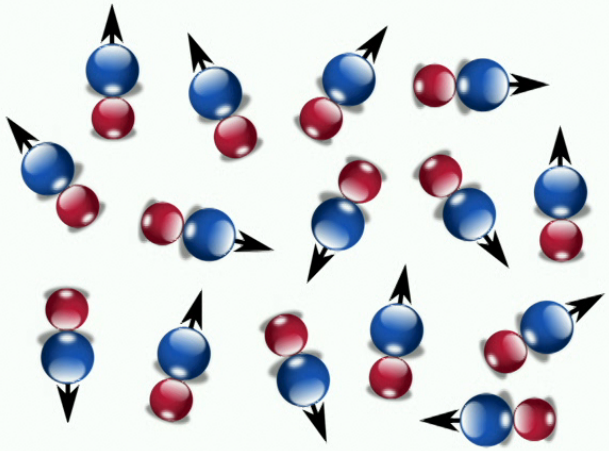
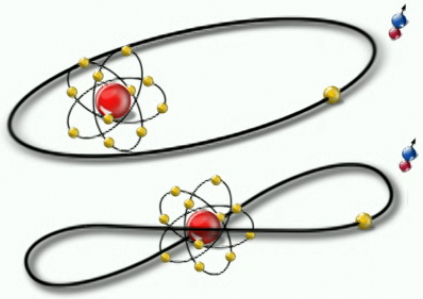
$$\mathbf{F}_{ryd}(\mathbf{R},\mathbf{r}) = e\frac{\mathbf{R}}{R^3} + \frac{\mathbf{r}-\mathbf{R}}{|\mathbf{r}-\mathbf{R}|^3}$$

Field-free

$$H = B\mathbf{N}^2$$



Rydberg atom + heteronuclear molecule



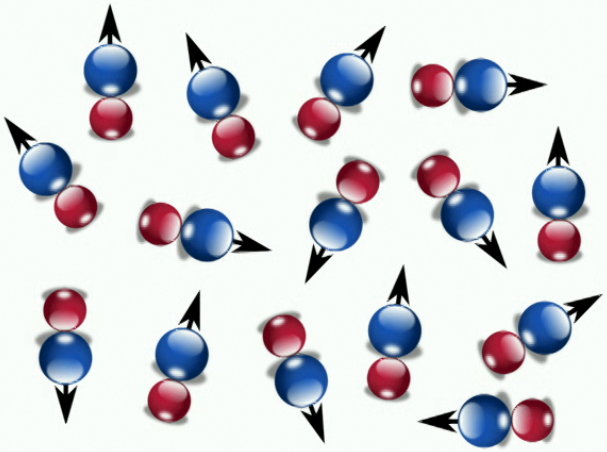
Core & Rydberg electron electric fields:

$$\mathbf{F}_{ryd}(\mathbf{R},\mathbf{r}) = e\frac{\mathbf{R}}{R^3} + \frac{\mathbf{r}-\mathbf{R}}{|\mathbf{r}-\mathbf{R}|^3}$$

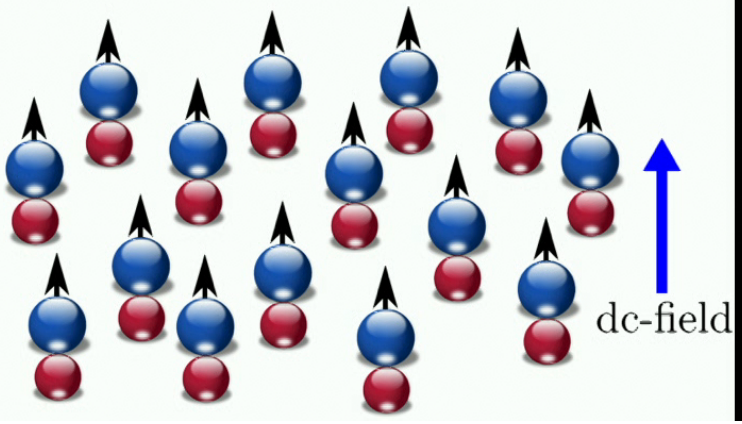
Field-free → dc electric field

Orientation: the molecule-fixed axes are confined and the dipole moment has a well-defined direction

$$H = B\mathbf{N}^2$$



$$H = B\mathbf{N}^2 - \mathbf{d}\cdot\mathbf{F}$$



Our Rydberg molecule

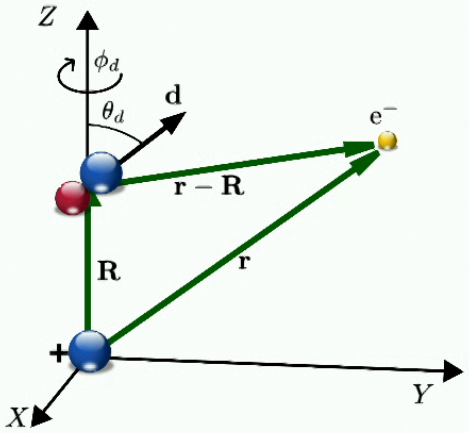
The total Hamiltonian $H = H_A + H_{mol} + V_{n,e}$

The Rydberg electron Hamiltonian

$$H_A = -\frac{\hbar^2}{2m_e}\nabla_r^2 + V_l(r)$$

The Hamiltonian of the molecule

$$H_{mol} = B\mathbf{N}^2 - \mathbf{d}\cdot\mathbf{F}(\mathbf{r},\mathbf{R})$$



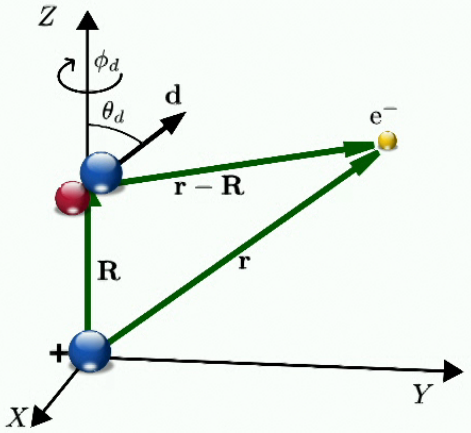
$$\mathbf{F}(\mathbf{r},\mathbf{R}) = e\frac{\mathbf{R}}{R^3} + e\frac{\mathbf{r}-\mathbf{R}}{|\mathbf{r}-\mathbf{R}|^3}$$

Fermi-Teller critical dipole: d<1.63D.

The electron-RbCS scattering $V_{n,e}(\mathbf{r},\mathbf{R}) = 2\pi A_s(k)\delta(\mathbf{r}-\mathbf{R})$

Our Rydberg molecule

The total Hamiltonian $H = H_A + H_{mol} + V_{n,e}$



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$$\mathbf{F}(\mathbf{r},\mathbf{R}) = e\frac{\mathbf{R}}{R^3} + e\frac{\mathbf{r}-\mathbf{R}}{|\mathbf{r}-\mathbf{R}|^3}$$

Fermi-Teller critical dipole: d<1.63D.

The electron-RbCS scattering $V_{n,e}(\mathbf{r},\mathbf{R}) = 2\pi A_s(k)\delta(\mathbf{r}-\mathbf{R})$

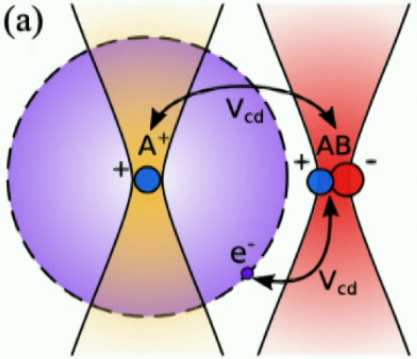
By a basis set expansion in terms of the coupled basis

$$\psi_{nlNJM_J}(\mathbf{r},\Omega_d) = \sum_{m_l=-l}^{m_l=l}\sum_{M_N=-N}^{M_N=N}\psi_{nlm}(\mathbf{r})\,Y_{NM_N}(\Omega_d)\langle lm_l NM_N|JM_J\rangle$$

- $Y_{NM_N}(\Omega_d)$ the field-free rotational wave function of KRb
- $\psi_{nlm}(\mathbf{r})$ the Rydberg electron wave function

Experimental setup

Ultracold molecules:
KRb (K.K Ni, Harvard)
RbCs (S. Cornish, Durham)



Experimental setup

RbCs rotational states

B=0.5 GHz
d=1.25D

Cesium Rydberg states

State	$E_{n,l} - E_{70,l\geq 4}$ [GHz]
$\mathrm{Cs}(n = 70, l \geq 4)$	0.000
$\mathrm{Cs}(70f)$	-0.642
$\mathrm{Cs}(74s)$	-0.948
$\mathrm{Cs}(72d)$	-9.124
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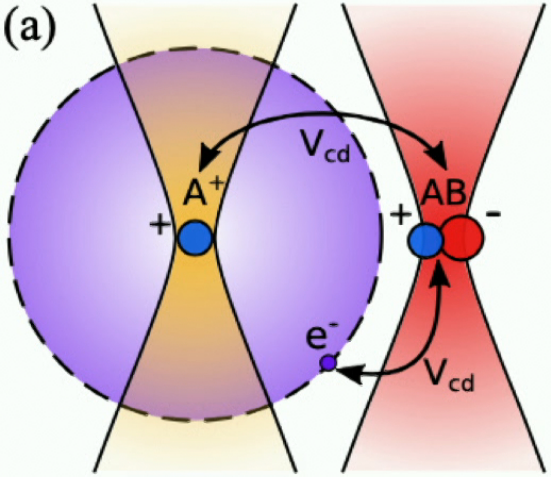
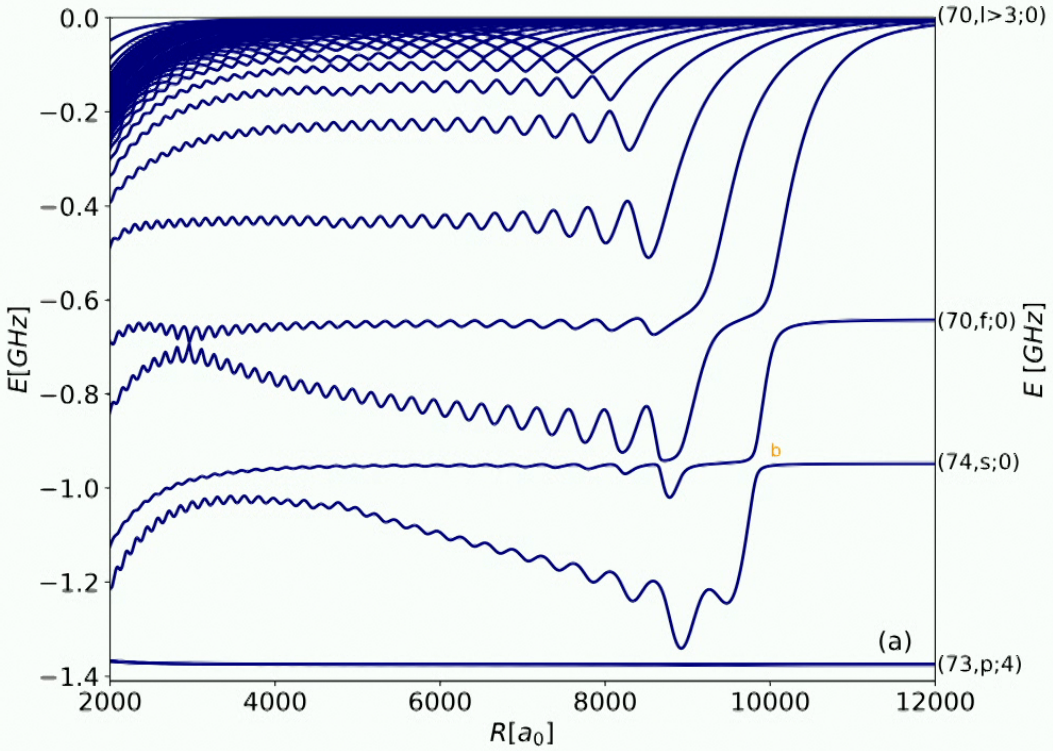


Rydberg molecule: Cs-RbCs

Energy limit for R>>1

$$E_{nl} + BN(N+1)$$

Electronic structure of Cs(n=70)-RbCS



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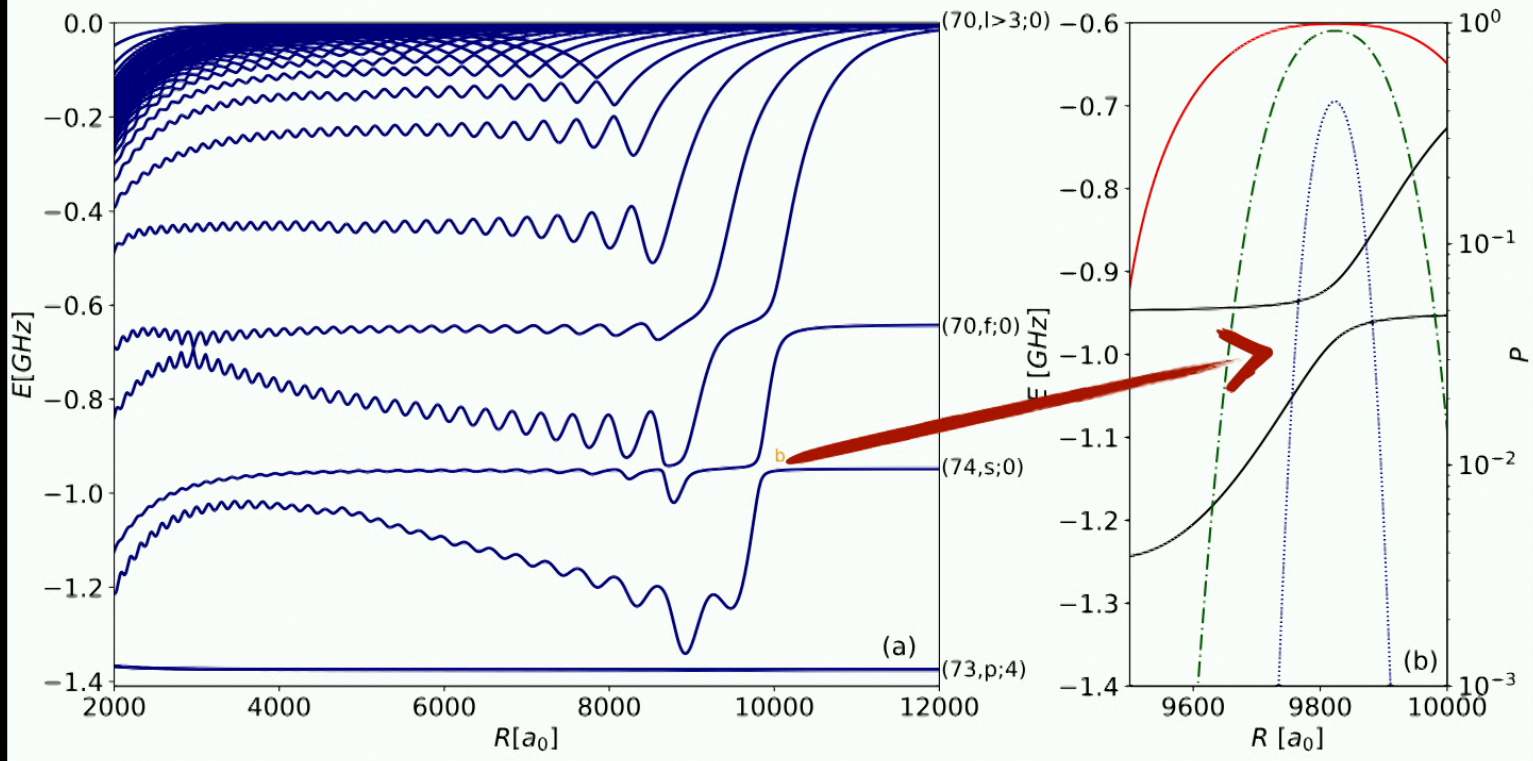

$v = 2.2 \times 10^2, 2.2 \times 10^3, 2.2 \times 10^4 \text{ cms}^{-1}$

$$P = e^{-\frac{\pi \Delta E}{4\hbar g v}}$$

$$g = |\langle \Phi_i | \frac{d}{dR} | \Phi_j \rangle|$$

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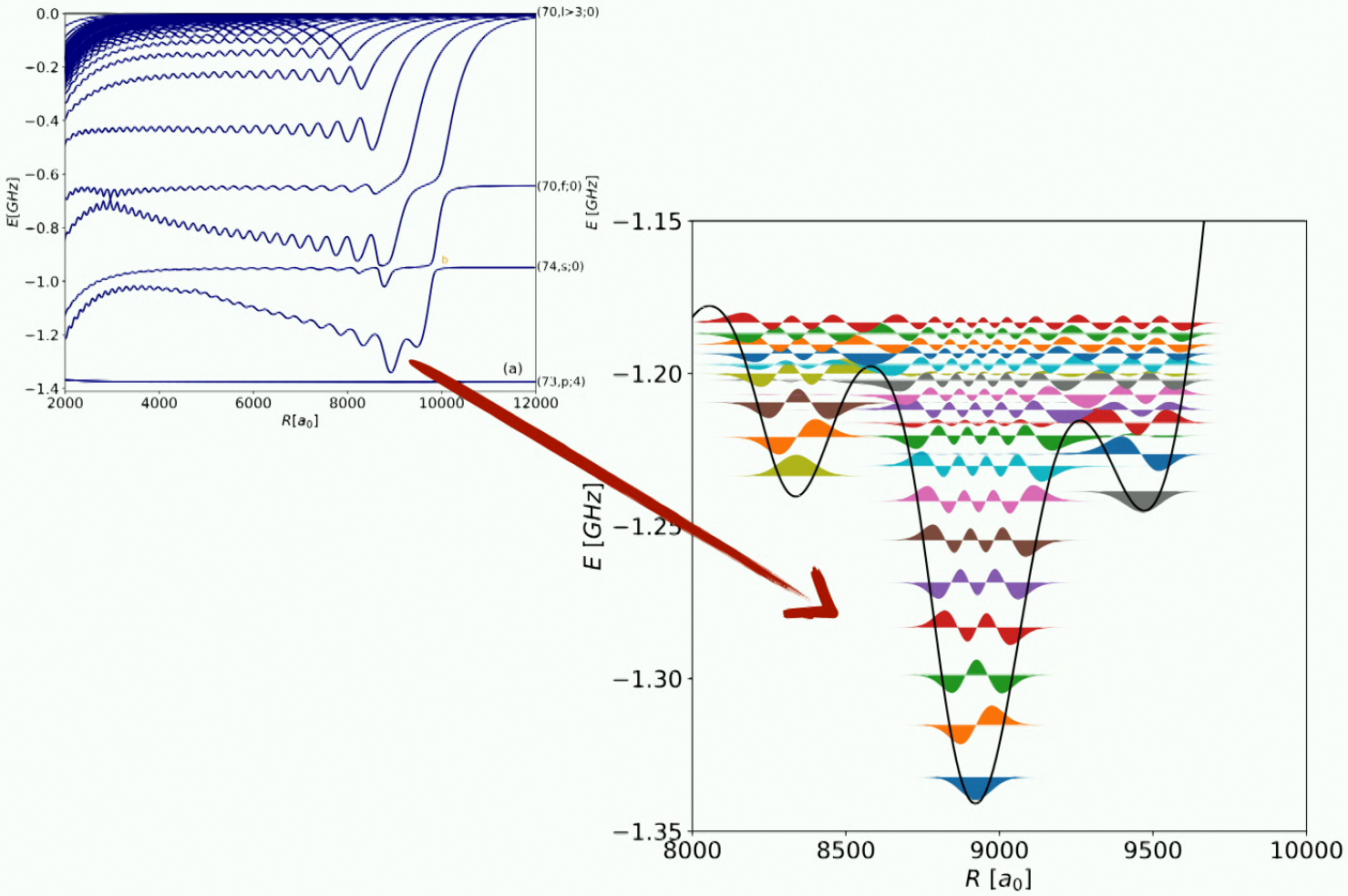

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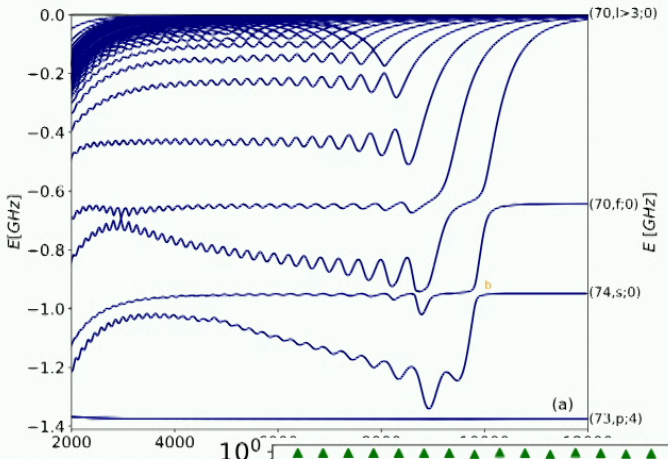

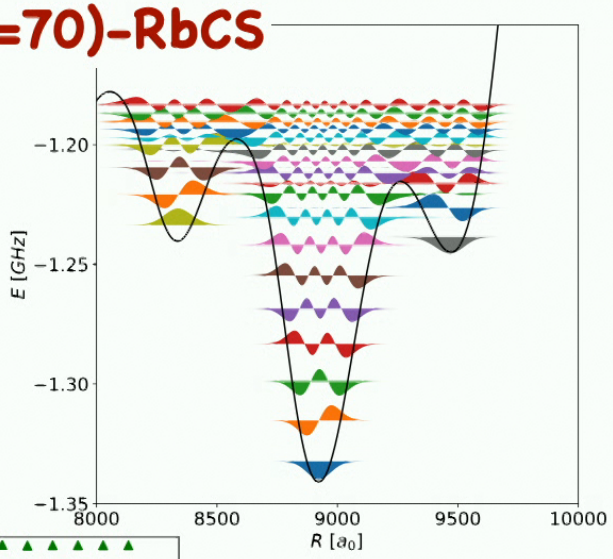

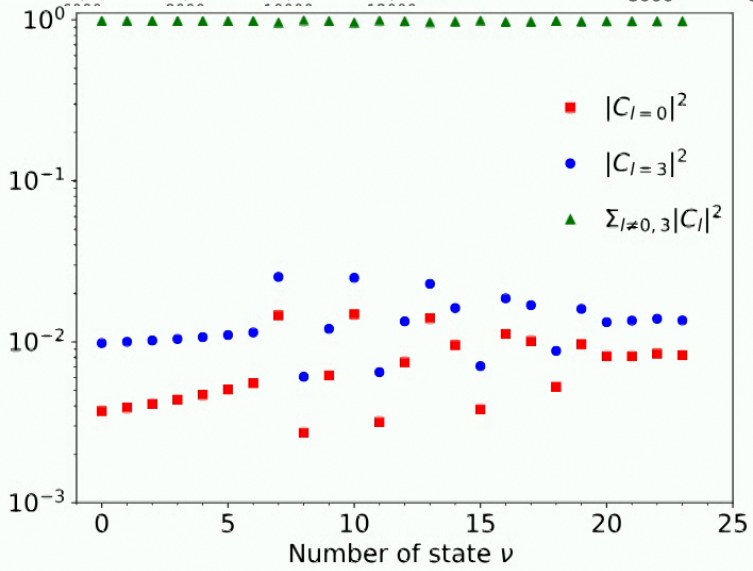

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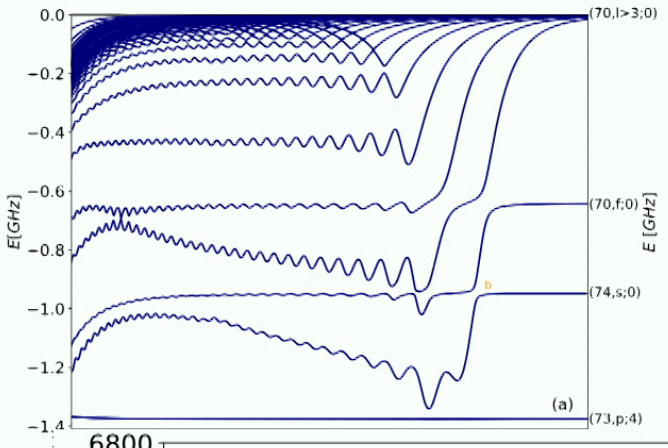

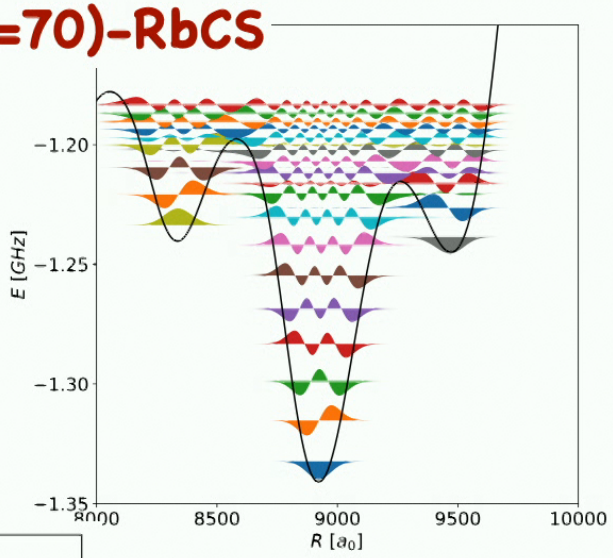

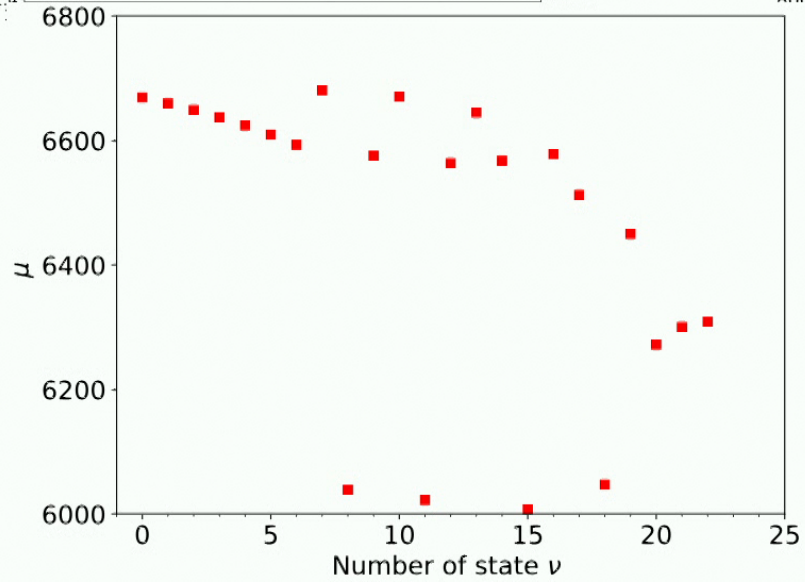

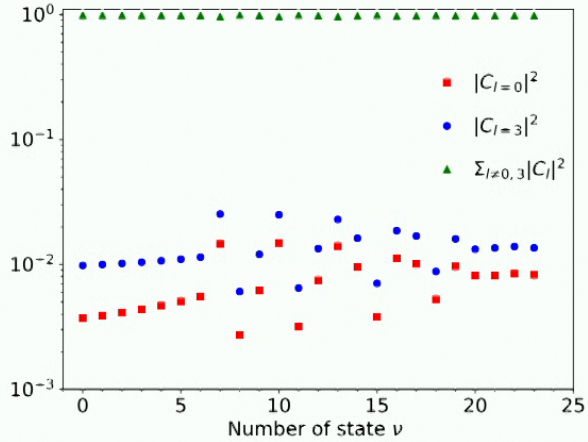

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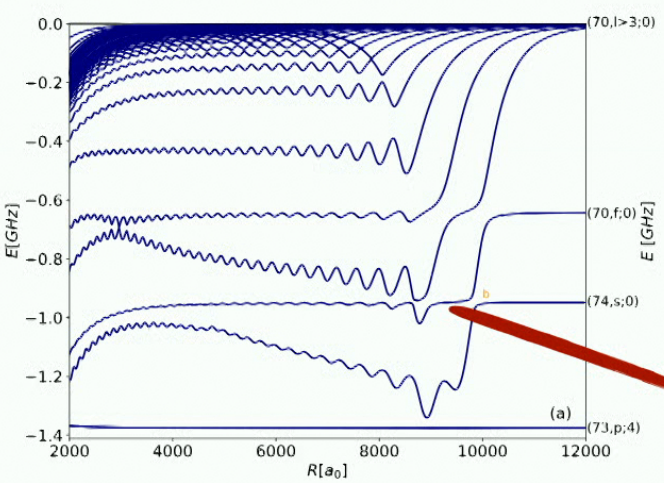

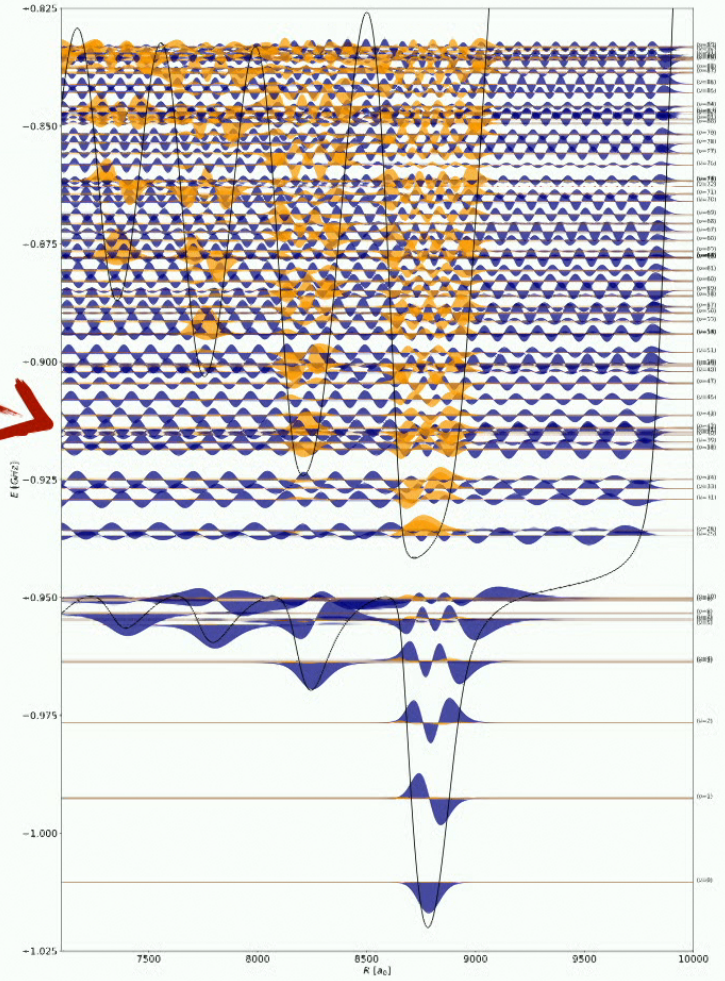

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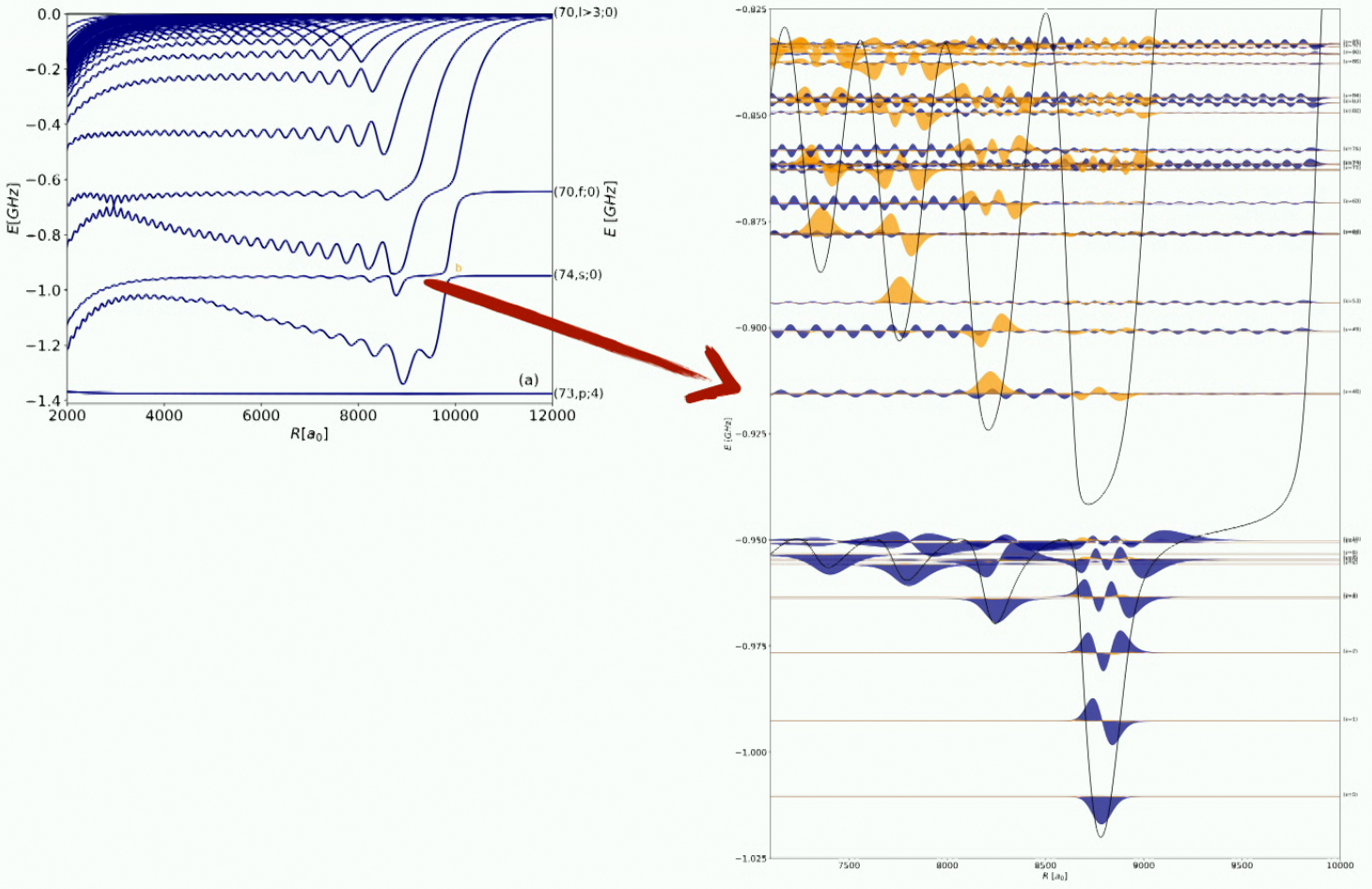

Experimental setup

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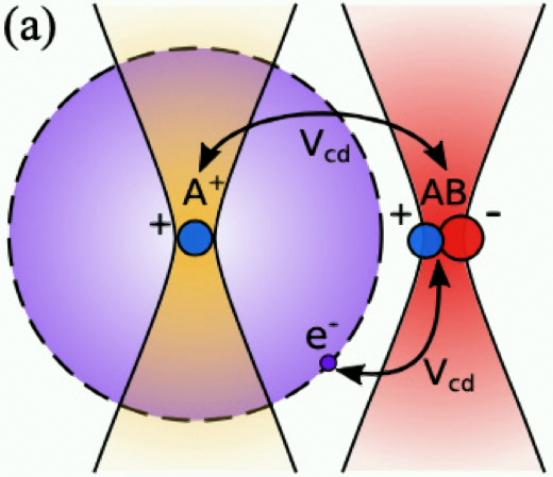

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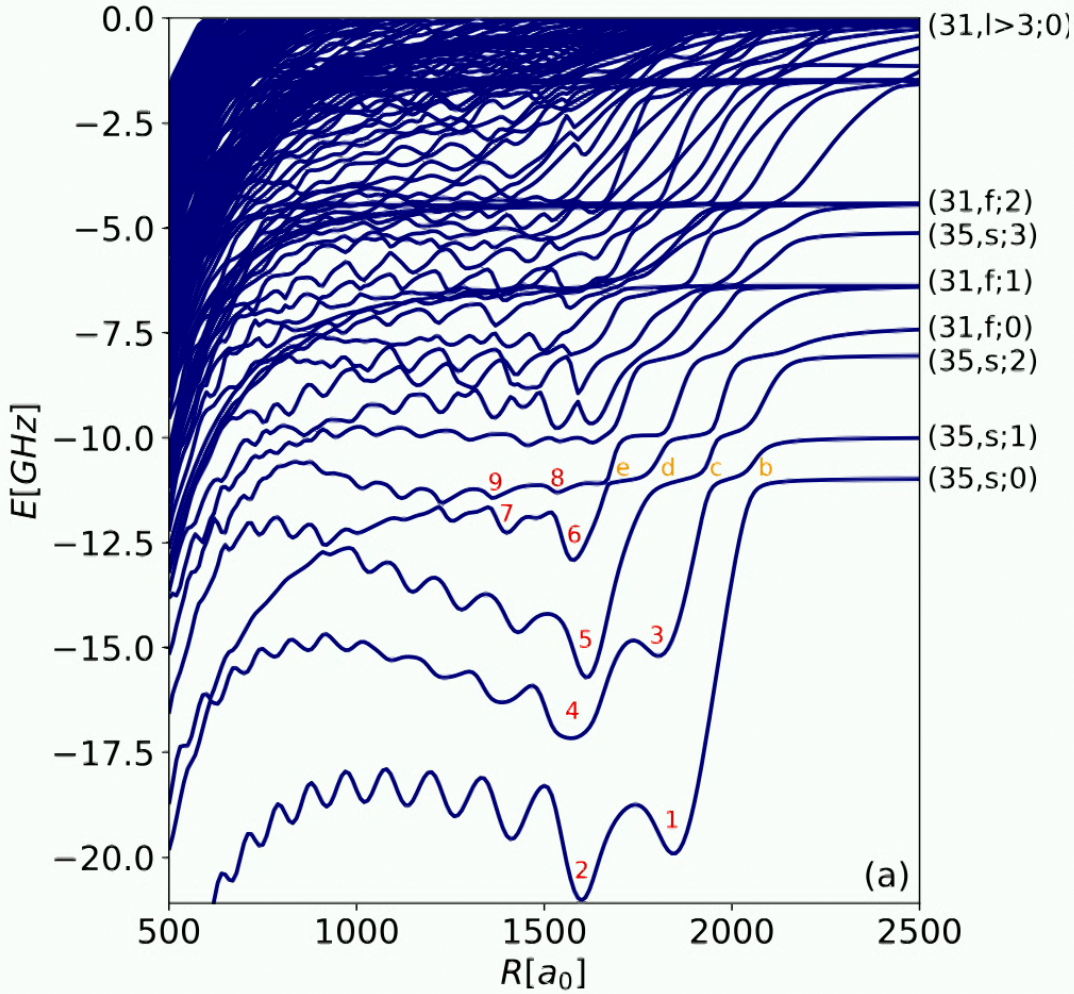
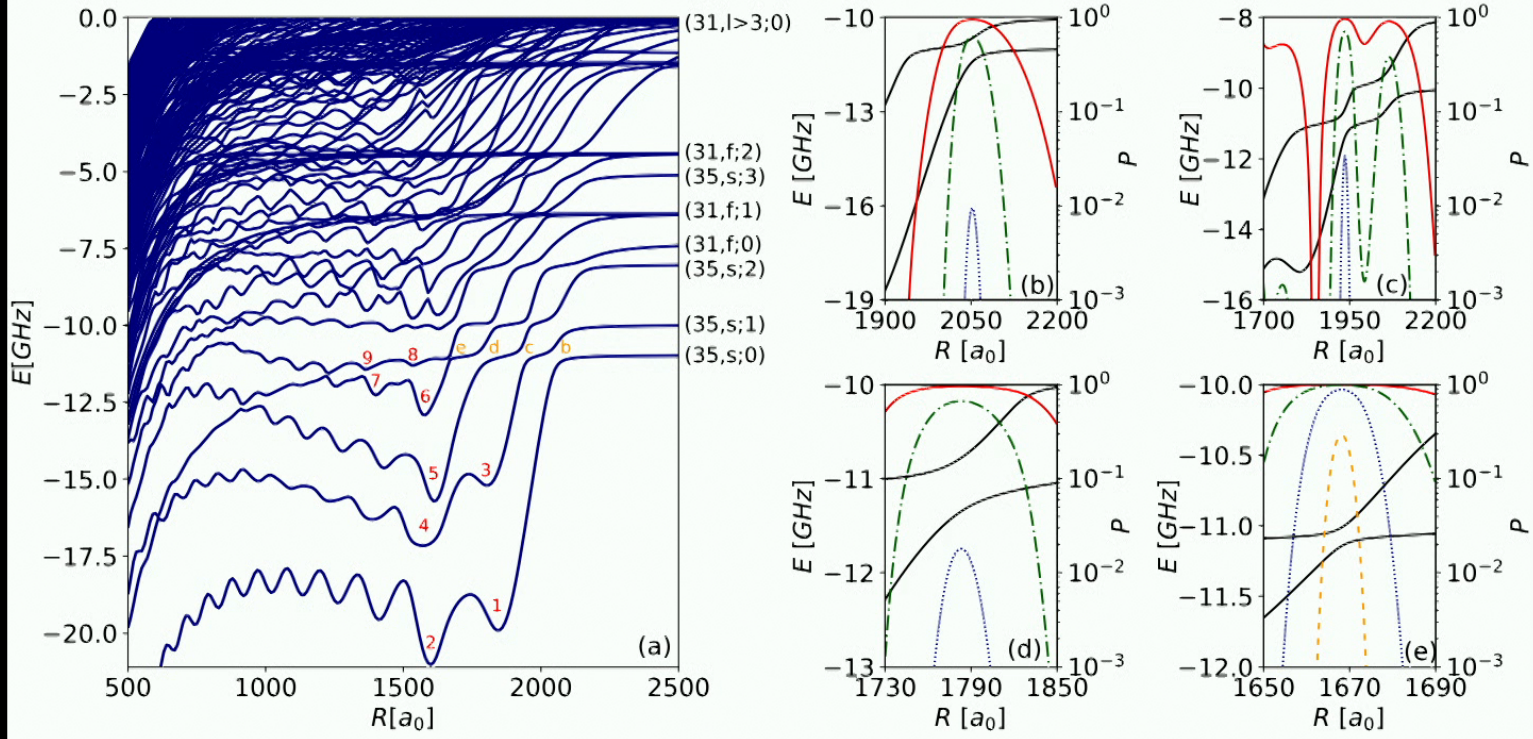
$\mathrm{Cs}(74s)$	-0.948
$\mathrm{Cs}(72d)$	-9.124
$\mathrm{Cs}(73p)$	-11.175

State	$E_{n,l} - E_{31,l\geq 4}$ [GHz]
$\mathrm{Cs}(n = 31, l \geq 4)$	0.000
$\mathrm{Cs}(31f)$	-7.359
$\mathrm{Cs}(35s)$	-10.977
$\mathrm{Cs}(33d)$	-106.468
$\mathrm{Cs}(34p)$	-130.787

Electronic structure of Cs(n=31,l>3)-RbCS



Electronic structure of Cs(n=31,l>3)-RbCS



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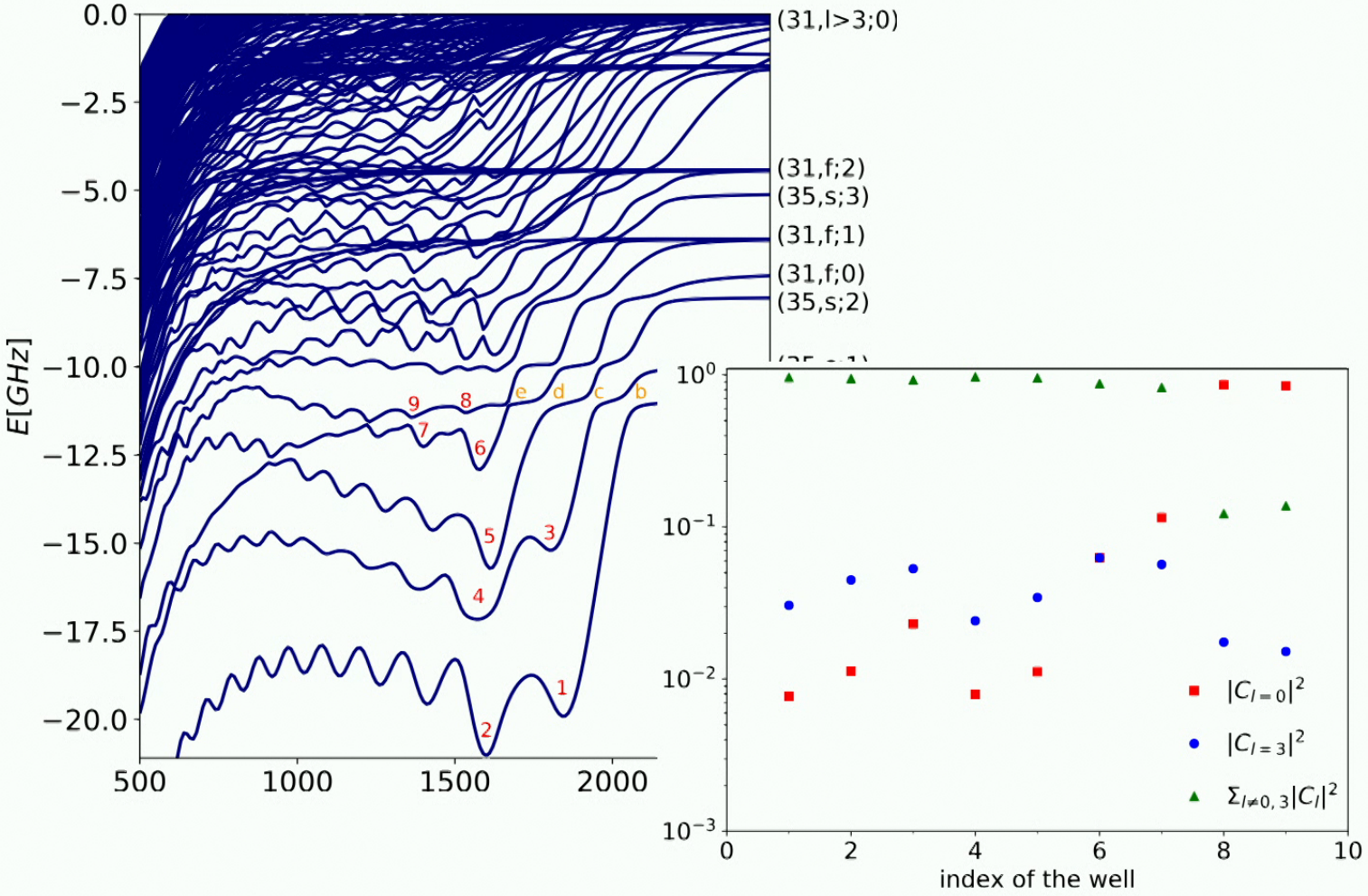

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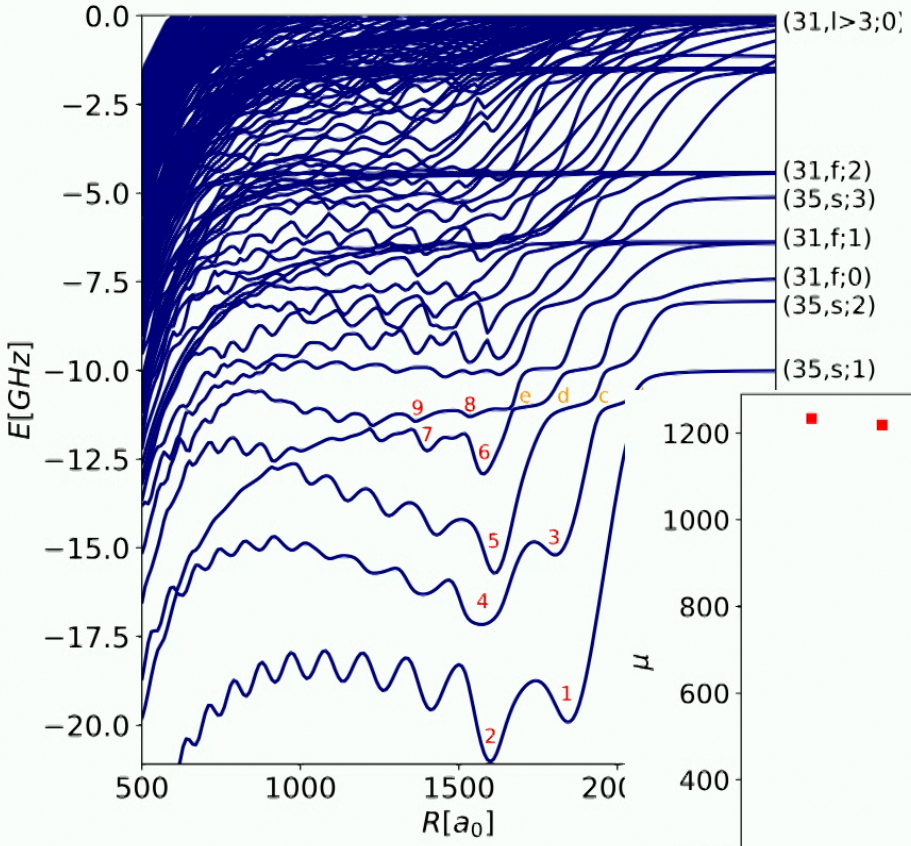

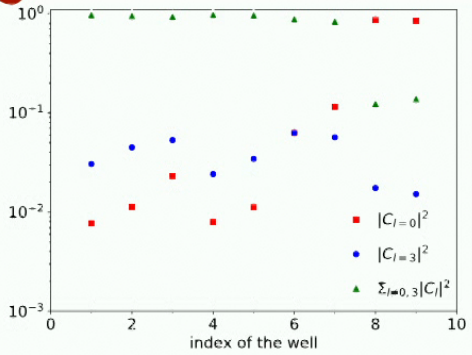

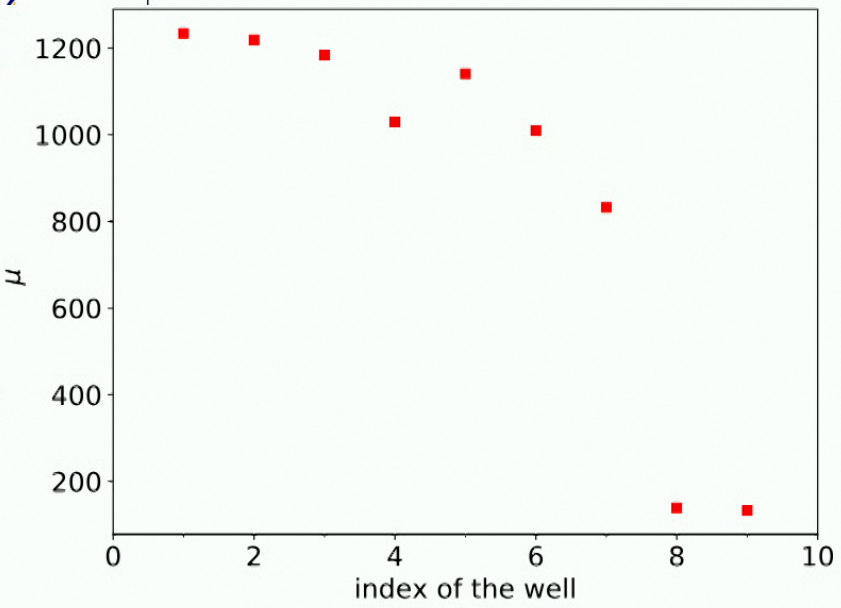

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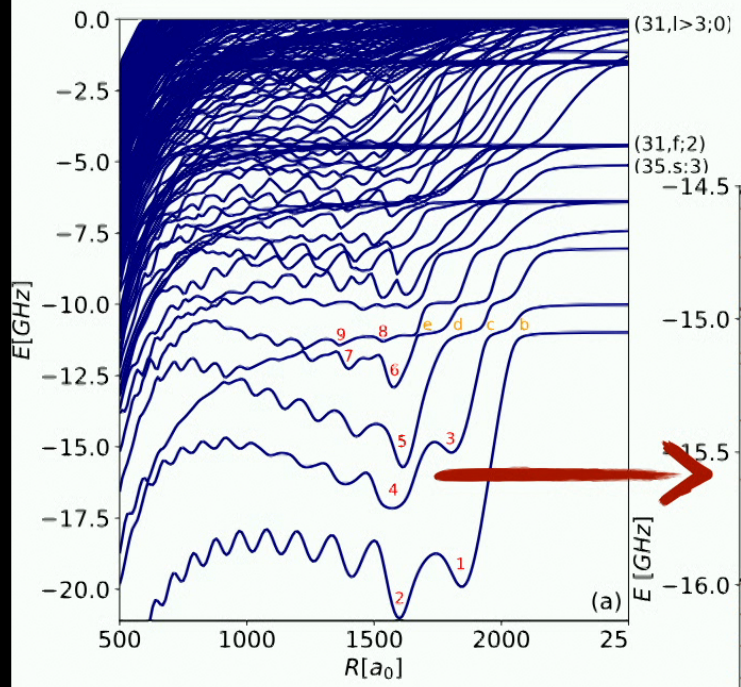
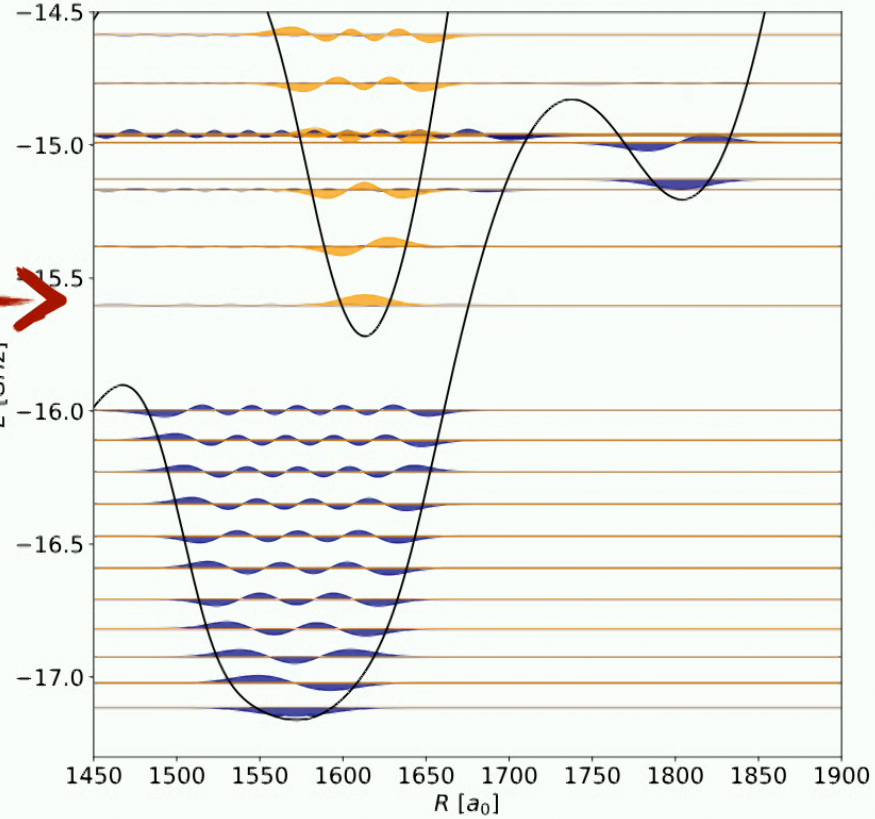
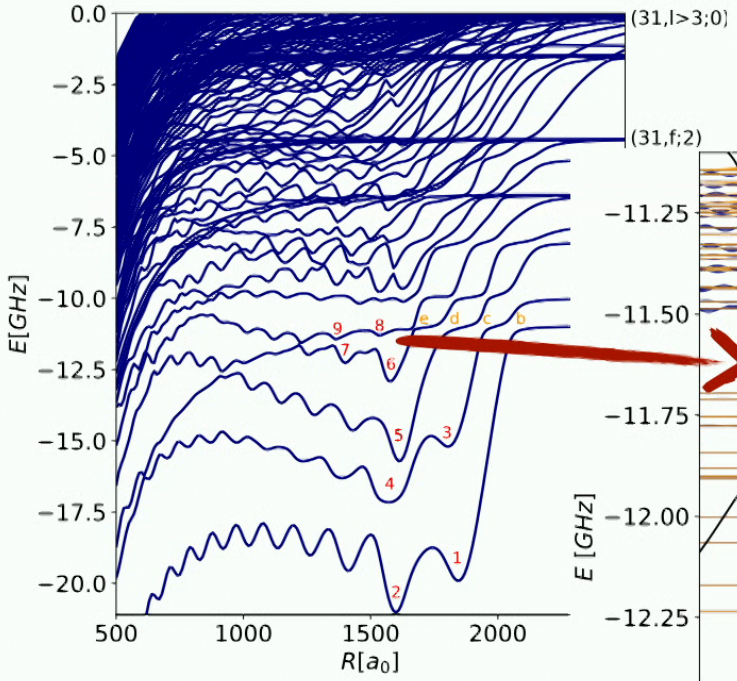


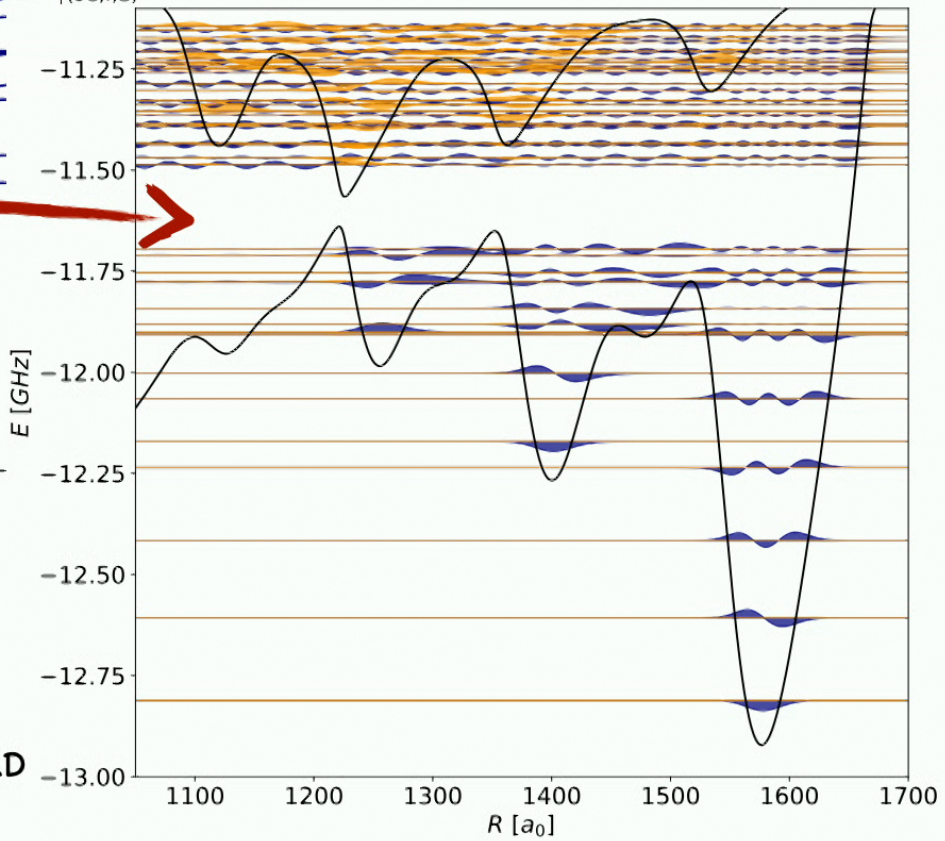


Large vibrational splitting

Dipole moments ~1kD

Electronic structure of Cs(n=31,l>3)-RbCS





Large vibrational splitting

Dipole moments ~1kD to ~0.1kD

$NO(^2\Pi)$- NO(nl) bi-molecule

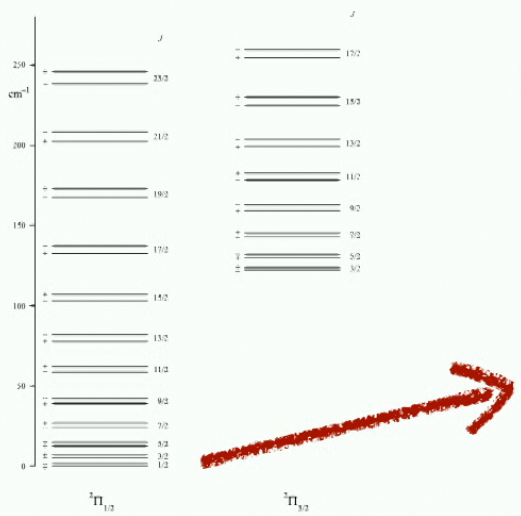
In March 2020 (!), we tried to find out if such molecules could exist

Janine Shertzer

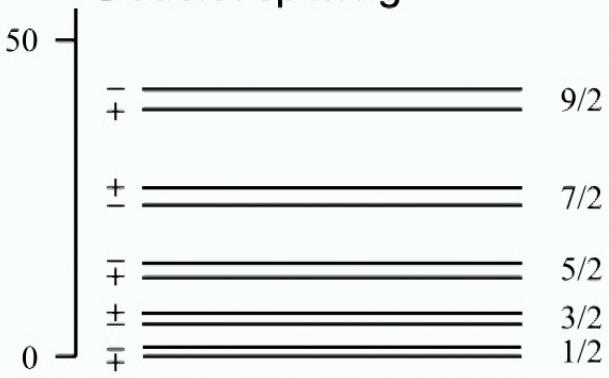
Hossein R. Sadeghpour

$NO(^2\Pi)$

A simple (two-level) Λ-doublet molecule!



Doublet splitting

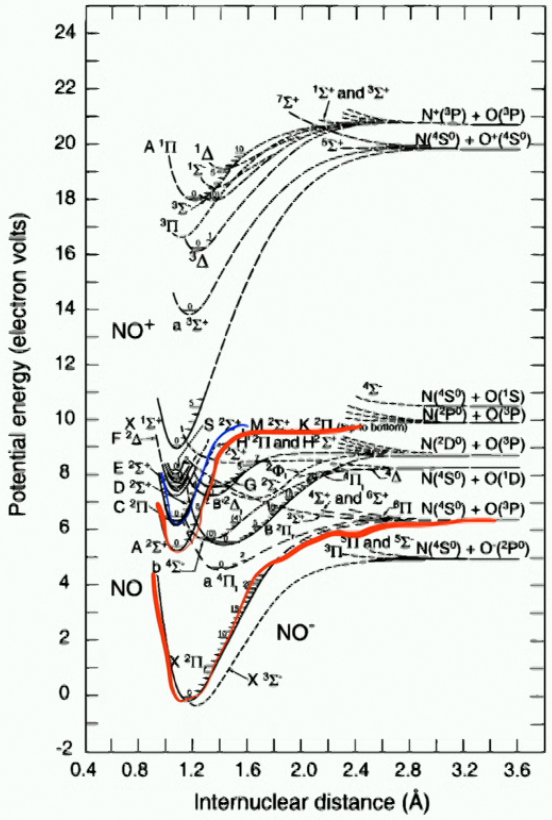


$$H_{NO} = E_+|+\rangle\langle+| + E_-|-\rangle\langle-|$$

$B = 50.848$ GHz

$E_\Lambda = 205.95$ MHz

d= 0.15 D



NO*

$A^2\Sigma^+$ (l=0 94%, l=1 1%, l=2 5%)

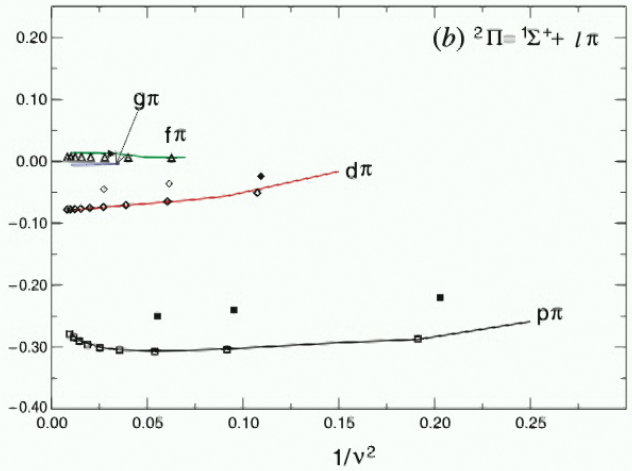
$np, nf\ (\Delta N = 0,2)\ \ excitations$

$ng\ near\ NO^+(X^1\Sigma)$

NO(ng) states are longest lived; fraction of μs (R. Field 2022)

NO*

NO quantum defects



M.Hiyama & M. S. Child JPB **35,** 1337 (2002)

J. Phys. B: At. Mol. Opt. Phys. **29** (1996) 3747–3761. Printed in the UK

R-matrix calculation of the bound and continuum states of the e^-–NO^+ system

I Rabadán and J Tennyson†
Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

$$E_{nl} = \frac{Ry}{(n-\mu_l)^2}$$

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS
J. Phys. B: At. Mol. Opt. Phys. **36** (2003) 4547–4559 PII: S0953-4075(03)66009-8

***Ab initio* R-matrix/multi-channel quantum defect theory study of nitric oxide: II. Analysis of valence/Rydberg interactions**

Miyabi Hiyama and Mark S Child
Physical and Theoretical Chemistry Laboratory, South Parks Road, Oxford OX1 3QZ, UK

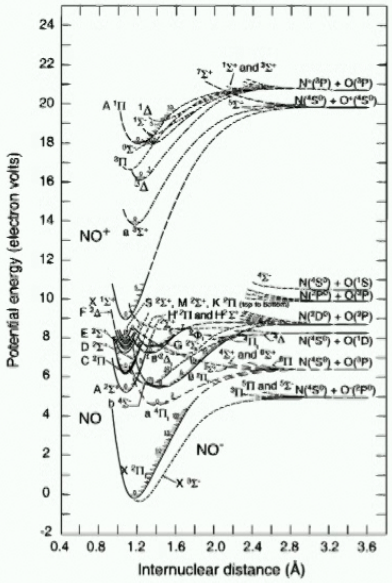
We're interested in high-l (longer lifetimes), and not the core penetrating l states

$$H = H_{\text{Ryd}} + H_{\text{NO}} + H_{e\text{-NO}}$$

$$H_{\text{NO}} = E_{+}|+\rangle\langle+| + E_{-}|-\rangle\langle-|$$

NO Rydberg molecular potent

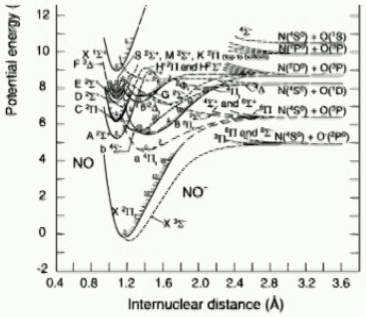
$$U_{n\Lambda}(R_{\text{NO}}) = U^{+}(R_{\text{NO}}) - \frac{1}{2(n-\mu_{\Lambda})^2}$$



NO($X^2\Pi$) has electron affinity (EA)

$$EA = NO^-(^3\Sigma, \nu = 0, J = 0) - NO(^2\Pi, \nu = 0, J = 0) = 33\ meV$$

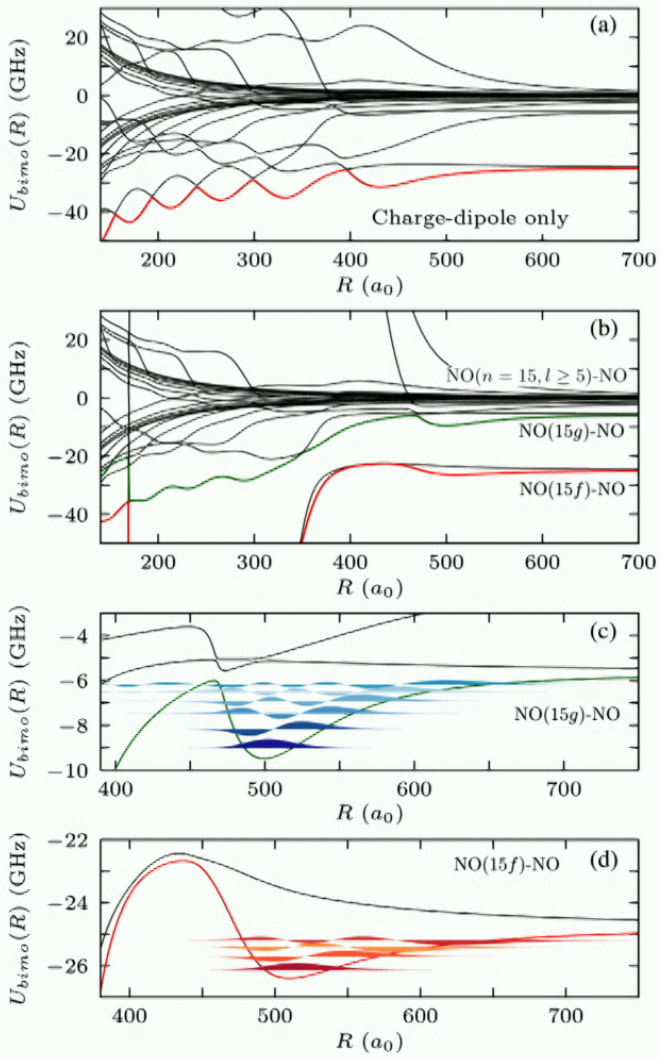
$$a_S(0) = -\tan(\delta_S(k))/k$$ Scattering length



resonance in elastic channel

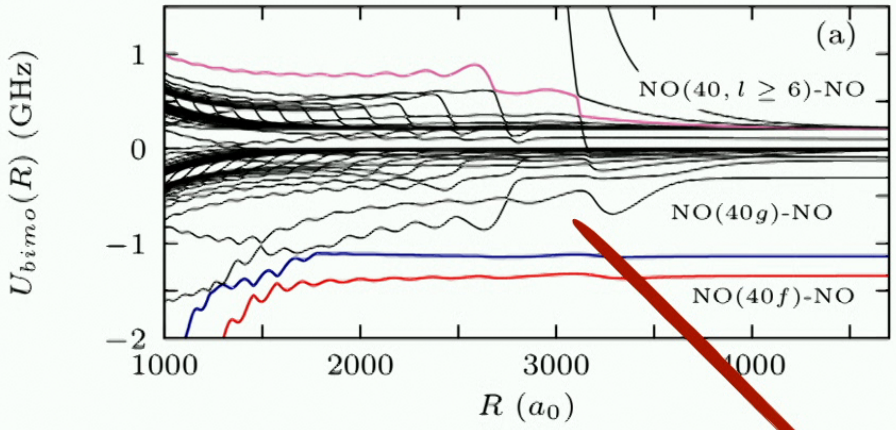
$$\{e^- + \mathrm{NO}[\mathrm{X}^2\Pi_{1/2}(\nu = 0)] \rightarrow \mathrm{NO}^-[\mathrm{X}^3\Sigma(\nu' = 1)]\}$$ 100 meV

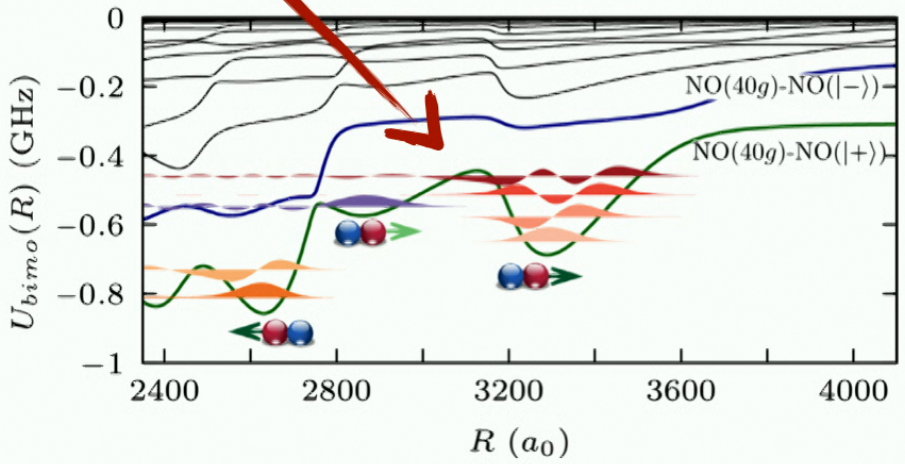
$$a_P^3(0) = -\tan(\delta_P(k))/k^3$$ Scattering volume



$NO(nf, ng) - NO(X^2\Pi_{1/2})$

$NO(nf, ng) - NO(X^2\Pi_{1/2})$



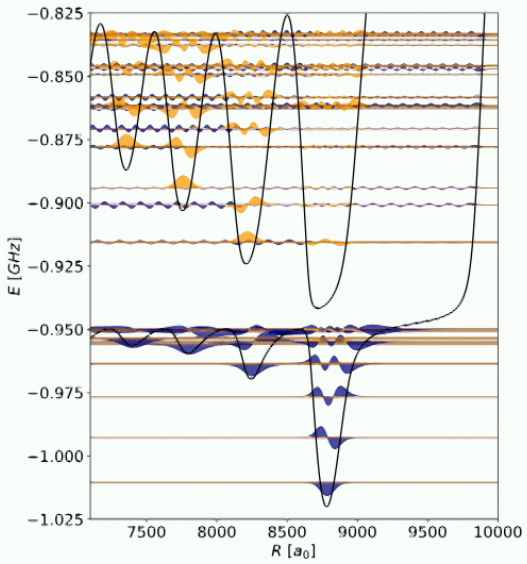


$B_{NONO^*} = 0.1 - 1\ MHz$

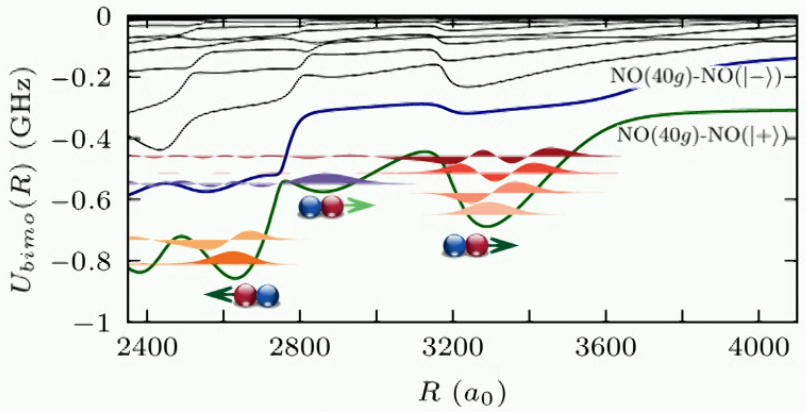
Conclusions

Rydberg molecules:
Rb-KRb (K.K Ni, Harvard)
Cs-RbCs (S. Cornish, Durham)

RGF, et al JPB **53**, 074002 (2020),
PRA **96**, 052509 (2017),
New J. Phys. **17**, 013021 (2015)

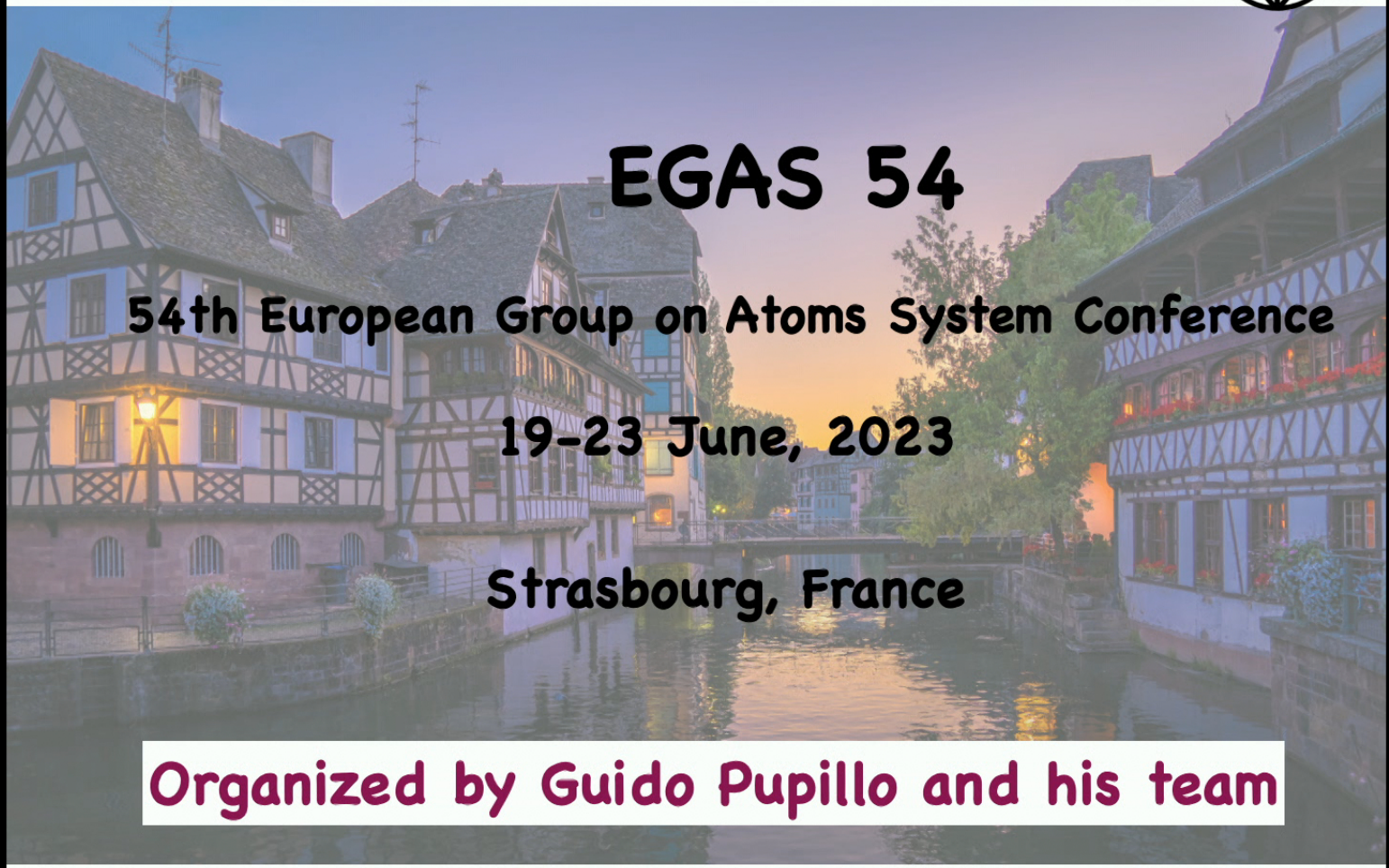


Rydberg bimolecules: NO-NO
T. Pfau & H. Kübler, Stuttgart



RGF, J. Shertzer, H.R. Sadeghpour, PRL 126, 043401 (2021)

EGAS



EGAS 54

54th European Group on Atoms System Conference

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Organized by Guido Pupillo and his team


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