Université Claude Bernard Lyon 1

INSTITUT LUMIÈRE MATIÈRE

cnrs dépasser les frontières

WAVEFUNCTION MICROSCOPY: SIMPLE ATOMS UNDER MAGNIFICATION

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EGAS

46th Conference of the European Group on Atomic Systems

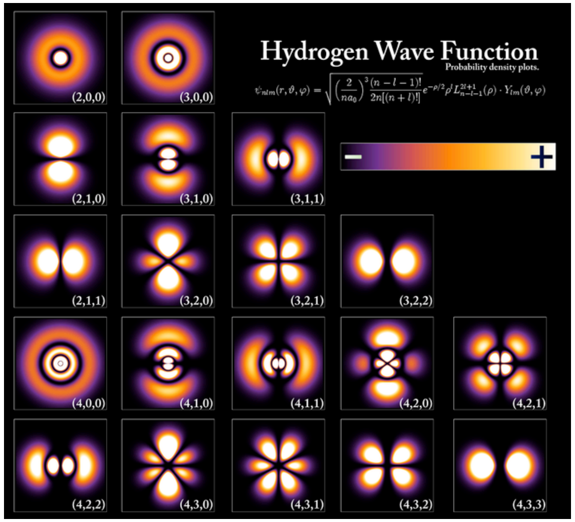
EPS

LILLE - FRANCE

July 1-4, 2014

GENERAL GOAL

WAVEFUNCTION: KEY CONCEPT FOR DESCRIBING ATOMS AND MOLECULES

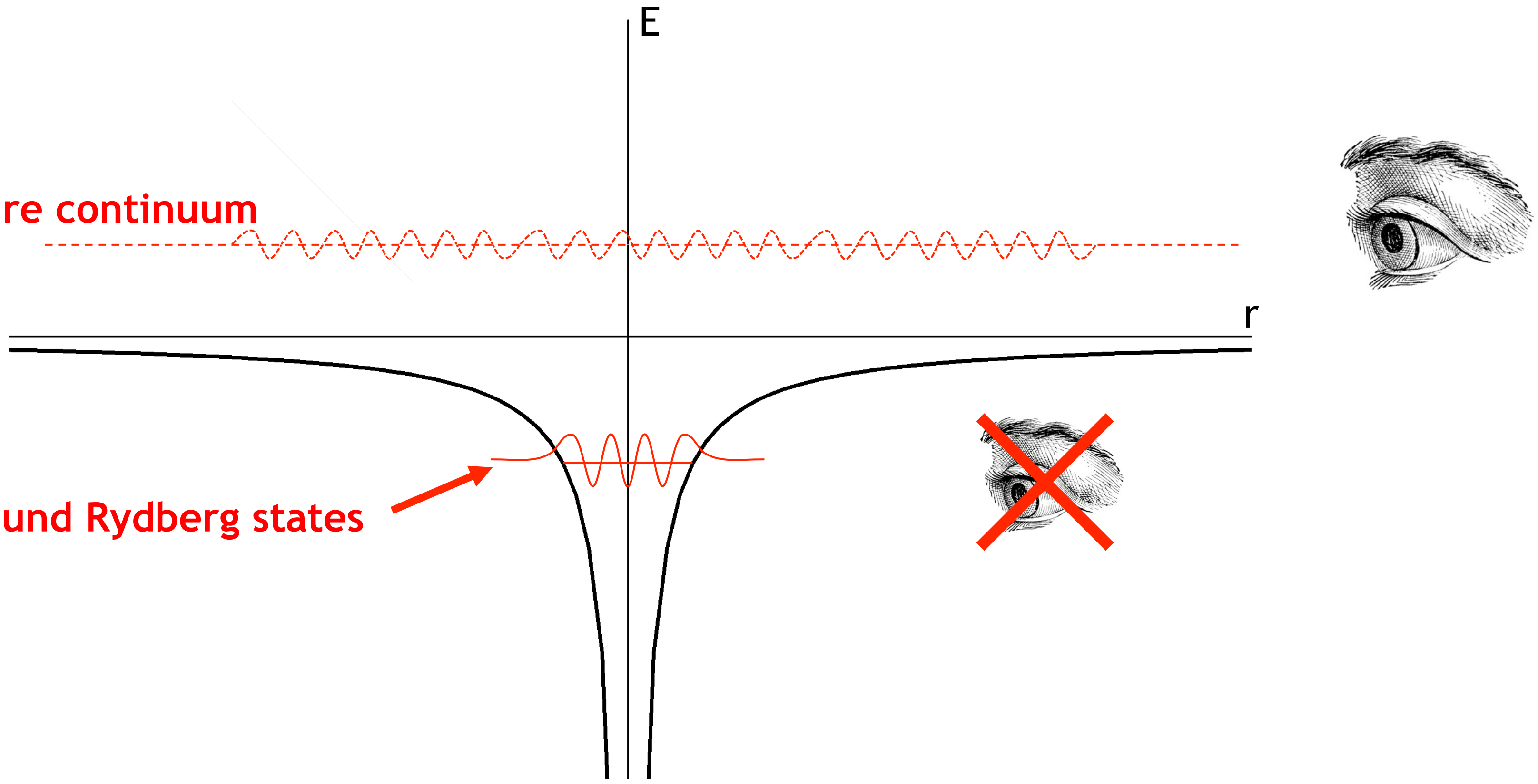


- **Direct look at the wavefunction = link between nano and macro-world… between quantum and classical physics.**
- **One of the current goals of quantum physics:**
  - *Presentation R. Dörner yesterday*
  - *Tomography: J. Itatani et al., Nature 432, 867 (2004).*
  - *Weak measurements: S. Kocsis et al., Science 332, 1170 (2011); J.S. Lundeen, Nature, 474, 188 (2011).*
  - **Direct visualization:** present work is a preliminary attempt… limited to the Wavefunction modulus…

ATOMIC (RYDBERG) WAVEFUNCTION - NO FIELD

F=0   Coulomb potential: V=-1/r

E > 0 pure continuum



E < 0 bound Rydberg states

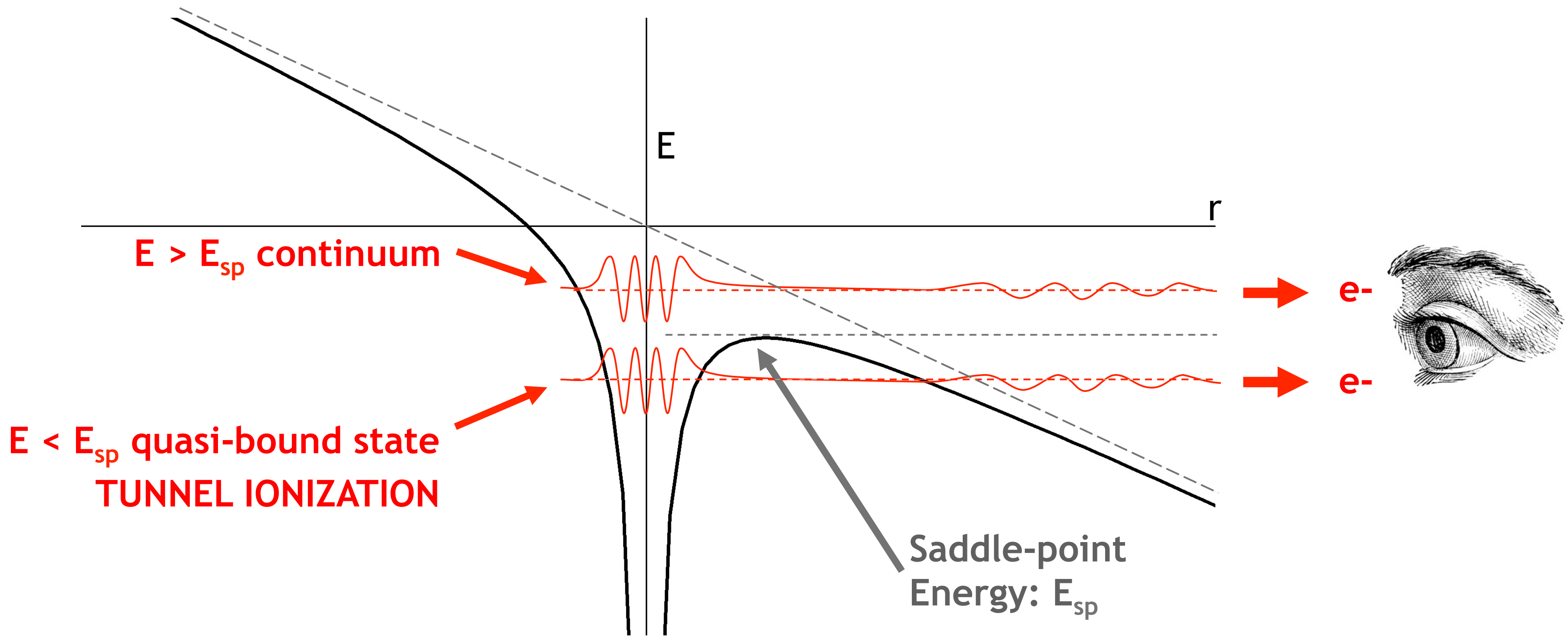
➔ Photoionization in the continuum

=

Continuum wavefunction, no specific atomic structure

ATOMIC (RYDBERG) WAVEFUNCTION - ELECTRIC FIELD

$F \neq 0$ Coulomb + Stark potential: $V=-1/r - Fz$



- ✧ Photoionization in the presence of an external electric field: electron wavefunction propagates at ∞
- ✧ Measuring the spatial distribution of the electron flux with a position sensitive detector = observing the square modulus of the transverse component of the wavefunction
- ✧ **Tunnel ionization of quasi discrete Rydberg states = unique opportunity to visualize directly the electron wavefunction.**

HYDROGEN STARK EFFECT

Hamiltonian is separable in parabolic coordinates

- ❑ **Quasi-discrete Stark State (n_1, n_2, m)**
- ○ n_1 : number of nodes of $\psi(\xi)$
- ○ n_2 : number of nodes of $\psi(\eta)$
- ○ $n = n_1 + n_2 + |m| + 1$

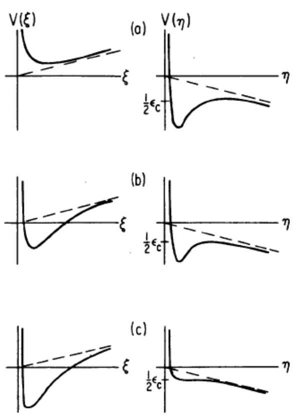


FIG. 1. Qualitative plots of the potentials $V(\xi)$ and $V(\eta)$ in Eqs. (3a) and (3b) for $m>1$, $F \gtrsim 0$, and sample values of $\beta_1 = 1-\beta_2$: (a) $\beta_1 \approx -0.1$, (b) $\beta_1 \approx +0.4$, and (c) $\beta_1 \approx 0.9$. --- pure-Stark potentials $+\frac{1}{4}F\xi$ and $-\frac{1}{4}F\eta$. The top of the potential hump in η, $\frac{1}{2}\epsilon_c$, and the potential well coalesce in (c), where $\beta_2 \approx \beta_{crit} \sim 0.1$.

PHYSICAL REVIEW A VOLUME 24, NUMBER 5 NOVEMBER 1981

Hydrogenic Stark effect: Properties of the wave functions

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(Received 2 March 1981)

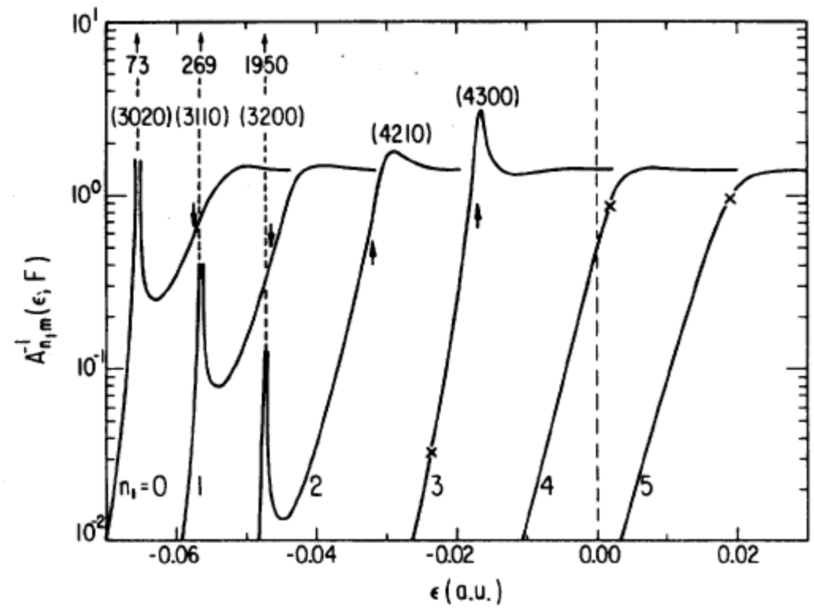


FIG. 7. Amplitude ratio $A^{-1}_{n_1m}(\epsilon,F) \propto \chi_2(\eta\sim 0)/\chi_2(\eta\sim\infty)$, on log scale, vs ϵ for $m=0$, $F=0.001$ (same as Figs. 3 and 6), and $n_1 = 0-5$. Peaks correspond to quasibound levels, below and above threshold ϵ_c (arrows). ×: $\beta_2(\epsilon,F;n_1,0)=0$; $A^{-1}_{n_1 0} \xrightarrow[\epsilon\to\infty]{} \sqrt{2}$.

Partial (n_1) photoionization cross-section

THOUGHT EXPERIMENT

Proposed in the 80's by Fabrikant, Demkov, Kondratovitch & Ostrovsky

J. Phys. B: At. Mol. Opt. Phys. **23** (1990) 3785–3809. Printed in the UK

Resonance and interference phenomena in the photoionization of a hydrogen atom in a uniform electric field: IV. Differential cross sections

V D Kondratovich† and V N Ostrovsky‡

† Vinnitsa Polytechnical Institute, Vinnitsa 286021, USSR
‡ Leningrad State University, Leningrad, 198904, USSR

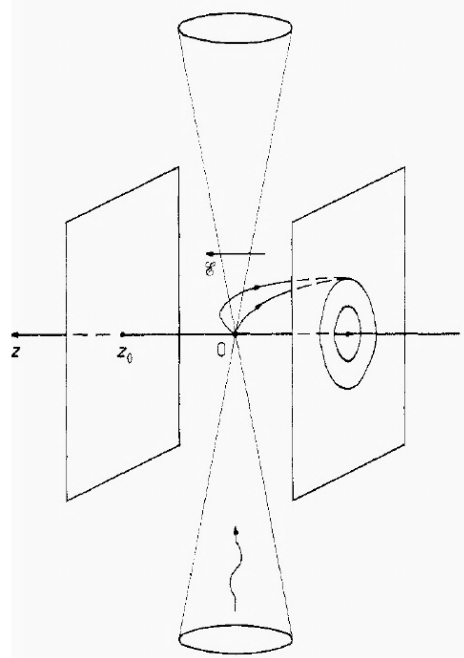
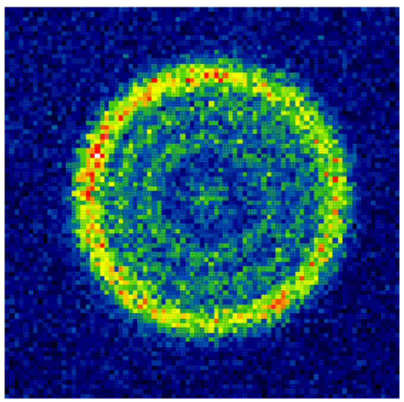


Figure 3. A possible experimental set-up for observation of the interference picture. The photoionization (photodetachment) occurs in the focus of the laser rays between the condenser plates. The distance between the plates is equal to $2z_0$. The plates generate the homogeneous electric field of the strength $\mathscr{E}$. The beam of atoms or the beam of negative ions can be directed perpendicular or parallel to the plates. Two trajectories leading from the ionization point O to the same point on the screen are pictured. Also the interference pattern circles are pictured in the plane screen perpendicular to the electric field direction.

V.D. Kondratovitch and V.N. Ostrovsky, J. Phys. B 17 (1984) pp. 1981 and 2011, 23 (1990) pp. 21 and 3785.

PHOTOIONIZATION vs. PHOTODETACHMENT

In the case of **photodetachment** classical trajectories as well as the reduced action are simple expressions and allow an analytical semiclassical treatment of photodetachment microscopy.



BALLISTIC MATTER WAVES WITH ANGULAR . . .

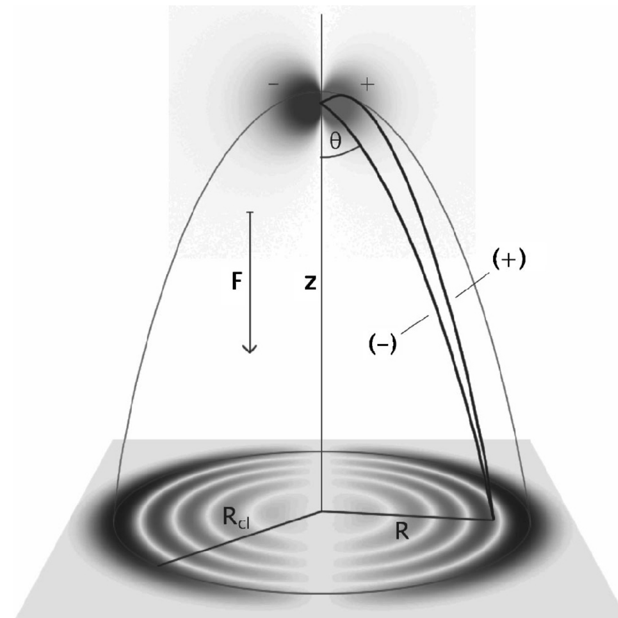
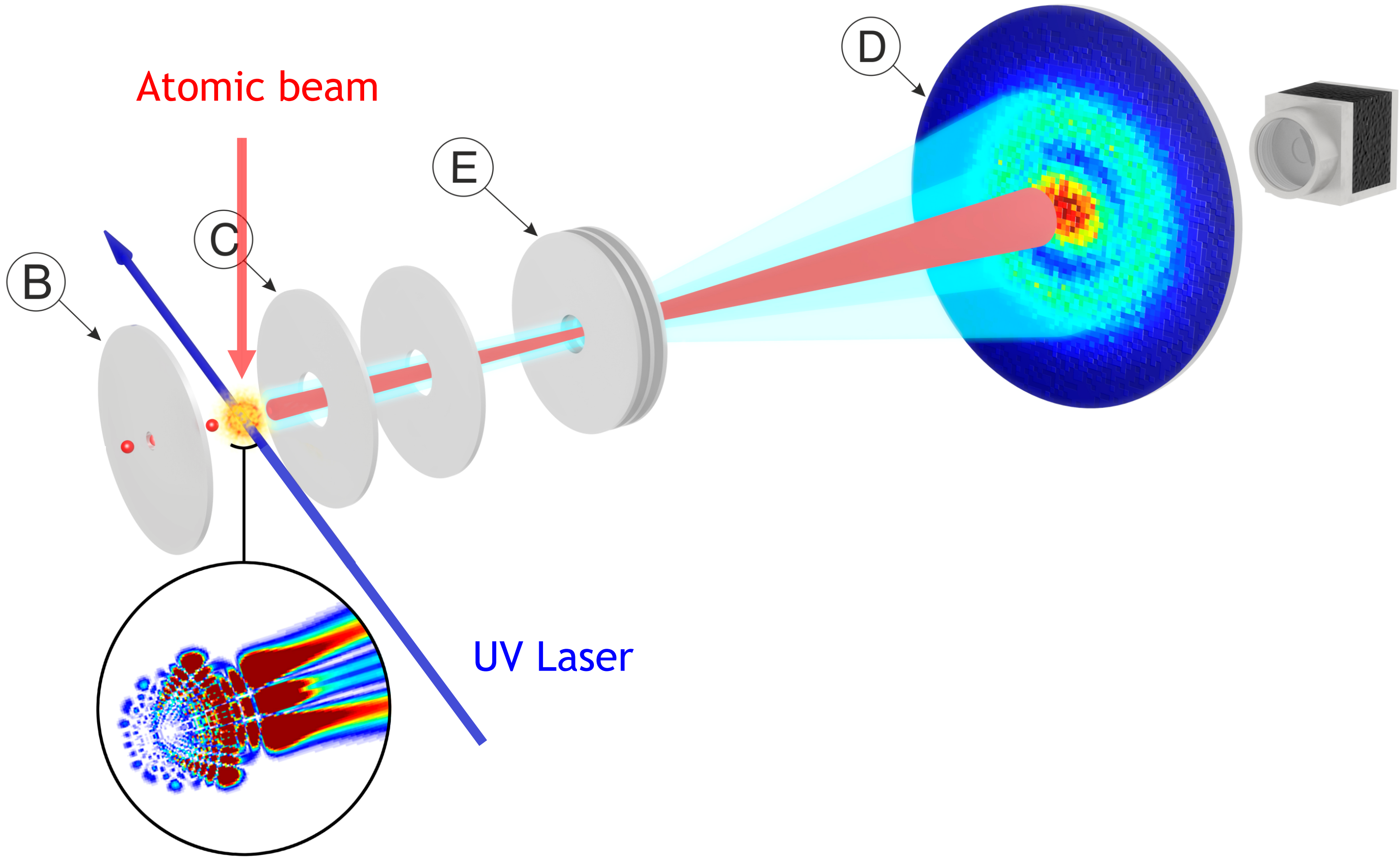


FIG. 1. Semiclassical ballistic motion in the far-field limit. Within the disk $R<R_{cl}$, the two parabolic paths (bold) emitted under opposite angles θ, $\pi-\theta$ will join the point source with the destination. Unlike the fast path $(-)$, the slow path $(+)$ undergoes a reflection at the parabolic turning surface $\alpha_{-}=0$. The accumulated phases, together with their initial "atomic" phases inherited from the source, determine the exact shape of the interference pattern on the screen.

C. Blondel, C. Delsart and F. Dulieu, Phys. Rev. Lett. 77 (1996) p. 3755 + C. Blondel GRC Photoionization & Photodetachment 2012

C. Bracher, T. Kramer and M. Kleber, Phys. Rev. A67 (2003) 043601.

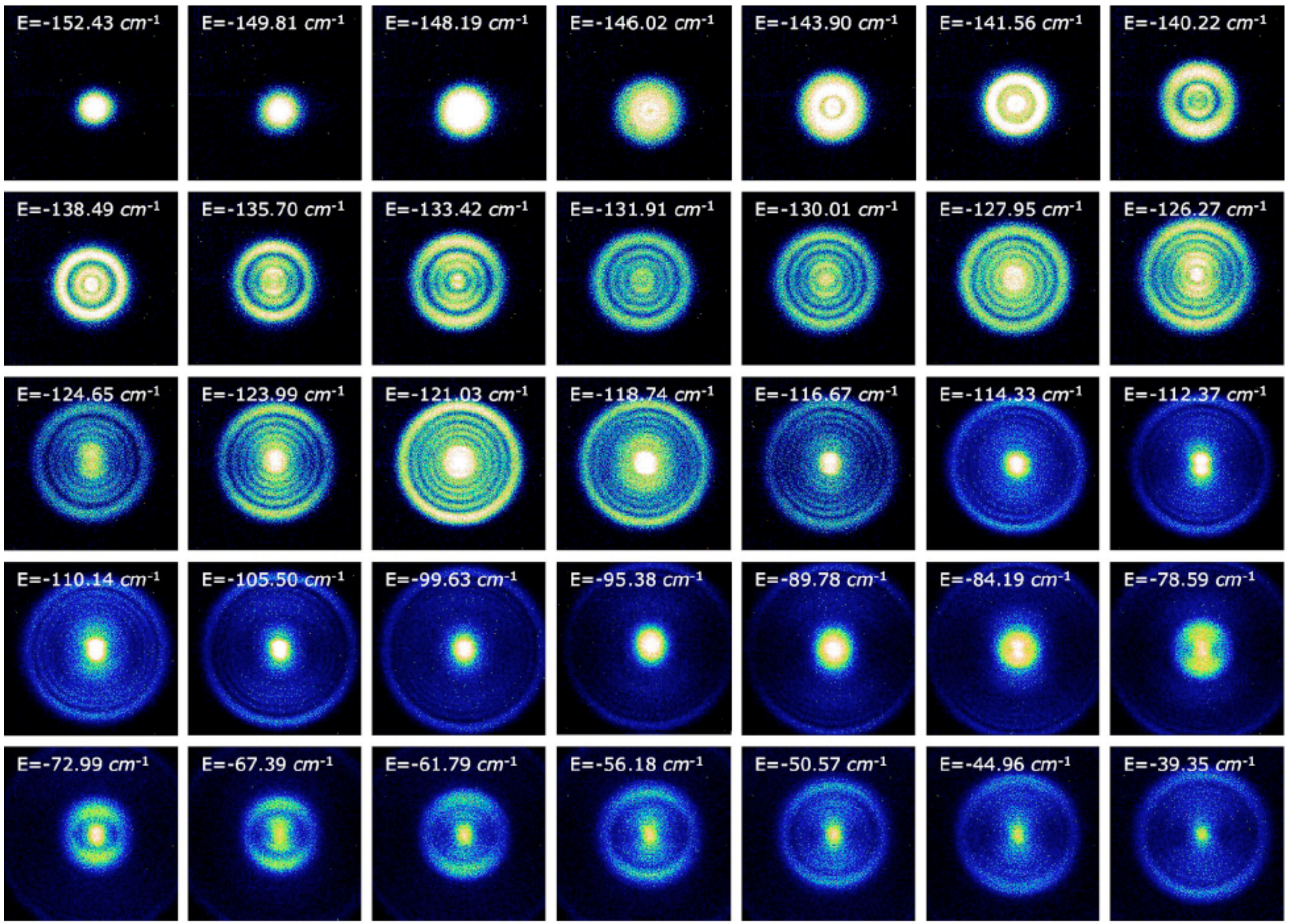
EXPERIMENTAL SET-UP



The atomic beam is crossed at right angle by the narrow-band ionization laser in the interaction region of a Velocity-Map-Imaging spectrometer. Electrons are accelerated between the repeller (B) and the extractor (C). An Einzel lens is used to magnify the image (up to ×20). Photoelectrons are detected on a MCP equipped with a phosphor screen, followed by a CCD camera.

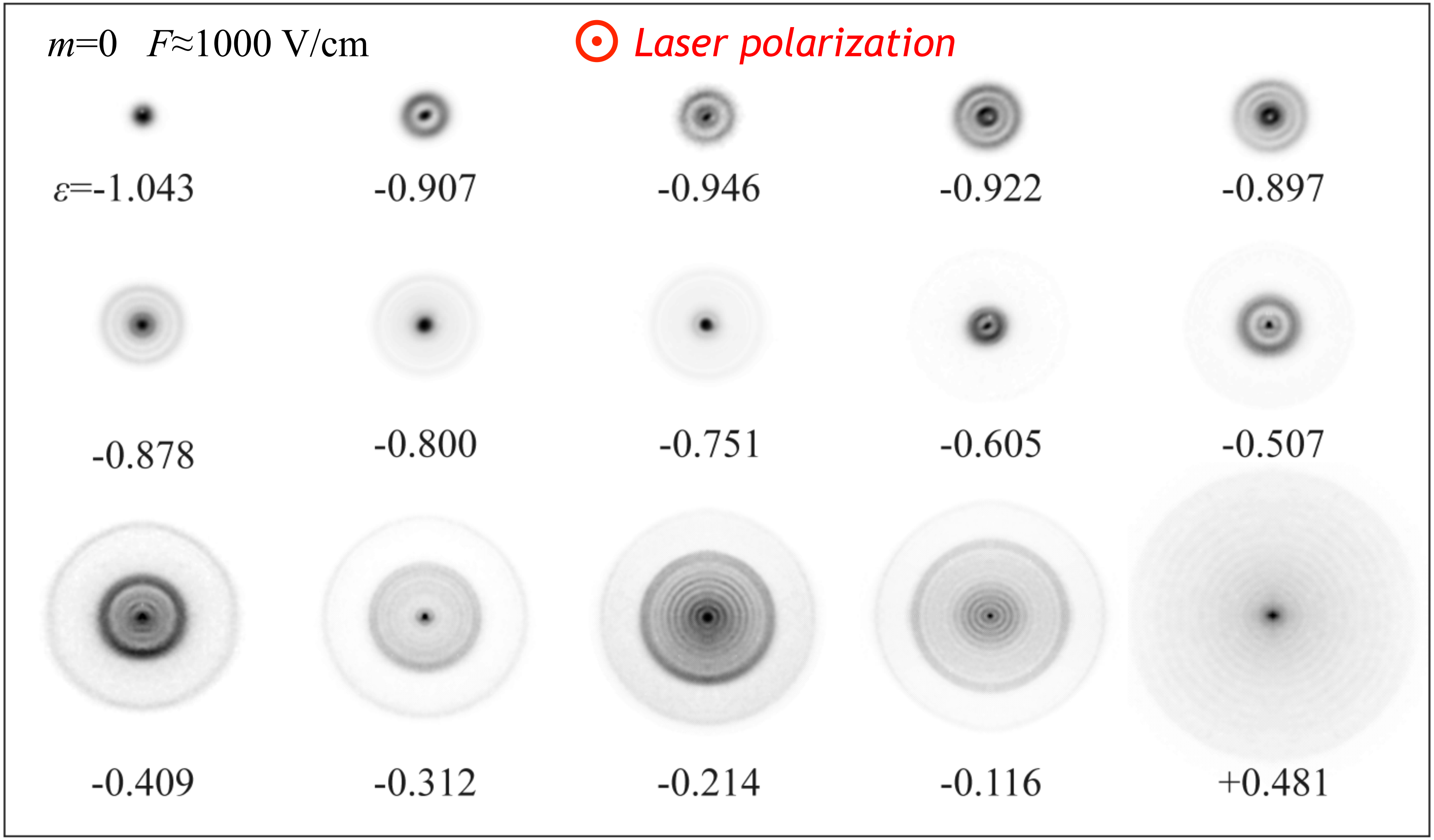
XENON ATOMS: NO RESONANCE EFFECTS

F= 615 V/cm   m=0 E_{sp} = -151.8 cm^{-1}



C. Nicole & al. Phys. Rev. Lett. 88, 133001 (2002); F. Lépine & al., Phys. Rev. A **70**, 033417 (2004)

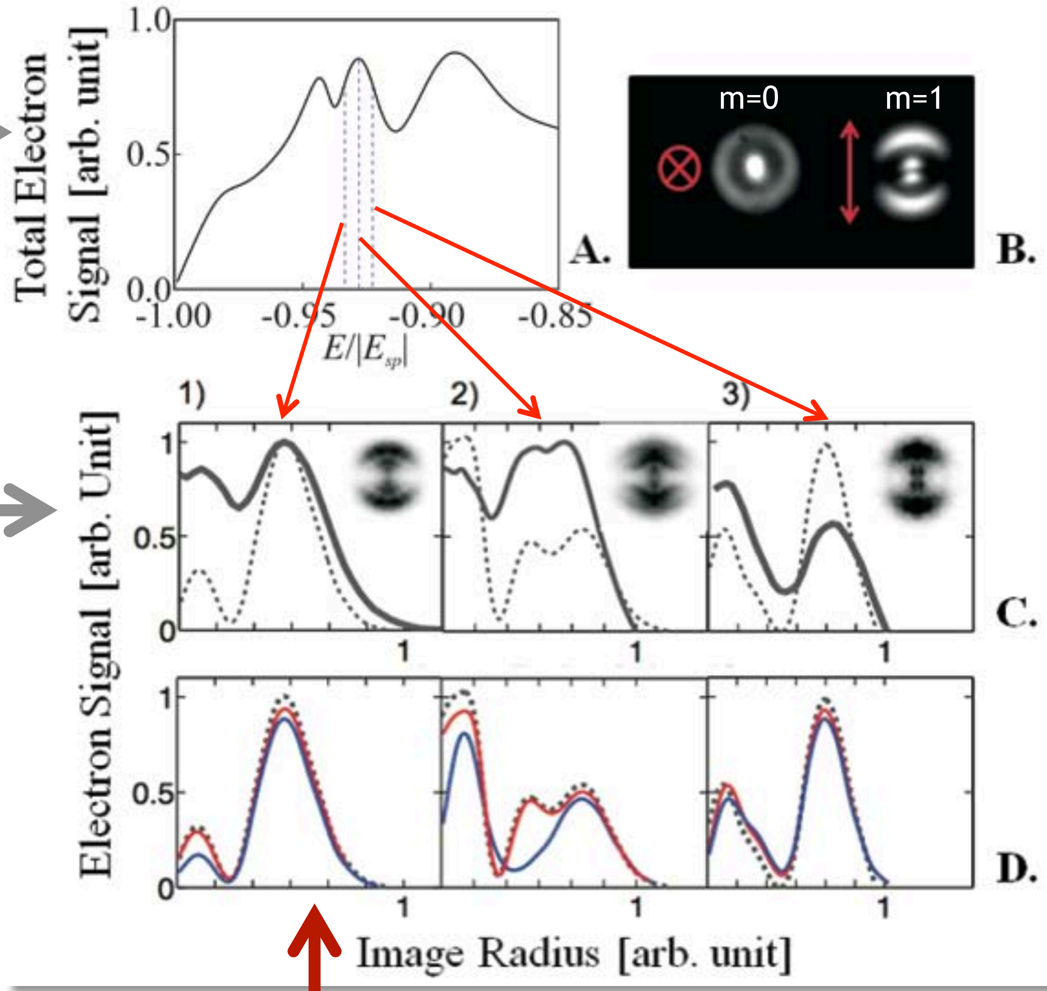
LITHIUM ATOM: RESULTS m=0 (one-photon λ≈230 nm)



Energy evolution of *m*=0 images for *F*=1000 V/cm. The energy is indicated below each image in terms of the dimensionless energy parameter $\varepsilon = E/|E_{sp}|$.

LITHIUM ATOM: RESONANCE EFFECT

Measured ionization yield (F=1010 V/cm), |m|=1). Images, shown below, are measured across one resonance near threshold.



Experimental (continuous line) and calculated (dotted line) radial distributions obtained from one-photon ionization of Li (|m|=1) F=1010 V/cm :

(1) 1 cm^{-1} below resonance; λ=230.917 nm

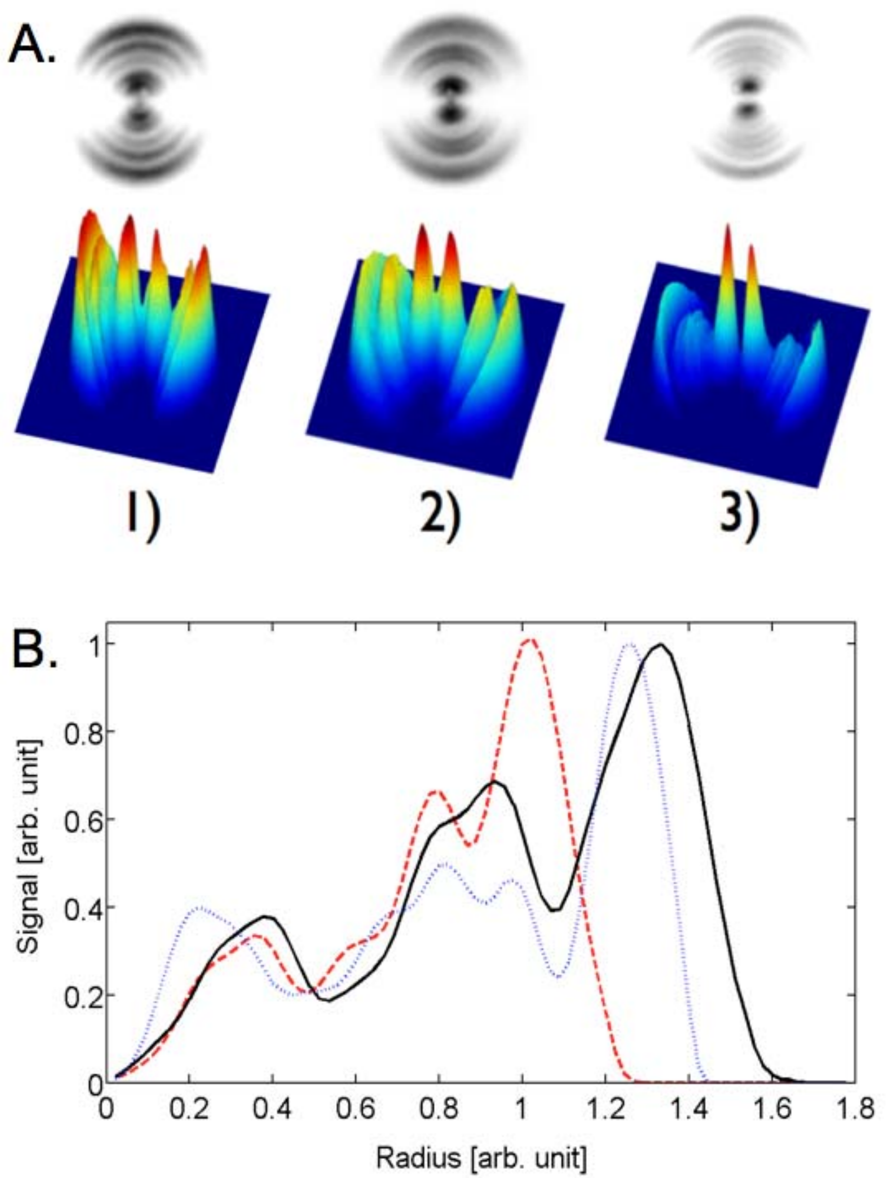
(2) On resonance, 14 cm^{-1} above E_{sp}; λ=230.912 nm)

(3) 1 cm^{-1} above resonance; λ=230.906 nm

The interferogram (2) is a direct image of the (n_1=2, |m|=1) state.

Calculated radial distributions for H (red line), Li (black-dotted line) and Cs (blue line).

LITHIUM ATOM: RESONANCE EFFECT



2D and 3D representation of experimental images - one-photon ionization of Li - *F*=1010 V/cm, around the (n_1=6, *m*=1) resonance

1) 35.0 cm^{-1} above E_{sp}

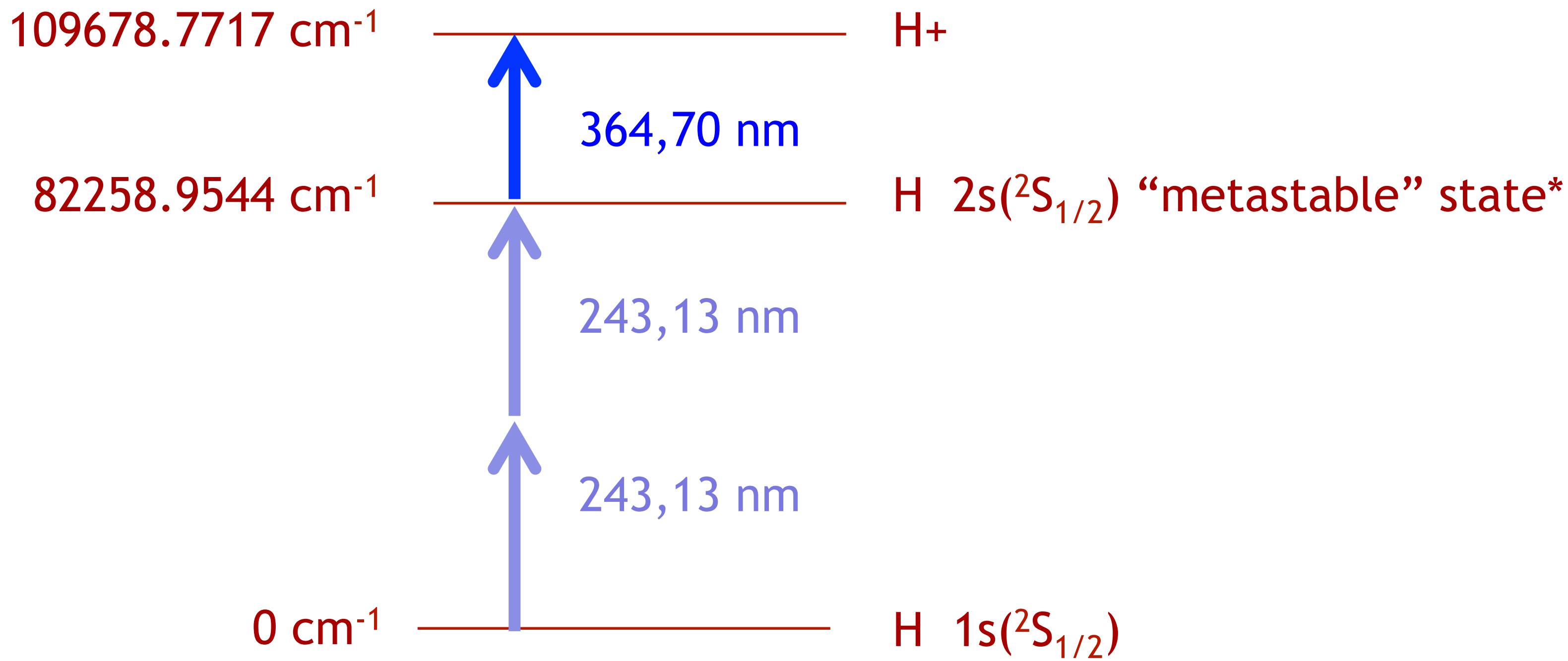
2) 40.8 cm^{-1} above E_{sp} (λ=230.769 nm);

3) 45.4 cm^{-1} above E_{sp}

Radial distribution. The radius is scaled to the classical radius. The image is larger on-resonance (black continuous curve) and smaller at both lower (red-dashed line) and higher (blue-dotted line) photon energies. This is attributed to tunneling ionization.

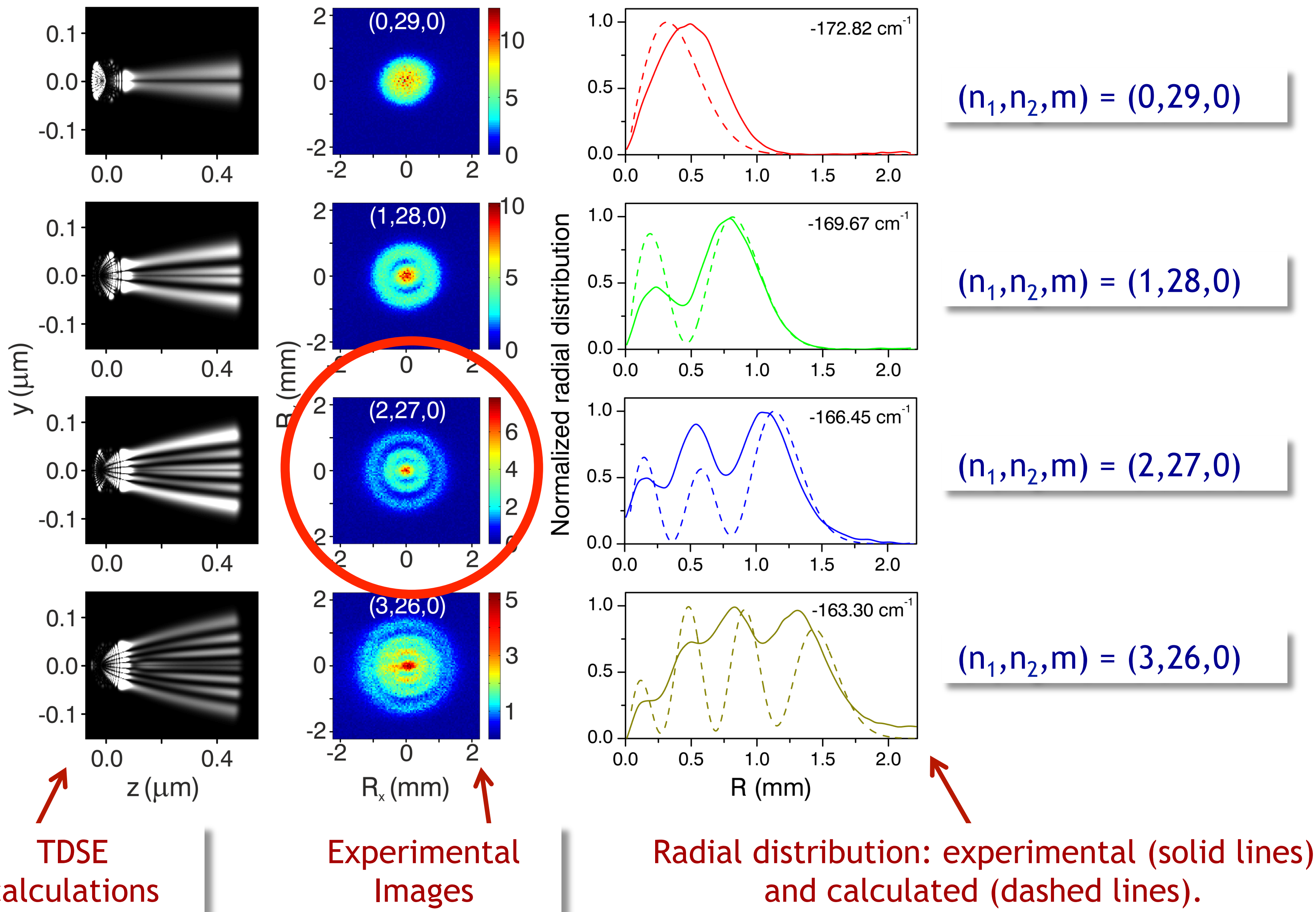
HYDROGEN ATOM

- Production of atomic hydrogen $H_2S + h\nu = H + SH$
- Photoionization in a dc electric field



Lithium atom: S. Cohen et al. Phys. Rev. Lett. 110, 183001 (2013)
Hydrogen atom: A.S. Stodolna et al. Phys. Rev. Lett. 110, 213001 (2013)

HYDROGEN ATOM: NODAL STRUCTURE



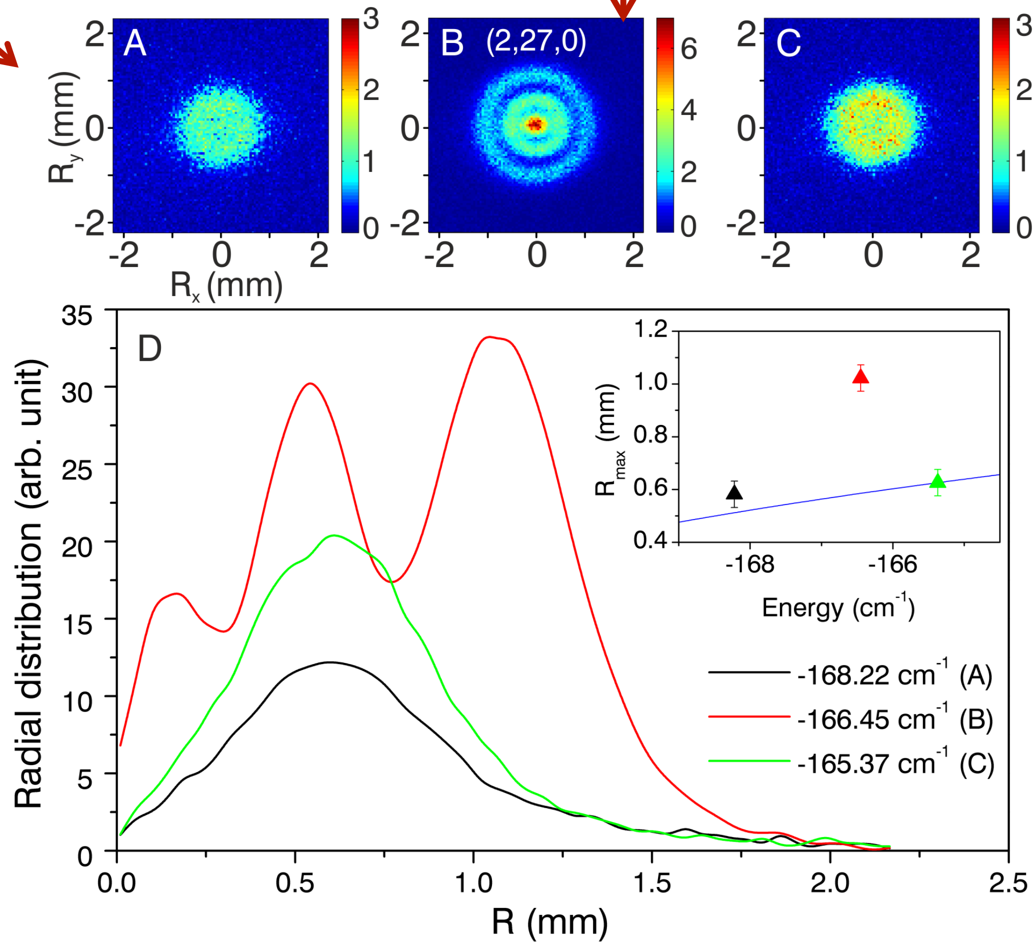
HYDROGEN ATOM: RESONANCE & TUNNELING

Evidence for resonance effects and ionization by tunneling through the Coulomb + static field potential.

1.8 cm^{-1} below resonance

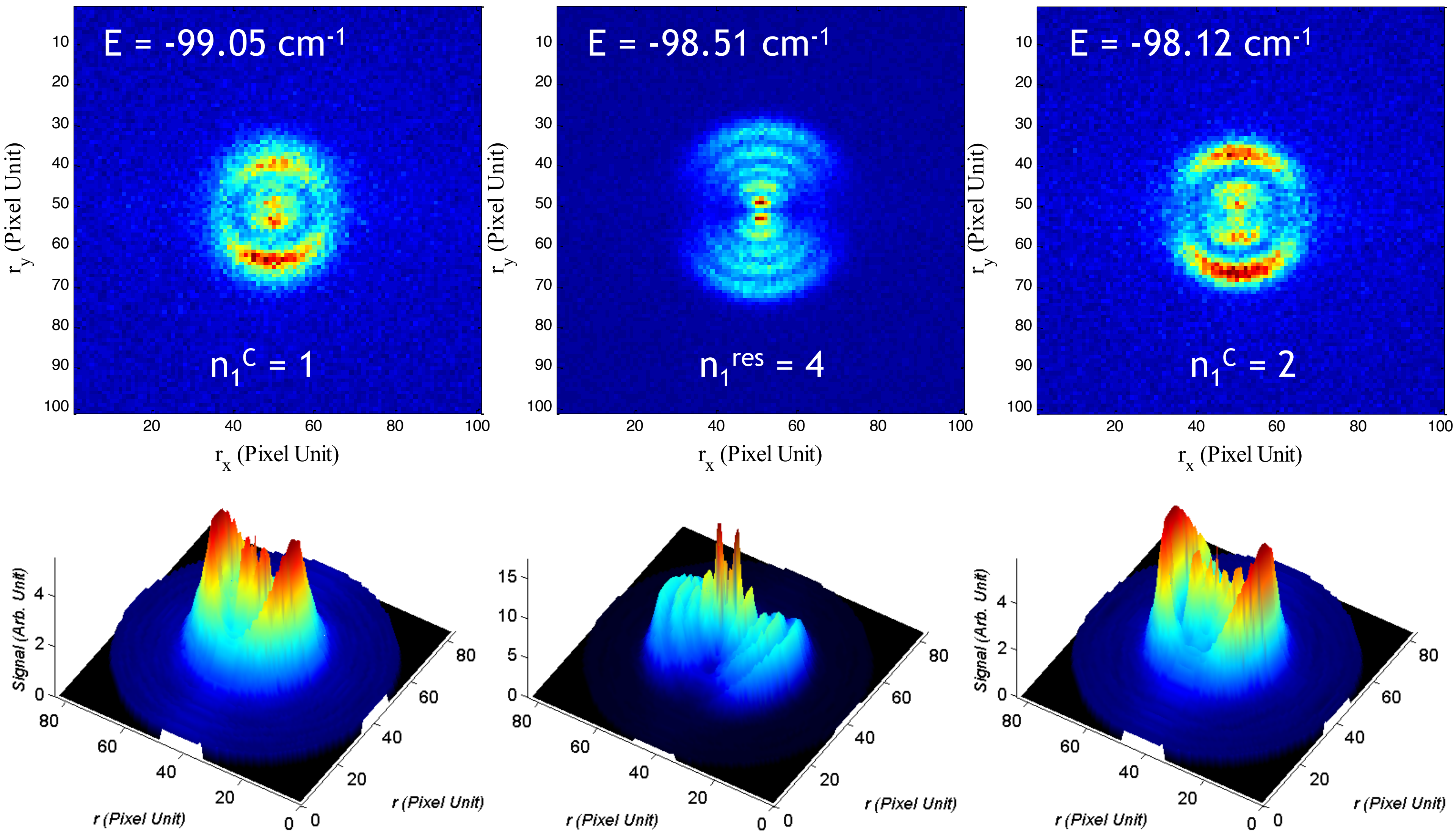
$(n_1,n_2,m)=(2,27,0)$ resonance

1.1 cm^{-1} above resonance



Exp. radial distributions

HYDROGEN ATOM: VICINITY OF A RESONANCE



CONCLUSION & OUTLOOK

- Direct observation of the wavefunction square modulus in the macroscopic world
- Photoionization microscopy on xenon: continuum only
- Clear resonance effects in light atoms (H, Li)
- Photoionization microscopy on lithium and hydrogen: quasi-bound states wavefunction

- Improve theoretical description for complex atoms
- Effect of magnetic field…
- Photoionization microscopy of molecules
- Slow Dynamics: building-up of the electron current
- Simple double-slit experiment: generalization to Microscopic interferometry…
- **Complete wavefunction: amplitude and phase… Which strategy?**

ACKNOWLEDGEMENTS

F. LÉPINE, A. OLLAGNIER (Ph.D. 2007), M. HARB (Ph.D. 2010), T. BARILLOT (Ph.D. 2013) iLM- CNRS & Université Lyon 1 (Li exp., wavepacket propagation).

M.J.J. VRAKKING, C. NICOLE, A.S. STODOLNA, A.ROUZÉE, A. GIJSBERTSEN, J.H. JUNGMANN - FOM AMOLF Amsterdam & MBI Berlin (Xe & H exp.)

S. COHEN - Univ. Ioanninna Greece (Li experiments)

F. ROBICHEAUX - Purdue University Al. (wavepacket propagation)

L. ZHAO, J.B. DELOS & coworkers - College of William & Mary (Coupled-channel theory)