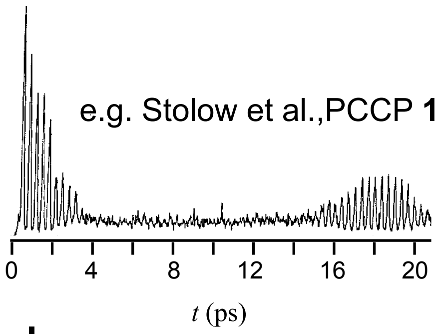
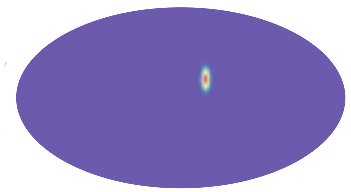
# Possible manifestations of quantum disordered dynamics in the arrested relaxation of a molecular ultracold plasma

## Eigenstate Thermalization Hypothesis




e.g. Stolow et al.,PCCP **13**, 18447 (2011)



Superpositions of states in small quantum systems evolve in quantum beats with periodic revivals.

Quenched observables of very large isolated quantum systems relax to states of maximum entropy.

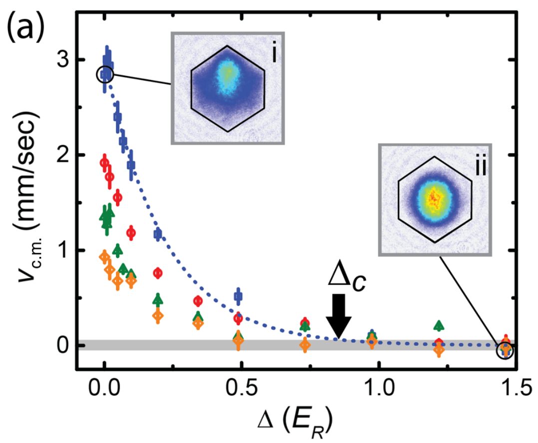
- ETH explains: Even a brief time average of any superposition in a dense manifold of states fills phase space, looks thermal. Eisert et al. *Nature Physics* **11**, 124 (2015).
- Unimolecular rate models assume energy randomization (RRKM Theory).

But, coherent control can localize energy in an excited molecule. Disordered landscapes can suppress transport in complex ensembles: Preserve spatial order and retain energy in highly excited superpositions of states.

- As a paradigm, many-body localization (MBL) in the dynamics of complex systems compares with coherent control in molecules.
- In MBL, local observables retain a memory of initial conditions for arbitrarily long times.
- Potential to preserve quantum information {Nandkishore & Huse, *Annu. Rev. Condens. Mat. Phys.* **6** 15-38 (2015); Abanin, Altman, Bloch & Serbyn, arXiv:1804.11065 (2018)}

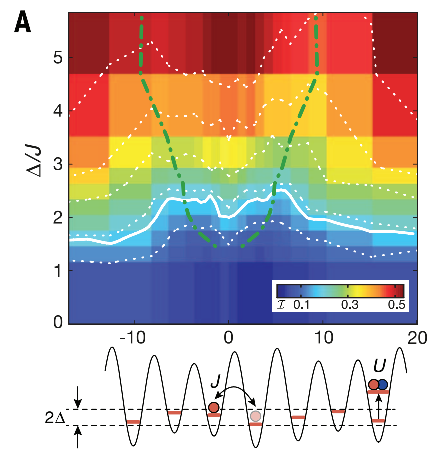
# Experimental quantum systems that fail to thermalize command a great deal of interest.

Kondov, et al., *PRL*, **114** 083002 (2015)



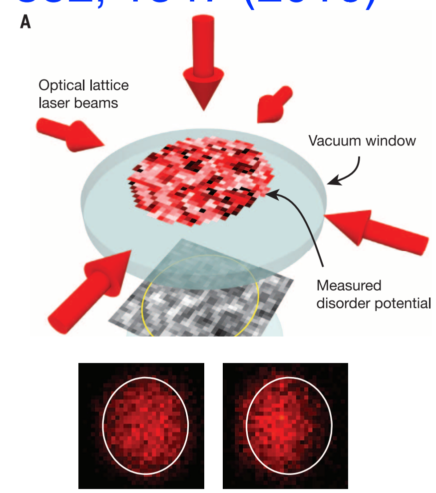

$^{40}$K response to a magnetic impulse

Schreiber, et al. *Science* **349**, 842 (2015)



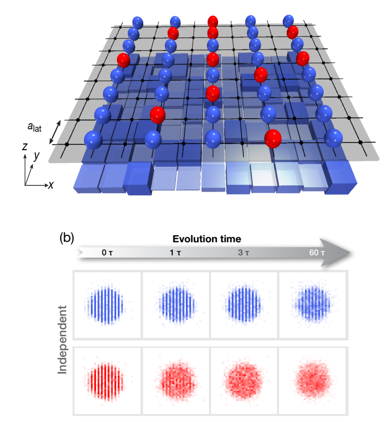

$^{40}$K CDW even to odd lattice sites

Choi et al. *Science* **352**, 1547 (2016)



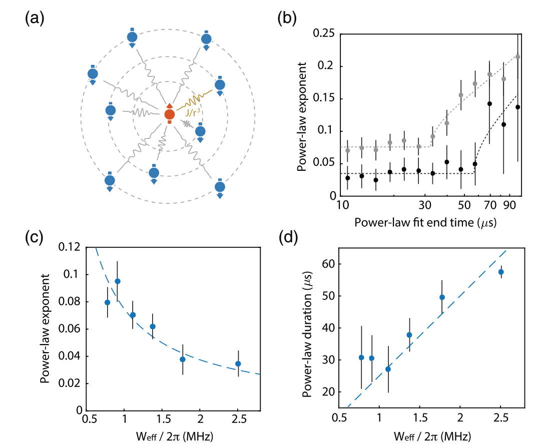

$^{87}$Rb relaxation of density distribution

Rubio-Abadal et al., 1805.00056 (2018)




$^{87}$Rb relaxation of spatial order

Kucsko , et al., *PRL*, **121,** 023601 (2018)




Critical thermalization in 3D NV diamond

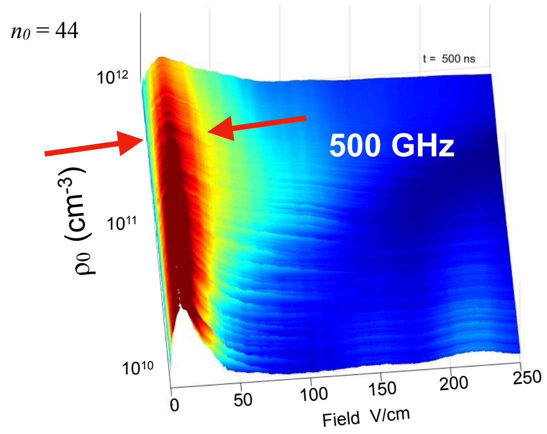
Highly engineered examples confirm the principle of many-body localization (MBL). Feature weak dipolar coupling, intricate experimental design & interpretation.

For systems with stronger interactions, can many-body localization arise naturally?

- In the MBL phase, local operators define local integrals of motion (LIOM).
- The LIOM determine how far any particular excitation can propagate.
- In a quench, can locally emergent conservation laws act to guide the self-assembly of a spatially evolving quantum system to form a global many-body localized state?

# Can the constraints of localization guide self-assembly to an MBL state?

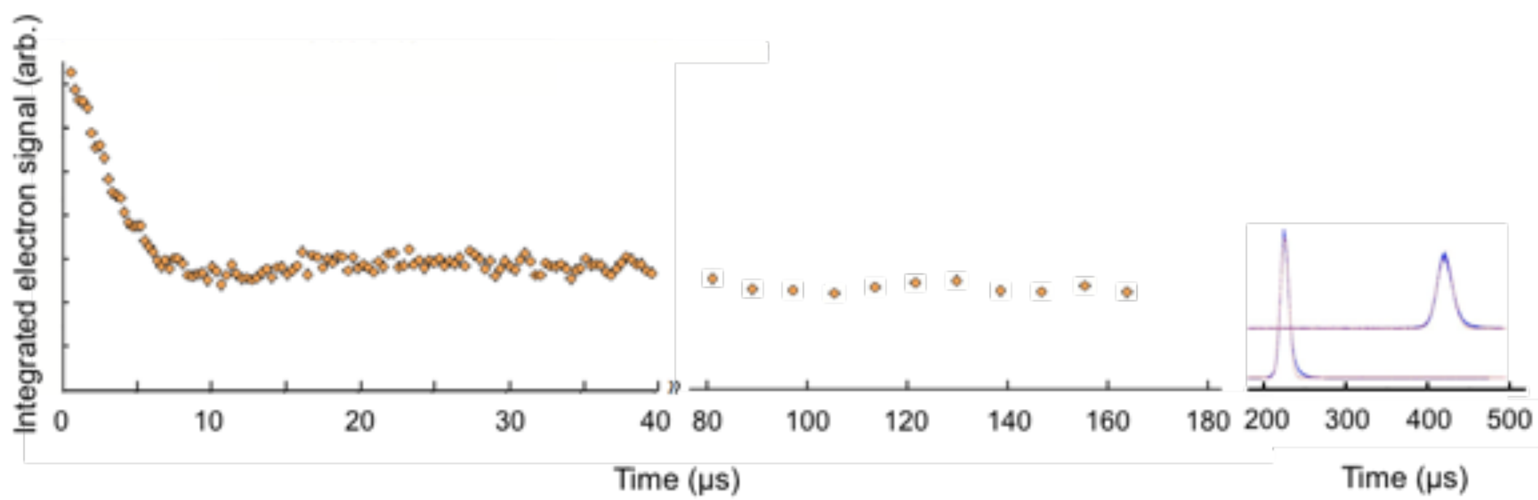
Experiment: Arrested relaxation in an isolated <u>molecular</u> ultracold plasma




An ultracold plasma evolves from a molecular Rydberg gas of nitric oxide

Bifurcates. Irreversibly disposes energy to a reservoir of mass transport.

Quenches to form a strongly coupled, quasi-neutral, plasma in a state of arrested relaxation, far from thermal equilibrium



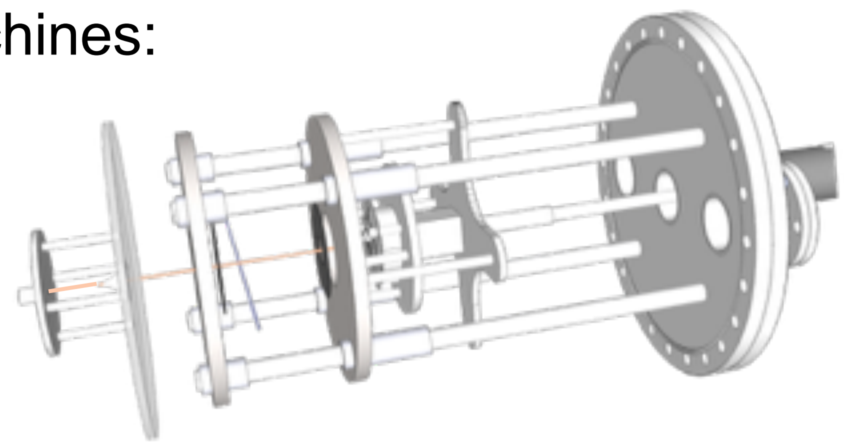

Results invite the theoretical question whether an observed long lifetime and evident very low electron temperature reflect self-assembly to a state of many-body localization far from thermal equilibrium.

Quick overview of the experimental results, model for interpretation

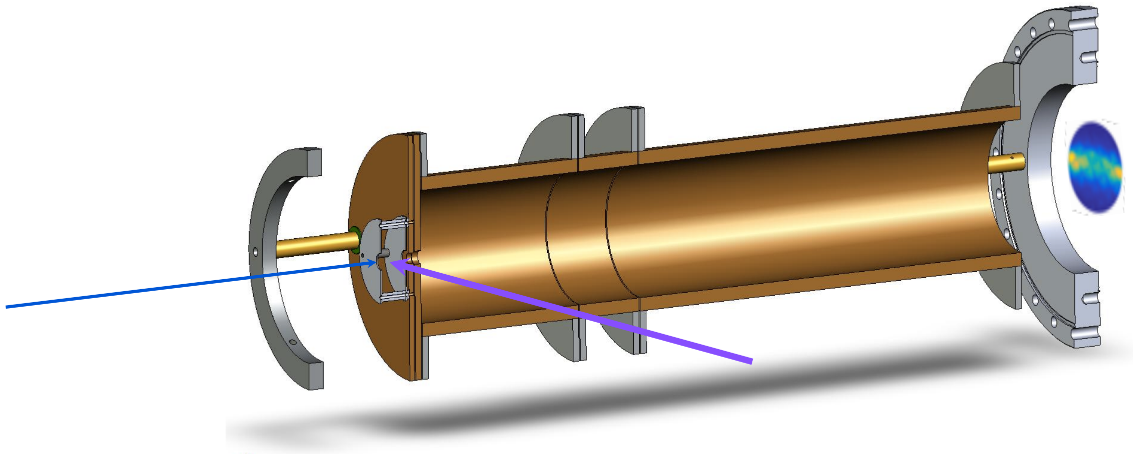
Defined conditions of initial density and temperature

# Differentially pumped skimmed supersonic molecular beam

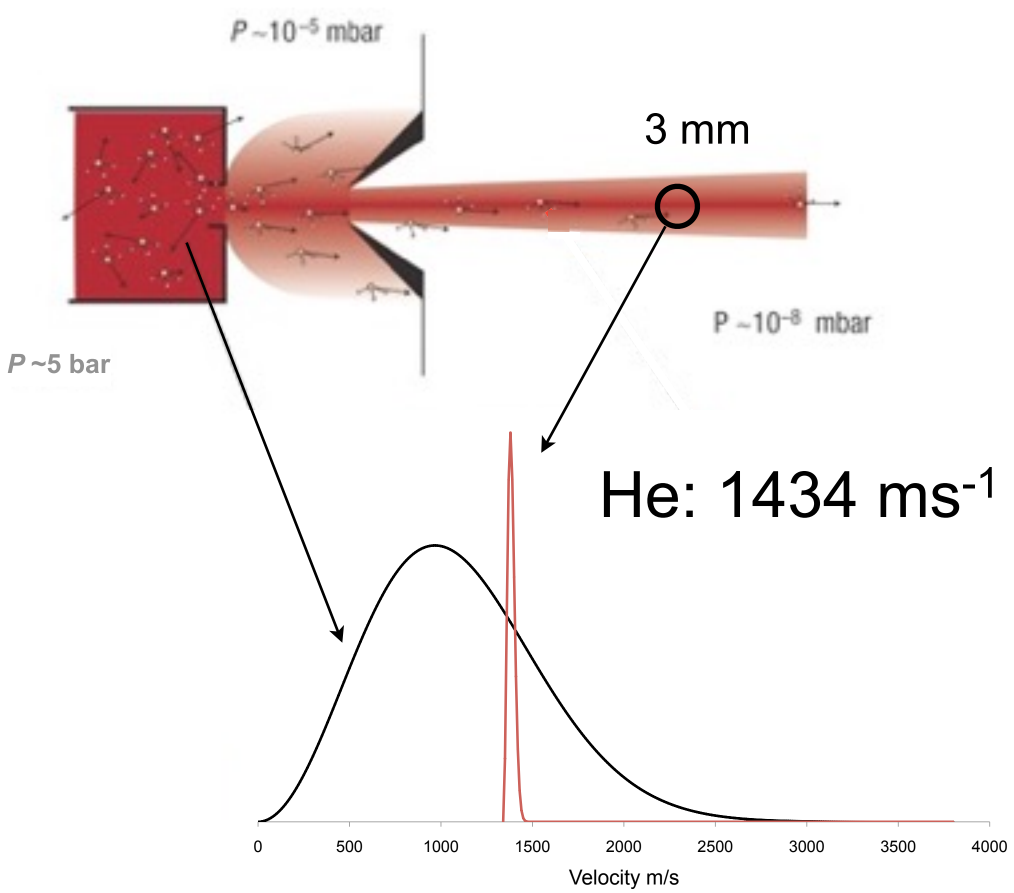
Two machines:



Moving grid



Plasma TV

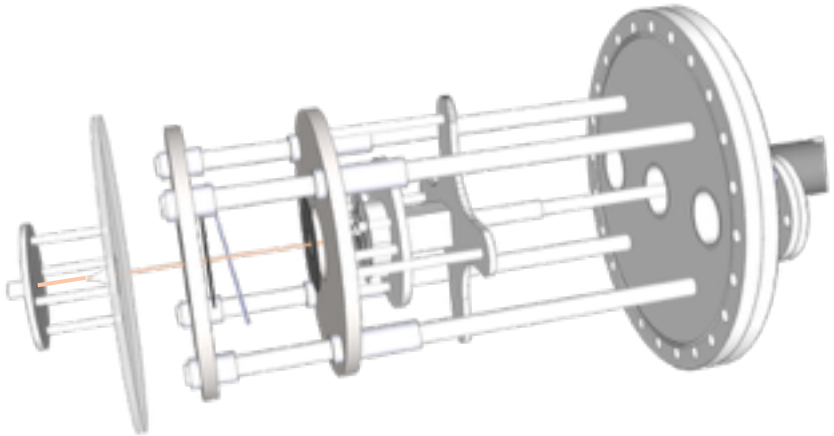


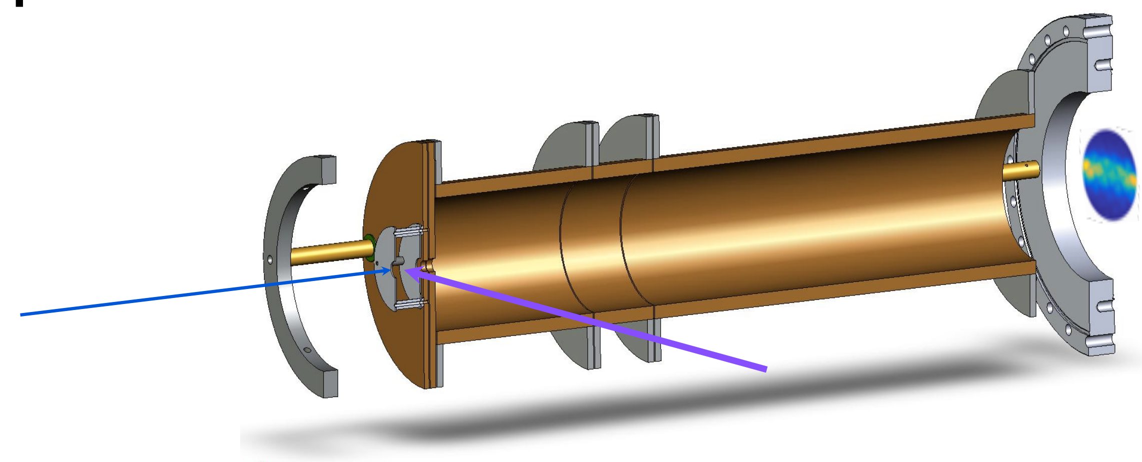

- NO 10% in 5 bar He, Ar
- 0.5 mm nozzle, 1 mm skimmer
- $\rho_{NO} = 1.6 \times 10^{14}\ cm^{-3}$
- $\rho_{NO^*} \approx 5 \times 10^{12}\ cm^{-3}$
- $T_{||}^{\infty} = 500$ mK
- $T_{\perp}^{\infty} \approx 5$ mK

M Schulz-Weiling, et al., J. Phys B **49,** 193001 (2016)

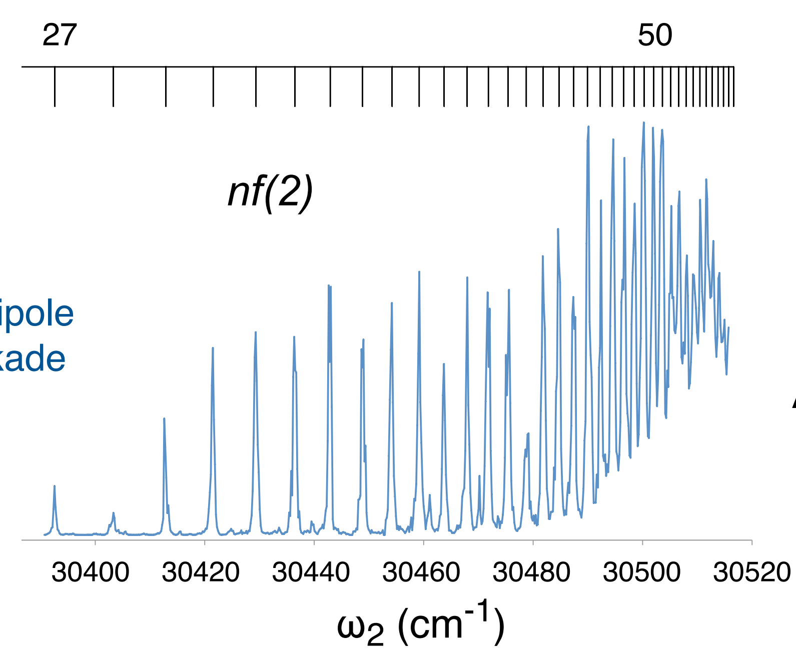
# Selected initial quantum state Experimental control of initial density

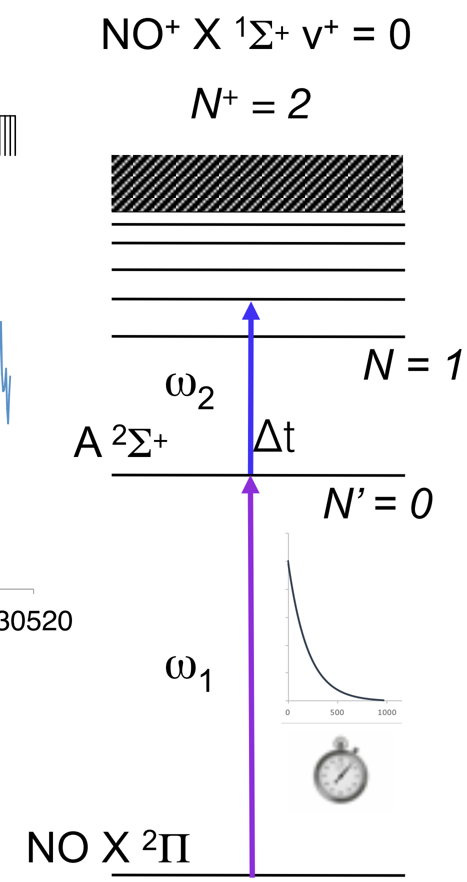
# Experimentally observed dynamics of avalanche and quench with a theoretical interpretation





No dipole blockade



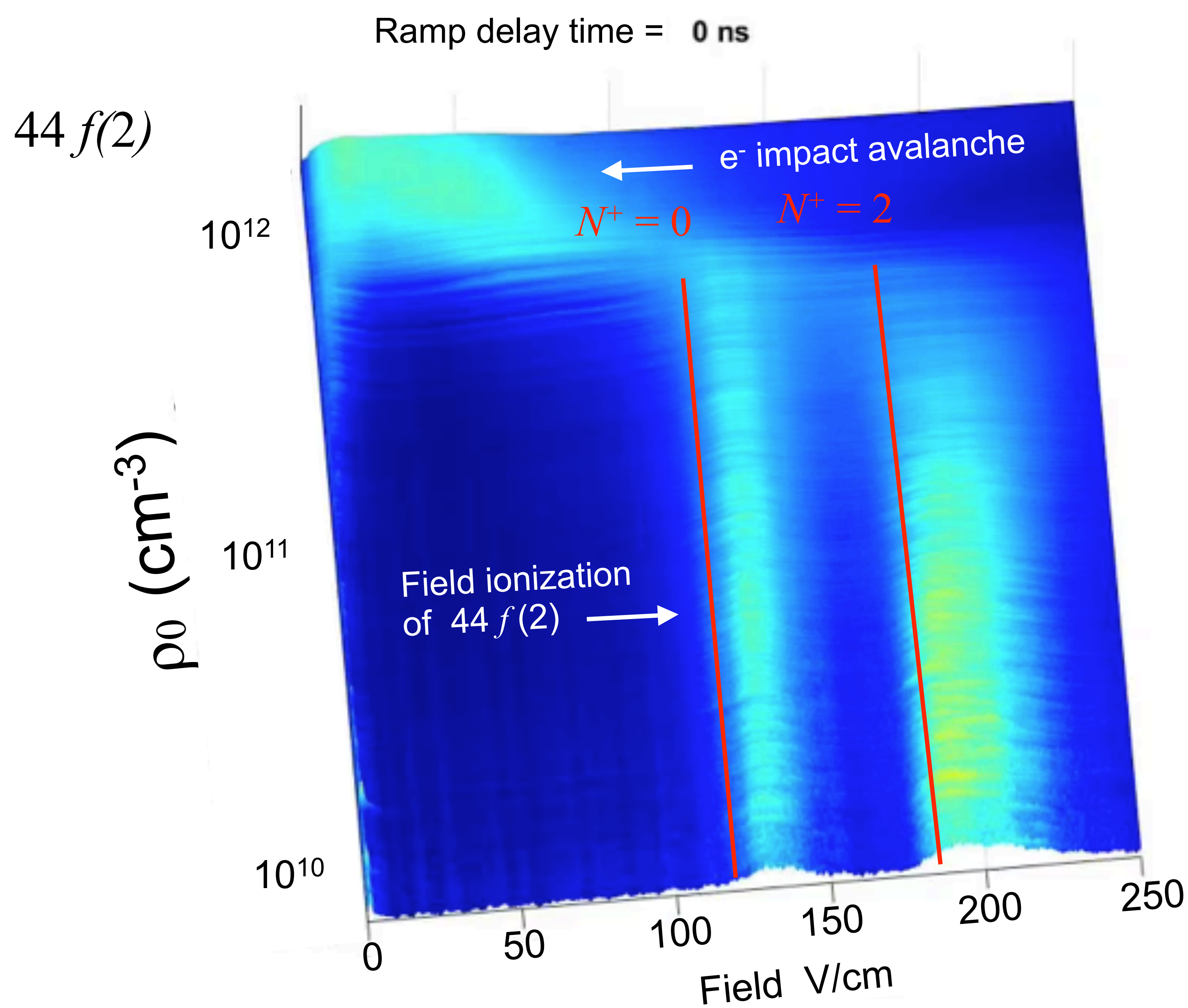



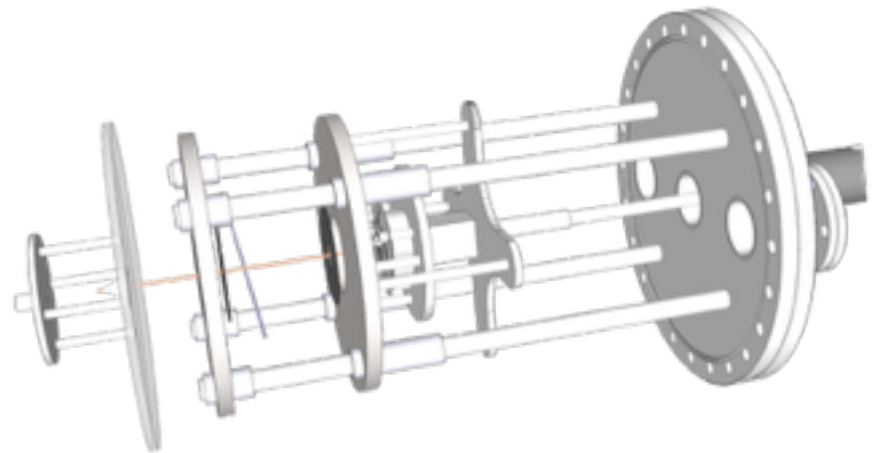

Double-resonant state selection

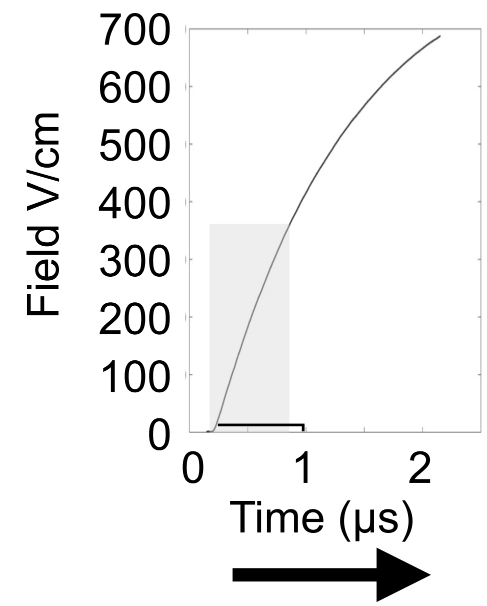
Quantum control of initial state and density

# Short-time dynamics of electron-impact avalanche ionization

# SFI captures the avalanche in progress

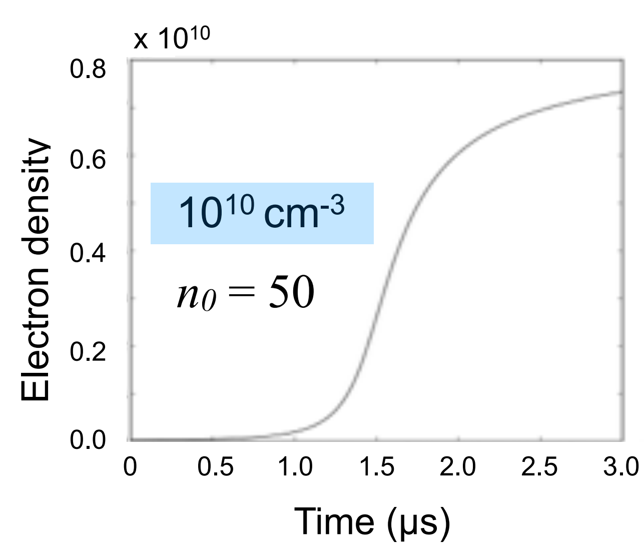





Vary ramp delay in steps

Coupled rate-equation model

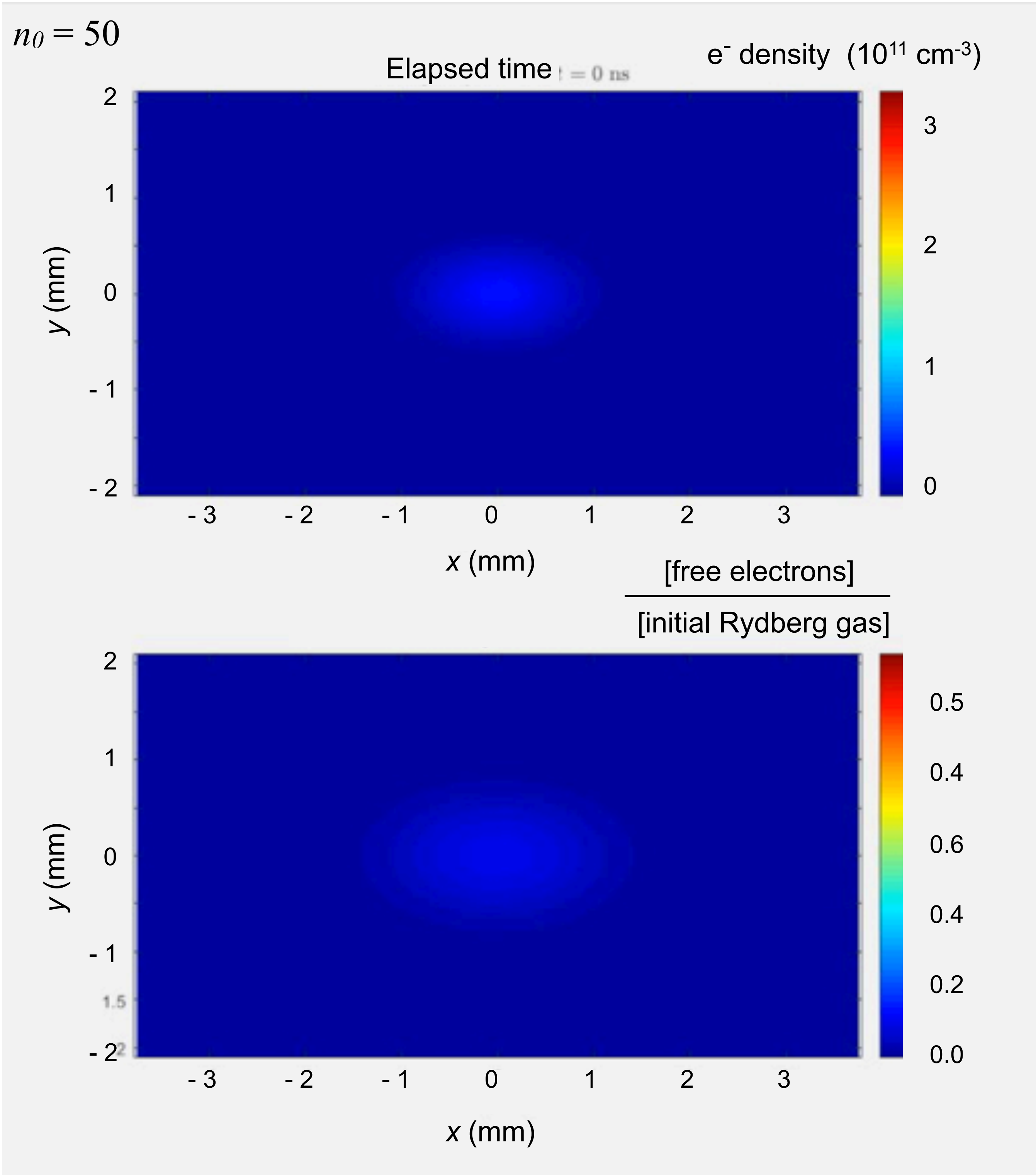


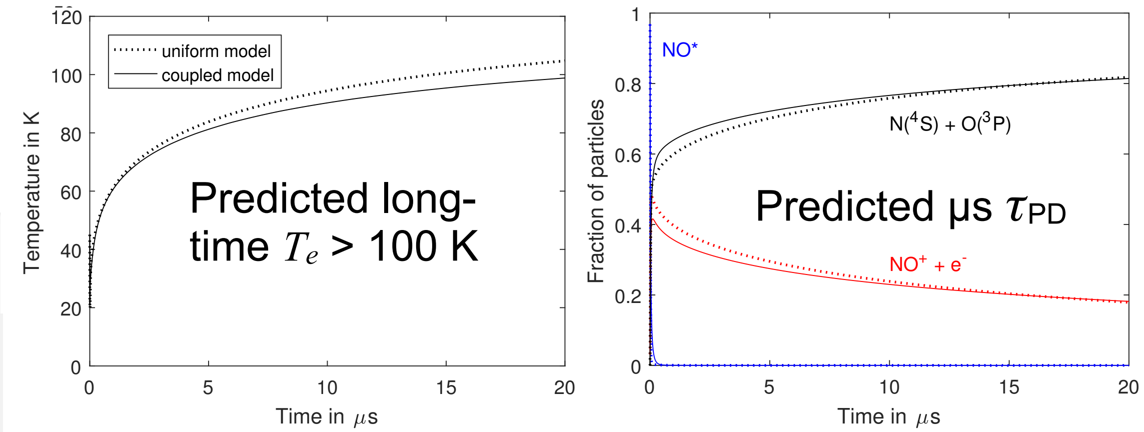

- Shot-to-shot total electron signal accurately classifies Rydberg gas density.
- Observed relaxation rate conforms well with classical simulations at all (uniform) densities.

J. Phys. B **45** (2012) 175302, J. Keller et al, *to be published*.

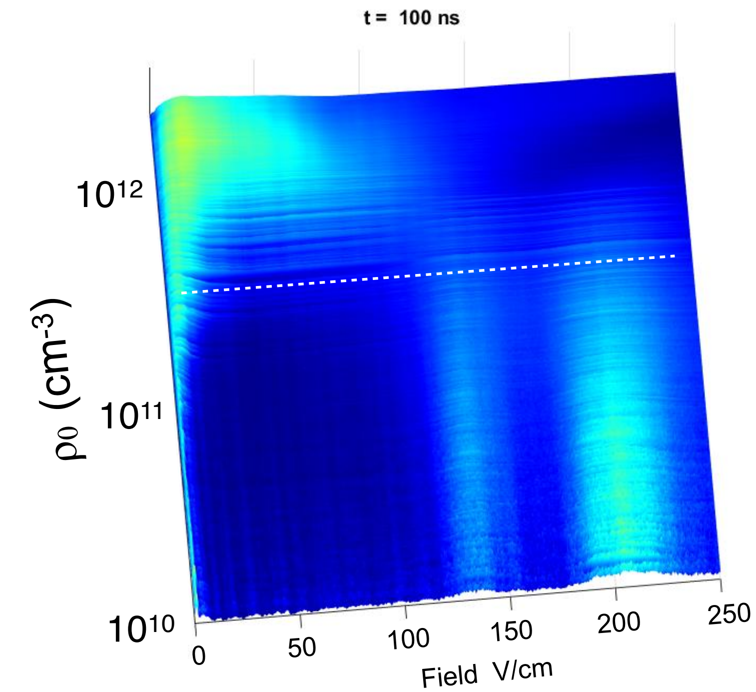
# Shell-model coupled rate-equation simulation

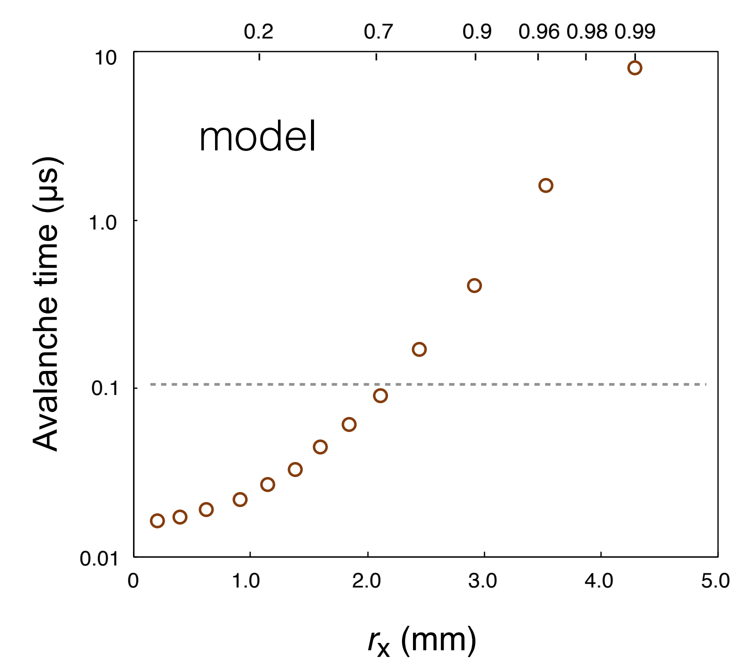
$5 \times 10^{11}\ \mathrm{cm^{-3}}$




$5 \times 10^{11}\ \mathrm{cm^{-3}}$



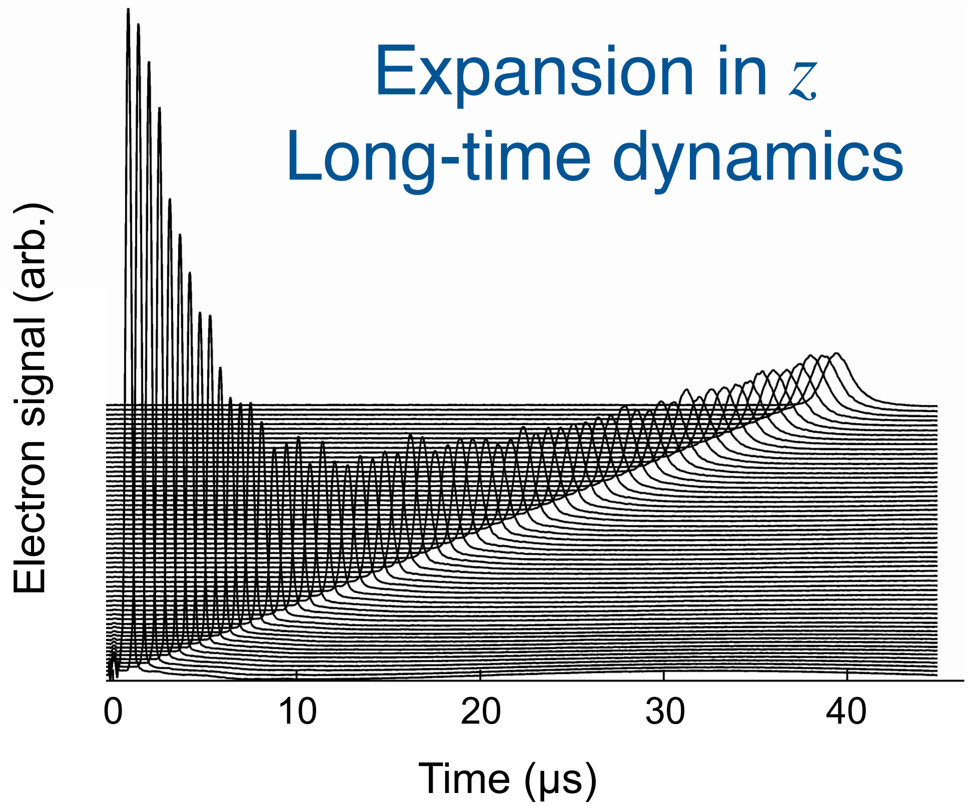




We can measure electron temperature and decay dynamics by examining the signal as a function of propagation time in $z$.

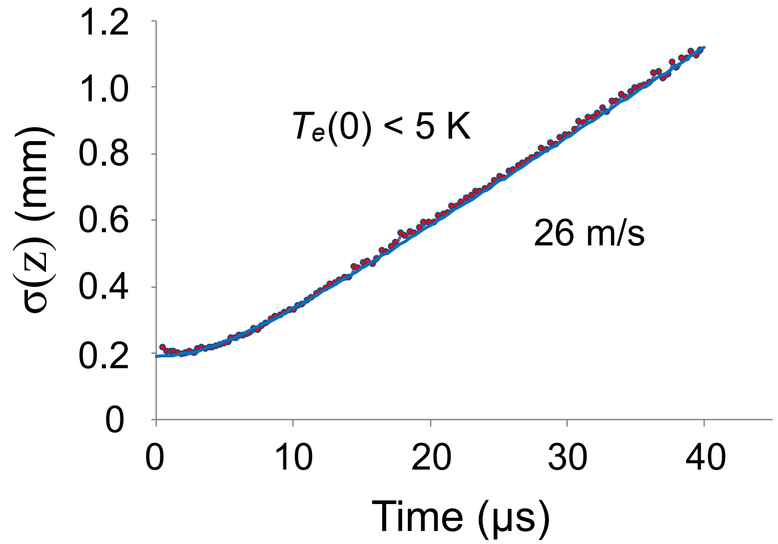
Long-time dynamics:
Ambipolar expansion and predissociation

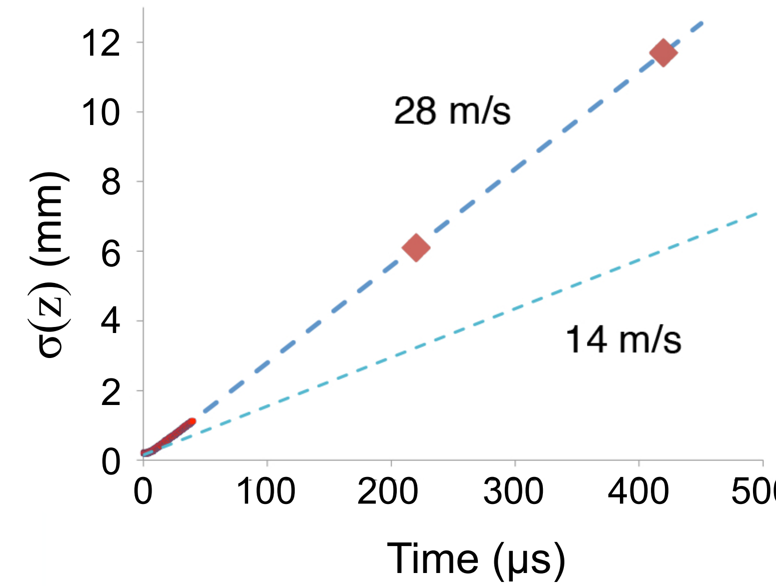
Measure $T_e$ and plasma lifetime

# Expansion in $z$
# Long-time dynamics



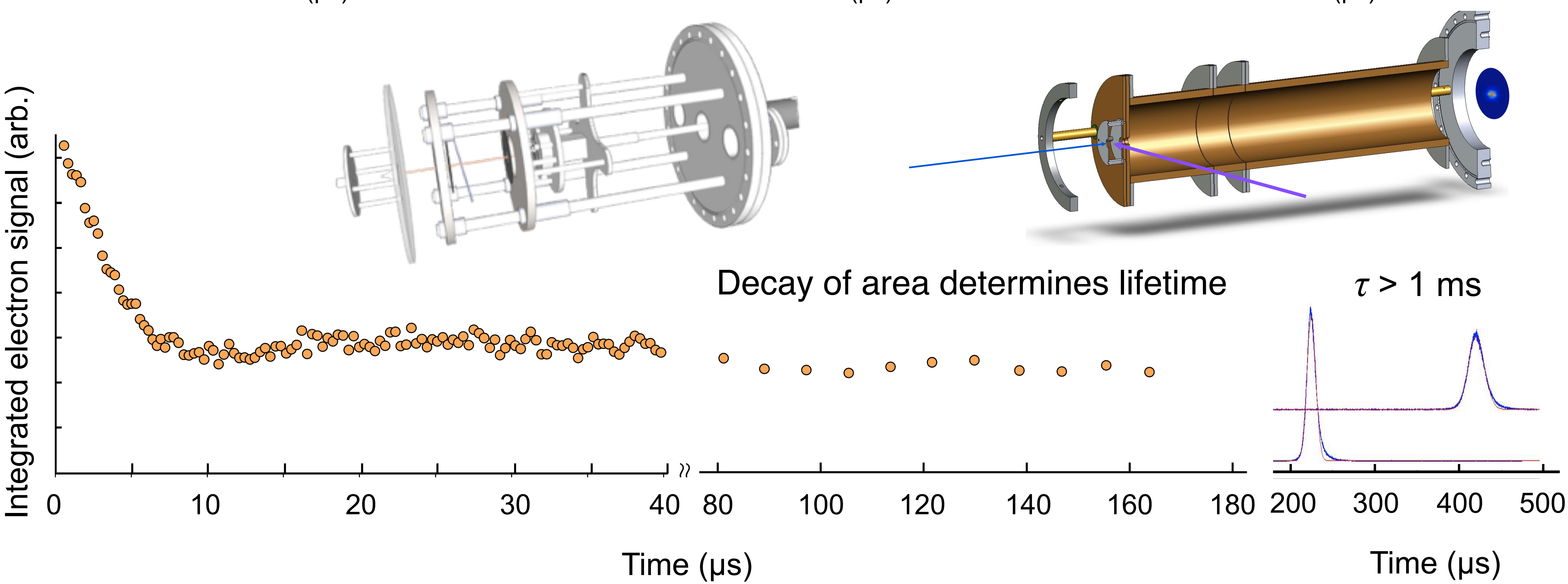

# Vlasov self-similar expansion measured in $z$
# Ion and electron thermal energy




Decay of area determines lifetime

$\tau$ > 1 ms




Confirm long life & find the missing electron energy in the long-time dynamics projected in $x$ and $y$

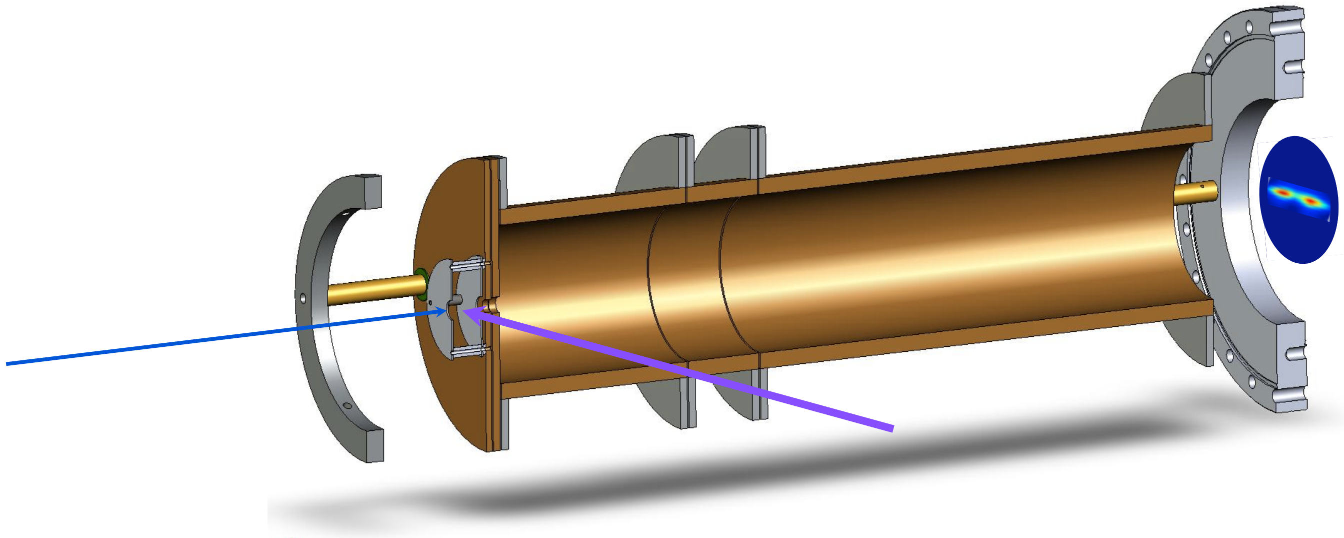
Long-time dynamics evident in plasma images projected in the *x,y* plane

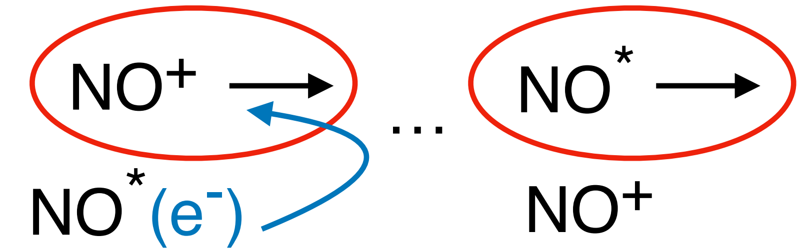
Bifurcation and quench

$$T_e \longrightarrow v_i \longrightarrow v_x$$

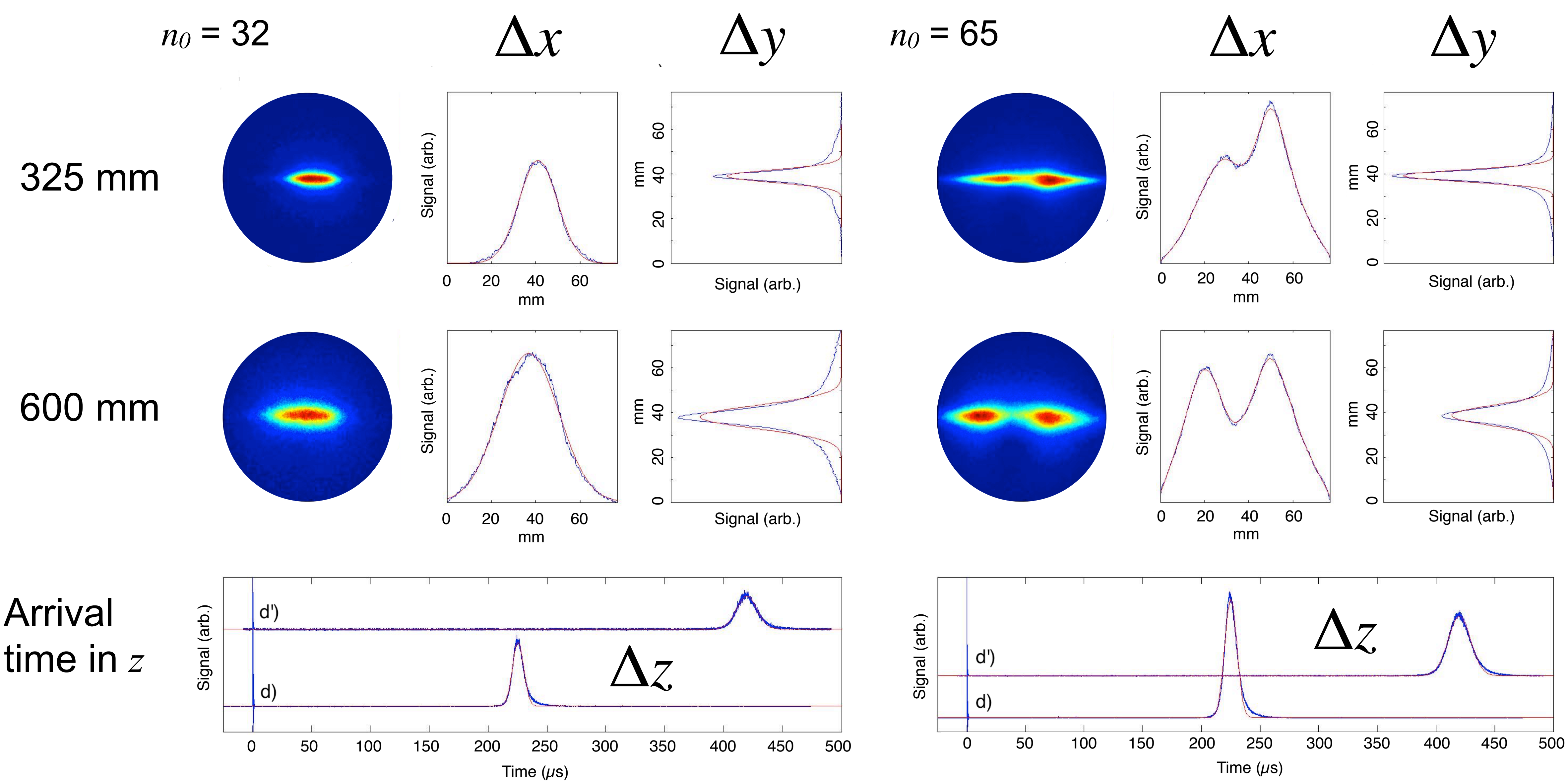
Canonical density and internal energy

Apparent effect of ion - Rydberg resonant charge exchange

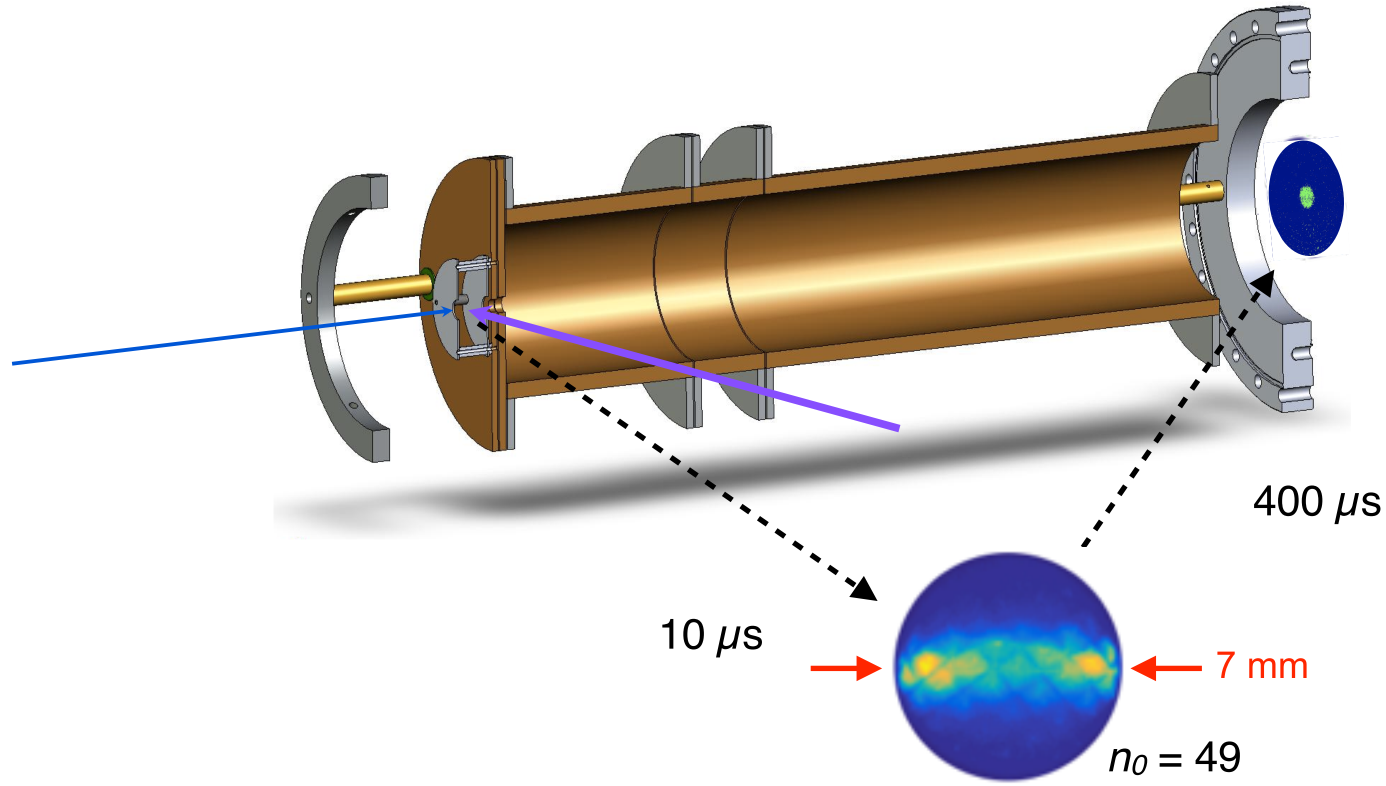




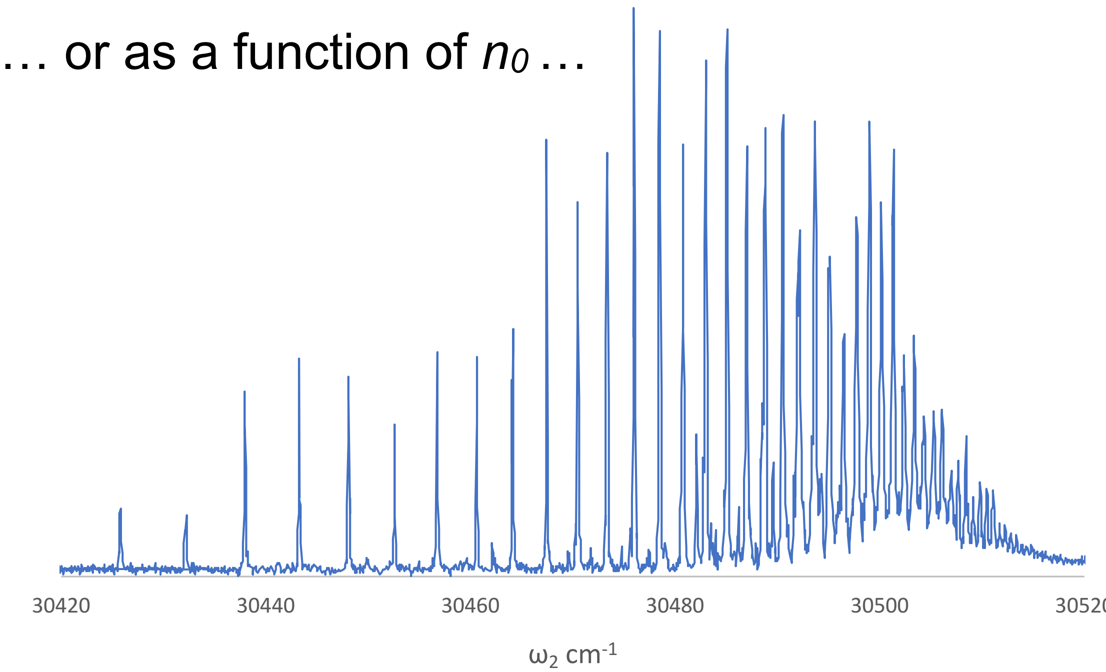

Quenches $T_e$, equalizes velocities, quenches $T_i$



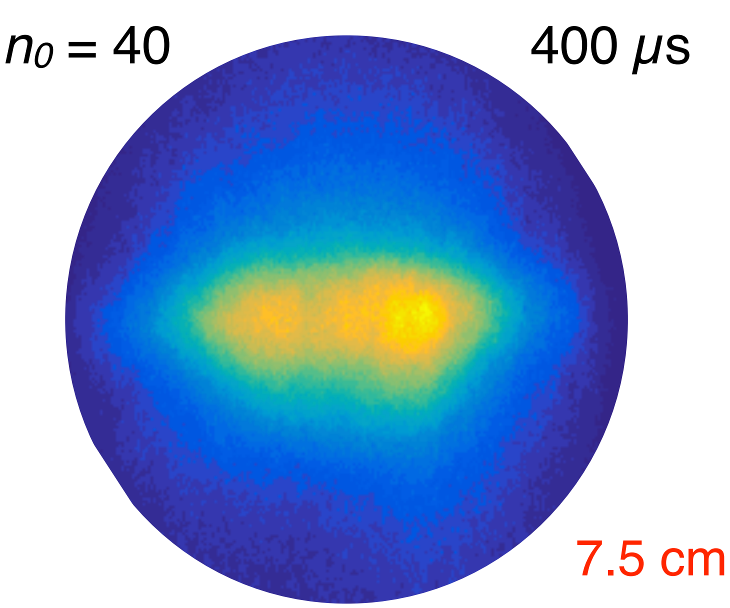

The recoil energy depends on the avalanche amplitude.

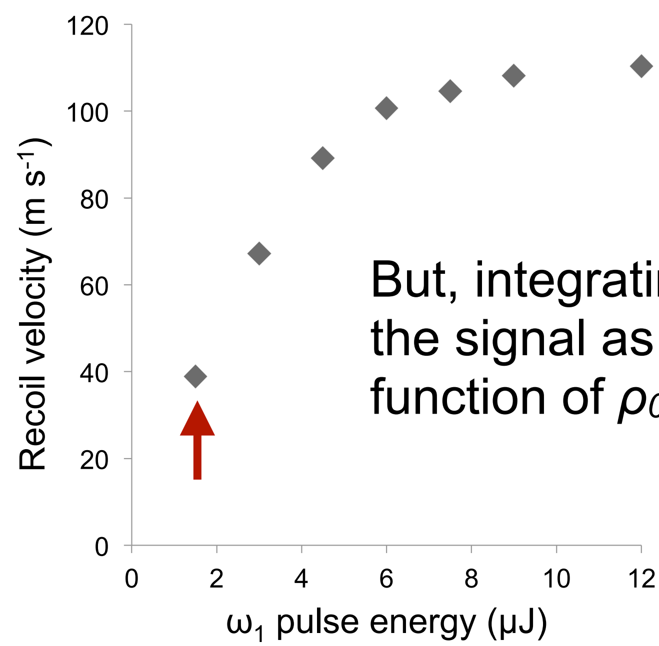



… or as a function of $n_0$ …



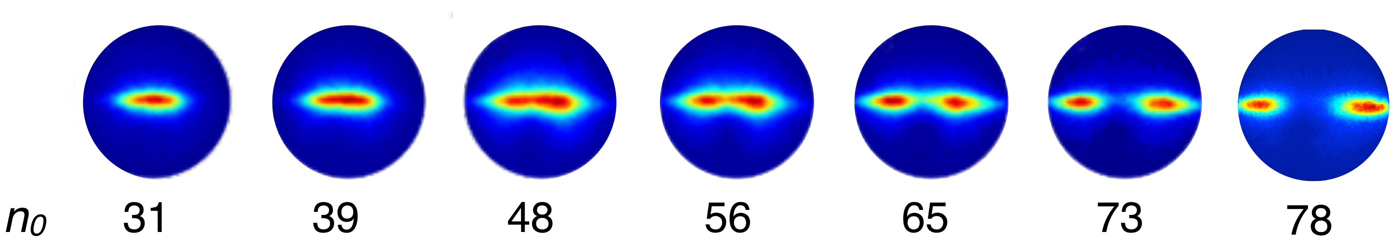

The velocity of bifurcation depends sensitively on initial Rydberg gas density …



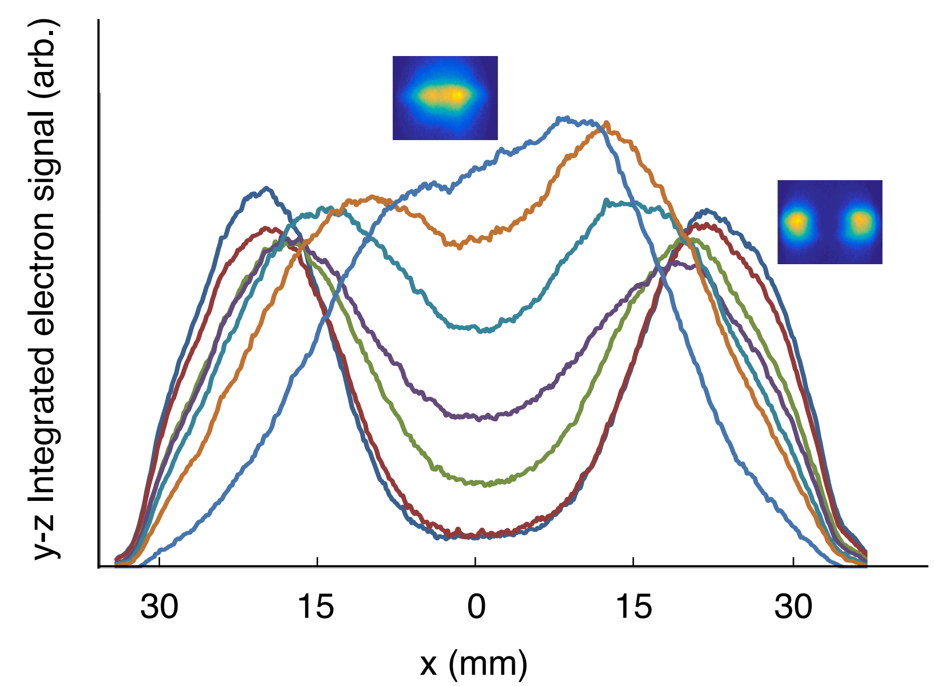




But, integrating the signal as a function of $\rho_0$ …

… and principal quantum number.

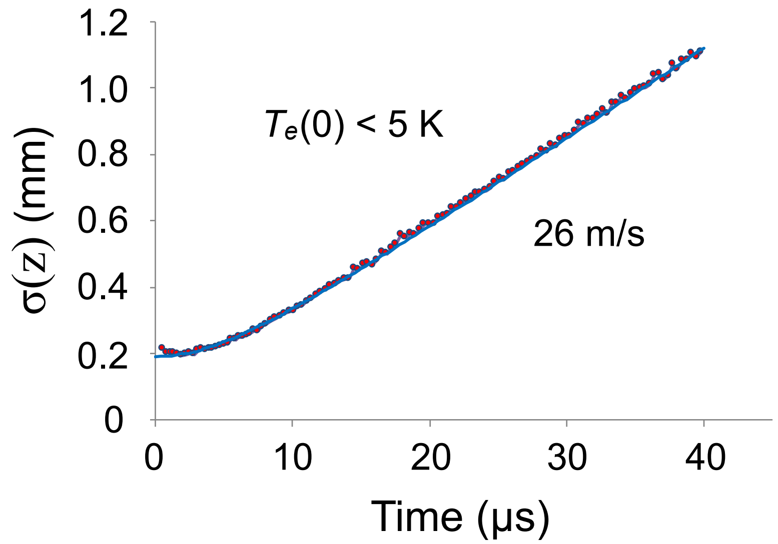


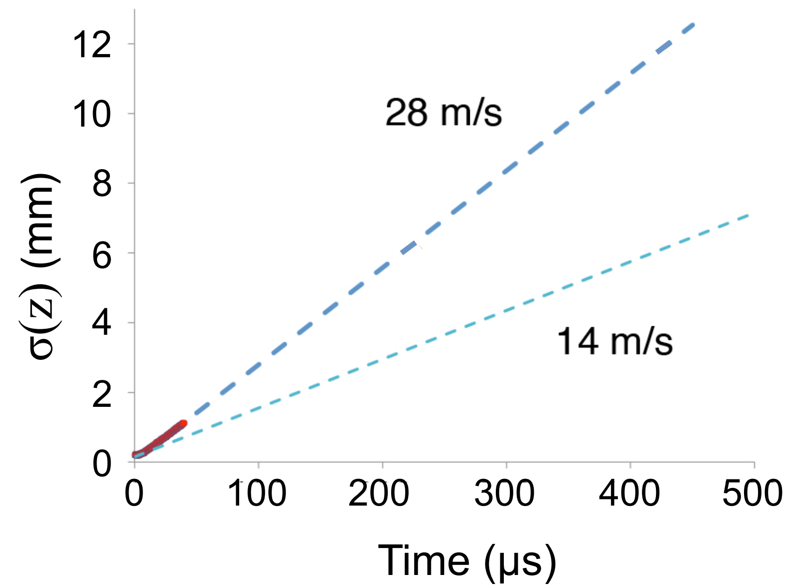

… we find that regardless of initial density, $\rho_0$ or principal quantum number, $n_0$, the plasma self-assembles to form the same final density, and …

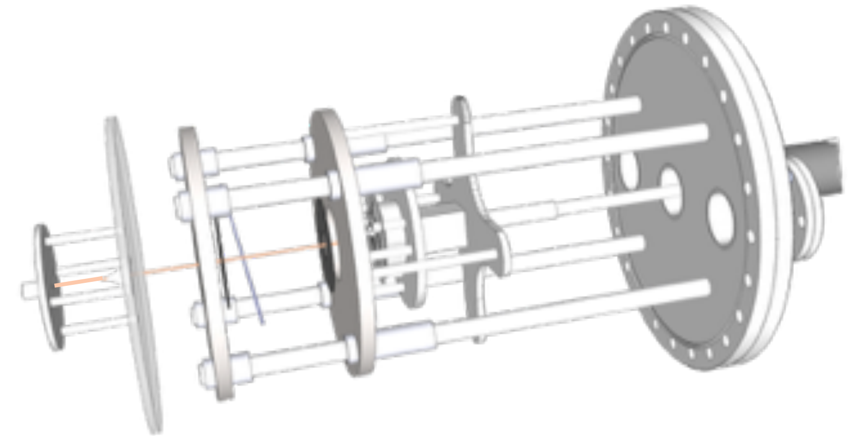



Electron signal, integrated in *y-z*, as a function of *x*

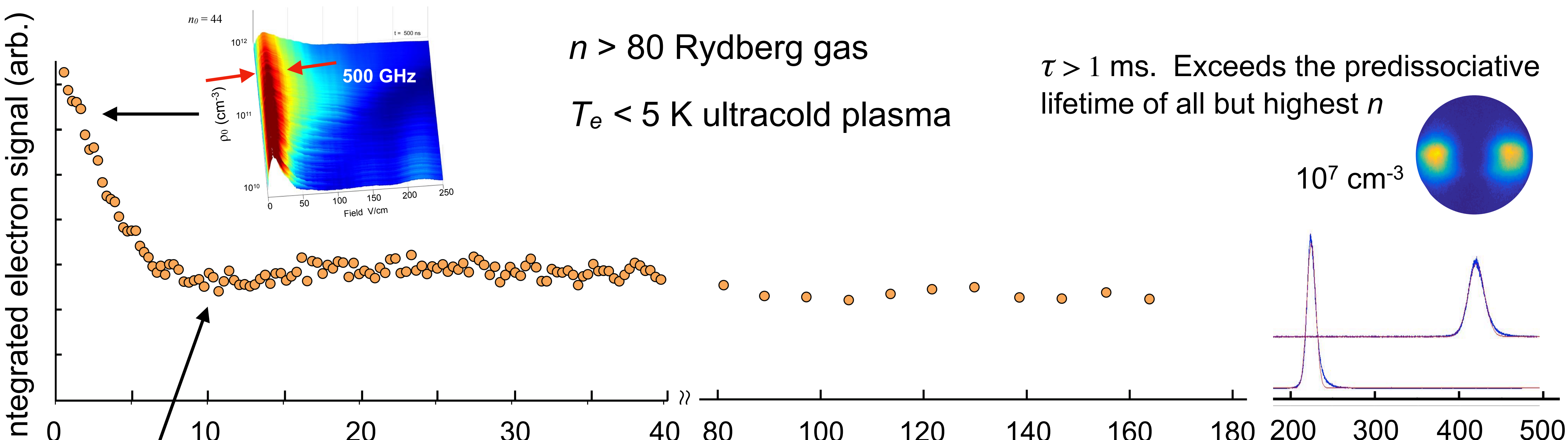
… the same internal energy …

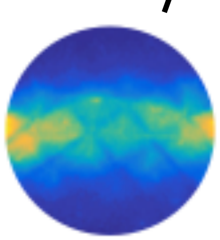


… and the same long lifetime.



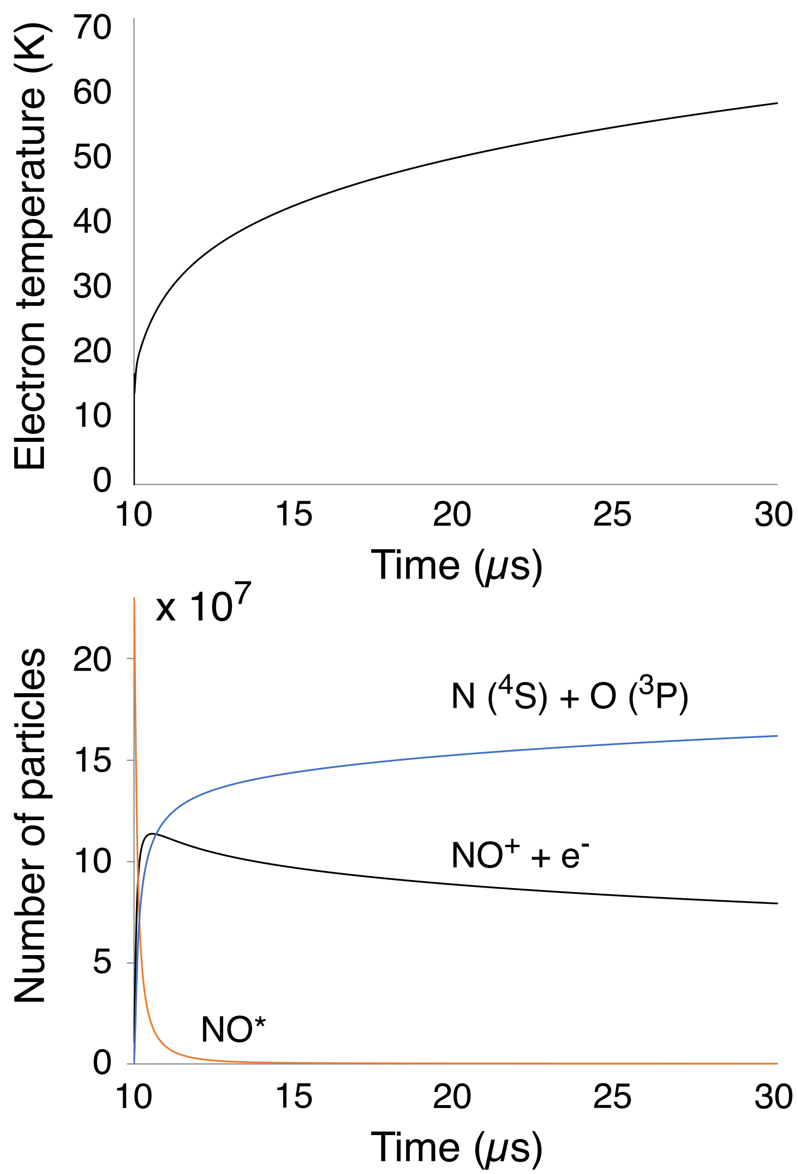



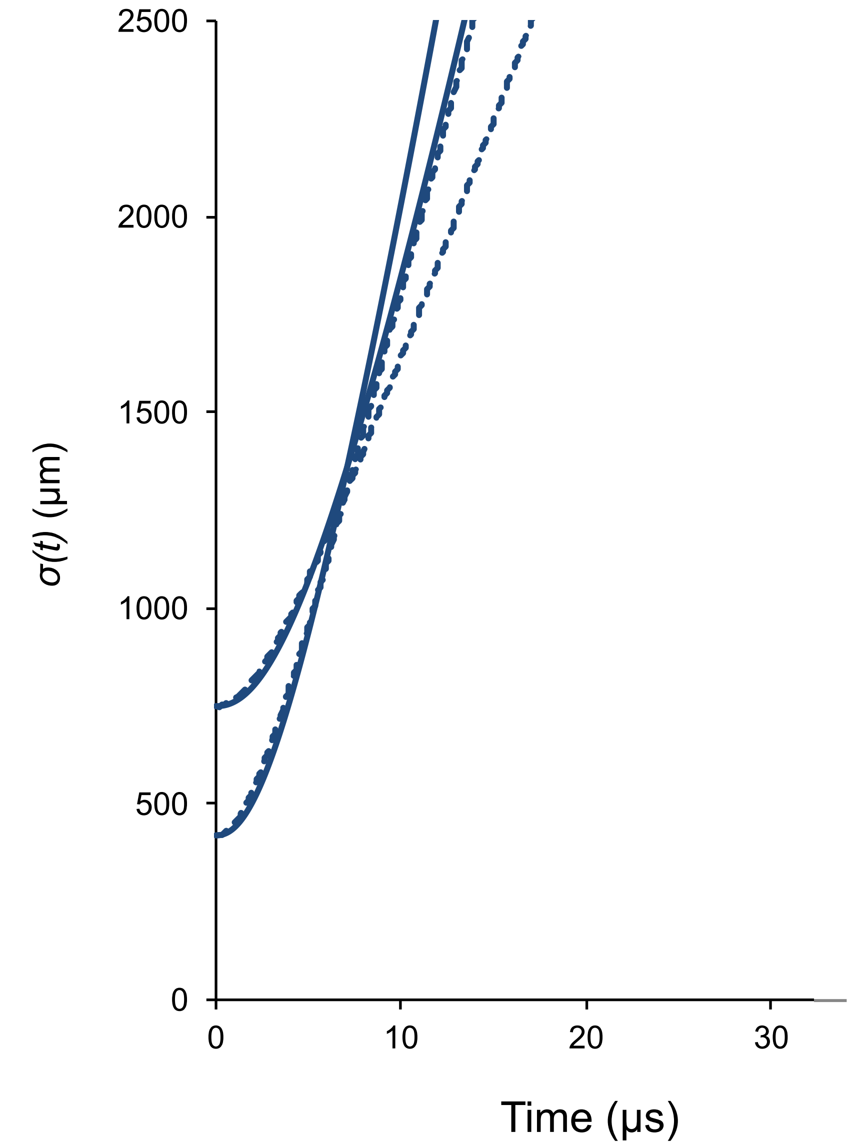

But, when we test these possible scenarios by classical simulations …

# Simulated evolution of a quenched gas of high-n Rydberg states.

A Rydberg molecule with $n$ = 80 has an orbital radius of 0.3 μm. For $\rho$ = 4 x $10^{10}$ $cm^{-3}$ simulation models predict Penning ionization and avalanche on a microsecond timescale with $T_e$ increasing to 50 K or more.

$n_0$ = 80 Rydberg gas

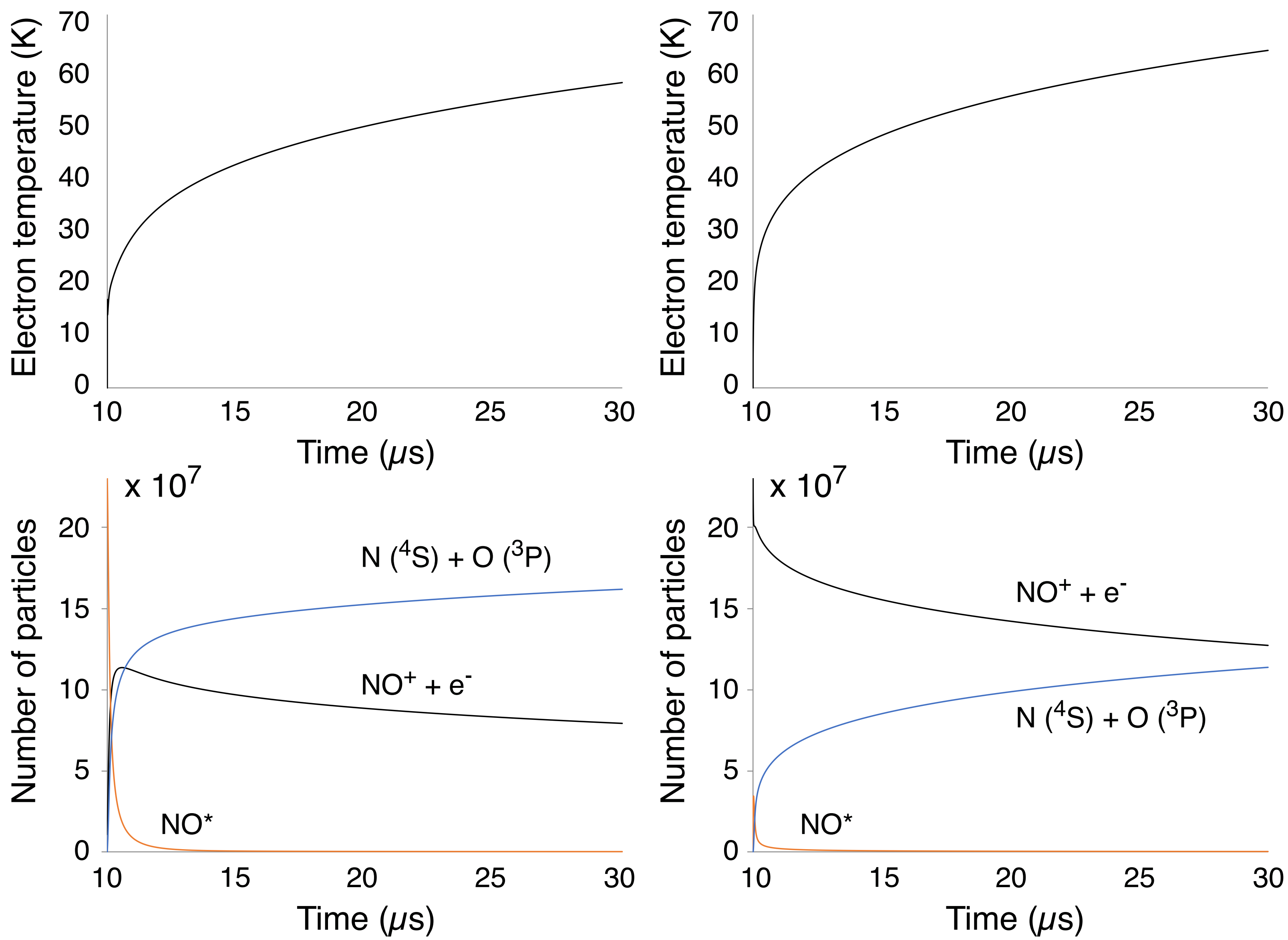


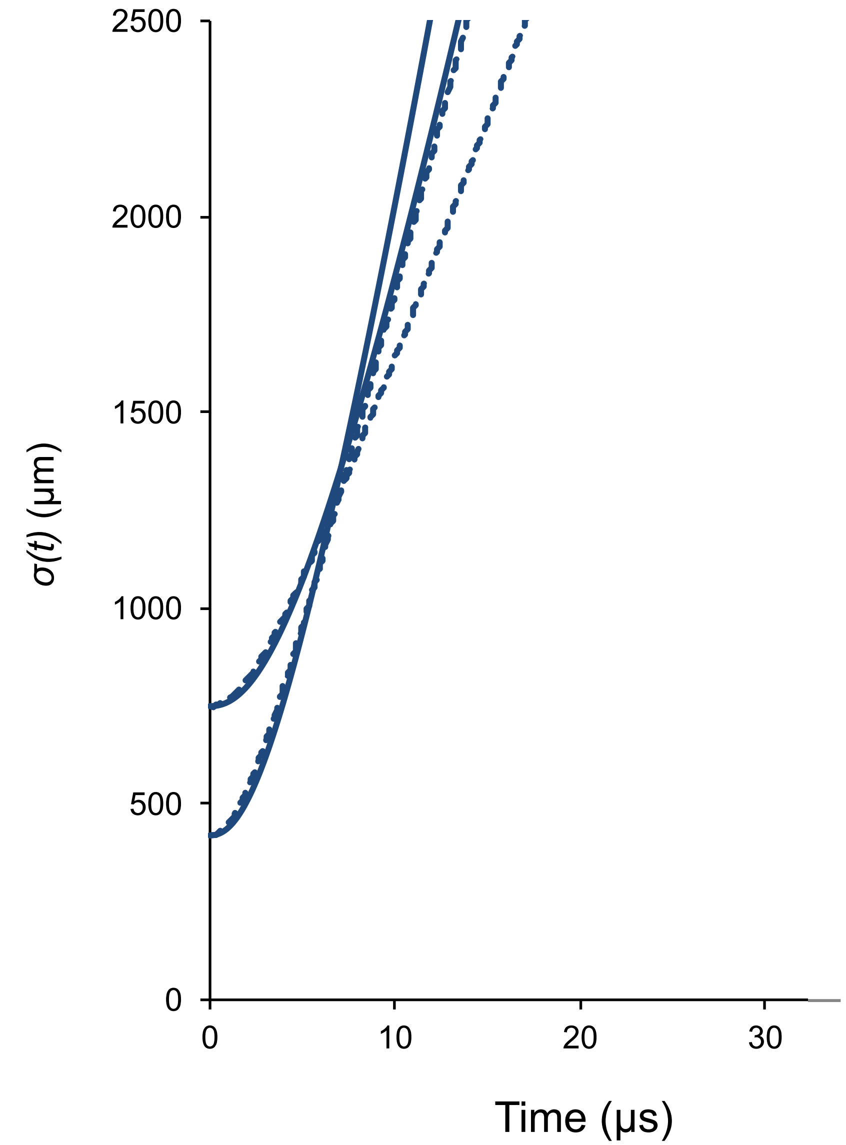



# Simulated evolution of a quenched ultracold plasma with $T_e(0)$ = 5 K

Slow expansion indicates $T_e < 5$ K. For $\rho = 4 \times 10^{10}$ cm$^{-3}$, $a_{ws}$ = 1.7 μm, and $\Gamma_e$ = 2. Coupled rate-equation models and MD simulations call for correlation energy release, three-body recombination, Rydberg relaxation and <u>significant electron heating</u>.

$n_0$ = 80 Rydberg gas $\qquad$ $T_e$ < 5 K ultracold plasma



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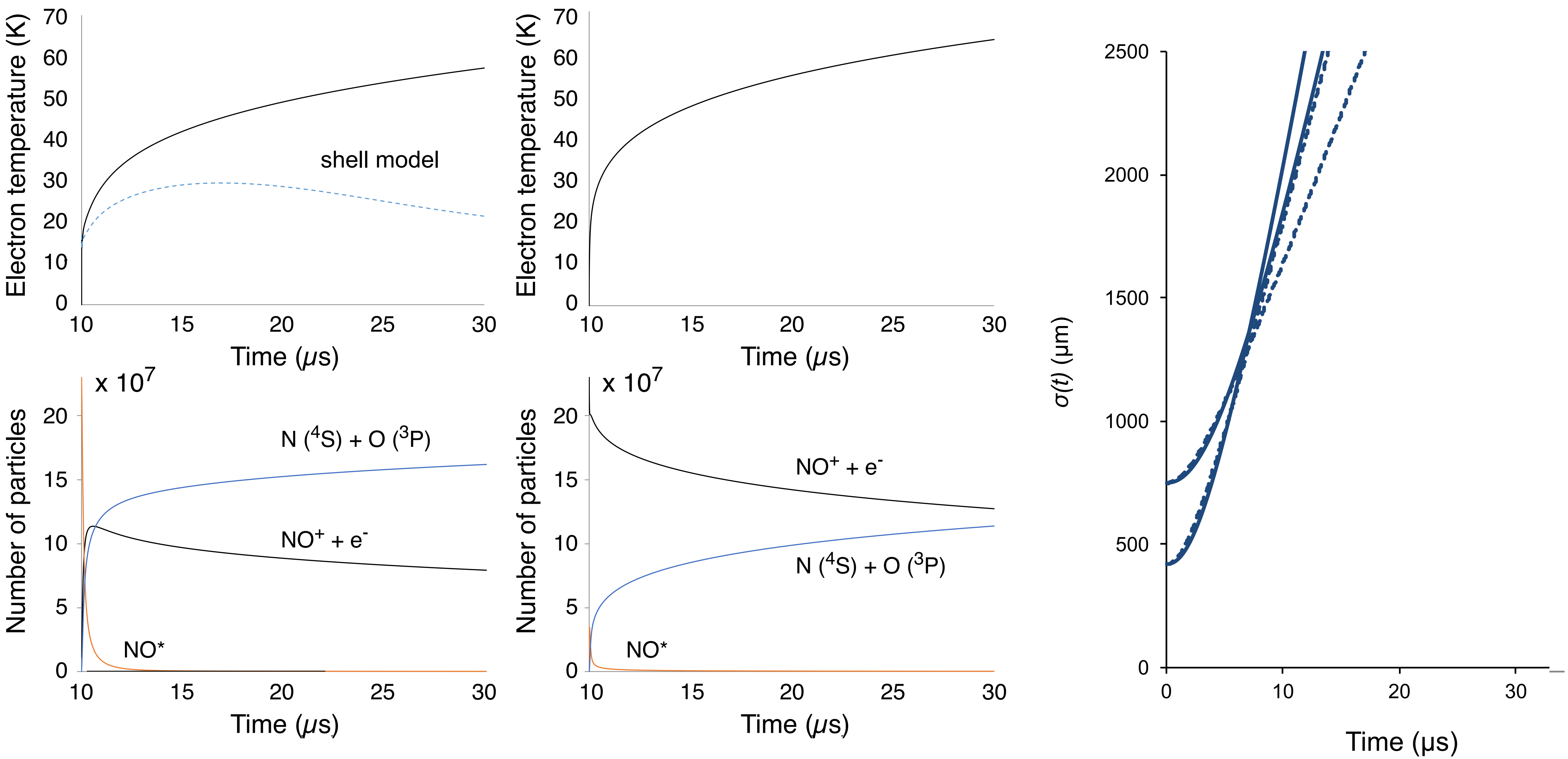
$n_0$ = 80 Rydberg gas

$T_e$ < 5 K ultracold plasma



# Simulated evolution of a quenched ultracold plasma with $T_e(0)$ = 5 K

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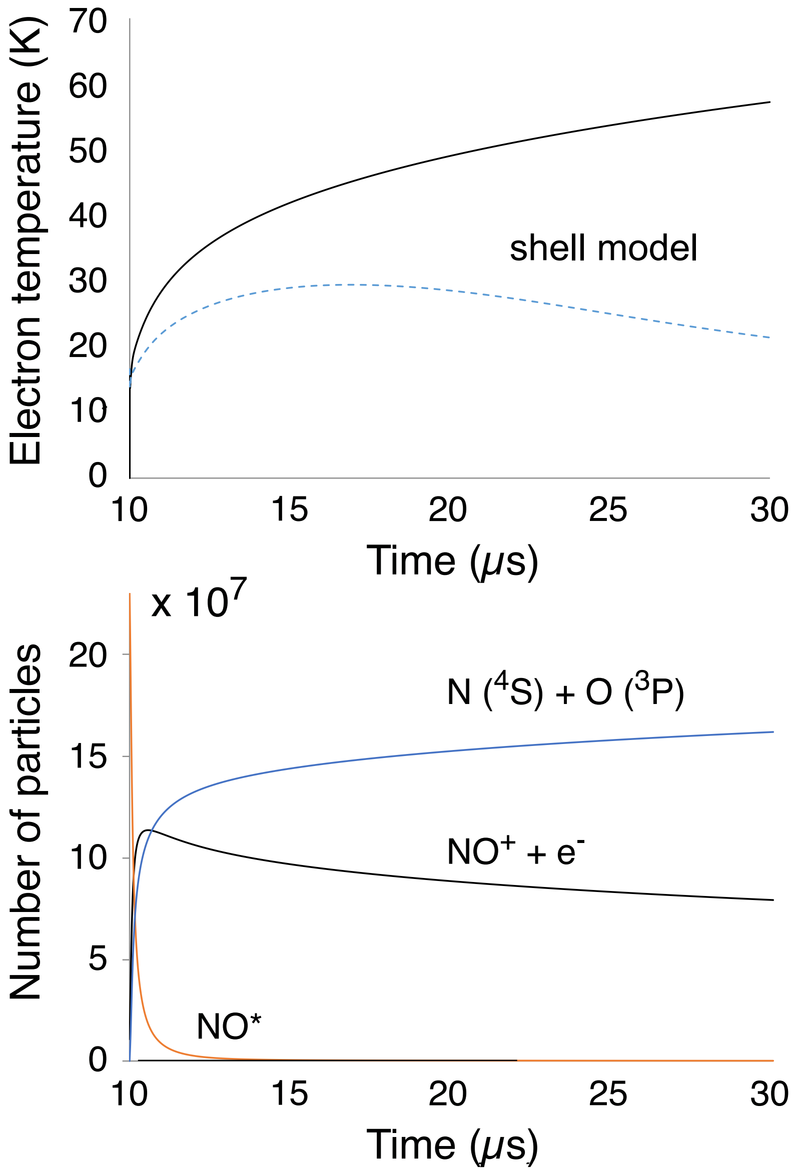
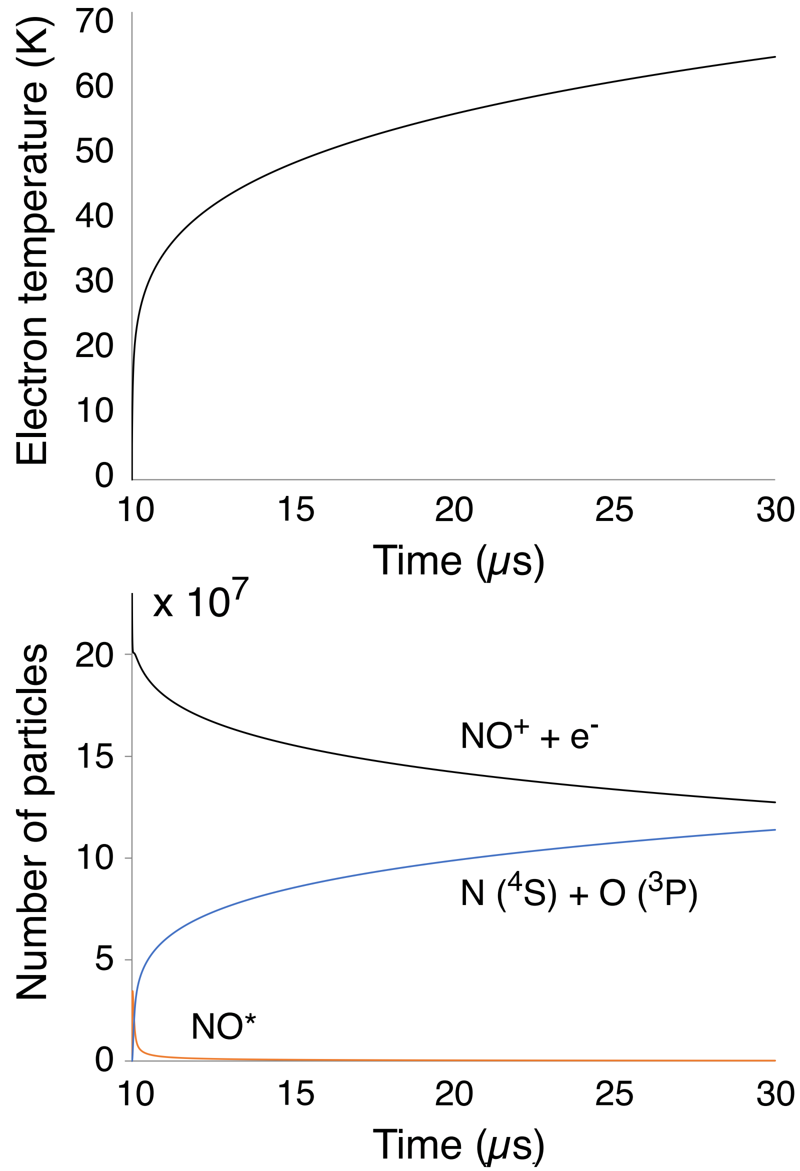
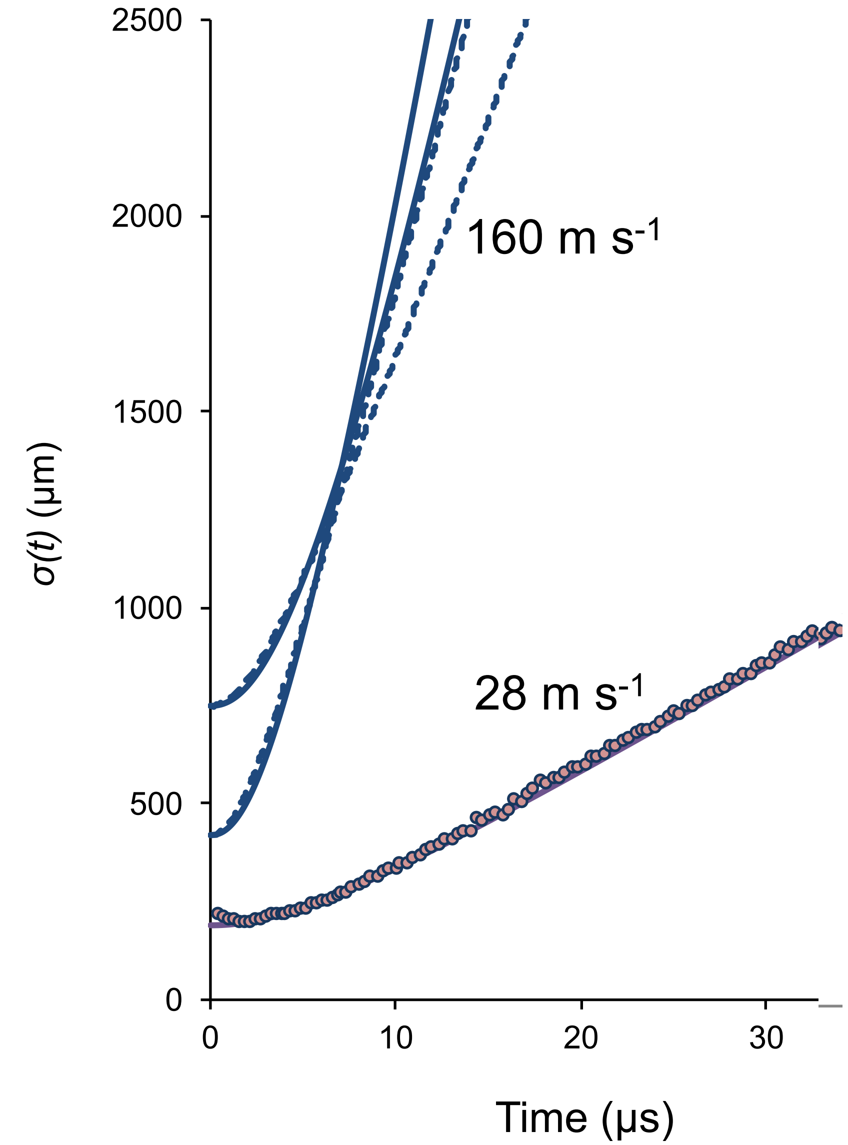
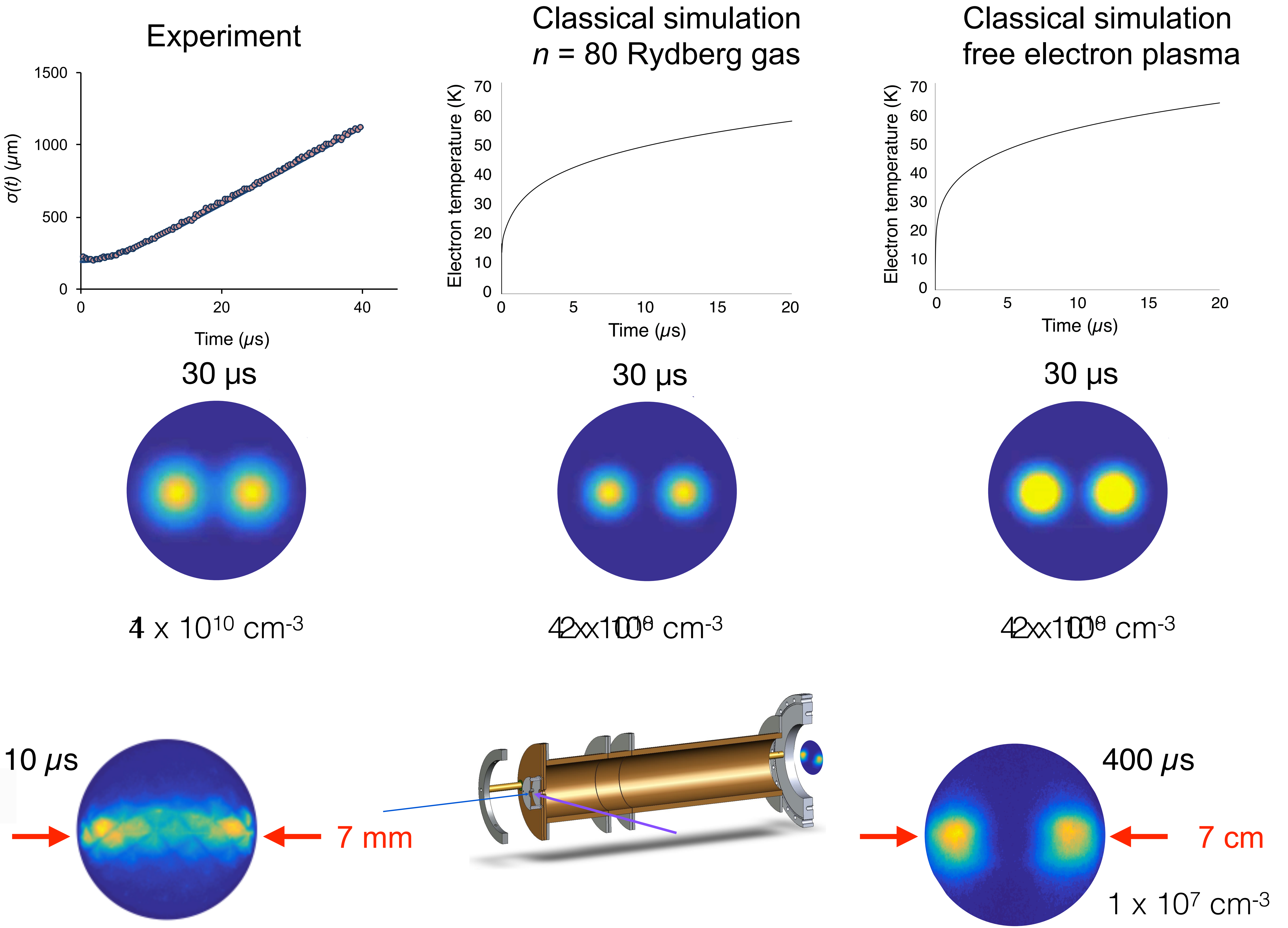



$T_e$ < 5 K ultracold plasma

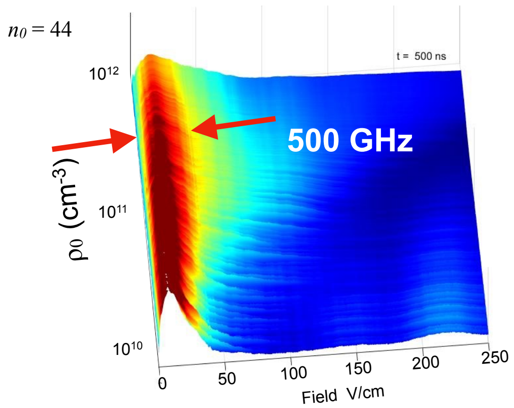




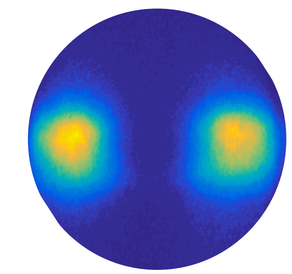
Experiment
Classical simulation
n = 80 Rydberg gas
Classical simulation
free electron plasma
σ(t) (μm)
1500
1000
500
0
0
20
40
Time (μs)
Electron temperature (K)
70
60
50
40
30
20
10
0
0
5
10
15
20
Time (μs)
Electron temperature (K)
Time (μs)
30 μs
30 μs
30 μs
10 μs
7 mm
400 μs
7 cm
1 x 10^7 cm^-3

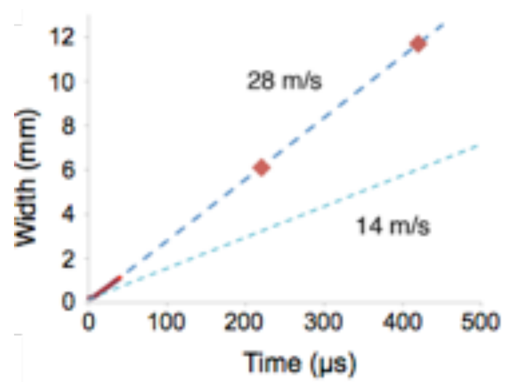
# Properties of the arrested state determined by experiment



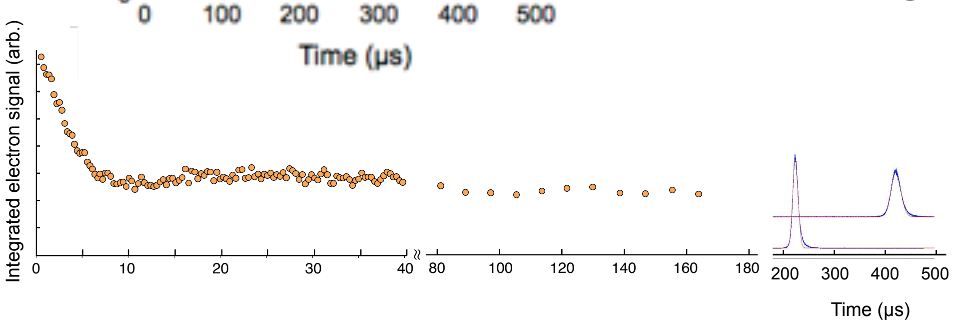

***Avalanched***, as clearly measured by field ionization spectrum. Evidence for electrons bound by low energy to individual ions (Rydberg molecules, $n_0$ > 80) or to multiple ions (plasma electrons).

***Quenched***, as demonstrated by bifurcation to separating volumes with very little internal energy





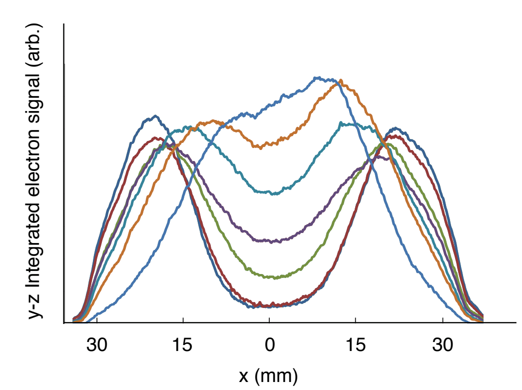

***Cold***, as confirmed by very slow plasma expansion, indicating in particular a low electron temperature.



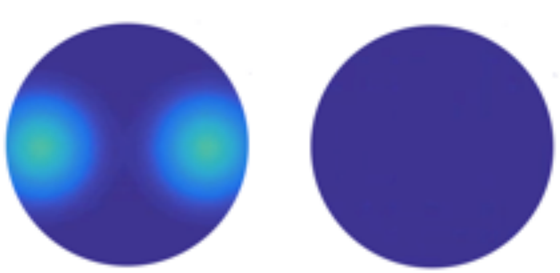

***Stable***, little if any dissociation of nitric oxide observed

$NO^+ + e^- \longrightarrow NO^* \longrightarrow N\,(^4S) + O\,(^3P)$ after 10 μs,

despite predissociative lifetime of 1 μs averaged over $l$ for $n_0$ = 80.




***Universal***, self-assembles to an arrested phase of common density and internal energy, regardless of starting conditions.

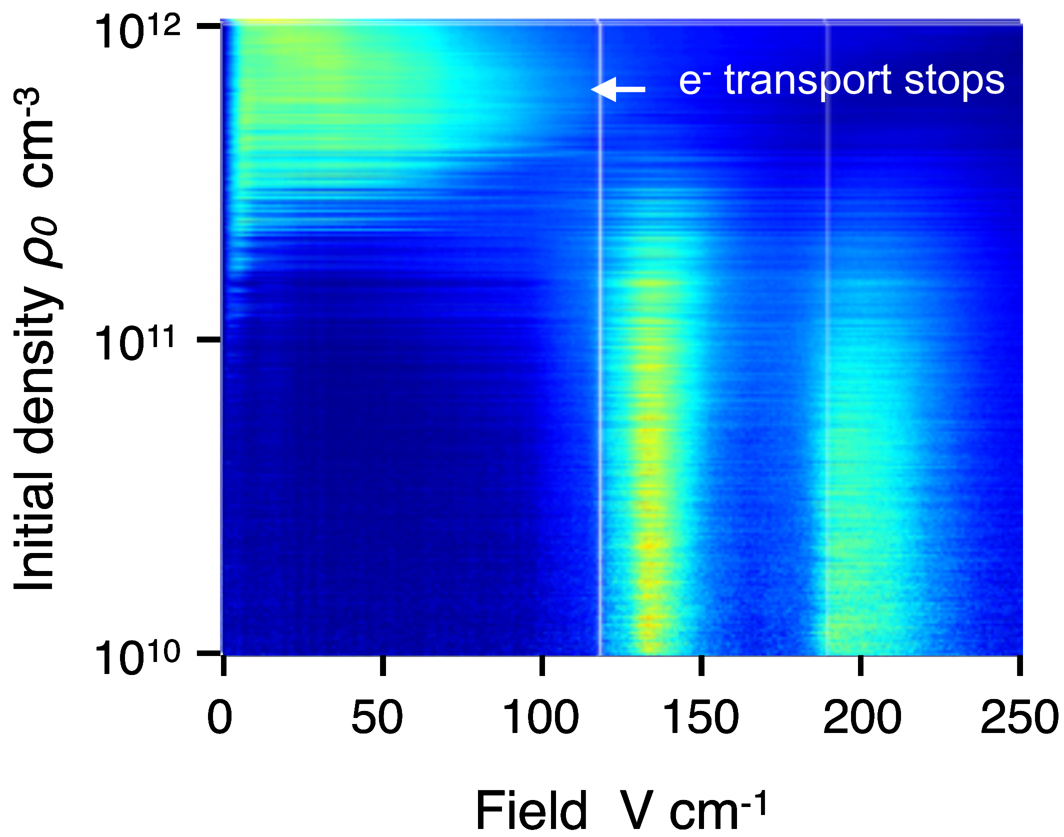


***Non-classical***. Observed long-time dynamics of this phase do not accord with classical coupled-rate equation simulations.

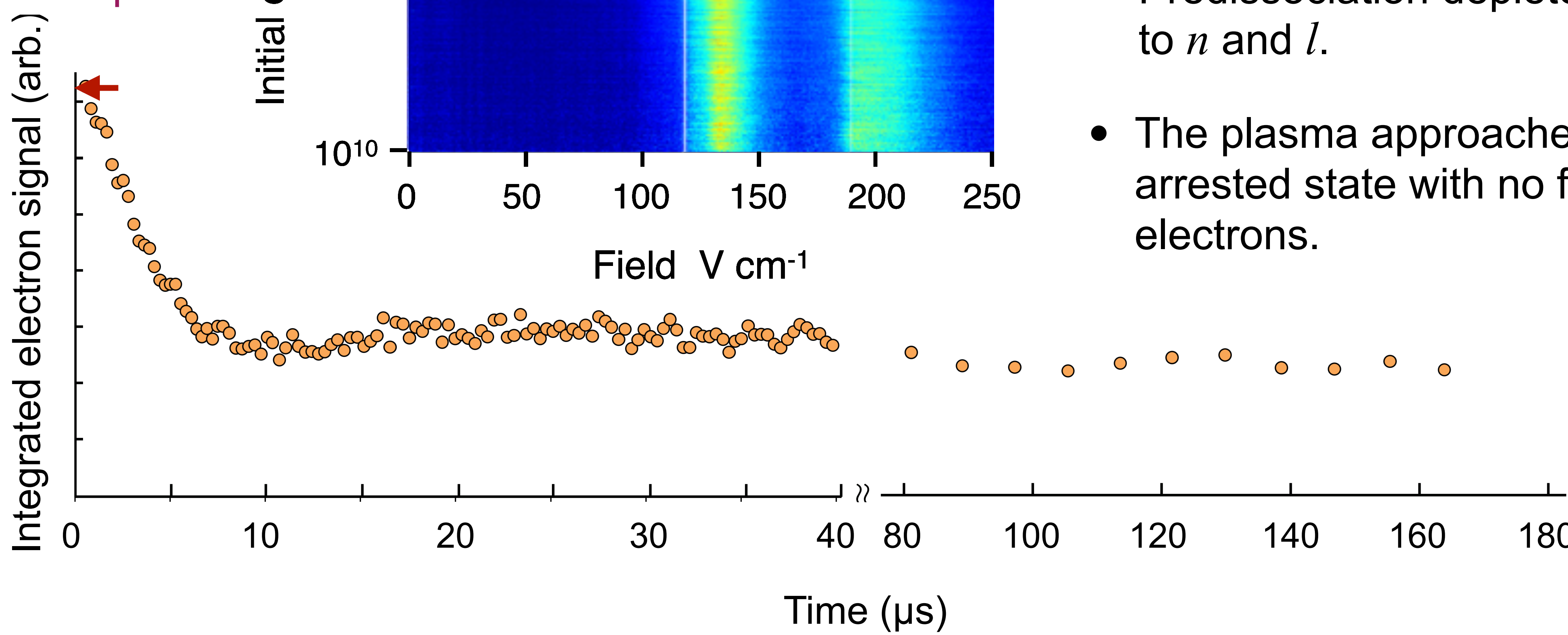
# Path to the arrested state

- At high density avalanche proceeds on a ns timescale: Initial energy transport by electron - Rydberg collisions (**classical**).

44 *f(2)*




□ □ □ 4 □ μs



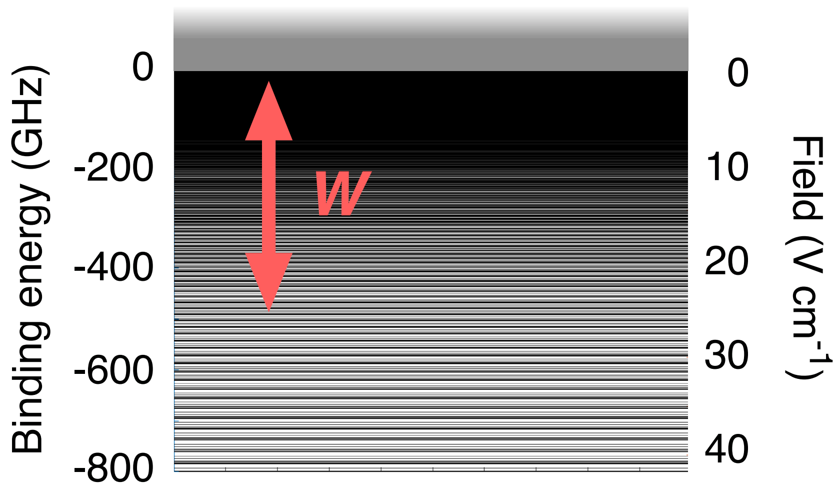

- Watch for effects of electron - NO* collisional *l*-mixing. 44*f*(2) bands move to higher field.
- Transport driven by electron - Rydberg collisions.
- After 150 ns, transport at high density stops.
- Predissociation depletes according to *n* and *l*.
- The plasma approaches an arrested state with no free electrons.

The delayed SFI spectrum signifies Rydberg electrons weakly bound to a single $NO^+$ ion, or the exciton-like state of an electron bound to a more distant $NO^+$ ion immersed in an $NO^+$ - $e^-$ dielectric.

# Energy transport in a basis of Rydberg molecules and $NO^+$ -- $e^-$ excitons

## Basis states

high positive energy : (expansion)



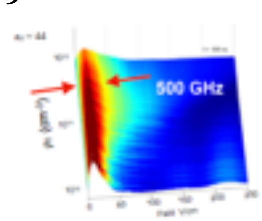

high negative energy : (dissociation)

$$H_{\rm d} = \sum_i \left( \frac{\mathbf{P}_i^2}{2m} + h_i \right) + \sum_{i,j} V_{ij}^{\rm dd}$$

$h_i$, complicated for a Rydberg molecule, extends with slightly greater complexity to describe an $NO^+$ - $e^-$ exciton in a back-ground ion-$e^-$ dielectric

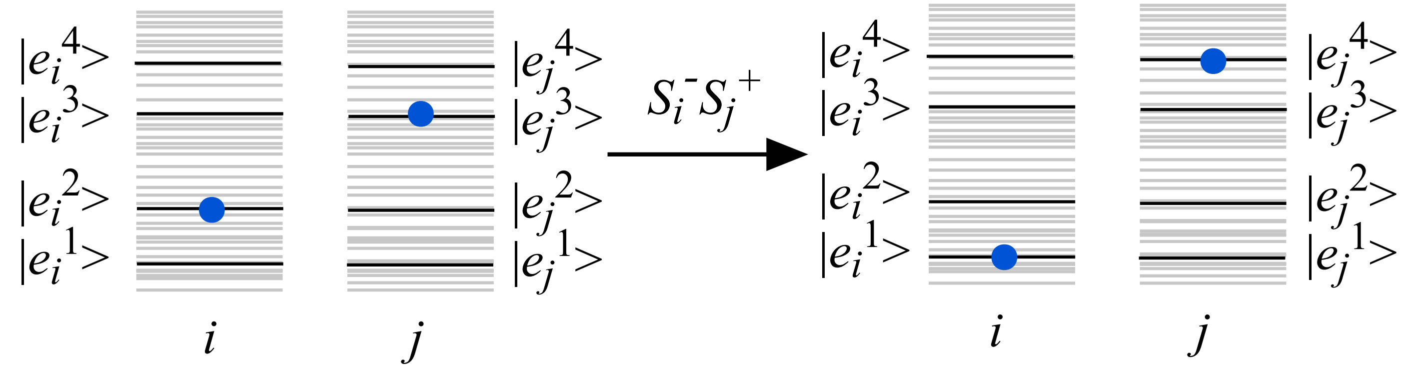
$$\{ |e_1\rangle, |e_2\rangle, |e_3\rangle \dots \}$$

Energies, $\epsilon_i$, span $W$




## Dipole-dipole coupling drives flip-flop state mixing interaction

Conceptualize in low order:




$$J_{ij} = t_{ij}/r_{ij}^3 \qquad U_{ij} = D_{ij}/r_{ij}^6 \approx J_{ij}^2 \widetilde{J}/W^2$$

$$|\downarrow_i, \uparrow_j, \uparrow_k\rangle \xrightarrow{\hat{S}_i^+ \hat{S}_j^-} |\uparrow_i, \downarrow_j, \uparrow_k\rangle \xrightarrow{\hat{S}_j^+ \hat{S}_k^-} |\uparrow_i, \uparrow_j, \downarrow_k\rangle \xrightarrow{\hat{S}_k^+ \hat{S}_i^-} |\downarrow_i, \uparrow_j, \uparrow_k\rangle$$

Dipoles $i$ and $j$ couple via $k$ in a third-order process giving rise to a self-interaction described by an Ising term with an amplitude, $U_{ij}$

Burin, *Phys Rev B* **92** 104428

Collect in a spin model with dipole-dipole and Ising terms:

$$H_{\rm eff} = \sum_i \epsilon_i \hat{S}_i^z + \sum_{i,j} J_{ij}(\hat{S}_i^+ \hat{S}_j^- + h.c.) + \sum_{i,j} U_{ij} \hat{S}_i^z \hat{S}_j^z$$

To gauge properties, assume a state of localization and then ask whether delocalizing perturbations destabilize this phase

Sous & Grant, *PRL* **120** 110601, arXiv:1808.07479

# Energy transport in a basis of Rydberg molecules and $NO^+$ -- $e^-$ excitons

Spin flip-flop interactions form resonantly coupled pair states $|\downarrow_i, \uparrow_j\rangle \leftrightharpoons |\uparrow_i, \downarrow_j\rangle$

Long-range dipole-dipole and Ising interactions form off-diagonal matrix elements that allow energy to propagate $|\downarrow_i, \uparrow_j\rangle \leftrightharpoons |\uparrow_i, \downarrow_j\rangle \longleftrightarrow |\downarrow_k, \uparrow_l\rangle \leftrightharpoons |\uparrow_k, \downarrow_l\rangle$

In a shell from $R_{xy}$ to $2R_{xy}$, a central spin finds $N_{xy}$ resonant interactions: $\frac{t_{ij}}{r_{ij}^3} \geq |\epsilon_i - \epsilon_j| \in W$

$$N_{xy} = (\rho R_{xy}^3)\frac{t/R_{xy}^3}{W} = \rho\frac{t}{W}$$

Critical. Does not diverge with system size.

Ising interactions mix resonant pairs. For a density of pseudospins $\varrho = \rho N_{xy}$, a central pair finds $N_z$ resonant interactions in a shell from $R_z$ to $2R_z$:

$$N_z = \varrho R_z^3 \frac{D/R_z^6}{t/R_{xy}^3} = \rho^2 \frac{t}{W} R_z^3$$

for an extended-pair relation between $R_{xy}$ and $R_z$. Diverges as system volume. Yao et al., *PRL* **113** 243002

Ising interactions sufficient to delocalize system when $U(R) \approx D\varrho^2$ exceeds $J(R) \approx \frac{J}{\rho R^3}$ .

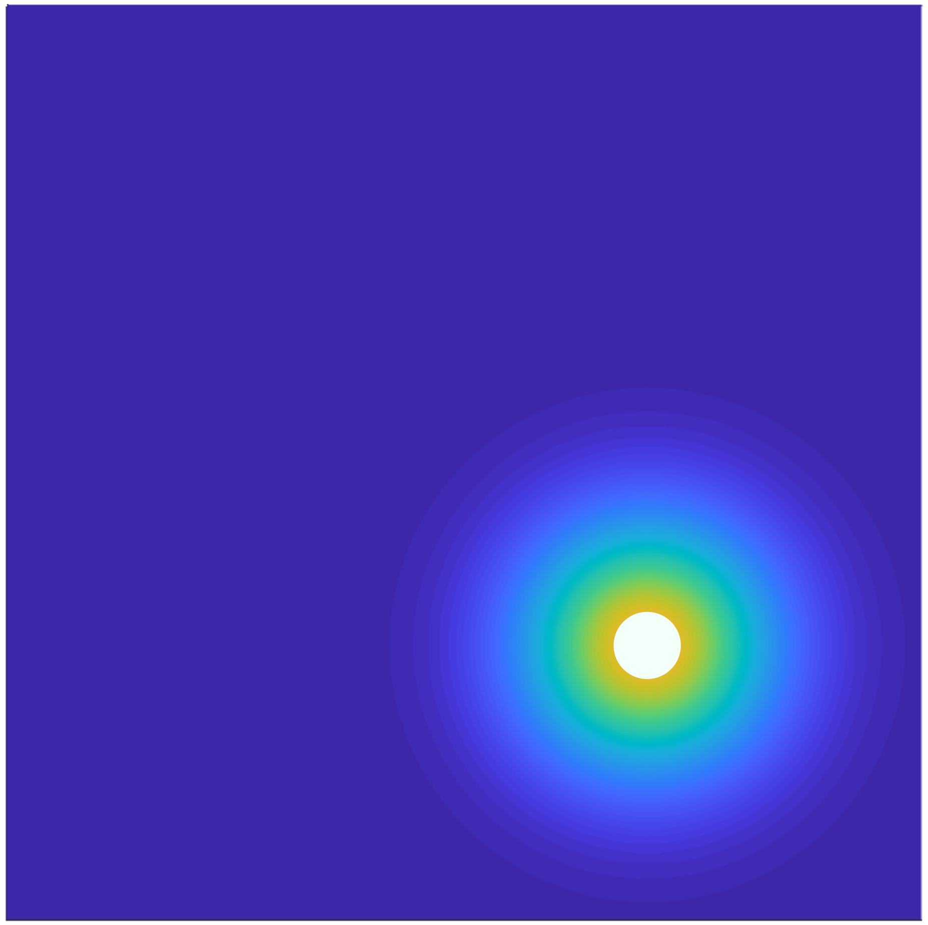
A system of density $\rho$ extended to a distance $R_c$ such that $U(R_c) = J(R_c)$ contains $N_c$ dipoles:

$$N_c = \left(\frac{W}{J}\right)^4$$

For $J = 2$ GHz, $W = 500$ GHz, $N_c = 3 \times 10^9$

Upon arrest, the ultracold plasma contains 100 times fewer than $N_c$ dipoles

# Inevitable fluctuations create locally thermalized volumes - Griffiths regions.



**The molecular ultracold plasma intrinsically opposes delocalization.**

Consider a thermal inclusion in an MBL bulk.

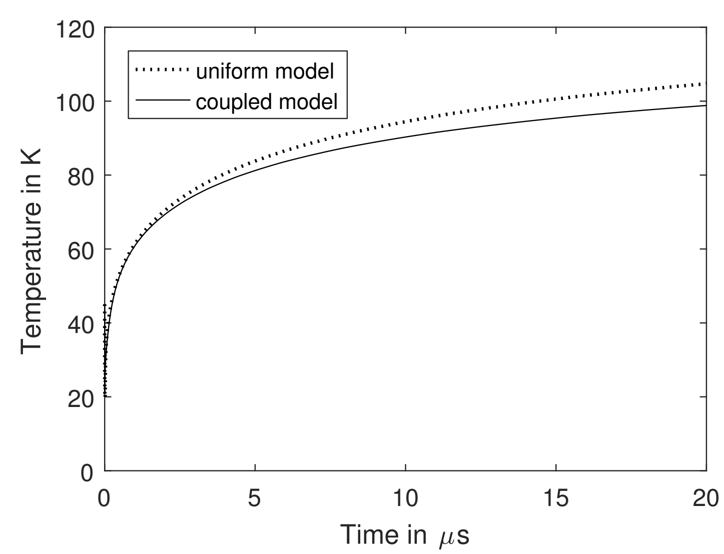
Thermal core mixes *l*-bits according to $e^{-R/\zeta}$,
$\zeta$ = decay length

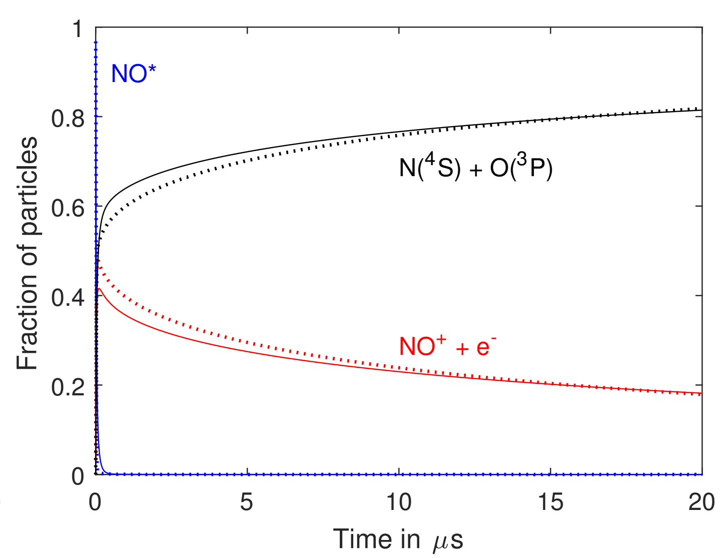
As a classical region, the thermal ultracold plasma inclusion has a characteristic signature: Avalanche dynamics owing to the collisions of Rydberg molecules with free electrons.

High-frequency electron-Rydberg collisions increase $T_e$, and drive Rydberg population to lower $n$, where predissociation rapidly causes the plasma to dissipate as neutral N($^4$S) + O($^3$P).

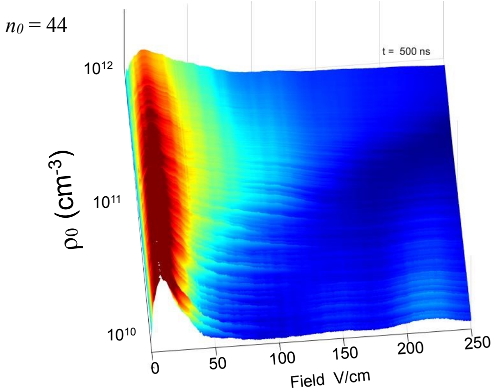
This loss of plasma ions and energetic molecules reduces the density of dipoles, and creates a weakened bath (sparser distribution of increased level spacings).

In this state of diminished mixing, the Griffiths region dissipates to a void of no consequence.

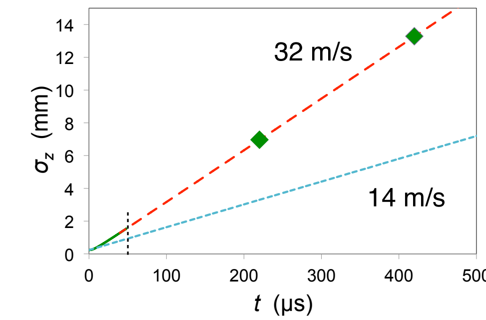


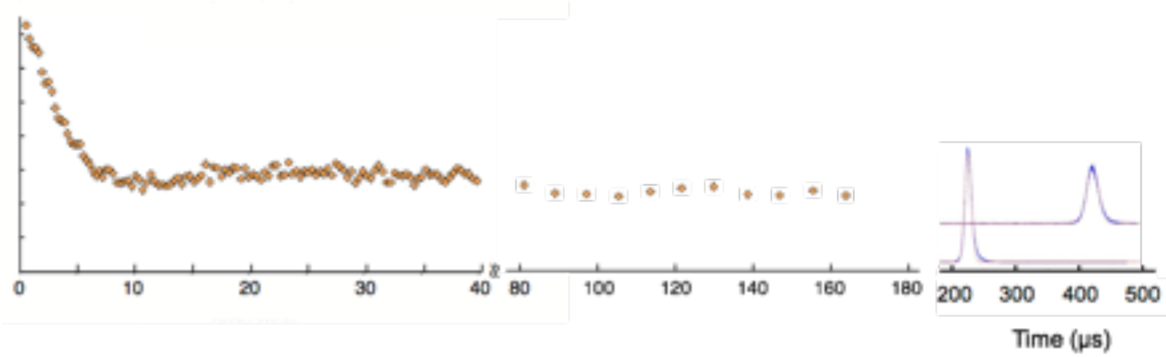
# Conclusions



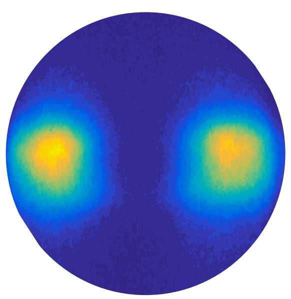

Fast avalanche to plasma in NO.



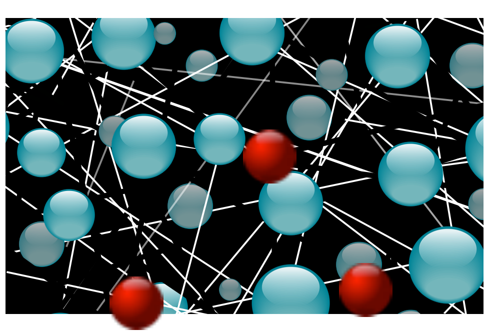

Quench. Anomalously slow plasma expansion.




Predissociation halts.

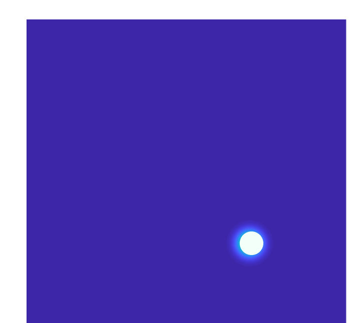


Images confirm liquid-like behaviour in which plasma irreversibly sequesters energy in a reservoir of mass transport



Dynamics suggest a robust process of self-organization to reach a state of arrested relaxation, far from thermal equilibrium. Disorder on a scale that appears to inhibit energy transport from Rydberg molecules to electrons.

Predissociation in the molecular ultracold plasma may act to diminish the delocalizing power of Griffiths regions



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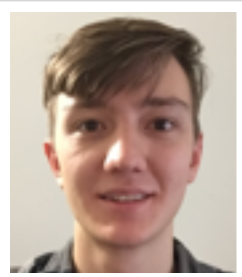
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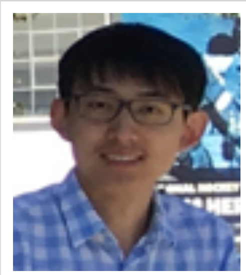
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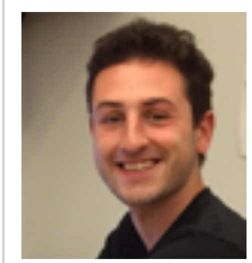
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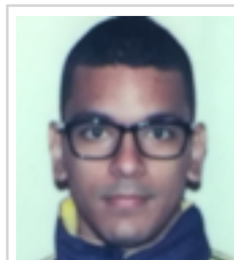
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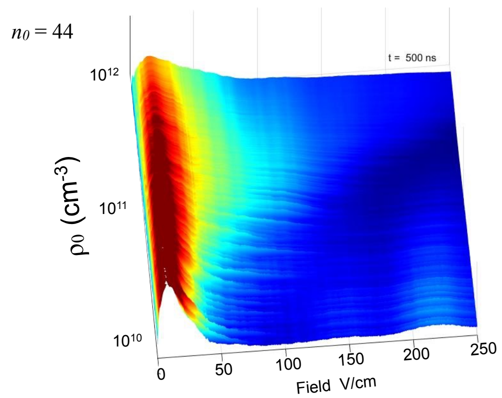
# Conclusions

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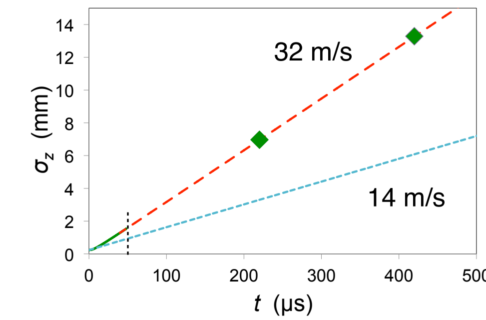


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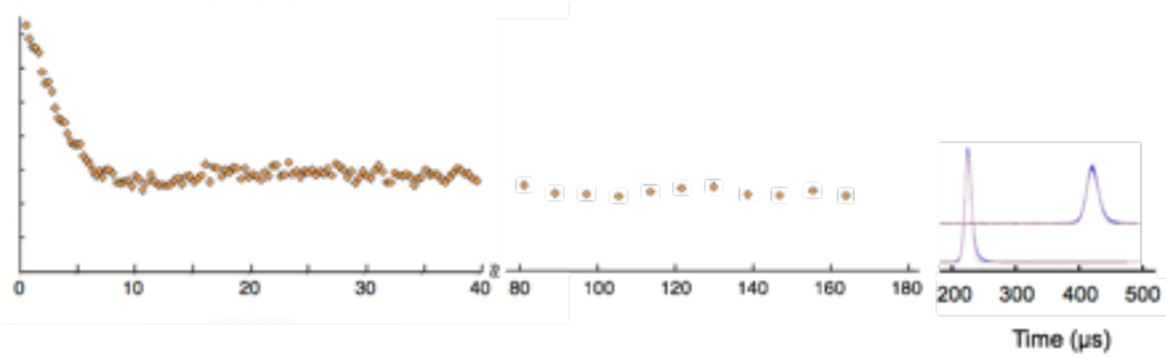


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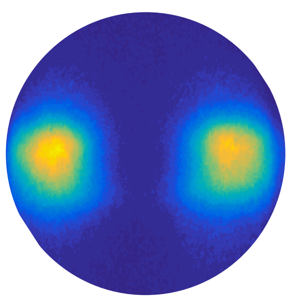

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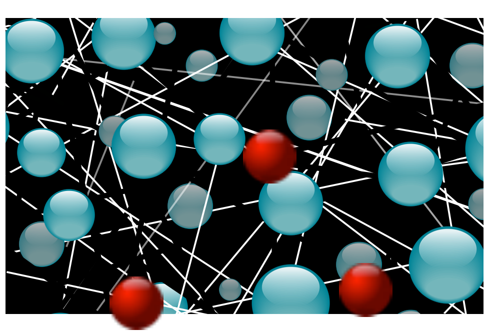

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