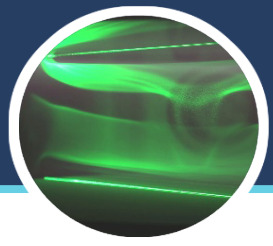


# NON-LINEAR PHENOMENA IN DUSTY PLASMAS: MIXED-MODE OSCILLATIONS

*M. Mikikian*

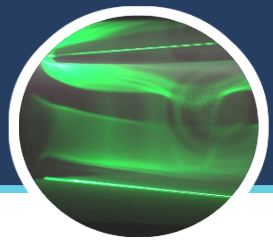
*GREMI, UMR7344 CNRS/Univ. Orléans, F-45067 Orléans, France*





# OUTLINE

- ❑ **Dusty (or complex) plasmas?**
- ❑ **Nanoparticle formation in a few words ...**
- ❑ **Experimental setup**
- ❑ **The "void"**
- ❑ **Void instabilities: Heartbeat**
- ❑ **Last beats: Mixed-Mode Oscillations**



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- ☐ Void instabilities: Heartbeat
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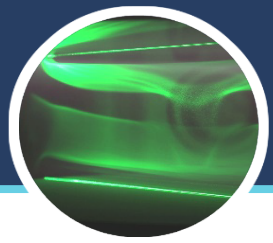


## Dust particles, nanoparticules = solid bodies from a few nm to cm


$$n_i = n_e + Z_d n_d$$

## → Modifications of plasma properties





# DUSTY (OR COMPLEX) PLASMAS?

**Presence of reactive gases**

**$\text{SiH}_4$ ,  $\text{CH}_4$ ,  $\text{C}_2\text{H}_2$**

**→ Succession of chemical and physical reactions**

**Material sputtering**

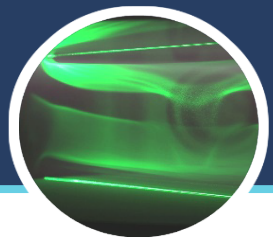
**Tungsten, Graphite, ...**

**All surfaces exposed to a plasma can be sputtered**

**Mainly by ion bombardment**

**Injection**

**To study dusty (complex) plasmas, many experiments are performed by injecting manufactured (calibrated) dust particles**



# DUSTY (OR COMPLEX) PLASMAS?

In nature

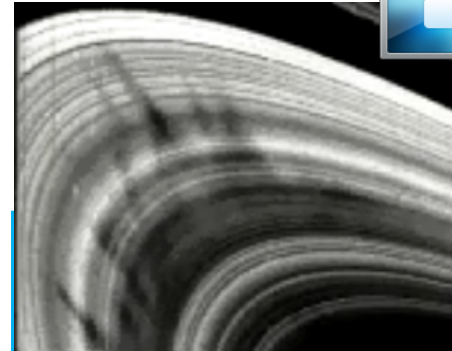
In astrophysics

- Comets
- Planetary rings

Hale-Bopp

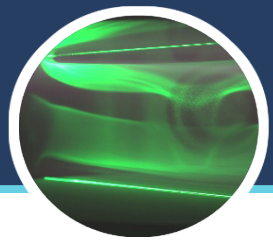


Hale-Bopp  
Taken 3/30/97 near Red Rock Canyon Park  
Celestron/EPOCH 8" f/11.5 Schmitz camera  
© 1997 Loke Kun Tan



Saturn

for ex. C.K. Goertz Rev. Geophys. (1989)

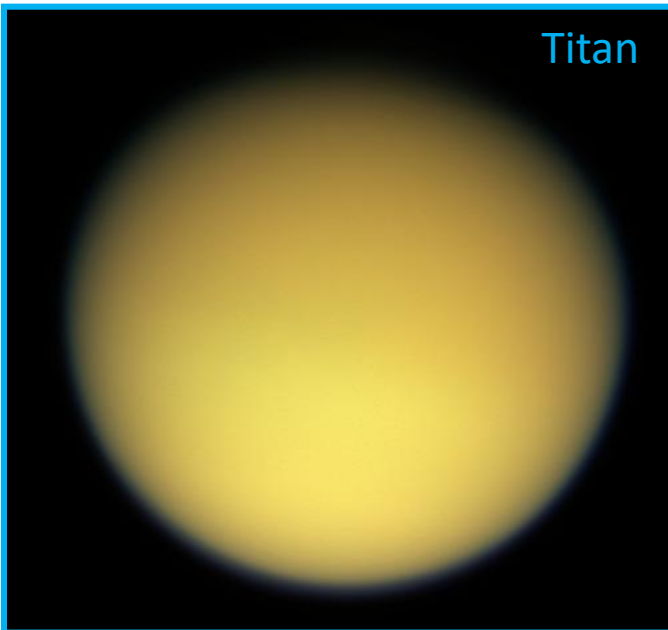


# DUSTY (OR COMPLEX) PLASMAS?

## In nature

### In astrophysics

- Planetary atmospheres ( $\text{CH}_4$ )
- Interstellar clouds (dark nebulae)



Titan

for ex. Szopa et al. Planet. Space Sci. (2006)



Horsehead Nebula

for ex. Kovacevic et al. ApJ (2005)

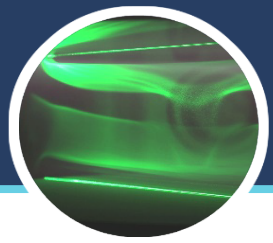
### In the atmosphere

- Noctilucent clouds
- Polar Mesospheric Summer Echoes (PMSE)



for ex. Scales et al. New J. Phys. (2004)





# DUSTY (OR COMPLEX) PLASMAS?

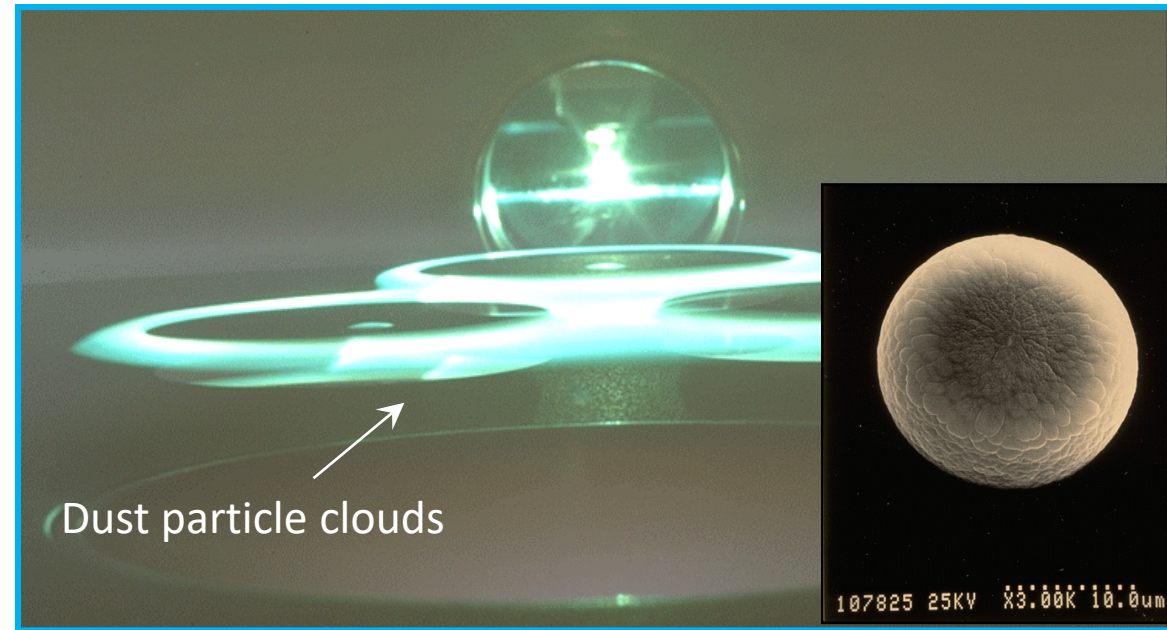
## In industry

### *Dust particles harmful*

(End of 80s – beginning of 90s)

Surface processing in microelectronics  
(cleanliness is a major requirement)

- Thin film deposition
- Etching

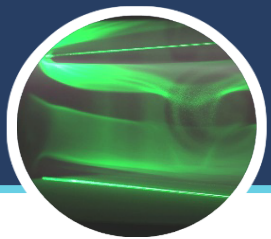


G. Selwyn et al. J. Vac. Sci. Technol. A (1989)  
R.M. Roth et al. Appl. Phys. Lett. (1985)

Use of reactive gases  
Sputtering



Dust particle formation



# DUSTY (OR COMPLEX) PLASMAS?

## In industry

*Dust particles useful*  
(End of 90s)

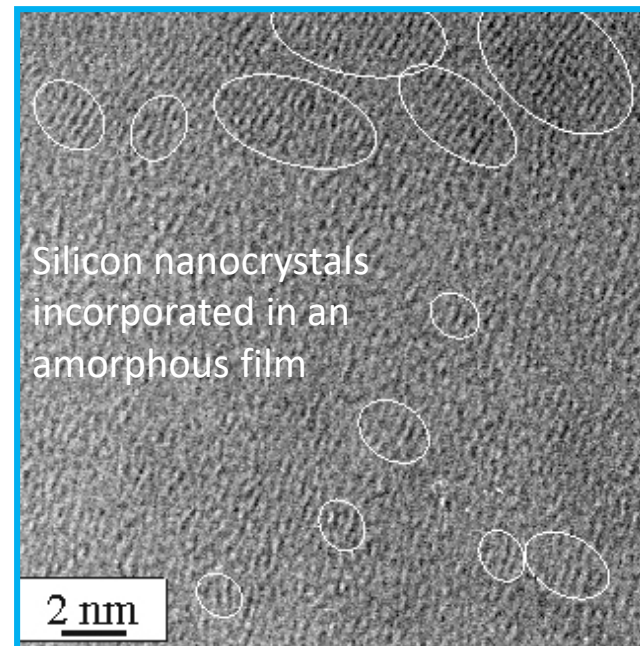
**Dust particle formation (silicon, carbon)**

**Design of new materials  
Nanotechnologies**

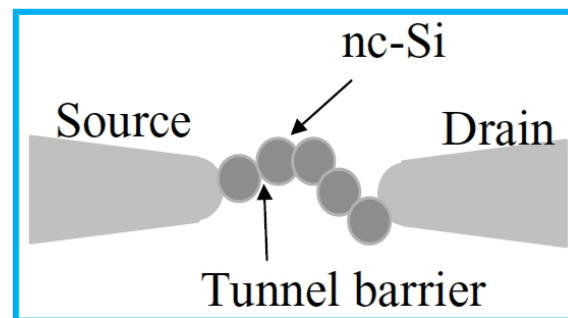
- Nanostructured materials
- Solar cells (polymorphous silicon)  
P. Roca i Cabarrocas J. Vac. Sci. Technol. A (1996)
- Single electron devices (transistor, memories)

## CHALLENGE

**Control the formation and deposition of  
grown nanoparticles (size, structure,  
number, position)**

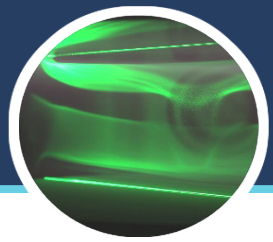


G. Viera et al. J. Appl. Phys. (2002)



Transistor using silicon nanocrystals  
A. Dutta et al. Jpn. J. Appl. Phys. (2000)





# DUSTY (OR COMPLEX) PLASMAS?

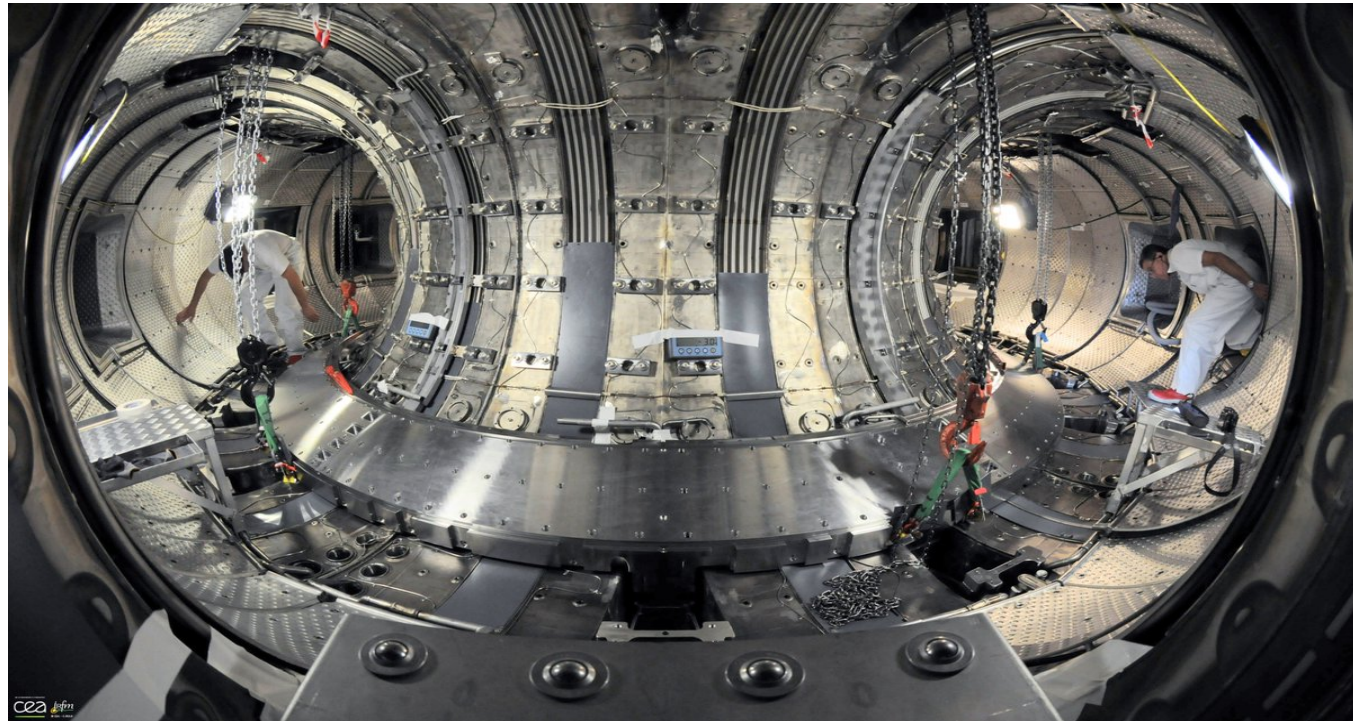
## In fusion devices

### *Dust particles harmful*

Wall erosion

Impurities in the plasma

Radio-toxicity of dust particles

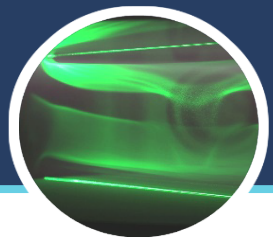


J. Winter, Plasma Phys. Control. Fusion (2004)

C. Arnas et al. J. Nucl. Mater (2009)

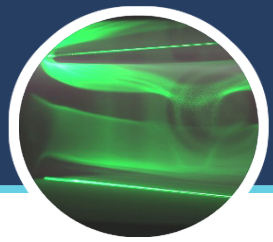
S.I. Krasheninnikov et al. Plasma Phys. Control. Fusion (2008)

D.L. Rudakov et al. Rev. Sci. Instrum. (2008)

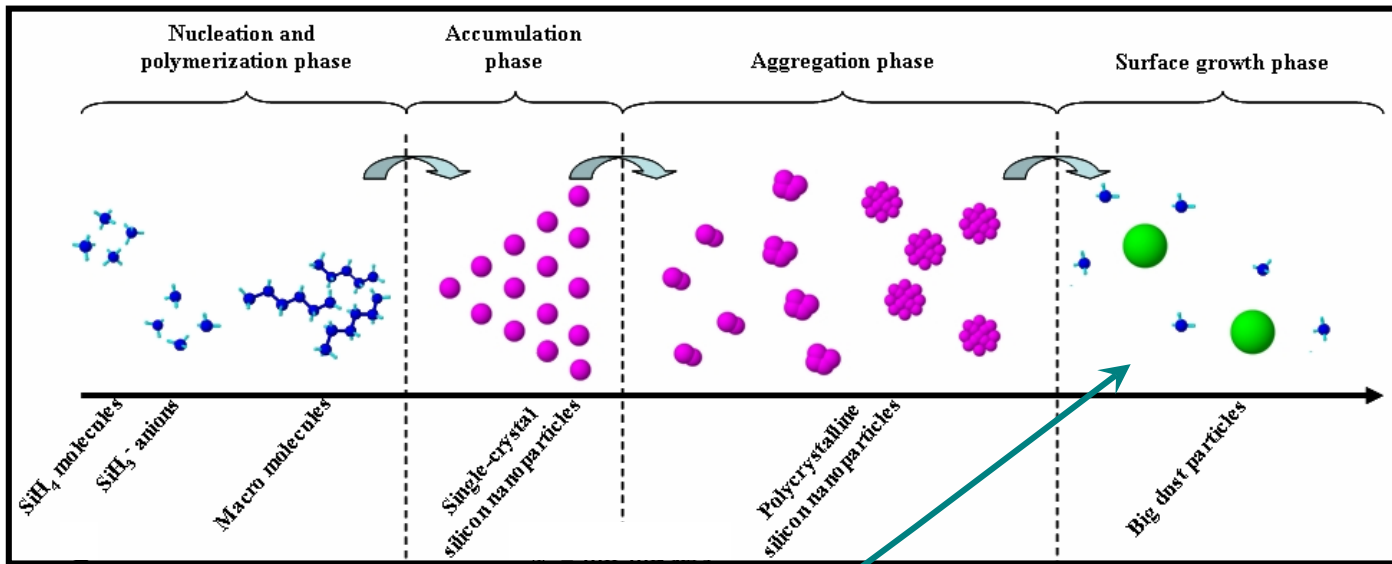


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# NANOPARTICLE FORMATION



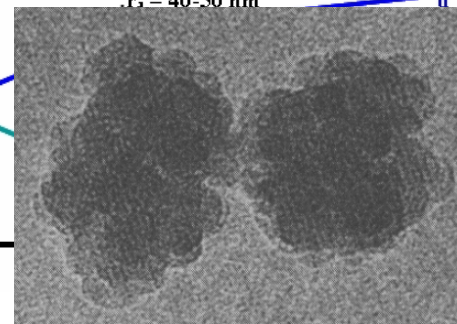
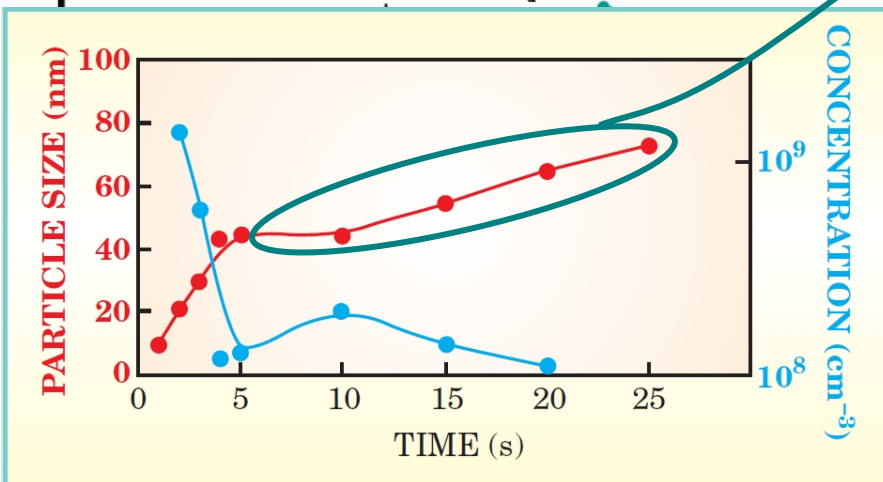
Cavarroc PhD Thesis (2007)

see also Bouchoule *et al.* Pure Appl. Chem. (1996)

Aggregation is still an open debate due to the dust particle charge

Ravi *et al.* Phys. Rev. E (2009)

Michau *et al.* J. Appl. Phys. (2017)

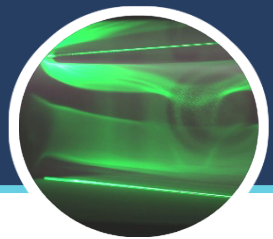


Bouchoule *et al.* Plasma Sources Sci. Technol. (1993)

from Merlino *et al.* Phys. Today (2004)

Nanoparticle electrical charge is a crucial parameter for the whole dusty plasma system



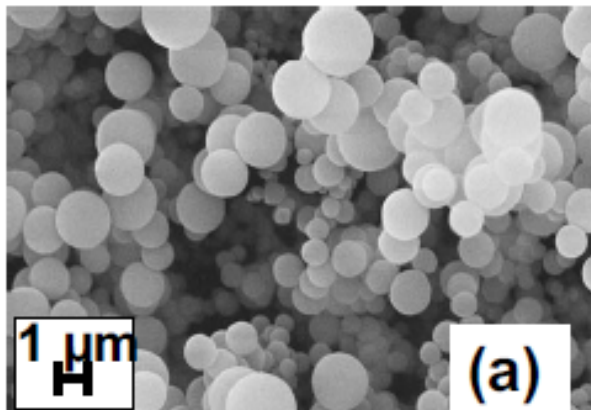


# NANOPARTICLE FORMATION



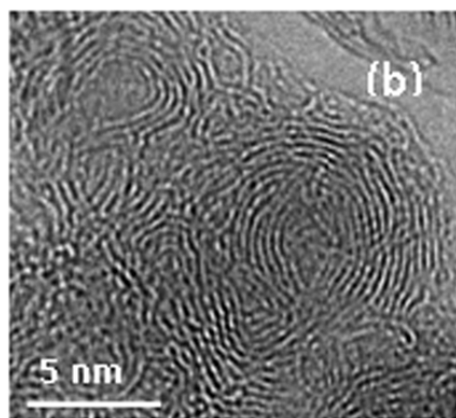
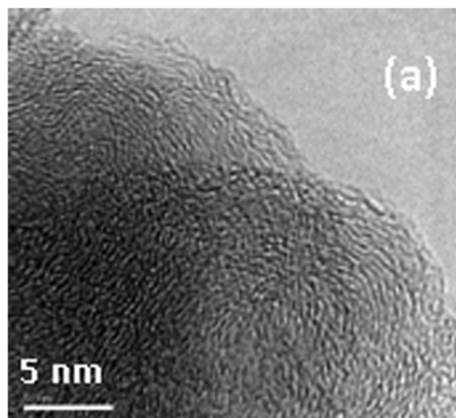
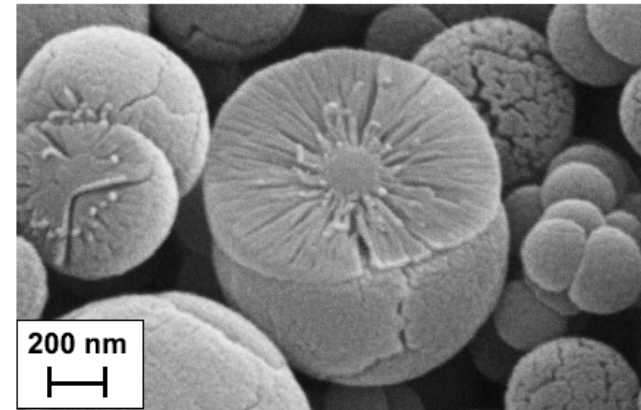
Ar/C<sub>2</sub>H<sub>2</sub> ( $r_d=180$  nm)

Berndt et al. Contrib. Plasma Phys. (2009)



CH<sub>4</sub>/N<sub>2</sub>

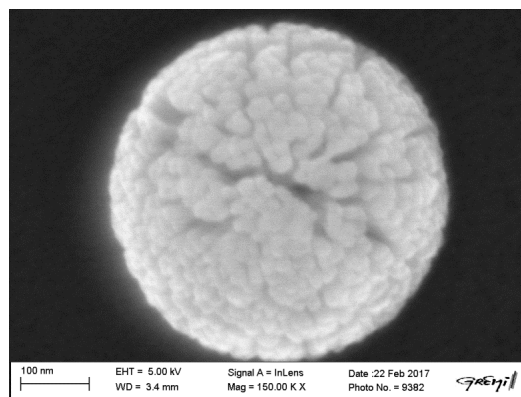
Massereau-Guilbaud et al. J. Appl. Phys. (2009)



Graphite DC sputtering

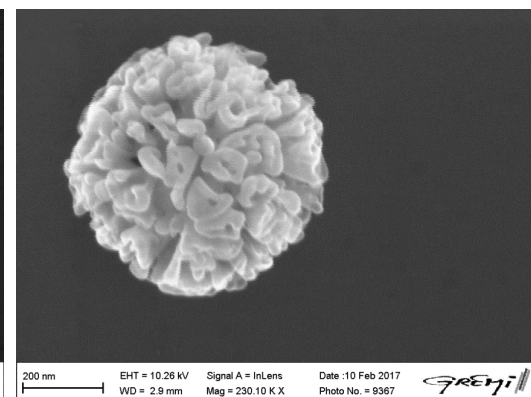
Ar/CH<sub>4</sub>/H<sub>2</sub>  $\mu$ wave

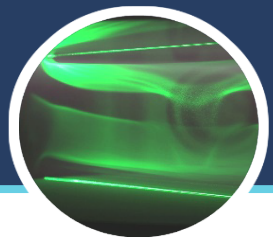
C. Arnas et al. Plasma Phys. Control. Fusion (2010)



Carbon based film sputtering (Ar/C<sub>2</sub>H<sub>5</sub>OH)

Labidi et al. AIP Conf. Proc. (2018)



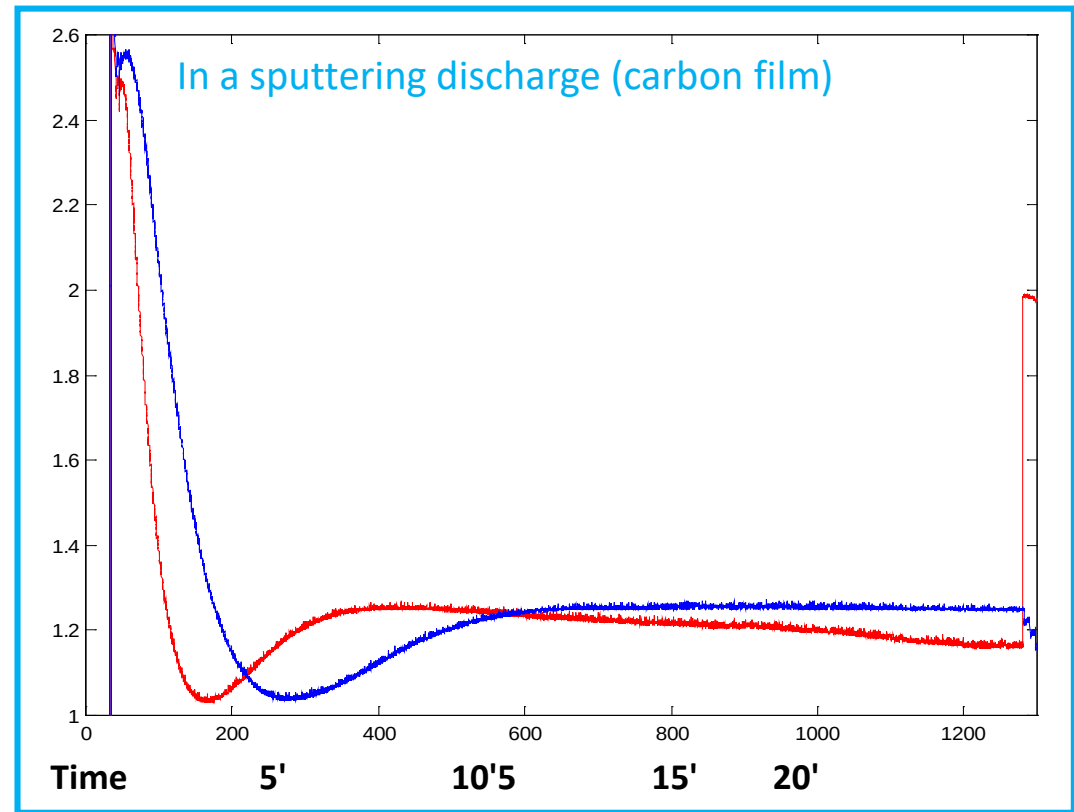
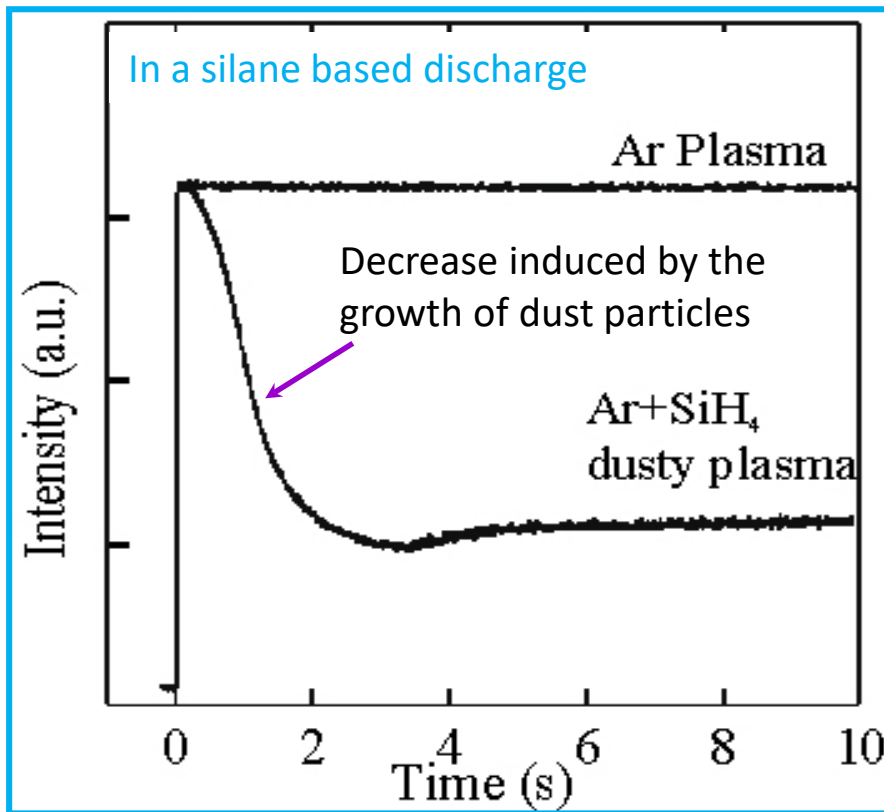


# NANOPARTICLE FORMATION

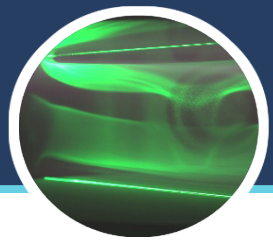
Charging process of nanoparticles is "universal", whatever the chemistry

All types of electrical measurements are able to detect nanoparticles

Example of the discharge current (see also Boufendi *et al.* Appl. Phys. Lett. (2001))



M. Mikikian *et al.* New J. Phys. **5**, 19 (2003)

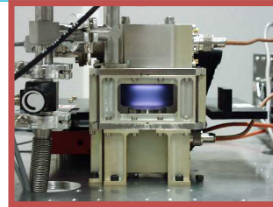


# NANOPARTICLE FORMATION

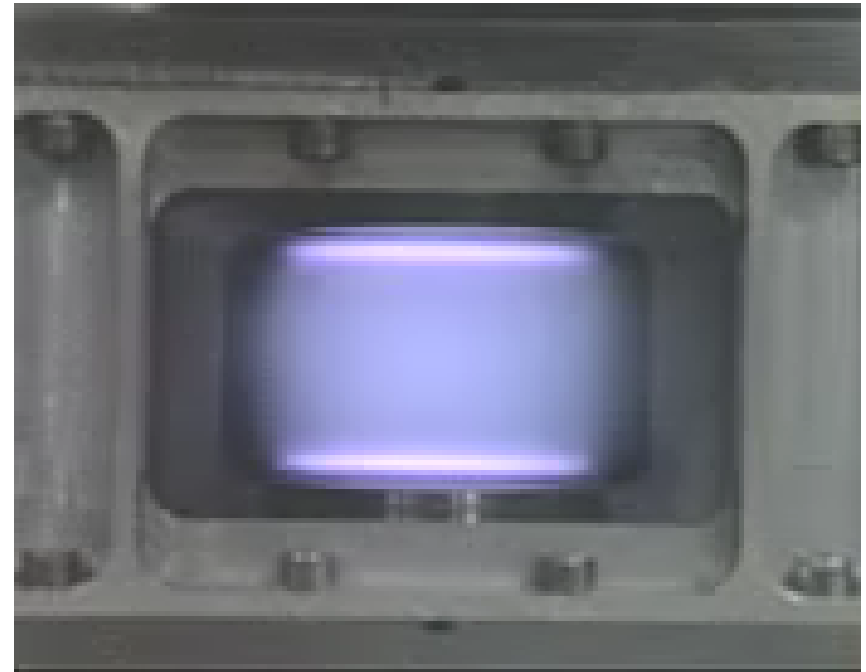
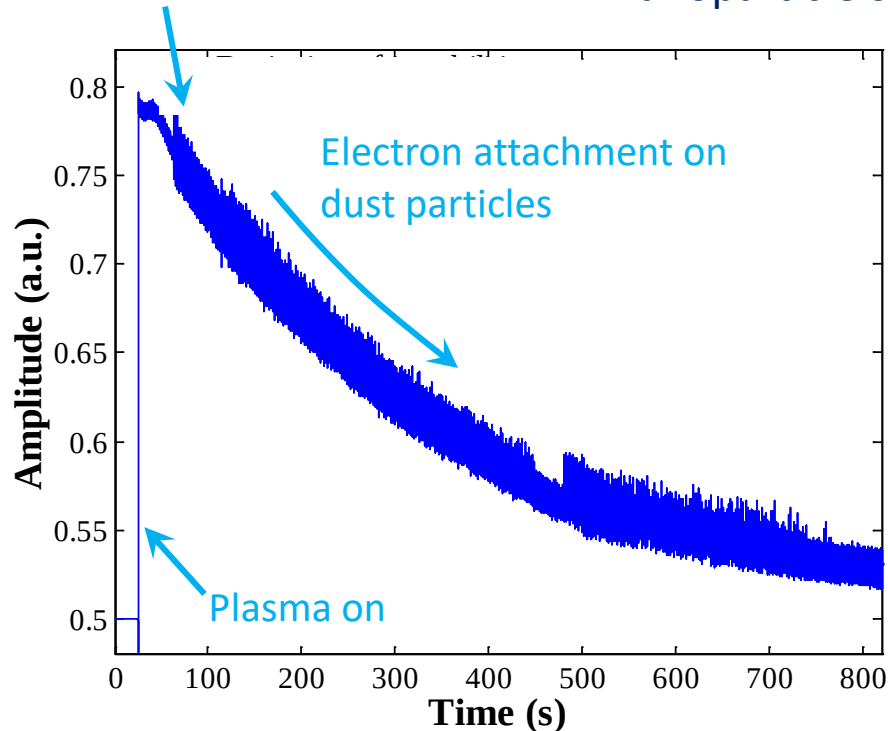
When nanoparticle density is large, huge loss of electrons

Plasma equilibrium is disturbed → Low frequency Instabilities

Frequencies from a few Hz to a few kHz depending on the nanoparticle size, density and charge



Instability beginning

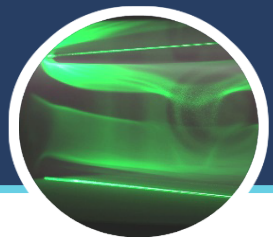


Mikikian *et al.* Phys. Plasmas **13**, 092103 (2006)

Tawidian *et al.* Plasma Sources Sci. Technol. **23**, 065009 (2014)

Mikikian *et al.* Eur. Phys. J Appl. Phys. **49**, 13106 (2010)

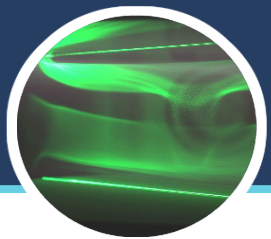
Mikikian *et al.* IEEE Trans. Plasma Sci. **36**, 1012 (2008)



# OUTLINE

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- ☐ Void instabilities: Heartbeat
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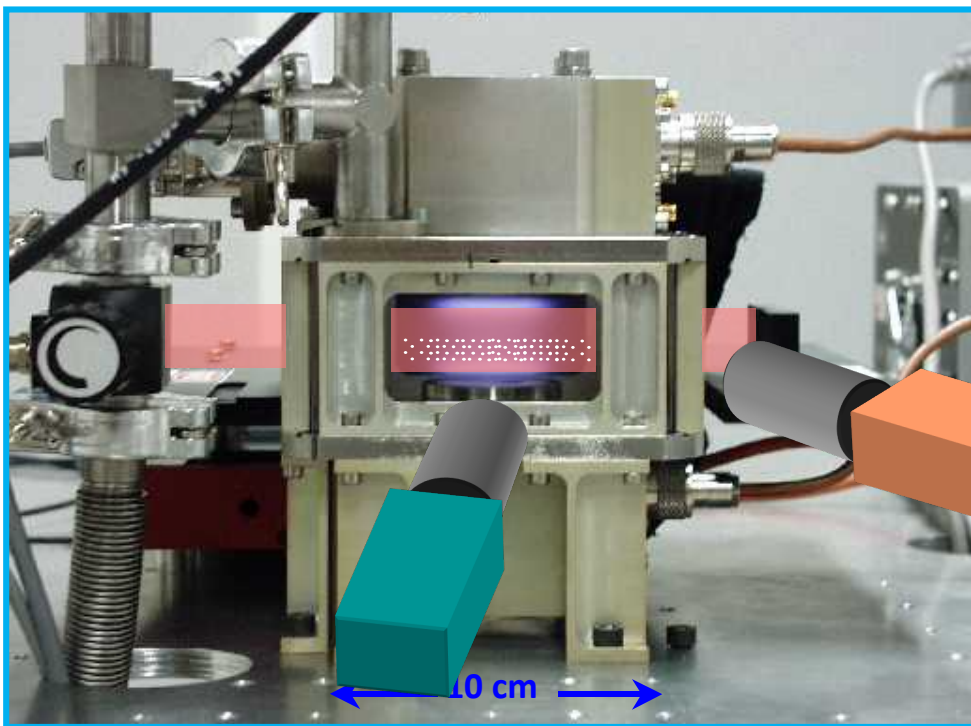
# EXPERIMENTAL SETUP

RF discharge (13.56 MHz) in push-pull mode

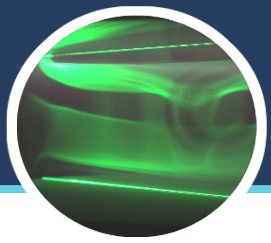
Electrode diameter = 4 cm, Inter-electrode distance = 3 cm

Ar or Kr plasma at pressure 0.1 → 2 mbar (no gas flow), power 0 → 4 Watts

**Nanoparticle formation by sputtering a polymer layer** (final diameter ~ 50→800 nm)



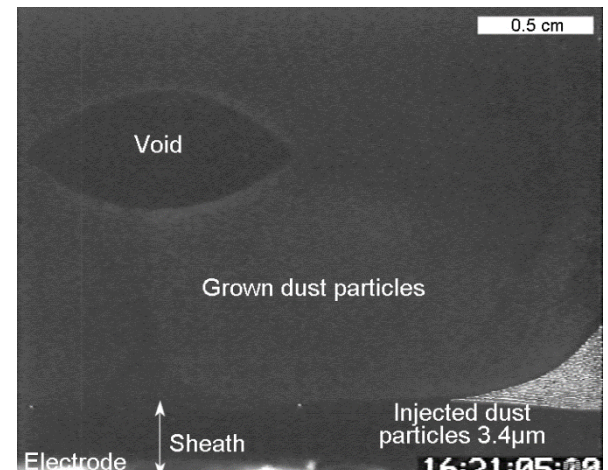
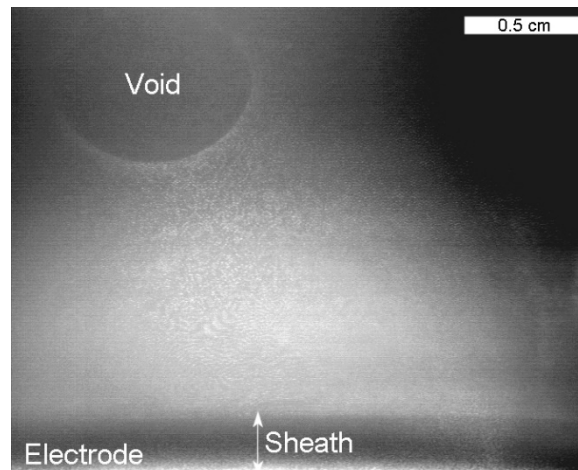
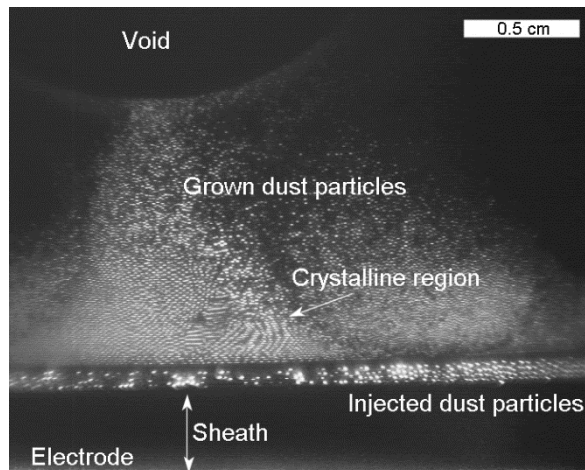
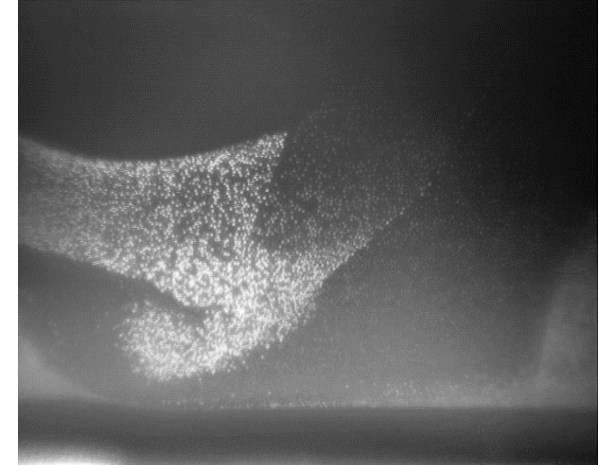
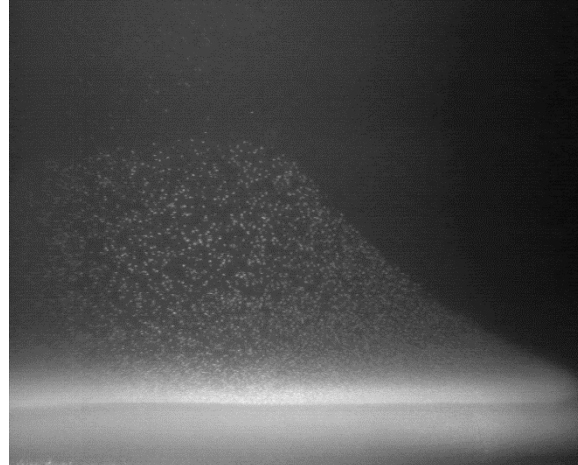
- Electrical probe (V, I)
- Optical Emission Spectrometer
  - F=500 mm
  - Gratings 600, 1200, 1800, 2400 lines/mm
  - CCD detector 400x1340 pixels
- Laser Light Scattering
  - Laser sheet
  - Normal and high-speed imaging
- Laser Induced Fluorescence
- Absorption Spectroscopy



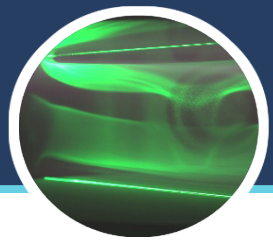
# EXPERIMENTAL SETUP

## Forces : shape of the dust cloud

Gravity, Electric force, Ion drag, Neutral drag, Thermophoresis, Interactions



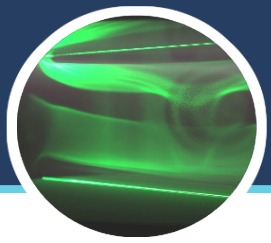
M. Mikikian, Habilitation (2008)



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- ☒ **The "void"**
- ☐ Void instabilities: Heartbeat
- ☐ Last beats: Mixed-Mode Oscillations

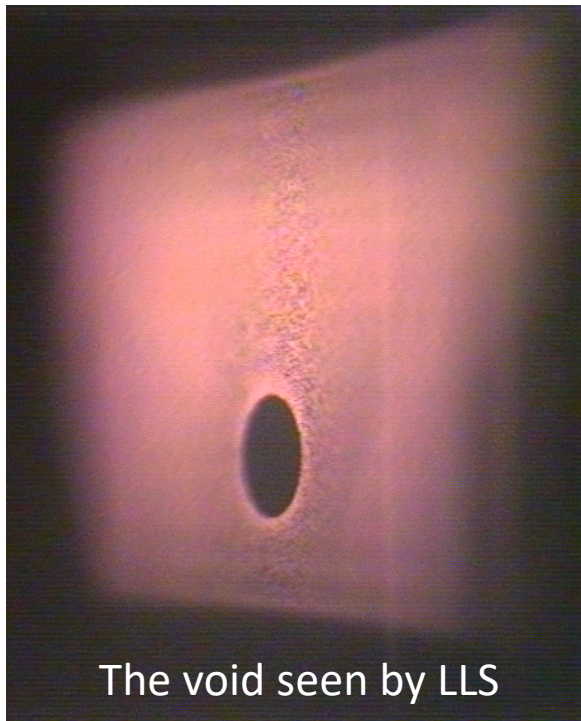




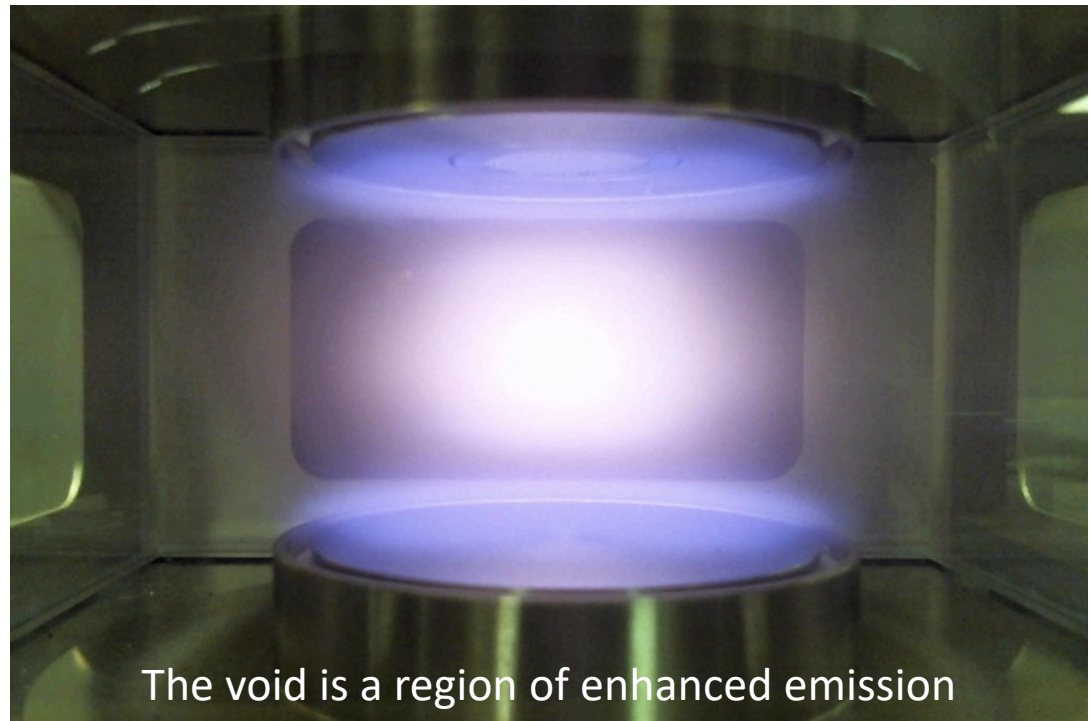
# THE VOID: A DUST-FREE REGION

**In typical cc-rf discharges a dust-free space appears close to the plasma center**

This void is induced by the ion drag force and appears when dust particles reach a sufficiently large size (charge)



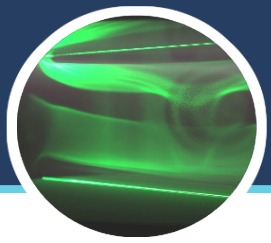
The void seen by LLS



The void is a region of enhanced emission

Mikikian *et al.* Eur. Phys. J. Appl. Phys. **49**, 13106 (2010)

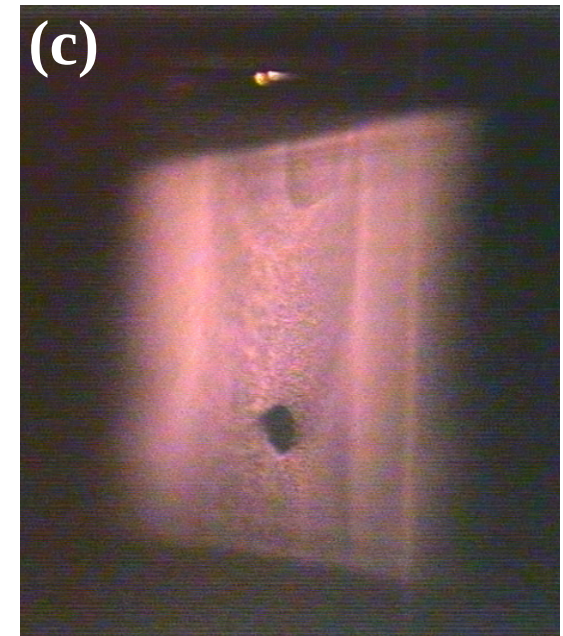
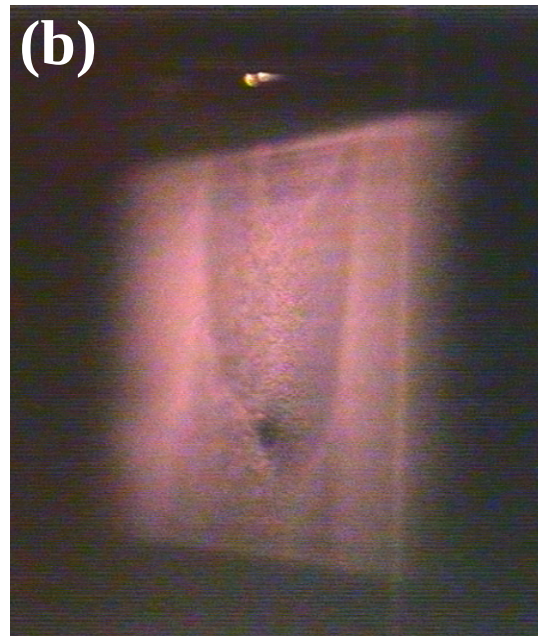
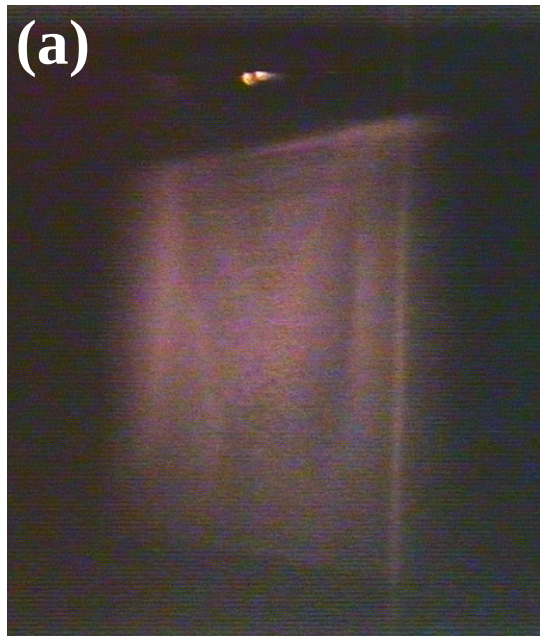




# THE VOID: A DUST-FREE REGION

In typical cc-rf discharges a dust-free space appears in the cloud when dust particles reach a sufficiently large size (charge)

**This void is induced by the ion drag force** (J. Goree et al. Phys. Rev. E (1999))



M. Mikikian et al. Pure Appl. Chem. (2010)

See also

J. Dorier et al. J. Vac. Sci. Technol. (1995)

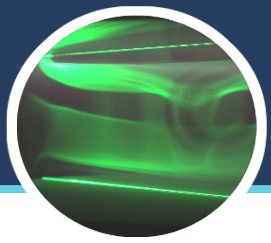
G. Praburam et al. Phys. Plasmas (1996)

G.E. Morfill et al. Phys. Rev. Lett. (1999)

M.R. Akdim et al. Phys. Rev. E (2001)

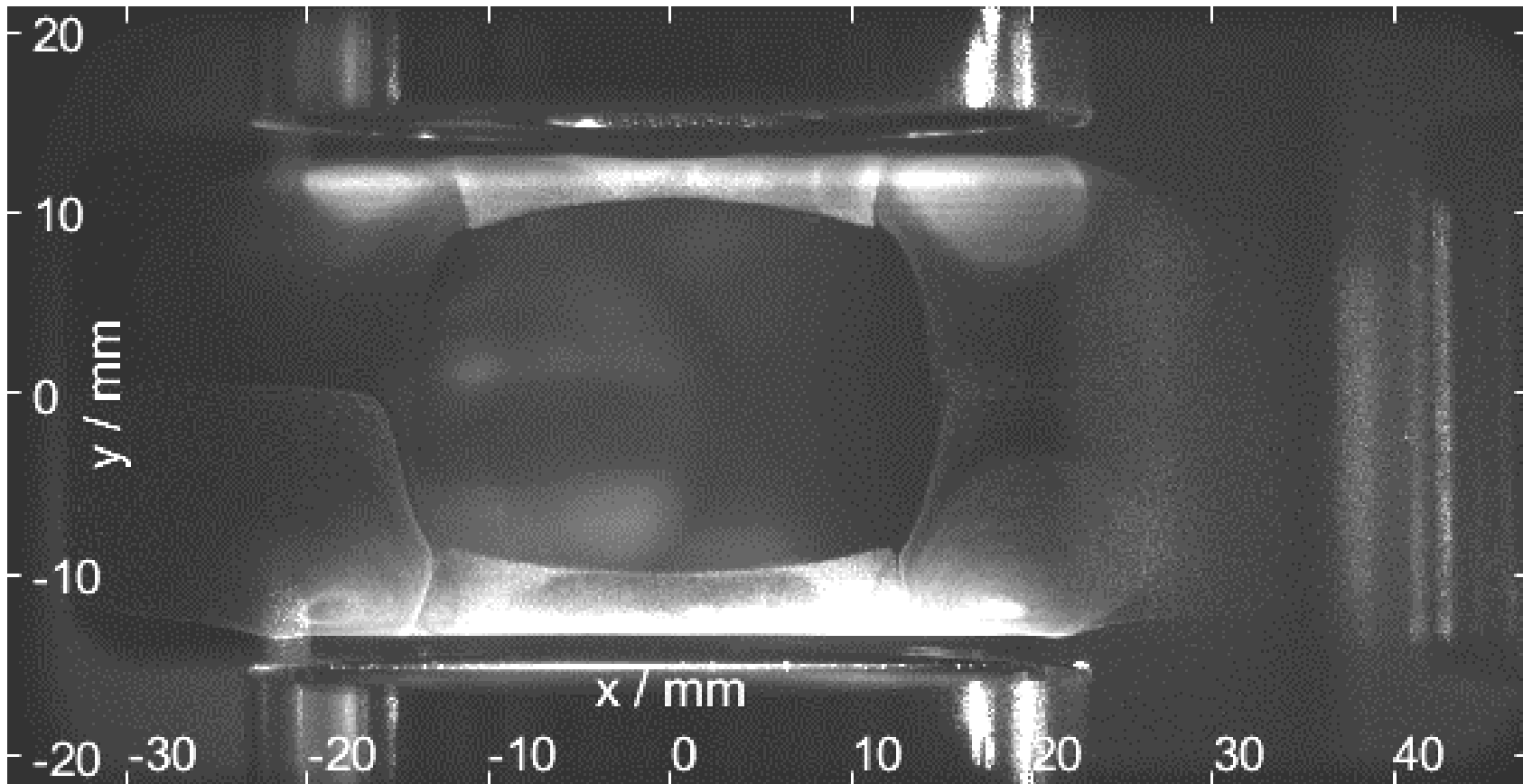
S.V. Vladimirov et al. Phys. Plasmas (2005)

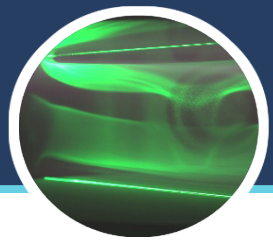
V. Land et al. New J. Phys. (2007)



# THE VOID: A DUST-FREE REGION

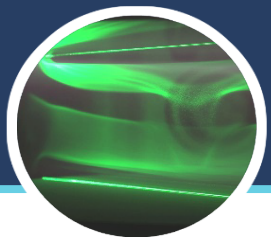
In the void, conditions are fulfilled for the nucleation of new nanoparticle generations





# OUTLINE

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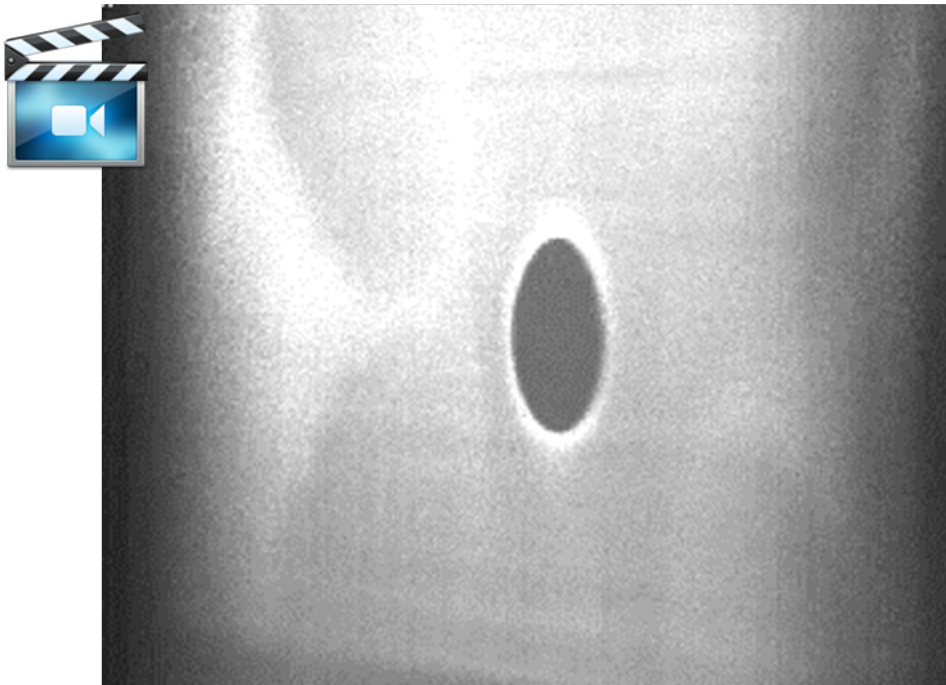


# VOID INSTABILITIES: HEARTBEAT

Void sustained by 2 forces of opposite directions (ion drag/electric force)

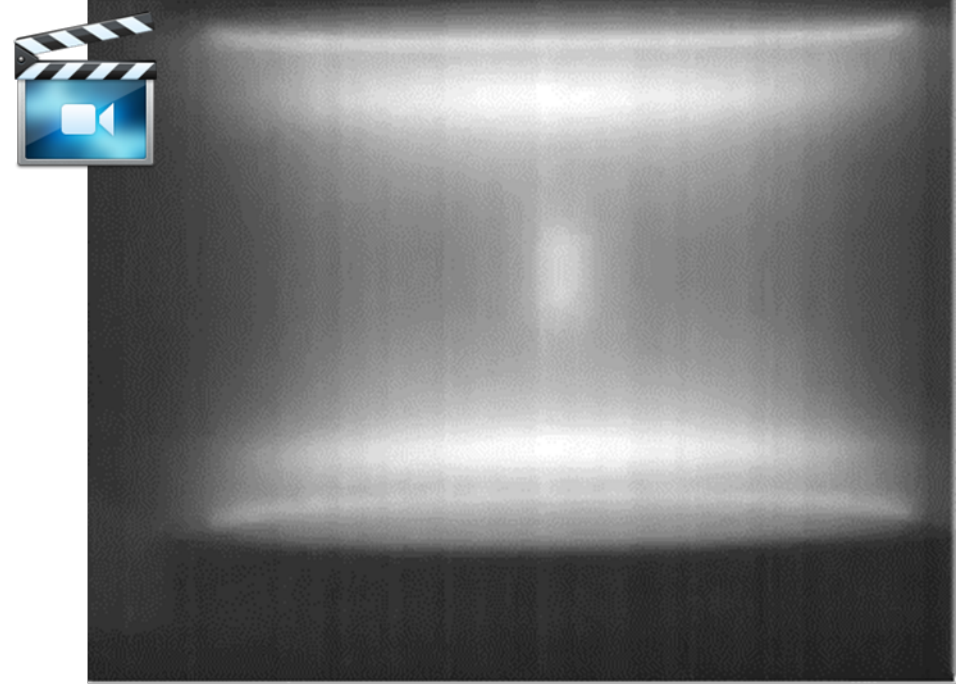
Heartbeat instability = Self-excited oscillations of the void size ( $\sim 20$  Hz)

Dust cloud observed with Laser Light Scattering



Side view

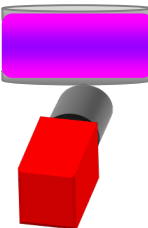
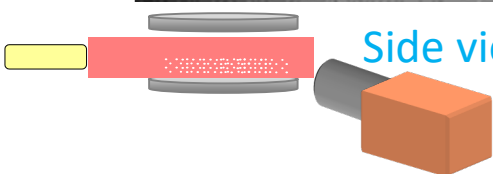
Direct observation of plasma glow



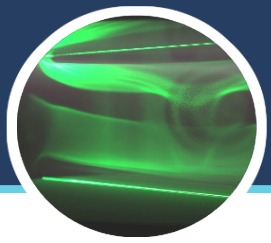
Front view

High-speed imaging  $\sim 1800$  fps

M. Mikikian et al. New J. Phys. (2007)  
M. Mikikian et al. Phys. Rev. Lett. (2010)

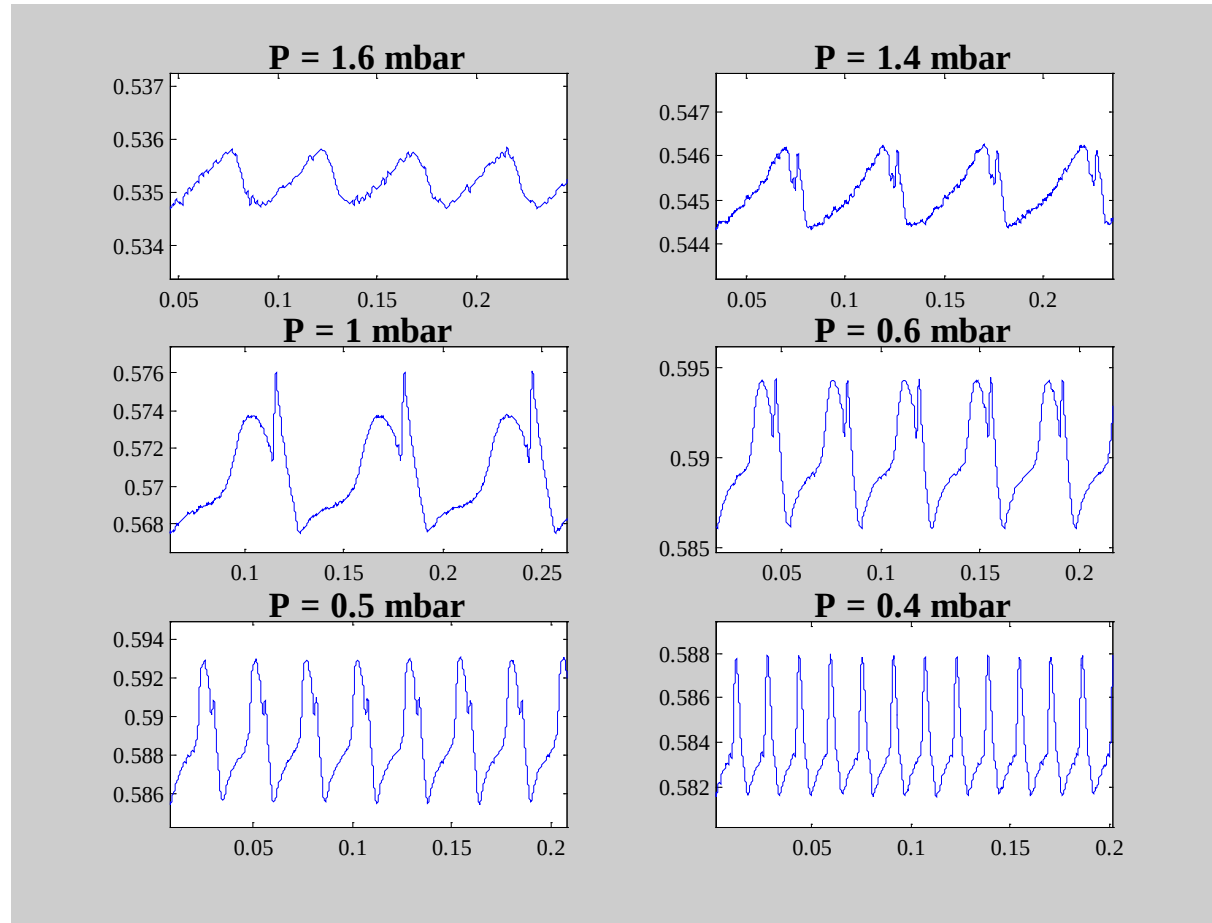




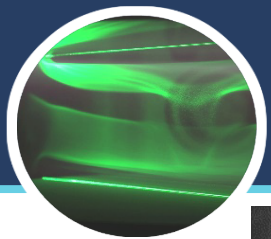


# VOID INSTABILITIES: HEARTBEAT

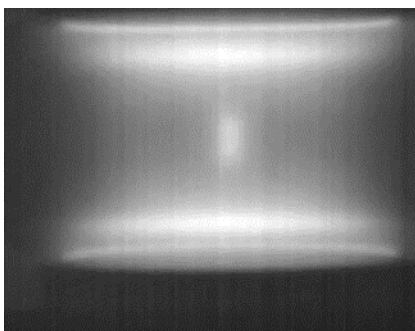
Signature on electrical measurements (discharge current)  
Strongly nonlinear behavior



M. Mikikian et al. Phys. Plasmas (2004)

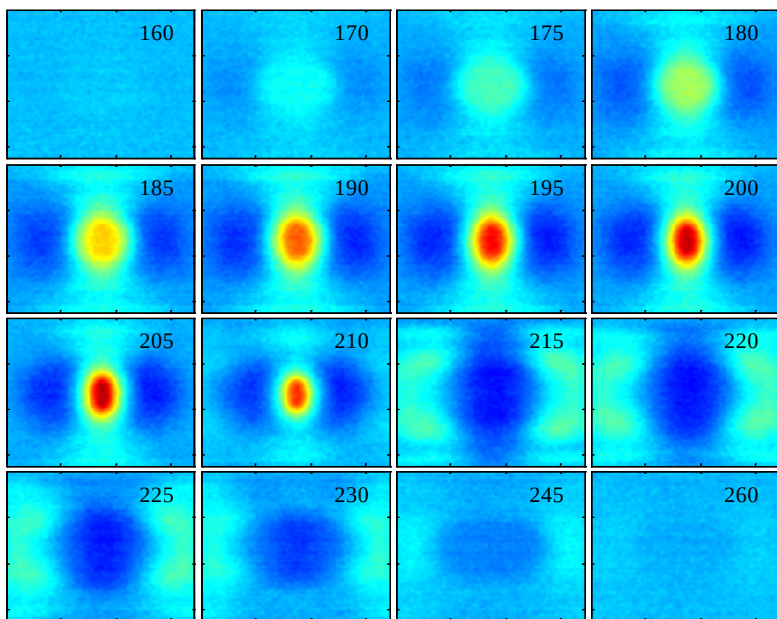


# VOID INSTABILITIES: HEARTBEAT



**Direct observation  
of the plasma glow**

Evolution of plasma glow during a single contraction-expansion sequence



Evolution of the central line profile during a single contraction-expansion sequence

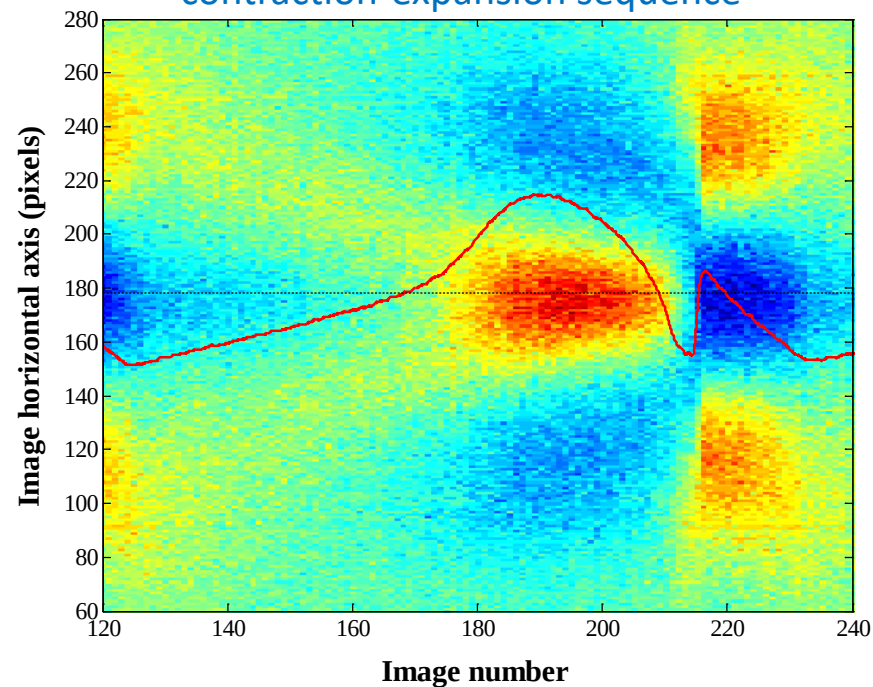
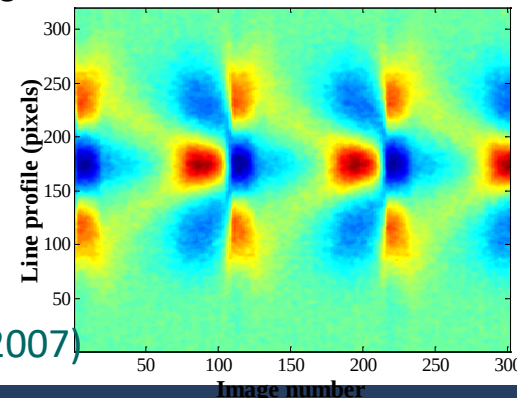
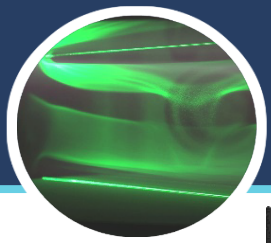


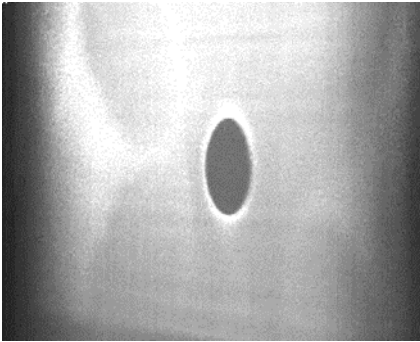
Image number



M. Mikikian, L. Couédel, M. Cavarroc, Y. Tessier, L. Boufendi, New J. Phys. **9**, 268 (2007)

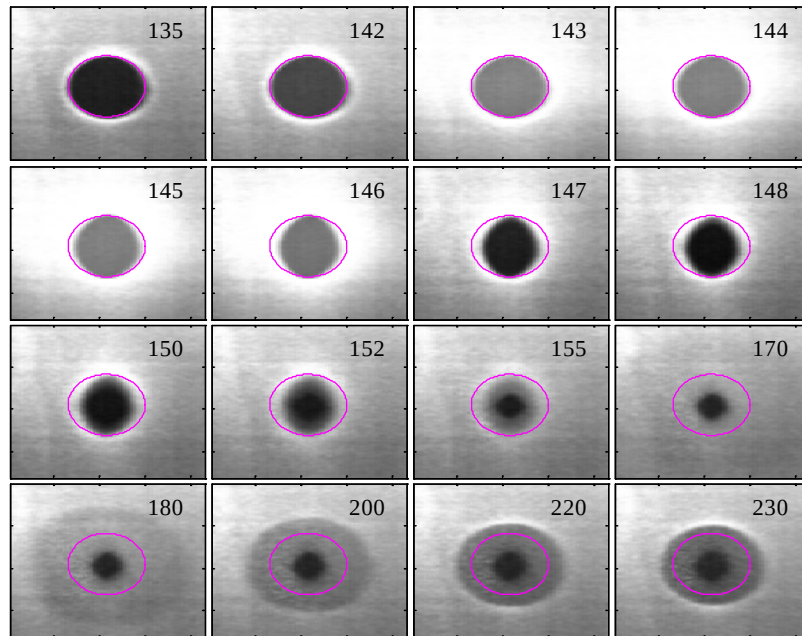


# VOID INSTABILITIES: HEARTBEAT

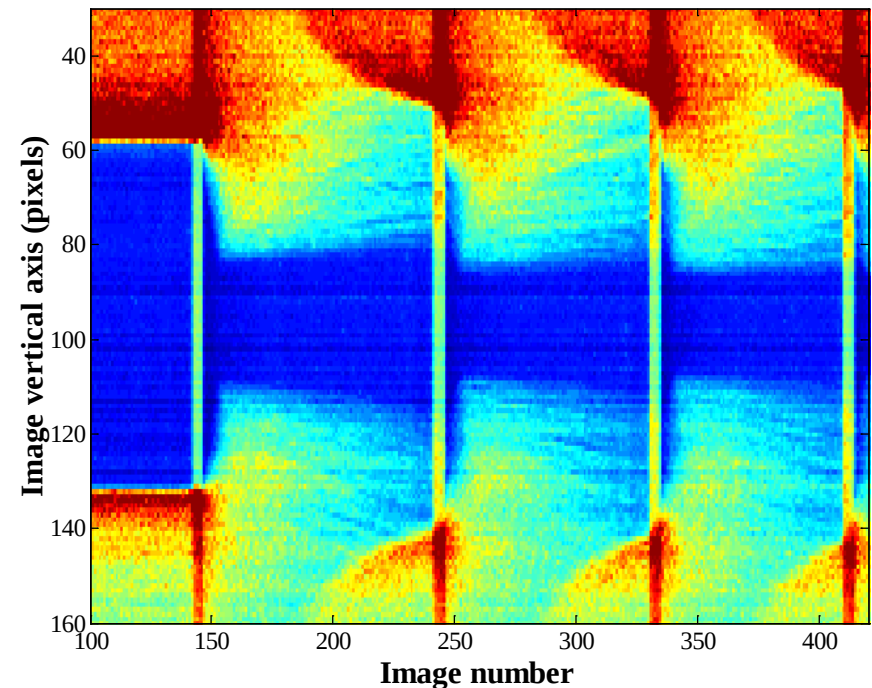


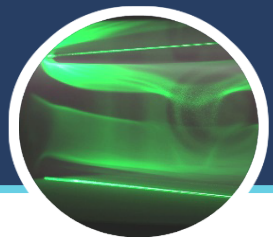
**Dust cloud observed with Laser Light Scattering**

Evolution of dust cloud during one contraction-expansion sequence



Evolution of the central column profile during one contraction-expansion sequence

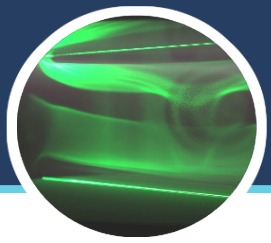




# OUTLINE

- ☐ Dusty (or complex) plasmas?
- ☐ Nanoparticle formation in a few words ...
- ☐ Experimental setup
- ☐ The "void"
- ☐ Void instabilities: Heartbeat
- ☐ **Last beats: Mixed-Mode Oscillations**





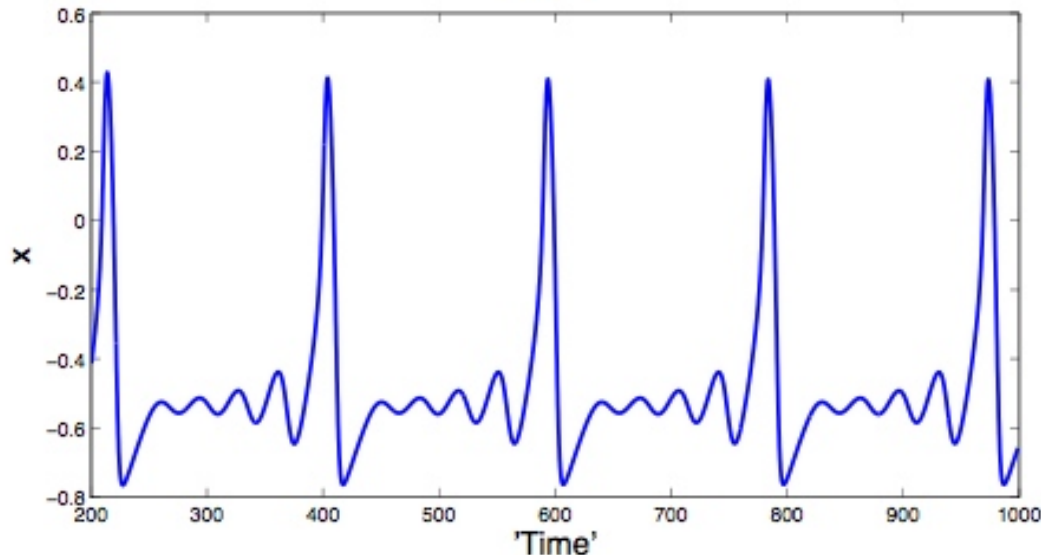
# MIXED-MODE OSCILLATIONS

**MMOs = Alternation of small and large amplitude oscillations**

**Small amplitude oscillations = subthreshold oscillations**

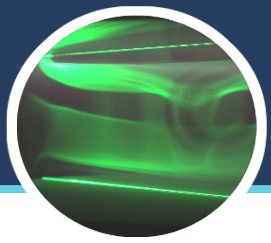
**High amplitude oscillations = relaxation mechanisms (spikes)**

$L^S = 1^4$



**MMO notation:**

**A pattern with L large amplitude oscillations  
and S small amplitude ones is noted  $L^S$**



# MIXED-MODE OSCILLATIONS

**Chemistry:** Belousov-Zhabotinskii (BZ) reaction (oxidation of malonic acid in sulfuric acid solution with bromate ions and a catalyst)

Electrochemical systems (copper-phosphoric acid)

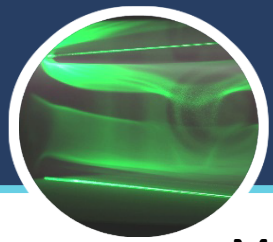
**Neuronal science:** Models of neuronal activity

Hodgkin-Huxley

FitzHugh-Nagumo

Morris-Lecar

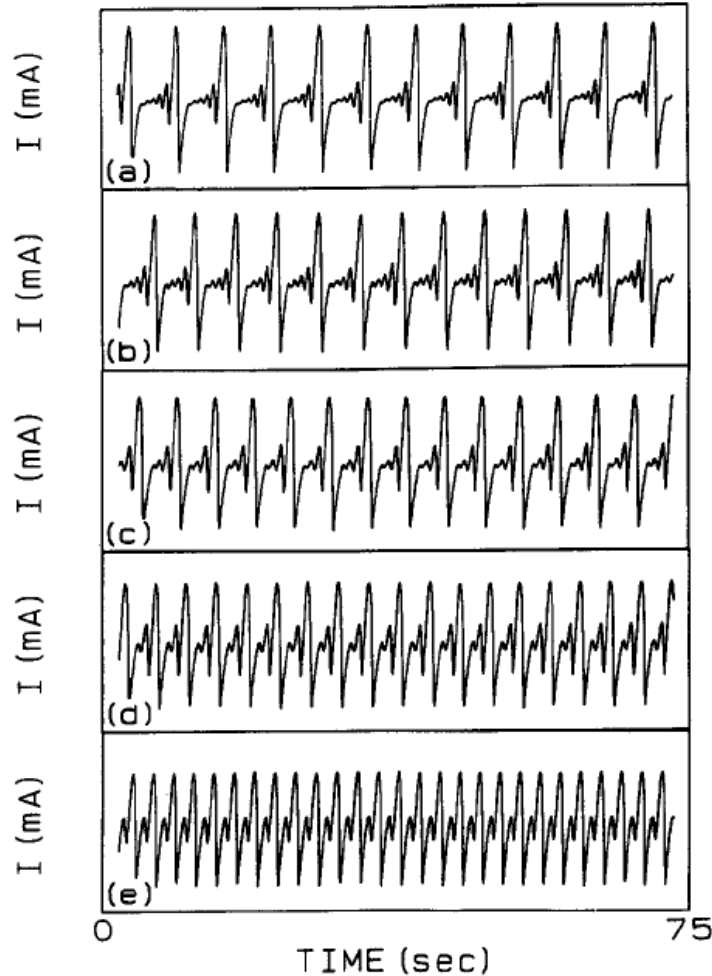
**Dynamical system theories (bifurcation):** Canards (French Ducks)  
Subcritical Hopf-homoclinic bifurcation  
Shilnikov homoclinic orbits



# MIXED-MODE OSCILLATIONS

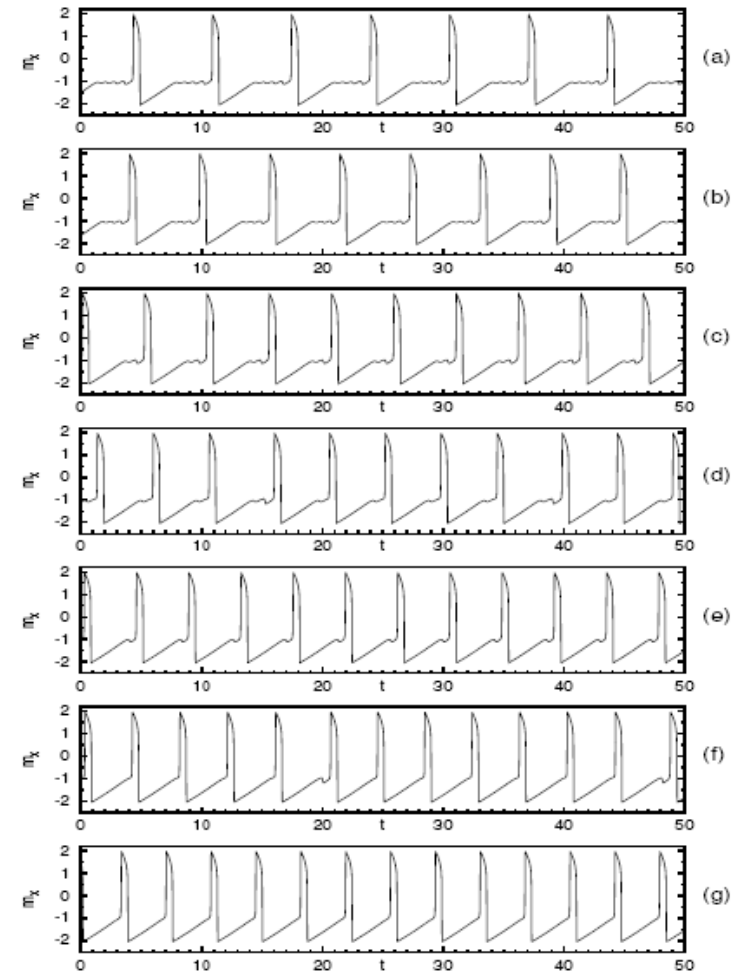
MMOs in electrochemical systems

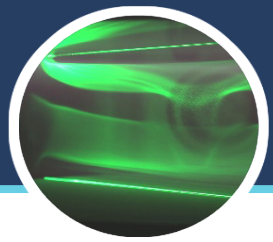
Schell et al., J. Chem. Phys. (1989)



MMOs in giant squid axon (FHN model)

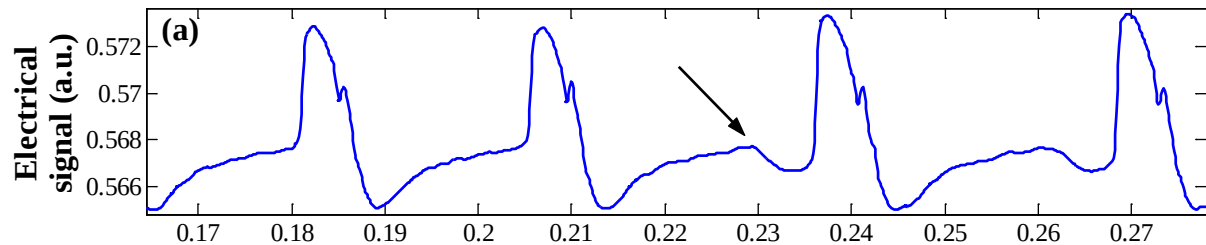
Zaks et al., Chaos (2005)



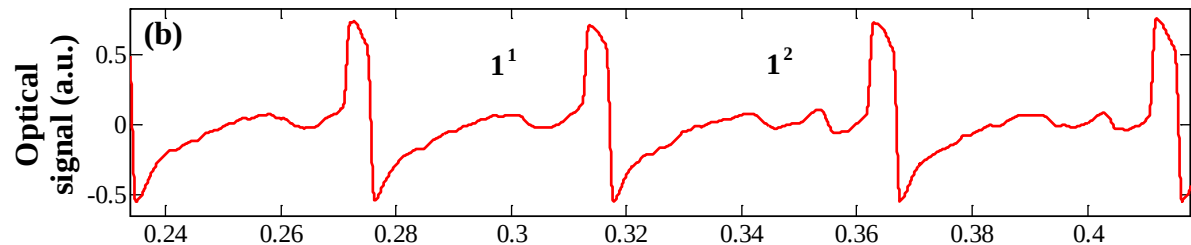


# HEARTBEAT INSTABILITIES: LAST BEATS...

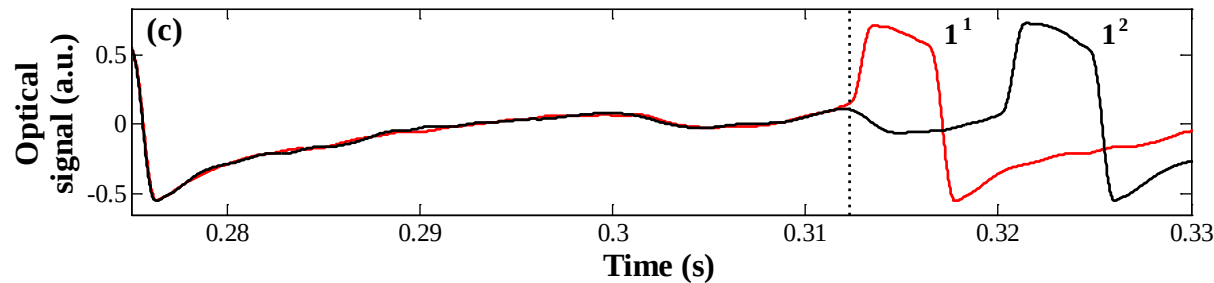
When the instability is about to stop, failed peaks occur  
Signal shape is typical of MMOs



Transition between  $1^0$  and  $1^1$  states in discharge current

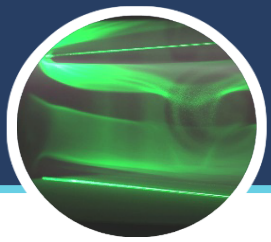


Transition between  $1^1$  and  $1^2$  states in plasma emission intensity



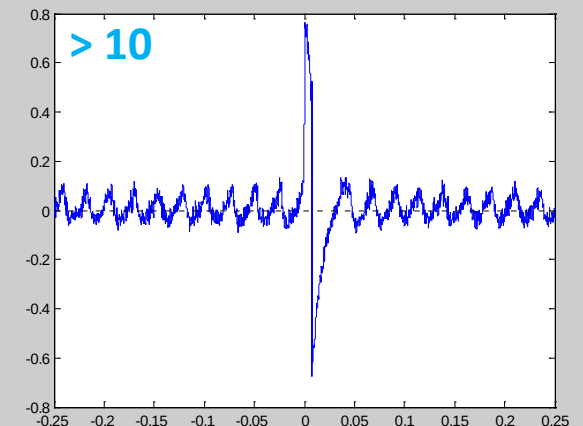
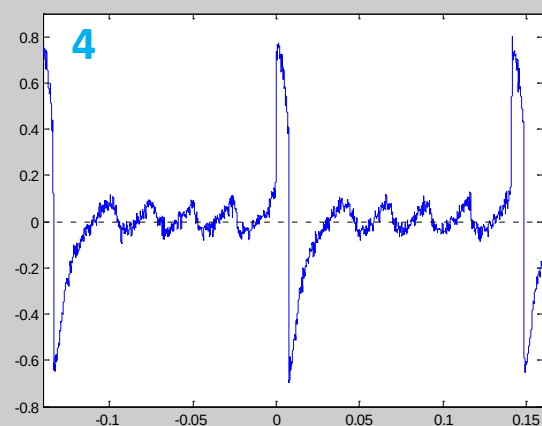
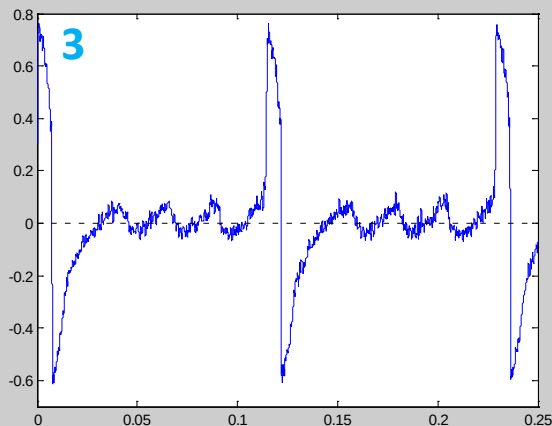
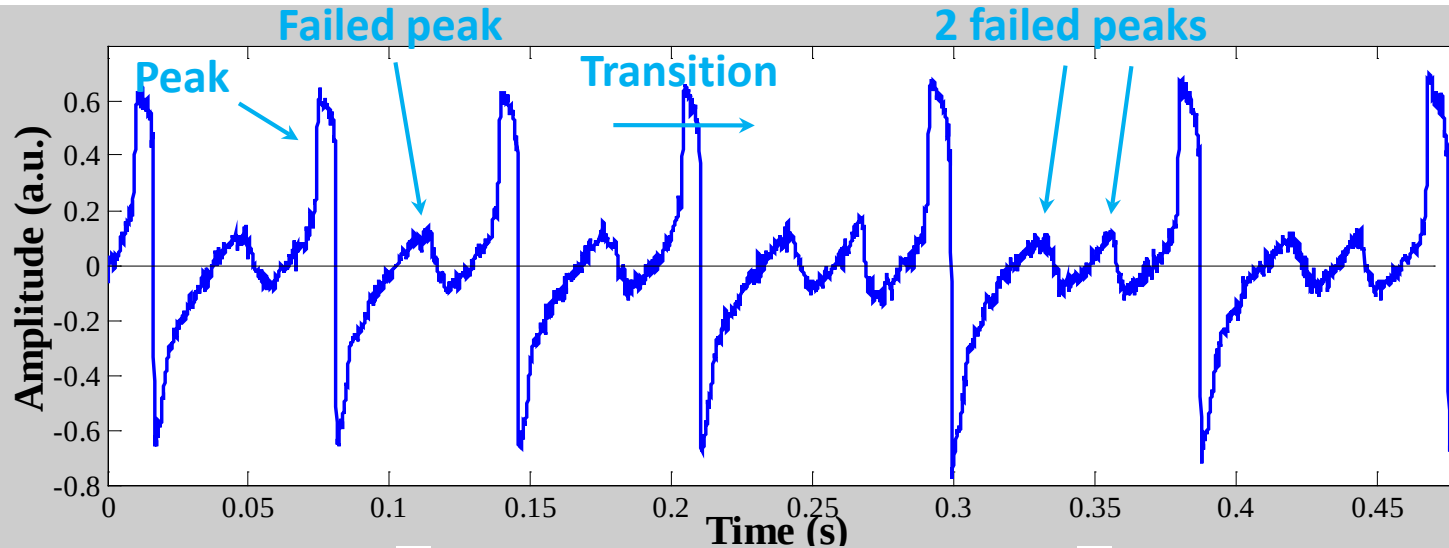
M. Mikikian, M. Cavarroc, L. Couédel, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **100**, 225005 (2008)

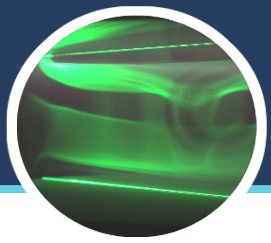




# HEARTBEAT INSTABILITIES: LAST BEATS...

Signature on electrical measurements (discharge current)





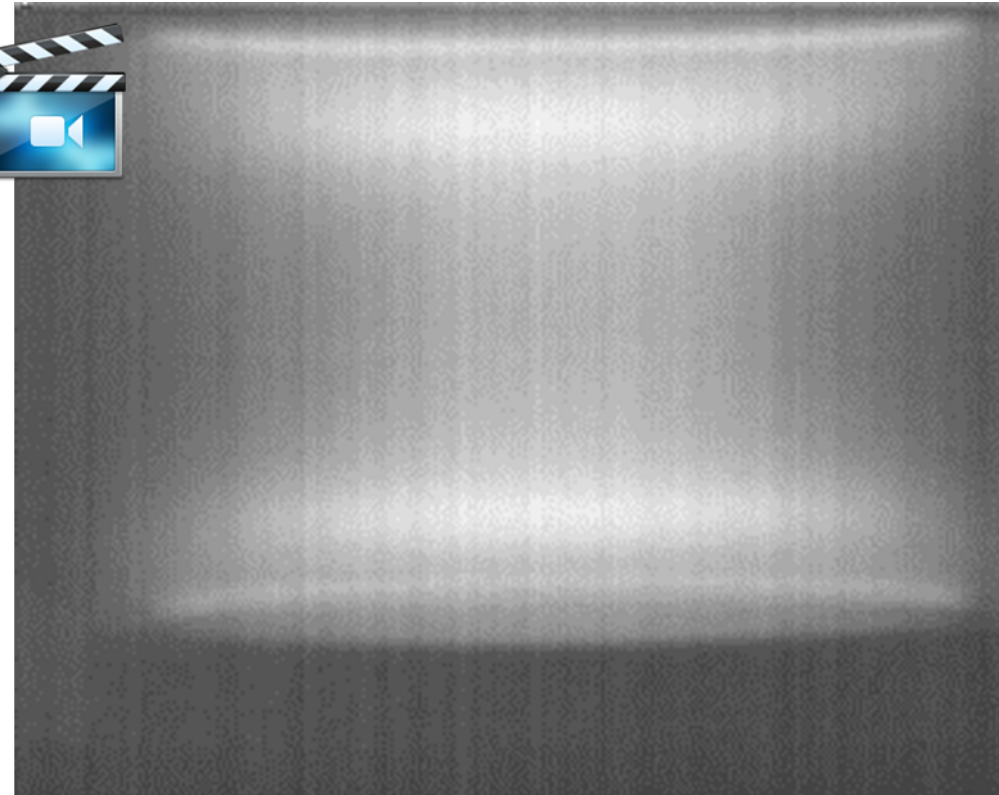
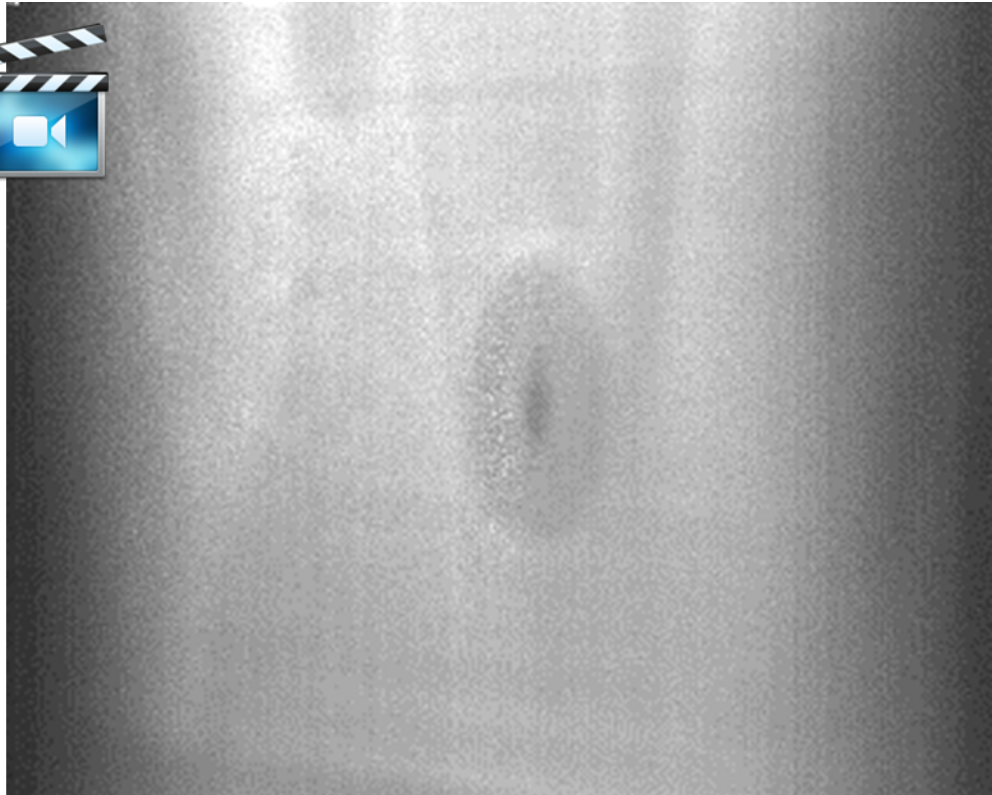
# HEARTBEAT INSTABILITIES: LAST BEATS...

**Contraction = Increase of central plasma emission**

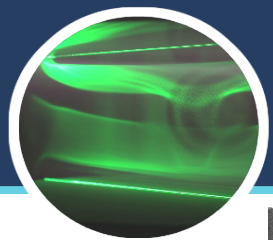
**Failed peaks = Failed contractions**

**2 failed peaks**

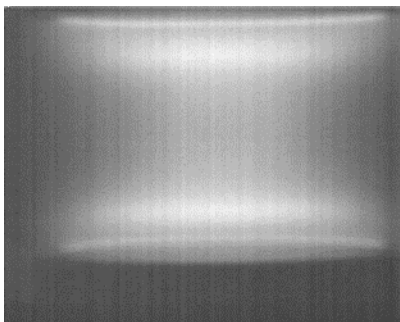
**1 failed peak**



M. Mikikian, L. Couédel, M. Cavarroc, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **105**, 075002 (2010)

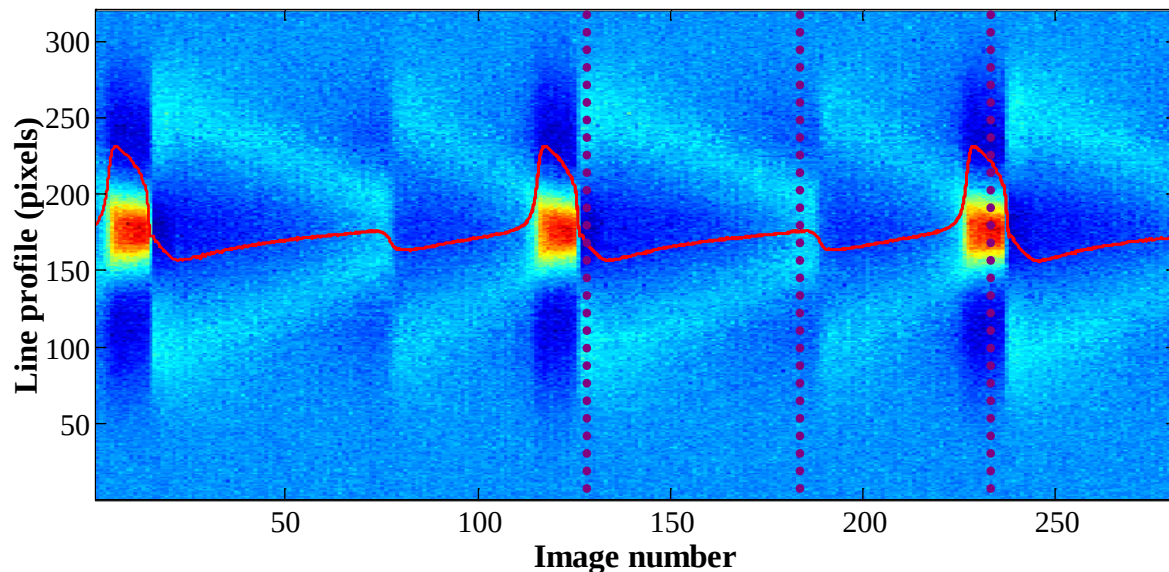
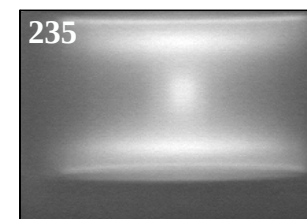
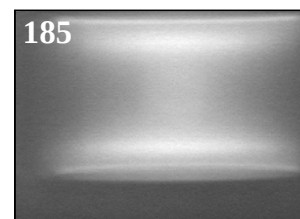
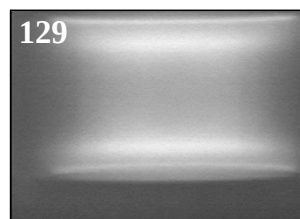
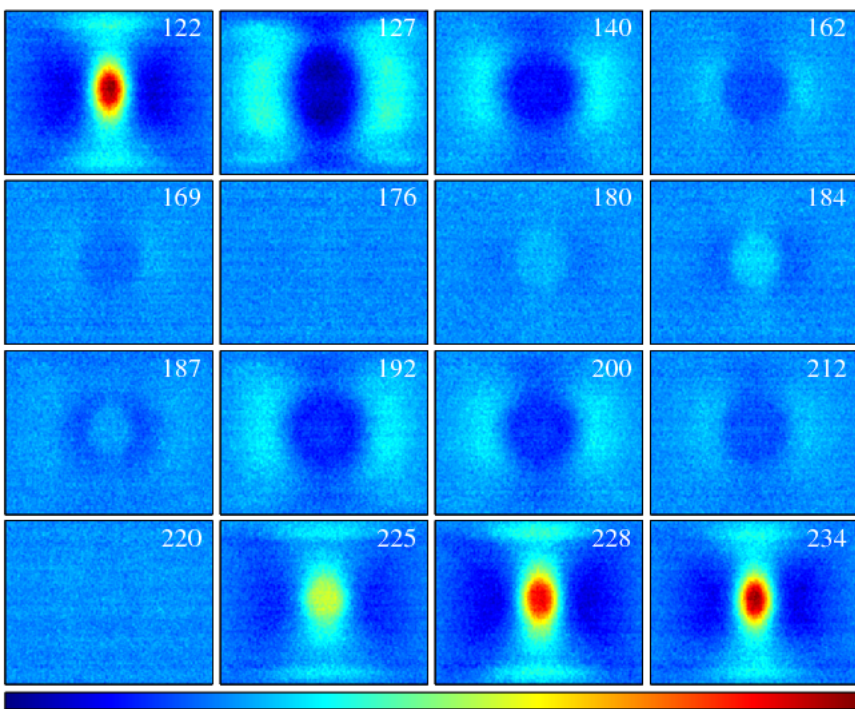


# HEARTBEAT INSTABILITIES: LAST BEATS...



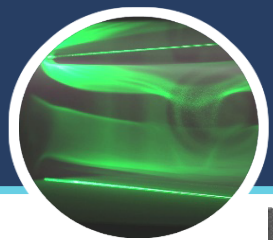
**Contraction = Increase of central plasma emission**

**Failed peaks = Failed contractions**

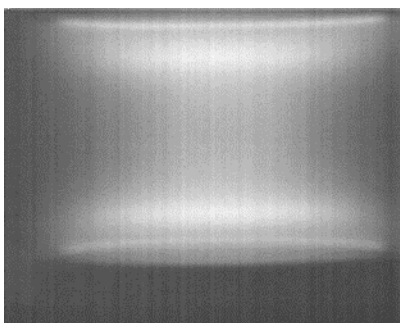


M. Mikikian, L. Couédel, M. Cavarroc, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **105**, 075002 (2010)



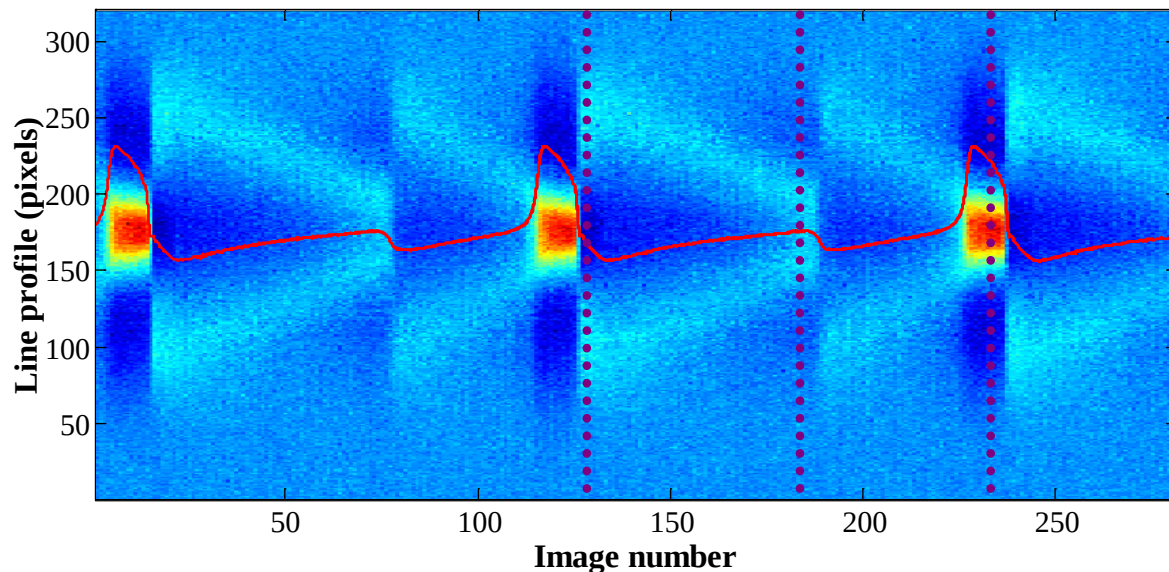
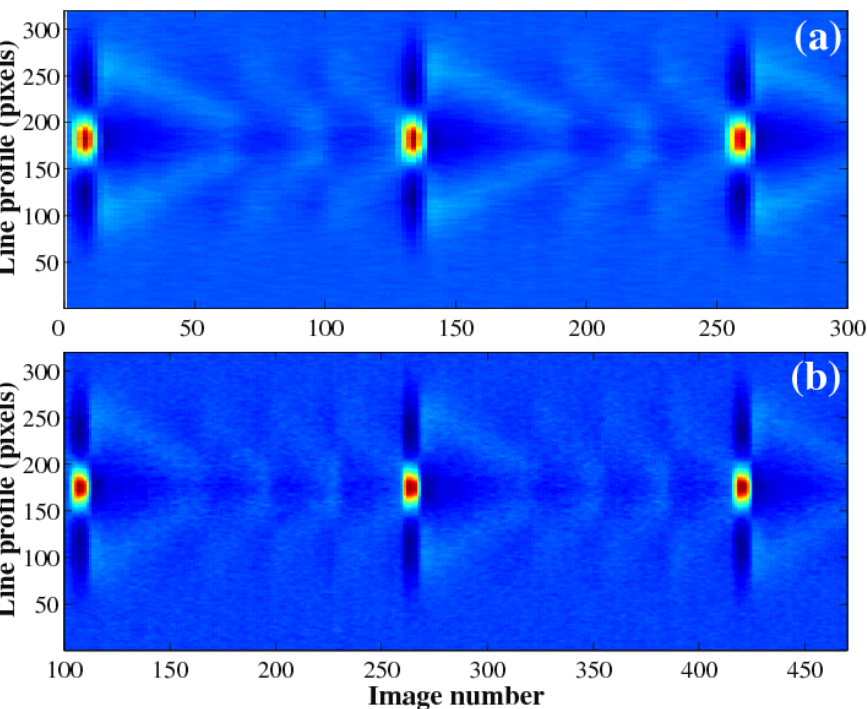
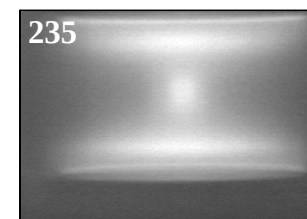
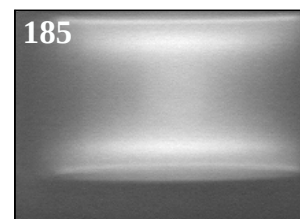
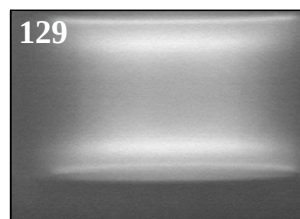


# HEARTBEAT INSTABILITIES: LAST BEATS...

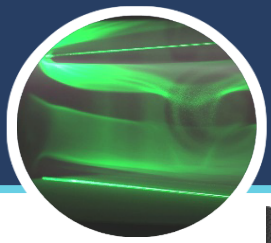


**Contraction = Increase of central plasma emission**

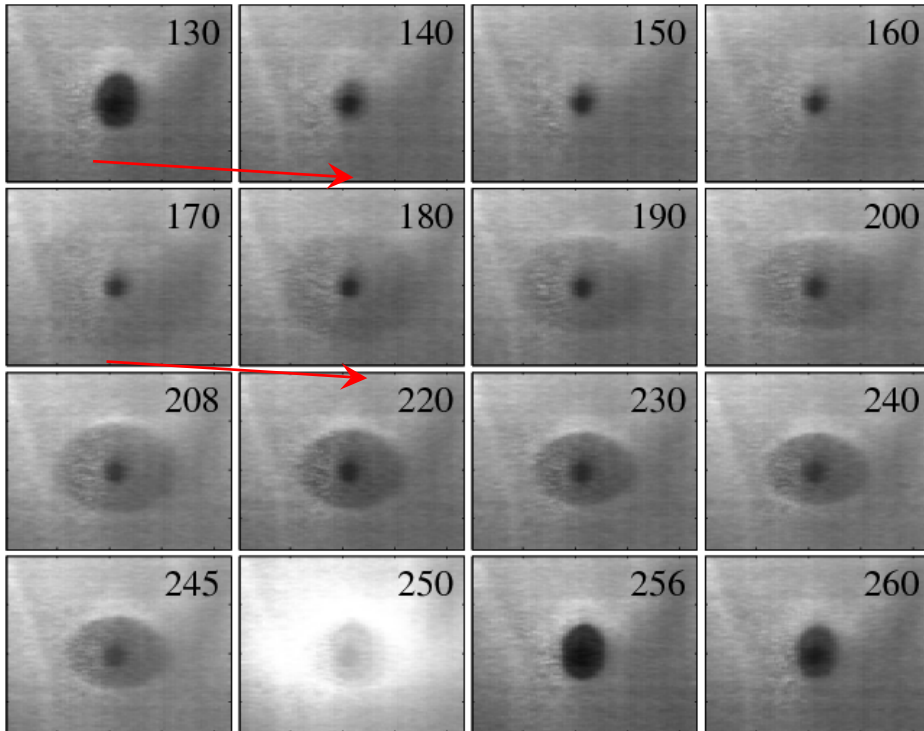
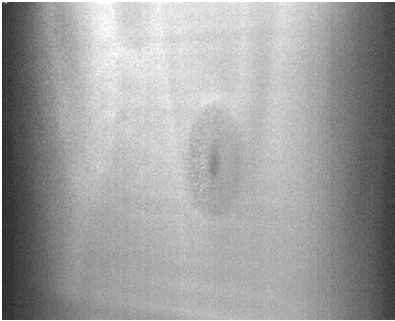
**Failed peaks = Failed contractions**



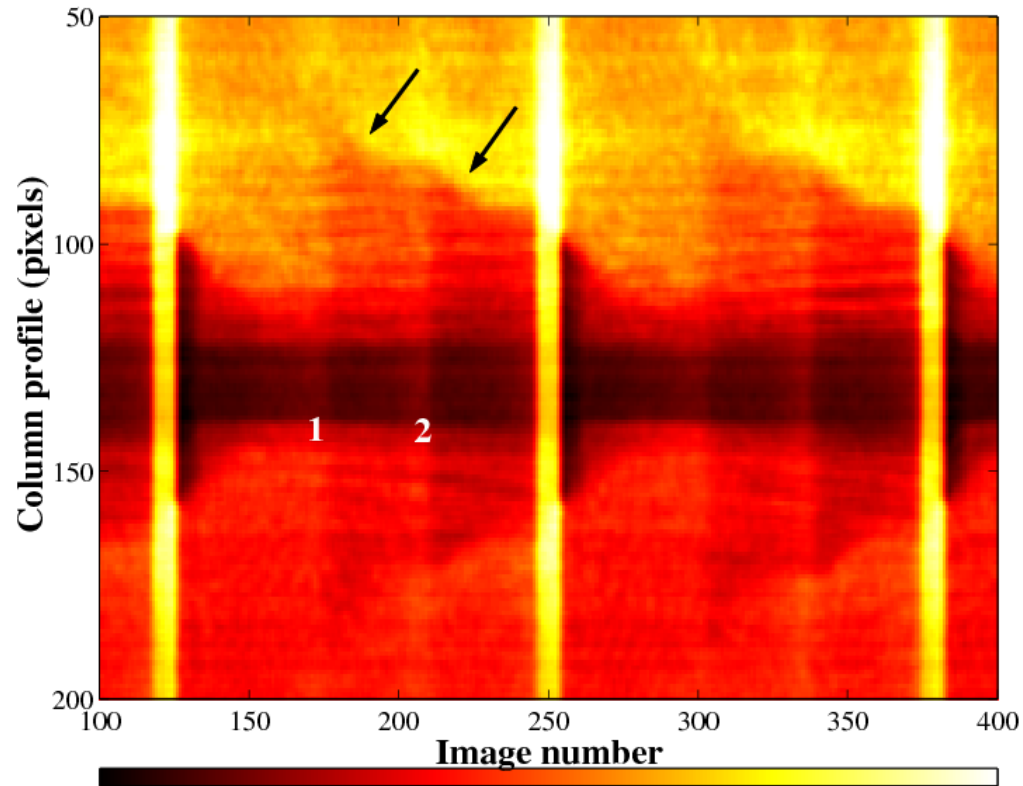
M. Mikikian, L. Couédel, M. Cavarroc, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **105**, 075002 (2010)



# HEARTBEAT INSTABILITIES: LAST BEATS...

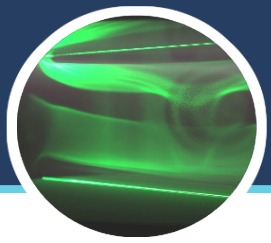


Central column profile with 2 failed peaks



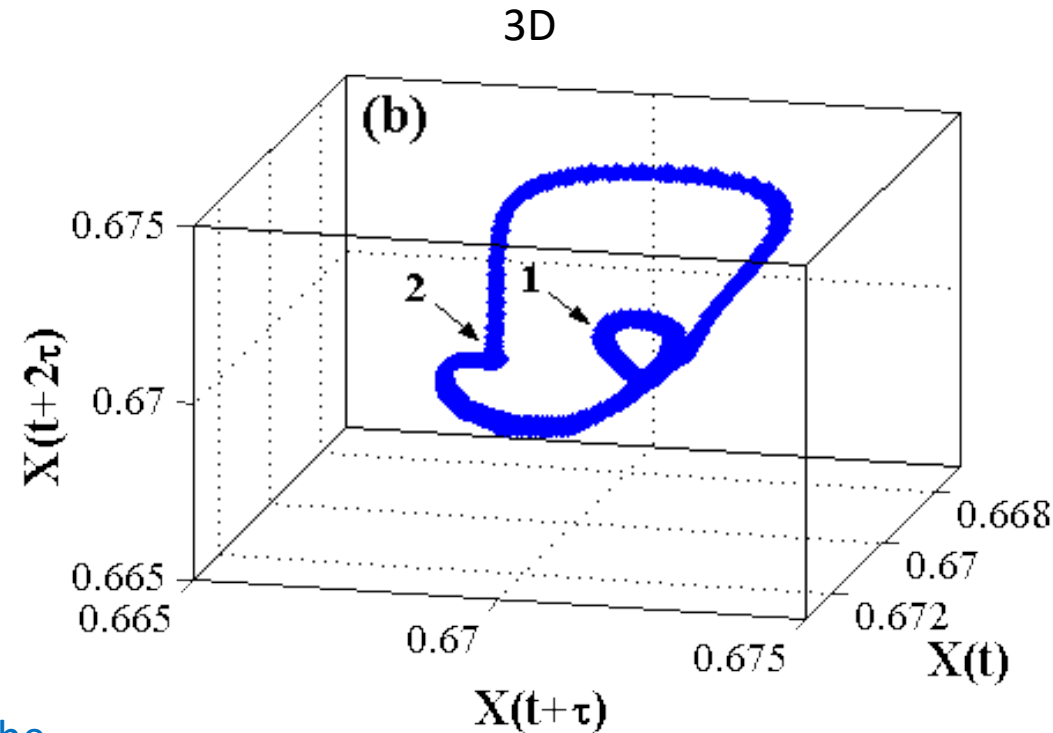
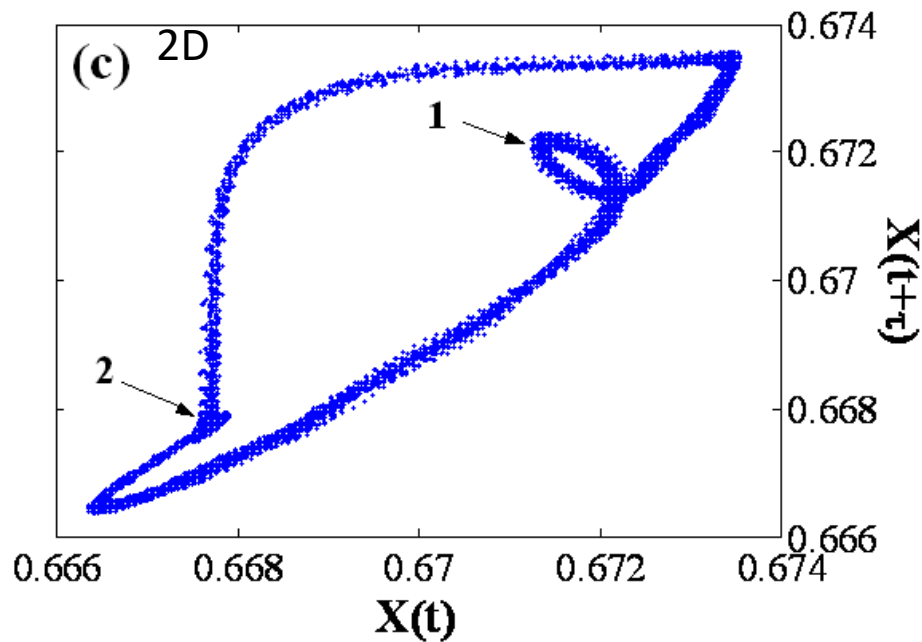
M. Mikikian, L. Couédel, M. Cavarroc, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **105**, 075002 (2010)





# HEARTBEAT INSTABILITIES: LAST BEATS...

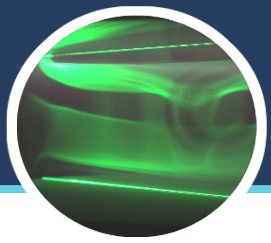
Phase Space  $X(t+\tau)$  versus  $X(t)$



Number of tiny loops in point 2 corresponds to the number of small amplitude oscillations

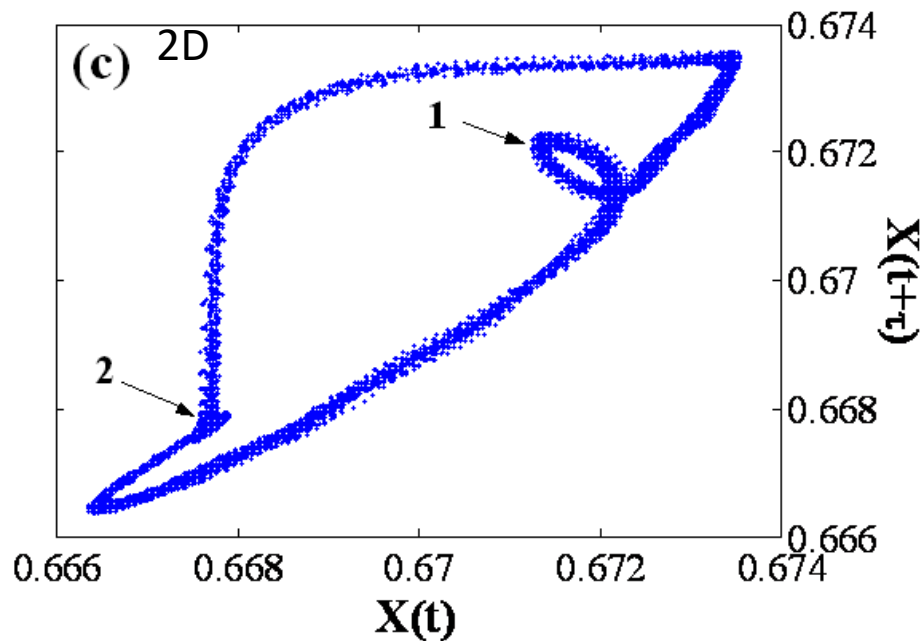
Large loops correspond to spikes

M. Mikikian, M. Cavarroc, L. Couëdel, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **100**, 225005 (2008)



# HEARTBEAT INSTABILITIES: LAST BEATS...

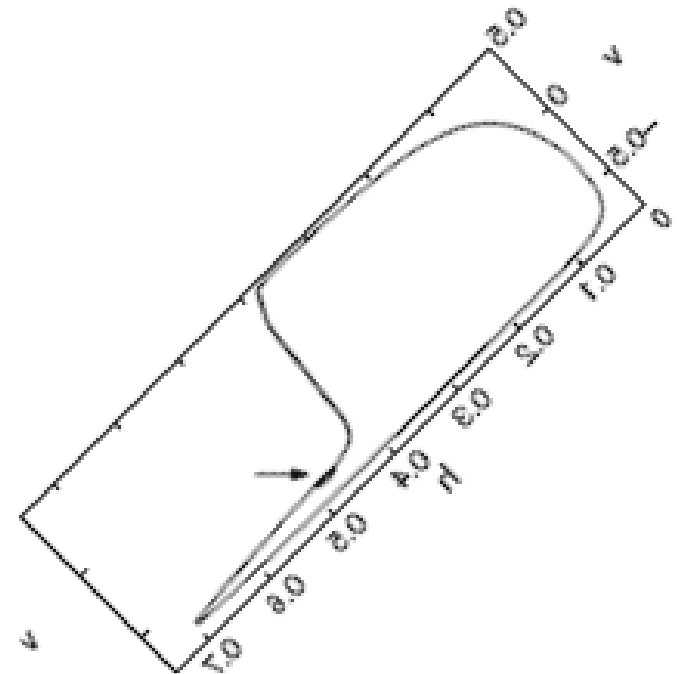
## Phase Space $X(t+\tau)$ versus $X(t)$



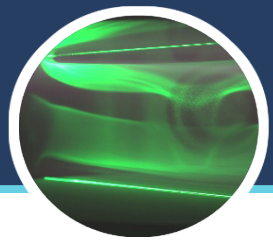
Number of tiny loops in point 2 corresponds to the number of small amplitude oscillations

Large loops correspond to spikes

MMOs in Hodgkin-Huxley model of squid giant axon, Rubin et al., Biol. Cybern. (2007)



M. Mikikian, M. Cavarroc, L. Couëdel, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **100**, 225005 (2008)

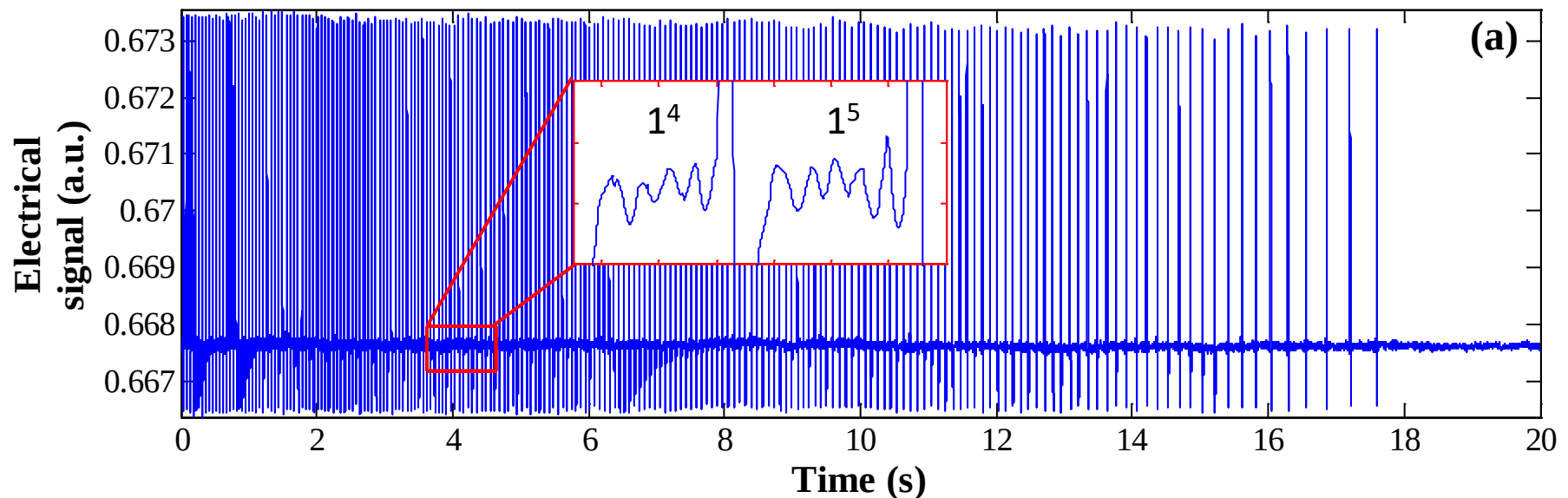


# MIXED-MODE OSCILLATIONS

Failed peaks are more and more numerous as the instability end approaches

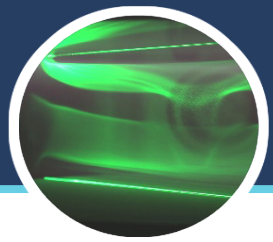
Transitions between  $L^S$  and  $L^{S+1}$  states

Very close to the end, transitions between  $L^S$  and  $L^{S+n}$  states with  $n$  from 2 to 5  
→ Increasing period in between spikes



M. Mikikian, M. Cavarroc, L. Couédel, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **100**, 225005 (2008)

M. Mikikian, H. Tawidian, T. Lecas, O. Vallée, Comptes-Rendus 15<sup>ème</sup> Rencontre du Non-Linéaire, Paris, 15-16 mars 2012, éd. E.Falcon, C.Josserand, M.Lefranc, C.Letellier, Non Linéaire Publications, p. 145-150

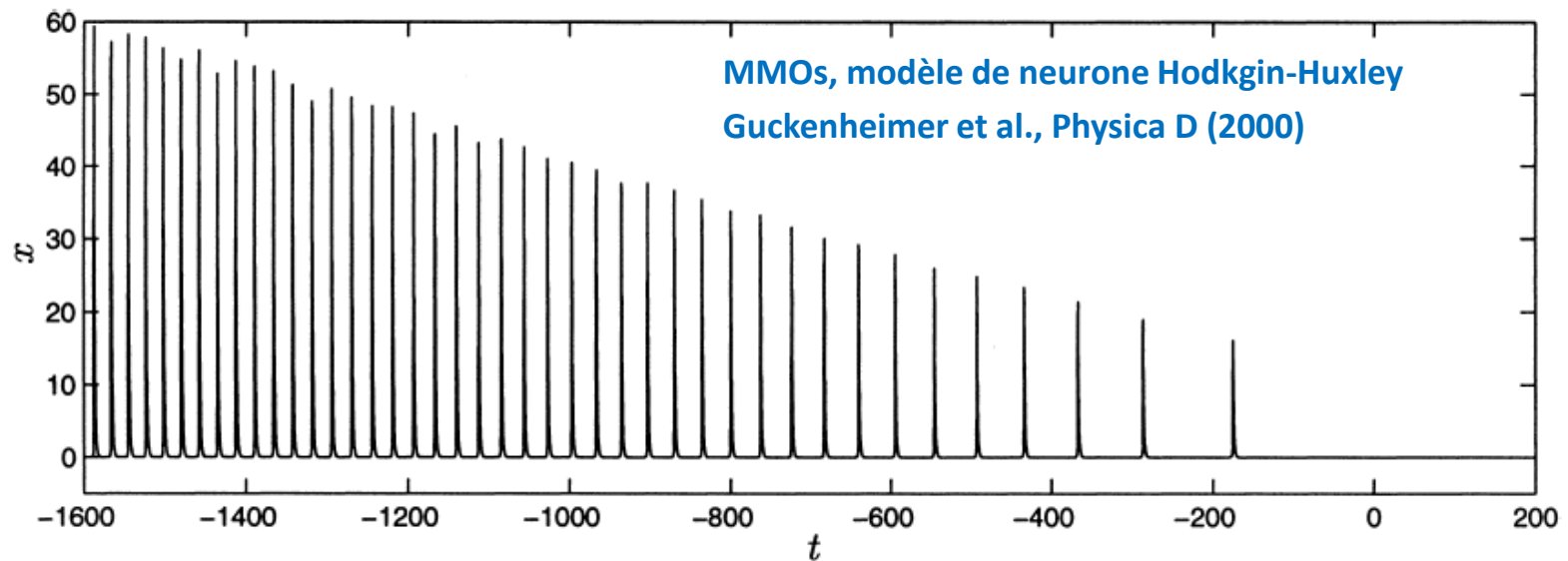


# MIXED-MODE OSCILLATIONS

Failed peaks are more and more numerous as the instability end approaches

Transitions between  $L^S$  and  $L^{S+1}$  states

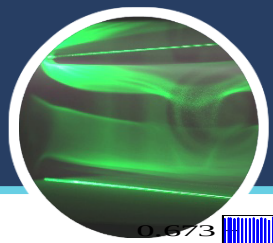
Very close to the end, transitions between  $L^S$  and  $L^{S+n}$  states with  $n$  from 2 to 5  
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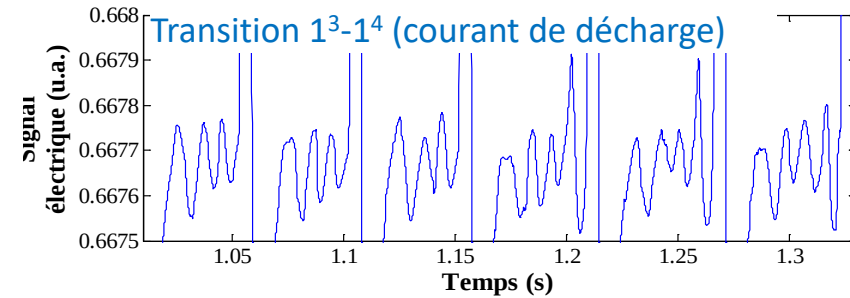
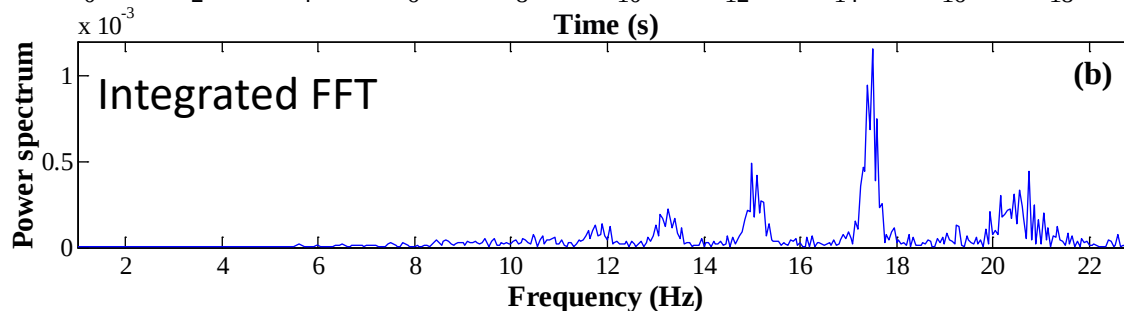
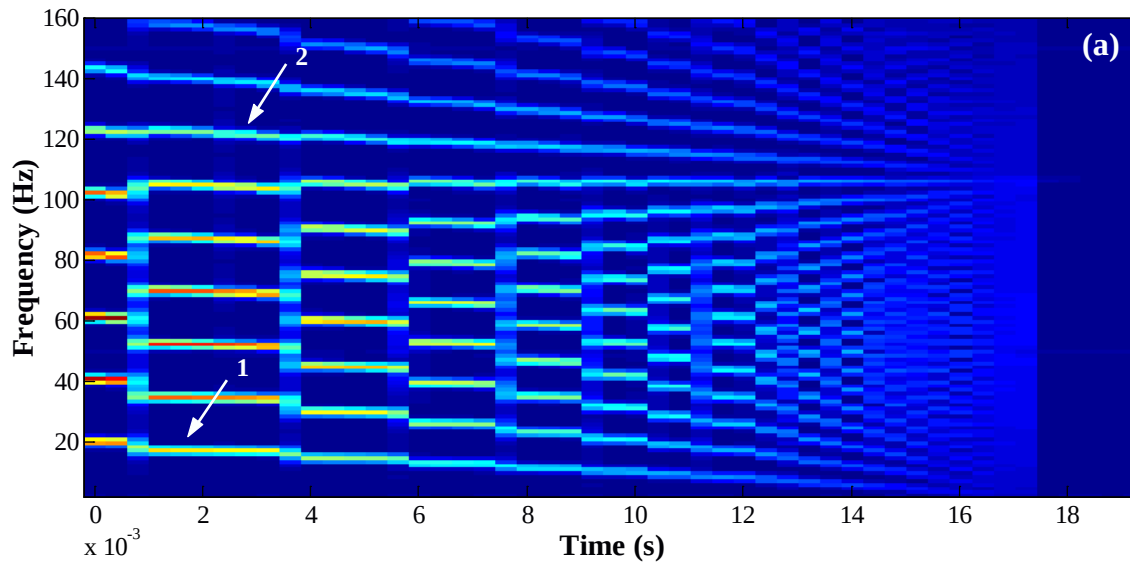
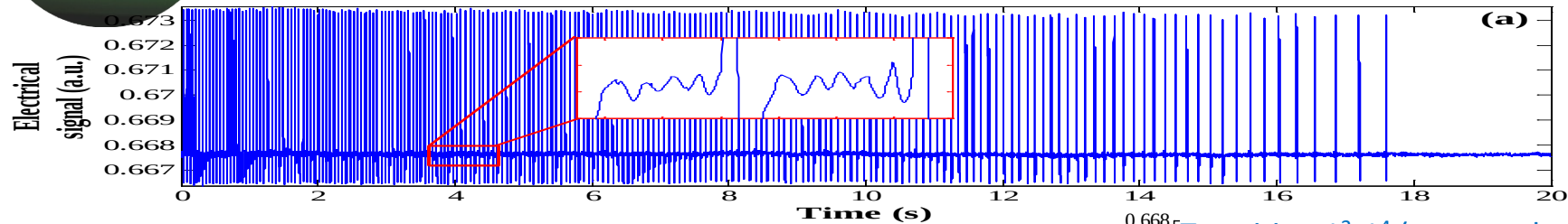
M. Mikikian, M. Cavarroc, L. Couëdel, Y. Tessier, L. Boufendi, Phys. Rev. Lett. **100**, 225005 (2008)

M. Mikikian, H. Tawidian, T. Lecas, O. Vallée, Comptes-Rendus 15<sup>ème</sup> Rencontre du Non-Linéaire, Paris, 15-16 mars 2012, éd. E.Falcon, C.Josserand, M.Lefranc, C.Letellier, Non Linéaire Publications, p. 145-150





# HEARTBEAT INSTABILITIES: LAST BEATS...



**Temporal evolution of the frequency**

→ Spectrogram

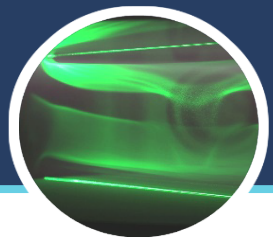
Main spike frequency (1)

Small amplitude oscillation frequency (2)

**Due to occurrence of more and more failed peaks, decrease by steps of :**

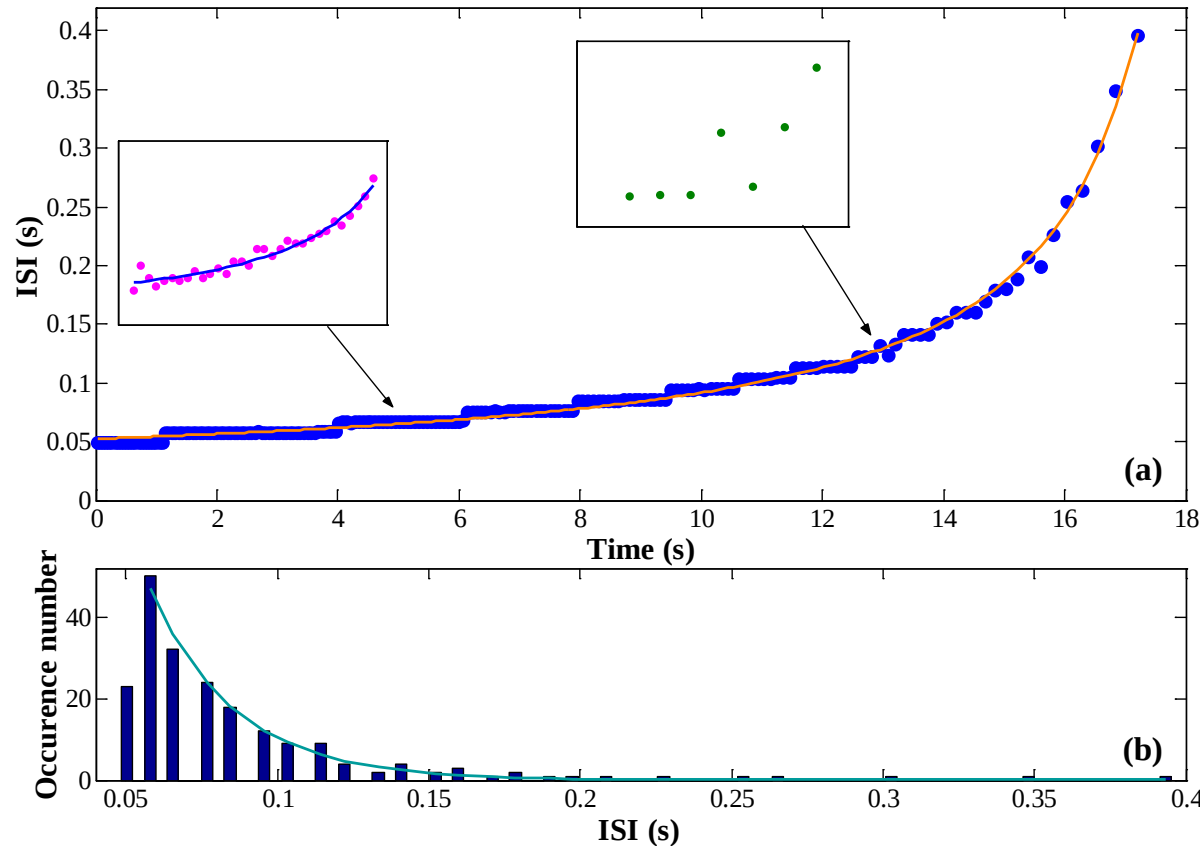
Spike frequency

Duration of each new  $L^S$  state



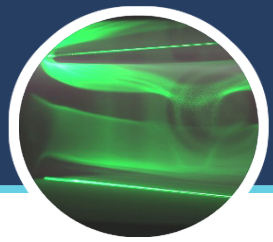
# HEARTBEAT INSTABILITIES: LAST BEATS...

Statistics on Interspike intervals is often used in neuroscience for characterizing neuronal activity



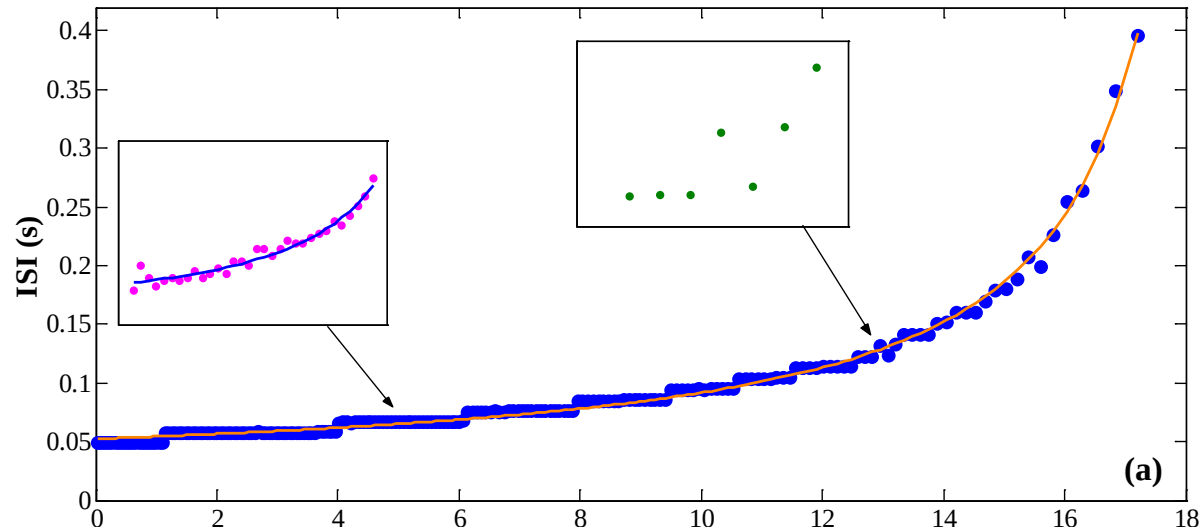
ISI increases as the end approaches  
(frequency decreases)

Each  $L^S$  state is characterized by a small  
but progressive increase of the ISI  
Transition to a new  $L^{S+1}$  state occurs  
after the progressive increase of the ISI  
in the  $L^S$  state



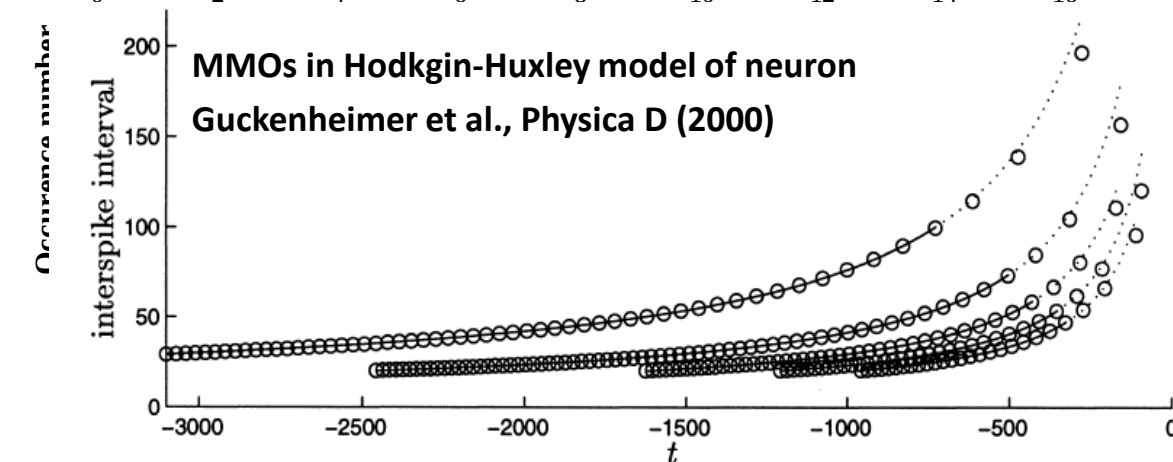
# HEARTBEAT INSTABILITIES: LAST BEATS...

Statistics on Interspike intervals is often used in neuroscience for characterizing neuronal activity

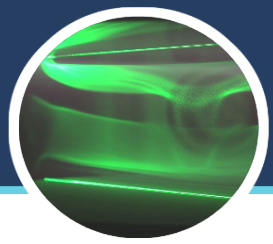


ISI increases as the end approaches  
(frequency decreases)

Each  $L^S$  state is characterized by a small but progressive increase of the ISI  
Transition to a new  $L^{S+1}$  state occurs after the progressive increase of the ISI in the  $L^S$  state

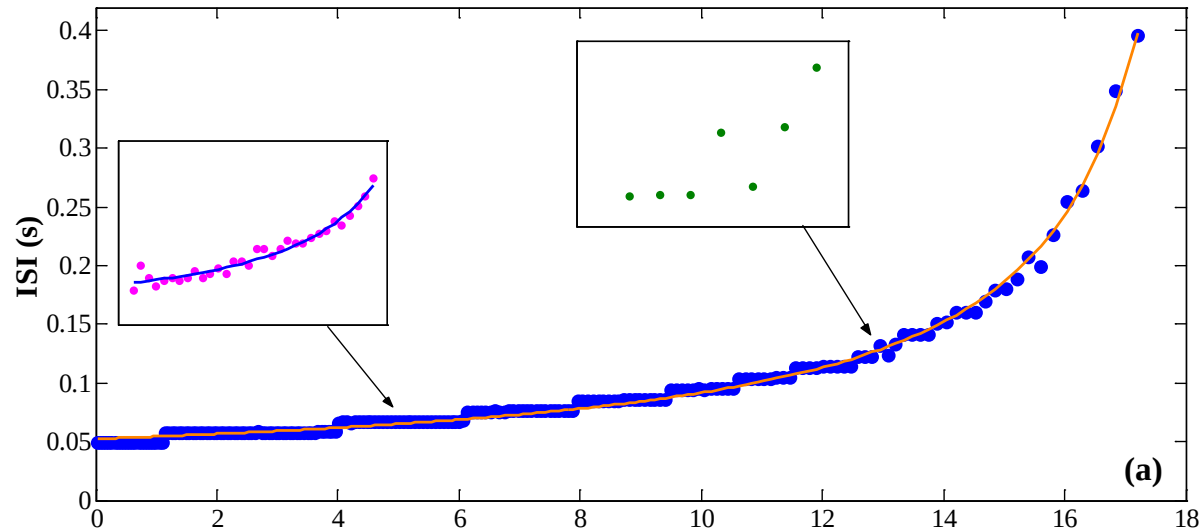


Mikikian et al. Phys. Rev. Lett. **100**, 225005 (2008)

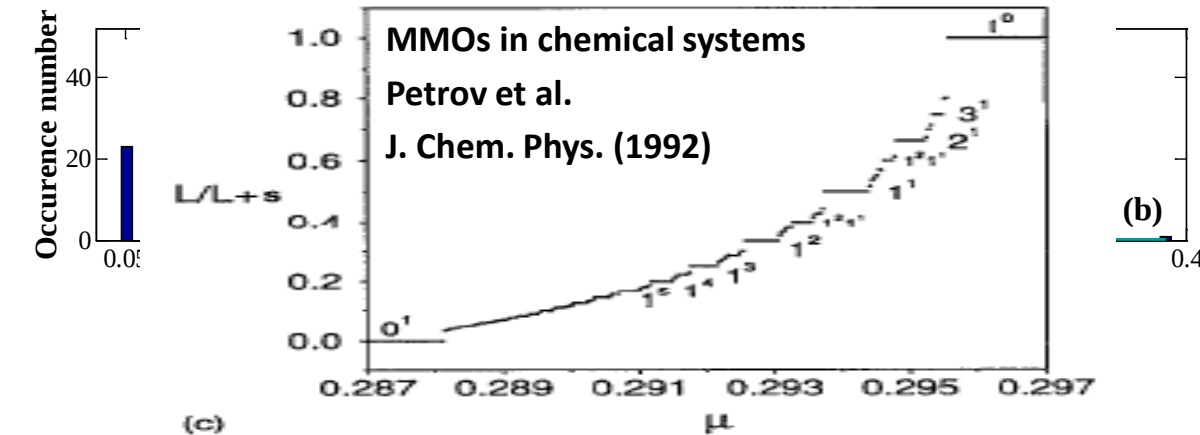


# HEARTBEAT INSTABILITIES: LAST BEATS...

Statistics on Interspike intervals is often used in neuroscience for characterizing neuronal activity

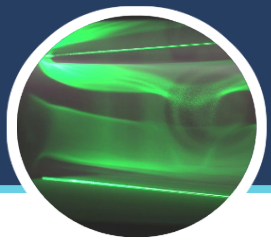


ISI increases as the end approaches  
(frequency decreases)



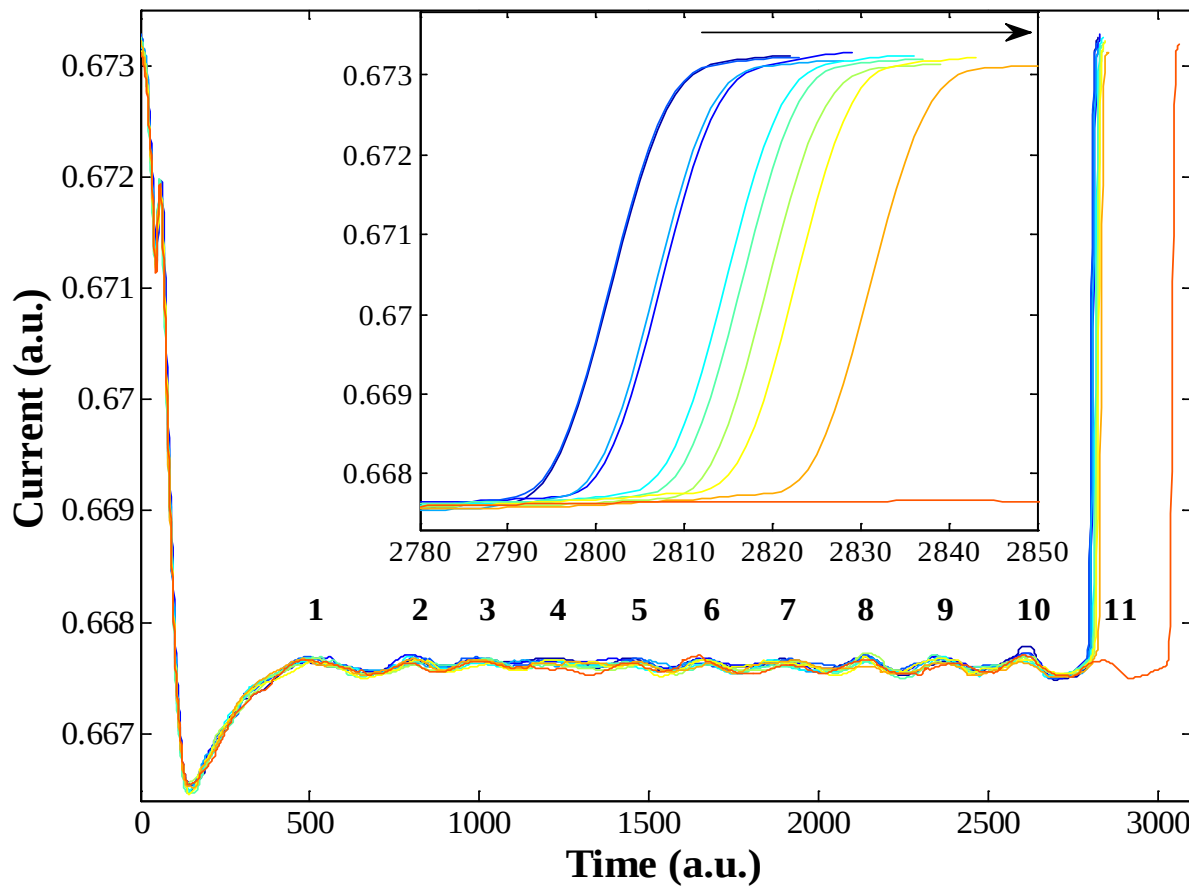
Step behavior in nonlinear systems :  
Devil's staircase  
Farey trees

Mikikian et al. Phys. Rev. Lett. **100**, 225005 (2008)



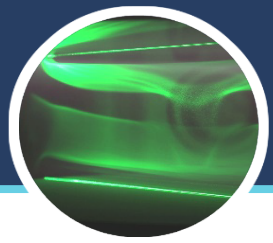
# HEARTBEAT INSTABILITIES: LAST BEATS...

Superimposition of states  $1^{10}$  and transition to  $1^{11}$



M. Mikikian, H. Tawidian, T. Lecas, O. Vallée, Chaos, Complexity and Transport, Proc. CCT'11, eds. X. Leoncini, M. Leonetti, World Scientific Publishing, Singapore, p. 85-93 (2012)





# HEARTBEAT INSTABILITIES: LAST BEATS...

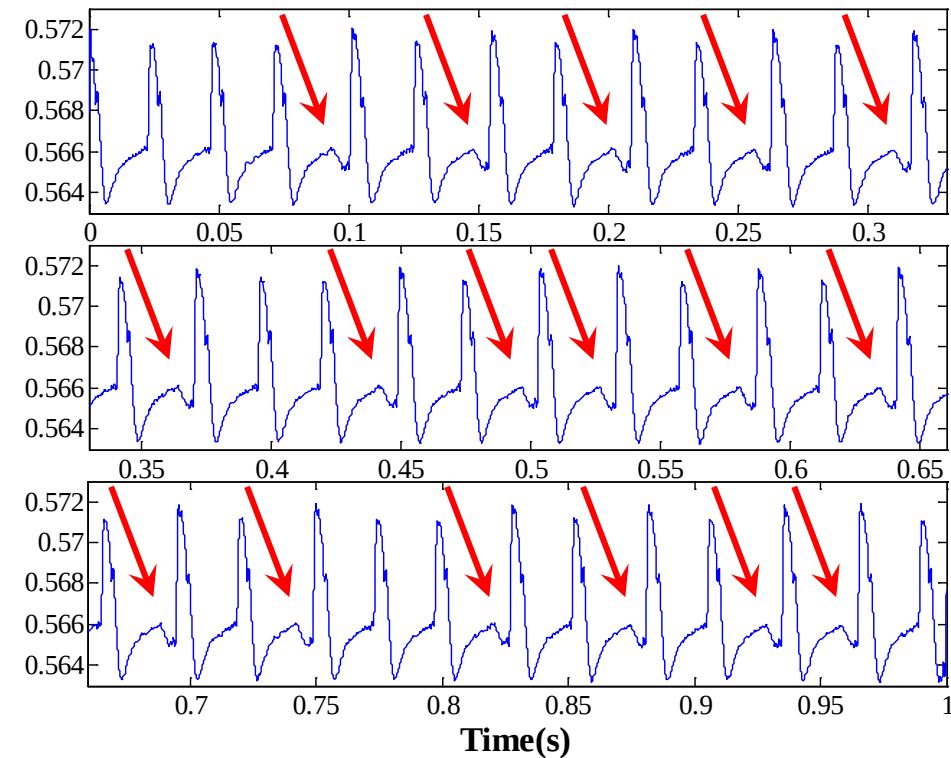
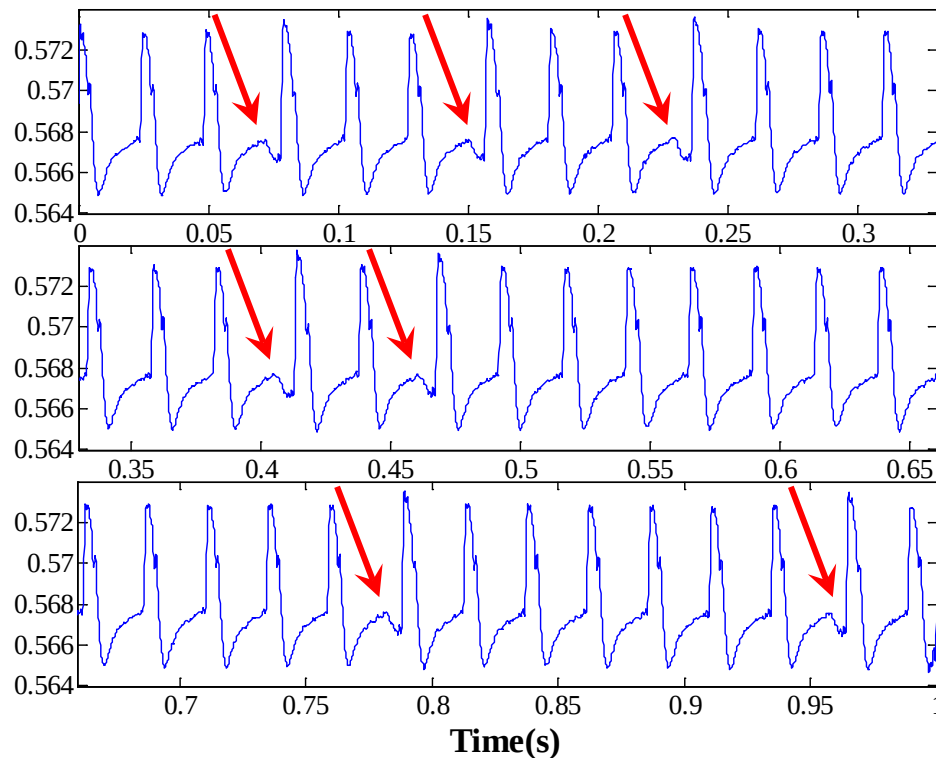
**Alternation of two states differing by one failed peak**

Progressive occurrence of  $L^{S+1}$  patterns during a  $L^S$  state

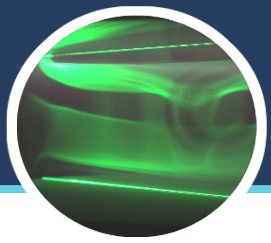
**Incomplete devil's staircase**

Intermediate states related to the parent states by the Farey arithmetic?

More intermediate states with slower varying signals to complete the staircase

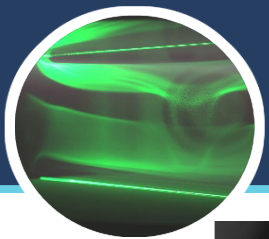


M. Mikikian, H. Tawidian, T. Lecas, O. Vallée, Chaos, Complexity and Transport, Proceedings CCT'11, eds. X. Leoncini, M. Leonetti, World Scientific Publishing, Singapore, p. 85-93 (2012)



# TO BE DONE FOR PERISTOCH

- Reproduce the experiments and acquire more new data like MMO
- Evaluate the role of the external electrical circuit
- Find any dependence with the signal amplitudes (thresholds?) or with the noise level
- Determine a set of equations modelling main void characteristics: from existing void theories or from existing MMO models
- Comparisons modeling/experiments



# THANK YOU !

