Molecular excitation by chirped laser radiation in ladder climbing and autoresonance regimes

Gilad Marcus, Arie Zigler and Lazar Friedland

Racah Institute of Physics, Hebrew University, Jerusalem, Israel

Outlines for the Lecture

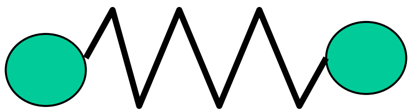
- Definition of the problem

- Ladder climbing and the Auto-Resonance concepts

- Ladder-climbing experiment on HF molecule
  - Radiation source for
  - Excitation of the molecule

Basics of Auto-Resonance

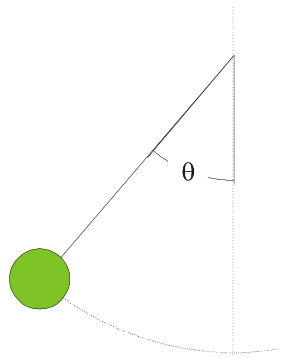
Harmonic Oscillator



$$m\ddot{r} = -kr$$

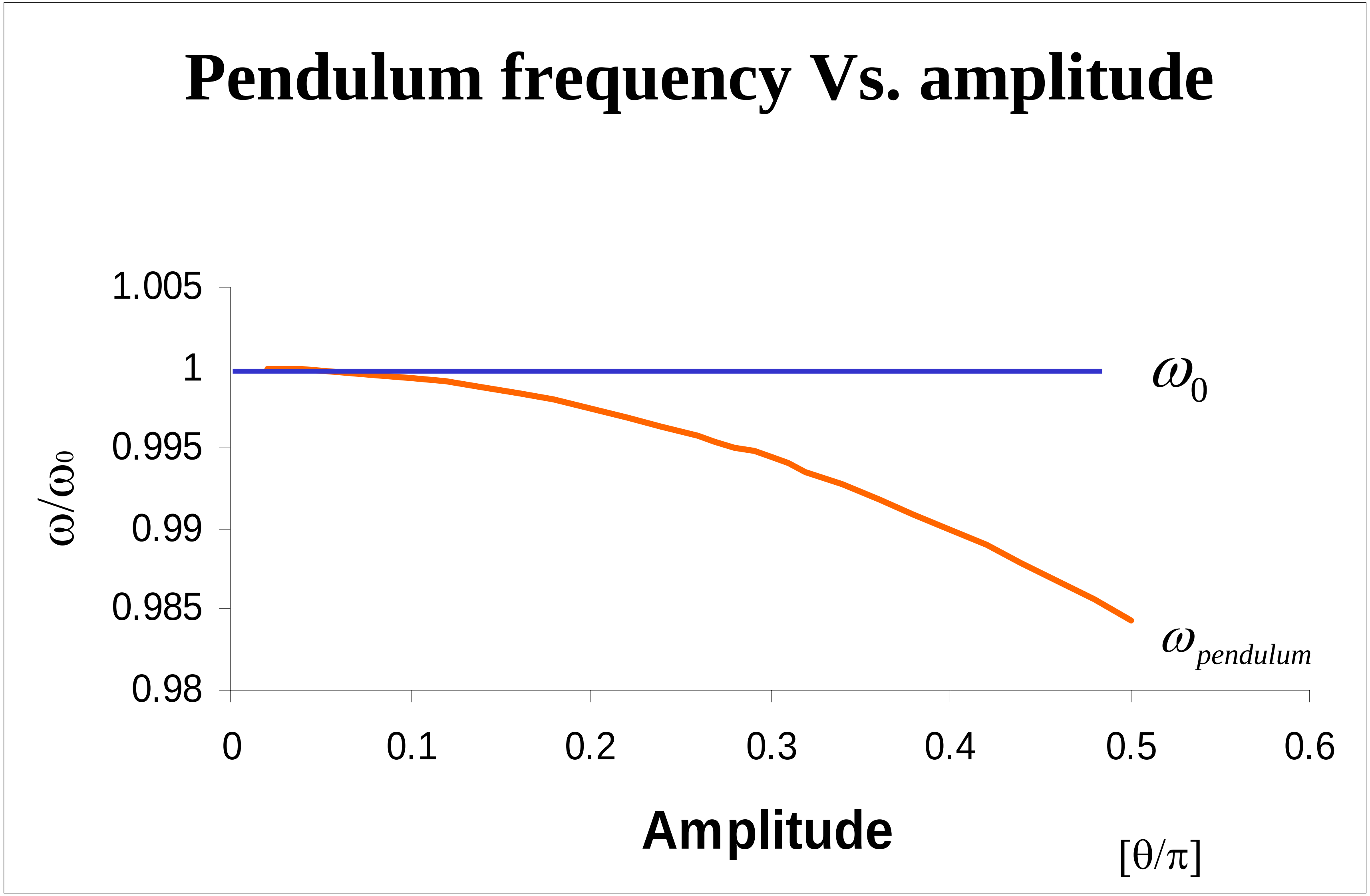
$$\omega_0 = \sqrt{\frac{k}{m}}$$

Anharmonic Oscillator



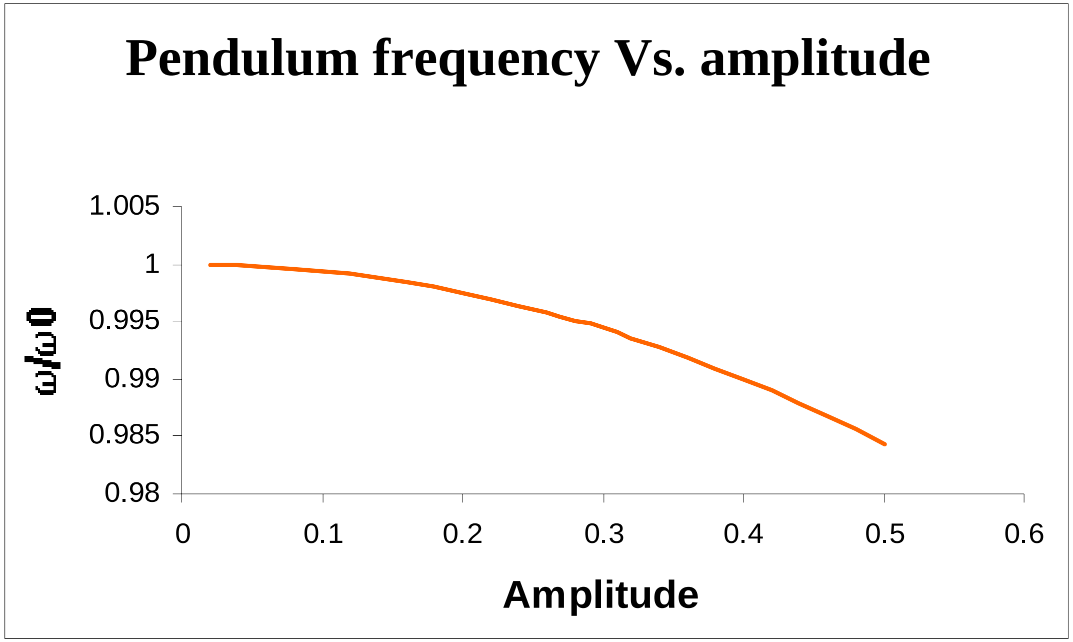
$$l\ddot{\theta} = -g \cdot \sin(\theta) \cong -g\theta + \frac{g\theta^3}{3!}$$

$$\omega_0 = \sqrt{\frac{g}{l}} + f(\theta_0)$$



Pendulum frequency Vs. amplitude
1.005
1
0.995
0.99
0.985
0.98
0
0.1
0.2
0.3
0.4
0.5
0.6
ω/ω0
ω0
ωpendulum
Amplitude
[θ/π]

- How to excite nonlinear systems into high energy ?
- Changing the drive frequency will keep it in resonance.

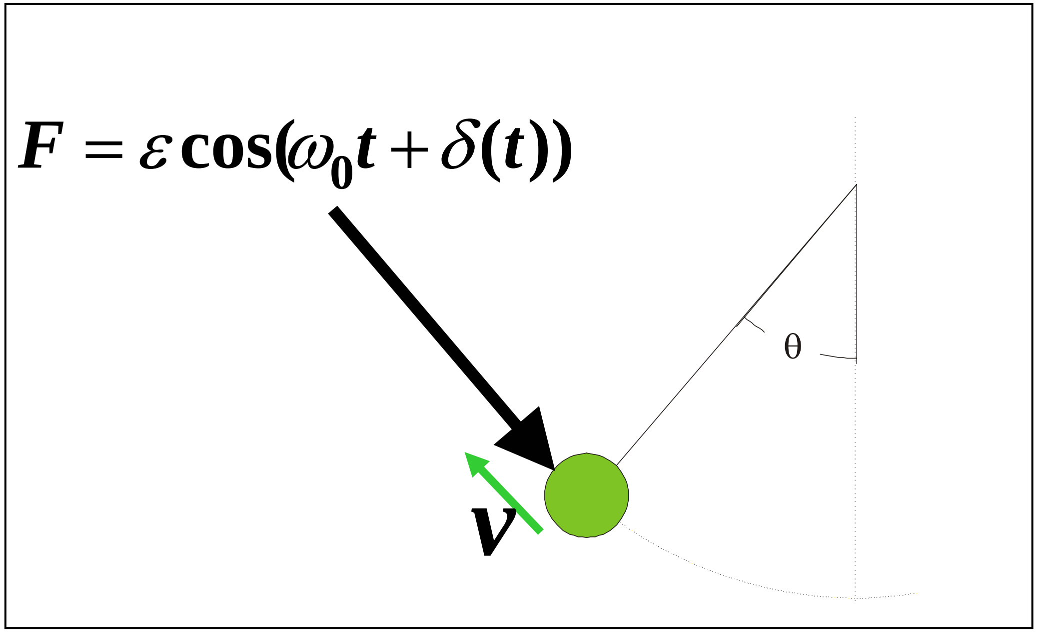


- How to excite nonlinear systems into high energy ?
- Changing the drive frequency will keep it in resonance.

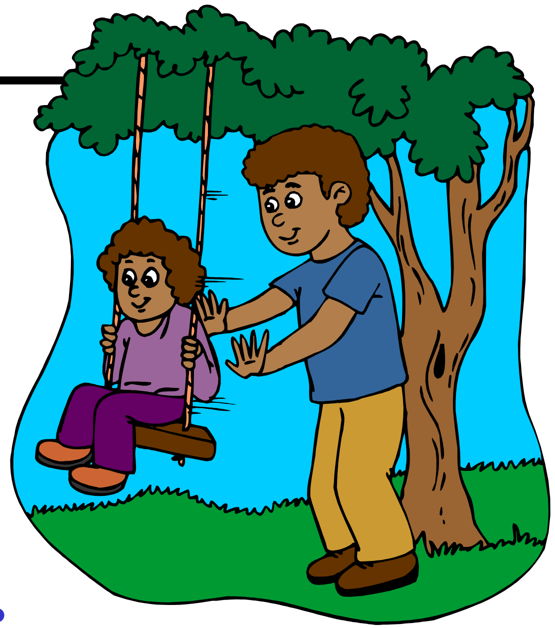
but we also have to

continually adjust

the phase



Few method to excite nonlinear oscillator



1. **Feedback control.**

   (requires a real time feedback)

2. **Exact tailoring of the force.**

   (requires pre-knowledge of the system)

3. **Ladder-climbing & autoresonance**

Auto-Resonance

- **The drive frequency is slowly changed**
  (slow chirp)

- The oscillator is automatically phase locked
  (provided that the force exceed a certain threshold)

- The energy of the oscillator is a function of the drive frequency

Threshold-chirp relation

$$\varepsilon > \varepsilon_{th} = 0.82m\sqrt{\omega_0}\left(\frac{\alpha^{3/4}}{\beta_c^{1/2}}\right)$$

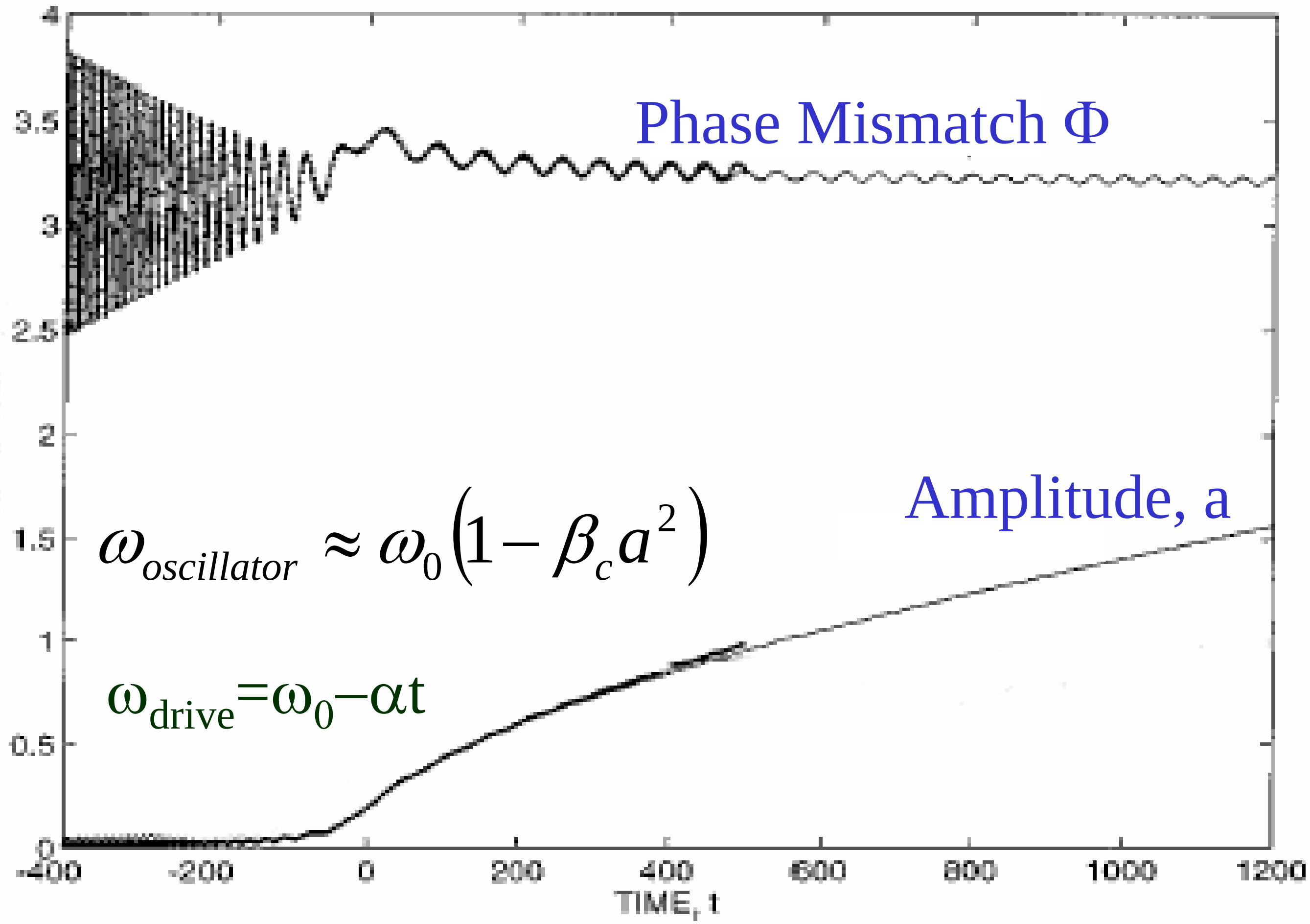
$$\varepsilon = drive\ \ force$$

$$\alpha = chirp - rate$$

$$\beta_c = Oscillator\ \ - nonlinearity$$

$$\omega(a) = \omega_0\left(1 - \beta_c a^2\right)$$

Auto-Resonance simulation



{ L. Friedland et al. Phys. Plasmas. 5 (645) }

Ladder climbing in a quantum systems

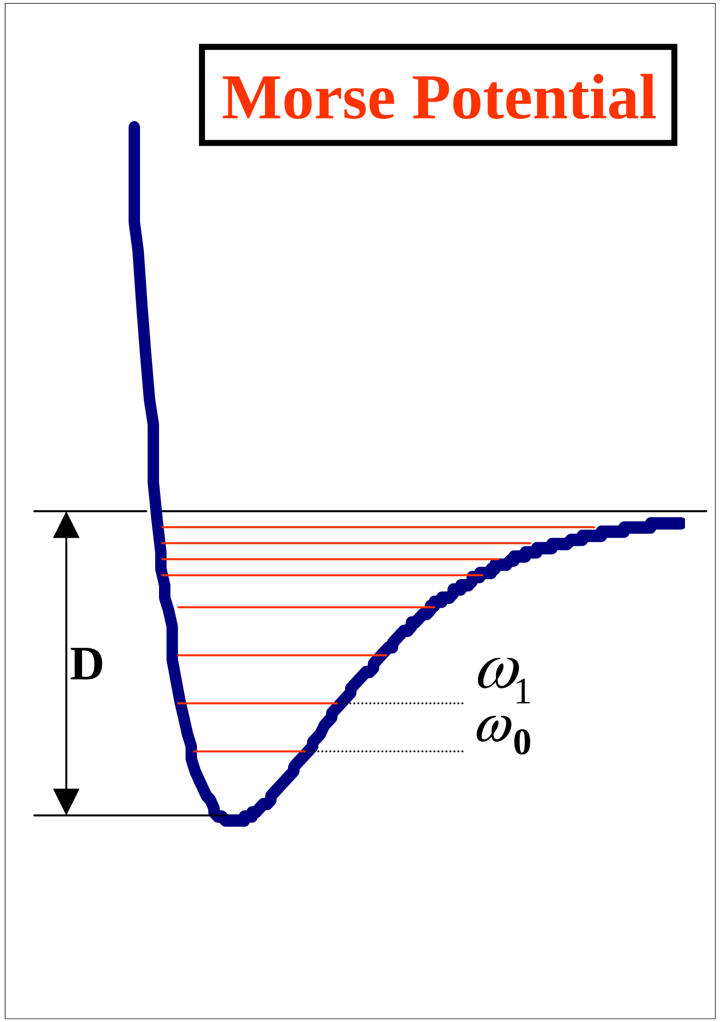
- Energy levels in Morse potential.

$$E_n \approx \hbar\omega_0\left[(n+1/2) - \beta_q (n+1/2)^2\right]$$

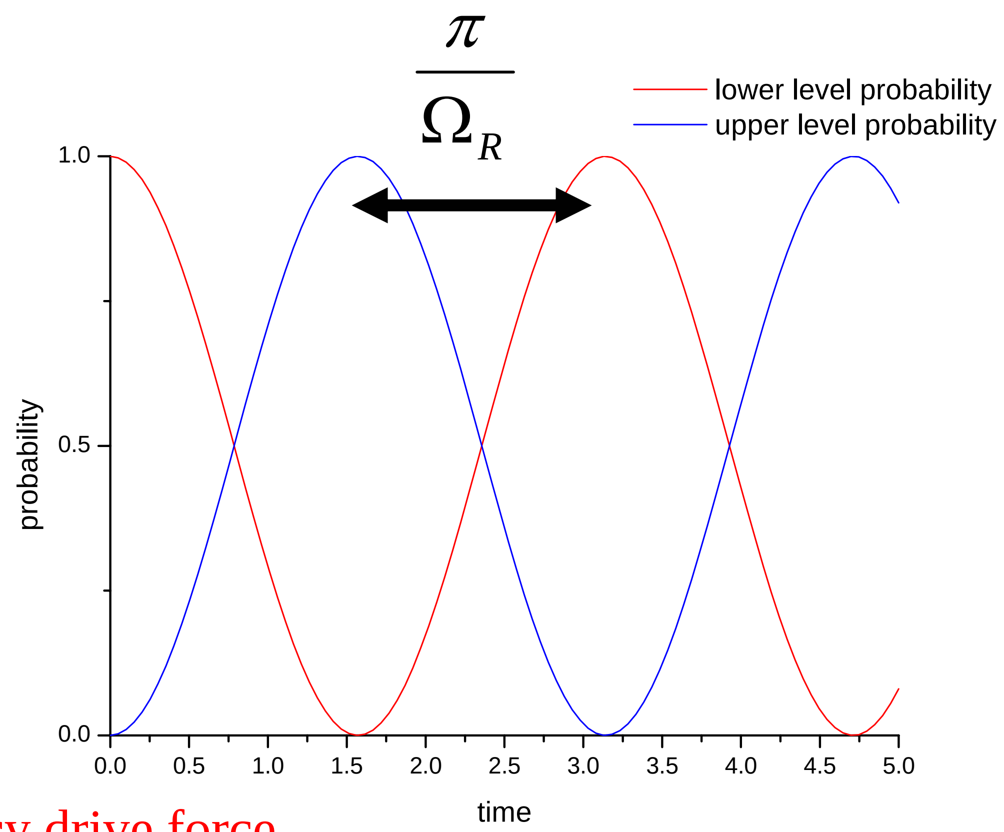
$$\omega_n = \omega_0 (1 - 2\beta_q n)$$

$$\omega_{n+1} < \omega_n$$

Ladder of energy levels with decreasing gaps.



Two levels with constant frequency drive



constant frequency drive force

$$E = E \cdot \exp(i\omega_n t)$$

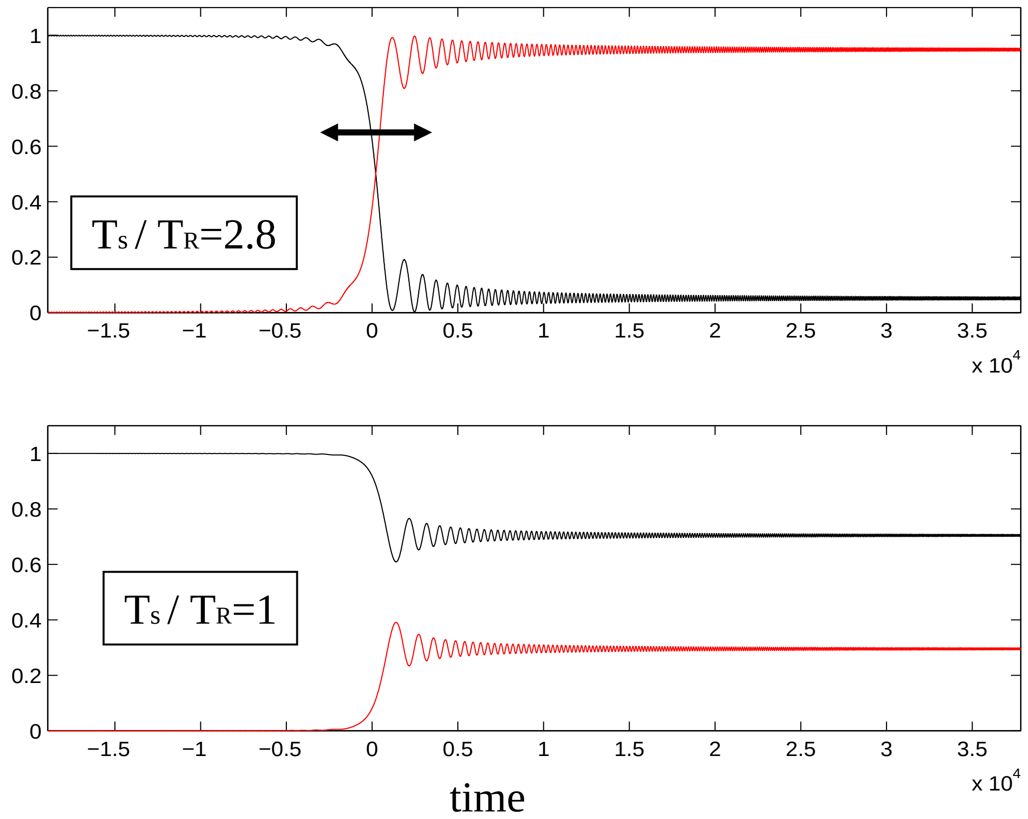
$$\hbar\Omega_{Rabi} = \mu E$$

Two level with chirped drive

$$T_S = 1/\sqrt{\alpha}$$

Efficient conversion when $\Omega_R^{\ 2}/\alpha > 2$

$$T_S / T_R > \sqrt{2}$$



chirped drive force

$$E = E \cdot \exp(i(\omega_0 - \tfrac{1}{2}\alpha \cdot t)t)$$

The validity of the two level approximation

$$\varepsilon\sqrt{\hbar / 2m\omega_0} < 2\hbar\omega_0\beta_q$$

Which means – the width of resonance is small enough to include only two levels

Characteristic times

$$T_R = 1/\Omega_R = \sqrt{2m\hbar\omega_0}/\varepsilon$$

$$T_S = 1/\sqrt{\alpha}$$

$$T_{NL} = 2\omega_0\beta_q/\alpha = 2\hbar\beta_c/m\alpha$$

The limit between quantum mechanics and classicality

$$\varepsilon\sqrt{\hbar / 2m\omega_0} > 2\hbar\omega_0\beta_q$$

In terms of the three characteristic times:

$$T_s^2 / \left(T_R T_{NL}\right) > 1$$

The condition for efficient ladder-climbing

$$\Omega_R / \alpha > \sqrt{2}$$

In terms of the three characteristic times:

$$T_s / T_R > \sqrt{2}$$

Efficient classical autoresonance

$$\varepsilon_{th} = 0.82m(\omega_0 / \beta_c)^{1/2}\alpha^{3/4}$$

In term of the characteristic times:

$$T_R^2 / \left(T_S T_{NL}\right) < 1.48$$

P_1-P_2 parameters

$$P_1 = T_S / T_R \qquad\qquad P_2 = T_{NL} / T_S$$

Quantum limit:

$$T_s^2 / \left(T_R T_{NL}\right) < 1 \;\Rightarrow\; \boxed{P_1 < P_2}$$

Efficient transfer between 2 levels:

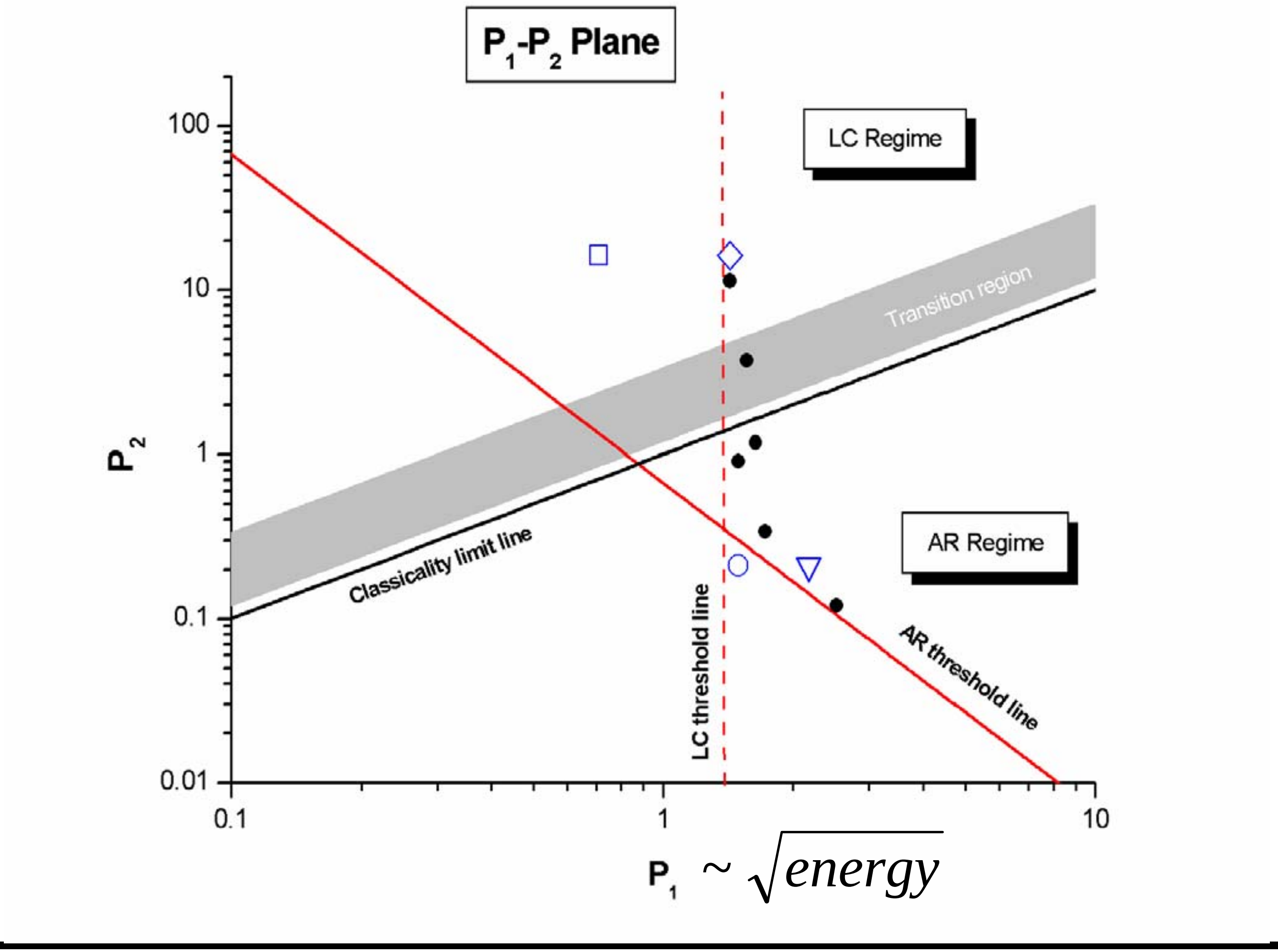
$$T_s / T_R > \sqrt{2} \;\Rightarrow\; \boxed{P_1 > \sqrt{2}}$$

Efficient Autoresonance:

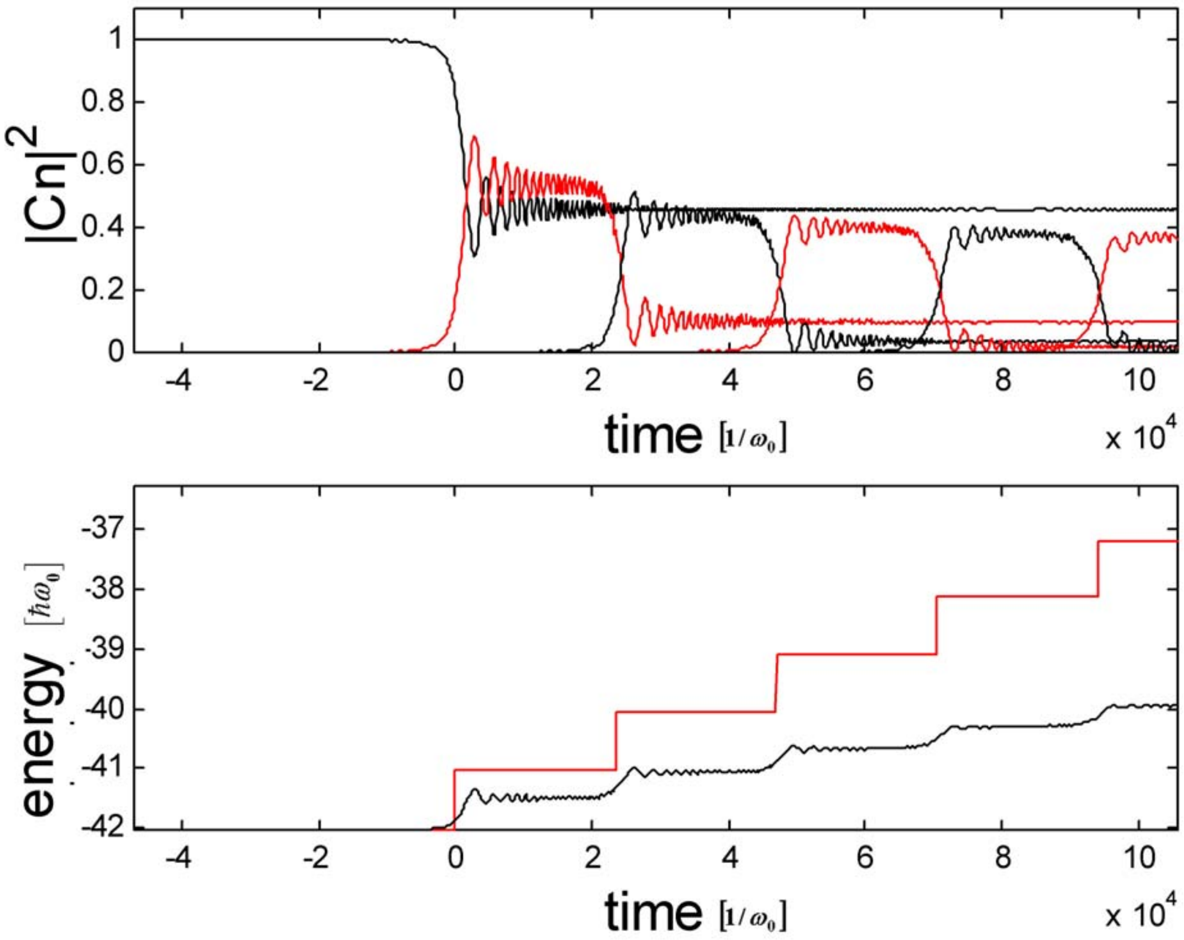
$$T_R^2 / \left(T_S T_{NL}\right) < 1.48 \;\Rightarrow\; \boxed{P_2 > 0.67 / P_1^2}$$

P_1-P_2 parameters

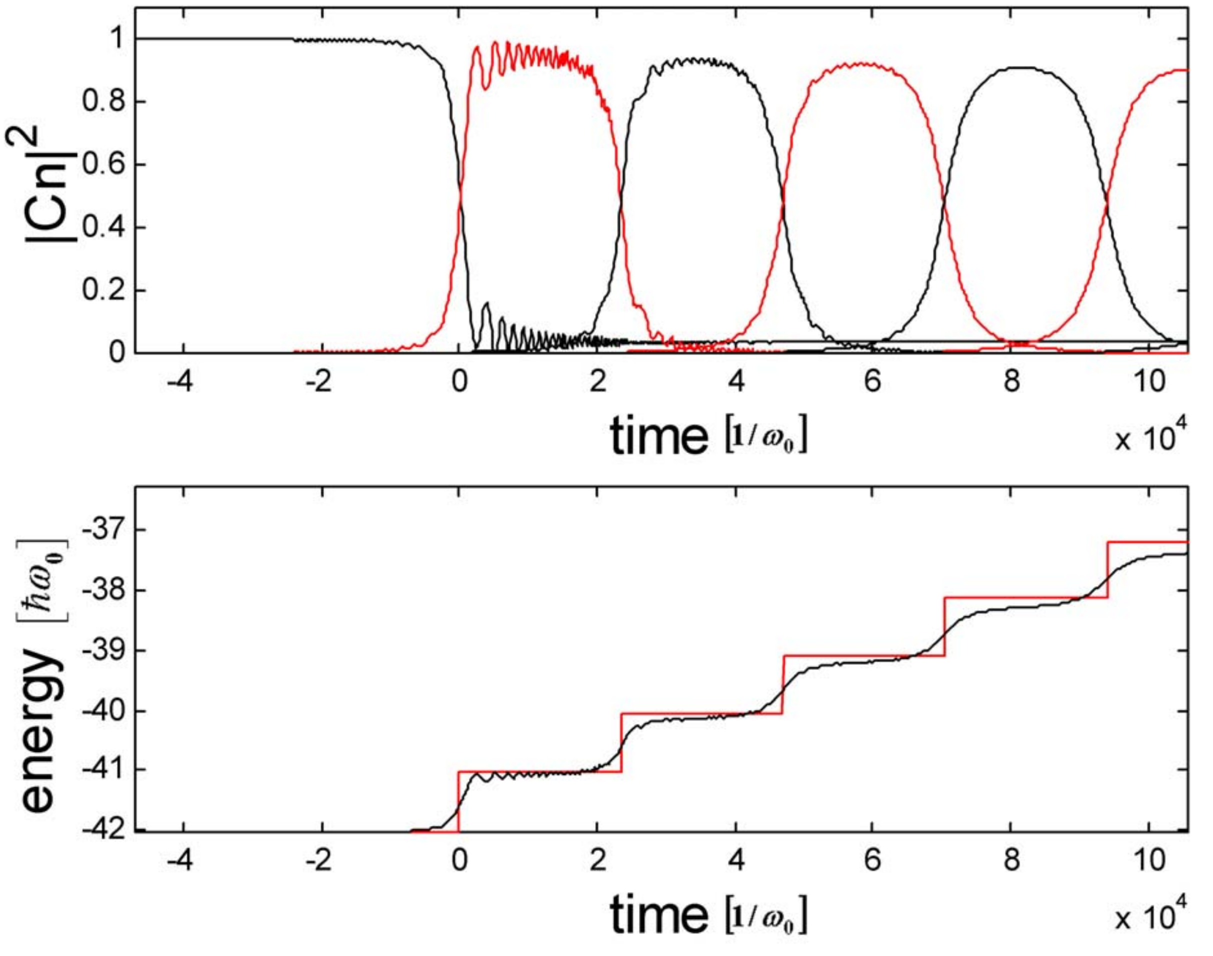
$$P_1 = T_S / T_R \qquad P_2 = T_{NL} / T_S$$



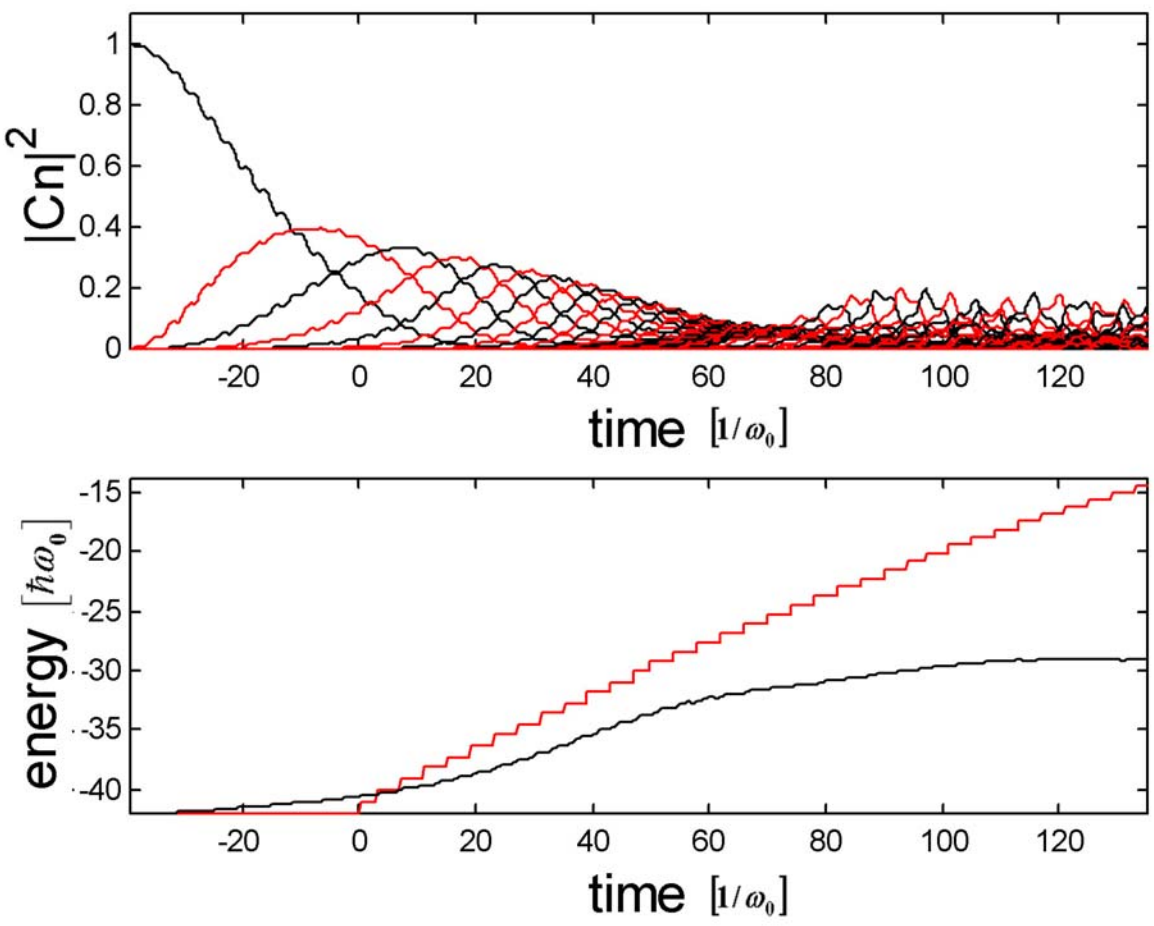
Ladder climbing-below threshold



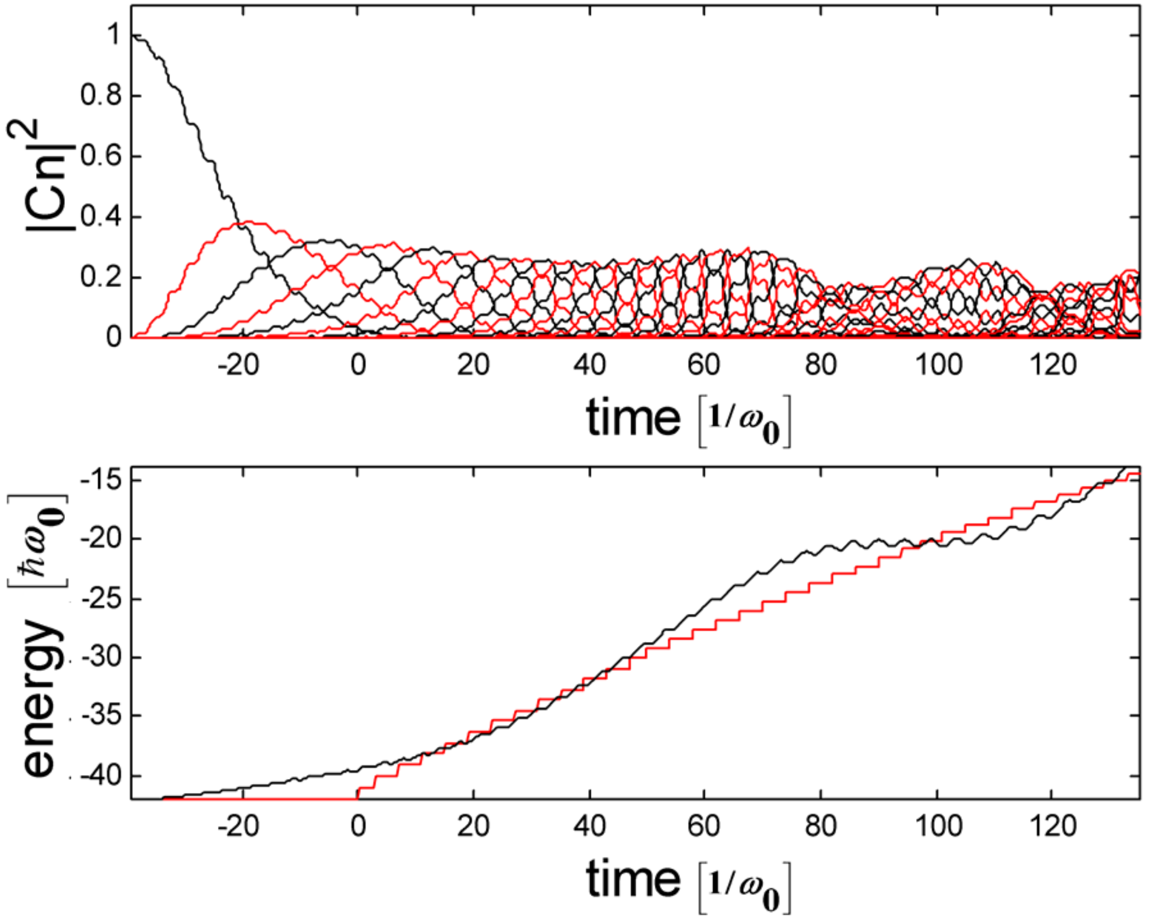
Ladder climbing-above threshold



Autoresonance-below threshold

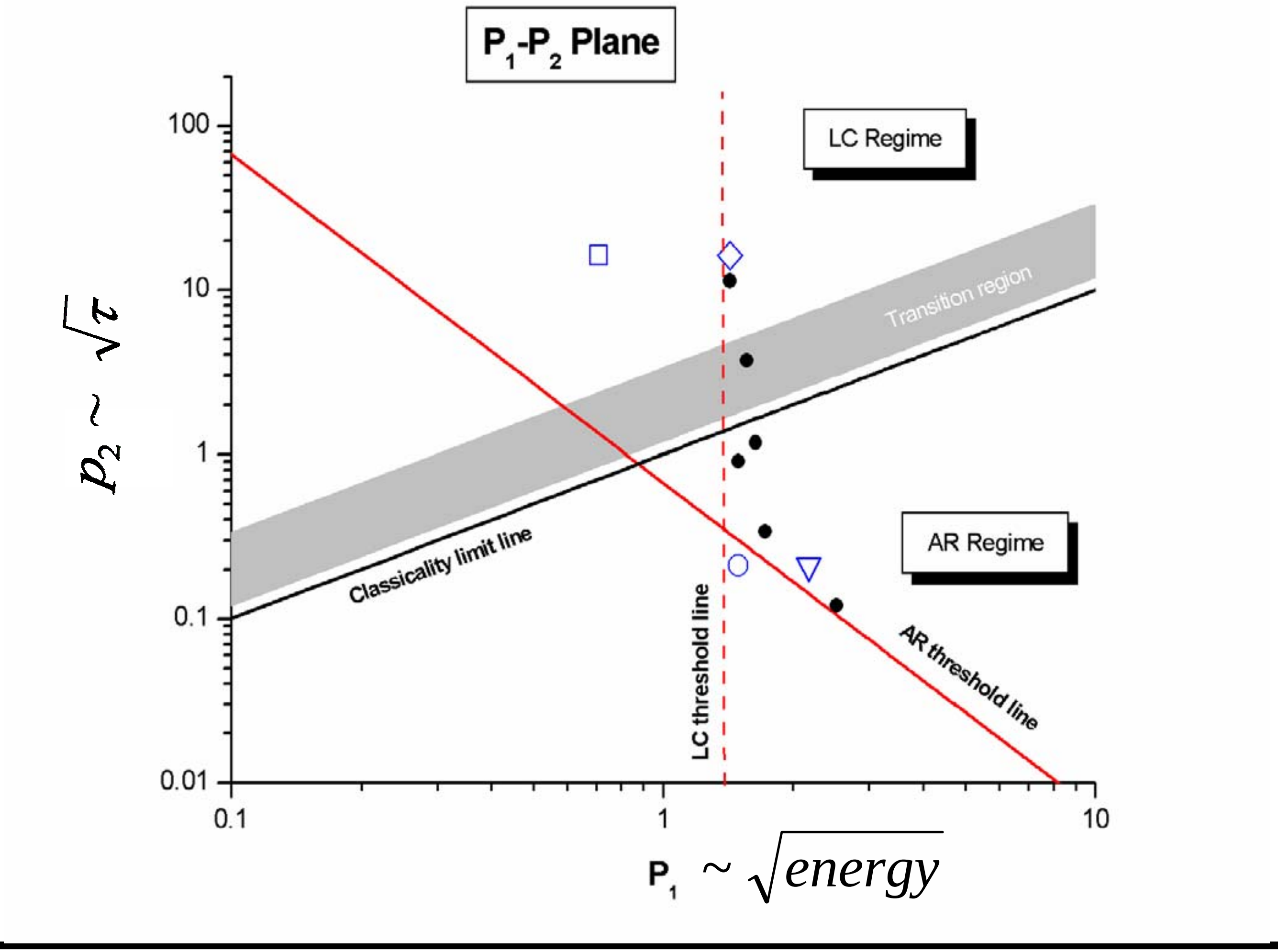


Autoresonance-above threshold



Design consideration for experiment

$$P_1 = T_S / T_R \qquad P_2 = T_{NL} / T_S$$



Experiment with HF molecule:

requirements from the radiation source

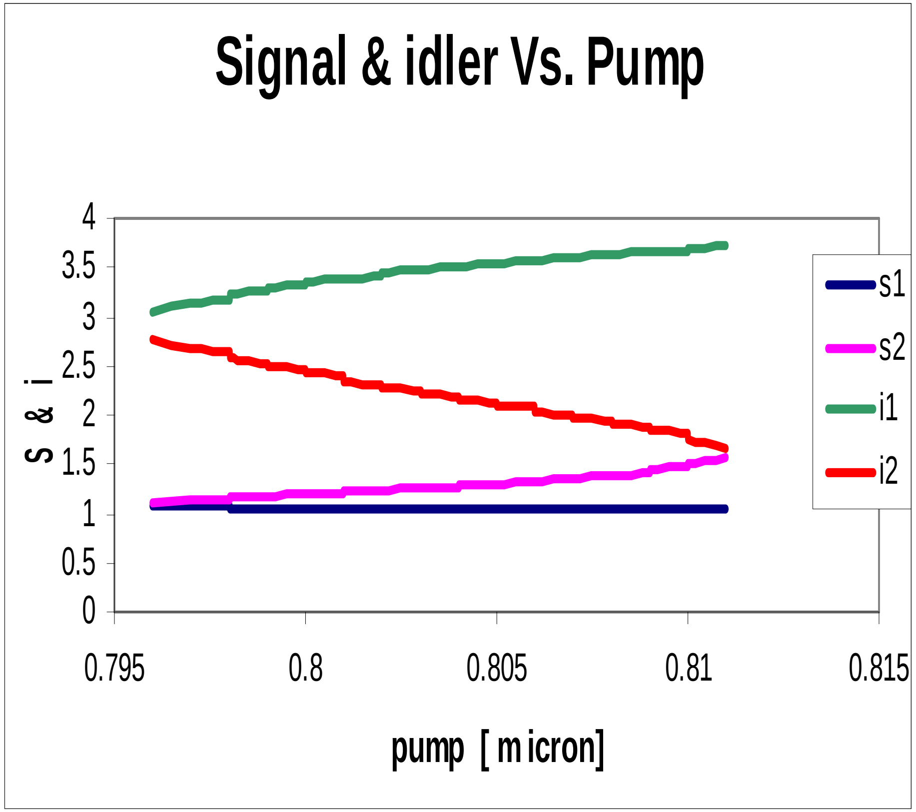
$\lambda_0 = 2.54\mu$ **In the IR regime**

$(\frac{\Delta\omega}{\omega_0}) \approx 16\ \%$ **To bring the population to the 4th level**

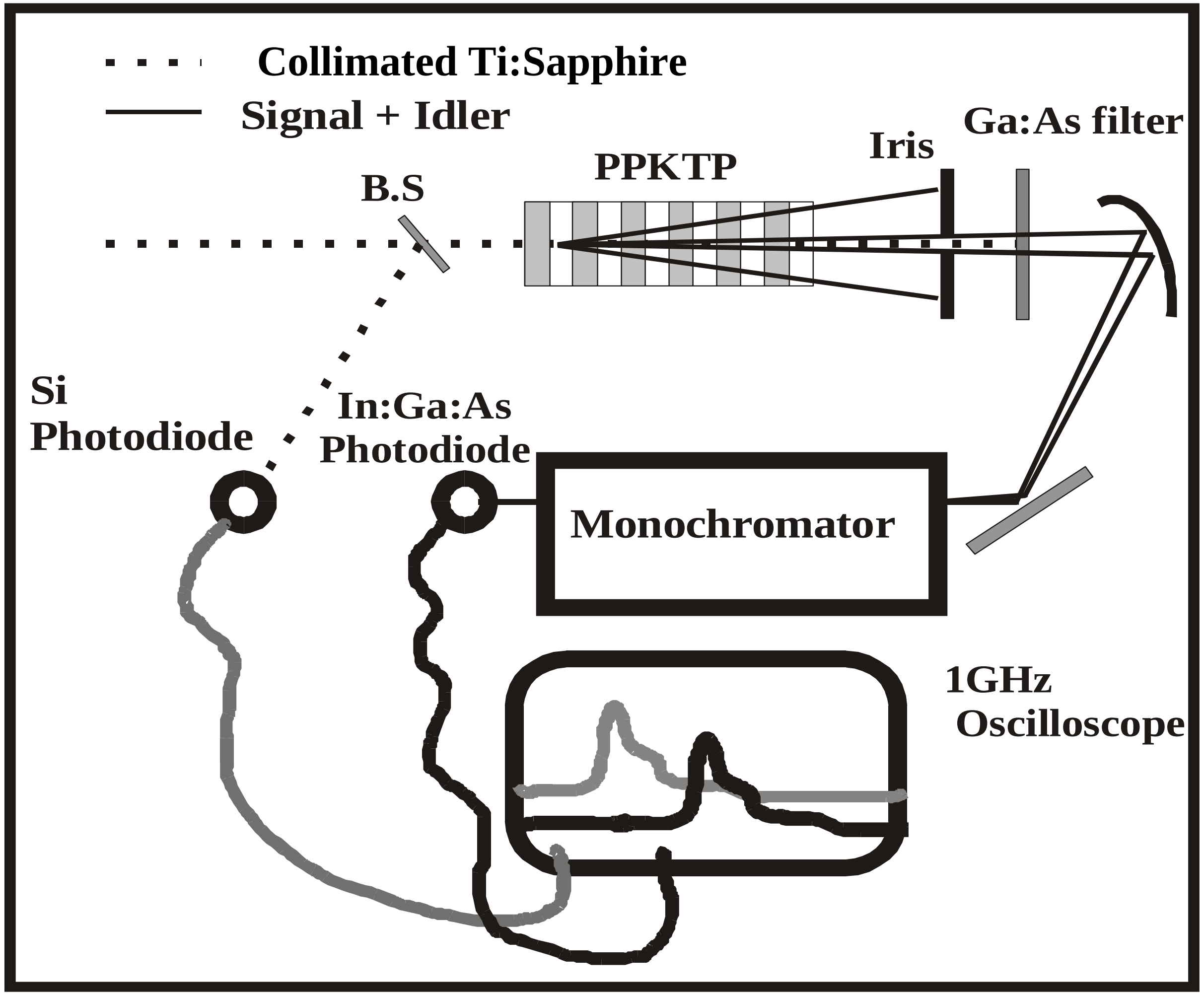
$\Phi_{th} \approx 700\ \frac{mJ}{cm^2}$ **Ladder climbing threshold**

Theoretical curve of phase matching for PPKTP with period of 27.1μ pumped by wide-band Ti:Sapphire Laser

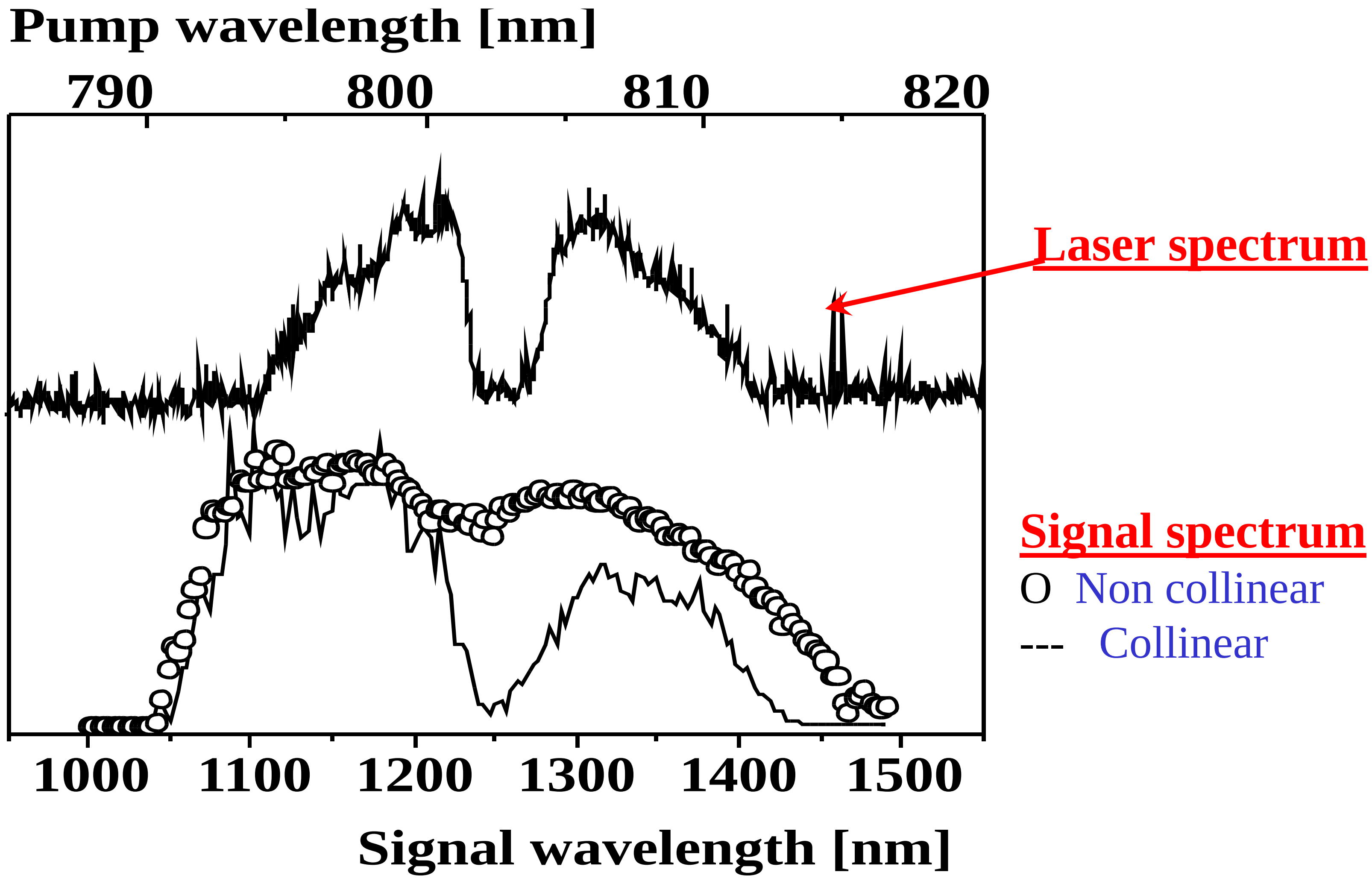
- Idler spectrum 2-3μ
- Signal spectrum 1-1.5μ



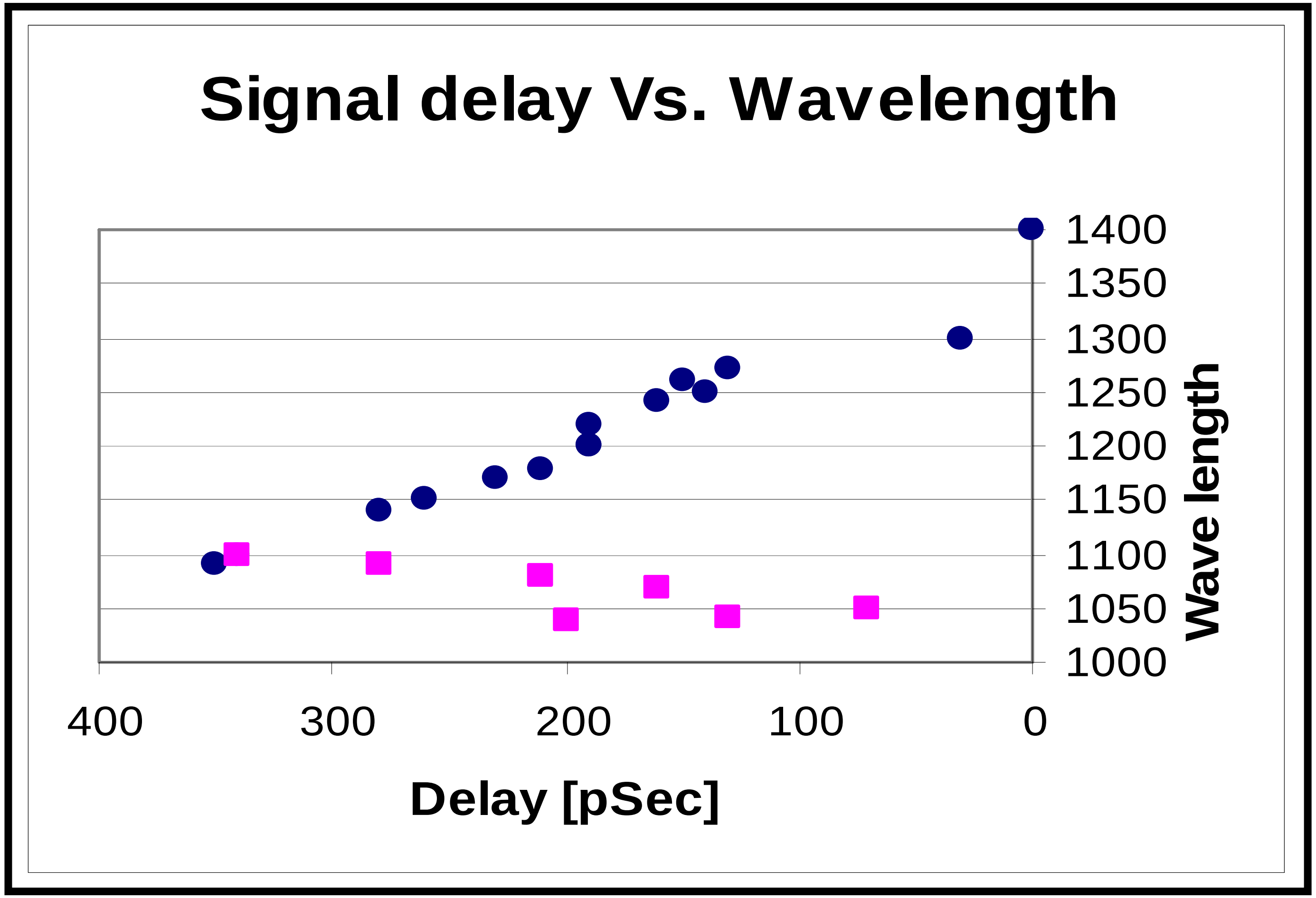
The Experiment



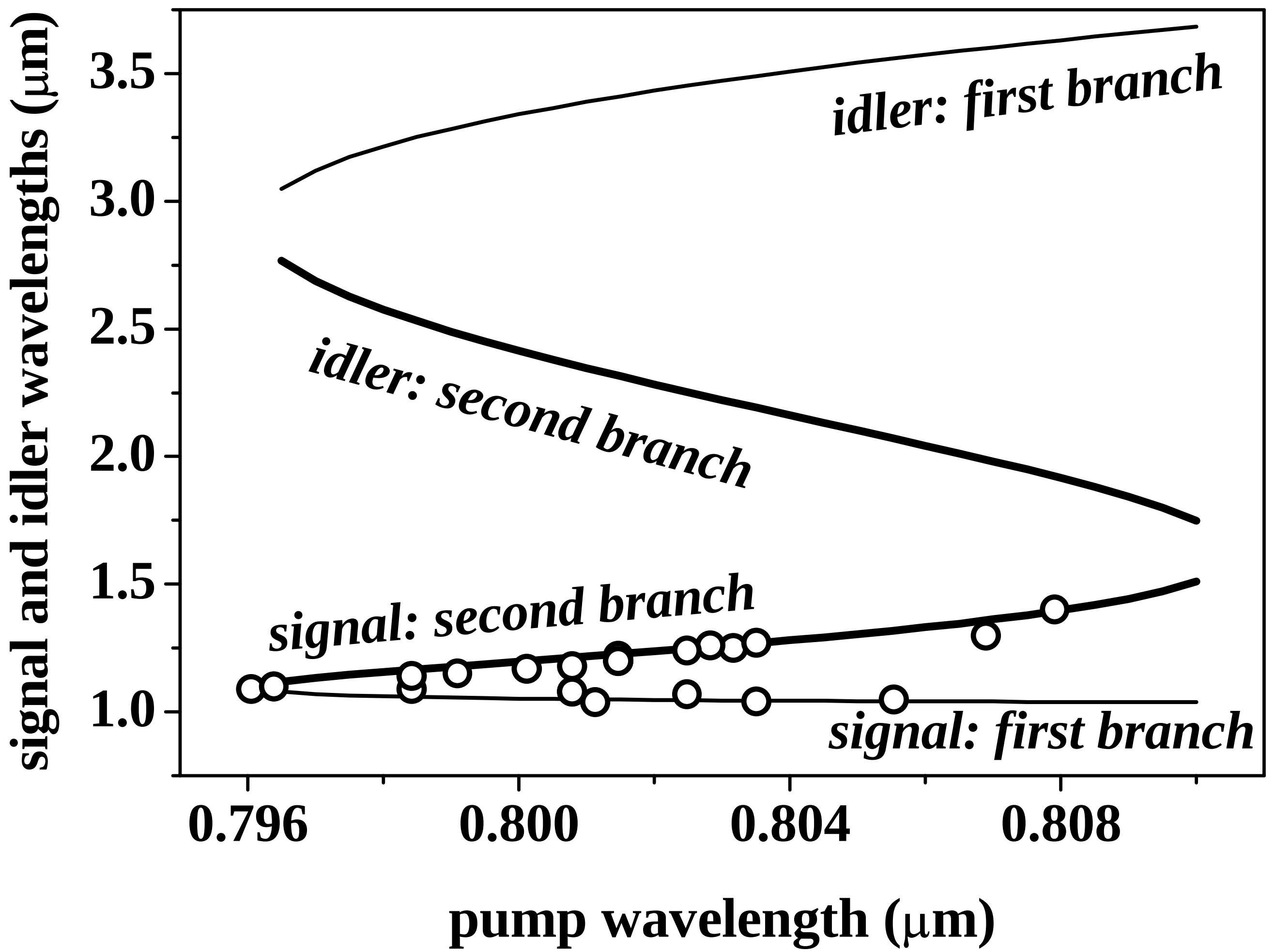
The Signal & Laser spectrum



Delay as a function of wave-length



Chirp measurement



IR specifications

- Bandwidth – 25%
- Pulse length – 185 psec
- Spot size 60µ x 700µ
- Energy – up to 200µJ

P_1-P_2 parameters

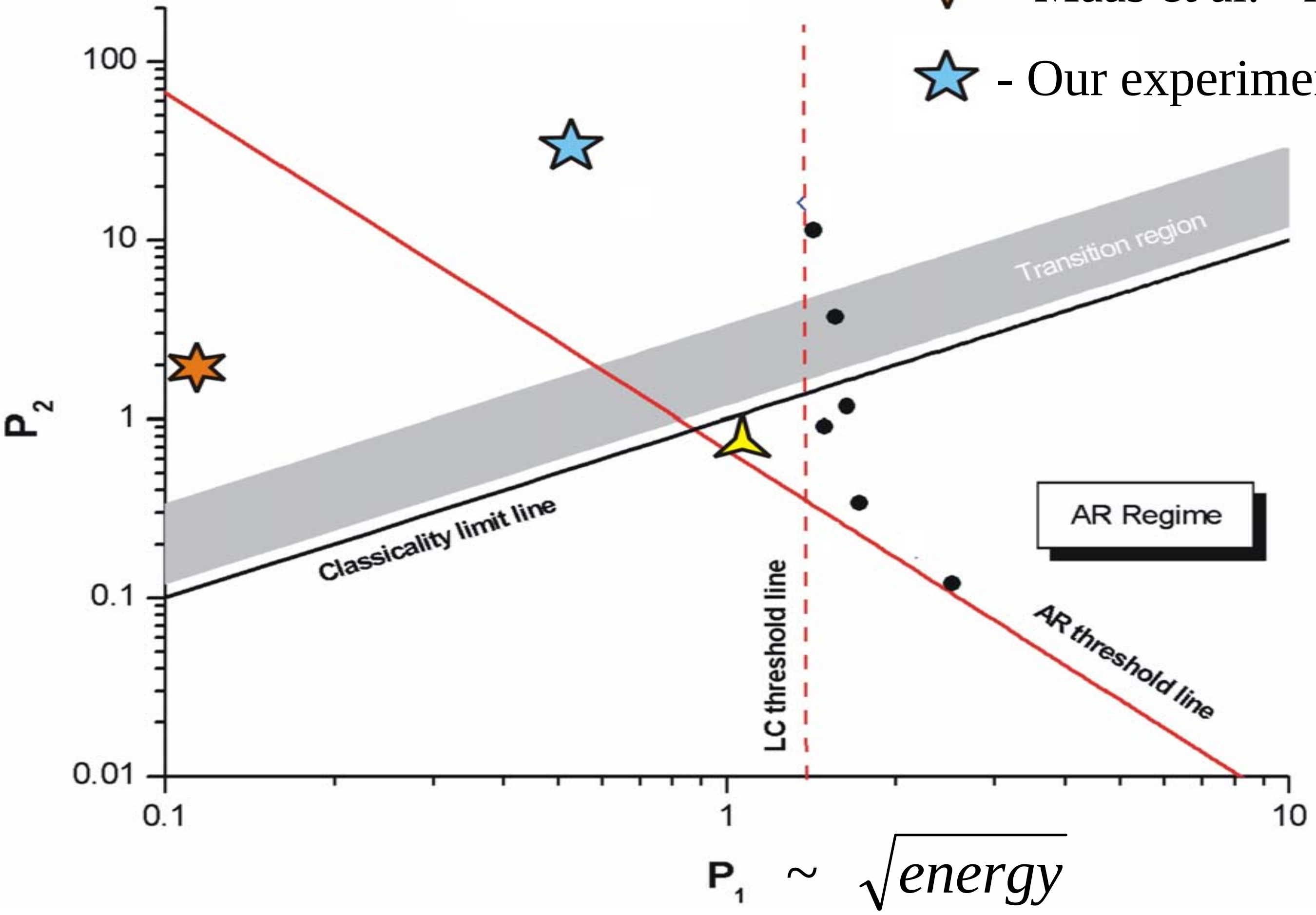
$$P_1 = T_S / T_R$$

$$P_2 = T_{NL} / T_S$$

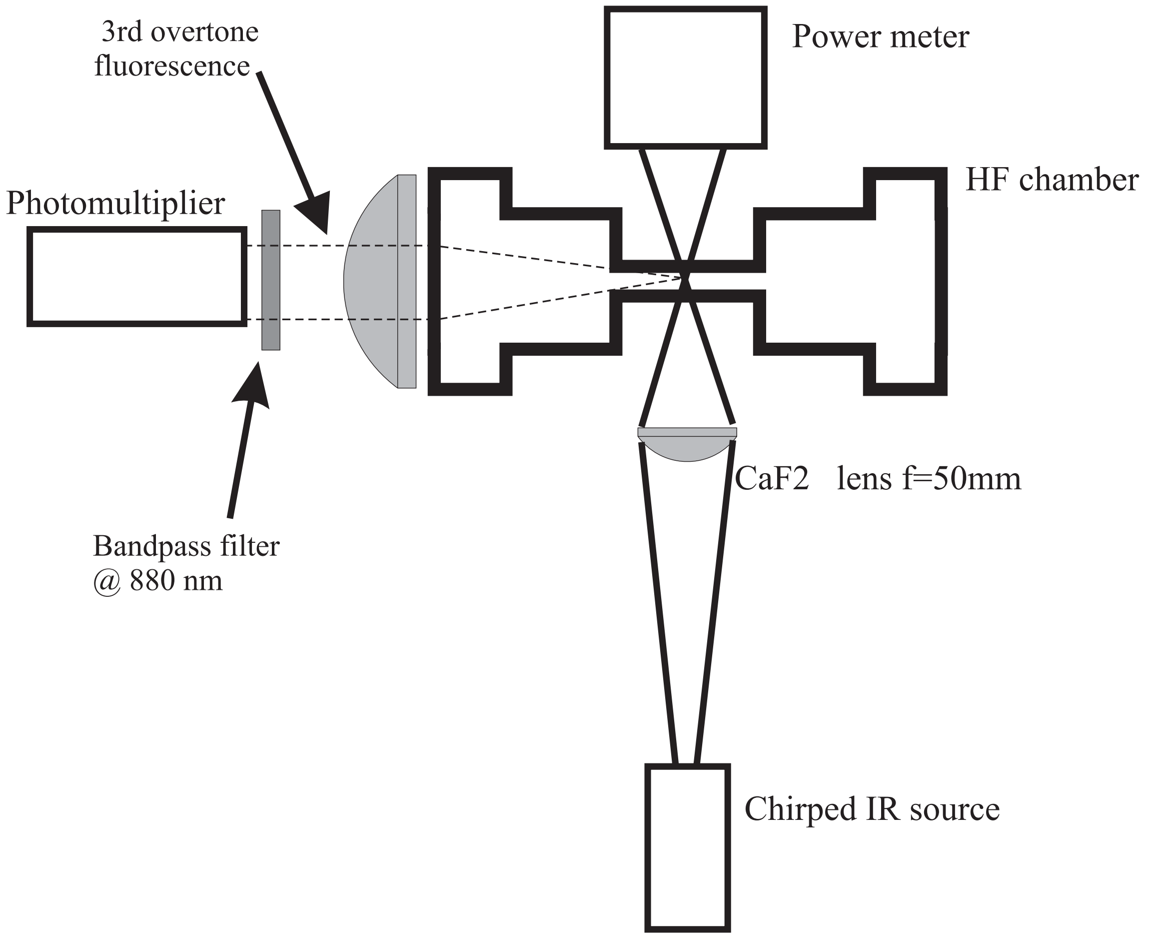
- Witte et al. – $Cr(CO)_6$

- Maas et al. - NO

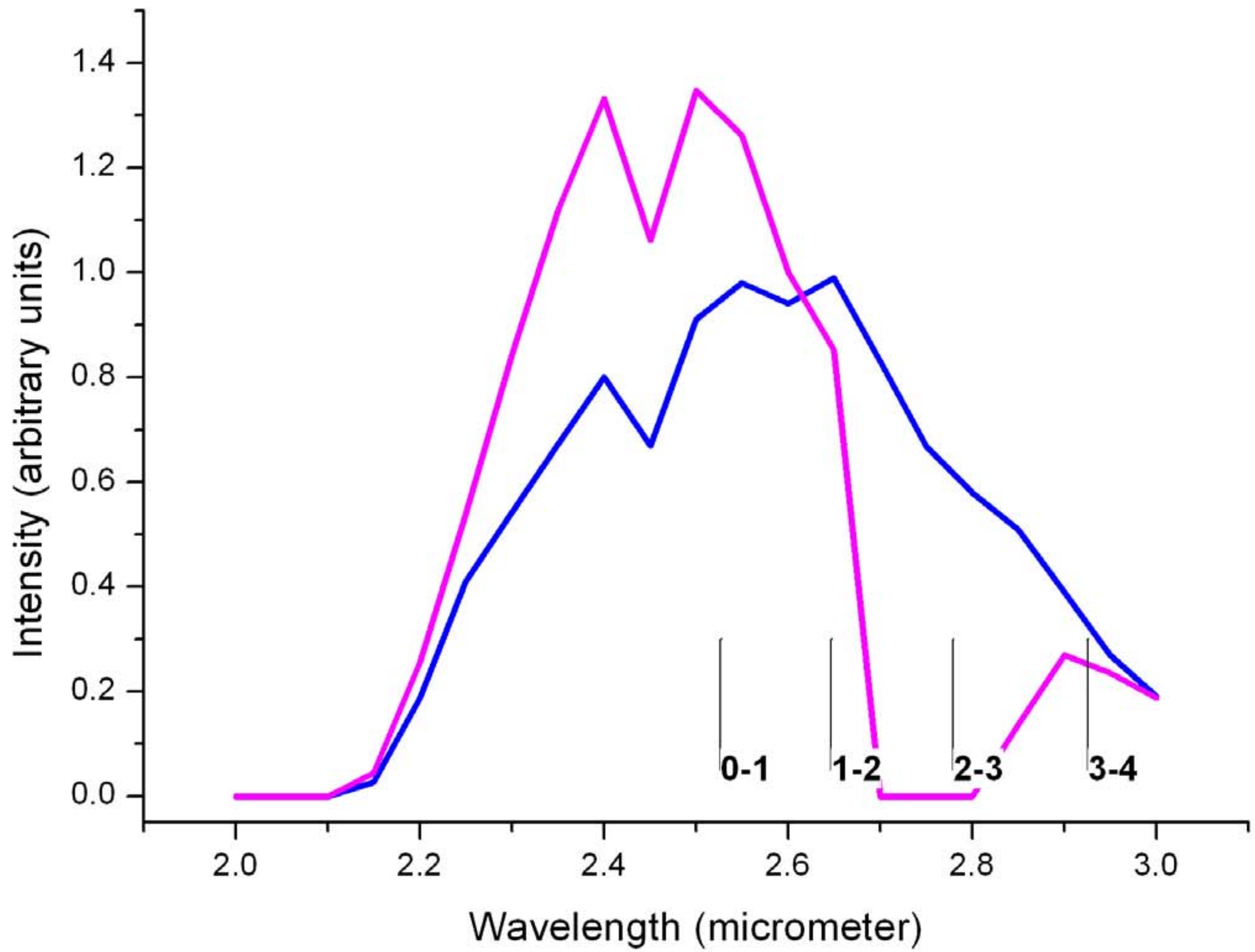
- Our experiment - HF



Demonstration of ladder climbing on HF molecule.



IR spectrum



Intensity (arbitrary units)
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0-1
1-2
2-3
3-4
2.0
2.2
2.4
2.6
2.8
3.0
Wavelength (micrometer)

HF experiment results

Conditions:	Avg. Number of photons	Standard deviation
10 torr HF	**11.16**	**4.6**
10 torr Air	**0.5**	**0.7**
Continually evacuated	**0.33**	**0.57**
10 torr HF & filtered spectra	**1.5**	**1.1**

Summary

- We have shown theoretically a smooth transition from ladder-climbing to autoresonance
- We have generated a chirped, ultra wideband radiation source in the IR
- We have demonstrated ladder-climbing on HF molecule

Plans for the future

- Improving the optics to allow us to be above the threshold
- Check the transition from quantum-mechanics to classicality.
- Other molecules

The end