- **Quantum control of laser – atom / molecule interaction processes and products**

This is a long standing activity of the group, dating back to the 1990 with **objective** the study and exploitation of coherent manipulation of atomic systems & processes. It includes

- Phase sensitive control
- Phase insensitive control

Over the years the activity has generated an number of **achievements** in several control schemes summarized bellow:

1) Demonstration of what is called **Laser Induced Continuum Structure (LICS).** [see some relevant earlier publications of the group] *Phys. Rev. Lett.* **67**, 3669 (1991); *Phys. Rev. Lett.* **70**, 3004 (1993); *Phys. Rev. Lett.* **74**, 2431 (1995)]
The basic scheme of the effect which is illustrated in Fig. 1 is the coupling of two bound states to each other and to the same continuum through two different electromagnetic fields. The resulting couplings scheme is in principle identical to the phenomenon of autoionization, thus resulting to asymmetric "Fano"-type line-shapes.
The scheme has been studied in terms of control application, namely the control of ionization branching ratios when the decay is in multiple continua [*Phys. Rev.* A **57**, 2915 (1998); *Phys. Rev.* A **66**, 013406 (2002)] and in molecular ionization [*J. Phys.* B **32**, 4485 (1999)]

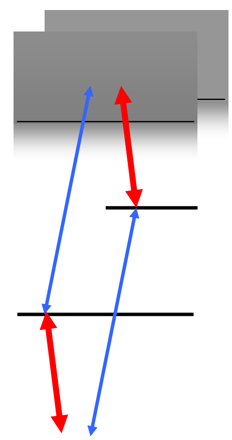

Fig. 1 Basic LICS scheme

2) Experiments and applications in **two color phase control.** Two color phase control occurs in schemes where excitation of the same bound or continuum state is through two channels of different non-linearity e.g. three photon vs single photon (third harmonic) excitation (see Fig. 2). Since the excitation probability is the result of the interference of the two channels, it depends on the relative excitation phase governed by the relative phase of the two fields. Thus externally varying the relative phase of the fields excitation probabilities can be manipulated in a controlled way.

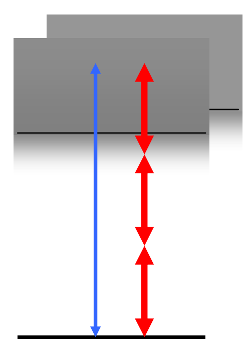

Fig. 2 Two color phase control

The experiments performed at IESL include

- Phase control in four-photon schemes. [*J. Phys.* B **29**, 3599 (1996)]
- Phase control of the autoionization rate.
- Phase control of third harmonic generation. [*Opt. Commun.* **152**, 83 (1998)]
- New convenient technique for phase sensitive studies and applications.

3) Coherent control through feedback - optimized tailored fs pulses. A process is optimized through appropriate tailoring of broad band radiation of fs laser pulses. Pulse-shape tailoring is based on the spectral phase modulation of different spectral components. Phase modulation occurs using a liquid crystal array as spatial light modulator through which the by an optical grating spatially dispersed radiation (4f optical arrangement) is transmitted. The afterwards through a second grating recombined spectral components form a pulse with the desired temporal shape.

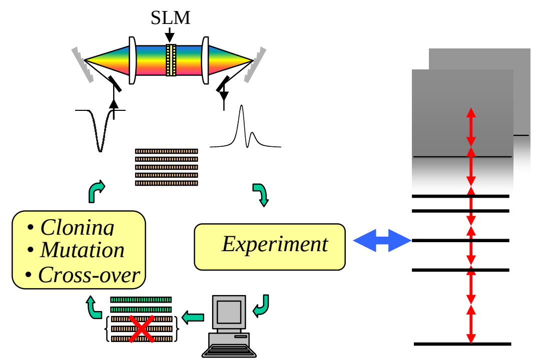

Fig. 3 Feedback - optimized tailored fs pulses.

Optimal shaping occurs through feed-back from the experiment and optimization through an evolutionary optimization algorithm. Evolution successively improves the pulse shape towards the aimed result. The set up is shown in Fig. 3

An application of the method, in the control of branching to different molecular ionic vibrations during ionization, has been recently demonstrated at FORTH-IESL [*J. Chem. Phys.*, **118**, 595 (2003)]. The experiment was in molecular NO and is shown in Fig 5. The ionic vibrational spectrum resulting from a not shaped Gaussian pulse consist of practically only the v_+=2 peak is shown in (a). Optimization of the v_+= 0 and v_+= 1 or the v_+=4 and v_+=5 vibrations has been achieved through pulse shaping. The results and the second order autocorrelation traces of the shaped pulses are shown in (b) and (c) respectively.

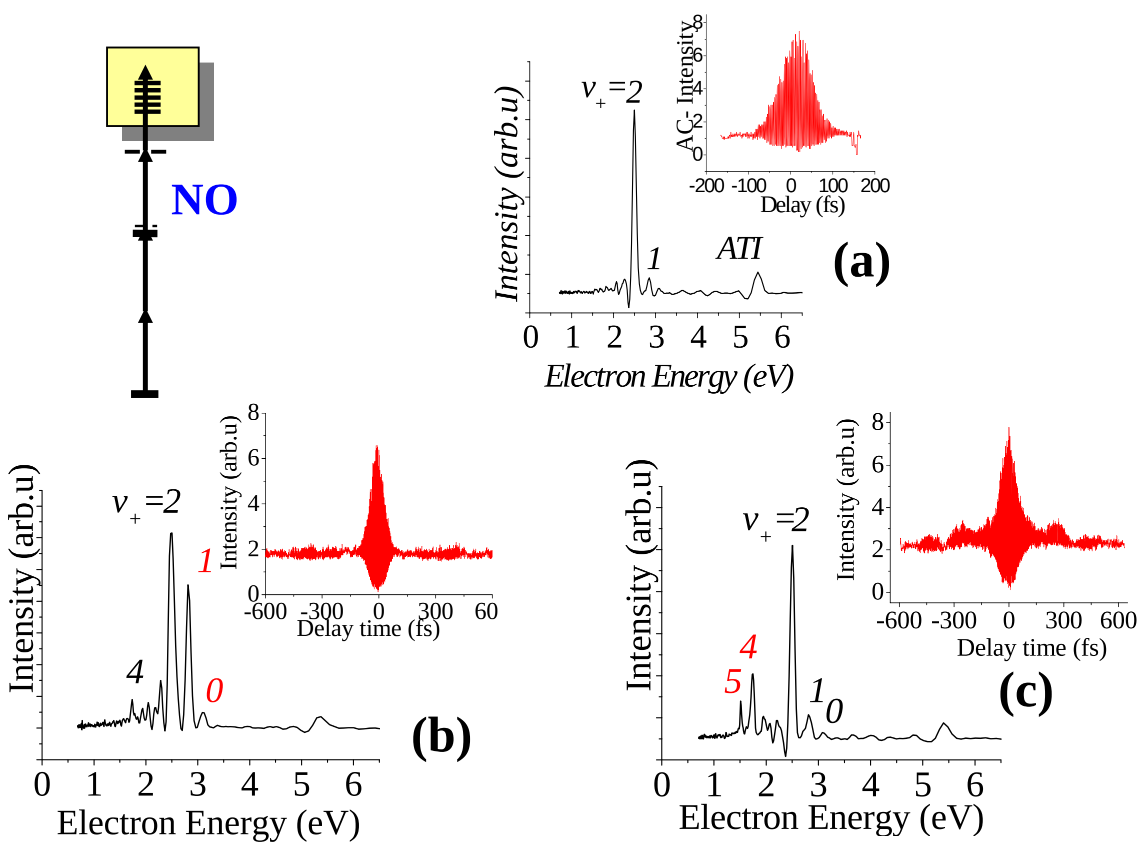

Fig. 4 Coherent control of molecular ionization branching ratios.