Wave packet engineering using a phase-programmable femtosecond optical source

I. Matsuda, K. Misawa, and R. Lang
Tokyo University of Agriculture and Technology
kmisawa@cc.tuat.ac.jp

A phase-programmable femtosecond optical source is indispensable for interactive control of photonic and electronic wave packets. In the present paper, we describe wave-packet shaping in the excited state of organic dye molecules. The phase-programmable femtosecond optical source is composed of a femtosecond pulse oscillator, phase modulator and phase analyzer as shown in Fig. 1 [1]. A femtosecond pulse with a spectral band width as broad as 100 nm is converted onto the Fourier plane. After a phase shift is provided to each spectral component by a spatial light modulator on the Fourier plane, the pulse is reconstructed. The output from the phase modulator is characterized by frequency resolved optical gating (FROG). The temporal profile and phase information of the femtosecond pulses can be obtained from the FROG measurement. A desired phase pattern can be realized through the iterative adjustment of the phase mask by analyzing the phase information. Figure 2 show the spectrum and phase dispersion for positively-chirped ($\Phi''(\omega)$=500 fs^2), transform-limited (0 fs^2) and negatively-chirped pulses (–500 fs^2).

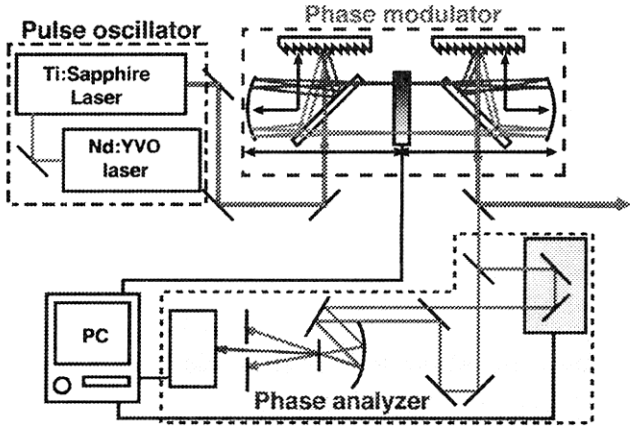

Fig.1: Schematic diagram of the phase-programmable femtosecond optical source.

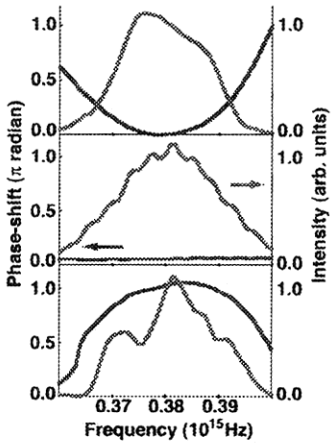

Fig.2: Spectrum and phase dispersion of positively-chirped (top), transform-limited (middle) and negatively-chirped pulses (bottop).

Wave-packet shaping by means of the phase-programmed femtosecond pulses is observed in a cyanine dye molecule (IR-140). Figure 3 shows the difference luminescence spectra of the positively-chirped (PC) and negatively-chirped (NC) excitations from the transform-limited excitation. The luminescence intensity is increased and decreased in case of PC and NC excitations, respectively. This chirp-dependent luminescence can be explained in terms of intra-pulse pump-dump process [2]. NC pulse induces narrow spatial distribution of the excited wave packet, while it is easily broadened and quickly escapes from the Franck-Condon window in PC case. The overlap integral between the excited- and ground-state wave packets determines the remaining excited state population, and as a result, the luminescence intensity.

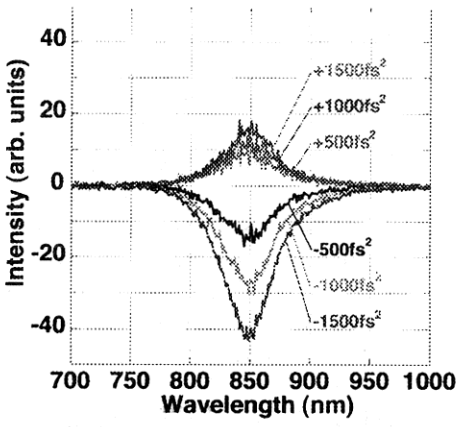

Fig.3: Difference luminescence spectra from IR-140 dye by the positively and negatively-chirped excitations with respect to the transform limited pulse.

[1] Isao Matsuda, Kazuhiko Misawa, and Roy Lang, Abstracts of International Workshop on FST 2001, p.186 (2001).
[2] K. Misawa and T. Kobayashi, J. Chem. Phys. 113(17), 7546-7553 (2000).