III-L Wave Packet Engineering Using a Phase-Programmable Femtosecond Optical Source

We proposed "wave packet engineering" which realizes mutual conversion between phase information of photonic and quantum wave packets by means of light-matter interaction. A phase-programmable femtosecond optical source is indispensable for such interactive control of photonic and quantum wave packets. We demonstrate control of quantum wave packets in organic molecules and semiconductors using phase-programmed pulses.

III-L-1 Single Molecular Phase-to-Amplitude Converter

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Control of quantum wave packets has been recently studied in various systems such as atoms, molecules, and semiconductors using chirp-controlled and phase-locked double pulses.[1)] In the present study, we observed wave-packet shaping by means of the phase-programmed femtosecond pulses in a cyanine dye molecule. The intra-pulse phase pattern of the pulses was converted to the amplitude of luminescence from the cyanine molecules.

The phase-programmable femtosecond optical source is composed of a femtosecond pulse oscillator, phase modulator and phase analyzer as shown in Figure 1. A femtosecond pulse with a spectral band width as broad as 160 nm is converted onto the Fourier plane. After a phase shift is provided to each spectral component with 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. The center wavelength, pulse energy, duration, and phase-shift division of the source output were 802 nm, 0.64 nJ, 14 fs, and $6\pi/700$ radian, respectively. Figure 2 shows the spectra and phase dispersions for positively-chirped (Φ" = 500 fs^2), transform-limited (0 fs^2) and negatively-chirped pulses (–500 fs^2).

Ethanol solution of a cyanine dye (IR-140) at a concentration of 4×10^{-4} M is circulated in a 0.5-mm thick quartz cell. The luminescence spectra of spontaneous emission are measured to evaluate the remaining excited-state population. 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 population, and as a result, the luminescence intensity.

In conclusion, we observed the remarkable dependence of luminescence intensity and the excited-state population in IR-140 molecules on the chirped pulse from the phase-programmable femtosecond optical source. The observed shaping of quantum wave packet opens a new possibility to process the intra-pulse phase information.

References

1) *Quantum Control of Molecular Reaction Dynamics*, R. J. Gordon and Y. Fujimura,Eds. (World Scientific, 2001).
2) K.Misawa and T.Kobayashi, *J.Chem.Phys.* **113**, 7546 (2000).

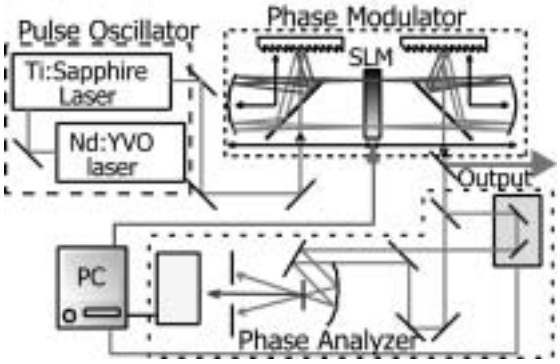

Figure 1. Schematic diagram of the phase-programmable femtosecond optical source.

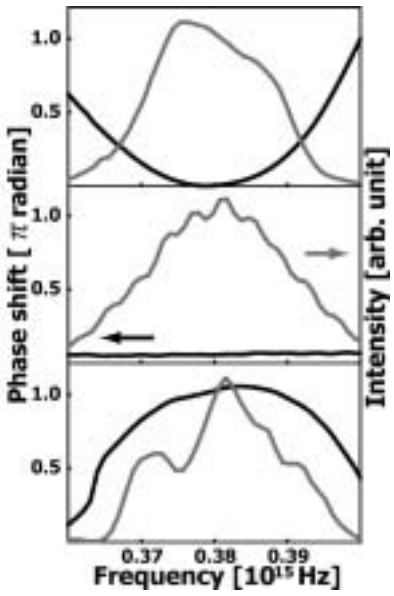

Figure 2. Spectra and phase dispersions of positively-chirped (top), transform-limited (middle) and negatively-chirped pulses (bottom).

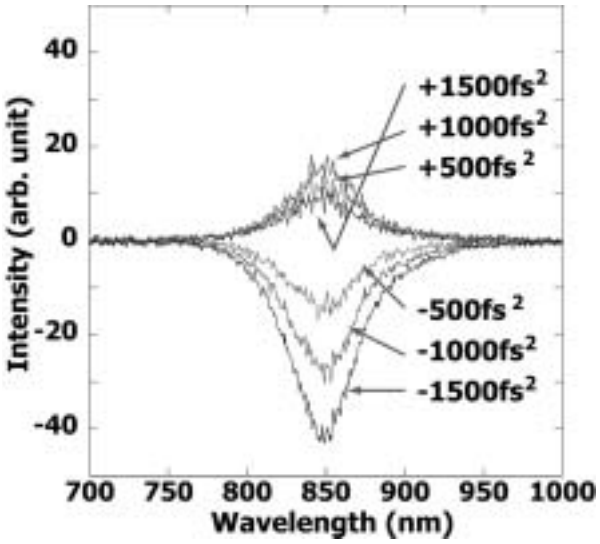

Figure 3. Difference luminescence spectra from IR-140 dye by the positively and negatively-chirped excitations with respect to the transform limited pulse.