Subnanosecond Transient Absorption Measurement by Using a Randomly-Interleaved-Pulse-Train (RIPT) Method

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Although transient absorption (TA) method has been powerful tool to study primary processes in various fields of photochemistry, there remains difficulty to measure in the time region from subnanosecond to several tens of nanosecond. To overcome it, we have recently proposed a novel technique, RIPT (Randomly-Interleaved-Pulse-Train) method and constructed a prototype of TA spectrometer (Fig. 1) based on the RIPT method[1]. For each pump pulse, many monochromatized probe pulses impinge upon the sample, and the corresponding pump-probe time delay is passively determined shot by shot with an accuracy of several tens of picoseconds. By repeated pumping with automatically varying time delay, a TA temporal profile that covers a wide dynamic range from subnanosecond to milliseconds is simultaneously obtained. By scanning wavelength, this simple and single apparatus acquires not only wide time range TA profiles, but also broadband TA spectra from visible to near-infrared regions. Furthermore, the RIPT method has the capability of eliminating fluorescence signals, which often pollute TA curves. We confirmed that the RIPT method has excellent time resolution of less than 100 ps by using a picosecond laser for the pump light and a picosecond supercontinuum light source for the probe light.

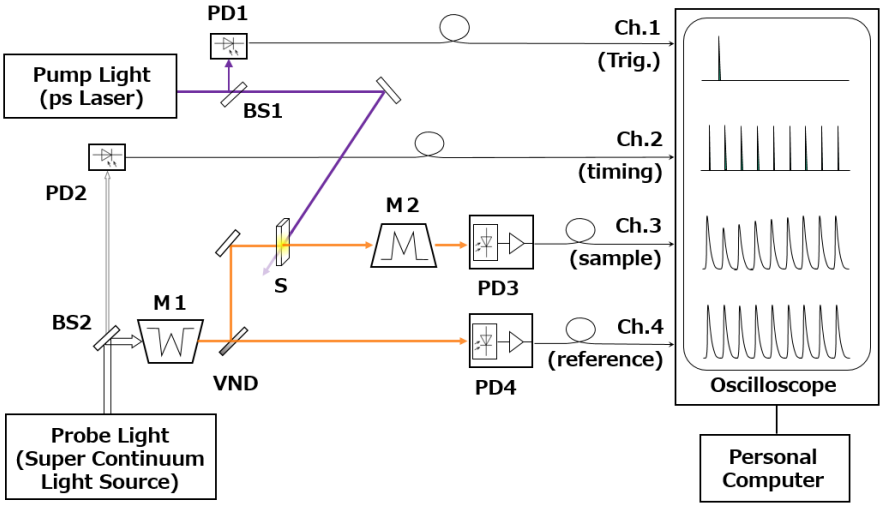

Figure 1. System layout of transient absorption spectrometer based on the RIPT method.

We have been applying the RIPT method to various samples, not only weakly luminescent compounds such as fullerene, benzophenone, titanium dioxide, and materials for artificial photosynthesis[2], but also highly luminescent compounds like laser dyes, quantum dots, and metal-complexes, etc. (Fig. 2-6). Most of the results provide us with what has hardly been obtained from conventional methods, namely, ultrafast pump-probe method or nanosecond flash photolysis, since the RIPT method bridges the gap time region (1 ns ~ 20 ns) in-between the time ranges covered by the conventional methods, and is also applicable to wide wavelength range from visible to near-infrared regions. Furthermore, the TA measurement free from luminescence signals is a unique feature of the RIPT method. Recently, the highly sensitive (< 1 mOD) TA measurements has successfully been made possible by the RIPT method. Pump intensity dependence of transient absorption signal for CdSeS/ZnS quantum dot in THF.

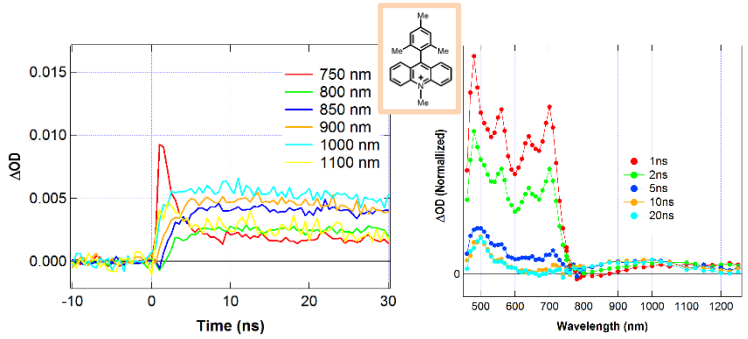

Figure 2. Observation of photoinduced electron-transfer dynamics in a donor-acceptor linked dyad, Acr^{+}-Mes (9-mesityl-10-methylacridinium ion dyad)

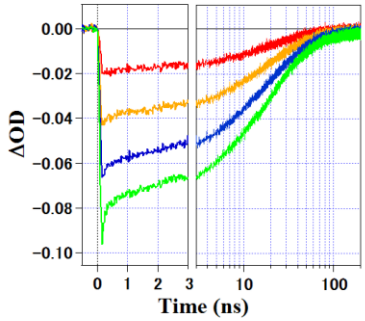

Figure 3. Pump intensity dependence of transient absorption signal for CdSeS/ZnS quantum dot in THF.

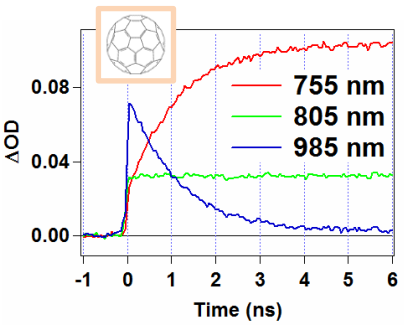

Figure 4. Transient absorption temporal profile of fullerene in toluene at 755, 805, and 985 nm.

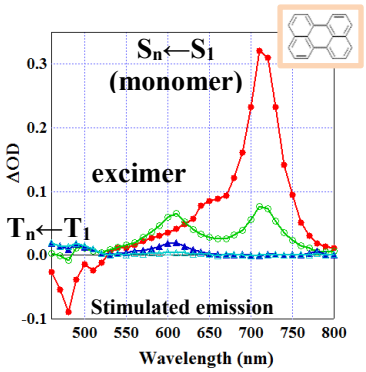

Figure 5. Transient absorption spectra of perylene in toluene at 0, 5, 25, 50 ns after the excitation.

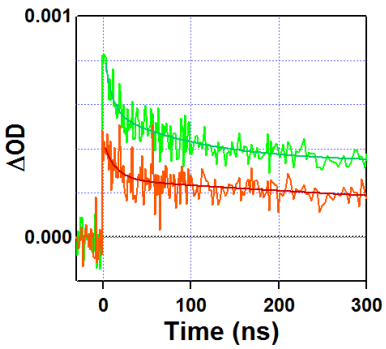

Figure 6. High sensitive transient absorption measurement of titanium dioxide when varying pumping energy. λ = 900 nm.

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References:

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