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Real-Time Time-Frequency Imaging of Ultrashort Laser Pulses Using an Echelon Mirror

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We demonstrate real-time time-frequency imaging for the autocorrelation traces of ultrashort laser pulses using an echelon mirror fabricated on a Ni block with 500 steps; the echelon mirror is employed to generate spatially encoded time delays for the probe pulses. By using the frequency-resolved optical gating (FROG) technique with the echelon mirror, the time-frequency images of ultrashort laser pulses were successfully mapped in real-time. The chirp characteristics of the laser pulses were also evaluated with the phase-retrieval procedure on a single-shot basis. Our technique provides significant advantages over conventional autocorrelation and FROG techniques, such as single-shot detection of time-frequency images, a small spot size at a nonlinear crystal, chirp-free characteristics of echelon mirrors, and ultrafast measurement capabilities by simply replacing the nonlinear crystal with samples. Hence, we believe that it becomes a powerful spectroscopic tool for monitoring ultrashort laser pulses and for investigating ultrafast dynamics of materials.

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1. Introduction

Since the invention of ultrashort lasers, various types of spectroscopic techniques have been developed for revealing ultrafast transient phenomena. [1)] Furthermore, by combining these spectroscopic techniques with either the pulse-shaping technique [2–5)] or chirped laser pulses [6–8)] coherent controls of photochemical reactions and of molecular and solid-state dynamics have been extensively studied. However, most of these studies are limited to reversible transient processes because conventional ultrafast spectroscopy requires many repetitions of pump-probe sequences to encompass the temporal and spectral regions of interest. Therefore, ultrafast measurements and coherent controls of irreversible transient processes (especially in solids) are still open issues. In order to measure the irreversible transient signals, many techniques based on single-shot detections have been proposed. [9–18)] These techniques have successfully overcome several practical problems such as the photodegradation of samples after many repetitions of pump-probe sequences, long measurement times, and reductions in the long-term stability for laser systems. In most of these techniques, the probe pulses are manipulated to have a spatially encoded time delay, and as a result, they have the capability of conducting single-shot measurements without scanning the time delay. However, experimental difficulties still exist; for example, in the dual-echelon technique, [11,12)] the transmitted pulse passing through the echelons causes a spatially dependent chirp that may distort the temporal profile. Further, in pump-probe imaging spectroscopy, [13–15)] a large sample is required to obtain a sufficiently long time delay for the probe beam, and the intersecting angle between the pump and probe beams affects the time resolution of the system. Therefore, these techniques might not be applicable to broadband spectroscopy on small samples with extremely ultrashort laser pulses or with white-light continuum.

To perform ultrafast measurements as well as achieve coherent control of irreversible transient processes, the development of simple, quick, and accurate single-shot methods to characterize the laser pulses themselves is also indispensable. Ideally, this characterization should be carried out using the same optical setup and pathway as those used for the actual ultrafast measurements. For the characterization of ultrashort laser pulses, spectral phase interferometry for direct electric field reconstruction (SPIDER) [19,20] and frequency-resolved optical gating (FROG) [21,22] are the two most popular techniques. A highly simplified device for ultrashort laser diagnostics (grating-eliminated no-nonsense observations of ultrafast incident laser light e-fields; GRENOUILLE) has also been proposed as a single-shot FROG technique. [23] However, although these methods are useful for the characterization of laser pulses, they might be less suitable for longer pulses such as expanded chirped or shaped pulses. Moreover, these techniques cannot be easily applied to the measurement of the ultrafast dynamics of materials, and as a result, laser pulses are typically monitored using optical arrangements different from those used for the actual sample measurements.

In this paper, we propose a new technique for the time-frequency two-dimensional (2D) imaging of ultrashort laser pulses implemented on a single-shot basis. The key to our technique is the use of an echelon mirror with a microstep structure to provide temporal delays for the probe laser pulses. By using the echelon mirror, we will demonstrate a simple and fast FROG apparatus to characterize ultrashort laser pulses using the phase-retrieval procedure. Our new technique has great technical advantages over the conventional autocorrelation and FROG techniques: time-frequency 2D imaging based on single-shot detections with wide temporal and spectral ranges, a small spot size at a nonlinear crystal, chirp-free characteristics of the echelon mirror, and capability for direct measurements of

molecular and solid-state dynamics by simply replacing the nonlinear crystal with samples. In addition, our new technique does not require any mechanical scanning delay unit to produce time delays between pump and probe beams, and therefore, small and compact experimental setups for the ultrafast measurements can be easily achieved. This capability is a significant advantage for limited experimental conditions such as those found inside ultrahigh vacuum chambers [24,25]. Hence, we believe that our new technique is a powerful spectroscopic tool not only for monitoring ultrashort laser pulses but also for investigating the ultrafast dynamics of materials.

2. Experimental Methods

Figure 1(a) shows the concept of our technique for obtaining time-frequency 2D autocorrelation traces (or FROG traces) of ultrashort laser pulses. Images of the echelon mirror obtained by using a conventional laser microscope are also shown. [26] The echelon mirror is fabricated on a Ni block ($10 \times 10 \times 10\ \mathrm{mm}^3$) with 500 steps using a precise micro-machining technique, and it is employed to generate a spatially encoded time delay for the probe pulses. Each step has a step width d' of 20 μm and a step height h of 5 μm; further, the surface roughness, <0.2 μm, is sufficient for our purposes. This echelon mirror can generate temporal steps of ~34 fs and an overall time delay of ~17 ps.

The echelon mirror plays a role of not only a spatially encoded time delay optics but also of a *reflective grating* with a periodicity of $d = (d'^2 + h^2)^{1/2}$. The incident and diffraction angles of the probe pulses against the normal of the echelon surface are denoted as α and β, respectively; the diffraction condition can then be written as

$$d\left(\sin\alpha + \sin\beta\right) = m\lambda \quad (m = 0, \pm1, \pm2, \pm3, \ldots), \tag{1}$$

where λ is the wavelength of the probe pulse and m is the diffraction order. Since the probe pulse has a finite spectral width $\Delta\lambda$ due to the ultrashort pulse duration, the diffracted light from each stair is spectrally dispersed with increasing distance from the echelon, as shown in Fig. 1(a). In order to suppress the spectral dispersion at the entrance slit of a spectrometer, we placed focusing and imaging lenses (f_1 and f_2) between the echelon and spectrometer, and subsequently, we obtained a clear image of the second harmonic (SH) autocorrelation signal generated at a nonlinear crystal (beta-BaB_2O_4; BBO) at the slit. Thus, in this imaging configuration, the echelon mirror does not act as a spectral-resolving optics but instead as a spatial encoding optics for the time delay.

The spot size Δx at the nonlinear crystal as a result of the spectral dispersion can be written as

$$\Delta x \approx \frac{mf_1}{d\cos\beta}\Delta\lambda, \tag{2}$$

where f_1 is the focal length of the focusing lens. Here, the diffraction limit of the spot size is neglected because Δx is considerably larger than the diffraction limit. To obtain small spot sizes at the nonlinear crystal and as clear images at the entrance slit of the spectrometer as possible, the entrance slit of the spectrometer and echelon surface should be aligned facing each other (i.e., $\beta \approx 0^\circ$). When the 12th order of the diffraction was used (m = 12), we could approximately satisfy this condition. Under our experimental conditions, (d = 20.6 μm, α = 28°, β = 0°, m = 12, f_1 = 100 mm, and $\Delta\lambda$ = 12 nm for pulse durations of ~150 fs), the spot size Δx is calculated to be 0.79 mm; this value is also confirmed by direct observations of the spot. Since the echelon mirror also works as a reflective grating, the probe beam diffracted by the echelon mirror is spectrally dispersed. Subsequently, the wavelength-resolved components are aligned along the spot size Δx at the nonlinear crystal. Therefore, a pump beam having

homogeneous intensity, whose spot size (typically 1 mm) is larger than Δx=0.79 mm, was focused on the nonlinear crystal to cover the entire probe beam area. Hence, undesirable effects of the spectral dispersion at the nonlinear crystal on the generated SH signals are suppressed, thereby leading to accurate pulse measurements.

The time resolution of the system depends on the geometrical configuration, that is, the intersecting angle θ between the gate and probe pulses at the nonlinear crystal. [10,13–15] Therefore, the gate pulse is focused onto the nonlinear crystal from the top against the diffracted probe pulse by making it pass through the center of the echelon; in this manner, reductions in the time resolution are avoided. The intersecting angle for the probe pulse that is diffracted around the center of the echelon is nearly 0, and hence, the time resolution around the center is as short as the pulse duration. On the other hand, the intersecting angle for the probe pulse that is diffracted from the edge of the echelon is given by $\theta \approx D/2f_1$, and here, the time resolution decreases by $\sim\Delta x\theta/c$, where c is the speed of light and D = 10 mm, i.e., the echelon size. Under our experimental conditions, the maximum value of $\sim\Delta x\theta/c$ was estimated to be ~130 fs. When we measure the longer laser pulses or the dynamics on a several-picosecond time scale, we need to use the overall length of the echelon; the reduction in the time resolution (~130 fs) is considerably smaller than the overall time delay of the echelon (~17 ps). On the other hand, when we measure the shorter laser pulses or the ultrafast dynamics (typically, <1 ps), we only need to use the center part of the echelon, resulting in no reductions in the time resolution coming from the geometrical configuration. Therefore, in both cases, the reductions in the time resolution due to the geometrical configuration are not serious for monitoring the ultrashort laser pulses from femtosecond laser sources with pulse durations longer than ~100 fs. In fact, the time resolution of our FROG apparatus is somewhat

limited by the pulse duration of the laser pulse itself and/or the echelon steps (~34 fs).

For measuring various types of transient signals with different temporal and spectral ranges, suitable echelon mirrors with different step widths, heights, and step numbers can be selected. Several echelon mirrors with different temporal and spectral ranges have been already fabricated and diagnostically tested to confirm their performances. For example, an echelon mirror with 80 μm step width, 10 μm step height (~67 fs) and 500 step numbers (~33 ps) has the ability to measure transient signals longer than 10 ps with a wide spectral range of ~200 nm. The results will be reported elsewhere.

As an initial demonstration, we measured the time-frequency 2D autocorrelation traces of nearly Fourier transform-limited (TL) ultrashort laser pulses with different pulse durations. Figure 1(b) shows the experimental setup. As the light source, we used a commercial femtosecond Ti:sapphire laser with repetition rate of 76 MHz, center wavelength of 800 nm, and pulse duration of 150 fs. The pulse duration of the laser pulses could be varied by narrowing the spectral width of the laser pulses using a pair of grating and variable slit and then tuned from 150 fs to 6 ps. The laser pulses were divided into probe and gate beams by using a beam splitter (BS). Further, the probe beam was expanded to a diameter of ~2 cm by a combination of negative and positive focal lenses to encompass the entire area of the echelon mirror and to achieve a homogeneous intensity at the echelon; the gate beam was directed through an optical delay stage to adjust the timing between the gate and probe beams. Subsequently, both the beams were focused onto a 1-mm-thick BBO crystal with a focusing lens f_1 to generate the SH light. The SH generation (SHG) signal reflecting the temporal correlation between the probe and gate pulses was imaged onto an entrance slit of a spectrometer coupled with a charge coupled device (CCD) detector (1024 × 252 pixels) using

an imaging lens f_2 and a periscope to obtain the spectrally resolved information. The focal lengths of the focusing and imaging lenses were 100 mm and 200 mm, respectively; the image of each step width (20 μm) of the echelon was transferred onto each pixel (24 μm square) of the CCD detector. The time delay of the probe pulse was aligned toward the direction of the entrance slit, while the spectrum of the probe pulses was recorded perpendicular to the direction of the slit. Finally, in this manner, we were able to obtain the time-frequency 2D image of the SHG signal (SHG-FROG) of the ultrafast laser pulses in real-time.

3. Results and Discussion

Figure 2(a) shows the observed echelon-based SHG-FROG traces for the laser pulses having a pulse duration of 150 fs with different time delays. The time delay was tuned by moving the optical delay stage. When the stage moves by 2 ps, the image moves vertically by 54 pixels on the CCD detector (shown by dotted lines). This result shows that the vertical axis of the CCD detector actually corresponds to the time axis as we had anticipated. From the moving distance, the time resolution per pixel can be estimated to be 37 fs/pixel; this value is in good agreement with the temporal step of the designed echelon mirror. Figure 2(b) shows the observed FROG traces for the laser pulses with different spectral widths of 0.34, 0.59, and 3.4 THz. The pulse duration becomes long as the spectral width decreases, implying that the laser pulses satisfy a Fourier TL relationship between time and frequency.

We will now compare the pulse durations measured by our technique with those by a conventional autocorrelator (Spectra-Physics 409). Figure 3 shows the obtained pulse durations as a function of the spectral width. The pulse duration was estimated from the

FROG and autocorrelation traces, while the spectral width of the laser pulse was directly measured by using a spectrometer. The hatched area shows the region below the spectral resolution of the spectrometer used here. The pulse durations measured by our technique using an echelon mirror are in good agreement with those measured by the conventional autocorrelator, showing that our technique works well for the pulse characterization above 150 fs. The Fourier TL relationships for the laser pulses having Gaussian and rectangular spectral shapes are shown by solid and dotted curves, respectively. Since the spectral width is limited by a variable slit [see Fig. 1(b)], the spectral shape of the laser pulses changes from the Gaussian to the rectangular as the spectral width narrows. In this manner, the observed behavior follows the Fourier TL relationship with the Gaussian and rectangular shapes when the pulse widths are broad and narrow, respectively. By using our new technique with the echelon mirror, we could successfully map the FROG traces of ultrafast laser pulses with different pulse durations in real-time.

Next, we measured the FROG traces for both negatively and positively chirped (NC and PC, respectively) laser pulses along with that of the TL pulse to characterize the chirp behavior of the laser pulses. For this measurement, we used a conventional Ti:sapphire regenerative amplifier system (Regen) with pulse duration, center wavelength, and repetition rate of 100 fs, 800 nm, and 1 kHz, respectively, as the light source [see Fig. 1(b)]. The NC and PC laser pulses are obtained by moving the position of the built-in compressor of the regenerative amplifier. Figure 4(a) shows the FROG traces for NC, nearly Fourier TL, and PC pulses with a single-shot accumulation. They can be reproduced well by their retrieved FROG traces using a FROG software (Femtosoft) within 1% error, as shown in Fig. 4(b).

The spectral phase of the laser pulses ψ is commonly written in terms of their Taylor

series expansions as follows: [27, 28)]

$$\psi(\omega) = \psi(\omega_0) + \frac{\partial \psi}{\partial \omega}(\omega - \omega_0) + \frac{1}{2}\frac{\partial^2 \psi}{\partial \omega^2}(\omega - \omega_0)^2 + \frac{1}{6}\frac{\partial^3 \psi}{\partial \omega^3}(\omega - \omega_0)^3 + ... \qquad (3)$$

Here, the second-order derivative corresponds to the group delay dispersion (GDD), while the third-order derivative causes asymmetry of the spectral image between the NC and PC pulses. We performed the phase-retrieval procedure by considering the zero- to third-order derivatives of the Taylor series expansion. From the retrieved FROG traces in Fig. 4(b), we could obtain the values of -1.14×10^4 fs^2 and 3.82×10^5 fs^3 for the NC pulses and 1.63×10^4 fs^2 and 3.39×10^5 fs^3 for the PC pulses. We also experimentally and numerically estimated the group velocity dispersion (GVD), which is defined by the GDD per unit length. The GVD value measured by our technique (6.36×10^4 fs^2/cm) is in quite good agreement with that calculated from the geometrical configuration of the built-in compressor (6.19×10^4 fs^2/cm). [29)]

The spectral profile and phase shift of the NC, nearly Fourier TL, and PC pulses obtained from the FROG traces (Fig. 4(a)) are shown in Fig. 5(a). The phase-shift profiles of the NC and PC pulses follow the quadratic dependence. On the other hand, the phase-shift profile of the Fourier TL pulse is not ideally flat but has the shortest pulse duration of ~100 fs. The retrieved spectral profiles are not very smooth; this might be due to numerical analysis issues with the FROG algorithm. Next, we compare the pulse durations obtained from the FROG traces with those measured by a conventional single-shot autocorrelator (SSA; Positive Light SSA-F). Figure 5(b) shows the autocorrelation traces obtained from our apparatus (solid lines) and SSA (dotted lines). The pulse duration obtained from our apparatus is almost equal to that from SSA.

These experimental results show that our new SHG-FROG apparatus using an echelon

mirror works well to quantitatively evaluate both the pulse duration and the chirp characteristics of the ultrashort laser pulses.

4. Conclusions

In conclusion, we have proposed and demonstrated a new technique for the time-frequency 2D imaging of ultrashort laser pulses using an echelon mirror that is fabricated on a Ni block with 500 steps to generate a spatially encoded time delay for the probe pulse. By using the SHG-FROG technique with the echelon mirror, the time-frequency 2D images for the ultrashort laser pulses with pulse durations of 0.15 to 6 ps were successfully measured in real-time. The chirp characteristics and spectral phases of the laser pulses were also evaluated on a single-shot basis. Our technique has significant technical advantages over the conventional autocorrelators and FROG techniques, such as single-shot detection of the time-frequency 2D images with wide temporal and spectral ranges, a small spot size at a nonlinear crystal, chirp-free characteristics of the echelon mirror, and capability for ultrafast measurements. Hence, it is a powerful spectroscopic tool not only for monitoring ultrashort laser pulses but also for studying ultrafast dynamics of materials.

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Figure Captions

Figure 1

(a) The concept of our apparatus to generate a spatially encoded time delay for probe pulses. The second harmonic signal generated at a nonlinear crystal is focused on an entrance slit of a spectrometer. Images of the fabricated echelon mirror taken by a conventional laser microscope are also shown. The surface roughness of the echelon which is examined on a stripe from points A to A', is less than 0.2 μm. (b) Experimental setup for the time-frequency two-dimensional (2D) imaging of ultrashort laser pulses using an echelon mirror. BS: beam splitter; WP: half-wave plate; CL: cylindrical lens; FL: bandpass filter; BBO: beta-BaB_2O_4. The light source can be switched from a Ti:sapphire oscillator (Laser) to a Ti:sapphire regenerative amplifier system (Regen).

Figure 2

(a) Second harmonic generation frequency-resolved optical gating (SHG-FROG) traces for the laser pulses with pulse duration of 150 fs for different time delays. (b) SHG-FROG traces for the laser pulses for different spectral widths 0.34, 0.59, and 3.4 THz.

Figure 3

Pulse durations of the laser pulses as a function of spectral width measured by our apparatus with an echelon mirror (solid circles) and a conventional autocorrelator (open circles). The solid and dotted curves indicate the Fourier transform-limit relationships having Gaussian and rectangular spectral shapes, respectively. The hatched area shows limitations of the spectral resolution of the spectrometer.

Figure 4

(a) Measured and (b) retrieved FROG traces for the negatively chirped (NC) (top), nearly Fourier transform-limited (TL) (middle), and positively chirped (PC) (bottom) laser pulses. The images are obtained with a single-shot accumulation.

Figure 5

(a) Retrieved spectral profiles and phase shifts for the negatively chirped (NC) (top), nearly Fourier transform-limited (TL) (middle), and positively chirped (PC) (bottom) laser pulses estimated from the frequency-resolved optical gating (FROG) traces shown in Fig. 4(a). (b) Autocorrelation traces measured by our apparatus with an echelon mirror (solid line) and by a conventional single-shot autocorrelator (SSA) (dotted line).

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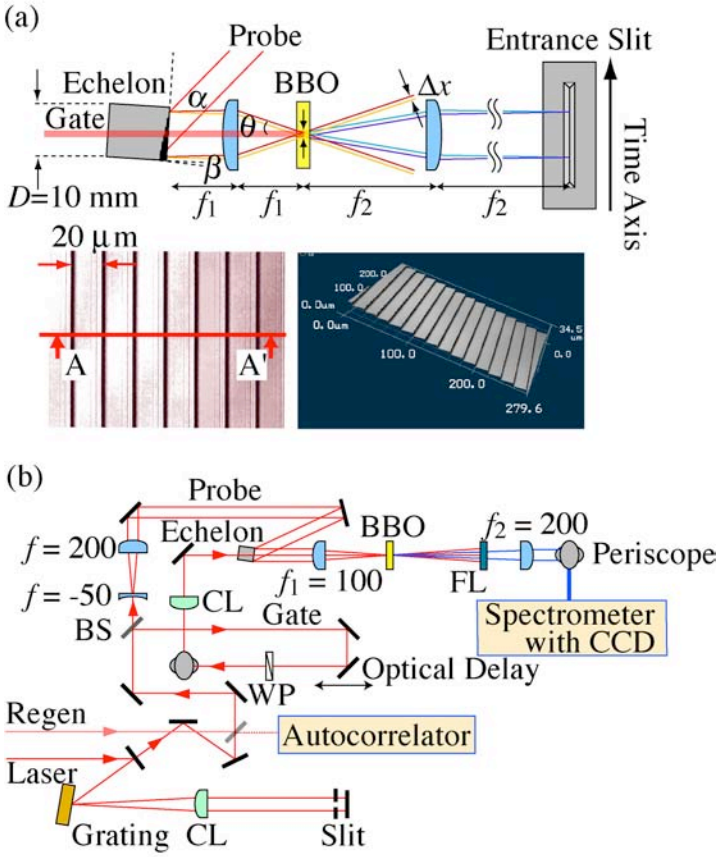
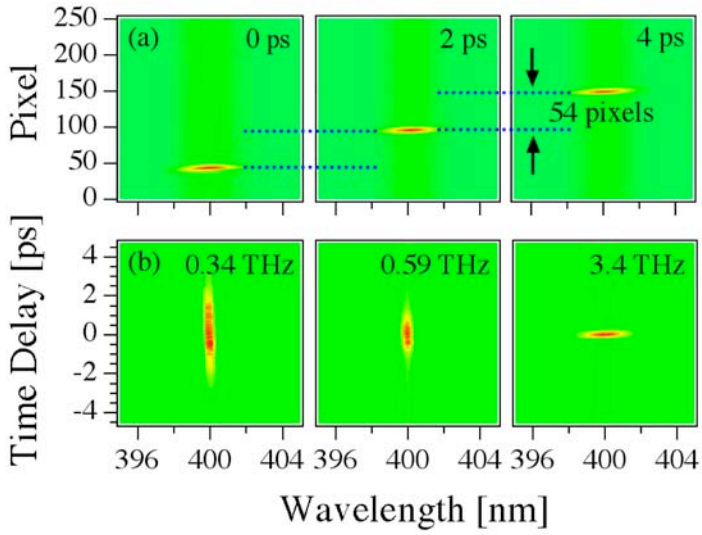


Figure 1 I. Katayama, H. Sakaibara, and J. Takeda



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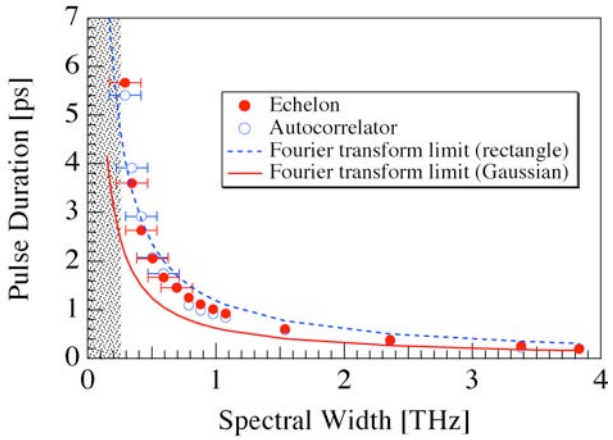

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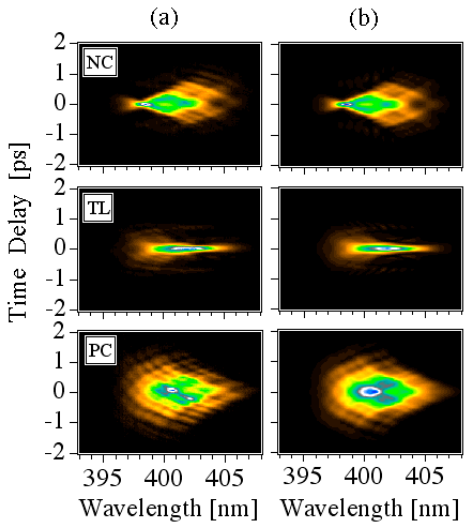

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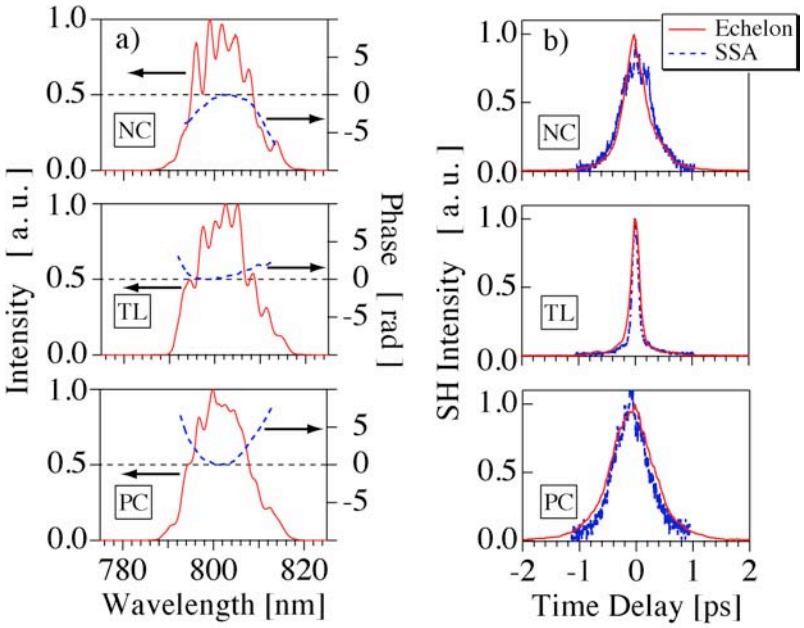

Figure 5 I. Katayama, H. Sakaibara, and J. Takeda