Spectral phase measurements for broad Raman sidebands by using spectral interferometry

Takayuki Suzuki,[1,2,*] Naoki Sawayama,[1] and Masayuki Katsuragawa[1,2]

[1]*Department of Applied Physics and Chemistry, The University of Electro-Communications, 1-5-1 Chofugaoka, Chofu-shi, Tokyo, Japan*

[2]*PRESTO, Japan Science and Technology Agency, 4-1-8 Honcho, Kawaguchi, Saitama, Japan*

**Corresponding author: t-suzuki@pc.uec.ac.jp*

Received August 25, 2008; revised October 22, 2008; accepted October 24, 2008; posted October 28, 2008 (Doc. ID 100487); published November 24, 2008

We report a method of spectral phase measurement for a femtosecond pulse train composed of a discrete Raman spectrum. The method is based on an idea of spectral interference. Making use of a small portion of two Raman pump laser radiations, we produce two sum-frequency spectra that spectrally interfere with each other. Based on this interfering sum-frequency spectra, we successfully derive the spectral phase and reconstruct the temporal profile. The reliability of this method is also confirmed by measuring the spectral phase changes caused by intentionally inserted silica plates of various thicknesses and comparing them to theoretical predictions. © 2008 Optical Society of America

OCIS codes: 120.3180, 120.5050, 190.4223, 190.5650, 320.5520, 320.7160.

Significant progress has recently been made regarding adiabatic excitation in Raman processes of molecular rotational and/or vibrational states to produce maximal coherence [1]. The maximal Raman coherence strongly modulates an incident light and generates broad high-order sidebands (Raman sidebands) [2–5]. One of the useful applications of such broad Raman sidebands is to produce ultrashort pulses [6–8].

To produce ultrashort pulses, generally spectral phase manipulation is a key issue. Such phase manipulation for Raman sidebands has been performed using a chirped mirror pair [7] or a spatial light modulator [8,9]. In those experiments pulse widths were measured by correlation techniques based on second-harmonic generation or four-wave mixing. The second-harmonic autocorrelation, however, gives only approximate temporal profiles and does not provide any spectral phase information. On the other hand, because the four-wave mixing scheme requires a pulse-shaping technique or frequency filtering, which adds some unknown dispersion [10], it can optimize the spectral phases but generally does not provide their original values.

In this study we fully characterize the ultrashort pulse train generated from Raman sidebands. The idea is based on the concept of the spectral phase interferometry for direct *e*-field reconstruction (SPIDER) technique [11], but modified to adapt the technique to *ultrashort pulses composed of discrete spectral lines*.

The point of our technique is that, instead of the chirped pulse used in the original SPIDER method, we combine a small portion of two Raman pump radiations with the Raman sidebands to produce two sum-frequency spectra. Here, the sum-frequency spectra are shifted from one another by $\Delta\Omega=\Omega_0-\Omega_{-1}$ (where Ω_0 and Ω_{-1} are the frequencies of the two excitation laser fields), which is exactly the same as the frequency spacing of the Raman sidebands. Each spectral line of one of the sum-frequency spectra, then, is completely overlapped by a line in the other spectrum. It should be noted that this scheme is more efficient than the original SPIDER technique, since there is no need to divide the Raman sidebands. *This scheme is applicable not only to Raman sideband spectra but also to any broad spectra composed of discrete spectral lines if their phases are mutually coherent*. For discrete spectra other than Raman sideband spectra, one may generate a two-color spectrum from the original spectrum by frequency filtering and use it in place of the two fundamentals in this experiment.

Next, we consider how the spectral phase appears in the interfering sum-frequency spectra. We assume that all spectral lines are monochromatic and can be expressed as delta functions. The nth pair consists of two sum-frequency lines $\Omega_0+\Omega_{n-1}$ and $\Omega_{-1}+\Omega_n$, where $\Omega_n=\Omega_0+n\Delta\Omega$ is the frequency of the nth-order Raman sideband. The intensity of this sum-frequency component can be expressed as

$$I_n^{\mathrm{SH}}=|E_{n-1}E_0e^{i\Omega_0\tau}+E_nE_{-1}e^{i\Omega_{-1}\tau}|^2=|E_{n-1}E_0|^2+|E_nE_{-1}|^2+2|E_{n-1}E_nE_0E_{-1}|\cos(\Delta\Omega\tau-\Delta\phi_n),\quad(1)$$

where $E_n=|E_n|e^{i\phi_n}$ is the complex amplitude of the nth-order Raman sideband; τ is the delay time of the excitation laser fields with respect to the Raman sidebands; ϕ_n is the phase of the nth-order sideband defined with respect to the phases of the laser fields $\phi_0,\phi_{-1}\equiv 0$ (which is equivalent to adding an arbitrary offset and a linear term of spectral phase); and $\Delta\phi_n=\phi_n-\phi_{n-1}$ is the phase difference. As expressed in Eq. (1), the intensity I_n^{SH} varies sinusoidally as a function of the delay τ with a fixed period of $2\pi/\Delta\Omega$, independent of n. We can therefore derive the phase difference $\Delta\phi_n$ by measuring the phase of the periodic change of I_n^{SH}. It should be noticed that the delay τ appears as $\Delta\Omega\tau$ in Eq. (1), not as $\Omega\tau$ as seen in typical interferometry. Since $\Delta\Omega$ is 1 order of magnitude smaller than Ω in this case, the resolution required

0146-9592/08/232809-3/$15.00

for the optical delay stage is merely on the order of a precision realized easily by a mechanical stage employing a differential micrometer.

The experimental setup is shown in Fig. 1. We used a dual-wavelength injection-locked pulsed Ti:sapphire laser [12] to drive a Raman transition in parahydrogen. We set the two frequencies Ω_{-1} and Ω_0 to $2\pi\times 371.7982$ THz (806.3312 nm) and $2\pi\times 382.4210$ THz (783.9331 nm), respectively, so that their difference, 10.6228 THz, was slightly detuned from the lowest rotational Raman resonance of parahydrogen. The pump pulses had a total energy of 4 mJ and pulse widths of 6 ns. The pump pulses were loosely focused into the molecular sample by a spherical lens ($f=800$ mm). The peak intensity was estimated to be ~ 4 GW/cm^2. Parahydrogen molecules were confined in the cell with a number density of 3×10^{20} cm^{-3} and cooled down to the liquid nitrogen temperature to populate the rovibrational ground state.

As shown in Fig. 1 the excitation pulses were partially reflected from the surface of a 5-mm-thick silica plate just before the sample cell and introduced into an optical delay stage to be used as a two-color pulse for generating sum frequencies, which were shifted by $\Delta\Omega$ with each other. Both the Raman sidebands and the two-color pulse were collimated by lenses with a focal length of 300 mm. Making use of the surface reflection of a silica plate, we attenuated the Raman sidebands to balance the total energy with the two-color pulse. The sidebands and the two-color pulse were sent parallel to one another into a concave mirror to be simultaneously focused at a 20 μm thick β-barium borate (BBO) crystal. The sum-frequency spectra were produced with various delays τ, and their interference was observed with a spectrometer.

Figure 2(a) shows typical interference spectra of the sum-frequency spectra with various delays $\tau=0$, 20, 40, 60, and 80 fs and the inset shows the Raman sideband spectrum to be characterized. The peak values of the six spectral lines are plotted in Fig. 2(b) against delay τ (filled circle). Different pairs of the sum-frequency components interfere constructively or destructively at different delay times depending on

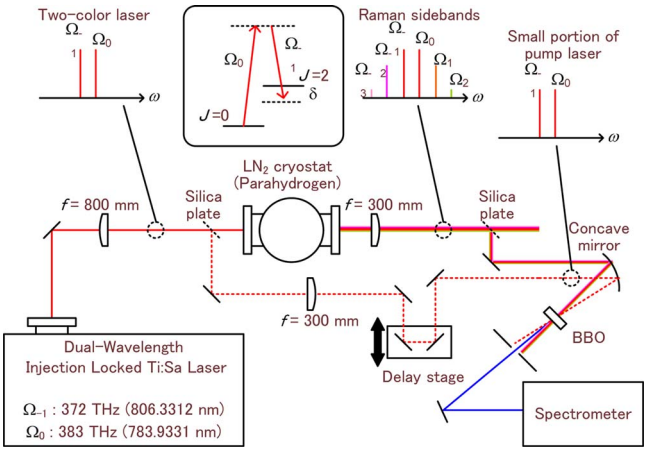

Fig. 1. (Color online) Schematic of experimental setup for broad Raman sideband generation and its spectral phase measurement system.

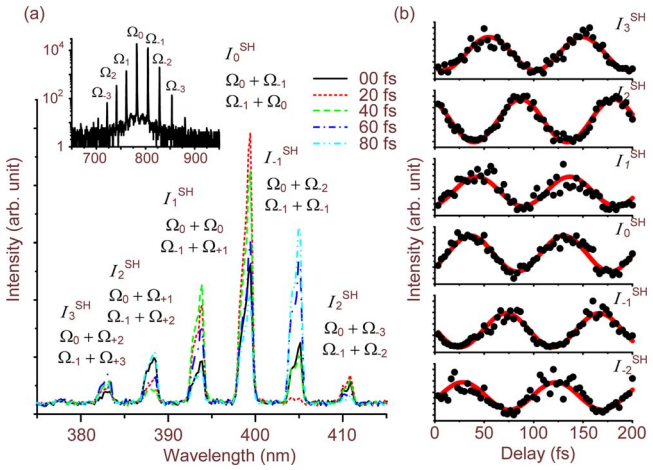

Fig. 2. (Color online) (a) Sum-frequency spectra with various delays. The intensities of the spectral lines periodically change with different phases reflecting the phase differences $\Delta\phi_n$. The symbols above the spectral lines designate corresponding frequency components of the fundamental. (b) Intensity variations of I_n^{SH} with respect to the delay τ (filled circle) and their fitting curves (solid curves).

the phase differences, as expressed in Eq. (1). The results show that the interfering intensities I_n^{SH} periodically change with the same period of 94 fs ($=2\pi/\Delta\Omega\equiv T$), independent of n. The solid curves in Fig. 2(b) are sinusoidal fits with the fixed period of 94 fs.

By measuring the phases of the periodic change of I_n^{SH} we obtain the phase differences $\Delta\phi_n$ between the corresponding two sidebands Ω_n and Ω_{-1}. Figure 3(a) shows the spectral phase (open circles) derived by summing the phase differences $\Delta\phi_n$ as well as its intensity spectrum of the Raman sidebands (solid curve). The ultrashort pulses generated from the Raman sidebands were thus fully characterized. Figure 3(b) shows the temporal profile reconstructed from the results of Fig. 3(a). The pulse width and the repetition period were measured as 52 and 94 fs, respectively.

Generally, nonlinear processes such as sum-frequency generation are affected sensitively by fundamental intensity fluctuations. In fact, as shown in Fig. 2(b), weak sum-frequency components that are produced from high-order Raman sidebands have intensity fluctuations. Our scheme, however, does not

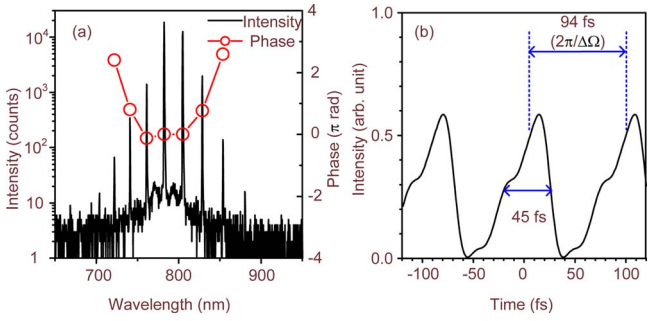

Fig. 3. (Color online) (a) Raman sideband spectrum (solid curve) and its spectral phase (open circles). (b) Reconstructed temporal profile of the Raman sideband.

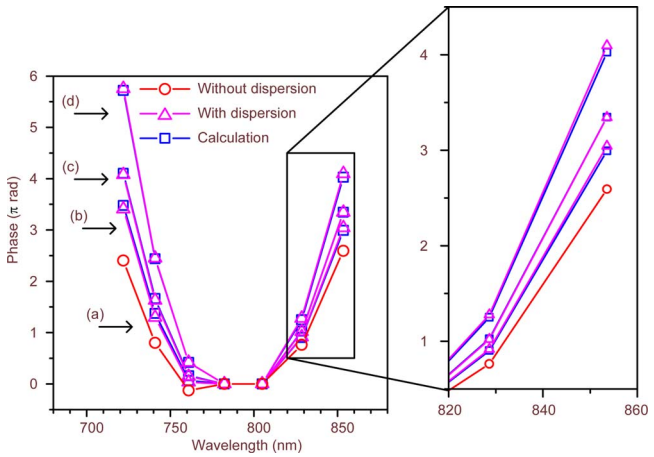

Fig. 4. (Color online) Spectral phases of the Raman sideband spectra (a) without, and with (b) 3, (c) 5, and (d) 10 mm thick silica plates. The open circles and open triangles indicate experimental results, and the open squares indicate results derived with a known refractive index dispersion of silica.

use the absolute values of I_n^{SH} for the spectral phase derivation procedure. The only important information is the phase of the periodic change of I_n^{SH}. Therefore we can still identify the spectral phase well under such intensity fluctuations. On the other hand, acceptance bandwidth and angular width become the main restrictions for the use of BBO crystals or similar methods. For an extremely broad spectrum and monocycle pulses, we may perform the phase measurement by dividing the measurement into multiple parts with different incident angles to a nonlinear crystal. This procedure extends its applicable bandwidth to the full range of a crystal to produce sum-frequency components.

Finally, to verify the reliability of our method we measured the spectral phases with fused silica plates inserted into the optical path of the Raman sidebands to add known dispersion. We used silica plates of three different thicknesses; 3, 5, and 10 mm. Figure 4 shows the results of the phase measurements (a) without (open circles), and with the (b) 3, (c) 5, and (d) 10 mm thick silica plates (open triangles). The theoretical predictions are also shown in (Fig. 4, open squares); these were obtained by calculating the refractive index dispersion of the silica plates using the Sellmeier equation [13] and adding them to the previous result without the silica plates [Fig. 3(b)]. The measurements and theoretical predictions were in good agreement, as seen in the enlarged graph in Fig. 4. The observation errors on ϕ_n originate in the sinusoidal-fitting errors on $\Delta\phi_n$ in Fig. 2(b), and were sufficiently small ($\sim$0.05 rad each). The reliability of our spectral phase measurement method was thus confirmed.

We should emphasize that this scheme can be rapidly executed compared with other methods that require iterative algorithms, such as the frequency-resolved optical gating (FROG) technique [14]. Moreover, if the intensity of the sum-frequency components are stabilized, and once the intensity fringes versus the delay are obtained for all the peaks, the spectral phase can, in principle, be estimated from only two trials of measurements. By normalizing the minimum and the maximum of I_n^{SH} to -1 and 1, respectively, the spectral phase differences can be derived up to their signs with a single measurement as the arccosine of the normalized sum-frequency intensities. To obtain the signs it is necessary to measure the spectrum again with a delay of $T/4$, for example.

In summary, we performed a spectral phase measurement of broad Raman sidebands composed of discrete spectral components, based on the idea of spectral interference. We confirmed the reliability of this method by measuring the phase changes produced by silica plates with various thicknesses and comparing them to theoretical predictions. Our method is robust against intensity and delay fluctuations. Furthermore, it is rapidly executed because it does not use iterative algorithms.

References

1. S. E. Harris and A. V. Sokolov, Phys. Rev. A **55**, R4019 (1997).
2. A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **85**, 562 (2000).
3. J. Q. Liang, M. Katsuragawa, F. Le Kien, and K. Hakuta, Phys. Rev. Lett. **85**, 2474 (2000).
4. M. Katsuragawa, J. Q. Liang, F. Le Kien, and K. Hakuta, Phys. Rev. A **65**, 025801 (2002).
5. D. D. Yavuz, D. R. Walker, G. Y. Yin, and S. E. Harris, Opt. Lett. **27**, 769 (2002).
6. S. E. Harris, D. R. Walker, and D. D. Yavuz, Phys. Rev. A **65**, 021801(R) (2002).
7. M. Katsuragawa, K. Yokoyama, and T. Onose, Opt. Express **13**, 5628 (2005).
8. M. Y. Shiverdin, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **94**, 033904 (2005).
9. W.-J. Chen, Z.-M. Hsieh, S. W. Huang, H.-Y. Su, C.-J. Lai, T.-T. Tang, C.-H. Lin, C.-K. Lee, R.-P. Pan, and A. H. Kung, Phys. Rev. Lett. **100**, 163906 (2008).
10. S. N. Goda, M. Y. Shverdin, D. R. Walker, and S. E. Harris, Opt. Lett. **30**, 1222 (2005).
11. C. Iaconic and I. A. Walmsley, Opt. Lett. **23**, 792 (1998).
12. M. Katsuragawa and T. Onose, Opt. Lett. **30**, 2421 (2005).
13. I. H. Malitson, J. Opt. Soc. Am. **55**, 1205 (1965).
14. R. Trebino and D. J. Kane, J. Opt. Soc. Am. A **10**, 1101 (1993).