Generation of ultrashort optical pulses using multiple coherent anti-Stokes Raman scattering in a crystal at room temperature

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We demonstrate Fourier synthesis of multiple coherent anti-Stokes Raman scattering signals in a $LiNbO_3$ crystal at room temperature. The signals up to the 20th order (470–800 nm) were generated by two crossing femtosecond Ti:sapphire laser pulses. Angle dispersion of the signals was compensated into one white-continuum beam by modifying a conventional $4f$ configuration. Spectral phase of the signal was measured by spectral phase interferometry for direct electric-field reconstruction. Isolated pulses with 25 fs duration at 1 kHz were generated only by appropriately aligning the angle-dispersion compensator. This result opens the possibility of the generation of subfemtosecond pulses in the visible region. © *2008 American Institute of Physics*.
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Generation of an intense one-optical-cycle pulse is attractive not only for the production of an isolated attosecond pulse in the extreme ultraviolet region by high harmonic generation[1] but also for the observation of ultrafast phenomena in any scientific field. One of the promising methods to generate such a pulse is the Fourier synthesis of Raman sidebands. Zhavoronkov and Korn have generated an isolated sub-4-fs pulse with a 400 nm center frequency by the "pump-probe technique" in a SF_6-gas-filled hollow fiber.[2] However, the bandwidth was restricted to ~100 nm. In contrast, for the generation of a pulse whose spectrum covers the whole visible region, which is more difficult and challenging, the "two-color resonant excitation" using near-infrared pulses seems to be more suitable. Shverdin *et al.* have generated a train of 1.6 fs pulses using discrete Raman sidebands of cooled D_2 gas.[3] However, its repetition period is too short (11 fs) because spectra of the adjacent Raman sidebands do not overlap at all, owing to the narrow Raman peaks of the gas.

On the other hand, it has been found that multiple coherent anti-Stokes Raman scattering (CARS) signals with broad spectral widths can be generated even in common crystals, such as $KTaO_3$, $KNbO_3$, TiO_2,[4] $PbWO_4$,[5] etc., by the excitation with two-color femtosecond laser pulses at room temperature, where all the spectra of adjacent CARS peaks overlap so that the entire spectrum with an almost two-octave bandwidth is continuous. This property gives us two advantages over the cases in a gas. One is the potential of the generation of an "isolated" ultrashort pulse. The other is that the spectral phase can be directly measured by some methods, such as spectral phase interferometry for direct electric-field reconstruction (SPIDER).[6] This is important not only from the viewpoint of the ultrashort optical pulse technology but also from the viewpoint of physics in quantum optics because it is an interesting issue whether or not discrete phase shifts of the signals due to Raman-coherence formation in a crystal can be directly observed.

However, refractive-index dispersion of solids is generally so large that the two collinear input beams cannot efficiently generate CARS signals owing to the phase-matching limitation in crystals with a finite thickness. Hence, two input beams are forced to be noncollinear with some crossing angle, and multiple CARS signals are generated with considerable angle dispersion. We have found that the angle-dispersion compensation can be achieved as follows:[7] provided the two laser beams with frequencies and wave vectors, $(\omega_1,\boldsymbol{k}_1)$ and $(\omega_2,\boldsymbol{k}_2)$, are used to excite a crystal, the nth-order CARS signal is generated in the direction of $\boldsymbol{k}_n \equiv \boldsymbol{k}_1 + n(\boldsymbol{k}_1 - \boldsymbol{k}_2)$, by which the angle and the wavelength of the multiple CARS signals are calculated to have a linear relationship, being similar to that between the wavelength of light and the diffraction angle of a grating. Hence, by substituting one of the gratings in a conventional $4f$ pulse shaper with a crystal [Fig. 1(a)], all the signals are expected to be diffracted into the same direction. In the present letter, we demonstrate the angle-dispersion compensation of the multiple CARS signals from a $LiNbO_3$ crystal, the characteriza-

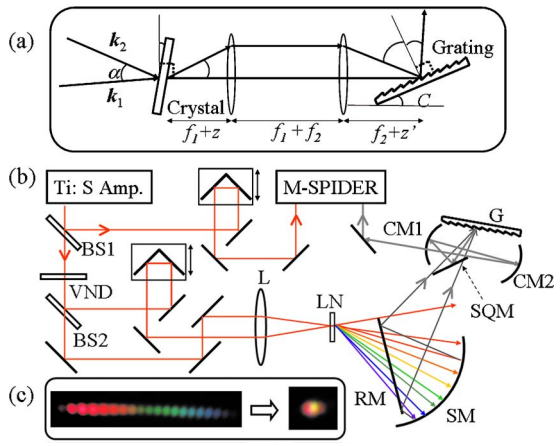

FIG. 1. (Color online) (a) Conceptual scheme of angle-dispersion compensation. (b) Experimental setup for angle-dispersion compensation, phase characterization, and pulse compression. (c) Photographs of multiple CARS signals before (left) and after (right) the angle-dispersion compensation.

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tion (observation) of their spectral phase by the modified (M) SPIDER,[8] which is the same as the cross-correlation (X) SPIDER, and the generation of an isolated optical pulse, which has been achieved by making full use of the property of the modified $4f$ configuration.

The experimental setup is shown in Fig. 1(b). The YZ surface of a $LiNbO_3$ crystal with a 0.5 mm thickness (LN) was simultaneously irradiated by two fundamental laser pulses with energies of 6 and 11 μJ, and a crossing angle of 1.9° in the air, which were from one multipass Ti:sapphire laser amplification system (center wavelength: 810 nm, duration: 30 fs, repetition rate: 1 kHz). The polarization of the input pulses was chosen to be parallel to the Y axis so that the signal intensity became the strongest. Two-color pulses whose frequency difference is set to be the frequency of a Raman-active phonon are generally used to generate CARS signals. Similarly, two slightly chirped (+100–200 fs^2) fundamental femtosecond pulses can also generate such signals by appropriately adjusting the relative time delay between them, because the bandwidth of the laser pulses ($\sim$800 cm^{-1}) is broad enough to cover frequencies of most of the Raman-active optical phonons of the crystals. The CARS signals were reflected back by a spherical mirror with a focal length of 100 mm (SM) and a rectangular mirror (RM), and focused on a grating with a groove density of 1200 lines/mm and a blaze wavelength at 500 nm (G). To tune the magnification factor of M defined as f_1/f_2 arbitrarily [see Fig. 1(a)], only one spherical mirror was used. In this case, M is the ratio of the distance between the crystal and the spherical mirror, and that between the spherical mirror and the grating. The M value was carefully chosen so that the signals were diffracted into the same direction; in the present study, the value is 0.74. To minimize the spatial chirp of the collimated signal beam, the position of the grating was carefully adjusted using a stage. The diffracted beam was picked up by a small square mirror (SQM) and collimated both horizontally and vertically by two cylindrical mirrors [CM1 (2) with a 100 (250) mm focal length] each. Figure 1(c) shows photographs of the CARS signals taken just behind the crystal (left) and after the angle-dispersion compensation (right). The beam size was approximately 4 mm in diameter at a position of 3 m from the grating so that the collimation accuracy is in the order of several mrad. Fundamental pulses reflected by a beam splitter (BS1), were passed through a 10 cm long TF5 glass twice and served as referential pulses in the M-SPIDER measurement. A pulse to be measured was divided into two pulses with a relative time delay of 1078 fs in a Michelson interferometer and upconverted by the referential pulse in a β-barium-borate (BBO) crystal with a 50 μm thickness. The interferogram of the upconverted pulses with a spectral shear of 5.09 THz (170 cm^{-1}) was observed by a spectrometer with an intensified charge coupled device. A SPIDER signal was observed with good visibility from 357 to 397 nm, even at minima of adjacent CARS peaks. The wavelength range corresponds to that of the collimated CARS beam from 637 to 777 nm since the spectral shift given by the referential pulse was 369 THz.

Figure 2 shows the intensity spectrum and the spectral phase of the angle-dispersion compensated CARS beam measured by the M-SPIDER. As eye-guided by red (blue) vertical lines, the intensity spectrum has peaks with a spacing of 198 (299) cm^{-1} (both of E, LO) in the frequency range

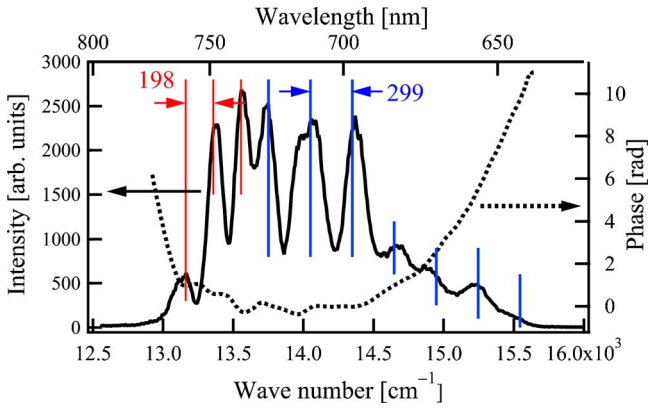

FIG. 2. (Color online) Intensity spectrum (black solid curve) and spectral phase (black dotted curve) of the angle-dispersion compensated multiple CARS signals from a $LiNbO_3$ crystal. The input-pulse energies are 6 and 11 μJ.

lower (higher) than 13 750 cm^{-1}. Though the entire spectrum of the collimated beam reached 470 nm, a frequency component of the CARS signals whose SPIDER signal was not observed was eliminated before the angle-dispersion compensation in order to reasonably compare the temporal profile of the generated pulse with that of the Fourier transform-limited (TL) one.

By carefully adjusting the rotation angle and the position of the grating, the spectral phase became almost flat in the frequency range from 13 000 to 15 600 cm^{-1}. Here, we can see many steplike changes in the spectral phase, which will be discussed later. The temporal profile of the generated pulse and that of the TL one are shown in Fig. 3. Unlike the cases of gases,[3,9] except a study using the pump-probe technique with femtosecond laser pulses in SF_6 gas,[2] an isolated pulse was generated owing to its continuous spectrum. The duration of the observed pulse is 25 fs while that of the TL one is 11 fs. This difference seems to come from the incompletely flattened spectral phase. The energy of the compressed pulse is estimated to be a few hundred nJ, because the total energy of the multiple CARS signals was 1 μJ at most, and it decreases by 50%–60% after the angle-dispersion compensator whose throughput is mainly determined by the diffraction efficiency of the grating.

Now we discuss our experimental results. First, we discuss how the compensations of both the angle and temporal dispersions of the multiple CARS signals are achieved in the experimental setup. The position and the angle of the diffractive grating (G) play the most important role. It is known

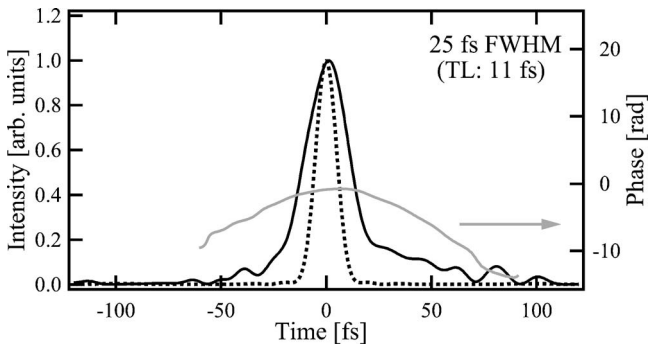

FIG. 3. Temporal intensity profile of the compressed pulse (black solid curve) and that of transform-limited one (black dotted curve). Solid gray curve shows temporal phase profile of the compressed pulse.

that angle dispersive elements, such as prisms and gratings in the configuration shown in Fig. 1(a), introduce group delay dispersion (GDD) in such a manner as,[10]

$$\frac{d^2\Psi}{d\Omega^2} = -\frac{1}{c}(z'M^2 + z)\left[\left(2\frac{da}{d\Omega} + \Omega\frac{d^2a}{d\Omega^2}\right)\sin a + \Omega\left(\frac{da}{d\Omega}\right)^2 \cos a\right]. \quad (1)$$

Here, a is the diffracted angle of a ray with an angular frequency Ω, and is measured from the direction of the center-frequency component. z is the distance between the crystal and the focal point, and z' is the distance between the focal point and the grating. [see Fig. 1(a)]. Since $da/d\Omega$ is a function of the angle of the grating, the GDD depends on both the angle and the position of the grating. According to the calculation using parameters of $z=0$ μm, $z'=\pm 300$ μm, and $M=0.74$, the applied GDD is ∓ 200 fs^2 at 700 nm. If we rotate the grating by 1°, the GDD changes by 5 fs^2. Thus, we can understand that the spectral phase of the collimated beam is adjustable by slightly moving the grating. In practice, the manual adjustment of the optical system on the GDD was quite delicate than the simulation. We think this is because z cannot be assumed to be zero.

Next, let us examine the spectral phase profile in more detail and discuss what prevents further pulse compression in the present study. Though the spectral phase is almost flat within 1 rad fluctuation in the range from 13 200 to 14 500 cm^{-1}, then, it increases a little steeply to 11 rad. We think that this is not due to the fundamental problem of the collimated CARS beam but due to the non-negligible higher-order dispersion applied by the modified $4f$ configuration. Hence, it will be certain that we can generate TL pulses with this scheme by using a programable multichannel spatial light modulator[11] or well-designed chirped mirrors.

Finally, let us discuss the origin of the ripples observed in the spectral phase profile in Fig. 2. This is related to the possibility of the observation of the discrete phase shift due to Raman coherence. There, ripples with spacings of 155 and 369 cm^{-1} are seen, which correspond to the frequencies of TO (E) phonons. On the other hand, the spectral resolution determined by the spectral shear is 340 cm^{-1} according to the Nyquist limit, so that one might think that such phonon frequencies are not observable. However, a simple simulation shows that some information on periodicity can still be retrieved even if the sampling step is nearly equal to the frequency of interest, although this does not necessarily rule out the ambiguity of the resolution and may lead to measurement of incorrect periods due to the aliasing effect.

In conclusion, we generated an isolated 25 fs optical pulse using multiple CARS signals from a single crystal of $LiNbO_3$ at room temperature. The crystal was noncollinearly pumped by two fundamental pulses from one Ti:sapphire laser amplification system. The angle and the temporal dispersions were compensated for by modifying a conventional $4f$ configuration.

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