Generation of Single Intense Short Optical Pulses by Ultrafast Molecular Phase Modulation

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(Received 31 December 2001; published 6 May 2002)

Pulses with durations below 4 fs have been generated using the method of ultrafast molecular phase modulation. A laser pulse shorter than the molecular vibrational or rotational period obtains spectral broadening during propagation along a hollow waveguide filled with previously impulsively excited Raman active gases. The induced time dependent phase, frequency, and frequency chirp are controllable by changing the delay between excitation and probe pulse within the molecular vibrational period.

DOI: 10.1103/PhysRevLett.88.203901 PACS numbers: 42.65.Re, 33.80.–b

The technology of intense short optical pulse generation using nonlinear methods has been revolutionized in pulse width and energy over the past few years with the help of self-phase modulation (SPM) as an effective process to produce additional spectral components. The SPM technique leads to a pulse shortening down to 4.5 fs [1] with energies in the sub-mJ-range and has opened now the way to attosecond science through high harmonic generation in the vacuum ultraviolet region [2,3]. The disadvantage of ultrafast phase modulation caused by SPM is its self-induced character; i.e., the induced time dependent phase always follows the pulse intensity shape due to the quasi-instantaneous character of the electronic Kerr effect in the atomic gases [4]. Moreover, the further scaling of SPM technique into the high intensity regime to produce ultrawide spectra [5] appears to be problematic, since the phase behavior becomes much more complicated, and its complete compensation becomes practically not attainable.

Recently a new mechanism for ultrashort optical pulse generation named molecular phase modulation (MPM) has been demonstrated with the basic idea to employ a linear spectral broadening that generally takes place in optical medium with temporally modulated dielectric parameters [6]. This mechanism is based on impulsive excitation [7] of molecular motion in Raman-active gases, where a first short and high intensity pulse excites impulsively a coherent molecular motion (vibration or rotation) in a molecular gas and a second, relatively weak delayed pulse obtains a strong phase modulation due to refractive index variation caused by this molecular motion. In the proposed scheme the deteriorating effects caused by nonlinear self-action and ionization are confined within the strong pump pulse; therefore the nonlinear cascade scattering taking place in conventional stimulated Raman scattering (SRS) is replaced for the delayed pulse to a linear one [6]. In [8] was shown that for the delayed pulse with a duration longer than the excited molecular vibration ($\tau_i \geq 100$ fs) the spectum of the output radiation exhibits individually resolved Stokes and anti-Stokes sidebands. For SF_6 as the Raman-active medium the output radiation could be compressed to a train of pulses with period of 43 fs and an individual pulse width of 5.8 fs [8,9].

In this paper we demonstrate for the first time the application of molecular phase modulation for a generation of single, intense, and extremely short pulse using a second delayed pulse with a duration considerably shorter than the molecular vibrational or rotational period. Contrary to SPM the time dependent phase induced by molecular phase modulation can be controlled by changing the delay between the pump and the probe pulses.

The physical phenomena connected with stimulated Raman scattering have been extensively investigated during last decades, and their main properties are now well established [6–12]. A synthesis of a pulse train with nearly single-cycle waveforms was demonstrated [13,14] by effective generation of multiple Raman components under adiabatic excitation of D_2 with nanosecond pulses. The models become more complicated when intense fs pulses are involved in the process, since the high order nonlinear effects as well as a group velocity dispersion (GVD) should be taken into account [6,8,15,16].

One of the important physical origins of SRS is connected with the vibrational period of molecular motion $T_v = 2\pi/\Omega_v$, where Ω_v is molecular vibrational frequency. If the laser pulse has a duration of $\tau_p \ll T_v$, the SRS proceeds in the "impulsive" regime. Under this condition the spectrum of the pulse initially contains spectral components shifted at Ω_v, so that SRS will occur as a kind of coherent, self-conversion process with up to 100% efficiency of transformation [6]. In gases commonly used for SRS experiments the vibration frequencies lie in the regions of several hundreds cm^{-1}, so that the laser pulses below 100 fs can easily satisfy the condition of "impulsive" excitation. The amplitude of molecular movements or the SRS gain in impulsively excited gases is determined by the full laser pulse energy and is not sensitive to the temporal structure of the pulse [17]. After interaction with a pulse for a time interval short enough to neglect the relaxation decay the molecules exhibit freely dumping oscillation in the form

$$Q(t) \approx Q_0 \exp(-t/T_2) \sin(\Omega_v^d t), \quad (1)$$

where $\Omega_v^d = \Omega_v - (1/T_2)^2 \approx \Omega_v$ and $T_2 \gg T_v$ is the dephasing time. The wave of excitation is formed in the

medium and spreads with the group velocity of the pump pulse. If the second relatively weak pulse, which does not change the state of molecular motion, is delayed by $\tau_d > \tau_p$ it will propagate in the preliminary excited media in a "linear" scattering regime [6]. The solution of the wave equation for the delayed pulse field takes the form

$$E_i(z, \tau_i) = E_i(0, \tau_i) \exp[-iG_i z \sin(\Omega_v \tau_i)], \quad (2)$$

where $E_i(0, \tau_i)$ is the amplitude of the injected field with a duration of τ_i, G_i has a sense of Raman gain, and the group velocity mismatch between pump and probe pulses is neglected. The phase of the resulting field shows a periodic temporal modulation, but the result of this modulation will depend on the injected pulse duration τ_i. When the pulse duration is $\tau_i > T_v$ and the pulse covers more than one period of molecular vibration, its phase obtains a periodic sinusoidal modulation and in the spectral domain sideband frequencies at $w_{ni} = w_i \pm n\Omega_v$ ($n = \pm 1, 2, \ldots$) will be observed. The pulse compression could be performed with positive or negative GVD that originates from the periodic character of phase modulation function [8,9]. In the case when the pulse duration is $\tau_i < T_v$ the delayed pulse covers only a part of sinusoidal modulation function, so the generated phase ϕ, frequencies $\partial\phi/\partial t$, and chirp $\partial^2\phi/\partial t^2$ will depend strongly on which part of the function is imposed on the pulse. We can easily choose the most "suitable" one by changing the delay between pump and probe pulses within the molecular modulation period.

The experiments on MPM were performed in a 1 m length and 128 μm diameter hollow waveguide filled with H_2 or SF_6 gases. A Ti-sapphire laser system generates 30 fs pulses with energies up to 0.8 mJ at $\lambda_0 = 800$ nm. About 0.5 mJ was used to excite impulsively Raman-active gases inside the waveguide, where a pump pulse intensity as much as 10 TW/cm^2 could be reached. Another part of the beam was used to produce the probe pulse at 400 nm in a 50 μm thick BBO (beta-barium-borate) crystal. The probe pulse was conveniently taken at the 2nd harmonic to distinguish between the pump and the probe in the experiment. The probe pulse had a duration of 27 fs, but if a considerably shorter pulse is required it could be produced by compressing the probe pulse, which experiences strong SPM in an additional hollow waveguide filled with Ar. The energy of the probe pulse before injection to the waveguide was typically about 10 μJ. The rather high energy of the transmitted probe pulse (between 1–2 μJ typically) enabled us to measure the pulse width and FROG (frequency resolved optical gating) traces using a low dispersion method based on self-diffraction (SD) in a 50 μm thick sapphire plate.

At first we used hydrogen at a pressure of 1.3 bar as a Raman-active media. In ordinary conditions H_2 contains a mixture of 25% para-H_2 and 75% orto-H_2, and as it was shown in [9] the impulsively excited Raman spectrum mostly exhibits the active rotational modes of orto-H_2 with $\Omega_r = 587$ cm^{-1} and $T_{\text{rot}} = 57$ fs. The second probe pulse was injected in the waveguide with about 400 fs delay relative to the maximum of the pump pulse. With an excitation intensity of about 10 TW/cm^2 the spectrum exhibits a strong broadening up to the 26 nm FWHM. Figure 1 shows the measured probe pulse spectra for different delays between the pump and probe pulses with and without the strong excitation. We found that the shape and the center frequency of the probe pulse spectrum changes strongly within the delay of one molecular period. The expected changes in the generated spectrum can be estimated from the first time derivative of the time dependent phase as $\partial\omega = -\partial\phi/\partial t = -G_i z \cos(\Omega_v \tau_i)$, so the change of the $\phi(t)$-function slope imposed on the probe pulse leads also to a change in the generated frequencies. The spectral shape in Fig. 1(c) resembles typical SPM behavior that can occur when the probe pulse is centered at the local extremum of the modulation molecular function. In this case the part of the periodic modulation function imposed on the probe pulse is close to the laser pulse intensity shape with a duration of one-half of the molecular period and the spectrum has a similar symmetric shape. Figure 2 shows the FROG traces for different experimental conditions. For the pulse without the excitation as it emerges from the waveguide after the fused silica window the FROG trace on Fig. 2(a) reveals a positive almost linear chirp with the pulse duration of 40 fs. An additional positive GVD of about 320 fs^2 was introduced. Figure 2(b) depicts the FROG trace for the pulse experiences 6 bounces off negatively GVD mirrors centered at 400 nm with about -40 fs^2 per bounce. The reduction of the chirp and a corresponding pulse shortening down to 30 fs is clearly seen. Figure 2(c) shows the FROG trace in the presence of impulsive excitation and with the delay tuned to generate a 8.7 fs pulse as it was measured by the SD autocorrelator. The additional phase modulation imposed to the probe

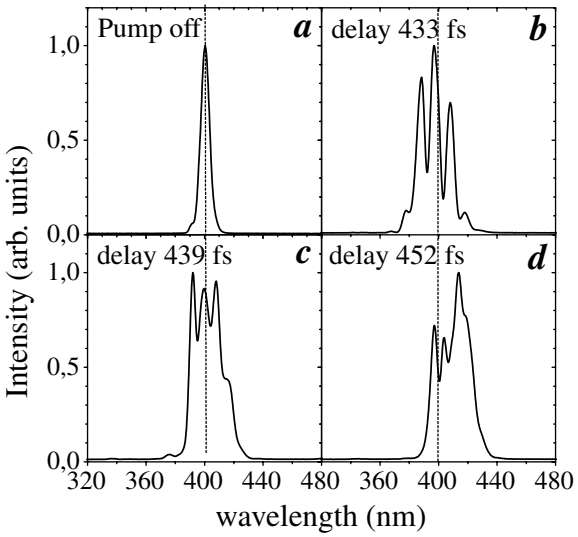

FIG. 1. Probe pulse spectrum in dependence on the delay between the pump and the probe pulses without (a) and with (b), (c), (d) rotational excitation in H_2.

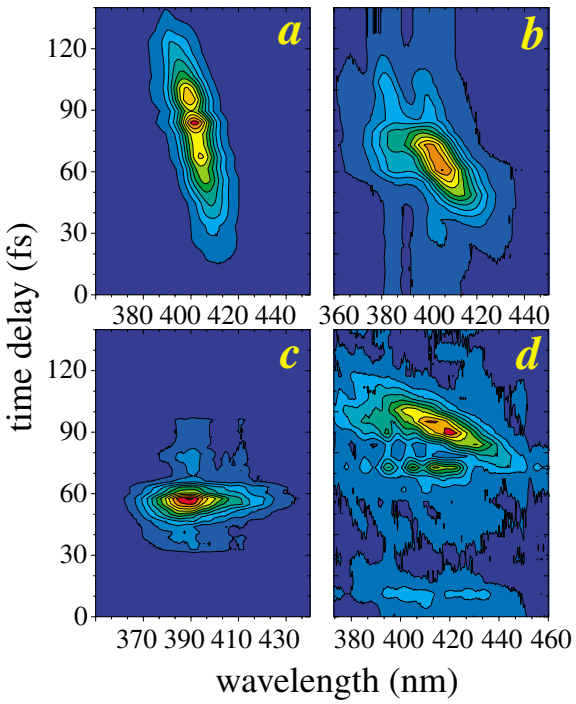

FIG. 2 (color). FROG traces of the probe pulse at different experimental conditions (see text) for rotational excitation in H_2 and compression.

pulse by the molecular modulation process is negative in that case. The mathematical reconstruction of the pulses using the "Femtosoft"-FROG program gives FWHM pulse width of 9.0 fs with the time-bandwidth product of 1.3 from time-bandwidth limit case for the measured spectrum. The reconstructed pulse's phase shows quadratic behavior for the FROG traces on Figs. 2(a) and 2(b), but changes to a linear one with a phase difference of 0.3π for Fig. 2(c). Figure 2(d) shows the trace in the case where the delay is changed by half a period. Additional chirp and satellite pulses appear due to the positive phase modulation imposed by the molecular modulation process. Both the spectra and the FROG measurements prove that the probe-pulse phase can be continuously tuned between negative and positive values by changing the delay between the pump and probe pulses.

The highly symmetric, spherical top SF_6 molecule exhibits no rotational Raman spectrum, but a strong, symmetric vibrational A_{1g} mode with $\Omega_v = 773\ \mathrm{cm}^{-1}$ and $T_v = 43$ fs. The electronic absorption band of SF_6 lies far above of the pump and probe wavelengths, so only the A_{1g}- Raman mode is efficiently excited by impulsive pumping with 30 fs pulses. Since the 27 fs probe pulse is not short enough for the experiments with SF_6, a probe pulse with 15 fs duration at 400 nm was generated using SPM. The experiments were performed at a pressure of 800 mbar. For such SF_6 gas pressure was found [18] that the combined action of the positive gas dispersion with the plasmalike waveguide dispersion can realize group velocity matching between the pump at 800 nm and the probe at 400 nm. It is important that during the propagation along the waveguide the probe pulse should interact with the same part of the molecular modulation function spread with the group velocity of the pump pulse. Additional negative dispersion after the output window of the gas chamber was introduced with 6 bounces off negative GVD mirrors. Figure 3 shows the SD-autocorrelation traces and the corresponding spectra in dependence on the delay between the pump and the probe pulses. The figures show clear spectral shifts to shorter or longer wavelength depending on the delay. A quasicontinuous spectrum depicted at Figs. 3(d) and 3(f) with a width of almost 100 nm has been generated under optimum conditions. The shortest SD-autocorrelation trace of 4.65 fs has been obtained with a delay of 242 fs. This delay has been found experimentally, and it corresponds to the best phase compensation for the dispersion of the whole system. Figures 3(a) and 3(b) resemble Figures 3(g) and 3(h) in spectrum and pulse width since the delay between them is close to one molecular period of 43 fs. The rather complicated spectral shape of the phase modulated pulses did not allow us to make a successful spectral phase retrieval using a SD FROG algorithm as in the case of H_2. Such investigations

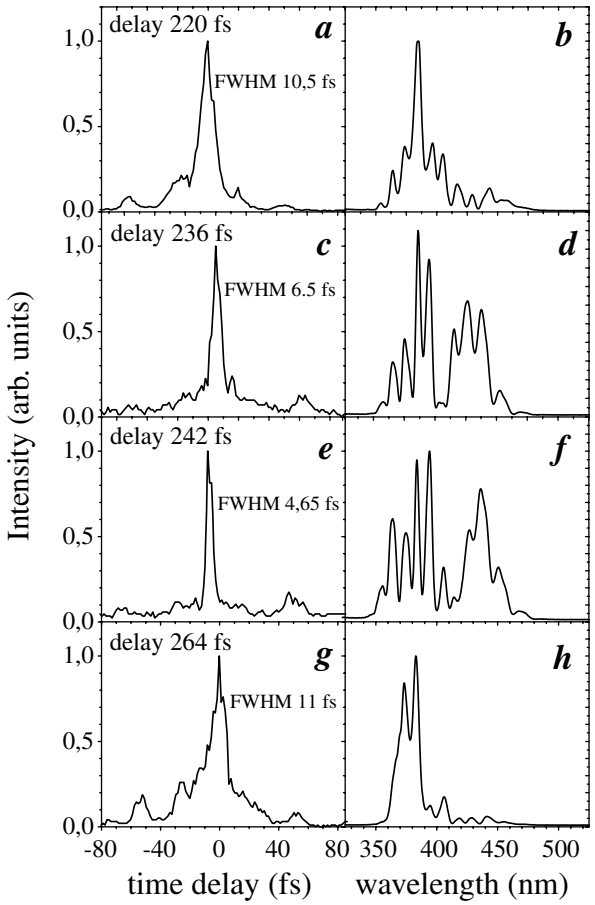

FIG. 3. Self-diffraction autocorrelation traces and corresponding spectra for different delays between the pump and the probe pulses for vibrational excitation in SF_6.

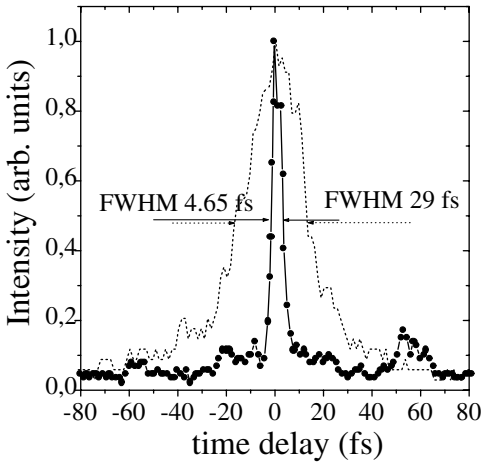

FIG. 4. Self-diffraction autocorrelation traces with (solid line) and without (dotted line) impulsive excitation of SF_6. Deconvoluted pulse duration is 3.8 fs (see text).

are in progress but may include necessary changes or updates in the retrieval program itself. Figure 4 shows the SD-autocorrelation trace of the probe pulse without and with the impulsive excitation for delay of 242 fs, where the probe pulse is compressed with a factor of 6. The deconvolution factor of 1.22 has been found by performing a Fourier transform of the square root of the spectrum depicted in Fig. 3(f). This gives the shortest theoretically possible pulse shape with a FWHM of 3.5 fs. This intensity shape has been used to calculate the self-diffraction ($\chi^{(3)}$) trace, which shows a FWHM of 4.2 fs. The ratio of these two values has been defined as the deconvolution factor. To estimate the influence of higher order phase distortions on the deconvolution factor, the best computer fits with the measured SD-trace have been achieved by different combinations of third and fifth order spectral phases. The calculated deconvolution factors changed in this case by an amount less than 4%. For the autocorrelation trace depicted in Fig. 4 this procedure leads to a deconvoluted pulse width of less than 4 fs (3.8 ± 0.15 fs). The change of the delay within one-half of the vibrational period from the optimum position for pulse shortening lengthens the pulse, and some satellites have appeared as well. This confirms that the phase modulation of the probe pulse changes with a temporal scanning over the time dependent modulation function generated by the molecular vibration. The parameters of the compressed pulses were reproducible within 10% in an experimental time interval of 10–20 min. The measured spectra can be reproduced qualitatively in computer simulations using Eq. (2) for the modeling of the probe pulse field as in [17].

In conclusion, we have demonstrated a new method of single extremely short intense pulse generation using ultrafast molecular phase modulation in a hollow waveguide filled with impulsively excited Raman active gas. For a probe pulse considerably shorter than the period of molecular motion a quasicontinuous spectrum with a width of almost 100 nm has been obtained. A continuous phase control of the second delayed pulse has been performed by changing the delay between the pump and the probe pulses within the period of molecular motion. As a result sub-4-fs pulses with energies of 1.5 μJ at 400 nm were generated.

We acknowledge the support by Deutsche Forschungsgemeinschaft and thank Dr. A. Nazarkin for valuable discussion.

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