

Mirror-dispersion-controlled sub-10-fs optical parametric amplifier in the visible

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Pulses from an optical parametric amplifier in the visible are compressed to sub-10-fs duration by a delay line made exclusively from chirped dielectric mirrors. We employ what we believe are the first ultrabroadband visible chirped mirrors, which provide negative group-delay dispersion up to the blue-green spectral region. The setup is compact and reliable, and we used it to observe, for what is to our knowledge the first time in organic molecules, coherent oscillations with periods as short as 16 fs. © 1999 Optical Society of America
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Recently, optical parametric amplifiers (OPA's) pumped by the second harmonic of a Ti:sapphire laser have shown the capability to generate visible light pulses of sub-10-fs duration. In a noncollinear geometry with a suitable pump-signal angle, wavelength-independent phase matching is achieved in β -barium borate (BBO),¹ which results in ultrabroad amplification bandwidths. Using this concept, several groups of researchers demonstrated visible OPA's with amplified bandwidths in excess of 100 THz, pulse durations well below 10 fs, and microjoule-level energy.²⁻⁶ These systems consist of three components: a white-light seed generation stage, a parametric gain stage, and a pulse compressor. The most critical element is the pulse compressor, which must correct the phase over broad bandwidths and have high throughput. Several compressor schemes have been proposed that included such components as Brewster-cut prisms,^{2,3} thin-prism sequences,⁴ and prism-grating⁵ and prism-chirped-mirror⁶ combinations. The presence of prisms in all configurations makes the compressor more sensitive to alignment, which increases the complexity of the system. Chirped-mirror-only dispersion compensation, previously demonstrated with laser oscillators,⁷ optical parametric oscillators,⁸ and pulse compressors,⁹ has the main advantage of greatly simplifying the system design, allowing for compactness, reproducibility, and insensitivity to misalignment, which are of paramount importance in practical application. Until now chirped mirrors, including those used in the research described in Ref. 6, were designed to provide negative group-delay dispersion (GDD) in the near-infrared and red spectral regions for frequencies lower than 500 THz.

Here we report on a noncollinear visible OPA system with a compressor made exclusively from chirped dielectric mirrors that provides negative GDD over the whole visible range, up to 600 THz (blue-green

spectral region). These mirrors exploit a recently introduced design¹⁰ that allows one to achieve a group delay (GD) that varies nearly linearly with frequency over bandwidths as broad as 200 THz. The resultant system is simple, compact, and reliable and generates sub-10-fs pulses in the visible spectral range.

The noncollinear OPA that we used has been described elsewhere.^{3,4} Briefly, the system is pumped by the second harmonic of a Ti:sapphire laser and seeded by the white-light continuum generated in a 1-mm-thick sapphire plate. Parametric gain is achieved in a single pass through a 1-mm-thick BBO crystal, cut at $\theta = 32^\circ$, by use of type I phase matching. The amplified pulses have $\approx 2\text{-}\mu\text{J}$ energy, peak-to-peak fluctuations of $<7\%$, and TEM₀₀ beam quality. As was shown in Refs. 3 and 4, the bandwidth of the amplified pulses depends on the alignment of the BBO crystal and on the pump-seed angle. For the research reported here we used a pump-seed angle slightly smaller than the optimum, thus allowing us to tune the OPA by varying the pump-seed delay or slightly tilting the BBO crystal. The amplified beam is collimated by a spherical mirror and sent to the chirped mirror compressor and then to a balanced noncollinear autocorrelator by use of silver mirrors and a 20- μm -thick BBO crystal.

The ultrabroadband visible chirped mirror system (VIS-CM) that we used was designed by a semianalytical method.¹⁰ A simple formula permits a starting structure to be determined that leads to the required GD and reflectance characteristics after limited computer optimization. Because the bandwidth constitutes an input parameter in the procedure, the design of ultrabroadband mirrors becomes straightforward. The VIS-CM¹¹ provides high reflectivity [$R > 99\%$; see Fig. 1(a)] over a range of almost 200 THz and an average GDD of -28 fs^2 , with fluctuations of $\pm 14\text{ fs}^2$. In Fig. 1(a) we plot the relationship between calculated

GD and frequency for the VIS-CM and (as squares) the experimental data, measured with a white-light interferometer. The differences between theoretical and experimental data can be attributed to imperfect knowledge of the coating material's dispersion at short wavelengths. We believe that these are the first chirped mirrors to provide negative GDD in the 500–600-THz frequency range: The mirrors used in the research reported in Ref. 6, in fact, have negative GDD only for frequencies lower than 500 THz, and for higher frequencies phase correction requires the use of prisms. The performance of the VIS-CM was compared, in the red spectral region, with that of existing mirror systems designed for hollow fiber pulse compressors⁹ and working in the near-infrared spectral region (NIR-CM). The characteristics of these mirrors, which provide an average GDD of -40 fs^2 from 350 to 500 THz, are shown in Fig. 1(b).

Because of the use of reflecting optics in the system and of a thin sapphire plate, the chirp of the amplified pulses is relatively low. A characterization of the white-light chirp that uses optical Kerr gating³ shows that, from 500 to 750 nm, it nearly coincides with the GD introduced by the sapphire plate; by adding to it the GD introduced by the BBO crystal we obtain an estimate of the GD of the amplified pulses. The GD required from the compressor is shown in Fig. 2: We can see that, over the full OPA gain bandwidth, its frequency dependence deviates from linear behavior because of third-order dispersion. The curve is, however, fairly linear over bandwidths of the order of 100 THz, so it can be reproduced by a sufficient number of bounces on the chirped mirrors. As shown in Fig. 2, good matching in the blue and in the red can be achieved by use of, respectively, 14 and 9 bounces on the VIS-CM; in the red, similar results can be obtained with 6 bounces on the NIR-CM.

Experimentally, optimum compression of the blue-green pulses [see the spectrum in the inset of Fig. 3(a)] was achieved with 18 bounces on the VIS-CM: The autocorrelation [Fig. 3(a)] yields a FWHM pulse duration of 9.5 fs, assuming a sech^2 pulse shape, to be compared with 8.5 fs obtained from a Fourier transform of the pulse spectrum. The red pulses [see the spectrum in the inset of Fig. 3(b)] could be compressed to 8.5-fs duration after 14 bounces on a VIS-CM [solid curve in Fig. 3(b)]; in this case the transform-limited pulse duration was 6.5 fs. For this wavelength range, better results were obtained with the NIR-CM, and a pulse duration of 7.5 fs was measured after 7 bounces [dashed curve in Fig. 3(b)]. The number of bounces is higher than expected from the theoretical GD, in accordance with the somewhat lower experimental values. The measured pulse widths are quite close to the transform-limited values, indicating the high quality of the compressor, and were easily reproducible on a day-to-day basis without any compressor adjustment. The chirped mirror compressor therefore adds reliability as well as compactness to the system and considerably simplifies its operation in ultrafast-spectroscopy experiments.

The system described here was used extensively for ultrafast spectroscopy measurements¹²: We easily

compensated for any additional dispersion met downstream from the OPA, such as from cuvette windows, by increasing the number of bounces on the chirped mirrors. As an example, we report on pump-probe experiments on samples of polydiacetylene, a system previously investigated by use of a high time resolution.^{13,14} Two sub-10-fs pulses (center wavelength of 540 nm) were used to excite and probe the material; after the sample, the probe pulse was spectrally filtered by a 10-nm-bandwidth interference filter at 600 nm. The differential transmission signal, plotted in Fig. 4 as a function of pump-probe delay, at this

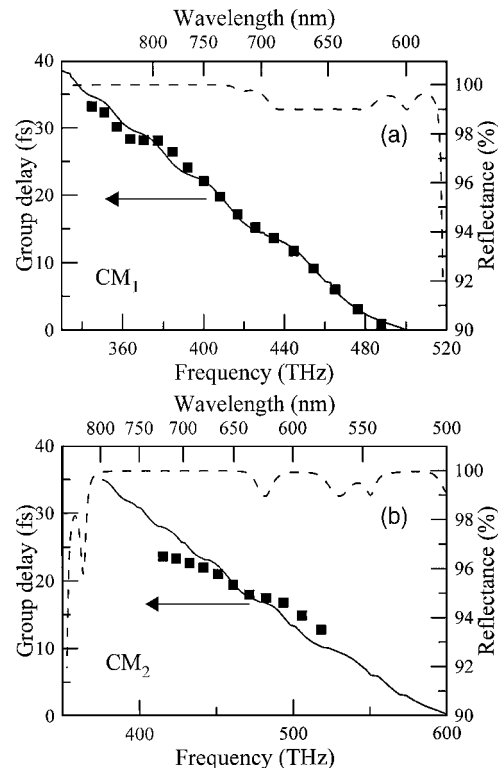


Fig. 1. (a) Dashed (solid) curves, calculated reflectance and (GD) versus frequency for a VIS-CM; filled squares, experimental data obtained by white-light interferometry. (b) Same as (a) but for a NIR-CM.

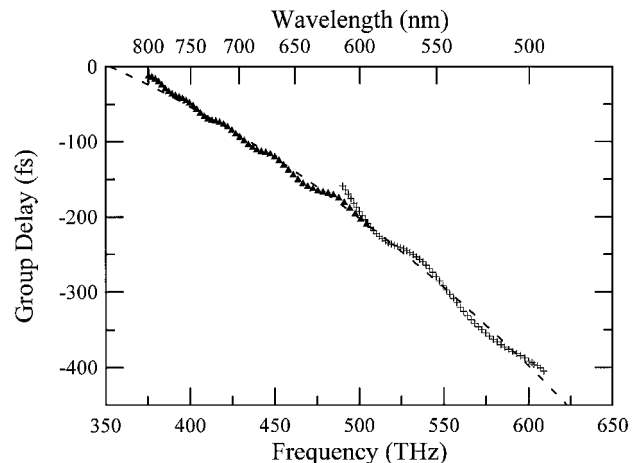


Fig. 2. Dashed curve, desired GD of the compressor versus frequency; crosses (triangles), GD after 14 (9) bounces on a VIS-CM.

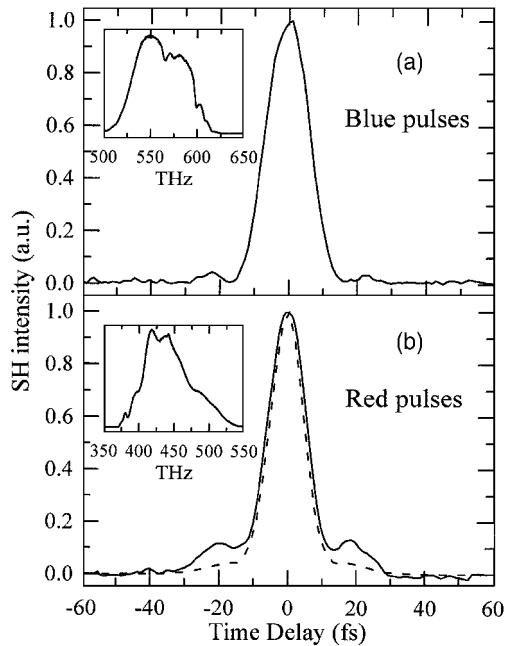


Fig. 3. Autocorrelation traces of (a) the compressed blue pulses (spectrum in the inset) and (b) the compressed red pulses (spectrum in the inset) for a VIS-CM (solid curves) and a NIR-CM (dashed curve). SH, second harmonic.

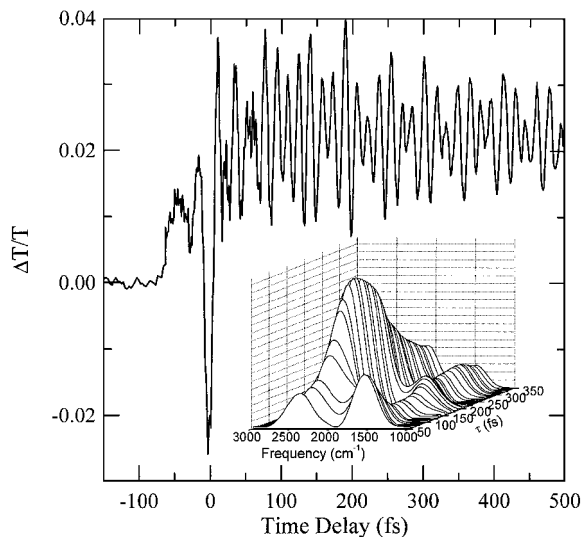


Fig. 4. Differential transmission versus pump-probe delay for a film of polydiacetylene pumped by an ultrashort blue pulse. Inset, sliding window Fourier transform of the oscillatory component of the signal.

wavelength is strongly modulated by a complex oscillatory pattern because of coupling of the optical transition to the $C=C$ (frequency, 1500 cm^{-1} ; period, 22 fs) and $C\equiv C$ (frequency, 2100 cm^{-1} ; period, 16 fs) vibrational modes. The inset of Fig. 4 shows a sliding window Fourier transform¹⁵ of the oscillatory component of the signal, which allows the time evolution of the coupled vibrations to be determined. This analysis shows that the $C\equiv C$ mode becomes dominant ≈ 150 fs after the excitation, strongly supporting the hypothe-

sis¹³ that photoexcitation gives rise to a transient butadienic configuration (with the $C=C$ mode dominant), which is quickly followed (within ≈ 150 fs) by return to a stable acetylenic configuration. These data provide new insight into the photophysics of polydiacetylene and show the potential of our spectroscopic system for investigating real-time evolution of the geometric structure of molecules.

In conclusion, we have demonstrated a compact visible OPA system that uses a dispersive delay line that employs only chirped mirrors. The use of the chirped mirrors considerably simplifies the compression stage and allows sub-10-fs pulses to be generated in a reproducible and reliable manner. Currently available mirrors are designed to correct second-order dispersion and are able to correct the phases of the amplified pulses only over bandwidths of ≈ 100 THz. This research provides a further stimulus for the development of ultrabroadband chirped mirrors, which, if designed to also correct third-order dispersion, will allow the required GD to be achieved over the entire OPA gain bandwidth.

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11. The VIS-CM consists of alternating SiO_2 and TiO_2 layers upon a quartz substrate, with thicknesses (in nanometers), starting from the substrate, of 160/117/102/95/139/62/146/82/123/78/146/72/126/75/136/68/112/58/162/81/123/77/148/70/133/64/113/50/150/64/117/73/126/67/89/45/104/65/107/63/92/36/93/64/113/43/48/65/110/48/26/71/124/25/40/76/114.
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