Ultrafast three-dimensional submicrometer-resolution readout of coherent optical-phonon oscillations with shaped unamplified laser pulses at 20 MHz

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Received October 27, 2011; revised January 28, 2012; accepted January 28, 2012; posted January 30, 2012 (Doc. ID 157236); published April 27, 2012

An ultrafast three-dimensional readout of coherent optical-phonon oscillations from a diamond film is demonstrated using temporally and spectrally shaped ultrashort laser pulses, delivered by a compact, oscillator-only laser system. This system integrates a long-cavity ytterbium-fiber-laser-pumped 30 fs Cr:forsterite oscillator with a photonic-crystal-fiber soliton frequency shifter and a periodically poled lithium niobate spectrum compressor, providing coherent Raman excitation and time-delayed interrogation of optical phonons in diamond at a 20 MHz repetition rate with a submicrometer spatial resolution. © 2012 Optical Society of America

OCIS codes: 190.5560, 190.4380.

Coupling light fields to optical phonons is the key to a high-precision local control over the properties of materials and for the development of innovative technologies of optical data processing. Methods of efficient and selective optical excitation and probing of coherent collective modes in condensed-phase systems are in high demand for material micro- and nanoprocesing [1], ultrafast photonics [2], optical information technologies [3], and studies of complex functions in living systems [4]. Ultra-short laser pulses provide a powerful tool for the coherent excitation of optical phonons in technologically relevant solid-state systems, such as silicon, diamond, and polymer materials [5–9]. Among these materials, diamond is especially important, due to a unique combination of its properties, including a wide band gap, biocompatibility, chemical robustness, high thermal conductivity, and high breakdown voltage, which makes it a promising material for a broad range of advanced optical technologies, including nanoprobe-based biosensing [10], ultrafast nonlinear optics [2,8], and quantum information processing [3].

Here, we demonstrate an ultrafast three-dimensional readout of coherent optical-phonon oscillations from a diamond film using temporally and spectrally shaped ultrashort laser pulses, delivered by a compact, high-repetition-rate oscillator-only laser system. Our approach is based on coherent excitation of Raman-active modes with a frequency $\Omega_p$ by pump and Stokes fields with central frequencies $\omega_1$ and $\omega_2$ chosen in such a way as to meet the condition of a two-photon, Raman-type resonance, $\omega_1 - \omega_2 = \Omega_p$. The third (probe, or interrogating) field with a central frequency $\omega_3$ is then scattered off the coherence induced by the pump and Stokes fields to give rise to the anti-Stokes signal at the central frequency $\omega_a = \omega_1 - \omega_2 + \omega_3$ through coherent anti-Stokes Raman scattering (CARS) [11], thus reading out the information stored in optical-phonon oscillations.

In experiments, we employ a two-color version of CARS with the pump and probe fields delivered by the same light source, implying that $\omega_1 = \omega_3$ and $\omega_a = 2\omega_1 - \omega_2$. Our laser system is based on a home-built ytterbium-fiber-laser-pumped mode-locked Cr:forsterite laser oscillator [12], which delivers laser pulses with a central wavelength of 1.25 $\mu$m and a pulse width of 40 fs. The extended-cavity design of the Cr:forsterite laser is used to increase the output laser energy up to 18 nJ at a pulse repetition rate of 20 MHz. The laser output is launched into a waveguide channel in a periodically poled lithium niobate (PPLN) crystal, which delivers a spectrally compressed second-harmonic output [13,14], needed as a probe pulse. Typically, for input laser pulses with an energy of 10 nJ and a bandwidth of 300 cm$^{-1}$, the waveguide channel with a pitch of 10.9 $\mu$m provides the efficiency of second-harmonic generation (SHG) at the level of 30%, yielding 623 nm pulses with a bandwidth of 60 cm$^{-1}$. These pulses are used as pump and probe fields in the CARS scheme. The fundamental-wavelength output of the PPLN crystal (nonconverted 1.25 $\mu$m radiation) is separated from its second harmonic with a beam splitter and is launched into a photonic-crystal fiber (PCF), designed to provide efficient wavelength conversion of 1.25 $\mu$m laser pulses through soliton self-frequency shift. The PCF delivers tunable ultrashort light pulses within a wavelength range of 1.35–1.80 $\mu$m. These pulses are then frequency-doubled in a 2 mm thick barium borate (BBO) crystal, yielding Stokes pulses for coherent Raman interrogation of optical phonons.

Envelope shaping of pump and probe pulses in our experimental scheme is based on the sensitivity of SHG phase matching to the nonlinear phase shift induced in a nonlinear crystal by the fundamental field. Due to the self-phase modulation of the fundamental field and cross-phase modulation of the second harmonic, the phase mismatch between the fundamental field and its second harmonic become nonuniform over the fundamental pulse [15], giving rise to a double-pulse structure of the second-harmonic output of the PPLN crystal. Light pulses were characterized in our experiments by the interference autocorrelation (IA) and cross-correlation frequency-resolved optical gating (XFROG) techniques. Figures 1(a) and 1(b) show the IA traces representing the 311 nm second-harmonic output of a 0.5 mm thick...
BBO crystal measured as a function of the delay time between two replicas of the second-harmonic output of the PPLN crystal for two different peak powers of 1.25 μm pulses at the input of the PPLN crystal. As can be seen from these measurements, the increase in the peak power of fundamental pulses tends to modify the temporal envelope of the second-harmonic output toward a clearly resolved double-pulse waveform. The best fit for the IA [Fig. 1(b)] and XFROG [the inset to Fig. 1(a)] traces of the second-harmonic PPLN output measured with 8.5 nJ, 1.25 μm pulses at PPLN input is achieved [Fig. 2(a)] with a double-spike pulse shape of the second-harmonic PPLN output, where each individual spike has a Gaussian envelope with an individual pulse width τ₀ = 180 fs, a time interval between the spikes τₚ = 400 fs, and the first-to-second-spike intensity ratio η ≈ 1:8. The ratio η increases with the growth in the peak powers of 1.25 μm pulses at the input of the PPLN crystal.

Coherent Raman excitation of the 1332 cm⁻¹ zone-center Γ(25−) (F₂₂g) symmetry optical phonon in our scheme is provided by one of the two 623 nm, 180 fs pulses generated by the PPLN crystal and the 680 nm, 120 fs, 50 pJ second harmonic of the wavelength-shifted soliton PCF output. The delay time τ₀ between the Stokes and pump pulses (the inset in Fig. 2) is controlled using a tunable optical delay line. The second pulse of the PPLN output serves as a time-delayed probe, providing optical interrogation of coherent phonon oscillations through the generation of the anti-Stokes field centered at 575 nm. The anti-Stokes signal at ωₙ is generated by the interfering fields produced through the ωₙ = 2ω₁ - ω₂ CARS and a nonresonant coherent signal originating from the ωₙ = 2ω₁ - ω₂ four-wave mixing process. Since the nondispersive nonlinear-optical susceptibility responsible for the nonresonant component of the CARS signal translates into a fast decaying nonlinear-optical response function in the time domain, a time-delayed probe is known [10] to efficiently suppress the nonresonant background in CARS. In our experimental scheme, both pump and time-delayed probe pulses are provided by the PPLN crystal, which combines functions of a frequency doubler and pulse shaper, converting Cr:forsterite laser pulses into a double-pulse output [Fig. 2(a)] with a variable intensity ratio η.

The CARS spectra of the zone-center Γ(25−) (F₂₂g) symmetry optical phonon in a 20 μm thick synthetic diamond film are presented in Fig. 2(b). The optical-phonon decay time in diamond films used in our experiments is estimated as τ₂ = 5.3 ps [8]. Therefore, when the Stokes pulse is applied simultaneously with the pump pulse, τₚ = 0, and the probe pulse interrogates coherent oscillations of the optical phonon excited by the pump and Stokes pulses with a delay time of τₚ = 400 fs, the nonresonant part of the CARS signal is strongly suppressed, and the probe field reads out virtually undamped (τₚ ≪ τ₂) oscillations of the optical phonon. This regime of coherent optical phonon interrogation yields the maximum-contrast CARS spectra [curve 1 in Fig. 2(b)]. In the regime where the Stokes—pump delay time τ₀ becomes close to the time interval τₚ between the probe and pump pulses, the second pulse in the pair of spikes delivered by the second-harmonic PPLN output plays the role of both the pump and probe field, as the first, lower-intensity pulse of this pair interacts with the system at negative delay times, having virtually no effect on the coherent Raman response. This regime implements coherent Raman interrogation with an unshaped, single-pulse waveform at ω₁, as opposed to coherent Raman scattering with a double-pulse PPLN output at ω₁ with τₚ ≠ 0 and a variable intensity ratio η. In the scheme with an unshaped, single-pulse waveform at ω₁, the spectra of the CARS signal from the diamond film feature a strong interference of the resonant and nonresonant components [curve 2 in Fig. 2(b)]. The ratio of the resonant CARS signal to the nonresonant background achieved with the temporally shaped, double-pulse output of the PPLN crystal used as pump and probe pulses with τₚ = 0 is 21 times higher [cf. curves 1 and 2 in Fig. 2(b)] than in the case of τₚ = τₚ, which corresponds to CARS with an unshaped pump/probe field.

Due to its nonlinear nature, the coherent Raman process is tightly confined to the laser-beam waist area, which makes CARS a powerful tool for three-dimensional imaging [16]. Here, we show that coherent Raman scattering can selectively interrogate coherent phonon modes in the bulk of solids in three dimensions with a submicrometer resolution in the transverse plane. Figures 3(a) and 3(b) present the maps of the CARS signal measured by scanning a diamond film in the xy plane perpendicular (Fig. 1) to the light beams (the z axis) focused with an NA = 0.65 objective on the film surface [Fig. 3(a)] and 10 μm below the film surface [Fig. 3(b)].
in the regime of maximum CARS contrast, i.e., with \( \tau_d = 0 \). The features observed in these images include local variations in film density (e.g., two bright spots in the lower parts of the maps, submicrometer-scale surface ripples, and larger-scale defects (e.g., the dark spot at the central part of the maps. For comparison, in Fig. 3(c), we present a bright-field microscopy image of the diamond film. In this image, the large-scale defect, seen as a dark spot at the central part of the CARS images appears as a strongly scattering feature in the lower left corner. The contrast, sharpness, and visibility of film ripples on the bright-field microscopy image are much lower than those provided by CARS microscopy with an objective of the same NA [cf. Figs. 3(a) and 3(c)], indicating an enhanced spatial resolution in CARS images. The local variations in film density, seen as bright areas in CARS images [Figs. 3(a) and 3(b)], can be visualized in three dimensions by scanning the light beams with respect to a sample along the \( x, y \), and \( z \) axes, illustrating the capability of coherent Raman scattering of locally reading out the excitation stored in optical phonon modes, converting the data stored in these phonon modes into an optical signal.

The spatial resolution of the coherent Raman interrogation of optical phonons was assessed using a synthetic diamond film sample with submicrometer surface ripples, induced by thermal instabilities building up at the stage of diamond film synthesis. As can be seen from the CARS map of this film [Fig. 3(d)] and a one-dimensional cut of this map [Fig. 3(e)] along the direction \( \xi \) shown by the dashed line in Fig. 3(d), measured with \( \tau_d = 0 \), the coherent Raman technique can resolve submicrometer features on the film surface. The scale of the finest features resolved in these experiments (the feature centered at \( \xi = 8 \mu m \) in Fig. 3(c)) is estimated as \( 0.7 \mu m \).

To summarize, we have demonstrated an ultrafast 3D readout of coherent optical-phonon oscillations from a diamond film using temporally and spectrally shaped ultrashort laser pulses, delivered by a compact, oscillator-only laser system. In a broader context, our experiments demonstrate that simple pulse-shaping functions can be advantageously integrated into a compact platform for coherent Raman applications using a combination of periodically poled nonlinear crystals and specifically designed nonlinear fibers. With regard to the capability of synthesizing broadly tunable optical field waveforms, PPLN pulse shapers demonstrated in this work cannot compete with adaptively controlled spatial light modulators. However, as an ultracompact component of a system for high-spatial-resolution, high-repetition-rate coherent Raman metrology, PPLN pulse shapers integrated with PCFs offer important advantages for microspectroscopy, bioimaging, quantum information processing, as well as time-resolved studies and control of ultrafast molecular vibrations and optical phonons using the coherent Raman effect. The approach demonstrated in this work is by no means limited to the study of optical phonons in solids. The wavelength tunability of the soliton PCF output allows this technique to be applied to a broad variety of Raman resonances in biomolecules, including the \( C=H \) stretching vibration. The work toward integrating the system demonstrated in this work into a compact platform for brain imaging is now in progress.

This research was partially supported by the Seventh European Framework Programme (CROSS TRAP 244068 project) and the Russian Foundation for Basic Research.

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