Conversion of high-power 15-fs visible pulses to the mid infrared

J.-P. Likforman

Steacie Institute of Molecular Sciences, National Research Council of Canada, 100 Sussex Drive, Ottawa K1A 0R6, Canada, and Laboratoire d’Optique Appliquée, Ecole National Supérieure de Techniques Avancées-Ecole Polytechnique–Centre National de la Recherche Scientifique, Unité Mixte de Recherche 7639, F-91761 Palaiseau Cedex, France

M. Mehendale and D. M. Villeneuve

Steacie Institute of Molecular Sciences, National Research Council of Canada, 100 Sussex Drive, Ottawa K1A 0R6, Canada

M. Joffre

Laboratoire d’Optique Appliquée, Ecole National Supérieure de Techniques Avancées-Ecole Polytechnique–Centre National de la Recherche Scientifique, Unité Mixte de Recherche 7639, F-91761 Palaiseau Cedex, France

P. B. Corkum

Steacie Institute of Molecular Sciences, National Research Council of Canada, 100 Sussex Drive, Ottawa K1A 0R6, Canada

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We measure the efficiency of converting high-power 15-fs 0.8-μm pulses to the mid infrared in GaAs and GaSe as well as the pulse duration and the spectrum of the infrared radiation that is produced. Free-carrier production limits the conversion efficiency in GaAs to approximately 5 × 10⁻⁷, allowing us to produce 2.5-pJ, 30-fs pulses spanning the spectral range from 6 to 14 μm. In GaSe we obtain, in a moderately saturated regime, a conversion efficiency of 7.5 × 10⁻⁵, limited by two-photon absorption, allowing us to produce pulses of 100-fs duration containing 10 nJ of energy. © 2001 Optical Society of America

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The unique applications of short infrared pulses will require nonlinearity. We can estimate the intensity needed for nonlinear interactions with quantum wells and dots, for example, by asking when the Rabi frequency ωr equals a typical dephasing time (or laser pulse duration). ωr = μE/ℏ, where E is the peak electric field amplitude, μ = qR/2, q is the electronic charge, and R is the quantum well dimension. Substituting R = 50 Å (1 Å = 0.1 nm) we reach ℏωr = 10–100 meV when E = 5 × 10⁴ – 5 × 10⁷ V/cm, which is equivalent to an intensity of 2.5 × 10⁸–2.5 × 10¹⁰ W/cm². Reaching such high intensities was the initial aim of our experiment. We achieve pulses that are powerful enough to be focused to ~10¹¹ W/cm². Terahertz experiments with Rydberg atoms have shown that high-power pulses are important not only on their own but even more so in conjunction with short visible pulses. A visible pulse excites an electronic wave packet, and terahertz radiation pushes electron wave packets around, leading to quantum manipulation and control.¹

Optical parametric amplification is widely used for generating tunable mid-infrared radiation.²,³ However, because the idler and the signal are generally not phase locked, their relative phase is not fixed, and the infrared electric field oscillation is randomly phased from shot to shot. However, the infrared carrier phase is an important parameter.⁴,⁵ In contrast, nonlinear conversion by difference-frequency generation (including optical rectification) with a single optical pulse produces infrared pulses that have a defined carrier-envelope phase and that also are perfectly phased with respect to the envelope of the pulse from which they are generated. Nonlinear conversion by difference-frequency mixing was used previously to produce low-intensity mid-infrared pulses by use of high-repetition-rate laser systems.⁶–¹¹ To date, the broadest pulse spectrum of which we are aware, extending from 7 to 15 μm, was obtained by use of optical rectification in gallium arsenide (GaAs) of 15-fs pulses delivered by a titanium:sapphire (Ti:S) oscillator.⁷ The infrared energy produced has been small, partly because of the low energy of the input pulse but also because of the small effective length of the conversion process caused by the absorption of the incident visible pulse and the absence of phase matching. Phase-matched difference-frequency mixing inside the laser’s spectral width has been developed with a cavity-dumped Ti:S oscillator and gallium selenide (GaSe) as the nonlinear medium to produce 2.5-pJ, 100-fs pulses.¹²

We used a single, intense ultrashort visible pulse to produce 10-nJ, 100-fs mid-infrared pulses through difference-frequency mixing (phase matching) and 2.5-pJ, 30-fs pulses through optical rectification (no phase matching). We studied the limitations of the frequency-conversion processes for two different nonlinear crystals: GaAs and GaSe. We characterized the infrared pulses in time by bringing together the older technology of semiconductor switching that has been used for femtosecond infrared generation with the newer technology of optical rectification and difference-frequency generation for producing short pulses at a wavelength of 10 μm.⁷,⁸,¹¹ We also characterized the infrared pulses in frequency by using a diffracting Fourier-transform interferometer.¹³

Our home-built laser source consists of a Ti:S oscillator (λ = 0.8 μm) regeneratively amplified at a
repetition rate of 310 Hz to produce 750-\(\mu\)J, 40-fs pulses. A pulse compression system\(^1^4\) in a 50-cm-long hollow core fiber (core diameter, 250 \(\mu\)m) filled with argon gas (pressure, 1 atm) and two pairs of 20\(^\circ\) apex angle fused-silica prisms separated by 8 m produced 140-\(\mu\)J, 15-fs pulses. A half-wave plate placed before the fiber and a Brewster's angle germanium slab following the fiber permitted dispersion-free control of the power in the 15-fs pulse.\(^1^5\)

To produce the infrared pulse, we focused the visible pulse on the nonlinear crystal, using gold-coated spherical mirrors. For our time measurement and saturation studies, the spatial profile at the infrared generator is Gaussian and has a FWHM of 1000 \(\mu\)m. To extract the maximum infrared energy from GaSe we use a smaller focal spot diameter of 300 \(\mu\)m (FWHM). We confirm that self-phase modulation in air did not affect the spectrum and divergence of the pulse. The infrared beam is focused on our HgCdTe detector of spectral range 5.5–14 \(\mu\)m. To prevent saturation of the detector, we used calibrated germanium filters to attenuate the infrared beam.

To determine what limits the energy conversion efficiency in (1T0) GaAs to \(\sim 5 \times 10^{-7}\), we measured the energy of the infrared pulse as function of the energy density of the visible pulse (Fig. 1(a)). We observed a deviation from an expected quadratic law at a pump energy density of 500 \(\mu\)J/cm\(^2\). Because free carriers are produced by the absorbed incident visible pulse, 500 \(\mu\)J/cm\(^2\) corresponds to a carrier density of \(\sim 8 \times 10^{18}\) cm\(^{-3}\) in the first 2 \(\mu\)m of the sample. Therefore, saturation occurs at a carrier density associated with an abrupt fall to zero of the index of refraction (critical density) in the semiconductor for 13-\(\mu\)m radiation. The lower-frequency components of the pulse are then reflected on the plasma layer, reducing the detected signal. The actual spectrum of the infrared pulse should extend well beyond 14 \(\mu\)m, which is the cutoff frequency of our detector.

One-photon absorption of the visible pulse in GaAs is a serious limitation for producing intense infrared pulses. Starting with a pump pulse at longer wavelength (below the bandgap) should increase the conversion efficiency. GaSe has a similar second-order nonlinear coefficient to GaAs but a bandgap of 2 eV, so single-photon transitions are not possible. Furthermore, phase-matched difference-frequency mixing is possible in GaSe. We generated infrared radiation in a 1-mm-thick GaSe crystal cut at 0\(^\circ\). Type I phase matching was achieved for an external angle between the input beam and the crystal surface of \(\sim 45^\circ\). The maximum infrared energy from GaSe was obtained when negative linear chirp was introduced in the pump pulse. A factor-of-2 gain in pulse energy was then achieved. This is in qualitative agreement with a previous measurement of infrared pulses generated with a Ti:S oscillator.\(^1^1\) Under these conditions we produced 50 nJ of infrared energy by using a 140-\(\mu\)J visible pump pulse, giving a maximum conversion efficiency of \(3.6 \times 10^{-4}\). In Fig. 1(b) we plot the infrared pulse energy as function of the visible pulse energy density. Saturation starts at a fluence of 2 mJ/cm\(^2\). We confirmed that this is the intensity where two-photon absorption becomes important\(^1^6\) be measuring the visible pulse transmission through the nonlinear crystal.

The spectrum of the infrared pulses was measured with a temporal interferometry technique developed in Refs. 7 and 13. Two infrared pulses were generated from two noncollinear nonoverlapped visible pulses and observed through an aperture. We then recorded the temporal interferogram (Fourier transform of the spectrum) by varying the time delay between the two visible pulses. Spectra of the infrared pulses generated in GaAs and GaSe are shown in Fig. 2. In the case of GaAs the spectrum is extremely large and extends from 5.5 to 14 \(\mu\)m, limited only by the bandwidth of our detector. For GaSe, the spectrum is narrower owing to selection by the phase-matching conditions.

The pulse duration is measured in a pump–probe experiment with a bulk germanium (Ge) sample. The intensity of the infrared probe transmitted through the sample is plotted in Fig. 3 as function of time delay with the visible pump pulse. The visible beam

![Fig. 1. (a) Infrared intensity (solid curve) obtained in GaAs as a function of the incident visible pulse energy density. Dotted curve, expected quadratic law in the absence of saturation. (b) Same as for (a) but for an infrared pulse generated in GaSe.](image)

![Fig. 2. Infrared spectrum of pulses generated in GaAs or GaSe. Inset, visible pulse intensity spectrum.](image)
creates an electron–hole plasma in a very small thickness (~0.2 μm). For the measurement we used a pump fluence of 1 mJ/cm², resulting in a carrier density of ~10²⁰ cm⁻³. The pump beam induces a change in the index of refraction of Ge large enough for the plasma layer to become reflective for 10-μm radiation. The intensity-dependent index of refraction of Ge can be calculated by use of Drude model for the dielectric constant. We calculated the transmission change for the infrared pulse by using refraction laws. Figure 3 (solid curves) show the experimental results obtained for an infrared pulse generated in GaAs [Fig. 3(a)] above its saturation fluence and in GaSe [Fig. 3(b)] generated with 5 mJ/cm². In these experimental conditions the conversion efficiency was 7.5 × 10⁻⁵, allowing us to produce 10-nJ pulses. The dotted curves show the calculated transmitted signal for a 30-fs FWHM Gaussian infrared pulse for GaAs and a 100-fs FWHM pulse in GaSe. We estimate an error of ±10% in the measurement of the pulse duration. Our results for GaAs and GaSe show some deviation between experiment and calculation. For GaAs at positive delays (infrared arrives first), we always observe a small increase in transmission, followed by a decrease. Although the decrease can be caused by an infrared prepulse or a visible postpulse, the increase can be caused only by carrier dynamics. In GaSe we find a discrepancy at negative delays. Inasmuch as we do not observe the same effect in our GaAs results, the increase cannot be due to a prepulse in our 0.8-μm pulse. We must conclude that we produce a postinfrared pulse. Although the origin of such a pulse remains unclear, this result is consistent with the infrared spectrum observed in Fig. 2, which exhibits the spectral oscillations that are characteristic of two pulses separated by ~500 fs. Other measurement methods such as intensity autocorrelations are highly complex for a single-period pulse. It is hard to obtain a large phase-matching bandwidth and also to separate the fundamental (or single-photon process) from its second harmonic (or two-photon process).

Although we have achieved high-power infrared pulses, our results do not approach a limit. Simply scaling our results to higher intensities and larger apertures means that 10–100 times more intense infrared pulses are achievable. Furthermore, we may be able to increase the conversion efficiency itself. For example, reducing the visible intensity inside the infrared generator by chirping the visible pulse would, similarly to chirped pulse amplification, reduce two-photon absorption.

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