Generation of infrared supercontinuum covering 3–14 μm in dielectrics and semiconductors

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Infrared supercontinua spanning the range 3–14 μm were observed when an intense pulse generated from a CO_2 laser was passed into GaAs, AgBr, ZnSe, and CdS crystals. These supercontinua have been qualitatively compared with theoretical predictions.

A fundamental and long-standing problem in photonics is the development of a laser source capable of emitting light covering a wide spectral range. Such a laser source would have numerous applications from ranging to imaging.[1] More than 15 years ago, the first white-light supercontinuum was observed by Alfano and Shapiro[2] when a 1.06- or 0.53-μm picosecond pulse propagated through condensed matter. Since that time, there have been numerous experimental[3–7] and theoretical[8–10] studies. Supercontinua have been extensively used in time-resolved spectroscopy. The spectral broadening so far realized covers the near-UV (0.19 μm) (Ref. 4) to the near-IR (2 μm) (Ref. 3) spectral region. In this Letter, we report the first observation to our knowledge of a supercontinuum spanning the 3–14-μm wavelength region.

The experimental arrangement is shown in Fig. 1. It consists of a picosecond CO_2 laser, appropriate filters, focusing and recollimating optics, a spectrometer, and an IR detection system. The design and performance of the gigawatt picosecond CO laser system have been described in detail by Corkum.[11,12] It has a near-transform-limited bandwidth of about 0.04 μm for a 2.5-psec pulse. For this experiment, the laser was operated at 9.3 μm with pulse durations of either 2.5 or 8 psec. When required, the incident beam could be attenuated with CaF_2 filters (ND). The CaF_2 filters, together with a long-pass filter (BF1) with a cutoff wavelength of 8.6 μm, ensured that there were no Raman or harmonic components on the incident beam. The full energy incident upon the nonlinear sample was 600 ± 200 μJ for both the 2.5- and the 8-psec-duration pulses. The incident beam was focused by an R = 1 m concave radius-of-curvature Au-coated mirror to an estimated spot size of 500 μm. This corresponds to a maximum power density of $\sim 10^{11}$ W/cm.^{2}

Measurements were made with the aid of two spectrally selective devices. A Jarrell-Ash spectrometer, used with a spectral resolution of $\sim$0.1 μm provided detailed spectral information. However, further suppression of the laser line was required. This was accomplished by filters BF2. For the anti-Stokes measurements BF2 was a MgF_2 plate; for the Stokes measurements BF2 consisted of two long-pass filters with cutoff wavelengths of λ = 10.58 μm and λ = 10.94 μm. The input signal level was detected on a fast pyroelectric detector (Molectron P5-00). The supercontinuum was monitored on an Infrared Associates 5-MHz HgCdTe detector on the Stokes side and a 150-MHz Au-doped Ge detector on the anti-Stokes side. Both signals were displayed on a Tektronix 7834 storage oscilloscope.

Nine different types of dielectric and semiconductor were tested. Sample crystals were purchased from Janos (KBr, KCl, NaCl), Crystal Specialties (GaAs),
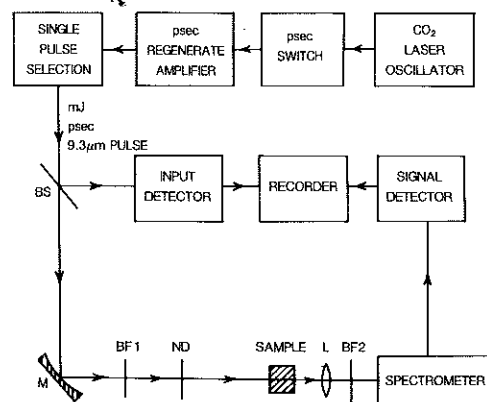

Fig. 1. Schematic of the ultrafast IR supercontinuum experiment. BS, beam splitter; BF1, bandpass filters to remove input noise; BF2, bandpass filters to remove laser line; M, focusing optics; L, signal-collection optics; ND, input power-attenuation filters.
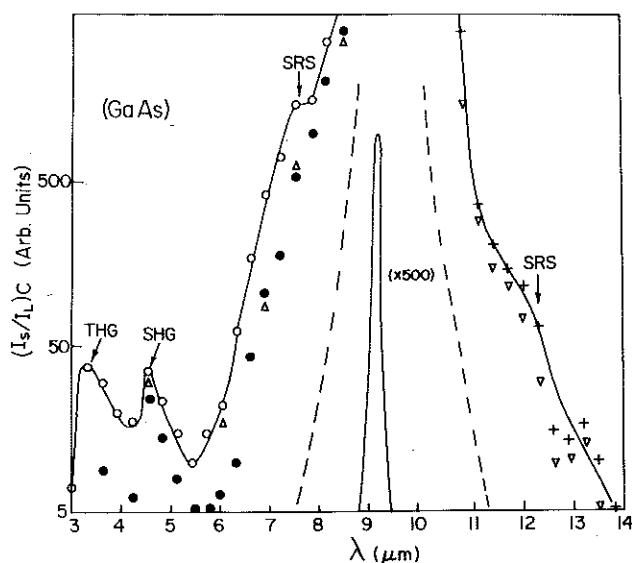
Fig. 2. Supercontinuum spectra from a 6-cm-long Cr-doped GaAs crystal. O 2.5-psec pulse duration, full power; △, 2.5-psec duration, half power; ●, 8-psec pulse duration, full power; +, 8-psec pulse duration, half power; ▽, 8-psec pulse duration, quarter power. SRS, stimulated Raman scattering; I_s, signal energy; I_L, relative input laser energy; C, correction factor for the spectral sensitivity variation of filters, grating, and detectors. The solid lines are guides. The dashed lines are the noise levels. The laser pulse displayed was obtained when sample was removed and attenuated roughly 500 times. Each data point corresponds to an average of at least three shots. The standard deviation of each point is ±10%. The lower excitation laser power used in the Stokes side was due to the extra filter added in the experiment to block out noise. The 3-μm signal could be a combination of the harmonic frequency broadening, the primary continuum extension, and other mixing processes.

II-VI (Ge), Buck Scientific (AgBr), Infrared Optics (ZnSe, CdS), and Microwave Associates (GaAs) and used without further processing. These samples were selected so that the absorption losses were small throughout the 2–14-μm wavelength region.

Spectral broadening was observed from Cr-doped or undoped semi-insulating GaAs, CdS, ZnSe, and AgBr.

With the present experimental arrangements, no measurable broadening was observed with NaCl, KCl, KBr, and Ge crystals. Spectral broadening in Ge has been reported because of rapid plasma production when viewed with greater spectral sensitivity.[11] Visible plasma and damage tracks were observed in the NaCl, KCl, and KBr crystals.

The supercontinuum spectra measured from a 6-cm-long Cr-doped GaAs and a 3.8-cm-long AgBr crystal are plotted in Figs. 2 and 3, respectively, for different laser pulse durations and intensities. The signals were normalized for the input laser energy and the spectral sensitivity of filters, grating, and detectors. Each point represents the average of three shots. The salient feature of curves displayed in Figs. 2 and 3 is that the spectral broadening spans the wavelength regions from 3 to 14 μm. The wave-number spread on the anti-Stokes side is much greater than on the Stokes side.

The spectral broadening mechanism of the supercontinuum can originate from several nonlinear optical processes. These include self-phase modulation,[2–4,6,7] four-wave parametric effect,[2,13] high-order harmonic generation,[14] and stimulated Raman scattering.[15,16] In Fig. 2 the supercontinuum from the GaAs has two small peaks at 4.5 and 3.3 μm. These arise from the second-harmonic generation (SHG) and third-harmonic generation (THG), respectively. Small plateaus are located at 7.5 and 12 μm. These arise from the first-order anti-Stokes and Stokes simulated Raman scattering combined with self-phase modulation (SPM) about these wavelengths. The SPM is attributed to an electronic mechanism.

A theoretical analysis,[8–10] which assumes that SPM and four-wave mixing are the dominant nonlinear mechanisms, predicts that the intensity and spectral broadening of the supercontinuum is a function of the parameter

$$S = n_2 |E|^2 z / c\tau, \tag{1}$$

where n_2 is the nonlinear index of refraction, E is the rms electric field of the laser pulse, z is the sample length, and τ is the pulse duration.

Qualitatively, the theory indicates that short high-power pulses should have a higher conversion efficiency into the continuum and a broader spectrum than the long, low-power pulses. It also predicts that for broad spectra ($S > 0.2$) the continuum should experience a larger wave-number shift to the blue than to the red.

From data displayed in Fig. 2, we find a maximum anti-Stokes spectral broadening of $\Delta\omega_a = 793$ cm^{-1}. Including SHG and THG, it spans to 2000 cm^{-1}. On the Stokes side we observe $\Delta\omega_s = 360$ cm^{-1}, yielding a value of $\Delta\omega_a/\Delta\omega_s \sim 2.2$. For AgBr we observe from Fig. 3 that $\Delta\omega_a = 743$ cm^{-1} and $\Delta\omega^s = 242$ cm^{-1}, yielding $\Delta\omega_a/\Delta\omega_s \sim 3$. Δw_a and Δw_s are defined to be the anti-
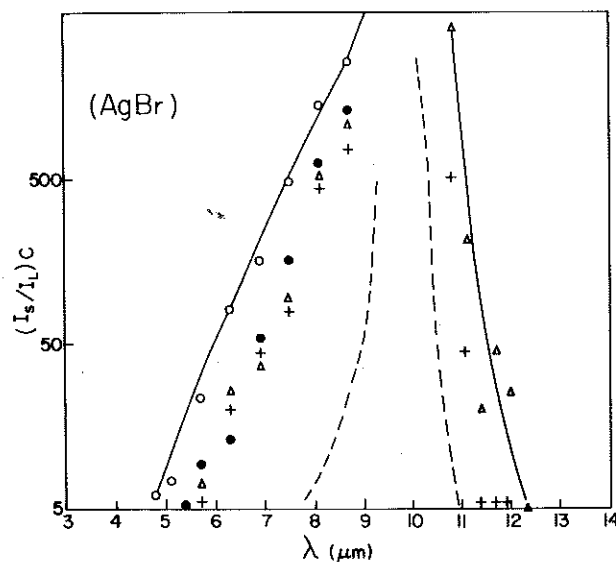

Fig. 3. Supercontinuum spectra from a 3.8-cm-long AgBr crystal. O, 2.5-psec duration, full power; ●, 2.5-psec duration, half power; △, 8-psec duration, full power; +, 8-psec duration, half power.

Stokes and Stokes frequency spreads from 9.3 μm to the furthest detectable wavelengths, respectively.

Any detailed comparison of theory and experiment is beyond the scope of this Letter. A comprehensive theory would have to include contributions that are due to group-velocity dispersion because we find experimentally that our 6-cm-long Cr-doped GaAs crystal broadens a low-power pulse ~5 psec because of linear dispersion. It would also have to include harmonic generation and stimulated Raman scattering. In addition, as noted on the figures, the Stokes and anti-Stokes measurements were performed with different combinations of intensities and pulse widths. Still, both Fig. 2 (without the harmonic components) and Fig. 3 show some wavelength symmetry about the laser line. This indicates the asymmetry in wave numbers. This is a signature of SPM. If we use a value of $n_2 = 10^{-20}$ (mks) (Ref. 17) for GaAs, a pulse duration of 8 psec, $Z = 6$ cm, and $E = 1.5 \times 10^9$ V/m, we obtain a value of $S = 0.56$. From Ref. 8, the theoretical value of $\Delta\omega_a / \Delta\omega_s = 2.8$. This is in qualitative agreement with the experimental results. In this regard, it is interesting to note that in scaling from visible experiments, n_2 for GaAs is approximately 2 orders of magnitude greater than that for quartz or water. We find that the intensity of the incident picosecond pulse can be correspondingly decreased by approximately 2 orders of magnitude by using semiconductors with large nonlinearity.

In conclusion, we have demonstrated the generation of supercontinua spanning the infrared from 3 to 14 μm. By using high-power picosecond 5-μm pulses recently produced by SHG of CO_2 radiation,[18] it seems clear that supercontinua will soon be available at any wavelength range from 0.9 to 14 μm.

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