Upconversion linearity in proustite for short 10.6-μm pulse diagnostics

P. A. Jaanimagi and M. C. Richardson

Division of Physics, National Research Council of Canada, Ottawa K1A 0R6, Canada

N. R. Isenor

Department of Physics, University of Waterloo, Waterloo N2L 3G1, Canada

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Infrared upconversion is the only currently available diagnostic technique for 10-μm CO_2 laser pulses that provides both linearity and picosecond time-resolution capability. We report an investigation of the linearity for upconversion of nanosecond CO_2-laser pulses in proustite pumped by a Nd:YAG laser. This was examined over three orders of magnitude of infrared intensity, and peak pump quantum-conversion efficiencies of 30% were obtained.

Infrared upconversion has become a widely accepted technique, with many applications in laser-pulse diagnosis, infrared imaging, low-level signal processing, and infrared spectroscopy of astronomical sources. It has considerable potential in studies of ultrashort CO_2-laser pulses, such as those utilized in laser-fusion investigations, primarily because it offers a sensitive linear detection system, which, with the incorporation of picosecond chronography, provides a detection bandwidth far in excess of that of other transient infrared recording systems. However, in order to be considered an unambiguous infrared measurement technique, it should maintain linear conversion over a wide range of infrared intensity. An examination of the limitations of the linearity of the upconversion process predicts saturation of the conversion efficiency through depletion of the pump radiation. Additional mechanisms that can affect linearity include optical breakdown and other absorptively induced effects. These result in a deviation from linearity at high conversion efficiencies, thus limiting the useful dynamic range for diagnosing the shape of ultrashort CO_2-laser-pulse phenomena. This saturation of the sum-frequency output intensity has been experimentally demonstrated in the present investigation.

Although a large number of nonlinear crystals have been utilized to upconvert 10-μm radiation,[1] the most commonly used material has been proustite (Ag_3AsS_3),[2] in which quantum-conversion efficiencies as high as 11% have been reported.[3] However, to date, no detailed study of the linearity of upconversion in proustite has been reported. An investigation of 10.6-μm radiation upconversion into the visible range in the crystal $AgGaS_2$ recently claimed linear quantum-conversion efficiencies up to 40%,[4] from 10 to 0.57 μm.

In this Letter we report an investigation of the linearity of upconversion of nanosecond 10.6-μm pulses in proustite using a pulsed Nd:YAG (1.06-μm) pump laser with a view to the utilization of this technique in schemes for the precise analysis of ultrafast infrared radiation. Whereas previous investigations of upconversion have concentrated on the maximum conversion efficiency of the low-frequency signal, here the emphasis is on maximizing the usable operating range over which linear upconversion is obtained for a given infrared signal. This usable operating range is limited at one extreme by the minimum upconverted signal detectable by conventional picosecond streak cameras and at the other extreme by nonlinearities that are due to pump depletion and other effects. The region of nonlinearity has been investigated in which peak quantum-conversion efficiencies from 1.06 to 0.967 μm of ~30% have been obtained for 10.6-μm input radiation.

The principal source of nonlinearity in an upconversion scheme is from the depletion of photons in either the pump or infrared beams. The analytic solution from three-plane-wave interaction theory[5] for perfect phase matching in a lossless medium, assuming I_1 constant, yields the relations,

$$I_3(z) = I_2(0)(\omega_3/\omega_2)\sin^2(\pi z/l), \tag{1}$$

$$I_2(z) = I_2(0)\cos^2(\pi z/l), \tag{2}$$

where z is the distance traveled in the crystal and l is the interaction length given by

$$l = \frac{2\pi}{d_{\text{eff}}}\left(\frac{n_1 n_2 n_3}{2\omega_2\omega_3}\right)^{1/2}(c\epsilon_0)^{3/2} I_1^{-1/2}, \tag{3}$$

I_1, I_2, and I_3 being intensities in (W cm^{-2}) of the input (10.6-μm), pump (1.06-μm), and upconverted signals, respectively, and ω_1, ω_2, ω_3 and n_1, n_2, n_3, their corresponding angular frequencies and indices of refraction in proustite. d_{eff} is the effective nonlinear susceptibility given by $d_{\text{eff}} = d_{22}\cos^2\theta_{\text{PM}}$, where $d_{22} = 2 \times 10^{-22}$ (MKS units) for proustite, and the phase-matching angle $\theta_{\text{PM}} = 20°$.

To ensure linear conversion, I_2 must remain relatively independent of z, or $\cos^2(\pi z/l) \sim 1$, $\pi L/l \ll 1$, where L is the length of the crystal. If this is satisfied, then the

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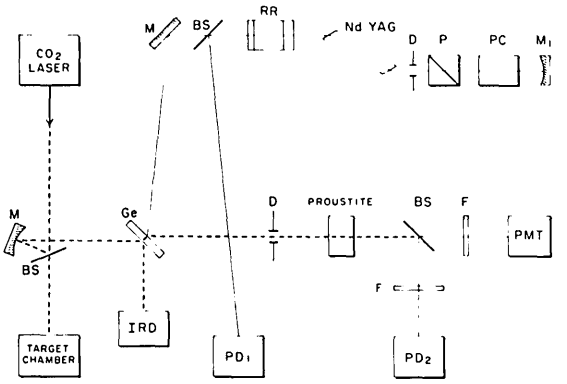

Fig. 1. Experimental configuration for short 10-μm laser-pulse upconversion. PC, Pockels cell; P, polarizer; RR, resonant reflector; D, diaphragm; M, M_1, fully reflecting mirrors; BS, beam splitter; PD_1, PD_2, photodiode; IRD, infrared detector; PMT, photomultiplier; F, 0.96- or 1.06-μm interference filter.

term $\sin(\pi z/l)$ in Eq. (1) may be replaced by its argument, $(\pi z/l)$. Then the upconverted signal I_3 is linear with I_1 and I_2 as

$$I_3(L) = \frac{(\omega_3 d_{\text{eff}} L)^2}{2n_1 n_2 n_3} (c\epsilon_0)^{-3} I_1(0) I_2(0). \quad (4)$$

By expanding the sine function in powers of its argument, the nonlinear dependence of $I_3(L)/I_2(0)$ due to depletion can easily be characterized as

$$I_3(L)/I_2(0) = \frac{(\omega_3 d_{\text{eff}} L)^2}{2n_1 n_2 n_3} (c\epsilon_0)^{-3} I_1(0) \times \left[1 - \frac{1}{3}\left(\frac{\pi L}{S}\right) I_1(0) + \frac{2}{45}\left(\frac{\pi L}{S}\right)^4 I_1^2(0) - \ldots\right], \quad (5)$$

where

$$S = \frac{2\pi}{d_{\text{eff}}} \left(\frac{n_1 n_2 n_3}{2\omega_2 \omega_3}\right) (c\epsilon_0)^{3/2}.$$

For this study, the master oscillator of the COCO-II CO_2-laser system[6] was used. This actively mode-locked, ultraviolet, preionized laser[7] produces a short train of 1.0-nsec-duration (FWHM) pulses, each separated by 25 nsec, oscillating on the $P(20)$ transition of the 10.4-μm CO_2-laser band. The experimental configuration used in investigating the linearity of upconversion is shown in Fig. 1. A Pockels-cell Q-switched Nd:YAG laser, oscillating within a single longitudinal and single transverse TEM_{00} mode, provided unfocused pump intensities of up to 1 MW/cm^2 in a 2.5-mm-diameter beam with a Gaussian pulse duration of 50 nsec (FWHM). This laser was synchronized with the output of the CO_2 laser through the triggering system of the COCO-II laser. The mixing crystal had a 1-cm-square cross section, was 0.5 cm long, and was cut with crystal axes oriented for type-II phase matching. The two laser outputs were orthogonally polarized and, with the utilization of a Ge reflector, were arranged to propagate collinearly through the proustite crystal at an angle of 20° to the optic axis. The resulting sum frequency was isolated with the aid of a 100-Å bandwidth interference filter centered at 0.967 μm and detected with a fast photomultiplier (RCA C70102B), a photodiode (ITT FW127A), or a picosecond streak camera,[8] all employing S-1 response photocathodes. The input 1.06-μm radiation and the CO_2-laser radiation were monitored by a calibrated photodiode and a calibrated photon drag detector, respectively. The cross section of each beam at the proustite crystal was carefully measured and complete overlap ensured by maintaining the CO_2 laser beam cross section significantly greater than that of the Nd:YAG laser.

Upconversion linearity measurements were made by varying the intensity of the CO_2-laser pulse with the aid of calibrated Mylar attenuators. The full CO_2-laser pulse train was used to provide a maximum number of data points per shot by temporally correlating at the crystal the pulse waveforms of the CO_2 laser, the Nd:YAG laser, and the upconverted signal recorded with photomultiplier. After the difference in beam cross sections and the reflection losses of the various optical elements, including the crystal, were accounted for, the results were plotted, along with the linear and nonlinear dependences given by Eqs. (4) and (5), respectively, in Fig. 2. This shows the variation of the sum-frequency signal, normalized to the incident 1.06-μm intensity, as a function of the fraction of the CO_2-laser intensity interrogated by the Nd:YAG beam in the proustite crystal. Each data point represents the average of 4–8 individual measurements. Peak conversion efficiencies of 30% were obtained at a CO_2 intensity of ~14 MW/cm^2.

It can be seen that the conversion efficiency remains linear up to CO_2 intensity levels of a few MW/cm^2, in agreement with the theory for pump depletion, with a rolloff at higher input levels. However, the falloff in conversion efficiency is greater than predicted. This discrepancy may possibly be due to the gradual onset of other mechanisms, such as optical breakdown,[9] two-photon absorption,[10] or absorptively induced index-of-refraction changes causing phase mismatch.

The quantum-conversion efficiency (10.6 to 0.967 μm) at the peak levels was ~0.2%, and thus I_1 remains unaffected by the interaction.

The depletion of the 1.06-μm radiation by the upconverted signal at high CO_2-laser intensities is clearly

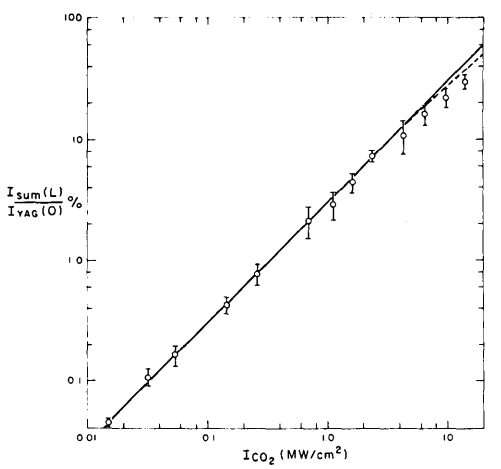

Fig. 2. Upconversion efficiency as a function of 10.6-μm pulse intensity. Linear conversion (solid line), theoretical nonlinearity (dashed line).

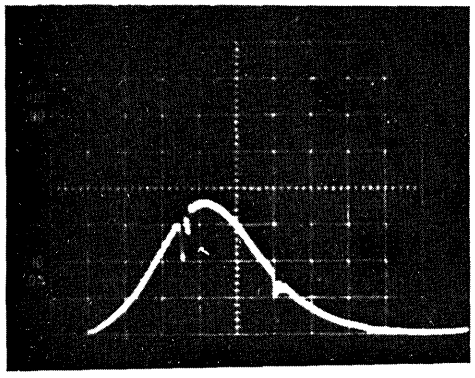

Fig. 3. Depletion of the 1.06-μm pump pulse by the upconversion process. Time scale, 10 nsec div^{-1}.

illustrated in Fig. 3. This figure shows an oscillogram of the 1.06-μm radiation transmitted by the proustite crystal as recorded by a fast photodiode through an interference filter of 100-Å bandwidth. These measurements also provided direct corroboration of the conversion efficiencies deduced from the calibrated monitors.

In summary, the present work has related the linearity of upconversion and its limitations at high efficiencies with the simple theoretical dependence resulting from pump depletion. For application to an ultrafast infrared diagnostic scheme, it is clear that the range of linearity surpasses the dynamic range of current streak cameras[11] operating in the picosecond domain. The measurement of ~70-psec, 10.6-μm pulses has already been reported with a system having 40-psec resolution,[12] and, in a separate report, we shall describe a system having temporal resolution of ~5 psec at present being utilized to diagnose high-power nanosecond pulses used in laser-produced plasma studies.[8]

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