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GENERATION OF INTENSE TUNABLE PICOSECOND PULSES IN THE
FAR-INFRARED

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Abstract

Continuously tunable far-infrared (FIR) picosecond pulses have been generated from 20-200 cm^{-1} through the parametric interaction of two visible pulses. Pulse energies were generated up to 3 nJ, which are sufficient for time-resolved spectroscopy. The quantum efficiency of 0.1-0.3% was surprisingly constant over this frequency range considering the large variations in FIR absorption, diffraction, and phonon resonance enhancement. The depletion of the visible pulse and the dependence of the FIR energy on pump power were typical of a saturated process. However, the FIR energy showed unexpectedly large pulse-to-pulse fluctuations. Understanding the origin of these fluctuations may lead to substantial improvements in the average efficiency.

The investigation of dynamical processes in the far-infrared (FIR) has been seriously limited by the lack of intense, tunable, narrow-band picosecond sources. Optical rectification has been used to generate broadband picosecond¹ and femtosecond² pulses. Recently there has been interest in the generation of picosecond pulses from free electron lasers,³ but success has not yet been demonstrated in the FIR. A variety of techniques have been successful in generating longer FIR pulses.^{4,5} In particular, tunable nanosecond FIR pulses have been generated by difference frequency mixing of two visible dye lasers.^{6,7} The recent development of high-power, picosecond dye lasers has allowed the extension of this technique to the picosecond domain. This letter reports the generation of high-intensity, narrow-bandwidth, picosecond FIR pulses by this technique.

In these experiments, two visible pulses, the first 1-2 ps long and the second 5 ns long, were mixed in a LiNbO_3 crystal to generate FIR pulses at a 10 Hz repetition rate. Since mixing only occurred while both pulses were present, picosecond FIR was generated. The 0.2 mJ picosecond pulses at 589 nm were generated by an amplified, synchronously-pumped dye laser.⁸ A commercial YAG-pumped dye laser produced the 20 mJ nanosecond pulses, which were tuned from 590-596 nm. The pulse-to-pulse energy fluctuations were $\pm 10\%$ for the picosecond pulses and $\pm 6\%$ for the nanosecond pulses. A single doubled-YAG beam was split to pump both the nanosecond dye laser and the picosecond amplifier, thus insuring

synchronization of the two pulses. The two beams were superimposed in the LiNbO_3 crystal, and focused slightly to give spot sizes of 0.6 mm and 1.2 mm for the picosecond and nanosecond beams respectively. Smaller spot sizes caused crystal damage. Note that the instantaneous nanosecond intensity (350 MW/cm^2) is much lower than the picosecond intensity (70 GW/cm^2).

Non-collinear phase-matching was achieved by the method of Yang, et al.⁶ The two visible beams, as well as the FIR, were polarized along the optic axis of the LiNbO_3 to utilize the largest non-linear coefficient.⁹ Since the FIR spectral band is below the optical phonon modes of the crystal, the FIR refractive index¹⁰ ($n = 5.5$) is much larger than the visible index ($n = 2.3$). Therefore the phase-matching can be accomplished geometrically, without using birefringence. Tuning was achieved by changing both the angle between the visible beams (from 5 to 50 mrad) and the frequency of the nanosecond laser. An advantage of the phase-matching scheme used here over collinear phase matching is that the direction of the optic axis does not have to be changed as the FIR is tuned. Thus only one crystal was needed for the entire tuning range. Because the FIR was produced at a 68 degree angle from the visible beams, a corner of the 4 mm cubic crystal was removed to avoid total internal reflection and allow the FIR to exit.⁶

The FIR was produced with a significant divergence due to the ~1 mrad convergence of the visible beams, the diffraction of the FIR, and the bandwidth of the picosecond pulse. This divergence was increased by refraction at the crystal surface to > 1 srad. In order to collect this large solid angle, the small end of a Winston concentrator¹¹ was placed in contact with the exit face of the crystal. The concentrator collected and collimated the emerging FIR pulse which then passed through frequency band limiting filters to a composite bolometer operated at 1.5 K. The bolometer was calibrated using blackbody sources at 77 and 300 K. A step-scanned Michelson Fourier spectrometer was used to measure the spectrum of the FIR pulses.

Figure 1 shows the median collected FIR energy at frequencies from 20-200 cm^{-1} . The energies were typically 3 nJ per pulse, which gave detector signals which were several orders of magnitude larger than detector noise. The quantum efficiency for conversion of photons from the visible picosecond pulse to detected FIR photons varied from 0.1% to 0.3%. Surprisingly, there was no strong trend in the energy or the quantum efficiency as the frequency changed by a factor of ten. The variation in the amount of diffractive loss in the FIR should be large over this range. Furthermore, an optical phonon mode in LiNbO_3 at 252 cm^{-1} would be expected to strongly influence the quantum efficiency through changes in the FIR absorption and phonon resonance enhancement.^{12,13}

The spectrum in Fig. 2 shows that the FIR was generated in a narrow bandwidth at the frequency difference of the visible pulses. The FIR bandwidth of $<10 \text{ cm}^{-1}$ was similar to that of the visible picosecond pulses. This bandwidth is consistent with a 1 ps FIR pulse. Since the crystal face is cut perpendicular to the FIR wavefront, the time required for the interaction region to propagate through the crystal does not broaden the FIR pulse. However, consideration of the transverse dimensions of the interaction region suggests that the actual FIR pulse length was closer to 10 ps.

The simple theory of parametric mixing predicts that photons from the high frequency picosecond pulse are split into photons at both the lower, nanosecond frequency and into FIR photons.¹⁴ As long as the nanosecond intensity is low relative to the picosecond intensity, the FIR energy will depend exponentially on the picosecond intensity and linearly on the nanosecond intensity. If the mixing continues until the picosecond pulse is significantly converted, the process will saturate. The FIR energy will then depend linearly on the picosecond intensity and will be insensitive to the nanosecond intensity.

Experimentally, a small amount of FIR was generated, even when only the the picosecond pulse entered the crystal. At the same time, a small amount of visible light was observed to be shifted down in frequency from 0 to $>300 \text{ cm}^{-1}$, but peaked near a shift of 150 cm^{-1} . This is interpreted to be parametric fluorescence

resulting from the amplification of quantum noise.¹⁵ Optical rectification may have also contributed to the FIR signal.^{1,2} These observations indicate the strength of the non-linear interactions at the intensities used.

When both the nanosecond and the picosecond pulse were passed through the full 4 mm length of the crystal, the picosecond pulse was depleted by 35-50%. With the nanosecond pulse at full intensity, the FIR energy was observed to depend linearly on the visible picosecond intensity for more than an order of magnitude above a threshold at 20 μ J. This threshold may represent the beginning of the saturated regime. Finally, the FIR was insensitive to the nanosecond pulse energy; when the nanosecond intensity was reduced by a factor of 7, the FIR intensity only dropped by a factor of 2 and the visible picosecond pulse was still strongly depleted. All these observations are consistent with a very strong non-linear interaction, leading to strongly saturated FIR generation.

However, strongly saturated mixing should lead to high quantum efficiencies. Even considering losses such as reflections at the crystal faces and FIR absorption¹² by the crystal, the observed quantum efficiencies were surprisingly low. Furthermore, strong pulse-to-pulse fluctuations in the FIR energy were observed, whereas saturated mixing should be quite stable. Figure 3 shows a histogram of these fluctuations. All pulses with energy greater than twice the median energy are shown in the cross-hatched bar at

the right. Some pulses were observed with as much as five times the median pulse energy. Comparable fluctuations were not seen in the strong depletion of the optical picosecond pulse which occurred when FIR was being generated. Given the observed weak dependence of the FIR energy on the visible pulse energies, the FIR fluctuations cannot result from the small visible pulse fluctuations assuming smooth visible pulses of uniform length. Also, measurements of the transmission of the visible beams through a pinhole showed that wandering of the beams from pulse-to-pulse was not responsible for the fluctuations.

Both the low quantum efficiency and the large pulse-to-pulse fluctuations are inconsistent with the simple theory of saturated mixing. Two of the possible explanations for this disagreement are:

- 1) The FIR was generated efficiently by a saturated process, but was then depleted by further non-linear processes. The fluctuations in these processes caused the fluctuations in the FIR and were responsible for the discrepancy between depleted picosecond energy and measured FIR energy. However, no reduction in the fluctuations was observed when the beam intensities were reduced.

- 2) The nanosecond pulse was not bandwidth limited, implying that it had significant amplitude modulation. Since the picosecond pulse was not long enough to average these fluctuations, it may

have seen pulse-to-pulse intensity variations in the temporal overlap with the nanosecond pulse that were larger than are implied by the total pulse-to-pulse energy variations. Since the FIR energy was relatively insensitive to the nanosecond energy, these modulations would have to be very large.

Neither of these explanations is entirely satisfactory, but the presence of occasional high efficiency pulses can be seen as encouraging. Their presence implies that a fundamental limit to conversion efficiency has not yet been reached. If the source of these fluctuations can be understood, it may be possible to consistently generate FIR pulses with the efficiencies which are now seen only erratically. Even without this improvement, intense, continuously tunable, narrow bandwidth FIR pulses have been produced which can be a valuable spectroscopic tool.

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Footnotes and References

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1. K. H. Yang, P. L. Richards, and Y. R. Shen, Appl. Phys. Lett. 19, 320 (1971); J. R. Morris and Y. R. Shen, Opt. Commun. 3, 81 (1971).
2. D. H. Auston, K. P. Cheung, J. A. Valdmanis, and D. A. Kleinman, Phys. Rev. Lett. 53, 1555 (1984).
3. E. D. Shaw and C. K. N. Patel, in Free Electron Generators of Coherent Radiation, S. F. Jacobs, H. S. Pilloff, M. Sargent III, M. O. Scully, and R. Spitzer, eds. (Addison-Wesley, London, 1980), p. 665.
4. For examples see Nonlinear Infrared Generation, Y. R. Shen, ed. (Springer-Verlag, New York, 1977).
5. J. M. Yarborough, S. S. Sussman, H. E. Purhoff, R. H. Pantell, and B. C. Johndon, Appl. Phys. Lett. 15, 102 (1969); M. A. Piestrup, R. N. Fleming, and R. H. Pantell, Appl. Phys. Lett. 26, 418 (1975).
6. K. H. Yang, J. R. Morris, P. L. Richards, and Y. R. Shen, Appl. Phys. Lett. 23, 669 (1973).
7. J. R. Morris and Y. R. Shen, Phys. Rev. A15, 1143 (1977).
8. M. Berg, A. L. Harris, J. K. Brown, and C. B. Harris, Opt. Lett. 9, 50 (1984).
9. I. P. Kaminow and W. D. Johnston, Phys. Rev. 160, 519 (1967).
10. A. S. Barker, Jr. and R. Loudon, Phys. Rev. 158, 433 (1967).

11. W. T. Welford and R. Winston, The Optics of Non-Imaging Concentrators (Academic Press, Inc., New York, 1978), pp. 47-68.
12. See, for example, D. R. Bosomworth, Appl. Phys. Lett. 9, 330 (1966). Our own measurements show the absorption coefficients to be a factor of 10 larger than Bosomworth's.
13. C. H. Henry and C. G. B. Garrett, Phys. Rev. 171, 1058 (1968).
14. N. Bloembergen, Non-linear Optics (W. A. Benjamin, Inc., Reading, Mass., 1965), Sec 4-4.
15. A. Yariv, Quantum Electronics (John Wiley and Sons, Inc., New York, 1975), Sec 17.7.

FIGURE CAPTIONS

Figure 1. Total detected energy of the picosecond FIR pulse as a function of frequency. The quantum efficiency, η , for conversion from the visible to the FIR varies from 0.1% to 0.3%.

Figure 2. Average spectrum of the FIR generated with the laser difference frequency centered at 70 cm^{-1} .

Figure 3. Distribution of the pulse-to-pulse fluctuations of the FIR energy. The cross-hatched region represents all of the pulses with more than twice the median energy.

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FIGURE 1

FIGURE 2

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FIGURE 3

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