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Generation of Far Infrared Radiation by Picosecond Light Pulses in $LiNbO_3$

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ABSTRACT

We have observed far infrared radiation generated by picosecond pulses in $LiNbO_3$ with several different phase-matching conditions. The output spectra, analyzed by a far-infrared Michelson interferometer and by a Fabry-Perot interferometer agree well with theoretical calculations. The laser pulse-width deduced from these measurements was about 2 psec in comparison with 5 psec obtained from two-photon fluorescence measurements.

The generation of coherent radiation in the far-infrared (IR) by optical beating has been of considerable interest.[1-5] Because of its broad spectral content, a picosecond laser pulse can generate far IR in a nonlinear crystal as a result of beating between its various frequency components. In this paper, we report our recent experimental results on far IR generation in a $LiNbO_3$ crystal using a mode-locked Nd:glass laser.[6] We have investigated the cases with crystal orientations corresponding to phase matching at zero frequency and at finite frequencies. In both cases, the experimental results agree well with theoretical calculations.[7]

The mode-locked laser output consisted of a train of about 30 pulses between the half-power points of the envelope with 6-nsec separation between pulses. The total energy contained in the pulse train was about 20 mJ. The pulsewidth measured with two-photon fluorescence techniques was about 5 psec. A lens with 30 cm focal length was used to focus the laser beam into the $LiNbO_3$ crystal which was 26 cm from the lens, and 1 mm of black polyethylene was used at the output to prevent laser light from reaching the far IR detectors. The far IR output was then split into two beams by a mylar beam splitter. One of the beams was used for spectral analysis in either a Michelson interferometer[8] (MI) or a Fabry-Perot interferometer[9] (FPI) and the other for normalization. They were separately detected by two n-type InSb (Putley) detectors[10,11] operated at 1.5 ± 0.05°K in a magnetic field of 5.4 kG. The entire far IR system was evacuated to avoid water vapor absorption. The two signals detected were amplified[3] and displayed on a Tektronix 556 dual beam oscilloscope. The ratio of the two signals was then computed. To obtain the true spectrum from the Michelson interferogram, the Fourier transform of the interferogram was divided point by point by the instrumental function (the spectral sensitivity of the spectrometer-detector system).[8] This instrumental function was measured by using a Hg arc lamp as a blackbody source. The same source was also used to align the MI and locate the zero of path difference.[8]

Two crystal orientations were studied. In the first, a 0.775 mm thick $LiNbO_3$ crystal was oriented with the normally incident laser beam polarized along the $\hat{c}$-axis and propagating along $\hat{b}$. In this configuration, the nonlinear susceptibility χ_{33} ($= 1.57 \times 10^6$ esu) contributes to the generation of far IR polarized along $\hat{c}$ and phase matching occurs at zero difference frequency. The MI was used to investigate the far IR spectrum. The interferogram was sampled at intervals of 0.2 mm out to a maximum of 5 mm which limited the resolution[8] to 2 cm^{-1}. Four laser shots were averaged for each sample. The final spectrum computed with linear apodization[8] and corrected by the instrumental function is shown in Fig. 1(a). Because of the frequency-dependent reflection coefficient[8] of the 5-mil mylar beam splitter in the MI, the reliable range of the spectrum measured was from 3 to 22 cm^{-1}. The resulting spectrum contains peaks at 2.5, 6.5 and 10.5 cm^{-1} with descending amplitude. The theoretical calculation[7] assuming a 1.8 psec Gaussian laser pulse is also shown in Fig. 1(a) for comparison. The agreement is good.

In the second case, a 1.524 mm thick $LiNbO_3$ crystal was cut with its $\hat{c}$-axis 16° away from the normal to the plane surfaces and the $\hat{a}$-axis at 30° from the plane containing the normal and the $\hat{c}$-axis. The normally incident laser beam was polarized to have equal components in the ordinary and the extraordinary rays. In this configuration, the ordinary far IR output produced by $X_{24} = 1.54 \times 10^{-6}$ esu and $X_{22} = 2.2 \times 10^{-7}$ esu, is phase-matched at 13.5 cm^{-1} and 6.7 cm^{-1}, corresponding to far IR propagating in the forward and the backward directions respectively. Extraordinary far IR was produced by X_{24}, X_{31}, X_{22}, and X_{33}, but was rejected in our experiments by a grid polarizer. The experimental result measured with the MI is shown in Fig. 1(b). The backward phase-matched peak[7] at 6.5 cm^{-1} was observed, but the forward phase-matched peak expected at 13.5 cm^{-1} was not distinguishable from the background noise. Theoretical curves for 1.8 and 2.3 psec Gaussian laser pulses are shown for comparison. It is seen that the relative strength of the 13.5 cm^{-1} peak is very sensitive to the variation of the pulsewidth, which changes with the laser operating conditions. In a separate experiment, we used a far IR Fabry-Perot to analyze the spectrum. Since the spectrum is expected to contain only two narrow phase-matching peaks, a Fabry-Perot which has higher peak transmissivity than the MI should be more suitable. The experimental results are shown in Fig. 2. The first, third, and fifth peaks in the figure arise from the forward phase-matched peak at 13.5 cm^{-1}, while the second and fourth peaks have contributions from both the forward and the backward phase-matched peaks. Since the resolving power of the Fabry-Perot[8,9] was limited to about 4 due to the 40° spreading angle of the far IR radiation from the crystal, the theoretical widths (~ 2 cm^{-1}) of the phase-matched peaks were not resolved. The solid curve in Fig. 2 is the corresponding theoretical calculation of the interferometer fringes assuming Gaussian laser pulses with a 1.8 - psec pulsewidth. It appears to be in satisfactory agreement with the experimental results. As a separate check using the Fabry-Perot interferometer, we rotated the $LiNbO_3$ crystal to phase match at $\theta = 14°$. The result was similar to Fig. 2 except that the phase-matched peaks appear at 11 cm^{-1} and 5.5 cm^{-1} as expected from the theory.

The total far IR energy detected from X_{33} in the first case (phase matching at zero frequency) was of the order of 1 erg, which is about 20 times larger than that detected in the second case (phase matching at finite frequencies). Both agree to within an order of magnitude with a theoretical estimate based on a mode-locked laser train of 30 pulses and a peak power of .2 GW/cm^2. We also measured the relative far IR power generated by X_{31} and X_{33}. The measured ratio $\frac{X_{33}}{X_{31}} \simeq 4$ was in satisfactory agreement with the calculated value[12] of 3.5. The peak power of the picosecond far IR pulses in the first case was of the order of 200 W, which could be increased to 5 kW by increasing the laser peak power to 1 GW/cm^2. In the second case, the spectral width and the tunability can not compete with the two ruby laser system previously investigated,[2,3] but if the laser pulse width could be as short as 0.5 psec, this system would be capable of producing tunable far IR pulses from 3 to 40 cm^{-1}. These pulses can be used to investigate transient and nonlinear phenomena in the far IR region.

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FIGURE CAPTIONS

Fig. 1a. Far-infrared spectrum generated by mode-locked pulses in $LiNbO_3$ phase-matched at zero frequency. The experimental points were obtained from the Michelson interferogram and the solid curve from a theoretical calculation assuming Gaussian laser pulses with a 1.8 psec. pulsewidth. The spectrum can be understood as a product of (1) the spectral content of the mode-locked pulses, (2) a radiation efficiency curve, and (3) a phase-matching curve centered at zero frequency (see Ref. 7).

Fig. 1b. Far-infrared spectrum generated by mode-locked pulses in $LiNbO_3$ oriented to have forward and backward phase-matching at 13.5 cm^{-1} and 6.7 cm^{-1} respectively. The experimental points were obtained from the Michelson interferogram. The solid and dashed curves were calculated by assuming Gaussian laser pulses with a pulsewidth of 2.3 psec and 1.8 psec respectively. In this experiment, our laser condition was somewhat different than in the other experiments, and it was very likely that the output pulses were longer.

Fig. 2. Fabry-Perot fringes of far-infrared radiation generated by mode-locked pulses in $LiNbO_3$ simultaneously phase-matched at 13.5 cm^{-1} and 6.7 cm^{-1}. The circles are experimental points and the curve was calculated from the dashed theoretical spectrum in Fig. 1b corresponding to Gaussian laser pulses with a 1.8 psec pulsewidth.

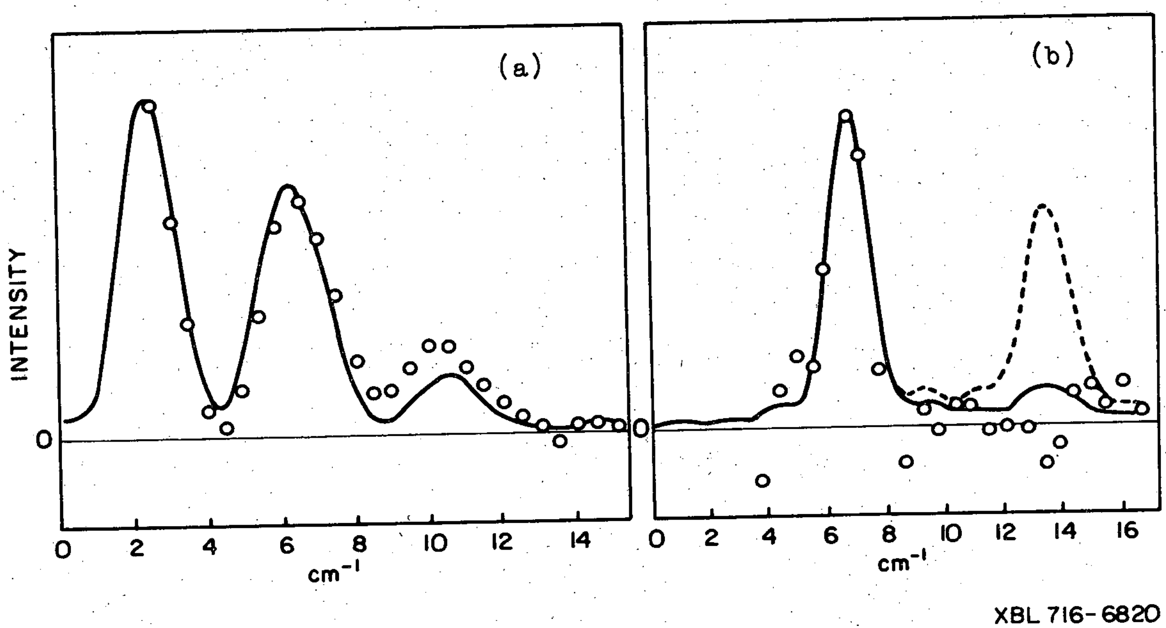
(a)
(b)
INTENSITY
0
0 2 4 6 8 10 12 14
cm⁻¹
0 2 4 6 8 10 12 14 16
cm⁻¹
XBL 716-6820

Fig. 1

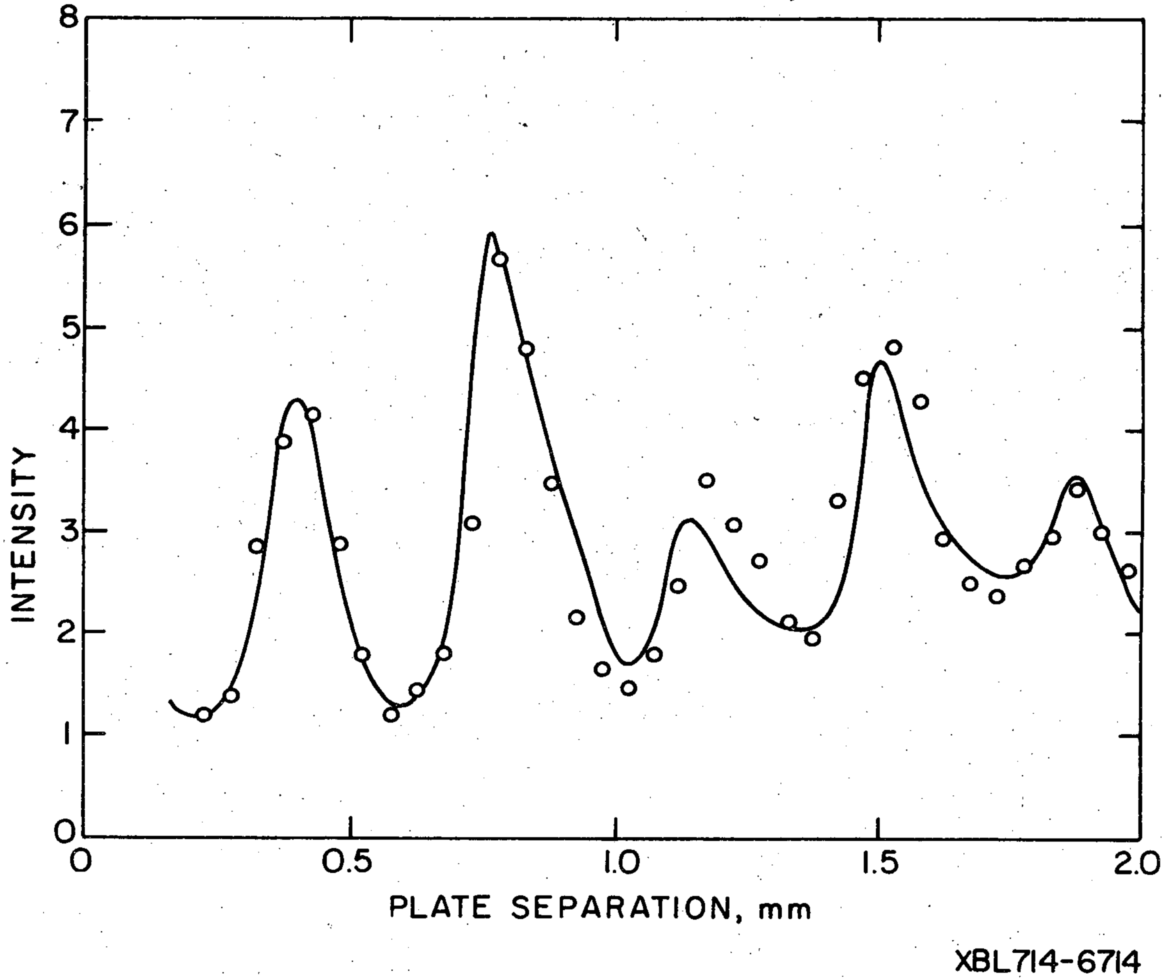
8
7
6
5
4
3
2
1
0
INTENSITY
0
0.5
1.0
1.5
2.0
PLATE SEPARATION, mm
XBL714-6714

Fig. 2