Tunable Far-Infrared Radiation Generated from the Difference Frequency between Two Ruby Lasers

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Far-infrared radiation generated from the difference frequency between two temperature-tuned ruby lasers has been observed over the frequency range from 1.2 to 8.1 cm^{-1}. Lithium niobate and quartz were used as mixing crystals. The conversion efficiency was measured as a function of angle around the phase-matched direction. The expected spectral content and frequency of the far-infrared radiation has been verified using a far-infrared Fabry-Perot interferometer.

SOON after optical second-harmonic generation was discovered, it was suggested by several persons[1] that difference-frequency generation in a nonlinear crystal using two temperature-tuned lasers would provide a tunable source of coherent far-infrared radiation. In this paper we describe the first observation of this tunable narrow-band far-infrared radiation. Fixed-frequency far-infrared radiation has been reported by two groups: Zernike and Berman[2] detected broadband radiation near 100 cm^{-1} resulting from the mixing of an unknown number of modes from a pulsed neodymium-glass laser. Yajima and Inoue[3] used the R_1 and R_2 lines of a single ruby laser to generate a fixed difference frequency, $\nu = 29$ cm^{-1}. In neither case was a spectral analysis reported. We have used two, simultaneously Q-switched, temperature-tuned ruby lasers to generate radiation between 1.2 and 8.1 cm^{-1}. By using sum-frequency generation to normalize the pulse-to-pulse variations, we have measured the far-infrared frequency directly and found it to be in agreement with the known temperature coefficient[4] of the ruby-laser frequency. We have also measured the variation of the far-infrared power with orientation of the $LiNbO_3$ crystal near the phase-matching angle. Difference-frequency generation was observed in quartz and $LiNbO_3$ and a comparison is made of their electro-optical coefficients as calculated from their relative efficiencies.

Consider two cylindrically symmetric beams of finite transverse radius a traversing a crystal of length l. The field intensities of the beams ($i=1, 2$) are

$$\mathbf{E}_i(\mathbf{r},t) = \tfrac{1}{2}[\mathcal{E}_i \exp(ik_i z - i\omega_i t) + \text{c.c.}].$$

A nonlinear polarization of frequency $\omega = \omega_1 - \omega_2$ will be produced in the cylinder of length l and radius a by the interaction of the two electric fields with the medium.

$$\mathbf{P}(\mathbf{r},t) = \tfrac{1}{2}(\chi^{(2)} \mathcal{E}_1 \mathcal{E}_2^* e^{i(k+\Delta k)z - i\omega t} + \text{c.c.}),$$

where $k_1 - k_2 = k + \Delta k = (\omega/c)n + \Delta k$ and where n is the index of refraction at the difference frequency ω. By integrating over the contributions of the cylindrical polarization wave in the far-field approximation, we obtain the total far-infrared power W collected in the detection system. We neglect the effect of the boundary by assuming that the detector is buried in the dielectric medium.

$$W = \frac{n\omega^4}{4c^3} |\chi^{(2)}|^2 |\mathcal{E}_1|^2 |\mathcal{E}_2|^2 l^2 (\pi a^2)^2 \times \int_{\phi=0}^{\phi_m} \sin\phi \, d\phi \left[\frac{\sin\eta}{\eta}\right]^2 \left[\frac{2J_1(\zeta)}{\zeta}\right]^2, \quad (1)$$

where $\eta = \frac{1}{2}kl(1 + \Delta k/k - \cos\phi)$, $\zeta = ka \sin\phi$, ϕ is the angle between the incoming beam and the generated radiation, and ϕ_m is the maximum angle collected in the detection system.

Equation (1) is valid for single-mode lasers. A beam with divergence Ω and area A contains $N = A\Omega/\lambda^2$ modes. Under the condition of small difference frequencies and limited collection angle (which existed in our experiments), the measured signal arises only from each mode of one laser interacting with one mode from the other laser. Therefore, the detected power is reduced by a factor of $1/N$ from that predicted by Eq. (1).

In our experiment, the two lasers were simultaneously Q-switched by using the same rotating mirror in both optical cavities.[5] The mode purity was controlled by using a resonant reflector as the output mirror and by

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using a saturable dyecell (Eastman 10220). One of the lasers was cooled by circulating ethyl alcohol at $T \geq -40$°C and the other was operated at room temperature. The two laser beams were made coincident and accurately parallel (within 1 min of arc) by careful adjustment of a beam splitter. No focusing lens was used. The polarizations of the lasers were made accurately perpendicular (vertical and horizontal) by the use of external polarizers. Each laser typically delivers a power of 1 MW over an area of 0.2 cm^2 with an angular divergence of 1.5 mrad and a pulse duration of 3×10^{-8} sec. The power is usually distributed into two frequency modes separated by 0.2 cm^{-1}.

The far-infrared signal was detected using a crystal of n-type InSb (Putley[6] detector) at $T=1.3$°K in a magnetic field of 5500 Oe. It was biased with a constant voltage of 0.25 V and the current was measured using an operational amplifier with a feedback resistor $R_F=205$ kΩ. The response time of this system is 2 μS. The sensitivity of the detector was measured using a blackbody at 200°C and a filter passing 0–50 cm^{-1}. This showed the average noise equivalent power in a 5×10^5-Hz bandwidth to be 10^{-6} W. However, since the sensitivity is certainly not uniform in this energy region[6] and since there are inevitable local system resonances at these long wavelengths, the absolute values of the infrared power may be in error by more than an order of magnitude. For this reason, emphasis was on relative powers in our measurements.

The nonlinear crystal was mounted on a rotatable table directly in front of the light pipe leading to the detector. A black polyethylene filter was used to reject unwanted radiation.

The infrared power generated is proportional to the integrated overlap in space and time of the two laser beams. Since this overlap varies from shot to shot, it is desirable to obtain an independent measurement of it for use as normalization.[7] This was done by monitoring the intensity of the sum frequency generated in a crystal of potassium dihydrogen phosphate (KDP). The discrimination of the sum frequency from the second-harmonic signal was achieved by using the scheme of Maier *et al.*[8] and Armstrong.[9] A discrimination factor better than 50 against second-harmonic radiation was obtained. Because of the small k vector of the far-infrared radiation, fluctuations in beam alignment and angular-mode distribution are expected to be more critical for difference-frequency than for sum-frequency generation. The far-infrared difference-frequency signals were found to be proportional to the sum-frequency signal within a factor of 2.

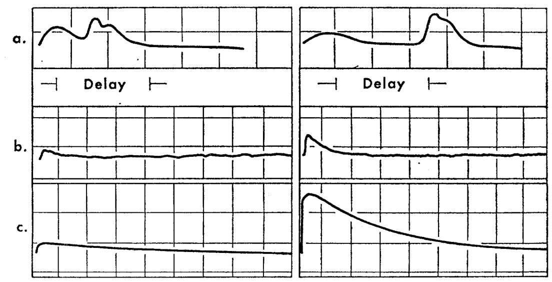

FIG. 1. Typical oscilloscope traces showing correlation between the time overlap of laser pulses and the strength of sum- and difference-frequency signals. The laser signals are displayed on a single trace (a) at a sweep rate of 50 nsec/div, with the cooled laser signal delayed by 125 nsec. Difference-frequency signals (b) and sum-frequency signals (c) are displayed at a sweep rate of 5 μsec/div. The pulse widths of (b) and (c) are characteristic of the time response of the detectors used. When there is considerable time overlap (as on the right), the sum- and difference-frequency signals are clearly much larger.

Typical infrared signals are shown in Fig. 1, where they are compared with the sum-frequency signal and the signals from the individual lasers. Satisfactory correlation is observed between the difference-frequency signal, the sum-frequency signal, and the laser timing.

The variation of the far-infrared power as the 1.5-cm $LiNbO_3$ crystal is rotated through the phase-matched direction is shown in Fig. 2. The experimental points are compared with the theoretical curve plotted assuming that the output of each laser is split equally between two frequencies separated by 0.2 cm^{-1}. The position of the peak in Fig. 2 agrees within experimental accuracy with the phase-matching angle of 9.5° from the optic axis computed using $n_e=2.189$ and $n_0=2.273$ (at the laser frequencies)[10] and $n_0=6.55$ (at 8.1 cm^{-1}).[11]

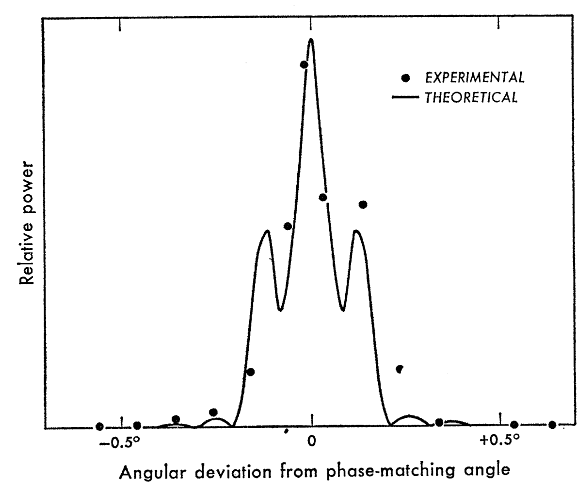

FIG. 2. Variation of the power of the difference-frequency signal as a function of the angular deviation from the phase-matched angle. The angles refer to the inside of the 1.5-cm $LiNbO_3$ crystal used.

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The measured far-infrared power from a 0.047-cm $LiNbO_3$ crystal at the phase-matching peak is about 1 mW. This is in order-of-magnitude agreement with the value calculated from Eq. (1) with a collection half-angle of 30°. For the 1.5-cm crystal, the measured peak power is 2×10^{-2} W, which is two orders of magnitude lower than what is expected. This discrepancy is most likely due to crystal inhomogeneity,[12] which would reduce the efficiency of optical mixing in long crystals. All the long crystals we used suffered damage after several hundred shots. The validity of quantitative comparisons with Eq. (1) is also limited by the unrealistic boundary conditions used in its derivation. Neglected effects include radiation from the edges of the crystal and multiple reflections at the faces.

The far-infrared wavelength was measured using a Fabry-Perot interferometer with electroformed metal mesh mirrors.[13] Typical transmission curves are shown in Fig. 3. The solid curve is obtained from the Airy formula by integrating over the finite collection angle so as to fit the decrease in Q with increasing order number. The wavelengths used were 3% [Fig. 3(a)] and 5% [Fig. 3(b)] smaller than those predicted from the known temperature dependence of the ruby-laser frequency. The finesse was computed from the geometry of the mesh. The fit shows unambiguously that we are observing a difference frequency with a bandwidth less than the $\sim$1 cm^{-1} resolution of our interferometer. The linewidth of the two frequency modes (separated by 0.2 cm^{-1}) from each laser is less than 0.02 cm^{-1}, leading to a predicted linewidth of less than 0.04 cm^{-1} for each of the three far-infrared frequencies produced.

We also compared the far-infrared power generated from a 0.047-cm-thick crystal of $LiNbO_3$ with that from a 1-cm-thick crystal of quartz. Using Eq. (1), the ratio of the electro-optic coefficients $r_{22}(LiNbO_3)/r_{62}(\text{quartz})$ is estimated to be 8.5. According to other measurements,[14] the ratio is 3.7. Because of the uncertainties in our measurement, this agreement must be considered satisfactory.

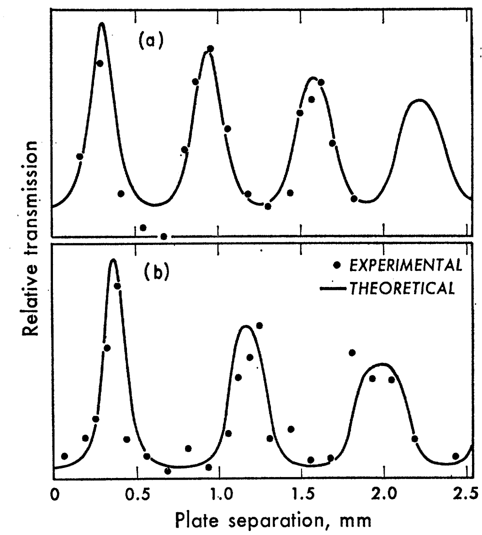

FIG. 3. Fabry-Perot scan of the difference-frequency output. The upper scan (a) is for a temperature difference $\Delta T=60°C$ of the two lasers. For the lower scan (b), $\Delta T=47°C$. The theoretical curves are Airy functions calculated from the geometrical properties of the Fabry-Perot reflectors and averaged to account for the 30° collection half-angle.

The tuning range was limited to frequencies less than 8.1 cm^{-1} by the cooling system used. This range could be extended to $\sim$20 cm^{-1} by using liquid nitrogen as a coolant. If the warmer laser were operated on the R_2 line, then the range could be extended to $\sim$50 cm^{-1}. The use of a tunable dye laser, stimulated Raman radiation, or parametric sources would, of course, extend this range throughout the infrared.

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