Acoustically Phase-Matched Noncollinear Optical
Second Harmonic Generation in Tellurium

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Introduction
The acoustically phase-matched noncollinear optical second harmonic generation (SHG) as a novel technique to realize an efficient optical frequency doubler is experimentally demonstrated for the first time in Te. The phenomenon is interpreted as being due to the mixing of the incident light and the acoustooptically scattered light.

Principle
The possibility of this technique has been theoretically predicted by Harris et al and Nelson et al.\(^1\),\(^2\) The basic principle for the technique is to compensate the phase-mismatch \(\Delta k\) in the conventional SHG by interacting the acoustic wave \(\vec{k} (=\Delta k)\). The output second harmonic (SH) wave \((\omega_c, \vec{k}_c)\) will be produced by mixing the two input infrared waves \((\omega_A, \vec{k}_A)\) and the one input acoustic wave \((\Omega, \vec{k})\). Exactly speaking, the output frequency \((\omega_c = 2\omega_A + \Omega)\) differs from the SH frequency \(2\omega_A\) by the small amount of \(\Omega\). In case of Te there are two kinds of interactions, one is direct mixing of the three input waves and the other is indirect mixing process. The latter indirect process arises from acousto-optic scattering (frequency of the scattered light: \(\omega_B = \omega_A + \Omega\)) followed by optical mixing \((\omega_c = \omega_A + \omega_B)\). Accordingly, the phase-matching condition for the direct process is

\[
\vec{k}_c = 2\vec{k}_A + \vec{k}
\]

(1)

On the other hand, to realize the indirect process, it is essential to satisfy the following two conditions simultaneously:

\[
\vec{k}_B = \vec{k}_A + \vec{k}
\]

(2-a) (for the acoustooptic scattering) and

\[
\vec{k}_c = \vec{k}_A + \vec{k}_B
\]

(2-b) (for the optical mixing). Theoretically, by satisfying Eqs. (2-a) and (2-b), the efficiency of the indirect process can be larger than that of the direct process by several orders of magnitude. To our knowledge, however, this indirect process has never been experimentally exploited up to date.\(^3\)

Experimental Results
The experiments were carried out in the geometry as shown in Figs. 1 and 2. The pulsed longitudinal acoustic wave propagating in the x direction \((v=2290\text{m/sec})\) was generated by the LiNbO\(_3\) transducer. The 10.6\(\mu\)m beam from a pulsed CO\(_2\) laser was focused into the Te crystal which traveled tilting from the optic axis by \(\theta_A=16.3^\circ\) as an ordinary wave. The confocal parameter of the beam within the crystal was 80mm. The peak power of the laser beam was about 50W. To satisfy the first phase-matching condition (2-a), the acoustic frequency was tuned to be at 88MHz. Thus, the intermediate wave \((\omega_B)\) was generated as an extraordinary wave by the anisotropic Bragg scattering due to the photoelastic constant \(P_{41}\). At the frequency of 88MHz, the second condition (2-b) was
automatically satisfied and the output SH wave \((\lambda_C=5.3\mu m)\) appeared as an ordinary wave in the direction of \(\theta_C=18.6^\circ\). Though the phase-matching condition (1) for the direct process was also automatically fulfilled, it was revealed that the efficiency for the indirect process was about \(3 \times 10^7\) times as dominant as that for the direct process. The SH power was separated from the unconverted fundamental by a sapphire plate filter. Fig.3 shows the acoustically phase-matched SH wave \((\omega_C)\), where the polarizer was set so as to pass only the ordinary wave component. When rotating the polarizer by \(90^\circ\) so as to pass the extraordinary wave, no SH power was detected as in Fig.4. When the acoustic power was switched off, or when the acoustic frequency was changed, the SH power again became zero. These experimental evidences prove that the detected output was due to the above-described nonlinear process.

Conclusion As an example for the novel technique for SHG in Te, the experiments using the longitudinal acoustic wave propagating in the \(x\) direction was successfully demonstrated. There are also several other acoustic waves propagating in other directions which satisfy Eqs. (2-a) and (2-b). Therefore, the restriction for the crystal orientation is much more relaxed than that in the conventional collinear index-matching technique. It is considered that the technique is especially suitable for Te since the optical nonlinearity and the acoustooptic figures of merit are anomalously large in the material.\(^4\) Besides, it is possible to apply this technique to a parametric oscillator in which the oscillation frequency can be varied by changing the acoustic frequency.\(^1\)

References

Fig.1 Wave vector diagram in \(\text{Te}\).

Fig.2 Experimental arrangement.

Fig.3 The wave form of SH pulse. The polarizer was set so as to pass the ordinary wave.

Fig.4 The polarizer was rotated by \(90^\circ\) so as to pass the extraordinary wave.