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Noncritical Detection of Tunable CO_2 Laser Radiation into the Green by Upconversion in Silver Thio-Gallate

G. C. Bhar[1], U. Chatterjee[1], P. K. Datta[1], S. Das[1], R. S. Feigelson[2], and R. K. Route[2]

[1] Burdwan University, Laser Laboratory Physics Department, Burdwan 713 104, India
[2] Centre for Material Research, Stanford University, Stanford, CA 94305, USA

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Abstract. Tangential near-noncritical phase matching is reported for upconversion in silver thiogallate ($AgGaS_2$) using a dye laser as pump. A large acceptance angle for noncollinearity as desirable from a device view point is realised. Weak temperature tunability is also exploited to realise noncriticality.

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Infrared detection by optical frequency mixing in a nonlinear crystal has been an attractive technique but its development was limited due to the non-availability of appropriate nonlinear crystal. With the development of quality $AgGaS_2$ crystal [1, 2] and its subsequent applicability in various other nonlinear devices, the field of infrared detection by upconversion has been studied from various view points [3–6]. These include infrared detection under critical and noncritical phase-matching, imaging and infrared spectrograph. Attractive features include its not-too-large birefringence, wider transmission in the visible coupled with large infrared transmission facilitating thereby use of dye and other visible lasers and availability in good optical quality. We demonstrate and interpret noncollinear tangential phase matching for the first time in this crystal in detection of tunable infrared radiation from CO_2 laser using a Nd : YAG laser-pumped dye laser as pump source. This offers wide tolerance in angular acceptance. We also report the effect of infrared heating of nonlinear crystal in obtaining stable and optimum operation for tunable upconversion.

Several noncollinear frequency mixing devices were previously studied [7–10] as an aid to automatic separation of the generated output from the interacting laser beams without significant reduction in the overall nonlinear conversion efficiency in the process. Tangential phase-matching is a special case of noncollinear phase matching whereby a large angular acceptance can be realised and so is very much desirable from a device view point. Beyond this a double peak results out of intersection of interpenetrating sum-frequency ellipsoid of wave normal with that of the signal frequency ellipsoid (as illustrated on a two-dimension plane in Fig. 1). Although such a wide angular acceptance situation was realised in

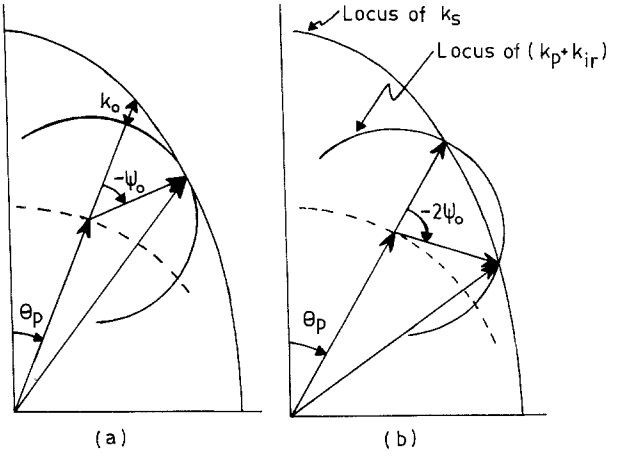

Fig. 1. a Diagram showing the tangential phase-matching situation. **b** Diagram showing the critical phase-matching situation $k_0 = 0$ and $\psi_{ir} = 0$ and $-2\psi_0$ for $\Delta k = 0$

critical tangential phase-matching case by Warner [11] and Koyanagi [12] in proustite crystal for 10.6 μm upconversion using, respectively, ruby laser and Nd : YAG laser as pump, it has not so far been explored in other crystals and also for other nonlinear frequency mixing processes. We have explored tangential phase-matching for upconversion not only in $AgGaS_2$ crystal but also in other nonlinear crystals for different frequency mixing processes and obtained interesting results. In $AgGaS_2$ upconversion some experimental results were, however, reported by Itabe and Bufton [13], and Koidl et al. [14] near a 90° phase-matching but without offering plausible explanation. Although the former researchers claimed the situation to be collinearly phase-matched but it is actually noncollinear since the collinear pump and

infrared beams would turn noncollinear inside the crystal on crystal rotation due to different values of refractive indices and so the situations are near equivalent.

Experimental

For small photon conversion efficiencies, the sum-frequency output is proportional to $[\sin(\Delta kL/2)/(\Delta kL/2)]^2$ where $\Delta k = \mathbf{k}_s - \mathbf{k}_p - \mathbf{k}_{ir}$ and L is the length of the crystal. From Fig. 2 we can write

$$\Delta k = k_s(\theta_p + \psi_s) - k_p(\theta_p)\cos\psi_s - k_{ir}(\theta_p + \psi_{ir})\cos(\psi_{ir} - \psi_s). \quad (1)$$

For typical negative uniaxial crystal, the degree of ellipticity of the k-surface for an extraordinary ray is such that ψ_{ir} will be small enough for $\sin\psi_{ir}$ to be replaced by $\psi_s = [k_{ir}(\theta_p)/k_s(\theta_p)] \cdot \psi_{ir}$, and expanding $k(\theta_p + \psi)$ in terms of θ_p and ψ using Taylor's theorem, we can write

$$\psi_{ir} = -\psi_0 \pm \sqrt{\psi_0^2 - A(k_0 - \Delta k)}. \quad (2)$$

For a type-I ($\mathbf{k}_s^e = \mathbf{k}_p^0 + \mathbf{k}_{ir}^0$) interaction

$$A = \frac{2[n_s^e(\theta_p)]^2 \lambda_p \lambda_{ir}^2}{\lambda_s[2\pi\lambda_{ir} n_{ir}^0(\theta_p) n_p^0(\theta_p) n_s^e(\theta_p) + \lambda_s \lambda_p n_{ir}^0(\theta_p) \cdot P]}, \quad (3)$$

where

$$P = \frac{3\pi}{2\lambda_s}[n_s^e(\theta_p)]^5 \left[\frac{1}{(n_s^e)^2} - \frac{1}{(n_s^0)^2}\right]^2 \sin^2 2\theta_p - \frac{4\pi}{\lambda_s}[n_s^e(\theta_p)]^3 \left[\frac{1}{(n_s^e)^2} - \frac{1}{(n_s^0)^2}\right] \cos 2\theta_p,$$

$$\psi_0 = \frac{A}{2} \cdot \frac{\lambda_s}{\lambda_{ir}} \cdot \frac{n_{ir}^0(\theta_p)}{n_s^e(\theta_p)} \cdot Q;$$

$$Q = -\frac{\pi}{\lambda_s}[n_s^e(\theta_p)]^3 \left[\frac{1}{(n_s^e)^2} - \frac{1}{(n_s^0)^2}\right] \sin 2\theta_p$$

and

$$k_0 = 2\pi[(n_s^e/\lambda_s) - (n_{ir}^0/\lambda_{ir}) - (n_p^0/\lambda_p)]_{\theta_p}.$$

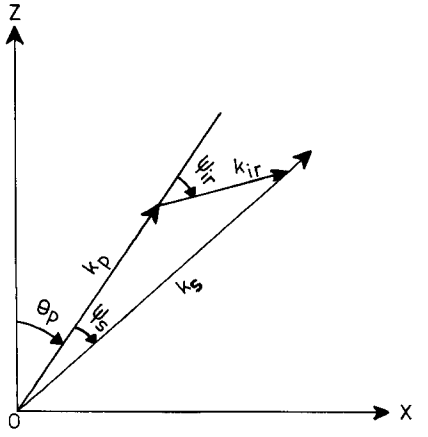

Fig. 2. Diagram illustrating the relationship between the crystal axes OZ, OX and the wave propagating vectors of the pump k_p signal k_{ir} and sum frequency k_s

If $\psi_0^2 = A(k_0 - \Delta k)$, there is only one solution to (2), corresponding to the tangential phase matching situation. Therefore, if we propagate the infrared beam at an angle $-\psi_0$ from the pump beam, tangential phase matching will be achieved at the appropriate value of θ_p.

We have found from an analysis of noncollinear phase-matching that such a noncritical tangential situation can be realised, whenever normal phase-matching is permitted for any critical phase-matching case [15]. In $AgGaS_2$ with a 10.6 μm signal for a 1.064 μm Nd pump laser such tangential matching is found to occur for noncollinear angle $-\psi_0$ of $1 \cdot 31°$ and $\theta_p = 37 \cdot 7°$ while for ruby laser it is obtainable at $-\psi_0$ of $1 \cdot 11°$ and $\theta_p = 52 \cdot 17°$. Figure 3a shows the computed results of noncritical tangential phase-matching in $AgGaS_2$ with different pump wavelengths of the dye laser as running parameters at a 10.7 μm signal.

The experimental arrangement consists of a tunable dye laser serving as the pump while a tunable cw CO_2 laser serving as the signal source. The arrangement is about the same as that described in [5] except in this case the crystal is type-I 90° cut and is a 8 millimeter cube. The CO_2-laser beam as well as the dye-laser beams are horizontally polarised. The crystal is mounted on a circular table capable of rotating in a vertical plane. Both the pump and signal beam enter the crystal in a vertical plane, and the arrangement is such that the CO_2 laser beam is made to propagate above the dye laser beam. The CO_2 beam is folded and focussed on the nonlinear $AgGaS_2$ crystal by a plane mirror and a concave mirror. The upconverted signal is detected by a silicon PIN photodiode (SGD 040 of EG & G) and is observed on a 100 MHz storage oscilloscope (Iwatsu model TS 8123). The signal on the oscilloscope is also monitored by detecting the pump dye-laser beam by another silicon PIN photodiode. The strong pump beam is blocked by two narrow-band interference filters (Microcoatings make) having 56% peak transmission at 560 nm and a bandwidth of 10 nm, whereas the CO_2 laser radiation is blocked by a sapphire plate (3 mm thick). The rejection outside passband is sufficient so that two filters along with a Glan polariser reduce the residual pump beam below the noise level of the detector, 5 mV. Collinear noncritical 90° phase-matching occurs near the dye-laser wavelength of 597 nm. While searching for tangential matching a small noncollinearity is introduced. With a small non-collinear angle at an appropriate pump wavelength phase-matched upconversion begins to occur which can be clearly seen through naked eyes after rejecting of the unconverted pump beam. With increase of pump wavelength the generation increases to an optimum broad angular peak. With still further increase of the pump-laser wavelength the upconversion peak is divided into two and is separated out, as shown in Fig. 3b. Similar results are obtained for other CO_2 laser wavelengths. The location of the broad peak and its separation is found according to theoretical prediction. Because of narrowness of the separated peaks it is difficult to locate the detector exactly on the peak. That is why peaks appear unequal experimentally sometimes. No detection could, however, be made at the trough where the intensity is very near to the

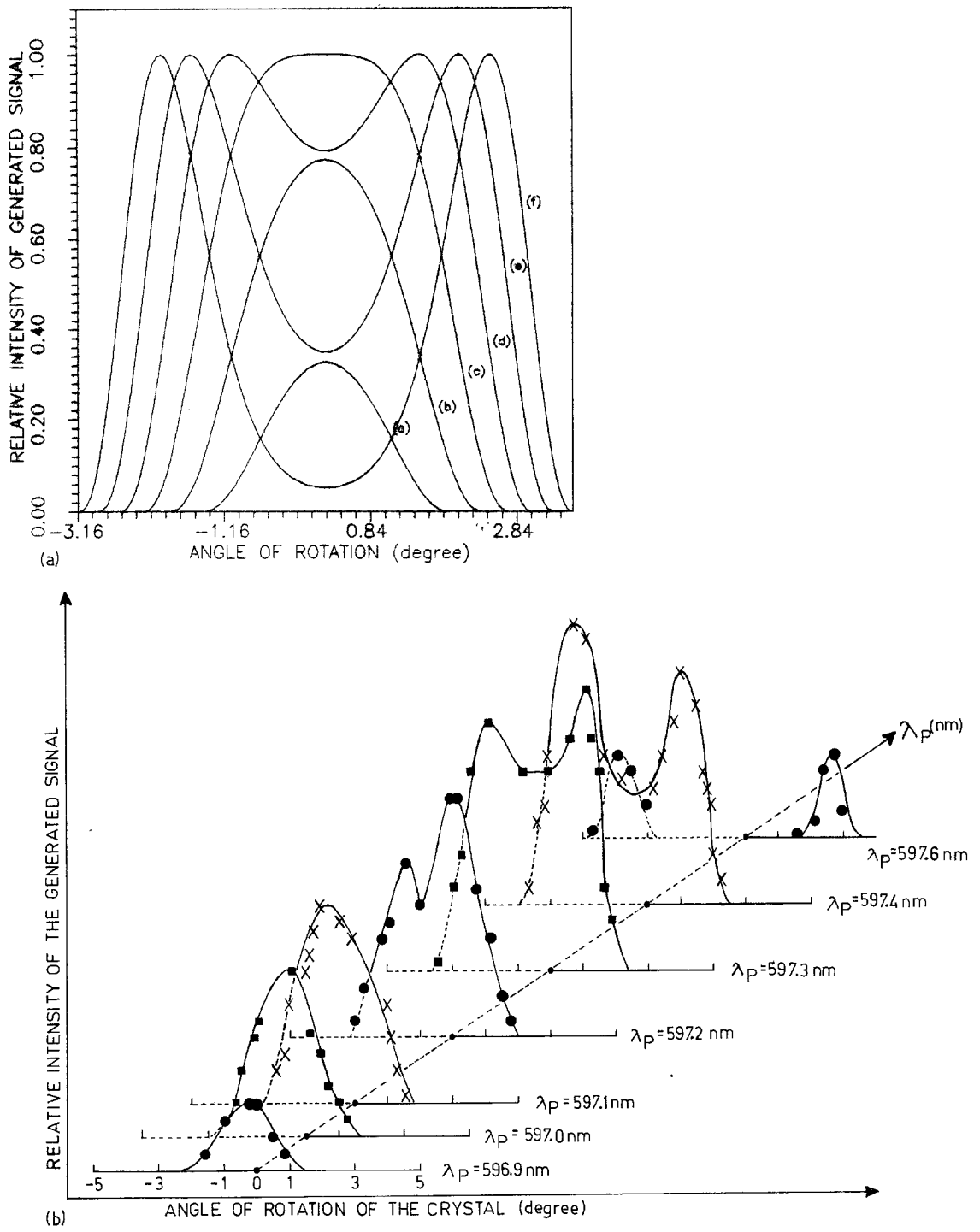

Fig. 3. **a** Relative, theoretically upconverted signal intensity (computed) with crystal rotation for different pump dye laser wavelength using Sellmeier's coefficient data of Bhar and Smith [16]. *a–f* corresponds to dye laser wavelength of 596.9, 597.0, 597.1, 597.2, 597.3, and 597.4 nms, respectively. **b** Relative upconverted observed signal intensity with crystal rotation from the normal position on changing the pump dye laser wavelength (λ_p). This angle of crystal rotation is related to the noncollinear angle inside the crystal. The computed graph for dye laser wavelength 597.6 nm is omitted in **a** for clarity

base line. We have measured the phase-matched acceptance angle very near to the optimum single-peak conversion. Both the experimentally measured value and the theoretically predicted value are 2.3°. The measured pump laser spectral bandwidth is 0.21 nm.

The refractive indices and birefringence of $AgGaS_2$ is temperature dependent but it is not so strong as in other ferroelectric crystals like ADP, $LiNbO_3$. However, in the course of the experiment we have noted weak but significant temperature sensitivity [16, 17]. In our experiment with intracavity chopped 8 watt cw CO_2 laser the temperature of the $AgGaS_2$ crystal is found to rise, as a consequence of this, the crystal requires a small but finite angular resetting from that of the room-temperature

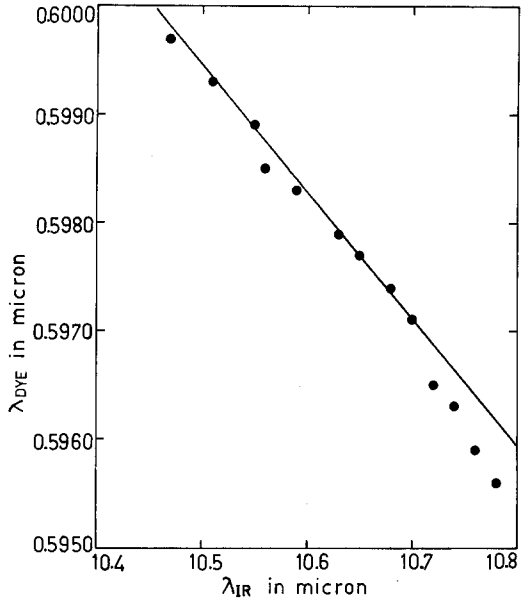

Fig. 4. Noncritical (90°) tuning of an infrared signal with the pump dye-laser wavelength. Smooth curve is theoretically predicted based on the Sellmeier data of Bhar and Smith [16], while ●●● are the experimental points

phase-matched position by a few seconds till arriving to this steady temperature which has been measured to be 38° C. Using the tunability of both the pump dye laser and the signal CO_2 laser we have measured the tuning characteristics for 90° phase-matching situation. The experimental points are presented in Fig. 4. Based on thermo-optic coefficients data of $AgGaS_2$ temperature-tuning characteristics for $\theta = 90°$ have been evaluated; it is represented as near-straight line in the figure showing fairly good agreement with experimental data.

Conclusion

In conclusion, we have demonstrated and analysed for the first time noncritical tangential phase matching in this potential $AgGaS_2$ crystal for upconversion detection of CO_2 laser radiation offering wide angular acceptance and so is potentially important from device viewpoint.

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