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Towards Enhancing the Efficiency of Nonlinear Optical Generation

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Abstract

The chapter dwells on two novel approaches towards enhancing the efficiency of nonlinear optical generation. The former is to enable the unabsorbed pump beam to pass through the crystal repeatedly. Integration of an unstable cavity containing the crystal with the stable pump cavity made this possible. The Q of the unstable cavity could be maintained high as the output coupler of the pump laser, itself served as the entrance mirror of this cavity. The unstable nature of the cavity kept the crystal from being exposed to high flux while ensuring longer interaction length. Although this scheme demonstrated in mid-IR region its advantage should persist across UV, visible, and near-IR regions too. The enhancement of conversion efficiency is effected in the second scheme by way of illuminating the crystal with alternate high and low regions of intensity along its length as against the uniform illumination case maintaining the same average intensity as in the conventional operation. The advantage is attributed to the square dependence of the second harmonic on the intensity of the pump. A simple modification of the existing experimental setup involving integration of an additional optical element with the pump cavity allowed exploitation of interference effect to realise such a non-uniform illumination condition.

Keywords: non-linear optical conversion, unstable cavity, interference, CO₂ laser, dichroic optics

1. Introduction

There has been a constant endeavour to increase the number of available coherent sources allowing wider coverage of the electromagnetic spectrum. To this end, nonlinear optical conversion of the emission of a laser inside an appropriate nonlinear crystal by ensuring that the fundamental and generated waves are phase matched has emerged as one of the most attractive methods across UV [1], visible [2], infrared [3], and mid-infrared [4] regions of the electromagnetic spectrum. As the non-linearity of the crystal is responsible for effecting this conversion, an increase in the intensity of the pump radiation to which the crystal is subjected to increases the conversion efficiency too albeit in a non-linear fashion. Single crystals, specifically grown to provide a reasonable interaction length, invariably suffer from low optical damage threshold. This thus puts an upper limit on the pump intensity to which the crystal can be exposed to causing a corresponding reduction in the conversion efficiency. A significant under-utilisation of the pump beam is thus the end result. The crystals employed for the conversion in the mid infrared region have

inherently high refractive index and the problem thus gets further compounded as the entrance and the exit faces of the crystals need to be essentially anti-reflection coated to arrest losses due to Fresnel reflection. The pump intensity therefore, needs to be further reduced as the optical damage threshold of dielectric coatings is usually lower than the crystal bulk. This drawback can be surmounted by increasing the interaction length of the pump beam with the nonlinear medium giving due consideration to the thermal de-phasing effect that occurs along the length of the crystal [5]. Increasing the length of the crystal brings about a steep rise in its cost and therefore is not an economically viable option. Attempts have been made to use a number of crystals instead, either in tandem [6] or in parallel [7] to circumvent this problem. These schemes however, suffer from an inherent disadvantage as they present too many crystal surfaces off which the pump photons escape through Fresnel reflections. To be noted here that the same crystal has also been used in the past to enhance the interaction length by allowing the pump beam to make two [8] or multiple passes [9] through it. These methods have not gained much popularity as the cavity configuration employed in the former case limited the operation to a non-collinear phase matched mode while in the latter case it resulted in enhancing the second harmonic (SH) conversion of the SH wave itself. In case of frequency doubling of near infrared cw pump to the visible, the schemes that have gained importance use the crystal in the intra-cavity mode [10] or external cavity resonant enhancement mode [11]. Ring cavity configuration that has an inherent advantage of blocking any feedback into the pump cavity has generally been employed here. The applicability of these schemes for pulsed second harmonic conversion (SHG) is challenging due to the high intra-cavity flux that prevails in a pulsed laser. Literature on similar schemes for SHG in the mid infrared region is scanty primarily due to the possibility of thermal lensing effect that may lead to crystal damage. This has restricted the operation to quasi-cw regime with adequate precautions to forbid Q-switched lasing [12] while in the case of pulsed operation, the intra-cavity flux has been brought down by using appropriate attenuators [13].

Another approach has been to increase the intensity of the pump beam itself. That the generated SH output increases in a non-linear fashion with the intensity of the pump radiation to which the crystal is exposed is a fact known since the time SHG was reported more than half a century ago [14]. A direct consequence of this fact is that if the crystal can in some way be subjected to alternate high and low regions of pump intensity along its conversion length that results in an average intensity I_{av} , there would be a net gain with respect to SHG as compared to the conventional situation where the same crystal is subjected to a uniform pump intensity of I_{av} . These two cases are illustrated in **Figure 1**. In the first case (**Figure 1a**) the crystal of length ' l ' is illuminated by a pump beam of uniform intensity ' I ' along its length. In the second case the incident pump intensity ' I ' is redistributed as alternate periodic intensity packets of ' $2I$ ' and ' 0 ' longitudinally along the crystal thus maintaining the same average intensity ' I ' as before (**Figure 1b**). The square dependence of second harmonic conversion on the incident pump intensity can be represented mathematically for the two cases as follows:

For the case of **Figure 1a**: $SH(\text{output}) \propto l \times I^2$.

For the case of **Figure 1b**: $SH(\text{output}) \propto [(l/2) \times 0] + [(l/2) \times (2I)^2] \propto 2l \times I^2$.

This clearly suggests that the generated SH, in the second case, is enhanced by a neat 100% as against the first case when the crystal is illuminated uniformly. A non-linear crystal placed inside a Fabry-Perot or a bidirectional ring cavity experiences flux from both ends and therefore is one of the most obvious ways of creating such a situation of non-uniform illumination. The interference of the forward and reverse beams creates alternate high (anti-nodal) and low (nodal) regions of intensity in the crystal and therefore should result in an enhancement of the SHG.

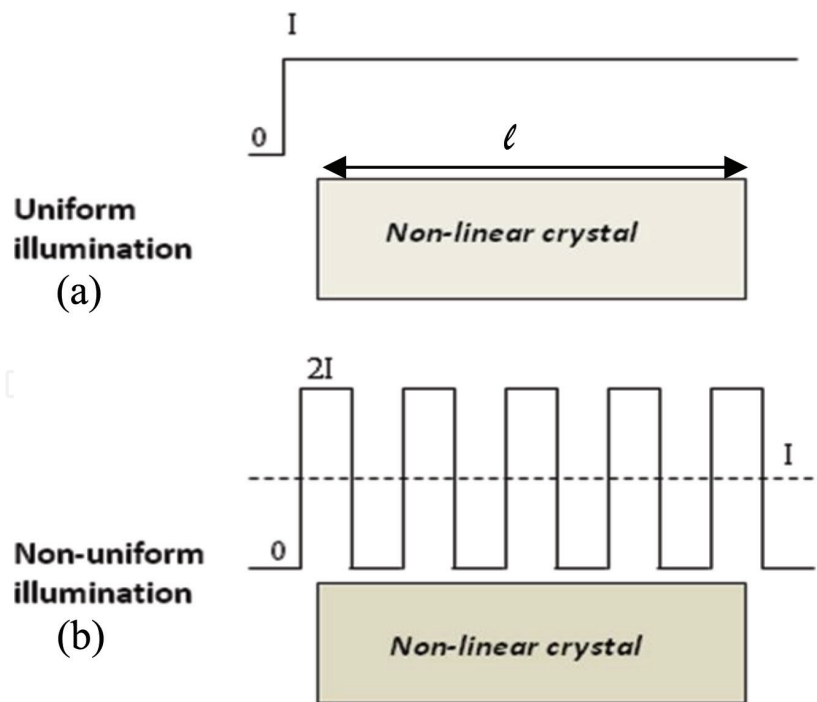


Figure 1.
A non-linear crystal exposed to the pump radiation. (a) Uniform illumination of intensity 'I'. (b) Periodic illumination with intensity packets of '2I' and '0' thus maintaining the same average intensity 'I' as before.

This chapter dwells on the recent advances made by our group in these two areas viz., enhancing the conversion efficiency by way of (a) increasing the interaction length between the pump and the non-linear medium and, (b) exploiting the effect of non-uniform illumination of the non-linear medium.

2. Enhancing the SH conversion efficiency by increasing the interaction length between the pump and the non-linear medium

By way of constructing a coupled plano-convex cavity external to the pump laser (Figure 2) that allowed to and fro passes of the unabsorbed pump through the crystal, we conceived a novel way to increase the effective interaction length between the non-linear medium and the pump beam [15]. An ideal situation demands that the coupling optics offers high transmission at the pump wavelength and high reflection too at the same wavelength to enable multiple passes through the crystal; a conflicting requirement indeed that is inherently taken care of in the above

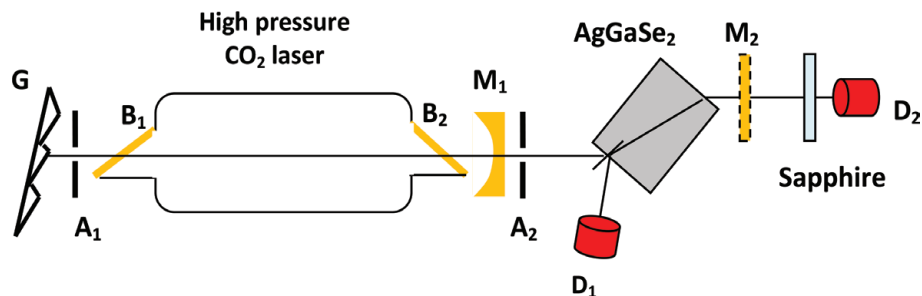


Figure 2.
Schematic diagram of the experimental setup for second harmonic conversion of the emission of a CO₂ laser in a AgGaSe₂ crystal. G: Plane blazed grating, A₁ and A₂: Adjustable apertures, B₁ and B₂: ZnSe Brewster plates, M₁: 70% R ZnSe concave mirror, D₁ and D₂: Energy/power detectors, M₂: Dichroic mirror. (a) In case of single pass second harmonic generation, dichroic mirror M₂ is absent. (b) In case of multi-pass second harmonic generation, dichroic mirror M₂ in conjunction with pump laser output coupler M₁ forms the unstable external cavity.

scheme. As the output coupler of the pump laser itself functioned as the entrance mirror of the external cavity, its quality factor could be maintained high allowing at the same time, efficient transportation of the pump beam into it. Further, the intra-cavity photon flux could be maintained within acceptable level due to the unstable nature of the external cavity. This reduced the risk of optical flux induced crystal damage besides eliminating the possibility of feed back into the pump cavity.

2.1 Experimental

The experimental demonstration of this scheme was effected in the second harmonic generation of the 10 micron emission of a pulsed CO₂ laser. A commercial uncoated 17 mm thick AgGaSe₂ crystal served as the non-linear medium for this conversion process. A rise in the energy conversion efficiency by ~300% and even higher peak power conversion efficiency has been achieved by making the unconverted pump go through the crystal time and again. The increase in the effective length of the crystal should in principle, allow the performance of a thin crystal in such a cavity configuration to match that of a thick crystal in the conventional operation although at a lower level of optical flux, that in turn, precludes the possibility of its damage even in the pulsed operation.

The schematic of the experimental lay out is depicted in **Figure 2**. In the first set of experiments (**Figure 2a**), the pulsed emission of a commercial multi-atmosphere TE-CO₂ laser was made use of to affect SHG in an uncoated AgGaSe₂ crystal (cross-section 10 × 10 mm and length 17 mm). A plane master grating (150 lines/mm) and a concave (7 m ROC) 70%R ZnSe output coupler separated by 105 cm formed the passively stabilised pump laser cavity. For this experiment, the laser was operated on 10P (34) line for which the second harmonic phase matching occurred at an external angle of incidence of ~34°. Usage of an intra-cavity adjustable aperture A₁ allowed the operation of the pump laser on the TEM₀₀ mode. The energy incident on the crystal was controlled by varying the charging voltage of the laser. An external adjustable aperture 'A₂' allowed maintaining the pump beam cross-section on the crystal entrance to ~4.5 mm diameter so as to ensure its clear passage through the non-linear medium. Monitoring of both the energy and the power of the incident pump pulse was possible by probing its Fresnel reflection off the incident face of the crystal. The energy and power profile of the generated SH beam were measured after blocking the unconverted pump beam that also emerged along with the SH beam through the crystal by means of a sapphire plate. The CO₂ laser, by virtue of its multi-atmosphere operation, possessed inherently very high gain and thus emitted pulses of relatively short duration (FWHM ~110 nsec, **Figure 3**). In the present experiment, the maximum intensity was restricted to ~2.5 MW/cm².

2.2 Results and discussion

In order to find the efficiency of the single pass non-linear conversion process as a function of the input pump energy, we gradually increased the input and measured the corresponding SH energy and the dependence is as shown in **Figure 4**. The parabolic nature of this dependence clearly reveals the square proportionality of the SH intensity on the pump intensity. As would be seen, ~8.46% is the maximum internal SH energy conversion efficiency that was obtained maintaining the pump intensity below the damage threshold of the crystal. Understandably therefore, significant fraction of the pump photons stays unconverted and emerge together with the SH beam and the same was measured using detector D₂ when the sapphire plate is removed. Effective utilisation of the pump beam is possible by making it to pass through the crystal time and again. To this end, a Fabry-Perot cavity was constructed

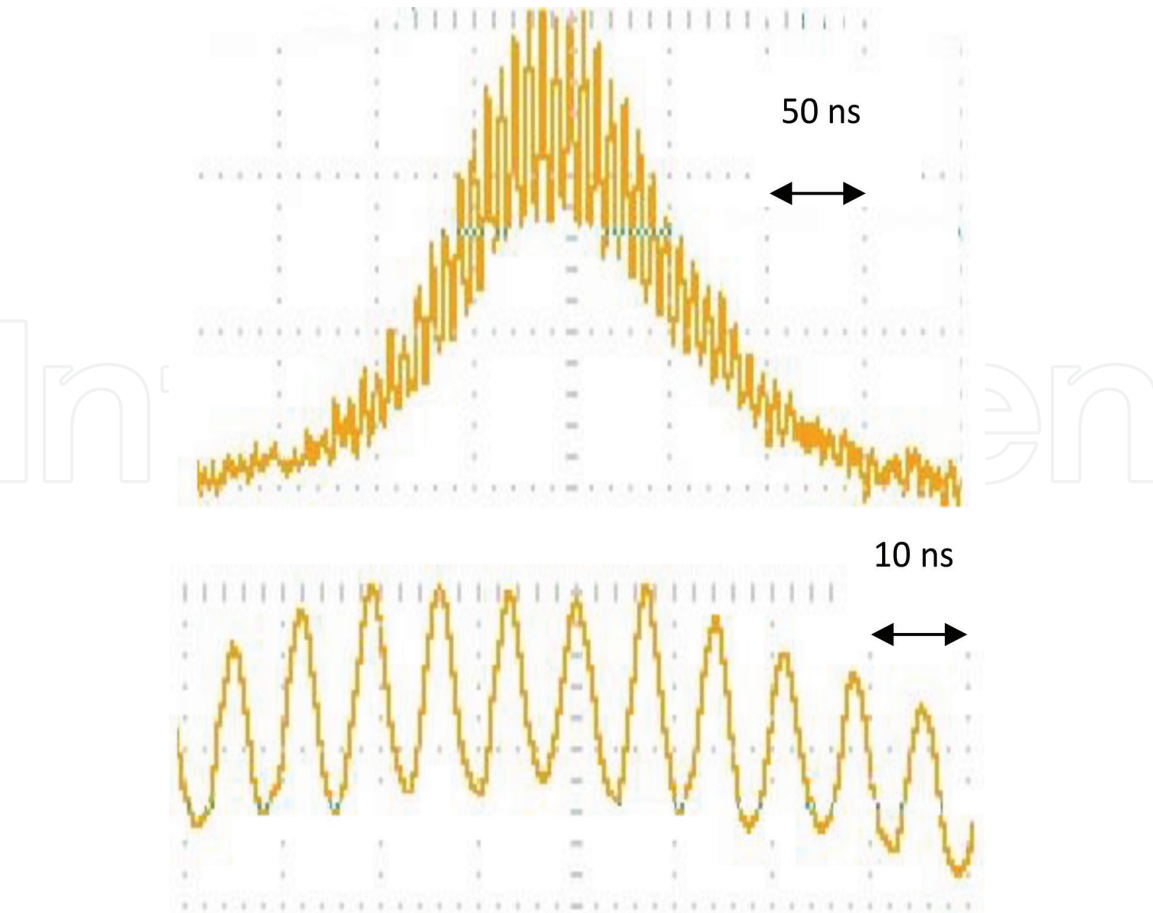


Figure 3.
Typical temporal profile of the emission of the pump CO₂ laser. FWHM value of ~110 ns is evident from the upper trace. The beating of two longitudinal modes at a period of ~7 ns is apparent from the lower trace. Absence of any beat at a longer period indicates operation on multi-longitudinal modes belonging to the same transverse family.

that contained the crystal and comprised of the output coupler (M_1) of the pump laser of plano-concave geometry [plane surface AR coated @ 10.6 μm and the concave

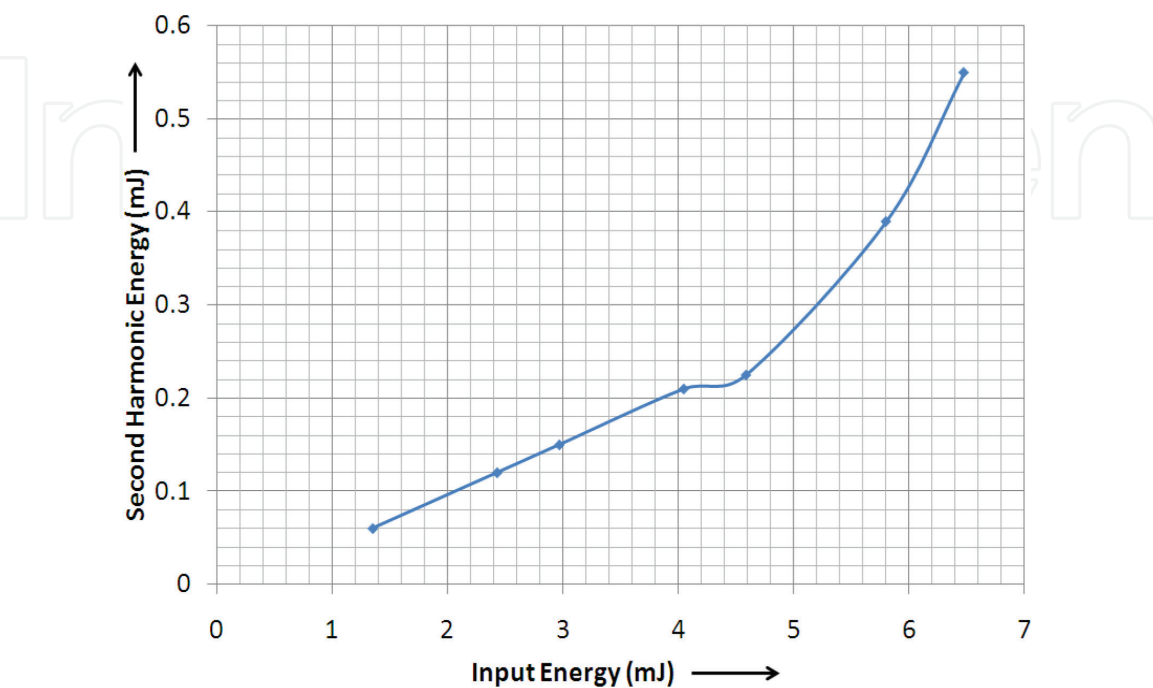


Figure 4.
Dependence of single-pass SH output on the energy of the pump pulse.

surface (7 m ROC) dielectric coated for 70% R @ 10.6 μm] and a plane ZnSe dichroic mirror M_2 (R > 90% @ 10.74 μm , T > 90% @ 5.37 μm) (refer to **Figure 2b**). The length of this external cavity (~1.21 m) was such as to push the $g_1 \times g_2$ value viz., 1.17 beyond the region of stability. There was a remarkable enhancement in the generation of SH output when M_2 was fine tuned to ascertain its parallelism with the convex face of mirror ' M_1 '. Performance of this multi-pass cavity with respect to the generation of SH was characterised by varying the pump energy incident on the crystal and measuring the corresponding energy of the SH beam emerging through ' M_2 ' (**Figure 5**). When the cavity is perfectly aligned, the pump photons coming through the Mirror M_1 are in phase, at every instant, with the fraction of the unconverted pump that is reflected off it. This increases the effective energy input to the crystal and that, in turn, results in a correspondingly increased SH output. This fact is amply clear from **Figure 4** in conjunction with **Figure 5**. It is apparent that for a maximum input pump energy of ~6.5 mJ, the single pass SH output is ~0.55 mJ (**Figure 4**) while according to **Figure 5**, the same input of 6.5 mJ gets enhanced to ~9.2 mJ due to cavity effect. The corresponding SH multi pass output is ~1.625 mJ, almost a three-fold increase when compared to the single pass case. Considering 9.2 mJ as the input energy, the SHG efficiency can be estimated to be ~17.66% - a clear ~209% improvement as against the single pass case. To be noted here that the pump energy has actually been maintained at ~6.5 mJ and therefore the conversion efficiency has risen by ~295% as a matter of fact. In these experiments, both pump and SH beams suffered significant Fresnel reflection losses during their repeated back and forth passage through the crystal that was not anti-reflection coated. Further, as the pump laser output coupler M_1 is only 23% reflective at 5.35 μm , a major part of the SH generated in the reverse direction escapes through this mirror. The dramatic improvement in the SH conversion efficiency that has been obtained in the multi-pass case is thus by no means an optimised one. Increasing the reflectivity of the rear mirror at the SH wavelength in addition to employing a crystal with broadband anti reflection coating on both its entrance and exit faces should be able to fully exploit the decided advantage of a multi-pass case. We also note here that this scheme does not suffer from the conventional single pass walk off [16] between the pump and the SH beams as mirror M_2 is almost transparent to the SH beam thereby providing feedback only to the pump beam. As the second

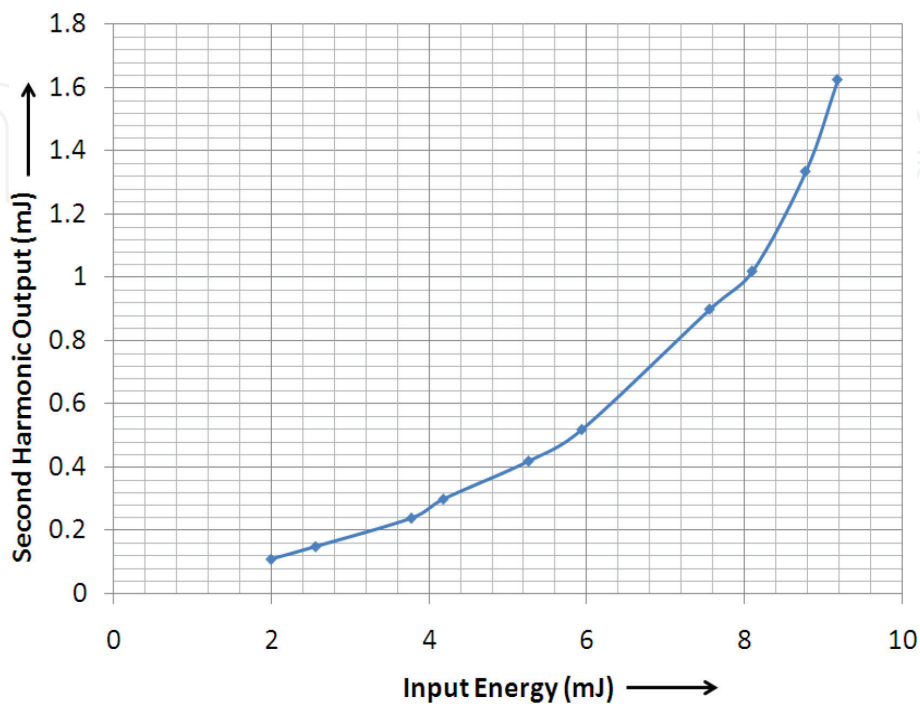


Figure 5.
Dependence of multi-pass second harmonic output on the effective input pump energy following cavity effect.

harmonic beam generated in the forward direction alone is extracted in this configuration, its spatial quality is practically same as that of a single pass case. Therefore no special effort was expended to monitor the spatial quality of the SH beam. However, the visual observation of a clear well defined spot when the generated beam was focussed by a 10 cm focal length CaF_2 lens on a graphite plate bore testimony to its satisfactory spatial character.

Towards comparing the SH power conversion efficiency in the single and multi-pass cases, we monitored the temporal profiles of the pump and the corresponding SH pulses with the external cavity in aligned and misaligned conditions. In order to obtain smooth temporal profiles devoid of mode beating, we captured the power profiles in all the four cases with oscilloscope set in bandwidth limited mode and the same are displayed in the traces of **Figure 6** from where the single pass internal peak power SH conversion efficiency can be readily estimated to be $\sim 10.48\%$. The power conversion efficiency is thus greater than the energy conversion efficiency (8.45%) of the SHG process. This is because the peak power always exceeds the average intra-pulse power of the pump beam and higher is the intensity at the pump wavelength, better is the SH conversion. This observation is in general concurrence with the finding of several researchers [13, 17, 18]. When the cavity is perfectly aligned, the photon flux at the entrance face of the crystal comprises of two components at any point of time; (i) the photons constituting the output of the pump laser and (ii) the photons constituting the fraction of the unconverted pump beam that is reflected off the convex surface of the output coupler of the pump laser. When the cavity is aligned, these two components fall in step and an overall rise in the power level of the input pulse is thus the end result. A comparison of the input power profile traces for aligned and misaligned conditions as recorded in **Figure 6** clearly substantiates this fact. The rise in the input power level, in turn, leads to an enhanced SH conversion yielding a peak power conversion efficiency of $\sim 22.36\%$, more than twice that is possible by single-pass conversion. Actually though, since the pump laser output has remained the same for both the aligned and misaligned cases, the effective SH peak power conversion efficiency stands at 35.8%, a neat enhancement of 341% due to the cavity effect.

In the next set of experiments, we captured the temporal profiles of the pump and the corresponding cavity enhanced second harmonic pulses by setting the oscilloscope at its highest bandwidth (Tektronix MSO 3054) and the same are depicted in **Figure 7a**. That the emission of the pump laser is on multimode is evidenced by the rich modulation present in the temporal profile of the pump as well as the corresponding SH pulses. The lower trace of **Figure 7b** depicts the time expanded temporal profile of the pump pulse where an oscillation of period ~ 7 ns arising out of the beating of two longitudinal modes, matching with the round trip

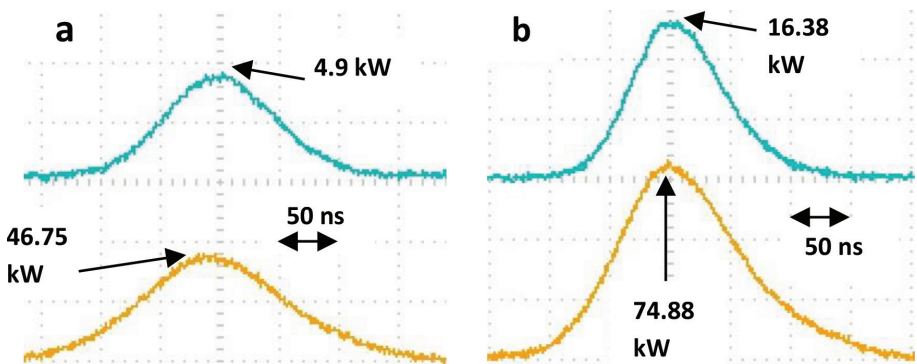


Figure 6.
Temporal profiles of the fundamental (bottom trace) and the corresponding second harmonic (top trace) captured in bandwidth limited mode; a: Single pass conversion, b: Multi-pass conversion.

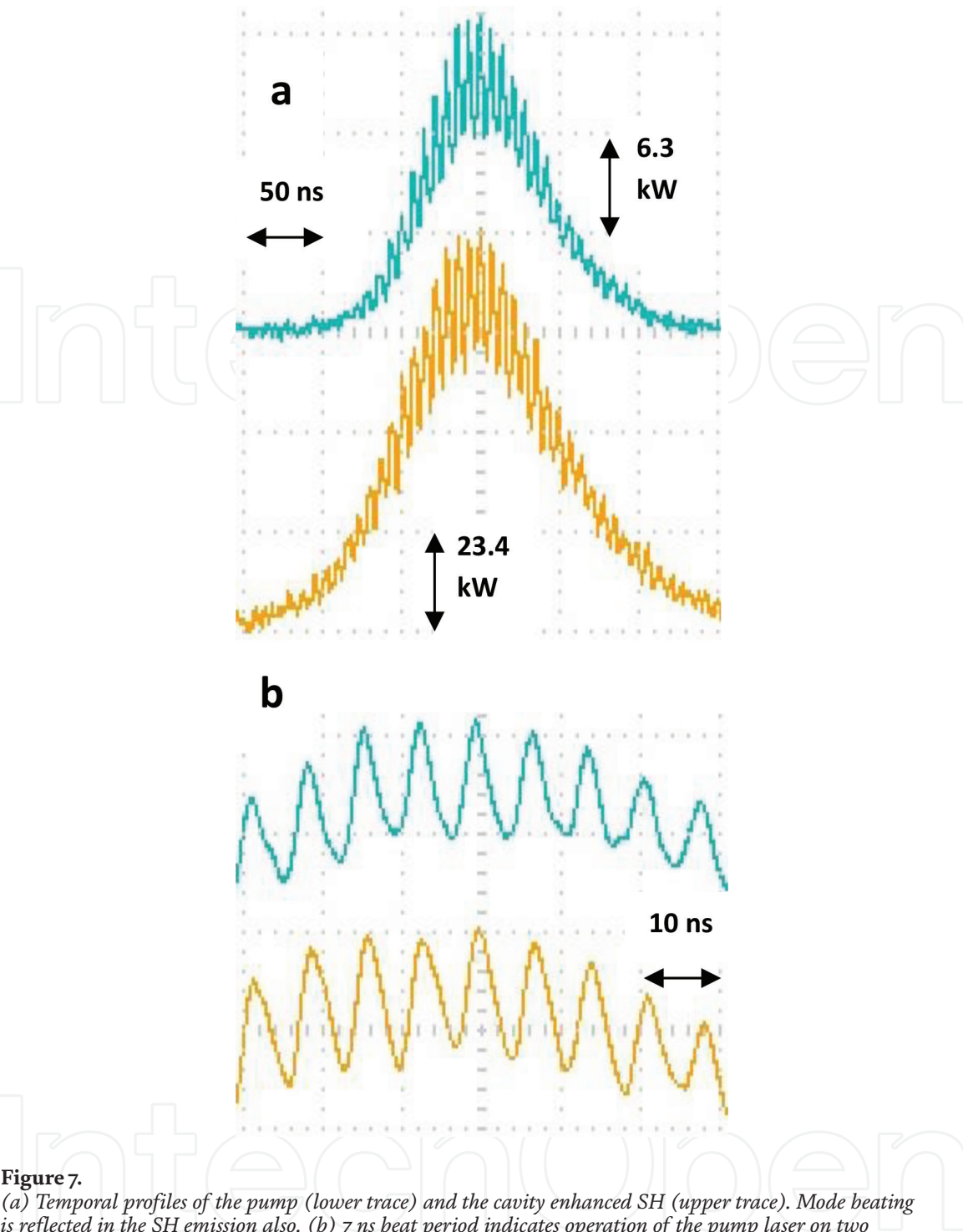


Figure 7. (a) Temporal profiles of the pump (lower trace) and the cavity enhanced SH (upper trace). Mode beating is reflected in the SH emission also. (b) 7 ns beat period indicates operation of the pump laser on two longitudinal modes (lower trace), the same is also reflected in the SH pulse (upper trace).

of 105 cm long cavity, is seen. Upon comparison with the typical temporal profile of the emission of the pump laser (**Figure 3**), it becomes obvious that the integration of the pump laser with this external unstable cavity does in no way bring in any new feature in its temporal profile or alter the beat period. This clearly implies that the coupling of the external cavity with the pump cavity has no effect on the dynamics of the pump laser. A comparison of the time expanded second harmonic temporal profile (upper trace of **Figure 7b**) with that of the temporal profile of the pump (lower trace of **Figure 7b**) readily establishes their phase and amplitude synchronisation: a signature of the instantaneous nature of the SHG process.

2.3 Conclusion

A thoughtful integration of a stable pump cavity with an unstable external Fabry-Perot cavity has resulted in remarkable enhancement in the SH conversion efficiency

even in case of pulsed operation of the laser. Although coupled external resonant enhancement has found application for the non-linear conversion process in the cw operation of the pump laser over visible region, it has not gained popularity in the mid-infrared (MIR) region owing to the possibility of damage to the MIR crystals that are not only expensive but also scarce. An unstable cavity that has the intrinsic ability to limit the intra-cavity flux there by safe-guarding the crystal from optical damage even in case of pulsed operation has been shown to offer a practical solution to this problem. We note here that the performance of this scheme can be further improved by employing a crystal with its end faces broad band AR coated, appropriate choice of the cavity parameters and control of cavity lengths. Although feasibility of this scheme has been demonstrated in the case of SHG in the MIR region, the same should, in principle, be valid for application across the near IR, visible and the UV regions of the electromagnetic spectrum as well for both cw and pulse operations.

3. Enhancing the SH conversion efficiency by non-uniform illumination of the non-linear medium

As explained in the introduction to this chapter, if a non-linear crystal can in some way be subjected to alternate high and low regions of pump intensity along its conversion length the conversion efficiency can be shown to increase 100% as against the case of conventional uniform illumination maintaining the same average intensity. We provide experimental validation of this hypothesis wherein a significant enhancement in the SH conversion efficiency has been achieved by subjecting the crystal to non-uniform illumination. Such a situation could be realised by shining the crystal from both ends as against the conventional operation of illuminating it from one end. This was readily possible by placing the crystal inside a Fabry Perot cavity wherein the interference of the forward and the reverse beams creates a periodic intensity modulation along its length. The coherent input beam was derived from the emission of a high pressure CO₂ laser while an AgGaSe₂ crystal was made use of to affect its frequency doubling. Subjecting the crystal to alternate high and low intensity of coherent pump radiation requires placing it inside a high 'Q' cavity that, at the same time, should allow significant transport of the pump energy into it. As in the previous case, integration of the pump laser cavity with the external Fabry-Perot cavity allowed efficient transport of the pump beam into the crystal while at the same time maintaining high Q of the external cavity at the pump wavelength. The only work that we came across and that explicitly connects SHG with cavity interference, albeit with a totally different central theme, is of Wu and Kimble [19] wherein two fundamental beams generate one or two SH coherent beams under non-collinear phase matched condition and the focus has been to study the phase dependence of the pump and the generated waves.

3.1 Experimental

The experimental system utilised here is identical to the one used towards increasing the interaction length between the pump and the non-linear medium and the same would therefore, not be repeated here, and the reader may refer to **Figure 2** of Section 2.1 and its description therein instead. To be noted here that **Figure 2a** depicts the case of uniform illumination while **Figure 2b** represents the case of non-uniform illumination. The CO₂ laser was tuned to the 10P(32) line giving rise to emission at 10.72 μm and consequently phase matching for frequency doubling was found to occur for an external angle of incidence of $\sim 36^\circ$. The cross-section of the pump beam on the crystal entrance face was restricted to ~ 5.0 mm diameter that allowed its clear passage through the crystal. Although the crystal

was AR coated over broad range covering 5–10 micron on both input and exit faces for normal angle of incidence (AOI), the small Fresnel reflection from the entrance face of the crystal, that was inevitable at oblique AOI, was utilised to monitor both energy and temporal profile of the pump pulse. The energy and temporal profile of the SH beam were monitored after blocking the residual pump beam, that also emerged with it through the exit face of the crystal, by a sapphire plate. By virtue of its multi-atmosphere operation, the CO₂ laser possessed intrinsically very high gain and thus delivered a pulse of relatively short duration (FWHM ~110 nsec).

3.2 Results and discussion

Towards finding the efficiency of the SHG process as a function of the pump energy for the conventional case of uniform illumination (**Figure 2a**), we gradually increased the input and monitored the corresponding SH energy and the dependence is as shown in **Figure 8**. The maximum SH energy conversion efficiency can be estimated from this figure as ~8.0%.

In the next set of experiments we subjected the crystal to alternate regions of high and low intensities along its length. This was readily possible by constructing a Fabry-Perot cavity comprising of the output coupler of the pump laser 'M₁' (R ~80%@10.72 μ m, T ~20%@5.36 μ m) and a plane dichroic mirror 'M₂' (R > 90%@10.72 μ m, T > 90%@5.36 μ m) located at the exit end of the crystal (refer to **Figure 2b**). The pump energy incident on the crystal, as measured by Detector D₁, showed a dramatic increase as 'M₂' was fine tuned to establish its parallelism with 'M₁', resulting, in turn, in a corresponding improvement in the measured SH output. In effect, there are now two inputs to the crystal; (a) Forward Input: the actual input on the entrance face in the forward direction that comes directly from the pump laser and (b) Reverse Input: the pump, that stays unconverted after its passage through the crystal, gets reflected off 'M₂' and shines on the exit face of the crystal from the opposite direction. When the cavity is perfectly aligned, the interference of these two components creates alternating nodal and anti-nodal intensity regions inside the cavity and partly contributes towards the observed dramatic enhancement of SH conversion by the crystal. At every instant, the reverse

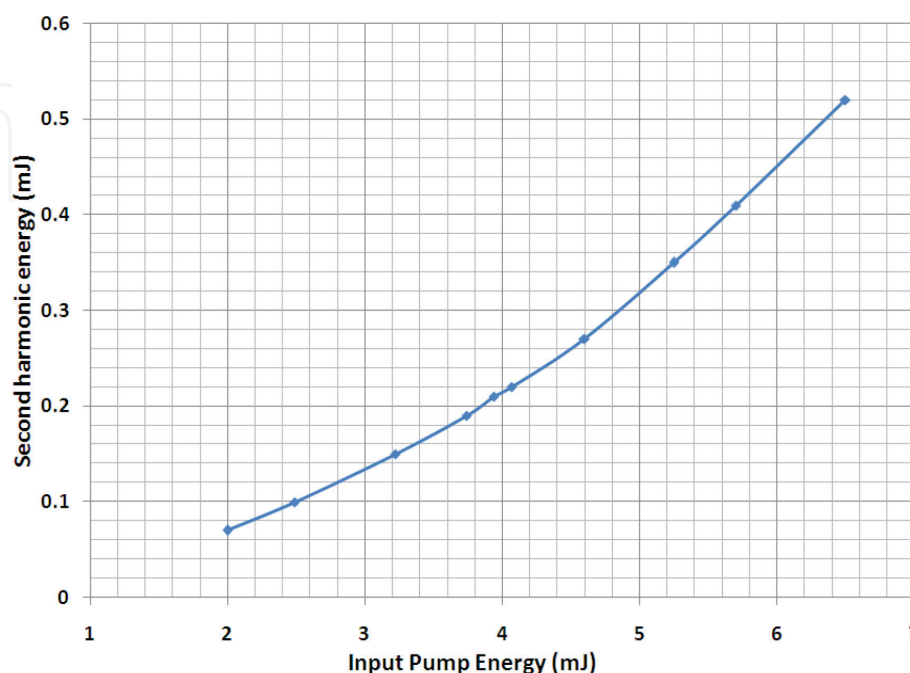


Figure 8. Second harmonic output as a function of the input pump energy in the conventional operation wherein the crystal is uniformly illuminated by the pump beam along its conversion length.

component, after traversing through the crystal, is reflected off M_1 and falls in step with the pump photons emerging through it resulting in an effective increase in the energy incident on the entrance face of the crystal as measured by the detector D_1 . At this point, towards gaining a deeper insight into this process, we gradually varied the pump (forward) input and measured both, the corresponding reverse input and the generated SH. The difference in the energy measured by D_1 with M_1 aligned and misaligned gives the measure of the reverse input. **Figure 9** depicts the dependence of the reverse input on the forward input to the crystal while **Figure 10** shows the SH output as a function of the total effective input to the crystal which is now the sum total of the forward and the corresponding reverse components. It is apparent from **Figure 9** that the reverse input does not exactly bear a linear relationship with the forward input and this behaviour owes its origin to the square dependence of the SH output on the intensity of the input at the fundamental wavelength as is evident from **Figure 8**. The square dependence basically means that as the pump intensity rises, increasingly higher fraction of it gets converted into SH and thus less of it is left to constitute the reverse input to the crystal. This explains the observed departure from the linear dependence of the reverse input on the forward input to the crystal.

The increase in the effective input to the crystal in case of an aligned cavity due to addition of forward and reverse components leads to the generation of higher SH output as revealed in **Figure 10**. For instance, the maximum pump input of 6.5 mJ in case of uniform illumination (**Figure 8**) gets enhanced to 10.34 mJ (**Figure 10**) in the aligned cavity condition giving rise to almost 2.54 fold increase in the SH conversion efficiency. However a closer examination of **Figure 10**, in conjunction with **Figure 8**, reveals a wealth of information, hitherto unexplored, that constitutes the central theme of this study and is captured in the traces of **Figure 11**. It is clearly evident from this figure that SH output in case of non-uniform illumination of the crystal is significantly higher compared to the case of its uniform illumination even when the total input to the crystal is maintained the same. Let us consider a typical input of 4.1 mJ that in case of uniform illumination generates 0.22 mJ (refer to **Figure 8**) of SH at a conversion efficiency of ~5.36%. It can be readily estimated from **Figure 9** that this input of 4.1 mJ in case of non-uniform illumination comprises of a forward component of 2.5 mJ and a reverse component of 1.6 mJ. Thus,

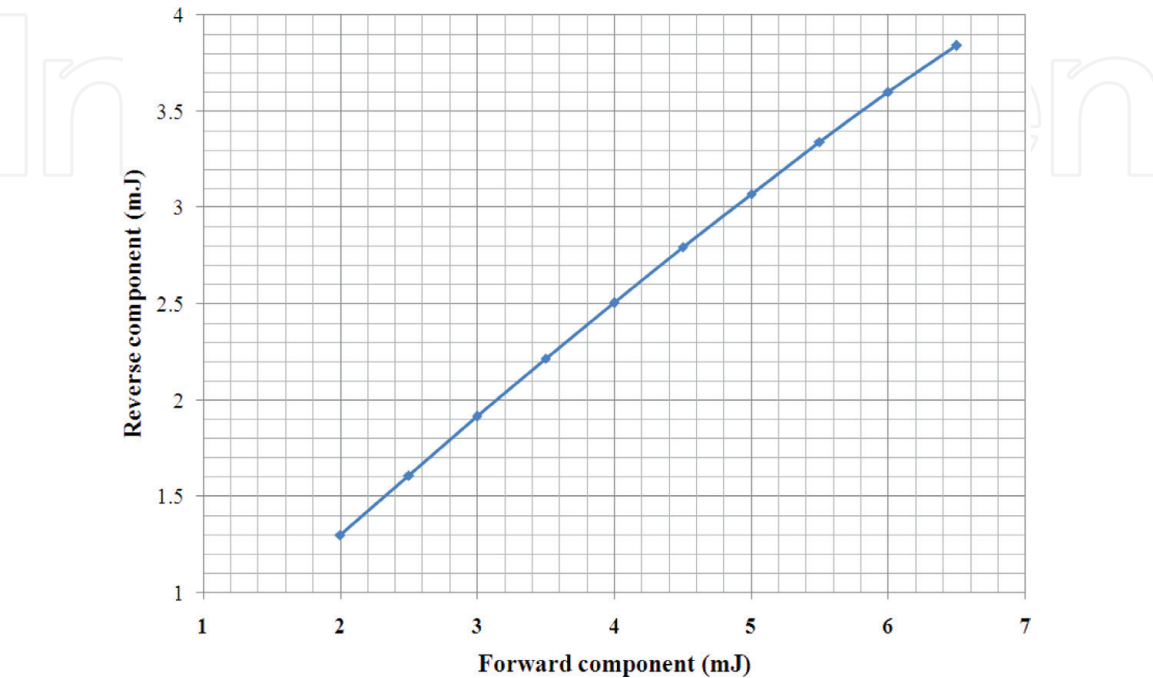


Figure 9.
Dependence of the reverse input to the crystal as a function of the forward component.

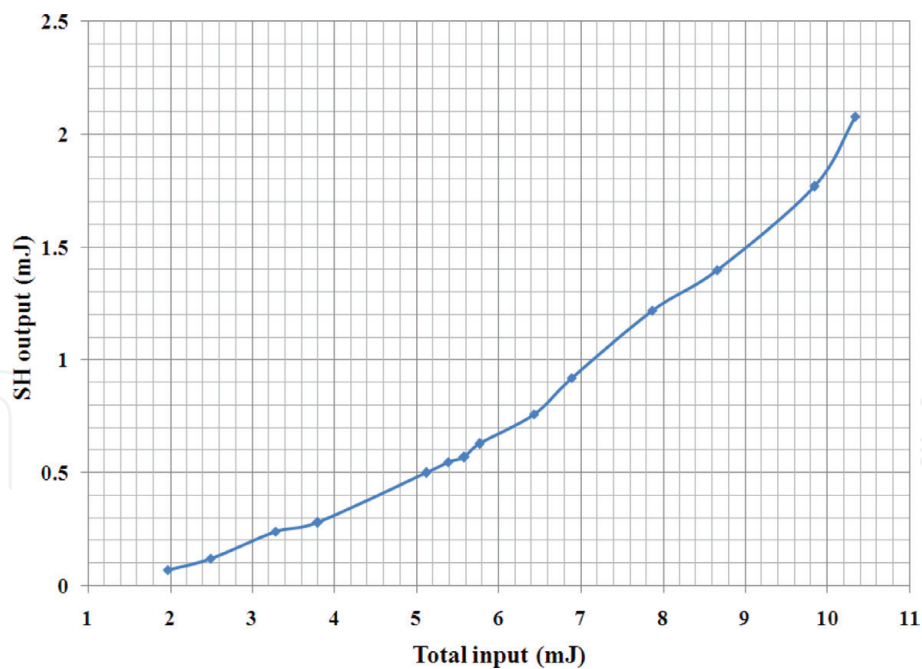


Figure 10.
Dependence of second harmonic output on the effective input pump energy in case of non-uniform illumination.

when the same total input of 4.1 mJ is made to shine on the crystal as two separate beams of 2.5 and 1.6 mJ from opposite directions by taking advantage of a cavity, a SH output of 0.325 mJ (refer to **Figure 10**) is generated at an efficiency of 7.93%; a clear advantage of ~48% in the SH conversion efficiency by going for non-uniform illumination. As discussed before this is attributed to the alternate high and low intensity regions seen by the crystal as a result of the interference of the forward and reverse beams travelling through the crystal in the latter case.

In order to estimate the expected advantage of the situation when the crystal is non-uniformly illuminated over the case of uniform illumination, we used the

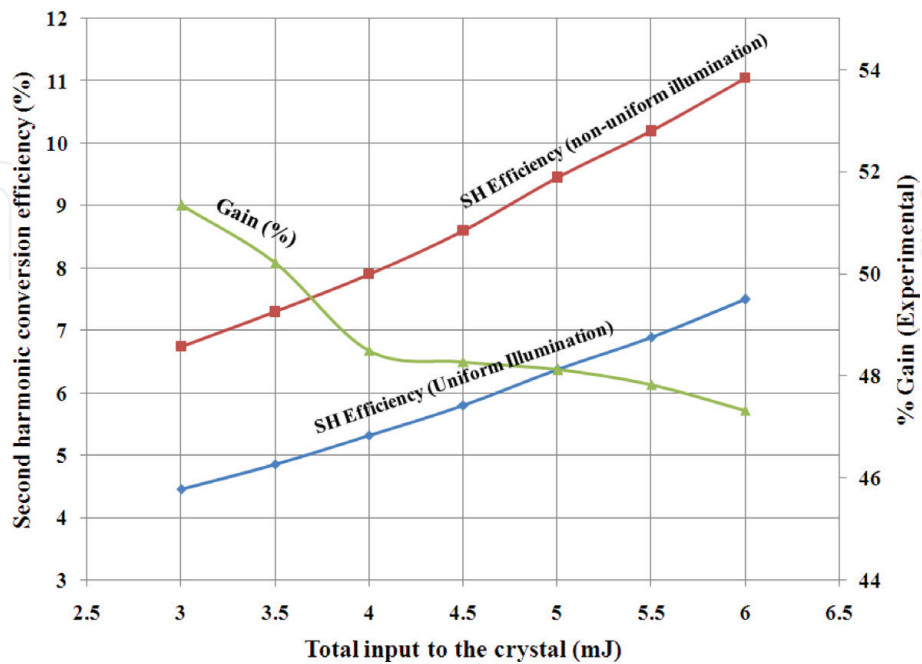


Figure 11.
Experimental SH conversion efficiency as a function of the total input to the crystal is shown for both uniform and non-uniform illumination cases. The % gain of SH conversion in case of non-uniform illumination over the uniform illumination case, defined as $[(SH_{NUI-EFF}-SH_{UI-EFF})/SH_{UI-EFF}] \times 100$, is also shown here as a function of the overall input to the crystal.

Uniform illumination		Non-uniform illumination					Overall Estimated Gain (%)*	
Input (E _{UI}) (mJ)	Estimated SH SH _{UI-EST} α E _{UI} ²	Forward input (E _F) (mJ)	Reverse input (E _R) (mJ)	Total input (E _{NUI} = E _F + E _R) (mJ)	Standing Wave Intensities			Estimated SH SH _{NUI-EST} α [E _{AN} ² + E _N ²]/2
					Anti-node (E _{AN} = E _F + E _R + 2√(E _F × E _R))	Node (E _N = E _F + E _R − 2√(E _F × E _R))		
3.33	11.09	2.0	1.33	3.33	6.59	0.07	21.72	95.85
4.14	17.11	2.5	1.64	4.14	8.18	0.09	33.46	95.6
4.61	21.25	2.8	1.81	4.61	9.11	0.11	41.50	95.29
4.94	24.40	3.0	1.94	4.94	9.76	0.12	47.64	95.23
5.9	34.81	3.6	2.3	5.9	11.65	0.14	67.87	94.97
6.53	42.64	4.0	2.53	6.53	12.89	0.17	83.09	94.86

^{*}Estimated overall gain (%) = $[(SH_{NUI-EST} - SH_{UI-EST}) / SH_{UI-EST}] \times 100$.
To be noted that the total input to the crystal in the two cases viz., uniform and non-uniform illumination has been always maintained same.

Table 1.
Estimation of the % gain obtainable in case of non-uniform illumination over the uniform illumination through reconstruction of the standing wave parameters from the experimental data recorded in **Figures 2, 3 and 4**.

data available from **Figures 8** and **10** in conjunction with the dependence of reverse input on forward input (**Figure 9**) for the reconstruction of the standing wave parameters. This is recorded in the **Table 1** above. It would be seen from this table that the advantage expected for the non-uniform illumination shows a definite reduction, although very marginal, with increasing input intensity. This reduction is because, with increasing intensity, E_R/E_F gradually reduces as is evident from **Figure 9** and discussed earlier. The experimentally measured advantage also recorded in **Figure 11** as a function of input intensity shows the same trend. The experimentally measured advantage of the non-uniform illumination, however, is seen to be considerably lower than the estimated value. This is due to the fact that a major fraction of the SH generated in the reverse direction escapes through the output coupler 'M₁' of the pump laser. Usage of a coupler that offers high reflectivity at both fundamental and generated wavelengths will help square the full advantage of the non-uniform illumination case.

To be noted here that the enhancement in the second harmonic conversion efficiency achieved by way of placing the non-linear medium inside a cavity, basically comprises of two components arising out of: (i) increased effective length of interaction between the pump and the non-linear medium and, (ii) non-uniform illumination of the non-linear medium. The above study helps decouple these two components. In the above example where the input was maintained at 4.1 mJ for both uniform illumination (meaning $E_F = 4.1$ mJ and $E_R = 0$) and non-uniform illumination (meaning $E_F = 2.5$ mJ and $E_R = 1.6$ mJ), the added advantage arising out of increased interaction length has been annulled. Thus the enhancement in the SH conversion efficiency (viz., ~70%) is entirely attributable to the modulation of intensity arising out of interference of forward and reverse beams. In case of a non-uniform illumination with $E_F = 4.1$ mJ, the corresponding $E_R = 2.6$ mJ as evident from **Figure 9**. The SH output now is 0.85 mJ as against 0.22 mJ for uniform illumination and the advantage gained here comprises of both the above components. From the discussion above it is amply clear that the component of gain due to increase in interaction length between the pump beam and the non-linear medium is ~126%. The modest gain obtained due to non-uniform illumination of the active medium is attributable to the inequality of the forward and the reverse components in the present study.

4. Conclusion

In conclusion, we conceived the advantage in SH generation by a nonlinear crystal when it is illuminated with alternate high and low regions of intensity along its length as against the conventional case of its uniform illumination with the same average intensity. Exploitation of interference effect by placing the crystal inside a Fabry Perot cavity has allowed the imposition of such a non-uniform illumination condition on to the crystal along its conversion length. The decided advantage of the non-uniform illumination over uniform illumination has been experimentally established under conditions of equal intensity exposure in the two cases. We believe that this advantage was always present in intra-cavity or resonantly enhanced frequency doubling generation processes but stayed unrecognised as the motivation of these works was to enhance the conversion efficiency by increasing the effective interaction length of the crystal and the advantage gained was thus automatically attributed in totality to this. Carefully planned experiment here has allowed us to decouple the advantage due to interference (as seen in **Figure 11**) from the total advantage as recorded in the data of **Figure 10** that also included the gain due to increased interaction length. While we have achieved the spatial variation of intensity by the exploitation of interference effect, we do not rule out the


possibility of achieving the same effect by some other means, e.g., a train of ps or fs mode locked pulses will manifest as spatial intensity variations in the sub mm to sub-micron scale appropriate to derive this advantage in a crystal of finite length. Advantage can be derived from even chaotic pulse trains wherein the temporal oscillations occur in the similar time scales as above. However, it is to be noted that the restriction on the maximum period of the spatial variation of the intensity is imposed by the crystal thickness while there is no restriction on the minimum period. As a matter of fact smaller is the periodicity of bright and dark intensity regions, better will be the heat diffusion and thus will be preferred from the point of view of handling higher intensity.

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