Self-Phase-Matching Mechanism for Efficient Harmonic Generation Processes in a Ring Pump Beam Geometry

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A new type of phase matching based on the ring-shaped pump beam for efficient harmonic generation is observed. The ring-shaped beam is shown to provide self-organized phase matching for a wide range of refractive index variation, which may automatically compensate the spatially nonuniform phase mismatch created by the optical Kerr effect. Self-phase-matching is expected to be very efficient for frequency tripling of broadband and/or ultrashort pulses, as well as for high order harmonic generation. The high conversion efficiency $\sim 1.5\%$ of 1.06 μm radiation tripling in a pure Rb vapor for a very short interaction length ~ 4 cm is obtained, when a disk-shaped beam has been replaced by a ring-shaped beam.

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It is well known that the efficiency of any frequency mixing and harmonic generation processes is strongly dependent on the phase-matching condition $\Delta\mathbf{k}=\sum_i \mathbf{k}_i=0$. There are two types of phase matching known to date: (a) the collinear phase matching when the difference between the phase velocity of fundamental and created waves is compensated either by birefringency of nonlinear crystal [1] or by the mixture of atomic vapor with the positively dispersive buffer gas [2]; and (b) the noncollinear phase matching when the wave vectors of fundamental and created waves are propagated under a certain angle, and is widely used for various CARS (coherent anti-Stokes Raman spectroscopy) techniques [3]. It should be mentioned that the ring-shaped beam geometry has been utilized in an unstable resonator spatially enhanced detection CARS [4] technique in order to simplify the alignment and improve the spatial resolution. The general feature of all these phase-matching mechanisms is their exceptional sensitivity to the input parameters' variation (wave frequencies and inclination angles, vapor and buffer density, etc.).

In this Letter the theoretical analysis and experimental demonstration of a new mechanism of phase matching for the harmonic generation processes in a centrosymmetric medium is provided. Actually, this mechanism is shown to be insensitive to the input parameters' variation.

Suppose we use for the third harmonic generation the ring-shaped pump beam, instead of the disk. Holding on the cone angle $\Theta=d/f$ (d is the ring diameter and f focal length of the lens) we consider the phase-matching condition for fixed point A with two arbitrary points B,B^* situated symmetrically around point A. The position of these points is determined by the angle Φ at the ring plane [Fig. 1(a)]. The phase-matching condition $\Delta\mathbf{k}=0$ along the z axis and at the ring plane is

$$\begin{aligned} &3k(\omega)\cos\Theta/2=k(3\omega)\cos\phi/2\,, \\ &k(\omega)\sin\Theta/2(1+2\cos\Phi)=k(3\omega)\sin\phi/2\,. \end{aligned} \tag{1}$$

Because of the pump beam axial symmetry the third harmonic (TH) beam will also be ring shaped with the cone angle ϕ, Eq. (1). Assuming the angles Θ,ϕ to be small ($\Theta,\phi\ll 1$) one may easily obtain the relation between ϕ and Θ as a function of vapor density (refractive index)

$$\frac{N}{N_0}=1-\left(\frac{\phi}{\Theta}\right)^2;\ \text{or}\ \phi=\sqrt{\Theta^2-8\Delta n}\,, \tag{2}$$

where $n(\omega)=1+\beta_\omega N$, $n(3\omega)=1+\beta_{3\omega}N$ are the refractive indices for the pump frequencies and TH frequencies, respectively, $\Delta n=n(\omega)-n(3\omega)=(\beta_\omega-\beta_{3\omega})N$, and $N_0=\Theta^2/8(\beta_\omega-\beta_{3\omega})$ is the maximal density value for which the phase-matching condition can be fulfilled

The second relation obtained from Eq. (1),

$$\phi=\tfrac{1}{3}\Theta(1+2\cos\Phi)\,, \tag{3}$$

determines the position of points B,B^* to be phase

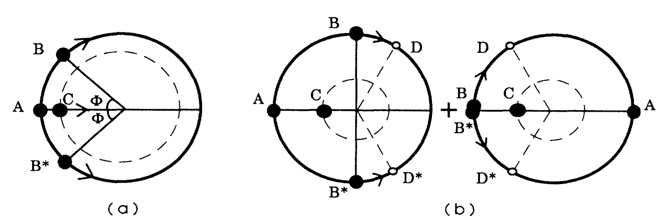

FIG. 1. The plane of input ring-shaped pump beam. (a) One photon from point A and two from points B,B^* are phase matched, creating TH at point C. When the vapor density is increased points B,B^* move towards points D,D^* (arrows on the ring) holding the phase-matching condition to be continuously satisfied. Point C (TH) consequently moves towards the ring center. (b) When points B,B^* cross the vertical ring diameter another configuration of three phase-matched points appears, which results in exactly the same direction of the TH photon. When points B,B^* reach the position D,D^* point C comes to the ring center.

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matched with point A [Fig. 1(a)]. Thus, the angle ϕ varies from $\phi=\Theta$, at $N\to 0$ (collinear phase matching, the points B,B^* coincide with A, $\Phi=0$), to $\phi=0$, when $N\to N_0$ [$\Phi=2\pi/3$ points B,B^* reach the points D,D^*, Fig. 1(b)].

Thus, for the ring-shaped pump beam the phase-matching condition is provided automatically within the whole range of Δn refractive index exchange,

$$0\le \Delta n \le \Theta^2/8\,, \tag{4}$$

without variation of other input parameters (buffer gas pressure, inclination angle, etc.), as is required for traditional types of phase matching. In other words, the variation of the Δn value within this range will not destroy the phase-matching condition because each point on the ring is able to find the corresponding pair of points to be phase matched (Fig. 1).

Moreover, in the range determined by

$$\Theta^2/9\le \Delta n\le \Theta^2/8,\quad 8/9\le N/N_0\le 1 \tag{5}$$

for each Δn (density) value there are two values of Φ angle available. This means that there are two different configurations of phase-matched points which result in the same direction of the TH photon. Since all points on the ring have the same phase, constructive interference between these two ways and consequently coherent enhancement of the TH signal occur (Fig. 2). The cone angle of the TH signal varies from $\phi=\Theta/3$ when $\Delta n=\Theta^2/9$ to $\phi=0$ when $\Delta n=\Theta^2/8$. In this range the rate of conversion becomes practically insensitive to the Δn value variation (Fig. 2).

Let us now consider what conclusions and advantages we may expect from such unusual properties of self-phase-matching. According to Eq. (5) the variation of Δn allowed by self-phase-matching (SPM) is ($\Delta n_{\max}=\Theta^2/8$),

$$\delta(\Delta n)_{\rm SPM}=0.12\Delta n_{\max}=1.5\times 10^{-2}\Theta^2\,. \tag{6}$$

For ring geometry the length of the interaction with the

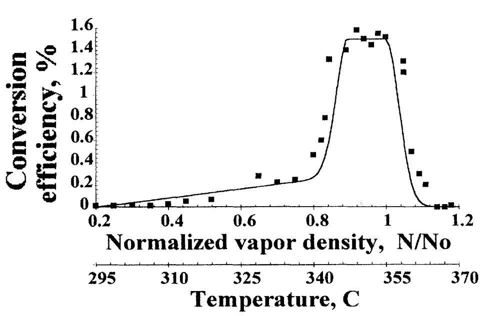

FIG. 2. Measured third harmonic (0.35μ) energy conversion efficiency vs Rb vapor density (temperature): $I_\omega\sim 4\times 10^{10}$ W/cm^2, $L_{\rm int}\sim 4$ cm, $d_0\sim 0.4$ mm, $\Theta\sim 20$ mrad, N_0(Rb)$\sim 7\times 10^{16}$ cm^{-3} ($P\sim 4.4$ torr). Theoretical curve (solid line) is based on Eq. (7) (interference effect is taken into account).

medium is determined by input angle Θ, lens focus, and ring thickness a: $I_{\rm ring}=8f\lambda/\pi a\Theta$. Substituting it into Eq. (6) we obtain [for the collinear phase matching, $\delta(\Delta nL)_{\rm coll}=\lambda/6$] $\delta(\Delta nL)_{\rm SPM}=(d/25a)\lambda$. Taking $a=0.1$ cm, $f=100$ cm, and $d=2$ cm ($\Theta=20$ mrad) we obtain a substantial enlarging of the phase-matched width, $\delta(\Delta nL)_{\rm SPM}/\delta(\Delta nL)_{\rm coll}\sim 5$, which is further shown to have crucial significance.

The TH generation (THG) conversion efficiency has the form (in a plane wave and small signal approach [5])

$$\eta=\left[\frac{12\pi^2\omega}{c^2}\chi_T^{(3)}(3\omega)NLI_\omega\right]^2\sin^2\frac{\Delta kL/2}{(\Delta kL/2)^2}\,, \tag{7}$$

where $\chi_T^{(3)}(3\omega)$ is the third-order susceptibility responsible for the TH generation process. I_ω—pump beam intensity (in W/cm^2), N—vapor density. The maximal value of the product $(NI_\omega L)$ is determined by the phase-matching condition, Eq. (6), where the expression for the nonlinear part of the refractive index should be substituted

$$\delta n(I)=n_2|E(\omega)|^2=1.3\times 10^{-2}\chi_S^{(3)}(\omega)I_\omega N\,, \tag{8}$$

where $\chi_S^{(3)}(\omega)$ is the nonlinear susceptibility responsible for the Kerr effect. Comparing the ring and collinear regimes one obtains the same enlarging of the ratio $(NI_\omega L)_{\rm SPM}/(NI_\omega L)_{\rm coll}\sim 5$.

The main problem limiting the energy conversion efficiency of THG has been recognized as the phase mismatch induced by the nonlinear part of the refractive index [5,6]. This influence is twofold (a) because of the nonuniform transverse intensity distribution, which does not allow us to fulfill the phase-matching condition simultaneously for whole spatial parts of the beam, and (b) because of the variation of the $\Delta n(I)$ term during the propagation and intensity conversion.

The substantial enlarging of the allowed range of the Δn value variation is expected to result in the following:

(1) Self-organization and self-compensation of the spatiotemporal phase mismatch induced by the nonlinear part of the refractive index (Kerr effect [5,6]). It is obvious that such self-compensation will be followed by the angular broadening of the TH beam cone angle. Thus, from Eq. (2) $\Delta\phi\sim 4\delta(\Delta n)/\phi$, which for the experimental data used ($I_\omega\sim 4\times 10^{10}$ W/cm^2, $N\sim 8\times 10^{16}$ cm^{-3}, $\phi\sim 6$ mrad) will provide $\phi\sim 4$ mrad, and instead of ring narrowing described before, the empty part of the cone will be filled in, if $\phi\le 4$ mrad.

(2) The self-phase-matching increases substantially the spectral bandwidth acceptance for efficient and nondestructive THG processes. In order to show this one should take into account the frequency-dependent linear part of the refractive index, $\Delta n_0=2\pi{\rm Re}(\chi_\omega^{(1)}-\chi_{3\omega}^{(1)})\sim 1/\Delta_{10}$, where $\Delta_{10}=\omega_{10}-\omega$ is the one-photon detuning. Using the expression $\delta(\Delta n)=d(\Delta n)/d\omega\,\delta\omega=\Delta n_0\delta\omega/\Delta_{10}$ and substituting it into Eq. (6) one obtains

$$(\delta\omega)_{\mathrm{SPM}} \sim 0.12\Delta_{10}, \quad (\delta\omega)_{\mathrm{coll}} \sim \lambda\Delta_{10}/6L\Delta n_0 . \tag{9}$$

Using the following data, $\Delta n_0(\mathrm{Na}) = 3\times10^{-22}N$, $\Delta n_0(\mathrm{Rb}) = 7\times10^{-22}N$, and $\Delta_{10}(\mathrm{Na}) = 7576\ \mathrm{cm}^{-1}$, $\Delta_{10}(\mathrm{Rb}) = 3340\ \mathrm{cm}^{-1}$ [5,6], and taking $N(\mathrm{Na,Rb}) = 8\times10^{16}\ \mathrm{cm}^{-3}$ (5 torr), we may estimate the spectral bandwidth acceptance for the frequency tripling process for self- and collinear-phase-matching ($L = L_{\mathrm{SPM}} \sim 10$ cm),

$$\begin{aligned} &\mathrm{Na:}\ \ \Delta\omega_{\mathrm{coll}} \sim 500\ \mathrm{cm}^{-1};\ \ \Delta\omega_{\mathrm{SPM}} \sim 900\ \mathrm{cm}^{-1}, \\ &\mathrm{Rb:}\ \ \Delta\omega_{\mathrm{coll}} \sim 100\ \mathrm{cm}^{-1};\ \ \Delta\omega_{\mathrm{SPM}} \sim 400\ \mathrm{cm}^{-1}. \end{aligned} \tag{10}$$

It is important to mention that [Eq. (9)] the ring spectral bandwidth acceptance is actually independent of the vapor column density, NL. In order to estimate the minimal pump pulse length which can be triplet one should calculate the walkoff in the medium (the difference between group velocity of pump and TH frequencies) $\Delta\tau = L(1/u - 1/c)$, where $u^{-1} = dk(\omega)/d\omega[dn(3\omega)/d\omega = 0]$. Thus, $\Delta\tau = L\Delta n_0\omega/c\Delta_{10}$. Substituting into this expression $L = L_{\mathrm{ring}}$ and Δn_0 values, one has $\Delta\tau\delta\omega_{\mathrm{SPM}} \sim \pi$. Thus, the minimal pulse length can be easily estimated by $\delta\omega_{\mathrm{SPM}}$ spectral bandwidth acceptance.

(3) We expect self-phase-matching to be useful for high harmonic generation processes because the collinear phase-matched allowed Δn variation $\delta^{(m)}(\Delta n) \sim \lambda/2mL$ (m is the order of the nonlinear process), and consequently the bandwidth acceptance, becomes in this case even narrower.

(4) Automatic fulfillment of the condition $b\delta(\Delta k) = 2$, required for a focused Gaussian beam (b is the confocal parameter [7]). According to Eq. (6) $0 \le \delta(\Delta k) \le 1.2$, thus the condition $b\delta(\Delta k) = 2$ may be fulfilled for $1 \le b \ll \infty$.

The preliminary experimental verification of the ring type phase matching for efficient third harmonic generation has been carried out with pure Rb and Na vapors. The new sapphire, sealed-off cell, $d = 15$ mm and $l = 16$ cm, has been utilized (Fig. 3). This kind of cell does not require either cooling or pumping, and might be heated up to 1000 °C, providing the homogeneous (ΔT along the cell varies within several degrees) and high density $P \sim 1000$ torr column of atomic vapor [8]. We used the Quantel mode-locked Nd:YAG laser ($\lambda = 1.064\ \mu$m), which could provide an output pulse energy up to 30 mJ and a pulse length $\tau \sim 30$ ps. The central part of the output radiation with $d = 11$ mm was blocked by circle shield $d = 9$ mm. The ring with thickness 1 mm was focused ($f = 50$ cm) into the cell with pure Rb(Na) vapor. The beam diameter in the waist was 0.4 mm, the length of interaction measured without the cell was $l_{\mathrm{int}} \sim 4$ cm, the input cone angle $\Theta \sim 20$ mrad, and the pulse energy did not exceed 4 mJ. The filters KG-5, UC-1 are used to cut ir and visible radiation. The first photodiode PD1 has measured the input 1.064 μm intensity (Molectron detector). Photodiode PD2 has been calibrated by TH from the nonlinear crystal, measured by the Molectron detector.

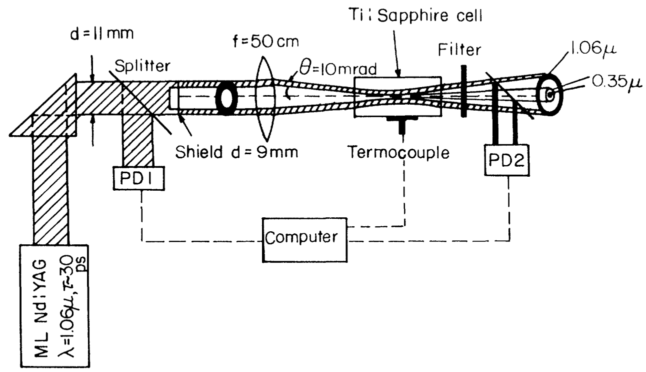

FIG. 3. Disk/ring-shaped pump beam experimental setup for THG in an alkali metal vapor. The pump beam $d = 11$ mm was blocked by the shield $d = 9$ mm getting out the ring with thickness 1 mm, which is focused ($f = 50$ cm) into the sealed-off sapphire cell with pure Rb (Na) vapor inside. The photodiodes PD1 and PD2 measure the input $\lambda = 1.06\ \mu$m and output $\lambda = 0.35\ \mu$m pulse energies, respectively. The filter cut the ir radiation. The inner ring- and disk-shaped UV radiation is related to the vapor density variation and several types of the phase-matching condition fulfillment.

First, the temperature dependence of the TH beam spatial evolution in Rb vapor was investigated. For $T = 290$ °C ($P = 0.9$ torr) the weak TH ring propagating along with the pump radiation (collinear phase matching) appears. Between $T = 290$ °C and $T = 348$ °C ($P = 3.9$ torr) we observe the TH signal to gradually increase in intensity and decrease in cone angle. For $T = 348$ °C ($P = 3.9$ torr) the sharp amplification of the TH intensity with cone angle $\phi \sim 7$ mrad is observed, which precisely corresponds to the condition of Eqs. (2) and (5). The further temperature growth is followed by filling in the empty part of the ring, without a significant decrease in the outside ring diameter. For $T = 362$ °C ($P = 4.5$ torr) the ring is transformed into the disk with cone angle $\phi \sim 5$ mrad. Such broadening of the TH angular distribution being in good agreement with theoretical consideration is a clear manifestation of the dynamic refractive index exchange due to the nonlinear Kerr effect (see Fig. 1). After reaching $T = 372$ °C ($P = 5.3$ torr) the TH intensity decreases rapidly, without significant exchange of its spatial distribution. The measured rate of conversion versus vapor density (temperature) shown in Fig. 2 is in a good qualitative agreement with theory [Eq. (7)], where the interference effect $I_\omega \rightarrow 2I_\omega$ should be taken into account. The wide plato $\delta T \sim 25$ °C clearly demonstrates the range of interference and the TH signal enhancement.

A control ring/disk experiment (the shield was taken away) with the same beam diameter and same pulse energy was performed. In this case very weak temperature dependence of TH intensity and shape was obtained. In the non-phase-matched regime $\Delta kL \gg 1$ and since Δk

$\sim N$, Eq. (7) should be density (temperature) independent.

The maximal ratio $\eta_{\rm ring}/\eta_{\rm disk} \sim 4$ for Rb and 5 for Na has been achieved. It should be mentioned that for a disk-shaped beam the whole cell length ~ 16 cm was involved in the interaction. Thus, the ratio $\eta_{\rm ring}/\eta_{\rm disk}$ might be much larger for the cell about 4 cm (this is the length of overlapping for SPM).

When the pure collinear geometry phase matching has been carried out (the pump beam was first narrowed by a telescope up to $d = 0.6$ cm and then focused into the cell) the negligible TH signal has been obtained.

Comparing with the collinear phase-matching experiments for tripling with Rb+Xe, $\eta = 10\%$ [6] and $\eta = 2.8\%$ [5], for $L = 50$ cm, the great advantage of SPM is indicated. Further efficiency growth is expected by enlarging the beam diameter up to 20 mm and the focal length up to 100–150 cm, which will increase the interaction length up to 15–20 cm, giving rise to an increase in efficiency by at least the order of magnitude [9]. In order to provide it one needs the length of the cell to be $l \geq 30$ cm (with the short $l = 16$ cm cell available we could not reach these parameters because of the cell windows' damage).

In summary we have described and demonstrated the new type of phase matching for efficiency sum-frequency mixing processes (THG) based on the ring-shaped pump beam. It may hold the phase-matching condition for a wide range of refractive index variation. It may in turn lead to the self-compensation of the phase mismatch induced by the nonlinear Kerr effect. The substantial enlarging of the spectral bandwidth acceptance of the SPM enables the efficient frequency tripling of broadband ($\delta\omega \sim 500$ cm^{-1}) and ultrashort ($\Delta\tau \sim 10$–20 fs) laser pulses.

The experimental measurement of THG of $\lambda = 1.064$ μm ps radiation in a pure Rb and Na vapor has shown a factor of 4–5 of improvement, when instead of disk shaped the ring-shaped pump beam has been utilized and high conversion efficiency $\eta \geq 1.5\%$ has been achieved.

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