Simultaneous generation of sum, difference, and harmonics of two laser frequencies by spread spectrum phase matching

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We demonstrate the simultaneous generation of sum, difference, and harmonics of two laser frequencies (which can be tunable) by spread spectrum phase matching in a nonlinear $Sr_{0.61}Ba_{0.39}Nb_2O_6$ crystal. The phase matching is obtained by inducing in the crystal domain gratings with broadband periodicity. Conversion efficiencies up to $\sim 1\%$ were observed. We also give a calculation that shows the effect of the spread spectrum domains on the mixing.

The intensive recent activity in nonlinear parametric processes is motivated by their unique offer in providing light oscillations with very large tuning ranges. The study and much of the advances benefitted from many new nonlinear crystals with large nonlinear coefficients. The phase-matching requirement,[1] however, remains a critical and a limiting factor in such nonlinear processes. The common method to solve it is to use the crystal birefringence, where the difference between the refractive indices for beams with different polarizations that propagate along specific crystal directions compensate the refractive index mismatch due to dispersion. Then, a degree of wavelength tunability of the input and the generated beams can be obtained by changing the angle of the beam propagation in the crystal (and to some extent by changing the temperature). Another way to overcome the problem is quasiphase matching, where "artificial" spatial modulation of the optical nonlinearity[1] or the refractive index provides the missing **k** vector. This method restricts the phase matching to a specific input wavelength. In a recent work,[2] we have shown a new method for obtaining broadband second harmonic generation (SHG) by controllable spread spectrum quasiphase matching. It was accomplished by forming quasirandom domain gratings in $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN) crystals. The broad spectrum of the domain grating period provided the broadband capability. We have also demonstrated[3] second harmonic generation for prespecified discrete or continuous beam wavelengths by tailored quasiphase matching.

In this work we demonstrate the simultaneous generation of sum, difference and harmonics of two laser frequencies by spread spectrum phase matching in $Sr_{0.61}Ba_{0.39}Nb_2O_6$. The broad phase matching is obtained by broadband domain grating **k** vector, induced in the nonlinear crystal. Conversion efficiencies of up to $\sim 1\%$ over a broad tunable range were observed. The advantage of the method is in its capability to tailor the gratings' bandwidth to desired bands of mixing and still have the feature of adding up the generated light along significant interaction lengths. Otherwise, such a simultaneous multiband frequency mixing can be achieved only "locally" where phase matching can be violated. Then, however, the interaction length is limited and only very "thin" crystals can be used. Our method can be also used for optical parametric oscillation with very broad spectral widths (especially needed in very short pulses and continuum generation) and large tuning ranges.

The needed grating number Δk_g for quasi phase matching, in a collinear configuration of three wave mixing with frequencies ω_1, ω_2, ω_3, is $\Delta k_g = \Delta\beta \equiv (\omega_1 n_1 \pm \omega_2 n_2 - \omega_3 n_3)/c$, where $\omega_1 \pm \omega_2 = \omega_3$ (the + and − signs correspond to the sum and difference generation, respectively), $n_i = n(\omega_i)$ are the refractive indices and c is the vacuum speed of light. We use the Sellmeier dispersion relation[4] for the dependence of the refractive index on the wavelength, $n_e^2(\omega) = 1 + S_i\lambda_{0i}^2/[1-(\omega\lambda_{0i}/2\pi c)^2]$, with the parameters of Ref. 4 for $Sr_{0.61}Ba_{0.39}Nb_2O_6$: $S_i\lambda_{0i}^2 = 3.8022$, $\lambda_{0i} = 0.1962$ μm and $n_e(\omega)$ is the index of refraction for the extraordinary polarization. As in the SHG experiment[2] we use here the second-order nonlinear coefficient d_{33} (corresponds to $\chi_{zzz}^{(2)}$) which is about twice larger than $d_{31}(\chi_{zxx}^{(2)})$. Figure 1 shows the domain grating period needed for exact quasiphase matching as a function of the input wavelength λ_1 (in nm) for second harmonic, sum and difference frequency generations. The second input beam wavelength λ_2 is identical to the first one in

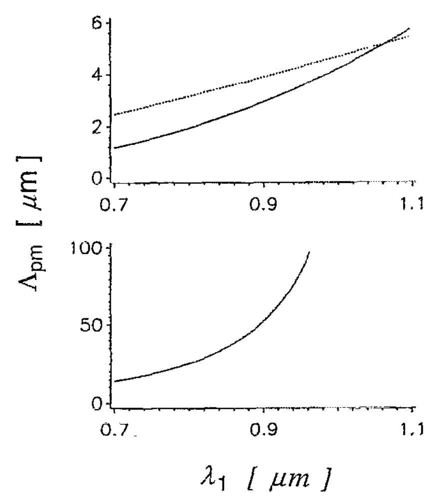

FIG. 1. The period of the domain gratings needed for quasiphase matching as a function of the input wavelength λ_1: In the upper figure, for second harmonic (solid curve) and sum frequency (dashed curve) generations; and in the lower figure, for difference frequency generation. The second beam wavelength is identical to the first one in the SHG case and equals 1064 nm in the sum and difference frequency generations cases.

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the SHG case and 1064 nm in the sum and difference frequency cases.

We give a brief analysis of the parametric mixing process in a spread-spectrum domain grating structure. The output wave can be calculated by summing up the contributions from all of the domains. These are $2N$ antiparallel domains with a c axis along the $\pm z$ direction ("transversal" domain grating). The light beams propagate along the x axis. Helmfrid *et al.*[5] and Fejer *et al.*[6] have calculated the influence of randomly varying domain lengths on the second harmonic generation. We use similar mathematical techniques and take advantage of the fact that in our case the domain lengths are highly scattered to obtain a simple analytic solution and to study its asymptotic behavior. The contribution of each domain is known[1] and we sum up all of them and evaluate the average for the suitable distribution function (of the domain boundary positions or their lengths) to obtain the overall generated output, in a similar way given of Ref. 5. Then, we have for the intensity, in the undepleted pump approximation,

$$I_{\omega_3}=\frac{C}{\Delta\beta^2}E\left\{\sum_{m=0}^{2N}\sum_{n=0}^{2N}(-1)^{m+n}a_m a_n e^{i\Delta\beta(x_m-x_n)}\right\}, \quad (1)$$

where $[x_{m-1},x_m]$ are the boundaries of the mth domain, a_n equals 1 for $n=0$ and $n=2N$ and equals 2 otherwise, $E\{\ \}$ denotes expectation, $C=[\omega_3^2\epsilon_0^2(\chi^{(2)})^2(\mu/\epsilon_0)^{3/2}/(2n_{\omega_1}n_{\omega_2}n_{\omega_3})]I_{\omega_1}I_{\omega_2}$, $\chi^{(2)}$ is the relevant nonlinear coefficient and $I_{\omega_1},I_{\omega_2}$ are the intensities of the two input waves.

We assume that the domain length is a normally distributed random variable with an average $\Lambda/2$ and variance σ. We also assume that the domain lengths are uncorrelated and that the number of domains is large [in our experiment $2N \approx 2l/\Lambda=(10\text{ mm})/(2.6\ \mu\text{m})\approx 3850$]. From Eq. (1) we can obtain

$$I_{\omega_3}=\frac{C}{\Delta\beta^2}\left((8N-2)+8\sum_{m=1}^{2N}(2N-m+1)\times e^{(-m\Delta\beta^2\sigma^2/2)}\cos(m\phi_0/2)\right), \quad (2)$$

where $\phi_0=\Delta\beta\Lambda-2\pi$ is the average phase deviation from phase matching. The domain periodicity Λ_{PM} for phase matching is given by $\Lambda_{PM}\Delta\beta=2\pi$. For the case of large variance of the domain lengths, $(2N\Delta\beta^2\sigma^2\gg 1)$, we have

$$I_{\omega_3}=\frac{8C}{\Delta\beta^2}\frac{l}{\Lambda}\frac{(1-|S|^2}{\{1+|S|^2-2\ \text{Re}(S)\}}, \quad (3)$$

where $S\equiv\exp[(1/2)(-\Delta\beta^2\sigma^2+i\phi_0)]$. The output intensity of the generated wave is plotted in Fig. 2 as a function of the Λ_{PM} for $\Lambda=2.6\ \mu$m and several values of the variance σ. Since Λ_{PM} is dependent on the input wavelength, the graph gives a picture of the behavior of a crystal with a given domain structure (Λ and σ) as the input wavelengths are varied. A similar qualitative behavior with large σ was experimentally obtained in our former work on SHG[2]. In that experiment we had $\Lambda\approx 2\ \mu$m. One can see in the figure that when the domain lengths are highly spread the crystal has a broadband response and the conversion efficiency drops. We also find that the peak of the conversion efficiency is slightly shifted to higher Λ_{PM} as σ increases.

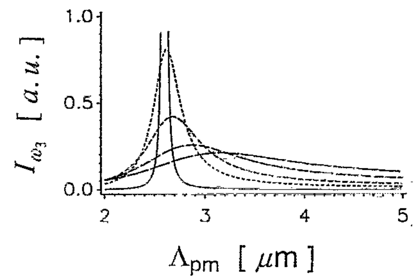

FIG. 2. Calculated output intensities (in arbitrary units) of the generated wave as a function of Λ_{PM} (which depends on the input wavelengths) for $\Lambda=2.6\ \mu$m and several values of the variance $\sigma=\Lambda/40$, $\Lambda/10$, $\Lambda/7$, $\Lambda/5.5$, $\Lambda/4$.

Asymptotic behavior of Eq. (3) for $\sigma^2\Delta\beta^2\gg 1$ and $\Lambda>\Lambda_{PM}$ is given by $I_{\omega_3}=(2C/\pi^2)l\Lambda_{PM}^2/\Lambda$. A similar result was obtained by Kurtz *et al.*,[7] who studied SHG in crystal powders. The proportionality to the length of the crystal is an indication that the light intensities generated by the domains are added up, compared to the case of ordered domains where the fields are coherently added, and the output intensity is proportional to l^2. For the case that $\Delta\beta^2\sigma^2\ll 1$ (but $2N\Delta\beta^2\sigma^2>1$) and $\Lambda\ll\Lambda_{PM}$ Eq. (3) gives $I_{\omega_3}=2C(l/\Lambda)\sigma^2(1+\Lambda^2\Delta\beta^2/16)$. In Ref. 7 the asymptotic expression for this region is $I_{2\omega}\propto l\Lambda/\Lambda_{PM}^2$.

We now turn to the experiment. Our $Sr_{0.61}Ba_{0.39}Nb_2O_6$ crystal had a nominal doping of 0.05 wt %Ce and dimensions of $5\times5\times5$ mm^3. The method for obtaining ordered domain gratings or random domain parameters in the crystal was described in our former works.[2,3] We add here that in the first stage of the fixing process as light is shined on the crystal, the simultaneously applied voltage can have an arbitrary polarity with respect to the crystal c axis. We also note that by fixing a confined volume in the crystal an induced refractive index difference can be obtained with the enhancement of an applied voltage during the frequency mixing experiment. It may be used for waveguiding the beams and increasing the conversion efficiency.

For the parametric mixing, the crystal was illuminated by two beams, one from a Ti-sapphire laser, with a wavelength tuning range of 700 to 900 nm, and a second from a Q-switched Nd:Yag laser which was also used to pump the Ti-sapphire laser. Both lasers had pulse durations of about 10 ns, a repetition rate of 10 Hz, a pulse energy of about 1 mJ and a spot size of about 2 mm. For $\chi_{zzz}^{(2)}$ the beams propagated along the principle axis which was perpendicular to the c axis (to avoid beam "walkoff"). The waves generated by the parametric mixing had extraordinary polarization and a far-field spread in the direction perpendicular to the plane which contained the incident waves and the c axis. The

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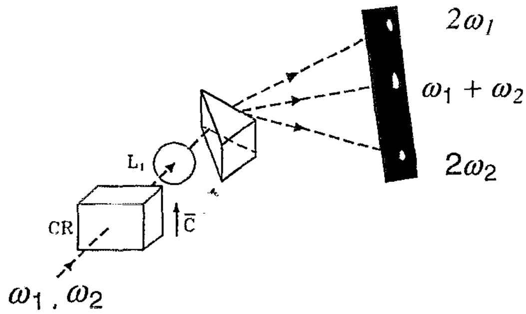

FIG. 3. Schematic of the frequency mixing experiment with a photograph of the output, split by a prism and focused by a cylindrical lens. CR is the crystal with the indication of the c axis and L is a lens.

spreading results from the microdomains in the crystal, as occurs in the broadband SHG experiment.[2]

An analysis of the output spectrum by a monochromator showed three new beams generated in the visible region; the second harmonic of the incident Ti-sapphire beam ($2\omega_1$), second harmonic of the Nd:Yag beam ($2\omega_2$), and a parametric mixing of the two incident beams ($\omega_1+\omega_2$). Figure 3 shows a photograph of these beams, split from the output by a prism and focused by a cylindrical lens. In this specific experiment, the wavelength of the incident Ti-sapphire beam was (840 nm) and the generated wavelengths were as follows: 420, 532, and 469 nm (corresponding to $2\omega_1$, $2\omega_2$, and $\omega_1+\omega_2$, respectively). The conversion efficiencies for all the generated waves was of the same order of magnitude, typically about 0.1% to 1%. The output intensity ratios of the above three generated frequencies were 1:0.89:1.58, respectively. This qualitatively agrees with the theoretical behavior in Fig. 2 for $\sigma=\Lambda/5.5$. No significant dependence of the conversion efficiencies was observed after changing the wavelength of the Ti-sapphire laser between 800 and 870 nm.

We also observed the frequency difference $\omega_1-\omega_2$, which is in the wavelength regime of $\approx 3.5\ \mu m$. The conversion efficiency in this process is expected to be lower than that obtained for the former cases. However, according to the analysis above, the broadband domain grating can give significant conversion efficiency even for $\Lambda \ll \Lambda_{PM}$. Here, the detection and background elimination necessitated more careful arrangements. We used a mercury cadmium teluride detector which was cooled to 77 K. A slab of 5-mm width germanium ($E_g=0.6$ eV) was placed in front of the detector to block the waves which are not in the far IR regime. In addition, we put in front of the detector a bandpass dielectric filter with a transmission wavelength around 3.465 μm. Then, we tuned the input Ti-sapphire wavelength and found a maximum measured intensity for 814.5 nm. This corresponds to a different frequency generation ($\omega_1-\omega_2$) with a wavelength of 3.473 μm. The intensity dropped to half the maximum value for detuning of the Ti-sapphire wavelengths by 1.5 nm (full width at half-maximum=3 nm). This corresponds to detuning of the generated different frequency by 55 nm. The drop to 0.1 of the maximum value occurred for a detuning of the Ti-sapphire wavelengths by 6 nm, corresponding to a detuning of the generated different frequency by 218 nm. These data fit the specification of the filter.

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