第 15 卷 第 6 期
1995 年 6 月

光 学 学 报
ACTA OPTICA SINICA

Vol. 15, No. 6
June, 1995

Femtosecond Pulse —— Second and Third Harmonic Generation with BBO

Yao Jianquan
(*Department of Precision Instrument Engineering. Tianjin University, Tianjin* 300072)
Liu Hang
(*Department of Electrical Engineering, University of Pennsylvania, Philadelphia, PA* 19104, *USA*)
Ashok Puri
(*Department of Physics, University of New Orleans, New Orleans, LA* 70148, *USA*)

Abstract Harmonic generation in BBO by femtosecond laser pulses is studied, taking into account both group velocity mismatch, lowest order group velocity dispersion (GVD) and second order GVD. The lowest and second order GVD of BBO as a function of wavelength is calculated. Second and third harmonic radiation with femtosecond laser pulses is computed by numerically solving the improved coupled wave equations. The effects of the lowest oder GVD and second order GVD on fundamental and harmonic pulses are analyzed. The compensation of group velocity mismatch for third harmonic generationis considered.
Key words group velocity dispersion, femtosecond pulse, harmonic generation.

1 Introduction

The experimental generation of femtosecond pulse from Ti : sapphire lasers has been of much interest recently[1~4]. The wave length range of such pulses is from 0. 67 μm to 1. 1 μm, and may be extended by using nonlinear frequency conversion. A new nonlinear crystal. β-Barium Borate (β-BaB_2O_4, BBO) is very good to be used in second and third harmonic generation to extend the wavelength of Ti : sapphire laser pulses to ultraviolet (UV)/blue because of its large nonlinear optical coefficient and wide range of transparency from near infrared down to ultraviolet [5~9]. The Ti : sapphire laser system with BBO second and third harmonic up-conversion offers a tunable UV/blue femtosecond coherent source and has several other advantages such as its high efficiency and structure simplicity, comparing with the CPM laser system. However, a femtosecond pulse has a wide bandwidth and BBO has a relatively large group velocity dispersion, which tend to broaden and distort the incident fundamental and generated harmonic pulses. One must take into account not only phase mismatch, group velocity mismatch but also the lowest- order even the second- order GVD for femtosecond pulse harmonic generation. In this paper, The lowest and second order GVD of BBO as a function of wavelength is calculated. Then we study harmonic generation of femtosecond pulses in BBO, taking into account both group velocity mismatch, lowest order GVD and second order GVD.

Received 25 February 1994; revised 18 May 1994

Second and third harmonic radiation in BBO with femtosecond pulses are computed by numerically solving the improved coupled wave equations. The effects of the lowest order GVD and second order GVD on fundamental and harmonic pulses are analyzed. The compensation of group velocity mismatch of fundamental and second harmonic waves in sum frequency process for third harmonic generation is considered. The possible influences of higher order nonlinear effects such as self-phase modulation and cross-phase modulation are also discussed.

2 Dispersion feature of BBO crystal

When ultrashort pulses propagate in crystals, its propagation constant $\beta(\omega)$ can be expended in a Taylor series about the central frequency ω_0 [9,10].

$$\beta(\omega) = n(\omega)\frac{\omega}{c} = \beta_0 + \beta_1(\omega-\omega_0) + \frac{1}{2}\beta_2(\omega-\omega_0)^2 + \frac{1}{6}\beta_3(\omega-\omega_0)^3 + \cdots, \tag{1}$$

here, $\beta_0 = n(\omega_0)\omega_0/c = 2\pi n(\lambda_0)/\lambda_0$, $\beta_1 = (d\beta/d\omega)_{\omega_0} = (n-\lambda dn/d\lambda)_{\lambda_0}/c = 1/v_g$, v_g is group velocity, $\beta_2 = (d^2\beta/d\omega^2)_{\omega_0} = (\lambda^3 d^2n/d\lambda^2)_{\lambda_0}/2\pi c^2$ corresponds to the lowest order GVD coefficient, $\beta_3 = (d^3\beta/d\omega^3)_{\omega_0}$ the second order GVD coefficient, and $\lambda_0 = 2\pi c/\omega_0$ is the central wavelength in free space. The first term in (1) gives a phase delay which is responsible for phase matching in harmonic generation. The second term corresponds to the whole group delay of the pulse envelope, which is a decisive factor in the performance of an optical converter when interactions between pulses shorter than 100 fs are considered[10]. The group velocity index can be defined as $m = c/v_g = (n-\lambda dn/d\lambda)_{\lambda_0}$, which is analogous to the refractive index $n = c/v$, here v is the phase velocity. The third and the forth terms describe the pulse spreading and distortion during propagation in a dispersive medium, which generally lead to longer pulses to be generated in a parametric process. The refractive index n, group velocity index m, the lowest order and second order GVD coefficients of BBO can be calculated by using Sellmeier' s relations[8],

$$n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2 \tag{2}$$

$$n_e^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2 \tag{3}$$

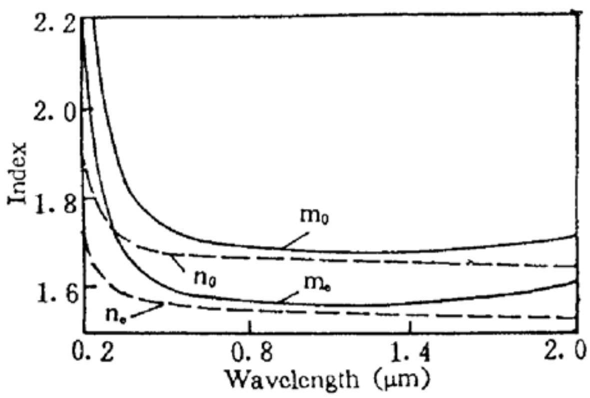

Fig. 1 Refractive n and group m indices in BBO

where wavelength λ is in microns. In Fig. 1, we show the calculated wavelength dependence of refracive and group indices n and m for ordinary and extraordinary waves in BBO. The group velocity indices exhibit stronger dispersion than the refractive indices. The conversion efficiency of a nonlinear process will be optimized if both group and phase matching are simultaneously achieved. i. e. $k(2\omega) - 2k(\omega) = 0$, and $(dk/d\omega)_\omega - (dk/d\omega)_{2\omega} = 0$ or $v_g(\omega) - v_g(2\omega) = 0$. In general practical situations, however, this is impossible[10]. The group velocity mismtch in harmonic generation can lead to a walk-off, which means that the fundamental and harmonic pulses will propagate with different velocities. In turn, it may produce a lengthening of the generated harmonic pulses, limit the usable crystal length and reduce the conversion efficiency[11]. In Fig. 2(a), the lowest order GVD coefficients of BBO for ordinary and extraordinary rays is plotted, and second-order GVD coefficients shown in Fig. 2(b). For an ordinary ray with a wavelength longer than 1.46 μm, and an extraordinary ray with a wavelength longer than 1.29 μm, the lowest order GVD $d^2\beta/d\omega^2$ is negative. The GVD's have important effects on the propagation of ultrashort pulses in the media. They lead to pulse reshaping and distortion. For example, an unchirped pulse will increase its duration and generate chirp when it propagates through an dispersive medium. It should be noted that the chirp may be compensated to Fourier transform limit by propagating the pulse in a medium with GVD sign opposite to the sign of pulse chirp. This is also the principle of ultrashort pulse compression. The lowest order GVD generally dominates, however, for femtosecond pluses, second order GVD contributes to asymmetry in pulse power spectrum and temporal shaping because of wide bandwidth[12].

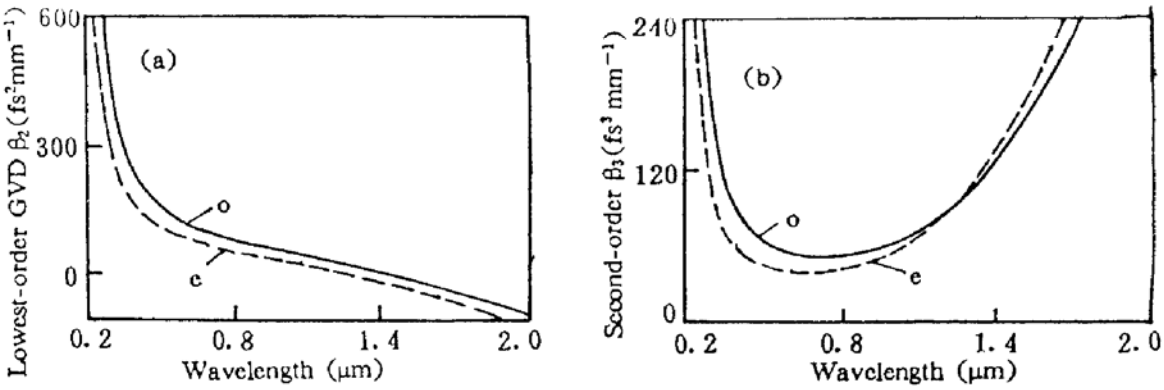

Fig. 2 Lowest order GVD coefficient (a) and second order GVD coefficient (b) in BBO

3 fs Pulse-Coupled Wave Equations of SHG in BBO and the Solution

Theoretically, the coupled wave equations can be used to describe harmonic generation of ultrashort pulse. Supposing quasi-monochromatic wave, the electric field of the pulses in the retarded frame of the incident fundamental wave, $\zeta = z$, $\tau = t - \beta_0^{(1)} z/\omega_0^{(1)}$ is written

$$E^{(N)}(\tau,z) = A^{(N)}(\tau,z)\exp\left[i(\omega_0^{(N)}\tau - (\beta_0^{(N)} - \beta_0^{(1)})z)\right] \tag{4}$$

where the envelope of the electric fields $A^{(N)}$ is assumed to be slowly varying compared to the phase $\exp\left[i(\omega_0^{(N)}\tau - (\beta_0^{(N)} - \beta_0^{(1)})z)\right]$. $\omega_0^{(N)}$ and $\beta_0^{(N)}$ are the central frequency and wavevector, respectively. $N = 1,2,3\cdots$ mean the incident fundamental, second harmonic, third harmonic waves $\cdots$ respectively. We further assume that the absorption of the fundamental and harmonic waves are neglected and the cross section of the nonlinear medium is infinite relative to that of laser beam. Then, the coupled wave equations of second harmonic generation (SHG) including group velocity mismatch, lowest and second order GVD's, are written,

$$\frac{\partial A^{(1)}}{\partial z} - \frac{i}{2}\beta_2^{(1)}\frac{\partial^2 A^{(1)}}{\partial\tau^2} - \frac{1}{6}\beta_3^{(1)}\frac{\partial^3 A^{(1)}}{\partial\tau^3} = -\frac{i8\pi d_{eff}\omega^{(1)}}{cn^{(1)}}A^{(2)}A^{(1)*}\exp(-i\Delta\beta_0 z) \tag{5}$$

$$\frac{\partial A^{(2)}}{\partial z} + \Delta\beta_1^{(2)}\frac{\partial A^{(2)}}{\partial\tau} - \frac{i}{2}\beta_2^{(2)}\frac{\partial^2 A^{(2)}}{\partial\tau^2} - \frac{1}{6}\beta_3^{(2)}\frac{\partial^3 A^{(2)}}{\partial\tau^3} = -\frac{i8\pi d_{eff}\omega^{(1)}}{cn^{(2)}}A^{(1)^2}\exp(i\Delta\beta_0 z) \tag{6}$$

Here, $\Delta\beta_1^{(2)} = \beta_1^{(2)} - \beta_1^{(1)}$ is the group velocity mismatch, β_2 and β_3 are the lowest and second order GVD coefficients, respectively, and $\Delta\beta_0 = 2\beta_0^{(1)} - \beta_0^{(2)}$ is the phase mismatch. The condition of phase match is $\Delta\beta_0 = 2\beta_0^{(1)} - \beta_0^{(2)} = 0$, i.e. $n^{(1)} = n^{(2)}$. d_{eff} is the effective nonlinear optical constant, and $n^{(1)}$ and $n^{(2)}$ are the refractive indices of fundamental and second harmonic waves. The BBO is a negative uniaxial crystal with point symmetry 3 and its effective nonlinear optical constants are given by[8].

$$d_{eff}(\mathrm{I}) = (d_{11}\cos 3\phi - d_{22}\cos 3\phi)\cos\theta + d_{31}\sin\theta \tag{7}$$

$$d_{eff}(\text{II}) = (d_{11}\sin 3\phi + d_{22}\cos 3\phi)\cos^2\theta \tag{8}$$

where θ and ϕ are polar coordinates referring to $z(=c)$ and $x(=a)$, respectively, $d_{11}=1.6$ pm/v, and the values of d_{31} and d_{22} are two orders smaller than that of d_{11} [7]. For type- I SHG ($o(\omega)+o(\omega) \rightarrow e(2\omega)$) , at a central wavelength of 0. 8 μm which can be generated by a Ti : sapphire laser, the phase matching angle is acquired to be 29. 2° by using the phase matching condition. If selecting $\phi=180°$ in crystal cut, d_{eff} is 1. 4 pm/v.

Considering that the incident pulses are unchirped hyperbolic secant pulses generated by a Ti : sapphire the coupled wave equations do not lend themselves to analytical solutions, we use numerical

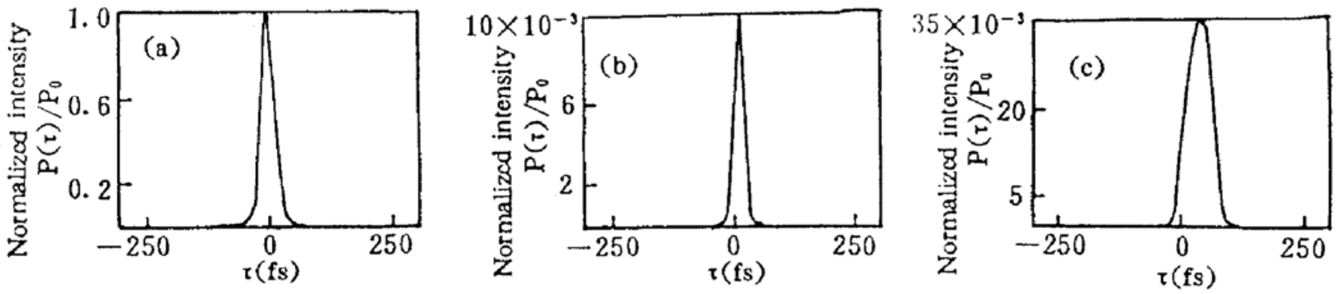

Fig. 3 SHG of 30 fs pulse (peak intensity: 1. 0×10⁸ W/cm²) (a) incident fundamental pulse, (b) and (c) SHG pulse using 0. 1 mm (b) and 0. 4 mm (c) BBO crystals

approach to solve them on computer[12]. Fig. 3 (a) shows the incident fundamental pulse shape for comparison, Fig. 3 (b) and Fig. 3 (c) show the generated harmonic pulse shapes with a BBO crystal of 0. 1 mm and 0. 4 mm, respectively. The incident pulses are 30 fs (FWHM) with a wavelength centered at 0. 8 μm, a pulse repetition rate of 88 MHz, a focused beam cross sectional area of 1 mm², and an average power of 300 mW, which corresponds to pulse peak intensity P_0 of 1. 0 × 10⁷ w/cm². The walk-off and duration of the second harmonic pulse increase with increasing crystal thickness because of group velocity mismatch and GVD′s. We show that the pulse duration as a function of the thickness of BBO in Fig. 4. The group velocity mismatch is dominant, however, GVD′s have an effect to increase the pulse duration and distortion in femtosecond pulse harminic generation because of its wide bandwidth, and generating chirps in the fundamental and second harmonic pulses. As matter of fact, the higher order nonlinear effects such as self-phase modulation and cross-phase modulation may also contribute to pulse chirp and broadening of their spectra, but they can be neglected here because the incident fuundamental pulses are not amplified, their peak intensity is not high enough, and the crystals is very thin in our case.

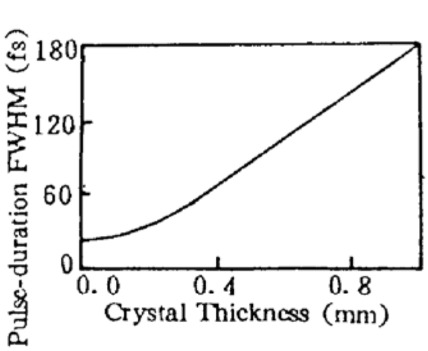

Fig. 4 The relation between pulse duration FWHM (fs) and crystal thickness (mm) (input pulse 30 fs)

4 fs Pulse-Coupled Wave Equations of THG in BBO and the Solution

The third harmonic can be generated with the sum frequency process by passing the generated second harmonic and remaining fundamental waves through another BBO crystal. The coupling wave equations of sum frequency can be written,

$$\frac{\partial A^{(1)}}{\partial z} - \frac{1}{2}\beta_2^{(1)}\frac{\partial^2 A^{(1)}}{\partial\tau^2} - \frac{1}{6}\beta_3^{(1)}\frac{\partial^3 A^{(1)}}{\partial\tau^3} = -\frac{i8\pi d_{eff}\omega^{(1)}}{cn^{(1)}}A^{(3)}A^{(2)*}\exp(-i\Delta\beta_0 z) \tag{9}$$

$$\frac{\partial A^{(2)}}{\partial z} + \Delta\beta_1^{(2)}\frac{\partial A^{(2)}}{\partial\tau} - \frac{i}{2}\beta_2^{(2)}\frac{\partial^2 A^{(2)}}{\partial\tau^2} - \frac{1}{6}\beta_3^{(2)}\frac{\partial^3 A^{(2)}}{\partial\tau^3} = -\frac{i8\pi d_{eff}\omega^{(2)}}{cn^{(2)}}A^{(3)}A^{(1)*}\exp(-i\Delta\beta_0 z) \tag{10}$$

$$\frac{\partial A^{(3)}}{\partial z} + \Delta\beta_1^{(3)}\frac{\partial A^{(3)}}{\partial\tau} - \frac{i}{2}\beta_2^{(3)}\frac{\partial^2 A^{(3)}}{\partial\tau^2} - \frac{1}{6}\beta_3^{(3)}\frac{\partial^3 A^{(3)}}{\partial\tau^3} = -\frac{i8\pi d_{eff}\omega^{(3)}}{cn^{(3)}}A^{(1)}A^{(2)}\exp(+i\Delta\beta_0 z) \tag{11}$$

where $\Delta\beta_1^{(2)} = \beta_1^{(2)} - \beta_1^{(1)}$ and $\Delta\beta_1^{(3)} = \beta^{(3)} - \beta_1^{(1)}$ are velocity mismatches between second harmonic and fundamental waves, and between third harmonic and fundamental waves, respectively. The phase match condition is $\Delta\beta_0 = \beta_0^{(1)} + \beta_0^{(2)} - \beta_0^{(3)} = 0$, *i. e.* $n^{(1)} + 2n^{(2)} = 3n^{(3)}$. Numerical methods are also necessary to solve these equations. The fundamental wave pass through a BBO then the generated second harmonic and remaining fundamental wave propagates through the second BBO. The polarizations of output second harmonic and fundamental waves from the first BBO are perpendicular for type- I SHG, but we can insert a $\lambda/2$ wave plate to make them have the same direction of polarization , then use type- I sum frequency process $[o(\omega) + o(2\omega) \rightarrow e(3\omega)]$ in the second BBO due to its much higher nonlinear optical constant than that of type- II sum frequency. The phase match angle is 44. 2° from phase matching condition and $d_{eff} = 1.15$ pm/v from (7). There is also a walk-off between second harmonic wave and fundamental wave from the first BBO crystal. In experiments, we can easily change the optical distances of these two waves to the second BBO to compensate the walk-off. The second harmonic pulse is behind of the fundamental pulse for 10 fs for 0. 1 mm BBO and for 44 fs for 0. 4 mm BBO, respectively. Fig. 5 shows the shapes of the generated third harmonic pulses by numerically solving the above coupled wave equations. The thicknesses of the first BBO and second BBO are both 0. 1 mm in Fig. 5(a), 0. 4 mm and 0. 1 mm, respectively, in Fig. 5(b) and both 0. 4 mm in Fig. 5(c). The solid curves in FIg. 5 are those which there are no walk-off compensation of second harmonic and fundamental waves before entering the second BBO. If we make the peaks of second harmonic and fundamental pulses overlap at the front of the second BBO by compensation, the third harmonics are shown in dashed curves of Fig. 5. However, their group velocities are also different in the second BBO, and the second harmonic pulse will delay about 33 fs and 133 fs at the output of 0. 1 mm BBO and 0. 4 mm BBO, respectively , comparing with

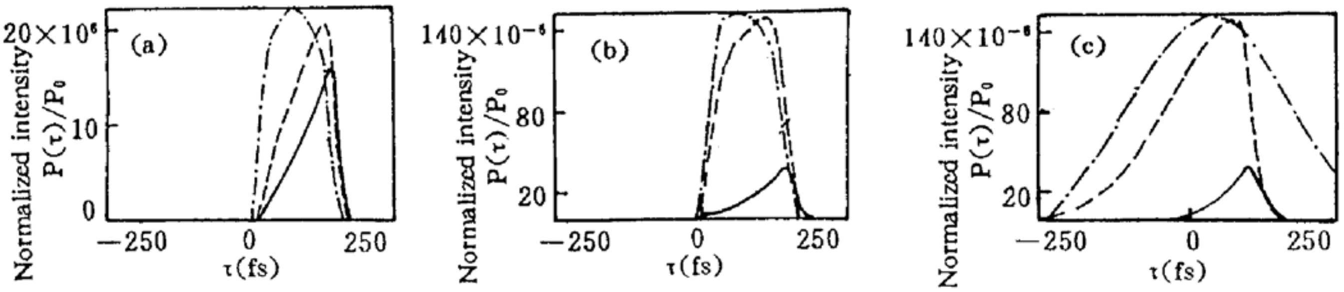

Fig. 5 Third harmonic pulse shapes generated by sum frequency mixing of second harmonic and fundamental pulses using both BBOs thickness 0. 1 mm (a) the first BBO 0. 1 mm and second BBO 0. 4 mm (b) and both BBOs 0. 4 mm for no group velocity compensation (solid curves), just compensation (dashed curves), and over compensation (dash-dot-dashed curves)

fundamental pulse if their peaks are overlapped when they enter the second BBO. We can solve this problem by over compensation before the second BBO. The dash-dot-dashed curve show the third harmonic pulses when the walk-off is compensated and the second harmonic pulses are ahead of fundamental pulses for half of possible walk-off in the second BBO. As shown in Fig. 5. , The broadening and distortion in the third harmonic generation are much greater that those in the second harmonic generation becaure there are group velocity mismatches not only between the second harmonic and fundamental waves but also between the third harmonic and fundamental waves, and between the third and second harmonic wave, and GVD's for the third harmonic pulses are much larger. The limitation on the crystal thickness is more strict in the third harmonic generation. Compared Figs. 5(a)、(b) and (c), the thickness of the first BBO does not affect the duration of the third harmonic pulses very much, but the thickness of the second BBO is dominant 0.4 mm for the BBO and 0.1 mm for the second BBO is a good combination in the third harmonic generation by considering the power, duration, and conversion efficiency. The duration of the output third harmonic pulse is 160 fs, with a peak intensity of 1.5×10^{3} W/cm^{2} in this combination. A shorter pulse could be generated if the thinner BBO crystal were used. Fig. 6 shows the power spectrum of fundamental wavea (a, b) and second harmonic waves (c,d) for $L_0=0.1$ mm (a,c) and 0.5 mm (b,d) BBO crystals with GVD by Fourier trasformation of intensity-time feature.

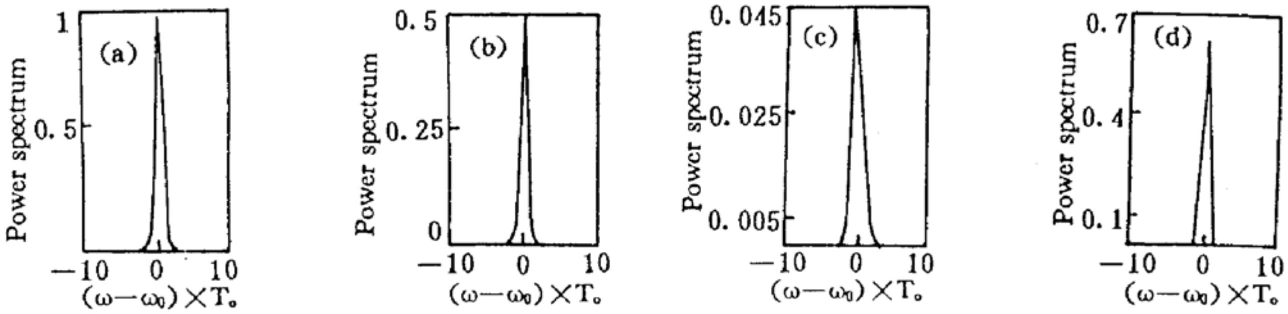

Fig. 6 The power spectrum of fundamental wave (a,b) and second harmonic wave (c,d) for $L_0=0.1$ mm (a,c) and 0.5 mm (b,d) BBO crystals with GVD ($T_0=50$ fs)

In summary, we studued harmonic generation in BBO by femtosecond laser pulses, taking into account both group velocity mismatching, lowest order GVD and second order GVD. The wavelength dependence of the lowest and second order GVD's of BBO is calculated. The pulse shapes of second and third harmonic radiation is computed by numerically solving the improved coupled wave equations. The effects of the lowest order GVD and second order GVD on fundamental and harmonic pulses are analyzed. The thickness of the second BBO dominates the duration of the third harmonic, and the compensation of group velocity mismatch for third harmonic generation is found to be important.

参 考 文 献

[1] Bob Proctor, Frank Wise, Generation of 13 fs pulses from a mode-locked Ti : Al_2O_3 laser with reduced third-order desipersion. *Appl. Phys. Lett.*, 1993, (5) : 470~472

[2] F. Krausz, Ch. Spielmann, T. Brabec *et al.*, Generation of 33 fs optical pulses from a solid-state laser. *Opt. Lett.*, 1992, **17**(3) : 204~206

[3] C. P. Huang, M. T. Asaki, S. Backus *et al.*, 17 fs pulses from a self-mode-locked Ti : sapphire laser. *Opt. Lett.*, 1992, **17**(18) : 1289~1291

[4] D. E. Spence, P. N. Kean, W. Sibbett, 60 fs pulse generation from a self-mode-locked Ti : sapphire laser. *Opt. Lett.*, 1991, **16**(1) : 42~44

[5] C. Chen *et al.*, Optical properties and growth of new UV frequency doubling crystal β-BaB_2O_4, *Sci. Sinica (Ser. B)*, 1984, (7) : 598~604

[6] D. C. Edelstein, E. S. Wachman *et al.*, Femtosecond ultraviolet pulse generation in β-BaB_2O_4. *Appl. Phys. Lett.*, 1988, **52**(26) : 2211~2213

[7] D. Eimerl. L. Davis *et al.*, Optical, mechanical, and thermal properties of barium borote. *J. Appl. Phys.*, 1987, **62**(5): 1968～1983

[8] K. Kato, Second-harmonic generation to 2048 Å in β-BaB_2O_4. *IEEE J. of Quant. Electron.*, 1986, **QE-20**(7): 1013～1014

[9] R. W. Boyd, *Nonlinear Optics*, Academic, U. S. A., 1992

[10] I. V. Tomov, R. Fedosejevs, A. A. Offenberger, Up-coversion of sub-picosecond light pulses. *IEEE J. of Quant. Electron.*, 1982, **QE-18**(12): 2048～2055

[11] A. Cutolo, L. Zeni, Self-induced mismatching effects in harmonic generation with ultra-short laser pulses. *Opt. & Laser Technol.*, 1991, **23**(2): 109～114

[12] H. Liu, A. Puri, effect of higher-order group-velocity dispersion on ultrashort pulse propagation in optical fibers. *Optics Comm.*, 1993, **99**(5/6): 375～379

BBO 晶体中飞秒脉冲的二次和* 三次谐波振荡产生

姚建铨
（天津大学精仪系，天津 300072）
刘 航
（宾夕法尼亚大学电机工程系，宾夕法尼亚. PA19104，美国）
Ashok Puri
（新奥尔良大学物理系，新奥尔良 LA70148，美国）

摘　要　同时考虑了群速色散失配、最低阶群速色散(GVD)和二阶群速色散。研究了在 BBO 晶体中飞秒激光脉冲的谐波产生，计算了 BBO 晶体的作为波长函数的晶体的最低阶和二谐群速色散，借助求解改进的耦合波方程的数值计算，得到了飞秒脉冲的二次及三次谐波辐射，分析了最低阶及二阶 GVD 对于基波脉冲和谐波脉冲的影响，最后考虑了三次谐波产生中群速失配的补偿。

关键词　群速色散，　飞秒脉冲，　谐波产生.

* 收稿日期：1994 年 2 月 25 日；收到修改稿日期：1994 年 5 月 18 日