Simultaneous High-Efficiency Second- and Third-Harmonic Generation in a 1-D Semiconductor Photonic Crystal

Ping Xie, Z. Q. Zhang, and K. S. Wong

Abstract—We show that very high-efficiency second- and third-harmonic conversions can be generated simultaneously in 1-D semiconductor photonic crystals that possess both $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities. This is theoretically demonstrated in a CdS–SiO₂ photonic crystal. The total conversion efficiency can be as large as $\sim 30\%$ for both second- and third-harmonic generations in a small sample with $\sim 14 \mu\text{m}$ thickness and an input power of $\sim 300 \text{ MW/cm}^2$. Such high conversion efficiency is the result of 1) the improved phase matching induced by the Kerr effect and 2) the formation of coupled localized states for the fundamental and second- and third-harmonic waves. From our calculation results, we have also observed the tristability of the output intensity as a function of the input power for each of the fundamental, second-harmonic and third-harmonic waves. The origin of the tristability can be understood from the merging of two bistabilities of the fundamental and higher harmonic waves.

Index Terms—Kerr effect, multiharmonic generation, multilayer photonic crystal, phase matching.

I. INTRODUCTION

RECENTLY, nonlinear photonic crystals (PCs) have attracted much attention partly due to their important applications in the enhancement of various nonlinear optical effects [1]. In $\chi^{(2)}$ -nonlinear PCs, many studies, both theoretical and experimental, have been devoted to the enhancement of second-harmonic (SH) generation [2]–[7]. The idea in these studies was to utilize both the field localization of the band-edge state and the good phase matching provided by the periodicity of the PCs. The periodicity of a PC is known to give rise to geometrical dispersion, which can cancel the material dispersion and produce good phase matching for SH generation. In $\chi^{(3)}$ -nonlinear PCs, it has been shown that, by changing the light intensity at a wavelength near a band edge, the band edge shifts [8], [9] and the bistability phenomenon can be observed as the result of the Kerr effect [10], [11]. Similarly, when the wavelength lies inside the gap near the band edge, a stable, self-organized localized state can be formed [12]–[18]. Recently, significant enhancement of third-harmonic (TH) generation has also been found by utilizing both field localization

and good phase matching near the band edge or inside the gap of a $\chi^{(3)}$ -nonlinear PC [19]–[21]. It was also found that the formation of coupled localized states of the fundamental and TH waves plays an important role in the enhancement of TH generation [21].

Simultaneous SH and TH generation has also attracted much attention recently because it can provide a means to obtain multi-wavelength coherent sources [22]–[25]. Centini *et al.* [26] have shown theoretically and Luo *et al.* [27] have demonstrated experimentally that, via a $\chi^{(2)}$ three-wave mixing process, simultaneous SH and TH generation with high conversion efficiency can be achieved in a 1-D $\chi^{(2)}$ -nonlinear PC.

It is known that most semiconductors possess both nonnegligible $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities. Therefore, in nonlinear PCs made of semiconductors, one must take into account the effects due to both nonlinearities. Recently, simultaneous enhancement of SH and TH generation has been observed in 1-D ZnSe–ZnMgS semiconductor PCs [28]. However, in that study, the input power used was below the threshold required for the occurrence of bistability or multi-stability. Thus, the coupled localized state was not utilized in the generation of the SH and TH waves and the Kerr effect was negligibly small [21]. In our work, in order to achieve much higher conversion efficiency, we theoretically study the enhancement of simultaneous SH and TH generation arising from the coupled localized state of the fundamental, SH and TH waves. Since the formation of a coupled localized state can produce large a Kerr effect on all three waves, which, in turn, gives rise to three equivalent wave vectors. The PC, therefore, has to be designed in a way such that the Kerr-induced wave vectors can be utilized to cancel the low-field phase mismatches due to both material dispersion and geometrical dispersion of the PC so that highly efficient SH and TH generations can be achieved. In this work, we demonstrate that the above idea is feasible. In order to have a low threshold input power, we consider here CdS–SiO₂ PCs rather than ZnSe–ZnMgS PCs. Due to the large dielectric contrast between CdS and SiO₂, this system has a larger photonic band gap than the system studied in [28] has. It, therefore, has more localized wavefunction at the band edge, leading to lower threshold power. We show explicitly that a conversion efficiency of $\sim 30\%$ can be achieved for both SH and TH generations in a small sample with $\sim 14 \mu\text{m}$ thickness and an input power of $\sim 300 \text{ MW/cm}^2$. Furthermore, due to the formation of the coupled localized states, tristability is observed numerically in all three waves. We show that the tristability is a result of the merging of two bistabilities of the fundamental and higher

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P. Xie is with the Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China (e-mail: pxie@aphy.iphy.ac.cn).

Z. Q. Zhang and K. S. Wong are with the Department of Physics, The Hong Kong University of Science and Technology, Kowloon, Hong Kong, China (e-mail: phzzhang@ust.hk; phkswong@ust.hk).

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harmonic waves. It should be mentioned that Konotop and Kuzmiak [29] also studied simultaneous generation of SH and TH waves in a PC with both $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities. Since they considered an infinite system, the effect that arose from the field localization was neglected.

II. COUPLED-WAVE EQUATIONS

Consider a 1-D nonlinear PC with N periods. One layer in each period is linear, with a refractive index n_0 and width l_0 . The other layer is nonlinear, with a weak-field refractive index n , a second-order nonlinear susceptibility $\chi^{(2)}$, and a third-order susceptibility $\chi^{(3)}$. The width of the nonlinear layer is l . A pump wave $\tilde{E}_1$ of frequency ω is normally incident upon the sample along the z axis. Due to the $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities, SH and TH waves can be generated. We represent the fundamental, SH and TH waves by $\tilde{E}_1(z, t) = E_1(z)e^{-i\omega t} + c.c.$, $\tilde{E}_2(z, t) = E_2(z)e^{-2i\omega t} + c.c.$ and $\tilde{E}_3(z, t) = E_3(z)e^{-3i\omega t} + c.c.$, respectively, where $E_\alpha^\pm(z, t) = E_\alpha^+(z) + E_\alpha^-(z)$ and $E_\alpha^\pm(z) = A_\alpha^\pm(z)e^{\pm i\kappa_\alpha z}$ ($\alpha = 1, 2, 3$). Here E_α^+ and E_α^- , respectively, denote the forward and backward propagating components.

By substituting the above fields into Maxwell's equation and using the slowly varying-amplitude approximation for the functions, $A_\alpha^\pm(z)$, we find the following coupled wave equations for the fundamental, SH and TH waves in the nonlinear layer with both $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities

$$\begin{aligned} \frac{dA_1^\pm}{dz} = & \pm i \frac{6\pi\omega}{n(\omega)c} \chi_{\text{Kerr}}^{(3)}(\omega) [|E_1|^2 + 2|E_2|^2 + 2|E_3|^2] A_1^\pm \\ & \pm i \frac{4\pi\omega}{n(\omega)c} \chi^{(2)} (A_1^{\pm*} A_2^\pm e^{\pm i\Delta k_1 z} + A_2^{\pm*} A_3^\pm e^{\pm i\Delta k_2 z}) \\ & \pm i \frac{6\pi\omega}{n(\omega)c} \chi^{(3)} (A_1^{\pm*2} A_3^\pm e^{\pm i\Delta k_3 z} + A_2^{\pm*2} A_3^\pm e^{\mp i\Delta k_4 z}) \end{aligned} \quad (1a)$$

$$\begin{aligned} \frac{dA_2^\pm}{dz} = & \pm i \frac{12\pi\omega}{n(2\omega)c} \chi_{\text{Kerr}}^{(3)}(2\omega) [2|E_1|^2 + |E_2|^2 + 2|E_3|^2] A_2^\pm \\ & \pm i \frac{4\pi\omega}{n(2\omega)c} \chi^{(2)} (A_1^{\pm*} e^{\mp i\Delta k_1 z} + 2A_1^{\pm*} A_3^\pm e^{\pm i\Delta k_2 z}) \\ & \pm i \frac{24\pi\omega}{n(2\omega)c} \chi^{(3)} A_1^\pm A_2^{\pm*} A_3^\pm e^{\pm i\Delta k_4 z} \end{aligned} \quad (1b)$$

$$\begin{aligned} \frac{dA_3^\pm}{dz} = & \pm i \frac{18\pi\omega}{n(3\omega)c} \chi_{\text{Kerr}}^{(3)}(3\omega) [2|E_1|^2 + 2|E_2|^2 + |E_3|^2] A_3^\pm \\ & \pm i \frac{12\pi\omega}{n(3\omega)c} \chi^{(2)} A_1^\pm A_2^{\pm*} e^{\mp i\Delta k_2 z} \\ & \pm i \frac{6\pi\omega}{n(3\omega)c} \chi^{(3)} (A_1^{\pm*3} e^{\mp i\Delta k_3 z} + 3A_1^{\pm*} A_2^{\pm*2} e^{\mp i\Delta k_4 z}) \end{aligned} \quad (1c)$$

where $\Delta k_1 = k_2 - 2k_1$, $\Delta k_2 = k_3 - k_1 - k_2$, $\Delta k_3 = k_3 - 3k_1$, and $\Delta k_4 = k_3 + k_1 - 2k_2$ denote the phase mismatches in the case of a weak field. It should be pointed out that (1) is more complete than [28, eq. (1)], in which a number of higher order terms were ignored due to low conversion efficiencies of both SH and TH generations. However, in this work, all these higher-order terms cannot be ignored due to the presence of large SH and TH conversion efficiencies as will be shown later. It should be pointed out that the Kerr nonlinearities appearing

in (1), i.e., $\chi_{\text{Kerr}}^{(3)}(\omega)$, $\chi_{\text{Kerr}}^{(3)}(2\omega)$ and $\chi_{\text{Kerr}}^{(3)}(3\omega)$ are in general different. However, the $\chi^{(2)}$ and $\chi^{(3)}$ susceptibilities for the frequency conversions appearing in the second and third terms on the right-hand sides of (1a)–(1c) are taken as frequency independent. This assumption leads to a flux conservation in our calculations, i.e., $I_0 = \sum_{i=1}^3 (I_i(0) + I_i(L))$, where $I_0 \equiv |E_1^+(0)|^2$ is the input energy density and $I_1(0) \equiv |E_1^-(0)|^2$, $I_2(0) \equiv |E_2^-(0)|^2$, $I_3(0) \equiv |E_3^-(0)|^2$, $I_1(L) \equiv |E_1^+(L)|^2$, $I_2(L) \equiv |E_2^+(L)|^2$ and $I_3(L) \equiv |E_3^+(L)|^2$ are various outgoing energy densities at two output surfaces. In the case of a homogeneous medium, it is straightforward to show that this assumption also leads to a constant total energy density as required by energy conservation, i.e., $d(n(\omega)|A_1^\pm|^2 + n(2\omega)|A_2^\pm|^2 + n(3\omega)|A_3^\pm|^2)/dz = 0$.

Assuming $A_\alpha^\pm = B_\alpha^\pm e^{i\kappa_\alpha z}$ ($\alpha = 1, 2, 3$), where

$$\kappa_1 = \frac{6\pi\omega}{n(\omega)c} \chi_{\text{Kerr}}^{(3)}(\omega) [|E_1|^2 + 2|E_2|^2 + 2|E_3|^2] \quad (2a)$$

$$\kappa_2 = \frac{12\pi\omega}{n(2\omega)c} \chi_{\text{Kerr}}^{(3)}(2\omega) [2|E_1|^2 + |E_2|^2 + 2|E_3|^2] \quad (2b)$$

$$\kappa_3 = \frac{18\pi\omega}{n(3\omega)c} \chi_{\text{Kerr}}^{(3)}(3\omega) [2|E_1|^2 + 2|E_2|^2 + |E_3|^2] \quad (2c)$$

Equation (1) can be rewritten as

$$\begin{aligned} \frac{dB_1^\pm}{dz} = & \pm i \frac{4\pi\omega}{n(\omega)c} \chi^{(2)} (B_1^{\pm*} B_2^\pm e^{\pm i\Delta k'_1 z} + B_2^{\pm*} B_3^\pm e^{\pm i\Delta k'_2 z}) \\ & \pm i \frac{6\pi\omega}{n(\omega)c} \chi^{(3)} (B_1^{\pm*2} B_3^\pm e^{\pm i\Delta k'_3 z} + B_2^{\pm*2} B_3^\pm e^{\mp i\Delta k'_4 z}) \end{aligned} \quad (3a)$$

$$\begin{aligned} \frac{dB_2^\pm}{dz} = & \pm i \frac{4\pi\omega}{n(2\omega)c} \chi^{(2)} (B_1^{\pm*2} e^{\mp i\Delta k'_1 z} + 2B_1^{\pm*} B_3^\pm e^{\pm i\Delta k'_2 z}) \\ & \pm i \frac{24\pi\omega}{n(2\omega)c} \chi^{(3)} B_1^\pm B_2^{\pm*} B_3^\pm e^{\pm i\Delta k'_4 z} \end{aligned} \quad (3b)$$

$$\begin{aligned} \frac{dB_3^\pm}{dz} = & \pm i \frac{12\pi\omega}{n(3\omega)c} \chi^{(2)} B_1^\pm B_2^\pm e^{\mp i\Delta k'_2 z} \\ & \pm i \frac{6\pi\omega}{n(3\omega)c} \chi^{(3)} (B_1^{\pm*3} e^{\mp i\Delta k'_3 z} + 3B_1^{\pm*} B_2^{\pm*2} e^{\mp i\Delta k'_4 z}) \end{aligned} \quad (3c)$$

where

$$\Delta k'_1 = \Delta k_1 + \Delta \kappa_1 = (k_2 - 2k_1) + (\kappa_2 - 2\kappa_1) \quad (4a)$$

$$\Delta k'_2 = \Delta k_2 + \Delta \kappa_2 = (k_3 - k_1 - k_2) + (\kappa_3 - \kappa_1 - \kappa_2) \quad (4b)$$

$$\Delta k'_3 = \Delta k_3 + \Delta \kappa_3 = (k_3 - 3k_1) + (\kappa_3 - 3\kappa_1) \quad (4c)$$

$$\Delta k'_4 = \Delta k_4 + \Delta \kappa_4 = (k_3 + k_1 - 2k_2) + (\kappa_3 + \kappa_1 - 2\kappa_2). \quad (4d)$$

Note that κ_i ($i = 1, 2, 3$) in (2) can be interpreted as equivalent wave vectors arising from the Kerr effect, while $\Delta k'_\beta$ ($\beta = 1, 2, 3, 4$) in (4) now denotes the phase mismatch in the case of a strong field, which is the sum of the phase mismatches in the case of a weak field and the Kerr-effect-induced phase mismatch, i.e., $\Delta \kappa_\beta$. As we will see in the next section, the formation of a coupled localized state can produce nonnegligible $\Delta \kappa_\beta$, which compensates Δk_β and results in good phase matching for $\Delta k'_\beta$.

III. RESULTS AND DISCUSSION

The fields $E_1^\pm(z)$, $E_2^\pm(z)$, and $E_3^\pm(z)$ inside the sample, which satisfy the coupled wave equations in the nonlinear layers described by (1)–(4) and the Fresnel law at each interface of two layers, can be solved numerically by using the iterative method described elsewhere [17], [18], [21]. In order to ensure a high accuracy, in our calculations, we require that the relative values of the quantities, $|(E_i^{(n)}(z)|^2 - |E_i^{(n-1)}(z)|^2)/|E_i^{(n)}(z)|^2$ ($i = 1, 2, 3$), are smaller than 10^{-6} for all values of z inside the sample, where $|E_i^{(n-1)}(z)|$ and $|E_i^{(n)}(z)|$ represent the values in the $(n - 1)$ th step and the n th step, respectively, in the iterative procedure. In the case of TH generation with $\chi^{(2)} = 0$, we are able to obtain the same results as shown in [21]. In order to achieve efficient SH and TH generation, in general, the wavelength of the fundamental wave, λ_1 , should be close to the band edge and, at the same time, the nonlinear layers should be thick. The former requires $n_0 l_0 \cong nl \cong \lambda_1/4$, while the latter requires that l/l_0 be as large as possible. Furthermore, in this study, the high conversion efficiencies are obtained through the formation of coupled localized states. We, therefore, further require that the wavelengths of SH and TH waves are also close to the band edges of other gaps as will be discussed later. In our calculations, we consider a CdS–SiO₂ PC with $N = 35$ periods, i.e., 35 layers of CdS and 35 layers of SiO₂. The widths for the SiO₂ and CdS layers are taken to be $l_0 = 210$ nm and $l = 200$ nm, respectively. Note that small variations in l_0 and l will have little effect on the general results presented in this work. SiO₂ layers can be considered to be linear because their $\chi^{(2)}$ and $\chi^{(3)}$ are negligibly small compared with those of CdS. CdS layers are nonlinear, with nonnegligible values of $\chi^{(2)}$ and $\chi^{(3)}$. The refractive index for SiO₂ is taken to be $n_0 = 1.5028 + 5.0227 \times 10^{-3}/\lambda^2 - 8.2382 \times 10^{-5}/\lambda^4$ [26] and the weak-field refractive index for CdS is taken to be $n = \sqrt{8.35 + (0.465/(\lambda^2 - 0.256^2)) + (4.01/(\lambda^2/150^2 - 1))}$ [30], where $\lambda = 2\pi c/\omega$ is the wavelength in vacuum, in units of μm . The value of $\chi_{\text{Kerr}}^{(3)}$ for CdS is taken to be $\chi_{\text{Kerr}}^{(3)} = 1.2 \times 10^{-15} \text{ cm}^2/\text{V}^2$ at $\lambda \approx 1672$ nm, $\chi_{\text{Kerr}}^{(3)} = 2.5 \times 10^{-15} \text{ cm}^2/\text{V}^2$ at $\lambda \approx 836$ nm and $\chi_{\text{Kerr}}^{(3)} = -3.5 \times 10^{-15} \text{ cm}^2/\text{V}^2$ at $\lambda \approx 557$ nm [31], where the wavelengths $\lambda \approx 1672$ nm, 836 nm and 557 nm are close to the fundamental wavelength, SH wavelength and TH wavelength in this work, respectively. The value of $\chi^{(2)}$ for CdS is taken to be $\chi^{(2)} = 3.5 \times 10^{-9} \text{ cm/V}$ [32]. However, there is no reliable experimental data on $\chi^{(3)}$ in TH conversion. In principle, it should be close to the value of $\chi_{\text{Kerr}}^{(3)}$ in the Kerr effect. In this work, we study the SH and TH conversions for two different values of $\chi^{(3)}$.

A. Properties of the Linear Sample

The transmission spectrum of the linear sample is shown in Fig. 1(a). The corresponding effective refractive index, n_{eff} , is shown as a solid curve in Fig. 1(b), where, for comparison, we also plot the refractive indexes of CdS (dotted line) and SiO₂ (dashed line). In Fig. 1(a), we see that the wavelength at the long-wavelength band edge of the first gap is $\lambda_{\text{edge}}^{(1)} =$

Fig. 1. Transmission spectrum (a) and effective refractive-index dispersion (b) of a linear CdS–SiO₂ PC with $l_0 = 210$ nm and $l = 200$ nm. The dotted and dashed curves in (b) are the refractive index dispersion of CdS and SiO₂, respectively.

1665.73 nm, that of the second gap is $\lambda_{\text{edge}}^{(2)} = 799.575$ nm and that of the third gap is $\lambda_{\text{edge}}^{(3)} = 561.05$ nm. In Fig. 1(b), we see that when the fundamental wavelength, λ_1 , is chosen near $\lambda_{\text{edge}}^{(1)}$, the phase matching for both SH and TH conversions is much improved compared with that in the bulk nonlinear CdS medium due to compensation from the geometrical dispersion arising from the periodicity of the PC. However, due to the existence of the large first band gap, the phase mismatch for the SH conversion, i.e., $(\Delta k_1)_{\text{eff}} = 4\pi/\lambda[n_{\text{eff}}(\lambda/2) - n_{\text{eff}}(\lambda)]$, becomes negative. In the TH conversion, since the material dispersion is too steep at the short wavelength, the geometrical dispersion cannot completely balance the material dispersion and, therefore, produces a positive phase mismatch, i.e., $(\Delta k_3)_{\text{eff}} = 6\pi/\lambda[n_{\text{eff}}(\lambda/3) - n_{\text{eff}}(\lambda)] > 0$.

B. Simultaneous SH and TH Generation

It is known that the Kerr effect can induce a shift in the band edge or the formation of the gap localized modes [8]–[18], which are represented by the first terms on the right-hand sides of (1a)–(1c). Thus, in order to make use of the field localization of the band-edge state to enhance the SH and TH conversions, we should take the fundamental wavelength, λ_1 , to be slightly larger than $\lambda_{\text{edge}}^{(1)}$ because $\chi_{\text{Kerr}}^{(3)} > 0$ at $\lambda_1 > \lambda_{\text{edge}}^{(1)} = 1665.73$ nm. In Fig. 2, we show some typical results of the normalized output intensities of the fundamental beam (in two upper panels), the SH beam (in two middle panels) and the TH beam (in two lower panels) for two different values of TH conversion susceptibility, $\chi^{(3)}$, at a fixed

Fig. 2. (a)–(c) and (d)–(f) are, respectively, the results obtained when $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)}(\lambda_1) = 1.2 \times 10^{-15} \text{ cm}^2/\text{V}^2$ and $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)}(\lambda_1/3) = -3.5 \times 10^{-15} \text{ cm}^2/\text{V}^2$ are chosen as the TH conversion susceptibility, $\chi^{(3)}$, at a fundamental wavelength of $\lambda_1 = 1672.2 \text{ nm}$ in the PC with $l_0 = 210 \text{ nm}$, $l = 200 \text{ nm}$, $\chi^{(2)} = 3.5 \times 10^{-9} \text{ cm/V}$. (a) and (d): Transmission $I_1(L)/I_0$ (solid lines) and reflection $I_1(0)/I_0$ (dotted lines) of the fundamental wave. (b) and (e): SH conversion efficiencies $I_2(L)/I_0$ (solid lines) and $I_2(0)/I_0$ (dotted lines). (c) and (f): TH conversion efficiencies $I_3(L)/I_0$ (solid lines) and $I_3(0)/I_0$ (dotted lines). The inset of (c) shows the logarithmic plot of $I_3(L)/I_0$ shown in (c).

fundamental wavelength, $\lambda_1 = 1672.2 \text{ nm}$. Fig. 2(a)–(c) is obtained when the TH conversion susceptibility $\chi^{(3)}$ is chosen to be $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)}(\lambda_1) = 1.2 \times 10^{-15} \text{ cm}^2/\text{V}^2$, which has a positive value. Fig. 2(d)–(f) are obtained when the TH conversion susceptibility, $\chi^{(3)}$, is chosen to be $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)}(\lambda_1/3) = -3.5 \times 10^{-15} \text{ cm}^2/\text{V}^2$, which has a negative value. It should be mentioned that the magnitudes of $\chi^{(3)}$ chosen here are on the same order of magnitude as those found in other large bandgap semiconductors [33]. In Fig. 2, we have defined $I_j(L) = |E_j^+(L)|^2$, $I_j(0) = |E_j^-(0)|^2$ ($j = 1, 2, 3$) and $I_0 = |E_1^+(0)|^2$. Fig. 2 clearly shows that both SH and TH beams can be generated efficiently and simultaneously, independent of the sign of $\chi^{(3)}$. For example, Fig. 2(b) and (c) shows that the total SH conversion efficiency, i.e., $I_2(L)/I_0 + I_2(0)/I_0$, can reach about 9.1%, while the total TH conversion efficiency, i.e., $I_3(L)/I_0 + I_3(0)/I_0$, can reach about 20% at an input power of $I_0 \approx 300 \text{ MW/cm}^2$. Thus, about 30% of the input power is converted to SH and TH generation. Here we should mention that the surface damage threshold of CdS is about 20 GW/cm^2 for a laser with a 35 ps pulsewidth at wavelength of $1.06 \mu\text{m}$ [34]. Thus, either the excitation power or the total intensity inside the CdS medium (as seen in Figs. 2–4) considered in our calculations is well below the damage threshold of the material we considered.

Fig. 3. Results for TH conversion with $\chi^{(2)} = 0$ and $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)}(\lambda_1) = 1.2 \times 10^{-15} \text{ cm}^2/\text{V}^2$ at fundamental wavelength $\lambda_1 = 1672 \text{ nm}$ in a PC with $l_0 = 210 \text{ nm}$ and $l = 200 \text{ nm}$. (a) shows the transmission $I_1(L)/I_0$ (solid line) and reflection $I_1(0)/I_0$ (dotted line) of the fundamental wave and (b) shows the TH conversion efficiencies $I_3(L)/I_0$ (solid line) and $I_3(0)/I_0$ (dotted line). The inset is the log plot of $I_3(0)/I_0$.

Fig. 4. Intensity distributions normalized by I_0 for a fundamental beam (dotted lines) and a TH beam (solid lines).

C. Physical Origin of High SH and TH Conversion Efficiencies

The high conversion efficiencies shown in Fig. 2 for both SH and TH generations are mainly due to the presence of a nonnegligible Kerr effect in (2), which greatly improves the phase matching for both SH and TH conversions. Since $\chi_{\text{Kerr}}^{(3)}(\lambda_1/2) > \chi_{\text{Kerr}}^{(3)}(\lambda_1) > 0$ and $\chi_{\text{Kerr}}^{(3)}(\lambda_1/3) < 0$ for $\lambda_1 \approx 1672 \text{ nm}$, from (2a)–(2c) we see that the Kerr effect induces a positive $\Delta\kappa_1 = \kappa_2 - 2\kappa_1$ and a negative $\Delta\kappa_3 = \kappa_3 - 3\kappa_1$, therefore, canceling the phase mismatches, $(\Delta\kappa_1)_{\text{eff}} < 0$ and $(\Delta\kappa_3)_{\text{eff}} > 0$, appearing in the case of weak fields. It is worth mentioning that the high conversion efficiency of TH generation shown in Fig. 2 is not due to a cascading effect of the $\chi^{(2)}$ nonlinearity. To show this, we have done another calculation with $\chi^{(2)} = 0$ and $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)} = 1.2 \times 10^{-15} \text{ cm}^2/\text{V}^2$. The results are shown in Fig. 3. It is clearly seen that the conversion efficiency of TH

generation is very similar to that shown in Figs. 2(c) and 2(f) where $\chi^{(2)} \neq 0$. The maximum TH conversion efficiency is about 25% at $I_0 \approx 326 \text{ MW/cm}^2$, which is close to the total energy conversion efficiency ($\sim 30\%$ at $I_0 \approx 300 \text{ MW/cm}^2$) in the case of nonzero $\chi^{(2)}$ [Fig. 2(a)–(c)].

It was mentioned earlier that the presence of a nonnegligible Kerr effect is a result of the formation of coupled localized states. To show such a coupled localized state, for simplicity, we plot in Fig. 4 the intensity distributions of λ_1 (dotted lines) and $\lambda_1/3$ (solid lines) normalized by input intensity, I_0 , at point A (or A') in Fig. 3. Both distributions are strongly localized and are nearly symmetric to the center of the sample. Since their magnitudes are comparable to each other, this suggests that there exists a significant energy exchange between λ_1 and $\lambda_1/3$ in the coupled localized state. This energy exchange also greatly enhances the TH conversion. In the presence of $\chi^{(2)}$ nonlinearity shown in Fig. 2 the coupled localized state consists of three components. In addition to the fundamental and TH waves shown in Fig. 4, there also exists a localized wave for the SH beam inside the sample.

D. Physical Origin of the Tri-Stability

Finally, we discuss the interesting tristability found in Figs. 2 and 3. The tristability occurs for all three waves. In fact, if tristability occurs in any one of the SH and TH waves, it will also occur in the fundamental wave. This can be seen in Fig. 3 where SH wave does not exist. The existence of tristability originates from the merging of two bistabilities: one arises from the formation of a localized state at the fundamental wavelength and the other arises from the formation of another localized state at higher harmonics. The merging of the two bistabilities produces a tristability, which is accompanied by the formation of a coupled localized state shown in Fig. 4. To demonstrate this, for simplicity, we have done calculations at three different values of fundamental wavelengths, i.e., $\lambda_1 = 1670.7$, 1671 , and 1671.5 nm for the case of $\chi^{(2)} = 0$ and $\chi^{(3)} = \chi_{\text{Kerr}}^{(3)}(\lambda_1)$. The results of $I_3(L)/I_0$ are shown in Fig. 5(a)–(c). Fig. 5(a) shows that, at $\lambda_1 = 1670.7$ nm, $I_3(L)/I_0$ exhibits two bistabilities. As λ_1 increases away from $\lambda_{\text{edge}}^{(1)} = 1665.73$ nm, both bistable regions become wider and the position of the right bistability shifts to the left as shown in Fig. 5(b). As λ_1 further increases, the two bistable regions merge, leading to the occurrence of tristability as shown in Fig. 5(c). The left bistability in Fig. 5(a) and (b) is caused by the shifted band edge of the first gap of the fundamental wave and, thus, as λ_1 moves away from $\lambda_{\text{edge}}^{(1)}$, the threshold of the input intensity required to shift the band edge and excite a localized state increases, leading to a wider bistable region [17], [18]. The bistability on the right is caused by the shifted band edge of the third gap or the formation of a localized state at the TH wavelength inside the third gap. This is because $\lambda_1/3 = 557.4$ lies to the left of $\lambda_{\text{edge}}^{(3)} = 561.05$ nm, which is the long-wavelength band edge of the third gap shown in Fig. 1. The reason that the position of the right bistability shifts to the left when λ_1 is increased is explained as follows: an increase in λ_1 corresponds to an increase in $\lambda_1/3$. Thus, $\lambda_1/3$ moves closer to the band edge at $\lambda_{\text{edge}}^{(3)}$. As a result, the threshold for the excitation of the localized state

Fig. 5. Results for the TH conversion efficiency, $I_3(L)/I_0$, versus I_0 with $\chi^{(2)} = 0$ and $\chi^{(3)} = 1.2 \times 10^{-15} \text{ cm}^2/\text{V}^2$ at various λ_1 in the PC with $l_0 = 210$ nm and $l = 200$ nm. (a) $\lambda_1 = 1670.7$ nm, (b) $\lambda_1 = 1671$ nm, (c) $\lambda_1 = 1671.5$ nm.

becomes smaller [17], [18]. If λ_1 is further increased, the tristability shown in Fig. 5(c) disappears and only bistability can be observed. This is more apparent when $\lambda_1/3$ becomes larger than $\lambda_{\text{edge}}^{(3)}$. In this case, the bistability can only occur in the fundamental wave. This is also true when $\lambda_1/3$ is located far away from the band edge and in the pass band. In this case, only the first bistable region can occur. In other words, no tristability will occur in this case. To test this, we have done another calculation using different sample parameters so that $\lambda_1/3$ lies well inside the pass band. The result of our calculation shows only the bistability due to the fundamental wave, which confirms the above explanation.

IV. SUMMARY

In this work, we have theoretically studied SH and TH conversions due to coupled localized states in a 1-D semiconductor PC made of CdS–SiO₂, which possess both $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities. By using realistic optical parameters, we have calculated both SH and TH conversion efficiencies. Our numerical results show that both the SH and TH beams can be generated simultaneously and very efficiently in a small sample of $\sim 14 \mu\text{m}$ thickness. The high conversion efficiencies are the

results of both improved phase matching induced by the Kerr effect and strong field localization of the fundamental, SH and TH waves in the coupled localized states. The accompanying phenomenon of tristability phenomenon is also discussed.

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Ping Xie received the Ph.D degree in optics from the Institute of Physics, Chinese Academy of Sciences (CAS), Beijing, China, in 1994.

After receiving his Ph.D. degree, he was with CAS as an Assistant Professor. From 2000 to 2001, he worked in the Faculty of Engineering, Hokkaido University, Hokkaido, Japan, as a JSPS Postdoctoral Fellow. From 2002 to 2003, he worked in the Department of Physics, Hong Kong University of Science and Technology, Hong Kong, as a Visiting Scholar. Since 2003, he has been working in CAS as an Associate Professor. From 1991 to 2001, he mainly did research on photorefractive nonlinear optics. He is currently working on nonlinear optics in photonic crystals and on molecular motors such as kinesin, myosin, dynein, etc.

Z. Q. Zhang received the B.S. degree from Tunghai University, Taiwan, R.O.C., in 1968 and the Ph.D degree from the University of Pennsylvania, Philadelphia, in 1974, both in physics.

He joined the Physics Department of the Hong Kong University of Science and Technology (HKUST), Hong Kong, in 1994, where he is currently a Professor. His research interests include wave propagation in complex media, wave localization, and nonlinear optical effects in photonic crystals, etc.

K. S. Wong received the B.Sc. (hons.) degree in physics from King's College, University of London, London, U.K., in 1983 and the D.Phil. degree in solid-state physics from Clarendon Laboratory, University of Oxford, Oxford, U.K., in 1987.

He joined the Physics Department of the Hong Kong University of Science and Technology (HKUST), Hong Kong, in 1991, where he is currently an Associate Professor. His main research interests include ultrafast lasers and nonlinear optics, time-resolved spectroscopy, semiconductor and polymer physics. He has published over 80 peer-reviewed journal publications and holds three U.S. patents.

Prof. Wong is a member of the Optical Society of America and Vice-Chairman of the Physical Society of Hong Kong.