

# Quasi-phase-matched backward second-harmonic generation by complementary media in nonlinear metamaterials

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High efficiency of the second-harmonic and sum-frequency generation can be obtained in optical superlattice by using the conventional quasi-phase-matched (QPM) method. Although this trick can be played on the acoustic wave, the media with negative nonlinear parameters are not common in acoustics. Furthermore, the QPM method used in acoustic metamaterials has been less studied. In this work, a protocol is provided to realize the QPM method by using nonlinear complementary media in acoustic metamaterials in order to obtain large backward second-harmonic generation. Compared with the conventional method, the method gains a broader bandwidth and can be used in both acoustic and electromagnetic waves. © 2012 Acoustical Society of America.

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## I. INTRODUCTION

In the last decade, metamaterials have attracted the interest of thousands of researchers, and this because of their special characteristics, such as negative refraction, invisible cloaking, resolutions breaking the limits of diffraction, etc.<sup>1–7</sup> All of the preceding properties are based on linear assumption. After the linear characteristics of the metamaterials became better understood, researchers expanded the research area to the nonlinear situation and began to find unique properties that had not been explored previously, e.g. backward second-harmonic emission, backward second-harmonic localization, etc.<sup>8–10</sup> To realize these properties, high backward second-harmonic conversion is expected. To ensure that energy storage is positive, the dispersion and lossy should be the characteristics of the metamaterials;<sup>11</sup> this means it is difficult to get a highly efficient second-harmonic conversion. One way proposed to achieve the efficient backward second-harmonic generation is to use the so-called phase-matched condition (PMC) to find a special frequency point in which the momentum conservation condition is satisfied.<sup>12</sup> But only some special selected frequencies can match the PMC; this greatly reduces the application of the unique characteristics of the nonlinear metamaterials. The quasi-phase-matched (QPM) method was applied to electromagnetic metamaterials to obtain large forward second-harmonic waves.<sup>13</sup> This raises some questions: Is there any other way to realize large backward second-harmonic generation in acoustic metamaterials in a very wide range of frequency? Can the QPM method be applied in the acoustic metamaterials? To the best of our knowledge, these questions are still unanswered despite their important applications as well as the significant efficiency of the high order harmonic conversion in acoustics.

## II. THEORY

Backward second-harmonic generation has been discovered in nonlinear metamaterials,<sup>9,10</sup> and there will be many future applications. In acoustics, backward second-harmonic generation can be used to realize the second-harmonic imaging in medical ultrasound. Developing a good way to obtain large backward second-harmonic generation will promote the application of the nonlinear metamaterials. Media with negative nonlinear are not common in acoustics; therefore the conventional QPM method used in optics is hardly used in acoustics. In this article, we propose a new way to realize the high efficiency of backward second-harmonic generation by the QPM method with nonlinear complementary media. The concept of the complementary media was proposed after metamaterials had been discovered. In the linear case, one of the most important applications is the perfect lens as proposed by Pendry.<sup>1</sup> Complementary media (also called anti-object) can “cancel” the corresponding space; the result is just like an empty space embedded in the cancelled space and can achieve the invisible cloak. The key idea of the complementary media is coordinate transformation. A wave accumulates its wave path when it crosses a normal medium, and the wave will experience a negative wave path when it crosses a negative refraction medium. By connecting these two kinds of media together in the wave path, the phase of the wave can be restored and the corresponding region seems nonexistent.<sup>4</sup> Consider the source positioned on the left  $x = -L$  and an acoustic wave path consisting of two media the mass densities of which are  $\rho_0$  and  $-\rho_0$ , bulk moduli are  $\kappa_0$  and  $-\kappa_0$ , and lengths are  $L$  and  $d$ , respectively, as shown in Fig. 1(a). When a wave illuminates from left to right, the wave path is first increased by  $nL$ , where  $n$  is the refractive index, when passing the normal medium, then decreased by  $nd$ , when passing the negative index medium. The final result is just like the wave passing a normal media through length  $L-d$ . If we set  $L = d$ , then the phases at  $x = -L$  and  $x = d$  are equal. It looks as if the wave travels through empty space if the attenuation of the metamaterials

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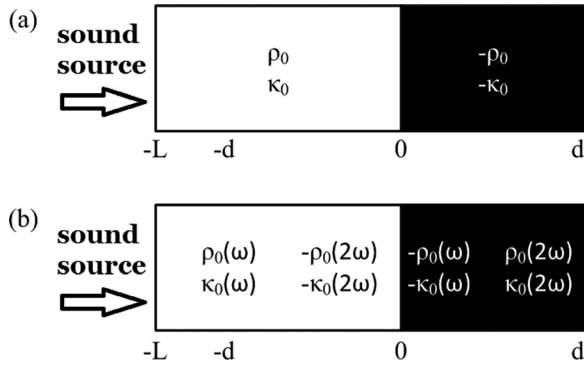


FIG. 1. (a) A pair of linear complementary media. (b) A pair of nonlinear complementary media.

is neglected. Now we replace the previous media with quadratic nonlinear media the properties of which at the fundamental frequency are the same as before. But at the second-harmonic frequency, their mass densities become  $-\rho_0$  and  $\rho_0$ , and their bulk moduli become  $-\kappa_0$  and  $\kappa_0$ , respectively, as shown in Fig. 1(b). The left media behave with right-handed properties at angular frequency  $\omega$  but left-handed properties at angular frequency  $2\omega$ , and the right media behave left-handed at  $\omega$ , but right-handed at  $2\omega$ . So we can still call this complementary media and in this pair of media, backward second-harmonic emission can be expected.<sup>14,15</sup> Now we can consider whether the second harmonic will be cancelled or enhanced through the nonlinear complementary media. Assume that  $L = d$ . First, we consider the second harmonic generated by the left media. At  $x = -L$ , the second harmonic generated by the infinitesimal element at point  $x$  equals  $A \cos[2\omega(t + t') - 2k_1x]dx$ . Here,  $A$  is proportional to the nonlinearity parameter and the square of the amplitude of the sound pressure,  $t' = (x + L)/c_2$ ,  $k_1 = \omega/c_1$ , and  $k_2 = 2\omega/c_2$  represent the wave number of the fundamental frequency and the second harmonic, respectively. We choose the sign of  $t'$  to be positive because its energy flux is in the negative direction though the wave vector of the second harmonic is positive in these kinds of metamaterials.<sup>9</sup> So the energy accumulates in the negative direction. For simplicity, we assume that the quasi-linear approximation, such that  $A$  is taken to be a constant and the total length of the structure to be relatively short. In our assumption, the phase matching condition is established, so we can make superposition of the infinitesimal second harmonic and obtain the pressure at the second-harmonic frequency at  $x = -L$ :  $P_{2\omega} = A \cos(2\omega t + 2k_1L)L$ . Now we consider the second harmonic generated by the right media. The phase of the fundamental wave at point  $x$  equals the phase at point  $-x$  because of the complementary media effect. So the phase of the second harmonic generated at point  $x$  is also the same as the phase at point  $-x$ . In such media, the second harmonic is generated backward, and the energy flux of the second harmonic accumulates in the negative direction. When the second harmonic generated at point  $x$  propagates to the point  $-x$ , the phase does not change because of the complementary media effect at  $2\omega$ . Finally all the second harmonics are added at points  $x = -L$ , and the

sound pressure at the second-harmonic frequency should be  $2P_{2\omega}$  instead of zero. So the pair of the complementary media is not regarded as empty if the nonlinear effect is considered because the second harmonic is generated. On the contrary, we can take advantage of it to realize the QPM method in fluids to enhance the second-harmonic generation.

In the PMC media, the phase of the second harmonic generated at any point is the same when superposed at  $x = -L$  as shown in Fig. 2(a), where the direction of the small arrow represents the phase of the second harmonic generated at  $x$  propagating to the position  $x = -L$ . All the arrows are in the same direction, meaning that the second harmonic is enhanced as shown in Fig. 2(b). Now we extend the analytical procedure to the phase-mismatched situation. As shown in Fig. 2(c), the parameters of the left media are  $\rho_1$  and  $\kappa_1$  at  $\omega$  and  $-\rho_2$  and  $-\kappa_2$  at  $2\omega$ , and the right media behave  $-\rho_1$  and  $-\kappa_1$  at  $\omega$  and  $\rho_2$  and  $\kappa_2$  at  $2\omega$ , respectively. Because of dispersion, the phase of the second harmonic generated at different locations is different when propagating to the position  $x = -L$ . Defining coherence length  $l_0 = \pi/|k_2 - 2k_1|$ , if we change the media to its complementary media each time the wave passes through  $l_0$ , then the QPM condition can be satisfied. Setting  $L = l_0$ , we can obtain the phase of the second harmonic generated at each position represented by the direction of the small arrows [Fig. 2(c)]. Making superposition of the infinitesimal second harmonic, we have  $P_{2\omega} = 4A \cos(2\omega t + 2k_1l_0)l_0/\pi$  at  $x = -L$  if there is only one period for the structure. Figure 2(d) is obtained from the vector superposition of the small arrows of Fig. 2(c). The length of the big arrow represents the amplitude of the second harmonic and its direction represents the phase of the second harmonic. If we cascade  $n$  period of this structure, the total length of which is  $2nl_0$ , then the total

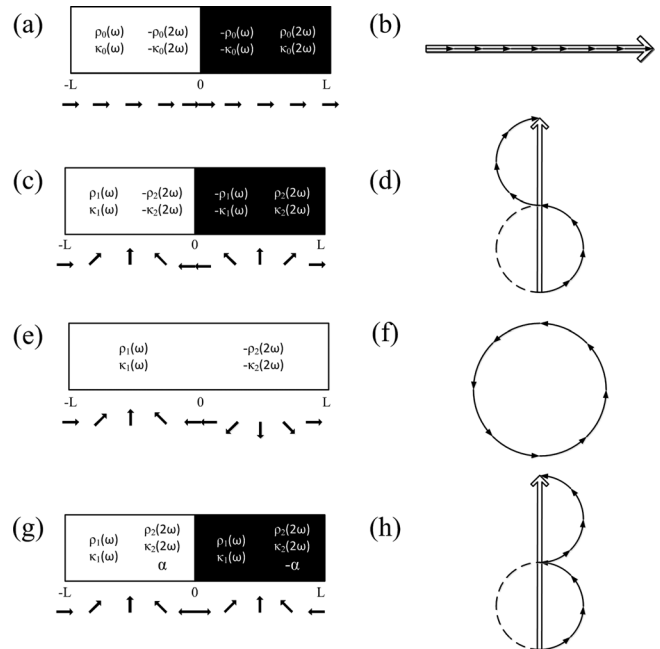


FIG. 2. (a) A pair of nonlinear complementary media without dispersion and (b) its second harmonic. (c) A pair of nonlinear complementary media with dispersion and (d) its second harmonic. (e) A normal media with dispersion and (f) its second harmonic. (g) A pair of normal nonlinear polarization media consists together and (h) its second harmonic.

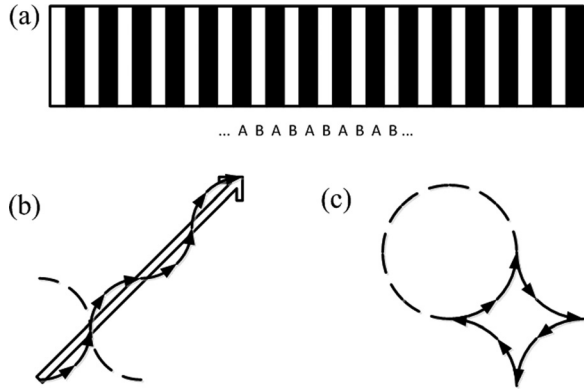


FIG. 3. (a) A pair of nonlinear complementary media or antiparallel  $180^\circ$  domains is arranged in the periodic structure, according to ABABABAB... and (b) its second harmonic. (c) The second harmonic in the structure with a pair of antiparallel  $180^\circ$  domains instead of the nonlinear complementary media.

second harmonic is  $P_{2\omega} = 4A \cos(2\omega t + 2k_1 l_0) n l_0 / \pi$  at the initial point ( $x = -L$ ). As shown in Fig. 2(e), if we use normal media instead of complementary media, after a wave passes two times the coherence length without using the QPM method, then all the second harmonic can be added in a circle; as shown in Fig. 2(f) the total second harmonic turned out to be zero. If we allow the length of the media shown in Fig. 2(e) to be  $2n l_0$ , the second harmonic is still zero. Our scheme is to arrange a pair of nonlinear complementary media periodically, in the order ABABABAB..., as shown in Fig. 3(a), to obtain large second-harmonic generation. We can also calculate it through the conventional QPM method as shown in Fig. 2(g). As shown in Fig. 2(g), the parameters of the left and the right media are  $\rho_1$  and  $\kappa_1$  at  $\omega$  and  $\rho_2$  and  $\kappa_2$  at  $2\omega$ , in common, but the nonlinearity parameters of the left and the right media are  $\alpha$  and  $-\alpha$ , respectively. In this case, a forward second-harmonic wave appears, and the second harmonic at  $x = L$  is  $P_{2\omega} = 4A \cos(2\omega t + 2k_1 l_0) l_0 / \pi$  as shown in Fig. 2(h), the amplitude of which is the same as that of our QPM method. The difference between our trick and the conventional QPM method<sup>16,17</sup> is that our trick is played by introducing nonlinear complementary media to restore the phase difference caused by dispersion, which doesn't require the existence of the antiparallel  $180^\circ$  domains or negative nonlinearity parameter.

$$\eta = f\left(\frac{D}{2l_0}\right) = f(l) = \frac{\sqrt{\left(\int_0^L \text{sign}\left(\sin\frac{2\pi x}{l}\right) \cos 2\pi x dx\right)^2 + \left(\int_0^L \text{sign}\left(\sin\frac{2\pi x}{l}\right) \sin 2\pi x dx\right)^2}}{L} \quad (1)$$

where sign is the sign function reflecting the phase of the second harmonic with respect to the periodic length of the structure.  $L$  is the total length of the structure, in our calculation, we choose  $L = 50$ . The first and the second integrations represent the real part and imaginary part contributions of the second-harmonic wave, respectively.

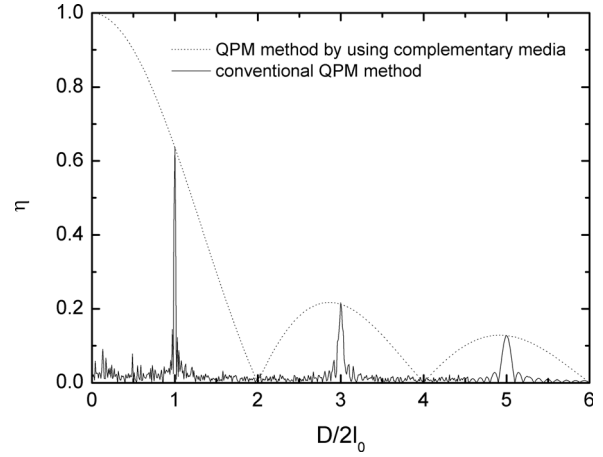


FIG. 4. The relationship between the efficiency coefficient and the period  $D$  of the structure with respect to  $2l_0$ .

To compare the difference between our scheme and the conventional QPM method, we set the period of the structure described in Fig. 3(a) to be  $l_0$  and the length of each single element to be  $l_0/2$ . First, we apply our QPM method by introducing a couple of nonlinear complementary media. After a wave passes  $2l_0$ , the second harmonic turns out to be  $P_{2\omega} = 4\sqrt{2}A \cos(2\omega t + 2k_1 l_0) l_0 / \pi$  at the initial point, as shown in Fig. 3(b). Turning our view to the conventional QPM method, if we introduce a pair of antiparallel  $180^\circ$  domains in our structure marked A and B, instead of the complementary media, as shown in Fig. 3(a), then after a wave passed  $2l_0$ , the second harmonic turns out to be zero as shown in Fig. 3(c). We define the normalization coefficient as  $\eta = |P_{2\omega}| / AL$ , indicating the efficiency of the second-harmonic emission, where  $P_{2\omega}$  is the amplitude of the second harmonic at source point,  $L$  is the length the wave has passed. If we set the period to be  $2l_0$ , as described in the last paragraph, the efficiency coefficient of our method and conventional QPM method turns to be  $2/\pi$  equally. If we set the period to be  $l_0$ , as described in this paragraph, then the efficiency coefficient turns out to be  $2\sqrt{2}/\pi$  and zero, respectively. Figure 4 shows the relationship between the period  $D$  of the structure and the efficiency coefficient.

For the conventional QPM method:

For the QPM method by using complementary media:

$$\eta = f\left(\frac{D}{2l_0}\right) = f(l) = \left| \frac{\sin \frac{\pi l}{2}}{\frac{\pi l}{2}} \right|. \quad (2)$$

From Fig. 4, we find that our method can be realized in a very wide range of the period of the structure; this implies our method can realize large second-harmonic emission at a very broad bandwidth. The physical nature of the conventional QPM method is coordinate translation, while our QPM method is coordinate reversion [as shown in Figs. 2(c), 2(d), 2(g), and 2(h)]. When using our QPM method, the phase of the second harmonic generated at point  $x$  is the same as that at point  $-x$  when superposed as the directions of the small arrows shown in Fig. 2(c). But when using the conventional QPM method, the phase of the second harmonic generated at point  $x$  is the same as that at point  $x-L$  when superposed, as the directions of the small arrows as shown in Fig. 2(g), and most of the second harmonic generated at  $x$  and  $-x$  are cancelled out by each other when the period of the structure is smaller than  $2l_0$ . This is why our QPM method can enlarge the bandwidth of the second-harmonic generation. In addition, we can see that if in our method we reduce the period of the structure, then we can obtain larger second-harmonic generation. But it is worth mentioning that the period of the structure to realize the QPM method cannot be very small because the lattice period to realize the double negative materials must be one order smaller than the wavelength in the media, so the period of the structure to realize the QPM method must be one order larger than the lattice period of the double negative materials; this confirms the assumption of the homogeneous medium.

### III. RESULTS AND DISCUSSIONS

To demonstrate the performance of our QPM method, we set  $\rho_1 = 998 \text{ kg/m}^3$  and  $\kappa_1 = 2.19 \text{ GPa}$ ,  $\rho_2 = 1594 \text{ kg/m}^3$ , and  $\kappa_2 = 1.40 \text{ GPa}$ ,  $f = 0.1 \text{ MHz}$  as described in Fig. 2(c). Then we have  $c_1 = 1481 \text{ m/s}$ ,  $c_2 = 938 \text{ m/s}$ ,  $k_1 = 424 \text{ m}^{-1}$ ,  $k_2 = 1340 \text{ m}^{-1}$ , and  $l_0 = 6.39 \text{ mm}$ . We set  $A = 1 \text{ N/m}^3$ , the period of the structure to be  $2l_0 = 12.78 \text{ mm}$ , and the total length of the media to be  $20l_0 = 127.8 \text{ mm}$ . Figure 5(a)

shows the fundamental field of the wave in the periodic structure media consisting of nonlinear complementary media. Figure 5(b) shows the second-harmonic field by the QPM method with nonlinear complementary media. Figure 5(c) shows the second-harmonic field without any QPM method. Comparing these figures shows that using the QPM method with nonlinear complementary media, large second harmonic can be obtained, which is one or two orders larger than that generated without any QPM method.

Our method can also be used in forward second-harmonic generation as long as one of the media is characterized with positive refraction at both the fundamental and second-harmonic frequencies, and its complementary media is characterized with negative refraction at fundamental and second harmonic, simultaneously. Furthermore, if the perfect phase match cannot be realized with complementary media at both the fundamental and second harmonic, the QPM method can also be effective if the phase difference at the fundamental frequency can be restored with the complementary media, and the right-handed medium should then be nonlinear and the left-handed medium should be linear.

Apparently, all the metamaterials in the real world are lossy, but the influence on the high order harmonic is very small.<sup>18</sup> So we neglected the attenuation of the metamaterials in our model. Our scheme is sufficiently simple for experimental realization in practice, and it is crucial to seek an efficient method to realize the nonlinear metamaterials. A periodic structure has been proposed to realize the double negative and double positive properties in different frequency ranges.<sup>19</sup> Also if the structure is filled by media with strong nonlinearity, such as the porous medium and ultrasound contrast agents,<sup>20</sup> large second harmonic can be obtained.

### IV. SUMMARY

In conclusion, we have proposed a brand new QPM method to obtain large second-harmonic generation in a very

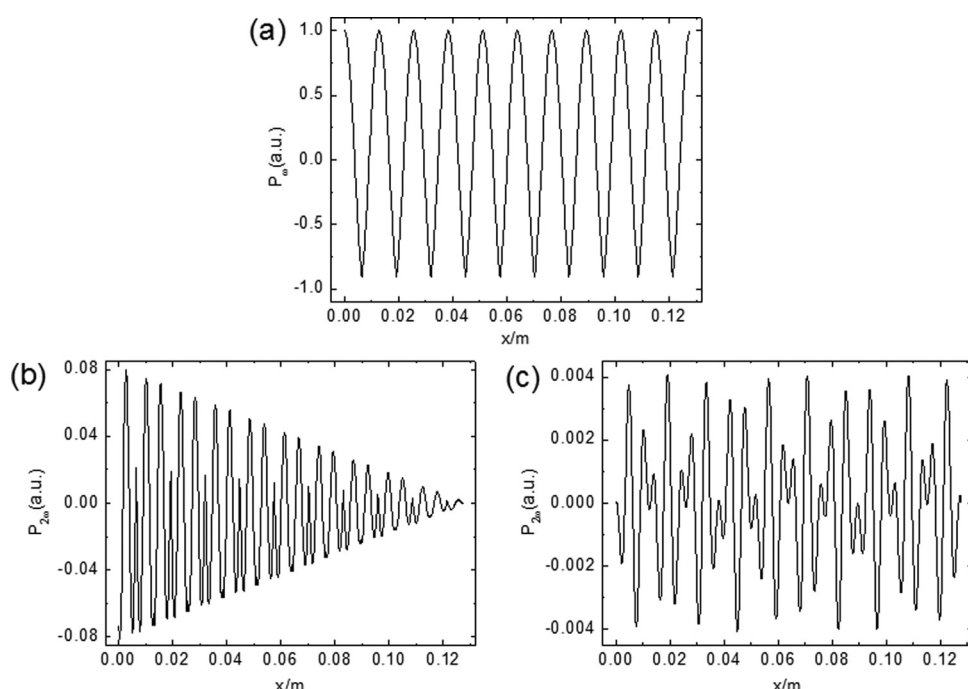


FIG. 5. (a) The field of the fundamental wave in the periodic structure consisting of nonlinear complementary media. (b) The distribution of the backward second-harmonic field generated by the QPM method with the nonlinear complementary media. (c) The distribution of the backward second-harmonic field generated without any QPM method.

wide bandwidth. Our method can be widely used in industrial and medical fields where the large high harmonic is expected. Moreover, we have discussed that the complementary media become invalid if the nonlinear effect is considered. That is to say, if the “perfect” cloak effect can be realized one day, nonlinear technology may be a good choice to detect the object.

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