Adaptive pumping for spectral control of broadband second-harmonic generation

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Second-harmonic generation (SHG) is always a significant frequency conversion process in nonlinear optics for many great applications but can be limited when broadband spectral laser sources are involved, e.g., femtosecond pulses. The conversion efficiency can be high, but the spectral control is hard because of the phase-matching (PM) limitation. Recently, a random quasi-phase-matching (QPM) scheme was proposed to make use of highly nonlinear materials that are difficult to be phase matched under traditional configurations. The spectral control is even harder in anisotropic random materials, and the coherence is completely lost. Here, we proposed an approach to solve this problem by coherent light control via feedback-based wavefront shaping. We utilized this method for spectral control of broadband SHG, which can be efficient even in strongly scattering media. Randomly selected wavelengths in the broadband spectra were enhanced with a good selectivity, and the direction was also controlled in a three-dimensional (3D) configuration. This technique paves the way for convenient spatial and spectral control of both linear and nonlinear emissions and a local enhancement of their conversion efficiency, indicating great progress in both random and ultrafast nonlinear optics.

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As is well known, broadband ultrafast lasers with high-peak and low-average powers are very popular in nonlinear optics, such as second-harmonic generation (SHG) or sum-frequency generation, because of the high conversion efficiency and low thermal damage [1,2]. However, traditional nonlinear processes pumped by broadband lasers usually provide narrowband or disordered nonlinear spectra because of the phase-matching (PM) limitation and defect [Fig. 1(a)]. Recently, a random quasi-phase-matching (QPM) scheme was proposed in the nonlinear material, where the PM or QPM limitation is loosened and the conversion efficiency increases linearly with the interaction length \( L \) [3–5]. This technique was aimed at making use of the nonlinear materials with a large coefficient but difficult to be phase matched under traditional configurations [Fig. 1(b)]. However, the spectra of the nonlinear emission were hard to control in this randomized domain structure. Moreover, many random nonlinear materials are anisotropic or porous, leading to detrimental scattering during the frequency conversion [6–8]. In this case, the directionality is completely lost, as shown in Fig. 1(c). However, a flexible control of the nonlinear emission in spectrum—direction as well as a locally enhanced conversion efficiency—is always desired.

Fig. 1. Concept of the SH emission spectral control in various systems. (a) The narrowband SH spectrum emitted from a nonlinear crystal (e.g., LN crystal) with PM limitation. (b) The broadband SH spectrum emitted from a random nonlinear crystal (e.g., strontium barium niobate crystal). (c) Without shaping, the SH signals were scattered and disordered in the spectrum at all spatial angles. (d) With an appropriate SLM phase mask applied, the SH signals coherently superpose at the selected wavelengths and locations.
In this Letter, we proposed an approach for spectral control of the SH scattered from 3D superline lithium niobate (LN) nanocrystal powder pumped by a broadband femtosecond laser via a feedback-based wavefront shaping method. This is a powerful method for manipulating disordered light from scattering media, which can be used for focusing and imaging through turbid media for many potential biological applications [9–12]. Great progress, such as complex imaging, perfect focusing, noninvasive imaging, and sub-wavelength manipulation, has been achieved in the last decade [13–20]. However, the turbid media used in these studies are usually linear materials. Nonlinear processes can also play an important role in deep focusing or imaging [21]. Our previous work has proved the possibility of manipulating the nonlinear signals generated directly from a scattering nonlinear medium for further applications [22]. Besides, it has been demonstrated recently that this powerful method works not only on a space domain, but is also active in time or frequency domains [14]. This fact is of great significance for spectral control in ultrafast broadband lasers and random lasers [23–25]. Although it is not limited to, here we introduced this method to random nonlinear optics. First of all, we theoretically proved the feasibility of SH spectral control by optimizing the pump profile. During the experimental demonstration, spectra at randomly chosen wavelengths in fundamental wave (FW) and SH broadband emission were both successfully optimized to sharp peaks with a linewidth of around 0.6 nm in full width at half-maximum (FWHM). To prove the flexibility of our manipulation, we experimentally realized a multi-peak optimization and a multi-space-angle optimization under similar configurations. Our method is efficient, especially in the case of strong scattering, which reveals that the defect or the scattering is not necessarily an obstacle but may help in the optimization.

The concept of the SH spectral control via feedback-based wavefront shaping is shown in Fig. 1. First, we presented a simple description of this random nonlinear process theoretically. It is hard to describe the whole process directly via a simple theory, such as the Huygens–Fresnel principle or coherent superposition for nonlinear optical interactions, because strong scattering and a nonlinear process mix up in the medium [26]. Here, we separate the two processes for simplicity without changing the main characteristics of the medium. The pump first enters a nonlinear crystal at the PM angle to generate efficient nonlinear signals. Then we put a linear scattering medium next to the rear surface of the crystal to disorganize the emitted signals. Thus, the multispectral transmission matrix (MSTM) for linear spectrum manipulation is valid for describing both FW and SH scattered from the medium [25]. The total output field at the detected location at the wavelength of $\lambda_k$ reads

$$E_{\text{out}}(\lambda_k) = \sum_{i=1}^{\text{SLM}} h_{ik} e^{i\phi_{ik}} E_{\text{in}}(\lambda_k),$$

where $E_{\text{in}}(\lambda_k)$ represents the input field and $h_{ik} e^{i\phi_{ik}}$ represents the coefficients of the MSTM with $\phi_{ik}$ being the spectral phase component. The summation represents the integration of all spatial light modulator (SLM) pixels. Finally, the spectrum is given by the combination of all wavelengths ($\lambda_k$).

Without shaping, the spectrum (SH or FW) is disordered at all space angles because different wavelengths encounter different scattering and incoherently superpose at free space [Fig. 1(c)]. If an SLM is applied to shape the pump profile appropriately, $\phi_{ik}$ could be optimized accordingly, leading to a coherent superposition of the signals at $\lambda_k$. Generally speaking, those signals with wavelengths far from $\lambda_k$ would not be enhanced simultaneously because of the dispersion. Thus, optimized spectra with sharp peaks at the selected wavelengths can be achieved based on the optimization [Fig. 1(d)].

The experimental setup for demonstration is illustrated in Fig. 2(a). The light source was a Ti:sapphire broadband femtosecond laser (80 fs duration, 80 MHz) with a frequency span of 780–820 nm. All spectra were measured using a high-resolution optical spectrometer (SR500, Andor, resolution 0.1 nm). A half-wave plate and a Glan–Taylor polarizer were used after the source for polarization and power control because of the response characteristics of the SLM. The pump power was 200 mW and the beam was spatially expanded to fit the SLM aperture of around 10 mm $\times$ 10 mm. A lens was then applied to focus the shaped pump into the LN scattering layer with a dimension of 30 mm $\times$ 30 mm $\times$ 0.1 mm [Fig. 2(b)]. A complex SHG process then occurred at all of the pumped volumes. The scattered SH signals were detected in the forward direction by the spectrometer through a low-pass filter. The distance between the rear sample surface and the probe was 200 mm. The SLM and spectrometer were both connected to a computer with a genetic algorithm for optimization.

Here, LN nanocrystal powder was used as the scattering medium for its large second-order susceptibility and high refractive index ($n \approx 2.2$). It was prepared by the solid state reaction method using niobium pentoxide and lithium acetate dihydrate as reactants [27]. Then the powder was deposited onto an indium-tin oxide (ITO) coated glass substrate by the electrophoresis method [28]. The maximum size of the particle was around 400 nm (average size: 200 nm) measured by the scanning electron microscope [Fig. 2(c)], smaller than the coherent interaction length $L_c$ in the LN crystal (about several microns). This fact ensures that every single particle emits maximum SH signals [3]. The scattering mean free path of the LN powder layer was estimated to be $l \approx 1.1 \lambda$ for 532 nm light from a coherent backscattering experiment [22]. The genetic algorithm was chosen for its robustness to the noisy environments [29]. The SH intensity at a selected wavelength was recorded after each iteration of the SLM phase mask. This intensity served as the cost function for further feedback. In our experiments, 512 $\times$ 512 active pixels on the SLM were divided into 128 $\times$ 128 segments for a faster optimization speed.
Some typical results of FW and SH spectral control are shown in Fig. 3. Before optimization, the FW with a broadband spectrum, as shown in the inset of Fig. 3(a), turned into a disordered spectrum after passing through the scattering sample [red curve in Fig. 3(a)]. The cost function for feedback was set as the normalized spectral intensity (the intensity before optimization was set as one) at the wavelength of 799 nm. After optimization, a broadband FW spectrum with a sharp peak at 799 nm was achieved [blue curve in Fig. 3(a)]. The linewidth of the peak was 0.8 nm in FWHM. The enhancement factor for this peak, which is the ratio of the intensities before and after optimization, was estimated to be 10.3 [Fig. 3(b)]. When considering the nonlinear frequency conversion, we shifted attention to the SH waveband and set the normalized intensity at 396 nm as the cost function. A broadband SH spectrum with a sharp peak at 396 nm was achieved after 200 iterations. The enhancement factor and the linewidth were 8.4 and 0.6 nm, respectively.

For comparison, the FW spectrum was also recorded and shown in the Fig. 3(c) inset. It remained a disordered spectrum with no obvious enhancement at the wavelength of 792 nm. This fact proved that the SH enhancement at 396 nm was not caused by the local enhancement of the pump, but a real coherent control. The interesting thing is that there was an enhancement of the total spectrum in the SH operation. This is mainly caused by a spatial focusing process similar to our previous work on spatial SH focusing [22]. This phenomenon provides a possibility of spatiotemporal manipulation for further potential applications. All the manipulations are repeatable except for some small variation on the enhancement factors.

Other than the generation of one peak in the broadband spectrum, our system can also do a multi-peak optimization for complex applications [14, 22]. For example, Figs. 4(a) and 4(b) present a two-peak optimization at the wavelengths of 397.6 nm and 400.2 nm. The cost function was originally set as the average intensity at these wavelengths. After optimization, two sharp peaks emerged with an enhancement factor and a linewidth of both peaks around 6.8 and 0.5 nm, respectively. Interestingly, there was also a total spectrum enhancement similar to the previous result [Fig. 3(c)]. For demonstration, we set a complex cost function in a three-peak operation to weaken this enhancement. As shown in Fig. 4(c), a three-peak optimization at 397.2 nm, 398.7 nm, and 400.2 nm was realized but with a lower total spectrum enhancement. The peak enhancement factor and the linewidth was 3.2 and 0.4 nm, respectively. All generation numbers were set to be 200 for comparison. It is obvious that the more complex manipulation, e.g., complex targets and complex cost functions, the lower the enhancement. Even so, the resolution of these results is still maintained at a good level. Actually, the individual peak intensity in multi-peak optimizations might not be exactly controlled. For better results, the SLM segments should be divided more precisely and the time cost could be increased depending on different applications and algorithms [30].

Furthermore, a multi-space-angle optimization was also realized, indicating a possible control of the spectral emission at different space angles. The experimental setup is illustrated in Fig. 5(a) where we substituted a two-in-one fiber probe for the original one. The integrated spectrum of two individual spectra detected from two different space angles was recorded by one spectrometer, as shown in Fig. 5(b) before and after optimization. The selected wavelength was 401.3 nm and the enhancement factor was 8.1 after 300 iterations [Fig. 5(b) inset]. The two individual spectra were also recorded and shown in Figs. 5(c) and 5(d). They were both automatically optimized to a profile similar to the integrated spectrum via a saturated operation, which means the enhancement factor increased to a saturated value based on the genetic algorithm. We experimentally achieved a similar spectrum emission at different space angles only by one detector. Actually, it will be more operable and visualized to respectively manipulate the spectra at different space angles by multiple detectors. For example, the enhanced
wavelengths of these spectra could be individually set and the cost function could be set relative to the peak’s intensity. As a result, both the emission spectra and directions can be expected to be manipulated with high flexibility.

In conclusion, adaptive pumping for spectral control of broadband FW and SH wave generated and scattered from nonlinear turbid media pumped by a broadband femtosecond laser via feedback-based wavefront shaping was demonstrated. Randomly selected wavelengths in the broadband spectra were successfully optimized to sharp peaks with a high FWHM resolution at desired spatial angles. We also experimentally realized a multi-peak optimization and a multi-space-angle optimization, indicating the flexible control of the emission spectra and directions. To make the manipulation more precise, the cost function could be appropriately improved at the cost of time. Some special experimental setups for great improvement of the optimization and imaging speed can also be considered for possibilities in vivo applications [31,32]. This work paves a way in spatial and spectral control of both broadband linear and nonlinear emissions with high selectivity and flexibility. Actually, the enhancement of the SHG was a localized effect at the chosen wavelength in the particular direction by constructive interference. But it is still significant for counteracting the scattering effect of random nonlinear materials, indicating significant progress in both random and ultrafast nonlinear optics.

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