Broadband parametric amplification in ultrafast fluorescence application

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Broadband parametric amplification is applied for the femtosecond ultrafast fluorescence research. The time resolution depends on duration of pump pulse, which works both as pump source and gate. For spectrum application, a special phase-matching angle exists for broadband amplification correspondence to the pump wavelength. The broadband amplification range from 500 to 750 nm was found for β-barium borate (BBO) crystal, pumped at 400 nm. Large dye fluorescence amplification of 10^7 is realized. The generation of cone emission is selected as reference for system tuning. It is supposed to be a new method for ultrafast study in fluorescence comparable to fluorescence up-conversion and optical Kerr gate techniques.

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Two main ultrafast time-resolved fluorescence spectroscopy techniques are fluorescence up-conversion[1] and optical Kerr gate[2] technique[3]. The former one based on sum frequency of fluorescence and ultrafast gate laser pulse. This up-conversion method has its own limitations, including the strong requirement system alignment, and the spectrum bandwidth limitation that comes from the strict phase match condition. The nonlinear crystals are quite sensitive to its alignment towards the input signal and gate pulse. For the detection of whole spectrum range, each wavelength needs to be scanned separately, though recently, broadband up-conversion technique[3] was explored. The optical Kerr gate technique allows a large bandwidth in comparison with the up-conversion technique. This technique uses solvents with large third-order optical nonlinearity, such as CS_2, as working media, to provide transparency in detection range. The shortcomings of this technique is the gate width. The solvents provide Kerr response with sub-picosecond or picosecond time scale, which removes the femtosecond resolution of the system. For the above two techniques, the efficiency is always a problem. They convert part of signals into the detector. These two techniques are all proved insufficient while investigating ultra-weak fluorescence phenomena, which are even more widely existing in nature. To increase the system sensitivities in order to have these signals detectable, one way is to spend more cost on detecting system and more time on integration. Another way is to amplify signal optically. The optical parametric amplification (OPA) is the way to amplify signals with broad tunable range and high efficiency.

We will report our recent research on ultrafast fluorescence technique, which applies femtosecond parametric amplification. These OPA processes are widely used as tunable light source from visible to infrared (IR) range[4], limited by crystal transparency and pump wavelength. β-barium borate (BBO) is one of the high efficiency crystal. While pumped with second harmonic (SH) of Ti:sapphire laser source at 400 nm, the tunable range is from visible to IR around 3 μm. It is very useful while this single crystal can produce such wide wavelength of several octaves for application. This high efficient amplification was also used for extra weak signal detection with cross-correlation frequency resolved optical gating (XFROG) technique[5,6].

While pumped with higher energy above threshold, superfluorescence will appear around axis of pump beam. It satisfies phase matching condition of three-wave mixing in angular dispersion. This superfluorescence is also called as cone emission, since it presents ring pattern on screen, surrounding the pump beam. Some interesting phenomena such as higher amplification rate and SH generation due to modulation instability[7] will occur.

In our previous report[8], we introduced our research on characterizing femtosecond laser pumped broadband amplification in visible range under minimized dispersive cone emission condition. The femtosecond laser system (Spitfire, from Spectrum-Physics) was used as light source, with pulse duration of 120 fs at 800 nm, running at 1 kHz. The laser pulse passed through a 1-mm-long BBO crystal to generate SH at 400 nm. Then the SH was kicked away from fundamental and sent through a delay line. It was used as pump pulse, which focused into a second 1-mm-long BBO crystal with cutting angle of 24° for type I phase matching. This second crystal was carefully placed before the pump focus, where the pump power was suitable for cone emission generation, but lower than the damage threshold. The left fundamental passed through another (the third) BBO crystal to generate pump pulse at 400 nm for the excitation of dye solution. The dye solution flowed through a quartz cell of 1 mm in thickness. We chose sample dye of DCM in ethanol, with concentration of 0.3 mmol/L. The fluorescence was then collected and focused down as seed for parametric amplification later.

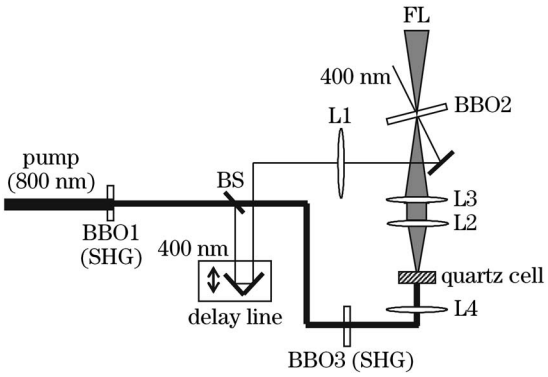

Fig. 1. Experimental setup. BS: beam splitter; L: lens; FL: fluorescence; SHG: second harmonic generation.

By varying the delay line, the seed was amplified in time delay. The experimental scheme is shown in Fig. 1.

When pumped with pulse energy higher than threshold at 400-nm wavelength, the colored cone emission happens. The best phase-matching angle is around 31.5°, which is the angle between pump and optical axis of crystal. At this angle, wide spectrum range, from 500 to 750 nm, is overlay at uniform spatial direction, which is 3.85° towards pump beam. Outside crystal, with surface diffraction, this angle is measured at 6.4°.

The solution of DCM showed bright fluorescence at wavelength from 530 to 720 nm, bottom to bottom, in Fig. 2. The reason to choose this dye is its absorption at 400 nm, while its emission covers large portion of broadband superfluorescence range. The amplified spectrum is also shown in Fig. 2. At the beginning of time delay, when the pump pulse just passed sample cell, the transient fluorescence spectrum is of blue shift towards static emission. Both of the transient and static spectra start from 530 nm.

As shown in Fig. 3, four wavelengths are selected from the amplified full spectrum. They are 570, 590, 610 and 630 nm. Their beginning processes of 2 ps are shown in the figure. Clear difference was found between different wavelengths. The fluorescence at 570 nm starts to decrease after excitation. At longer wavelength after 600 nm, the dynamics goes upwards. There are several other factors need to be considered for raw data in this figure. One of them is the relative amplitude. It should be done on integration on each wavelength,

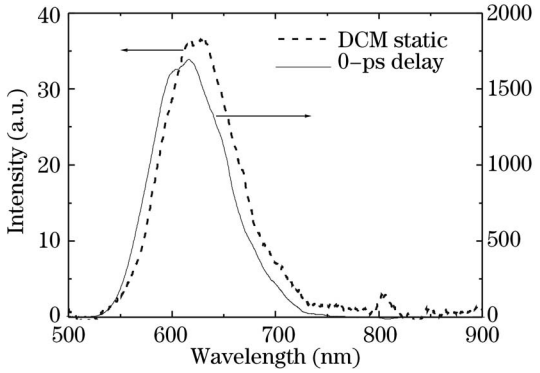

Fig. 2. The spectrum of static DCM dye and the transient amplified spectrum just after the excitation at 400 nm.

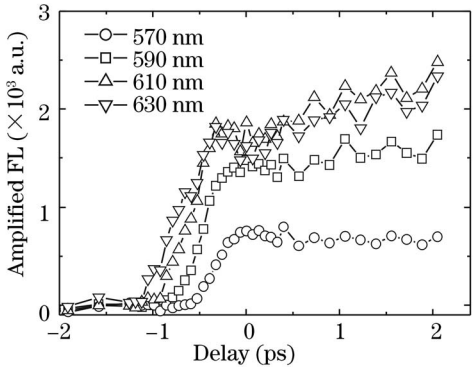

Fig. 3. The ultrafast dynamics of first 2 ps at 4 different wavelengths.

and then compared with static fluorescence spectrum. However, for DCM dye, its dynamics is longer than our delay line, which makes the normalization difficult. Another factor is the group velocity dispersion between each wavelength as present in the figure. This is easy to be corrected, by manually shift each wavelength to get same starting point of rising, where the front part of laser pulse starts to excite and all wavelengths start to radiate at same time.

In order to have larger amplification rate, this parametric amplification process should prove higher amplification rate, but avoid any saturation as possible. As tunable light source, this parametric process is required to work in its saturation region, in order to generate stable output. In our research, we should avoid this saturation. To have higher pump energy, the cone emission will occur as an accompanying effect for the fluorescence amplification. In our research, it is found that when above the cone emission generation threshold, the amplification is more efficient. As shown in Fig. 4(b), the superfluorescence appears after 20 μJ of pump energy. However, it is pretty weak. It becomes stable after 40 μJ and increases linearly with pump energy. While seeding the nonlinear crystal under different pump intensities, three sections of amplification could be found on Fig. 4(a). The first one is the low pump energy under 30 μJ. Here the superfluorescence just appears. The amplified signal increases with pump energy, but with lower rate than the second section. At second section, the superfluorescence becomes stable and brighter. With pump energy under 80 μJ, the seed is amplified with higher rate and increases linearly. While above 80 μJ of pump energy, the amplification is saturated and no more signal energy can be achieved. For this section, the superfluorescence is extremely bright. Therefore, proper pump energy of second section should be selected for linear amplification and high amplification rate.

However, the pump energy value mentioned here is not ruler for selecting proper pump light pulse. For example, the nonlinear crystal is placed out of focus of pump beam. It means the area of pump spot on crystal is widely adjustable. Therefore, it might be expected that the intensity per unit area could be used as standard. It may be true. However, as our observation, the if the crystal is place at focus of pump beam, no matter what is the pump energy, superfluorescence will not form properly. If no proper superfluorescence ring could be found,

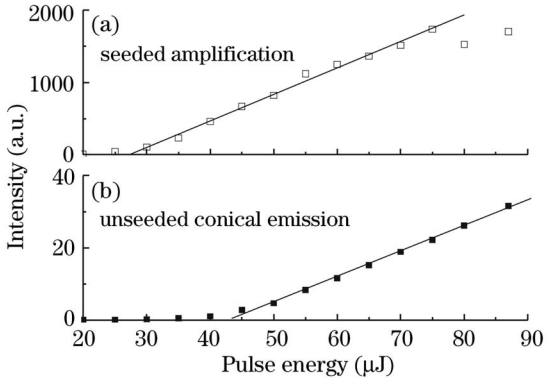

Fig. 4. The dependence of amplified seed and superfluorescence on pump pulse energy.

the seeded amplification could not be expected either. Therefore, we prefer to have another standard instead of pulse energy. We use the threshold of superfluorescence generation as reference, i.e., we tune pump energy according to the superfluorescence. The proper pulse energy will produce bright cone emission, but it will not reach extra bright region.

The intensity of seeding is another factor for the saturation. In Fig. 4, the seed comes from supercontinuum of water cell, which is quite bright. It is very short in duration, which is about a few picosecond. So it is quite strong within the gate time of 120 fs. The saturation reaches about 40 times intensity of superfluorescence. For real research on dye fluorescence, the amplified signals are only a few times, typically 2–5, higher than background superfluorescence. Therefore, the seed-intensity induced saturations are of less consideration. The linear amplification of seed under specified pulse energy was also verified. In our investigation, the amplification reaches to about 10^7. However, this rate could be higher if we pump the nonlinear crystal with higher energy. It will also bring larger superfluorescence background.

In conclusion, a broadband amplification condition inside nonlinear crystal was found. A full spectrum amplification of dye molecules is successfully performed. The conditions for this technique are investigated. We demonstrate that the superfluorescence background should be used as standard reference instead of any specified number on pump pulse energy. The linear dependences on pump energy and seed intensity towards output are verified. Basically, this technique is initially established and ready for further improvement.

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