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Compact optical scheme for the generation of ultrafast mid-IR laser pulses

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Abstract. Several optical techniques that are often employed to generate mid-IR pulses are discussed. These are based on a difference frequency generation (DFG) process for mixing signal and idler pulses generated from optical parametric oscillators that are synchronously pumped by Ti:Sapphire laser. Here, we proposed a new optical scheme that improves the day-to-day operation stability and the ease of manoeuvring spectrum anywhere in the 2 to 10 μm range. The demonstrated scheme is very compact and cost-effective, and we believe this optical scheme will be helpful for researchers working with mid-IR pulses.

Keywords. Silver thiogallate crystal ($AgGaS_2$ or AGS); optical parametric amplifier; signal and idler difference frequency mixing; mid-IR generation; collinear difference frequency generation mixing.

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1. Introduction

Infrared linear spectroscopic investigations on materials are ubiquitous and an essential tool for all branches of sciences and industry [1–3]. Over the past two decades, ultrafast mid-IR pulses have emerged as a powerful tool for nonlinear spectroscopy. In nonlinear infrared spectroscopy, pump-in visible/near IR and probe-in mid-IR are recently emerging in exploring the exciting physics of charge carriers in semiconductors and fascinating 2D materials [4,5]. Over the last two decades, two-dimensional infrared spectroscopy (2DIR) has become a powerful tool for monitoring the fast-changing vibrational couplings in molecules [6], molecular dynamics [7], energy transfer mechanisms [8], solvent dynamics [9,10], etc. An indirect way to access the vibrational levels is by Raman spectroscopy using visible pulses. High contrast Raman microscopy imaging is possible by pulse shaping difference frequency generation (DFG) [11]. Recently, it has been found that IR pulses are capable of doing molecular sensing through chemically sensitive microscopy [12].

To do this exciting research, one has to generate the mid-IR femtosecond laser pulses in the 2 to 10 μm range. In the last two decades, many approaches have been successfully demonstrated for direct IR generation, like supercontinuum generation [13,14], quantum cascaded laser [15], optical parametric oscillator [16,17], rare-earth-doped fluoride fibre laser [18–20] and transition-metal-doped chalcogenide laser [21,22]. However, many research labs use the commercially available Ti:sapphire regenerative amplifier, which produces femtosecond laser pulses from 10 fs to 150 fs, with a central wavelength of 800 nm at 1 kHz repetition rate. The optical parametric amplifiers (OPA) generate a pair of signal and idler pulses from a white light seed and a pump of 800 nm or the fundamental frequency of the laser. These signal and idler pulses are tunable from 1100 nm to 2700 nm by changing the angle of the BBO nonlinear crystals. The most popular way to generate IR light from these lasers is by the DFG between two near-infrared pulses (signal and idler). Difference frequency generation is a nonlinear optical process in which the generated light (mid-IR) frequency is the difference between two near IR pulses (signal and idler) obtained from an OPA. This IR generation from DFG crystals can be due to the collinear [23,24] or non-collinear [25,26] mixing of the signal and the idler. In our scheme here,

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we used collinear mixing of the signal and the idler in $AgGaS_2$ (silver thiogallate) DFG crystal using type-l nonlinear mixing of the signal (extraordinary) and the idler (ordinary) to generate mid-IR (ordinary) pulses.

This work discusses the existing optical schemes adopted, including the commercial set-up, to generate IR pulses using collinear mixing of the signal and the idler in DFG crystals. We will present the advantages and disadvantages of each scheme and experimentally demonstrate a new optical scheme for the IR generation which is repeatable, cheaper and compact that allows day-to-day stability to tune the wavelengths. By adopting this optical scheme, researchers can save their day-to-day alignment time for tuning the IR pulses, and it can be repeated with motorised components.

2. Experimental details

In our experiment, a Ti:sapphire ultrafast laser (Maitai, Spectra Physics) which produces 800 nm central wavelength, 75 fs pulse duration and 80 MHz repetition rate laser pulse was used as the seed laser. The oscillator output pulses were stretched and amplified by a regenerative amplifier (Spitfire Ace), pumped by a 1 kHz green laser (Ascend). The final amplified output laser pulse has 6 mJ energy per pulse with a time duration of 75 fs pulse width. The 1.2 mJ/pulse was used as the input for the commercially available optical parametric amplifier (OPA) TOPAZ prime from light conversion. We have commercial OPA (light conversion) to generate tunable signal and idler pulses, which were used for DFG generation to produce mid-IR (2 to 10 μm) pulses. The crystal we used for DFG was $AgGaS_2$ (2.5 mm thickness from ESKMA Optics $\theta = 37.1°$ and $\varphi = 45°$ with both sides coated with broadband antireflection coating 1.25–2.2/2–8 μm, Type-I nonlinear mixing with signal (e), idler (o) and IR (o) [23]. The 2–2.5 mm crystal lengths are optimal for 100 to 150 fs. For smaller pulse duration, thinner crystals are better. $AgGaS_2$ has nearly 70% transmittance between 2 and 10 μm which decreases for larger wavelengths. For longer wavelength (12–18 μm) IR generation, one can use GaSe or $AgGaSe_2$ crystals [27,28]. After generating IR pulses from the $AgGaS_2$ crystal, we need to place a long pass filter to remove the residual signal and idler (part#713M, Spectrogon AB, Sweden, transmission for IR >70%); this filter ensures the complete removal of the signal and the idler. To control the mid-IR intensity, we used a variable germanium-based optical density (OD) filters kit (supplied by Edmund Optics #64363) and CaF_2 beam splitters (Edmund Optics) before the detection. A CaF_2 lens with 50 mm focal length was used to focus the IR light into the slit of the spectrometer (ANDOR Shamrock SR-303i). The spectrometer has tripod grating available with 100 lines/mm (at a blaze angle of 5200 nm), 150 lines/mm (at 4000 nm) and 900 lines/mm (at 550 nm). The output of the spectrometer was detected by a single-pixel MCT detector (mercury cadmium telluride, Infrared Associates, USA), which is connected to a laser-synchronised BOXCAR integrator (SR-280). The DC output of BOXCAR was given to a data acquisition system (NI-BNC-2110) for the automated collection of data points, controlled by LabView programming. The power meter, Newport 843-R, was used for measuring power. Also we presented the noise measurements in the supplementary information, in which we detected a small fraction of IR pulses. This IR beam was split into two equal parts for balanced detection measurements and collected by two detectors (reference I_R and signal I_S). These two outputs were detected simultaneously, and we performed the mathematics $[(I_S - I_R)/I_R]$ in ref. [29].

3. Salient features of the widely used DFG optical schemes

First, we would like to introduce the different schemes adopted by researchers for the generation of mid-IR pulses using DFG crystals. A pair of tunable signal and idler pulses are the pre-requirements for generating mid-IR pulses. Several optical schemes and crystals for optical parametric amplifiers generate a unique tunable pair of signal and idler using the 800 nm fundamental beam. Typically, the signal wavelength is from 1100 nm to 1600 nm and the idler wavelength is from 1600 to 2700 nm. The most popular nonlinear crystals for generating signal and idler are BBO and BiBo crystals for OPG/OPA [30–32].

3.1 *Commercial DFG scheme*

In our laboratory, we have commercial OPA Topaz prime along with an IR mixer just after the OPA, as shown in figure 1. The IR crystal is mounted on a rotational stage to tune the phase-matching angle for generating IR pulses based on the input signal and idler spectrums. The wavelength compensator is used before the crystal to match the signal and idler temporally. However, this compensator is designed for only one particular wavelength as the user cannot change it.

This DFG set-up has a few disadvantages in day-to-day operation. The position of the DFG crystal must be fixed from the OPA and it cannot be kept at the desired location on an optical table. As a result, if the mid-IR experiments are far from IR generating crystal, you need

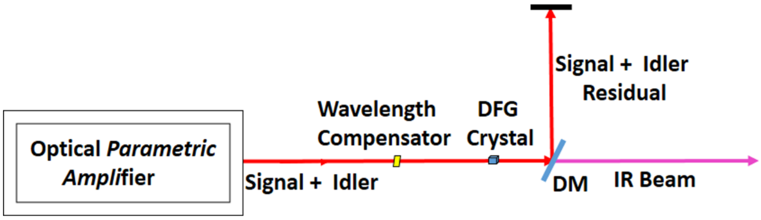

Figure 1. Optical lay-out of the commercial set-up of difference frequency generation (DM denotes dichroic mirrors).

to create a larger volume of purging system to remove the carbon dioxide and water vapour in the path of the IR beam. Also, due to the minute misalignment as a consequence of thermal and environmental changes in the laboratory, the signal and idler beams coming from OPA can make a small angle with each other. This can lead to a slight spatial separation at the DFG crystal, resulting in an inefficient conversion to generate the IR signal. Both signal and idler will not have complete temporal overlap over the entire tunable range of the spectrum and it can change daily based on the OPA alignment. This results in a spatial and temporal mismatch of the signal and idler pulses in the crystal and IR beam quality and stability will vary from day-to-day operation. In this commercial set-up configuration, researchers cannot control the signal and idler beams. In advanced experiments where a high signal-to-noise ratio is essential, one must control the signal and idler temporal and spatial overlap for producing stable mid-IR pulses. Of course the advantage of this commercial system is that there is no need for any optical alignment. However, it lacks control in IR signal generation efficiency and its day-to-day instability is a significant concern.

One must precisely overlap the signal and idler pulses in the DFG crystal both spatially and temporally for efficient nonlinear conversion. This can be achieved only when the signal and idler are independently aligned. For this purpose, we need to split the signal and idler and recombine them back in the crystal, which can be achieved in several ways.

3.2 *Optical scheme with two dichroic mirrors*

Many research groups use this optical scheme for mid-IR generation (figure 2) [22,23]. The first dichroic mirror splits the signal and idler and recombines them again using another dichroic mirror. One of the arms has a delay control for the exact time overlap of the signal and idler pulses. This method makes the signal and idler pulses overlap in the DFG crystal for stable IR generation. However, using two expensive dichroic mirrors, extra mirrors and optomechanics make the set-up expensive. Also, energy losses will be more at each reflection and it will have a larger footprint on an optical table.

3.3 *Single dichroic mirror scheme*

To overcome the disadvantages of cost, energy losses and size of the DFG unit, a single dichroic mirror is used, as shown in figure 3. Similar to the previous optical scheme, signal and idler pulses are coming from an OPA and separated by a dichroic mirror and recombined on the same dichroic mirror at another point vertically downwards or sideward, returning with a slight angle (from M4 and M5 mirrors in figure 3).

Most laboratories have a single dichroic mirror to separate the signal and idler, supplied by the commercial OPA. This set-up can be readily made in any laboratory with commercial OPA. The disadvantage of this set-up is that when we change the wavelength of the signal and the idler, for tuning the IR wavelength, there will be a change in the temporal overlap. To compensate for this temporal overlap, one has to play with the delay in one of the arms. Since the returning beam makes an angle downwards/sideward with the original input beam, the recombination of the signal and idler will not be at the same point; it will disturb the spatial overlap of the signal and the idler. One must readjust the mirror to overlap the signal and the idler on the crystal to generate the maximum IR output. Due to this, tuning of the desired IR wavelength has to be done iteratively for every 100 to 200 nm step. The complete IR signal may be lost if larger steps are adopted. Typically, the researcher needs to spend a lot of time in generating the desired spectrum and maximising the stability and energy of mid-IR pulses.

4. New optical scheme with vertical rooftop prisms and single dichroic mirror

We adopted a new and very simple design to overcome the disadvantage of the previous schemes, as shown in figure 4. Here, the signal and the idler coming from OPA are separated by a dichroic mirror and they are retraced back to the same dichroic mirror by using retro mirrors without making any angle. They recombined at a different point vertically downward and were picked up by

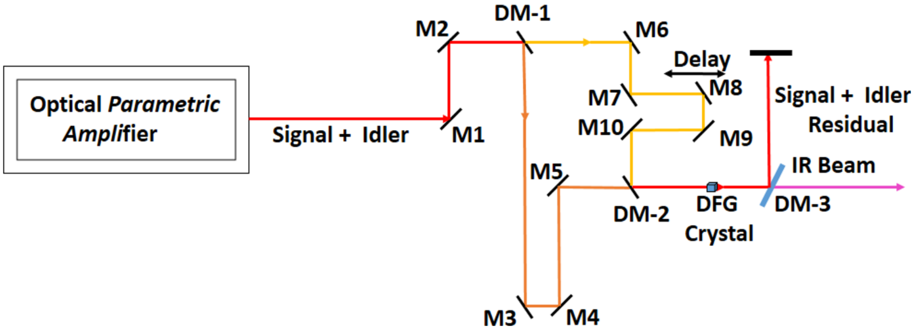

Figure 2. Optical lay-out of the DFG set-up using three DMS (M1 to M10 denote mirrors).

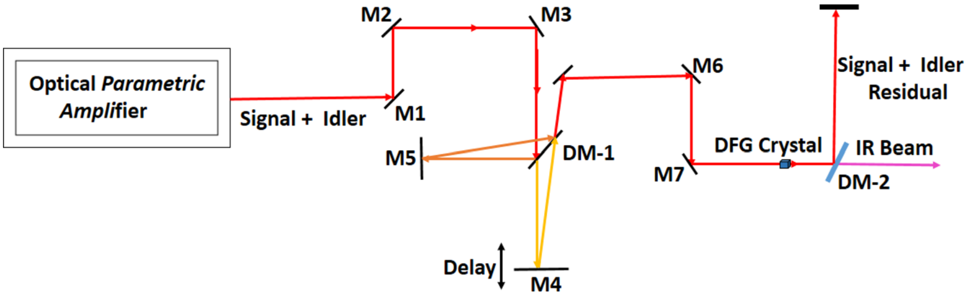

Figure 3. The optical lay-out of the DFG set-up using two DMs (M denotes mirrors).

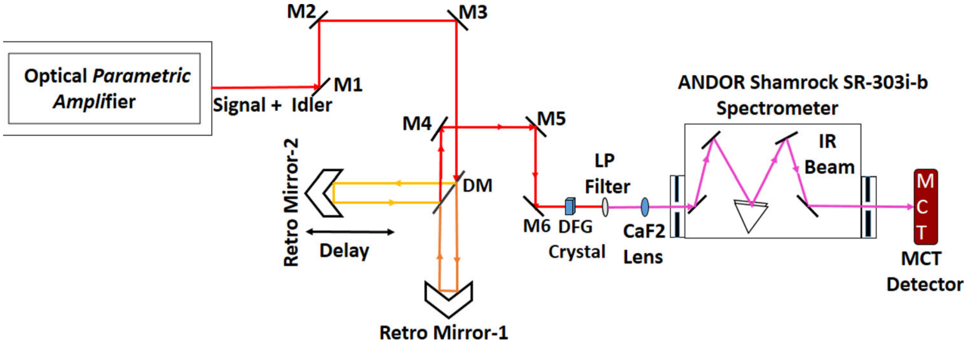

Figure 4. Optical lay-out of a home-built DFG set-up using the retro mirror (DM denotes the dichroic mirror, LP is the long pass filter, M are the mirrors, CaF_2 lens is the calcium fluoride lens and MCT is the HgCdTe detector).

another mirror. The time delay in one of the arm provides a tool for precisely adjusting the temporal overlap of the signal and the idler pulses in the DFG crystal. Because of the retro-reflector configuration, the change of the delay does not disturb the spatial overlap of the signal and the idler. This is the key for easily tuning the spectrum anywhere in the 2 to 10 μm region. We can move the spectrum from anywhere in the range without losing the IR energy for any given pair of the signal and the idler pulses; only the delay needs to be adjusted for maximum IR output.

The DFG crystal can be placed very far from the beam splitter and near the sample for IR experiments. This will reduce the purging volume for mid-IR experiments. Effective purging is one of the most important factors for attaining the best signal-to-noise ratio for sensitive mid-IR experiments. By adjusting the angle of the crystal, one can maximise the phase-matching condition for generating stable IR pulses. We collected the IR spectrum at several places from 2 to 8 μm. The normalised spectrum is shown in figure 5. The stability and repeatability of the IR pulses are in excellent correlation with the delay of the retro-reflector. The output energy produced at different wavelengths is presented in figure 6. The measured energy is good and much stable in our daily operations. The IR output can vary depending on the amplifier output, which generally varies due to thermal conditions, alignment and environmental factors. However, this set-up allows one to get maximum IR energy with minimum adjustments on a given day.

This set-up is very cost effective compared to the commercial system but yet it gives more stable mid-IR pulses

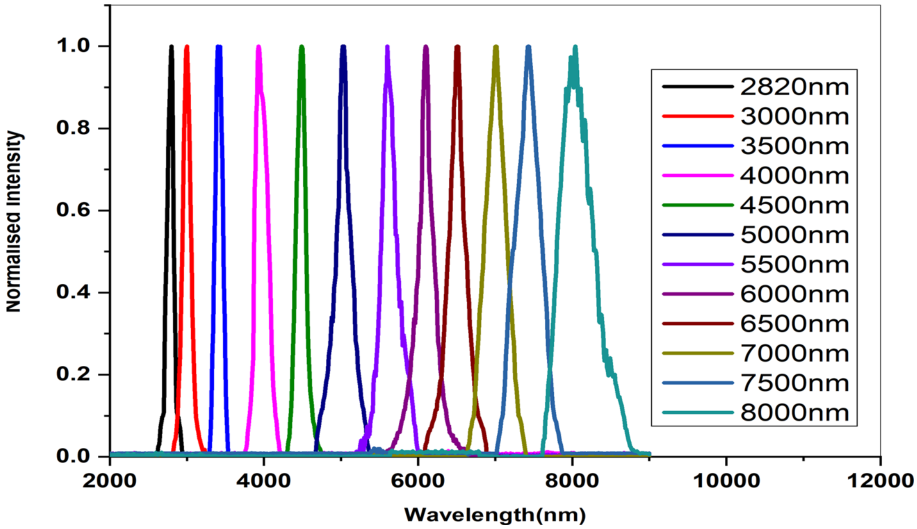

Figure 5. Spectral tuning of the IR pulses over the range of 2–8 μm using a home-built DFG.

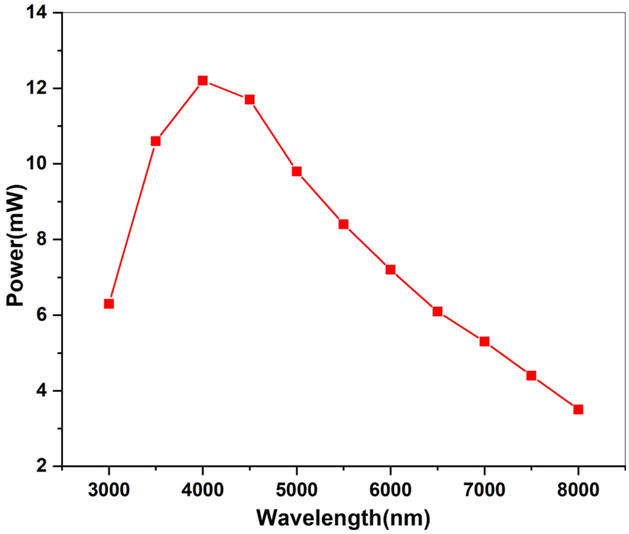

Figure 6. Experimentally obtained IR power curve with 1.1 W fundamental OPA input.

and one can easily build it with ease. For all our IR pump probe experiments, we need to use the OD filters ranging from as high as 6 OD at 4 μm and 1.5 OD for 8 μm, in spite of having two 50:50 IR beam splitters to reach the MCT detectors either directly or through the spectrometer. The IR power measurements are shown in figure 6. These are done after the DFG crystal and the long pass filter. The IR generation efficiency depends on the fundamental pump pulse parameters. They do change slightly in normal day-to-day operation. But this set-up allows one to maximise the IR power generation by adjusting the delay and overlap of the signal and idler pulses. This is not possible in the commercial set-up.

5. Conclusion

We demonstrated a new optical scheme to generate the tunable mid-IR pulses through difference frequency generation of the signal and the idler pulses generated from OPA. We also presented the other optical schemes which are widely used in the laboratories for the generation of mid-IR pulses. The proposed new DFG scheme is compact and cost-effective compared to other schemes. One of the major advantages of this set-up is the ease of moving the spectrum anywhere in single step. Also, the produced IR pulses are very stable, repeatable with delay positions and can be maximised with minimum expertise or effort. This optical scheme can be easily implemented and will help researchers to save time in getting stable IR pulses in their daily operation.

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