# Efficient generation of broadband MIR radiation by difference–frequency generation in LiGaS$_2$

*Bo-Han* Chen[1,2,*], *Tamas* Nagy[3], *Peter* Baum[1,2,4,*]

[1]Ludwig-Maximilians-Universität München, Am Coulombwall 1, 85748 Garching, Germany
[2]Max-Planck -Institute of Quantum Optics, Hans-Kopfermann-Str. 1, 85748 Garching, Germany
[3]Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2A, 12489 Berlin, Germany
[4]Universität Konstanz, Universitätsstraße 10, 78464 Konstanz, Germany

**Abstract.** We report a surprisingly broadband and efficient middle-infrared (MIR) pulse generation in LiGaS2 (LGS) by invoking a simultaneous interplay of intra-pulse difference frequency generation, self-phase-modulation and dispersion.

## 1 Introduction

Short and broadband optical pulses at mid-infrared (MIR) or multi-THz frequencies have a key importance in various research fields, for example in molecular fingerprint spectroscopy [1], all-optical electron pulse compression [2] and high-field experiments in condensed matter [3-5]. A common way to produce MIR few-cycle pulses is intra-pulse difference frequency generation (DFG) in a nonlinear crystal. One of the most favorable materials is LiGaS$_2$ (LGS). Although its effective nonlinearity is rather low, the material has on the other hand a wide transparency range, a high damage threshold and a particularly weak two-photon absorption. However, temporal walk-off between the two required driving polarizations limits the overall efficiency and spectral bandwidth of the MIR output. In this work, we demonstrate that broadband and efficient MIR generation is still feasible even with long LGS crystals [6].

## 2. Concept and experiment

Fig. 1(a) shows the experimental setup. 1-ps pulses from a regenerative thin-disk laser are first compressed down to 30 fs through 2 stages of nonlinear spectral broadening processes, pulse compression, and then focused into a LGS crystal for MIR generation. Fig 1(b) shows the generation mechanism: the temporal walk-off is continuously compensated by nonlinear spectral broadening and dispersion of the driving pulses during propagation, in such a way that the high-frequency components of the extraordinary/faster part of the pump

---

[*] Corresponding author: bohan.chen@physik.uni-muenchen.de, peter.baum@uni-konstanz.de

pulse and the low frequency components of the ordinary/slower part of the pump pulse have a continuous overlap all the time. It allows us to use substantially longer crystals than the expected walk-off length, in order to simultaneously achieve higher MIR frequencies and more output power.

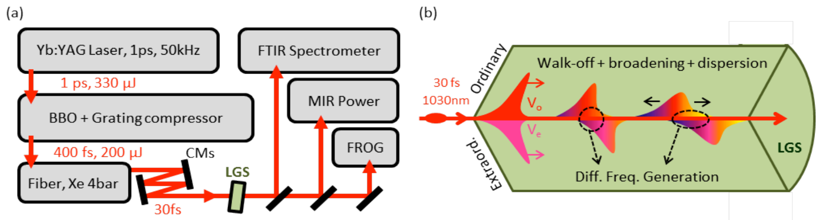

**Fig. 1.** (a) Experimental setup and, (b) concept for simultaneous, cascaded spectral broadening and intra-pulse difference-frequency generation [6].

## 3. Experimental results

To support the mechanism proposed in Fig. 1(b), we provide experimental evidences spectrally and temporally, which are shown in figure 2(a)–2(d). First of all, with higher input power the short wavelength side of generated MIR spectrum becomes stronger than the longer wavelength side, see Fig 2(a). This is an indication that the incoming spectrum is getting broader, leading to higher difference frequencies. It is also clear that with longer LGS the input spectrum gets broader, as shown in Fig. 2(b). Fig. 2(c) and 2(d) show the retrieved temporal shape from SHG-FROG measurement of the driving pulse after passing through LGS. In the case of low power, the pulses of two polarizations get separated, while in the case of high power they are still overlapping and interacting with each other, as we expected from the picture shown in Fig. 1(b).

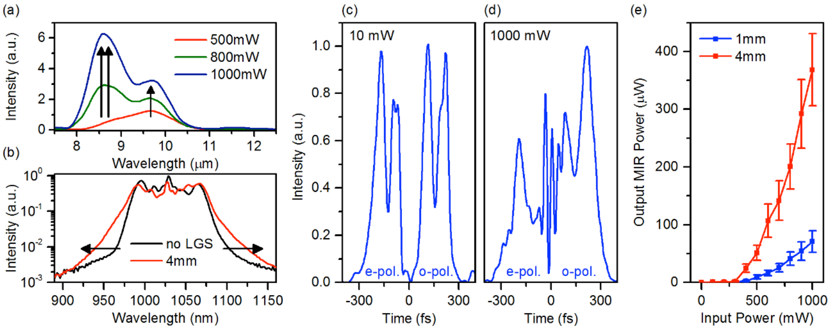

**Fig. 2.** Evidence in the spectral and temporal domain. (a) MIR spectra with different input power. (b) Optical spectrum in air and after 4mm LGS. Retrieved FROG pulse shape with an input power of (c) 10 mW, and (d) 1000 mW. (e) Output power with a 1-mm (blue) and a 4-mm (red) LGS crystal. The 4mm LGS performs much better, although it is 8 times longer than the walk-off distance.

In a final measurement, we compared the overall performance of a 1-mm and a 4-mm LGS crystals. Figure 2(e) shows the power dependence of these two crystals. As one would expect from a $\chi(2)$ process, a quadratic-like power dependence is measured, but the 4-mm crystal performs substantially better. With such nanojoule pulses and MV/m E-fields, as now available at 7.5-11 $\mu$m, it is well sufficient for our intended applications in femtosecond and attosecond electron microscopy [2, 7, 8], but also for many other applications.

## 4. Conclusion

We conclude with three remarks. First, in a way, our reported combination of nonlinear-optical self-phase modulation with nonlinear-optical difference frequency mixing is an example of cascaded or multi-process nonlinear optics [9, 10], a concept which seems to be generally useful. Second, it is clear now why somewhat broader MIR spectra than expected have been reported before [11]. Third, there should be other optical materials for which the above picture also applies, namely whenever self-phase modulation and dispersion create enough temporal effects to compete with walk-off. Novel optical applications might be ahead.

## 5. References

1. A. Schliesser, N. Picque, and T. W. Hansch, Nat. Photonics, **6**, 440–449, (2012).
2. C. Kealhofer, W. Schneider, D. Ehberger, A. Ryabov, F. Krausz, and P. Baum, Science, **352**, 429–433, (2016).
3. S. Ghimire, A. D. DiChiara, E. Sistrunk, P. Agostini, L. F. DiMauro, and D. A. Reis, Nat. Phys., 7, 138–141, (2011).
4. O. Schubert, M. Hohenleutner, F. Langer, B. Urbanek, C. Lange, U. Huttner, D. Golde, T. Meier, M. Kira, S. W. Koch, and R. Huber, Nat. Photonics, **8**, 119–123, (2014).
5. M. Hohenleutner, F. Langer, O. Schubert, M. Knorr, U. Huttner, S. W. Koch, M. Kira, and R. Huber, Nature, **523**, 572-575, (2015).
6. B.-H. Chen , T. Nagy and P. Baum, Opt. Lett., **43**, 1742-1745, (2018).
7. A. Ryabov and P. Baum, Science, **353**, 374–377, (2016).
8. Y. Morimoto and P. Baum, Nat. Phys., **14**, 252–256 (2018).
9. C. Homann, M. Bradler, M. Forster, P. Hommelhoff, and E. Riedle, Opt. Lett., **37**, 1673–1675, (2012).
10. Hanieh Fattahi, Alexander Schwarz, Sabine Keiber, and Nicholas Karpowicz, Opt. Lett., **38**, 4216–4219, (2013).
11. I. Pupeza, D. Sanchez, J. Zhang, N. Lilienfein, M. Seidel, N. Karpowicz, T. Paasch-Colberg, I. Znakovskaya, M. Pescher, W. Schweinberger, V. Pervak, E. Fill, O. Pronin, Z. Wei, F. Krausz, A. Apolonski, and J. Biegert, Nat. Photonics, **9**, 721-724, (2015).