A vibrational sum-frequency generation spectrometer enabled by a high-power few-cycle MID-IR OPCPA from 7 – 11 µm

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Motivation

Monitoring blood glucose levels is a critical task in the prevention and management of many common diseases, in particular, diabetes. The vibrational modes of glucose span from 900 to 1400 cm^{-1} (11 – 7 µm). Conventional methods of glucose detection, using long pulse or continuous lasers across these vibrational modes, have limited sensitivity. However, ultrashort MID-IR pulses offer much higher sensitivity [1] and can identify different cellular components using hyperspectral imaging [2].

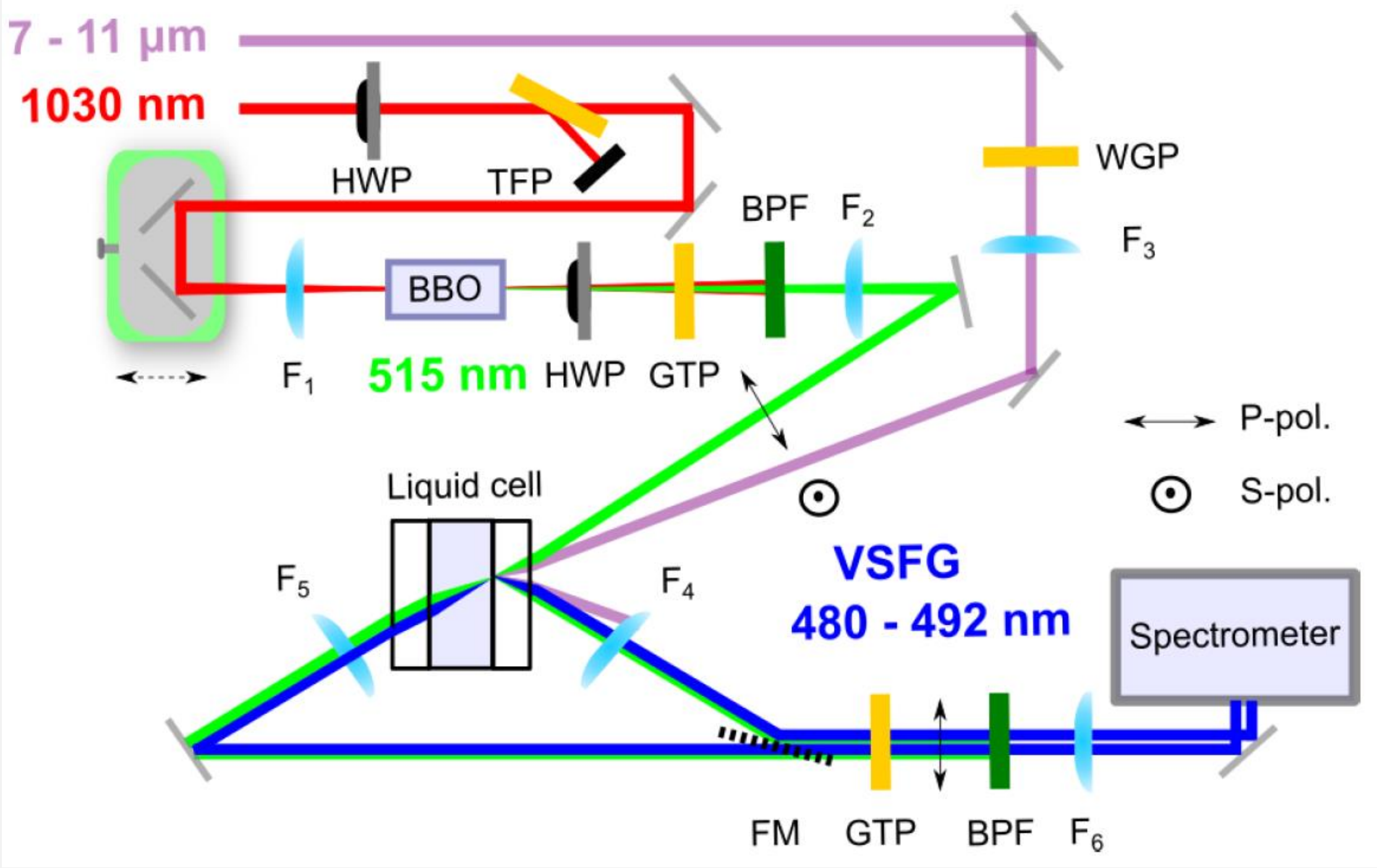
In this poster, Class 5 Photonics together with the group of Prof. T. Laarmann from DESY have developed a ultra-broadband vibrational sum-frequency generation (VSFG) spectrometer enabled by a White Dwarf-HE-MIR from Class 5 Photonics [3]. This laser generates broadband pulses (7 – 11 µm) centered at 9 µm with a pulse duration of 114 fs and high average power of 245 mW and pulse energy of 1.2 µJ [4]. This development prepared the ground for observing distinct glucose absorption bands using VSFG spectroscopy at physiological relevant low concentrations.

Optical Layout

White Dwarf – HE – MIR

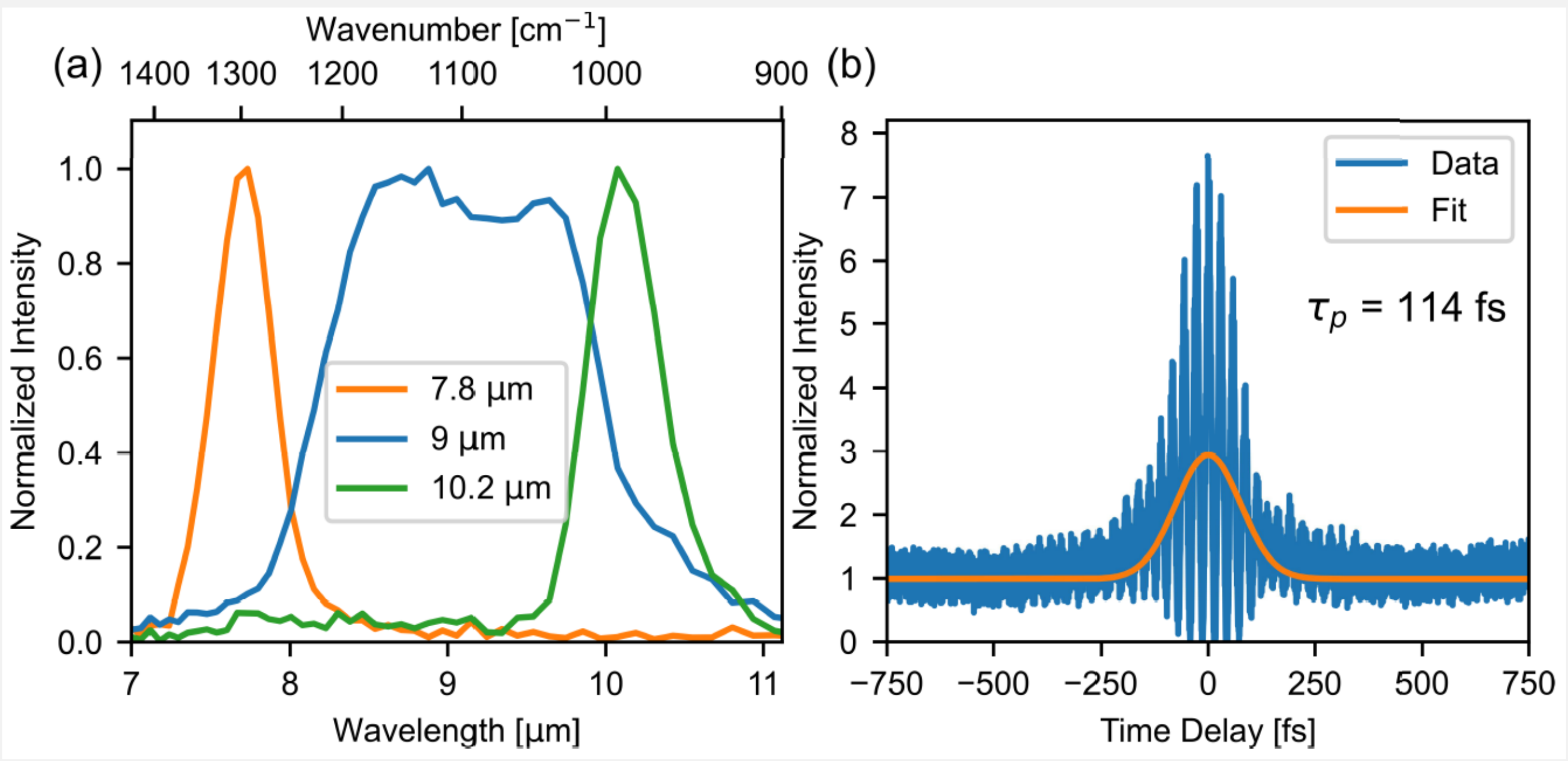
Outputs:
7-11 µm (100 fs, 3 W)
1030 nm (< 1ps, 10 W)

Rep. Rate: 100 kHz – 1 MHz



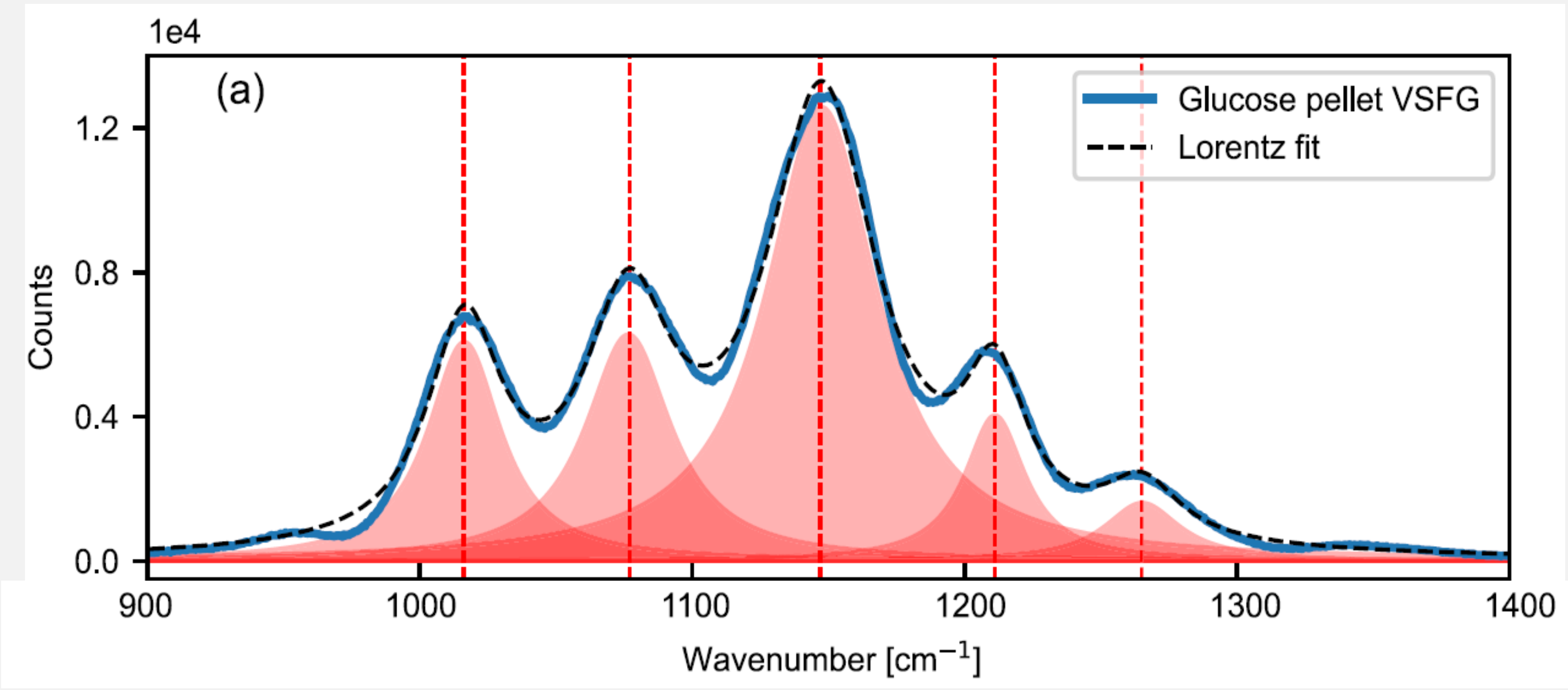
Schematic of the ultra-broadband VSFG spectrometer, pumped by a White Dwarf - High Energy – Mid Infrared (WD-HE-MIR) laser from Class 5 Photonics: HWP – half-wave plate, TFP – thin-film polarizer, BBO – BaB_2O_4, GTP – Clan-Thompson polarizer, WGP – wire grid polarizer, BPF – bandpass filter, F – lenses, FM – flip mirror.

Laser Performance: White Dwarf – HE – MIR



(a) Wavelength-tunable MID-IR pulse spectra generated from the negatively chirped signal NOPA scheme at 7.8 µm, 9 µm, and 10.2 µm central wavelength. (b) The second-order interferometric autocorrelation trace (blue data points) of the broadband, 9 µm MID-IR pulse fitted with a Gaussian function (orange line) reveals a pulse duration τ_p = 114 fs (for details, see ref. [4]).

VSFG spectrum of glucose pellets



Recorded VSFG spectrum from glucose pellets (blue line) fitted with Lorentzian functions (dotted black lines). The observed resonances (light red areas) can be assigned to certain vibrational modes indicated by red dashed vertical lines.

The spectrometer was tested with a sample of naturally occurring D-glucose powder (Sigma-Aldrich), hand-pressed into a pellet. To prevent the glucose surface from melting, the excitation power levels in the visible at 515 nm and in the MID-IR are restricted to 50 mW and 40 mW, respectively. A distinct cumulative VSFG spectrum is obtained with an exposure time of 30 s, displaying 5 prominent modes ranging from 900 to 1400 cm^{-1}. Notable, the observed VSFG coincide with IR and Raman active modes. Further details are given in reference [3].

Vibrational Sum-Frequency Generation (VSFG)

VSFG involves the combination of infrared (IR) and visible (VIS) laser beams with intensities I_{IR} and I_{VIS}, respectively. If the broadband IR pulse (ω_{IR}) is resonant with a vibrational mode in a molecule (v_0 -> v_1) and a visible beam (ω_{VIS}) interacts as the vibrational mode decays back to the ground state, then a sum frequency wave ($\omega_{SFG}=\omega_{IR}+\omega_{VIS}$) is generated. In this process, the spectral resolution is determined by the spectral bandwidth of the visible beam. Further details are given in reference [3].

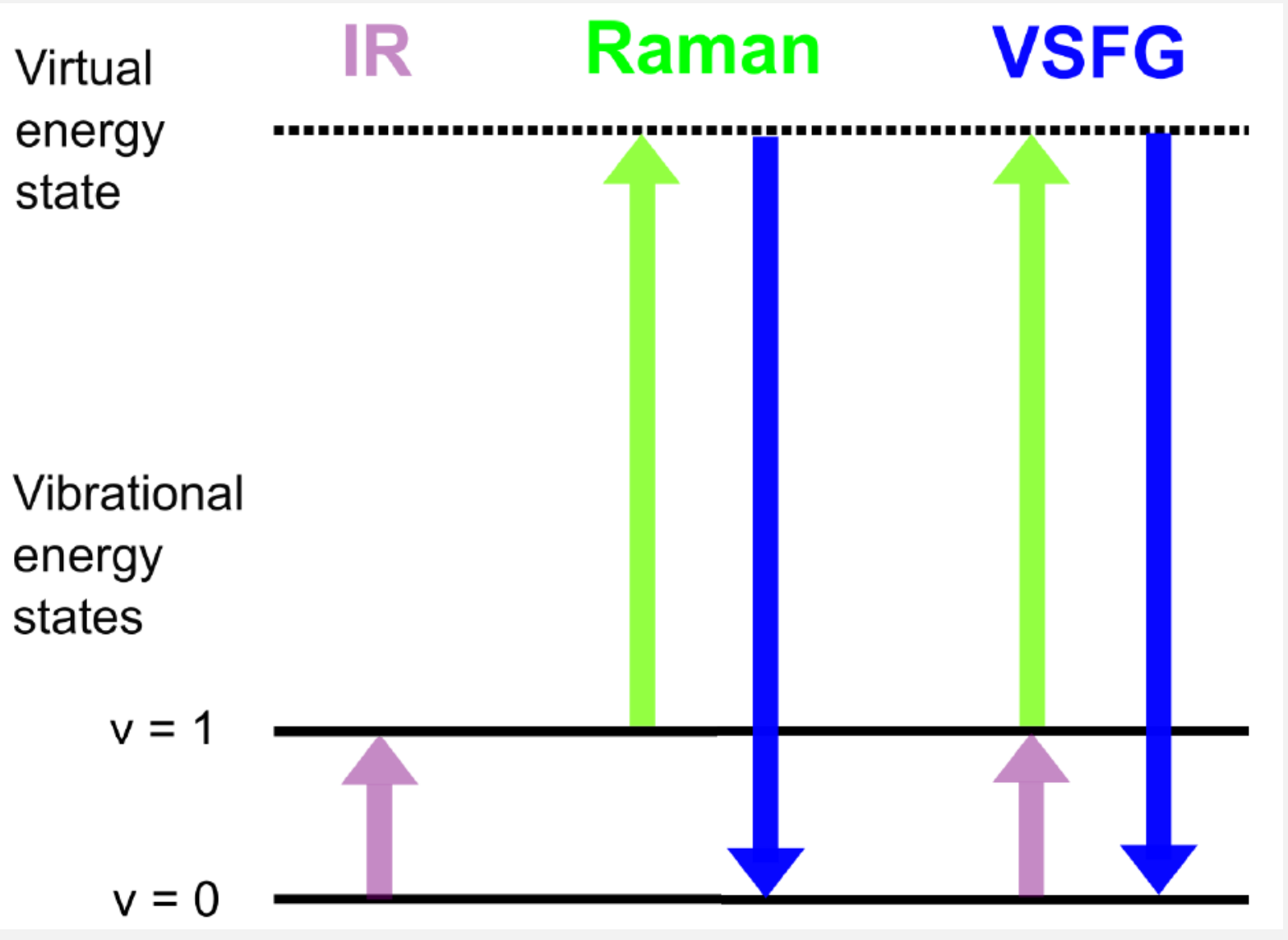
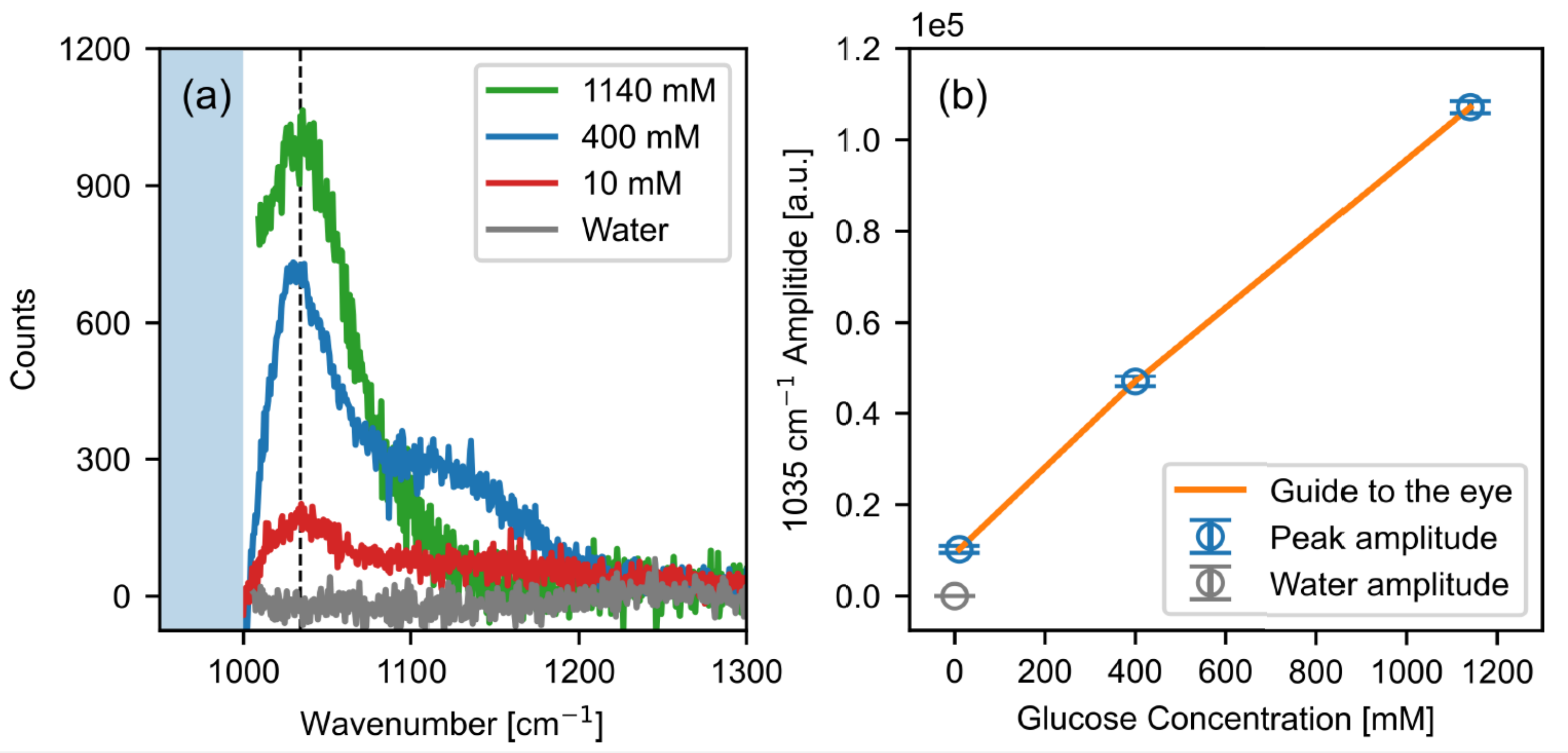


Illustration of vibrational sum-frequency generation (VSFG) with contributions arising from IR absorption of vibrational modes (v = 0, 1), and Raman scattering.

VSFG spectra of glucose in aqueous solutions



(a) Recorded VSFG spectra from aqueous glucose for different concentrations (1140, 400, 10 mM) of the solute in the transmission mode of signal detection. A pure water solvent spectrum is shown in gray. (b) The Lorentzian amplitude of the 1035 cm^{-1} C-O stretch vibration indicated in (a) by a dashed vertical line is given as a function of glucose concentration.

Different concentrations of glucose powder are dissolved in 100 ml of deionized water, where the solution is placed in a liquid cell of 5 mm thick. Note, IR spectroscopy on aqueous glucose is challenging, because of the high absorption of liquid water. Here, we make use of the highly transparency of water in the spectral range of 480 to 492 nm, which allows for measuring of the transmitted VSFG signal. The key-technological component allowing for biologically relevant applications in aqueous solutions is the White Dwarf-HE-MIR laser.

In a future development, we are developing this system towards a time-resolved VSFG spectroscopy and VSFG microscopy.

References

[1] H. Huber, et al, "Detection sensitivity of field-resolved spectroscopy in the molecular fingerprint region", *CLEO/Europe-EQEC* pg. 1 (2017)
[2] Y. Zhao, et al, "High-speed scanless entire bandwidth mid-infrared chemical imaging", *Nat. Commun.* **14**(1) 3929 (2023)
[3] C. Luo, et al, "Spectral fingerprinting of aqueous glucose with ultra-broadband vibrational sum-frequency generation spectroscopy at bio-relevant low concentration", *Opt. Express,* **33,** 6080 (2025)
[4] M. Namboodiri, et al, "Versatile few-cycle high-energy MID-IR OPCPA for nonlinear optics, spectroscopy and imaging", *Opt. Continuum* **1**(5) 1157-1164 (2022)

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