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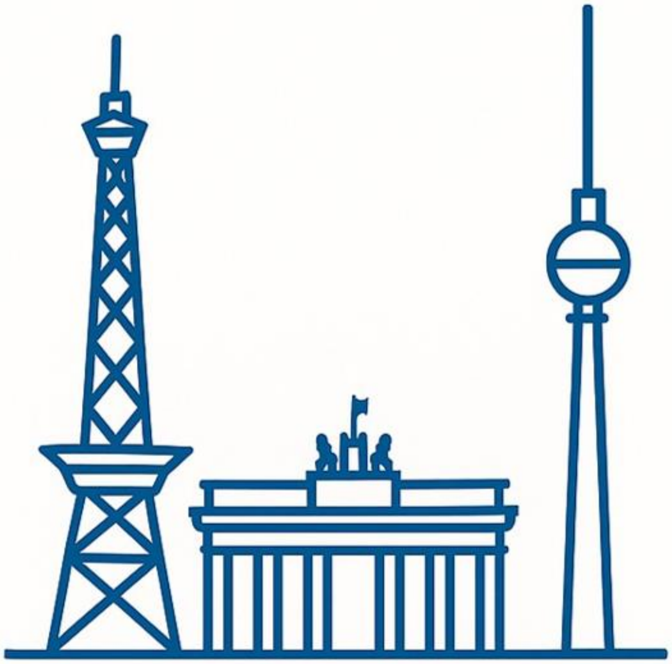

BERLIN ECONOS 2026

29 March – 1 April 2026

Federal Ministry of Research, Technology and Space
Grant-ID: 03RU2U033F

DFG Deutsche Forschungsgemeinschaft
German Research Foundation
Grant-ID: 580147127

MAX PLANCK SCHOOL of photonics

Program

Sunday, 29 March 2026

13:00 **Registration**

15:00 **Welcome**

15:10 **Keynote I - Gerd Leuchs**

Influence of the correlations of the light field on nonlinear light matter interaction

16:10 Coffee Break

16:30 **Keynote II - Markus Antonietti**

Light as a Reagent – Artificial Photosynthesis Well Beyond Nature

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Monday, 30 March 2026

Session I - Precision Spectroscopy

09:00 **Nathalie Picqué** - invited

Nonlinear dual-comb spectroscopy

09:40 **Antoine Espaignol**

Impulsive molecular alignment of acetylene through vibrational resonance

10:00 **Francesco Vergari**

Interferometric effects in multiple fs/ps rotational coherent Raman scattering

10:20 **Francesco Mazza**

Atomic coherent anti-Stokes Raman spectroscopy in metal vapours

10:40 Coffee Break

Session II - Molecular Interfaces and Imaging

11:10 **Michael Schmitt** - invited

Advances in Nonlinear Multimodal Imaging and Hyperspectral Coherent Raman Spectroscopy for Precision Oncology

11:50 **Nasim Mirzajani**

Mapping In-plane Orientational Order and Correlation Lengths in Molecular Films using Azimuthal-Scanning Vibrational Sum-Frequency Generation Microscopy

12:10 **Nick Lemberger**

Automated correction of optical attenuation in stimulated Raman scattering imaging

12:30 **Sighart Fischer**

Interpretation of the primary chiral charge separation on the basis of second order spin-orbit-coupling

12:50 **Lunch**

14:00 **Industrial Session**

14:30 **Poster Session**

16:30 **Free Time**

Tuesday, 31 March 2026

Session III - Combustion Diagnostics

09:00 **Robert Lucht** - invited

Application of Femtosecond CARS for Measurements in High-Pressure Reacting Flows

09:40 **Clément Pivard**

55Hz rate fs/ps-CARS spectroscopy for combustion thermometry

10:00 **Michael Scherman**

High-speed fs/ps-CARS of H_2 for energetics and propulsion studies

10:20 **Jonas Hölzer**

Rotational coherent anti-Stokes Raman scattering evaluation of collisional dephasing in nitrogen ammonia gas mixtures

10:40 Coffee Break

11:10 **Waruna Kulatilaka** - invited

Femtosecond, Two-Color, Two-Photon, Polarization Spectroscopy for Quenching-Free Detection of Atomic Hydrogen

11:50 **Jeanna Buldyreva**

Rotational-CARS thermometry of ethylene relying on accurate linewidth calculations

12:10 **Alexander Fellows**

Imaging Structural Anisotropy and Heterogeneity in Molecular Films with Sum-Frequency Microscopy

12:50 **Lunch**

14:00 **Christian Eggeling** - invited

Advanced super-resolution optical microscopy for cellular research

14:40 **Carsten Fallnich**

Photothermal stimulated Raman scattering with two beams only

15:00 **Timea Koch**

Ultrabroadband CARS Detection of Solvation-Induced Vibrational Changes in Liquid Mixtures

15:20 Coffee Break

16:00 **Social event:** Guided Tour "100 years of Science in German Oxford"

17:30 **Steering Committee** Meeting

19:00 Conference Dinner, **Wirtshaus "Fischerhütte"**, Fischerhüttenstraße 136, 14163 Berlin

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Wednesday, 1 April 2026

09:00 **Alexis Bohlin -** invited

The shaping of aeroengines with diagnostics

09:40 **Tom Lippoldt**

In-situ analysis of thermal decomposition of calcium oxalate by simultaneously applying Thermogravimetric Analysis and Ultrabroadband Coherent Anti-Stokes Raman Spectroscopy

10:00 **Steffen Gommel**

Sub-Cycle Response of Indium Tin Oxide film at its Epsilon-Near-Zero Region by Fieldoscopy

10:20 Coffee Break

10:50 **Alexandros Gerakis** - invited

Thermodynamic characterization of neutrals, ions, electrons and nanoparticles in high speed flows and gas discharges with coherent Rayleigh Brillouin scattering

11:30 **Closing session**

12:00 **Lunch**

13:30 **End of ECONOS 2026**

Abstracts Talks

Nonlinear dual-comb spectroscopy

Nathalie Picqué,

Max-Born Institute, Berlin, Germany

nathalie.picque@mbi-berlin.de

Optical frequency combs provide precise rulers in frequency space, enabling direct measurements of optical frequency differences and straightforward links between microwave and optical frequencies. They underpin precision laser spectroscopy, tests of fundamental physics, and optical atomic clocks. One of their most powerful applications beyond metrology is dual-comb interferometry [1-3]. Dual-comb interferometry uses two frequency combs with slightly different repetition frequencies to form an interferometer without moving parts. Unlike conventional interferometers, dual-comb systems perform direct frequency measurements and are therefore not limited by geometric constraints on resolution. They enable broadband, high-resolution, and rapid measurements and have become powerful tools for spectroscopy and distance metrology.

Although dual-comb spectroscopy has mainly been applied to linear spectroscopy of small molecules, frequency combs are typically generated by ultrashort-pulse mode-locked lasers whose high peak powers enable nonlinear optical interactions. This makes it possible to perform broadband nonlinear and multidimensional spectroscopy without mechanical delay lines. Techniques demonstrated so far include nonlinear Raman spectroscopy, two-photon excitation, pump–probe measurements, four-wave mixing, transient absorption, and two-dimensional spectroscopy using asynchronous sampling.

Combining nonlinear spectroscopy with dual-comb techniques also enables Doppler-free measurements. Recent dual-comb interferometers based on self-referenced fibre lasers record spectra spanning more than 1 THz with 1 MHz line spacing, corresponding to resolving powers exceeding 2×10^8 in a single acquisition. This opening intriguing prospects for broadband precision spectroscopy.

This talk will provide a brief introduction to optical frequency combs and dual-comb spectroscopy and will discuss results and prospects in nonlinear dual-comb spectroscopy.

1. N. Picqué, T.W. Hänsch, Dual-comb spectroscopy, *Nature Reviews Methods Primers*, in press (2026).
2. N. Picqué, T.W. Hänsch, *Photoniques* **113**, 38-42 (2022).
3. N. Picqué, T.W. Hänsch, *Nature Photonics* **13**, 146-157 (2019).

Impulsive molecular alignment of acetylene through vibrational resonance

Antoine Espaignol, Pierre Béjot, Edouard Hertz, Franck Billard and Olivier Faucher

Université de Bourgogne Europe, ICB Institut Carnot de Bourgogne,

Dijon, France

We study the impulsive alignment of acetylene (C_2H_2) molecules induced by resonant vibrational excitation with ultrashort infrared laser pulses. Two vibrational bands are considered: the fundamental ν_3 mode at 3 µm and the combination band $\nu_1 + \nu_3$ at 1.5 µm. Theoretical results are compared with experimental data. Simulations show that excitation of the ν_3 band produces a strong and long-lived planar alignment typical of resonant mechanisms, similar to effects recently reported in iodine [1]. By contrast, excitation through the $\nu_1 + \nu_3$ band is largely dominated by nonresonant coupling because of its smaller transition dipole moment, leading to only a minor resonant contribution. Measurements performed at 1.5 µm corroborate the prevalence of nonresonant dynamics. Overall, these results emphasize the key role of transition dipole strength in resonant alignment and demonstrate the broader potential of vibrationally driven alignment for molecular control. Recent exploration of the 3 µm region will be discussed.

1. M. Bournazel, A. Espaignol, D. Singh, R. K. Bhalavi, F. Billard, P. Béjot, E. Hertz, O. Faucher. Persistent Ground-State Planar Alignment of Iodine Molecule through Resonant Excitation. *Physical Review Letters* **2024**, *133* (13)

Interferometric effects in multiple fs/ps rotational coherent Raman scattering

Francesco Vergari[1], *Francesco Mazza*[2], *Ali Hosseinnia*[3], *Michele Marrocco*[4]

1. *Dipartimento di Fisica, Università di Roma “La Sapienza”, Roma I-00185, Italy*
2. *Chair of Optical Diagnostics in Energy, Process and Chemical Engineering, RWTH Aachen University, 52062 Aachen, Germany*
3. *Luxembourg Institute of Science and Technology, L-4422, Belvaux, Luxembourg*
4. *ENEA, via Anguillarese 301, Roma I-00123, Italy*

We use fs/ps rotational coherent Raman scattering as a selective method to detect molecular coherence in gas mixtures through interferometric mixing of scattering events arising from a sequence of pump, control, and probe pulses. Compared to an earlier approach based on ns probing [1, 2], the reduced spectral resolution of this work may spoil the interferometric capability. However, limited temporal resolution becomes a critical constraint when attempting to resolve spatial phase effects reported in a recent study using purely fs probing [3]. To examine these competing factors, we investigate a room-temperature CO_2/N_2 gas mixture. The pulses were generated and arranged as illustrated in the left panel of Fig. 1. The recorded spectra exhibit alternating constructive and destructive interference, clearly revealing the direct impact of temporal phase mixing on the spectral response (right panel of Fig. 1). We further assess the potential emergence of spatial interference patterns under conditions of restricted temporal resolution.

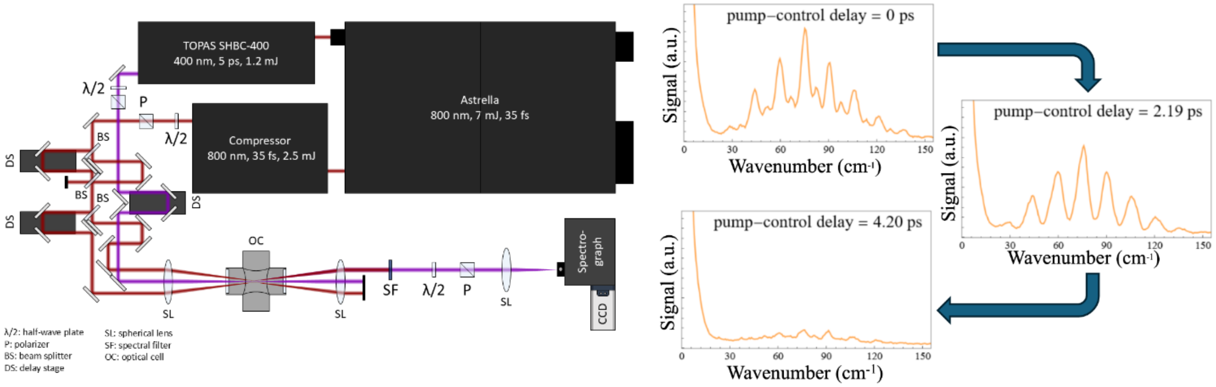

Figure 1. (left) experimental set-up, (right) interferometric effects in N_2 spectra for a probe delay of 4.20 ps (half revival).

2. A. Hosseinnia, M. Raveesh, A. Dominguez, M. Ruchkina, M. Linne, J. Bood, Opt. Express 30 (2022) 32204
3. M. Raveesh, A. Dominguez, M. Ruchkina, M. Linne, J. Bood, A. Hosseinnia, Opt. Express 31 (2023) 38064
4. F. Vergari, F. Mazza, M. Marrocco, A. Hosseinnia, Opt. Lett. 51 (2026) 604

Atomic coherent anti-Stokes Raman spectroscopy in metal vapours

Francesco R. Mazza,[1,2] *Ashutosh Jena*,[1] *Abhishek Kushwaha*,[1] *Ali Hosseinnia*,[3] *and Isaac Boxx*[1]

1. *Chair of Optical Diagnostics in Energy, Process and Chemical Engineering, RWTH Aachen University, Schinkelstraße 8,*
2. *Faculty of Aerospace Engineering, Delft University of Technology, Kluyverweg 1, Delft, 2629 HS, The Netherlands*
3. *Luxembourg Institute of Science and Technology (LIST), 4422 Belvaux, Luxembourg*

The application of coherent Raman anti-Stokes spectroscopy (CARS) to atomic species is largely underexplored. By exploiting the gyroscopic coupling of the spin (S) and orbital (L) angular momenta of the valence electrons of the target atoms [1], it is possible to generate a fine-structure CARS signal in atomic vapours [2].

Here we perform CARS measurements on atomic iron (Fe) in a laser-induced plasma. The ground state Fe atom has four unpaired electrons in the *3d* shell resulting in total spin and orbital angular momenta, $L = 2$ and $S = 2$, respectively. Spin orbit-coupling results in the fine structure shown in Fig. 1(a), with five energy levels with decreasing total angular momentum, $J = L+S, \ldots, |L-S| = 4, \ldots, 0$.

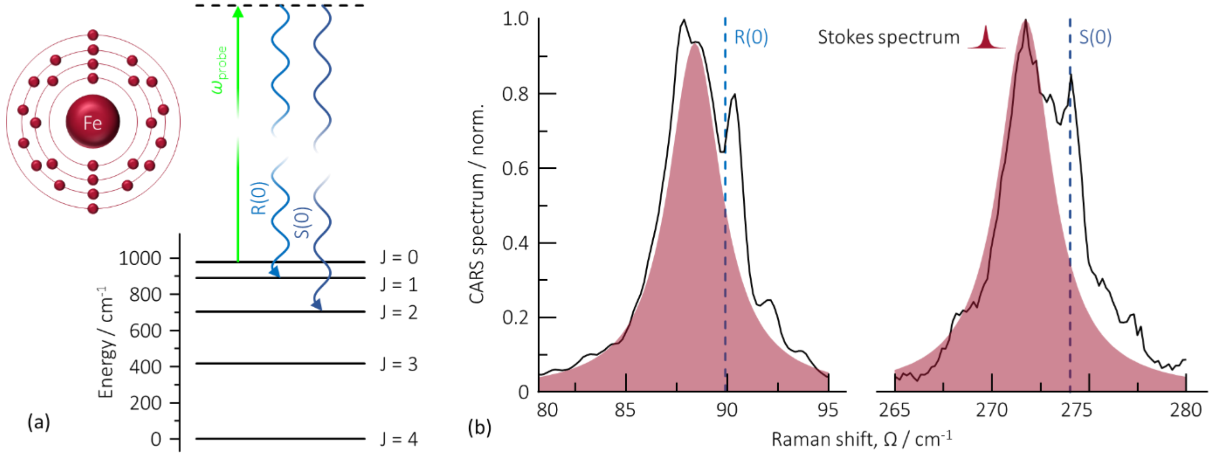

Figure 1. (a) Fine structure of the ground state Fe atom, (b) Fe CARS spectral lines: R(0) at 89.9 cm^{-1} and S(0) at 274.1 cm^{-1}.

The experimental setup comprises an injection-seeded frequency-doubled Nd:YAG laser providing the pump and probe beams and an OPO acting as the relatively broadband (<5 cm^{-1}) Stokes source. The OPO output wavelength is tuned to target different lines in the fine-structure CARS spectrum of Fe, and we assign four lines to the R- and S-branch spectra in Fig. 1(b). We furthermore discuss possible uses of atomic CARS diagnostics to metal chemistry in combustion and plasma applications.

1. C. Penney, J. Opt. Soc. Am. 59 (1969) 34
2. R. E. Teets, J. H. Bechtel, Opt. Lett. 6 (1981) 458

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At the Pulse of Light

Label-free Imaging
CARS, SRS by picoEmerald FT

- Tunable picosecond light source
- 100x faster tuning / 10x faster imaging
- One box, hands-free, shot noise limited
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  Raman tags, Fingerprint region, CHx

Living Drosphila Embryo

Fast, multi-color
chemical imaging

Tunable Light Sources
Ultrafast, UV to MID-IR Systems

- Automated Wavelength Tuning
- UV to MID-IR (190 nm ... >20 µm)
- Femto- & Picosecond
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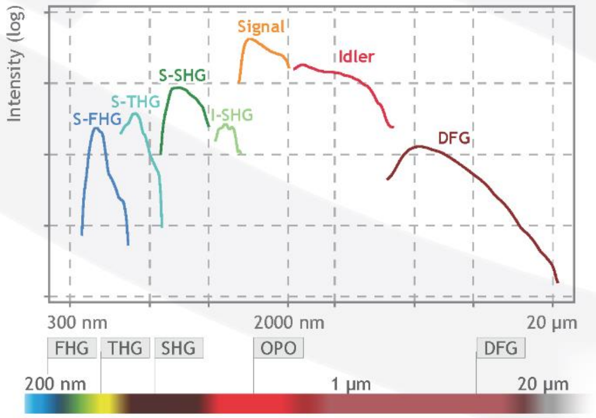
- High Degree of Customization



Laser Diagnostics
for Ultrafast Systems

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- Autocorrelators for 200 nm – 12 µm
- High Contrast Pulse Characterization
- FROG & SPIDER
- Microscopy Autocorrelator
- Spectrometers up to 6.3 µm

Advances in Nonlinear Multimodal Imaging and Hyperspectral Coherent Raman Spectroscopy for Precision Oncology

Michael Schmitt[1], *Juergen Popp*[1,2]

1. *Institute of Physical Chemistry and Abbe Center of Photonics, Friedrich-Schiller-University Jena, Helmholtzweg 4, 07743 Jena, Germany*
2. *Leibniz Institute of Photonic Technology, Member of Leibniz Health Technologies, Albert-Einstein-Straße 9, 07745 Jena, Germany*

Nonlinear optical spectroscopy has emerged as a powerful platform for label-free biomedical imaging by combining molecular specificity with high spatial resolution. In particular, coherent Raman techniques such as coherent anti-Stokes Raman scattering (CARS) provide chemically selective contrast based on vibrational resonances, while two-photon excited fluorescence (TPEF) and second harmonic generation (SHG) complement the information by probing endogenous fluorophores and non-centrosymmetric structures such as collagen.

Recent developments in fiber-based multimodal implementations have enabled the integration of CARS, TPEF, and SHG into compact endomicroscopic platforms, allowing nonlinear spectroscopic imaging directly at the surgical site. By targeting the CH-stretch region, CARS delivers rapid contrast related to lipid and protein distribution, enabling real-time differentiation of tissue states. The combination of these nonlinear contrast mechanisms provides simultaneous morphological and biochemical information at subcellular resolution without exogenous labeling.

Beyond single-resonance imaging, hyperspectral coherent Raman imaging (CRI), including broadband CARS (BCARS) and broadband stimulated Raman scattering (BSRS), acquires full vibrational fingerprints at each pixel. This spectroscopic extension enables multiplexed chemical mapping, digital histopathology, and refined tissue subtyping based on subtle biochemical variations. Advanced data-driven analysis, including deep learning approaches, further enhances the extraction of diagnostically relevant spectral features and supports objective tissue classification.

Together, nonlinear multimodal imaging and hyperspectral coherent Raman spectroscopy demonstrate how vibrational contrast mechanisms, combined with modern photonic instrumentation and computational analysis, open new perspectives for data-driven precision oncology and real-time optical biopsy.

Acknowledgement: Financial support of the EU, the “Thüringer Ministerium für Bildung, Wissenschaft und Kultur”, the “Thüringer Aufbaubank”, the Federal Ministry of Research, Technology and Space Germany (BMFT), the German Science Foundation, and the Carl-Zeiss Foundation are greatly acknowledged.

Mapping In-plane Orientational Order and Correlation Lengths in Molecular Films using Azimuthal-Scanning Vibrational Sum-Frequency Generation Microscopy

Nasim Mirzajani, Ben John, Martin Wolf, Martin Thämer, Alexander Fellows

1. *Fritz Haber Institute of the Max Planck Society, Berlin, Germany*

Self-assembled molecular films are ubiquitous in nature, with highly ordered, anisotropic packing structures which govern their macroscopic properties and functional behavior. Local anisotropy in these systems is frequently accompanied with pronounced heterogeneity and long-range in-plane packing order extending from molecular to microscopic length scales. Experimental access to this structural complexity remains a formidable challenge. Recent advances have established phase-resolved sum-frequency generation (SFG) microscopy as a direct probe of thin-film structures, however, many properties have thus far remained unaddressed, including the width and shape of microscopic orientational distributions. We present a method to overcome this limitation by implementing an azimuthal-scanning approach in SFG microscopy, enabling access to the full in-plane orientational distribution, far beyond average molecular orientation. Specifically, comparison of the complete set of rotational frequencies arising from the azimuthal dependence with simulated data illustrate how these frequencies respond differently to in-plane orientational disorder, deviations from perfect crystallinity, and more complex packing motifs such as bimodal arrangements. We apply this framework to a model membrane consisting of a phase-separated mixed phospholipid monolayer, revealing that molecules within condensed domains exhibit micron-scale orientational correlations despite substantial in-plane orientational diversity, including a finite distribution width and markedly different orientations of the two lipid tail groups. Overall, this example highlights the potential of this method for future investigations into how packing structure governs functional behavior in lipid membranes. Beyond this specific system, the theoretical concepts introduced here are readily extendable to a wide range of systems, from molecular assemblies to phononic materials, and thus open new directions for structural elucidation at interfaces.

1. Mirzajani, N., John, B., Wolf, M. et al. Faraday Discuss., 2026, Accepted Manuscript
2. Fellows, A.P., John, B., Wolf, M. et al. Nat Commun 15, 3161 (2024).
3. Fellows, A.P., John, B., Wolf, M. et al. J. Phys. Chem. Lett. 2024, 15, 43, 10849–10857
4. Khan T, John B, Niemann R, et al. Opt Express. 2023 Aug 28;31(18):28792-28804.

Automated correction of optical attenuation in stimulated Raman scattering imaging

Nick S. Lemberger[1], Kristin Wallmeier[1], Carsten Fallnich[1,2]

1. Institute of Applied Physics, University of Münster, Corrensstraße 2, 48149 Münster, Germany
2. Cells in Motion Interfaculty Centre, University of Münster, Münster, Germany

Stimulated Raman scattering (SRS) is a label-free and chemically-selective microscopy technique. The contrast in SRS is based on probing the imaginary part of the third-order nonlinear susceptibility which is related to molecular vibrational resonances. However, SRS is typically detected as an intensity modulation on either the pump or Stokes beam with a lock-in amplifier, and therefore not distinguishable from signal variations caused by optical loss across the sample. When detecting SRS as stimulated Raman gain (SRG) on the Stokes beam with both the pump and Stokes beam experiencing optical loss, then the SRG signal strength can be expressed as follows: $\mathrm{SRG} \propto \mathrm{Im}(\chi_R^{(3)})\, I_p t_{p,1}\, I_s \mathrm{t}_{s,1} \mathrm{t}_{s,2}$, where $\mathrm{Im}(\chi_R^{(3)})$ denotes the imaginary part of the resonant third-order nonlinear susceptibility, and I_s and I_p the Stokes and pump intensity, respectively. Additionally, $t_{p,1}$ and $t_{s,1}$ denote the transmission factors of the pump and Stokes beam through the microscope up to the focal point in the sample and $t_{s,2}$ the transmission factor of the Stokes beam from the focal point in the sample to the photodetector.

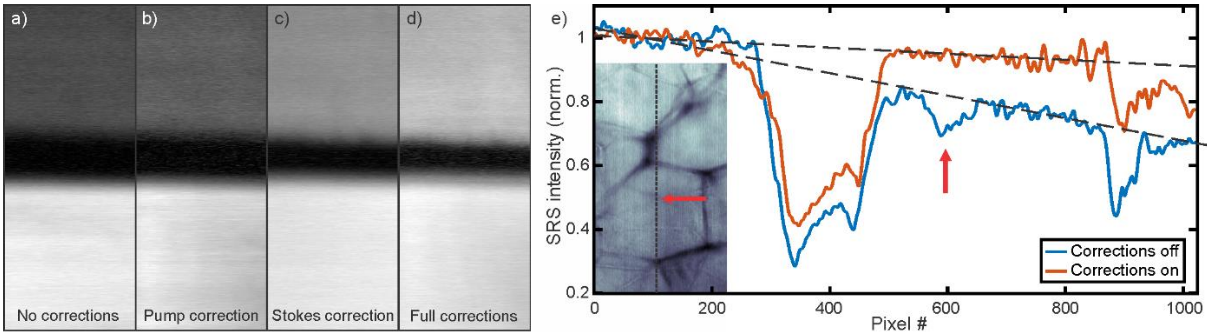


Fig. 1 a-d) SRS images of a homogeneous oil sample at 2850 cm^{-1} with an ND filter partially inserted into the scan plane, imprinting a shadow on image a). The filter is becoming almost invisible when the pump and Stokes corrections are both applied in image d). e) Profile view of an SRS image of sliced chicken fat tissue imaged at 2850 cm^{-1} (insert, cut along dashed line) with corrections turned off (blue line) and on (red line).

In order to overcome the issue of transmission loss artifacts in SRS microscopy and correct for both pump as well as Stokes losses, we implemented a transmission loss correction scheme for SRS microscopy by extending an autobalanced detector [1] with an additional PID loop and a confocal detection setup for pump reflection [2]. When both pump and Stokes corrections were activated, the lossy shadow of the filter on the SRS image vanished, as shown in Fig. 1 a)-d). Additionally, the SRS profile view in Fig. 1 e) of sliced chicken fat tissue imaged at 2850 cm^{-1} (see insert) demonstrated, how a loss artifact (dip in blue line at pixel 600) as well as an image gradient (compare long-dashed lines) could be removed or significantly reduced, respectively, from the expected homogeneous lipid distribution.

1. N. Lemberger, K. Wallmeier, C. Fallnich, Opt. Express 33 (2025) 62
2. N. Lemberger, K. Wallmeier, C. Fallnich, Opt. Express 34 (2026) 5

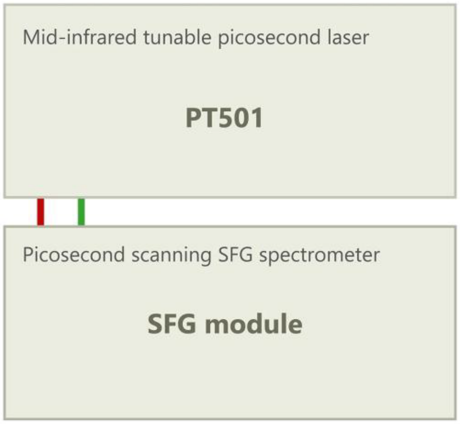
New two boxes SFG spectrometers

Ekspla is a developer and manufacturer of advanced laser systems and spectroscopy equipment. The company's expertise lies in high-resolution laser spectroscopy, where the most suitable radiation bandwidth to meet the resolution requirements for spectroscopy of solids is typically 2–6 cm^{-1}, corresponding to the bandwidth of Ekspla's picosecond tunable laser systems.

The presentation will describe the architecture and applications of Ekspla's broadly tunable laser sources, which cover an exceptionally wide spectral range from the deep ultraviolet (190 nm) to the mid-infrared (18 µm). Several of these laser sources can be optically synchronized with each other or with external lasers using phase-locked loop (PLL) techniques or wavelength shifting in photonic crystal fibers, enabling coherent anti-stokes raman scattering,advanced pump–probe, sum-frequency generation, and other multi-wavelength experiments.

The presentation will also outline Ekspla's portfolio of fixed-wavelength picosecond lasers, mid-IR laser sources, and tunable laser systems designed for both scientific applications. Further topics include laser systems for Scanning Near-field Optical Microscopy (SNOM) and high-intensity laser systems used in ultrafast and nonlinear optics research. Finally, Ekspla's sum-frequency generation (SFG) spectrometers will be presented, providing a complete solution for experiments focused on surface and interface investigations. Ekspla can offer both picosecond and femtosecond SFG spectrometers.

The new two-box spectrometers feature a wider spectral range, better spectral resolution, and the sweep function that make measurements several times faster. Due to compact two-box approach the whole system occupies two times less space in the lab. Air cooled laser system is more user friendly.



Schematic layout of SFG Classic spectrometer.

/ Spectral range 2.3 – 16 µm 625 – 4300 cm^{-1}

/ Repetition rate 100 Hz

/ Spectral resolution <3 cm^{-1}

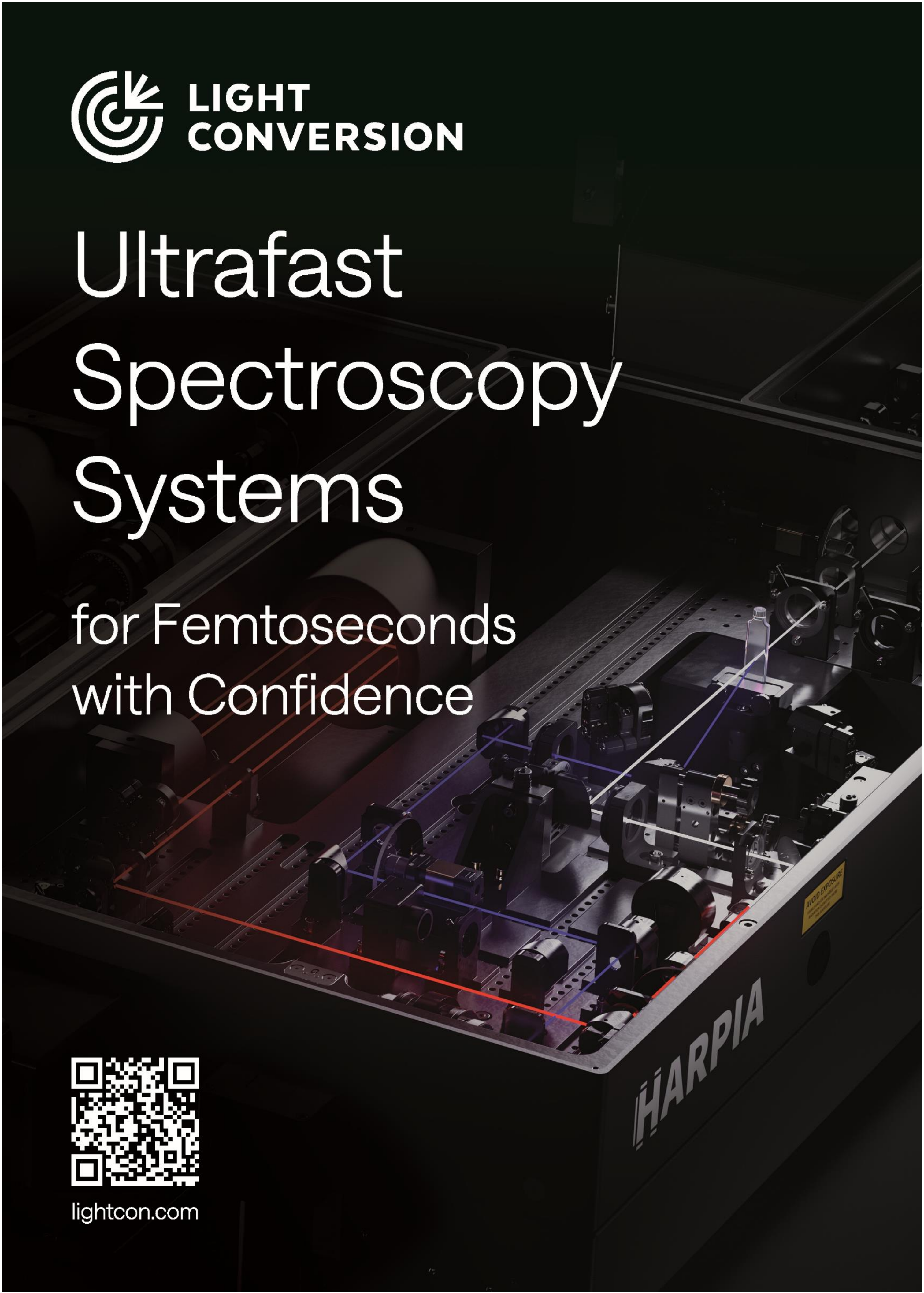
Interpretation of the primary chiral charge separation on the basis of second order spin-orbit-coupling

<u>Sighart F. Fischer</u> and Philipp O.J. Scherer

Department of Physics, Technical University of Munich

We analyze the primary dynamics of the photoinduced charge separation for the reaction centre from Rps.viridis on the fs time scale in order to elucidate the fundamental conditions created by nature leading to a very efficient suppression of a recombination. We propose by means of Time-dependent HF calculations within the Tamm-Dancoff approximation and semi-empirical dynamics (consistent with spectral observations) that the initial relaxation of bond lengths of the special pair in the excited state is driven by a strong electron density shift towards the central region of the dimer. The following dynamics can be characterized as the formation of an internal dimer charge–transfer state with dominant electron density on the dimer part PM. It acts as electron donor-site for the chiral nature of the charge separation with BL as bridging state. The non-exponential dynamics on the longer ps time scale is characterized by the competition between the localization forces and the positive charges on PL versus the chiral transfer to the acceptor HL. The population increase of HL- induces an external force, which helps to optimize the energetics of the BL- state for all component of the broad BL- distribution. The interaction between neighbors can be described as second order Spin-Orbit-Phonon-induced Super-exchange coupling. The dynamics can be algebraically described as the evolution of an initially prepared Gaussian energy distribution with time dependent coupling.

1. Sighart. F. Fischer, Chem. Phys. Letters. 91 (1982) 367
2. Sighart. F. Fischer, H. Köppel, Chem.Phys.59 (1981) 279-287
3. P. O. J. Scherer, S. F. Fischer, Chem. Phys. Letters. (1989) 2345
4. P. O. Scherer, C. Scharnagl, Sighart F. Fischer, Chem. Phys. 179 (1995) 333-341
5. Thomas P. Fay, J. Phys. Chem. Letters. 12. 2021 1407-1412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AVOID EXPOSURE

Application of Femtosecond CARS for Measurements in High-Pressure Reacting Flows

Robert P. Lucht[1]

1. *School of Mechanical Engineering and School of Aeronautics and Astronuatics, Purdue University, West Lafayette, Indiana, USA*

The use of femtosecond CARS for measurements in high-pressure combustion experiments is discussed. We have used 5 kHz, single-laser-shot chirped-probe-pulse (CPP) femtosecond (fs) coherent anti-Stokes Raman scattering (CARS) to perform temperature measurements in a hydrogen/oxygen rocket chamber with a co-annular injector. The measurements were very challenging due to the short time duration, on the order of one second, of the experiment, and the effect of the windows on the femtosecond laser beams. In addition, probing the hydrogen molecule to perform temperature measurements presented issues not encountered previously with nitrogen CPP fs CARS.[1] The incorporation of collisional narrowing effects in time-dependent modeling of femtosecond CARS will also be discussed. Recent experiments on line CARS will be described.

1. Z. Chang, R. M. Gejji, M. Gu, and R. P. Lucht, “H_2 Chirped-Probe-Pulse Femtosecond Coherent Anti-Stokes Raman Scattering Thermometry at High Temperature and Pressure,” *Optics Express* **32**, 13701-13719 (2024). DOI: 10.1364/OE.519992

55 Hz rate fs/ps-CARS spectroscopy for combustion thermometry

Clément Pivard, Rosa Santagata, Michael Scherman

DPHY, ONERA, Université Paris-Saclay, F-91120 Palaiseau, France

Coherent Anti-Stokes Raman Scattering (CARS) is a powerful nonlinear spectroscopic technique for probing in-situ reacting flows at high repetition rate [1-3]. It allows non-intrusive, spatially localized measurements of temperature and chemical composition. We present a compact fs/ps-CARS laser bench system based on a Yb:KGW femtosecond laser and a MOPA architecture [4] which delivers high-energy pulses at 55 kHz repetition rate. The probe pulse temporal profile is optimally shaped, featuring a sharp rise (<1 ps) and long decay (>10 ps). This particular shape ensures efficient interrogation of the Raman coherences with strong rejection of non-resonant background. High speed acquisition of the CARS spectra has been achieved using a scientific CMOS detector. Single-shot 55 kHz CARS spectra of N_2 Q-branch were successively recorded in a laminar CH_4/air flame on a McKenna burner (Fig1a). This achievement represents more than a tenfold improvement in acquisition rate compared to state-of-the-art of fs/ps-CARS thermometry in combustion [2] in continuous-mode operation. The experimental spectra exhibit a sufficient spectral resolution and signal-to-noise ratio to retrieve temperature with high sensitivity, achieving a precision of σ_T = 50 k at <T> = 2000 K. Continuous temperature monitoring was performed at various locations of the flame (Fig. 1b). It enabled the assessment of the measurement precision in stable combustion zones and the tracking of temperature variations over a dynamic range exceeding 500 K in regions exhibiting unsteady behavior. This demonstrates the system's ability to resolve transient phenomena and capture high-frequency instabilities (Fig 1.c). This capability opens new opportunities to study rapidly evolving processes, instabilities and turbulence in reacting flows over a large analysis bandwidth.

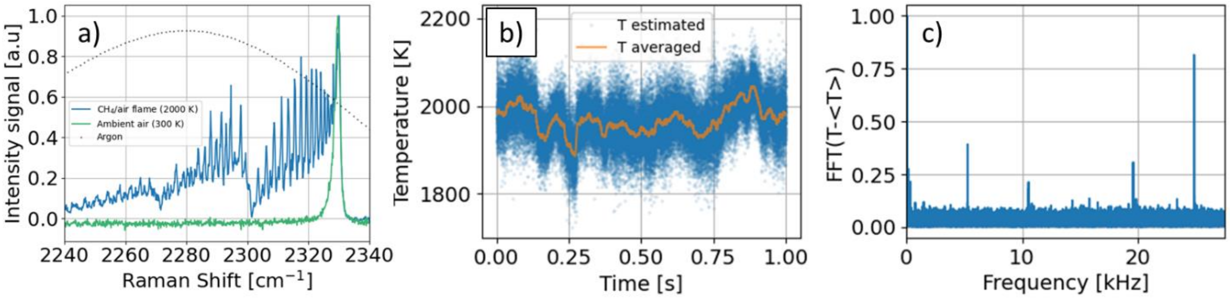

Figure 1.a) Averaged CARS spectra in flame at 2000 K and in ambient air at 300K. b) Temperature monitoring performed at 55 kHz inside the flame during 1 second 1c) Fourier transform analysis of temperature monitoring over 1 second.

[1] A. K. Patnaik, I. Adamovich, J. R. Gord, and S. Roy, Plasma Sources Sci. Technol. **26** (2017) 103001. A.

[2] J. Barros, M. Scherman, E. Lin, N. Fdida, R. Santagata, B. Attal-Tretout, and A. Bresson Opt. Express **28** (2020) 34656.

[3] Pivard, C., Scherman, M., Santagata, R., Pilla, G., Pelletier, G., & Le Pichon, T. (2025). High-speed fs/ps-CARS thermometry for supersonic H2/air combustion studies. Experiments in Fluids, 66(4), 79

[4] R. Santagata, , M. Scherman, M. Toubeix, M. Nafa, B. Attal-Tretout, and A. Bresson Opt. Express **27** (2019) 32924.

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High-speed fs/ps-CARS of H_2 for energetics and propulsion studies.

Michael Scherman[1], *Laila Dakroub*[1,3], *Nelly Dorval*[1], *Denis Packan*[1], *Clément Pivard*[1], *Rosa Santagata*[1], *Brigitte Tretout*[1], *Renaud Jalain*[2], *Leopold Dru*[2], *Lucien Vingert*[2], *Yves Mauriot*[2], *Sean McGuire*[3]

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Hydrogen combustion is encountered in both power generation and propulsion applications, for instance as a fuel for sustainable aviation or space rockets (e.g. Ariane6 H_2/O_2 engine), or as a reaction product in CH_4/O_2 rocket combustion. It is therefore a valuable tracer for probing complex physics in many reacting flows. In this work, femtosecond/picosecond coherent anti-Stokes Raman scattering (fs/ps-CARS) is employed to measure H_2 temperature [1] and concentration in two different experimental configurations.

The CARS system [2] is first applied to probe methane plasmalysis, an alternative hydrogen production pathway that avoids CO_2 emissions by converting methane into hydrogen and solid carbon. High-speed (1 kHz) single-shot measurements of H_2 temperature and concentration are demonstrated in a low-pressure CH_4 DC glow discharge [3]. Time-resolved measurements during plasma ignition, steady-state operation, and extinction phases provide insight into the dynamics of methane conversion.

Second, the technique is used to characterize an over-expanded supersonic jet at the exit of a CH_4/O_2 rocket combustor (ONERA MASCOTTE test bench). 1 kHz CARS spectra of hydrogen produced in the flame are acquired in the post-combustion region and temperature profiles are retrieved by scanning the measurement volume during burst operation.

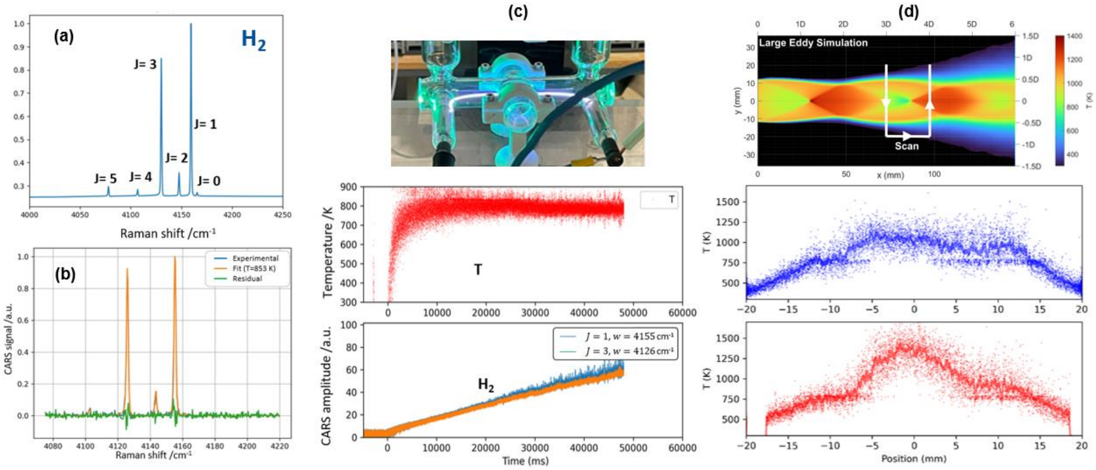

Figure 1. (a) Typical H_2 CARS spectrum and (b) best fit of single-shot spectrum. (c) Monitoring of H_2 in a CH_4 Glow discharge, and (d) at the exit of a supersonic rocket combustor.

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Rotational coherent anti-Stokes Raman scattering evaluation of collisional dephasing in nitrogen ammonia gas mixtures

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To reduce CO_2 emissions and facilitate technically simple transport of hydrogen, ammonia is considered both as an easily liquified hydrogen carrier, but also as a zero-carbon fuel for retrofitting existing combustion processes [1]. To optimize ammonia related technical processes, optical diagnostics and particularly rotational coherent anti-Stokes Raman scattering (RCARS) play a key role by non-invasively measuring space and time resolved temperature and concentration. However, the different chemical situation requires adaptation of RCARS to provide reliable data. As was recently shown, accurate RCARS diagnostics are particularly sensitive to inclusion of accurate S-branch Raman linewidths considering the correct spectral branch, temperature, pressure and collider environment [2]. In this work, we study the collision induced signal decay in ammonia-nitrogen gas mixtures with time-resolved RCARS under defined temperature and mixture composition to obtain the S-branch Raman linewidths. The collisional decay of nitrogen (Figure 1) and resultingly its Raman linewidths show to be significantly influenced by ammonia collisions, which apparently speed up collisional dephasing compared to N_2-N_2 collisions. This is expected to translate into diagnostic errors if not accurately accounted for. These errors in thermometry and species concentration measurements can be avoided if our newly obtained linewidth data are used for evaluation.

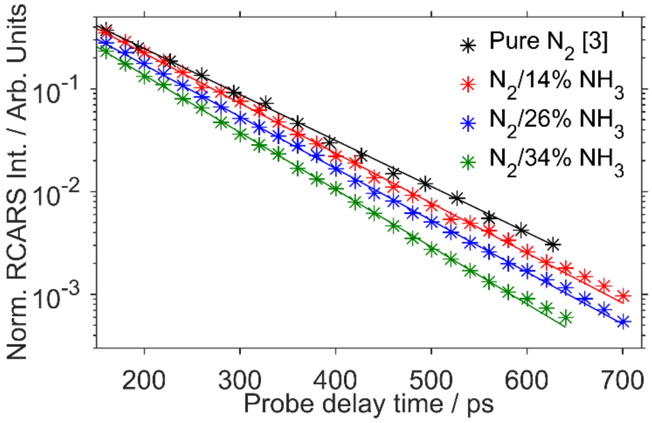

Figure 2: Normalized RCARS intensity of the nitrogen J''=18 line in the pure gas (by Meißner et al. [3]) and different N_2-NH_3 gas mixtures at 500 K and 1 bar with fits of a monoexponential decay functions.

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Femtosecond, Two-Color, Two-Photon, Polarization Spectroscopy for Quenching-Free Detection of Atomic Hydrogen

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Two-photon laser-induced fluorescence (TPLIF) detection of highly reactive atomic species in flames and plasmas is challenging due to, i) low signals resulting from weak two-photon cross sections, ii) laser photolytic interferences, iii) collisional quenching, iv) strong background emissions, and v) the necessity of deep ultraviolet (DUV) excitation laser beams that are hard on optics and can also be readily absorbed by some surrounding species. Challenges i & ii have been addressed by employing high-peak-power, low-energy ultrashort femtosecond (fs) laser pulses for TPLIF detection of atomic species such as H and O. In all fluorescence experiments, quantitative species detection is challenging due to unknown quenching environments and cross-sections. Recently, our group has developed fs two-color, two-photon polarization spectroscopy (fs-TCPS), which addresses the challenges iii–v, along with i &ii, and demonstrates its application to H detection in flames. By using fs pulses, excitation and detection occur on time scales significantly faster than typical species collision timescales in gas-phase media; hence, no quenching corrections are needed. Unlike weak, incoherent fluorescence signals, fs-TCPS produces a coherent signal along the probe beam path.

For H-TCPS experiments, both pump and probe beams were generated from the same laser source. First, the excitation wavelength, laser energy, and polarization dependence of the 243-nm pump beam were characterized. The 243.0-nm pump beam excites the n=1-->-->n=2 two-photon transition, followed by probing the population anisotropy in the excited state via n=2-->n=4 single-photon transition at 486 nm. A quadratic dependence of the pump energy was observed below ~7 µJ/pulse, and the maximum TCPS signal was achieved at linear vertical polarization. The probe beam was vertically polarized. The accuracy of H-TCPS measurements was first confirmed by comparing with predicted H-atom concentrations in a range of CH_4-air and H_2-air flames. The quenching-independent nature of fs TCPS was tested in two different CH_4-O_2 flames diluted with either Ar or He. Both flames have the same predicted H concentrations, but the Ar-diluted flames have a ~14.5% higher total quenching rate. Comparison of simultaneously recorded TCPS and two-color TPLIF signals revealed nearly identical H-TCPS profiles in both flames, but systematic differences in TPLIF profiles. In all experiments, the coherent probe signal was detected ~2 m away from the flame, but could be extended further, as needed, for highly luminous flames, plasmas, or practical combustors with limited optical access. Red-shifting the excitation wavelength away from the DUV also enabled measuring H in ammonia flames, for example.

Overall, fs-TCPS scheme represents a major advancement in collision-independent, coherent detection of atomic species concentrations in flames and plasmas.

Rotational-CARS thermometry of ethylene relying on accurate linewidth calculations

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Accurate modeling of collisional linewidths is a key requirement for Raman-based thermometry of complex molecules such as ethylene, which plays an important role in both combustion and atmospheric chemistry. As an asymmetric-top molecule with small values of rotational constants, ethylene exhibits very dense Raman spectra strongly affected by collisional broadening and line-mixing effects, making reliable temperature retrieval particularly challenging.

In this work, we present an improved linewidth model for Raman spectra of pure C_2H_4, building upon a previous Coherent Anti-Stokes Raman Spectroscopy (CARS) study [1]. In that work, isolated-line widths, assumed to be transition-independent, were found to increase with temperature, which does not follow the well-established experimental decrease [2-4]. To improve physical consistency, transition-dependent collisional half-widths and their temperature-dependence exponents have been computed using a semi-empirical approach and validated against infrared absorption data [5] and incorporated in the linewidth model, with the subsequent empirical adjustment of the interference contributions. The refined linewidth formulation incorporated into the existing CARS spectral model is validated on experimental spectra over a wide temperature range 293–804 K. The new approach yields reduced residuals compared to previous modeling (Figure) and restores physically meaningful trends with isolated-line widths decreasing with both rotational quantum number and temperature.

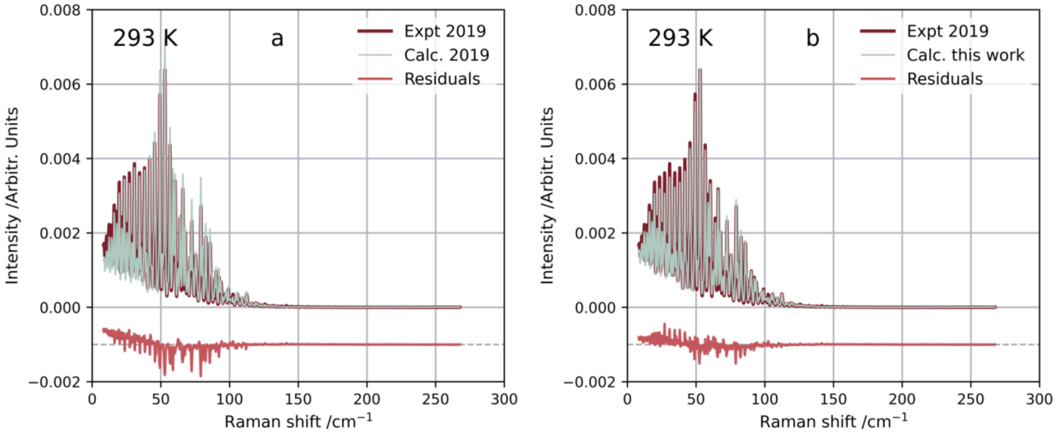

Figure. Comparison of theoretical and experimental [1] rotational CARS spectra of ethylene at room temperature.

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Imaging Structural Anisotropy and Heterogeneity in Molecular Films with Sum-Frequency Microscopy

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Molecular organisation is one of the central factors governing the macroscopic behaviour and functionality at interfaces, with important examples ranging from systems in atmospheric and ocean science to physiological assemblies. Knowledge of the specific arrangement of molecules within such self-assembled films is thus crucial for our understanding of the origins of their physicochemical properties. Yet, experimentally accessing information on the local orientation, conformation, and specific packing structures at molecular interfaces, as well as how this is heterogeneously expressed, is a substantial experimental challenge.

In this work, we introduce vibrational sum-frequency generation (SFG) microscopy as a technique for precisely characterising the molecular-to-macroscopic structural details in anisotropic molecular assemblies. Due to its selection rules, SFG is highly sensitive to the absolute orientation of molecules as well as their degree of orientational order, making it uniquely suitable for characterising the packing structure in these systems. Through a revised imaging geometry and detection system, we demonstrate on mixed phospholipid monolayers a substantially improved signal-to-noise ratio, now achieving the sub-monolayer sensitivity at the sub-micron length-scale that is required for obtaining structural insights. [1-2] Thereafter, through polarisation- and azimuthal-dependent analysis of the hyperspectral images, we extract detailed information on the molecular structure within the phase-separated lipid monolayers, yielding images of the density and composition, molecular tilt angles, and the directionality and distribution within their chiral in-plane packing structure.[3-5] Overall, with such detailed insight into the molecular-level structure, SFG microscopy is poised to make significant breakthroughs in our understanding into the interfacial assemblies that are crucial across a wide range of systems.

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Advanced super-resolution optical microscopy for cellular research

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The understanding of cellular structures and molecular organization and interactions dynamics therein are key to the understanding and treatment of diseases. However, the direct and non-invasive observation of such events in the living cell is often impeded by principle limitations of conventional optical microscopy, for example with respect to limited spatio-temporal resolution. A remedy is fluorescence-based super-resolution microscopy (1-3], for which the Nobel Prize in Chemistry was awarded in 2014. Here, we present the principles and recent advances of such tools such as super-resolution STED or MINFLUX microscopy. We highlight how these approaches can reveal novel aspects of molecular organization and interaction dynamics in living cells and exemplify it with some examples like virus infection.

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Photothermal stimulated Raman scattering with two beams only

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Photothermal stimulated Raman scattering (PT-SRS) [1] is an advanced development that combines the vibrational excitation of stimulated Raman scattering (SRS) with photothermal microscopy [2]. During SRS excitation, heat is accumulated within the sample, which results in a local refractive index change, acting as a thermal lens [3]. This lens effect can be measured via a change of divergence of a probe beam, e.g., by an aperture behind the sample that transfers the change in beam divergence into an intensity change. We present PT-SRS without an additional probe beam by exploiting the refractive index change experienced by the Stokes beam itself, avoiding setup complexity and increased power on the sample.

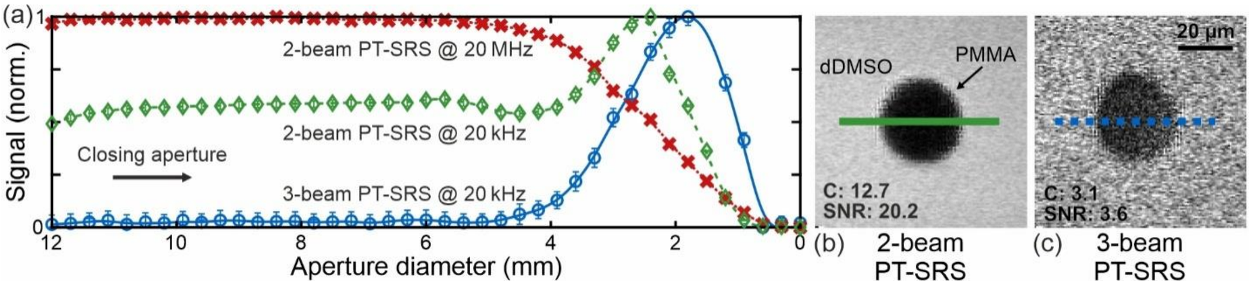

Fig. 1: (a) Comparison of conventional SRS and PT-SRS with (3-beam) and without (2-beam) additional probe beam. (b), (c) Microscope images of a poly(methyl methacrylate) (PMMA) bead in deuterated dimethyl sulfoxide (dDMSO) for different PT-SRS modalities.

In order to verify the presence of a PT-SRS signal in addition to the SRS signal on the Stokes beam, the influence of decreasing diameter of the iris aperture on the signal was measured with a lock-in amplifier (Fig. 1). Conventional SRS (red curve) showed a signal for an opened aperture and with closing the aperture only a decrease in signal could be observed, demonstrating the absence of a photothermal contribution due to the high modulation frequency. In contrast, for low modulation frequency the signal increased in magnitude (green curve) when the aperture size was reduced, indicating the additional signal contribution from PT-SRS on top of the SRS signal on the Stokes beam. For comparison, the PT-SRS signal with additional probe beam was detected and occurred for a decreased aperture size only (blue curve). Microscope images of a PMMA bead in dDMSO showed improved contrast (C) and signal-to-noise ratio (SNR) for PT-SRS without probe beam by a factor of 4.1 and 5.6, respectively, demonstrating the superiority of the presented simplified approach. Furthermore, specific experimental conditions including various detection schemes and power levels could be identified that are advantageous for either conventional SRS or PT-SRS.

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Ultrabroadband CARS Detection of Solvation-Induced Vibrational Changes in Liquid Mixtures

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Ultrabroadband coherent anti-Stokes Raman scattering (BCARS) is a versatile tool used e. g. in combustion and biomedical research, providing high signal strength for the analysis of complex chemical systems on short time scales. Here, we employ a time-delay BCARS scheme based on a custom ORPHEUS OPCPA (Light Conversion, Lithuania) to demonstrate fast diagnostics of liquid mixtures and chemical interactions. The setup uses a broadband pump/Stokes pulse with maximum energy of 100 µJ centered at 800 nm, and a delayed narrowband probe at 513 nm, covering Raman shifts of 500 - 5000 cm^{-1}. Adjustable repetition rates between 1 Hz and 100 kHz allow flexible operation from single-shot measurements to rapid signal averaging.

We investigate dilution series of dimethyl sulfoxide (DMSO) in water and mixtures of DMSO with formic acid. Raman signatures of DMSO remain clearly identifiable down to concentrations of 1 %. In mixtures with formic acid, characteristic peaks of both components are observed, while the DMSO S=O stretch near 1040 cm^{-1} is strongly suppressed, indicating interaction between the two molecules. Independent Raman microscope measurements confirm these observations.

These results illustrate how ultrabroadband BCARS can provide rapid chemical diagnostics by monitoring intensity changes and spectral redistribution of vibrational modes. The approach enables fast detection of concentration changes and intermolecular interactions in liquid systems, highlighting the potential of high-repetition-rate BCARS for time-resolved chemical analysis and process monitoring.

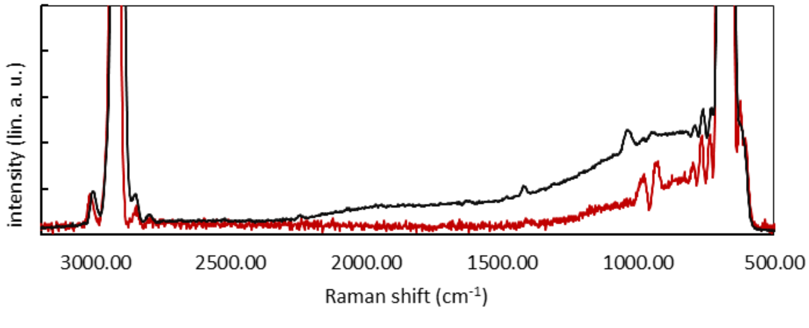

Figure 1: CARS spectra of pure DMSO (black line) and 40% DMSO mixed with 60% formic acid (red line). The significant peak at 1040 cm^{-1} is strongly suppressed due to the solvation reaction.

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The shaping of aeroengines with diagnostics

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In this talk, I will present the objectives of our diagnostics [1,2] to perform quantitative measurements in fluids and solids that are key to advancing the performance of aeroengines. We use standard metrology solutions following established protocols as well as innovative methods and tools to be used in scenarios where standardized equipment fails to deliver the desired reliability in results. Our team's ambition is to support the development of propulsion and power systems by providing information on important mechanisms and parameters operating in harsh environments and industrial conditions. At one of our measurement problems, we also find that particularly non-linear diagnostics deliver the data we need. [3]

1. Synchrotron X-rays (e.g. P21 Swedish Material Science beamline, Petra III, DESY Hamburg) for determining surface integrity and bulk properties during machining of difficult-to-cut materials.
2. Laser ultrasonics for quality assurance in additive manufacturing of aeroengine components.
3. Non-linear optical spectroscopy (coherent anti-Stokes Raman scattering) for spatially resolved measurements of temperature and species concentration in plumes of aeroengines.

In-situ analysis of thermal decomposition of calcium oxalate by simultaneously applying Thermogravimetric Analysis and Ultrabroadband Coherent Anti-Stokes Raman Spectroscopy

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Coherent Anti-Stokes Raman Spectroscopy (CARS) is a well-established technique in combustion diagnostics, providing insights into molecular dynamics. Simultaneously, Thermogravimetric Analysis (TGA) is employed to monitor minor mass changes in solid feedstock materials. While both techniques are well established individually, their combination can greatly enhance the understanding of ongoing chemical reactions involving solid feedstock materials in gaseous atmospheres [1].

In the present study, we establish a new platform by combining ultrabroadband CARS and TGA. A custom-built, dual output OPCPA system delivers near transform limited pulses of ~7 fs, spanning a spectral range ≥5000 cm^{-1}, acting as the pump/Stokes pulse. A ~0.5 ps, narrow bandwidth pulse at 514 nm acts as a probe, delayed by 3.2 ps to suppress the non-resonant background. Both pulses are collinearly focused into a custom-built gas reactor, equipped with optical ports as well as TGA. The reactor is capable of achieving environmental conditions up to 30 bar and 800 °C. To show the capabilities of the new platform, the well-known three step process of thermal decomposition of calcium oxalate monohydrate [2] is examined. 250 mg of $CaCo_2O_4 \cdot H_2O$ are heated to 800 °C at a rate of 5 K/min under constant N_2 gas flow. The respective mass change, measured by TGA, is shown in Figure 1(a). Simultaneously, the probe region of the CARS setup is located in close proximity to the sample material, to detect the decomposition gasses, shown in Figure 1(b).

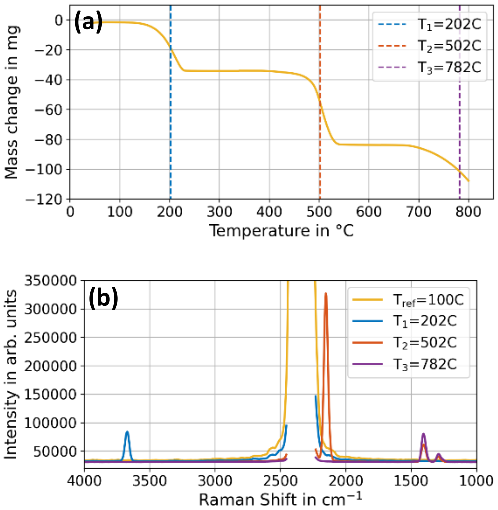

Figure 3: (a) TGA measurement of sample mass change of $CaCo_2O_4 \cdot H_2O$. (b) CARS signal above the sample at different Temperatures. The central N2 signal was blocked for T=202 °C, T=502 °C, T=782 °C for better visibility of the detected reaction gasses.

Our simple example measurement shows the capabilities of this new platform. Adapting it to the investigation and improvement of combustion processes or catalytically enhanced gas conversions offers multiple fruitful applications.

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Sub-Cycle Response of Indium Tin Oxide film at its Epsilon-Near-Zero Region by Fieldoscopy

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Epsilon-near-zero (ENZ) media exhibit exceptionally strong optical nonlinearities, enabling ultrafast modulation for applications including photonic switching, temporal interfaces, and energy-transfer schemes. [1] Refractive-index changes induced under intense excitation at ENZ wavelengths enable rapid tuning of transmittance, reflectance, and nonlinear frequency conversion. [2] Characterizing the rise time of the transient nonlinear response is key to high-speed optical modulation and to understanding the underlying physics. We excite a 400 nm indium tin oxide (ITO) film deposited on a 250 µm SiO_2 substrate at its epsilon-near-zero (ENZ) frequency using a focused 14 fs laser pulse with variable intensity. Employing fieldoscopy [3], we resolve the electric field of the transmitted pulse with sub-100-attosecond temporal precision (Fig. 1a), enabling direct access to the ultrafast response of the ITO film. With increasing intensity, we observe a pronounced spectral redshift and a corresponding shift of the transmitted pulse's center-of-mass frequency (Fig. 1b, c). By temporally referencing the local extrema and zero crossings of the main field oscillations to the low-intensity response at 51 GW/cm² (Fig. 1d), we uncover a sub-cycle, intensity-dependent rise time in the nonlinear response of the ITO film (Fig. 1e).

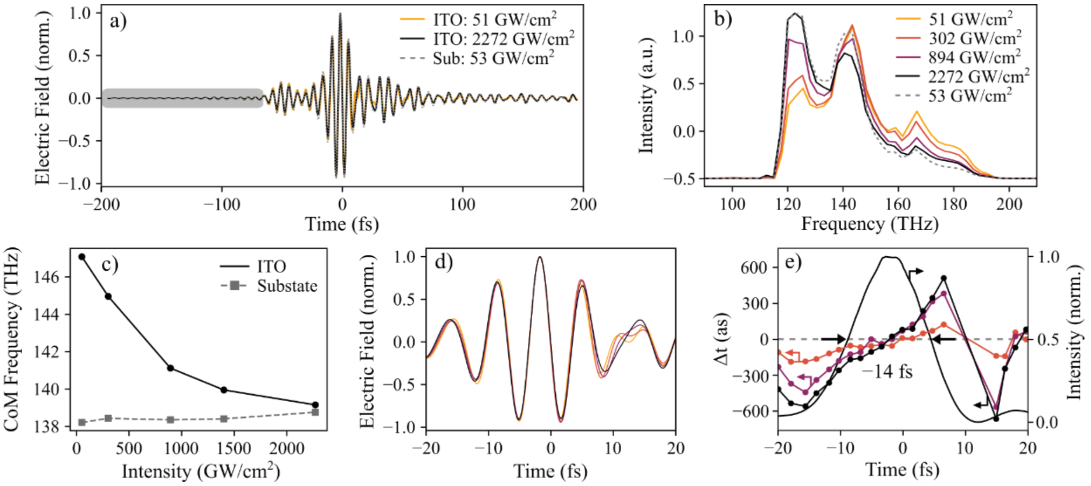

Fig. 1. (a) Transmitted electric field through the ITO film and the bare substrate, shown by the solid and dotted lines, respectively. (b) Transmitted intensity spectra. (c) Center-of-mass (CoM) frequency of the transmitted spectra. (d) Main-pulse electric field. (e) Sub-cycle temporal shifts Δt of field extrema and zero crossings referenced to 51 GW/cm².

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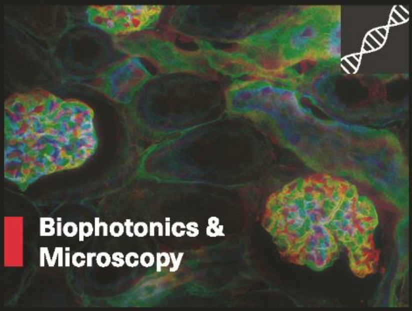
Biophotonics &
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Poster Abstracts

Femtosecond/Picosecond CARS of Gas-Phase Hexamethyldisiloxane (MM): Coherence Dynamics in a Heavy Non-Ideal Vapor

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Heavy organosilicon vapors such as hexamethyldisiloxane (HMDSO, commonly denoted MM) are increasingly employed in studies of non-ideal compressible flows and vapor-phase material processing. In these regimes, classical ideal-gas assumptions break down and key thermodynamic and optical properties, including temperature-dependent refractive index, and molecular concentration, remain insufficiently characterized. Recent observations of rarefaction shock waves in dense vapors further highlight the nonclassical gas-dynamic behavior of such fluids [1], for which validated non-ideal models are still lacking. The absence of quantitative laser diagnostics for such heavy vapors therefore limits experimental validation of non-ideal gas dynamic models.

In this work, we investigate the third-order nonlinear vibrational response of gas-phase MM at 450 K and 0.4 bar using femtosecond/picosecond coherent anti-Stokes Raman scattering (CARS). A temperature- and pressure-controlled gas cell was developed to ensure stable vapor phase operation of MM. The 35 fs output of a Ti:Sapphire amplifier at 800 nm was employed directly as broadband pump/Stokes excitation, providing sufficient spectral bandwidth to coherently excite the symmetric Si–O–Si bending mode at 521 cm^{-1} and the SiC_3-related band at 664 cm^{-1}. A narrowband picosecond probe enabled delay-resolved detection of the $\chi(3)$ nonlinear response.

Time-delay measurements reveal rapid vibrational dephasing with characteristic decay times of approximately 7 ps. Notably, this decay is comparable to that of the non-resonant four-wave mixing background, indicating that the resonant vibrational polarization does not significantly outlive the electronic contribution. This behavior contrasts with the longer-lived coherences typically observed in small diatomic molecules and reflects the dense vibrational manifold and strong intramolecular coupling characteristic of heavy polyatomic systems. These results extend time-domain coherent Raman spectroscopy to complex heavy vapors under non-ideal thermodynamic conditions and represent a first step toward quantitative optical diagnostics of temperature and concentration in non-ideal gas flows, with implications for determining refractive index and acoustic properties in such media.

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Combining spectroscopies for theoretical modelling of C_2H_4 lineshape parameters and line-mixing effects in rotational Raman bands

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Raman-based non-intrusive thermometry of molecular gases relies strongly on lineshape parameters of spectroscopically active transitions. The case of ethylene is particularly challenging since this asymmetric-top molecule with small rotational constants produces spectra with very dense rotational structure even at sub-atmospheric pressures. Closely spaced lines make lineshape measurements extremely difficult, so that a limited number of C_2H_4 self-broadening coefficients are reported in the literature and are largely limited to infrared absorption.

We present a semi-empirical theoretical approach based on extensive use of infrared-absorption linewidths [1,2] to determine and validate a set of model parameters [3]. Further, this set is employed for calculating linewidths of rotational Raman transitions involved in rotational CARS thermometry [4], leading to improved agreement between experimental and theoretical spectra in a wide temperature range 293–804 K. The non-negligible line-mixing effects are modelled purely empirically at this step. To move toward a relaxation-matrix description of ethylene Raman spectra with line interferences, an Energy-Corrected Sudden approach to building the full (essentially non-diagonal) relaxation matrix is outlined. The determination of the ECS-parameters set is traditionally based on the linewidths of transitions in the isotropic Raman Q-branch (unavailable for C_2H_4), so that a third series of linewidth calculations becomes necessary. In this way, we highlight the connections between infrared absorption (tensorial rank $p = 1$), anisotropic Raman scattering ($p = 2$), and isotropic Raman scattering ($p = 0$). We also demonstrate the importance of combining these approaches to achieve rotational Raman spectra modelling from limited linewidth data measured in infrared.

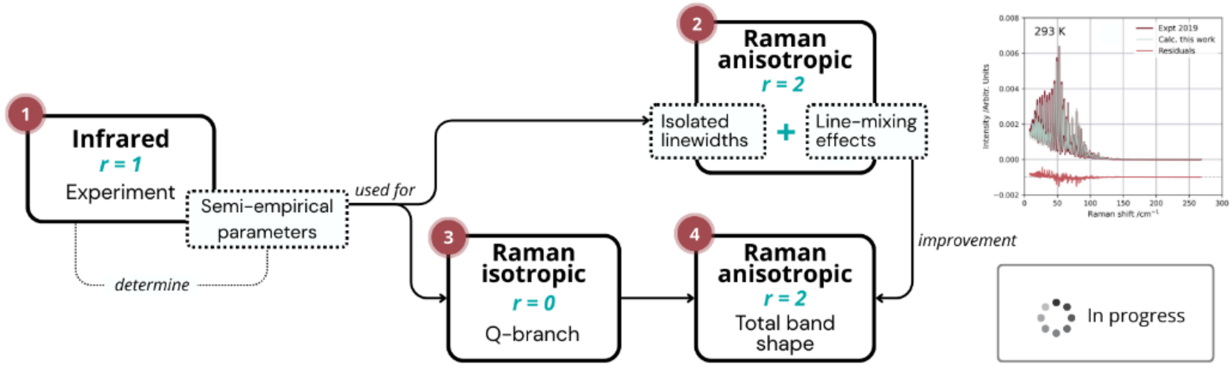

Figure. Scheme of connections between different spectroscopies required for accurate modelling of C_2H_4 Raman spectra.

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Experimental Determination of the Collisional Broadening Effect of NH_3 on the S-Branch Raman Linewidth of N_2

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Ammonia (NH_3) is a promising carbon-free combustion fuel that can replace hydrocarbons to reduce CO_2 emissions. However, optimization of NH_3 combustion is required to improve energy conversion efficiency and suppress NO_x formation[1]. Accurate determination of combustion parameters using rotational coherent anti-Stokes Raman scattering (RCARS) spectral models require precisely determined Raman linewidth[2]. At present, collisional broadening parameters for NH_3-containing mixtures are not available to our knowledge. In this work, we experimentally investigate collisional broadening of N_2 S-branch Raman linewidths by NH_3 in N_2/NH_3 binary mixtures with NH_3 concentrations up to ~50%, at atmospheric pressure and temperatures ranging from 295 K to 870 K. Although the NH_3 rotational CARS signal is too weak to permit direct linewidth determination using time-domain RCARS, its effect on N_2 linewidths is substantial. A pronounced increase in magnitude of N_2 linewidths with increasing NH_3 fraction is observed, indicating accelerated coherence decay and more efficient energy transfer in N_2–NH_3 collisions compared to N_2–N_2 collisions. Therefore, incorporating these NH_3-induced broadening effects into refined spectroscopic models is expected to significantly improve the accuracy of thermometry and species concentration measurements in gas-phase combustion diagnostics.

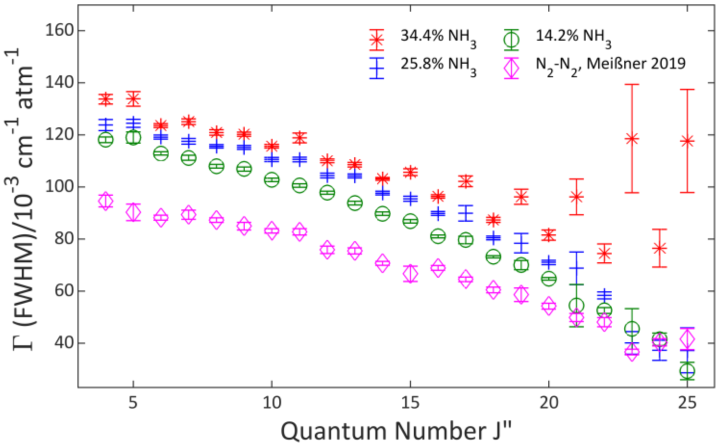

Figure 4: S-branch rotational Raman linewidths of N_2 in N_2–NH_3 binary mixtures at 295 K for the selected mixture compositions, compared with the pure N_2 S-branch Raman linewidths from Meißner et al. [3].

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Non-resonant surface enhanced hyper Raman scattering (SEHRS) of aromatic amino acids with gold and silver nanostructures

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Among the nonlinear vibrational spectroscopy techniques, hyper Raman scattering (HRS) is relatively less popular due to its inherent weakness. However, because of the nonlinearity, the confined local optical fields in plasmonic structures are more beneficial for enhancing HRS compared to its one-photon counterpart, spontaneous Raman scattering. SEHRS can thus give access to complementary vibrational information that is inaccessible with surface enhanced Raman scattering (SERS). Specifically, SEHRS has been shown to be more sensitive towards surface interactions and adsorbate orientation, making it an excellent approach to study metal-protein systems. Nevertheless, finding a plasmonic metal substrate suitable for SEHRS is challenging because of the large gap between excitation and detection wavelengths and the strong influence of metal-molecule interaction in the process.

In this work, we present non-resonant SEHRS spectra of aromatic amino acids excited at 1064 nm, obtained with spherical gold nanoparticles of different sizes as plasmonic substrate. We compare them to their SERS spectra also excited off-resonance at 633 nm. We observed spectral differences regarding the relative signal intensities and particular vibrational modes arising from different selection rules, as well as from the functional groups specific to the different amino acids. [1] We also report SEHRS from silver substrates and compare them with the data obtained using gold nanoparticles. The data were found to be in good agreement with previous work that used silver nanostructures for SEHRS [2], but they also indicate that the spectra are sensitive regarding the specific molecule-nanostructure interactions. The complete vibrational (SEHRS+SERS) profile of the amino acids obtained in this work will enable us to study supramolecular structures and proteins in complex bioenvironments.

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Femtosecond 2-photon-induced sideways lasing

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There is a strong need for sensitive methods capable of measurements at remote locations, i.e., remote sensing techniques. Optical methods producing highly directional signals are expected to have a significant impact on both fundamental and applied research, including industrial applications. It has been shown that constituents of air can act as an active laser medium, generating a laser-like beam without cavity mirrors from a remote location in the atmosphere. Despite extensive research on cavity-free lasing concepts, the underlying physical mechanisms are still not fully understood, particularly under conditions relevant for practical applications. Over the past decade, our research group has explored lasing based on femtosecond (fs) multiphoton excitation of atoms in flames, with emphasis on forward and backward lasing [1], but also an initial investigation of the feasibility of generating sideways lasing [2]. The inset of Fig. 1 illustrates laser-optical configurations for forward/backward lasing (a) and sideways lasing (b).

In the present work, sideways lasing experiments are performed in a Xe cell, where 30-fs laser pulses at 225.5 nm two-photon excite Xe atoms, producing lasing at 869.2 nm. Two counter-propagating fs laser sheets create an ultranarrow (~10 µm) gain volume at their temporal and spatial overlap. The fluorescence image in Fig. 1 (Top View) shows this narrow gain region, while the Side View image, recorded with a streak camera in focus mode (no streaking), captures the emission from the side. The right panel shows a highly time-resolved streak camera image of the sideways emission. A weak fraction of the excitation pulse was detected simultaneously (Excitation). The ~70 ps delay between excitation and emission and the short emission duration (~20 ps) strongly suggest that the emission originates from superfluorescence, for which a certain time is needed to build up the macroscopic atomic coherence, resulting in a very short emission pulse.

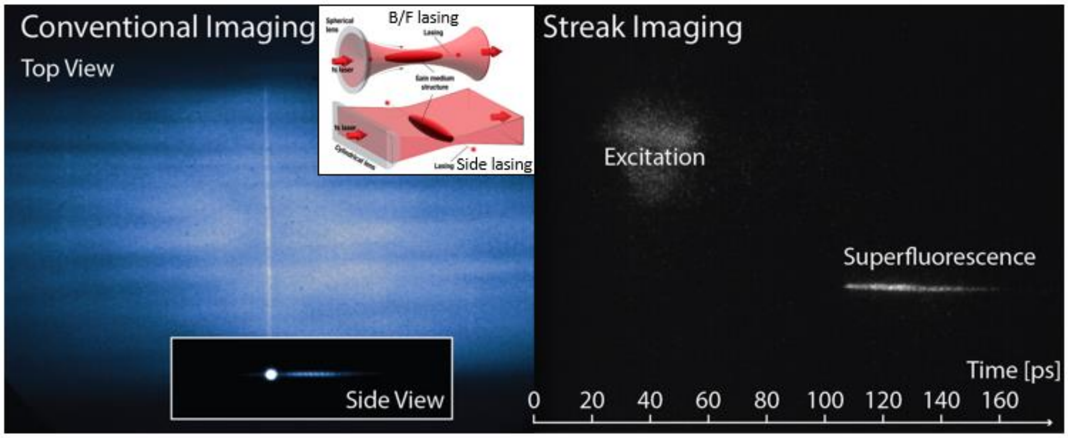

Fig. 1 *Left: Fluorescence from the gain volume imaged from the top and the side. Right: Streak camera image of the sideways emission (Superfluorescence) together with a weak fraction of the excitation pulse (Excitation).*

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