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RESEARCHER SHORT TERM MOBILITY REPORT

By: Stefan Wabnitz

Reference: CNR 2015 Short Term Mobility (STM) programme
Beneficiary: Stefan Wabnitz, Università degli Studi di Brescia, associate INO Naples Research Unit
Subject: Final report of Prof. Stefan Wabnitz activity at MPQ Laboratory of Université Paris 7-Paris Diderot- Period of stay: 24/9-13/10/2015.
Date of report: 27/10/2015
Work topic: Preparation and submission of a FET-Open European project on integrated mid-infrared optical parametric oscillator frequency comb sources (MIROIR), and design of AlGaAs microring resonators for low power comb generation based on second and third order nonlinearities.

I- Introduction and background

MIR spectroscopy is a powerful technique for univocally identifying and quantifying molecular species in a given environment, providing a tool for non-intrusive diagnostics of composite systems of physical, chemical or biological interest, in gas, liquid or solid phase. Spectrometers working in the MIR region can be deployed in a large number of roles, like research, minimally invasive medical diagnostics, environmental monitoring, industrial real-time process control, and security applications. Furthermore they could be used to assess the quality of pharmaceutical and food products, in addition to facilitating process control during production.

Optical frequency combs (OFCs) are light sources with a spectrum containing thousands of equally spaced laser lines. Originally based on femtosecond Ti:sapphire lasers, bulk OFCs have revolutionized precise optical frequency measurements by making it possible to directly link any optical frequency to a microwave clock. Nowadays they are the indispensable equipment for many other applications, ranging from synchronization of telecommunication systems to astronomical spectral calibration and biomedical or environmental spectrometry. To date, most of the comb spectroscopies rely on table-top femtosecond systems. As an alternative, Kerr microresonator combs based on third-order nonlinear materials have shown the technological possibility to reduce the size of a sensing device[1], although complete chip-scaling is hampered by the still too high pump power required for their operation.

Currently OFCs are directly available in the visible (VIS) and near-infrared (NIR) spectral range, and are typically transferred to the MIR through nonlinear frequency mixing. On the other hand, the availability of OFCs with direct broad spectral emission in the MIR region would allow for the simultaneous monitoring of multiple trace gas species or quantitative detection of liquid and solid samples.

The collaboration between INO-CNR and the MPQ Laboratory of Université Paris 7, and the FET-Open project proposal MIROIR which has been jointly prepared and submitted to the European Commission, is based on the scientific breakthrough achieved with the recent demonstration by INO CNR of OFC generation in second-order $\chi^{(2)}$ nonlinear cavities[2]. Such quadratic frequency combs represent a new paradigm of comb generation, opening the way to a novel class of highly efficient and versatile frequency comb synthesizers in miniaturized devices. Inherently more efficient than Kerr OFCs, $\chi^{(2)}$ combs configured in micrometric structures can be operated with pump powers at the microwatt scale, way better than the present state-of-the-art of Kerr microresonators. Still at an early stage, $\chi^{(2)}$ combs can take advantage of the availability of different materials with a strong quadratic nonlinearity. Moreover, three-wave mixing makes it possible to

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[1] T.J. Kippenberg et al., "Microresonator-Based Optical Frequency Combs," Science **332**, 555 (2011).

[2] I. Ricciardi, S. Mosca, M. Parisi, P. Maddaloni, L. Santamaria, P. De Natale, and M. De Rosa, "Frequency comb generation in quadratic nonlinear media," Phys. Rev. A **91**, 063839 (2015).

generate OFCs simultaneously in different frequency regions.

II- Objectives of the work motivating the visit

Members of the Electromagnetic Fields and Photonics group at the University of Brescia associated with INO-CNR have many years of experience in the modelling of optical beam propagation in guided wave structures, and in developing the theory of ultrashort nonlinear pulse propagation for applications to communications and broadband light sources. The Nonlinear and Quantum Optics group of INO-CNR in Naples has renowned know-how and experience in the field of nonlinear optics and in the development of highly coherent sources based on nonlinear frequency mixing in periodically poled crystals, with applications to spectroscopy.

The MPQ Laboratory has strong expertise in the physics and the engineering of laser and nonlinear photonic devices on chip. In particular, Prof. Leo's group is active in the design, fabrication and characterization of semiconductor microcavities for parametric and quantum light generation, frequency conversion and optomechanical phenomena.

The objective of the visit and the subsequent long-term collaboration between INO-CNR (and the associated people at the University of Brescia) and Université Paris 7 (as formulated in the MIROIR project) is to develop $\chi^{(2)}$ microresonators in the AlGaAs material platform, for the on-chip implementation of a new revolutionary class of comb-based spectrometers. AlGaAs has a strong quadratic nonlinearity, a broad transparency window in the MIR, and a bandgap that can be engineered by varying the alloy composition to eliminate two-photon-absorption. Moreover, this platform enables monolithic integration of laser source and detector on the same chip. While $\chi^{(2)}$ mixing processes have already been demonstrated in AlGaAs microdisks[3], the fabrication of bus-microring mesa structures with low vertical index contrast and large depth/width aspect ratios is challenging, and it also limits their effective nonlinearity.

III- Report of activities

IIIa) FET Open project preparation

The first part of the visit has been dedicated to the preparation and joint submission, as a coordinator, of a FET-Open European project on integrated mid-infrared optical parametric oscillator frequency comb sources (MIROIR). The participants to this project are: the CNR, the MPQ group of the Université Paris Diderot (MPQ, the Julius Maximilians Universitaet Wuerzburg (UWU), and the Danmarks Tekniske Universitet (DTU). The project has duration of 36 months and a total requested budget of 2526576 euros. The MIROIR consortium is well-balanced, including world-leading universities and a non-profit research organisation, with forefront skills in:

- Fundamental and applied nonlinear optics (theory experiment), and spectroscopy (CNR)
- Design, fabrication and characterization of hybrid nonlinear photonic devices (DTU)
- Semiconductor lasers and technology in the GaAs platform (UWU)
- Design, fabrication and characterization of monolithic photonic devices (MPQ)

Therefore all participants possess a world-class record in one or more of the project domains. More specifically, the consortium owns patents for semiconductor lasers (UWU), leadership in integrated OPOs[4] and microdisk SHG[3] (MPQ), and is world leader in AlGaAs-on-insulator nonlinear microring resonators (DTU). Finally, a crucial competitive advantage of the consortium is the breakthrough paper by CNR

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[3] S. Mariani, A. Andronico, A. Lemaître, I. Favero, S. Ducci, and G. Leo, "Second-harmonic generation at 1,55 μm in AlGaAs microdisks", Opt. Lett. 39, 3062 (2014); P.S. Kuo et al., "Second-harmonic generation using 4-quasi-phasematching in GaAs whispering-gallery-mode microcavity," Nature Commun. **5**, 3109 (2014).

[4] M. Savanier, C. Ozanam, L. Lanco, X. Lafosse, A. Andronico, I. Favero, S. Ducci, and G. Leo, "Near-infrared optical parametric oscillator in a III-V semiconductor waveguide," Appl. Phys. Lett. 103, 261105 (2013). Featured on the journal cover.

researchers, recently published[2], and the associated development of the theory behind the principle of quadratic comb generation[5]. UWU, MPQ and DTU will carry out complementary technological approaches in a fully synergic fashion. UWU will provide epitaxy and laser fabrication in the GaAs platform, MPQ oxidation and electro-optical characterization in the same platform, and DTU the fabrication in the bonded platform. Partners will systematically share samples and test protocols.

The detailed objectives of the MIROIR project are:

- Development of a novel theoretical framework, describing the generation and control of stable frequency combs in nonlinear quadratic microcavities.
- Development of a new technological platform for the fabrication of waveguides and passive components for the MIR range; two variants of the platform will be investigated: AlGaAs-on-sapphire and AlGaAs-on-AlOx.
- Demonstration of frequency comb generation in microresonator using an integrated and electrically pumped laser pump.
- Demonstration of generation and control of stable frequency combs in nonlinear quadratic microcavities.
- Demonstration of low-phase noise and widely tuneable optical comb generation in the MIR range.
- Demonstration of the feasibility of the MIROIR frequency combs for a dual-comb spectroscopy dedicated to real-time spectroscopy in the liquid and solid-state phases.

Since their appearance at the turn of the millennium, laser frequency combs have revolutionized precise optical frequency measurements with an impact on telecommunication and spectroscopic systems. Mode-locked laser-based OFCs have been joined by continuously pumped Kerr microresonators, exploiting third-order nonlinearity, and by quantum cascade lasers. Microresonator-based Kerr combs in planar integrated platforms involve micro or millimetre size devices, but still require pump powers typically in the 0.1-1 W range. The lack of femtosecond oscillators directly emitting in the MIR spectral region has slowed down the development of convenient instrumentation, however progress has been reported in the efficient nonlinear conversion of synthesized combs from NIR to MIR by means of optical parametric oscillation (OPO) or difference frequency generation[6].

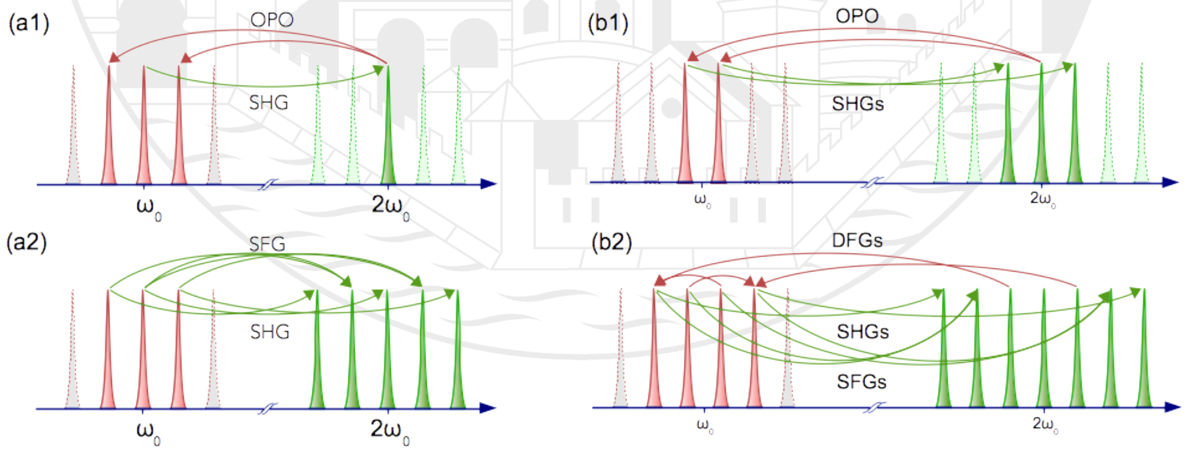

***Figure 1**: Basic principle of quadratic comb generation.*

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[5] F. Leo, T. Hansson, I. Ricciardi, M. De Rosa, S. Coen, S. Wabnitz, M. Erkintalo, "Walk-off-induced modulation instability, temporal pattern formation, and frequency comb generation in cavity-enhanced second-harmonic generation," submitted to Physical Review Letters (2015).

[6] A. Schliesser et al., "Mid-infrared frequency combs," Nature Photon. **6**, 440 (2012).

Very recently, experiments in bulk free-space resonators have demonstrated that **frequency combs can also arise entirely through second-order $\chi^{(2)}$ nonlinear effects**,[2] and our theoretical analysis shows that $\chi^{(2)}$ combs can be potentially operated with pump powers as low as few microwatts. The basic principle of the MIROIR comb generation via three wave mixing (TWM), pumped at the fundamental frequency (FF) ω_0, is outlined in Fig. 1. Above a threshold power (see section 1.4), the second-harmonic (SH), at $2\omega_0$, gives rise to an internally pumped OPO with new oscillating fields that are frequency symmetric around the FF [Fig. 1(a1)]. Subsequently, thresholdless cascaded second-order processes create new oscillating fields, forming an equally spaced comb structure around both the FF and the SH [Fig. 1(a2)]. Similarly, combs can be generated starting directly from a degenerate OPO configuration [Fig. 1(b1-b2)]. Such parametric quadratic OFCs represent in fact a new paradigm of comb generation, not only for the alleviated pump requirements, but also for the fundamentally different underlying physical mechanism. They are relatively immune to material dispersion and open the way to a novel class of highly efficient and versatile frequency comb synthesizers based on second-order nonlinear materials in miniaturized devices. One of the ambitions of our project is the development and experimental testing of a theoretical framework, which lays the foundation for a deep comprehension of the MIROIR comb-generation technique; equally important, providing a toolbox for fully designing and controlling the properties and performance of targeted miniaturized devices.

AlGaAs, the material of our choice, exhibits a large quadratic nonlinearity. To date, however, AlGaAs whispering-gallery-mode (WGM) microresonators include either a thick layer stack on GaAs substrate[7], or a suspended structure on a narrow pedestal[3]. The former has low vertical index-contrast that limits device effective nonlinearity and requires very challenging fabrication processes, while the latter lacks a robust waveguide-to-resonator coupling scheme. In the MIROIR project, we will develop **AlGaAs-on-insulator (sapphire or AlOx) platforms** offering high index contrast and the potential to implement complex designs like in silicon-on-insulator photonics[8]. Very recently, DTU has demonstrated an **AlGaAs-on-silica platform for nonlinear applications in NIR**.[9] They have obtained low-loss nano-waveguides with ultra-high effective nonlinearities and micro-resonators with quality factors orders of magnitude higher than the state-of-the-art in the AlGaAs platform. **The first demonstration of Kerr comb generation** based on cubic $\chi^{(3)}$ effects in AlGaAs material was also obtained by DTU with threshold power at the milliwatt level[9]. As quadratic $\chi^{(2)}$ nonlinearity is orders of magnitude larger than cubic $\chi^{(3)}$ nonlinearity, the proposed AlGaAs-on-sapphire and AlGaAs-on--AlOx platforms, combining the strong light confinement and high quadratic nonlinearity, will allow us to dramatically further reduce the threshold power of the MIROIR $\chi^{(2)}$ comb down to the **microwatt level.** The platform development will pave the way towards a fully integrated comb system where comb generation is ignited by on-chip light sources as shown in Figure 2.

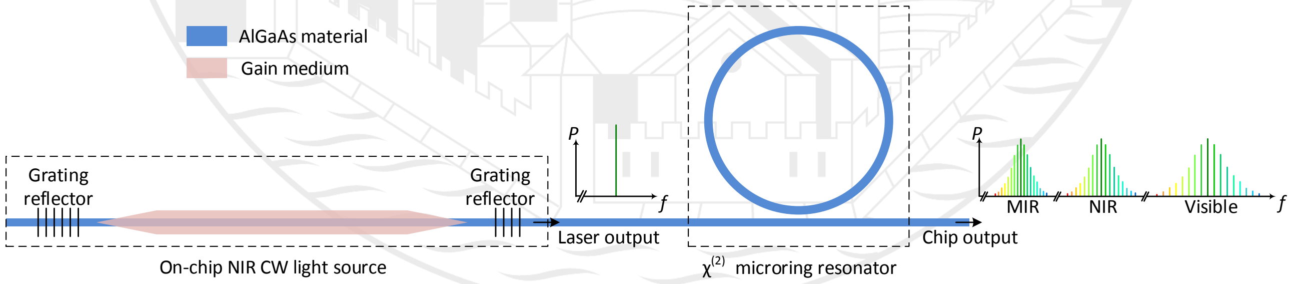

***Figure 2**: Schematic drawing of a fully-integrated MIROIR comb generator consisting of an on-chip NIR continuous wave (CW) light source and nonlinear $\chi^{(2)}$ microring resonator.*

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[7] P. Kultavewuti, V. Pusino, M. Sorel And J. S. Aitchison, "Low-power continuous-wave four-wave mixing wavelength conversion in AlGaAs-nanowaveguide microresonators" Opt. Letters 40, 3029 (2015).

[8] N. Morais, O. Stepanenko, A. Lemaitre, C. Gomez, T. Hansson, G. Leo, and S. Wabnitz, CLEO Europe, CD_P_20 (2015).

[9] M. Pu et al., "AlGaAs-on-Insulator Nonlinear Photonics," arXiv:1509.03620; M. Pu et al., "Ultra-Low Threshold Power on-chip optical parametric oscillation in AlGaAs-on-insulator microresoantor," CLEO, JTh5A.9 (2015).

IIIb) Device design and modelling of frequency comb nonlinear dynamics

The second part of the visit was dedicated to the development of theoretical models and the numerical design of microring resonator frequency comb sources based on both cubic and quadratic nonlinearities. The motivation for the use of the AlGaAs platform it the strong quadratic $\chi^{(2)}$ (d_{14}=110 pm/V) and cubic $\chi^{(3)}$ ($n_2 \approx 1.55\times10^{-13}$ cm^2/W) material response, which make the nonlinear dynamics of GaAs devices richer than, e.g., silicon or silicon-nitride based devices, and might permit to effectively generate a single frequency comb from the visible to the near infrared and even the MIR (see Fig.2). To this end we have designed bandgap engineered AlGaAs microrings, with special care for suppressing two-photon absorption and the related free carrier generation.

With the ridge guiding structures sketched in Fig. 3a-3b, MPQ fabricated the bus-resonator device of Fig. 3c by molecular-beam epitaxy, electron-beam lithography and ion etching. We resorted to this race-track geometry because it allows for a better compromise between technological constraints and critical coupling.

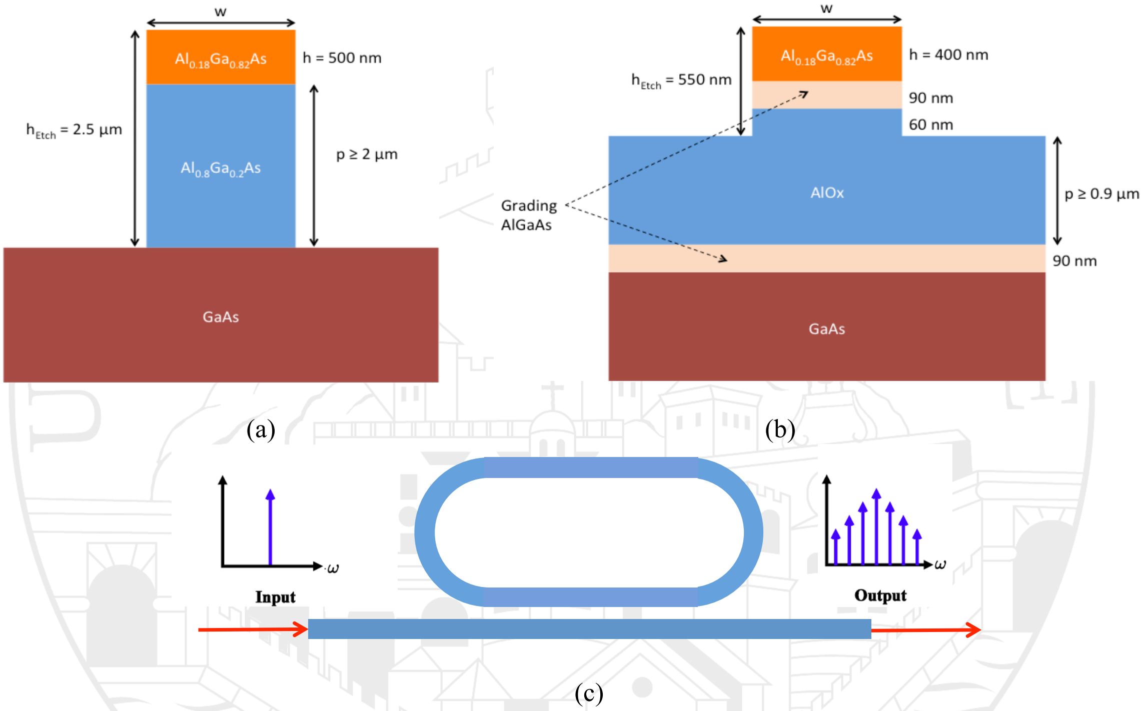

Figure 3 *(a) Structure 1 with Al$_{0.8}$Ga$_{0.2}$As substrate: deeply etched ridge because of the weak index step between core and substrate; (b) Structure 2 with aluminum oxide substrate: shallower ridge thanks to much higher weak index step between core and substrate; (c) planar bus-ring geometry for FWM-based frequency comb source.*

In the numerical simulations, first of all we carried out an optimization of waveguide core aspect ratio for dispersion engineering, in order to reduce the dispersion around 1500 nm to values close to zero. As can be seen from Fig. 4, where the waveguide height h is kept constant to 500 nm, optimal waveguide width is determined to be close to w=600 nm.

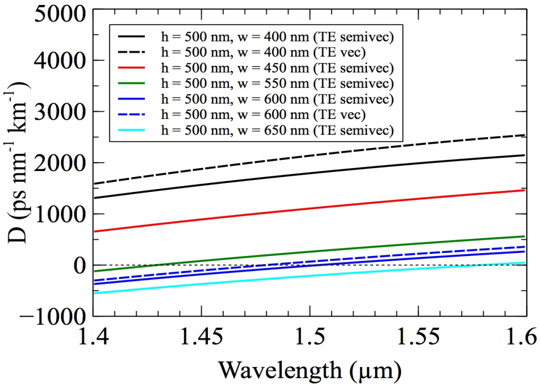

Figure 4: *Variation of waveguide dispersion vs. height h and width w of the ring cross-section.*

Next, we numerically simulated by means of the generalized Lugiato-Lefever equation including the AlGaAs cubic nonlinear response only, the generation of coherent frequency combs associated with the presence of a single cavity soliton for both structures 1 (Fig.5) & 2 (Fig. 6). As can be seen, for a quality Q factor of 10^5, for both structures coherent frequency combs with a bandwidth in excess of 200 nm are predicted at the moderate CW pump power of 50 mW.

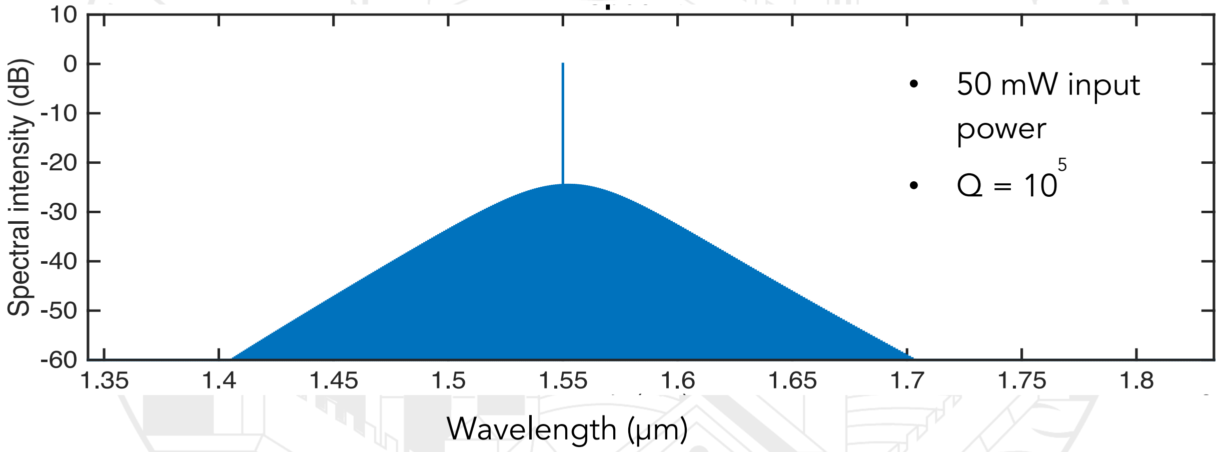

Figure 5 *Optical frequency comb for structure 1, with gap=250 nm, L=300 μm, R=50 μm: FSR = 117 GHz.*

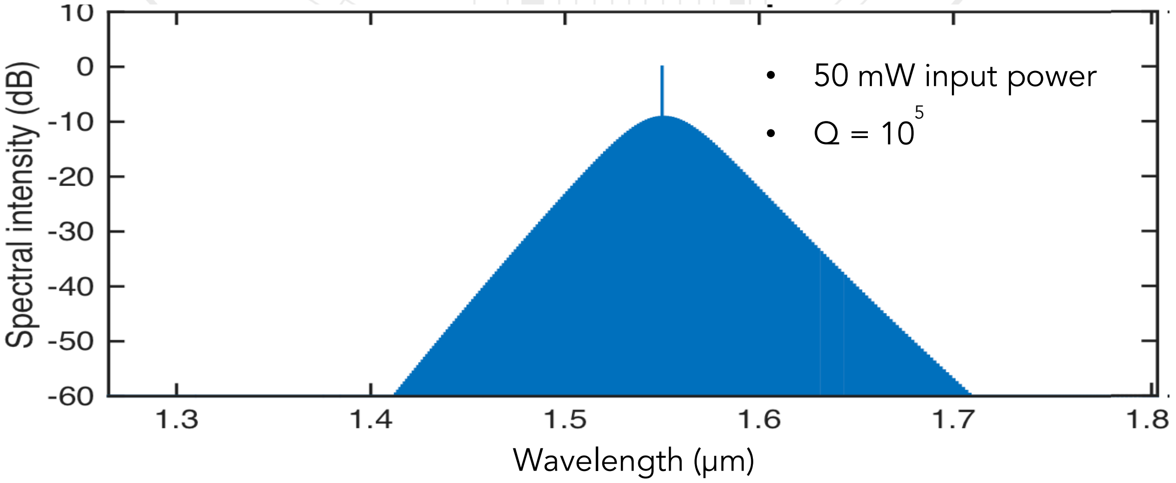

Figure 6 *Optical frequency comb for structure 2, with gap=100 nm, L=61 μm, R=50 μm: FSR = 171 GHz*

The MPQ Lab carried out the fabrication of both structures, grown by molecular beam epitaxy, with a special care for structure 2, which includes two transition regions with a grading in the Al molar fraction between the $Al_{0.98}Ga_{0.02}As$ thick layer to be oxidized and its adjacent layers. Oxidation is a critical point, which is handled with a state-of-the-art oxidation oven including an in situ optical control.

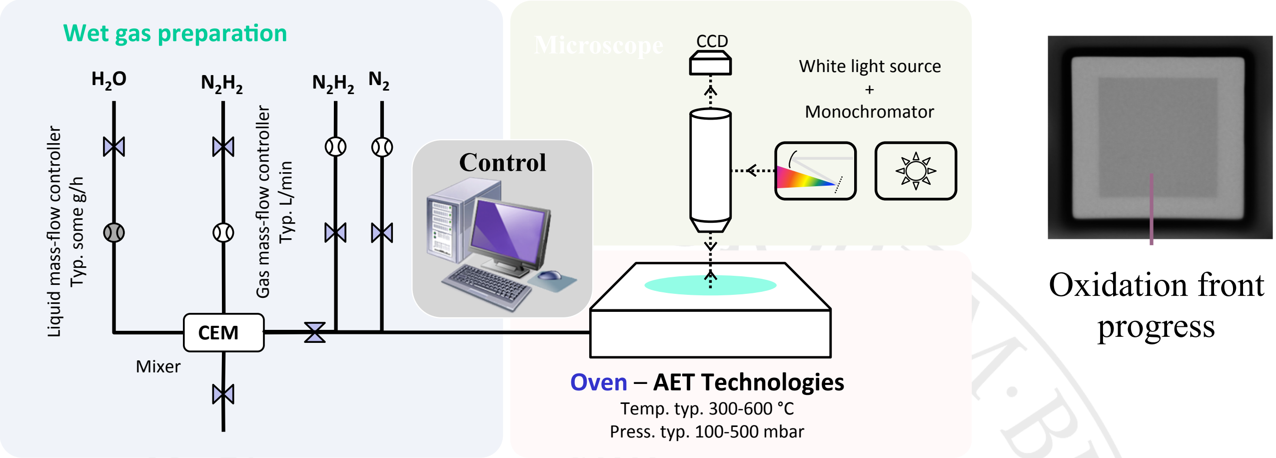

***Figure** 7 Details of fabrication process: Electronic lithography: MaN-2403 photoresist, AZ 726MIF developer; Dry etching: ICP-RIE Sentech SI500 system using $SiCl_4$; Oxidation: T=390°C, pressure =500 mbar, oxidation time = 30'*

A preliminary characterization of the fabricated devices (see Fig. 8) was carried out. The measurement of optical losses in the waveguides has however revealed a too high level of linear losses, which prevented the measurement of mode spectra even in the linear regime. Therefore a new round of fabrication is presently underway in order to reduce the waveguide losses. This prevented so far the comparison between theoretical and experimental spectra also in the nonlinear regime where frequency comb generation has been predicted.

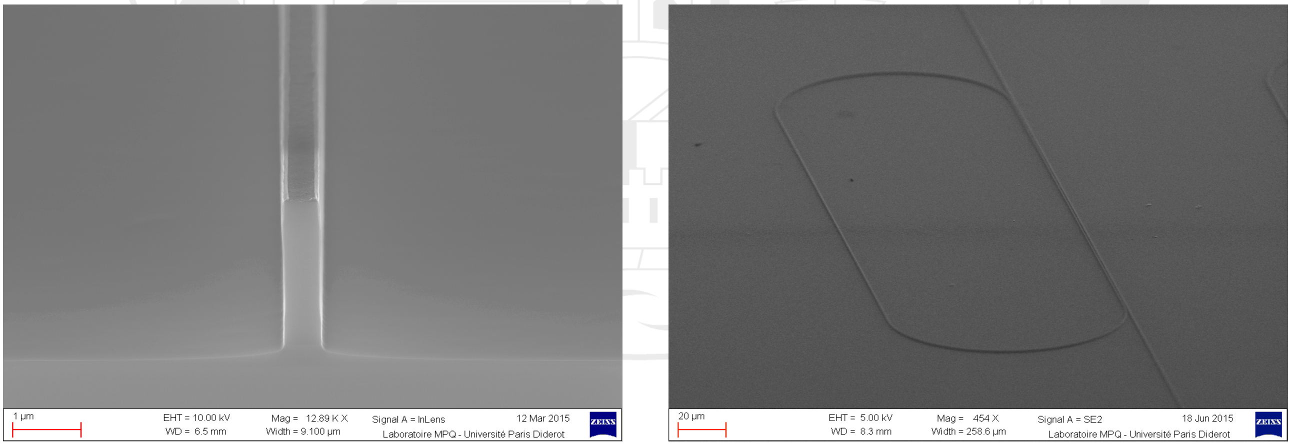

***Figure 8** Scanning electron microscope images of AlGaAs microrings.*

In the meanwhile, we have developed a new model for the numerical generation of frequency combs in a microresonator with both cubic and quadratic nonlinearity, based on the single field approach.

We model the dynamics of optical frequency comb generation by means of the Ikeda map, combining the generalized nonlinear envelope equation (GNEE)[10], for describing the evolution of the envelope of the electric field within a waveguide with both quadratic and cubic nonlinearity, together with boundary conditions that relate the fields between successive roundtrips and the input pump field:

$$A^{m+1}(t,0) = \sqrt{\theta}A_{in} + \sqrt{1-\theta}e^{i\phi_0}A^m(t,L), \tag{1}$$

$$\left[\partial_z - D + \frac{\alpha}{2}\right]A^m(t,z) = i\rho_0\left(1 + i\tau_{sh}\frac{\partial}{\partial t}\right)p_{NL}(t,z,A^m) \tag{2}$$

Here the independent variables are the evolution variable z, which is the longitudinal coordinate measured along the waveguide, and t which is the (ordinary) time. The first equation is the boundary condition that determines the intracavity field $A^{m+1}(t,z=0)$ at the input of roundtrip m+1 in terms of the field from the end of the previous roundtrip $A^m(t,z=L)$ and the pump field A_{in}. The path length of the resonator is assumed to be equal to L. Additionally, θ is the transmission coefficient between the resonator and the bus waveguide, and $\phi 0 = 2\pi\, l - \delta_0$ is the linear phase-shift, with δ_0 the detuning from the cavity resonance (assumed to correspond to the longitudinal mode number l = 0) which is the closest to the pump frequency. Eq.(2) is written in a reference frame moving at the group velocity, the dispersion operator is

$$D = \sum_{m\geq 2}\frac{i^{m+1}}{m!}\left(\frac{\partial^m\beta}{\partial\omega^m}\right)\frac{\partial^m}{\partial t^m}. \tag{3}$$

and the nonlinear polarization is written as the sum of quadratic and cubic contributions

$$p_{NL}^{(2)} = \frac{\varepsilon_0\chi^{(2)}}{2}\left[2|A|^2\exp(i\omega_0 t) + A^2\exp(-i\omega_0 t)\right] \tag{4}$$

$$p_{NL}^{(3)} = \frac{\varepsilon_0\chi^{(3)}}{4}\left[3|A|^2A + A^3\exp(-2i\omega_0 t))\right] \tag{5}$$

We numerically solved Eqs.(1)-(2) for describing frequency comb generation processes in a microresonator. In order to impose phase-matching among the waves at the center of the respective combs, we included in Eq.(2) a periodic square wave spatial modulation of the second-order nonlinear coefficient with different values of the quasi-phase-matching (QPM) period Λ. We numerically simulated the spectral and temporal dynamics of parametric frequency comb generation by solving Eq.(2) in the frequency domain as a set of coupled ordinary differential equations for the resonator modes, with computationally efficient evaluation of three and four-wave mixing terms in the time domain via the fast Fourier transform method. We used a quantum noise input (one photon per mode), and considered a microresonator operating in the critical-coupling regime. The advantage of the GNEE approach is that it includes all parametric processes at once, namely second-harmonic generation, optical parametric generation and oscillation, and sum-frequency

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[10] S. Wabnitz, A. Picozzi, A. Tonello, D. Modotto, and G. Millot, J. Opt. Soc. Am. B **29**, 3128-3135 (2012).

generation. Extensive simulations are under way for different type of microresonator materials, and will be the subject of a forthcoming publication.

IV- Conclusions

The STM has been essential for the successful preparation and completion of the FET-Open European project on integrated mid-infrared optical parametric oscillator frequency comb sources (MIROIR). In fact the day-to-day close interaction with Prof. Giuseppe Leo in the final week before submission was absolutely necessary given the very strict timeline in front of us, in particular in order to prepare a sound implementation work plan and timing of the project activities. Moreover, the STM has permitted us to substantially advance in the collaboration on frequency comb generation based on the AlGaAs platform, thanks to the development of appropriate numerical design and simulation codes that will be of invaluable help as soon as the first experimental results will be available by the MPQ lab.

V- Acknowledgements

I am very grateful to Prof. Giuseppe Leo for his warm hospitality at Université Paris Diderot, and to Dr. Maurizio De Rosa for his precious support.