Interview with Nathalie Picqué

Specialist in laser frequency combs, scientific director at the Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy - Berlin, Germany.

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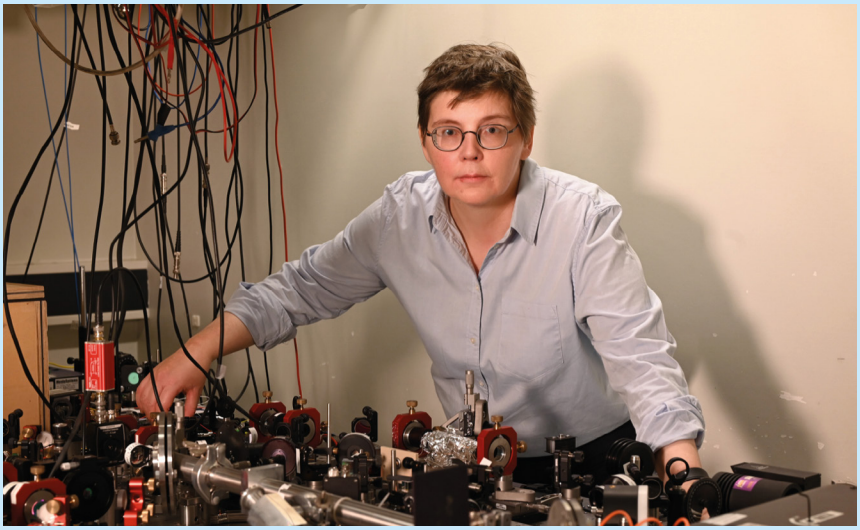

Charlotte Huber, Max Planck Institute of Quantum Optics

How did you become interested in science?
When I was in secondary school, I was passionate about archaeology. My parents were scientists and they managed to get me an appointment with an archaeologist. They told me that it was almost impossible to secure an academic position in that field and my dream collapsed. I decided to study science by default, thinking it would open me opportunities. I took my science high school leaving exam (baccalaureate series C), then went on preparatory classes to the Lycée Saint-Louis, and then to the Ecole Normale Supérieure in Paris for a magistere degree in chemistry. Meanwhile, I realised that I was more attracted in the physics side of chemistry and decided to switch to physics. That's why I enrolled in the Masters in "Lasers and Matter" at the Pierre and Marie Curie University, École Polytechnique and the University of Paris-Sud.

Did this Master in Lasers and Matter strongly influence the rest of your career?
Yes, I really enjoyed the lectures. For the first time, I started to really engage with the material being taught instead of having my own agenda. I was particularly interested in quantum mechanics, non-linear optics and lasers, and molecular spectroscopy. Part of the training involved studying research papers and presenting them to others, and this was my favourite activity. I did a short internship at an institute called the Laboratoire de Physique Moléculaire et Applications in Orsay (now merged with other institutes as ISMO). My internship focused on pushing the limits of the precision of a Fourier transform spectrometer, a widely used spectroscopy technique based on the Michelson interferometer.

Did this experience lead you to pursue a PhD?
It seemed to me that by developing instrumental concepts, I might be able to measure something that had never been measured before. I developed a passion for the high-resolution Fourier transform spectrometer I was developing and decided to pursue my doctoral thesis in the same team. The ideas were to develop measurement techniques combining high temporal and spectral resolution to observe unstable molecules, or selective techniques to preserve only transitions of paramagnetic or ionic species. My supervisor was Guy Guelachvili, a pioneer in Fourier spectroscopy. He had been involved in the first high-resolution Fourier spectroscopy in the 1970s at the Laboratoire Aimé Cotton under the direction of Pierre and Janine Connes. My graduate research has been one of the happiest times of my life. The team was very supportive and I had great freedom to carry out original research projects. Our aim was to explore new topics, but we did not try to publish our results in high-impact journals. I defended my thesis in December 1998.

What did influence your choice for finding your post-doctoral position?
I wanted to learn new things, especially about frequency metrology and precision measurements with lasers. That's why I joined Massimo Ingucio's group at LENS in Florence as a post-doctoral fellow, where the fine structure splitting was being measured more and more precisely in order to better determine the fine structure constant. I learnt a lot during my 1.5 year stay and it was there that I first heard about frequency combs from Theodor Hänsch. In 2001 I was offered a position at the CNRS (French National Centre for Scientific Research), which was ideal for me as I was very keen to explore my own ideas on optical frequency combs for Fourier spectroscopy. I joined the Laboratoire de Photophysique Moléculaire in Orsay (which has since merged with other institutes to form ISMO).

What was your CNRS research project?
I wanted to develop experimental spectroscopy techniques that combine lasers and Fourier spectroscopy. Fourier spectroscopy offers a wide spectral range, while lasers offer high resolution and high sensitivity. My aim was to combine the two to have all these features in a single instrument. At that time, I was already ready to work with optical frequency combs and had come up with the dual comb scheme for Fourier spectroscopy, among other crazy ideas. But I was part of a team that had no laser and no expertise in ultrafast optics. My applications for funding were systematically turned down. Broadband spectroscopy with frequency combs was a very bold idea. It was a time when people were suspicious of frequency combs as frequency rulers, so proposing to use two of them for interferometry was obviously ahead of its time. My projects couldn't start as I wanted them to because of a lack of funding. I started to build the lasers myself, in my spare time in addition to pursuing other, less intriguing but funded, projects. In 2006, graduate student Julien Mandon and I saw our first femtosecond pulses from our home-built mode-locked oscillator. Out of ignorance, I had chosen to build a Cr:YAG laser, which turned out to be a complex system to build and work with. Nevertheless, it was a great way to learn about ultrafast laser physics and I had managed to attract some excellent students who contributed greatly to the success of the project.

How did this home-made laser source impact your work in spectroscopy?
Once the frequency comb generator worked, we were able to analyse it with our Michelson interferometer and quickly demonstrated the feasibility of our approach. We obtained the first experimental results on a technique now called frequency comb Fourier transform spectroscopy. The first paper reporting these results was published in Optics Letters in early 2007. At the same time, another group in the US was starting to explore similar ideas. I became painfully aware that we were not alone and did not have an infinite amount of time to explore our ideas, that it was necessary to publish quickly, and that without dedicated funding I could not compete with groups in the best US institutions. At the end of 2006, I contacted Theodor Hänsch, whom I knew from my post-doc in Italy and who had just been awarded the 2005 Nobel Prize in Physics for the invention of frequency combs as frequency rulers in metrology. In early 2008, I went on a six-month sabbatical to his institute, the Max Planck Institute of Quantum Optics (MPQ) in Garching near Munich, with two PhD students. This stay significantly accelerated our work, as we were able to benefit from the infrastructure and expertise at MPQ. We were able to greatly improve the dual-comb interferometer we had first developed in Orsay, and we were the first to measure spectra with resolved comb lines over the full span of the combs. Ironically, I received my first dedicated funding in France for this project in 2008.

Was it during this period that you considered moving to Germany?
I realised that the working conditions and the scientific environment at the Max Planck Institute for Quantum Optics were exceptional. At the end of my sabbatical, Theodor Hänsch offered me a position at the MPQ and I was very excited about the opportunity. There was a transition period where the CNRS allowed me to go back and forth, followed by a secondment from the CNRS. I moved to Garching full-time in 2011. This has certainly greatly evolved my approach to research, as it is unique to get to know two very different institutions such as the CNRS and the Max Planck Society so closely. This has made me think a lot about how I organise myself and what I prioritise.

This opened up a very stimulating period for your research activities.
Yes, it was an incredible time, with many directions to explore and abundant resources. Frequency comb interferometry turned out to be a great platform for exploring new insights. Initially there were only a few groups involved in this research, but there was intense competition. Now the community has grown, with more than two hundred groups contributing.

What were some of your most significant scientific results during this time?
It all started out of curiosity. I had noticed that by using two frequency combs with slightly different frequencies, the time delay between pulses in a pulse pair, one from each comb generator, would increase from pulse pair to pulse pair. This mimicked the effect of a scanning Michelson interferometer, where you send a pulse train while scanning one of the two arms. So it was obvious to me that you could do Fourier transform spectroscopy with such a system. Initially, I saw the main advantages of the dual comb interferometer as being the absence of moving parts and the ability to scan the delay between pulses much faster than with a mechanical interferometer. So that was the aim of our first demonstrations. Once we had the comb sources, it produced results incredibly quickly. However, we recognised that the frequency comb sources we needed weren't exactly the same as those needed for metrology. We had to develop sources that emitted in regions of interest for molecular spectroscopy, particularly in the mid-infrared, because no sources were available. We then became involved in the development of frequency comb sources in this spectral region. We also realised that the methods for measuring interference in dual comb spectroscopy required long coherence times between the two combs. Fast feedback loops are required. There was a lot of groundwork to be done to make the technique efficient, and after the first convincing proofs of principle, it took years to explore different variants that would allow high quality spectra to be obtained, especially in the mid-infrared and, more recently, in the near-ultraviolet

What were your main scientific projects?
I had originally planned to work on linear absorption spectroscopy, where the spectrum of small molecules is sampled by the narrow lines of the comb. When the combs are stabilised and the dual-comb

spectrometer resolves the comb lines, the frequency scale becomes self-calibrated and the signature of the spectrometer, which we call the instrumental line shape, becomes negligible compared to the molecular profiles. I thought it would be exciting to explore this potential. Then other insights followed in quick succession: because the laser sources we used emit ultrashort pulses, we can exploit nonlinear phenomena at the sample, and we played with several schemes using two-photon excitation or coherent Raman effects. With two-photon excitation, we were able to largely cancel out the Doppler broadening of the lines. In dual-comb spectroscopy, as in other techniques of Fourier spectroscopy, all spectral elements are recorded on a single detector. Another intriguing idea was to replace the detector with a matrix of detectors. We could then perform hyperspectral imaging and even digital holography.

You were recently appointed scientific director at the Max Born Institute in Berlin.
Yes, and I currently spend a lot of time commuting between Munich and Berlin because the new laboratories in Berlin are not yet ready. I now have exceptional resources to carry out my research, with ample space for state-of-the-art laboratories, positions for collaborators and generous funds for equipment and consumables. This is extremely exciting and, in the meantime, creates some new challenges: I have always spent a lot of time in the lab; my schedule was organised to spend every afternoon with the students in the lab, and I will need to organise myself to maintain a strong involvement in the lab.

Have you developed international collaborations?
Over the years, our work has benefited greatly from collaborations with specialists all over the world. For example, in our work on integrated optics, we have had fruitful collaborations with Jelena Vučković's group at Stanford, Kerry Vahala at Caltech, Alex Gaeta and Michal Lipson at Columbia, Bart Kuyken at Ghent University and Marko Lončar at Harvard. I have also kept in touch with French scientists and, just to give an example, we have interesting ongoing work with my brilliant former post-doc, Lucas Deniel, who is now a "maître de conferences" in Lannion.

What scientific challenges do you want to address in the framework of your new position?
There is nothing I love more than exploring new insights and ideas, setting up new experiments and exploiting the interplay between fundamental physics and photonics technology. I love the intellectual rollercoaster ride that is the uncertainty in the early stages of an experiment, when we gradually discover all the problems to be solved and oscillate between doubt, excitement, confusion and hope. This guides my endeavours. In each new project I try to add ingredients that are new to me, so that I am out of my comfort zone.
I have been surprised to find that frequency combs and dual-comb interferometry have been a rich enough platform to continually fuel new ideas, and I now feel that the most interesting ones are yet to come. Dual comb interferometry is the only broadband spectroscopy technique where the resolution depends only on time. This is fundamentally different from all spectrometers where the resolution depends on length. I realised this only some years ago, and the consequences have not yet been exploited. For me, this opens up new questions such as: Can we bridge the gap between broadband spectroscopy and frequency metrology? Can we perform frequency metrology over a wide spectral band? What are the accuracy limits of these techniques? Can we contribute to fundamental physics tests? Can we search for new physics by pushing the limits of precise laser spectroscopy of simple molecules? We are currently developing Doppler-free dual-comb spectroscopy and ultraviolet dual-comb spectroscopy to answer these questions. Another consequence is more relevant to applied physics: our instruments can be very compact. But how compact can they be? Can we put high-resolution spectrometers on a chip? More generally, what other interferometric techniques would benefit from a dual-comb system?