MINIATURIZING GREENHOUSE GAS ANALYSIS WITH FIBER FABRY-PEROT MICROCAVITIES

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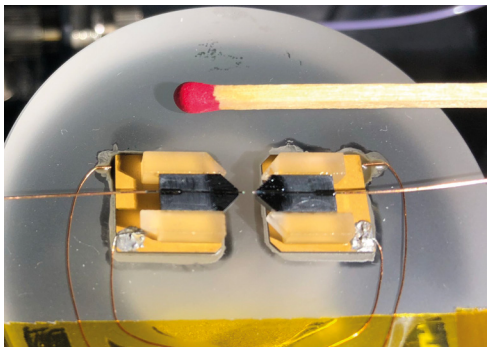

Fiber Fabry-Perot (FFP) cavities with laser-machined mirrors combine exceptional optical properties with a rugged, miniaturized, inherently fiber-coupled design. Originally developed for research on quantum technologies, they have found applications far beyond that field and are now being used to realize miniaturized, mobile greenhouse gas analysers, as this article explains.

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Reducing anthropogenic greenhouse gas (GHG) emissions is a number one requirement for a more sustainable world. Carbon dioxide is responsible for more than two thirds of the global warming, followed by methane, which has a lower concentration in the atmosphere, but has global warming potential more than 80 times that of the same quantity of CO_2. Globally, we know quite well what causes these emissions. For CO_2, fossil fuel combustion is by far the biggest culprit, but also concrete production for example. Methane is released in stages of natural gas extraction, processing and transport (pipeline leaks), but also from agriculture. Under the pressure of evidence, GHG reduction targets are agreed upon, but putting them into practice requires reliable emission measurement. This is why measurement technology has a key role to play here. For example, a recent report [1] of the United Nations Environment Programme puts improved gas leak detection on top of its list of recommended measures to reduce methane by 45 % by 2030. The EU methane regulation which entered into force in August 2024 requires operators in the oil and gas sector to detect leaks and monitor source-level emissions on a regular basis. On the research side, oceanic CO_2 concentration needs to be measured globally with increasing spatial and temporal coverage. In all these measurement situations, small, mobile gas analyzers can provide substantial gains in productivity, coverage and data quality, but this requires instruments which are only starting to exist.

While there is a wide range of technologies for gas analysis in general, very few of them are suitable for precise, quantitative measurement of low concentrations in the ppm (parts per million) range and below, as required for GHG detection and abatement. For example, electrochemical and MOS sensors are cheap and small, which is why they are used in

CO_2 detectors for classrooms or air conditioning systems. But they are prone to cross-sensitivity: their reading may be affected by other gases than the CO_2 they are supposed to measure, such as hydrogen or even water vapor. This makes them unsuitable for more quantitative analysis. The gold standard for quantitative trace gas analysis is laser spectroscopy, mostly by absorption: a laser source is scanned across a specific resonance of the desired molecule; the attenuation of the laser beam is measured as it passes through a sample volume. Resonance frequencies are the fingerprint of a molecule, very specific to each molecular species, making it possible to largely avoid the cross-sensitivity problem. Furthermore, the relationship between the signal (absorption) and the desired quantity (molecular concentration) is governed by simple physical laws, enabling reliable quantitative measurement in real-life conditions, where factors such as gas composition, temperature, pressure and humidity may all vary widely. This is more challenging for methods such as photoacoustic spectroscopy, which has excellent sensitivity, but a detection mechanism relying on acoustic wave conduction between the sampling region and the microphone.

Laser absorption spectroscopy, in its turn, is still a broad term that covers a variety of techniques, ranging from simple, single-pass absorption of a diode laser to powerful and sophisticated methods such as dual-comb spectroscopy. For trace gas analysis, one problem to solve is the extremely weak signal resulting from the low concentration: at the typical ~330pm CO_2 concentration in air, a laser beam which is resonant with one of the relatively strong CO_2 resonances at 2µm wavelength is attenuated by only 0.5 % per meter of path length. In spite of this very weak absorption, today's spectroscopic trace gas analyzers are impressively powerful instruments, easily reaching and even surpassing • • •

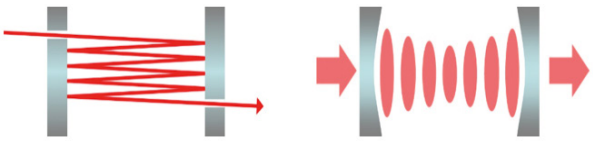

Figure 1. Two ways to increase optical path length for high-sensitivity spectroscopic detection. In a multipass cell (left), the laser beam is reflected multiple times between the mirrors, hitting a different mirror spot at every reflection. Mirrors need to be carefully aligned so that the beam leaves the cell through the exit hole after a high number of passes through the cell. In practice, this limits the number of passes to less than 100 typically. In a Fabry-Perot cavity (right), there is no exit hole. All reflections are exactly superposed and the transmitted beam results from constructive interference of the very weak transmission of each of the partial beams. With high-reflectivity, low-loss mirrors, the effective path length can be more than 100000 times the cavity length.

the sensitivity and uncertainty requirements for greenhouse gas monitoring and abatement in many cases. They reach this performance by using mirrors to multiply the optical path length. This can be done in two ways, using either a multipass cell or an optical cavity (Figure 1). With established technology, however, both cavities and multipass cells are massive, macroscopic objects, requiring sturdy construction to achieve the required stability, measuring tens of centimeters and weighing several kilograms. They also require a dedicated optical system for precisely matching the small spatial mode of the laser diode to the macroscopic cavity mode profile. The end result is a large and heavy instrument, usually weighing tens of kilograms and consuming upwards of 100 W electrical power. Mobile surveys with such instruments typically require trucks or airplanes.

Unsurprisingly, researchers and manufactures are aware of this drawback and are making efforts to reduce the size and weight of their cells and cavities. However, there are limits to what can be done without a radical change in technology. In the case of multipass cells, physics itself imposes limits to miniaturization: here, the last reflected beam should fully pass through the exit hole, while the neighbouring one should still be fully reflected. Due to diffraction, this condition becomes harder to fulfill as the mirror area is shrunk down for miniaturization. The situation is different in cavities, where all passes hit the mirrors at the same spot, so that radical miniaturization is conceivable in principle, while maintaining much higher pass numbers in the hundreds of thousands. However, this requires new approaches to fabricate the miniature, ultralow-loss, concave cavity mirrors. As we will see below, this problem has been recently solved by research advances achieved in a completely different area: this is where gas analysis meets quantum technologies.

FIBER FABRY-PEROT MICROCAVITIES

Quantum technologies have been a main driver for intense development of optical microcavities that has been going on since the early 2000's [2]. This is because qubits – the elementary building blocks of all quantum technologies – often rely on strong and controlled coupling between single photons and single quantum emitters. The latter can be solid-state objects such as quantum dots, defect centers in diamond, or indeed gas-phase atoms and molecules. This research has turned out highly original solutions, many of which have found applications beyond the application they were intended for. Well-known examples are photonic bandgap cavities, silica microspheres, and toroid resonators. However, for most of those microscale technological marvels, the optical field is concentrated with-in the solid, glass or semiconductor structure. As a consequence, gaseous emitters can only couple to it through the evanescent field that is confined to a minuscule layer close to the surface (typically less than 200nm), which leads to unwanted surface interactions and drastically restricts the detection region. For these emitters, free-space cavities such as the Fabry-Perot (cf. Fig. 1) have an obvious advantage, because the photonic field fills the entire free-space area between the mirrors, unperturbed by surface effects. This is true not only for quantum technologies, but also for gas spectroscopy, where the Fabry-Perot's entire mode volume is accessible to gas flow. Yet, traditional technologies do not allow for miniaturization of Fabry-Perot cavities without sacrificing the desired very high finesse (tens to hundreds of thousands). The reason for this is as simple as it is fundamental: high finesse means low loss upon reflection – as low as a few ppm to reach a finesse of 100000. Reaching such an extremely low loss requires correspondingly low surface roughness – below 0.2nm rms for a mirror in the near infrared. When it comes to mirror polishing, the only way to reach this ultralow roughness used to be a technique known as superpolishing, which works well for large radius of curvature of a few centimeters or more, but fails for the small, strongly curved mirrors that would be needed for a submillimeter-scale Fabry-Perot. Things have changed with the advent of fiber Fabry-Perot cavities with CO_2 laser-machined micromirrors [3,4], which turned out to be a game changer.

The idea of laser-machined micromirrors is very simple: focus a high-power laser spot onto a glass surface to obtain laser ablation, and the result will be a concave depression centered at the laser spot. What was less obvious was whether this could be combined with the required ultralow surface roughness. Laser ablation on dielectrics has a

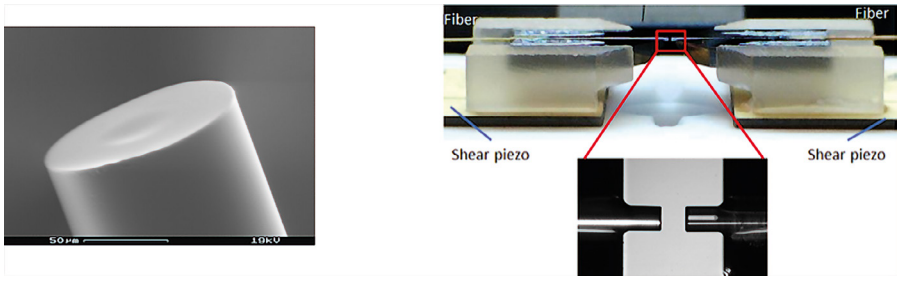

Figure 2. Left: Scanning electron micrograph of a laser-machined, ultralow-roughness mirror shape on an optical fiber. The fiber diameter is 125 µm. Right; Photograph of a complete FFP microcavity. The cavity length in this example is 120 µm; cavities have been realized with lengths ranging from less than 3 µm to more than 1.5 mm.

tendency to create sub-wavelength ripples on the machined surface, limiting the surface quality – our group rediscovered this well-known fact in our first attempts to machine optical fiber surfaces with femtosecond laser pulses. But it turned out that there is an exception: with the right parameters, we found that CO_2 laser pulses can produce ultralow-roughness concave depressions in fused silica – a readily available optical glass. Figure 2 (left) shows a scanning electron micrograph of a concave mirror shape machined in this way. Radii of curvature as low as 10 µm have been reached with this technique, with a surface roughness that consistently reaches 0.2 µm rms. These remarkable features are the consequence of two physical processes occurring simultaneously: ablation is one, and is responsible for evaporating material to create the overall concave structure. At the same time, melting occurs in a thin surface layer, allowing surface tension to dress the liquid surface into a low-roughness state. The fact that these two processes can occur simultaneously is a lucky consequence of fused silica's material parameters in the regime of CO_2 laser wavelengths. The original publications [3,4] use 10.6 µm lasers, 9.3 µm lasers were later shown to work as well [5]. Machining mirror surfaces in this way solved the micro-mirror problem: once the substrate has been machined, it can be coated with high-reflectivity dielectric coatings using existing, commercially available coating processes, yielding mirrors with absorption and scatter losses as low as a few ppm, approaching the values achieved with traditional superpolishing.

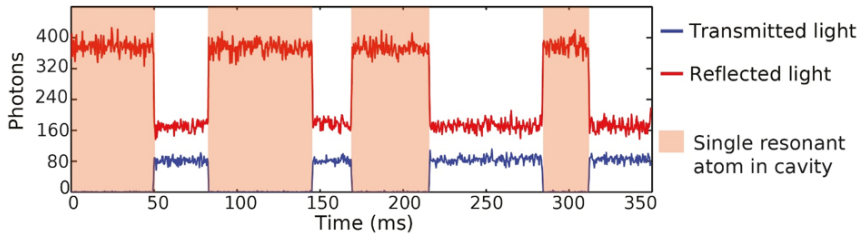

Figure 3. Single-atom detection with a fiber Fabry-Perot cavity. When a single, laser-cooled atom is trapped in the cavity and the cavity and probe laser are tuned to the atomic resonance, the cavity switches from full transmission to full reflection due to the presence of the atom. Figure adapted from [9].coupled average powers.

Good luck did not stop there. Fused silica happens to be the glass from which most optical fibers are made. Cleaved optical fiber • • •

Figure 4. Left: The Fiber Fabry-Perot tunable cavity available from Mirega.com. Right: With its 100 m optical path length in a mm-scale form factor and <100g weight, this device enables a drone-mounted trace gas analyzer (artist's view).

tips thus provide an ideal substrate on which to realize the newly discovered low-roughness micromirrors. This gives rise to a microcavity with exceptional properties: finesse values above 200000 have been achieved, while at the same time, coupling light into the cavity is as easy as plugging together optical fiber connectors. Indeed, the problem of aligning the incoming beam with the cavity mode is now solved at fabrication time, rather than on the user's optical bench. By appropriately matching the mirror parameters, fiber type and cavity length, excellent mode matching can be accomplished without additional optical elements, and cavity transmission losses below 1.5 dB have been reached. In cases where cavity parameters cannot be chosen in that regime, permanent low-loss mode matching can still be achieved by integrating a stretch of gradient-index fiber, as shown in [6].

Recently, laser-machined micromirrors have been further improved [7] by going beyond the single-shot process that was used in the original version. With a single laser shot, the resulting depression has a Gaussian shape, rather than the desired spherical one. Close to the center, the Gaussian can be approximated by a sphere, and this is why these mirrors still give good results if the beam diameter is small enough. However, the Gaussian shape limits the usable cavity length and gives rise to nonstandard higher-order modes. In the improved version, mirrors are machined by applying dozens of laser pulses to the same surface in a carefully optimized pattern. The result is a close approximation of the desired spherical shape over a much larger area. This is what has enabled millimeter-long fiber Fabry-Perots [7], and we now routinely apply this process to produce our fiber mirrors. It is a key factor in the application to gas detection, as we will see below.

SINGLE-ATOM DETECTION AND MOLECULAR SPECTROSCOPY WITH FFPS

Fiber Fabry-Perot microcavities excel in ultrasensitive detection of atoms down to the single-particle level [8]. Figure 3 shows how cavity transmission changes from full transmission to full reflection due to the presence of a single resonant atom trapped in the cavity mode [9]. Experiments like this brought FFP microcavities to the attention of many researchers. Today, FFPs are employed in research applications ranging from single-photon sources and quantum network nodes to optomechanics and biomolecule detection, and they are considered a promising technology for future atom-based quantum simulations. A novel FFP-based scanning microscopy technique has been demonstrated [10] and is now commercially available from the startup Qlibri. Interestingly, different applications exploit very different elements of the FFP's rich feature set: strong coupling to quantum particles, extremely large free spectral range and narrow resonances making them a remarkable optical filter, built-in fiber coupling, miniature size and robustness making them suitable for field applications. A review article [11] describes many of those features and applications as of 2021, but the field is moving rapidly and the list of applications continues to grow.

Some experiments have already demonstrated different forms of gas spectroscopy with FFP cavities. Early on, a beautiful series of experiments focused on Raman spectroscopy [12]. The FFP was used in a doubly resonant configuration, where both the pump beam and the Raman emission were resonant with the cavity. CO_2 in air was measured in this way, resulting in a cavity-enhanced emission signal that was enhanced by an impressive factor of 10^7 with respect to what would have been observed without a cavity under comparable conditions. More recently, absorption spectroscopy of oxygen has also been demonstrated [13]. However, these were laboratory experiments not intended to leave the well-controlled conditions of a research lab.

MINIATURIZING TRACE GAS ANALYSIS WITH FFPS

Empowered by the FFP microcavity, the vision of a compact and robust, drone-mountable cavity-enhanced greenhouse gas analyzer is now getting fulfilled. The challenge has been taken up by the newly founded startup Mirega SAS in collaboration with Laboratoire Kastler Brossel in Paris, with support from SATT Lutech and an ERC Proof-of-Concept grant of the EU. The first, crucial step was to develop the FFP cavity itself from its laboratory implementation into a high-TRL product. As a stand-alone product, it acts as a tunable optical filter with extremely narrow bandwidth

and an exceptionally large stopband. It is available as a standard product in the telecom C-band wavelength range. Other wavelengths and filter parameters will follow next year.

Most importantly however, this device has removed the main miniaturization roadblock for mobile, cavity-enhanced trace gas analysis. Based on the FFP technology, constructing an MVP-level portable CO_2 analyzer became a straightforward task, requiring a very small number of external components. We have built two of these devices which now routinely detect CO_2 in our lab. Characterization of their detection properties (sensitivity, drift etc) is under way. The optical circuit is entirely fiber-coupled with no need for free-space components. The project received another boost when this analyzer was selected for support by the Nova incubator by GRTgaz, a major European operator in pressurized gas transport. As part of this program, a methane version of our analyzer will be tested in GRTgaz's test and calibration laboratory in November. In parallel with this work, we are also pushing forward the software development that will turn the analyzer into a turnkey product.

Beyond its primary advantage, the FFP analyzer comes with additional benefits. Power efficiency is one of them, because the microscopic cavity and gas volume can be temperature stabilized with very low heating power, and because a micropower MEMS pump is enough to ensure a stable gas flow. Another advantage is the extremely small cavity volume, which is on the order of a nanoliter. This enables even the smallest gas samples to be analyzed, as required for dissolved gas analysis, for example. Moreover, it significantly increases the analyzer's response time, because the gas exchange rate is very fast. Last not least, the microscopic, monolithic construction drastically reduces sensitivity to vibrations and shock. With all these features under its tiny hood, the FFP analyzer has a lot to offer to the gas analysis market. Greenhouse gas abatement requires a multitude of new technologies; we hope this one can make its miniature contribution.

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