Discussion and future work
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8.1 Discussion

Optical imaging beyond the diffraction limit is challenging. Ideally, one would like to have extremely high imaging resolution and measure many properties simultaneously, with a user-friendly instrument that, possibly, could work well also in harsh environments.

Optical fibers are flexible, robust and inexpensive, and, as such, may be considered as the ideal building block for versatile sensors that may address the needs described above. In chapter 3, we showed that, on the basis of this idea, one can design a fiber-based probe that can be used for simultaneous AFM and SNOM analysis. This approach allows for combined structural and near-field optical characterization, which improves the imaging speed and makes data correlation easier. Our fiber-based integrated probe, dubbed as “ferrule-top AFM+SNOM cantilever”, minimizes drifts, makes measurements in liquids or other difficult environments possible without the need for special precautions, and benefits from an unprecedented plug-and-play design.

In chapter 4, 5 and 6 we then reported a series of experiments that show the advantages of ferrule-top AFM+SNOM probes. In chapter 4 we demonstrated how these probes can be used for AFM+SNOM analysis in air and in water with an optical resolution of 160 nm. Chapter 5 elaborates on the possibility to work in critical environments. Finally, in chapter 6, a ferrule-top AFM+SNOM probe is used in VCSEL reliability experiments.

The probes used in these chapters have a very sharp tip for the best possible resolution. However, this limits the light throughput and may hamper the applicability in chemical analysis. The sharper the tip, the lower the transmission. For vibrational spectroscopic methods, such as Raman spectroscopy, a larger tip aperture is required resulting in a spatial resolution that is not much better than available with far-field methods using short excitation wavelengths. In addition, most scanning methods are relatively slow and designed for small sample areas.

To take a next step in combining AFM and spectroscopic analysis, in chapter 7 we introduced a method to screen a large sample area for reactivity using a combination of AFM and Raman spectroscopy. In this method the high-resolution topography data obtained by the AFM and the spectroscopic information of the Raman microscope is decoupled. Through data analysis techniques, both streams of information can be combined and correlated afterwards. Scanning a large area of 100 x 100 µm² in a relatively short time and obtaining high-resolution morphological data and reactivity information is therefore possible. Our method allows these measurements to be finished within a few hours, which would take multiple days using SNOM (if the
Thus, decoupling morphological and optical imaging improves sensitivity and speed, and reduces costs since conventional instruments can be used that might be already available in the lab. However, it also might complicate data analysis and sample preparation to be able to correlate both streams of information.

In conclusion, we have presented a set of methods that can be used to apply nanoscale spectroscopy in special conditions. The use of ferrule-top AFM+SNOM probes enables high-resolution optical imaging in different environments, such as elevated temperatures or liquids, without the need for special precautions. Chapter 7 shows a different approach to obtain high-resolution topographic and spectroscopic information by decoupling both techniques. In future work these methods could be combined into one technique that is able to cover all the advantages of both approaches.

### 8.2 Future work

In this work, some methods have been presented that lead to optical analysis at the nanometer scale. The research described in this thesis mainly focuses on odd situations that are difficult to tackle with conventional methods. Measurements in liquids or at elevated temperatures always complicate the analysis, as well as combining information from different instruments at sub-micrometer precision. Ferrule-top and fiber-top technology provides an excellent tool for (optical) surface analysis in harsh conditions. Unfortunately, the approach illustrated has still some limitations, the most relevant of which are:

1. Complexity and cost of probe fabrication;
2. Limited sensitivity;
3. Limited imaging speed.

The fabrication process for example is still complicated and expensive due to the required FIB milling step. In section 3.4.2 a first step towards cost reduction and simplification was proposed. Avoiding FIB milling would reduce production cost and complexity significantly. The spin-off company Optics11 already commercialized ferrule-top technology for nano-indentation and atomic force microscopy, showing that it is possible to make probe fabrication commercially attractive. Eliminating the FIB cut, we could probably develop similarly cost effective AFM+SNOM probes.

In addition, light throughput remains fundamentally limited by the size of the aperture, and therefore limits the sensitivity and spatial resolution. Switching to...
apertureless SNOM, such as tip enhance Raman spectroscopy (TERS), might be a solution, although this approach may introduce other challenges.

In chapter 7 a method to separate the structural and chemical analysis is introduced. This screening technique is designed to image a large sample area and obtain chemical information about a large number of nanoscale hotspots simultaneously. The technique could be extended with high-resolution SNOM imaging at selected locations. Some efforts in that direction have been made, but did not show significantly improved resolution yet.

Apart from the details illustrated above, it is probably undeniable that the most fascinating challenge in this research line is the integration of the large area screening with a fiber-based solution that could take the advantages of all the methods described in this thesis.