Summary

Scattering-type near-field optical microscopy (s-SNOM) is a novel method to study nanoscale systems with spatial resolution well below the diffraction limit. Although such devices are commercially available, it still requires significant expertise from both technical and theoretical side to confidently utilize its capabilities in state-of-the-art research. In my thesis I introduced the concept of s-SNOM and the details on how to use it to characterize carbon nanotubes and how to acquire additional information compared to classical microspectroscopy. As I showed, the contrast mechanism is more complex than in conventional microscopy and the interpretation of the s-SNOM signature of nanoparticles has to be conducted with care due to multiple sources of artifacts. Supplemented by quasi-electrostatic models, we can use s-SNOM to identify different component of the samples with 20 nm resolution even at infrared wavelengths.

I showed that s-SNOM can be successfully applied to unambiguously distinguish individual carbon nanotubes based on their metallicity. Semiconducting nanotubes give ignorable near-field phase contrast compared to metallic ones, due to their low charge carrier concentration. I validated this finding with the help of the extended finite dipole model (EFDM) model which qualitatively describes the contrast of the nanotubes even with dielectric functions acquired from bulk measurements.

Based on the facts that s-SNOM is extremely sensitive to local metallic components of the sample and the semiconducting nanotubes only provide low phase contrast, I applied s-SNOM to observe the formation of nickel clusters inside carbon nanotubes. I could identify the nano-sized metal clusters and found quantitative agreement of the near-field phase spectrum with model calculations based on my modified EFDM model. I found that most of the nanotubes express phase signatures of nickel clusters which corresponds to high filling ratio, also supported by electron microscopy measurements.

Afterwards, I turned to more complex and more exotic phenomena to study quantum plasmons in metallic carbon nanotubes based on the fact that optical near fields can support high in-plane momentum required to launch propagating surface waves. By realizing scanning polariton interferometry I studied the interaction of nanotube plasmons and substrate phonons. I imaged the standing wave patterns of the forward propagating and back-reflected plasmons and retrieved their dispersion relation. I found a surprisingly effective hybridization between nanotube plasmons and phonon polaritons. The new hybrid plasmons-phonon states express significant mode splitting and a coupling strength reaching the ultrastrong coupling regime. I corroborated my findings with the coupled harmonic oscillator model to both fit the dispersion curves and also the phase contrast spectrum.

The first part of my thesis uncovers the power of s-SNOM as a characterization method for carbon nanomaterials, while the second part truly highlights that carbon nanotubes still hold uncovered potential in the field of nanophotonics. Our results provide evidence to further examine carbon nanotube plasmons for enhanced vibrational spectroscopy of small molecules attached to or encapsulated into carbon nanotubes.