1. PHD PROJECT DESCRIPTION (4000 characters max., including the aims and work plan)

Project title: Plasmon Enhanced Molecular Spectroscopy with Optical Frequency Combs

1.1. Project goals

The goal of the project is to combine the ultrahigh spectrotemporal resolution and broadband operation offered by optical frequency combs with spatial confinement provided by plasmonic nanoparticles. The ultimate goal is the realization sensitive molecular spectroscopy at extremely low concentrations.

1.2. Outline

Optical frequency combs enable high-precision spectroscopic measurements combined with high frequency and temporal resolution, the latter in the tens of microseconds range, due to the relative phase stability of tens of thousends of their spectral components [1,2].

Metallic nanostructures supporting plasmon polaritons are able to concentrate electromagnetic fields into subwavelength spatial domains [3]. For this reason, they can be exploited to enhance light-matter interaction strength. A molecule positioned in plasmonic hot spots interacts with significantly enhanced fields, and may considerably modify the optical properties of the nanoantenna [4]. This influence is critically depenent on the molecular position and orientation, meaning that plasmonic nanoparticles can be used as sensors capable to localize a single-molecule with a nanometre precision [5].

Recently, the two technologies have been combined allowing insights into modulation dynamics of nanoparticles with spatial precision at the Angstrom level [6]. In this project, our goal is to exploit further this combination for highly sensitive spectroscopy of molecules, based on the precise phase information provided by the comb together with the strong resonant enhancement of nanoparticles.

1.3. Work plan

- **12 months: Nanostructured sample characteristics**

simulations of near field distributions and scattering spectra for basic plasmonic nanoparticle geometries, design optimization in the context of experimental requirements.
exemplary tools: COMSOL Multiphysics, CST Studio Suite, predeveloped Matlab scripts

- **12 months: Preliminary experimental steps**

Prepariation and test runs of an experimental setup designed to include nanostructures and gas/liquid samples.

- **12 months: Experiment with nanostructured samples**

Frequency-comb-based spectroscopy experiment on samples designed in the first year of the project.

- **12 months: Additional problems and thesis preparation**

1.4. Literature

[1] *Nobel Lecture: Passion for Precision,* T.W. Hänsch, Reviews of Modern Physics 78, 1297 (2006).
[2] Broadband Optical Cavity Mode Measurements at Hz-Level Precision With a Comb-Based VIPA Spectrometer, G. Kowzan et al. Sci. Rep. 9, 8206 (2019).
[3] *Nanoantennas for visible and infrared radiation, P.* Biagioni, H. Jer-Shing, B. Hecht. Reports on Progress in Physics 75, 024402 (2012).
[4] *Strong coupling of optical nanoantennas and atomic systems,* K. Słowik, R. Filter, J. Straubel, F. Lederer, C. Rockstuhl, Physical Review B, 88, 195414 (2013).
[5] *Optical detection of single non-absorbing molecules using the surface plasmon resonance of a gold nanorod*, P. Zijlstra, P. Paulo, and M. Orrit. Nature Nanotechnology 7, 379 (2012).
[6] *Plasmonic dynamics measured with frequency-comb-referenced phase spectroscopy* Anh, Nguyen Duy, Byung Jae Chun, Sungho Choi, Dong-Eon Kim, Seungchul Kim, and Young-Jin Kim, Nature Physics (2019).

1.5. Required initial knowledge and skills of the PhD candidate

- Masters degree in physics or related fields (to be acquired by October 2020).
- Basics of quantum mechanics and atomic physics.
- Optional but welcome: experience in laboratory works or electromagnetic modelling.
- Good level of spoken and written English.

1.6. Expected development of the PhD candidate's knowledge and skills

The candidate will acquire and master their knowledge on:
- classical electromagnetism, in particular aspects of nanophotonics,
- atomic and molecular optics,
- methods of high-precision spectroscopy.

The candidate will acquire the following skills:
- modelling of the spectral response of plasmonic nanoparticles,
- spectroscopic measurements of molecular samples,
- experimental data analysis.

The candidate will get experience with commercial numerical tools widely used in academia and industry, as well as with the femtosecond light sources used in spectroscopy and frequency metrology.