λ/2 Fabry Pérot micro-resonators in single molecule spectroscopy

Alfred J. Meixner[1,*], *Frank* Wackenhut[1], *Alexander* Konrad[1], *Michael* Metzger[1], and *Marc* Brecht[1,2]

[1]Eberhard Karls University, Institute of Physical and Theoretical Chemistry, 72076 Tübingen, Germany
[2]Reutlingen University, Reutlingen Research Institute, Process Analysis and Technology (PA&T), 72762 Reutlingen, Germany

Abstract. Embedded in a tuneable λ/2-FabryPérot micro-resonator the radiative relaxation of a dye molecule or quantum dot can reproducibly be modified allowing to determine their quantum yield, control Förster energy-transfer or localize them with nanometer precision.

1 Introduction

Small optical micro-resonators are structures that confine light to volumes with dimensions on the order of a wavelength and hence have become increasingly important for controlling and studying light-matter interaction in integrated optics [1]. The spontaneous emission rate of an atom or molecule depends on both the transition dipole moment (TDM) between the electronically excited state and the electronic ground state as well as on the density of optical modes of the electromagnetic field in the surrounding medium. In an optical subwavelength micro-resonator with piezo-controlled mirror separation the density of optical modes can be varied with respect to the free space in a controlled manner, and hence, also the radiative coupling of the dipole transition of the emitter to the optical field can be tuned.

2 Results

We demonstrate controlled modification of the vibronic relaxation dynamics of a single fluorescent molecule fixed in a solid host matrix by varying the distance between the cavity mirrors. While it is well known from quantum electrodynamics nowadays that the spontaneous emission is not an intrinsic property of an atom but can be modified by tailoring the electromagnetic environment, the situation for a molecule or quantum dot embedded in a solid host is more complex since the radiation rate and relaxation pathways depend on the balance between the radiative (the far-field) and non-radiative (the near-field) relaxation dynamics, the latter being sensitively dependent on the interaction with the local chemical environment. Hence, by controlling the electromagnetic environment of a single molecule or quantum dot, we can select the vibronic transition where fluorescence will mostly occur [2] and we can tune its radiative transition and determine its fluorescence quantum yield [3] as shown in Fig. 1).

---

[*] Corresponding author: alfred.meixner@uni-tuebingen.de

Using a tuneable optical micro-resonator with subwavelength spacing, we demonstrate controlled modulation of the radiative transition rate of a single molecule, which is measured by monitoring its fluorescence lifetime [4]. By comparing the experimental data with a theoretical model, we extract both the pure radiative transition rate as well as the quantum yields of individual molecules. We observe a broad scattering of quantum yield values from molecule to molecule, which reflects the strong variation of the local interaction of the observed molecules with their host environment. Finally, we will also discuss the possibility of controlling Förster resonance energy transfer in a tuneable micro-resonator [5, 6, 7] and nanometer precise localization [8].

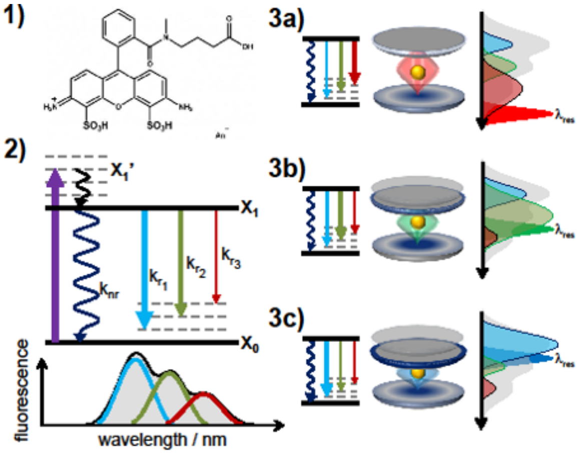

Fig. 1. Adopted from ref. [5] (1) Chemical structure of a typical dye molecule. (2) Jablonski-like diagram with an electronic ground state X_0 and excited state X_1. After pulsed excitation (violet arrow) into the excited vibronic level X_1' the system relaxes to the purely electronic level X_1. The electronic relaxation occurs either non-radiatively or radiatively into possible vibronic levels of the electronic ground state. Each fluorescent transition can be described by a spectrally broadened Gaussian band due to thermal energy level fluctuations and phonon interactions with the environment. (3a–3c) Tuning the micro-resonator with the on-axis spectral emission to the emission wavelength of a specific vibronic transition enhances resonant and suppresses off-resonant radiative transitions.

References

1. H. Yokoyama, K. Ujihara, *Spontaneous emission and laser oscillation in microcavities* (CRC Press, 1995)
2. A. I. Chizhik, F. Schleifenbaum, R. Gutbrod, A. M. Chizhik, D. Khoptyar, A. J. Meixner, J. Enderlein, J. Phys. Rev. Lett., **102**, 073002 (2009)
3. A. I. Chizhik, A. M. Chizhik, D. Khoptyar, S. Bär, A. J. Meixner, J. Enderlein, Nano Lett., **11**, 1700 (2011)
4. A. I. Chizhik, A. M. Chizhik, A. M. Kern, T. Schmidt, K. Potrick, F. Huisken, Phys. Rev. Lett., **109**, 223902 (2012)
5. F. Schleifenbaum, A. M. Kern, A. Konrad, A. J. Meixner, et al. PCCP, 16, 12812 (2014)
6. A. Konrad, M. Metzger, A. M. Kern, M. Brecht, A. J. Meixner, Nanoscale, **7**, 10204 (2015)
7. A. Konrad, A. L. Trost, S. Skandary, M. Hussels, A.J. Meixner,N. V. Karapetyan, M. Brecht, Phys. Chem. Chem. Phys., **16**, 6175 (2014)
8. R. Gutbrod, D. Khoptyar, M. Steiner, A. M. Chizhik, A. I. Chizhik, S. Bär, A. J. Meixner, Nano Lett., **10**, 504 (2010)