Stable single molecules for quantum optics and all-optical switches

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Summary

The field of single-molecule spectroscopy and microscopy at room temperature relies mainly on the dilution of a dye in a solid or solvent such that only one fluorescent molecule is expected within the diffraction-limited spot size. At room temperature the absorption cross section is low because the excitation line is broadened due to electron decoherence. However, when the molecule in a non-interacting solid host is cooled down to 1.5 K (as was done in the research described in this thesis), the suppression of decoherence processes leads to a narrowing of the excitation line (down to the lifetime-limited or Fourier-limited width) and to an increased absorption cross section. Finally, because the purely electronic transition of the molecule occurs without the creation or annihilation of phonons (zero-phonon line), the single molecule can be described as a single-quantum two-level system.

Part of this thesis concerns the spectroscopic properties (the distribution of excitation linewidths, the frequency stability, the saturation profile) of single dibenzoterrylene (DBT, chapter 2), terrylene (chapter 3) and perylene (chapter 6) molecules embedded in different solid hosts in order to characterize these new host-guest systems for single-molecule spectroscopy. By looking at the distribution of the excitation linewidths of selected single molecules, obtained at zero intensity, we correlate the experimental observations with physical phenomena in the local environment of the single fluorophore.

From recording intensity-time traces from a single molecule and by looking at the corresponding auto-correlation functions, we were able to study dynamic processes that occur in the surrounding of the dye molecule at different time scales. Results of such studies are described in chapters 2 and 5.

Further in chapter 5, we use the lifetime-limited linewidths of DBT in an anthracene crystal to perform experiments on a host-guest system that is externally disturbed in a controlled way. The resonant excitation line serves as an ultra-sensitive vibrational sensor to detect the tiny oscillations of an electrically driven quartz tuning fork. Moreover, the stable single molecules reported in this thesis are suitable isolated quantum systems, which could be used as single photon sources, as optical transistors, as all-optical switches based on non-linear optical phenomena and for optical read-out of the quantum state of nano-mechanical oscillators.

In order to prepare for a proposed experiment on a two-color optical transistor, we need the transition energy from the ground singlet state to the excited triplet state. For perylene embedded in anthracene this transition was measured at 778 nm. For terrylene this transition has never been observed because of the low probability of intersystem crossing. In the work reported in chapter 4, we tried to induce the singlet to triplet transition in this molecule by using an electron beam instead of optical excitation. Besides electron-energy-loss spectroscopy, other techniques were used to study the vibrational and electronic properties of terrylene. Despite all efforts, the energy of the triplet state of terrylene remains unknown.