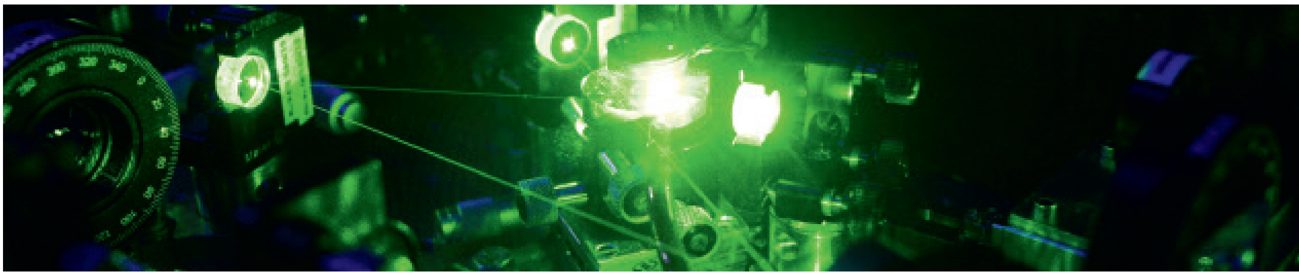

Ideas taking shape – worldwide.

Single molecule spectroscopy using tunable lasers

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The fluorescence excitation spectra of single organic molecules in a solid state crystal are measured at cryogenic temperatures using a single frequency tunable laser light source based on optical parametric oscillator technology. This laser exhibits promising features as a light source for spectroscopy applications, including a broad tuning range from 450 to 650 nm, narrow linewidth < 1 MHz and mode-hop-free tuning over > 25 GHz. This applications note presents the experimental setup, measured spectra and discusses the applicability of this kind of laser for high-resolution spectroscopy.

Introduction

Investigations of molecular processes and their characteristics play an important role in many scientific disciplines, like medicine, biology, chemistry, and physics. Fluorescence spectroscopy of single molecules gives fundamental insights into their properties and the influence of their surroundings [1,2]. However, studying single molecules in a host material is a challenging task with high demands on the detectors and on the laser light sources [3]. This report examines the applicability of a new tunable continuous-wave (cw) laser light source for various spectroscopy methods.

Experiment

The experimental setup is illustrated in Fig. 1. A frequency-tunable laser beam is focused onto a sample containing dibenzanthanthrene (DBATT) molecules hosted in a naphthalene crystal. The laser beam is tuned to resonances of the DBATT molecules, leading to their excitation and subsequent fluorescence. The fluorescence light is filtered and measured with a single photon detector, SPCM-AQR from Perkin Elmer. A High-Finesse WS/6-200 wavemeter monitors the frequency of the excitation laser. A more detailed description of the experimental methods is given, e.g. in [3].

Suitable excitation lasers

Single molecules in a solid state crystal can be regarded as nearly ideal two-level systems with natural linewidths in the range of 10 - 50 MHz. Due to imperfections in the crystal the transitions of individual molecules are inhomogeneously distributed over more than 1 THz. With a narrow-band laser tuned to an individual transition single molecules can be isolated from the ensemble. Therefore, the laser is a crucial component for the excitation of single molecules: It requires a linewidth below the natural linewidth of the molecules and a mode-hop-free tunability over a broad spectral range to address many molecules. Furthermore the flexibility in choosing the center wavelength for different kinds of molecule-host combinations, an output power well above 200 mW, and low intensity noise are other relevant parameters. Commonly, dye lasers, Ti:Sa lasers, or tunable diode lasers are used for such experiments. Ti:Sa lasers are restricted to the wavelength ranges 700 – 1000 nm and 350 – 500 nm when frequency-doubled. Dye lasers require a change of the dye or even of the pump laser used, when switching between wavelength ranges. Diode lasers are typically restricted to small tuning ranges and low output powers in the visible wavelength range. Optical parametric oscillators (OPO) offer an attractive alternative: The wavelength coverage of OPOs can be designed according to the experimental requirements, OPOs are solid-state systems which do not rely on consumables such as dyes, and they deliver relatively high output powers.

In this applications note, the commercially available cw tunable OPO, C-WAVE, was used. This OPO covers the wavelength ranges 450 – 650 nm (frequency doubled SHG option) and 900 – 1300 nm. The output power is in the 500 mW range and the linewidth is below 1 MHz. C-WAVE can be swept mode-hop-free over more than 25 GHz. Changes of the center frequency of several GHz as well as large variations of the wavelength are fully computer controlled. The good beam profile allows high coupling efficiencies into optical fibers and therefore a simple integration into existing setups.

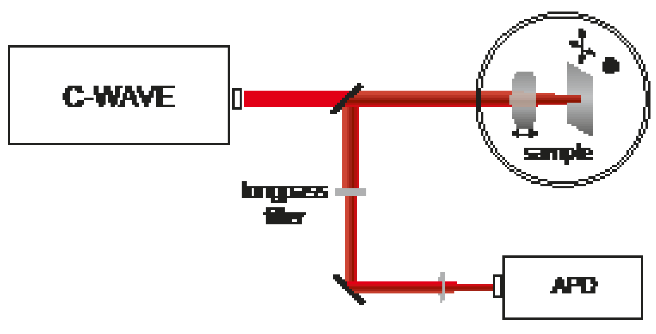

Figure 1. Experimental setup: Confocal microscope including a cryostat. A fiber guides the laser light to the experimental setup where it is cleaned up with filters and focused onto the sample. The red-shifted fluorescence light is collected, filtered, and analyzed with an avalanche photodiode (APD).

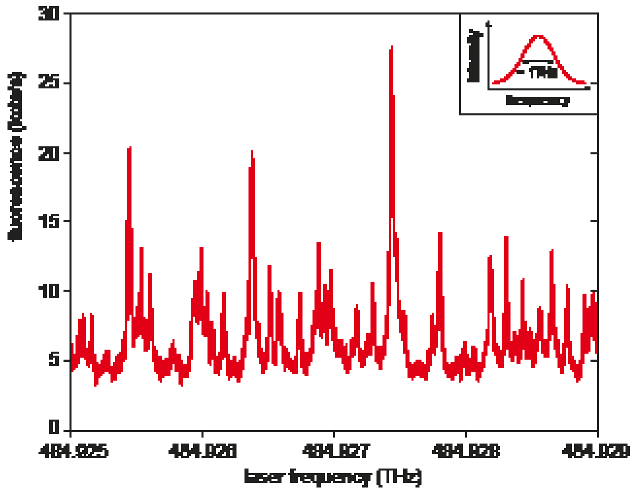

Figure 2. Fluorescence intensity of DBATT molecules hosted in a naphthalene crystal versus excitation frequency. Inset: Schematics of the inhomogeneous broadening of DBATT molecules with a linewidth of 1 THz.

Results

Fluorescence spectra of DBATT molecules have been measured at a center wavelength of 618 nm and over a range of 1 THz by stitching several mode-hop-free frequency sweeps. Figure 2 shows a zoom in one of the mode-hop-free scans. The molecules' redshifted fluorescence signal is measured with the APD while the laser frequency is scanned. The measurement shows several narrow spectral features corresponding to individual DBATT molecules [4]. With C-WAVE it is also possible to lock the laser frequency with wavemeter-precision to one single molecule resonance in order to study its photo-physics and photon dynamics.

Summary

The presented setup enables the measurement of fluorescence excitation spectra of single molecules in transparent host materials. Both laser light sources, Ti:Sa lasers and C-WAVE, are well suited for measuring narrow spectral features over wide frequency ranges. A combination of these laser sources allows a nearly complete wavelength coverage from 450 – 1300 nm with fully automatic wavelength control and no need to change laser media or perform realignments. This makes the presented setup a user friendly, flexible and sensitive spectrometer for characterizing single molecules, color centers and semiconductor quantum dots.

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