

Optimized Distributed Feedback Dye Laser Sensor for Real-Time Monitoring of Small Molecule Diffusion

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Abstract: Nanoimprinted distributed feedback dye laser sensors featuring multilayer slab waveguides are presented. A simple yet precise analytical model is used to optimize the lasers in order to give highest sensitivity and it is found that the thickness of a high index TiO₂ top layer is the most important parameter for optimization. Using such laser sensors in an imaging spectroscopy setup, real-time label-free monitoring of sugar molecule diffusion in water is demonstrated. This method could potentially pave the way towards the analysis of small molecule diffusion in various media, e.g. protein signaling processes in tissue.

Organic dye-based distributed feedback (DFB) lasers have gained significant interest as widely tunable laser light sources in the visible wavelength regime. They are inexpensive and feature simple fabrication, low threshold, and single-mode emission.¹ Recently, they have been found to be highly sensitive refractive index and label-free bio-sensors where a change in the cladding refractive index results in a laser wavelength shift which is used as sensor signal.^{2,3} Label-free sensing is very promising for the measurement of diffusion properties of small molecules in different media. These properties could not be measured with label-based methods as the labels would affect the diffusion. On a longer term, label-free monitoring of signaling proteins in tissue could be achieved this way leading to deeper understanding of the pathways that exist in tissue.⁴

Figure 1a shows a scheme of a 2nd order DFB dye laser sensor featuring a Pyrromethene 597 doped Ormocomp layer structured by UV nanoimprinting with a grating period of $\Lambda = 384$ nm.⁵ In addition, the structure exhibits a thin TiO₂ top layer which is deposited by ion beam evaporation and significantly enhances the sensitivity of the sensor. This is proven by simulations with a simple yet precise model for calculating the emission wavelength of multilayer DFB lasers.⁶ In contrast to choosing the DFB emission wavelength and solving for the corresponding propagation constant, here the propagation constant, β , is chosen to be $\beta = m\pi/\Lambda$, where m is the Bragg order. Thus, a more straightforward calculation yields the emission wavelength directly and the approach avoids several common approximations.

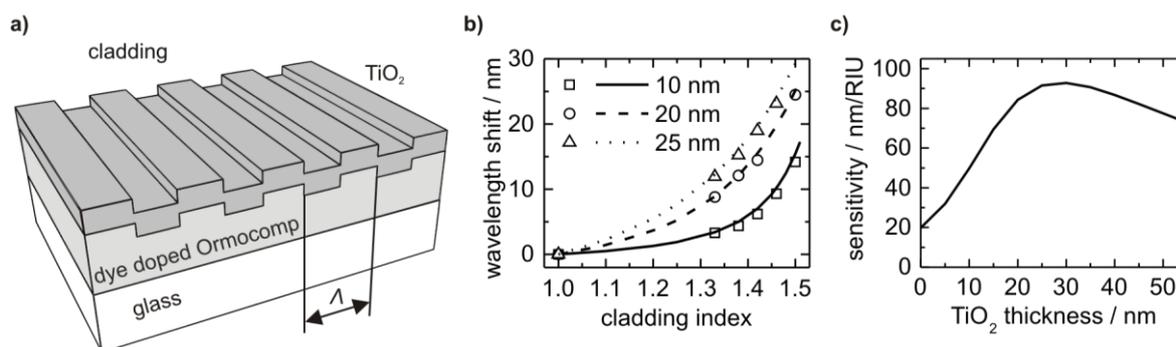


Fig. 1: a) Schematic of DFB dye laser sensor. b) Measured (symbols) and calculated (lines) wavelength shift for varying TiO₂ thickness. Ormocomp thickness is 400 nm. c) Calculated sensitivity as function of TiO₂ thickness.

Figure 1b shows the simulated and measured emission wavelength shift of DFB lasers with TiO₂ thickness of 10 nm, 20 nm, and 25 nm. Very good agreement between experimental data and simulations is evident. Thus, using this model is valid for optimizing the sensitivity of the lasers which is defined as wavelength change per refractive index unit. The plot in Fig. 1c shows the sensitivity as function of the TiO₂ layer thickness. It can be seen that the sensitivity can be improved

by a factor of 5 in comparison to a laser without TiO₂ top layer and that there is an optimal thickness of approx. 30 nm. This way, a detection limit of $8 \cdot 10^{-6}$ RIU is achieved.⁵

With these optimized lasers real-time monitoring in time and one dimension of space of sugar molecules diffusion in water is feasible. In order to achieve this, a microscope setup with an imaging spectrometer^{7,8} is used, see Fig. 2a. Here, the green pump laser light (ns pulses at 532 nm) is focused onto the DFB dye laser sensor. The laser emits laser light of longer wavelength. For analysis the pump light is blocked by a dielectric mirror and the DFB laser light is used to project an image of the device surface onto a CCD for position control, Fig. 2b. Via a beamsplitter, the same image of the device surface is projected onto the slit of an imaging spectrometer (Acton SP-2756 imaging spectrograph with PIXIS00B digital CCD camera). The laser is oriented such that the imaged grating lines are parallel to spectrometer slit. Analysis of the spectrometer CCD image gives the emission wavelengths as function of the y-position where the spectrum of each row is analyzed and the emission wavelength is found with a center-of-mass approach.⁵ 12 CCD images per second are taken. Each image gives one column with emission wavelengths for 100 y-positions corresponding to 100 rows on the CCD. In order to prove the functionality of the setup, a fluidic well on top of the laser is filled with water and a small piece of sugar is added. Thereupon, the sugar is dissolved in the water and the refractive index of the solution increases. The surface plot in Fig. 2c shows how the wavelength is changing as the sugar concentration is increasing. It is expected that this method can be used to analyze the diffusion of small molecules in various media on the milli- and micrometer scale and in real-time.

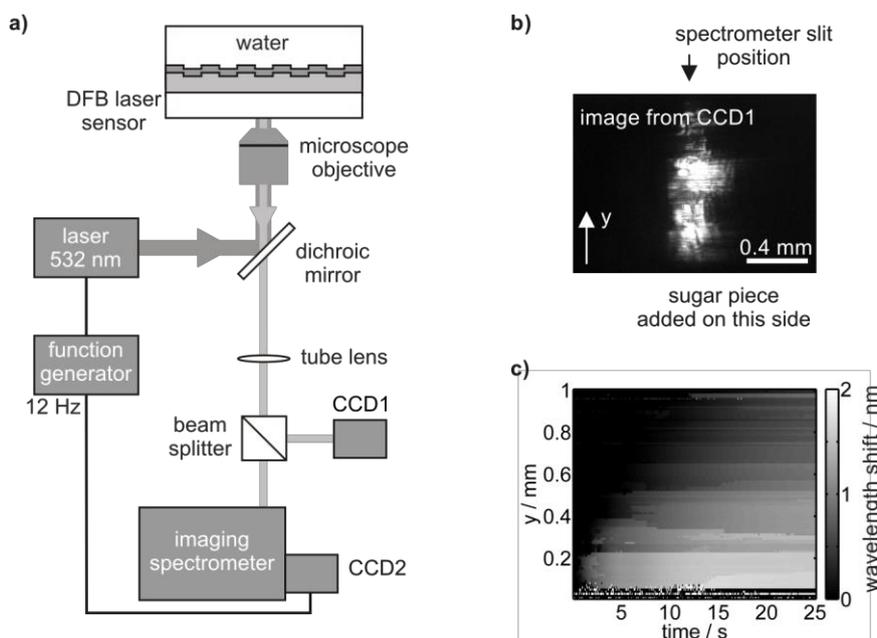


Fig. 2: a) Schematic of setup for real-time monitoring of sugar diffusion in water with DFB laser sensor. b) Image of laser emission on the surface indicating the position of the spectrometer slit. c) Wavelength shift occurring when a piece of sugar is added to water plotted as function of y-position and time as obtained from the imaging spectrometer data.

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