

Analysis of Molecular Transformations and Light Manipulation with Optical Microresonators

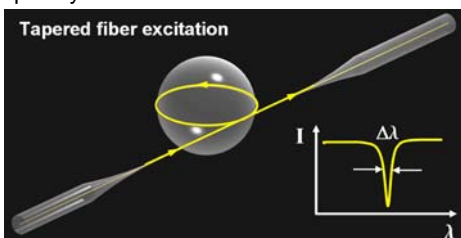
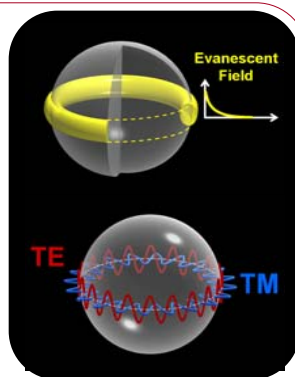
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Optical Microcavities

- High-Q whispering gallery modes (WGMs) ($Q \sim 10^6$).
- Highly sensitive to refractive index (polarizability) perturbations.
- Evanescent field probing of ultra-thin molecular layers.
- Two probing polarizations (TE and TM). Capable of probing optical anisotropies in molecular self-assemblies.
- Sensitive to molecular binding events and changes within the structure of immobilized molecules.
- Binding events and conformational changes conveyed as shift in resonant frequency of WGMs.



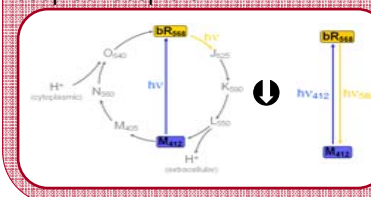
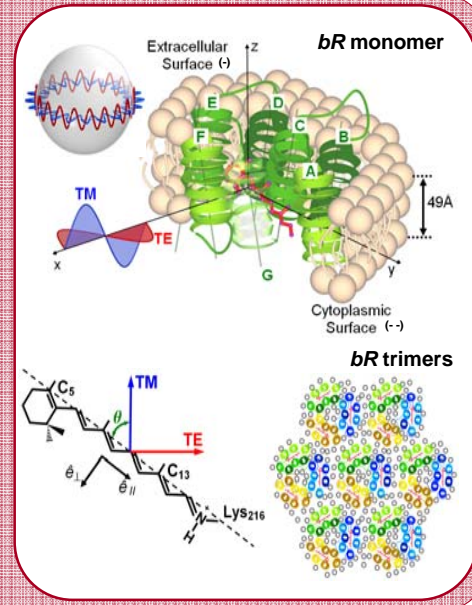
Resonant shift:

$$\frac{\Delta\lambda}{\lambda} \propto \frac{\sigma_{mol} \alpha_{mol}}{R}$$

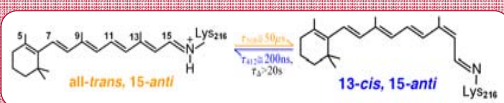
σ_{mol} : molecular surface density
 α_{mol} : molecular polarizability
 R: resonator size

Bacteriorhodopsin

- Model biological membrane system.
- Known 3D structure. Highly oriented, optically anisotropic system. Large polarizability changes upon isomerization.
- Self-assembles as a monolayer.
- Technologically important biological photochrome. Applications in photonics and optoelectronics.
- Shares structure with G-protein coupled receptors.



Trans-cis isomerization and orientation of the retinal



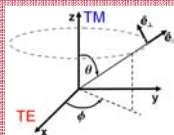
- Photoinduced *all-trans* to *13-cis* isomerization of the chromophore retinal monitored off-resonance with a 1,310nm probe.
- Novel pump-probe spectroscopy tool.
- Direct measurements of polarizabilities of molecules in complex proteo-lipid environments.

Average retinal polarizability change:

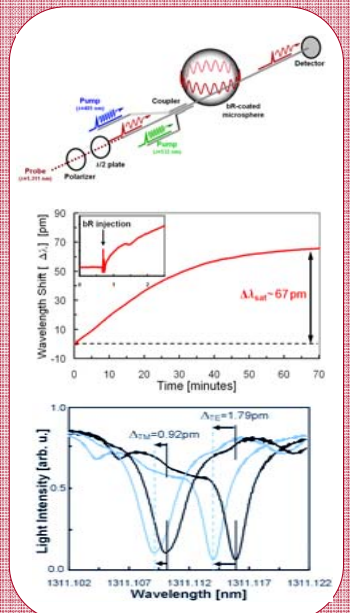
$$\langle \Delta\alpha_{iso} \rangle = \frac{1}{3} (\langle \Delta\alpha \rangle_{TM} + 2\langle \Delta\alpha \rangle_{TE}) \cong -384 \text{ au}$$

- Direct orientation measurements of molecular self assemblies

TE/TM resonant shift ($\beta = \frac{\partial\alpha_x}{\partial\alpha_z} \approx -10, \langle \theta \rangle = 61^\circ$):



$$\frac{\Delta\lambda_{TE}}{\Delta\lambda_{TM}} \approx \frac{1}{2} \left(\frac{1 + \cos^2\langle\theta\rangle + \beta \sin^2\langle\theta\rangle}{\sin^2\langle\theta\rangle + \beta \cos^2\langle\theta\rangle} \right) \approx 2$$



All-optical switching in the near-infrared

- All-optical resonant coupler.
- Optical functionality provided by ultra-thin molecular layer.
- Operation at frequencies far from *bR* molecular transitions.
- Full-linewidth shift achieved by multiple *bR* layering (*bR*/PDAC).
- Switching speed limited by the speed of the photochromic transitions ($\sim 50\mu\text{s}$).
- Faster all-optical switching at arbitrary optical frequencies with molecular monolayers is possible.

