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# Strong light-matter coupling for optical switching through the fluorescence and FRET control

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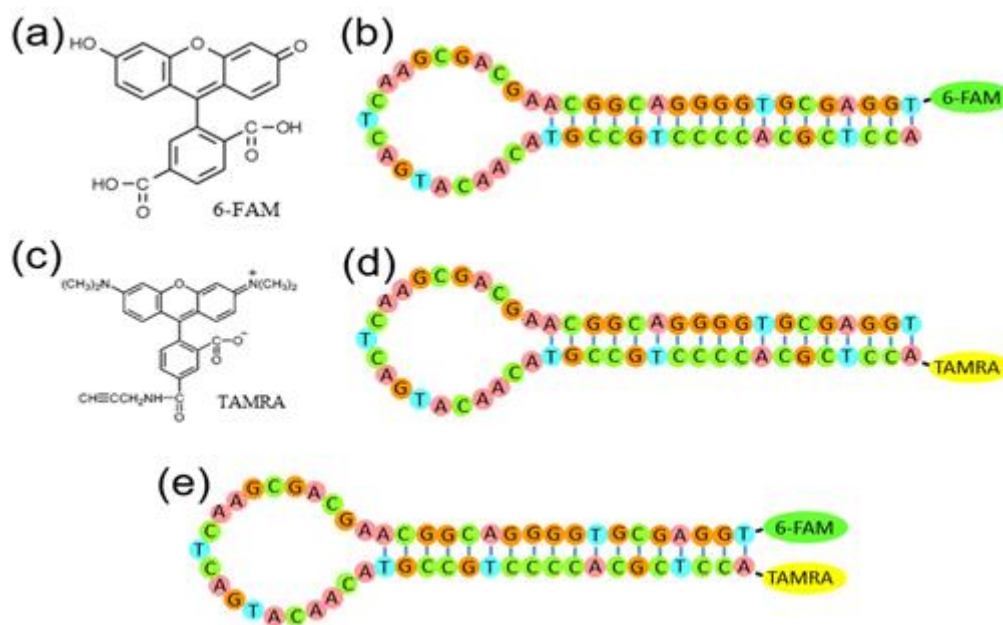
**Abstract.** Resonant interaction between excitonic transitions of molecules and localized electromagnetic field forms the hybrid polaritonic states. Tuneable microresonators may change the light-matter coupling strength and modulate them from weak to strong and ultra-strong coupling regimes. In this work we have realised strong coupling between the tuneable open-access cavity mode and the excitonic transitions in oligonucleotide-based molecular beacons with their terminus labelled with a pair of organic dye molecules demonstrating an efficient donor-to-acceptor Förster resonance energy transfer (FRET). We show that the predominant strong coupling of the cavity photon to the exciton transition in the donor dye molecule can lead to such a large an energy shift that the energy transfer from the acceptor exciton reservoir to the mainly donor lower polaritonic state can be achieved, thus yielding the chromophores' donor-acceptor role reversal or "carnival effect". The data show the possibility for confined electromagnetic fields to control and mediate polariton-assisted remote energy transfer. Obtained results open the avenues to quantum optical switching and other applications.

## 1. Introduction

The FRET effect only occurs when several conditions are satisfied: (i) the donor emission spectrum should overlap with the acceptor absorption spectrum; (ii) the donor and acceptor fluorophores should be in a favourable mutual orientation, and, (iii) since the FRET efficiency is inversely proportional to the sixth power of the distance between the fluorophores, the distance between the donor and the acceptor should not exceed the Förster limit (10 nm). When these conditions are satisfied, the FRET effect results in a decreased donor fluorescence emission accompanied by a simultaneously increased acceptor fluorescence emission. Under the strong light-matter coupling regime [1], in the tuneable microcavities [2,3], both donor and acceptor excitonic states could be coupled to the same microcavity optical mode, which may act as a mediator. This mediation can make it possible not only to increase the efficient distances of the energy transfer to values ten times larger than the Förster limit (to more than 100 nm) [4] but also to ensure an up to sevenfold increase in the rate of energy transfer [5]. This increase in the energy transfer rate leads to a significant decrease in donor fluorescence in the presence of the acceptor. As a result, the energy transfer efficiency, which is characterized by the ratio between the intensities of the donor fluorescence in the presence and absence of the acceptor, will be also significantly increased. It has been reported recently that under the strong coupling regime the energy transfer efficiency may be increased from 0.55 to 0.90 [5].



In the reporting study [6], we have investigated the dependence of the photoluminescence (PL) properties of the oligonucleotide-based molecular beacons with a donor–acceptor pair of closely located FAM and TAMRA organic dyes in a tuneable microcavity in the strong coupling regime and have analyzed the dependence of the polaritonic state population on the detuning of the optical microcavity.



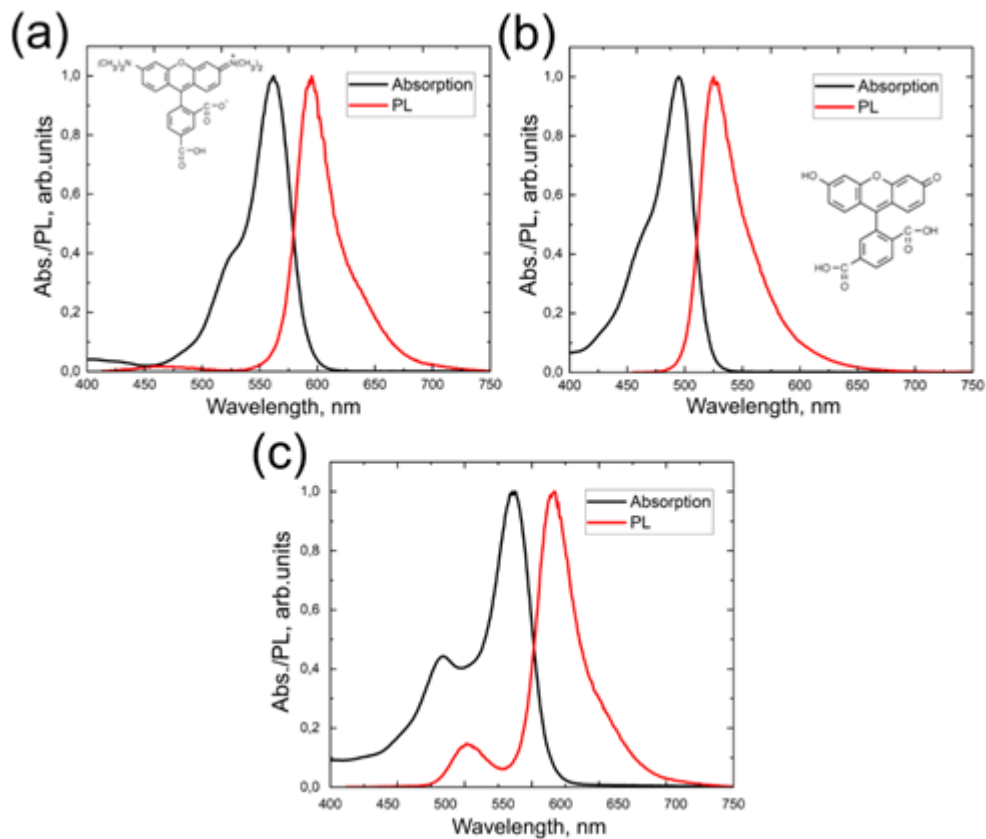
**Figure 1.** The structures of the dyes and molecular beacons under study. The donor (a) and acceptor (c) dye chemical structures. Schematics of the structures of molecular beacons: the donor-only-labelled hairpin (b), the acceptor-only-labelled hairpin (d), and the donor–acceptor double-labelled hairpin (e).

## 2. Materials

6-Carboxyfluorescein (FAM) was selected as a donor dye, and carboxytetramethylrhodamine (TAMRA), as an acceptor dye. The most common way to fix the distance between chromophores required for efficient FRET is to use oligonucleotide-based molecular beacons, because, in this case, the distance between the donor and the acceptor is of the order of the diameter of the DNA double helix, which is 2 nm.

In this study, the donor and acceptor dye molecules were conjugated with a self-complementary oligonucleotide, 5'-TGG AGC GTG GGG ACG GCA AGC AGC GAA CTC AGT ACA ACA TGC CGT CCC CAC GCT CCA-3'.

Donor-only- and acceptor-only-labelled hairpins were also obtained and used as controls (Figure 1). The oligonucleotide sequence of 57 oligonucleotide residues forming a hairpin with an 18-bp stem was so selected as to ensure hairpin stability and a small distance between the donor and the acceptor. The molecular weights of the donor-only-, acceptor-only-, and donor–acceptor double-labelled hairpins were MFAM=17477.47, MTAMRA=17568.68, and MFAM\_TAMRA=17945, respectively. All components were obtained from Evrogen (Evrogen Joint Stock Company, Moscow, Russia) and diluted with phosphate-buffered saline (PBS) to a concentration of 100  $\mu$ M. For experiments, we used 10  $\mu$ L of the solution of each component.



**Figure 2.** The absorption and photoluminescence spectra of the solutions of the TAMRA dye (a); FAM dye (b); FAM and TAMRA dyes (c).

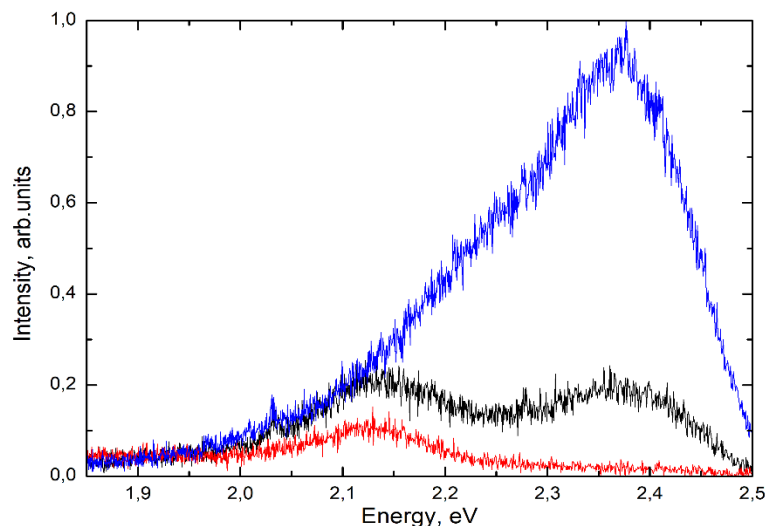
The photoluminescence and absorption spectra of the compounds were first studied outside of the cavity (Figure 2). Here, we used different excitation wavelengths, depending on the absorption spectrum of each compound.

In order to estimate the FRET efficiency via dipole–dipole interaction, we placed the solutions containing the beacons labelled with FAM, TAMRA, and FRET pair alternately on the lower mirror of the microcavity without the upper one and excited them non-resonantly with a 450-nm laser. In this way, we ensured the same experimental conditions as in the experiments with the medium placed inside the cavity. The measured photoluminescence (PL) spectra are shown in Figure 3.

It can be seen from these spectra that FAM-only-labelled hairpins exhibited a high PL intensity (probably due to the high quantum yield of FAM, 97%) with a peak at 525 nm, while the TAMRA-only counterpart exhibited a low-intensity PL. These results correspond to a more efficient excitation of FAM at 450 nm (~20% of the maximum at 495 nm) compared to TAMRA (~2% of the maximum at 546 nm). The hairpins labelled with both the donor and the acceptor exhibited a significant decrease in the donor PL intensity and a simultaneous increase in the acceptor PL intensity, a result that indicates energy transfer from the donor to the acceptor. From these data, the FRET efficiency can be estimated using the following expression:

$$E = 1 - \frac{F_{DA}}{F_D}, \quad (1)$$

where  $E$  is the efficiency of FRET,  $F_{DA}$  and  $F_D$  are the donor fluorescence intensities in the presence and absence of the acceptor, respectively. The above data lead to an estimated FRET efficiency value of 80%.



**Figure 3.** The photoluminescence spectra of the solutions containing molecular beacons labelled with FAM alone (blue), TAMRA alone (red), and both FAM and TAMRA (black). All samples were located outside the microcavity. The photoluminescence was excited at 450 nm. The concentrations of the dye molecules in the sample solutions were about 100  $\mu\text{M}$  in all experiments.

### 3. Results

#### 3.1. Experimental setup

The experimental setup is shown in Figure 4. Briefly, our versatile tuneable microcavity cell (VTMC) is composed of plane and convex mirrors that form an unstable  $\lambda/2$  Fabry–Perot microcavity. One mirror is made convex in order to satisfy the plane-parallelism condition and minimize the mode volume. The plane mirror is mounted on top of a Z-piezopositioner to provide fine tuning of the microcavity length in a range of up to 10  $\mu\text{m}$  with a nanometer precision, and the landing procedure is carried out with a DRV3 high-precision differential micrometer (Thorlabs), which is indirectly connected to the convex mirror.

The alignment of the plane-parallelism point and the sample is performed by moving the convex mirror in the lateral direction by means of an XY precision positioner. A sample is deposited directly onto the plane mirror, which consists of standard (18 $\times$ 18 mm) glass coverslips with a  $\sim$ 35-nm layer of aluminum metallization on their upper side. The VTMC is mounted onto an inverted confocal microspectrometer consisting of an Ntegra-base (NT-MDT) with a 100X/0.80 MPLAPON lens (Olympus) on a Z-piezopositioner, an XY scanning piezostage, and a homemade confocal unit. The fluorescence spectra of all samples were excited with an L450P1600MM 2.1-W, 450-nm laser (Thorlabs) with an LDS5-EC power supply (Thorlabs); for the transmission spectra, an MCWHF2 white LED (Thorlabs) with a homemade optical condenser was used. It should be noted that the laser power was far from saturation in our experiments.

The recording system included an Andor Shamrock 750 monochromator equipped with an Andor DU971P-BV CCD (Andor Technology Ltd.) and two 488-nm RazorEdge<sup>®</sup> ultrasteep long-pass edge filters (Semrock).

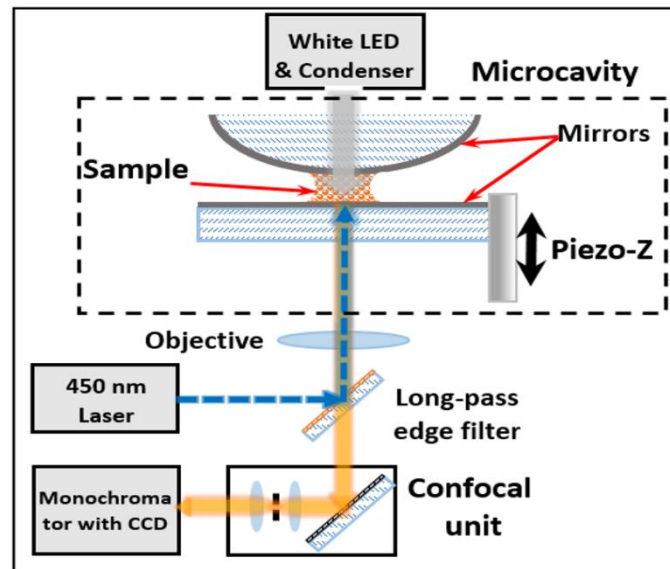


Figure 4. The experimental setup.

### 3.2. Accounting for changes in pumping intensity.

For correct calculation of the lower polariton branch population, it was necessary to account for changes in pumping intensity during the tuning of the cavity length. When the exciting field was in resonance with one of the cavity eigenmodes, a significant rise of the field intensity inside the cavity was observed. To account for this effect, we used a numerical model developed for calculating the spectral and spatial properties of the microcavity electromagnetic modes by the finite elements method. It should be noted that higher transverse modes of the microcavity were also taken into account. This was necessary because, during the excitation, the radiation of the pumping laser was focused by an objective lens with quite a large numerical aperture ( $NA=0.95$ ), and the pumping radiation excited the higher transverse modes (Figure 5a).

In the transmission experiments, the illuminating light had an approximately planar wavefront (Figure 5b); therefore, there were no signs of higher transverse modes in the transmission spectra.

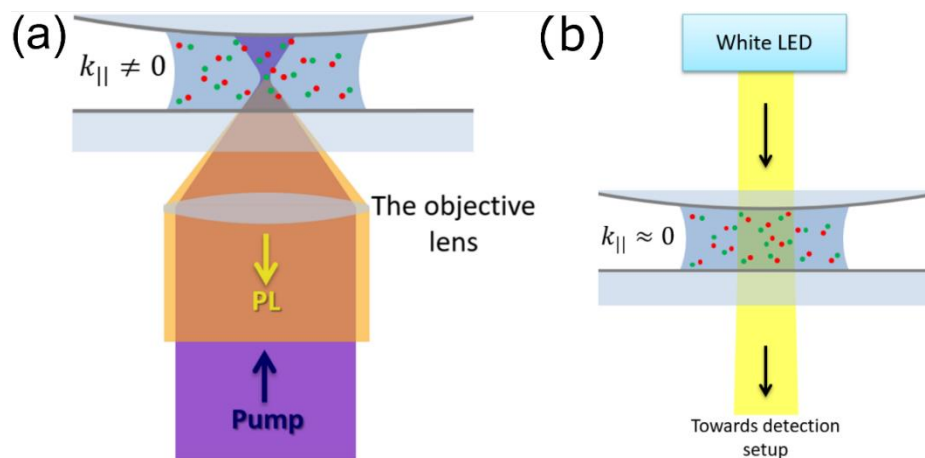


Figure 5. The principal configuration of the excitation (a) and transmission (b) experiments.

#### 4. Conclusion

In the reporting study [6], we have investigated the dependence of the photoluminescence (PL) properties of the oligonucleotide-based molecular beacons with a donor–acceptor pair of closely located FAM and TAMRA organic dyes in a tuneable microcavity in the strong coupling regime and have analyzed the dependence of the polaritonic state population on the detuning of the optical microcavity. These results show the way to realization of the donor-acceptor roles reversal thus opening the perspectives for the highly efficient quantum optical switching realization.

#### Acknowledgements

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#### References

- [1] Dovzhenko D S, Ryabchuk, S V, Rakovich Yu P and Nabiev I R 2018 Light–matter interaction in the strong coupling regime: configurations, conditions, applications. *Nanoscale* **10** 3589.
- [2] Mochalov K E, Vaskan I S, Dovzhenko D S, Rakovich Yu P and Nabiev I 2018 A versatile tuneable microcavity for investigation of light–matter interaction. *Rev. Sci. Instr.* **89** 053105.
- [3] Dovzhenko D, Martynov I, Samokhvalov P, Osipov E, Lednev M, Chistyakov A, Karaulov A and Nabiev I 2020 Enhancement of spontaneous emission of semiconductor quantum dots inside one-dimensional porous silicon photonic crystals. *Optics Express* **28** 22705.
- [4] Zhong X, Chervy T, Zhang L, Thomas A, George J, Genet C, Hutchison J A and Ebbesen T W 2017 Energy transfer between spatially separated entangled molecules. *Angew. Chem. Int. Ed.* **56** 9034–9038.
- [5] Zhong X, Chervy T, Wang S, George J, Thomas A, Hutchison J A, Devaux E, Genet C and Ebbesen T W 2016 Non-radiative energy transfer mediated by hybrid light-matter states. *Angew. Chem. Int. Ed.* **55** 6202–6206.
- [6] Dovzhenko D, Lednev M, Mochalov M, Vaskan I, Rakovich Y, Nabiev I 2020 Polariton-assisted donor–acceptor role reversal in resonant energy transfer between organic dyes strongly coupled to electromagnetic modes of a tuneable microcavity. arXiv:2009.03429 [cond-mat.mes-hall]