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Plasmon effect on triplet-singlet energy transfer in the dye-doped Langmuir-Blodgett films

The effect of silver plasmonic nanoparticles on triplet-singlet energy transfer in the donor-acceptor pair of organic dyes was studied. Layered dye films were prepared on the surface of glass and island silver films using the Langmuir-Blodgett method. Amphiphilic analogs of Rose bengal and polymethine dye were used as donor and acceptor of energy. A polymer monolayer was deposited between monolayers of donor and acceptor molecules to exclude the competing triplet-triplet energy transfer. The spectra of delayed fluorescence and phosphorescence of donor-acceptor films and the delayed luminescence lifetime of donor in these films have been measured. It is shown that a threefold increase in the fluorescence intensity and a twofold increase in the donor phosphorescence intensity are observed on silver island films. The successful triplet-singlet energy transfer is evidenced by both the quenching of donor centers and appearance of sensitized delayed fluorescence of the acceptor with the duration close to the donor triplet lifetime. In the presence of silver nanoparticles, in addition to enhancement in the intensity of the donor emission, an increase in the efficiency of triplet-singlet energy transfer was observed. The obtained results can be used in various optical devices.

Keywords: Rose bengal; polymethine dye; silver island film; plasmon; delayed fluorescence; phosphorescence; triplet-singlet energy transfer; Langmuir-Blodgett films.

Introduction

Intermolecular energy transfer is one of the main processes in various applications of photonics [1, 2], optics, optoelectronics [3, 4, 5] and photovoltaics [6, 7]. The electron excitation energy transfer between molecules is one of the possible ways to influence the rate of photochemical reactions. The mechanism, the theoretical description of which was first proposed by Förster in the 1940s, allows distances up to 10 nm to be measured and it is based on the nonradiative dipole-dipole interaction of the giving and receiving molecules [8].

The triplet-singlet energy transfer was predicted by Förster [8] and confirmed by Ermolaev and Sveshnikova [9], who discovered this type of energy transfer using several phosphorescent donors and fluorescent acceptors in solid media. Thus, it was experimentally shown that Förster's theory can be applied to donor-acceptor pairs undergoing triplet-singlet energy transfer [9, 10].

The triplet-singlet energy transfer is widely used to obtain high-efficiency fluorescent organic light-emitting devices [3, 11, 12], to create biochemical and biophysical sensors [12], and can be used as a method to increase the rate of light emission from excited triplet states [13]. In [11], the authors used nonradiative energy transfer to increase the efficiency of a fluorescent red organic light-emitting device in four times. The triplet-singlet energy transfer is used in a new emitter concept for organic light-emitting diodes (OLEDs) called as thermally activated delayed fluorescence (TADF) [12, 14]. In [15], simultaneous and efficient energy transfer from both donating singlet and triplet states of a single photoluminescent molecular species was demonstrated, showing that cooperation between these two exciton transfer channels is possible.

At present, there are few publications devoted to the study of the effect of plasmonic nanoparticles on the triplet-singlet energy transfer. In [16], a decrease in the efficiency of triplet-singlet energy transfer on the surface of a silver island film was observed. And in [17], the influence of the plasmon effect of Ag nanoparticles (NPs) on singlet-singlet (S-S) and triplet-singlet (T-S) energy transfer in the same donor-acceptor pair of organic molecules was studied, and it was shown that the plasmon effect affects both S-S and T-S energy transfer.

In this work, the Langmuir-Blodgett (LB) technology was used to obtain structured films. The LB method is a useful and well-established tool for the fabrication of ultrathin films. The composition and thickness of such films can be precisely controlled at the molecular level. In addition, the orientation of fluorescent probes to the metal surface is fixed and the probes are in a monolayer, which is favorable for studies of metal-enhanced processes [18–20].

In the present work, we studied the effect of plasmonic nanoparticles on the T-S energy transfer in langmuir films in a pair of Rose bengal and polymethine dye. In contrast to [17], where the donor and acceptor molecules are arranged randomly, in the present work, the molecules are arranged structured using the Langmuir-Blodgett technology.

Experimental

Amphiphilic analogs of the dyes of Rose Bengal (RB) and indotricarbocyanine (PD) [21] were used as an energy donor (D) and acceptor (A), respectively. The structural formulas are shown in Fig. 1.

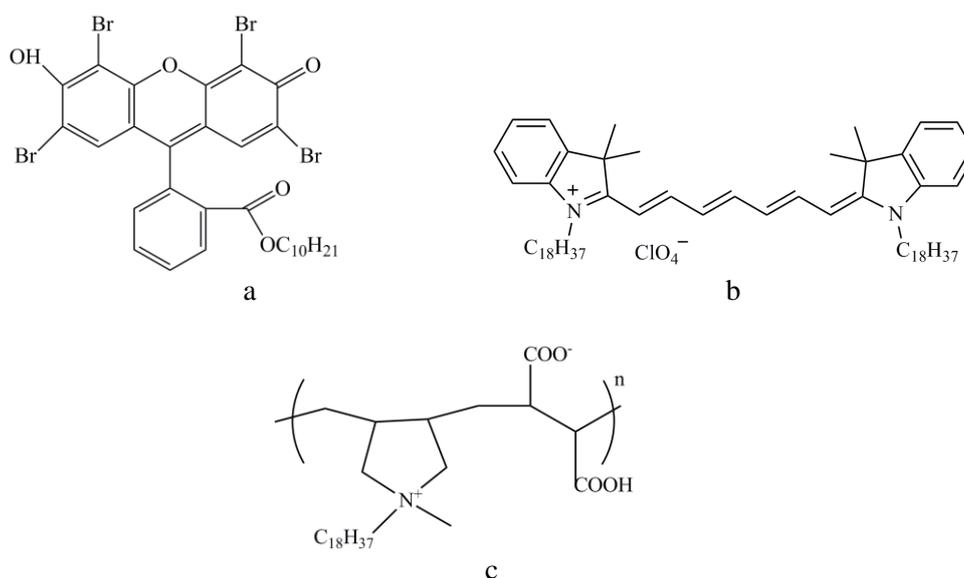


Figure 1. Structural formulas: a) RB; b) PD; c) PDOAM

Silver island films (SiFs) were prepared by magnetron sputtering, as described in [20]. After silver deposition, the films were annealed at a temperature of 240°C in a muffle furnace for 30 minutes. MIRA 3LMU scanning electron microscope (SEM, Tescan) was used for the studying of morphology and structure of the films. As shown by SEM (Fig. 2a), clusters of Ag particles with a radius of 40–50 nm are formed after annealing. The absorption spectrum of the SiF has a maximum at a wavelength of 435 nm (Fig. 2b).

Samples were prepared using the Langmuir-Blodgett (LB) method in a KSV Nima trough. In LB films, the distance between molecules can be changed up to their direct contact. The subphase was deionized water purified with AquaMax water purification system.

To prepare the samples, the dyes were dissolved in chloroform and mixed in the required ratios with the amphiphilic polymer poly (N,N-dialyl-N-octadecylamine-alt-maleic acid) (PDOAM) [21]. A mixed solution of amphiphilic polyampholyte and a dye makes it possible to obtain more stable and condensed films on the water surface. The relative concentration of dyes was 20 and 80 mol% for RB and PD, respectively. Transfer of monolayers to solid substrates was carried out according to the Z-type with the vertically method at a surface pressure of $\pi=35$ mN/m for RB, $\pi=30$ mN/m for PD. The π -A isotherm of PD is shown in Fig. 3. A pol-

polymer monolayer was deposited between the acceptor and donor monolayers (Fig. 2c). The polymer provides a distance between the donor and the acceptor of more than 2 nm. This makes it possible to eliminate the competing triplet-triplet energy transfer as an additional channel for energy transfer from the donor to the acceptor. Polymer monolayers were transferred to the substrate at a surface pressure of $\pi=35$ mN/m in a Z-type. It is known that a certain distance from the surface of plasmonic NPs is required to obtain the maximum effect [20, 22]. Therefore, in this work, several samples were prepared with different distances from the SiF surface (Fig. 2c). In one case, donor molecules were deposited directly onto the SiF surface; in the other case, three monolayers of PDOAM were deposited between SiF and donor molecules. In total, 3 RB monolayers and 1 PD monolayer were deposited in each film.

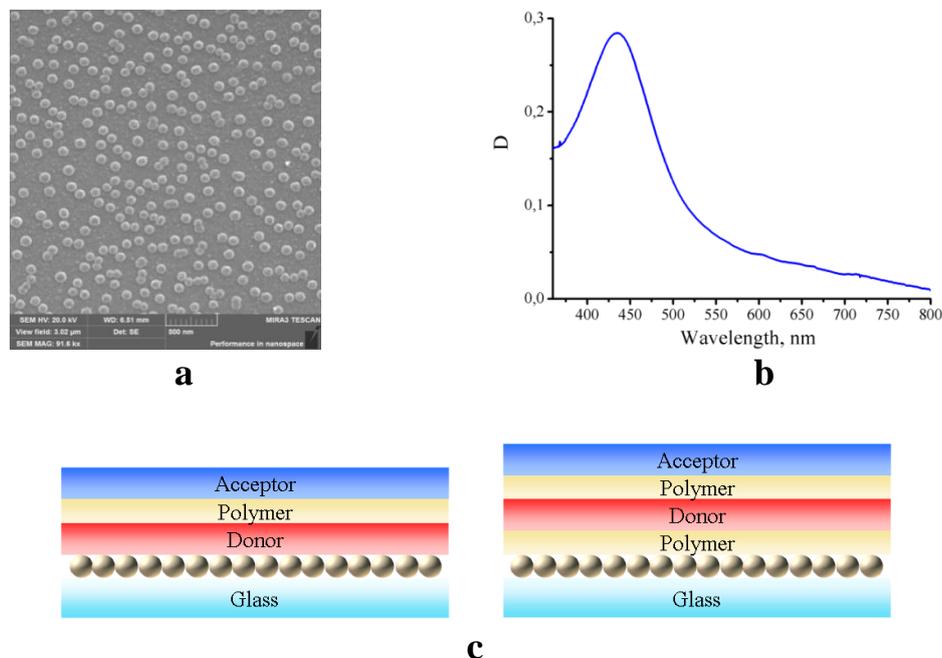


Figure 2. SEM image (a) and absorption spectrum (b) of SiF, and schemes of multilayer samples (c)

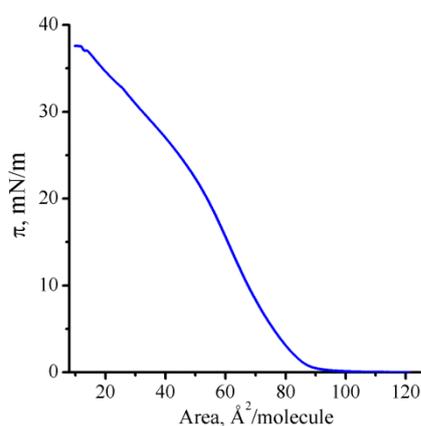


Figure 3. Surface pressure-area (π -A) compression isotherm of PD

The absorption spectra of the samples were measured using a Cary 300 spectrophotometer (Agilent Technologies). The emission spectra and lifetime were performed on a Cary Eclipse spectrometer (Agilent Technologies). Delayed fluorescence (DF) and phosphorescence measurements were carried out using a cryostat OptistatDN (Oxford Instruments). Phosphorescence mode: delay time = 0.1 ms; gate time = 0.5 ms. The DF and phosphorescence lifetimes were determined using the Cary Eclipse software using the following equation:

$$I(t) = I_0 \exp(-kt),$$

where, I_0 , $I(t)$ — luminescence intensity at zero and arbitrary times, respectively; $k=1/\tau$ — is the rate constant of the luminescence decay calculated from the graph plotted in the coordinates $\ln(I_0/I)$ versus t .

The energy transfer efficiency (E_{ET}) was estimated according to equation [23-25]:

$$E_{ET} = 1 - \frac{\langle \tau_D \rangle}{\langle \tau_{0D} \rangle}$$

where $\langle \tau_D \rangle$, $\langle \tau_{0D} \rangle$ — average fluorescence lifetime of RB in donor-acceptor and donor films, respectively.

The energy transfer rates in the presence of plasmonic NPs (k_{ET}^{pl}) and without them (k_{ET}) were estimated according to the following formulas) [17, 23-24]:

$$E = \frac{k_{ET}}{\tau_{0D} + k_{ET}} \quad \text{and} \quad E_{ET}^{pl} = \frac{k_{ET} + k_{ET}^{pl}}{\tau_{0D} + k_{ET} + k_{ET}^{pl}},$$

where E_{ET}^{pl} is the energy transfer efficiency in the presence of plasmon NPs.

Results and Discussion

The normalized absorption and luminescence spectra of RB and PD in the LB films are shown in Fig. 4. When excited in the absorption bands of RB, the fluorescence spectrum of RB has a maximum at a wavelength of $\lambda_{max}^{fl} = 562$ nm. The DF and phosphorescence spectra of RB at room temperature have a maximum approximately at 565 nm and 695 nm, respectively. The absorption spectrum maximum of PD is exhibits at 755 nm, and the fluorescence spectrum — at 785 nm. The absorption and fluorescence spectra have a universal shape characteristic of dyes of the polymethine series [26-27]. The overlap integral of the phosphorescence spectrum of RB and absorption of PD is equal to $I = 2.87 \cdot 10^{-12} \text{ M}^{-1} \text{ cm}^3$ [17].

The optical density of the dye LB film on glass at the absorption band maximum is about ~ 0.006 for RB and ~ 0.01 for PD. It is difficult to estimate the optical density of LB films on the SiF surface due to the effects of light scattering by Ag islands.

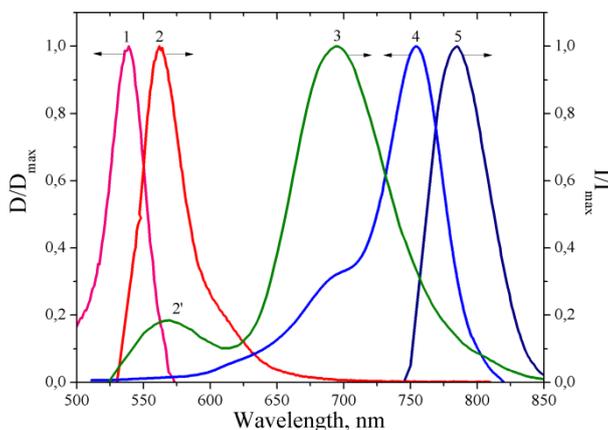


Figure 4. Normalized absorption (1, 4) and fluorescence (2, 5) spectra of RB (1, 2), PD (4, 5) films and delayed fluorescence (2') and phosphorescence (3) spectrum of RB

The fluorescence spectra of the LB films of the donor and the donor-acceptor pair on a glass substrate and on the SiF with a polymer are shown in Figure 5. The fluorescence intensity of the donor increased in 1.8 times directly on the SiF, and in 2.8 times on the SiF coated with three polymer monolayers compared with the control sample on the glass. In the presence of an acceptor, the intensity of the donor decreased, and a sensitized fluorescence of the acceptor appeared in the range of 700–800 nm. When the acceptor is excited with a wavelength of $\lambda_{exc} = 530$ nm in the absence of donor molecules, no radiation is detected. These data indicate that energy transfer proceeds from excited singlet RB molecules to unexcited acceptor molecules.

The influence of the SiF on the singlet–singlet energy transfer by the inductive–resonant mechanism was considered in previous works [17, 18, 25].

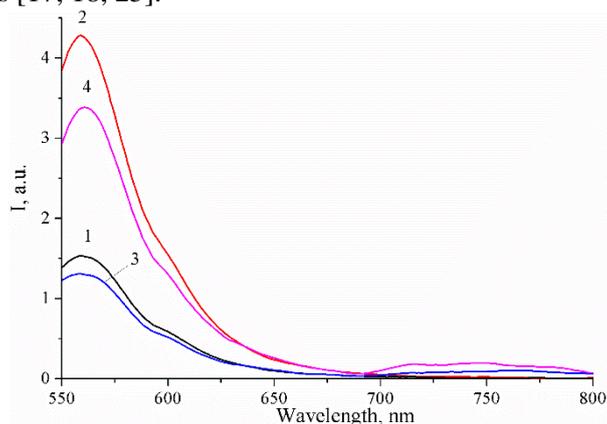


Figure 5. Fluorescence spectra of donor films (1, 2) and donor-acceptor films (3, 4) at $\lambda_{exc}=530$ nm on glass (1, 3), SiFs (2, 4) and SiFs coated with polymer monolayers (3)

The results of measurements of DF and phosphorescence of deoxygenated films of a donor and a donor-acceptor pair on glass and SiFs are shown in Figure 6a. When RB films on glass were photoexcited at $\lambda_{exc}=532$ nm, the maximum of the DF band of dye is recorded at approximately $\lambda_{max}=565$ nm, and the phosphorescence maximum at $\lambda_{max}=695$ nm. The lifetimes of phosphorescence and DF of RB are practically the same and equal to 0.3 ms.

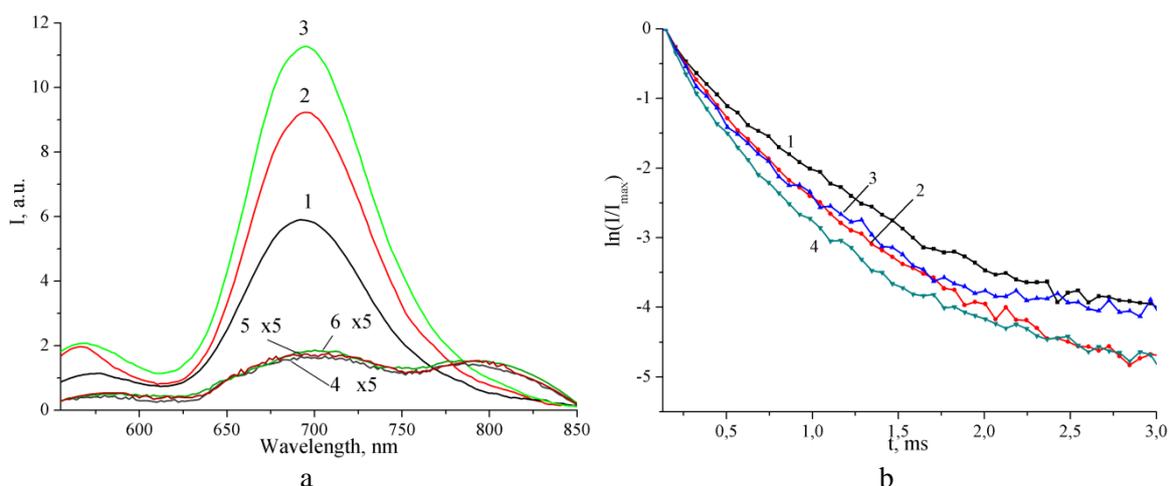


Figure 6. Spectra of DF and phosphorescence (a) and phosphorescence decay kinetics (b) of donor (1, 2, 3) and donor-acceptor films (4, 5, 6) at $\lambda_{exc}=530$ nm on glass (1, 4), on SiFs (2, 5) and SiFs coated with polymer monolayers (3, 6)

In the presence of an acceptor, quenching of the intensity of both DF and donor phosphorescence is observed, and the donor lifetime was decreased to 0.22 ms (Figure 6b). When a pure PD film was excited at $\lambda_{exc}=532$ nm, no radiation was registered. Because of the great overlapping of the sensitized luminescence of PD with the long-wavelength wing of the donor band, acceptor luminescence lifetimes, were recorded taking into account the donor phosphorescence contribution. The lifetime of acceptor in this case was equal to ~0.1 ms.

The observed quenching of the delayed luminescence RB and the sensitized luminescence of PD indicate the T–S energy transfer from triplet donor molecules to acceptor molecules in the ground electronic state.

In the presence of silver NPs, the intensity of DF and phosphorescence was increased almost 1.6 times directly on the SiFs and 1.9 times on the SiFs coated with three polymer monolayers. In this case, the duration of the luminescence is slightly reduced. In the presence of an acceptor, a similar spectrum is observed on SiFs, as on glass (Table 1).

Table 1

Intensity of DF and phosphorescence of donor (D), sensitized luminescence of acceptor, phosphorescence lifetime of donor τ_D^{Ph} ($\lambda_{reg}=695$ nm), T–S energy transfer rate in the presence (k_{ET}^{pl}) and without SiF (k_{ET}) in donor-acceptor (DA) films, $\lambda_{exc}=530$ nm

Samples	I_D^{DF} , r.u.	I_D^{Ph} , r.u.	I_A^{Fl} , r.u.	τ_D^{Ph} , ms	s^{-1} OR $k_{ET}^{TS pl}$ k_{ET}^{pl} s^{-1}	$\frac{k_{ET}^{pl}}{k_{ET}}$
On glass substrates						
D	1.2	5.9	–	0.295	–	–
DA	0.1	0.34	0.3	0.220	$1.13 \cdot 10^3$	–
On SiF						
D	1.9	9.2	–	0.230	–	–
DA	0.1	0.35	0.3	0.165	$1.32 \cdot 10^3$	1.17
On SiF with polymer						
D	2.1	11.2	–	0.240	–	–
DA	0.1	0.35	0.3	0.178	$1.2 \cdot 10^3$	1.06

In the presence of Ag NPs, an increase in the rate of spin-forbidden energy transfer is observed. For a sample in which donor molecules were located directly on the SiF, the rate of T–S energy transfer increases by almost 1.2 times. Even though at 6–8 nm from the SiF, the maximum intensity of the donor radiation was observed, the rate of T–S energy transfer from the energy donor to the acceptor is lower. This can be explained by the fact that molecules located far from the SiF are less affected by the plasmon field.

Conclusions

The effect of plasmonic silver NPs on the T–S energy transfer in a donor–acceptor pair in planar nanostructures has been studied. Ag island films exhibit a threefold increase in the fluorescence intensity and a twofold increase in the phosphorescence intensity of the energy donor. An increase in the T–S energy transfer efficiency was registered recorded in the plasmon field of Ag NPs. In this case, the maximum growth in the rate of T-S energy transfer was observed for a sample deposited directly on the SiF surface.

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Боялған Ленгмюр-Блоджетт қабыршақтарындағы триплет-синглеттік энергия тасымалдауына плазмон әсері

Мақалада органикалық бояғыштардың донор-акцептор жұбындағы триплет-синглеттік энергия тасымалдауына плазмондық күміс нанобөлшектерінің әсері зерттелген. Ленгмюр-Блоджетт технологиясын қолдана отырып, шыны төсемелері мен күміс аралдық қабыршақтарына бетіне қабатты бояғыш қабыршақтары дайындалды. Энергия доноры және акцепторы ретінде бенгалдың қызғылт және полиметиндік бояғыштарының амфифильді аналогтары пайдаланылды. Бәсекелес триплет-триплеттік энергияның тасымалдауын болдырмау үшін донорлық және акцепторлық молекулалардың моноқабаттары арасында полимер моноқабаты орналастырылды. Донор-акцепторлық қабыршақтардың баяуланған флуоресценция және фосфоресценция спектрлері және осы қабыршақтардағы донордың ұзақ мерзімді люминесценциясының өмір сүру ұзақтығы өлшенді. Күміс қабыршақтарында донордың флуоресценциясының қарқындылығы үш есе және фосфоресценция қарқындылығы екі есе артуы байқалатыны көрсетілген. Триплет-синглет энергия тасымалдануы донор сәулеленуінің өшуімен де, донорлық триплеттердің өмір сүру ұзақтығына жақын сенсбилизацияланған акцептордың баяуланған флуоресценцияның пайда болуымен де дәлелденеді. Күміс нанобөлшектері болған жағдайда энергия донор қарқындылығын арттыруынан басқа, триплет-синглеттік электрондық энергия тасымалдау эффективтілігінің жоғарылауы байқалады. Алынған нәтижелерді әртүрлі оптикалық құрылғыларда қолдануға болады.

Кілт сөздер: бенгалдың қызғылты, полиметиндік бояғыш, күміс аралдық қабыршағы, плазмон, баяуланған флуоресценция, фосфоресценция, триплет-синглеттік энергия тасымалдау, Ленгмюр-Блоджетт қабыршақтары.

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Влияние плазмонного эффекта на триплет-синглетный перенос энергии в окрашенных пленках Ленгмюра–Блоджетт

Исследовано влияние плазмонных наночастиц серебра на триплет-синглетный перенос энергии в донорно-акцепторной паре органических красителей. По технологии Ленгмюра-Блоджетт были приготовлены слоистые пленки красителей на поверхности стекла и островковых пленок серебра. В качестве донора и акцептора энергии были использованы амфифильные аналоги бенгальского розового и полиметинового красителя. Между монослоями молекул донора и акцептора был нанесен монослой полимера для исключения конкурирующего триплет-триплетного переноса энергии. Измерены спектры замедленной флуоресценции, фосфоресценции донорно-акцепторных пленок и время жизни длительной люминесценции донора в этих пленках. Показано, что на островковых пленках серебра наблюдается трехкратное увеличение интенсивности флуоресценции и двухкратное увеличение интенсивности фосфоресценции донора. Об успешном триплет-синглетном переносе энергии свидетельствует как тушение донорных центров, так и появление сенсibilизованной замедленной флуоресценции акцептора с длительностью, близкой ко времени жизни триплетов донора. В присутствии наночастиц серебра, помимо усиления интенсивности излучения донора, наблюдается рост эффективности триплет-синглетного переноса электронной энергии. Полученные результаты могут быть использованы в различных оптических устройствах.

Ключевые слова: бенгальский розовый, полиметиновый краситель, островковая пленка серебра, плазмон, замедленная флуоресценция, фосфоресценция, триплет-синглетный перенос энергии, пленки Ленгмюра-Блоджетт.