

Versatile Tunable Microresonator for the Light-Matter Interaction Studying in the Strong-Coupling Mode

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The interaction between an ensemble of molecules and an electromagnetic field in a highly limited volume makes it possible to control the properties of a substance and, therefore, is an exceptionally promising area of research. The most common way to achieve weak or strong light-matter coupling is to place an ensemble of molecules in a micron-sized resonator. In such a system, the interaction of light with matter appears in the form of a change in the spectral response of the system, which depends on the strength of the coupling between the ensemble of molecules and the modes of the resonator. Currently, there is no general and user-friendly approach that allows studying a lot of different samples in a wide optical range using the same resonator setup. The present paper describes the design of a device that makes it possible to overcome this disadvantage, speed up and facilitate the study of the light-matter interaction, and also obtain weak and strong light-matter coupling modes for a large number of samples in the UV, visible, and IR regions of the optical spectrum. The device developed here is based on the tunable unstable $\lambda/2$ Fabry-Perot microresonator, consisting of flat and convex mirrors, which satisfy the condition of plane-parallelism at least at one point of the curved mirror and can significantly reduce the mode volume. The device was used to study the effect of the strong-coupling regime on the fluorescent properties of the Rhodamine 6G (R6G) dye embedded in a boron nitride nanoparticles matrix. It was found that the use of boron nitride (h-BN) as a carrier matrix has an orienting effect on the dye molecules, that results in an increase of the light-matter coupling strength at a lower resonator mode energy required.

Keywords: microspectroscopy, optical microresonator, strong coupling, boron nitride.

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Introduction

Resonant interaction between ensembles of molecules and an electromagnetic (EM) field in a very limited volume makes it possible to control the properties of a bound substance and is an extremely promising area of research. For the first time, the possibility of controlling the spontaneous emission of a quantum system when it interacts with an EM field in a limited volume was described by Purcell [1]. The interaction in such a system is described as the binding of a substance placed in a resonator with quantized modes of the EM field in such a way that the properties of the substance can be manipulated [2–6]. In the resonator, the binding strength depends on the rate of exchange of light energy with matter compared to irreversible energy losses arising due to the short lifetime of the bound substance in the excited state. In this case, a weak coupling mode is a mode in which an ensemble of molecules with discrete modes of the EM field is excited and bound, but energy losses dominate. In this mode, the lifetime of the excited states changes due to the limitation

of the mode volume of the field in the resonator. This mode is called the Purcell effect, which can be used to enhance or attenuate spontaneous radiation. In contrast to the Purcell effect, the strong coupling mode occurs when the rate of energy exchange between light and matter is high compared to the rate of damping of the excitation of bound molecules of matter, i.e. there is a coherent coupling of the photonic modes of the resonator and excitons of matter. Also, depending on the type of resonator, excited plasmons can act instead of photonic resonator modes, while coherently coupled excitons and plasmons, or photonic modes generate new optical excitations — polaritons arising due to the strong coupling of these two oscillator systems. Thus, studies of nanostructures representing complexes of J aggregates and gold nanoparticles have shown that in such systems a strongly bound state of hybridized modes is detected not only in the extinction spectra, but also in the photoluminescence [7,8]. It is noteworthy that in the photoluminescence signal of such systems, the evolution of two peaks corresponding to the upper and lower polariton branches was traced, as the

plasmon energy is rebuilt from the exciton resonance. In addition, studies have previously been conducted on the effect of the near field of silver nanoparticles, which are nanoscale resonators, on the photophysical properties of semiconductor quantum dots of the compositions CdSe/ZnS and InGaAs, in an AlGaAs matrix under stationary and pulsed excitation of [9,10]. At the same time, when studying the parameters of the photoluminescence spectra of quantum dots under excitation in the plasmon band region, an increase in the absorption of quantum dots in the near field of silver nanoparticles [9,10] was found.

One of the most interesting features of the strong light-matter coupling regime is the potential for changing the fundamental properties of matter molecules, which opens up new prospects in biology, chemistry and other fields of science [11]. In particular, in the paper of Coles and others [12], a microresonator consisting of a film of polyvinyl alcohol with a thickness of 205 nm with chlorosomes embedded in it (photosynthetic antenna complexes of green serobacteria) was demonstrated, which was placed between two translucent mirrors with a thickness of 40 nm. In the described system, the magnitude of the Rabi splitting of the electronic levels of photosynthetic antenna complexes that determine the pathways of energy transfer during photosynthesis reached 150 MeV. The results of this study showed that the strong interaction of light and matter can be used to modify energy transfer pathways in photosynthetic organisms. In addition, when using a microresonator consisting of a 620-nm layer of a mixture of *J* aggregates of two different dyes in a matrix placed between two metal mirrors, it was shown that in the strong coupling mode, energy can be transferred by the resonance transfer mechanism to distances much greater than the Foerster radius [13].

No less interesting is the effect of a strong connection between the photonic mode of the microresonator in the IR range and the vibrational modes of the substance, which can be used, for example, to change the rate of chemical reactions and influence the processes of enzymatic catalysis in biology and medicine. Thus, in experiments with a layer of polyvinyl acetate with a thickness of 2 μ m placed between two gold mirrors with a thickness of 10 nm deposited on germanium substrates, Rabi splitting of vibrational transitions reaching 20.7 MeV [14] was observed. Later, the same approach was used to demonstrate that strong coupling at oscillatory transitions can also be used to enhance Raman scattering [15]. The successful continuation of these works was the discovery of a strong connection of the vibrational modes of bovine serum albumin in the mid-IR range [16], which may be important for clarifying the role of vibrational dynamics in enzymatic catalysis in biology and in experiments on hydrogen-deuterium exchange. In the paper [17], the same group established the influence of a strong bond

on the rate of chemical reactions involving the Si-C bond.

Over the past two decades, advances in the development and creation of new microresonators have had a huge impact on the study of the interactions of light with matter. Various types of microresonators were used to achieve a strong coupling regime accompanied by changes in the properties of the bound substance with different values of the values of the Rabi splitting [18–22]. The choice of a microresonator for a particular experiment depends on several factors, such as design properties (ease of manufacture and use) and internal parameters of the microresonator (Q factor, modal volume). In this respect, the Fabry-Perot microresonator offers undeniable advantages: simplicity of design, versatility and the ability to provide a very small mod volume [23–26]. Earlier in the paper [27], a device based on a tunable unstable $\lambda/2$ Fabry-Perot microresonator was proposed to create conditions for a strong „light-substance“ connection. The design of the device includes a convex mirror to ensure a minimum modal volume, as well as to meet the condition of plane parallelism at least at one point of such a mirror, which eliminates the need for strictly parallel displacement of two parallel mirrors relative to each other. Adjustment of the optical path length of such a microresonator is carried out by three piezoactuators that change the vertical position of the upper convex mirror. However, this design has several disadvantages that limit its use: (i) sample replacement is time-consuming, (ii) matching the sample with the working area of the microresonator is difficult, (iii) the device can only work in a narrow wavelength range.

In this paper, we present a universal tunable microresonator, which is a modification of the above approach. Structurally, the device is a flexible and simple tunable microresonator that allows you to obtain an electromagnetic field with a minimum modal volume with an operating range in the UV, visible and IR spectral regions, which makes it possible to quickly replace the sample, which makes the configuration very convenient for studying the interaction of light with matter.

Construction of a universal tunable microresonator

Fig. 1,*a* shows a diagram of a universal tunable microresonator, demonstrating its general functionality. The proposed design is based on the use of two mutually positioned plates (1 and 2), on which the resonator mirrors are mounted. The tunable unstable Fabry-Perot microresonator consists of a convex mirror (3, a plane-convex lens with a radius of curvature of 77.3 mm and a diameter of 25.4 mm, covered with a layer of Al with a thickness of 60 nm and a layer of SiO₂ with a thickness of 20 nm, mounted on top plate) and a flat mirror (4, a cover glass with a similar coating, mounted on the bottom plate) and is the basis of the entire structure. The convex

mirror is not rigidly fixed to the upper plate by means of a magnetic holder (5) having three points of contact with the upper movable plate, and is installed in the corresponding slots. The use of a non-rigid attachment of the upper mirror is dictated by the need to prevent the possibility of damage to the mirror coatings in the event of their direct contact during vertical positioning. The upper plate (1) can be precisely moved in *XY* directions using two linear positioners (6) to align the working area of the microresonator with respect to the sample. The coarse tuning of the resonator length is carried out using a high-precision differential regulator (7, DRV3. 8 mm High Precision Differential Micrometer, Thorlabs, USA), in increments of 1 μm . The lever of the regulator action correlates with the lens attachment point as 1:4 to increase the accuracy of vertical positioning up to 250 nm. The plane of the upper plate can be adjusted using two screws, which also serve as attachment points for the upper plate. Permanent magnets are used to fix and clamp the upper plate. This fixation makes it easy to remove the upper plate, providing access to the sample holder, which consists of a steel ring with a lower flat mirror glued to it, on which the sample is directly applied. In turn, the sample holder is loosely (using permanent magnets) attached to the magnetic plate of the Z piezo positioner (8), which allows you to roughly change the position of the sample in the field of view of the lens of the inverted confocal microspectrometer (9), as well as quickly change sample. The main function of the Z piezo positioner — fine tuning of the optical length of the microresonator with an accuracy of up to a nanometer. The Z piezo positioner is controlled by an atomic force microscope (AFM) controller, which is very well suited for this purpose due to the high dynamic range of voltages used, which makes it possible to work both in the full voltage range (vertical movement of a flat mirror up to 10, μm) and in a narrow range voltages for fine tuning of the microresonator within a few nm. The bottom plate (2) serves as the basis of the entire microresonator design. All of the above components are attached to it, and it can be installed on a micrometric *XY* positioner of an optical confocal inverted microspectrometer. The installation is carried out using three micrometer support screws (10), which, in turn, allow the alignment of the plane of the entire microresonator relative to the optical axis of the microspectrometer.

Optical scheme of using a universal tunable microresonator

Fig. 1, *b* shows the optical scheme of a universal tunable microresonator in combination with an inverted confocal microspectrometer. The developed optical system is designed to integrate a tunable microresonator with both an inverted optical confocal microspectrometer, and an Optem Zoom 125 (5) direct video microscope, equipped with either a video camera or a

„white“ LED, depending on the microresonator setup procedure.

The optical system uses two types of microresonator lighting configuration.

1. Illumination from below with a collimated laser beam (microspectrometer lens (7) removed) is used both for pre-tuning the microresonator and for confocal microspectrometry — the microspectrometer lens is installed. In the first case, a collimated laser beam passing through a microresonator forms Newton rings, which can be observed in a direct microscope Optem Zoom 125 (Excelitas Technologies, USA) using a MOTIC 3.0 video camera. (Motic, Hong Kong). This mode is used to bring mirrors closer together and adjust the working area of the microresonator in the lateral plane. In the confocal microspectrometry mode, laser excitation and collection of secondary radiation (Raman scattering and/or fluorescence) is carried out through the mounted lens of an inverted microscope. The recorded secondary radiation from the sample (3) deposited on the surface of the lower mirror (2) passes through the confocal module and enters the Andor Shamrock 750 monochromator equipped with the Andor DU971P-BV CCD sensor (Andor Technology, Oxford Instruments, Great Britain). The confocal mode of microspectrometric measurements is used to artificially limit the area of registration of secondary radiation in the region of the existence of the resonator mode.

2. Vertical illumination (6) by a broadband light source („white“ MWWHF2 LED, Thorlabs, USA) is carried out through the optical path of the Optem Zoom 125 direct microscope. This mode is used for spectral characterization of the microresonator mode composition. The process of tuning the microresonator mode composition is carried out due to the smooth movement of the Z-piezopositioner (4). The spectral composition of the radiation passing through the microresonator is recorded in the same way as the secondary radiation from the sample. At the same time, both spectra can be recorded simultaneously, which makes it possible to adjust the resonator with high accuracy to the electronic transitions of the studied molecules. It is important to note, that such a method of controlling the optical length of the microresonator makes it possible to achieve the half-wave mode of its operation with high accuracy.

Studies of the effect of the strong coupling mode on the fluorescent properties of the dye P6J embedded in the matrix of boron nitride nanoparticles

At this stage, we have developed a technique for introducing molecules of the fluorescent dye P6J into boron nitride (h-BN) nanoparticles and forming densely packed layers of such nanoparticles on the surface of flat mirrors

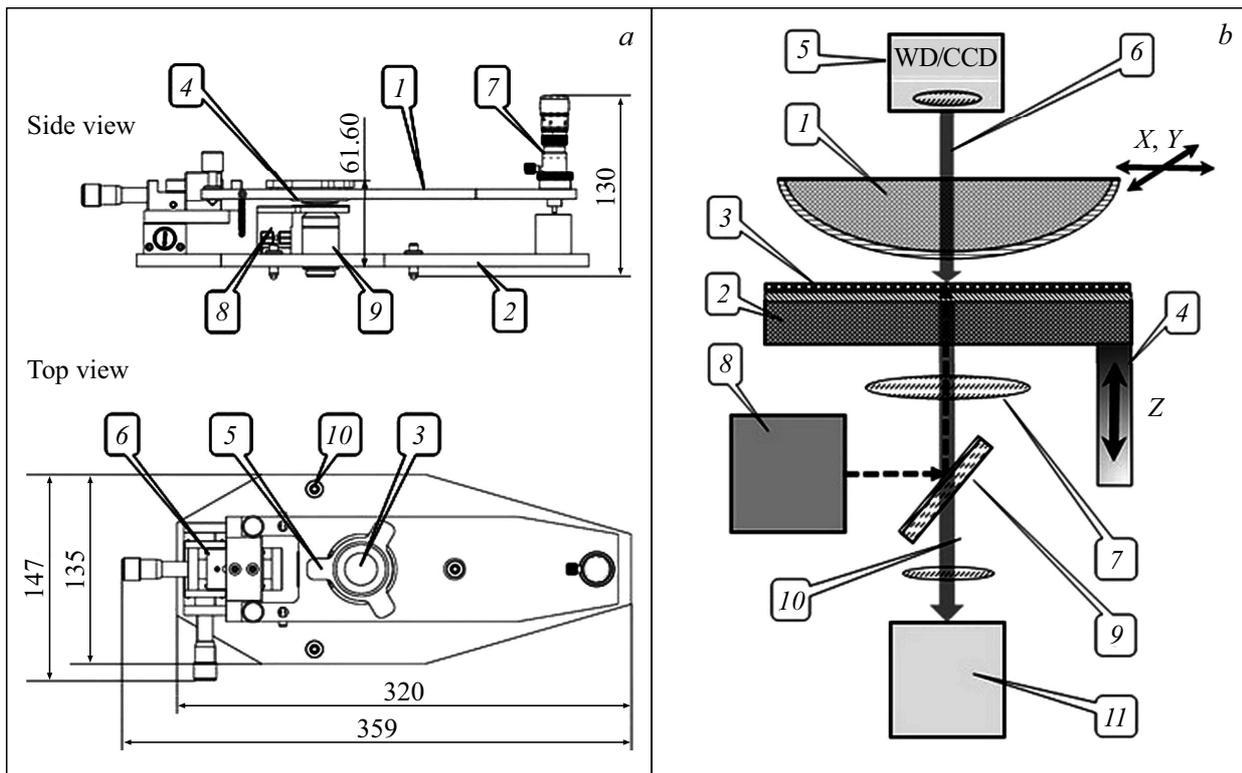


Figure 1. (a) Construction of a universal tunable microresonator: 1 — upper plate; 2 — lower plate; 3 — flat-convex metallized lens — upper mirror of the microresonator; 4 — metallized cover glass — bottom mirror of the microresonator; 5 — magnetic holder of the upper mirror; 6 — precision XY -positioner of the upper mirror; 7 — high-precision differential regulator; 8 — Z -piezo positioner; 9 — lens of inverted confocal microspectrometer; 10 — micrometer support screws. (b) Optical scheme of a universal tunable microresonator: 1 — flat-convex metallized lens — upper mirror of the microresonator; 2 — metallized cover glass — lower mirror of the microresonator; 3 — sample; 4 — Z -piezo positioner; 5 — a block containing „white“ LED, CCD and optical collimator; 6 — broadband radiation; 7 — inverted microscope lens; 8 — laser; 9 — optical low-frequency filter; 10 — analyzed radiation; 11 — confocal module and monochromator equipped with CCD.

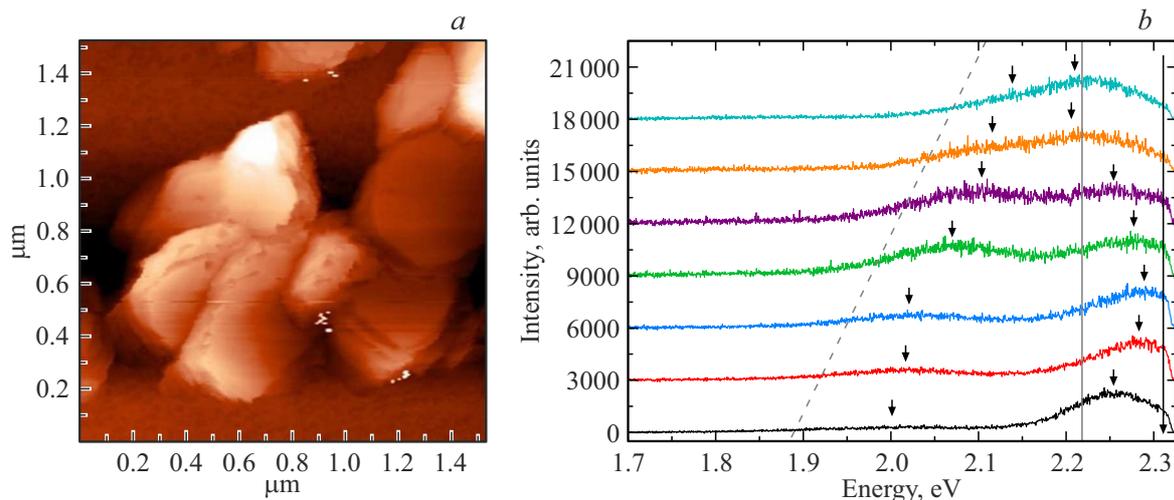


Figure 2. (a) Atomic force image of single clusters of h-BN nanoparticles doped with P6J dye. (b) Photoluminescence spectra (color curves) of the P6J-(h-BN)-PVP film associated with a tunable microresonator with a different distance between the reflecting mirrors (67%). From top to bottom, the distance has been increased from 988 to 1195 nm. The corresponding position of the transmission maxima is marked with a dashed gray line. A solid gray line indicates the maximum radiation of the unbound P6J film. The black arrows indicate the maxima of polariton emission.

of the tunable microresonator described in the previous sections. It was found that the use of polyvinylpyrrolidone (PVP) when introducing P6J into h-BN matrices makes it possible to isolate individual P6J molecules (the absence of a characteristic red shift in the fluorescence spectra of P6J), which significantly increases the quantum yield of fluorescence of the entire system. Densely packed layers of h-BN nanoparticles doped with P6J dye in the PVP matrix and deposited on the bottom mirror of the microresonator were characterized using AFM. It was found, that h-BN nanoparticles doped with P6J dye are arranged in the form of separate aggregates with heights reaching 60 nm. At the same time, the distribution of the obtained aggregates has a fairly narrow height distribution in the region of 43 nm, which means a sufficiently uniform coating in height, allowing the resonator mirrors to be brought closer to the distance necessary for the strong coupling mode. The orientation of the occurrence of single clusters of h-BN nanoparticles doped with P6J dye was also studied. Fig. 2, *a* shows an AFM image of such a cluster. It follows from the above figure that most of the doped h-BN nanoparticles are located almost parallel to the plane of the lower mirror of the resonator, which ensures that the dipole transition moment of the dye P6J coincides with the electric field strength vector in the resonator.

In the experiments to study the effect of the strong coupling mode on the fluorescent properties of P6J, embedded in a matrix of boron nitride nanoparticles, we varied the distance between the microresonator mirrors from 988 to 1195 nm with a small step of about 30 nm, while the free spectral range was 0.4 eV (Fig. 2, *b*). From the data presented in the figure, one can see an obvious anti-crossing, as well as large splitting energies that correlate with the difference in the optical properties of the electromagnetic modes of the resonator. The corresponding mode energy ranged from 1.88 to 2.07 eV, without overlapping the regions around the radiation maxima of the P6J film, while the binding force reached 208 MeV. For comparison with the case of undirected P6J, we conducted a study of the same dye, but embedded in the matrix from polymethylmethacrylate (PMMA). By the method of ultramicrotomy, thin slices of PMMA with embedded P6J were obtained and then transferred to the plane of the lower mirror of the resonator. In this experiment, the distance between the mirrors changed from 1790 nm to 1970 nm in increments of 30 nm. The corresponding mode energy varied from 2.1 to 2.3 eV, covering the region around the maxima of the P6J radiation encapsulated in the PMMA film. At the same time, the greatest splitting and, consequently, the binding force occurred when the photon energy of the resonator was strictly equal to the radiation maxima of P6J about 2.22 eV and reached 184 MeV.

Thus, it can be concluded that the introduction of boron nitride into the system leads to an increase in the binding force at a lower required resonator mode energy due to the orienting action of h-BN nanoparticles. At the same time, the increase in the binding energy in our case is of the

order of 15%. Since the magnitude of the binding energy is directly proportional to the scalar product of the dipole moment of the dye transition and the electric field intensity vector, it is obvious that this energy has the greatest value when the directions of both vectors coincide. Theoretical and experimental consideration of this issue is given in the papers [28,29]. In particular, in the paper [29], the change in the value of the Rabi splitting (binding energy) is considered depending on the degree of ordering of liquid crystal molecules placed in the cavity of the microresonator. Thus, during the transition from an isotropic (completely disordered) phase to an ordered smectic phase, in which the liquid crystal molecules and, accordingly, their dipole moments lie in the plane of the microresonator mirrors, the binding energy increases by 30%. Such an increase is very close to the theoretical limit of the increase of any value proportional to the scalar product of the vectors when moving from their random location relative to each other to a collinear location. It is known that the liquid crystal molecules in the smectic phase have the highest degree of order, which, in turn, determines such a strong increase in the binding energy in this experiment. In our case, we observed an increase in the binding energy, half as much as in the papers described above, which is nevertheless a remarkable result, since the increase in the binding force of 15% was achieved not in an idealized, but in a kind of universal system that allows orienting a fairly wide class of dyes.

Conclusion

In this paper, a new design of a universal tunable Fabry-Perot microresonator is proposed, adapted to a wide range of studies of the interaction of light with matter in the strong coupling mode during serial and systematic studies of a large number of samples. The developed microresonator opens the way for further progress in studying the effect of the coupling force of light and matter on the efficiency of the Raman scattering effect, the rates of chemical and biological reactions, conductivity, radiation generation, efficiency and characteristic distances of non-radiative energy transfer and other processes. With the help of the developed device, the effect of the strong coupling mode on the fluorescent properties of the P6J dye embedded in an orienting matrix of boron nitride nanoparticles was studied, which contributes to an increase in the binding force at a lower required resonator mode energy due to the orienting action of h-BN nanoparticles. The creation of the described system and the developed methods of its use is an important step towards numerous practical applications of the effect of the interaction of light and matter in the development of new materials with unique properties.

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Conflict of interest

The authors declare that they have no conflicts of interest.

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