Giant enhancement of two photon induced luminescence in metal nanostructure

Pavel N. Melentiev,[1,*] **Anton E. Afanasiev,**[1] **Arthur A. Kuzin,**[1,2] **Alexey V. Zablotskiy,**[2] **and Victor I. Balykin**[1,3]

[1]*Institute for Spectroscopy Russian Academy of Sciences, Phizicheskaya str., 5, Troitsk, Moscow, 142190 Russia*
[2]*Moscow Institute of Physics and Technology, Institutskiy per., 9, Dolgoprudniy, Moscow reg., 141700, Russia*
[3]*balykin@isan.troitsk.ru*
[*]*melentiev@isan.troitsk.ru*

Abstract: We experimentally demonstrate a drastic increase in the rate of radiative process of a nanoscale physical system with implementation of the three physical effects: (1) the size effect, (2) plasmon resonance and (3) the optical Tamm state. As an example of a nanoscale physical system, we choose a single nanohole in Au film when the nanohole is embedded in a photonic crystal of a specific type that maintains an optical Tamm state and as a radiative process - a nonlinear photoluminescence. The efficiency of the nonlinear photoluminescence is increased by more than 10^7 times in compare to a bulk material.

OCIS codes: (310.6628) Subwavelength structures, nanostructures; (190.4180) Multiphoton processes; (190.7110) Ultrafast nonlinear optics; (190.4350) Nonlinear optics at surfaces; (260.3800) Luminescence; (350.4238) Nanophotonics and photonic crystals.

References and links

1. G. Pake and E. Purcell, "Line shapes in nuclear paramagnetism," Phys. Rev. **74**, 1184 (1948).
2. E. Yablonovitch, "Inhibited spontaneous emission in solid-state physics and electronics," Phys. Rev. Lett. **58**, 2059 (1987).
3. J. P. Wilcoxon, J. E. Martin, F. Parsapour, B. Wiedenman, and D. F. Kelley, "Photoluminescence from nanosize gold clusters," J. Chem. Phys. **108**, 9137 (1998).
4. G. S. Kino, A. Sundaramurthy, P. J. Schuck, D. P. Fromm, and W. E. Moerner, in *Surface Plasmon Nanophotonics*, edited by M. L. Brongersma and P. G. Kik, (Springer, 2007), p. 125.
5. G. T. Boyd, Y. R. Yu, and Z. H. Shen, "Photoinduced luminescence from the noble metals and its enhancement on roughened surfaces," Phys. Rev. B **33**, 7923 (1986).
6. M. R. Beversluis, A. Bouhelier, and L. Novotny, "Continuum generation from single gold nanostructures through near-field mediated intraband transitions," Phys. Rev. B **68**, 115433 (2003).
7. P. Y. Yeh, A. Yariv, and A. Y. Cho, "Optical surface waves in periodic layered media," Appl. Phys. Lett. **32**, 104 (1978).
8. A. P. Vinogradov, A. V. Dorofeenko, S. G. Erokhin, M. Inoue, A. A. Lisyansky, A. M. Merzlikin, and A. B. Granovsky, "Surface state peculiarities in one-dimensional photonic crystal interfaces," Phys. Rev. B **74**, 045128 (2006).
9. C. R. Rosberg, D. N. Neshev, Y. V. Kartashov, R. A. Vicencio, W. Krolikowski, M. I. Molina, A. Mitchell, V. A. Vysloukh, L. Torner, and Y. S. Kivshar, "Nonlinear Tamm states in periodic photonic structures," Opt. Photonics News **17**, 29 (2006).
10. M. Kaliteevski, I. Iorsh, S. Brand, R. A. Abram, J. M. Chamberlain, A. V. Kavokin, and I. A. Shelykh, "Tamm plasmon-polaritons: possible electromagnetic states at the interface of a metal and a dielectric Bragg mirror," Phys. Rev. B **76**, 165415 (2007).

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

11. H. Eckardt, L. Fritsche, and J. Noffke, "Self-consistent relativistic band structure of the noble metals," J. Phys. F: Met. Phys. **14**, 97 (1984).
12. O. Gazzano, S. Michaelis de Vasconcellos, K. Gauthron, C. Symonds, J. Bloch, P. Voisin, J. Bellessa, A. Lematre, and P. Senellart, "Evidence for confined Tamm plasmon modes under metallic microdisks and application to the control of spontaneous optical emission," Phys. Rev. Lett. **107**, 247402 (2011).
13. S. V. Fomichev, D. F. Zaretsky, D. Bauer, and W. Becker, "Classical molecular-dynamics simulations of laser-irradiated clusters: nonlinear electron dynamics and resonance-enhanced low-order harmonic generation," Phys. Rev. A **71**, 013201 (2005).
14. P. N. Melentiev, T. V. Konstantinova, A. E. Afanasiev, A. A. Kuzin, A. S. Baturin, A. V. Tausenev, A. V. Konyashchenko, and V. I. Balykin, "Single nano-hole as a new effective nonlinear element for third-harmonic generation," Laser Phys. Lett. **10**, 075901 (2013).
15. A. Mooradian, "Photoluminescence of Metals," Phys. Rev. Lett. **22**, 185 (1969).
16. E. Dulkeith, T. Niedereichholz, T. A. Klar, J. Feldmann, G. von Plessen, D. I. Gittins, K. S. Mayya, and F. Caruso, "Plasmon emission in photoexcited gold nanoparticles," Phys. Rev. B **70**, 205424 (2004).
17. D. Nagesha, G. Laevsky, P. Lampton, R. Banyal, C. Warner, C. DiMarzio, and S. Sridhar, "In vitro imaging of embryonic stem cells using multiphoton luminescence of gold nanoparticles," Int. J. Nanomed **2**, 813 (2007).
18. N. Durr, T. Larson, D. Smith, B. Korgel, K. Sokolov, and A. Ben-Yakar, "Two-photon luminescence imaging of cancer cells using molecularly targeted gold nanorods," Nano Lett. **7**, 941 (2007).
19. A. Gobin, M. Lee, N. Halas, W. James, R. Drezek, and J. West, "Near-infrared resonant nanoshells for combined optical imaging and photothermal cancer therapy," Nano Lett. **7**, 1929 (2007).
20. P. N. Melentiev, T. V. Konstantinova, A. E. Afanasiev, A. A. Kuzin, A. S. Baturin, and V. I. Balykin, "Single nanohole and photoluminescence: nanolocalized and wavelength tunable light source," Opt. Express **20**, 19474 (2012).
21. K. Imura, T. Nagahara, and H. Okamoto, "Near-field two-photon-induced photoluminescence from single gold nanorods and imaging of plasmon modes," J. Phys. Chem. B **109**, 13214 (2005).
22. P. Biagioni, M. Celebrano, M. Savoini, G. Grancini, D. Brida, S. Mátéfi-Tempfli, M. Mátéfi-Tempfli, L. Duò, B. Hecht, G. Cerullo, and M. Finazzi, "Dependence of the two-photon photoluminescence yield of gold nanostructures on the laser pulse duration," Phys. Rev. B **80**, 045411 (2009).
23. T. V. Shahbazyan, "Theory of plasmon-enhanced metal photoluminescence," Nano Lett. **13**, 194 (2003).
24. P. N. Melentiev, A. E. Afanasiev, A. A. Kuzin, A. Zablotsky, A. S. Baturin, and V. I. Balykin, "Single nanohole and photonic crystal: wavelength selective enhanced transmission of light," Opt. Express **19**, 22743 (2011).
25. I .V. Treshin, V. V. Klimov, P. N. Melentiev, and V. I. Balykin, "Optical Tamm state and extraordinary light transmission through a nanoaperture," Phys. Rev. A, **88**, 023832 (2013).
26. V. M. Shalaev, C. Douketis, T. Haslett, T. Stuckless, and M. Moskovit, "Two-photon electron emission from smooth and rough metal films in the threshold region," Phys. Rev. B **53**, 11193 (1996).
27. P. N. Melentiev, A. E. Afanasiev, and V. I. Balykin, "Optical Tamm state on a femtosecond time scale," Phys. Rev. A **88**, 053841 (2013).
28. Monocrystalline Au films was supplied from www.phasis.ch

1. Introduction

The radiative properties of a single atom in free space are determined by its coupling to the continuum of radiation modes in vacuum. The spatial and frequency distributions of the modes depend upon the nature of the surrounding of atom environment which can be modified and controlled. This photon density of states can be increased or reduced with respect to its free-space value, leading to inhibited or enhanced atomic radiative properties. Textbook example from atomic physics is a change in the rate of spontaneous emission of an atom when it is placed in a resonant cavity (the Purcell effect) [1]. As was shown by Yablonovitch [2], such a capability is equally important in solid-state physics, where spontaneous emission arises in the form of radiative electron-hole recombination. Another approach to enhance radiative properties of quantum emitters is the use of their coherent collective interactions. As an example, the radiative properties of an ensemble of atoms can be enhanced by collective emission processes. This effect has been extensively studied since the early work of Dicke (1954) on superradiance.

In this Letter, we present results on the realization of a physical situation in which the radiation properties of a mesoscopic physical system (nanostructure) can be significantly changed. We experimentally demonstrate a drastic increase in the rate of radiative process (electron-hole recombination) of a nanoscale system due to simultaneous implementation of the three phys-

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

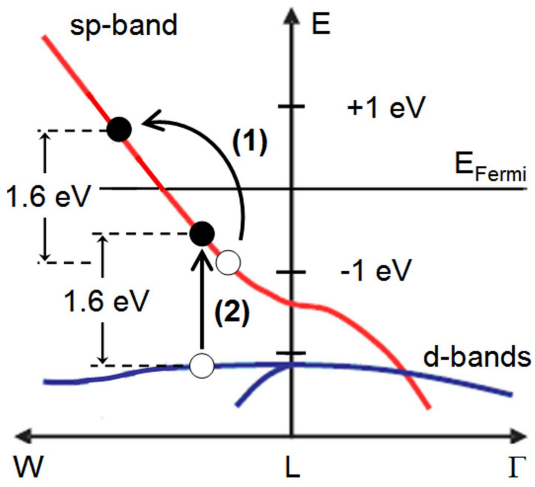

Fig. 1. Schematic of the band structure of Au near the L symmetry point of the first Brillouin zone [11]. Two photons excitation leads to the appearance of a hole in the *d* band. After nonradiative relaxation of the hole from the *d* band and electron from the *sp* band, an electron-hole recombination with spontaneous radiation of a photon (PL) can occur.

ical effects: (1) the size effect, (2) plasmon resonance and (3) the optical Tamm state (OTS). As a rule, all three physical processes listed above affect the radiative capability of a quantum or mesoscopic system separately. An example of a physical system in which the radiative rate may strongly depend on its size is a metal cluster that is capable of emitting photoluminescence (PL) [3]. In addition, as is well known from nanoplasmonics, a radiative system placed in an electromagnetic field can, in turn, enhance its radiative capabilities by means of excitation of plasmon resonances in a metal [4].

The radiative process under investigation is radiative electron-hole recombination in gold, initiated by two photon absorption, known as a *two photon induced luminescence* (2PPL) [5]. As a nanoscale physical system is used a *nanohole "embedded" in the OTS electromagnetic mode*. A nanohole plays role of plasmonic nanostructure, and due to the size effect and excitation of associated localized plasmonic resonance dramatically enhance efficiency of radiative electron-hole recombination in its vicinity. Excitation of local plasmon resonance leads to enhancement of local fields. The physics of size effect is based on realization of high wave vectors due to scattering of light on nano-sized object, needed to realize intraband transitions in 2PPL process [6].

The electromagnetic mode of the OTS plays a role of a resonant cavity mode. Over the last few years, OTSs have been intensively studied both theoretically and experimentally [7–9]. In literature another name for this phenomenon is also used - Tamm plasmon mode [10, 12]. In our research the OTS is realized at the interface between a photonic crystal and a metal. The key role of excitation of the OTS is to enhance a local field at the fundamental wavelength in the vicinity of a nanohole.

The reason to choose a nanohole as the nanoscale object is the following. The optical behavior of a nanohole in a conducting screen can be directly related to the properties of a nanoparticle (with the use of the Babinet principle) and characterized by the corresponding (magnetic and electric) dipole moments and can be modeled as a nanoscale nonlinear oscillator [13]. The basic advantage of the former over the latter is the implementation of a background-free light measurement, since, in the case of a nanohole, there is no background from the excitation ra-

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

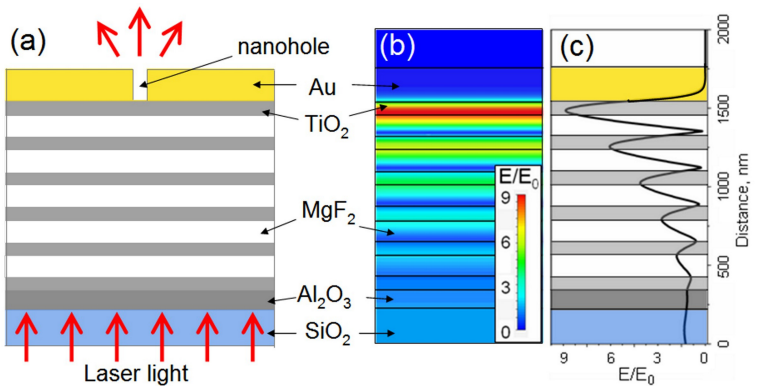

Fig. 2. (a) Schematic representation of the object of study, "PC-metal nanofilm", (b) computer simulation by the FDTD method of the spatial distribution of the electromagnetic field of the OTS excited at its resonance wavelength λ = 780 nm, and (c) its cross section.

diation, whereas, in the case of a nanoparticle, the PL is always accompanied by the intense excitation radiation. In addition, the damage threshold for nanoholes is always higher than for complementary nanostructures [14].

Photoluminescence in metals is an important physical process occurring in the interaction of radiation with metals, and its physical nature is determined by corresponding interband and intraband transitions [6, 15, 16]. For the first time 2PPL process was investigated from roughened metal surfaces [5]. Today one of the main physical reasons for interest in 2PPL in Au is the possibility to use Au nanoparticles as theragnostic biomarkers, since they provide at the same time the possibility for deep tissue imaging as well as for local heating in photothermal therapies [17–19].

Absorption of radiation in a metal with subsequent PL can occur only in a region with a thickness of about the skin-layer, i.e., on a nanosized scale. Possibility to excite local plasmonic oscillations in nanostructures made it possible to increase the efficiency of PL from nanoobjects. As a result the PL efficiency from spherical nanoparticles is about 10^{-6}, from nanorods can reach a value of 10^{-4} [6, 16]. Recently we have shown that the PL process from nanostructures in the form of nanohole made in Au film has the same efficiency as from Au spherical nanoparticles [20]. The physics of this drastic enhancement of PL quantum efficiency in the nanostructures compared to smooth metal films is appearance of a new channel of electron-hole relaxation, namelly excitation of collective electron oscillations which relaxate radiatively [16].

2. Experimental setup and samples

Fig. 1 shows the scheme of the 2PPL process upon excitation of gold by radiation at a wavelength of 780 nm (1.58 eV), which was used in this experiment. The first photon excites an electron via an intraband transition within the *sp* conduction band. The second photon excites an electron from the *d* band, and it recombines with the *sp*-hole in the conduction band. Subsequently, the hole created in the *d* band as a result of the two - photon process recombines with electron in the *sp* band, implementing the PL process [21, 22].

In a separate experiment we have found that quantum efficiency of the 2PPL from Au film with nanoholes depends on nanohole's diameter. The maximum efficiency of the process is realized for nanoholes with diameter about 60 nm (Fig. 3). This result corresponds to known size dependence of the 2PPL emission from metal spherical nanostructures: for nanostructures with diameter bigger than 60 nm the efficiency of the 2PPL process is damped due to retardation

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

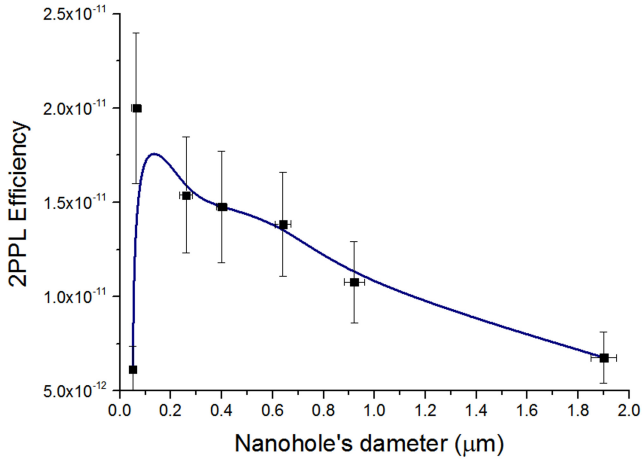

Fig. 3. The efficiency of 2PPL emission measured from nanoholes of different diameters made in 200 nm thick Au film on a quartz substrate.

effects, while for smaller diameters it is damped by excitation of the Auger plasmons [23].

To realize the main idea of the paper we chose the nanostructured object with the following parameters: (1) it is a nanohole with a diameter of 60 nm in a gold film with a thickness of 200 nm; (2) a nanohole with its plasmon resonance frequency that is close to the 2PPL frequency [16]; (3) the nanostructure is "embedded" in an electromagnetic field of an optical Tamm state [20, 24]; (4) the laser radiation frequency exciting 2PPL in the nanohole corresponds to resonance excitation of the OTS.

Schematically, the arrangement of the object of study is shown in Fig. 2(a). On a quartz substrate was built a photonic crystal (PC). The PC is a periodic dielectric structure: a 12-layer stack of alternating high/low-index dielectric layers [24]. A PC was coated on one side with an optically thick 200-nm Au layer. Deposition of the gold film allowed to create the optical Tamm state at the interface "PC-metal nanofilm" (the OTS resonance wavelength equals to 780 nm), Fig. 2(b). The chosen optical thickness of the Au film makes it possible to realize light reflectance at the PC/Au interface about 98%, which is necessary to form microcavity with $Q\sim100$, as well as strongly decrease light transmission trough the Au film. The OTS makes considerable enhancement of the amplitude of the light field incident on the system "PC-metal nanofilm" [24]. Thereafter a nanosized object, nanohole, is created in the gold nanofilm. As follows from works [24,25], nanostructuring of the film does not destroy OTS. Upon irradiation of a system "PC-metal nanofilm" by resonant optical radiation, an OTS arises on the inner surface of the metal film [24] and the nanohole became "embedded" in the OTS electromagnetic mode (as can be seen from Fig. 2).

Quantitative analysis based on comparison of 2PPL from smooth and nanostructured metal film (as well as rough metal films [26]) predicts that 2PPL efficiency from nanostructured metal film will be in 10^3 - 10^6 times higher compared to smooth metal film. Placement of the nanostructured metal film in to the OTS mode should further enhance the 2PPL yield, since local field amplitude at the fundamental wavelength is increased if the OTS is excited. Our calculations of electric field enhancement in the OTS without nanoholes (Fig. 2), with nanoholes [25] and measurements of transmission through nanoholes embedded in the OTS [24] show that in this case the electric field amplitude at the photonic crystal/metal interface is increased by about 4.5 times in compare with incident field. The 2PPL intensity is proportional to E^4, where E is the local electric field amplitude enhancement [21]. Hence excitation of the OTS should enhance

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

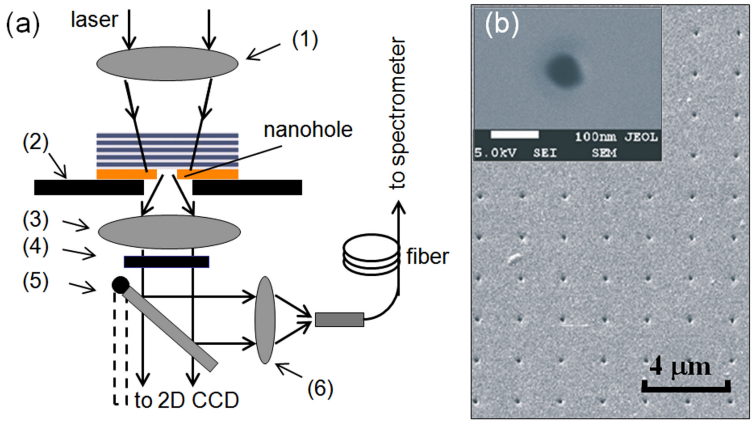

Fig. 4. (a) Setup for microscopic imaging and transmission spectroscopy: (1) 10x/0.25 objective that focuses excitation radiation, (2) 2D piezo stage with feedback sensors, (3) 40x/0.65 PL - collecting objective, (4) interference filter to cut off the excitation radiation, and (5) flipping mirror. (b) SEM image of a nanohole array made in an Au layer of a PC. An enlarged image of one nanohole is shown in the inset.

2PPL radiation process in about 400 times. Thus we estimate 10^5 - 10^8 - fold enhancement of the 2PPL for nanostructured metal film placed in the OTS mode compared to the free standing smooth metal film.

Figure 4(a) shows the schematic of the experimental setup for the laser excitation, microscopy, and spectroscopy of a nanohole "embedded" in an OTS field. A laser pulse of sufficiently long duration is required for the OTS to be established [27]. In the experiment, we used the laser pulse with a duration larger than the time of the OTS formation and it was τ =1.2 ps. The spectrum width was less than 2 nm, and the central frequency can be tuned in the spectral range 700 - 900 nm [27]. The laser light was focused into a spot with a diameter of 4 μm on the surface of a film with nanoholes. Peak intensity of laser radiation on the sample was 1.2×10^8 W/cm^2. This value is two orders of magnitude smaller than measured threshold intensity of nanoholes damage.

Microscopic and spectroscopic studies of a single nanohole were performed using a Nikon Eclipse Ti inverted microscope. The 2PPL from the nanohole was collected by a 40x microscope objective (NA = 0.65); its spatial distribution was measured with a 2D cooled CCD camera with avalanche gain (Hamamatsu, 9100-13) and its spectrum - by using a spectrometer equipped with a cooled CCD array. To suppress the excitation radiation, an interference notch filter as well as color filter were installed before the CCD array. Thus signal from excitation radiation was reduced by more than 16^{th} orders of magnitude.

At first we used two samples to study 2PPL from smooth Au surface: (1) 200 nm thick monocrystalline Au film [28], (2) 50 nm thick Au nano layer (50 nm) that was deposited on an ultra smooth SiO_2 surface. Measurements with monocrystalline Au film was carried out in the reflection microscopy mode. The Au nanolayer of 50 nm thickness possess high transmission of light (about 10%) at the frequency of PL, so that 2PPL can be detected in the transmission microscopy mode. Based on our measurements, we estimate the efficiency of the 2PPL process from the smooth gold surface to be lower than 10^{-16} for both samples. The tiny efficiency of 2PPL from the smooth gold nanofilm is explained by the existence of the dipole forbiddance for intraband transitions [26].

Then we prepared two samples with nanoholes in Au film: (1) an Au nano layer (200 nm)

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

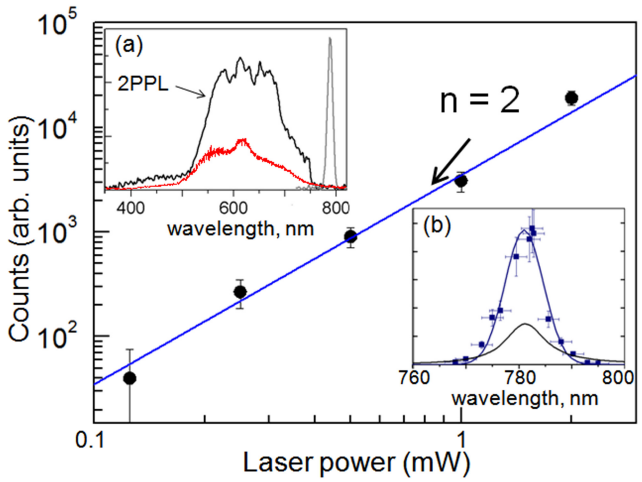

Fig. 5. Power dependence of the multiphoton PL emission measured from a single nanohole in Au nanofilm. The inset (a) presents: (i) the PL spectrum from the nanohole (black line); (ii) excitation laser (gray line), and (iii) transmission spectrum of nanohole (red line). The inset (b) presents measurements of spectral transmittance of laser light (black curve) as well as 2PPL signal (blue curve) from a single nanohole in "PC-metal nanofilm".

that was deposited on a SiO_2 substrate; (2) an Au nano layer (200 nm) on the PC. The arrays of nanoholes were produced by a focused ion beam. The spacing between nanoholes was 2 μm (Fig. 4(b)). An electron image of one of the 2D array of nanoholes is shown in the inset of Fig. 4(b). The nanoholes had a circular shape with a diameter of 58 ($\pm$5) nm. We note that the chosen distance between nanoholes is enough to neglect excitation by a sharply focused laser light a surface plasmons coupled to the array with nanoholes [24].

3. Experimental results

At first the measurement was made for a characterization of PL. Measurements of the dependence of the multiphoton PL signal on the power of the excitation laser radiation are shown in Fig. 5. It follows from the Figure that the multiphoton PL can be approximated well by a power dependence with a coefficient n = 2. The inset (a) of Fig. 5 presents the measured spectrum of 2PPL from a single nanohole made in Au film on a quartz substrate without the PC (black curve). The right narrow peak corresponds to the excitation radiation (gray curve). The PL peak is located at a wavelength of 610 nm. Its spectral position is determined by the nanohole plasmon resonance and the energy of the band gap of gold near the L symmetry point of the first Brillouin zone [21]. Excitation of localized plasmon resonance on the wavelengths of the 2PPL emission was confirmed by measuring transmission of light through the nanohole (red curve in Fig. 5(a)). Then we made measurements of dependence of 2PPL luminescence on laser radiation wavelength from both samples with nanoholes. The resonant dependence was found only for the sample *with a photonic crystal*. The inset (b) of Fig. 5 presents measurements of spectral transmittance of laser light (black curve) as well as 2PPL signal (blue curve) from a single nanohole in "PC-metal nanofilm". As can be seen from the figure, the efficiency of 2PPL process dramatically depends on excitation laser wavelength and has a maximum at the wavelength of the OTS, where the maximum transmittance of light through the nanohole is realized [24].

Figure 6 demonstrates a giant enhancement of 2PPL emission from nanohole. Figure 6(a)

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

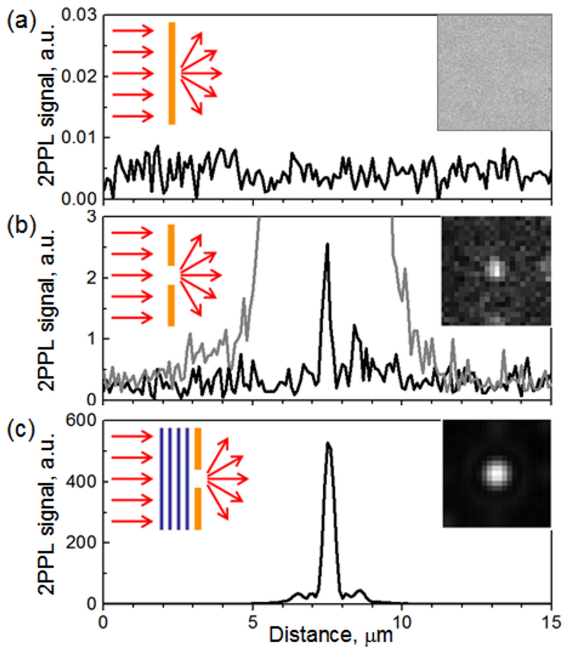

Fig. 6. 2PPL from a nanofilm and a single nanohole at identical parameters of laser irradiation (λ = 780 nm, P = 1 mW): (a) 2PPL from a nanofilm; (b) 2PPL from a nanohole in a metal film (black curve) and 2PPL from a nanohole in a "PC-metal nanofilm" (gray curve); (c) 2PPL from a nanohole in a "PC-metal nanofilm" in full scale. The insets show the schemes of excitation of nanoholes by the laser radiation and corresponding two - dimensional images.

shows results of measurements of 2PPL from an extremely smooth gold film. As can be seen there is no measurable 2PPL. Figure 6(b) show results of similar experiments for 2PPL measurement from a gold film with a nanohole. In this case a nanoobject (a nanohole) is created in the metal nanofilm, and, as a result, the size effect and the plasmon excitations lead to a strong enhancement of the 2PPL. The 2PPL efficiency was measured to be 2×10^{-11}. This value is as much as 2×10^5 higher than the measured 2PPL yield from smooth Au film and is in good accordance with theoretical predictions [26]. Figure 6(c) shows results of the experiments for 2PPL measurement from a nanohole that was placed inside OTS - the case of "PC-metal nanofilm". Placing of the nanohole in the OTS mode leads to a dramatic, 200 - fold, enhancement of the 2PPL signal compared to that from the nanohole in the gold film. This increase is mainly due to the local enhancement of the excitation field and is in a good correspondence with the numerical simulations of the electric field amplitude shown in Fig. 2. To emphasize this considerable increase in the 2PPL signal, in Fig. 6(b), along with the 2PPL signal from the nanohole in the gold film (black curve), the 2PPL signal from the nanohole in the "PC-metal nanofilm" is also presented (gray curve). For correct quantitative analysis of Fig. 6 it is important to take into account that signals of Fig. 6(b) and Fig. 6(c) corresponds to a diffraction limited spot ($\sim$ 600 nm in diameter). This radiation emits from a nanohole of 60 nm diameter. While signal of Fig. 6(a) is result of luminescence from a large surface of Au, determined by diameter of excitation laser light spot.

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015

4. Conclusion

Thus, in this work we demonstrated an enhancement of the radiative capability of the nanoobject when the three physical effects: the size effect, the plasmon resonance, and the optical Tamm state are simultaneously realized. The total increase in the efficiency of the 2PPL process is about 10^7. The maximum realized efficiency of 2PPL process is still small in compare to other emitters (such as molecules, quantum dots). But the gold nanostructures as emitters of light have important advantages: (1) optical stability (absence of blinking and bleaching), (2) fast optical response on a fs and sub fs timescale, compared to ns time scale response for molecules and quantum dots, (3) there is no saturation of photon flux in comparison to molecules and quantum dots, (4) a broad emission spectrum, (5) they can be produced by conventional nanolithography approaches. We note a possible practical importance of our work for the creation of nanolocalized broadband laser radiation sources [20].

Acknowledgments

The authors would like to thank A. Kamchatnov and S. Lukishova for helpfull discussions. This work was partially supported by the Russian Foundation for Basic Research (projects 12-02-00784, 13-02-01281), by the Program ”Extreme Light Fields” of the Presidium of the Russian Academy of Sciences, by the Grant of the Government of the Russian Federation for the Support of Scientific Investigations under the Supervision of Leading Scientists (Contract No. 14.B25.31.0019), and by the Russian Scientific Foundation (Contract No. 14-12-00729). This work was produced using equipment at the MIPT Center of Collective Usage (CCU MIPT) with the financial support from the Ministry of Education and Science of Russian Federation (grant id RFMEFI59414X0009).

#226544 - $15.00 USD Received 7 Nov 2014; revised 30 Jan 2015; accepted 18 Mar 2015; published 23 Apr 2015