Single nanohole and photoluminescence: nanolocalized and wavelength tunable light source

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Abstract: We are first to demonstrate a broadband, nanometer-scale, and background-free light source that is based on photoluminescence of a single nanohole in an Au film. We show that a nanohole with a diameter of as small as 20 nm in a 200-nm thick Au film can be used for this purpose. Further development of the localized source that involves the use of a photon-crystal microcavity with a Q-factor of 100 makes it possible to create a 30-fold enhanced, narrowband tunable light source and with a narrow directivity of the radiation.

OCIS codes: (230.6080) Optical devices, sources; (130.0250) Integrated optics, optoelectronics; (310.6628) Subwavelength structures, nanostructures; (050.1220) Apertures; (350.4238) Nanophotonics and photonic crystals; (310.4165) Multilayer design; (260.2710) Inhomogeneous optical media.

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#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012

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1. Introduction

During the last decade, considerable effort of the physical community is devoted to the elucidation of the physical origin of new properties of nanoobjects, such as (i) the interaction process of elementary excitations inside of nanoobjects; (ii) the coupling with radiation; (iii) the coupling with the environment (matrix, absorbed molecules, and other nanoobjects). Basically and technologically, this interest is motivated by the fact that nanoobjects are a new class of objects with new properties for applications.

Photoluminescence of nanoobjects from noble metals is precisely one of the examples of these unusual and unexpected properties. Interaction of light with a metal nanoobject is mainly governed by two basic physical processes: (1) free-electron effects and (2) collective oscillations [1]. Until recently, the third physical process that is involved in the interaction of radiation with the metal, namely, interband transitions, played an insignificant role. In 1969, Mooradian [2] revealed interband transitions for the first time in the course of his observations of photoluminescence on the surface of gold. The phenomenon of photoluminescence was actively studied in various metals [3]. One of the principal motivations of interest in photoluminescence was and remains the possibility of studying the band structure of metals [3]. The possibility of technological applicability of photoluminescence remained unlikely because of extremely low photoluminescence efficiency: optically excited metal surfaces show either no or very low luminescence, with about 10^{-10} of quantum efficiency of the process [2].

In noble metals, the photoluminescence extends from the infrared wavelength range to the deep ultraviolet [4]. The origin of this extremely broad luminescence spectrum lies in the nature itself of the electron–photon absorption mechanism by which the metal absorbs the radiation. In noble metals, there are two physical mechanisms that are responsible for the photoluminescence. The first of them is an interband transition, as a result of which an electron is excited from the valence band to the conduction band [2]. In this case, the photoluminescence in a metal can be described as a three-stage process: (i) the excitation of electrons to generate electron–hole pairs, (ii) the scattering of electrons and holes with partial energy transfer to the phonon lattice, and (iii) the electron–hole recombination that results in the photon emission. The second mechanism is an intraband (free-electron) transition, in which the electron excitation is described in terms of the model of radiation absorption by a quasi-free electron: one photon is absorbed by an electron in the presence of a third particle (to conserve energy and momentum) [5].

#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012

The radiation absorption and subsequent photoluminescence in a metal can occur only within a region on the order of a skin layer, i.e., on a nanoscale level; therefore, with the development of the technology of creation of nanostructured objects, interest in photoluminescence was rekindled but now with respect to nanosized structures. Further experiments demonstrated the phenomenon of an extreme increase in the quantum efficiency of photoluminescence from nanoobjects. The measured photoluminescence efficiency for spherical nanoparticles is on the order of 10^{-6} [6]. The measured value of this parameter for nanorods can be as high as 10^{-4} [7]. The efficiency of photoluminescence from molecular clusters can reach tens of percent. The mechanism by which the photoluminescence efficiency of Au nanoobjects extremely increases consists of the d–sp interband electron excitation and the subsequent nonradiative recombination of excited d-band holes with sp electrons, as a result of which particle plasmons are created. These plasmons subsequently emit photons, giving rise to the photoluminescence [6]. A further increase in the photoluminescence efficiency is possible in nanoobjects in the shape of nanorods, and it is referred to as the process of enhancement of incoming and outgoing electric fields via coupling to the surface plasmon resonance in the rods [7,8].

This work is devoted to the study of photoluminescence from a nanoobject in the shape of a single nanohole. In the work, we will demonstrate that the nanohole exhibits (1) an extremely broad photoluminescence band and (2) a considerable photoluminescence quantum yield; furthermore, we will show that (3) the combination of these two physical properties opens up the possibility of constructing of a *nanolocalized* broadband and wavelength tunable light source. We will also demonstrate the operation of this source using a microcavity based on a 1D photonic crystal.

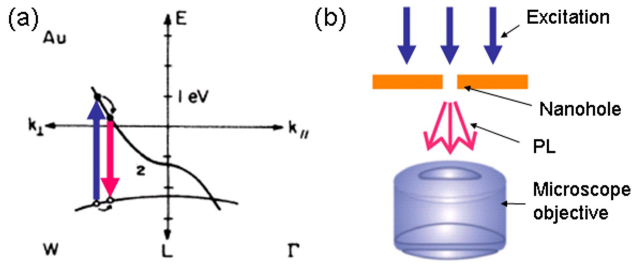

Fig. 1. (a) Schematic band structure of Au at the high-symmetry point L in Au close to the Fermi surface [3] showing the electron excitation and photoemission; (b) sketch of an experimental scheme for the photoluminescence excitation in gold in a close vicinity of the nanohole and for the collection of the photoluminescence radiation.

The choice of an object in the shape of a nanohole rather than a nanodisk as a nanolocalized radiation source has a considerable advantage. For thin PEC film it can be analized by taking advantage of Babinet's principle. According to Babinet's principle, reflected fields of a disk can be directly related to the properties of transmitted fields of its complementary hole [9,10]. The behavior of a nanohole in a conducting screen can be directly related to the properties of a nanodisk. The basic advantage of the former over the latter is the implementation of a background-free light source, since, in the case of a nanohole, there is no background from the excitation radiation, whereas, in the case of a nanoparticle (which, as a rule, is deposited on a dielectric surface), the photoluminescence is always accompanied by the intense excitation radiation and considerable background scattering from the surface. In addition, the damage threshold for nanoholes is always higher than for complementary nanostructures [11,12], because the heat removal of a metal film in which a nanohole is made is more efficient, which makes it possible to use substantially higher intensities of the excitation radiation.

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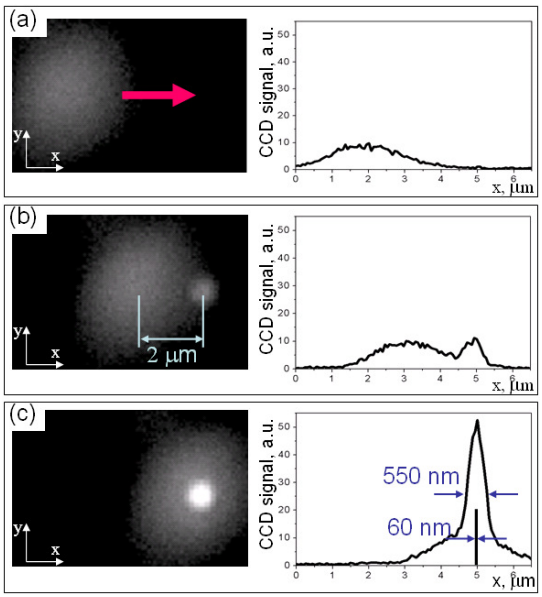

Fig. 2. Optical image of a nanohole excited by laser light: (a) nanohole is located 3 μm away from the center of the laser light spot (the arrow shows the location of the nanohole); (b) nanohole is located 2 μm away from the center of the laser light spot; and (c) the laser beam spot is superimposed directly on the nanohole.

Development of a single nanohole as a light source is of central interest in basic and applied nanosciences [13,14], since, in experiments performed on large ensembles, all parameters fluctuate from object to object, and only averaged properties are observed. This strategy enables us to demonstrate *spatially nanolocalized broadband and enhanced narrowband wavelength-tunable light sources*.

2. Experimental setup

The schematic of the setup that was used in experiments on the study of the photoluminescence from a single nanohole and construction of a nanolocalized light source on its basis is shown in Fig. 1. The experiment was performed as follows. A diode laser (λ = 406 nm, 3.04 eV photon energy) was used to excite d–sp interband transitions in a gold nanohole (Fig. 1(a)). The excitation beam with a power of 22 mW was focused by a 20 × /0.45 objective into a spot of 830 nm diameter (FWHM) on a sample with a nanohole that was arranged on the stage of an inverted microscope (Nikon Eclipse Ti). The photoluminescence radiation from the opposite side of the Au film with respect to the excitation beam (Fig. 1(b)) was collected with a 40 × /0.6 objective of the Nikon microscope and was analyzed with either (1) a 2D cooled CCD camera with avalanche gain (Princeton Instruments), or (2) a grating spectrometer with a high optical efficiency that was coupled to another cooled CCD camera (Princeton Instruments). A barrier interference optical filter with the spectral range 480–1100 nm was used to suppress the signal from the excitation beam. The experimental setup allowed obtaining a 2D optical image of the localized light source formed by a single nanohole with a spatial resolution better than 1 μm in a wavelength range from 480 nm to 825 nm. The samples preparation and the measurements were performed in a Class 100 cleanroom.

Nanoholes were formed by a focused ion beam (FEI Quanta 200 3D) in a gold film (with a thickness of 50, 100, or 200 nm) that was deposited on an ultrathin (40-nm thick) freestanding low-stress SiO_2 membrane [15]. The size of the nanohole was in a range from 20 nm to 1 μm. It should be noted that, in this work, measurements were performed with individual holes. Each hole had its own marker, which made it possible to find the required hole on the gold film and to perform measurements with it. A minimal spacing between holes was about 5 μm.

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The shape and the size of each individual nanohole were measured with a spatial resolution of about 5 nm using a JEOL JSM-7001F electron microscope.

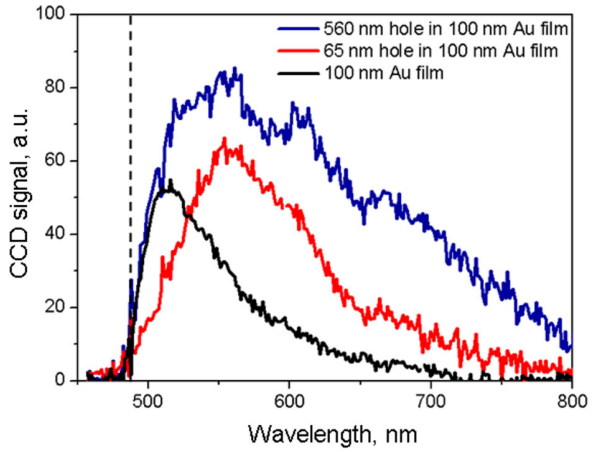

Fig. 3. Photoluminescence spectra from a 100-nm thick bare Au film (black) and from two holes of 65 (blue) nm and 560 nm (red) in diameter created in the same film.

The use of an ultrathin membrane as a substrate for the gold film has the following principal advantages: (1) the surface of the gold film that is adjacent to the membrane is smooth because of a high quality of the membrane surface [16], and (2) since the thickness of the membrane is small, the measured photoluminescence signal from the material of the membrane is three orders of magnitude lower than the measured luminescence signal from the smooth gold surface.

The sample was illuminated by the laser radiation from the side of the membrane. With this illumination method, surface irregularities on the rear side of the gold film do not form photoluminescent «hot spots», since they are exposed to the laser radiation that is strongly weakened by the gold film. This scheme of excitation makes it possible to considerably reduce the background photoluminescence signal from surface irregularities of the gold film.

3. Photoluminescence of individual nanohole

Figure 2 shows optical images of a nanohole with a diameter of 60 nm in a gold film 100 nm thick. Figure 2(a) shows an optical image for the case where the excitation laser radiation is focused onto the surface area of the gold film outside the nanohole (in this experiment laser was slightly defocused to a spot with a diameter of about 2 μm on the gold film surface). The center of the laser spot lies at a distance of 3 μm away from the nanohole. The arrow in the Fig. 2(a) shows the position of the nanohole. The light spot in the Fig. 2(a) with a diameter that is equal to the diameter of the laser beam corresponds to the photoluminescence from the gold film. Since the nanohole is not illuminated by the laser radiation, there is no photoluminescence signal from it. As the laser beam approaches the nanohole, a light spot is appeared in the optical image (Fig. 2(b)), which corresponds to the photoluminescence from the nanohole. This light spot is diffraction limited. Figure 2(c) corresponds to the case of exact focusing of the laser spot onto the nanohole. It can be seen from this figure that the photoluminescence signal from the nanohole dominates over the photoluminescence signal from the surface of the gold film. When comparing the photoluminescence signals from the nanohole and the surface of the gold film, it is necessary to take into account that the image in Fig. 2(c), which corresponds to the photoluminescence from the nanohole, is diffraction limited. Therefore, the ratio between the photoluminescence efficiencies of the nanohole and the gold film is, in fact, considerably higher compared to that resulted from the mere comparison of the signal amplitudes in Fig. 2(a) and Fig. 2(c).

#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012

We measured the efficiency of the photoluminescence process for holes of different diameters in a gold film with a thickness of 200 nm. In the film of this thickness, the photoluminescence signal from its surface is considerably weakened by the absorption in the film itself, which makes it possible to measure the photoluminescence from the nanohole alone. The efficiency is defined as $\eta = P_{hole} / (I_{laser} \times S_{hole})$, where P_{hole} is the power of the photoluminescence from the nanohole, I_{laser} is the intensity of the excitation laser beam in the vicinity of the nanohole, and S_{hole} is the area of the nanohole. In the experiment, both P_{hole} and I_{laser} were measured in the same optical scheme with the use of a 2D CCD camera. Our measurements showed that, for holes with a diameter from 20 nm to 1 μm, the photoluminescence efficiency is almost the same and is about 10^{-6}, which is approximately 10^4 times higher compared to the photoluminescence from the surface of the bare gold. These data agree well with measurement results for spherical gold nanoparticles [6,17]. The fact supports plasmonic nature of the measured high efficiency of the PL process in nanoholes.

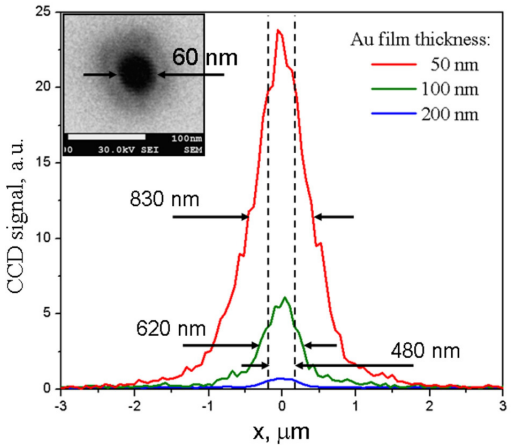

Fig. 4. Cross sections of photoluminescence images of a hole with a diameter of 60 nm created in Au films of different thicknesses: 50 nm (red), 100 nm (green), and 200 nm (blue). The insert shows the electron image of the nanohole.

Figure 3 shows the photoluminescence spectra that were measured for a 100-nm-thick bare Au film and for two nanoholes with diameters of 65 and 560 nm that were prepared in this film. The photoluminescence spectrum of the gold film surface was subtracted from the presented photoluminescence spectra of the nanoholes, since the photoluminescence from surface of the film of the chosen thickness gives rise to a background signal. The vertical dashed line in the figure indicates the cutoff wavelength of the barrier filter. The measured photoluminescence spectrum represents a supercontinuum in the wavelength range from 480 to 800 nm. A slight difference between the measured amplitudes of the photoluminescence signals for the chosen holes, the areas of which differ almost by two orders of magnitude, is explained by sharp focusing of the excitation laser radiation; namely, the effective intensity of the laser radiation focused onto the hole with a larger diameter was significantly lower compared to the hole with a smaller diameter.

From Fig. 3, it can be clearly seen that the photoluminescence spectra of the nanoholes distinctly differ in shape from the spectrum of the gold film surface, which means that the mechanisms of formation of photoluminescence signals from the nanoholes and from the surface are different. In the gold film, photoluminescence arises as a result of the excitation of an electron–hole pair with its subsequent recombination and photon emission event [2]. For nanoobjects, their photoluminescence arises upon excitation of interband transitions near the nanohole, which leads to the excitation of localized plasmons and corresponding photoluminescence radiation [6]. As can be seen from this figure, a maximal value of the

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photoluminescence signal for the nanoholes corresponds to a plasmon resonance in gold ~550 nm (2.2 eV). For the bare gold film, the peak energy of luminescence photons is related to the energy separation between d holes and the Fermi surface, which, in the proximity of the X symmetry point of the first Brillouin zone in gold, is roughly 690 nm (1.8 eV), and, near the L symmetry point [3], is 510 nm (2.4 eV).

For the future developments a femtosecond radiation can be used for excitation. In this case the photoluminescence spectrum from a nanohole can be spectrally even broader compared to the spectrum shown in Fig. 3. Strongly confined fields near a nanohole illuminated by laser radiation have wave vectors that are comparable with wave vectors of electrons in gold. Therefore, large field gradients that arise give rise to high-order multipolar transitions, which initiate sp-intraband electron transitions, which, in turn, are accompanied by spectrally broad infrared radiation that is extended [5] to the long-wavelength range up to ~1100 nm. The use of femtosecond radiation for the nanohole excitation makes the nanolocalized source with ultra short duration [18].

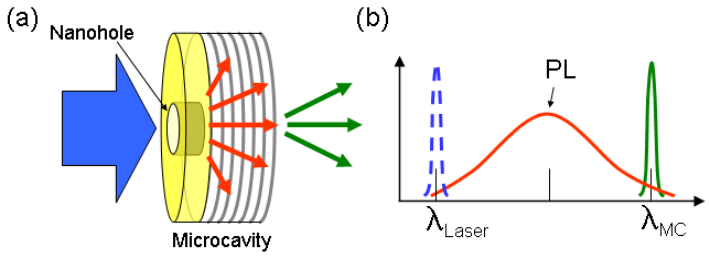

Fig. 5. (a) Sketch of an experimental scheme for the photoluminescence excitation in single nanohole in a gold film, comprising the last layer of photonic crystal microcavity, (b) schematic spectra of excited laser light (blue), photoluminescence of nanohole in Au film without microcavity (red), radiation from single nanohole imbedded in microcavity (green).

We performed investigations that were aimed at determining the degree of possible spatial localization of a source on the basis of photoluminescence. Figure 4 shows the effect of the gold film thickness on the degree of localization of a light source formed by a nanohole with a diameter of 60 nm. In this figure, the cross sections of 2D images of nanoholes in films with a thickness of 50, 100, and 200 nm are presented. For the 50-nm film, the spatial size of the photoluminescence region from the nanohole is comparable with that from the surface; i.e., this photoluminescence source is localized in the area of the laser radiation focal spot. For the 100-nm film, the photoluminescence signal from the nanohole dominates over the corresponding signal from the film surface; however, the latter forms a rather broad background. The nanohole in the film with a thickness of 200 nm enables implementation of a nanolocalized light source that is free from background, since, in this case, the gold film is optically thick, and this leads to a substantial weakening of the photoluminescence signal from the surface.

4. Photoluminescence of individual nanohole inside a photonic crystal microcavity

As is well known from the laser physics, a broad photoluminescence spectrum of active laser media makes it possible to generate narrowband and, simultaneously, tunable radiation. In this work, we developed a scheme for controlling the spectrum and the efficiency of a nanolocalized light source using a 1D photonic crystal (Fig. 5). Deposition of a photonic crystal onto the gold surface gives the possibility to create a photonic-crystal microcavity [19] (PCM) with a Q-factor of about 100. In this case the photoluminescence source (based on nanohole in Au film of the PCM) is brought inside the microcavity. The radiation of the localized light source at a PCM resonant wavelength is amplified in the microcavity, whereas the radiation at all other wavelengths is weakened by the 1D photonic crystal bandgap.

A PCM is formed by an optically thick Au film and a 1D photonic crystal. The PCM design consisted of a 12-layer stack of alternating high-index (TiO_2, n = 2.23) and low-index

#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012

(MgF_2, n = 1.38) dielectric layers with a thickness of λ/4n (λ = 730 nm) that were created on an optically thick 200-nm Au layer. The 12-layer stack of the dielectric layers formed a 1D-type photonic crystal, which ensured a low (about 2%) transmission of light in the spectral range between 650 and 800 nm (the bandgap of the photonic crystal).

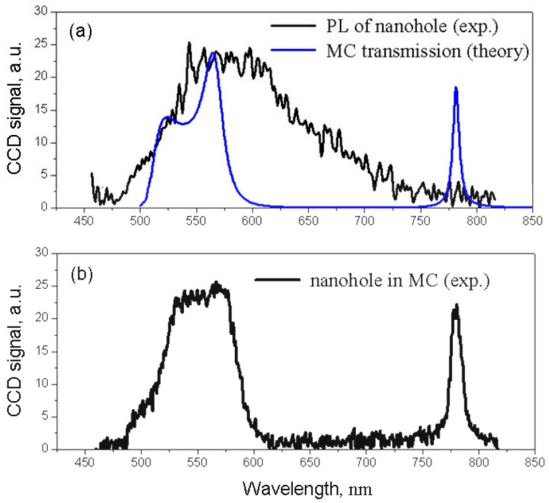

Fig. 6. (a) Photoluminescence spectrum (black) of a 60-nm nanohole in a 200-nm thick Au film without a PCM and calculated transmission spectrum (blue) for the nanohole incorporated into the PCM; (b) the measured photoluminescence spectrum of the nanohole incorporated into the PCM.

We demonstrate that the spectrum and the efficiency of the created localized light source can be controlled by coupling the nanohole's photoluminescence to the photonic crystal microcavity. The excitation laser impinges on a 60-nm nanohole, forming a localized photoluminescence source inside the PCM. Figure 6(a) presents the measured photoluminescence spectrum from a nanohole with a diameter of 60 nm that was prepared in a gold film 200 nm thick in the case where there is no photonic crystal (black curve) and the calculated transmission spectrum of a nanohole that was incorporated into a photonic crystal (blue curve). The transmission spectrum was calculated for a plane monochromatic wave that was incident normally on the film. A sharp resonance in the calculated spectrum at λ_{PCM} = 782 nm is a consequence of the cavity formation with the quality factor $Q = \omega / \Delta\omega$ of about 120. The dip in the spectrum in the wavelength range 650–750 nm is a consequence of weakening the radiation by the band gap of the 1D photonic crystal.

Figure 6(b) shows the photoluminescence spectrum that was measured from a nanohole in a gold film on which a photonic crystal was created. The spectrum has a resonance peak at a wavelength of λ = 782 nm, which corresponds to the resonance wavelength of the PCM. It is remarkable that, in the photoluminescence spectrum for the nanohole in the Au film without the photonic crystal (Fig. 6(a)), the signal at λ_{PCM} = 782 nm is almost at the noise level, while the use of the photonic crystal substantially increases the signal.

Figure 7 shows 2D optical images of a nanohole with a diameter of 60 nm in a gold film and of a nanohole with the same diameter in a gold film incorporated in the photonic crystal. The images were obtained at a wavelength of the resonance of the PCM using a bandpass filter at 780 nm and the transmission bandwidth of 10 nm. Comparison of the signals in Fig. 6(a) and Fig. 6(b) shows that the use of the photonic crystal makes it possible to enhance the photoluminescence signal from the nanohole by a factor of about 30. We'd like to note that intraband mechanism is dominant in the photoluminescence emission of Au at around 780 nm [5].

#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012

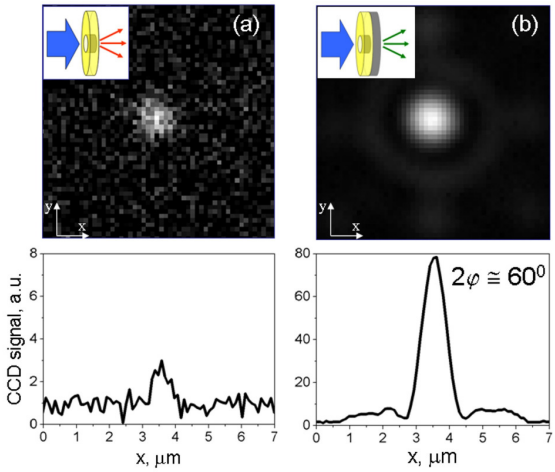

Fig. 7. 2D optical images of a 60-nm nanohole measured at the PCM resonance frequency: (a) nanohole in a 200-nm thick Au film, (b) nanohole incorporated into the PCM.

Another important consequence of the application of the photonic crystal is the manifestation of the directivity of the photoluminescence radiation from the nanohole at the resonance wavelength of the photonic crystal. The measurements of the amplitude of the photoluminescence signal from the nanohole with the photonic crystal performed with the use of objectives with different numerical apertures show that the majority of the photoluminescence at the wavelength λ_{PCM} = 782 nm is concentrated in a cone with an opening angle less than 60 degrees. This directivity is a result of the constructive interference at the PCM's resonance frequency of the photoluminescence, similar to the case of formation of radiation pattern of classical dipole in a cavity [20]. The microcavity transforms the free-space radiative modes on the exit side of the nanohole. Our calculations show that the measured 30-fold enhancement is attributed to increase of directivity of the PL emission from the nanohole. The total flux of the radiation remains the same.

5. Conclusion

In conclusion we have demonstrated that a nanohole in an Au film and electron interband excitations in gold near the nanohole define a nanolocalized background-free supercontinuum source of light that is considerably broader than spectrum of the source based on the use of atoms, molecules and quantum dots. We have shown that the use of a 1D photonic crystal with a nanohole in an Au film makes it possible to create a narrowband enhanced photon source that can be tuned over a wide spectral range. The use of the two-photon excitation of photoluminescence [21] in the nanohole can enhance the efficiency of the source of this type and enrich its short-wavelength part of spectrum. Another important perspective of future developments of the source is use of slits made in metal film instead of nanoholes. Nanoslits in metal film have higher polarizability compared to circular nanoholes, hence the quantum efficiency of the PL process in slits can be substantially higher comparing to nanoholes, in analogy to measurements of the PL process performed with Au nanoparticles having different geometry [6]. We expect that the use of high-Q cavities for the system «nanohole + photonic crystal microcavity» will substantially enhance effectiveness and directivity of the demonstrated light source which will find applications in nanophotonics, nanoplasmonics and optical microscopy with nanometer resolution [22–28]. New perspectives to control transmission of nanoopenings in thin metal films as well as the PL process come from application of the active hyperbolic metamaterials for loss compensations [29].

#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012

Acknowledgments

This work was partially supported by the Russian Foundation for Basic Research, grant no. 11-02-00804-a, 12-02-00784-a, and by the Program «Extreme Light Fields» of the Presidium of the Russian Academy of Sciences. The equipment of the CKP ISAN, CKP MIPT (project 16.552.11.7022), and REC "Nanotechnology" of the MIPT was used in this work.

#170176 - $15.00 USD Received 11 Jun 2012; revised 16 Jul 2012; accepted 16 Jul 2012; published 10 Aug 2012