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Two-dimensional dynamic photonic crystal creation by means of three non-coplanar laser beams interference in colloidal CdSe/ZnS quantum dots solution

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We demonstrated a simple way to create dynamic photonic crystals with different lattice symmetry by interference of three non-coplanar laser beams in colloidal solution of CdSe/ZnS quantum dots. Two-dimensional dynamic photonic crystal was formed due to the periodical changing of refraction and/or absorbtion of resonantly excited excitons in CdSe/ZnS quantum dots. The formation of dynamic photonic crystal was confirmed by the observed diffraction of the beams that have excited photonic crystal at the angles equal to that calculated for the corresponding two-dimensional lattice (self-diffraction regime).

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1. Introduction. Materials in which the refractive index (dielectric constant) is modulated on a length scale close to the wavelength necessary for operation are called photonic crystals. Multiple interference between waves scattered from each unit cell of the crystal results in photonic bandgap formation – a set of frequencies within which no propagating electromagnetic modes exist [1, 2].

Photonic crystals have demonstrated an attractive potential for numerous devices fabrication [3–5], which, for example, deal with high-capacity data storing and other related applications [6]. Considerable progress has been made in constructing two-dimensional structures with the use of conventional lithography technique [2], but this method does not provide the possibility to produce dynamic photonic crystals. Colloidal crystals may be used as templates to make submicrometer structures [7–12] but the use of closed-packed spheres severely restricts the range of lattices that may be produced [13] and allows almost no freedom to alter the structure of a unit cell. That's why formation of photonic crystals based on the quantum dots colloidal solution is an actual problem of great interest, because one can change the parameters of dynamic photonic crystal (spacial symmetry, unit cell shape and size, crystal dimension) by means of quantum dots concentration and dimensions changing from the one hand and by means of experimental geometry changing from the other hand. Moreover, obtained dynamic structure opens new horizons for careful analysis of relaxation processes in the QDs.

In the present paper we describe a technique, which gives possibility to form two-dimensional tunable dynamic photonic crystals with different lattice symmetry. With this technique we have made microperiodic dynamic semiconductor structure with strong nonlinear changing of refraction and absorbtion and analyzed the self-diffraction processes of three non-coplanar laser beams at the dynamic photonic crystal (diffraction grating). To our knowledge, we demonstrated for the first time that two-dimensional tunable dynamic photonic crystal (diffraction grating) can be formed in the colloidal quantum dots solution due to the periodical changing of refraction and/or absorbtion caused by three non-coplanar laser beams interference. To reach the best uniform contrast of the structure under investigation and for better understanding of the problems, specially raised by the interference of multiple non-coplanar beams we have also performed theoretical calculation of the periodic intensity field in the quantum dots solution.

2. Experimental setup. We used 35 picosecond pulses train from a mode-locked frequency-doubled Nd^{+3}:YAG-laser ($\lambda = 532\,$nm) to irradiate the cell with colloidal solution of CdSe/ZnS quantum dots. Three laser beams were created by splitting the laser output twice with dielectric beam-splitters. Three coherent non-coplanar laser beams with the same linear polarization and equal intensities $I_1 = I_2 = I_3 = I$ attain the cell with colloidal solution of quantum dots simultaneously. The beams geometry is shown schematically in Fig. 1.

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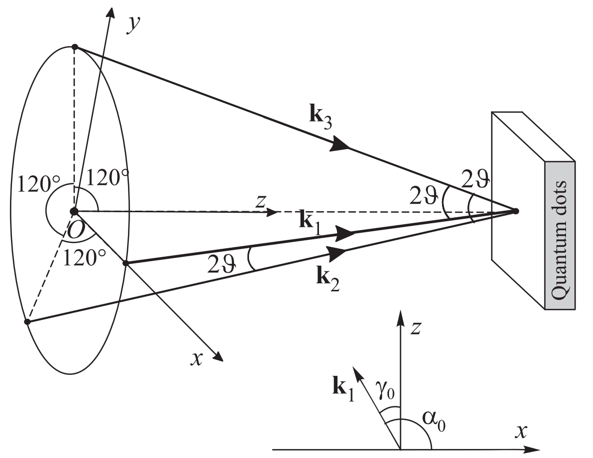

Fig. 1. Beams geometry. Wave vectors of the three laser beams are drawn as arrows

The intensity and linear polarization state of each beam were controlled by means of a half-wave plate and a dielectric polarizing beam-splitter; the polarizer is the last optical element before the cell with colloidal solution of CdSe/ZnS quantum dots [14, 15]. The diffraction pattern formed by transmitted and self-diffracted beams was registered with the help of Nikon $D70$ camera. Colloidal solution was based on the CdSe/ZnS quantum dots with radii about 2.4 nm and dimensions dispersion 20 % dissolved in hexane with concentration $10^{17}\,\mathrm{sm}^{-3}$. Linear absorbtion coefficient of the colloidal solution was $35\,\mathrm{sm}^{-1}$. Peculiarities of three laser beams interaction in the nonlinear optical medium were analyzed in the vicinity of resonant excitation of main exciton transition in CdSe/ZnS quantum dots [16, 17]. The CdSe/ZnS quantum dots colloidal solution was utilized due to the possibility of control under physical properties, which determine nonlinear system response on the resonance laser excitation.

3. Results and discussion. Non-stationary two-dimensional photonic crystal can be formed due to the interaction of three coherent non-coplanar plane waves ($E_i = E_{i0}\cos(\omega t - k_i r + \varphi_{0i})$, $i = 1, 2, 3$) in the colloidal solution of CdSe/ZnS quantum dots due to the spatial modulation of medium optical properties. The intensity distribution of the interference field of three plane waves of the same wavelength λ:

$$I = \sum_j E_{0j}^2 + \sum_{i<j} 2E_{0i}E_{0j}\cos\theta_{ij} \times \times \cos[(\mathbf{K}_i - \mathbf{K}_j)\mathbf{r} + \varphi_{0i} - \varphi_{0j}], \quad i, j = 1\text{–}3, \tag{1}$$

where $E_{0i(j)}$, $\mathbf{K}_{i(j)}$, and $\varphi_{0i(j)}$ are the amplitude, the wave vector and the initial phase of the $i(j)$th plane wave, respectively, and θ_{ij} is the angle between the polarization directions of the ith and jth waves. Due to experimental geometry in our case linear polarization state of each beam was the same, consequently, $\theta_{ij} = 0$ and $\varphi_{0i} - \varphi_{0j} = 0$. Fig. 2 shows calculated plane (see Fig. 2a) and three-dimensional (see Fig. 2b) intensity distribution, which induces hexagonal two-dimensional photonic crystal in colloidal solution of quantum dots.

Evaluated intensity distribution demonstrates that by means of three interacting coherent non-coplanar plane waves with equal amplitudes one can obtain two-dimensional dynamic photonic crystal with the periodicity in two directions, which is determined by the vectors $\mathbf{a}_1$ and $\mathbf{a}_2$ (Fig. 2a). Vectors $\mathbf{a}_1$ and $\mathbf{a}_2$ have equal length, which can be evaluated from the obtained intensity distribution (see Fig. 2):

$$|\mathbf{a}_1| = |\mathbf{a}_2| = \frac{\lambda}{\sqrt{3}\sin\theta}. \tag{2}$$

One can easily prove the presence of two-dimensional dynamic photonic crystal analyzing the results of self-diffraction of the waves, which form non-stationary two-dimensional photonic crystal. Diffraction patterns were registered with the help of Nikon $D70$ camera and demonstrate three transmitted beams holding the direction of incident beams and 21 additional beams. The presence of additional beams can be explained by the self-diffraction of three incident beams at the two-dimensional dynamic photonic crystal, caused by the periodical spatial changing of absorbtion of colloidal quantum dots, which takes place in the interference light field (see Fig. 2). Periodical changing of absorbtion during the resonant single-photon excitation of main exciton transition in colloidal solution of quantum dots by means of second harmonic picosecond pulses can be explained by the presence of competing and coexisting effects of states filling and Stark shift of the exciton absorbtion [18, 19]. Nonlinear absorbtion changing can also be accompanied by the nonlinear changing of refraction [20]. Small red shift of second harmonic wavelength from the resonant wavelength of colloidal quantum dots absorbtion may result in formation of two-dimensional dynamic photonic crystal (phase diffraction grating). It was estimated that induced changing of refractive index in the regions with maximum value of intensity can reach $\Delta n \simeq 10^{-3}$ – enough for formation of two-dimensional non-stationary photonic crystal. The presence of diffraction rings is the direct manifestation of self-diffraction at the induced transparency channel [19].

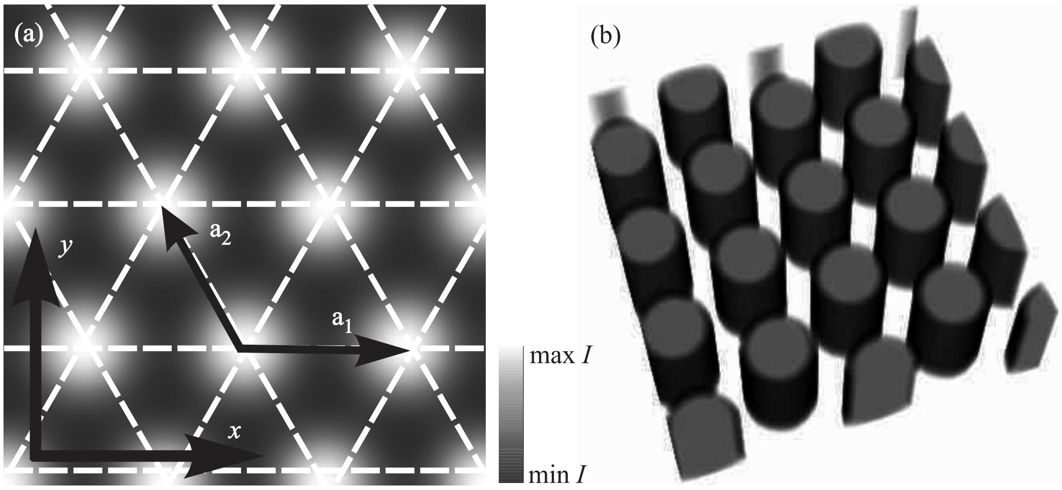

Fig. 2. (a) – Calculated plane surface of three-beam laser interference pattern. (b) – Calculated three-dimensional intensity distribution

m_2 \ m_1	–2	–1	0	1	2	3
–5				61		
–4			46.3		53.3	
–3		46.3		32		61
–2	61		23.6		32	
–1		32		11.6		46.3
0	53.3		11.6		23.6	
1		32		11.6		46.3
2	61		23.6		32	
3		46.3		32		61
4			46.3		53.3	
5				61		

Fig. 3. (Color online) Propagation angles of the laser beams self-diffracted on the induced two-dimensional dynamical photonic crystal for different diffraction maxima $(m_1; m_2)$

To determine the propagation angles of self-diffracted laser beams the Laue method for the two-dimensional diffraction grating was applied:

$$\begin{gathered} c_1(\cos\alpha - \cos\alpha_0) = m_1\lambda, \\ c_2(\cos\beta - \cos\beta_0) = m_2\lambda, \\ \cos^2\alpha_0 + \cos^2\beta_0 + \cos^2\gamma_0 = 1, \\ \cos^2\alpha + \cos^2\beta + \cos^2\gamma = 1, \end{gathered} \tag{3}$$

where α_0, β_0, and γ_0 are the angles, which incident beams form with the axis x, y and z correspondingly; α, β, and γ are angles of diffracted beams, $m_i \epsilon \mathbf{Z}$, $\mathbf{c}_1$ and $\mathbf{c}_2$ are the translation vectors for rectangular unit cell, which can be expressed through the translation vectors $\mathbf{a}_1$ and $\mathbf{a}_2$ for the hexagonal unit cell in the following way:

$$|\mathbf{c}_1| = |\mathbf{a}_1|,$$

$$|\mathbf{c}_2| = 2|\mathbf{a}_2|\cos\frac{\pi}{6}. \tag{4}$$

One can also find from Fig. 1 the following relations between the angles:

$$\beta_0 = \frac{\pi}{2},$$

$$\alpha_0 = \gamma_0 + \frac{\pi}{2},$$

$$\sin\gamma_0 = -\cos\alpha_0 = \frac{2\sqrt{3}}{3}\sin\theta. \tag{5}$$

Consequently, one can obtain the following system of equations for angles α, β, and γ:

$$\begin{gathered} \cos\alpha = \left(\sqrt{3}m_1 - \frac{2\sqrt{3}}{3}\right)\sin\theta, \\ \cos\beta = m_2\sin\theta, \\ \sin\gamma = \sqrt{cos^2\alpha + \cos^2\beta}. \end{gathered} \tag{6}$$

According to obtained equations the possible values of diffraction angles were calculated. The values of angle γ, obtained for $\theta = 10°$ are demonstrated in the

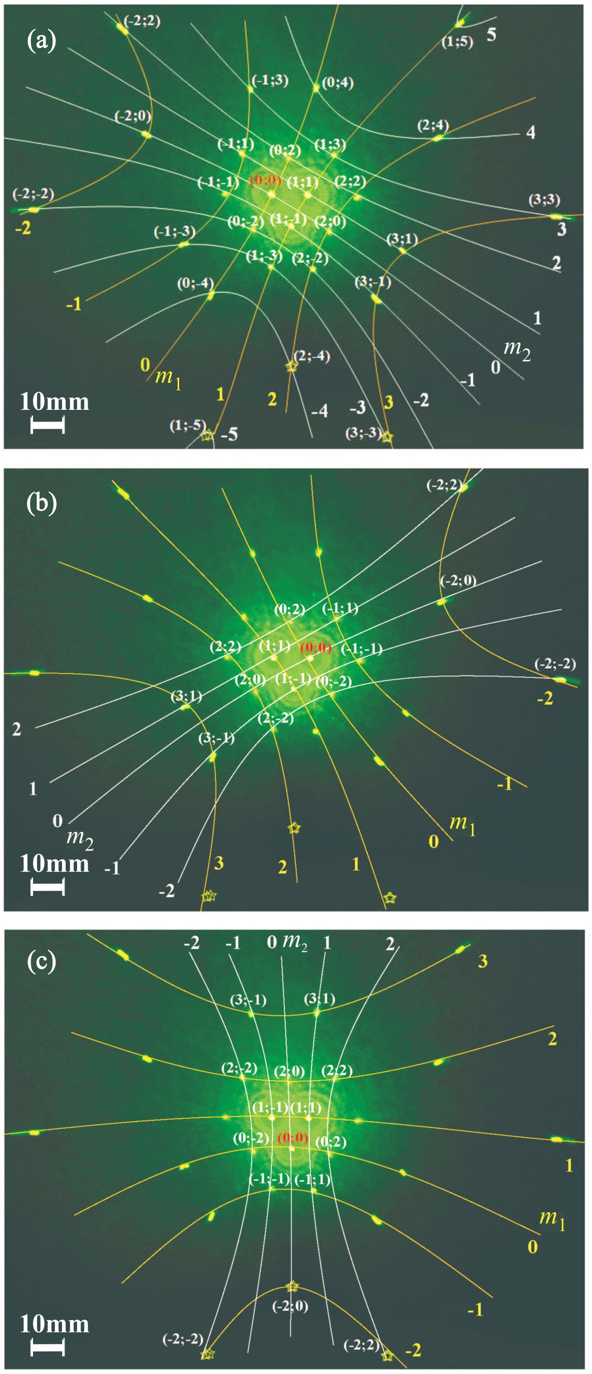

Fig. 4. (Color online) Experimental diffraction partens, obtained for self-diffraction of three laser beams on the induced dynamical two-dimensional photonic crystal. (a) – Diffraction maxima $(m_1; m_2)$ are denoted for the laser beam with wave vector $\mathbf{k}_1$. (b) – Diffraction maxima $(m_1; m_2)$ are denoted for the laser beam with wave vector $\mathbf{k}_2$. (c) – Diffraction maxima $(m_1; m_2)$ are denoted for the laser beam with wave vector $\mathbf{k}_3$. Hyperbolic intersections for all the figures are demonstrated for better diffraction pattern visualization

Fig. 3. Each diffraction maximum is denoted by the couple of indexes $(m_1; m_2)$, which correspond to the self-diffraction order according to axis x and y. One can find empty cells in the Fig. 3. Red cells are empty, because the values of angles α and β, calculated from system of Eqs. (6) do not correspond to the condition (3). For the transmitted waves directions, which correspond to the odd values of $m_1 + m_2$ (yellow cells in Fig. 3), the existence of diffraction maxima are restricted, because waves from the neighboring lattice points of induced two-dimensional photonic crystal have opposite phases, this means that structure factor is equal to zero. For the observed hexagonal induced photonic crystal structure factor is equal to zero, when indexes $m_1 + m_2$ accept odd values.

The values of self-diffracted beams angles were obtained from the experimentally measured diffraction patterns (see Fig. 4) with the help of the expressions:

$$\alpha = \arcsin\sqrt{\frac{L^2+b^2}{L^2+a^2+b^2}},$$
$$\beta = \arcsin\sqrt{\frac{L^2+a^2}{L^2+a^2+b^2}},$$
$$\gamma = \mathrm{arctg}\,\frac{\sqrt{a^2+b^2}}{L}, \qquad (7)$$

where $L = 35$ mm is the distance from the cell with colloidal quantum dots to the screen, and a and b are diffraction maxima coordinates in the x–y plane. Experimentally obtained values correspond to the values calculated with the help of the system of Eqs. (6). Diffraction patterns are demonstrated in the Fig. 4. The absence of diffraction maxima in the case when structure factor is equal to zero confirm that the induced dynamic two-dimensional photonic crystal is the hexagonal one. We marked by the stars diffraction maxima, which were not revealed in the experiment, but were evaluated from the system of Eqs. (6).

Diffraction pattern is symmetric in the screen plane with respect to rotation angle equal to 120°. Diffraction maxima for all the three transmitted laser beams coincide, but for the different values of indexes $(m_1; m_2)$. The most significant contribution to the single maximum belongs to the beam with the smallest value of a sum of corresponding indexes the value $(|m_1| + |m_2|)$.

4. Conclusion. We described for the first to our knowledge time a technique for two-dimensional tunable dynamic photonic crystals formation with different lattice symmetry by means of three interacting noncoplanar laser beams in colloidal solution of resonantly excited excitons in CdSe/ZnS quantum dots. With this technique we have made micro-periodic dynamic semiconductor structure with strong nonlinear changing of refraction and absorption and proved the photonic crystal formation by observed self-diffracted laser beams.

We performed theoretical calculation of the interference intensity field in the quantum dots solution and as an example demonstrated the formation of a photonic crystal with hexagonal lattice structure.

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