Analysis of exciton luminescence in GaN meso-cavity.

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We have considered the interaction of an exciton mode with photonic modes in a GaN structure several μ m in size. A technique for obtaining spectra from such structures has been demonstrated. The cavity shape and dimensions most optimal for efficient light/matter interaction have been selected. The theoretical spectrum for the selected cavity has been obtained and analyzed

Keywords: gallium nitride, exciton, meso-cavity, spectrum.

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Investigations in the field of the light/matter interaction are at present of a great fundamental and applied importance in physics. Among the elements in which efficient light/matter interaction can take place there are optical micro-cavities. Dimensions of such structures are of about the emission wavelength in the material. Incorporation of the emitter into the micro-cavity can give rise to varioustype effects connected with the light/matter interaction. An increase in the density of photonic states in the microcavity in the vicinity of the emitter location with respect to that in vacuum causes an increase in the rate of spontaneous emission. The increase or decrease in the cavity spontaneous emission rate is referred to as the Parcell effect [1]. In 1992, Weisbuch et al. observed an effect of strong coupling between quantum dot excitons and photonic eigenmodes of a planar micro-cavity [2]. Vast investigations performed in this field during last three decades have formed a new solid state physics paradigm, namely, quantum electrodynamics of crystals [3-5]. In micro-cavities, electromagnetic fields are localized in a volume comparable with the light wavelength. Strong coupling in the microcavity occurs between a quantum dot exciton and only one cavity mode [2]; in more complex structures (e.g., in coupled cavities) [6], interaction between a few excitons and cavity modes is possible. The Rabi splitting will be smaller than the distance between the cavity modes. To provide these conditions, it is necessary to fabricate smallsize structures (of about several μm). However, fabrication of structures of such dimensions is at present a complicated technological task for semiconductors of many types [7], for instance, for wide-bandgap semiconductors GaN possessing such useful properties as bright UV radiation, high strength of the exciton oscillator, chemical and thermal stability. At the same time, cavities several wavelengths in size in which the exciton mode interacts with a few optical modes of the system at once (meso-cavities) remain poorly studied.

The earliest studies of meso-cavities [8–10] demonstrated a number of new effects: nonlinear behavior of the system mode population; a specific nonmonotonic character of bistable behavior of the polariton population depending on pumping; the possibility of occurring of strong and weak coupling between the exciton mode and photonic eigenmodes of the system despite the high density of the cavity modes.

The Hamiltonian for a system describing the interaction between the exciton mode with energy $\hbar \omega_k$ and several photonic modes with energies $\hbar \omega_k$ will be expressed as follows [11]:

$$\hat{H} = \hbar \omega_x \hat{x}^+ \hat{x} + \sum_k \hbar \omega_k \hat{a}_k^+ \hat{a}_k + \sum_k \hbar g_k \left(\hat{a}_k^+ \hat{x} + \hat{a}_k \hat{x}^+ \right),$$
(1)

where \hat{x}^+ , $\hat{a}_k^+(\hat{x}, \hat{a}_k)$ are the exciton and photon creation (annihilation) operators, respectively. For these operators, the following commutation relations are valid: $[\hat{a}_k, \hat{a}_k^+] = 1$, $[\hat{x}, \hat{x}^+] = 1$, $\hbar g_k$ is the interaction energy of the exciton mode and *k*-th photonic mode. In the presence of dissipation, the system will be modified so as to describe the system via the Liouvillian for the density matrix containing terms accounting for the dissipation. Hence, the overall system state is describable by the density matrix $\hat{\rho}$, while its evolution is defined as follows: $\partial_t \hat{\rho} = \hat{\mathcal{L}} \hat{\rho}$, where $\hat{\mathcal{L}}$ is the Liouvillian with Lindblad terms describing the dissipation:

$$\begin{aligned} \hat{\mathscr{L}}\hat{\rho} &= \frac{\imath}{\hbar} [\hat{\rho}, \hat{H}] + \frac{\gamma_x}{2} \left(2\hat{x}\hat{\rho}\hat{x}^+ - \hat{x}^+\hat{x}\hat{\rho} - \hat{\rho}\hat{x}^+\hat{x} \right) \\ &+ \sum_k \frac{\gamma_k}{2} \left(2\hat{a}_k\hat{\rho}\hat{a}_k^+ - \hat{a}_k^+\hat{a}_k\hat{\rho} - \hat{\rho}\hat{a}_k^+\hat{a}_k \right), \end{aligned}$$
(2)

where γ_x , γ_k are the dissipation coefficients for the exciton and photonic mode, respectively.

To find the average number of particles in the exciton and photonic modes, as well as in their mixed states, it is necessary to solve a set of differential equations that can be derived from relation $\partial_t \langle \hat{O} \rangle = \text{Tr}(\hat{O}\partial_t \hat{\rho}) = \text{Tr}(\hat{O}\hat{\mathscr{L}}\hat{\rho})$, where \hat{O} is an arbitrary operator. Then, the set of equations

L, μm $\hbar\omega$, eV Q 1.5 3.5099 11501 2.0 3.4969 1791 2.5 3.4833 50068 3.0 3.4727 $1.13 \cdot 10^{6}$ 3.5 3.4519 $4.5 \cdot 10^{5}$

Energy $\hbar\omega$ and Q factor Q of the high-Q mode versus length L of the hexagonal cavity side

describing the temporal dynamics of the mode populations gets the following form:

$$\begin{cases} \partial_t n_{xx} = \sum_k i g_k (n_{kx} - n_{xk}) - \gamma_x n_{xx}, \\ \partial_t n_{ij} = i (\omega_i - \omega_j) n_{ij} - i g_j n_{ix} + i g_i n_{xj} - \frac{1}{2} (\gamma_i + \gamma_j) n_{ij}, \\ \partial_t n_{xi} = i (\omega_x - \omega_i) n_{xi} + \sum_k i g_k n_{ki} - i g_i n_{xx} - \frac{1}{2} (\gamma_x + \gamma_i) n_{xi}, \end{cases} \end{cases}$$

$$(3)$$

where $n_{xx} = \langle \hat{x}^+ \hat{x} \rangle$ is the average number of excitons, $n_{xi} = \langle \hat{x}^+ \hat{a}_i \rangle = n_{ix}^*$ and $n_{ij} = \langle \hat{a}_i^+ \hat{a}_j \rangle$ $(i \neq j)$ are the respective values in mixed states. The number of photons in the *i*-th mode is defined as $n_{ii} = \langle \hat{a}_i^+ \hat{a}_i \rangle$.

To calculate the meso-cavity emission spectra, the formalism presented in [12] was used. After standardization, the system emission spectrum (probability density of the fact that the photon emitted by the system has frequency ω) looks like:

$$S(\omega) = \frac{\sum_{i} s_{i}(\omega)}{\sum_{i} \int_{0}^{\infty} n_{ii} dt}.$$
(4)

The value of function $s_i(\omega)$ may be obtained using the firstorder correlation function. The procedure for calculating correlation functions for the $s_i(\omega)$ function is given in details in paper [12]. Thus, using the above described technique, it is possible to obtain emission spectra for systems where interaction between one exciton mode and several photonic modes takes place.

The majority of studies devoted to growing planar structures from GaN showed that the grown cavities have hexagonal shape due to specific features of the crystal lattice [13,14]. Using the technique of selective gas-phase epitaxy from metal-organic compounds, one can succeed in experimentally obtaining hexagonal strips of different lengths. However, the loss of the cavity shape symmetry results in a sharp decrease in the eigenmode Q factors. In this work, we have studied hexagonal symmetric structures obtained from GaN, which are, as shown experimentally, much easier to be grown than structures of other geometries. In addition, formation of a single high-Q mode able to efficiently interact with an exciton is much more probable in hexagonal structures [8].

Let us first determine the size of the hexagonal structure in which such a mode structure occurs where the high-Q mode energy is close to the exciton energy. The



Figure 1. Distribution of the electromagnetic field intensity within the ideal GaN hexagonal cavity with the side of $3 \mu m$ and refraction index n = 2.6267 calculated by using the COMSOL code. Energy is $3.4727 \text{ eV}, Q = 1.13 \cdot 10^6$.

eigenmodes and their Q factors were calculated for a twodimensional hexagonal meso-cavity by using the COMSOL Multiphysics code. The meso-cavity refraction index was $n_{cavity} = 2.6267$, absorption was absent. Refraction index of the environment was $n_{media} = 1$. The table lists the calculations of energy $\hbar \omega$ and Q-factor Q of the high-Q cavity mode for several lengths of the hexagonal mesocavity side L. As the table shows, the high-Q mode energy $\hbar \omega = 3.4727 \text{ eV}$ for side length $L = 3 \,\mu\text{m}$ is closest to the GaN exciton mode energy $\hbar \omega_x = 3.47 \,\text{eV}$ [13]. Fig. 1 presents the distribution of the electromagnetic field intensity within the cavity for the specified high-Q mode with $Q = 1.13 \cdot 10^6$. The table also shows that the mode Q factor essentially increases with the cavity size. However, in this case the energy interval between the modes (including high-Q ones) will decrease. Thus the distributions of optical eigenmodes for the most optimal meso-cavity length $L = 3 \,\mu m$ were calculated over energies and Q factors (see Fig. 2). As Fig. 2 shows, only one high-Q mode exists in the exciton mode energy range. Nevertheless, there are a few eigenmodes with Q factors only twice lower than Q factor of the main mode.

To calculate the selected meso-cavity spectrum, the following parameter values were taken: exciton mode energy $\hbar \omega_x = 3.47 \text{ eV}$, exciton dissipation coefficient $\gamma_x = 0.075 \text{ meV}$ [11], coupling constant $\hbar g_k \approx 50 \text{ meV}$ [15]. The photonic mode dissipation coefficients were $\gamma_k = \frac{\omega_k}{Q_k}$ (at the zero temperature). Initial number of excitons in the system was $n_{xx}^0 = 10$, that of photons was $n_{ii}^0 = 0$. Thus, we consider a system into which a certain number of excitons were initially injected. Since we consider a linear model, the number of initial excitons does not affect the dynamics of the photonic and exciton mode populations. Fig. 3 presents the calculated emission spectrum of the selected meso-cavity. To find out the way by which the energy was split between the high-Q photonic mode and



Figure 2. Energy and Q-factor distributions of the hexagonal meso-cavity eigenmodes. The dashed line represents the exciton mode energy $\hbar\omega_x = 3.47$ eV.



Figure 3. Theoretically calculated emission spectrum of the selected hexagonal meso-cavity.

exciton mode, manipulations with the coupling constant value were carried out. Finally, one can see that energies $\hbar\omega_{p1} = 3.49 \text{ eV}$ and $\hbar\omega_{p2} = 3.53 \text{ eV}$ relate to the polariton modes. The increase in the emission intensity of the $\hbar\omega_{p1}$ mode relative to emission intensity of mode $\hbar\omega_{p2}$, as well as energy shifts of the modes, is connected with the presence in the structure of other modes with lower Q factors. The results of the two-dimensional simulation are applicable to planar cavities in which the configuration of system modes is affected mainly by only the 2D shape of the cavity [16].

Hence, in this study the optimal GaN meso-cavity shape was selected taking into account specific features of procedures for obtaining these structures in experiments. The size of a GaN meso-cavity with the most suitable structure of the system eigenmodes was determined. The calculated structure of the photonic eigenmodes exhibited the existence in the exciton mode energy range of a single high-Q optical mode able to efficiently interact with the exciton. In the obtained spectrum, an enhancement of the emission intensity was observed for the polariton mode with energy $\hbar\omega_{p1} = 3.49$ eV.

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Conflict of interests

The authors declare that they have no conflict of interests.

References

- [1] E.M. Purcell, Phys. Rev., 69, 681 (1946).
- [2] C. Weisbuch, M. Nishioka, A. Ishikava, Y. Arakawa, Phys. Rev. Lett., 69 (23), 3314 (1992).
 DOI: 10.1103/PhysRevLett.69.3314
- [3] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J.M.J. Keeling, F.M. Marchetti, M.H. Szymańska, R. André, J.L. Staehli, V. Savona, P.B. Littlewood, B. Deveaud, L.S. Dang, Nature, 443 (7110), 409 (2006). DOI: 10.1038/nature05131
- [4] A. Amo, J. Lefrére, S. Pigeon, C. Adrados, C. Ciuti, I. Carusotto, R. Houdré, E. Giaobino, A. Bramati, Nature Phys., 5 (11), 805 (2009). DOI: 10.1038/nphys1364
- [5] M. Sich, D.N. Krizhanovskii, M.S. Skolnick, A.V. Gorbach, R. Hartley, D.V. Skryabin, E.A. Cerda-Méndez, K. Biermann, R. Hey, P.V. Santos, Nature Photon., 6 (1), 50 (2011). DOI: 10.1038/nphoton.2011.267
- [6] A. Armitage, M.S. Skolnick, V.N. Astratov, D.M. Whittaker, G. Panzarini, L.C. Andreani, T.A. Fisher, J.S. Roberts, A.V. Kavokin, M.A. Kaliteevski, M.R. Vladimirova, Phys. Rev. B, 57 (23), 14877 (1998). DOI: 10.1103/PhysRevB.57.14877
- [7] T. Takeuchi, S. Kamiyama, M. Iwaya, I. Akasaki, Rep. Prog. Phys., 82 (1), 012502 (2019).
 DOI: 10.1088/1361-6633/aad3e9
- [8] A.V. Belonovski, I.V. Levitskii, K.M. Morozov, G. Pozina, M.A. Kaliteevski, Opt. Express, 28 (9), 12688 (2020). DOI: 10.1364/OE.388899
- [9] G. Pozina, C. Hemmingsson, A.V. Belonovskii, I.V. Levitskii, M.I. Mitrofanov, E.I. Girshova, K.A. Ivanov, S.N. Rodin, K.M. Morozov, V.P. Evtikhiev, M.A. Kaliteevski, Phys. Status Solidi A, **217** (14), 1900894 (2019). DOI: 10.1002/pssa.201900894
- [10] A.V. Belonovski, K.M. Morozov, E.I. Girshova, G. Pozina, M.A. Kaliteevski, Opt. Express, 29 (13), 20724 (2021). DOI: 10.1364/OE.420277
- [11] A. Baas, J.P. Karr, H. Eleuch, E. Giacobino, Phys. Rev. A, 69 (2), 023809 (2004). DOI: 10.1103/PhysRevA.69.023809
- [12] F.P. Laussy, E. del Valle, C. Tejedor, Phys. Rev. B, **79** (23), 235325 (2009). DOI: 10.1103/PhysRevB.79.235325
- [13] G. Pozina, K.A. Ivanov, M.I. Mitrofanov, M.A. Kaliteevski, K.M. Morozov, I.V. Levitskii, G.V. Voznyuk, V.P. Evtikhiev, S.N. Rodin, Phys. Status Solidi B, **256** (6), 1800631 (2019). DOI: 10.1002/pssb.201800631

- [14] W.V. Lundin, A.F. Tsatsulnikov, S.N. Rodin, A.V. Sakharov, S.O. Usov, M.I. Mitrofanov, I.V. Levitskii, V.P. Evtikhiev, Semiconductors, 52 (10), 1357 (2018). DOI: 10.1134/S106378261810007X.
- [15] M.A. Kaliteevski, K.A. Ivanov, G. Pozina, A.J. Gallant, Sci Rep., 4, 5444 (2014). DOI: 10.1038/srep05444
- [16] A.V. Belonovskii, G. Pozina, I.V. Levitskii, K.M. Morozov, M.I. Mitrofanov, E.I. Girshova, K.A. Ivanov, S.N. Rodin, V.P. Evtikhiev, M.A. Kaliteevski, Semiconductors, 54 (1), 127 (2020). DOI: 10.1134/S1063782620010042.