Exciton-like electromagnetic excitations in non-ideal microcavity supercrystals

Vladimir Rumyantsev, Stanislav Fedorov, Kostyantyn Gumennyk, Marina Sychanova & Alexey Kavokin

1Galkin Institute for Physics & Engineering, Donetsk 83114, Ukraine, 2Mediterranean Institute of Fundamental Physics, 00047 Marino, Rome, Italy, 3Physics and Astronomy School, University of Southampton, Highfield, Southampton, SO17 1BJ, United Kingdom.

We study localized photonic excitations in a quasi-two-dimensional non-ideal binary microcavity lattice with use of the virtual crystal approximation. The effect of point defects (vacancies) on the excitation spectrum is investigated by numerical modelling. We obtain the dispersion and the energy gap of the electromagnetic excitations which may be considered as Frenkel exciton-like quasiparticles and analyze the dependence of their density of states on the defect concentrations in a microcavity supercrystal.

Photonic structures and metamaterials are presently in the focus of theoretical and experimental interdisciplinary studies, which span laser physics, condensed matter physics, nanotechnology, chemistry and information science. Many papers have been devoted to realization of light-emitting devices based on polaritonic crystals. In this context, semiconductor microcavities represent quantum confined optical systems featured by strong coupling between elementary crystal excitations (excitons) and the optical field. Photonic supercrystals can be built from spatially-periodic systems of coupled microcavities.

The physics of photonic supercrystals is in many ways similar to the physics of crystalline solids. Due to imperfections of the supercrystal lattice photonic gaps may contain impurity states, which are of crucial importance in realistic photonic structures. While the theory of impurity bands and excitons in semiconductor crystals has been developed in 1970–1980s, a similar theory for photonic crystals is yet to be constructed. In this work we carry out a theoretical study of exciton-like electromagnetic excitations in disordered photonic supercrystals composed by coupled microcavities.

Semiconductor microcavities are widely used in optoelectronic devices nowadays. Nanocavities in photonic crystals represent a particular case of microcavities characterized by a discrete photonic spectrum. Nanocavities with embedded quantum dots have been used to demonstrate the strong light-matter coupling regime in Ref. 11 and proposed for realization of quantum solitons coupled to lower-branch polaritons (LBPs). Refs. 3, 4 indicate also that chains of microcavities may be used for practical realization of quantum-information processing.

Recent progress in fabrication of reliable semiconductor microcavities with Bragg mirrors and embedded quantum wells led to demonstration of a Bose-Einstein condensation of exciton-polaritons and finding features of their superfluidity. In those specified systems polaritons can be treated as a quasi-equilibrium two-dimensional gas of interacting bosonic quasiparticles.

Basing upon the previously developed concept of photonic structures, Ref. 15 studies a non-ideal polariton supercrystal realized in a system of coupled microcavities, whose atomic subsystem contains impurity clusters. It is important to know the dispersion of electromagnetic eigenmodes in such non-ideal microcavity supercrystals in order to develop opto-electronic and quantum computation devices based on such structures. Here we study dispersions of localized electromagnetic excitations in an array of coupled microcavities, which form a non-ideal supercrystal containing numerous point-like defects.

Theoretical background

One of the methods of fabrication of polaritonic crystals is the trapping of two-level atoms in an ideal coupled resonator optical waveguide (CROW) or in a non-ideal photonic structure. Refs. 3, 8, 9, 15 study coupled cavities with dopant atoms. In the present work, we do not consider photon mode coupling with dopant atoms. Instead we concentrate on exciton-like electromagnetic excitations of the disordered multicavity structure. We consider a 2D lattice of microcavities, each characterized by a single confined optical mode. An overlap of optical
fields of the eigenmodes of neighboring microcavities is taken into account, so that photons are allowed to move along the surface of the microcavity array. For the sake of generality, we assume that each cell of the photonic supercrystal lattice may contain an arbitrary number of elements.

Hamiltonian $H$ of the model system we consider (for more details see also Ref. 3) writes:

$$H_{ph} = \sum_{n,x} E_{nx} \Psi_{nx}^+ \Psi_{nx} - \sum_{n,z,m} A_{nxm\beta} \Psi_{nx}^+ \Psi_{m\beta}.$$  \hspace{1cm} (1)

Subscripts $n$ and $m$ are two-dimensional integer lattice vectors, $x$ and $\beta$ numerate sublattices, whose total number is $\sigma$, $E_{nx} = \omega_{nx}$, where $\omega_{nx}$ is the frequency of photonic mode localized in the $n$-th site (cavity). Quantity $A_{nxm\beta}$ defines the overlap of optical fields of the $n$-th and $m\beta$-th cavities and the transfer of the corresponding excitation, $\Psi_{nx}^+$, $\Psi_{nx}$ are bosonic creation and annihilation operators describing the photonic mode. Hamiltonian (1) is formally identical to the tight-binding excitonic Hamiltonian in a semiconductor crystal\textsuperscript{10,17}, for which reason the studied electromagnetic excitations can naturally be referred to as exciton-like. It is worth stressing that we discuss photonic supercrystal excitations and no electronic transitions are involved. Nevertheless, it will be seen below that the dispersion relations of purely electromagnetic crystal excitations in the studied system are quite similar to the Frenkel exciton bands in molecular crystals\textsuperscript{16,20}.

Let us consider a topologically ordered non-ideal lattice of microcavities with point-like defects, namely vacancies and non-typical cavities. The considered photonic supercrystal lattice may contain an arbitrary number of cavities. The left-hand side of Eq. (7) is then a second-order determinant, which when equated to zero gives the solvability condition of the system (5)

$$\left| \langle E_{nx} \rangle \delta_{x\beta} - h_{nx} \langle A_{nxm\beta} \rangle \exp \left[ik (r_{nx} - r_{m\beta}) \right] \right| = 0.$$  \hspace{1cm} (6)

Here $r_{nx}$ is the radius-vector of a resonator belonging to the $n$-th elementary cell. The solvability condition of the system (5) yields the dispersion law $\omega_{1,2}(k)$ of electromagnetic excitations in the considered photonic supercrystal.

**Results and Discussion**

Consider localized electromagnetic excitations in a two-sublattice ($r=2$) system of cavities. The left-hand side of Eq. (7) then is a second-order determinant, which when equated to zero gives the following dispersion of photonic excitations:

$$\omega_{1,2}(k) = \frac{1}{2h} \left\{ L_{11}(k) + L_{22}(k) \pm \sqrt{L_{11}(k) - L_{22}(k)}^2 + 4|L_{12}(k)L_{21}(k)|^2 \right\}.$$  \hspace{1cm} (7)

Here $L_{11}(k) = E_1 - A_{11}(k)$, $L_{12}(k) = E_1 - A_{22}(k)$, $L_{21}(k) = -A_{12}(k)$ and $L_{22}(k) = -A_{21}(k)$ are the matrix elements of operator $L$.

To be more specific, let us consider a spectrum of electromagnetic excitations in a binary system where each sublattice contains only two types of cavities. In such a case, the quantities $\langle E_{nx} \rangle$ and $\langle A_{nxm\beta} \rangle$ are given by

$$\langle E_{nx} \rangle = \sum_{v(x)} E_v^x \frac{f_v^x}{n_x}, \langle A_{nxm\beta} \rangle = \sum_{v(x),\mu(\beta)} A_v^x \langle \mu(\beta) |(n - m) \rangle f_v^x \frac{f_\mu^\beta}{n_{\beta}}.$$  \hspace{1cm} (3)

where $n_x$, $m$, $\beta$ are occupation numbers of sites of a lattice. Using the VCA (similarly to the variational approach\textsuperscript{12,13}) yields

$$\langle E_{nx} \rangle = \sum_{v(x)} E_v^x C_v^x; \langle A_{nxm\beta} \rangle = \sum_{v(x),\mu(\beta)} A_v^x \langle \mu(\beta) |(n - m) \rangle f_v^x \frac{f_\mu^\beta}{n_{\beta}},$$  \hspace{1cm} (4)

where $C_v^x$ and $C_\mu^\beta$ are concentrations of the $v$-th and $\mu$-th types of cavities. The left-hand side of Eq. (7) is then a second-order determinant, which when equated to zero gives the following dispersion of photonic excitations:

$$\omega_{1,2}(k) = \frac{1}{2h} \left\{ L_{11}(k) + L_{22}(k) \pm \sqrt{L_{11}(k) - L_{22}(k)}^2 + 4|L_{12}(k)L_{21}(k)|^2 \right\}.$$  \hspace{1cm} (8)

$$\frac{1}{2h} \left\{ L_{11}(k) + L_{22}(k) \pm \sqrt{L_{11}(k) - L_{22}(k)}^2 + 4|L_{12}(k)L_{21}(k)|^2 \right\} = 0.$$  \hspace{1cm} (9)

where $C_1^1 \equiv C_1$ is the cavity concentration in the first sublattice, $C_2^1 \equiv C_2$ is the cavity concentration in the second sublattice,
defined by the type of the considered sublattices and the quantities of the same sublattice but different cells.

Figure 2 | Dispersion $\omega \pm (k, C_1^V, C_2^V)$ of electromagnetic excitations in the non-ideal two-dimensional two-sublattice system of microcavities for a) $C_1^V = 0.55, C_2^V = 0.1$; b) $C_1^V = 0.84, C_2^V = 0.2$, c) $C_1^V = 0.9468, C_2^V = 0.7$.

and thus the corresponding matrix elements of operator $\hat{L}$ take the form

$$L_{11}(k) \pm L_{22}(k) =$$
$$= \omega_1 (1 - C_1^V) = \omega_2 (1 - C_2^V) -$$
$$- 4 \left[ A_{11}(d) (1 - C_1^V)^2 \pm A_{22}(d) (1 - C_2^V)^2 \right]$$
$$\cos \frac{d(k_x + k_y)}{2} \cos \frac{d(k_y - k_x)}{2},$$

In (10) the overlap characteristic of optical fields $A_{11(22)}(d)$ defines the transfer probability of electromagnetic excitation between the nearest neighbors in the first (second) sublattice, and $A_{12(21)}(0)$ is the excitation transfer probability between cavities in the first (second) and second (first) sublattices in an arbitrary cell. Substitution of expressions (10) for $L_{12}(k)$ into Eq. (8) gives the dispersion law $\omega_+(k)$ for electromagnetic excitations (Fig. 2a,b,c). We performed calculation for modeling frequencies of resonance photonic modes in the cavities of the first and second sublattices $\omega_+ = \langle E_{bl} \rangle / \hbar = 6 \times 10^{15} \text{Hz}$ and $\omega_2 = \langle E_{bl} \rangle / \hbar = 8 \times 10^{14} \text{Hz}$ respectively and for the overlap parameters of resonator optical fields $A_{11(22)} / 2\hbar = 3 \times 10^{14} \text{Hz}$, $A_{22(11)} / 2\hbar = 5 \times 10^{13} \text{Hz}$ and $A_{12(21)} = A_{21(12)} / 2\hbar = 5 \times 10^{13} \text{Hz}$. The lattice period was set equal to $d = 3 \times 10^{-7} \text{m}$.

Figure 1 | Schematic of a non-ideal two-dimensional two-sublattice system of microcavities, $e_1$ and $e_2$ are the basis vectors of the square Bravais lattice. “V” denotes vacancies.
[The text from the image is not fully legible, but it appears to be discussing the dispersion of collective excitations in a microcavity lattice, focusing on the energy gap width and concentration dependence. The text includes mathematical expressions and references to figures showing isofrequency lines and cavity concentration dependence graphs.]

Figure 3 | Cavity concentration dependence of the photonic gap width $\Delta \omega (C_1^V, C_2^V)$ in the studied microcavity supersystem.

Figure 4 | Isofrequency lines for a), d) upper and lower surfaces in Fig. 2a; b), e) upper and lower surfaces in Fig. 2b; c), f) upper and lower surfaces in Fig. 2c. The frequency is measured in the units of $10^{15}$ Hz. Black diamonds indicate saddle points, which yield singularities in the corresponding densities of states (see Fig. 5).
solid curves show the densities of states \( g_\pm(\omega, C_1^V, C_2^V) \) for the surfaces \( \omega_+ \) and \( \omega_- \) in Fig. 2a. Dashed lines show the transformation of functions \( g_+ \) and \( g_- \) under varying \( C_1^V \) and \( C_2^V \). It turns out that in the region \( \Delta \omega(C_1^V, C_2^V) \neq 0 \) the density of states \( g_- \) is all but independent of \( C_1^V \), while \( g_+ \) is almost unaltered by variations in \( C_2^V \). This is explained by the smallness of the term \( L_{12}(k)L_{21}(k) \) as compared to \( |L_{11}(k) - L_{22}(k)|^2 \) in Eq. (8). Figs. 5bc and 5ef give examples of the typical \( g_+ \) and \( g_- \) curves for concentration values corresponding to \( \Delta \omega = 0 \) (here we took \( C_1^V = 0.84, C_2^V = 0.2 \) and \( C_1^V = 0.9468, C_2^V = 0.7 \)). Their evident non-monotonic and discontinuous character is similar to the analogous dependence \( g(\omega) \) obtained in Ref. 21 for phonon excitations.

**Conclusion**

A number of recent experimental works indicate that microcavity supercrystals may have interesting applications, in particular for creating the optical clockworks of unprecedented accuracy\(^{22-24}\). We have used the virtual crystal approximation to model the effect of lattice point defects (vacancies) on the spectrum of exciton-like electromagnetic excitations in a quasi-2D binary microcavity lattice. The energy spectrum of electromagnetic excitations affects the density of states of electromagnetic excitations and alters propagation of normal electromagnetic waves. The obtained dispersions of electromagnetic excitations are noticeably more complex than those of primitive lattices. This complexity is due to the non-ideality of the structure and to the presence of two sublattices. The latter entails multiple manifestations in experimentally observable integral characteristics of optical processes. Evaluation of excitation spectra in more complex photonic systems requires the use of more sophisticated computational methods. Depending on particular cases such can be the one- or multiple-node coherent potential method\(^{25}\) and the averaged \( T \)-matrix method\(^{26}\) along with their various modifications. Our study contributes to the modeling of novel functional materials with controllable propagation of electromagnetic excitations.

1. Cai, W. & Shalaev, V. Optical Metamaterials: Fundamentals and Applications (Springer, New York, 2010).
2. Razeghi, M. Technology of Quantum Devices (Springer, New York, 2010).
3. Alodjants, A. P., Barinov, I. O. & Arakelian, S. M. Strongly localized polaritons in an array of trapped two-level atoms interacting with a light field. J. Phys. B: At. Mol. Opt. Phys. 43, 095502 (2010).
4. Sedov, E. S., Alodjants, A. P., Arakelian, S. M., Lin, Y. Y. & Lee, R.-K. Nonlinear properties and stabilities of polaritonic crystals beyond the low-excitation-density limit. Phys. Rev. A 84, 013813 (2011).
5. Joannopoulos, J. D., Johnson, S. G., Winn, J. N. & Meade, R. D. Photonic Crystals: Molding the Flow of Light (Princeton University Press, Princeton, 2008).
6. Vahala, K. I. Optical microcavities. Nature 424, 839 (2003).
7. Kaliteevski, M. A. Coupled vertical microcavities. Tech. Phys. Lett. 23, 120–121 (1997).
8. Golubev, V. G. et al. Splitting of resonant optical modes in Fabry-Perot microcavities. Semiconductors 37, 832–837 (2003).
9. Lee, R. K., Painter, O., Kitaev, A., Scherer, A. & Yariv, A. Emission properties of a defect cavity in a two-dimensional photonic bandgap crystal slab. J. Opt. Soc. Am. B 17, 629–633 (2000).
10. Vučković, J., Loncar, M., Mabuchi, H. & Scherer, A. Design of photonic crystal microcavities for cavity QED. Phys. Rev. B 65, 016608 (2001).
11. Englund, D. et al. Resonant Excitation of a Quantum Dot Strongly Coupled to a Photonic Crystal Nanocavity. Phys. Rev. Lett. 104, 073904 (2010).
12. Kasprzak, J. et al. Bose-Einstein condensation of exciton polaritons. Nature 443, 409 (2006).
13. Balli, R., Hartwell, V., Snoke, D., Pfeiffer, L. & West, K. Bose-Einstein Condensation of Microcavity Polaritons in a Trap. Science 316, 1007–1010 (2007).
14. Amo, A. et al. Superfluidity of polaritons in semiconductor microcavities. Nature Phys. 5, 805–810 (2009).
15. Alodjants, A. P., Rumyantsev, V. V., Fedorov, S. A. & Proskurenko, M. V. Polariton dispersion dependence on concentration of admixture in imperfect superlattice of coupled microcavities. Int. Conf. Functional Materials. 195 (Ukraine, Crimea, Yalta, Haspra, 2013).
16. Agranovich, V. M. Theory of Excitons (Nauka Publishers, Moscow, 1968).
17. Kittel, C. Quantum theory of solids (Wiley, New York, 1987).
Acknowledgments

This work was supported by the European contract FP7-PEOPLE-2013-IRSES (Grant # 612660 "LIMACONA"). A.K. acknowledges support from the EPSRC in the framework of the Advanced Career Fellowship on Polaritonics.

Author contributions

V.R. proposed the idea and has written the bulk of the paper. V.R., S.F. performed analytical calculations. K.G. and M.S. performed the numerical simulations. V.R., S.F. and A.K. contributed to the discussion and paper writing.

Additional information

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Rumyantsev, V., Fedorov, S., Gumennyk, K., Sychanova, M. & Kavokin, A. Exciton-like electromagnetic excitations in non-ideal microcavity supercrystals. Sci. Rep. 4, 6945; DOI:10.1038/srep06945 (2014).