Composite Lattice Phases of Light and Low Symmetry Molecules

A B Kuklov[1] and V Fleurov[2]

[1] *Physics and Astronomy, CSI and the Graduate Center, CUNY, 10314, New York NY, USA*
[2] *School of Physics and Astronomy, Tel Aviv University, Tel-Aviv, Israel*
Contact Email: akuklov2@gmail.com

Some dye molecules used in lasers are characterized by a strong dipole active transition 1S-2S. In order to describe a system of such low symmetry molecules interacting with light, it is important to introduce the field of orientations of $\vec{n}(\vec{x})$ of the dipolar matrix element d of the optical transition. Local orientation of the molecular symmetry axis determines $\vec{n}(\vec{x})$. Inspired by the realization of the Bose-Einstein condensate of light in the system of the dye molecules [1,2], the analysis [3,4] of the role of the molecular orientations has discovered various novel phases and phase transitions in the limit of the macroscopic population of photons.

Here we argue that very interesting options for exotic phases can exist in the lattice of microcavities, each containing one or few such excited molecules in the limit when there is no macroscopic population of free photons. In this limit, virtual photons support the transport of the excitons between lattice sites. The resulting excitonic phases strongly depend on the geometry of the lattice and the resonators. In general, the operator of the amplitude of the exciton tunneling from a site i to a site j in the lowest order of the fine-structure constant has a form

$$t_{ij}c_j^\dagger c_i = \sum_{\alpha,\beta} V_{\alpha\beta}(\vec{r}_{ij}) n_\alpha(\vec{r}_i) n_\beta(\vec{r}_j) c_j^\dagger c_i \tag{1}$$

where $c_i^\dagger$, c_i are bosonic operators of the excitons; $\vec{r}_{ij}$ stands for the vector of a distance between the sites; the Greek indices refer to the spatial coordinates; the tensor $V_{\alpha\beta}(\vec{r}_{ij}) \sim d^2$ accounts for the lattice geometry. In the limit of $|\vec{r}_{ij}|$ much less than the resonant wavelength λ, one finds $V_{\alpha\beta}(\vec{r}) = d^2 \frac{3\hat{r}_\alpha \hat{r}_\beta - \delta_{\alpha\beta}}{r^3}$, where the indices refer to the $\hat{x}, \hat{y}, \hat{z}$ directions. In the situation when the lattice is immersed in a 2D resonator (two parallel mirrors like in Ref. [1,2]) and in the limit, $|\vec{r}_{ij}| >> \lambda$, the angular part of the amplitude becomes $V_{\alpha\beta}(\vec{r}) \sim \delta_{\alpha\beta}$, with the indices referring to the directions along the mirrors and the spatial dependence controlled by the detuning from the resonance. If the sites i, j are connected by a 1D micro-waveguide supporting the resonance mode $\mathrm{TE}_{(1,0)}$, the angular part in Eq.(1) becomes $V_{\alpha\beta}(\vec{r}_{ij}) \sim d^2 e_{\alpha,ij} e_{\beta,ij}$, where $e_{\alpha,ij}$ is a unit vector determining the polarization of the mode of the waveguide (that is, $\vec{e}_{ij}\vec{r}_{ij} = 0$). In this case, the excitonic transport becomes highly anisotropic and strongly dependent on $\vec{n}$, which may be useful in photonic applications.

Since $\vec{n}(\vec{x}_i)$ is not, in general, ordered, a lattice system of such excitons *cannot* be treated within the standard polaritonic approach of a coherent mixing between excitons and light. One feature inherently present in all such lattice models, where the excitons and the field of orientations are two dynamical variables, is the Z_2 gauge symmetry. Indeed, the amplitude (1) demonstrates the local (gauge) symmetry $\vec{n}(\vec{r}_i) \to -\vec{n}(\vec{r}_i)$ together with $c_i \to -c_i$. This excludes any coherence in either subsystem—that is, an order can only exist in a gauge invariant combination of both fields such as,e.g., the product $\vec{p}_i = \vec{n}(\vec{r}_i)c_i$. [The coherence, then, occurs in the combined density matrix $\langle p^\dagger_{\alpha,i} p_{\beta,j} \rangle$]. There are also other—higher order combinations which may demonstrate an order, while $\vec{p}_i$ is disordered. For example, it can be the tensor $n_\alpha(\vec{r}_i) n_\beta(\vec{r}_i)$ (nematic order) or the pairing order parameter $c_i c_j$. There is also a possibility for insulating phases such as checkerboard and valence-bond solids. It is important to note that the gauge symmetry makes this type of system strongly interacting, which renders the mean-field analysis unreliable. Accordingly, numerical simulations must be performed in order to identify possible phases.

References

[1] J Klaers, F Vewinger and M Weitz, Nat. Phys. **6**, 512 (2010)

[2] J Klaers, J Schmitt, F Vewinger and M Weitz, Nature (London) **468**, 545 (2010)

[3] V Fleurov and A B Kuklov, New J. Phys. **21**, 083009 (2019)

[4] V Fleurov and A B Kuklov, Phys. Rev. A **101**, 043836 (2020)