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Cooperative phases and phase transitions of Bose condensed light in dye filled cavities

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Abstract

Recent realization of Bose–Einstein condensation of light in 2D provides a new platform for studying novel phases and phase transitions. The combination of low effective mass of the confined light and the presence of the dye molecules with randomly oriented directions of the dipolar transition engages a competition between disorder and the tendency to forming algebraic off-diagonal order. The phase diagram of possible phases is constructed at the mean field level. One of the phases is the condensate of photon pairs induced solely by the orientational disorder. Such a *geometrical* mechanism of pairing has no analogy in other systems.

1. Introduction

About 25 years ago a new revolution in physics took place—three groups [1–3] realized Bose–Einstein condensates (BECs) of ultracold atoms. This has initiated a search for new phases of matter culminating in demonstrating Bose—Mott insulator quantum phase transition [4]. Since then many more proposals of exotic novel strongly interacting phases of matter have been put forward and realized experimentally (see in [5]). Among the exciting possibilities is the bosonic fermionization [6] induced by artificial spin–orbit coupling [7]. Realization of interacting topological insulators with ultracold atoms also appears to be within the reach [8]. To a great extent these achievements have stimulated the development of strongly interacting photonics where the role of atoms is played by photons (see in [9]).

Creation of BEC of light [10–14], has opened up a new chapter in the search for strongly interacting phases of light. In these experiments the thermalization of light is achieved through absorption and reemission of photons by dye molecules which represent a thermal bath—thanks to their manifolds of rovibrational states exchanging energy with the solvent [15, 16]. The equilibration of light occurs on much faster scale than the photon escape from the cavity. Thus, for all practical purposes the emerging phase of light becomes a thermodynamical phase where photons can be characterized by finite chemical potential—due to a significant difference between temperature and photon energies.

The most recent exciting development in this field includes realization of the lattice consisting of micro photonic condensates with the Josephson interaction between them [17]. It clearly opens up a pathway toward realization of strongly interacting phases of light—by means of independent tuning of the Josephson tunneling amplitude and onsite interaction.

At this juncture it is important to mention that there are some similarities and significant differences between the dye molecules in the resonator [10, 11, 13, 14] and excitons in the condensed matter systems [18, 19] (see also in [20]). In the exciton-polaritonic materials the effective interaction between photons is induced through a direct exciton–exciton scattering—thanks to the linear and coherent coupling between exciton and photon branches of the spectrum. Accordingly, the Gross–Pitaevskii (GP) equation including two polarizations of polaritons is straightforward to derive [21]. In the systems [10, 11, 13, 14] the dye ensemble is disordered and, thus, it cannot be coherently coupled to photons. (The coherency and interaction between dye molecules via exchanging 2D-photons was discussed in [22].) An option to include photon–photon interaction has been

suggested in [12]—by taking into account the Kerr effect. Another option is the thermal lensing effect in the dye subsystem [10, 17].

The experimentally achieved condensation of light is characterized by a weak effective interaction between photons [10, 11, 13, 14]. However, even infinitesimally small interaction changes dramatically the properties of BEC (see in [23]). According to [10], the dimensionless interaction constant $\tilde{g} \sim 10^{-3}$ and typical photon numbers $N \sim 10^5$, with the condensate size $D \sim 10 - 20$ μm. This corresponds to the healing length $l_h \approx 1/\sqrt{\tilde{g} n_{\rm ph}} \approx 1 - 2$ μm, where $n_{\rm ph} \sim N/D^2$ is the 2D density of the photonic condensate. This estimate shows that, despite the smallness of the interaction, the healing length is much smaller than the condensate size and, thus, the GP equation is a relevant description of the system.

As will be discussed below, to achieve algebraic condensation of photons in 2D at finite temperature T it is important to have polarizational anisotropy of the photon–photon interaction. In this respect, the thermo-optical effect, which is insensitive to the polarization [10, 17], cannot provide such an anisotropy. Here we will address this aspect by including the anisotropic photon–photon interaction induced by the dye molecules each represented as a two level system (TLS) with randomly oriented vector $\vec{d}$ of the dipolar transition between the ground and excited states. (In this analysis the role of the dye sub ensemble in the thermalization of photons is set aside.) Furthermore, the limit of low density of the dye molecules is considered so that their interaction through near field photons, as discussed in [22], can be safely ignored. The analysis is conducted for uniform condensate in the thermodynamical limit.

The principal focus of the study is on the *orientational* disorder of the field $\vec{d}$ in the dye ensemble. We argue that it should result in a new phase of the photonic matter—the *geometrically* paired photonic superfluid (PSF). More specifically, such a pairing is induced by spatial randomness in $\vec{d}$ so that no algebraic order can be detected in the one-photon density matrix, while the two-photon density matrix (TPDM) demonstrates the algebraic off-diagonal long range order (ODLRO). This result was first introduced in [24]. Here we provide more details and suggest a possible pathway toward its realization. Another important and unique aspect of the system arises from the possibility of the TLS centers to change their orientations and positions due to the interaction with the PSF. This can lead to the phase separation effect, implying that the condensation can proceed simultaneously with the formation of non uniformity. This aspect of the system will be considered in the follow up publication.

Our paper is organized as follows. In section 2 the relevant variables are introduced. The Hamiltonian and the effective action are discussed in section 3. Symmetries of the phases are analyzed in section 4, with the geometrically paired PSF introduced in section 4.4. Finally, the overview and discussion is presented in section 5. Appendices A, B, C, respectively, provide details on the derivation of the GP equation, on a possibility of the realization of the paired PSF and on the nature of the phase transition from this phase to the ordinary PSF.

2. Order and disorder parameters

The setup [10] used to achieve thermalization and eventual condensation of light is explained in figure 1.

The order parameter of light is a complex vector $\vec{E} = \vec{E}(\vec{x}, z)$ representing the amplitude of electric field in the rotating wave approximation (RWA), with $\vec{x} = (x, y)$ and z being coordinates along and perpendicular to the XY plane of the resonator [10], respectively. The geometry of the experiment [10] selects a single mode $\phi = \phi(z)$ along the resonator Z-axis so that the field can be represented as $\vec{E} = \phi(z)\vec{\psi}(\vec{x})$, where the complex vector $\vec{\psi} = (\psi_x, \psi_y)$ accounts for the transverse order and its long wave fluctuations in the XY-plane of the resonator. (There is also the E_z component present insuring the divergenceless nature of the photonic field as $\nabla_z E_z + \vec{\nabla}\vec{\psi}\phi(z) = 0$.) We will be using the normalization in which $\vec{\psi}^\dagger\vec{\psi}$ is the operator of the 2D density of photons. Then, in the standard SI units[3]

$$\phi(z) = \sqrt{\frac{2\hbar\omega_0}{\varepsilon_0 L_z}}\,\sin(q_0 z), \tag{1}$$

where $q_0 = \omega_0/c$ is the wavevector of the standing wave, with c being speed of light and $L_z = 7\pi/q_0$ is the 'height' of the resonator [10] (see figure 1), and ε_0 denotes the background electric permittivity of the medium. The goal is to obtain the effective action for the amplitude $\vec{\psi}$ in the long wave limit by eliminating TLSs and integrating out the direction along the Z-axis.

The TLS molecules can be accounted for by a coarse grained field of their dipolar transitions $\vec{d} = \vec{d}(\vec{x}, z) = (d_x, d_y, d_z)$ responsible for absorbing and emitting photons. Such a variable emerges because the dye molecules are characterized by low symmetry so that the dipolar transition is non-degenerate, (see in [11, 25]) and, thus, is described by a specific (positive or negative) direction determined by the spatial orientation of the molecule.

[3] This normalization follows from equating the total energy of electromagnetic field in the condensate to $\hbar\omega_0 N$ (in the limit $N \gg 1$).

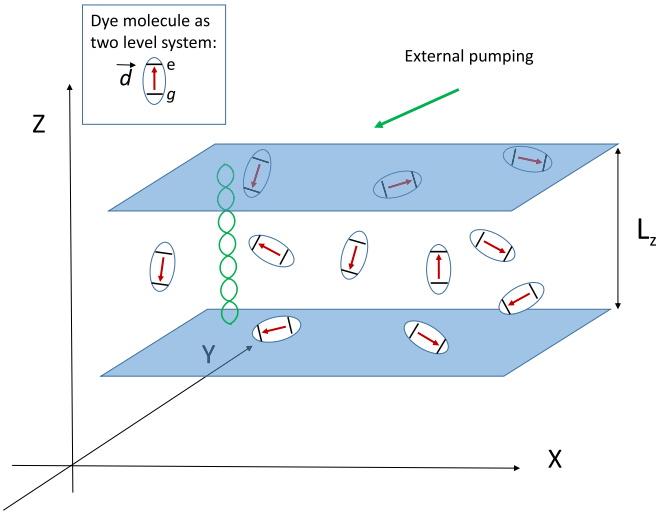

Figure 1. Sketch of the setup [10]. Two parallel planes depict cavity mirrors with the dye solution (randomly oriented ellipses with arrows) in between. (The mirrors in [10] have a curvature with the radius of a few meters, which creates a harmonic trap and ensures a true ODLRO of 2D light.) Externally pumped light forms standing wave along Z-direction (green chain of ellipses). Low energy perturbations of the photonic condensate are characterized by small momenta $\vec{k} = (k_x, k_y, 0)$ with the X, Y components only. Inset: dye molecule modeled as two level system characterized by the ground (g) and excited (e) states and the dipole moment of the transition $\vec{d}$.

In order to derive the effective low energy description for 2D vector field $\vec{\psi}(\vec{x})$ it is necessary to project the 3D field $\vec{d}$ onto the XY plane. Such a projection involves the integration of various tensors formed by $\vec{d}$ over z. Once projected onto the XY plane of the resonator, this field plays the role of essentially a static and random gauge field. It is also important to note that the orientational disorder of $\vec{d}$ precludes formation of the condensate of polaritons [26] (see in [27]). Despite that, there is a novel feature emerging—the geometrical pairing of photons due to the orientational disorder. This aspect of the system is one of the principal focuses of this work.

It is important to keep in mind the hierarchy of time scales. The shortest one is the thermalization time $\tau_t \sim 20$ ps [10]. It is about 2–3 orders of magnitude shorter than the photon life time in the cavity $\tau_{\rm ph} \sim$ few ns [10]. Thus, for all practical purposes the system can be treated as an equilibrium one despite external pumping at least on the time scale shorter than $\tau_{\rm ph}$ and in the limit of weak pumping. The translational and rotational dynamics of the dye subsystem is characterized by its own time scales. These are determined by dynamical viscosity η of the solvent (see in [28]). The rotational diffusion time $\tau_r \sim \eta a^3/T$, where $a \sim 1$ nm is a typical size of the dye molecule and T stands for temperature (in energy units). For typical viscosity $\eta \sim 10^{-3}$ Pa s, this time is $\tau_r \sim$ 1–10 ns. Thus, the orientational disorder in $\vec{d}$ can be considered as static on time scales $t \ll \tau_r$ only. Beyond this time the dipolar variable $\vec{d}$ must be treated as a dynamical (diffusive) degree of freedom. The translational diffusion of the dye molecules is characterized by a spread of times—from the travel time $\tau_{trc} \sim \eta R_n^2 a/T$ to its closest neighbor (about $R_n \sim 10$ nm apart) to the time $\tau_{\rm cl} \sim \eta R_{\rm cl}^2 a/T$ it takes to diffuse on some typical spatial scale $R_{\rm cl}$. Taking $R_{\rm cl} \sim 1 - 10$ μm (comparable to the size of the condensate in [10]), this range covers from $\tau_{trc} \sim 10^{-7} - 10^{-6}$ s to $\tau_{\rm cl} \sim 0.1 - 1$ s. Similarly to the rotational degree of freedom, center of mass positions of the molecules should be considered as dynamical (diffusive) degrees of freedom on times longer than τ_{trc}.

Here the main analysis is conducted within the approximation that the dynamics of TLS centers is frozen. That is, the focus is on the situation of short times t such that $\tau_t < t < \tau_{\rm ph} \ll \tau_r \ll \tau_{trc}$.

3. The Hamiltonian and the effective action

The Hamiltonian H consists of three parts

$$H = H_{\rm ph} + H_{\rm TLS} + H_{\rm int}, \tag{2}$$

where the first term accounts for the free photons inside the cavity; the second term describes the TLSs and the last one stands for the interaction between the photons and TLSs. Explicitly,

$$H_{\rm ph} = \int {\rm d}^2x \left[\frac{\hbar^2}{2m} \nabla_i \psi_j^\dagger \nabla_i \psi_j - \mu_0 \psi_j^\dagger \psi_j \right], \tag{3}$$

where $\int {\rm d}^2x ... = \int {\rm d}x{\rm d}y ...$; $m = \hbar\omega_0/c^2$ stands for the effective mass of photons (induced by the dimensional quantization), and μ_0 is the effective chemical potential of excitations introduced into the system by external pumping. These include photons and excited TLS (see more details in the appendix A. The summation over the repeated coordinate indices (X, Y) here and below is implied.

The interaction can be written in terms of the photonic electric field $\vec{E}$ and the TLS centers in the minimal form within the RWA as

$$H_{\rm int} = -\sum_{x,z} [\vec{d}\,\vec{E}\,\sigma^+ + {\rm h.c.}], \tag{4}$$

where the summation runs over the locations $(\vec{x},\ z)$ (with $\vec{x} = (x,\ y)$) of the centers and σ^+, σ^- are the Pauli matrices describing absorption and emission of photons, respectively, by each TLS. (The component $E_z \sim \vec{\nabla}\vec{\psi}/q_0$ is ignored in equation (4) in the limit of small momenta along X, Y directions.) The TLS energy can be accounted for by

$$H_{\rm TLS} = \sum_{\vec{x},z} \delta \cdot \sigma_z, \tag{5}$$

with δ standing for the detuning of the TLS energy ϵ_0 from the energy $\hbar\omega_0$ of the condensed photonic mode (shifted by the chemical potential), and σ_z being the Pauli matrix.

Since we excluded vibrons from the consideration, the quantity ϵ_0 should be attributed to Zero Phonon Line of the electronic transition in the dye molecule. In general, however, vibrons can change this interpretation. In what follows δ will be considered as a free parameter. More details on the dynamical properties of the system as well as on the derivation of the low energy action are given in the appendix A.

3.1. Free energy

In the presence of macroscopic occupation of photons forming the field $\vec{\psi}$, the quantum nature of $\vec{\psi}$, $\vec{\psi}^\dagger$ can be safely ignored. Such an approach is the basis for describing superfluids by classical fields $\vec{\psi}$, $\vec{\psi}^*$ within the GP equation (see in [29, 30]). This method is applicable in 2D at finite T as well (see in [30]) despite the absence of the true ODLRO which is replaced by algebraic off diagonal order (and which we will be loosely referring to as 'condensate').

In the presence of the condensate, the TLS contribution to the partition function can be calculated explicitly. Indeed, the thermal operator $\exp(-\beta(H_{\rm TLS} + H_{\rm int}))$, with $\beta = 1/T$, can be represented as a product $\prod_i \exp(-\beta H_i)$ over each TLS where H_i is the contribution from the ith TLS to the terms (5), (4). Then, using the identity $\exp(\vec{B}\vec{\sigma}) = \cosh(|\vec{B}|) + \sinh(|\vec{B}|)\vec{B}\vec{\sigma}/|\vec{B}|$ for Pauli matrices $\vec{\sigma}$ and real vector $\vec{B}$ and keeping in mind that all the components of $\vec{\sigma}$ are traceless, the contribution to the partition function $Z = Z[\vec{\psi},\ \vec{\psi}^*]$ evaluated with respect to the electronic degrees of freedom becomes

$$Z[\vec{\psi},\ \vec{\psi}^*] = \prod_i Z_i, \quad Z_i = \cosh\left(\beta\sqrt{\delta^2 + |\vec{d}(i)\vec{E}(i)|^2}\right), \tag{6}$$

where the coordinate dependence of $\vec{d}$ and $\vec{E}$ at the location of ith TLS is shown in a short hand manner $(\vec{x},\ z) \to (i)$.

At this point it is important to note that, depending on the time scale of the experiment, the effective action for the photonic field should be derived differently. At times shorter than the time of the molecular reorientation $\tau_r \sim 1 - 10$ ns, the contribution to free energy from ith TLS should be taken as $U_i = -T \ln Z_i$, with the orientation of $\vec{d}_i$ treated as a non-dynamical (quenched) disorder variable. This gives the total free energy from all TLS molecules

$$U_\psi([\vec{\psi}],\ [\vec{d}]) = -T\sum_i \ln\left[\cosh\left(\beta\sqrt{\delta^2 + |\vec{d}(i)\vec{E}(i)|^2}\right)\right]. \tag{7}$$

It is worth noting that equation (7) is essentially the same as the one obtained in [31] (in the case of the triple degenerate dipolar transition), although derived from a different perspective. (We don't include the diamagnetic term [32] because in the context of the pumped cavity it simply redefines the chemical potential and the detuning.) After calculating all averages of the photonic variables with the energy $U_\psi([\vec{\psi}],\ [\vec{d}])$, the final averaging over the quenched disorder should be performed.

At longer times, the molecules will be able to adjust their orientation in the presence of the photonic condensate $\vec{\psi}$. In this limit the molecular orientation should be integrated out from Z_i, that is, $Z_i \to \int {\rm d}\Omega_i Z_i$,

where Ω_i stands for the solid angle as a measure of the orientation of $\vec{d}$. Then, the free energy can be calculated as

$$U_\psi([\vec{\psi}]) = -T\sum_i \ln\left[\int d\Omega_i \cosh\left(\beta\sqrt{\delta^2 + |\vec{d}(i)\vec{E}(i)|^2}\right)\right]. \tag{8}$$

The detailed analysis of the situation described by equation (8) will be conducted elsewhere. As mentioned above, the present focus is on the short time limit, equation (7).

While considering low energy properties, it is reasonable to resort to a coarse grained description and, thus, to replace the summation $\sum_i ...$ over locations of TLSs by integration $\int dz dy dx n ...$ where $n = n(x, y, z)$ stands for the coarse grained density of TLSs (which is not necessarily uniform). At this point a comment about a possible degeneracy of the TLS transition is in order. If the TLS molecules were fully symmetric, the dipolar transition would be triple degenerate. Accordingly, each component of $\vec{E}$ would see no preferential orientation of the molecules, and a contribution to free energy of one molecule exposed to the condensate of light would depend on the product $d|\vec{E}|$, where $d = |\vec{d}|$. Then, the free energy U, equation (7), would depend only on the modulus $|\vec{E}|$. In other words, the term $|\vec{d}\vec{E}|^2$ in equation (7) should be replaced by $d^2|\vec{E}|^2$. Accordingly, the effective action (represented as a Landau expansion [29]) would depend on $|\vec{\psi}|^2$ and its higher powers. This implies the O(4) symmetry of the effective action. It is important that in 2D systems the symmetries higher than O(2) preclude condensation at any finite temperature even in the algebraic sense [33]. This aspect has been emphasized in the context of polariton condensation in [34] and remains valid in the case under consideration too. Thus, in our analysis it is important that the TLS transition is not triple degenerate.

Here we consider the limit $|\delta| \gg |\vec{d}\vec{E}|$ validating the separation of fast and slow variables. Thus, U in equation (7) can be expanded in $1/\delta$ up to the quartic order in $\vec{\psi}$ as

$$U_\psi = \int d^2x[-d_{ij}\psi_i^*\psi_j + d_{ijkl}\psi_i^*\psi_j^*\psi_k\psi_l], \tag{9}$$

where the tensors

$$d_{ij} = c_2\int dz n d_i d_j |\phi(z)|^2, \quad d_{ijkl} = c_4\int dz n d_i d_j d_k d_l |\phi(z)|^4 \tag{10}$$

are introduced (with $\phi(z)$ defined in equation (1)) and

$$c_2 = \frac{\tanh(\beta\delta)}{2\delta}, \tag{11}$$

$$c_4 = \frac{\tanh(\beta\delta)}{8\delta^3} - \frac{1}{8T\delta^2\cosh^2(\beta\delta)}. \tag{12}$$

It is worth mentioning that $c_4 > 0$ and varies from $c_4 = 1/(8\delta^3)$ for $\delta \gg T$ to $c_4 = 1/(12T^3)$ in the opposite limit.

The summations in equation (9) run over the X, Y directions only (because $\vec{\psi}$ has only X, Y components). Thus, d_{ij} and d_{ijkl} become 2D tensors. Then, it is convenient to introduce the representation

$$d_{ij} = \mu_\parallel n_i n_j + \mu_\perp m_i m_j \tag{13}$$

in terms of the local frame where d_{ij} is diagonal and $\mu_\parallel$, $\mu_\perp$ are its eigenvalues. The unit vectors $\vec{n}$, $\vec{m}$ can be represented as $\vec{n} = (\cos\theta, \sin\theta)$, $\vec{m} = (\sin\theta, -\cos\theta)$ by the angle $\theta = \theta(x, y)$ with respect to, say, the X-axis. These vectors satisfy the orthogonality condition $\vec{n}\cdot\vec{m} = 0$ so that the field $\vec{\psi}$ can be expanded as

$$\vec{\psi} = \vec{n}\Phi + i\vec{m}\Phi_\perp, \tag{14}$$

where Φ, $\Phi_\perp$ are complex coordinates of $\vec{\psi}$ in the local frame ($\vec{n}$, $\vec{m}$). Accordingly, the energy (9) together with the free photonic part (3) becomes

$$U = \int d^2x\left[\frac{\hbar^2}{2m}(|\vec{\nabla}\Phi_\perp + i\vec{\nabla}\theta\Phi|^2 + |\vec{\nabla}\Phi + i\vec{\nabla}\theta\Phi_\perp|^2) - \mu_\Phi|\Phi|^2 - \mu_{\Phi_\perp}|\Phi_\perp|^2 + U_4\right], \tag{15}$$

where $\mu_\Phi = \mu_0 + \mu_\parallel$, $\mu_{\Phi_\perp} = \mu_0 + \mu_\perp$; the free photonic chemical potential μ_0 was introduced in equation (3); and the specific form of the quartic term U_4 from equation (9) will be discussed below. The energy (15) represents the potential part of the full GP action S derived in appendix A, equation (A11).

4. Symmetries of the photonic condensate

In most cases the TLS transition is non-degenerate [11, 25, 35]. The case of a double degenerate transition is qualitatively the same as the one considered below as long as the orientation of $\vec{d}$ is not aligned with the XY plane. In this section, we will first consider the case of full isotropy of the tensors d_{ij}, d_{ijkl}, equation (10).

4.1. Emerging isotropy

At short times when the field $\vec{d}\,(x,\,y,\,z)$ can be treated as a static disorder variable, it is natural to assume that the integration along Z-direction in equation (10) returns isotropic tensors—because the distance L_z (which is $\approx$1.5 μm) is much larger than the inter TLS separation (of the order of few nm). This allows for averaging over isotropic 3D orientations of $\vec{d}\,(x,\,y,\,z)$ in equation (10), which gives

$$d_{ij} = \mu_{\parallel}\delta_{ij}, \quad d_{ijkl} = g\,(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}), \tag{16}$$

where $\mu_{\parallel} = \mu_{\perp} = \frac{c_2 d^2}{3}\int \mathrm{d}z n|\phi|^2$ and $g = \frac{c_4 \vec{d}^4}{15}\int \mathrm{d}z n|\phi|^4$, and c_2, c_4 are given in equations (11) and (12). It is worth mentioning that in the Kronecker symbols the indices run over X, Y, Z directions while the summation in equation (9) is performed only over X, Y. Accordingly, the substitution of equations (16) into (9) gives 2D density of the quartic part of U_ψ as

$$U_4 = g\,(2|\vec{\psi}|^4 + (\vec{\psi}^*)^2(\vec{\psi})^2), \tag{17}$$

or in terms of the representation (14):

$$U_4 = g\,(3(|\Phi|^4 + |\Phi_\perp|^4) + 4|\Phi|^2|\Phi_\perp|^2 - (\Phi^{*2}\Phi_\perp^2 + \text{c.c.})), \tag{18}$$

which should be used in equation (15) above. These expressions are obtained under the assumption of self-averaging of the molecular orientations despite the fact that they are frozen in at short time scales.

4.2. O(4) versus O(2) × Z_2 symmetry

At this point it is important to note that, given $\mu_{\parallel} = \mu_{\perp}$ and without the last term in U_4, equation (17), the symmetry of the free energy (15) is O(4). In this case, the vectors $\vec{n}$ and $\vec{m}$ in the representation (14) can be gauged away and taken as aligned with the XY axes. Thus, the gauge field $\vec{\nabla}\theta = 0$ in equation (15). As mentioned above, the formation of the off-diagonal algebraic order of O(4) symmetry is impossible at any finite T. The situation is changed by the term $\sim(\vec{\psi}^*)^2(\vec{\psi})^2$ which breaks the O(4) symmetry down to O(2) × Z_2. This makes the formation of the algebraic order possible (see [34]).

Introducing real $\vec{a}$ and imaginary $\vec{b}$ parts of $\vec{\psi} = \vec{a} + \mathrm{i}\vec{b}$, that is, $\Phi = a_x + \mathrm{i}b_x$, $\Phi_\perp = a_y + \mathrm{i}b_y$ in equation (14), the uniform term in equation (15) becomes

$$U = -\mu_{\Phi_\perp}(\vec{a}^2 + \vec{b}^2) + g\,(3(\vec{a}^2 + \vec{b}^2)^2 - 4(\vec{a}\times\vec{b})^2) - ((\vec{\eta} + \vec{\eta}^*)\vec{a} + \mathrm{i}(\vec{\eta}^* - \vec{\eta})\vec{b}), \tag{19}$$

where for the sake of generality, the term linear in $\vec{\psi}$ and $\vec{\psi}^*$ induced by the external pumping $\sim\vec{\eta}$ [10] has been introduced. In what follows this term will be ignored which corresponds to the limit of weak pumping. In section 5 the role of this term as a testing tool will be specifically addressed.

At the mean field level the condensation corresponds to $\mu_{\Phi_\perp} > 0$. The lowest energy of the functional at $\vec{\eta} = 0$ is achieved for $\vec{b}\perp\vec{a}$ and $\vec{a}^2 = \vec{b}^2 = \mu_{\Phi_\perp}/(8g)$. Explicitly, $b_x = a_y$, $b_y = -a_x$ or $b_x = -a_y$, $b_y = a_x$ so that

$$\psi_x = a_x \pm \mathrm{i}a_y, \quad \psi_y = a_y \mp \mathrm{i}a_x, \tag{20}$$

where the sign $\pm$ is correlated in both equations and it represents two directions of the circularly polarized light. Thus, the ground state of the condensed light is characterized by a spontaneous circular polarization—left or right handed. This corresponds to the Z_2 symmetry. Rotation of the vector $\vec{a}$ (together with $\vec{b}$) in the XY-plane implies O(2) (or U(1)) symmetry. Accordingly, the BEC transition belongs to the O(2) × Z_2 universality class, which, in general, differs from the Berezinskii–Kosterlitz–Thouless (BKT) scenario characteristic of O(2) symmetry. On the phase diagram, figure 2, this transition corresponds to the point $\mu_{\Phi_\perp} = \mu_0 + \mu_{\parallel} = 0$, $\Delta\mu = 0$ labeled as 'O(2) × Z_2'.

The emerging order is algebraic in 2D because of the gaussian rotational fluctuations of the vectors $\vec{a}$ and $\vec{b}$ locked to each other. Thus, the one-photon density matrix (OPDM) behaves as $\langle\vec{\psi}^*(\vec{x})\vec{\psi}(0)\rangle \sim 1/|\vec{x}|^{1/K}$ with some index $K > 0$ determined by the parameters and temperature (see in [36]).

It should also be emphasized that the product $S_z = \vec{a}\times\vec{b} \sim \mathrm{i}\vec{\psi}^*\times\vec{\psi}$ represents the order parameter characterized by Z_2 symmetry—similarly to 2D Ising model for the spin variable $S_z \sim \pm 1$. It forms trivially as long as O(2) symmetry is broken. There is another option—the O(2) × Z_2 transition may proceed as two successive transitions, with the S_z condensing first and O(2) to follow as temperature lowers further down.

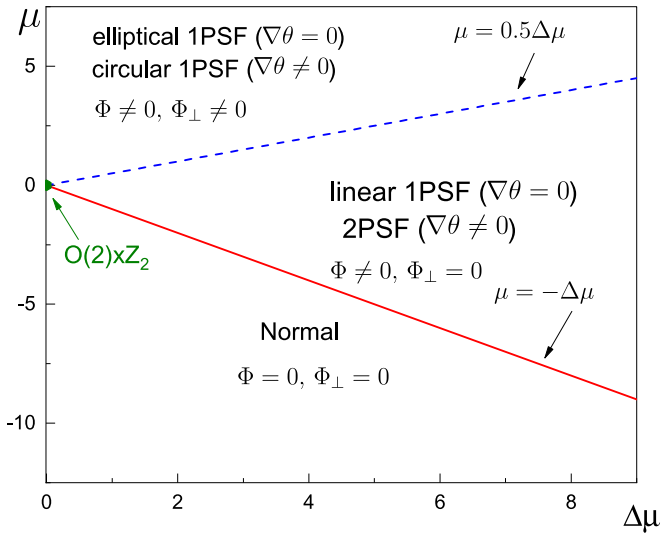

Figure 2. Mean field phase diagram ($\vec{\eta} = 0$, units are arbitrary). Here $\Delta\mu = \mu_{\Phi} - \mu_{\Phi_\perp}$ and $\mu = \mu_{\Phi_\perp}$. For the other notations see the text.

We note that the field theories with U(1) $\times$ Z_2 symmetry describe multi-band superconductors exhibiting spontaneous time reversal symmetry breaking [37–39]. These feature a variety of topological excitations [37] and also allow for a splitting of the transition into two [39]. A significant difference between these systems and the condensate of light lies in the structure of order parameters—scalars in [37–39] and a complex vector $\vec{\psi}$ for the light. This, in particular, determines the effect which is not present in superconductors—paired photonic condensate induced by non-Abelian gauge disorder. This effect, which represents the main result of our work, is described below. Here and below we will be using the abbreviation *m*PSF (*m*-photon superfluid) to characterize algebraic order occurring in *m*-photon ($m = 1, 2$) density matrix.

4.3. Dipolar anisotropy: the uniform case

Let us consider a situation when there is a residual dipolar anisotropy characterized by some finite field $\vec{d}_0(\vec{x})$ which is a result of the microscopic averaging of $\vec{d}\,(x, y, z)$. It can be induced by external fields (see appendix B) or form spontaneously as a part of the orientational disorder (on a time scale shorter than τ_r). Then, the tensor d_{ij} in equation (10) will deviate from the isotropic form (16) so that its eigenvalues $\mu_{\|}$, $\mu_{\perp}$ become unequal to each other. Without loss of generality we will assume that $\Delta\mu = \mu_{\Phi} - \mu_{\Phi_\perp} = \mu_{\|} - \mu_{\perp} > 0$.

Then, the order will form first in Φ while $\Phi_\perp = 0$ in the representation (14). As follows from the minimization of U in equation (15), this occurs in the range $\mu_{\Phi_\perp} < 0$ and $\mu_{\Phi} = \mu_{\Phi_\perp} + \Delta\mu > 0$.

If $\vec{d}_0$ is uniform in space, the emerging (algebraic) order is characterized by O(2) symmetry of the 1PSF linearly polarized along this vector (if ignoring $\vec{\eta}$ in equation (19)). This transition occurs along the solid line $\mu_{\Phi_\perp} = -\Delta\mu$ in figure 2. The dashed line, $\mu_{\Phi_\perp} = 0.5\Delta\mu$, corresponds to the condensation of the second field, $\Phi_\perp$, proceeding as Z_2 transition. As a result, the 1PSF becomes elliptically polarized above this line.

There is also a possibility of a non-uniform one-component condensation if the residual anisotropy $\vec{d}_0$, while persisting on a mesoscale, self averages to zero on the spatial scale of the cavity. Under this condition the 2PSF phase forms.

4.4. Non-uniform dipolar anisotropy: geometrically paired photonic condensate

Now let us consider a situation when $\vec{d}_0$ is not spatially uniform. One possibility how this can be achieved is discussed in appendix B. We start with the case $|\vec{d}_0|$= const. Then, the local eigenvalues $\mu_{\|}$, $\mu_{\perp}$ of d_{ij} are uniform while the orientation of the tensor is set by a non-uniform $\vec{\nabla}\theta \neq 0$. Since the threshold for the condensation is controlled by the quadratic part of the free energy (15), we will ignore anisotropy of the quartic term and will consider it isotropic as represented in equation (18).

It is important to note that a long wave structure of the field $\vec{\nabla}\theta$ without windings of the angle θ will not produce any significant deviation from the results discussed above in section 4.3 as long as the scale of the variations is larger than the healing length. The most interesting option occurs when $\vec{\nabla}\theta$ is characterized by topological defects—vortices. (These defects can be created by imposing external Gaussian–Laguerre beams—as

discussed in appendix B.) Let us consider a situation when there is a plasma of such proliferated (and frozen in) vortex-antivortex pairs of the field $\theta(\vec{x})$. If the distance between these vortices ξ_d is larger than the healing length l_h, the phase $\Phi \neq 0$, $\Phi_\perp = 0$ (realized in between the two lines in figure 2) persists. Then, setting $\Phi_\perp = 0$ in equation (15) the free energy becomes

$$U = \int \mathrm{d}^2x \left[\frac{\hbar^2}{2m} |\vec{\nabla}\Phi_\perp|^2 + \left(\frac{\hbar^2(\vec{\nabla}\theta)^2}{2m} - \mu_{\mu_\perp} - \Delta\mu \right) |\Phi_\perp|^2 + 3g|\Phi_\perp|^4 \right]. \tag{21}$$

Thus, the gauge effect of the field $\vec{\nabla}\theta$ vanishes. The remaining term $\sim(\vec{\nabla}\theta)^2$ contributes to a weak suppression of the local chemical potential—as long as $\xi_d \gg l_h$. According to the Harris criterion (see in [30]), the disorder produced by $\sim(\nabla\theta)^2$ in equation (21) is diagonal and, thus, is irrelevant at the transition marked by the solid line in figure 2. This means that the transition of Φ remains of the BKT type even in the presence of the disordered field $\sim(\nabla\theta)^2$. (The solid line in the phase diagram figure 2 shifts slightly upward by the term $\approx \frac{\hbar^2(\vec{\nabla}\theta)^2}{2m} \sim \hbar^2/(m\xi_d^2) \ll \mu_\Phi$).

It is important to note that, despite the emerging order in the field Φ, the one-photon density matrix (OPDM) becomes disordered—as long as θ contains frozen in windings. These windings are now imprinted onto the physical field $\vec{\psi} = \vec{n}\Phi$ through the factor $\vec{n}$. Accordingly, the OPDM becomes

$$\langle \psi_i^*(\vec{x})\psi_j(\vec{x}')\rangle = n_i(\vec{x})n_j(\vec{x}')\langle \Phi^*(\vec{x})\Phi(\vec{x}')\rangle. \tag{22}$$

Since $\vec{n}$ and Φ are not, practically, coupled, the averaging over the disorder $\langle ...\rangle_\theta$ in equation (22) can be applied to the factor $n_i(\vec{x})n_j(\vec{x}')$ only. At large distances n_i can be considered as gaussian random variables coarse grained over patches of size $\sim\xi_d$. This produces exponential behavior $\langle n_i(\vec{x})n_j(\vec{x}')\rangle_\theta \sim \exp(-|\vec{x}-\vec{x}'|/\xi_d)$ and, accordingly, $\langle \psi_i^*(\vec{x})\psi_j(\vec{x}')\rangle \sim \exp(-|\vec{x}-\vec{x}'|/\xi_d) \to 0$, as long as $|\vec{x}-\vec{x}'| \gg \xi_d$.

In drastic contrast, the TPDM $\rho^{(2)}_{ijkl} = \langle \psi_i^*(\vec{x})\psi_j^*(\vec{x})\psi_k(\vec{x}')\psi_l(\vec{x}')\rangle$ retains the algebraic order. The simplest way to see this is to consider the scalar product $\vec{\psi}^2$ which, after taking into account equation (14) becomes $\vec{\psi}^2 = \Phi^2$. That is, $\rho^{(2)}_{iikk} = \langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle \sim |\vec{x}-\vec{x}'|^{-2/K}$.

In general,

$$\rho^{(2)}_{ijkl} = \langle n_i(\vec{x})n_j(\vec{x})n_k(\vec{x}')n_l(\vec{x}')\rangle_\theta \cdot \langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle. \tag{23}$$

At distances larger than ξ_d, the averaging over the disorder gives $\langle n_i(\vec{x})n_j(\vec{x})n_k(\vec{x}')n_l(\vec{x}')\rangle_\theta = \delta_{ij}\delta_{kl}/4$. At distances $|\vec{x}-\vec{x}'| < \xi_d$ the factor $\langle n_i n_j n_k n_l\rangle_\theta$ becomes $\sim(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})/8$. Overall, however, $\rho^{(2)}_{ijkl} \propto \langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle \sim |\vec{x}-\vec{x}'|^{-2/K}$. More specifically, the component $\rho^{(2)}_{xxxx}$ changes from $(3/8)\langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle$ at $|\vec{x}-\vec{x}'| < \xi_d$ to $(1/4)\langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle$ in the opposite limit. For the component $\rho^{(2)}_{xxyy}$ the corresponding limits are $(1/8)\langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle$ and $(1/4)\langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle$. Finally, for the component $\rho^{(2)}_{xyxy}$ the limits are $(1/8)\langle (\Phi^*(\vec{x}))^2(\Phi(\vec{x}'))^2\rangle$ and $\to 0$. This behavior describes the 2PSF phase (labeled as '2PSF ($\vec{\nabla}\theta \neq 0$)' in figure 2). In this phase the order parameter becomes a complex tensor $\psi_i\psi_j$, while $\psi_i = 0$.

The condensation of the field $\Phi_\perp$ can restore the one-photon order by introducing vortices into the fields Φ, $\Phi_\perp$ which compensate partially the disorder created by $\theta(\vec{x})$. This mechanism corresponds to the transformation of the phase gradient $\vec{\nabla}\varphi$ of the fields Φ and $\Phi_\perp$ from being vortex free to acquiring windings as $\vec{\nabla}\varphi \to \vec{\nabla}\varphi = \vec{\nabla}\tilde{\varphi} - \vec{\nabla}\theta$, with $\tilde{\varphi}$ denoting the irrotational part of the phase. As a result, the OPDM of the physical field $\vec{\psi}$ acquires algebraic order of circularly polarized light. This phase occurs above the dashed line in figure 2 and it is labeled as 'circular 1PSF ($\vec{\nabla}\theta \neq 0$)'. More details on the 2PSF to 1PSF transformation are given in appendix C.

Finally, we note that including disorder in the modulus of $\vec{d}_0$ will affect the value of $\Delta\mu = \mu_\Phi - \mu_{\Phi_\perp}$ introduced in section 4.3. However, weak disorder fluctuations of $\Delta\mu$ should not modify the above conclusions —at least for the O(2) transition (see in [30]). Once the disorder becomes strong, a Bose glass phase can emerge. This requires that fluctuations of $\Delta\mu$ are comparable or stronger than μ_Φ itself.

5. Discussion

Our phenomenological analysis of condensed phases of light points out to multiple possibilities characterized by various symmetries and symmetry breaking patterns of PSF. The algebraic condensation of light in 2D can only occur if the TLS dipolar transition is not fully degenerate. Then, the resulting symmetry of the condensate is $O(2) \times Z_2$, and the algebraic long range order becomes possible. The orientational anisotropy in the TLS ensemble can break this transition into two.

The main prediction reported here is the geometrically paired photonic superfluid. At this point we emphasize that, in contrast to the paired condensates of photons [40] or ultracold atoms [41] requiring an effective attraction between photons, the pairing found here is of purely geometrical nature. It is induced by the non-Abelian disorder in the TLS ensemble, and it does not require any attraction between photons. This effect can be assigned into the class of phenomena named as *order by disorder* [42]. In some sense, there is a hidden algebraic 1PSF order in the non-observable field Φ while there is no algebraic order in the observable field $\vec{\psi}$ in the representation (14).

One option of detecting such a phase would be by controlling orientation of the dye molecules so that $\vec{d}$ of the dipolar transition can be ordered (see appendix B). Then, the non-observable field Φ will become observable because $\vec{n}$ in the representation (14) orders and, thus, restores the one-photon coherence in $\vec{\psi} \sim \vec{n}\Phi$. As discussed above, detecting 2PSF directly requires two-photon correlation spectroscopy.

It is worth mentioning that, in the presence of the pumping of light into the cavity (described by the bias $\vec{\eta}$ in equation (19)) the phases discussed above can be distorted and then destroyed. A strong external bias $\vec{\eta}$ may essentially lock the condensate polarization along the external field. (This what was likely observed in [43].) Upon lowering η the crossover to the spontaneous polarization should occur. This effect can serve as a measure of the interaction strength between photons. An estimate of the field strength for a crossover follows from comparing the last terms in equation (19) as $|\vec{\eta}| \approx g n_{\rm ph}^{3/2}$. A different situation occurs in the cases of the dipolar anistropy when the competition occurs between the residual $\vec{d}_0$ and $\vec{\eta}$. Thus, the phases will be destroyed if $|\vec{\eta}| \geqslant |\vec{d}_0|\sqrt{n_{\rm ph}}$.

The presented mean field analysis is only a first step toward detailed studies involving realistic geometries. It gives a qualitative assessment of the emerging possibilities. There are several important questions to answer. The nature of the 2PSF-1PSF transition involving the compensation of the frozen in vortices in the effective gauge field needs to be explored. To the best of our knowledge, there is no known analogy to this transition.

Another exciting aspect of the system is the possibility of dynamical interaction between PSF and essentially classical degrees of freedom of dye molecules—their translational and rotational diffusive dynamics. This poses fundamental questions about how classical diffusive bath affects off diagonal order and dynamics. Phase separation effect leads to forming regions of higher TLS density which invokes the necessity to consider the near field direct interaction between TLSs as initiated in [22]. In view of the emerging very promising experimental possibilities [17] going beyond BEC, these questions acquire much greater significance.

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Appendix A. GP functional

The Hamiltonian (2) conserves the total amount of photons and TLS molecules in their excited state. This can be explicitly seen after calculating the time derivative $\dot{\rho} = [\rho, H]/(\mathrm{i}\hbar)$ of the excitation density operator

$$\rho(\vec{x}, t) = \vec{\psi}^{\dagger}(\vec{x})\vec{\psi}(\vec{x}) + \sum_{\vec{x}_i, z_i} \frac{1}{2}\sigma_z(\vec{x}_i, z_i)\delta^{(2)}(\vec{x} - \vec{x}_i) \tag{A1}$$

which leads to the 2D continuity equation

$$\frac{\partial \rho}{\partial t} + \vec{\nabla}\vec{J} = 0, \tag{A2}$$

where

$$\vec{J} = \frac{\hbar}{2mi}[\psi_j^{\dagger}\vec{\nabla}\psi_j - \text{c.c.}] \tag{A3}$$

is the photon current 2D density; $\delta^{(2)}(\vec{x} - \vec{x}_i)$ is 2D delta-function defined in the space $\vec{x} = (x, y)$.

It is important to emphasize that, in the absence of a direct exchange interaction between TLS molecules, the energy transfer is carried by photons only while storage of the energy is due to both—photons and excitations of TLSs.

In order to describe (slow) dynamics of the condensed light, the Berry term $S_{\rm B}$ in the effective action $S = S_{\rm B} - \int \mathrm{d}t U$, where t is real time, should be constructed. Within the standard GP approach it is

$$S_B^{(0)} = \int \mathrm{d}t \int \mathrm{d}^2x\ \mathrm{i}(\hbar/2)\vec{\psi}^*\dot{\vec{\psi}} + \text{c.c.} \tag{A4}$$

This form guarantees the conservation of photons and leads to the continuity equation $\partial|\vec{\psi}|^2/\partial t + \vec{\nabla}\vec{J} = 0$.

Since the actual conservation is given by equation (A1), the GP equation following from $\delta S/\delta\vec{\psi}^* = 0$ must feature the continuity equation

$$\frac{\partial(|\vec{\psi}|^2 + \rho_{\mathrm{ex}})}{\partial t} + \vec{\nabla}\vec{J} = 0, \tag{A5}$$

where we have introduced the local density of the excited TLS centers ρ_{ex}.

Since TLSs represent fast degrees of freedom, ρ_{ex} can be found within the approximation of local equilibrium with respect to TLS ensemble as

$$\rho_{\mathrm{ex}} = n \cdot (1 + \langle\sigma_z\rangle)/2, \tag{A6}$$

where the averaging $\langle\sigma_z\rangle$ is done over the ground and excited TLS states in the presence of the condensed field $\vec{E}$ with the equilibrium Boltzmann weight. The diagonalization of H in equation (2) within this approximation gives

$$\rho_{\mathrm{ex}} = \frac{1}{2}\left[1 - \frac{\delta\tanh(\beta\epsilon)}{\epsilon}\right]n, \tag{A7}$$

where

$$\epsilon = \sqrt{\delta^2 + |\vec{d}\vec{E}|^2}\,. \tag{A8}$$

Then, the Berry term consistent with the conservation law (A5) follows as $S_B = S_B^{(0)} + S_e$ where

$$S_e = \frac{\mathrm{i}\hbar}{2}\int \mathrm{d}t \int \mathrm{d}^2x\mathrm{d}z \frac{(\vec{d}\vec{E}^*)(\vec{d}\dot{\vec{E}})}{|\vec{d}\vec{E}|^2}\rho_{\mathrm{ex}} + \text{c.c.} \tag{A9}$$

In order to project the form (A9) into the XY plane, the integration along Z-direction must be performed. This can be done straightforwardly as an expansion in $1/\delta$. Expanding ρ_{ex} up to the first term $\sim|\vec{d}\vec{E}|^2$ and, then, integrating over Z,

$$\rho_{ex} = \rho_0 + b(\mu_{\|}|\Phi|^2 + \mu_{\perp}|\Phi_{\perp}|^2), \tag{A10}$$

where $\rho_0 = \frac{1}{2}(1 - \tanh(\beta\delta))\int \mathrm{d}z n$ and the representation (14) has been used; $b = 2\delta c_4/c_2$.

Representing $\Phi = |\Phi|\mathrm{e}^{\mathrm{i}\varphi}$ and $\Phi_{\perp} = |\Phi_{\perp}|\mathrm{e}^{\mathrm{i}\varphi_{\perp}}$ and substituting into equations (A9) and (A10) and dropping full time derivative terms, the GP action S can be written as

$$S = \int \mathrm{d}t \int \mathrm{d}^2x[-\hbar((1 + b\mu_{\|})|\Phi|^2\dot{\varphi} + (1 + b\mu_{\perp})|\Phi_{\perp}|^2\dot{\varphi}_{\perp}) - U], \tag{A11}$$

where U is given in equation (15).

Appendix B. Inducing anisotropy

Equilibrium (weak) anisotropy in the cumulants of d_i can be created by external electric field $\vec{E}^{(\mathrm{ex})}$ imposed at optical frequency different from the photonic condensate—either by short pulses (as used to observe optical Kerr gating [44]) or by stationary illumination. Using techniques for creating optical vortices [45, 46] and their arrays [47], the field $\vec{E}^{(\mathrm{ex})}$ can induce non-uniform polarization of the dye molecules with imprinted windings in $\vec{d}$.

Let's estimate a magnitude of the effect in the equilibrium situation. The corresponding polarization energy contribution (in the second order of the perturbation) is $U^{(\mathrm{ex})} = -|\vec{E}^{(\mathrm{ex})}\vec{d}|^2/\delta^{(\mathrm{ex})}$, where $\delta^{(\mathrm{ex})}$ stands for the detuning of the optical field $\vec{E}^{(\mathrm{ex})}$ from the resonance. Then, the tensor $d_{ij} \sim \langle d_i d_j\rangle$ (in equation (10)) averaged over the orientations with the Boltzmann factor $\sim\exp(-U^{(\mathrm{ex})}/T)$ in the RWA becomes

$$d_{ij} \sim \langle d_i d_j\rangle = \frac{\vec{d}^2}{3}\left(1 - \frac{2|\vec{E}^{(\mathrm{ex})}|^2}{15\delta^{(\mathrm{ex})}T}\right)\delta_{ij} + \frac{\vec{d}^4}{15\delta^{(\mathrm{ex})}T}(E_i^{(\mathrm{ex})*}E_j^{(\mathrm{ex})} + \text{c.c.}). \tag{B1}$$

If the field $\vec{E}^{(\mathrm{ex})}$ is uniform, the case of the uniform anisotropy considered above in section 4.3 can be realized. The corresponding phase is labeled as 'linear 1PSF ($\nabla\theta = 0$)' in figure 2.

In order to create the anisotropy with windings discussed in section 4.4 the external field should be formed by combinations of Laguerre–Gaussian beams. As a demonstrative example, let's consider two circularly

polarized standing waves (along Z) with the orbital momenta $l = \pm 1$. Such two waves can be represented by the complex amplitudes

$$E_+ = E_x^{(\mathrm{ex})} + \mathrm{i}E_y^{(\mathrm{ex})} = u_+(r, z)\mathrm{e}^{+\mathrm{i}\theta}, \tag{B2}$$

$$E_- = E_x^{(\mathrm{ex})} - \mathrm{i}E_y^{(\mathrm{ex})} = u_-(r, z)\mathrm{e}^{-\mathrm{i}\theta}, \tag{B3}$$

where $u_\pm$ (r, z) are real amplitudes; $r = \sqrt{x^2 + y^2}$; and θ is the polar angle in the XY-plane. Then, the anisotropic part $\tilde{d}_{ij}$ of d_{ij}, equation (B1), (that is, $\tilde{d}_{ij} \sim (E_i^{(\mathrm{ex})*}E_j^{(\mathrm{ex})} + \mathrm{c.c.}) - |\vec{E}^{(\mathrm{ex})}|^2\delta_{ij}$) becomes

$$\begin{bmatrix} \tilde{d}_{xx} & \tilde{d}_{xy} \\ \tilde{d}_{yx} & \tilde{d}_{yy} \end{bmatrix} \sim u_+u_- \begin{bmatrix} \cos(2\theta) & \sin(2\theta) \\ \sin(2\theta) & -\cos(2\theta) \end{bmatrix}. \tag{B4}$$

This term selects the orientation of the vectors $\vec{n}$, $\vec{m}$ in the representation (13), (14) favoring condensation of Φ (while $\Phi_\perp = 0$). (The vectors $\vec{n}$, $\vec{m}$ introduced in equations (13), (14) are eigenvector of the matrix (B4) with the corresponding eigenvalues $\pm u_+u_-$). The resulting phase, then, is 2PSF (labeled as '2PSF ($\nabla\theta \neq 0$)' in figure 2).

Strength of the anisotropy can be estimated as the ratio of the anisotropic to the isotropic terms in equation (B1). Then, $\Delta\mu/\mu_\parallel$ introduced in section 4.3 can be estimated as

$$\frac{\Delta\mu}{\mu_\parallel} \approx \frac{\vec{d}^2}{\delta^{(\mathrm{ex})}T}|\vec{E}^{(\mathrm{ex})}|^2. \tag{B5}$$

This expression is valid as long as the condition $U^{(\mathrm{ex})} \ll T$ holds. For $E^{(\mathrm{ex})} \sim 100$ kV cm^{-1} (which corresponds to energy density $\sim 10^{-3}$ J cm^{-3}), $d \sim 1$ Debye, $\delta^{(\mathrm{ex})} \sim 0.1$ eV and room temperature, this condition is well satisfied, with the corresponding anisotropy value $\frac{\Delta\mu}{\mu_\parallel} \sim 10^{-4}$.

Appendix C. Transition 2PSF-1PSF

In the case when both fields are condensed, the mean field solution for (15) gives

$$\begin{aligned} \Phi &= \Phi_1 \exp(\mathrm{i}\varphi),\ \Phi_\perp = \Phi_2 \exp(\mathrm{i}\varphi), \\ \Phi_1^2 &= \frac{2\mu_{\Phi_\perp} + 3\Delta\mu}{16g},\ \Phi_2^2 = \frac{2\mu_{\Phi_\perp} - \Delta\mu}{16g} > 0, \end{aligned} \tag{C1}$$

which is valid for $\mu_{\Phi_\perp} > 0.5\Delta\mu$. Here the phase φ is to be determined from the minimum of the gradient part

$$\Delta U = \int \mathrm{d}^2x \frac{\hbar^2}{2m}[(\Phi_1\vec{\nabla}\varphi + \Phi_2\vec{\nabla}\theta)^2 + (\Phi_2\vec{\nabla}\varphi + \Phi_1\vec{\nabla}\theta)^2] \tag{C2}$$

of the energy functional (15).

Let us assume that the field $\vec{\nabla}\theta$ has just a single vortex. Then, the minimum of ΔU can be reached either for $\vec{\nabla}\varphi = 0$ (which corresponds to 2PSF) or for $\vec{\nabla}\varphi = -\vec{\nabla}\theta$, which corresponds to circulary polarized 1PSF. Comparison of these two energies leads to the condition

$$\frac{\Phi_2^2}{\Phi_1^2} < 7 - 4\sqrt{3} \tag{C3}$$

for the existence of the 2PSF. Using the solution (C1) in equation (C3) gives

$$\mu_{\Phi_\perp} < \frac{3\sqrt{3} - 5}{2\sqrt{3} - 3}\Delta\mu \approx 0.423\Delta\mu, \tag{C4}$$

which conflicts with the requirement $\mu_{\Phi_\perp} > 0.5\Delta\mu$ needed to have $\Phi_2^2 > 0$.

Thus, for $\mu_{\Phi_\perp} > 0.5\Delta\mu$ the minimum of ΔU is achieved for $\vec{\nabla}\varphi = -\vec{\nabla}\theta$, that is when the phase of the fields Φ, $\Phi_\perp$ contains antivortex. In this case the components of the full field $\vec{\psi}$ become

$$\psi_x = \left[\frac{\Phi_1 + \Phi_2}{2} + \frac{\Phi_1 - \Phi_2}{2}\mathrm{e}^{2\mathrm{i}\theta}\right]\mathrm{e}^{\mathrm{i}\tilde{\varphi}}, \tag{C5}$$

$$\psi_y = -\mathrm{i}\left[\frac{\Phi_1 + \Phi_2}{2} + \frac{\Phi_2 - \Phi_1}{2}\mathrm{e}^{2\mathrm{i}\theta}\right]\mathrm{e}^{\mathrm{i}\tilde{\varphi}}, \tag{C6}$$

where $\tilde{\varphi}$ is a non-winding part of φ (accounting for non-topological excitations—phonons of 1PSF). The terms $\sim\exp(2\mathrm{i}\theta)$ containing the winding field are washed out at large distances, and the coherent part of the total field $\vec{\psi}$ becomes

$$\psi_x = \frac{1}{2}(\Phi_1 + \Phi_2)\exp(\mathrm{i}\tilde{\varphi}),\ \ \psi_y = -\frac{\mathrm{i}}{2}(\Phi_1 + \Phi_2)\exp(\mathrm{i}\tilde{\varphi}), \tag{C7}$$

which describes 1PSF with circular polarization. Such a transformation renders the gauge field $\vec{\nabla}\theta$ irrelevant, and the algebraic part of the OPDM becomes $\sim\langle \exp(i[\tilde{\varphi}(\vec{x}) - \tilde{\varphi}(\vec{x}')])\rangle \sim |\vec{x} - \vec{x}'|^{-1/K}$.

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