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Modelling photon condensation in a fluorescent dye filled optical microcavity

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Abstract. In this paper, a mathematical model for photon condensation in an optical microcavity is considered based on Gross-Pitaevskii equation. The interaction of photons with fluorescent dye molecules in an optical microcavity resembles electron-ion interaction in a periodic lattice. The system is modelled analogous to Kronig-Penney model for electrons in a one-dimensional periodic lattice. The laser used as the light source gives rise to nonlinearity due to the deformation of electron orbits in the fluorescent dye. To accommodate the optical Kerr effect which arises due to this nonlinear interaction, the resultant Schrödinger equation is modified, which assumes the form of Gross-Pitaevskii equation. The model is validated using equation of state, whose virial coefficients can be evaluated from grand canonical ensemble scheme taking into account the interaction between photons and fluorescent dye molecules.

1. Introduction

Mathematical modeling forms an indispensable part of modern scientific analysis. Among the various mathematical techniques used for describing physical systems, differential equations are comprehensive and elegant. In differential equations, nonlinear partial differential equations are particularly useful in analyzing a myriad of nonlinear systems arising from hydrodynamics to fiber optics. Gross-Pitaevskii equation, which we consider here, is a nonlinear partial differential equation used to describe the dynamics of Bose-Einstein condensate of alkali atoms under ultracold temperatures near absolute zero [1]. Recently, Bose-Einstein condensate of photons was successfully realized in an optical microcavity filled with a fluorescent dye [2]. Here, we envisage the usage of Gross-Pitaevskii equation with a different physical setting to describe photon condensation.

The physical system that can demonstrate photon condensate consists of an optical cavity filled with a fluorescent dye as shown in figure 1. The optical cavity has a cavity length of the order of micrometers with highly reflecting dielectric mirrors on either side of the cavity. A laser light having a wavelength of 532 nm is used to excite the rhodamine 6G fluorescent dye inside the cavity. The excited fluorescent dye undergoes Stokes shift to produce radiation emission in yellow region of visible spectrum (580 nm) compared to the excitation wavelength in green region. The presence of cavity can alter the dynamics of radiation inside the cavity. The constrained cavity length supports only one longitudinal mode inside the cavity. Moreover, thermal equilibrium achieved between fluorescent dye and fluorescence emission enables the formation of photon condensate beyond a critical photon number. Here, the critical photon number is implicitly related to pump power of laser (~1.55 W) and type of fluorescent dye that is being used.

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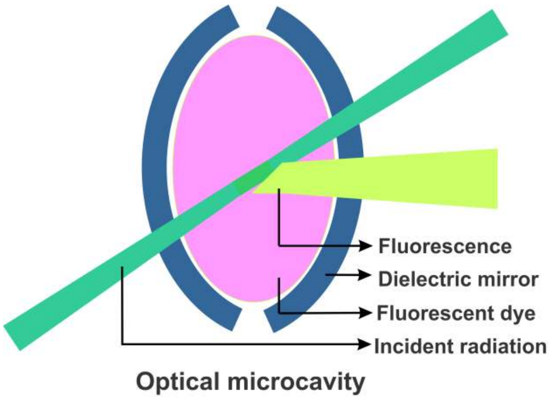

Figure 1. Optical microcavity filled with a fluorescent dye used to realize photon condensate [2].

We try to analyze the system based on the interaction of photons with the dye molecules. To this end, we consider the dye molecules as distributed periodically inside the cavity. This scenario is similar to the conditions which prevail for electrons in a periodic lattice. Here, the electrons are replaced by photons, whereas dye molecules take the role of lattice sites. The interaction between photons and dye molecules also have similar sort of interactions as that between electrons and lattice atoms. Along these lines, Kronig-Penney model for electron propagation through a one-dimensional periodic lattice can be used for describing the system. The intensity of input laser induces nonlinear effects, and the Schrödinger equation used for describing the system is modified to accommodate these effects. By considering such a nonlinear effect, namely the optical Kerr effect, the resulting nonlinear Schrödinger equation turns appropriate to describe the system completely. This equation which has the form of Gross-Pitaevskii equation is used for modeling photon condensation in an optical microcavity. Moreover, grand canonical ensemble which effectively describes the system under consideration is used to validate the theoretical scheme. From this analysis, a theoretical framework is developed to describe photon condensation in an optical microcavity.

2. Photons in a periodic lattice

From solid-state theory, it is well known that the electrons which move through a periodic lattice are susceptible to interaction with the ion cores in the lattice. This interaction between ion cores and electrons gives rise to a periodic variation of electron wavefunction given by Bloch theorem. For a one-dimensional periodic lattice, the solution of Schrödinger equation which describes the system was worked out by Kronig and Penney based on Bloch theorem [3]. This model was found to be useful for interpreting the band gap formation in a periodic lattice. Here, the system under consideration also consists of an array of dye molecules with which the incoming photons interact. Similar to the interaction between electrons and ions cores in the lattice which gives rise to a periodic variation in electron wavefunction, the presence of fluorescent dye molecules causes a periodic variation in photon wavefunction. So, it would be logical to analyse the system based on this approach.

Periodic lattice considered in Kronig-Penney model is shown in figure 2. This can also be extended into the case of photons propagating through a periodic lattice of fluorescent dye molecules. Invoking fermionization applicable in one-dimensional systems, the wavefunction of many-body strongly interacting bosons (photons in this case) can be represented in terms of absolute value of wavefunction of non-interacting fermions (electrons in this case) [4]. An expression for the same is given by,

$$\Psi_B(x_1,\cdots,x_N) = \left|\Psi_F(x_1,\cdots,x_N)\right|, \tag{1}$$

where $\Psi_B(x_1,\cdots,x_N)$ is the wavefunction for bosons, and $\Psi_F(x_1,\cdots,x_N)$ is the wavefunction for fermions. Due to the periodic nature of wavefunction in the lattice, in effect, according to this mapping, the photon wavefunction ends up in the same form as that of electron wavefunction. Hence, according to this mapping, it is permissible to use electron wavefunction in place of photon wavefunction in this scenario.

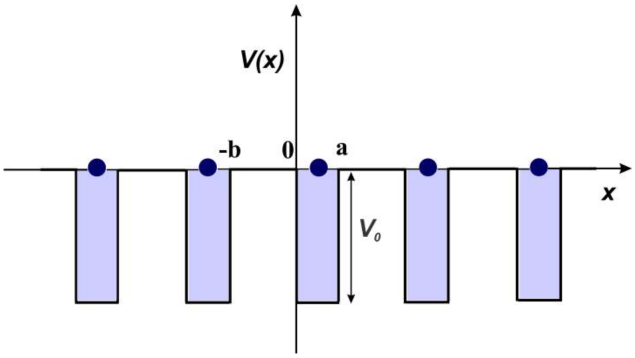

Figure 2. Periodic potential for electrons due to the presence of ion cores in a periodic lattice.

For modelling the system, the fluorescent dye molecules are assumed to be arranged periodically inside the optical microcavity as shown in figure 3(b). The incoming photons are susceptible for variation in its trajectory due to the presence of dye molecules. A graphical illustration of incident photons getting absorbed by the dye molecule, and subsequent emission of fluorescent radiation is depicted in figure 3(a). The overall variation in photon trajectory due to the presence of a one-dimensional array of dye molecules is shown in figure 3(c). Similar to modulation of electron wavefunction by a periodic lattice, the photon wavefunction also gets modulated by the presence of an array of dye molecules.

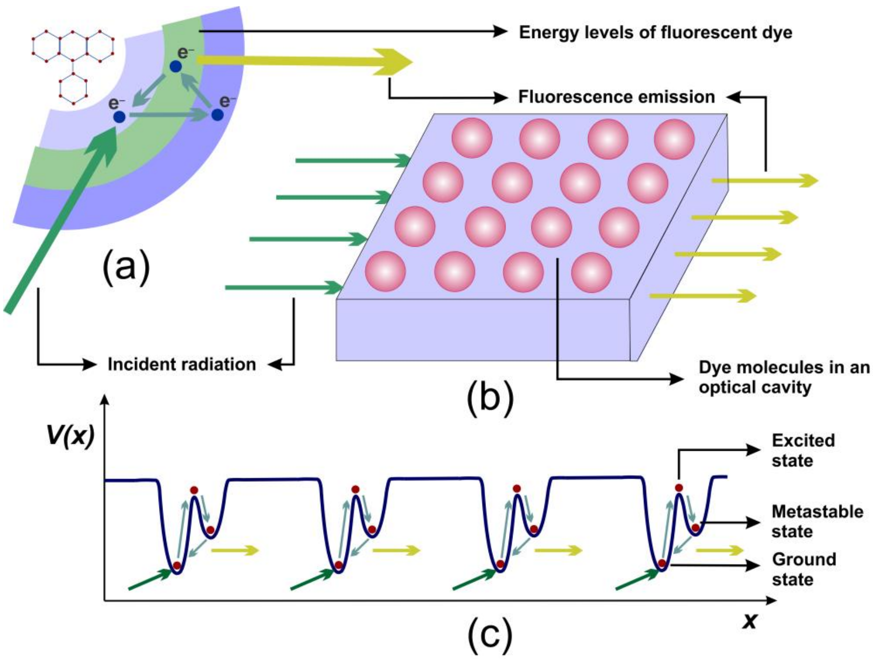

Figure 3. (a) Mechanism of fluorescence from a typical fluorescent dye. (b) An array of fluorescent dye molecules in an optical cavity. (c) Potential experienced by electrons in a one-dimensional array of fluorescent dye molecules which when interact with the incoming photons influence the photon trajectory through the lattice.

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3. Modelling using Gross-Pitaevskii equation

The Schrödinger equation which is used to describe the system is modified due to the presence of a periodic potential. This can be accommodated by incorporating the scheme adopted by Kronig and Penney. Moreover, due to the intensity of input laser light, deformation of electron orbits occurs, turning the system nonlinear [5]. By considering such a nonlinear effect, namely the optical Kerr effect, we try to model the system completely by taking into account of this nonlinear effect which can arise in the system. The variation in refractive index induced by intensity of incident radiation is known as optical Kerr effect. This has an after-effect which is characterized by focusing of the laser beam after passing through the medium, known as self-focusing [6]. Mathematically, nonlinearity arising due to optical Kerr effect is expressed as $\gamma|\psi|^2$, where γ is the nonlinear parameter, and $|\psi|^2$ is modulus square of electric field which corresponds to the intensity of laser [7].

Combining various parameters such as periodic potential and nonlinearity, the resulting Schrödinger equation assumes the form of Gross-Pitaevskii equation, given by,

$$i\hbar\frac{\partial\psi}{\partial t}=\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2}+V(x)+\gamma|\psi|^2\right]\psi. \tag{2}$$

The nonlinearity in this expression can support self-focusing mechanism, and can act as a balance against the spreading of wavefunction due to dispersion. In this way, this effect is a boon for stabilizing the wavefunction.

A possible solution of equation (2) can be obtained when dispersion is balanced by self-focusing nonlinearity, giving rise to a soliton type solution which is characterized by stabilized undistorted pulses preserving their shape in the propagation [8]. For the one-dimensional case considered here, such a solution is given by,

$$\psi(x,t)=\eta\,\mathrm{sech}[\eta(x-vt)]\exp[i(kx-\omega t)], \tag{3}$$

where η is amplitude, k is wavenumber, ω is angular frequency, and v is velocity of the soliton [9]. The dynamics of interaction between photons and fluorescent dye molecules within the system can be further investigated by considering the grand canonical ensemble which replicates the system under consideration.

4. Grand canonical ensemble

Grand canonical ensemble is a scheme proposed by Gibbs for describing an open isothermal system which can exchange both particles and energy between its microstates. The microstates of this ensemble are characterized by invariance in chemical potential, volume, and temperature. A graphical illustration of grand canonical ensemble is shown in figure 4. The system considered here can be described using this ensemble. Here, the interaction between photons and dye molecule is considered as a microstate of the ensemble.

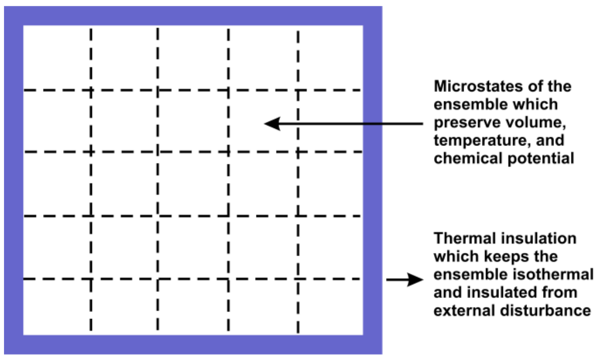

Figure 4. Schematic representation of grand canonical ensemble.

Due to the nonlinear nature of interaction between photons and dye molecules, it is necessary to go beyond the ideal non-interacting particle case. Therefore, it is useful to consider the equation of state based on virial coefficients which takes into account the interacting nature of particles found in real gases. According to this formalism, the equation of state is represented as a series in terms of virial coefficients, given by,

$$\frac{Pv}{RT}=1+\frac{B}{v}+\frac{C}{v^2}+\cdots, \tag{4}$$

where $\frac{Pv}{RT}$ is compressibility factor, v is molar specific volume, B is second virial coefficient, and C is the third virial coefficient [10]. To a good approximation, the terms up to second virial coefficient are sufficient to effectively describe the system under consideration.

Considering the interaction between photons and dye molecules as similar to hard sphere interaction, the expression for second virial coefficient turns out to be $B=\frac{2\pi}{3}N_A\sigma^3$, where N_A is Avogadro's number, and σ is radius of the dye molecule. Substituting this expression for second virial coefficient in equation (4), an expression is obtained for pressure inside the cavity,

$$P=\frac{RT}{v}\left(1+\frac{2\pi}{3}\frac{N_A\sigma^3}{v}\right). \tag{5}$$

Upon substituting the values for various parameters, $R = 8.314$ J K^{-1}mol^{-1}, $T = 300$ K, $v = 1.39\times10^3$ m^3 mol^{-1}, and $\sigma = 0.8$ nm, pressure inside the cavity turns out to be 1.79 N m^{-2}. To confine the condensate formed inside the cavity, an equivalent amount of radiation pressure should be supplied externally from the laser source. Experimentally reported pump power of laser at 1.55 W along with a beam diameter of 35 μm gives a radiation pressure in the same order at 3.46 N m^{-2}. Based on this assurance, the proposed theoretical scheme is validated as a mathematical model for describing the condensation of photons in an optical microcavity.

5. Conclusion

In this paper, we have considered a mathematical model for condensation of photons in an optical microcavity. The photons which propagate through an array of fluorescent dye molecules in an optical microcavity are analogous to electron-ion interaction in a periodic lattice. Photon–dye molecule interaction also follows the periodic Bloch function as in the case of Kronig-Penney model for electrons. For the one-dimensional case considered in modelling the system, the photon wavefunction turns out to be same as that of electron wavefunction due to the fermionization feasible in a one-dimensional system. Moreover, due to the large intensity of laser light that is being used, nonlinearity effects can arise in the system. One such a nonlinear effect, namely the optical Kerr effect, is taken into account for modelling the system. By accommodating this nonlinear effect, the system becomes nonlinear, and is described using a nonlinear partial differential equation.

Taking into account the periodic potential as well as the nonlinear term which arises due to optical Kerr effect, the resulting nonlinear partial differential equation assumes the form of Gross-Pitaevskii equation. A solution for this type of equation is feasible when there is a balance between dispersion and nonlinearity, resulting in soliton pulses which preserve their shape. The interacting nature of the system can also be interpreted using grand canonical ensemble theory. Based on this theory, an expression for pressure inside the cavity can be deduced using the equation of state having virial coefficients. The pressure inside the cavity based on this expression (1.79 Nm^{-2}) was found to be of the same order of radiation pressure from the laser source (3.46 Nm^{-2}). This gives an assurance for the validity of the model. In this way, this model based on periodicity of lattice and nonlinearity can effectively describe the condensation of photons in an optical microcavity.

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