**Evanescent quadrupole polariton**

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In the work we demonstrate the formation of new type of polariton on the interface between a cuprous oxide slab and a polystyrene micro-sphere placed on the slab. The evanescent field of the resonant whispering gallery mode (WGM) has a substantial gradient, and therefore effectively couples with the quadrupole 1S excitons in cuprous oxide. This evanescent polariton has a long life-time (1.7 ns), which is determined only by its excitonic component. The polariton lower branch has a well pronounced minimum. This suggests that this excitation can be utilized for BEC. The spatial coherence of the polariton can be improved by assembling the micro-spheres in a linear chain.

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**INTRODUCTION**

Although quadrupole excitons in cuprous oxide crystals are good candidates for BEC due to their narrow line-width and long life-time there are some factors impeding BEC [1, 2]. One of these factors is that due to small but non negligible coupling to the photon bath, one must consider BEC of the corresponding mixed light-matter states called polaritons [3]. The photon-like part of the polariton has a large group velocity and tends to escape from the crystal. Thus, the temporal coherence of the condensate is effectively broken [4]. One proposed solution to this issue is to place the crystal into a planar micro-cavity [5]. But even state-of-the-art planar micro-cavities can hold the light no longer than 10 µs.

Therefore in this work we propose to impede the polariton escaping by trapping it into a whispering gallery mode (WGM) of a polystyrene micro-sphere (PMS).

**THE MODEL**

We assume that the PMS of radius \( r_0 \) \( \mu \)m is placed at a small [14] distance \( \delta r_0 \ll r_0 \) from the cuprous oxide crystal (\( \epsilon_{Cu2O} = 6.5 \)). Some density of quadrupole 1S excitons (\( [QE] \), \( h\omega_{1S} = 2.05 \) eV, \( \lambda_{1S} = 2\pi/\omega_{1S} = 6096 \) Å) has been created by an external laser pulse. The corresponding polaritons move in the crystal by diffusion and can be trapped at the surface by the micro-sphere.

Because the evanescent field penetration depth \( \lambda_{1S}/2\pi (\epsilon_{Cu2O} - 1)^{1/2} = 414 \) Å is much bigger than the QE radius \( \lambda_{1S} = 4.6 \) Å the light-matter interaction can be considered semi-classically. For resonance coupling with a WGM its size parameter should be determined by the resonant wave vector in the cuprous oxide \( k_0 = 2.62 \times 10^7 m^{-1} \). For example, if one takes a polystyrene \( (\epsilon^2 = 1.59) \) sphere of radius \( r_0 = 10.7 \) \( \mu \)m then \( k_0 r_0 = 28.78350 \) and this corresponds to the 39TE1 resonance [6].

The light part of the polariton trapped inside the PMS moves as it would move in a micro-cavity of the effective modal volume \( V \ll 4\pi r_0^3/3 \). Consequently, it can escape through the evanescent field. This evanescent field essentially has a quantum origin and is due to tunneling through the potential caused by dielectric mismatch on the PMS surface. Therefore, we define the *evanescent* polariton (EP) as an evanescent light-matter coherent superposition.

The evanescent light has small intensity. Therefore it is not effective for the dipole allowed coupling. But it has a large gradient, so it can effectively couple through its quadrupole part.

Let us assume that the incident polariton wave vector is along the interface and runs in the \( z \) direction, the polarization of the polariton is along \( x \) direction. Therefore, in the system of coordinates centered at the sphere, the incident polariton light part can be written as [7]:

\[
E_i = E_0 i^l \frac{2l+1}{l(l+1)} (M_{1l} - iN_{1l}),
\]

where \( M_{1l} \) and \( N_{1l} \) are vector spherical harmonics corresponding to TE- and TM-polarized modes of angular momentum \( l \) and \( z \) component of the angular momentum is \( |n| = 1 \); \( E_0 \) is the amplitude of the electric field. The scattered field is given as:

\[
E_s = E_0 i^l \frac{2l+1}{l(l+1)} (ia_{1l} N_{1l} - b_{1l} M_{1l}),
\]

where \( a_{1l} \) and \( b_{1l} \) are scattering Mie coefficients (See the Appendix). Keeping only the resonant term the last expression yields:

\[
E_s = -E_0 i^l 0.05 b_{1,39} M_{1,39},
\]

While in the system of the coordinate, centered at the cuprous oxide, the plane wave is still given by the expression [4], the scattered field has to be changed according to the vector spherical harmonic addition theorem [8]:

\[
M_{1,39} = A_{1,39}^m (r_0 + \delta r) M_{ml} + B_{1,39}^m (r_0 + \delta r) N_{ml}
\]
Here $A_{1,39}^{ml}$ and $B_{1,39}^{ml}$ are the translational coefficients. Their explicit expression can be found, for instance, in [6, 9] and are explicitly listed in the Appendix.

The bulk (incident) and evanescent polaritons in cuprous oxide are formed through the quadrupole part of the light-matter interaction:

$$H_{int} = \frac{ie}{m\omega_{S}} E_{i,s} \cdot \mathbf{p}$$

Here $e, m$ are the electron mass and charge, and $\mathbf{p}$ is the electron momentum. For the quadrupole 1S transition in cuprous oxide the energy of the interaction can be written as:

$$g = \langle 3\Gamma_{5,1,2}^{++} | H_{int} | \Gamma_{1}^{+} \rangle = \langle 3\Gamma_{5,1,2}^{++} | H_{int} | \Gamma_{1}^{+} \rangle$$

Here we introduced the initial state of the system, which transforms as irreducible representation $1\Gamma_{1}^{+}$ of the cubic centered group $O_h$. The final state is the ortho-exciton state which transforms as $3\Gamma_{5,1,2}^{++}$ in Cartesian system or as $3\Gamma_{5,1,2}$ in the corresponding spherical basis.

Hence, using [1, 2, 3, 4], one can deduce that the coupling of the spherical harmonic compared to the plane wave ($g_{1,2} = 124 \, \mu eV$) is resonantly enhanced:

$$g = \langle 3\Gamma_{5,1,2}^{++} | H_{int} | \Gamma_{1}^{+} \rangle = \langle 3\Gamma_{5,1,2}^{++} | H_{int} | \Gamma_{1}^{+} \rangle$$

where $a_k$, $b_k$ are annihilation operators for light and the exciton, respectively. Therefore, considering that both the exciton and WGM of a single sphere are localized, the dispersion is reduced to:

$$\omega = \omega_{1S} \pm g_{1/2}/\hbar$$

The excitons are trapped in the minimum of the lower branch. Hence the corresponding WGM pattern can be observed. The dispersion above is similar to the quadrupole-dipole hybrid in the organic-inorganic heterostructures [2]. In the later case, the excited organic molecules create an evanescent field penetrating in the cuprous oxide.

The evanescent polariton provided by a single sphere gives the time coherence necessary for the observable BEC of the quadrupole exciton. But the spatial coherence is limited to a small region nearby to the sphere. To improve the spatial coherence one has to sacrifice the temporal coherence slightly by delocalizing the corresponding WGM. It can be done by using an array of spheres aligned along the $z$ direction and separated by the distance $\delta r_0$ (See Fig. 2).

**RESULTS AND DISCUSSION**

Around the resonance between WGM and the quadrupole exciton $\omega_{1S} \approx \omega_{1S}$ the EP branches are given by the eigenvalues of the following Hamiltonian:

$$H/\hbar = \omega_{1S} a_{k}^{\dagger} a_{k} + \omega_{1S} b_{k}^{\dagger} b_{k} + g_{1/2} (a_{k}^{\dagger} b_{k} + a_{k} b_{k}^{\dagger})$$

The coupling grows with mode number $l$, because the gradient of the evanescent field increases. At $l = 89$ the coupling becomes of the order of $meV$. The property of the scalable coupling factor can be utilized in practical applications such as non-linear optics and is the subject of our future work.

![Fig. 1](image1.png)

**FIG. 1:** The evanescent light - 1S quadrupole coupling ($g_{1/2}$) scaled to the bulk exciton-photon coupling ($g_{1,2}$). The size parameter $kr_0$ is denoted as $x$ and the PMS is placed directly on the cuprous oxide sample ($\delta r = 0$, See also Fig. 2).

![Fig. 2](image2.png)

**FIG. 2:** Schematic of formation of the evanescent polariton on linear chain of PMS. Actual dispersion is determined by the ratio of two coupling parameters such as exciton-WGM coupling and WGM-WGM coupling between the microspheres.
Recent experimental [10] and theoretical [11] studies have shown that the WGM can travel along the chain as "heavy photons". Therefore the WGM acquires the spatial dispersion, and the evanescent polariton has the form (See Fig.3):

\[
2\omega = \omega_{1l,k} + \omega_{1S} \pm \sqrt{(\omega_{1l,k} - \omega_{1S})^2 + 4g_{1l}^2/\hbar^2}
\]

\[
\omega_{1l,k} = \omega_{1S} + 2(g_{1l}^H/\hbar) \cos(x - x_1 + \pi/2)
\]

(8)

Here \(g_{1l}^H = \omega_{1S}b_{1l}A_{1l}^H(\delta r_1)\) is the nearest-neighbor inter-sphere coupling parameter.

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**APPENDIX**

In the appendix we list explicit expression for the Mie scattering coefficient:

\[
a_{ml} = \frac{n^2 j_{ml}(nx) [xj_{ml}(x)]' - j_{ml}(x) [nxj_{ml}(nx)]'}{n^2 j_{ml}(nx) [xh_{ml}^{(1)}(x)]' - h_{ml}^{(1)}(x) [nxj_{ml}(nx)]'}
\]

\[
b_{ml} = \frac{j_{ml}(nx) [xj_{ml}(x)]' - n^2 j_{ml}(nx) [nxj_{ml}(nx)]'}{j_{ml}(nx) [xh_{ml}^{(1)}(x)]' - n^2 h_{ml}^{(1)}(nx) [nxj_{ml}(nx)]'}
\]

Here \(n = \epsilon^2\) is the refractive index of the spheres; \(x = kr_0\) is the size parameter; \(j_{ml}, h_{ml}\) are the spherical Bessel and Hankel of the first kind functions respectively.

In the case of \(l \gg 1\) the calculation of the translational coefficients can be significantly simplified with the help of the so-called maximum term approximation [2].

\[
A_{l}^{l'} \approx -2l(-1)^{l'+1} \sqrt{\frac{l+l'}{\pi(l'+1)(l-1)}} \times
\]

\[
\frac{l^{l'}[(l')^{l'}]}{[(l'+1)^{l'+1} - (l-1)^{l-1}]} = h_{l+l'}^{(1)}(nx)
\]

\[
B_{l}^{l'} \approx \frac{x|x-j|}{l^{l'}} A_{l}^{l'}
\]

Here \(\eta\) defined as \(\eta = |r_0 + \delta r|/r_0 \geq 1\) is a dimensionless distance between the centers of the spheres.

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