Strong coupling of collection of emitters on hyperbolic meta-material

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Abstract

Recently, considerable effort has been devoted to the realization of a strong coupling regime of the radiation matter interaction in the context of an emitter at a meta surface. The strong interaction is well realized in cavity quantum electrodynamics, which also show that strong coupling is much easier to realize using a collection of emitters. Keeping this in mind, we study the radiation matter interaction in the context of an emitter at a meta surface. The strong coupling is much to realize using a collection of emitters. Keeping this in mind, we study the interaction is well realized in cavity quantum electrodynamics, which also show that strong coupling regime. The usefulness of collective excitations has been demonstrated for a very wide variety of systems: superconducting qubits [13]; magnons [14, 15]; cyclotron motion of surface electrons [16]; graphene ribbons [17]; porphyrin excitons [18]; J-aggregates [19–24], dye molecules [25, 26], quantum dots [27, 28] and nanoparticles [29]. Recently also, strong coupling for monolayer transition dichalcogenides in a dielectric cavity [30] and in a plasmonic cavity [31] has been realized.

In the last decade, hyperbolic materials [32, 33] have shown to be a versatile platform, allowing for broadband enhanced spontaneous emission [6–11], hyperbolic lensing [34–38], negative refraction [39, 40], broadband enhanced thermal emission [41–46], near-field thermophotovoltaics [47, 48], long-range heat and energy transfer [49–51], and thermal and nonthermal nanoantennas [52, 53], as well as strong coupling of emitters embedded in such materials [57]. Reviews dealing with wave propagation in hyperbolic metamaterials and applications are given in [58–60]. In this work, we investigate if hyperbolic metamaterial (HMM) could provide a good platform for realizing the strong coupling regime using the collective excitations of emitters. We show that strong coupling can be realized for densities of emitters exceeding a critical value. A way to detect strong coupling between emitters and hyperbolic metamaterials is to use the Kretschmann-Raether configuration. The strong coupling appears as the splitting of the reflectivity dip. In the weak coupling regime, the dip position shifts. The shift and splitting can be used to sense active molecules at surfaces.

Keywords: hyperbolic metamaterial, rabi splitting, kretchmann configuration

(Some figures may appear in colour only in the online journal)
how the Kretchmann-Raether [25, 26] configuration can be used for investigations on strong coupling on HMM.

Our work is organized as follows: in section 2, we show what prevents the observation of strong coupling for a single emitter and what is required for seeing the effects of strong coupling. In section 3, we introduce the Kretchmann-Raether configuration, the hyperbolic material used in that configuration and the material properties of the dye molecules. The Kretchmann-Raether configuration is the simplest way in which the features of the strong coupling can be explored. Finally, in section 4, we discuss the transition from the weak to the strong-coupling regime by studying the reflectivity in the Kretchmann-Raether setup as a function of the concentration of the dye molecules. In the weak coupling regime, the dip of the reflectivity shifts, whereas in the strong coupling regime, the reflectivity dip splits in two. These features can be used as sensors of molecules on surfaces [54, 55]. We summarize our main findings in section 5.

### 2. Rabi splitting for a single atom

First, let us dwell on the possibility to detect a single atom by means of Rabi splitting. To this end, we consider a single atom at position \( r_A \) close to the surface of a substrate. Here, we describe the atom by a classical dipole, because, for Rabi splitting, it is not essential to make a quantum approach. A nice comparison of the classical and quantum approach can be found in [5]. In the classical case, the dipole moment can be described as a harmonic oscillator obeying the standard equation of motion

\[
\left( \frac{d^2}{dr^2} + \omega_0^2 \right) p = \frac{e^2}{m} (E_{\text{ext}} + E_{\text{dip}})
\]

where \( p = e \mathbf{x} \), \( E_{\text{ext}} \) is the external field exciting the dipole moment of the atom, \( E_{\text{dip}} \) is the field generated by the dipole moment and \( \omega_0 \) is the atomic transition frequency. In Fourier space, it can be written as

\[
E_{\text{dip}} = \frac{\omega_0^2}{c^2 e_0} (G^{(\text{vac})}(\mathbf{r}_A, \mathbf{r}_A; \omega) + G^{(\text{sc})}(\mathbf{r}_A, \mathbf{r}_A; \omega)) \cdot \mathbf{p}.
\]

Introducing the vacuum and the scattering parts \( G^{(\text{vac})} \) and \( G^{(\text{sc})} \) of the dyadic Green function evaluated at the position of the atom. By going into Fourier space and inserting Green’s function, the equation of motion reads

\[
\hat{A} \cdot p = \frac{e^2}{m} E_{\text{ext}}
\]

with

\[
\hat{A} := (\omega_0^2 - \omega^2) \mathbb{I} - \frac{\omega_0^2 e^2}{c^2 e_0 m} \text{Im} G^{(\text{vac})}(\mathbf{r}_A, \mathbf{r}_A; \omega)
\]

\[
- \frac{\omega_0^2 e^2}{c^2 e_0 m} G^{(\text{sc})}(\mathbf{r}_A, \mathbf{r}_A; \omega).
\]

Here, we have neglected the divergent real part of the vacuum Green’s function. This corresponds to a renormalization of mass and frequency of the harmonic oscillator.

In order to determine the eigen frequencies of the coupled field-atom system, we need to solve equation (3). But it suffices to focus on the ‘zeroes’ of \( \hat{A} \) in order to find the eigen frequencies. If we focus, for example, on the \( zz \) component of \( \hat{A} \), only, then

\[
A_{zz} := (\omega_0^2 - \omega^2) I - \frac{\omega_0^2 e^2}{c^2 e_0 m} \text{Im} G^{(\text{vac})}(r_A, r_A; \omega) - \frac{\omega_0^2 e^2}{c^2 e_0 m} G^{(\text{sc})}(r_A, r_A; \omega).
\]

Here,

\[
\nu^{(\text{vac})} := \frac{\omega_0^2 e^2}{c^2 e_0 m} \text{Im} G^{(\text{vac})}(r_A, r_A; \omega)
\]

is essentially the emission rate of the atom in vacuum. The coupling constant of the atom-surface system can now be determined by assuming that the substrate itself has a surface mode resonance. Around this resonance, we can approximate the \( zz \) component of the scattered part of Green’s function as

\[
G^{(\text{sc})}(\omega) \approx \frac{-\chi}{\omega - \omega_c + i \kappa_c}.
\]

Here, \( \chi \) is the oscillator strength and \( \kappa_c \) is the damping constant of that surface resonance. Assuming further that the resonance frequency and that the excitation frequency of the atom are close to resonance, i.e. \( \omega_0 \approx \omega_c \) and \( \omega \approx \omega_0 \), we can determine the zeroes of \( A_{zz} \) easily. For the eigen frequencies of the coupled system, we then obtain

\[
\omega = \omega_0 - \frac{1}{2} \frac{\nu^{(\text{vac})} + \kappa_c}{2} \pm \sqrt{\frac{1}{4} - \frac{\nu^{(\text{vac})} - \kappa_c^2}{4}}
\]

where we have introduced the coupling constant

\[
g = \frac{e^2 \omega_0}{2mc^2 e_0} \chi.
\]

Obviously, we have a damping or energy loss due to the spontaneous emission into vacuum \( \nu^{(\text{vac})} \) and due to the losses in the surfaces wave. In the weak-coupling regime, i.e. for \( g^2 \ll (\nu^{(\text{vac})} - \kappa_c^2)/4 \), the eigen frequencies of the coupled system coincide with \( \omega_0 \). On the other hand, in the strong coupling regime when \( g \gg \kappa_c/2 \) and \( g \gg \nu^{(\text{vac})}/2 \), the degeneracy of the eigen frequencies of the coupled system is lifted and one can observe two distinct eigen frequencies which differ from \( \omega_0 \). Therefore, in order to observe this Rabi splitting, it is necessary to enter the strong coupling regime. If the atom is placed in the near-field of the surface mode resonance, then the condition \( g \gg \nu^{(\text{vac})}/2 \) is automatically fulfilled due to the Purcell effect. On the other hand, the condition

\[
\frac{2g}{\kappa_c} \gg 1
\]

is much harder to fulfill, because the damping of the surface waves becomes large close to the surface mode resonance. We have checked by numerical calculations that indeed this condition is hard to meet so that Rabi splitting for a single atom seems to be quite difficult, although for the coupling of single atoms to metal nanoparticles, a strong-coupling is theoretically possible [56]. However, it is known that the Rabi splitting scales like \( \sqrt{N} \) where \( N \) is the number of atoms [12].
Therefore, by increasing the number of atoms, Rabi splitting at some point becomes measurable.

In view of this, it is better to consider a dense medium whose linear response to electromagnetic fields can be described by the dielectric function

$$\epsilon(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\omega\gamma}. \quad (11)$$

We further assume that such a medium is in a plasmonic cavity with losses described by $\kappa_c$. The cavity mode is tuned such that $\omega_c \approx \omega_p$. Then, one finds in the strong coupling regime with $\omega_c \gg \gamma$, $\kappa_c$, the simple relation

$$\omega \approx \omega_c \pm \frac{\omega_p}{2} \quad (12)$$

for the new coupled mode frequencies if $\omega_p \ll \omega_c$ and $\omega_p \gg \gamma$, $\kappa_c$. First, we note that the Rabi splitting is proportional to $\omega_p$, and therefore proportional to $\sqrt{N}$ as expected [5, 12]. To see the Rabi splitting, the density of atoms described by $\omega_p$ has to be larger than the losses of the plasmonic cavity $\kappa_c$. This is in accordance with our previous result in (10) and makes it clear that, due to the losses of the metal, a large density of atoms is necessary to see Rabi splitting. Note that this gives an estimate of the new normal modes. Actual values are to be obtained from solutions of full Maxwell equations for the anisotropic medium.

3. Kretschmann-Raether setup

In the following, we will show that by using hyperbolic metamaterials we can use Rabi splitting for the sensing of very small concentrations of atoms or molecules. To this end, we consider the Kretschmann configuration as depicted in figure 1. The prism is assumed to be made of silica with refractive index $n = 1.5$. The plasmonic film is assumed to be a hyperbolic material of thickness $d = 50$ nm, which is realized by a multilayer structure of alternating layers of silver and TiO$_2$ using the same material parameters as in [50]. The effective medium response of such a multilayer material is that of a uni-axial medium with permittivity

$$\epsilon = \begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{||} \end{pmatrix}, \quad (13)$$

where $\epsilon_{\perp}$ is the permittivity along the optical axis which is parallel to the surface normal and $\epsilon_{||}$ is the permittivity perpendicular to the optical axis. In figure 2, the real values of both permittivities $\epsilon_{\perp}$ and $\epsilon_{||}$ are shown for a volume filling fraction of $f = 0.6$ of silver. The two hyperbolic frequency bands of type I ($\epsilon_{||} < 0$ and $\epsilon_{\perp} > 0$) and of type II ($\epsilon_{||} > 0$ and $\epsilon_{\perp} < 0$) can be nicely seen. The edges of the two bands are determined by the epsilon-near-zero (ENZ) $\lambda_{\text{ENZ}} = 414$ nm and the epsilon-near-pole (ENP) $\lambda_{\text{ENP}} = 513$ nm wavelengths. It could already be shown that large dipole-dipole interactions are achievable close to these wavelengths [50, 51]. One can expect to have a large Rabi splitting close to ENP and ENZ wavelengths as well.

To test this hypothesis, we consider a thick layer of dye molecules of rhodamin 6 (R6G) in ethylene glycol solution on a hyperbolic medium as the substrate, as depicted in figure 1. The resonance wavelength of the R6G molecules is around 532 nm, which closely matches the resonance ENP wavelength. Hence, we can expect to have a very effective coupling and Rabi splitting in this case. In order to determine the reflectivity in the Kretschmann configuration in figure 1, we use for the permittivity of R6G the model [61] which is given by equation (11) with

$$\omega_p^2 = \frac{C n_e^2}{m \epsilon_0} \quad (14)$$

with $\omega_0 = 3.5 \times 10^{15}$ rad/s, $\gamma = 2.07 \times 10^{14}$ rad/s, $h = 0.74$. The concentration $C$ of rhodamin dye molecules is given in units of $M = \text{mol/l} = 1000 \text{ mol/m}^3$. Therefore, a concentration of 0.002M corresponds to 1.2 $\times 10^{24}$ m$^{-3}$
and 0.1 M corresponds to 60 molecules in a volume of \((10 \text{ nm})^3\) or a number density of \(6 \times 10^{25} \text{ m}^{-3}\).

4. Transition from weak to strong-coupling regime

The reflectivity of p-polarized light \([62]\) for an angle of incidence of \(\theta = 48^\circ\) is shown in figure 3 for different concentrations of R6G using the effective medium theory for the hyperbolic material. It can be seen that for very low concentrations in the range of \(0 M-0.01 M\) (\(-\Delta \omega = 6 \times 10^{24} \text{ m}^{-3}\)), the minimum in the reflectivity shifts from 0.2 to 0.04, which is easily measurable. Note that there is a small blue shift. We find that for smaller angles like \(\theta = 47.5^\circ\), we have larger blue-shift, whereas for larger angles like \(\theta = 49^\circ\), we find a red-shift. Therefore, the reflection dip has a tendency to shift towards \(\omega_0\).

For large concentrations of 0.02M-0.1M \((1.2 \times 10^{25} \text{ m}^{-3} - 6 \times 10^{25} \text{ m}^{-3})\), the Rabi splitting sets in. Again, by measuring only the shift of the small wavelength minimum, one can easily measure the concentration of the dye molecules. Note that the separation between the two dips at 498 nm \((2.49 \text{ eV})\) and 586 nm \((2.12 \text{ eV})\) for a high concentration of 0.1M \((6 \times 10^{25} \text{ m}^{-3})\) is \(\Delta \lambda = 88 \text{ nm}\), which corresponds to 370 meV. Let us compare this value with the estimate given in equation \((12)\). For the R6G molecule at the highest concentration, we have \(\omega_p = 3.76 \times 10^{14} \text{ rad/s} \ll \omega_0 = 3.5 \times 10^{15} \text{ rad/s}\) and \(\omega_p > \gamma = 2.1 \times 10^{13} \text{ rad/s}\). Therefore, from the relation \((12)\), a Rabi splitting of about 247 meV can be expected. This is in agreement with the experimentally found values of 100–200 meV observed in \([25]\). The here observed value of 370 meV with the hyperbolic film is larger than the estimate for an ideal perfect cavity, which is due to the detuning between the ENZ frequency, which corresponds to \(\omega_0\) and the resonance frequency \(\omega_0\) of the dye molecule and the anisotropic character of the hyperbolic medium.

An angle dependent plot of the reflectivity in p-polarization for this configuration is shown in figure 4, in the presence of 0.1M \((6 \times 10^{25} \text{ m}^{-3})\) of R6G, compared to the case where no dye molecules are present in figure 4. The Rabi strong coupling between the dye molecules and the resonance of the hyperbolic metamaterial can be nicely seen. Obviously, the Rabi splitting can be found in a range of angles of incidence of about 45° to 55°, so that the effect is relatively robust with respect to variations in the angle of incidence.

Figure 3. Reflectivity of p-polarized light in the Kretschmann configuration for different concentrations \(C\) of R6G molecules.

Figure 4. Reflectivity of p-polarized light in the Kretschmann for different angles of incidence and wavelengths for a configuration of \(C = 0 M\) or \(n = 0 \times 10^{27} \text{ m}^{-3}\) (top) and \(C = 0.1 M\) or \(n = 6 \times 10^{25} \text{ m}^{-3}\) (bottom) of R6G molecules. The vertical dashed black line is positioned at the resonance wavelength of R6G molecule at 532 nm. The vertical solid white are positioned at the ENZ and ENP wavelength.
5. Conclusion

In summary, we have shown that by using hyperbolic metamaterials in a Kretschmann-Raether configuration, one can measure even small concentrations of R6G molecules of only a few mM. For large concentrations, one can easily observe the transition from the weak coupling to the strong coupling regime. The Rabi splitting is directly observable in the reflectivity signal. Therefore, hyperbolic materials are also interesting platforms for Rabi splitting measurements, as well as sensing applications. Finally, hyperbolic materials can also serve as a platform for studying strong coupling phenomena in J-aggregates and two-dimensional transition metal dichalcogenides like MoS2, which are promising candidates for room temperature excitonic devices in opto-electronics, photovoltaics and ultra-fast photodetection [63].

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