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Temporal dynamics of a quantum emitter with multiple excited states in the vicinity of an anisotropic metasurface

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Abstract. In this work we focus on studying the temporal dynamics of a quantum emitter with few (3) excited states, which allows to observe an interplay between different spontaneous emission channels. We show that if the quantization axis is being rotated with respect to the normal of a metasurface it is possible to observe a difference in the transfer dynamics from one state to another and vice versa.

1. Introduction

The topic of anisotropic vacuum has been attracting a lot of attention since the first time it was discussed in [1] by G. Agarwal. The author proposed an idea that one can exploit the interference between different emission channels of a quantum emitter due to the anisotropy of the surrounding. An interesting manifestation of this effect is that it is possible to suppress the total emission rate due to destructive interference between the decay channels, which was studied in [2]. In this paper we want to study further the topic of coupling the orthogonal transitions of a single quantum emitter through the field modes of a metasurface.

2. Theoretical Framework and results

In this work we investigate the time evolution of a four level inverse tripod-type atom with a single ground state $|g\rangle$ and three excited states $|e_{-1}\rangle$, $|e_0\rangle$, $|e_{+1}\rangle$ with the corresponding transition dipole moments \mathbf{d}_{-1} , \mathbf{d}_0 , and \mathbf{d}_{+1} as shown in Fig. 1, (a). These three dipole moment vectors form an orthonormal set in \mathbb{R}^3 , which leads to the fact that the three corresponding excited states are decoupled in the isotropic environment. However, they might interact with each other via the modes supported by a photonic or plasmonic structure. From this perspective a metasurface presents an interesting system to consider, since it can be anisotropic in the interface plane. The optical properties of it can be characterized by the tensor of the effective surface conductivity σ , which can be chosen to be diagonal in some coordinate system. To describe the optical properties of a metasurface we use the effective conductivity described by [3]:



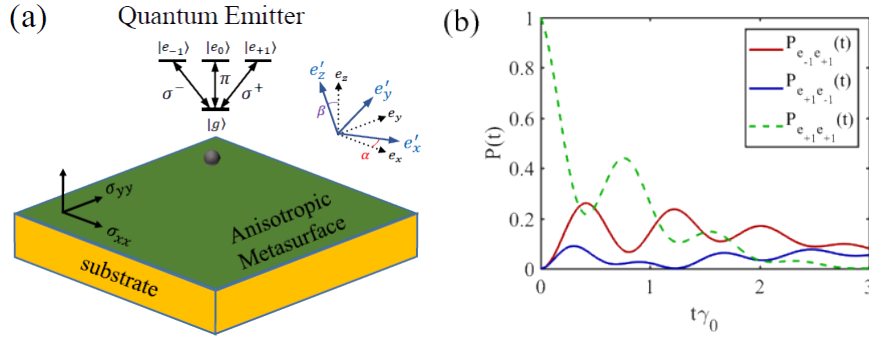


Figure 1. a) Schematic picture of the system: an atom with 3 excited states in the vicinity of an anisotropic metasurface. The dotted arrows represent the lab coordinate system, the blue solid arrows - the rotated on 2 Euler angles. b) Excitation transfer from e_1 to e_2 and vice versa along with the initially excited state population. The parameters are: $A_{xx} = A_{yy} = 1$, $\Omega_{xx} = 1$, $\Omega_{yy} = 3$, $\gamma_{xx} = \gamma_{yy} = 0.01$, $k_0 = 0.65\Omega_{xx}/c$, the rotation angles were $\alpha = \beta = \pi/4$, $\gamma = 0$. The substrate was considered as an infinite dielectric half-space with $\epsilon_2 = 4$. The atom-surface distance is $\Delta z = 0.05 c/\Omega_{xx}$.

$$\sigma = \begin{pmatrix} \sigma_{xx} & 0 \\ 0 & \sigma_{yy} \end{pmatrix}, \quad \sigma_{jj} = A_{jj} \frac{ic}{4\pi} \frac{\omega}{\omega^2 - \Omega_{jj}^2 + i\gamma_{jj}\omega}, \quad (1)$$

where A_{jj} is the normalization constant, Ω_{jj} is the resonance frequency, γ_{jj} is the bandwidth of the corresponding resonance.

In order to solve for temporal dynamics of the emitter's states we employ the approach of the resolvent operator [4, 5], when it is possible to find the matrix elements of the evolution operator $\hat{U}(t, 0)$ using:

$$\langle e_f | \hat{U}(t, 0) | e_i \rangle = \int_C \frac{dz}{2\pi i} e^{-izt/\hbar} \langle e_f | \hat{G}(z) | e_i \rangle, \quad (2)$$

where $\hat{G}(z) = (z - \hat{H})^{-1}$ is the resolvent operator of the full Hamiltonian \hat{H} . Using this relation we can find probabilities for the atom to be in the excited state $|e_f\rangle$ at time t given that it was in the state $|e_i\rangle$ initially by simply finding $P_{ef}(t) = |U_{efei}(t, 0)|^2$.

We should note that here we work in the single excitation domain since we are studying the excited states evolution of a single atom. In this single excitation subspace of a Hilbert space the resolvent can be found to be:

$$\langle e_f | \hat{G}(z) | e_i \rangle = \langle e_f | \left[z - \hat{H}_0 - \hat{\Sigma}(z) \right]^{-1} | e_i \rangle \approx [(z - \hbar\omega_0) \delta_{efei} + 4\pi k_0^2 \mathbf{d}_{ef}^* \mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega_0) \mathbf{d}_{ei}]^{-1}, \quad (3)$$

here \hat{H}_0 is the unperturbed Hamiltonian and $\Sigma(z)$ is the level shift operator, δ_{ij} is the Kronecker delta. The level-shift operator in the latter expression is written within two approximations: near-resonant interaction of the states ($z = \hbar\omega_0$) and taking into account only processes of the second order [6], leading to a form of a classical dipole-dipole coupling constants: $\Sigma_{efei} = -4\pi k_0^2 \mathbf{d}_{ef}^* \mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega_0) \mathbf{d}_{ei}$. Therefore, in order to describe the interaction of the emitter with the electromagnetic modes of the field we only need to construct the Green's function of a metasurface, which can be done by following the procedure outlined in [7].

Now we want to rotate the transition dipole moments \mathbf{d}_q with respect to the original coordinate system, in which σ tensor is diagonal. The rotation of dipole moments will alter the way excited states couple with each other, therefore, altering the time dynamics. Formally this can be done by first considering the level-shift operator matrix in the Cartesian coordinate system $\Sigma_{Cart}(\hbar\omega_0)$, which expresses the couplings of linear dipoles d_j , $j = x, y, z$. Then we can use the transformation matrix S , which transforms the Σ from the Cartesian coordinate system into the spherical tensor basis \mathbf{d}_q , $q = +1, 0, -1$. Finally, we can simply rotate the spherical tensors written in Cartesian coordinates with rotation matrix $M(\alpha, \beta, \gamma)$ (active representation, right-hand rule, $z'' - y' - z$ convention), where α, β, γ are the Euler angles:

$$\begin{aligned}\Sigma'_{Sph} &= T^{-1}(\alpha, \beta, \gamma) \Sigma_{Cart} T(\alpha, \beta, \gamma), \\ T(\alpha, \beta, \gamma) &= M(\alpha, \beta, \gamma) S.\end{aligned}\quad (4)$$

After performing this, we can express the evolution operator matrix elements (2) through the matrix elements of $T(\alpha, \beta, \gamma)$, and the diagonal entries of Σ_{Cart} as:

$$U_{kl}(t, 0) = \sum_{j=1}^3 T_{j,k}^* T_{j,l} e^{-i\Sigma_{jj}t/\hbar} = \sum_{j=1}^3 C_j^{(kl)} e^{-i\Sigma_{jj}t/\hbar}, \quad (5)$$

Note that for Σ_{Cart} index j labels the Cartesian coordinates as $1 - x, 2 - y, 3 - z$, and for the case of spherical basis - $1 - (-1), 2 - (0), 3 - (+1)$.

Let us focus on the case, when we study how excitation is transferred between the two excited states: $|\mathbf{e}_{+1}\rangle \rightarrow |\mathbf{e}_{-1}\rangle$ and the inverse process $|\mathbf{e}_{-1}\rangle \rightarrow |\mathbf{e}_{+1}\rangle$. For them we can write explicitly the corresponding coefficients before the exponents:

$$\begin{aligned}C_1^{(13)} &= -\frac{e^{2i\gamma}}{2} (\cos(\alpha)\cos(\beta) - i\sin(\alpha))^2, \\ C_2^{(13)} &= \frac{e^{2i\gamma}}{2} (\cos(\alpha) - i\cos(\beta)\sin(\alpha))^2, \\ C_3^{(13)} &= -\frac{e^{2i\gamma}}{2} \sin^2(\beta),\end{aligned}\quad (6)$$

and for the inverse process we have $C_j^{(31)} = (C_j^{(13)})^*$. One can notice from (6) that the Euler angle γ enters as an overall phase factor, therefore, the probabilities given by $|U_{kl}(t, 0)|^2$ are independent of it. This is natural since γ represents the rotation around the new quantization axis \mathbf{e}'_z and such a rotation should not change the behavior of the system. It is also important to note that in a general situation, for arbitrary α and β , the coefficients $C_k^{(ij)}$ are complex. It means that when finding the probabilities $|U_{kl}(t, 0)|^2$, the $e^{-i\Sigma_{jj}t/\hbar}$ terms will interfere in such a way that the oscillating parts of the dynamics for $|\mathbf{e}_{+1}\rangle \rightarrow |\mathbf{e}_{-1}\rangle$ and $|\mathbf{e}_{-1}\rangle \rightarrow |\mathbf{e}_{+1}\rangle$ will have different initial phases as can be seen in Fig. 1, (b). Indeed,

$$P_{e_{-1}e_{+1}}(t) = \sum_j \xi_{e_{-1}e_{+1}}^{(jj)} e^{-\gamma_{jj}t} + \sum_{i < j} 2\xi_{e_{-1}e_{+1}}^{(ij)} \cos\left([\Delta_{ii} - \Delta_{jj}]t - \phi_{e_{-1}e_{+1}}^{(ij)}\right) e^{-(\gamma_{ii} + \gamma_{jj})t/2}, \quad (7)$$

where $\Delta_{ii} = \text{Re}[\Sigma_{ii}]$, $\gamma_{ii} = -2\text{Im}[\Sigma_{ii}]$, $\xi_{e_{-1}e_{+1}}^{(ij)}$ are some real-valued constants, $\phi_{e_{-1}e_{+1}}^{(ij)}$ are the initial phases of oscillations. Notice that both $\xi_{e_{-1}e_{+1}}^{(ij)}$ and $\phi_{e_{-1}e_{+1}}^{(ij)}$ depend only upon the Euler

angles and have the following properties: $\xi_{e_{-1}e_{+1}}^{(ij)} = \xi_{e_{+1}e_{-1}}^{(ij)}$, but $\phi_{e_{-1}e_{+1}}^{(ij)} = -\phi_{e_{+1}e_{-1}}^{(ij)}$. The latter leads to a *phaseshift* in the oscillatory dynamics for the probabilities of the two processes under study. Also notice from (7) that in order to observe this effect the anisotropy of the structure is needed as if $\Delta_{ii} = \Delta_{jj}$, then the contributions from the phaseshifts can be simply absorbed in the definition of the constant $\xi_{e_1e_3}^{(ij)}$ and there will be no difference in the dynamics.

3. Conclusion

In our work we studied the dynamical behavior of an inverse-tripod type quantum emitter with three excited states interacting through the modes of an anisotropic metasurface. We demonstrated that if the quantization axis has an arbitrary orientation then it is possible to observe a non-reciprocity, which manifests itself in a phase delay in the oscillating part of the dynamics arising from the anisotropic properties of the structure.

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