Entanglement of bright and dark modes in PT-symmetric plasmonic nano-antennas

Revised 10/11/2019 14:44:00

Vasily Klimov^{1,2,#} and Anatole Lupu³

¹ *P.N. Lebedev Physical Institute, Russian Academy of Sciences,
53 Leninsky Prospekt, Moscow 119991, Russia*

² *National Research Nuclear University,
31 Kashirskoe shosse, Moscow 115409, Russia*

[#]*e-mail address: klimov256@gmail.com*

³ *Centre de Nanosciences et de Nanotechnologies, CNRS, Univ. Paris-Sud, Université
Paris-Saclay,*

C2N – 10 Boulevard Thomas Gobert – 91120 Palaiseau cedex, France

Abstract: The generic model of two coupled multimode plasmonic nanospheres with gain and loss forming a Parity-Time symmetric dimer is considered. Analytical solutions based on a quasi-static approximation are used to explore the behavior of the eigenmodes in such a coupled nano-antenna system as a function of their separation distances and their gain-loss level. It is shown that the introduction of the same amount of gain and loss modifies the composition of eigenmodes and breaks their orthogonality. The resulting entanglement of different spatial order and symmetry modes can be advantageously exploited to actively control the scattering properties of such multimode PT-symmetric nano-antennas.

Keywords: PT symmetry; loss compensation; dimer; eigenmodes; plasmonics; entanglement, nano-antenna, dark modes, bright modes

PACS: 95.55.Sh, 93.90.+y, 13.15.+g

I. INTRODUCTION

The advent of metamaterials in recent decades with a plethora of fascinating effects and applications has also sparked renewed interest in the physics of sub-wavelength resonant systems. In many cases, the interest of such resonant systems is motivated by the ability to obtain sharp characteristics in their spectral response. High Q resonances are highly desirable in a wide range of applications, such as RFID in wireless microwave technology [1] or biochemical sensing in THz domains [2] and optics [3, 4]. The reduction of radiation and absorption losses is the key to high Q resonance. However, achieving this goal becomes particularly challenging when considering operating in a near-IR or visible domain where material-related absorption losses are dominant. The conventional approach to solve this problem is to compensate losses by gain using certain active medium materials such as, for example, organic dyes [5] or semiconductor heterostructures [6]. The paradigm shift came with the introduction of the concept of Parity-Time (PT) symmetry in optics [7], which goes far beyond the mere idea of loss compensation. Here, the losses could rather be considered as an asset and have the same importance as the gain. The combination and modulation of gain and losses can lead to asymmetric transport [8], unidirectional invisibility [9], loss-induced suppression and revival of lasing [10], lasing coherent absorption in the same cavity [11], to mention only a few. The aim of the present work is to provide further impetus to this direction by considering the functionalities that can be obtained in a multimodal PT-symmetric coupled plasmonic nanoresonator system. On the basis of the generic model of two coupled plasmonic nanospheres, it is shown analytically and numerically that the introduction of a PT-symmetric gain-loss in such a system modifies the composition of eigenmodes and breaks their orthogonality.

The use of this concept in plasmonic has several promising outcomes. Having real eigenfrequencies in the Parity-Time symmetric phase means the possibility of considerably increasing the resonance quality factor, a feature highly desirable for sensing applications. Furthermore, this sensitivity can be greatly enhanced by taking advantage of the abrupt evolution of eigenfrequencies in the vicinity of the exceptional point [12-14]. The position of the operating point in the 2D parametric space, which can be adjusted by varying the resonance frequency of the resonators using heating elements [15-17], as well as the PT-symmetric loss-gain level, offers the ability to manipulate the trajectory on Riemann sheets in a growing number of topological photonics applications.

Moreover, even in the simplest case of PT-symmetric dimer formed of coupled single-mode resonators, the possibility of modifying the splitting of the eigenfrequencies by the gain-loss variation involves the modification of the spectral response of the scattered wave. This is an extremely useful feature that can be used for implementation in adjustable metasurfaces. The substantial difference from conventional approaches based on the variation of the resonators or its dielectric environment refractive index is that the amount of gain-loss to be supplied in the system depends of the coupling strength between the resonators. This means that the PT-symmetric gain-loss can be adjusted to the required level by judiciously choosing the separation distance between the coupled resonators.

An additional degree of freedom in eigenfrequency control can be achieved by considering PT-symmetric multimode dimers where the variation of the gain-loss level can be used to manipulate the modal composition of the dimer. It is namely this configuration encompassing all the properties detailed above that is considered in the present work.

The article is organized as follows. In Section II, the quasi-static dipole approximation employed to capture most of the physical image is used to find the electronic modes of the PT-symmetric dimer formed of two monomode coupled nano-antennas.

In section III the quasi-static approach is extended to the derivation of the eigenmodes in a PT-symmetric dimer composed of multimode nanospheres. This technique is then applied to section IV to study the evolution of the eigenmodes as a function of the hybridization between different order modes.

II. DIPOLE APPROXIMATION FOR PT-SYMMETRIC DIMER OF COUPLED SINGLE-MODE NANO-ANTENNAS

The sketch of the considered PT-symmetric nano-antennas dimer is shown in Fig. 1. It consists of two nanospheres of identical size, where the permittivity of the left and right nanospheres is linked by the PT-symmetric relation:

$$\varepsilon(x, y, z) = \varepsilon^*(x, y, -z) \quad (1)$$

or

$$\varepsilon_1 = \varepsilon' + i\varepsilon'', \quad \varepsilon_2 = \varepsilon' - i\varepsilon'' \quad (2)$$

Here the symbol $*$ stands for complex conjugate.

To find the eigenoscillations in such system we use quasi-static dipole approximation. To this end it is assumed that all the system dimensions, i.e. the radii of the nanospheres R as well as the distance D_{12} between their centers are much smaller than the wavelength λ_0 . In the weak coupling approximation when the following condition is fulfilled:

$$2R \ll D_{12} \ll \lambda_0 \quad (3)$$

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Fig. 1: Schematic of PT-symmetric nano-antenna consisting of two coupled resonant nanospheres, one with loss (left) and another with gain (right).

the nanosphere can be approximated as electric dipole with polarization:

$$\alpha_i = R^3 \frac{(\epsilon_i - 1)}{(\epsilon_i + 2)} \quad (4)$$

In PT-symmetric case $\alpha_1 = \alpha_2^* = \alpha$ and the system of self-consistent equations for the dipole moments of nanospheres $\mathbf{d}_1$ and $\mathbf{d}_2$ is then:

$$\mathbf{d}_1 = \alpha \{ \mathbf{E}(\mathbf{d}_2, \mathbf{r}_1) \}, \quad \mathbf{d}_2 = \alpha^* \{ \mathbf{E}(\mathbf{d}_1, \mathbf{r}_2) \}, \quad (5)$$

$$\mathbf{E}(\mathbf{d}_j, \mathbf{r}) = -\frac{\mathbf{d}_j}{|\mathbf{r} - \mathbf{r}_j|^3} + \frac{3(\mathbf{d}_j \cdot (\mathbf{r} - \mathbf{r}_j))(\mathbf{r} - \mathbf{r}_j)}{|\mathbf{r} - \mathbf{r}_j|^5} \quad (6)$$

The system of Eqs. (5) is having non-trivial solutions when its determinant Δ is equal to zero:

$$\Delta = \beta^2 \left((\varepsilon' - 1)^2 + \varepsilon''^2 \right) - \xi^6 \left((\varepsilon' + 2)^2 + \varepsilon''^2 \right) = 0 \quad (7)$$

where $\xi = D_{12}/R$. The parameter $\beta = -1$ corresponds to the eigenoscillations with dipole momentum perpendicular to the axis of the dimer, while $\beta = 2$ to the eigenoscillations with dipole momentum along the axis of the dimer.

Equation (7) determines the dispersion relation between ε' and ε'' , which graphical representation is shown in Fig. 2. It is equivalent to that of a circle with the center:

$$\varepsilon'^* = (-2\xi^6 + \beta^2) / (\xi^6 - \beta^2) \quad (8)$$

and radius:

$$r^* = 3r^3 |\beta| / (\xi^6 - \beta^2) \quad (9)$$

Each pair of points on the dispersion curve with coordinates $\{(\varepsilon'^* + r^* e^{i\varphi}), (\varepsilon'^* - r^* e^{-i\varphi})\}$ corresponds to $\varepsilon_{\pm}$ set of eigenvalues for effective permittivity of PT-symmetric dimer. Two sets of eigenvalues on the dispersion curve are worthy to be mentioned. The first one at the angle $\varphi = 0$ corresponds to the case of two coupled nanospheres in the absence of gain and loss ($\varepsilon'' = 0$). As known, in a system invariant with respect to spatial coordinates inversion (P symmetry) the eigenvalues are corresponding then to symmetric and antisymmetric modes.

The second particular eigenvalue on the dispersion curve occurs at $\varphi = \pi/2$. It corresponds to the exceptional point (EP) where both eigenvalues and eigenvectors coalesce at:

$$\varepsilon'' = \varepsilon''_{\max} = 3|\beta|\xi^3 / (1 - \beta^2 \xi^6) \quad (10)$$

The particular feature of Eq. (7) solutions occurring for $\varepsilon'' < \varepsilon''_{\max}$ is that eigenoscillations for $\varepsilon_{\pm}$ are undamped in spite of the presence of gain and loss in the system. Note however that except when $\varepsilon'' = 0$, the eigenvectors of the dimer are

nonorthogonal, which results in the entanglement of symmetric and antisymmetric modes.

Fig. 2: Solutions of the dispersion Eq. (7) for $\xi=D_{12}/R=2.4$. The red and blue curves correspond to the transversal ($\beta=-1$) or to the longitudinal ($\beta=2$) eigenoscillations respectively. The white circles are corresponding to the exceptional points. The black

square on the graph corresponds to the limit case when $\xi \rightarrow \infty$ while radius $r^* \rightarrow 0$ and the dispersion curve shrinks to a point ($\epsilon' = -2, \epsilon_1'' = \epsilon_2'' = 0$) corresponding to the dipole resonance of an isolated nanosphere.

III. EIGENOSCILLATIONS IN MULTIMODES PT-SYMMETRIC NANO-ANTENNAS: GENERAL THEORY

The analysis carried out so far was limited to the case of single-mode PT-symmetric nano-antennas. The situation is more complex when higher order modes have to be taken into account. The problem is that the validity of the dipolar approximation is limited to the case of weak coupling expressed by Eq. (2). With the enhancement of the coupling interaction between the nano-antennas, it becomes necessary to also take into account the higher order terms in the multipole decomposition of the field. When the distance between the nanospheres is not too small, the dispersion relation for each term of the multipole is also a circle with a center at:

$$\epsilon' = -\frac{n+1}{n} \quad (11)$$

where n stands for the order of multipole ($n=1$ for dipole (*cf.* Fig. 2), $n=2$ for quadrupole etc.).

The issue is that this approach based on multipole decomposition of fields becomes cumbersome with further enhancement of coupling between nano-antennas when phenomenon of mode hybridization starts to play an important role [18, 19]. For this reason, in the following for the description of eigenoscillations in a system of strongly coupled nano-antennas an approach based on bispherical harmonics will be used [20, 21, 22].

To this end, as in the previous section, we consider that the dimensions of the nanoparticles are small relative to the wavelength, that the retardation effects are

negligible, that the quasi-static approach is valid and that the radiation losses are not taken into account. In this case, to find eigenoscillations one should solve the Laplace equation for potential satisfying the conditions of continuity for potential and normal component of electric displacement vector on all boundaries:

$$\mathbf{E} = -\nabla\varphi, \Delta\varphi = 0, \quad (12)$$

$$[\varphi]_S = 0, [\varepsilon\partial\varphi/\partial n]_S = 0, \quad (13)$$

where $\mathbf{E}$ is the electric field vector, φ is the potential, ∇ is the spatial gradient, Δ is the Laplacian, and S is the nanoparticles surfaces.

In the case of two nanospheres, the equation can be conveniently solved in the bispherical system of coordinates (Fig. 3) [23,24].

Fig. 3. The coordinate lines of the bipolar system of coordinates.

The coordinates of the bispherical system $-\infty < \eta < \infty$, $0 < \xi \leq \pi$, $0 \leq \varphi < 2\pi$ are related to the Cartesian coordinates by the relations ([23,24]):

$$x = a \frac{\sin \xi \cos \varphi}{\cosh \eta - \cos \xi}, y = a \frac{\sin \xi \sin \varphi}{\cosh \eta - \cos \xi}, z = a \frac{\sinh \eta}{\cosh \eta - \cos \xi} \quad (14)$$

The equation $\eta = \eta_1 < 0$ defines a passive sphere of the radius $R_1 = a / |\sinh \eta_1|$ in the Cartesian system of coordinates. Its center is situated in the point $x_1 = y_1 = 0$, $z_1 = a \coth \eta_1$. The second (active) sphere may be defined analogously by the equation $\eta = \eta_2 > 0$. At that, its radius will be equal to $R_2 = a / \sinh \eta_2$, and the center will be situated in the point $x_2 = y_2 = 0$, $z_2 = a \coth \eta_2$. The dimensional constant a is half of the distance between the poles of the bispherical system of coordinates. It is defined by the positive root of the equation:

$$D_{12} = \sqrt{R_1^2 + a^2} + \sqrt{R_2^2 + a^2}$$

where $D_{12} = z_2 - z_1$ is an arbitrary distance between the centers of the first and second spheres ($D_{12} \geq R_1 + R_2$). Hereafter in this section, only the case of spheres with equal radii R_0 ($-\eta_1 = \eta_2 = \eta_0$), made of same material with the electric permittivity ϵ_1 , and placed in a medium with the electric permittivity ϵ_2 will be considered for simplicity.

In the bi-spherical system of coordinates variables can be separated, and the arbitrary solution of the Laplace equation may be written in the form [23, 24]:

$$\Phi = \frac{1}{a} \sqrt{\cosh \eta - \cos \xi} P_{m+n-1}^m(\cos \xi) e^{\pm \left(n + \frac{1}{2}\right) \eta \pm i m \varphi} \quad (15)$$

where $m = 0, 1, 2, \dots; n = 1, 2, 3, \dots$, and $P_n^m(\cos \xi)$ are the associated Legendre polynomials.

In the space outside the spheres ($-\eta_0 < \eta < \eta_0$), the most general solution may be sought as:

$$\Phi = \frac{1}{a} \sqrt{\cosh \eta - \cos \xi} \sum_{m=0}^{\infty} \sum_{n=1}^{\infty} P_{m+n-1}^m(\cos \xi) \times \cos(m\varphi) \times \left(\alpha_{mn} \frac{\cosh\left(\left(m+n-\frac{1}{2}\right)\eta\right)}{\cosh\left(\left(m+n-\frac{1}{2}\right)\eta_0\right)} + \gamma_{mn} \frac{\sinh\left(\left(m+n-\frac{1}{2}\right)\eta\right)}{\sinh\left(\left(m+n-\frac{1}{2}\right)\eta_0\right)} \right) \quad (16)$$

while inside the spheres only the solutions decreasing for $|\eta| \rightarrow \infty$ can be used according to the potential finiteness requirement. Thus, the potential inside the first ($\eta < -\eta_0 < 0$) and second ($\eta > \eta_0 > 0$) spheres can be expressed as:

$$\begin{aligned}\Phi^{(1)} &= \frac{1}{a} \sqrt{\cosh \eta - \cos \xi} \sum_{m=0}^{\infty} \sum_{n=1}^{\infty} e^{\left(m+n-\frac{1}{2}\right)\eta} P_{m+n-1}^m(\cos \xi) \times a_{mn}^{(1)} \times \cos m\varphi \\ \Phi^{(2)} &= \frac{1}{a} \sqrt{\cosh \eta - \cos \xi} \sum_{m=0}^{\infty} \sum_{n=1}^{\infty} e^{-\left(m+n-\frac{1}{2}\right)\eta} P_{m+n-1}^m(\cos \xi) \times a_{mn}^{(2)} \times \cos m\varphi\end{aligned}\quad (17)$$

In Eqs. (15) and (16), α_{mn} , γ_{mn} and $a_{mn}^{(1)}$, $a_{mn}^{(2)}$ are unknown coefficients, which are to be determined basing on the boundary conditions on the surfaces of both spheres.

From the condition of potential continuity on the surface of each nanosphere, we readily obtain the relation between $a_{mn}^{(1)}, a_{mn}^{(2)}$ and α_{mn}, γ_{mn} :

$$\begin{aligned}a_{mn}^{(1)} e^{-\left(m+n-\frac{1}{2}\right)\eta_0} &= \alpha_{mn} - \gamma_{mn} \\ a_{mn}^{(2)} e^{-\left(m+n-\frac{1}{2}\right)\eta_0} &= \alpha_{mn} + \gamma_{mn}\end{aligned}\quad (18)$$

The continuity condition of the normal component of electric induction results in an infinite chain of interlaced equations. In the case of two identical spheres ($-\eta_1 = \eta_2 = \eta_0$), these equations may be written in the following matrix form:

$$\begin{aligned}\left[\varepsilon_0 \hat{\mathbf{A}}_0 + \varepsilon_1 \hat{\mathbf{A}}_1 \right] \vec{\alpha} + \left[\varepsilon_0 \hat{\mathbf{\Gamma}}_0 - \varepsilon_1 \hat{\mathbf{A}}_1 \right] \vec{\gamma} &= 0 \\ \left[\varepsilon_0 \hat{\mathbf{A}}_0 + \varepsilon_2 \hat{\mathbf{A}}_1 \right] \vec{\alpha} - \left[\varepsilon_0 \hat{\mathbf{\Gamma}}_0 - \varepsilon_2 \hat{\mathbf{A}}_1 \right] \vec{\gamma} &= 0\end{aligned}\quad (19)$$

where: $\vec{\alpha} = (\alpha_{m,1}, \alpha_{m,2}, \dots, \alpha_{m,n}, \dots)$; $\vec{\gamma} = (\gamma_{m,1}, \gamma_{m,2}, \dots, \gamma_{m,n}, \dots)$ are the eigenvectors, and $\hat{\mathbf{A}}_0, \hat{\mathbf{A}}_1$ and $\hat{\mathbf{\Gamma}}_0$ are infinite matrices with nonzero matrix elements

$$\begin{aligned}
A_{0n,n-1} &= -(n-1) \tanh[y_{m,-1+n}\eta_0] \\
A_{0n,n} &= \sinh[\eta_0] + 2 \cosh[\eta_0] \tanh[y_{m,n}\eta_0] y_{m,n} \\
A_{0n,n+1} &= -(2m+n) \tanh[y_{m,1+n}\eta_0]
\end{aligned} \tag{20}$$

$$\begin{aligned}
A_{1n,n-1} &= -(n-1) \\
A_{1n,n} &= -\sinh[\eta_0] + 2 \cosh[\eta_0] y_{m,n} \\
A_{1n,n+1} &= -(2m+n)
\end{aligned} \tag{21}$$

$$\begin{aligned}
\Gamma_{0n,n-1} &= \coth[y_{m,-1+n}\eta_0] (n-1) \\
\Gamma_{0n,n} &= -\sinh[\eta_0] - 2 \cosh[\eta_0] \coth[y_{m,n}\eta_0] y_{m,n} \\
\Gamma_{0n,n+1} &= \coth[y_{m,1+n}\eta_0] (2m+n)
\end{aligned} \tag{22}$$

$$y_{m,n} = m + n - 1/2 \tag{23}$$

As it can be seen, in the case of P-symmetry ($\epsilon_1 = \epsilon_2$) the system of Eqs. (19) can be decomposed in symmetric and antisymmetric solutions for α and γ , respectively. This is no longer true for PT-symmetric case where symmetric and antisymmetric modes are entangled.

By introducing a state vector $\Psi = (\vec{\alpha}, \vec{\gamma})$ the equation on eigenvalues can be expressed as:

$$\epsilon_0 \begin{bmatrix} \hat{\mathbf{A}}_0 & \hat{\mathbf{\Gamma}}_0 \\ \hat{\mathbf{A}}_0 & -\hat{\mathbf{\Gamma}}_0 \end{bmatrix} \Psi - i\epsilon'' \begin{bmatrix} -\hat{\mathbf{A}}_1 & \hat{\mathbf{A}}_1 \\ \hat{\mathbf{A}}_1 & \hat{\mathbf{A}}_1 \end{bmatrix} \Psi = -\epsilon' \begin{bmatrix} \hat{\mathbf{A}}_1 & -\hat{\mathbf{A}}_1 \\ \hat{\mathbf{A}}_1 & \hat{\mathbf{A}}_1 \end{bmatrix} \Psi \tag{24}$$

It is convenient to cast Eqs. (24) in the form:

$$\epsilon_0 \begin{bmatrix} \hat{\mathbf{Q}}^- & \hat{\mathbf{Q}}^+ \\ \hat{\mathbf{Q}}^+ & \hat{\mathbf{Q}}^- \end{bmatrix} \tilde{\Psi} + i\epsilon'' \begin{bmatrix} \hat{\mathbf{A}}_1 & 0 \\ 0 & -\hat{\mathbf{A}}_1 \end{bmatrix} \tilde{\Psi} = -\epsilon' \begin{bmatrix} \hat{\mathbf{A}}_1 & 0 \\ 0 & \hat{\mathbf{A}}_1 \end{bmatrix} \tilde{\Psi} \tag{25}$$

where $\tilde{\Psi} = (\vec{\alpha} - \vec{\gamma}, \vec{\alpha} + \vec{\gamma})$ and $\hat{\mathbf{Q}}^\pm = (\hat{\mathbf{A}}_0 \pm \hat{\mathbf{\Gamma}}_0) / 2$.

As it can be seen Eqs. (23) and (24) represent non-Hermitian PT-symmetric Hamiltonians with real eigenvalues ϵ' [25]. They can be solved for instance with respect

to the real component ϵ' of permittivity while considering the imaginary part ϵ'' as parameter.

Search of the resonance electric permittivities in the proposed definition corresponds solving the so-called generalized eigenvalues problem for the systems of equations. The generalized problem of eigenvalues (which has the form $\hat{A}x = \lambda \hat{B}x$) is much more complex than the standard eigenvalues problem (which has the form $\hat{A}x = \lambda x$), and so its solutions describe a larger class of phenomena.

IV. ENTANGLEMENT OF BRIGHT AND DARK MODES IN MULTIMODE PT-SYMMETRIC PLASMONIC NANO-ANTENNAS

The previous section was devoted to the derivation of natural frequencies for multimode PT-symmetric strongly coupled nano-antennas. Meanwhile, though the determined solutions are exact, they can hardly be used to analyze the behavior of the system because of the necessity to operate with matrices of infinite size.

For this reason, in the following the analysis of multimode PT-symmetric nano-antennas is performed on the basis of numerical results obtained for a selection of relevant examples.

The initial point of our analysis concerns the example of multimode P-symmetric nano-antennas where $\epsilon'_1 = \epsilon'_2$ while $\epsilon''_1 = \epsilon''_2 = 0$. The spectrum of transverse eigenoscillations as function of separation distance between nanospheres is shown in Fig. 4.

As shown in Fig. 4, when the separation distance between nanospheres is relatively large, the interaction between nano-antennas is small enough in order to allow the use of hybridization approach [18, 19]. The situation is very similar to that described in Section II of the P-symmetric monomode dimer analysis. Each eigenmode corresponding to an isolated nanosphere is split into a symmetric (P-mode) and antisymmetric (L-mode). Thus, for example, the dipole mode of a nanosphere with $\epsilon = -2$ is split into a symmetric

P_1 -mode and antisymmetric L_1 -mode. Similarly, the quadrupole mode with $\epsilon=-3/2$ is split into a symmetric P_2 -mode and antisymmetric L_2 -mode. However, as long as the separation distance between the nanospheres is reduced, the mode with a different symmetry begin to cross. In addition, a different type of highly localized M-modes in the nanosphere's gap is emerging [20, 21].

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Fig. 4. Eigenvalues of P symmetric coupled nano-antennas as function of separation distance between them. The dotted lines indicate the separation distances between the nanospheres used in selected examples of numerical results analysis (see Fig.5)

As for P-symmetric dimers, the same trends can be observed during the transition from a monomode to a multimode PT-symmetric dimer. As shown in Section II on the example of monomode PT-symmetric dimer, the introduction of gain and loss leads to the

entanglement of symmetric and antisymmetric modes. The presence of higher order modes in multimode PT-symmetric dimers entails the entanglement of even more modes.

The evolution of the dispersion relation of eigenoscillations in multimode PT-symmetric dimer as a function of their dielectric permittivity for different separation distance between nanospheres is shown in Fig. 5. As can be seen when the separation distance between nanospheres is large, the dispersion relation appears as follows: a collection of nonintersecting ovals. Each oval corresponds to a different order mode in multipolar decomposition. Thus the oval centered at $\epsilon' = -2$ is that of the dipole mode discussed previously in Section II.

Fig. 5. Loci in the plan ϵ', ϵ'' of PT-symmetric multimode dimer eigenvalues as function of separation distance $D_{12}/(2R)$ between nanospheres.

The reduction of the separation distance between the nanospheres leads to the crossing of dispersion curves of second order (quadrupole) and third order (octupole) modes, which progressively transforms into two nested ovals. The number of entangled modes

for all points of the dispersion curve, with the exception of those on the real axes $\varepsilon'(\varepsilon''=0)$, now goes from two (L_2, P_2 or L_3, P_3) to four (L_2, P_2, L_3, P_3). The reduction of the separation distance between the nanospheres leads to the crossing of the dispersion curves of the second-order (quadrupole) and third-order (octupole) modes, which progressively transforms into two nested ovals. The number of entangled modes for all points of the dispersion curve, with the exception of those on the real axes, now goes from two (L_2, P_2 or L_3, P_3) to four (L_2, P_2, L_3, P_3). Further reduction of the separation distance leads to crossing with the first-order mode and a gradual transition to the situation when two ovals are nested in a larger oval. As in the case previously considered, the number of entangled modes increases to six ($L_1, P_1, L_2, P_2, L_3, P_3$). This hybridization process is shown schematically in Fig. 6.

Fig. 6. Diagram of the evolution of the dispersion curves of the eigenfrequencies of multimode PT-symmetric dimers as a function of the coupling force between nanospheres. a) Weakly coupled nanospheres. The dispersion curves corresponding to different order modes in multipolar decomposition do not intersect; b) Moderate coupling resulting in a cross and hybridization of second and third order mode dispersion curves; c) Strong coupling above the critical value leading to the formation of nested dispersion curves; d) Crossing and hybridization of the first order mode with the entanglement mode formed by hybridization of the second and third order modes; e) Higher strength coupling resulting in the nesting of two internal dispersion curves inside an external dispersion curve.

To illustrate the evolution of the entanglement of modes induced by the hybridization process, the spatial distribution of the complex potential representing the eigenfunctions of the PT-symmetric coupled nanospheres was calculated using the bisphere harmonic approach presented in section III.

The dispersion curves and the spatial distribution of the real and imaginary components of the complex potential of weakly coupled nanospheres (uncrossed ovals) are illustrated in Fig. 7a. As can be seen, the leftmost point on the dispersion curve ($\epsilon' \approx -2.2, \epsilon'' = 0$) corresponds to the purely antisymmetric mode L_1 , which for reasons of symmetry cannot be excited by a plane wave and is therefore dark. In contrast, the rightmost point of the dispersion curve ($\epsilon' \approx -1.87, \epsilon'' = 0$) represents the purely symmetric bright mode P_1 . The distribution of electrical charges for this bright eigenmode corresponds to the excitation of a transverse electric dipole. The situation is different for the rest of dispersion curve where $\epsilon'' \neq 0$. The spatial distribution of the complex potential now corresponds to the entanglement of symmetric and antisymmetric modes. Nevertheless, even in this case, we can still observe some reminiscences of the initial P-symmetric dimer bright and dark

modes. Thus, the distribution of the real component of the complex potential right to the exceptional point ($\epsilon' \approx -2.03, \epsilon'' = 0.18$) strongly resembles that of a bright mode. This resemblance is less pronounced for the points left to the EP.

The case of moderate coupling with quadrupole and third order hybridization is illustrated in Fig. 7b. From the leftmost point of the dispersion curve ($\epsilon' \approx -1.36, \epsilon'' = 0$) the eigenmodes composition evolves from purely antisymmetric L_3 (dark) to entangled L_3 and P_3 , then to entangled L_3, P_3, L_4 until terminating with purely symmetric P_3 mode (bright).

These results show that the PT-symmetric coupled nano-antennas can present a spectrum of real eigenvalues not only for the fundamental dipole mode, but also for the higher order modes. The hybridization process resulting from the decrease of the separation distance between the nano-antennas is accompanied by an increase in the number of entangled modes on the branch of the dispersion curve.

a)

b)

c)

Fig. 7. Dispersion curves and spatial distribution of real and imaginary component of the complex potential on the right and left side of the inset, respectively. a) Separation

distance between nanospheres $D_{12}/(2R) = 1.33$ corresponds to the case of weakly coupled nanospheres and non-crossing dispersion curves; b) Separation distance between nanospheres $D_{12}/(2R) = 1.319$. Moderately coupled nanospheres with crossing of second and third higher order modes dispersion curves; c) Separation distance between nanospheres $D_{12}/(2R) = 1.315$ corresponds to the case of strongly coupled nanospheres and imbricated dispersion curves.

The case of a strong coupling resulting in the formation of two nested ovals inside an outer oval is shown in Fig. 7c. The complex potential of the outer oval dispersion curve is determined by the real part of the dielectric permittivity of two closest to this eigenvalue dark and bright modes. It should be noted that the modes having an equal real part of the eigenvalue but different imaginary parts have a similar distribution of the complex potential. Another point to mention is that the increase of ϵ'' leads to the decrease in the number of branches for the dispersion curve. This behavior is very similar to that observed in a system of multimode coupled waveguides [26].

Also note that the value $\epsilon''_{\max}$ delineating the upper limit of gain and loss for which the eigenfrequencies are real decreases with the number of higher order modes supported by the nanosphere. Since lower order terms in multipolar field decomposition make an essential contribution to the scattering process, the implementation of an adjustable and reconfigurable scattering response obtained through a gain-loss modulation mechanism would need to introduce a limitation on the number of higher order modes in the system.

V. SUMMARY AND CONCLUSIONS

The generic model of two coupled multimode plasmonic nanospheres with gain and loss forming a PT-symmetric dimer is considered to develop an analytic approach based on quasi-static approximation for eigenmodes solutions. It is shown that the distinctive

feature of such PT-symmetric dimers is that, despite undamped oscillations, the eigenvectors of the dimer are not orthogonal, resulting in the entanglement of different multipole order modes with odd and even symmetries. The scattering properties and modal composition of such system with interleaved dark (odd) and bright (even) modes can be controlled by modes hybridization as a function of separation distance between nanospheres as well as the level of PT-symmetric gain-loss.

This opens promising avenues for the realization of active plasmonic metamolecules with tunable and reconfigurable scattering response. Their practical implementation can be obtained by considering core-shell geometry for plasmonic nanoresonators. For example, the core may be silver while the shell is either CAS 7149-49-7 (naphthalene-2,3-dicarboxaldehyde) dyestuff with an emission length of 331 nm [27, 28] or CAS 477934-10-4 (2- (benzoxazolyl) phenylbenzoate) with an emission wavelength of 361 nm [29].

In addition to the prospects for applications, the study of multimodal PT-symmetric dimers is also of fundamental interest. The presence of multiple phase transitions and branching points on the eigenfrequency diagram leads particularly relevant exploration of such systems in a more general context of non-Hermitian photonics [30, 31, 32].

ACKNOWLEDGEMENTS

Funding by the Russian Foundation for the Basic Research (grant №18-02-00315) and from the MEPhI Academic Excellence Project (grant from the Ministry of Education and Science of the Russian Federation No. 02.a03.21.0005) is acknowledged by V.K.

References

1. A. Vena, E. Perret, and S. Tedjini, “Chipless RFID tag using hybrid coding technique,” *IEEE Trans. Microwave Theory Tech.* **59**, 3356-3364. (2011).
2. R. Singh, W. Cao, I. Al-Naib, L. Cong, W. Withayachumnankul, W. Zhang, “Ultrasensitive terahertz sensing with high-Q Fano resonances in metasurfaces,” *Appl. Phys. Lett.* **105** (17), 171101 (2014).
3. B. Gallinet and O. J. F. Martin, “Refractive index sensing with subradiant modes: a framework to reduce losses in plasmonic nanostructures,” *ACS Nano* **7**, 6978 (2013).
4. V. V. Klimov, A. A. Pavlov, I. V. Treshin, I. V. Zabkov, Fano resonances in a photonic crystal covered with a perforated gold film and its application to bio-sensing *J. Phys. D: Appl. Phys.* **50**, 285101 (2017).
5. S. Xiao, V. P. Drachev, A. V. Kildishev, X. Ni, U. K. Chettiar, H.-K. Yuan, and V. M. Shalaev, “Loss-Free and Active Optical Negative-Index Metamaterials,” *Nature* **466**(7307), 735–8 (2010).
6. R. F. Oulton, V. J. Sorger, T. Zentgraf, R.-M. Ma, C. Gladden, L. Dai, G. Bartal, and X. Zhang, “Plasmon lasers at deep subwavelength scale,” *Nature* **461**, 629–632 (2009).
7. S. Longhi, “Parity-time symmetry meets photonics: A new twist in non-Hermitian optics,” *Europhysics Letters* **120**, 64001 (2018).
8. C. E. Rüter, K. G. Makris, R. El-Ganainy, D. N. Christodoulides, M. Segev, and D. Kip, “Observation of parity–time symmetry in optics,” *Nature Phys.* **6**, 192–195 (2010).
9. L. Feng, Y.-L. Xu, W. S. Fegadolli, M.-H. Lu, J. E. Oliveira, V. R. Almeida, Y.-F. Chen, and A. Scherer, “Experimental demonstration of a unidirectional reflectionless parity-time metamaterial at optical frequencies,” *Nat. Mater.* **12**, 108–113 (2012).
10. B. Peng, Ş. K. Özdemir, S. Rotter, H. Yilmaz, M. Liertzer, F. Monifi, C. M. Bender, F. Nori, L. Yang, “Loss-induced suppression and revival of lasing,” *Science* **346**, 328–332 (2014).

- 11.Z. J. Wong, Y. L. Xu, J. Kim, K. O'Brien, Y. Wang, L. Feng, and X. Zhang, "Lasing and anti-lasing in a single cavity," *Nature photonics* 10, 796-801 (2016).
- 12.A. Kodigala, T. Lepetit, and B. Kanté, "Exceptional points in three-dimensional plasmonic nanostructures," *Phys. Rev. B* 94(20), 201103 (2016).
- 13.M. Sakhdari, M. Farhat, and P. Y. Chen, "PT-symmetric metasurfaces: wave manipulation and sensing using singular points," *New Journal of Physics* 19(6), 065002 (2017).
- 14.H. Hodaei, A. U. Hassan, S. Wittek, H. Garcia-Gracia, R. El-Ganainy, D. N. Christodoulides, and M. Khajavikhan, "Enhanced sensitivity at higher-order exceptional points," *Nature*, 548(7666), 187 (2017).
- 15.H. Hodaei, M.-A. Miri, A. U. Hassan, W. E. Hayenga, M. Heinrich, D. N. Christodoulides, and M. Khajavikhan, "Parity-time-symmetric coupled microring lasers operating around an exceptional point," *Opt. Lett.* 40,4955–4958 (2015).
- 16.H. Hodaei, A. Hassan, W. E. Hayenga, M. A. Miri, D. N. Christodoulides, and M. Khajavikhan, "Dark-state lasers: mode management using exceptional points," *Opt. Lett.* 41, 3049-3052 (2016).
- 17.Q. Zhong, J. Ren, M. Khajavikhan, D. Christodoulides, S. Ozdemir, and R. El-Ganainy, "Sensing with exceptional surfaces: combining sensitivity with robustness," *arXiv:1810.09417* (2018).
- 18.E. Prodan, C. Radloff, N. J. Halas, and P. Nordlander, "A Hybridization Model for the Plasmon Response of Complex Nanostructures," *Science* 302(5644), 419–22 (2003).
- 19.E. Prodan and P. Nordlander, "Plasmon Hybridization in Spherical Nanoparticles.," *J. Chem. Phys.* 120(11), 5444–54 (2004).
- 20.V. V. Klimov and D. V. Guzatov, "Plasmonic Atoms and Plasmonic Molecules," *Appl. Phys. A* 89(2), 305–314 (2007).

- 21.V. V. Klimov and D. V. Guzatov, "Strongly Localized Plasmon Oscillations in a Cluster of Two Metallic Nanospheres and Their Influence on Spontaneous Emission of an Atom," *Phys. Rev. B* **75**(2), 024303 (2007).
- 22.V. Klimov, *Nanoplasmonics*, (Pan Stanford Publishing, Singapore 2014)586 p.
- 23.H. Buchholz, *Elektrische und magnetische Potentialfelder*, Springer , Berlin, 1957. Ch.2.
- 24.H. Feshbach, P. M. Morse, *Methods of Theoretical Physics*, Cambridge University Press ,1953. Ch.10.
- 25.C. M. Bender and S. Boettcher, "Real Spectra in Non-Hermitian Hamiltonians Having PT Symmetry," *Phys. Rev. Lett.* **80**, 5243 (1998).
- 26.H. Benisty, A. Lupu, and A. Degiron, "Transverse periodic PT symmetry for modal demultiplexing in optical waveguides," *Phys. Rev. A* **91**, 053825 (2015).
- 27.T. Ueda, F. Kitamura, R. Mitchell, T. Metcalf, T. Kuwana, and A. Nakamoto, "Chiral separation of naphthalene-2,3-dicarboxaldehyde-labeled amino acid enantiomers by cyclodextrin-modified micellar electrokinetic chromatography with laser-induced fluorescence detection," *Anal. Chem.* **63**, 2979-2981 (1991).
- 28.www.scbt.com
- 29.D. A. Powell, "Interference between the modes of an all-dielectric meta-atom," *Phys. Rev. Applied*, **7**(3), 034006 (2017).
- 30.H. Lourenço-Martins, P. Das, L. H. Tizei, R. Weil, and M. Kociak, "Self-hybridization within non-Hermitian localized plasmonic systems," *Nature Physics*, **14**(4), 360 (2018).
- 31.W. Yan, R. Faggiani, and P. Lalanne, "Rigorous modal analysis of plasmonic nanoresonators," *Phys. Rev. B*, **97**(20), 205422 (2018).