Parametric resonances in nonlinear plasmonics [Invited]

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In the context of nonlinear plasmonics, we review the recently introduced concept of plasmonic parametric resonance (PPR) and discuss potential applications of such phenomena. PPR arises from the temporal modulation of one or more of the parameters governing the dynamics of a plasmonic system and can lead to the amplification of high-order sub-radiant plasmonic modes. The theory of PPR is reviewed, possible schemes of implementation are proposed, and applications in optical limiting are discussed.

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Localized surface plasmon (LSP) resonances are a salient feature of the optical and electronic response of metallic nanoparticles. These modes can be externally excited by photonic or electronic scattering, leading to strongly localized electric fields in proximity of the nanoparticle's surfaces. An enhanced optical response is obtained when LSPs are resonantly excited by an incident field at the characteristic frequency of the dipolar eigenmode. The dynamics of these non-propagating coherent electronic oscillations show a strong dependence on the geometry of the particles, their composition, and the dielectric environment in which they are located. Yet, some features are common to all plasmonic configurations. In particular, LSP resonances in nanoparticles of any shape form an infinite discrete set of modes. In the simple case of particles of spherical shape with permittivity $\varepsilon_1(\omega)$ surrounded by a medium with permittivity ε_2, for a resonance of any order n, there are $2n+1$ degenerate states with complex frequency ωn, such that $\varepsilon_1(\omega n) = -(1+n)\varepsilon_2/n$. As shown in Fig. 1, for $n \gg 1$, the eigenmodes tend to occur for $\varepsilon_1(\omega n \gg 1) \sim -\varepsilon_2$. The increased modal density for $\varepsilon_1 \sim -\varepsilon_2$ is a general feature of all plasmonic structures. Such increased modal density in plasmonic particles of different shapes for $\varepsilon_1 \sim -\varepsilon_2$ arises because the spatial oscillation of the fields along the particle's surface occurs with a negligible local wavelength compared with the local radius of curvature of the metal–dielectric interface[1]. Accessing such spectrally dense sets of tightly bound resonant modes would greatly enhance nonlinear light–matter interactions at the nanoscale and foster new developments in nonlinear plasmonics[2].

The efficiency with which LSP resonances can be excited by an external incident field depends upon the spatial and spectral overlap between the excitation field and the specific plasmonic mode. For deeply subwavelength plasmonic particles, only the lowest-order mode of an electric dipolar nature is efficiently coupled to radiation states. In order for a particular mode to be efficiently excited, it is necessary for the incident field to be able to induce the appropriate polarization charge distribution. Such polarization charge distributions are illustrated in Fig. 2 for the first few modes of a sphere. From Fig. 2, it is apparent that to induce modes of order $n > 1$ the incident field would have to display strong spatial variations over deeply subwavelength regions. That is why the higher-order eigenmodes tend to be sub-radiant, and by reciprocity, they are nearly decoupled from free-space propagating fields. Therefore, exciting and detecting such higher-order modes requires either near-field scattering techniques[3] or the use of active media to promote surface plasmon amplification by stimulated emission of radiation (SPASER)[4].

A different mechanism to drive high-order LSP modes with a spatially uniform optical field relies on the recently introduced concept of plasmonic parametric resonance (PPR)[5]. In contrast with conventional localized plasmonic resonances, in which modes are excited directly by an external field of frequency and spatial profile matching those of a given mode of the plasmonic particle, PPR is a form of amplification in which a pump field transfers energy to a mode in an indirect way. In PPR, in fact, the modes of a plasmonic structure are amplified by means of a temporal

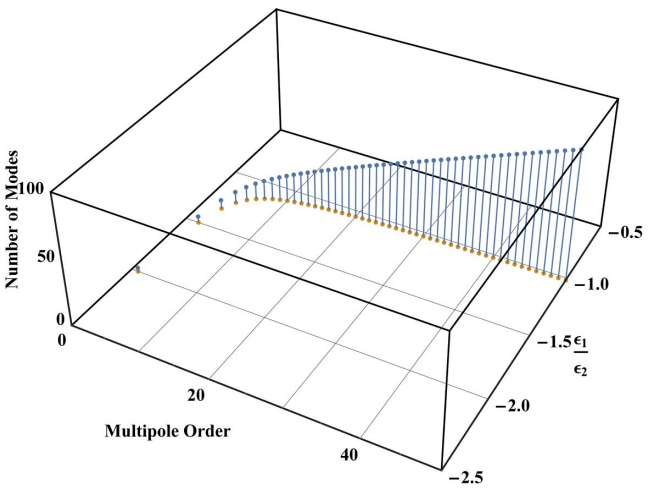

Fig. 1. Spectral distribution of the LSP resonances in a plasmonic sphere.

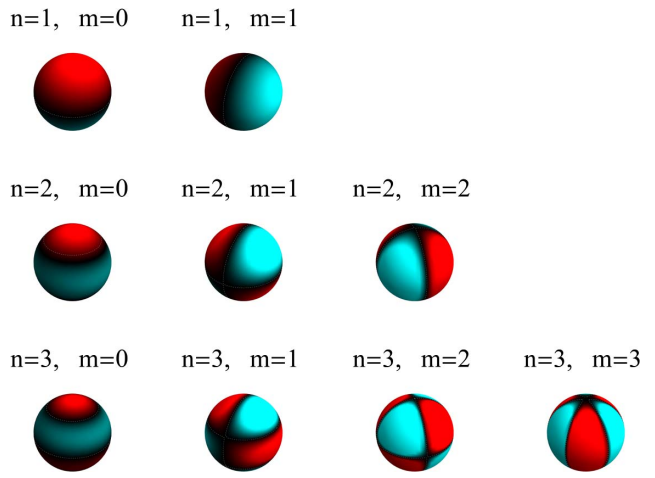

Fig. 2. Polarization charge density of the first few resonant modes of a plasmonic sphere.

modulation of the background permittivity caused by an appropriate pump field. Such permittivity variation translates into a modulation of the modal resonant frequency. Under specific pump conditions, amplification can occur. As shown in Ref. [5], among the unique characteristics of PPR is the possibility of accessing modes of arbitrarily high order with a simple spatially uniform pump, provided that such pump exceeds a certain intensity threshold—a characteristic of all parametric resonances.

In very general terms, a parametric resonance[6] occurs when one or more of the parameters controlling the evolution of a dynamical system undergo a temporal modulation of appropriate amplitude and frequency. When such conditions are met, the amplitude of the parametrically resonant mode increases exponentially with time as long as the parametric modulation continues. In formal terms, the temporal evolution of a representative dynamical variable $X(t)$ of a system with resonant frequency ω_0 and damping γ under the action of a stimulus $F(t)$ is given by Eq. (1) in the case of direct excitation, and by Eq. (2) for parametric excitation:

$$\frac{\mathrm{d}^2X(t)}{\mathrm{d}t^2}+\gamma\frac{\mathrm{d}X(t)}{\mathrm{d}t}+\omega_0^2X(t)=F(t), \tag{1}$$

$$\frac{\mathrm{d}^2X(t)}{\mathrm{d}t^2}+\gamma\frac{\mathrm{d}X(t)}{\mathrm{d}t}+\{\omega_0^2+2\omega_0\delta\omega[F(t)]\}X(t)=0. \tag{2}$$

In the parametric Eq. (2), the external stimulus $F(t)$ acts indirectly on the system by modifying the instantaneous resonant frequency by the amount $\delta\omega[F(t)]$.

A simple system described by Eq. (1) could be a simple harmonic oscillator, like a mass m attached to a spring with elastic constant $k=m\omega_0^2$. In this example, $X(t)$ represents the displacement of the mass from the equilibrium position. The restoring force provided by the spring can be described in terms of a potential energy $U=kX^2/2$. As illustrated in Fig. 3(a), the free evolution of the system is a damped harmonic oscillation. Now, let us assume that

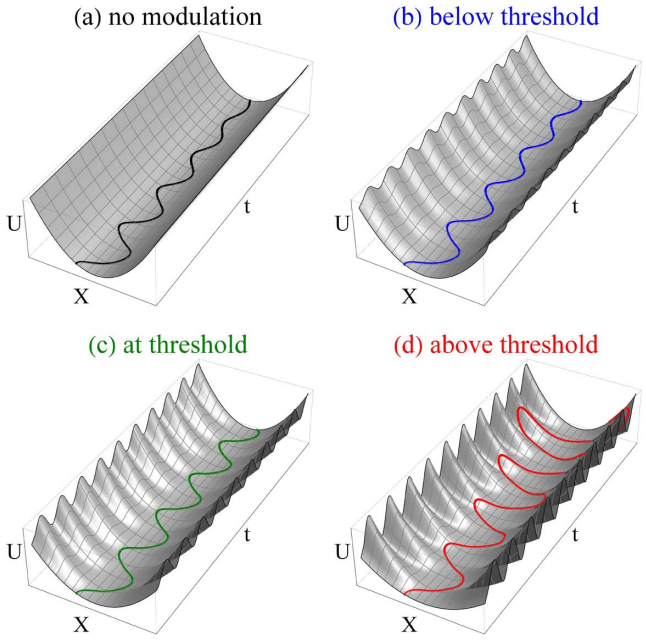

Fig. 3. (a) Time evolution of the position X of a harmonic oscillator in a parabolic potential U. (b)–(d) Time evolution of a parametrically driven oscillator with time-varying potential. (b) Below threshold, the oscillation decays. (c) At the threshold of parametric regeneration, the dissipations are exactly compensated. (d) Above threshold, the parametric gain causes the oscillations to grow exponentially.

the elastic constant of the spring is changed with time through some external mechanism. This new situation can be effectively described by Eq. (2). In the presence of such parametric modulation, the potential landscape varies with time, as shown in Figs. 3(b)–3(d), for the case of a sinusoidal modulation around the unperturbed potential. Under the appropriate conditions, the energy that is externally provided to change the potential of the system can be transferred to the harmonic oscillator to compensate or even overcome the dissipation mechanisms. If a certain parametric-modulation threshold is exceeded, the oscillation amplitude grows in time with an exponential envelope, as indicated in Fig. 3(d). In PPR, these concepts are extended and applied to heavily multimode optical resonators such as plasmonic particles.

In order to illustrate in general terms the principle of the operation of PPR in a plasmonic nanoparticle of arbitrary shape, the first step is to identify a dynamical variable obeying an evolution equation similar to Eq. (2). The first complication that arises is due to the fact that all the relevant physical quantities involved in such an electromagnetic problem are fields of some sort (like electric potential, electric field, magnetic field, polarization density, and current density), rather than simple kinematic variables like the position $X(t)$ considered in the previous harmonic oscillator example. This issue is addressed by performing a modal decomposition of the field of interest in terms of an appropriate complete and orthogonal set of basis functions. By doing so, the field is expressed as a sum of vector-field basis functions, each weighted with a

scalar time-dependent modal amplitude determined by the initial conditions and by the subsequent evolution. As shown in the following, such modal amplitudes can be used as dynamical variables to cast the PPR problem in the form of Eq. (2). Each modal amplitude will evolve in time, according to an equation of the same form as Eq. (1), with an appropriate natural frequency ω_n depending on the specific mode, the permittivity ε_1 of the particle, and the permittivity ε_2 of the surrounding medium. This approach immediately suggests that modulating one of these properties, say ε_2, will cast the problem in the desired form of Eq. (2). In the following, for the purpose of illustration of PPR, we will consider a system that is amenable to a close-form solution: a subwavelength plasmonic sphere in a homogeneous dielectric background medium.

We consider a sphere of radius R and relative permittivity ε_1 (medium 1) embedded in a uniform dielectric medium ε_2 (medium 2). The radius R is assumed to be much smaller than the free-space wavelength associated with any of the plasmonic eigenmodes of interest, so that a quasi-static approach is applicable for determining the spatial distribution of the electromagnetic field. The dispersion of ε_2 is neglected. Medium 1 is assumed to follow a Drude-like frequency-domain dispersion $\varepsilon_1(\omega)=\varepsilon_\infty-\omega_{pl}^2/(\omega^2+i\omega\gamma)$, with plasma frequency ω_{pl}, collision frequency γ, and a non-dispersive term accounting for high-frequency spectral features ε_∞. The dispersive term in the $\varepsilon_1(\omega)$ expression is associated with the equation of motion for the free-carrier polarization density $\mathbf{P}_1(\mathbf{r},t)$ within medium 1:

$$\frac{\partial^2\mathbf{P}_1(\mathbf{r},t)}{\partial t^2}+\gamma\frac{\partial\mathbf{P}_1(\mathbf{r},t)}{\partial t}=\varepsilon_0\omega_{pl}^2\mathbf{E}_1(\mathbf{r},t), \tag{3}$$

where $\mathbf{E}_1(\mathbf{r},t)$ is the electric field within medium 1. Medium 2 is assumed to be endowed with second-order nonlinearity with a dominant term $\chi^{(2)}_{zzz}$. Under such hypotheses, the polarization density $\mathbf{P}_2(\mathbf{r},t)$ in medium 2 can be expressed in terms of the total local field $\mathbf{E}_2(\mathbf{r},t)$ as follows:

$$\mathbf{P}_2(\mathbf{r},t)=\varepsilon_0(\varepsilon_2-1)\mathbf{E}_2(\mathbf{r},t)+\mathbf{P}_2^{NL}(\mathbf{r},t). \tag{4}$$

In Eq. (4), $\mathbf{P}_2^{NL}(\mathbf{r},t)=\varepsilon_0\chi^{(2)}\cdot\mathbf{E}_2(\mathbf{r},t):\mathbf{E}_2(\mathbf{r},t)$ is the nonlinear polarization density due to the quadratic nonlinearity of medium 2. The total electric field $\mathbf{E}_2(\mathbf{r},t)$ in medium 2 is the sum of all the fields due to the plasmonic modes of the particle and a spatially uniform incident field $\mathbf{E}_P(t)$, henceforth referred to as "pump".

In the quasi-static approximation, the polarization density in medium 1 can be expanded in terms of spherical harmonics $Y_{n,m}^{(e/o)}(\theta,\phi)$, defined and normalized as in Ref. [5]:

$$\mathbf{P}_1(t)=\sum_{n,m}\nabla\left\{\frac{\mathbf{r}^n}{R^{n-1}}\left[\mathbf{P}_{n,m}^{(e)}(t)\,Y_{n,m}^{(e)}(\theta,\phi)+\mathbf{P}_{n,m}^{(o)}(t)\,Y_{n,m}^{(o)}(\theta,\phi)\right]\right\}. \tag{5}$$

Performing similar expansions for all field quantities in terms of spherical harmonics and applying the boundary conditions at the particle's interface yields the following evolution equation for the polarization density amplitude associated with any of the electromagnetic angular momentum eigenmodes of the sphere:

$$\frac{\mathrm{d}^2\mathbf{P}_{n,m}^{(e,o)}(t)}{\mathrm{d}t^2}+\gamma\frac{\mathrm{d}\mathbf{P}_{n,m}^{(e,o)}(t)}{\mathrm{d}t}+\omega_n^2\mathbf{P}_{n,m}^{(e,o)}(t)=\omega_n^2\frac{S_{n,m}^{(e,o)}(t)}{n}. \tag{6}$$

In Eq. (6), the parameter ω_n is the resonant frequency of the eigenmodes of order n in the absence of nonlinear interactions and is given by

$$\omega_n=\sqrt{\frac{n\omega_{pl}^2}{n\varepsilon_\infty+(n+1)\varepsilon_2}}. \tag{7}$$

The term $S_{n,m}^{(e,o)}(t)$ on the right-hand side of Eq. (6) is the projection on the spherical harmonic $Y_{n,m}^{(e,o)}(\theta,\phi)$ of the nonlinear polarization density $\mathbf{P}_2^{NL}$ evaluated over the surface of the sphere:

$$S_{n,m}^{(e,o)}(t)=\oint_{r=R}Y_{n,m}^{(e,o)}(\theta,\phi)\mathbf{P}_2^{NL}(R,\theta,\phi,t)\cdot\hat{\mathbf{r}}\sin(\theta)\mathrm{d}\theta\mathrm{d}\phi. \tag{8}$$

Through Eq. (8), various eigenmodes are nonlinearly coupled to one another and to the pump field. The symmetry group of medium 2 and the spatial profile of the pump determine which specific three-wave mixing products contribute to the dynamics of a given eigenmode.

Let us consider the dynamics of the azimuthally uniform ($m=0$) resonant mode of order n in the presence of a spatially uniform time-harmonic z-polarized pump $E_P(t)$. In this case, the evolution of Eq. (6) assumes the following form:

$$\frac{\mathrm{d}^2P_{n,0}^{(e)}(t)}{\mathrm{d}t^2}+\gamma\frac{\mathrm{d}P_{n,0}^{(e)}(t)}{\mathrm{d}t}+[\omega_n^2-\alpha_1E_P(t)]P_{n,0}^{(e)}(t)=\alpha_2\left[P_{n,0}^{(e)}(t)\right]^2. \tag{9}$$

The expressions of the nonlinear interaction coefficients α_1 and α_2 are given by

$$\alpha_1=-\frac{4\sqrt{\pi}\chi_{zzz}}{n\sqrt{3}}\frac{\omega_n^4}{\omega_{pl}^2}G_{n,0}^{(e,e)};\ \ \alpha_2=\frac{\chi_{zzz}}{n\varepsilon_0}\frac{\omega_n^6}{\omega_{pl}^4}F_{n,0}^{(e,e,e)},$$

$$F_{n,0}^{(e,e,e)}=\int_0^{2\pi}\int_0^{\pi}\left\{\cos\theta\frac{\partial}{\partial z}\left[\frac{R^{n+2}}{r^{n+1}}Y_{n,0}^{(e)}(\theta,\phi)\right]_R\times\frac{\partial}{\partial z}\left[\frac{R^{n+2}}{r^{n+1}}Y_{n,0}^{(e)}(\theta,\phi)\right]_R Y_{n,0}^{(e)}(\theta,\phi)\right\}\sin\theta\mathrm{d}\theta\mathrm{d}\phi,$$

$$G_{n,0}^{(e,e)}=\int_0^{2\pi}\int_0^{\pi}\left\{\cos\theta\frac{\partial}{\partial z}\left[r\,Y_{1,0}^{(e)}(\theta,\phi)\right]_R\times\frac{\partial}{\partial z}\left[\frac{R^{n+2}}{r^{n+1}}Y_{n,0}^{(e)}(\theta,\phi)\right]_R Y_{n,0}^{(e)}(\theta,\phi)\right\}\sin\theta\mathrm{d}\theta\mathrm{d}\phi. \tag{10}$$

The PPR threshold is minimized[5] if the pump field oscillates at the second-harmonic frequency of the mode of interest. We start, therefore, by considering a spatially uniform monochromatic pump field of the form $E_P(t) = A_p \sin(2\omega_n t)$. In solving Eq. (9), we notice that so long as the condition $\left|P_{n,m}^{(e,o)}(t)\right| \ll \alpha_1 A_p/\alpha_2$ holds, which is the case at the initial stages of the parametric interaction, a solution can be easily obtained in terms of Mathieu functions[7,8]. More intuitive though is the following slowly varying envelope approximate solution:

$$P_{n,m}^{(e,o)}(t) = p(t)\cos[\omega_n t - \theta(t)]e^{-\frac{\gamma}{2}t},$$
$$p(t) = p_0\sqrt{\cosh\left(\frac{\alpha_1 A_p}{2\omega_n}t\right)};\ \theta(t) = \operatorname{arccot}\left[\exp\left(-\frac{\alpha_1 A_p}{2\omega_n}t\right)\right], \tag{11}$$

where p_0 is the initial modal amplitude. From Eq. (11), for $p(t)$, it is clear that the system enters the PPR regime provided that the pump electric-field amplitude A_p exceeds the threshold value $A_{PPR} = 2\gamma\omega_n/\alpha_1$.

It is worth pointing out a unique property of plasmonic parametric gain that emerges from the analysis above: a plasmonic mode of any order (m, n) can undergo PPR and be amplified by a spatially uniform modulation of the background permittivity, provided that the corresponding threshold is exceeded. This is in contrast with conventional LSP resonances, which, for a mode of order (m, n), requires a driving field with a matching spatial profile—a condition almost impossible to realize in practice for high-order plasmonic modes of deeply subwavelength particles. For these reasons, PPR is uniquely suitable to access plasmonic resonances of arbitrarily high order in deeply subwavelength structures.

The unique characteristics of PPR lend themselves to interesting applications in optical limiting. Recently, a new class of nonlinear absorbers termed plasmonic parametric absorbers (PPAs) has been proposed[9]. The key insight informing the PPA idea is that in the PPR process the pump field experiences an absorption rate that strongly depends on the intensity of the pump itself, creating two distinct regimes: one of weak absorption under low intensity illumination and one of strong absorption when the threshold of parametric resonance is reached or exceeded. Such a threshold condition separates distinct dynamics, so that $P_{n,m}^{(e,o)}(t)$ decreases exponentially for $A_p < A_{PPR}$ and increases exponentially for $A_p > A_{PPR}$. Such contrasting modal dynamics are reflected in the distinct absorption regimes that the pump is subjected to. As shown in Ref. [9], the power parametrically transferred from the pump to the resonant mode is given by

$$W_{abs}(t) = \frac{nR^3\alpha_1 A_p p_0^2}{32\varepsilon_0\omega_{pl}^2}\left[2\omega_n + \gamma\sinh\left(\frac{\alpha_1 A_p t}{2\omega_n}\right)\right]e^{-\gamma t}$$
$$\sim \frac{nR^3\alpha_1 A_p p_0^2\gamma}{64\varepsilon_0\omega_{pl}^2}\exp\left[(A_p - A_{PPR})\frac{\alpha_1 t}{2\omega_n}\right]. \tag{12}$$

Equation (12) highlights the fundamental trait of PPA, which is in stark contrast with linear absorption: in PPA, absorption is vanishingly small for incident fields below the PPR threshold and increases exponentially under high-intensity conditions.

Clearly, a saturation of the exponential behavior is expected, because, if nothing else, the absorbed power of Eq. (12) cannot exceed the finite power carried by the pump. In reality, a different mechanism limits Eq. (12) before pump depletion occurs. Such a mechanism is the resonance detuning due to additional three-wave mixing processes in Eq. (9) that we have neglected so far. As $\left|P_{n,m}^{(e,o)}(t)\right| \sim \alpha_1 A_p/\alpha_2$, Eq. (9) can only be integrated numerically. Nevertheless, the following asymptotic expressions as $t \to \infty$ for $P_{n,m}^{(e,o)}(t)$ hold for $A_p > A_{PPR}$:

$$P_{n,m}^{(e,o)}(t) \sim -\frac{\alpha_2 Q_1^2}{2\omega_n^2} + Q_1\cos(\omega_n t + \theta_1) + \frac{\alpha_2 Q_1^2}{6\omega_n^2}\cos(2\omega_n t + 2\theta_1),$$
$$\theta_1 = \frac{1}{2}\arccos\left(-\frac{A_{PPR}}{A_p}\right);\ Q_1 = \frac{\omega_n}{\alpha_2}\sqrt{\frac{6\gamma\omega_n}{5}\sqrt{\left(\frac{A_p}{A_{PPR}}\right)^2 - 1}}. \tag{13}$$

Within the range of validity of Eq. (13), the exponentially growing oscillations of the polarization density amplitude level off as $t \to \infty$ after a sequence of relaxation oscillations. In Fig. 4, the numerical solution of Eq. (9) (indicated in blue) is compared with the predictions of the asymptotic model in Eq. (13), shown in orange.

Based on Eq. (13), the average power $\bar{W}_P$ transferred from the pump to the plasmonic mode asymptotically approaches the value

$$\bar{W}_{abs}(t \to \infty) = \frac{3}{20}nR^3\frac{\gamma}{\varepsilon_0\omega_{pl}^2}\frac{\omega_n^3}{\alpha_2^2}\sqrt{\left(\frac{A_p}{A_{PPR}}\right)^2 - 1}. \tag{14}$$

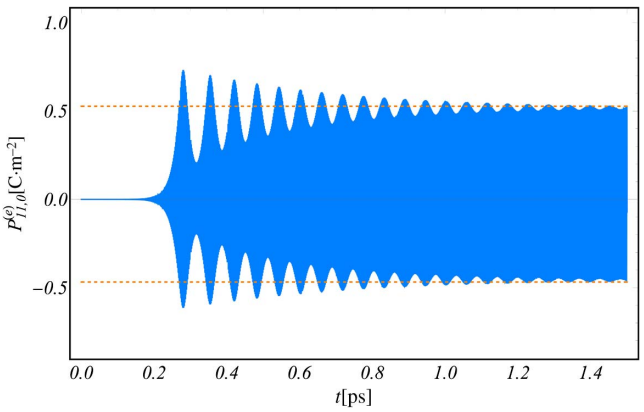

Fig. 4. Polarization density amplitude term $P_{11,0}^{(e)}$ of a silver sphere immersed in an MNA background medium. In order to better highlight the relaxation oscillations occurring in the system, we show a case in which the PPR threshold is exceeded by a large margin ($A_p = 20A_{PPR}$). The dashed lines show the oscillation limits predicted by the asymptotic Eq. (12).

Using the steady-state asymptotic estimate of Eq. (14) of the absorbed power, it is possible to obtain the PPR contribution to the particle absorption cross-section (in addition to the linear portion):

$$\sigma_{NL}=\frac{3nR^3}{40\varepsilon_0\sqrt{\varepsilon_2}}\frac{\omega_n^3}{\omega_{pl}^2}\frac{\alpha_1^2}{\alpha_2^2}\sqrt{\frac{I_{PPR}}{I_p}\left(1-\frac{I_{PPR}}{I_p}\right)},\quad I_p>I_{PPR}. \quad (15)$$

In Eq. (15), $I_p=A_p^2/(2\eta)$ is the incident intensity, and $I_{PPR}=A_{PPR}^2/(2\eta)$ is the PPR intensity threshold, where η is the intrinsic impedance of the background medium. If particles similar to the one described so far are dispersed with density N in the background medium, the nonlinear pump attenuation coefficient of the composite follows from Eq. (15) as $\alpha_{NL}=N\sigma_{NL}$.

Figure 5 shows how the normalized absorption cross-section of a silver sphere of radius $R=100\,\text{nm}$ in a 2-methyl-4-nitroanline (MNA)[10,11] host is affected by various modes undergoing PPR. The absorption cross-section is plotted against the normalized pump intensity, and, for each of the PPR modes considered in Fig. 5, the incident pump field is at twice the value of the corresponding resonant frequency given by Eq. (7). For the case at hand, all the possible PPR resonant wavelengths λ_n fall in the range $\lambda_\infty<\lambda_n\le\lambda_1$, where $\lambda_\infty=563\,\text{nm}$ and $\lambda_1=448\,\text{nm}$. As evident from Fig. 5, as soon as a pump field of frequency $2\omega_n$ exceeds the intensity threshold I_{PPR}, the particle's absorption cross-section increases dramatically due to the contribution of the mode of eigenfrequency ω_n undergoing PPR. This phenomenon is a form of reverse saturable absorption and could have interesting applications in optical limiting devices[12], especially given the design versatility of metallic nanoparticles for targeting different spectral regions.

For the purpose of illustration, in Fig. 6, we apply the analysis and the models described thus far to the practically relevant case of a pulsed pump. The pulse considered here is a 30 ps pulse of average power 0.5 W, focused to an

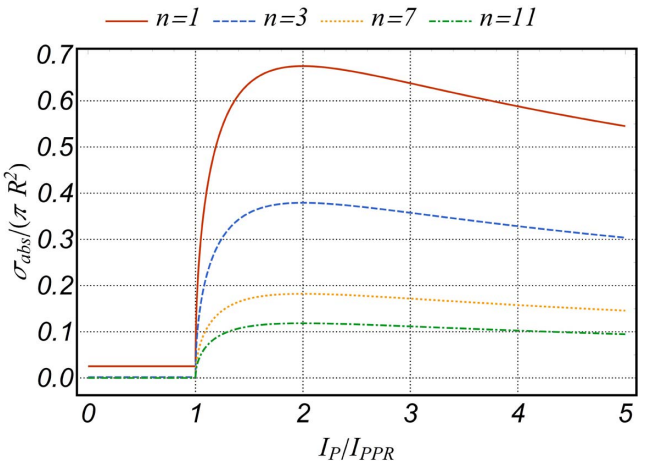

Fig. 5. Absorption cross-section of the plasmonic particle normalized to the geometrical cross-section as a function of the incident intensity.

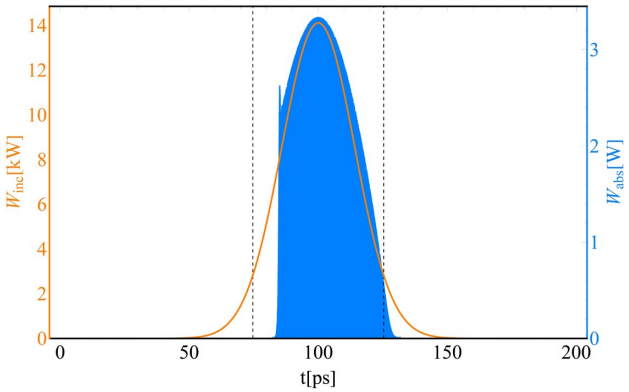

Fig. 6. Orange curve shows the instantaneous pump power W_{inc}. The blue curve shows the instantaneous pump power W_{abs} absorbed by the particle via PPR of the $n=11$, $m=0$ mode. The dashed vertical lines indicate the times at which the pump intensity is equal to the PPR threshold.

area of $25\,\mu\text{m}^2$, on parametric resonance with the $n=11$, $m=0$ mode ($\lambda_{11}=460\,\text{nm}$) of a silver particle of radius 100 nm embedded in an MNA background medium. The orange curve in Fig. 6 shows the total instantaneous power of the pump field. The blue curve shows the instantaneous absorbed power caused by the PPR process. The vertical dashed lines show the time interval in which the pump field exceeds the PPR threshold. A similar behavior is observed both in Figs. 4 and 6 at the onset of PPR, where the modal polarization (and the corresponding absorption) builds up exponentially to slightly surpass the steady-state value and then relaxes to such a value through a series of oscillations (only one is discernible in Fig. 6). Figure 6 confirms the dramatic increase in absorption that PPAs exhibit under high-intensity illumination.

In conclusion, we have illustrated the principles of the operation of PPR. Unlike conventional LSP resonances, all of the plasmonic modes of a nanostructure, including the strongly sub-radiant ones, can be resonantly excited by spatially uniform optical pumping, provided that the corresponding threshold is exceeded. Accessing such a high density of strongly localized states holds promise for enhancing nonlinear light–matter interaction at the nanoscale for the development of nonlinear optical metamaterials and for optical limiting applications. In the context of PPR, we have discussed the closely related theory of PPAs. PPAs exhibit a reverse saturable absorption behavior whereby an incident field that is parametrically resonant with one or more of the modes of a plasmonic particle experiences a strongly enhanced absorption whenever its intensity exceeds the relevant PPR threshold. Such effect makes PPAs very promising candidates for optical limiting applications, in addition to being of fundamental interest in the emerging field of nonlinear plasmonics.

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