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Time-varying metamaterials: dynamic transformation optics and parametric phenomena

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14. ABSTRACT
This research program explored the following aspects of the electromagnetic interaction with time-varying media:
1) The physical processes suitable to dynamically induce large permittivity changes in specifically selected regions of space. This research thrust led to the formulation of the theory of a new class of surface waves termed Accumulation-Layer Surface Plasmons (ASP).
2) The resonant eigenmodes of metallic nanostructures embedded in media with time-varying permittivity, and the conditions of parametric regeneration and parametric resonance. This research thrust led to the development of the theory of Plasmonic Parametric Resonance (PPR).
3) Optical limiting effects relying on plasmonic parametric resonance. In this research thrust a new class nonlinear saturable absorber termed Plasmonic Parametric Absorbers (PPA) was introduced and theoretically modeled.
4) The study of tunable extreme anisotropy. In this context we proposed a variety of electromagnetic and optoelectronic devices relying on metamaterials with tunable hyperbolic dispersion.

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Principal Investigator: **Alessandro Salandrino**

Reporting Period: **June 15, 2016 to June 14, 2019**

Abstract

In this research program we focused our investigation on the following aspects of the electromagnetic interaction with time-varying media:

1) The physical processes suitable to dynamically induce large permittivity changes in specifically selected regions of space.

2) The resonant eigenmodes of metallic nanostructures embedded in media with time-varying permittivity, and the conditions of parametric regeneration and parametric resonance.

3) Optical limiting effects relying on plasmonic parametric resonance.

4) The study of tunable extreme anisotropy.

In the context of our research on permittivity modulation processes we formulated an analytical description of the electromagnetic eigenmodes supported by electrically induced charge accumulation layers at the surface of transparent conductive oxides. The new class of surface plasmons termed **Accumulation-Layer Surface Plasmon (ASP)** was introduced. Near resonance ASPs display a vast effective index tunability that could be employed for the realization of reconfigurable waveguides, of transmission-line-based time-varying metamaterials, as well as of optoelectronic devices such as ultra-compact phase modulators. The suppression of ASP in the presence of epsilon-near-zero (ENZ) region was investigated. The theory of **modal-dichroism** in spatially-patterned accumulation layers was also developed, which is concerned with mode-orthogonality under a spatially non-uniform permittivity modulation.

In the context of our research on parametric interactions in nanostructures embedded in time-varying media we developed the theory of **Plasmonic Parametric Resonance (PPR)**. Our analysis showed that losses of localized surface plasmons in nanoparticles can be effectively compensated exploiting the temporal modulation of the permittivity of the host medium. Unlike in conventional localized surface plasmon resonances, all the plasmonic modes of a nanostructure, including the strongly subradiant ones, can be resonantly excited by spatially uniform optical pumping, provided that the corresponding threshold is exceeded. Accessing such high density of strongly localized states holds promise for enhancing nonlinear light-matter interaction at the nanoscale, and for the development of nonlinear optical metamaterials, as well as for optical-limiting applications.

Based on our PPR theory, we have explored its use in optical limiting applications. In this context we have developed the theory of **Plasmonic Parametric Absorbers**. Our theory indicates that plasmonic elements embedded in a host medium endowed with a second order optical nonlinearity can lead to a form of reverse saturable absorption, whereby the composite shows high transparency under low intensity illumination and becomes highly absorptive when the incident intensity exceeds a certain threshold. These findings show that the proposed system could be employed for the protection of devices or operators from accidental or intentional exposure to high intensity radiation.

Finally, we explored the possibility of electrically **tuning the anisotropy** of a composite medium. In this context we proposed a variety of optoelectronic devices relying on metamaterials with tunable hyperbolic dispersion.

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I. Introduction

This is the final performance report on the AFOSR 2016 Young Investigator Program, "Time-varying metamaterials: dynamic transformation optics and parametric phenomena", being carried out at the University of Kansas. The program was initiated in June 2016. The present report covers the period June 15, 2016 to June 14, 2019.

The overarching purpose of this research program is to formulate the theory of spatiotemporally modulated metamaterials, and to develop the design principles of devices relying on the unique properties of this novel class of metamaterials, with an emphasis on parametric schemes for loss compensation.

Our research progress is summarized in section II of this report. A list of the publications related to this effort is presented is section III.

II. Research Accomplishments

In this research program we focused our investigation on the following aspects of the electromagnetic interaction with time-varying media:

1. The study of physical processes suitable to dynamically induce large permittivity changes in specifically selected regions of space, and the analysis of the electromagnetic modes supported by such configurations. Our progress in this area is summarized in section II.1 of this report.

2. The study of the resonant eigenmodes of metallic nanostructures embedded in media with time-varying permittivity, with an emphasis on the conditions of parametric regeneration and parametric resonance. Our progress in this area is summarized in section II.2 of this report.

3. The study of optical limiting effects relying on plasmonic parametric resonance. Our progress in this area is summarized in section II.3 of this report.

4. The study of tunable extreme anisotropy in device applications. Our progress in this area is summarized in section II.4 of this report.

II.1 Electromagnetic and electronic properties of charge-accumulation layers

The realization of time-varying metamaterials requires physical mechanisms to dynamically modify the dielectric response of the media in which electromagnetic propagation and interactions take place. Modulating the free-carrier density in degenerately doped semiconductors is a particularly attractive strategy since very large changes of local permittivity can be obtained by electric bias. Such permittivity profiles depend on the free-carrier density at thermodynamic equilibrium within the regions of charge accumulation, or "accumulation layers". We formulated an analytical description of the electromagnetic eigenmodes supported by an accumulation layer in a planar structure. The new class of surface plasmons termed **Accumulation-Layer Surface Plasmon (ASP)** was introduced. Near resonance ASPs display a vast effective index tunability that could be employed for the realization of reconfigurable waveguides, of transmission-line-based time-varying metamaterials, as well as of optoelectronic devices such as ultra-compact phase modulators. The suppression of ASP in the presence of epsilon-near-zero (ENZ) region was investigated. The theory of **modal-dichroism** in spatially-patterned accumulation layers was also developed, which is concerned with mode-orthogonality under a spatially non-uniform permittivity modulation.

The prototype system studied is shown schematically in fig. 1, along with an example of electron density profile and the corresponding electric potential. The structure under consideration is the interface between an insulator and a transparent conductive oxide, such as Indium Tin Oxide (ITO), with an applied static potential that causes electrons to accumulate in proximity of the surface. The accumulation layer charge density profile was obtained by solving the Poisson's equation along with the Boltzmann's relations for the carrier density under thermodynamic equilibrium.

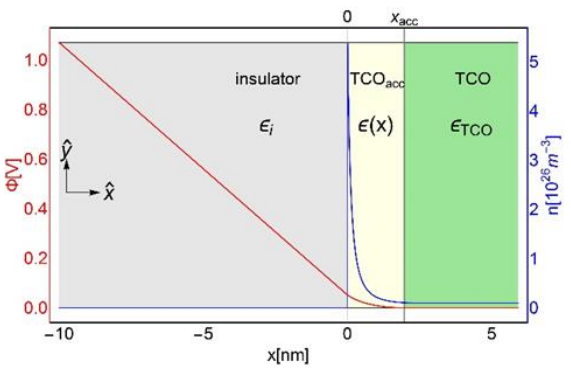

Figure 1 Electrostatic potential and charge density plotted as a function of position over the layout of a 1D Silicon-dioxide/Indium Tin oxide interface.

The unique characteristic of such system is that depending on the doping concentration and on the applied electrical bias three qualitatively distinct types of permittivity profile can be obtained at a given frequency, as shown in fig. 2:

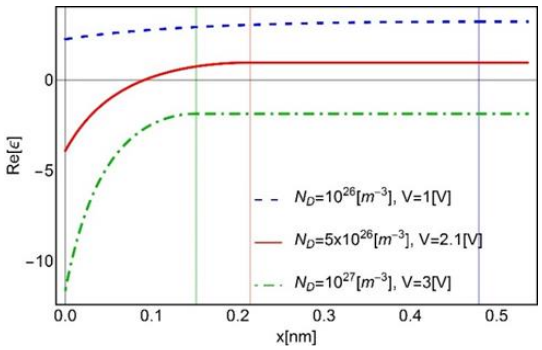

Figure 2 Permittivity profile within and around the accumulation layer in an ITO film with three different doping concentrations.

1. The dashed blue curve shows that below a certain doping concentration the ITO response is dielectric, with a graded positive permittivity.
2. The dash-dotted green curve shows that with a sufficiently high doping concentration the ITO response is plasmonic, with a graded permittivity profile that stays negative also outside of the accumulation layer.
3. The red solid curve shows an intermediate situation in which the ITO response is plasmonic, with a graded negative permittivity, within a portion of the accumulation layer, and dielectric everywhere else, with a graded positive permittivity.

A perturbation analysis of the electromagnetic properties of this system led us to analytical expressions for the mode profiles and for the dispersion relations of ASP in terms of doping concentration, applied electric bias, and temperature. An example of field profile of an ASP and the corresponding dispersion relation are shown in figure 3.

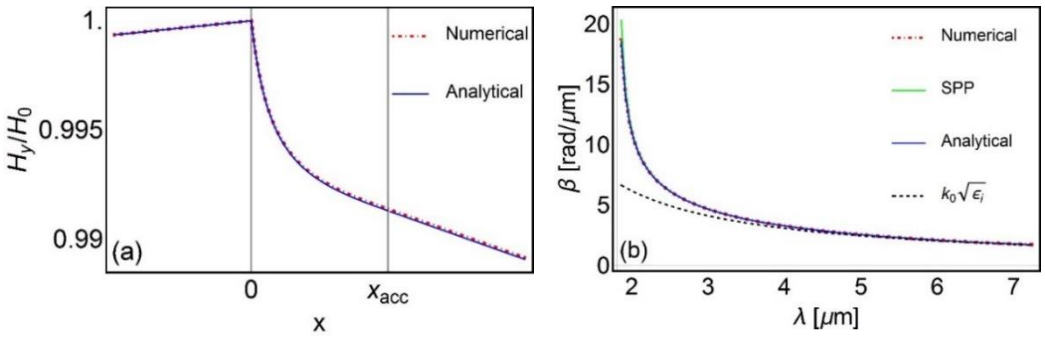

Figure 3 (a) Magnetic field profile and (b) dispersion of an ASP at SiO2-ITO interface.

ASPs have several remarkable properties that are here summarized:

1. The field profile within the charge accumulation layer significantly departs from the exponential decay observed in surface plasmon polaritons supported by planar metallic interfaces.
2. ASPs are supported if and only if the TCO permittivity remains negative outside the accumulation layer. ASPs are suppressed in the presence of epsilon-near-zero (ENZ) regions in the permittivity profile.
3. A resonance condition exists for ASPs, and it is dictated by the TCO permittivity outside the accumulation layer. Away from resonance the dispersion relation of ASPs is approximately equal to the dispersion relation of the surface plasmon polaritons that would exist in the absence of an accumulation layer. On resonance, a very large perpendicular electric field is developed at the interface and both the effective index and the propagation length become highly sensitive to the applied electric bias (fig. 4).

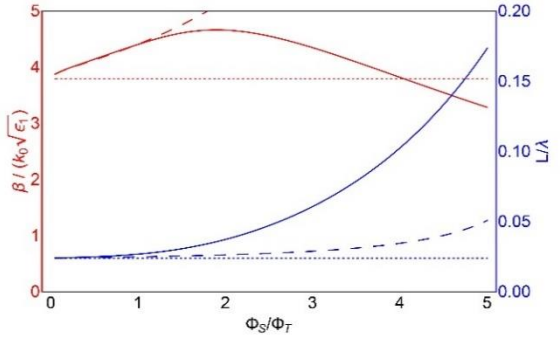

Figure 4. The solid curves represent the exact normalized propagation constant and propagation length of an ASP at a SiO2-ITO interface the near resonant wavelength for different induced surface potentials. The dashed curves represent the approximate perturbative solution.

The theory of **modal-dichroism** in spatially-patterned accumulation layers was also developed, which is concerned with mode-orthogonality under a non-uniform permittivity modulation. The unique electromagnetic characteristics of electrically-induced accumulation layers make them ideal building blocks for the realization of reconfigurable and time-varying metamaterials.

II.2 Parametric excitation and amplification of surface plasmons

In the context of our research on parametric interactions in nanostructures embedded in time-varying media we developed the theory of **Plasmonic Parametric Resonance (PPR)**. Our analysis showed that the absorption and radiation losses in localized surface plasmons in nanoparticles can be effectively compensated exploiting the temporal modulation of the permittivity of the host medium. Our findings hold promise for the compensation of losses in optical metamaterials using plasmonic nanoparticles as building blocks. Our results led to two invited conference presentations [C1] and [C2]. A manuscript [J3] is currently in preparation.

Plasmonic particles, in general, support an infinite discrete set of plasmonic resonances. In the simple case of a plasmonic nanosphere (figure 5) with frequency dispersive permittivity $\varepsilon_1(\omega)$ embedded in a background medium with constant permittivity ε_2, for a resonance of order $n \geq 1$ there are $2n+1$ degenerate angular-momentum states with complex frequency ω_n satisfying the condition $\varepsilon_1(\omega_n) = -(1+n)\varepsilon_2 / n$. For $n >> 1$ the eigenmodes tend to occur for $\varepsilon_1\left(\omega_{n>>1}\right) \sim -\varepsilon_2$. The increased modal density for $\varepsilon_1 \sim -\varepsilon_2$ is not specific to spherical particles but is rather a general feature of all plasmonic structures. The occurrence of such increased modal density in plasmonic particles of different shapes for $\varepsilon_1 \sim -\varepsilon_2$ is a consequence of the fact that the plasmonic mode becomes essentially unaffected by a finite curvature of the metal-dielectric interface supporting it. Accessing such spectrally dense set of

tightly bound resonant modes would greatly enhance nonlinear light-matter interactions at the nanoscale and offer new avenues to design nonlinear optical metamaterials.

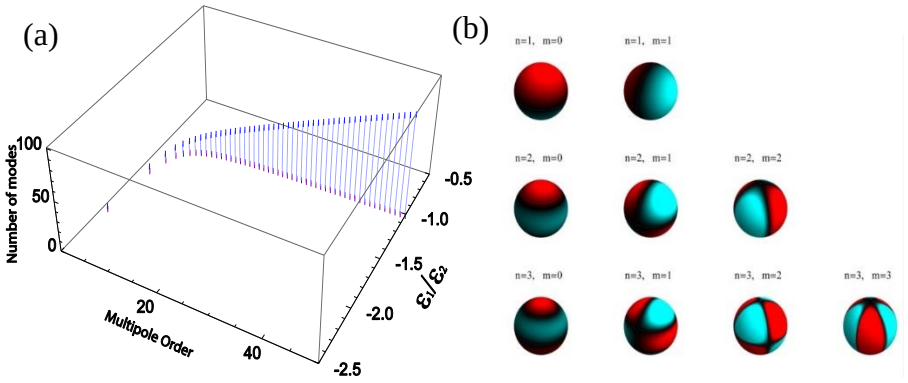

Figure 5. a) Spectral distributions of the resonant modes of a plasmonic nanosphere. b) Surface polarization charge density for the first few modes of a plasmonic nanosphere.

The efficiency with which such 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 electric dipolar nature is efficiently coupled to, and excited by, radiation states. The higher order eigenmodes tend to be subradiant, and by reciprocity they are nearly decoupled from free-space propagating fields. Therefore, exciting and detecting such higher order modes requires sophisticated near-field scattering techniques, or the use of active media to promote surface plasmon amplification by stimulated emission of radiation (SPASER). We investigated an alternative approach to deliver energy to the free-carriers of a plasmonic nanoparticle by introducing a different form of LSPR: the plasmonic parametric resonance (PPR). In analogy with parametric resonance in dynamical systems, PPR originates from the temporal modulation of one of the parameters governing the evolution of the state of the system.

Considering a metallic nanosphere embedded in a uniform medium, given an initial state of excitation of a resonant eigenmode, the polarization density within the structure evolves according to a damped harmonic oscillator equation, alternating states of high potential energy and high kinetic energy of the polarization charges, as shown in figure 6 for an octupolar mode. Since the natural frequency of oscillation of the eigenmodes depends on the permittivity of the particle and of the background medium, any change in such properties would result in a change in the modal frequency, which in turn would be accompanied by a change in the energy content of the system. Such change in energy depends on the instantaneous partition between kinetic and potential energy in the mode of interest at the instant of time when the permittivity modulation occurs. In particular our analysis indicated that:

1. An instantaneous background permittivity change that increases (decreases) the eigenmode frequency Ω_n results in an increase (decrease) in the energy W_n of the mode.
2. The amount of energy delivered to (or extracted from) the mode is proportional to the potential $U_n(t)$ energy of the polarization charges.
3. An instantaneous change in the permittivity of the background medium occurring when the system is in a state of pure kinetic energy $K_n(t)$ does not change the energy of the eigenmodes.

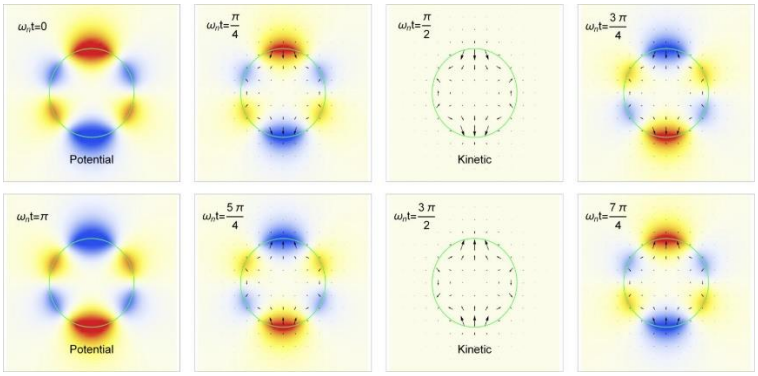

Figure 6. Temporal evolution of the potential and polarization currents of the octupolar mode of a plasmonic nanosphere

Based on these observations a suitably timed periodic modulation of the background permittivity can be devised to amplify the oscillation of a plasmonic eigenmode. The idealized stepwise modulation cycle shown in figure 7 was considered in our analysis in order determine a lower bound for the parametric regeneration threshold in terms of the eigenmode frequency change $d\Omega_n$, which can be simply expressed in terms of the resonance quality factor Q_n as: $d\Omega_n / \Omega_n = \pi / (2Q_n)$. An example of the evolution of the energy in the system is shown in figure 8(a) under perfect synchronization between modal oscillations and parametric modulation. In contrast figure 8(b) shows the case in which the initial phase between modal oscillation and parametric modulation is unfavorable to promote energy transfer to the system; yet when the parametric regeneration threshold is exceeded the modal oscillation spontaneously synchronizes to the pump and grows in amplitude.

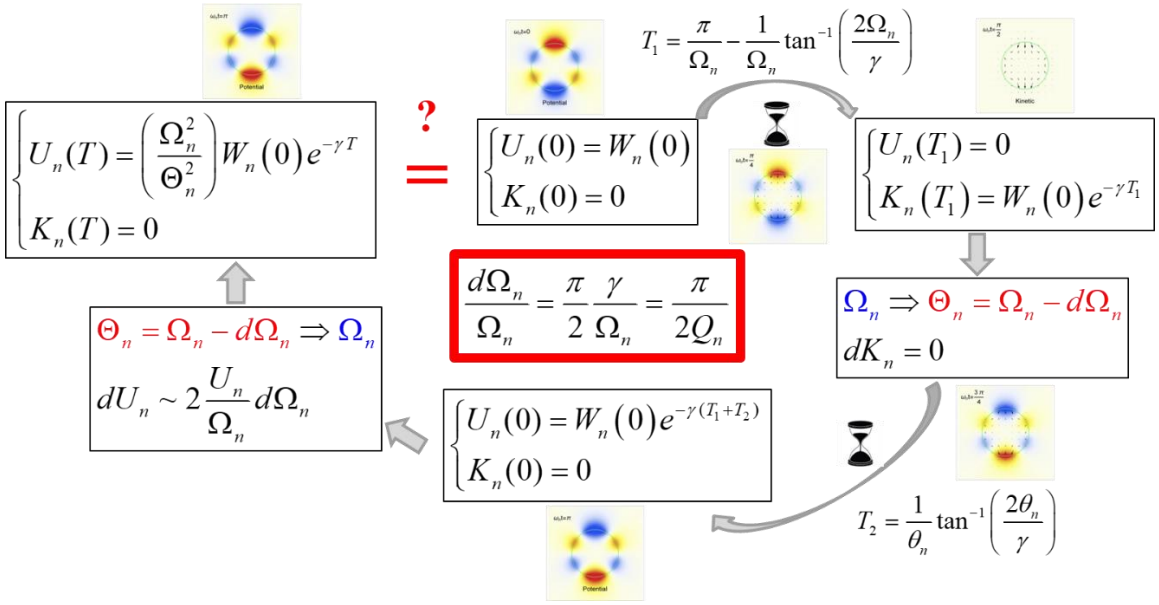

Figure 7. One parametric modulation cycle.

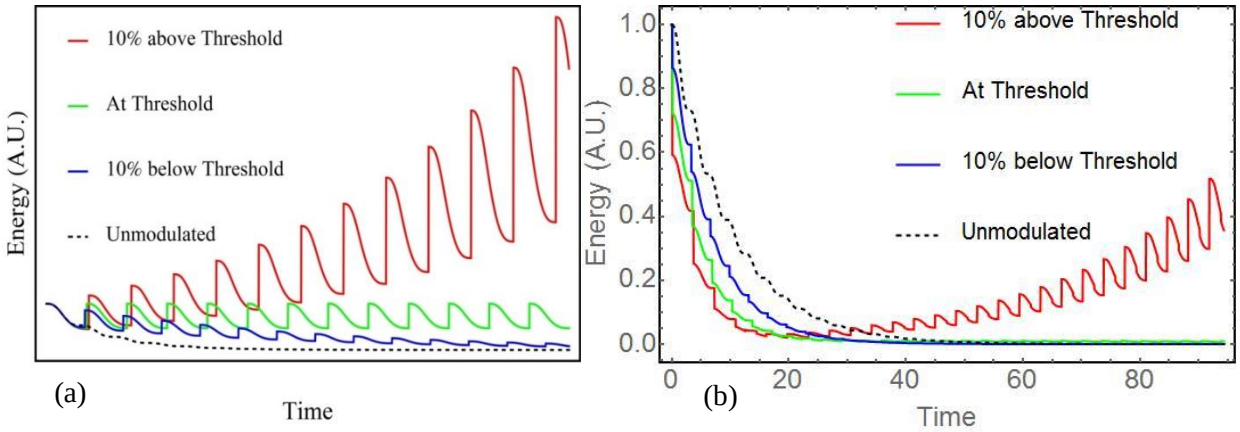

Figure 8. Evolution of the energy in a parametrically pumped resonant mode under (a) perfect and (b)unfavorable initial conditions.

We also analyzed the more realistic scenario of a harmonic modulation of the background permittivity mediated by second order nonlinearity. The analysis in that case is substantially more complicated due to the inherent anisotropy of the second order susceptibility, but yields qualitatively similar results. Although many of the dynamics that we described, such as the existence of a threshold and the tendency to self-synchronize to the pump, are shared by all parametrically resonant systems, a few unique features of both fundamental and practical relevance emerged from our analysis of plasmonic parametric resonance. In particular:

1. Sub-radiant resonant modes of arbitrarily high order can be amplified by a **spatially uniform pump.** This is in stark contrast with the mechanism of direct excitation of high-order mode, which would require the pump spatial profile to match the modal field distribution.
2. The frequency of the pump field responsible for the background permittivity modulation allows for the targeted excitation of specific subsets of angular-momentum-degenerate plasmonic modes.
3. Depending on the anisotropy of the second order susceptibility, a set of **selection rules** exist that can be exploited to transfer energy to one specific mode among those already selected by the pump frequency.
4. When the PPR threshold is exceeded the pump effectively experiences reverse-saturable-absorption – a feature of great interest for optical limiting applications.

In conclusion, we have introduced the concept of Plasmonic Parametric Resonance. Unlike in conventional localized surface plasmon resonance, all the plasmonic modes of a nanostructure, including the strongly subradiant ones, can be resonantly excited by spatially uniform optical pumping, provided that the corresponding threshold is exceeded. Accessing such high density of strongly localized states holds promise for enhancing nonlinear light-matter interaction at the nanoscale, and for the development of nonlinear optical metamaterials, as well as for optical-limiting applications.

II.3 Plasmonic Parametric Absorbers

The PPR theory that we have developed hints at some interesting possibility to use nonlinear plasmonic composites for optical limiting applications, which are of great relevance to the AFOSR mission. In this context we developed the theory of Plasmonic Parametric Absorbers.

Absorption, which is arguably the Achilles' heel of most metamaterials applications, in the case of absorbers is a desired effect that can be accurately tailored exploiting the properties of nanostructured

media. Since the seminal work of Landy et Al.[1] a great deal of attention has been devoted to the design of metamaterial absorbers [2] operating in a variety of spectral regions, from the terahertz range [3] up to the optical domain [4]. The degrees of freedom afforded by metamaterial structures have led to the realization of absorbers with unprecedented characteristics in terms of efficiency, spectral and/or angular bandwidth [5-8] or selectivity [9, 10]. Nonlinearity can greatly enrich the optical absorption phenomenology, and a host of applications can be envisioned exploiting the unique properties of nonlinear metamaterial absorbers, ranging from all-optical modulation schemes to optical-limiting devices.

Here we present the theory of a new class of nonlinear absorbers termed Plasmonic Parametric Absorbers (PPA) relying on the recently introduced concept of Plasmonic Parametric Resonance (PPR) [11]. In contrast with conventional localized plasmonic resonances, whereby 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 fashion. In PPR in fact the modes of a plasmonic structure are amplified by means of a temporal permittivity modulation of the background medium interacting with an appropriate pump field. Such permittivity variation translates into a modulation of the modal resonant frequency, and under specific conditions amplification can occur. As shown in [11], 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. As we will show in the following, it is such threshold behavior that can lead to a type of nonlinear metamaterial absorber with rather unique properties.

The key insight informing the PPA idea is that in the PPR process the pump field experiences an extinction 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 met or exceeded. The characterization of the pump field extinction is the main focus of our theoretical analysis. In addition, the theory of PPR is extended here to predict and quantify the effects of saturation dynamics in the high-intensity/high-absorption regime.

The system under consideration is a composite medium of identical subwavelength plasmonic particles dispersed in a dielectric host material. An electric field $\mathbf{E}_P(t)$, which will be referred to as "pump", is incident on the system. It is assumed that the electromagnetic response of the medium constituting the particles (henceforth referred to as "medium 1") is dominated by free-carriers effects. Under such assumption the dispersive component of the polarization density $\mathbf{P}_1(\mathbf{r},t)$ in medium 1 evolves in time according to the following differential equation:

$$\frac{\partial^2 \mathbf{P}_1(\mathbf{r},t)}{\partial t^2} + \gamma \frac{\partial \mathbf{P}_1(\mathbf{r},t)}{\partial t} = \varepsilon_0 \omega_{pl}^2 \mathbf{E}_1(\mathbf{r},t) \ , \qquad (1)$$

where $\mathbf{E}_1(\mathbf{r},t)$ is the electric field within medium 1. It is worth noting that equation (1) is equivalent to assigning to medium 1 a Drude-like permittivity with plasma frequency ω_{pl} and collision frequency γ. For completeness a nondispersive permittivity term ε_∞ will be also included to account for higher frequency spectral features of the dielectric response of medium 1. The background medium, henceforth "medium 2", is assumed non-dispersive with relative permittivity ε_2 and endowed with second order nonlinearity $\boldsymbol{\chi}^{(2)}$. 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:

$$\mathbf{P}_2(\mathbf{r},t) = \varepsilon_0(\varepsilon_2 - 1)\mathbf{E}_2(\mathbf{r},t) + \mathbf{P}_2^{NL}(\mathbf{r},t) \qquad (2)$$

In equation (2) $\mathbf{P}_2^{NL}(\mathbf{r},t) = \varepsilon_0 \boldsymbol{\chi}^{(2)} \cdot \mathbf{E}_2(\mathbf{r},t)\mathbf{E}_2(\mathbf{r},t)$ is the nonlinear polarization density due to the quadratic nonlinearity of medium 2.

For the purpose of illustration we shall consider subwavelength plasmonic spherical particles of radius R randomly distributed in a dielectric host possessing a second order optical nonlinearity with a dominant term $\chi_{zzz}^{(2)}$. This choice is merely for mathematical convenience, as it is amenable to an analytical treatment. A completely analogous formulation and qualitatively similar results would hold for different particles ensembles and different host media. The specific system analyzed to produce the results reported in Figures 1, 2 and 3 is a Silver sphere of radius $R = 100nm$ embedded in a 2-methyl-4-nitroanline (MNA) host medium, which is characterized by a dominant nonlinear susceptibility term $\chi_{zzz}^{(2)} = 500\,pm/V$ [12, 13].

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 [11]:

$$\mathbf{P}_1(t) = \sum_{n,m} \nabla \left\{ \frac{r^n}{R^{n-1}} \left[P_{n,m}^{(e)}(t) Y_{n,m}^{(e)}(\theta,\phi) + P_{n,m}^{(o)}(t) Y_{n,m}^{(o)}(\theta,\phi) \right] \right\} \qquad (3)$$

Performing similar expansions for all field quantities in terms of spherical harmonics and applying the appropriate 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:

$$\frac{d^2 P_{n,m}^{(e,o)}(t)}{dt^2} + \gamma \frac{dP_{n,m}^{(e,o)}(t)}{dt} + \omega_n^2 P_{n,m}^{(e,o)}(t) = \omega_n^2 \frac{S_{n,m}^{(e,o)}(t)}{n} \qquad (4)$$

In equation(4), the parameter ω_n is the resonant frequency of the eigenmodes of order n in the absence of nonlinear interactions and is given by:

$$\omega_n = \sqrt{\frac{n\omega_{pl}^2}{n\varepsilon_\infty + (n+1)\varepsilon_2}} \qquad (5)$$

The term $S_{n,m}^{(e,o)}(t)$ in the right-hand side of equation (4) 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, i.e.:

$$S_{n,m}^{(e,o)}(t) = \oint_{r=R} Y_{n,m}^{(e,o)}(\theta,\phi)\mathbf{P}_2^{(NL)}(R,\theta,\phi,t) \cdot \hat{\mathbf{r}} \sin(\theta) d\theta d\phi \qquad (6)$$

As a consequence of the term (6) various eigenmodes are nonlinearly coupled to each other and to the pump field. The symmetry group of medium 2, along with the spatial profile of the pump, determine which specific three-wave mixing products contribute to the dynamics of a given eigenmode.

Proceeding by way of example, we consider the dynamics of the azimuthally uniform ($m=0$) resonant mode of order n in the presence of a spatially uniform pump. In this instance the evolution equation (4) assumes the following form:

$$\frac{d^2P_{n,0}^{(e)}(t)}{dt^2}+\gamma\frac{dP_{n,0}^{(e)}(t)}{dt}+\omega_n^2P_{n,0}^{(e)}(t)=\alpha_1E_P(t)P_{n,0}^{(e)}(t)+\alpha_2\left[P_{n,0}^{(e)}(t)\right]^2 \quad (7)$$

The first term in the right hand side of equation (7) represents the three-wave mixing process responsible for the parametric interaction of the resonant mode with the pump. The second term in the right hand side of equation (7) accounts the sum and difference frequency generation processes due to the plasmonic mode itself interacting with the background medium. The expressions of the nonlinear interaction coefficients α_1 and α_2 are given by:

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

$$F_{n,0}^{(e,e,e)}=\int_0^{2\pi}\int_0^{\pi}\left\{\cos\theta\frac{\partial}{\partial z}\left[\frac{R^{n+2}}{r^{n+1}}Y_{n,0}^{(e)}(\theta,\phi)\right]_R\frac{\partial}{\partial z}\left[\frac{R^{n+2}}{r^{n+1}}Y_{n,0}^{(e)}(\theta,\phi)\right]_R Y_{n,0}^{(e)}(\theta,\phi)\right\}\sin\theta\,d\theta d\phi \quad (8)$$

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

The PPR threshold is minimized [11] if the system is driven at the second harmonic frequency of the mode of interest. We start therefore by considering a spatially uniform monochromatic pump field of the form $E_P(t)=A_p\sin(2\omega_n t)$. Under these conditions the linear interaction of the pump field with the subwavelength particle is essentially of electric dipolar origin. Importantly the pump field at $2\omega_n$ in the case at hand is strongly detuned from any resonant mode, including the dipolar one occurring at ω_1, so a relatively small pump absorption is expected. The linear absorption cross-section of a subwavelength of volume V at the pump frequency $2\omega_n$, in the absence of any nonlinear process (i.e. for $\alpha_1,\alpha_2=0$) can be written as follows:

$$\sigma_{2\omega_n}=\frac{36V\varepsilon_0\eta\left(\omega_n^2-\omega_1^2\right)^2\omega_{pl}^2\gamma}{(n-1)^2\left(4\omega_n^2-\omega_1^2\right)^2\omega_n^2} \quad (9)$$

It is straightforward to verify that, given the large detuning of the pump frequency $2\omega_n$ from the dipolar resonance ω_1, equation (9) yields a very small value for the absorption cross-section – much smaller in fact than the geometric cross-section of the particle. We turn then to the nonlinear interactions to show how the absorption cross-section dramatically increases when the threshold condition for PPR is met.

In solving equation (7) we notice at the outset that so long as the condition $|P_{n,m}^{(e,o)}(t)|<<\alpha_1A_P/\alpha_2$ holds, which is certainly the case at least at the initial stages of the parametric interaction, a solution can be easily

obtained in terms of Mathieu functions [14, 15]. More physically transparent though is the following slowly-varying-envelope approximate solution:

$$P_{n,m}^{(e,o)}(t) = p(t)\cos\left[\omega_n t - \theta(t)\right]\mathrm{e}^{-\frac{\gamma}{2}t}$$
$$p(t) = p_0\sqrt{\cosh\left(\frac{\alpha_1 A_p}{2\omega_n}t\right)}\ ;\ \ \theta(t) = \operatorname{arccot}\left[\exp\left(-\frac{\alpha_1 A_p}{2\omega_n}t\right)\right] \qquad (10)$$

where p_0 is the initial modal amplitude.

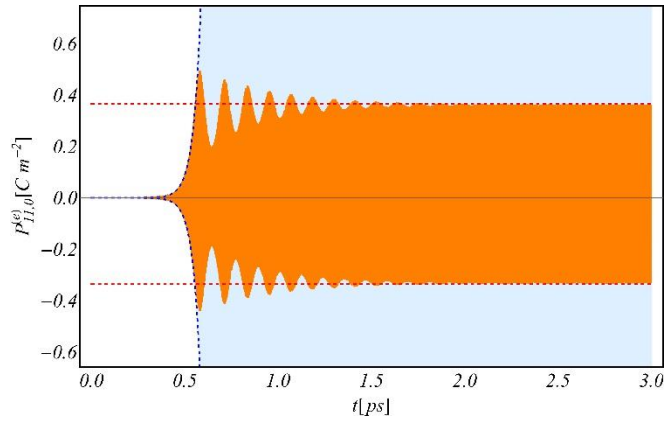

Figure 9 Polarization density amplitude term $P_{11,0}^{(e)}$ of a Silver sphere immersed in a MNA background medium. In order to better highlight the relaxation oscillations occurring in the system we show a case in which the PPR threshold is exceeded by a large margin ($A_p = 10A_{PPR}$). The dashed lines show the oscillation limits predicted by the asymptotic formula (12).

From equation (10) for $p(t)$ it is apparent that the system enters the PPR regime provided that the pump electric field amplitude A_p exceeds the threshold value:

$$A_{PPR} = 2\gamma\omega_n / \alpha_1 \qquad (11)$$

Such threshold condition separates distinct dynamics, whereby $P_{n,m}^{(e,o)}(t)$ 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 experiences. The power parametrically transferred from the pump to the resonant mode is given by:

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

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

Clearly a saturation of the exponential behavior is expected, because, if nothing else, the absorbed power (12) cannot exceed the finite power carried by the pump. In fact a different mechanism limits (12) before pump depletion occurs. Such mechanism is the resonance detuning due to the last quadratic term in equation (7) that we have neglected in our analysis so far. As $|P_{n,m}^{(e,o)}(t)| \sim \alpha_1 A_P / \alpha_2$ equation (7) can only be integrated numerically. Nevertheless the following asymptotic expressions as $t \to \infty$ for $P_{n,m}^{(e,o)}(t)$ hold for $A_p > A_{PPR}$:

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

Within the range of validity of equations (13) the exponentially growing oscillations of the polarization density amplitude level-off as $t \to \infty$ after a sequence of relaxation oscillations. In figure 9 the numerical solution of equation (7) (indicated in orange) is compared with the predictions of the analytical model (10) , shown in blue, and representing the PPR dynamics in the absence of saturation effects (i.e. $\alpha_2 = 0$). As evident from this comparison the PPR model (10) faithfully reproduces the dynamics of the actual system until the polarization density amplitude of the mode attains values close to asymptotic estimate(13), shown by the red dashed lines. At that point the exponential trend transitions into a train of amplitude oscillations that eventually relax towards a constant steady state.

Based on (13) the average power $\bar{W}_p$ transferred from the pump to the plasmonic mode approaches asymptotically the value:

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

Using the steady state asymptotic estimate (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 given by equation(9):

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

In equation (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 characteristic 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 equation (15) as $\alpha_{NL} = N\sigma_{NL}$.

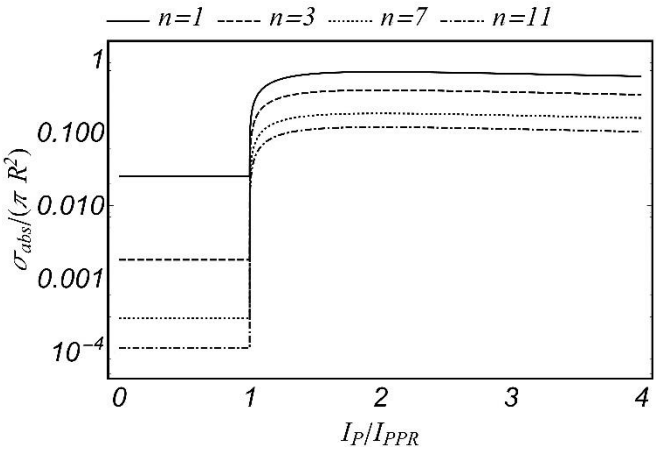

Figure 10 Absorption cross-section of the plasmonic particle normalized to the geometrical cross-section, as a function of the incident intensity.

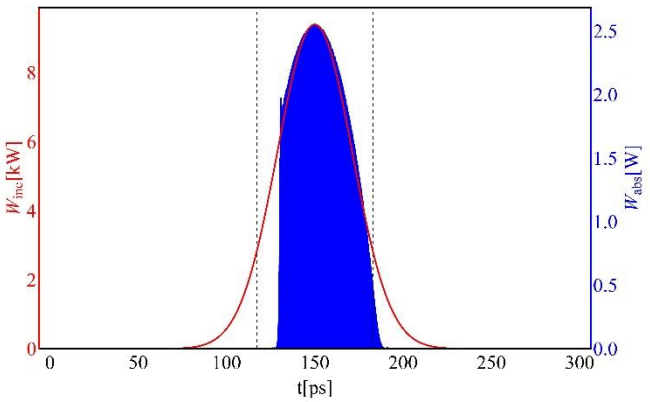

Figure 11. The red curve shows the instantaneous pump power W_{inc}. The blue curve show the instantaneous pump power W_{abs} absorbed by the particle via PPR of the n=11, m=0 mode. The dashed vertical lines indicate the times at which the pump intensity is equal to the PPR threshold.

Figure 2 shows how the normalized absorption cross-section of a silver sphere or radius $R = 100nm$ in a MNA 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 figure 2, the incident pump field is at twice the value of the corresponding resonant frequency given by equation(5). 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 $\lambda_\infty = 563nm$ and $\lambda_1 = 448nm$. As evident from figure 10, 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 form of reverse saturable absorption and could have interesting applications in optical limiting devices, especially given the design versatility of metallic nanoparticles for targeting different spectral regions.

For the purpose of illustration in figure 11 we apply the analysis and the models developed thus far to the practically relevant case of a pulsed pump. The pulse considered here is a 40ps pulse of average power 0.5W, focused to an area of $25\mu m^2$, on parametric resonance with the n=11 m=0 mode ($\lambda_{11} = 460nm$) of

a silver particle of radius 100nm embedded in a MNA background medium. The red curve in figure 3 shows the total instantaneous power of the pump field. The blue curve shows the instantaneous absorbed power mediated by the PPR process. The vertical dashed lines demarcate the time interval in which the pump field exceeds the PPR threshold. A similar behavior is observed both in figure 9 and figure 11 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 value through a series of oscillations (only one is discernible in figure 11). Figure 11 confirms the dramatic increase in absorption that PPAs exhibit under high intensity illumination.

In conclusion we have introduced the theory of Plasmonic Parametric Absorbers (PPA). In particular we have shown that 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 of being of fundamental interest in the emerging field of nonlinear plasmonics.

II.4 Tunable Extreme Anisotropy

In the context of time-varying metamaterials we explored the possibility of actively tuning the hyperbolic dispersion of a multilayer structure.

The unit cell of the proposed metamaterial is formed by two different materials of permittivity ε_1 and ε_2 with thickness d_1 and d_2 respectively as shown in figure 12.

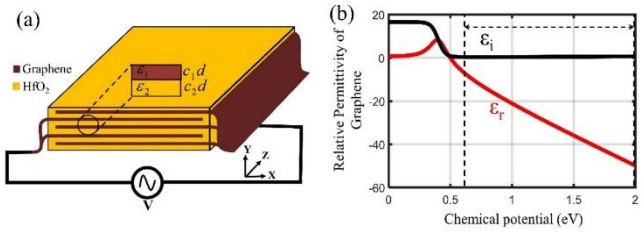

Figure 12 (a) Schematic diagram of metamaterial consists of periodic layer of HfO2 and mono-layer graphene and (b) Real and imaginary parts of single layer graphene permittivity ε_GR^∥ at 1550 nm wavelength as the function of chemical potential

In the proposed structure a HfO_2 layer and a monolayer graphene have been chosen to form the unit cell of MM. The schematic of the metamaterial is shown in figure 12(a). Graphene layers are interleaved for connection to the opposite sides of electrode, which allows electrical biasing to change the chemical potential. The standard mathematical model of HfO_2 permittivity ε_{HfO_2} in 1550nm wavelength window is used. The well-known Kubo's formula is used to represent the single layer graphene conductivity,

$$\sigma(\omega,\mu_c,\Gamma,T)=\frac{je^2(\omega-2j\Gamma)}{\pi\hbar^2}\times\left\{\frac{1}{(\omega-j2\Gamma)^2}\int_0^\infty \varepsilon\left[\frac{\partial f_d(\mathcal{E})}{\partial\mathcal{E}}-\frac{\partial f_d(-\mathcal{E})}{\partial\mathcal{E}}\right]d\varepsilon-\int_0^\infty\frac{f_d(\mathcal{E})-f_d(-\mathcal{E})}{(\omega-j2\Gamma)^2-4(\varepsilon/\hbar)^2}d\varepsilon\right\}$$

which is the combination of the inter-band and intra-band absorption represented by the 1st and the 2nd term, respectively. ω is the optical frequency, e is the electron charge, $\hbar=h/2\pi$ is the reduced Planck's constant, $f_d=1/\left(e^{(\varepsilon-\mu_c)/k_BT}+1\right)$ is the Fermi-Dirac distribution function. ε is the energy, k_B is the Boltzmann's

constant, T= 300K is the absolute temperature, Γ=5meV is the scattering parameter, and μ_c is the chemical potential. The conductivity predicted by Eq. (2) can be converted into an in-plane complex permittivity based on $\varepsilon_{GR}^{\parallel} = 1 - j\,\sigma/(\omega\varepsilon_0\delta_g)$, where ε_0 is the free space permittivity, and δ_g=0.34nm is the thickness of monolayer graphene. It is worth noting that the permittivity perpendicular to the graphene plane, $\varepsilon_{GR}^{\perp}$, is unity. $\varepsilon_{GR}^{\parallel}$can be tuned by charge doping or chemical potential change through externally applied voltage. The real and the imaginary parts of $\varepsilon_{GR}^{\parallel}$at 1550 nm wavelength as the function of its chemical potential are shown in figure 12(b). This provides the mechanism of tuning ε_T of the proposed metamaterial electronically. It is evident from figure 12(b) that within the chemical potential range from 0.57eV to 2eV as marked by two vertical dashed lines, the imaginary part of $\varepsilon_{GR}^{\parallel}$is nearly zero and remains almost invariant, while the real part of $\varepsilon_{GR}^{\parallel}$becomes more negative with the increase of chemical potential.

To achieve such a high level of chemical potential, a high electric field is needed that requires the dielectric material to have high electrical breakdown field, E_{bd}. Normally, bulk HfO_2 has an E_{bd} of lower than 1MV/cm. But when its thickness goes down to nanometer scale, E_{bd} becomes significantly higher and attains a breakdown electric field as high as E_{bd} ≈60MV/cm. The breakdown of nanometer-thick HfO_2 film is most likely initiated by bond rupturing rather than punctual defects as happens in bulk HfO_2.

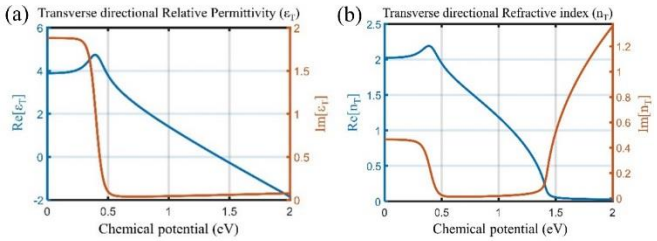

Figure 13 Real and imaginary parts of transverse directional (a) permittivity and (b) refractive index at 1550 nm wavelength as the function of chemical potential.

The extent of the tunability of the electromagnetic properties of the proposed metamaterial are shown in figure 13. Based on specific application and desired functionality, the proper range of chemical potential can be selected to design the devices with the maximum performance and tuning efficiency. In the article [J4] we have designed a variety of devices, including polarizers and directional couplers, relying on the proposed active metamaterial.

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III. Publications

Journal Papers

Published or accepted

[J1] Susobhan Das, Shima Fardad, Inki Kim, Junsuk Rho, Rongqing Hui, and Alessandro Salandrino, "Nanophotonic modal dichroism: mode-multiplexed modulators," Opt. Lett. 41, 4394-4397 (2016).

[J2] Shima Fardad, E. Alexander Ramos, and Alessandro Salandrino, "Accumulation-layer surface plasmons in transparent conductive oxides," Opt. Lett. 42, 2038-2041 (2017).

[J3] Alessandro Salandrino, "Plasmonic Parametric Resonance," Physical Review B (*rapid comm.*) 97, 081401(R), (2018)

[J4] Susobhan Das, Alessandro Salandrino, and Rongqing Hui, "Tunable hyperbolic photonic devices based on periodic structures of graphene and HfO2," J. Opt. Soc. Am. B 35, 2616-2624 (2018).

[J5] Shima Fardad and Alessandro Salandrino, "Plasmonic parametric absorbers," Opt. Lett. 43, 6013-6016 (2018).

[J6] Shima Fardad, Eric Schweisberger, Alessandro Salandrino, "Parametric Resonances in Nonlinear Plasmonics," (*invited)* Chinese Optics Letters – *accepted Aug 2019*

Conference Proceedings

Published or accepted

[C1] Alessandro Salandrino, and Eli D. Symm, "Accessing high-order resonances in plasmonic nanostructures," *(invited)* Proc. SPIE 9918, Metamaterials, Metadevices, and Metasystems 2016, 99182B

[C2] Alessandro Salandrino, and E. Alexander Ramos, "High Order Plasmonic Resonances in Time-Varying Media," *(invited)* SPIE Metamaterials, Metadevices, and Metasystems 2017

[C3] Alessandro Salandrino, "Plasmonic Parametric Resonace," *(invited)* 2018 IEEE Photonics Conference.

[C4] Alessandro Salandrino, and E. Alexander Ramos, "Accumulation-layer surface plasmons," *(invited)* SPIE Optics+Photonics 2018 - Metamaterials, Metadevices, and Metasystems.

IV. Honors and Awards

- 2016 Miller Scholar Award for Research, University of Kansas.
- 2019 Promotion to the rank of Associate Professor with Tenure.